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GLENN T. SEABORG

**Chief, Section C-1, Metallurgical Laboratory,
Manhattan Engineer District
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Lawrence Berkeley Laboratory
University of California

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PREFACE

This is volume III of a history of the research work of the many chemists who worked with me in the University of Chicago Metallurgical Laboratory during World War II. The work of these groups, which were affiliated under my direction in a unit which became known as "Chemistry Section C-I" as part of a broader Chemistry Division, was concerned with the development of chemical procedures for the extraction of plutonium, for the purification of plutonium and, in the later phases, for research on the isotopes of other heavy elements including other transuranium elements. Volume I, "History of Met Lab Section C-I, April 1942 to April 1943," was published as LBL special report PUB-112 in February 1977 and Volume II, "History of Met Lab Section C-I, May 1943 to April 1944" was published as LBL special report PUB-112, Volume II in May 1978.

This history is written on the basis of some 60 categories of information, including a wide range of Met Lab Progress Reports, notes on meetings, an almost complete file of all the Laboratory notebooks, personnel records, patent files, travel vouchers, organization charts, records of pile and cyclotron bombardments, health monitoring records, administrative bulletins, etc. Unfortunately, I did not keep a diary, but we were able to locate hundreds of notes I had taken to cover meetings, telephone calls, etc.; these were often written in a style that amounted to a code (in order to protect the secret nature of the information), but I found it possible to decipher them. This was one of the most important categories of information, as was the file of meeting notes which had been issued as informal reports and which usually included names of the attendees. Another welcome source was a very brief diary, actually an intermittent daily notation or a few words in a small pocket-size appointment book, kept by my wife Helen; this made it possible to include a large number of entries concerning our social life during most of the period 1942-1946 when we were in Chicago.

The style is such that the entry for each day is written as though it was entered in a diary on the basis of information available to me at the end of the day. There is more reliance on quotations from letters, written or received, than would be usual for an actual diary, which is justified on the basis that this kind of information comes closest to emulating a diary. An exception to this style of diary imitation is the footnotes that are included to give additional background material; these go beyond the activities of Chemistry Section C-I, covering meetings that I did not attend, and hence often include information that I could not have had on those entry dates.

Essentially all of the events, and the dates on which they are recorded, are based on the numerous categories of documentation; only a very small portion is based on memory alone, and even then it is usually associated with related information based on documentation. Chicago newspapers for this period, available in libraries, and weather records were used to embellish the narrative with some additional information on current events.

To help me in this writing task I wrote and talked to many who participated with me in the Met Lab experience to seek their recollections to augment the documented record. I am very grateful for this help. Since photography was not encouraged, the supply of illustrations is limited. I appreciate receiving early pictures of the Hanford Engineer Works from Orville Hill and photographs from the personal albums of Leon Levanthal, Tom and Betty Morgan, Fred and Edrey Albaugh, John and Lorraine Crawford, and Herman Robinson.

I am indebted to Joseph Katz, Carol Flaumenhaft, Sydney Gaarder, Bernard Saunders, Donald Stewart, John Farmakes, and Peter Seaborg for help in gathering and preparing material, and to Margie Hollander, Kathleen Van Der Haeghen, and Helen Seaborg for much help in putting this volume into publishable form.

MAY 1944

Monday, May 1, 1944

Willard asked Vernon to arrange for the release and shipment from Clinton Labs to Cunningham of a lanthanum fluoride precipitation to be made by Perlman on a one-gallon sample of the supernatant solution from the product precipitation in the first decontamination cycle at the Clinton plant. Our hope is to obtain a significant quantity of 93^{237}; recent experiments have indicated that a higher concentration of 93^{237} will be present during the early steps of the bismuth phosphate extraction-decontamination process than in the final Clinton product.

Hogness issued a summary of manpower distribution in the Chemistry Division here that shows how our staff of 92 men (total of 95, including Willard, Manning, and me) are being used:

		No. of Men	
		March	April
Thompson	Albaugh, extraction and		
Site W work	decontamination	13	13
31 men	Watt, concentration and isolation	8	9
	Dreher, process and development	9	8
Orlemann	Jensen, precipitation and		
Site Y work	extraction	6	5
32 men	Davidson, volatility	6	6
	Baumbach, metal production	16	16
	Orlemann, fluoride chemistry	4	4
Cunningham	Cunningham, basic chemistry	9	9
29 men	Dawson, recovery	11	10
	Ghiorso, instruments	10	10

Helen had coffee at Wilma's and then took a walk in the evening.

U.S. planes blasted targets in France, and the Soviets bombed Idritsa, a rail town near the Latvian border.

Tuesday, May 2, 1944

Roy C. Thompson, who has just received a Ph.D. degree in biochemistry from the University of Texas, started work in Section C-I. He is being hired as an Associate Chemist at $3300 per year and will be assigned to Albaugh's group.

Ralph James completed a series of bromate cycles (as in the neptunium-plutonium separation) on 49DC-31, a plutonium-containing fraction of the plutonium sample that was subjected to a 1,931 micro-

ampere-hour deuteron bombardment in the Berkeley cyclotron in March. He has been assuming that 95 might be more rapidly oxidized by $KBrO_3$ than is plutonium. His final lanthanum precipitate contained a total of 386 alpha-particle counts and 4 beta-particle counts per minute (he started with 5.5×10^6 alpha-particle counts per minute) and is still behaving like plutonium. Therefore if element 95 was formed in the bombardment, it does not oxidize more readily than plutonium.

Magnusson, who is working on the problem of isolating microgram amounts of Np^{237} (the long-lived isotope of neptunium), completed a series of oxidation and carrying experiments with Np^{239} tracer which demonstrates that oxidation of neptunium with dichromate does not occur at room temperature unless the fluoride ion is present. The oxidized state, after the addition of the fluoride, is VI as shown by its being carried with sodium uranyl acetate.

I had a meeting with Baumbach, Hagemann, Heath, Jensen, J. Katz, Manning, Orlemann, and Simpson to discuss purification and plutonium metal production problems. Commitments agreed upon include:

1. To press for the best measurements that can be made on the vapor pressure of plutonium metal in a tantalum crucible in one month. Simpson will need 10 mg of plutonium metal for this purpose.

2. Every effort is to be made to increase the scale on which plutonium metal is produced to the 10-mg scale and eventually the 100-mg scale. The best method for "mass production" of the metal will be decided on at a meeting next Monday. Other items discussed were preparation of fluorides for plutonium metal production, production of $PuBr_3$, preparation of the iodides, liquid phase halogenation of plutonium metal, preparation of fluorides, bomb reduction, electrolysis and the hot wire technique, need for another quartz balance, status of neutron counters, and the preparation of a pure plutonium compound.

At 6:30 p.m. there was a contamination accident in Room 36 of the New Chemistry Building when a centrifuge tube containing 200 mg of plutonium collapsed during centrifugation. Walling, Kelley, and Yett were in the room at the time. Small pieces of glass were thrown out of the centrifuge and some liquid was distributed around the inner wall of the centrifuge, but there was no evidence of any liquid on the floor. Not all of the plutonium has yet been recovered. There were no high alpha counts on the men's hands or clothing. Nose swab counts revealed: Walling 20 c/m, Kelley 8 c/m, Yett 150 c/m. Group leader Watt notified Nickson of the accident at 10:30 p.m. by telephone.

I held an evening meeting of the Council of our section at 7:45 p.m. in my office, attended by Cunningham, Dawson, Dreher, Ghiorso, Hindman, Katzin, Manning, Orlemann, Thompson, Watt, and Willard. I brought up the question of changing the Council meeting to 8:00 in the morning. Wednesday was finally selected as the best day. The Berkeley bombardment has been stopped (60 pounds of metal bombarded with high energy neutrons to produce

Np^{237}). Manning brought up the subject of the cleaning schedule. Aid to the dieners is to be given by the lab personnel at the time cleaning is done. It was suggested that the cleaning of walls and ceilings be done only once a week. The main stock of plutonium is to be locked in a safe, and cabinets with locks will be provided in each room for the stock in that room.

Helen spent the evening at Wilma's. I arrived home after the meeting with a migraine headache.

The mammoth battle between the President of the United States and Sewell Avery of Montgomery-Ward and Company is being heard in court. The issues are presidential power and whether or not the company falls in the category of war production.

Wednesday, May 3, 1944

I received from Thompson supplemental information on decontamination and isolation work for my use at the Clinton Steering Committee meeting day after tomorrow.

Hogness visited Simpson's laboratory today to check on the progress of our measurement of the vapor pressure of plutonium metal. He suggested that we give this project AAA-1 priority.

Kircher and I discussed Hogness's interest in a lifetime evaluation of rubber-lined equipment for use in HCl concentration procedures. Kircher does not think such a test is feasible with the equipment in our semiworks; furthermore he doubts whether an evaluation by us will carry enough weight to influence the decision of TNX personnel regarding the use of HCl concentration methods. We both agree that valuable information on the life of rubber-lined equipment can be obtained from industrial users. Kircher will write a letter to this effect to C. M. Cooper.

At 4:45 p.m. I received a phoned-in teletype from C. A. Prescott in Berkeley, who is head of the Radiation Laboratory Chemistry Department there and is an expert on making metal by the hot wire method using halides, saying he will arrive in Chicago by Streamliner, Sunday, May 7, and will be in town from 12:15 p.m. to 11:40 p.m. He asked if he could meet with me as per my invitation. Within the hour I replied by wire, explaining I shall return to Chicago from the South at 2:30 p.m. on Sunday, the day of his arrival. I suggested that we meet at 3:00 p.m. in the New Chemistry Building. Other people who plan to be there to confer with him include Manning.

At 7:45 p.m. in Room 133, Eckhart Hall, I spoke at the Technical Division's orientation lecture series for new employees. I gave a basic talk on the discovery of nuclear fission, our early work at Berkeley, and the nuclear reactions involved in the production of plutonium.

After the meeting Joe Hamilton dropped in at our apartment to see Helen and me. My headache was still with me.

At 11:30 p.m. Hogness, Gilbreath, and I left Chicago (from the Englewood Station) by Penn RR to Cincinnati, to continue on to Knoxville via the L&N RR.

There are reports in today's paper that a U.S. carrier task force destroyed 126 enemy planes and shelled Truk and its buttresses of Panape and Satawan last weekend.

The Project Council Policy Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Cantril, Chapman, Chipman, Compton, C. M. Cooper, Doan, Franck, Hamilton, Hilberry, Hogness, Jeffries, W. C. Johnson, McKinley, Mulliken, Pratt, Smyth, Spedding, Stearns, Vernon, Warner, C. J. Watson, W. W. Watson, and Wigner.

As part of Compton's "State of the Nation" message, he stated that Cockcroft is now in Canada to head up the Canadian development. Their plant will be a heterogeneous heavy water pile of 10,000 kw and a separations plant. It is not intended primarily as a war project.

Compton brought up the matter of reorganizing the management groups for the Metallurgical Project (Advisory Board, Technical Council, Administrative Committee). After discussion Compton summarized the comments as: (1) The Advisory Board is satisfactory as now set up [Conant, Jeffries, Thomas, Fermi, and Compton as chairman]. It is to meet every third Monday evening in the month. (2) Composition of the Technical Council might well be reviewed at this time. The Technical Council has the same personnel as the present Project Council with Allison as chairman. (3) Tuesday meetings should devote full time to information matters. (4) The Technical Council [executive session] should meet the first and third Wednesday mornings for "State of the Nation." The information sessions are to be held the first and third Tuesdays. (5) Administrative Committee members should name alternates. Present membership is Allison, Whitaker, Fermi, Latimer, Cooper, Stone, Hogness, Compton (chairman), Hilberry (vice-chairman). Meeting times have been set for the first and third Wednesdays after the executive sessions.

Cecil J. Watson, M.D., who is Associate Project Director for Health, gave a project-wide activities report for clinical medicine, medical research, and biological research for the period ending April 30 and gave a distribution of effort for April and May. Next, Cantril presented the April and May distribution of effort for Clinton Laboratories as pertaining to the Medical Section (S. T. Cantril, M.D., Director), Health Physics Section (H. M. Parker, Section Chief), and Biological Section (H. J. Curtis, Section Chief). He was followed by W. W. Watson who reported on the work of the General Physics Division at Chicago, Wigner who reported on the

Nuclear Physics Division at Chicago, and Doan who reported on the Physics Division at Clinton Laboratories.

Thursday, May 4, 1944

I arrived in Tennessee in the afternoon and am staying at the Vance Coopers'.

A Chinese bulletin estimated that 80,000 Japanese troops are attacking the Hankow-Peiping railway on Honan Peninsula.

Friday, May 5, 1944

In Oak Ridge. I attended the Chicago-Clinton Chemical Conference (Clinton Steering Committee meeting) along with Brown, Greager, Hogness, W. C. Johnson, Kirst, Squires, and later W. C. Kay. Topics discussed and agreements reached are as follows: (1) Coating problem. Hogness and I proposed the caustic sodium nitrate method of coating removal be tentatively adopted for the Hanford flowsheet. After discussion, it was agreed we would do further work on the method. (2) Extraction step. It was agreed that some of the five men working on basic process chemistry at Chicago will work on the extraction step. (3) Decontamination cycle. It was agreed that no more work will be done at either Chicago or Clinton on alternate oxidizing agents. Both Chicago and Clinton Chemistry Divisions will concentrate a large part of their decontamination studies on cerium and zirconium scavengers. (4) Concentration. In view of the high probability for the use of the method of plutonium fluoride precipitates in the 224 (concentration) building, work on the bismuth hydroxide method of concentration will stop.

Saturday, May 6, 1944

In Oak Ridge. I attended the second day of the Clinton Steering Committee meeting with Brown, Greagor, Hogness, Johnson, Kirst, Squires, and Kay.

The manpower distribution at Clinton and Chicago for the coming month was discussed and, at Chicago, will be approximately as follows:

Bismuth phosphate solubility	1 man
Decontamination studies	5 men
Basic chemistry of the process	5 men
Product precipitation studies	3 men
Effect of aluminum, zirconium, and NH_4^+ on extraction step	2 men
Concentration process for early operation at Hanford	2 men

Hanford isolation process	5 men
Uranous oxalate concentration method	1 man

Squires gave us information on the latest schedules for Site W. Chemical extraction plant shakedown, with dummy runs, to start between September 1 and October 15. The first pile will start up on September 15 and will reach 100,000 kw on October 1. The first pile material will be pushed about that date and cool for a month. By December, plutonium will be delivered to the 231 (isolation) building.

At about midnight, Hogness, Gilbreath and I caught the train (Southern RR) to Cincinnati where we will continue on to the NYC RR to Chicago. We are sharing a drawing room.

Mahatma Ghandi was released from detention this morning at Poona, India. He has been held by the British since August 1942. Press reports indicate that Ghandi is in poor health.

Sunday, May 7, 1944

I arrived in Chicago (Woodlawn Station) at 3:00 p.m.

I expected to meet Prescott at the New Chemistry Building at 3:00 p.m. but instead found a wire from him saying that he is in Nebraska on the Streamliner which is hopelessly delayed with a flat wheel.

Helen had coffee at Wilma's earlier in the day before I arrived.

Monday, May 8, 1944

During the time I have been gone from Chicago, the following events have occurred here at the Met Lab.

James, searching for plutonium isotopes other than Pu^{239} last Wednesday and Thursday, purified the original plutonium fraction (49DC-31) of plutonium sample 49DC (which was bombarded with deuterons in the Berkeley cyclotron) until a constant alpha/beta particle ratio was obtained. He finds that the ratio, as well as the complete spectrum of electrons and x-rays, is identical to unbombarded plutonium. Hence, from this experiment there is no evidence for the existence of isotopes of plutonium other than Pu^{238} and Pu^{239}.

Norm Davidson returned from a trip to New York and to Brown University in Rhode Island to review work on fluorides.

On Thursday my office received a copy of a letter from J. W. Kennedy to Captain Lavender in Washington concerning the patent negotiations on our discovery of element 94. Kennedy reviews our April 20 meeting and urges

Lavender to make the detailed study we discussed. He states again, as we did during the Washington meeting, that we inventors do not wish to appear difficult in these negotiations and prefer to accept Lavender's studied estimate of the actual value of the applications to any other course.

Katzin sent two memos to Moulton on Thursday giving additional information regarding patent cases enumerated in the record of invention forms MUC-PA-1036 (electrolytic oxidation and reduction of plutonium, reduction of fluorides and oxyfluorides by alkaline earths, precipitation of bismuth phosphate lanthanum solutions), and MUC-PA-830, S-1701 (decontamination of plutonium by electrolysis).

In a memo to Arnold on Thursday Manning listed the personnel of Section C-1 who require hand counting and nose counting for alpha, beta, and gamma-ray contamination.

Also on Thursday, Vernon sent a memo to Hilberry confirming that he will arrange to obtain the lanthanum fluoride precipitate containing Np^{237} from Clinton, as requested in Willard's May 1 memo to Vernon.

John D. Cockcroft, the new Director of the Montreal Laboratory, came to Chicago to see Allison last Thursday. Cockcroft and Allison are old friends; Allison spent a year at the Cavendish Laboratory, Cambridge University. Cockcroft's mission here was to discuss two topics in particular: (1) Future relations between the Montreal group and the Metallurgical Laboratory, and (2) Criteria for the selection of a heavy water pilot plant. Last month, on the 13th, Secretary of War Stimson held a meeting in his Washington office of the Combined Policy Committee, which consists of Anglo-Canadian and American representatives. It was decided to build a multi-million dollar pilot plant in Canada for the production of plutonium by the P-9 (heavy water) process. Organization of the Montreal group is as follows: Director of the Laboratory, Cockcroft; Director of the Physics Division, Halban; Division Heads — Placzek, Theoretical Physics; Paneth, Chemistry; and Newell, Engineering.

The evening meeting of the Purification and Metal Production Subsection of Section C-1, held Thursday at 7:45 in Room 209, Eckhart Hall, was attended by Frank, Fried, Hellman, Jasaitis, J. Karle, J. Katz, S. Katz, Manning, Robinson, Westrum, and others. Westrum described recent work on plutonium metal production. Tantalum wire furnaces with tantalum shields are now being used. Beryllium oxide crucibles are now being used exclusively. If barium is used, it is possible to reduce PuF_4 to plutonium metal with better than 80% yields. When prepared in the wet way, PuF_3 gives results which are extremely promising.

Jasaitis talked about studies on the melting point of plutonium metal. The best figure he has obtained with barium-reduced metal is 810°-830°C. Various metals have been tried in an attempt to observe alloy formation. There are indications that plutonium has an appreciable vapor pressure on heating to 1,700°C. From tantalum, a considerable amount of plutonium was volatilized onto a glass plate at this temperature.

S. Katz reported that the plutonium metal being produced is purer than 99.75% by spectrographic analysis. Plans are being made to determine the actual metal content by hydrogen evolution. Densities, determined by the modified Kirk method, are in the vicinity of 17 to 17.5 gm/cm^3.

Ray Frank reported that hardness of plutonium metal varies according to the refractory and the reducing agent used in preparation. The values fall into two groups, one in the vicinity of 150 on the Vickers scale and the other around 300.

Fried discussed the dissociation pressure of plutonium hydride. Values obtained for the dissociation pressure are 100°C, 19 mm; 200°C, 25 mm; 300°C, 43 mm. There are a number of anomalous results.

Karle and Hellman discussed bomb reduction and refractory studies with uranium, respectively.

Last Thursday Helen had lunch at Dagmar's and dinner at Wilma's.

At 10:30 a.m. last Thursday there was the regular weekly conference on the solvent extraction process, attended by Bird, Buffum, Dreher, Holt, A. C. Hyde, Kircher, Maloney, and Tepe. During the past week, runs 6 and 7 were completed, in which the ether extraction and aqueous extraction columns were operated independently. Product transfer was quantitative in both cases. Fission product transfer was 10% to the ether phase and 50% to the aqueous phase. An attempt will be made to operate the entire continuous system during the coming week. It was decided that future meetings will be scheduled from 10:30 to 11:30 a.m. every Thursday.

Last Friday Hagemann prepared plutonium bromide from plutonium metal using Br_2 specially purified to remove Cl_2 (obtained from Arnold). The preparation was made by treating a 75-microgram sample in an atmosphere of bromine in a quartz capillary at 350°-450°C for a total of 3 hours. The sample was sealed off in the capillary and submitted to Zachariasen as Sample H-15 for x-ray analysis.

My office received a memo to me from Allison on Friday asking if it would be possible for us to send 10 pounds of thorium carbonate to Montreal for use by Jules Guéron and A. G. Maddock. They are interested in the extraction of U^{233} from thorium.

Friday, a meeting on the basic process chemistry was attended by Ader, Albaugh, Bartell, Cunningham, Hindman, Howland, Malm, Morgan, O'Connor, S. Peterson, Sheft, C. Smith, S. G. Thompson, Watt, and Willard. Progress made in the program, proposed in document MUC-GTS-640 (summarizing the April 27 meeting on the basic process chemistry planning program, at which Albaugh, Cunningham, Hindman, Howland, Morgan, Thompson, Watt, and Willard were present) on problems arising in the bismuth phosphate extraction-decontamination process was reviewed. Plans were made as follows:

1. Further studies on poor carrying in the extraction step will be made.

2. There should be further study on the effects of variables on the carrying of Pu(III) and Pu(IV) by bismuth phosphate.

3. The mechanism of the inhibition by iron of carrying by bismuth phosphate will be investigated by preparing samples of bismuth phosphate precipitated in the presence of Fe(II) and of Fe(III) for x-ray analyses.

4. Crystal structure studies. Smith will prepare crystalline precipitates of phosphates with low ratios of bismuth to plutonium(IV) in order to clarify the mechanism of carrying plutonium(IV) by bismuth phosphate.

5. Work on solubilities of Pu(III) and Pu(IV) compounds and bismuth phosphate will be continued.

Helen had dinner at Wilma and Al Ghiorso's home last Friday night.

At 9:45 on Friday night, Manning, Arnold, and Ware entered Room 41 and found it in what Manning considered to be an inexcusable state of disorder. Among the objectionable conditions he found were ten or twelve drawers full of equipment open, nearly all cabinet doors open, gas burner flame on, paper towels scattered over the floor with only a slight tendency for the concentration to increase in the vicinity of the waste jars. There was no evidence of the occupants being in the area. Saturday Manning wrote a memo to the occupants of Room 41. In conclusion he said,

> The very considerable amount of time and money which is being expended in remodeling the building will be wasted so far as our goals of safety to personnel and purity are concerned unless far greater attention is paid to the elementary principles of laboratory housekeeping than was evidenced last night in Room 41. To insure success of our program each individual will have to assume a reasonable degree of responsibility for operating under safe and clean conditions.

Watt sent a memo to Nickson on Saturday about the high nose-swab counts taken of Yett on April 15 and April 22 (290 and 580 per minute). Watt concludes that the only likely source of alpha-particle exposure is a clinical centrifuge which was routinely being operated in the dust hood used by Yett.

Watt also described for Nickson last Tuesday's accident in Room 36, in which a centrifuge tube containing 200 mg of plutonium broke while in the centrifuge. He stated that so far 193 mg have been recovered with the figure subject to further revision. The room has been scrubbed thoroughly including the ceilings, walls and floor; a survey by Marilyn Jordan of the Radiation Protection group shows that the degree of contamination is not in excess of that reported prior to the accident.

Helen had lunch at Wilma's Saturday. She took care of the Ghiorso baby Kristine in the early evening, and then played cards with Al and Wilma later.

John G. Burr has been requested to transfer from Burton's Section C-II to our section to join Group 8 under Dawson.

I had a meeting today with Baumbach, Davidson, Heath, Jensen, Manning, Orlemann, Simpson, and Westrum to discuss purification and metal production problems. During the past week a total of about 175 mg of $PuCl_3$ was prepared in Davidson's group by the action of CCl_4 on dried $PuO_2 \cdot xH_2O$. A total of 50 mg of this material has been reduced in beryllia with barium, calcium, sodium, and potassium. Yields were poor in every instance. The fundamental difficulty is that $PuCl_3$ is so volatile that it distills through the inner crucible and, in some cases, even through the outer crucibles. For this reason it is considered undesirable to select $PuCl_3$ as the material for large-scale production by the present technique. It is, however, considered desirable to reduce $PuCl_3$ in bombs at the earliest possible date. Additional preparations of $PuCl_3$ by the "wet" method are to continue.

The following decisions were reached as to the work in the immediate future: (1) The starting material for metal production will be PuF_3 prepared by low-temperature dehydration of the precipitated material. These reductions will be made with barium. (2) $PuBr_3$ is to be reduced to plutonium metal as soon as material that can be characterized is obtained; a sample now in the hands of Zachariasen may be satisfactory.

Simpson said that within the next two or three days a tantalum oven suitable for the vapor pressure determinations of plutonium metal is expected to be available. Everyone is greatly concerned with the difficulty of avoiding the oxide coating on the metal surface that would seriously interfere with the vapor pressure measurements — perhaps adding barium to the plutonium will eliminate the difficulty although it may make the experiment uncertain.

The sample of plutonium bromide prepared by Hagemann on Friday was reported by Zachariasen late today as being $PuBr_3$. The structure is different from that of plutonium bromide obtained by treating plutonium with bromine that is not chlorine-free. This is satisfactory for metal production.

Helen worked in the Met Lab Information Division on the secret version of the "Table of Isotopes" which we are now compiling.

Reports today say that 4,500 Allied planes hammered targets in occupied Europe last night. Berlin, Bucharest, and other objectives were hit from a two-way assault from Great Britain and Italy.

Tuesday, May 9, 1944

At Ralph James' request, Art Jaffey performed a range measurement on the alpha particles from a portion of plutonium sample 49DC which has already been put through successive dichromate oxidations and LaF_3 precipitations in the search for element 95. Jaffey found no evidence of any alpha particles with a range different from that of Pu^{239}.

Hilberry sent the following list of the Project Technical Council members and observers to C. M. Cooper. He noted that administrative officers have been omitted since the objectives of the Council are now purely technical in nature. He also observed that any document containing the name of anyone on this list must be classified as secret.

PROJECT TECHNICAL COUNCIL

Project Office

A. H. Compton	N. Hilberry
R. S. Stone	R. S. Mulliken
A. J. Dempster	J. C. Stearns

Advisory Board

J. B. Conant	C. A. Thomas
Zay Jeffries	

Argonne Laboratory

E. Fermi

Clinton Laboratories

M. D. Whitaker	R. L. Doan
S. W. Pratt	S. T. Cantril
W. C. Kay	W. C. Johnson
O. H. Greager	D. M. Smith

Metallurgical Laboratory

S. K. Allison (Chairman)

H. C. Vernon	J. Franck
W. W. Watson	E. P. Wigner
L. Szilard	C. J. Watson
J. Chipman	C. M. Cooper (Secretary)

University of California

W. M. Latimer or E. D. Eastman

Observers

U. S. Engineers — A. V. Peterson
duPont — C. H. Greenewalt
Site Y — J. C. Warner
Ames — F. H. Spedding
Consultants — H. D. Smyth and R. C. Tolman

Helen worked at the Met Lab Information Division on the secret version of the "Table of Isotopes."

The Soviets have smashed through the main Axis defense line at Sevastopol. The Germans captured this city on July 2, 1942.

Wednesday, May 10, 1944

Ralph James completed the examination of the hydroxide soluble fraction (49DC-2) of plutonium sample 49DC which has undergone deuteron bombardment in the Berkeley cyclotron. He finds no rapid growth or decay of alpha activity. This indicates the absence of any new radioactive body unless it is of less than 5 minutes half-life.

I recommended to Richmond a merit increase of $5 per week for Edrey Smith, my secretary. I also said we might consider whether she might be eligible for promotion to the class "Secretary A" at this time as she is now supervising the work of five other secretaries and stenographers in my section and her work from all standpoints continues to be very good.

I wrote Clark J. Egan, Department of Chemistry, University of California, telling him I have seen his application blank which was referred to me after being forwarded from down South. I said we would like very much to have him come with us and that I am asking our Personnel Office to make him an offer.

I met with Albaugh, Cunningham, Dreher, English, Howland, S. Thompson, and Willard to discuss modifications of the basic process chemistry program and pertinent new information from Clinton Laboratories. Plutonium losses of 12-20% have occurred lately in the extraction step of the Clinton Plant even though bismuth phosphate extractions from the same solutions in the laboratory give high yields. It is possible that some IV to VI oxidation reactions proceed through the mechanism of IV disproportionating into III and VI, followed by oxidation of III to IV. Investigation of this will be part of the regular basic chemistry group under Hindman.

The following proposed new experiments on basic process chemistry were assigned: 1. *Extraction step difficulties.* Ader and Howland will test the effects of oxides of nitrogen. The rate of reduction of VI to IV by formic acid will be followed spectrophotometrically. 2. *Analysis for valence state of product carried from process solutions.* Malm will do this by potentiometric titration. 3. *X-ray crystallographic studies.* Smith will attempt to prepare more completely crystalline bismuth phosphate containing plutonium(IV) by mixing known crystalline bismuth phosphate with known crystalline plutonium(IV) phosphate and digesting the mixture.

Helen began attending the Office of Civilian Defense (OCD) class for recreation for children at the downtown YWCA. She worked at the YWCA in the afternoon.

The Soviets regained Sevastopol after a 24-day siege, clearing the Crimea peninsula of the invaders.

The U.S. withdrew control of (Montgomery) Ward's since the union elections have taken place.

Thursday, May 11, 1944

Vance Cooper is visiting the Met Lab from Oak Ridge.

Orlemann sent me the manpower assignments in Sub-section II as of May 11, and showing the following organization: Group 4, Purification and Analysis of Plutonium Compounds — Jensen (Group Leader), Reinhardt, Stein, Dixon, Brody. Group 5, Volatility and General Dry Chemistry of Plutonium Compounds — Davidson (Group Leader), Hagemann, J. Katz, Brody, Karle, Hyde. Group 5A, Chemistry of the Plutonium Fluorides — Orlemann (Group Leader), Heath, Florin, Meyer, Zvolner. Group 6, Plutonium Metal Production — Baumbach (Group Leader), S. Katz, Fried, Westrum, Robinson, Frank, Jasaitis, J. Karle, Hellman, Gerstein. High Vacuum Technique Sub-Group — Simpson, Phipps, Seifert, Sears, Gilpatrick, Johnson.

I acknowledged the receipt of Kennedy's May 2 letter. I explained that Underhill (Secretary of Regents, University of California) is in the East at the present time and that I have an appointment with him in Chicago on Saturday, May 20; Shane has asked that I talk with Underhill on this matter at that time. I invited Kennedy to participate in the conference if he comes to Chicago next week and can stay over until Saturday.

Thompson asked Vernon to secure Clinton process samples for use here.

I attended the evening meeting of the Basic Chemistry, Recovery, and Instrument Groups of our section at 7:45 in Room 209, Eckhart Hall. Others present were Ames, Arnold, Cunningham, Davidson, Dawson, Dreher, English, Hindman, Kirk, Kohman, Kraus, Krueger, La Chapelle, Manning, McLane, Meyer, Morgan, O'Connor, Orlemann, Peterson, Rosenfels, S. W. Sheel, Cliff Smith, Studier, R. Thompson, S. Thompson, V. Cooper, Willard, and others. After introductory remarks, I turned the meeting over to Cunningham.

Hindman gave the results of his spectrophotometric examination of solutions of Pu(III), (IV), and (VI) in order to determine the usefulness of spectrophotometry as an analytical tool for the quantitative estimation of the various valence states. O'Connor reported on the solubilities of Pu(III) and Pu(IV) phosphates under various plant conditions. He is surprised to find that the solubility curves for HCl and HNO_3 solutions are practically the same. Data were also given on the solubility of plutonium(IV) oxalate, which confirm the previously observed fact that Pu(IV) forms a strong oxalate complex.

Magnusson brought up the points of difference in the chemistry of plutonium and neptunium that would permit the separation of the two elements. He has found that, in the presence of fluoride ion, neptunium is oxidized to the sodium uranyl acetate carryable state by dichromate; earlier work in Berkeley suggested that dichromate oxidizes neptunium to an intermediate valence state not carried by sodium uranyl acetate. Apparently, the fluoride ion acts as a catalyst. I summarized the discussion which followed, stating that Magnusson's experiments do not disprove the existence of a +5 state for neptunium, but they do show that for the experiments cited the assumption of a +5 state is unnecessary. The following experiment also suggests a method for the separation of neptunium and plutonium: If a mixture of the two is treated for one hour at 75°C with 0.1 M $Cr_2O_7^{=}$, in 1.0 M HAc, the plutonium

can be carried by sodium uranyl acetate but neptunium cannot. Since this experiment was carried out in the absence of fluoride, it is possible that this surprising fact is due to slower reaction rates of neptunium with oxidizing agents of plutonium. Magnusson also reported that he has been unsuccessful in separating neptunium and plutonium by ether extraction of a solution containing 5 M NH_4NO_3 and 0.05 M $Cr_2O_7^{=}$ after 10 minutes digestion; these conditions are not sufficiently drastic to oxidize plutonium to the (VI) state.

Jaffey reported on range measurements on the alpha particles from 93^{237}. Since only 50 to 100 counts per minute were available for the determinations, a high geometry method (aluminum absorber method) had to be used. He found a value of 3.26 cm for the range. Cunningham mentioned that Ghiorso obtained approximately the same values for the range of these alpha particles.

Helen worked at home. Vance Cooper had dinner with us in our apartment, went to the meeting with me, and then spent the night with us.

James V. Forrestal has been nominated to succeed the late Frank Knox as Secretary of the Navy.

Friday, May 12, 1944

I met with Baumbach, Davidson, Heath, Jensen, Manning, Orlemann, and Simpson to discuss purification and metal production problems. The work during the past week was reviewed and decisions as to future policy were agreed upon. Significant points brought out were:

1. The needs for PuF_3 will be no larger than 0.5 gram during the next month.

2. The higher fluoride of plutonium has apparently been successfully transferred from nickel to glass by revolatilization at 300°C in nitrogen. A sample of the material is in the hands of Zachariasen for identification.

3. Densities of the order of 17 gm/cm^3 have been obtained for samples of metal prepared recently from either plutonium fluoride or chloride.

4. Beginning next week the scale upon which plutonium metal is produced will be increased to the 30-mg level. With present equipment it should be possible to increase the scale to 175 mg. As of today, one reduction bomb has been delivered to us, and it should be possible for work on bomb reductions to proceed.

5. To date all preparations of $PuBr_3$ have resulted in a mixture of $PuBr_3$, PuOBr, and an unknown phase. A commitment to provide 20 mg of $PuBr_3$ of at least 90% purity for metal production by next Friday was made.

6. Work should continue on developing a better method for the direct synthesis of $PuCl_3$ from oxide using CCl_4.

7. The status of metal vapor pressure work was described. The tantalum crucible has been obtained, and initial measurements around 500°C will be made using a 6-mg piece of fluoride-reduced plutonium.

At 10:30 a.m. in Room 261, Ryerson Laboratory, the weekly conference on the solvent extraction process was held, attended by Bird, R. S. Buffam, Dreher, Holt, A. C. Hyde, Kircher, and Maloney. It was noted that M. R. Fenske, Professor of Chemical Engineering at Pennsylvania State College, will become a consultant to the Project. Solutions are being prepared for a 13-hour run of the continuous system which should start on May 16.

The report "Chemistry Division Summary Report for April 1944" (CS-1658) was issued today. The work of Section C-I is summarized under three headings: Separations Process Studies, Purification and Metal Production, and Basic Chemistry.

Willard sent a memo to Allison and Vernon covering the Section C-1 review of drawings for the stainless steel laboratory hood to be used at Hanford. He also transmitted to Vernon drawings of an apparatus for punching platinum discs for counting samples, which du Pont wishes to duplicate for use at Hanford.

I saw Jasaitis who told me that today he observed melting of plutonium metal on a tantalum strip at 700°C or possibly even lower.

Helen attended the OCD class at the downtown YWCA. She and I played cards at Ghiorsos' in the evening. Vance Cooper again spent the night at our apartment.

Four thousand planes rained 7,000 tons of bombs on 19 Nazi centers, raiding rails in four nations.

Saturday, May 13, 1944

In a memo to A. C. Hyde, Cunningham describes the role the semiworks is to play in recovering Np^{237} from the 60 pounds of uranium metal (now at the West Stands) which has received about 100,000 microampere-hours with high energy neutrons at the Berkeley cyclotron. The plan is to have the semiworks dissolve the metal, reduce the solution with Fe^{+2}, and precipitate the neptunium with LaF_3. The precipitate will be turned over to Magnusson and La Chapelle for further chemistry. The efficiency of the operations in extracting neptunium will be determined by adding Np^{239} and analyzing for recovery of this tracer.

I received a six-page memo from Thompson giving the progress of work in Sub-section I and presenting some plans that he, English, and Albaugh made as a result of my trip to Oak Ridge last week. He mentions that 16 scavenger test runs are planned of which eight will involve the use of Ce(IV)-Zr(IV) scavengers and eight will involve the standard Hanford flow-sheet for control purposes. He also states that $PbSO_4$ appears to have

desirable properties as a scavenger for barium and strontium. In the slug recovery program the caustic-sodium nitrate method of coating removal has been used on a large batch of slugs and works very satisfactorily. An almost invisible film of scale remains, however, which must be removed before the slugs can be dissolved. He states that the semiworks is interested in C. M. Cooper's suggestion of redesigning the ether extraction column to allow for putting a concentrated ammonium nitrate solution in the top to scrub out those fission products which are carried through the ether phase.

Helen had lunch with Wilma. In the evening we went to the movies at the Frolic Theatre and saw "Women of the Town" with Claire Trevor and "Whistling in Brooklyn" with Red Skelton. Vance Cooper again spent the night with us.

Berlin has acknowledged that Allied troops in Italy have broken through in the Liri Valley but added that Allied troops have nowhere reached the "actual German defense system."

Sunday, May 14, 1944

Vance left to return to Oak Ridge. At 11:10 tonight Crawford will be leaving on a one-week visit to Clinton Labs.

I played golf during the day while Helen visited Wilma. In the evening Helen and I saw the movie, "Girl Crazy" with Mickey Rooney and Judy Garland at the Frolic Theatre.

Monday, May 15, 1944

Ralph James began working up the sample of sodium plutonyl acetate that has been in the Clinton pile for about a month. He hopes that, through the Szilard-Chalmers mechanism, any Pu^{240} formed would be found as Pu(IV) and thereby concentrated. He finds that a considerable quantity of the PuO_2^{++} has been reduced during the period of the irradiation as evidenced by its precipitation with HF. His first counting data on this reduced fraction show no detectable long-range alpha particles and hence no evidence for a new isotope of plutonium having alpha particles of range longer than those of Pu^{239}.

Janet Owen, one of our secretaries, resigned in order to join the Red Cross for overseas duty.

James B. Conant is visiting the Met Lab and inspected New Chem facilities including the apparatus built by Simpson and Phipps for measurement of the vapor pressure of plutonium metal.

Albaugh described for Thompson plans to have eight men in Group 1 participate in extensive experiments to test the Ce(IV)-Zr(IV) scavenger combination.

The heavy water pile at Argonne went critical today for the first time. It consists of a cylindrical tank six feet in diameter with 136 rods (each six feet long) inserted vertically into the heavy water moderator. Although 8,200 pounds of heavy water have been accumulated to date, which would fill the tank to a height of 125 cm, criticality was achieved when the water rose to a height of 122.5 cm (critical volume 3,214 liters; critical mass 3.44 metric tons). The discrepancy between the calculated and observed critical volumes is thought to be due to errors in the exponential experiment or to an underestimation of the effectiveness of the pile's reflector.

Beginning at 10:30 a.m. in Room 209, Eckhart Hall, I attended an all-day meeting with C. A. Thomas on the final purification and metallurgy of plutonium. Others present at the morning session were Allison, Ashcraft, Chipman, Derge, Eastman, L. S. Foster, Franck, Hogness, Jeffries, W. C. Johnson, C. S. Smith, Spedding, Thomas, and Warner. The afternoon session which began at 2:00, was attended by Allison, Ashcraft, Chipman, Derge, Eastman, L. S. Foster, Franck, Hogness, W. C. Johnson, C. S. Smith, Spedding, Thomas, and Warner; part of the time by Dempster, Hilberry, and W. W. Watson; and later by Jeffries, Compton, and Conant.

C. S. Smith described the metal reduction program at Site Y, indicating that lithium is the only reducing agent consistently successful under the range of conditions studied on the 50-mg scale. Vitrified BeO is the best crucible material and when it is used, lithium has successfully reduced PuF_3, PuF_4, and $PuCl_3$ with quite respectable yields. Reductions on the one-gram scale in the centrifuge have given satisfactory yields in vitrified BeO at about 1,100°C with either PuF_4 or $PuCl_3$ as starting material. The average density of five buttons is 17.2 gm/cm^3. The highest density material was heated in vacuo and observed to melt at 805°C. Smith also reported on work on electrolytic reduction in a mixed chloride electrolyte which produced only small spheres (weighing up to several milligrams). The fact that the spheres were obtained at 660°C suggests the metal or alloy produced has a melting point below this.

Eastman and Spedding reported for Berkeley and Ames, respectively. Following them I presented the work of our section. In covering our studies with plutonium metal, I reported that x-ray diffraction photographs by Zachariasen show that the plutonium metal produced by the action of barium and sodium on the trichloride has the same complex unknown structure previously obtained. Plutonium metal made by reduction of the tetrafluoride with calcium in beryllia also shows the typical complex structure. I stated that plutonium metal made by barium reduction of the tetrafluoride in beryllia has melted at 810°C in vacuo on tantalum. I reviewed the status of the vapor pressure of plutonium metal and made the following observations: An apparatus for the measurement, using a tantalum effusion unit, is now complete. Means have been provided for collecting ten different effusates without breaking the vacuum; it is hoped to obtain results in several weeks.

Among the decisions reached and assignments made are:

1. Experiments on production of plutonium by reduction with active metals and by electrolysis should be continued at high priority ratings.

2. Work on remelting plutonium and stand-ins should be continued at high priority ratings.

3. Work on the determination of the physical properties of plutonium and on the effect of impurities in plutonium on physical properties remains important. It seems important to obtain approximate values for the vapor pressure of plutonium within the next few weeks.

4. Problems on the production of pure halides of plutonium remain urgent.

5. Further work on plutonium hydride should have a low priority rating.

The meeting concluded with a summary of decisions reached and assignments to be carried out.

Watt sent a 13-page memo to Thompson transmitting information from Perlman which Watt considers to be of considerable importance to the experiments currently in progress in the Separation Processes Sub-section. The information covers such items as further work on carrying and dissolving properties of the beta form (difficultly soluble) of bismuth phosphate, use of scavengers, metathesis problems, solubility of plutonium peroxide, investigation of plutonium compounds suitable for storage and shipment, and investigation of the chemistry of plutonium.

Helen had lunch with Marie Barnes and attended the OCD class at the YWCA. Iz Perlman, visiting from Clinton, had dinner with us at home. Then he and I attended the Chemistry Division Seminar.

Top headlines today come from the Italian front and announce that Allied troops have cut the Cassino-Formia highway — the Germans' main lateral road of supply.

At 8:00 p.m. the Project Advisory Board had its first meeting (Eckhart Hall). In attendance were Compton (Chairman), Conant, Fermi, Jeffries, Thomas, and part time, Stone and Stafford Warren. Compton opened the meeting by explaining that the function of the Board is to give the best advice possible to him as Project Director. The problem of the Board was defined as the consideration of the course of action to be pursued by the Metallurgical Project which would be to the best interest of the DSM Project as a whole.

The major item for discussion was the Health Program. Compton presented a brief history of the growth of the Health Division, pointing out that as time has passed new health problems have continually appeared. He interpolated with a brief discussion of the subject of radioactive warfare, emphasizing the fact that no positive work is being done in this connection, but that knowledge important for such a field is being gathered as a by-product of the general Health Division Program concerned with the health problems and hazards incident to the operation of plant facilities as designed. The immediate problem is the determination of how extensive the health

program should be for the coming year, including both the health service functions and the necessary clinical and biological studies.

Dr. R. S. Stone then presented the health program. He gave a historic survey of the growth of the present health program. The first question that was faced at the start was whether or not 1/10 r per week should be accepted as the "tolerance dose" for radiation exposure and, if so, enforced. The earliest studies indicated that this tolerance dose was not on a sound basis and, consequently, needed study. Moreover, the accepted value was a choice among several and, as far as a legal basis for choice was concerned, the only foundation upon which such choice could be based was to carry through actual studies. The work at the National Cancer Institute fitted into the program at this point beautifully, and their cooperation was immediately obtained. The second problem that arose promptly was the hazard due to fission products. Again experimental investigation was required because of the paucity of information. A systematic study of the metabolism of fission products was needed and was programmed with Dr. Hamilton at the University of California commencing the work with tracer studies and with Dr. Cole at the Metallurgical Laboratory following through with studies utilizing significant quantities. This latter work is now coming to the fore with the steadily increasing quantities of fission products now being made available.

The expansion of Cole's section and the budget for the Health Program were discussed. Next, Stone talked about curative investigations of the Health Program. Up to the present time these have consisted of vacations, transfusions, and hope. Studies are now being instituted on the effect of vitamins and other curative agents. He then reviewed clinical tests being developed by Dr. C. J. Watson and his group on kidney and liver function, the role of phosphorous, etc., as a basis for tests of incipient radiation damage and also incipient toxic effects. The program can be summarized then in the following manner. In the early stages of the program two hazards were realized, radiation and toxicity. There was a considerable body of knowledge on radiation effects, and the necessary corollary studies were immediately undertaken and have been largely completed. In the case of toxicity, it was indicated that the hazard is serious. This led to a program of toxicity studies which is still continuing. The program then expanded as new hazards were discovered. At present it can be roughly presented as follows: 1) toxicity studies, 2) dust and gas problems, 3) tumor and fibrosis problems, and 4) product hazards studies.

Compton continued the meeting by quoting from a discussion he had with Curtis at Clinton indicating that the plutonium studies program is in a transition stage from acute to chronic investigations. The acute investigations are well in hand. It is hoped that acute hazards may be avoided so it becomes necessary to know what the chronic problem will be. At this point there was a general discussion of the problem. Compton then raised the question as to whether any real result would come out of the dust program. Dr. Stone answered that knowledge is required for a determination of what might happen

to Project personnel in case of accident and what the result would be in case of radioactive warfare. Conant commented on this, answering that as far as radioactive warfare is concerned, the results are either unimportant or else would be much too late.

Fermi made brief comments on beta- and alpha-particle dosages to the lungs. He feels that an extrapolation for beta ray effects from the known x-ray effects is safe, while in the case of alpha-particle radiation, the effects might be quite different. Dr. Stone emphasized that although a calculation is excellent, an experimental verification of the computations is really necessary. Dr. Thomas raised the question as to whether or not the studies in progress on the effects of polonium might not be sufficient to cover the effect due to 49. Dr. Warren pointed out that the important thing is the way in which the various materials are distributed in the lung and that this depends upon the nature of the material and its physical constitution. Actually the dusts tend to settle in patches leading to local damage.

The question was then raised as to whether the chronic plutonium experiments should be undertaken. Dr. Warren stated that he would like to see one alpha-particle emitter and one beta-particle emitter investigated together with x-ray and neutron studies. This would imply an increase in facilities of about that indicated for Cole.

The proposed distribution of plutonium to be produced at Clinton, as proposed by Whitaker, was reviewed and approved. Compton pointed out that the production schedule at Clinton will be affected by the shift to the Hanford process in July. The question of Fermi's activities was discussed. It was decided that his services are really needed by the Met Lab through the summer until the Hanford plant gets into operation and that a decision should then be reached as to whether Fermi's services are needed more by Chicago or by Los Alamos.

The meeting adjourned at 11:30 p.m.

Tuesday, May 16, 1944

In a memo to members of Group 1, Albaugh gave a detailed plan of the scavenger experiments to test the use of cerium and zirconium. Each experiment will be on a 100-ml scale and will be carried through the Hanford flowsheet to the plutonium precipitation in the crossover cycle.

Margolis and Blaedel described for Dreher their chemical experiments in ether extraction during the period April 27 to May 12, during which time they have determined the distribution of nitric acid in UN-H_2O-ether systems and the dependence of this distribution on UN, HNO_3, and dichromate concentrations.

At 9:30 a.m. in Room 209, Eckhart Hall, I attended a Project Council Information Meeting on Chemistry. Others present were Allen, Allison,

Ashcraft, Boyd, Burton, Chapman, Chipman, Compton, Conant, C. M. Cooper, Doan, Eastman, Fermi, Franck, Gensamer, Greager, Hogness, Jeffries, W. C. Johnson, McKinney, Mulliken, Perlman, Seitz, Selwood, C. S. Smith, Spedding Stearns, Stone, Sugarman, Sutton, Thomas, Vernon, J. Warf, Warner, W. W. Watson, Whitaker, Wigner, and Zachariasen. Allison opened the meeting and asked Hogness to introduce the Chicago Chemistry Division speakers. As the first speaker on the program I reviewed our work on Hanford problems. I mentioned that no very critical conditions have been found except possibly in the extraction step where there is evidence of oxidation. I said that not much doubt remains that the concentration and isolation step, as outlined, will work for Hanford. I mentioned that one run has been made to determine the yield of 93^{239} in the presence of Hanford concentrations of 94^{239}, fission products, and 93^{237}; it is not carried very well by bismuth phosphate, a yield of 5% being obtained through extraction plus one cycle. From this it appears that 35 days cooling should be sufficient at Hanford so far as protection from 93^{239} radiation is concerned. Next, I reviewed our work on Los Alamos problems including the production of plutonium metal and halides. I indicated that the highest densities yet obtained for the metal are about 20 gm/cm^3 with an average density of about 17 gm/cm^3. Our best guess is that the melting point of plutonium metal will be 750°C ± 100°C. I talked about our work on the carrying of Pu(III) and (IV) with bismuth phosphate and the interesting work on the chemistry of neptunium that shows it is carried by sodium uranyl acetate if HF is present (in dichromate-oxidized solution).

Perlman, in reporting on isolation studies at Clinton Labs, mentioned the possible shipment of dry plutonium from Hanford to Site Y. This set off a discussion led by Allison and Thomas who were of the opinion that we are supposed to ship plutonium as a concentrated solution. It was finally agreed that there has been no change in the official decision to ship in the form of a concentrated solution.

Warren Johnson stated that the following quantities of plutonium have undergone irradiation in the Clinton pile: 25 mg — in pile 30 days; 50 mg — in pile 40 days: 50 mg — in pile 10 days. Eastman reported that Gofman has developed a modification of the oxalate extraction-decontamination process and has tested the first steps on a one-liter scale with Site W concentrations. Later steps have been tested on the one-ml scale. Overall yield is at least 95% with a gamma decontamination factor of 6×10^5.

Helen took a walk with Wilma, then prepared dinner for us and our guest, Ermon Eastman.

Again today's top news comes from Italy where American and French troops continue to advance, seizing more towns and 2,000 prisoners.

Wednesday, May 17, 1944

At 8:00 a.m. I held a meeting in my office of the Council for our section, attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Hindman, Katzin, Orlemann, Simpson, Thompson, Watt, and Willard. I mentioned that the Argonne heavy water pile has been started with considerably less heavy water than expected. I reported that Fermi, using the one gram 94^{239} sample we prepared for him, has found evidence for the absorption of neutrons in 94^{239} to form 94^{240}; thus the properties of 94^{240} will be of critical importance.

Matters pertaining to the organization of Section C-I were discussed. Willard will leave June 1 to go to Hanford, and Watt will take his place as Associate Section Chief. Pye will then become group leader of the Concentration and Isolation Group (Group 2). A Group 6A (high vacuum) under Simpson is to be created. Another Group 5A under Heath is also to be formed.

I mentioned some of the points brought out in yesterday's Project Council Information Meeting, including Sugarman's finding that the so-called bremsstrahlung in beta-counting is due to scattering around the absorbers and that if the absorbers are put directly on top of the samples, good results in reducing bremsstrahlung are obtained. I also mentioned Spedding's finding that plutonium can be leached from beryllia with boiling nitric acid.

At the conclusion of the meeting I reviewed the roster of men available in the building for special services. These include: (1) Building engineer, Stackhouse; (2) Head janitor, Slattery; (3) Property, Lange; (4) Procurement, Donlan; (5) Building manager, Howard; (6) Shop, Emery; assistant, Edgerton; (7) Machine shop, drawings (Ryerson) Cantrell; and (8) Shop order priority, Selwood.

In accordance with a request made by Vernon on May 8 and referred to me by Hogness the next day, I submitted the following estimate of needs for uranium and uranium compounds in connection with the work of Section C-I for the next six months. The tabulations are as follows: Sub-section I [S. G. Thompson]: 30 lbs uranium metal (for process development work), 1500 lbs uranium metal (for semiworks operations exclusive of ether extraction), 500 lbs uranium in the form of inactive UNH (for ether extraction work), and 15 lbs uranium metal (for extraction-decontamination studies); Sub-section II [E. F. Orlemann]: 2 lbs uranium metal (for use in purification and metal production) and 10 lbs uranium metal in the form of various uranium compounds (for use in purification and metal production); Sub-section III [B. B. Cunningham]: 120 lbs uranium metal for special bombardments, i.e., production of 93^{237}, and 31 lbs uranium as various compounds (for use in studies relating to 93^{239} and extraction processes).

I summarized the total definite needs in the following way:

Uranium metal	1,667 lbs
Uranium as UNH	500 lbs
Uranium as other compounds	41 lbs
	2,208 lbs

I also noted that there may be need in Sub-section I for an additional 30 lbs of uranium metal for process development work and 250 lbs of uranium as inactive UNH for ether extraction work.

Helen went to her OCD class, had lunch with Dagmar, and coffee at Wilma's.

Today's top headlines indicate that the Allies continue to advance in Italy. Other reports say that the Japanese are making a new threat toward India.

The Project Council Policy Meeting was held at 9:00 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Eastman, Fermi, Greager, Hilberry, Jeffries, Johnson, Mulliken, Oppenheimer, Spedding, Stearns, Stone, Szilard, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. In his "State of the Nation" message, Compton gave the latest information on the Hanford schedule: The first pile unit will be operating at power in late August or early September. The first extraction plant is to be completed September 1, with first actual operations November 1. Problems still not completely solved include canning and film. Decontamination and purification are still not completely in hand.

Fermi initiated an extensive discussion of the difficulty of jacketing slugs which will withstand full power operation; the possibility of using long helium-filled tubes as a substitute was considered. Fermi pointed out that although the P-9 pile at Argonne has been brought to criticality, it has not been operated at power because of the lack of sufficient heavy water to cool it. This shortcoming will be remedied in another month.

Compton mentioned that the basis for the interchange of information with the Canadians is still under discussion. We are not at present free to exchange information on 49 and its chemistry. Whitaker stated that Clinton will be ready to test chemical separations with a full number of cells about June 15, and this will be followed by tests of the Hanford process. The last few groups of eight batches have given overall yields above 80%. Whitaker also raised the question of what to do with the 20 lbs of thorium in the pile. About 50 mg of U^{233} have been accumulated. Compton questioned whether it would be necessary to push it out at all, to which Whitaker replied that it is only a matter of gas pressure. Oppenheimer stated that he would be glad to study the U^{233} if the chemistry can be handled.

Eastman brought up the purity limits set for 49, indicating that if limits loosen up, the lag in passing on this information should be kept to a minimum; Oppenheimer promised to cooperate in releasing this information. Compton agreed there is a possibility of using plutonium in a way which would not require the high purity, but we are not yet in a position to have any confidence in it. Eastman mentioned that Gofman is leaving next month to enter the Navy for medical training at his own request.

Thursday, May 18, 1944

The weekly solvent extraction meeting was held at 10:30 a.m. in Room 261, Ryerson Laboratory.

"Chemical Research—Basic Chemistry of Plutonium, Report for Month Ending May 1, 1944," (CK-1587), was issued. The studies reported are as follows: Basic Chemistry. Howland has measured the potentials of the Pu(III)/Pu(IV) couple in acetate, fluoride, and phosphate solution and obtained values that range from -0.4 v to -0.8 v. Hindman has measured the potential of the Pu(IV)/Pu(VI) couple in 1 M H_2SO_4 at 25°C and estimates it to be -1.3 v. O'Connor has investigated the solubility of plutonium(IV) phosphate under a variety of conditions from the point of view of determining whether this compound at Hanford concentrations can precipitate as a pure phase. He finds that this cannot occur at any of the conditions of the extraction and decontamination process. This is also true of plutonium (III) phosphate.

Patton has carried out an investigation of the rate of gas evolution from a plutonium nitrate solution prepared in the manner proposed for shipment and storage at Site W. He finds that the rate is about four times the theoretical calculation of 1 ml per gram of plutonium per 24 hours. He has also determined the composition of plutonium(IV) 8-hydroxyquinolate and finds fairly strong evidence that the coordination number of Pu(IV) is eight. Ames has studied the absorption spectra of Pu(IV) in 1 M HNO_3 with a Beckman quartz spectrophotometer and finds that the principal absorption bands of probably analytical usefulness obey Beer's law in the concentration range up to 0.02 M plutonium.

Kraus has studied the rate of oxidation of Pu(IV) to Pu(VI) in nitric acid solutions in connection with possible oxidation to Pu(VI) in plant operation. The experiments suggest that the rate of oxidation falls off rapidly with increasing HNO_3 concentration. He is investigating the hydrolytic behavior of Pu(III) and Pu(IV) in order to anticipate difficulties likely to be encountered in storage of or operation with plutonium in weakly acid solution (1.0 N and less). Kraus has also investigated the pH dependence of the solubility of Pu(IV) in 1.0 M Na_2SO_4 and in perchloric acid. He has also determined the solubility of plutonium(IV) in 0.3-0.4 M Na_2SO_4.

Instruments. Jaffey has measured the range of 93^{237} alpha particles using the air-screen low geometry counter with aluminum absorbers. He has obtained a value of 3.26 ± 0.05 cm in air at 760 mm and 15°C.

Recovery and Services. Asprey, Stewart, and Studier have purified two relatively large batches of plutonium received from Clinton. In addition, they have rendered 475 mg of extremely pure plutonium for Ghiorso to use in studying the radiation spectrum of plutonium. Asprey, Britain, Fineman, Leventhal, Stewart, Studier, and Wetlaufer have recovered 205 mg of plutonium for reuse by various groups in Section C-I. Dawson, Asprey, Studier, and Britain have worked out an improved ether extraction method for plutonium recovery. Ferric ion is used to dissolve the LaF_3 by complex formation and to aid in salting out the plutonium. A single batch of ether may be used for several extractions by alternately freezing the solution being extracted and the receiving solution. In this way the ether can be poured back and

forth. Plutonium yields of 85 to 95% have been obtained. Beard and Britain have worked on a method of analysis for plutonium on filter paper used by the Health Group to collect air samples.

I attended the evening meeting of the Separations Processes Subsection of Section C-I at 7:45 in Room 209, Eckhart Hall. Others present were S. Thompson (who led the discussion), Albaugh, Bartell, Dreher, Fields, Fineman, Flox, Gilbreath, Gilpatrick, Greenlee, Hoekstra, Howland, Hyman, Jasaitis, Katzin, Kelley, Kirk, Larson, Lincoln, Margolis, Morgan, Peterson, Post, Phipps, Pye, Sedlet, Sheft, Simpson, R. Thompson, Walling, Watt, Willard, and others. Gilbreath reported on his studies of low acidity scavenging using lanthanum phosphate. This method gives promising results as far as increased decontamination is concerned but requires increased volumes and careful pH regulation. Hoekstra explained the differences between the proposed Hanford procedure and present Clinton plant and laboratory practice. Probably the most important difference is that about 10% less volume of acid is to be used to dissolve the precipitates at Site W than at Site X. Walling described concentration experiments (peroxide precipitation) simulating Site W conditions and carried out starting with two 400-ml bismuth phosphate by-product supernatants from Cell 3 at Clinton which had been spiked to Site W plutonium concentrations. An overall yield of 94.8% was obtained through the second cycle.

Pye reviewed another concentration experiment in which he metathesized a 7.5-g lanthanum fluoride sample from Clinton containing 1 gram of plutonium. Crud and light oils were present, and only a 78% yield was obtained after two peroxide precipitations. Dreher stated that the continuous ether extraction unit is ready for testing. Much work is yet to be done on the proper reducing agent to be used in the second column. Hyman described the present methods being employed for removal of the silicon-aluminum bond from uranium slugs. Lincoln reported that, in tests of the prereduction step, twenty-five 700-ml runs on semiworks-dissolved material gave 25 different extraction results after running through a formic acid reduction.

Helen played golf at Jackson Park and later had coffee at Wilma's.

"Nazis Pull Out of Cassino" is the headline in this morning's paper.

Friday, May 19, 1944

At 9:00 a.m. I attended a meeting at which Fermi reported on recent values of nuclear constants of U^{235} and Pu^{239}. Others present were Allison, H. L. Anderson, Burton, Castle, C. M. Cooper, Dancoff, Fermi, Goldsmith, Hughes, Morrison, Mulliken, Seitz, Snell, Sugarman, Szilard, Turkevich, W. W. Watson, Weinberg, Wigner, and Young. Fermi has recently measured the absorption cross section of Pu^{239} for thermal neutrons by the transmission method. His absorber consisted of the mixture of one

gram of plutonium oxide and two grams of graphite powder sample which we prepared for him. His measured value is 950 barns. Fermi also reviewed the present status of the branching ratio (neutron capture to fission) of U^{235} and Pu^{239}. He has found evidence pointing to a greater branching ratio in Pu^{239} than in U^{235}. This means 49 has a large n,γ cross section. He has measured the number of neutrons emitted in fission per neutron captured to be slightly less for Pu^{239} than for U^{235}; the number of neutrons emitted per fission is somewhat larger for Pu^{239}, being 2.8 as compared with 2.4.

I received a letter from E. C. Lingafelter of the Department of Chemistry, University of Washington, saying that he will not be able to join our group except possibly during the summer. He did not feel that he would be of use to us.

Watt sent me the latest flowsheet for the peroxide isolation process. In the covering memo he says, "Undoubtedly a number of additional modifications in the process will be made before it is actually put into use at Hanford. The present edition of the flowsheet, therefore, should be looked upon merely as a progress report which incorporates results of our most recent laboratory experience."

Dreher sent A. C. Hyde a recommended procedure for operation of the aqueous extraction column in the continuous solvent extraction process.

"Chemical Research — Separation Processes for Plutonium, Report for Month Ending May 1, 1944," (CN-1585), was issued. It includes the following information of major interest:

I. Extraction-Decontamination. Hoekstra, Ader, and Sheft have determined the effects of deviations from Hanford flowsheet conditions in small-scale tests of individual process steps using Site W concentrations of plutonium and fission products and glass equipment. They find that considerable variations can be tolerated in the extraction step, oxidation procedure, and by-product precipitation. Sedlet has tested the effect of Fe(III) on plutonium yield in the extraction step using Site W concentrations of plutonium and fission products on a 10-ml scale. Results show a gradual increase in plutonium loss from 1.1% to 12.8% as the Fe(III) concentration increases from zero to 0.06 M. Bartell, Malm, and Sheft have completed three Site W-level, 100-ml scale, process test runs carried through two bismuth phosphate cycles and a crossover cycle according to the Hanford flowsheet. The runs are designed to test the effect of varying ratios of steel surface to liquid volume and twice Hanford concentrations of fission products. The effects are not significant. Sedlet and Sheft have carried out plutonium precipitations at high acidities and at Clinton concentrations in order to test the effect on product carrying by bismuth phosphate and decontamination. Using Fe(II) as the reducing agent, they find that decontamination is considerably improved, but at the expense of a 3 to 4% plutonium loss. When U(IV) is used as the reducing agent, there is no improvement in decontamination. Greenlee has formulated optimum conditions for the use of Pb_3O_4 as an oxidizing agent in the Bismuth Phosphate Process on the basis of

small-scale, Hanford concentration runs, made in the presence of stainless steel. Also tested were $Na_2S_2O_8$ and $NaBiO_3$ which were found less promising.

II. Concentration-Isolation. Pye and Walling have studied Hanford concentration by the fluoride method in an effort to improve dissolution of the metathesis products. They find that the use of a metathesis reagent consisting of a mixture of KOH and K_2CO_3 provides a metathesis product that may be dissolved in HNO_3 leaving only a very small HNO_3-insoluble residue. By this procedure, plutonium losses are much smaller than when 15% KOH alone is used. Kelley and Yett have demonstrated the feasibility of recycling by-product solutions from peroxide precipitations to the lanthanum fluoride plutonium precipitation in the concentration (crossover) cycle. This procedure appears to be a means of recovering plutonium not initially separated as plutonium peroxide during early operation at Site W when plutonium concentrations may be as low as 30 grams per ton. Experiments are continuing. Haeckl finds that the use of $Bi(OH)_3$ in a concentration (crossover) cycle shows promise as a substitute for the fluoride concentration process. Goeckermann and Hopkins have worked on the establishment of optimum conditions for the precipitation of plutonium peroxide. They find that improved plutonium yields and more granular plutonium peroxide precipitates result from process solutions containing H_2SO_4 as well as HNO_3. They have demonstrated that plutonium peroxide precipitations under Hanford flowsheet conditions may be carried out in 25-12 stainless steel. In a single 25-ml experiment, an overall product yield of 99% is obtained. Asprey and Goeckermann, considering the storage and shipment of plutonium, find that Pu(IV) nitrate solutions containing as much as 1,200 grams of $Pu(NO_3)_4$ per liter (607 g plutonium per liter) can be produced by dissolving freshly precipitated plutonium peroxide in 16 N HNO_3.

III. General Problems in Process Chemistry. Lincoln and Dreher have studied the decontamination factor with respect to Np^{239} in the Bismuth Phosphate Process. Two runs on a 10-ml scale with tracer concentrations show 80% of the neptunium is carried in the extraction step and a 14% yield after one decontamination cycle. With Hanford concentrations of plutonium and neptunium, the yields of neptunium were 3.6% and 8.7%, respectively, in two runs. Thompson and Ghiorso have calculated the effects of Pu^{239} radiations on decontamination factors in the Hanford separations process and find that when solutions of fission activity, such as can be safely handled in the laboratory, are enriched with plutonium to Hanford concentrations, the limiting gamma-radiation decontamination factor that can be measured is of the order of 10^5 for uranium originally containing 50 micrograms of plutonium per pound and 10^6 for uranium originally containing 500 micrograms of plutonium per pound. Morgan, Katzin, and Thompson have made calculations which show that, considering the decontamination achieved by the Bismuth Phosphate Process with respect to Np^{239}, and the time required for processing in the canyons, it should be possible to start chemical processing of the metal at Hanford after 35 days of cooling.

IV. Basic Chemistry of the Separations Process. Hindman, Howland, Kraus, and Malm have investigated the following: a) Prereduction step. Oxidation of Pu(IV) to Pu(VI) may occur to a considerable extent. b) Extraction step. Pu(IV) has been found to be the only stable carryable

form. Solubility measurements indicate that a pure phosphate of either plutonium(III) or (IV) could not precipitate in the process solutions in the extraction step at Hanford concentrations of plutonium. c) Oxidation step before second product precipitation. Reduction by excess Fe(II) produced plutonium(III) momentarily, but this is re-oxidized to Pu(IV) on heating. d) Second product precipitation. Malm and Howland find that the mechanism of carrying Pu(IV) by bismuth phosphate cannot yet be elucidated, but they show by oxidimetric titration of co-precipitated Pu(IV) that it is carried in the (IV) state rather than the (III) state. C. Smith has prepared plutonium(III) phosphate crystals which have been examined by x-ray crystallography by Dr. Mooney. The examination shows that the crystal lattice is the same as the hexagonal bismuth phosphate. O'Connor has measured the solubilities of plutonium compounds in process solutions — extraction step and decontamination cycles.

"Chemical Research — Special Chemistry of Plutonium, Report for Month Ending May 1, 1944," (CK-1586), was also issued. The following investigations were summarized: A. Stein, working on the purification of plutonium compounds, has determined separation factors for iron, magnesium, and phosphorus in the case of various organic solvents; and for aluminum, beryllium, calcium, iron, lithium, magnesium, sodium, and silicon using organic reagents to complex the plutonium.

Volatility and General Dry Chemistry. J. Katz has prepared plutonium(III) chloride by a low temperature method: Pu(IV) chloride solution is reduced with a slight excess of HI, evaporated to dryness in a stream of HCl at 50°C, and dehydrated in the HCl stream by heating to 300°C. Zachariasen reports an x-ray pattern showing only the lines of anhydrous $PuCl_3$. An effort to duplicate this result was unsuccessful. I. Karle has prepared $PuCl_3$ from $PuO_2 \cdot xH_2O$ by reacting with CCl_4 vapor at 750°C. It gives a green sublimate of well-formed crystals; Zachariasen has reported an x-ray pattern showing only the lines of anhydrous $PuCl_3$. In air this material forms blue crystals that Zachariasen finds to be isomorphous with a hydrated $NdCl_3$.

Hagemann has reacted Br_2 with 100 micrograms of plutonium metal at 300°C and obtained a blue green $PuBr_3$ that is shown by Zachariasen to be isomorphous with $LaBr_3$, $CeBr_3$ and UBr_3. E. Hyde has studied the hydrobromination of plutonium compound, taking a Pu(IV) solution in HBr to dryness at 70°C in a stream of HBr. The Pu(IV) is reduced to Pu(III) under these conditions, and dehydration yields a green product which gives an unidentifiable x-ray diffraction pattern. Wolf has developed a procedure for the analysis of milligram quantities of plutonium halides by potentiometric titrations of the bromide and chloride.

Fluoride Chemistry. Meyer has prepared a total of 120 mg of PuF_4 for plutonium metal production during the past month by the reaction of HF with three different batches of the oxide at 600°C. An attempt to prepare PuF_4 by dehydration of $PuF_4 \cdot xH_2O$ gives a product which results in a very poor yield upon reduction to metal with barium. Similarly poor results are obtained with samples of Pu(IV) and Pu(VI) nitrates which are vacuum-dried and hydrofluorinated in the presence of O_2 at 600°C. Florin has studied the reaction of hydrogen-HF mixtures with PuF_3 and PuF_4 and finds that a mixture of hydrogen fluoride containing *ca.* 0.6% H_2 reduces PuF_4

to PuF_3 at 600°C. PuF_4 is not reduced by HF alone at 600°C. Florin has also studied the condensation and revolatilization of a plutonium higher fluoride made by the action of fluorine on PuF_4 at 600°C. When passed through a nickel tube, 94 percent condenses in the temperature range 50°C to 160°C.

Production and Properties of Plutonium Metal. S. Katz has used the pycnometric method to measure the density of plutonium metal samples made by barium reduction of PuF_4 in the following refractories with the results (in gm/cm^3): tantalum (18.8), ThO_2 (18.2), and BeO (17.4, 17.7, 19.2). Fried has determined the composition and hydrogen dissociation pressures of two preparations of plutonium hydride. In one case $PuH_{4.0}$ was found with a heat of formation of 6,000 calories per mole of $PuH_{4.0}$. The other preparation shows $PuH_{3.1}$ with a heat of formation of 8,500 calories per mole of $PuH_{3.1}$. Frank has found that plutonium metal in BeO, La_2O_3, ThO_2, and tantalum shows hardness values from 145 to 368 Vickers units. Jasaitis and Robinson have improved their apparatus for the melting point determinations of plutonium (better vacuum and provision for vibration of the tantalum strip). Using this apparatus, they find that plutonium metal prepared in BeO by barium reduction of PuF_4 melts as low as 810°C; the metal is shown to be of high purity by spectrochemical analysis.

Westrum has found that reductions of PuF_4 with barium in degassed BeO using three-minute firing at 1,230°C in vacuo have been consistently successful in over 20 cases, giving plutonium metal of higher density, greater purity, and greater hardness than plutonium metal prepared in ThO_2 crucibles. He also has found that compressed PuF_4 pellets of density 6.2 gm/cm^3 have yielded 72% and 86% plutonium metal when 5 to 8 mg quantities are reduced with barium in BeO. Satisfactory reductions of PuF_4 with barium have been made in lanthana and tantalum crucibles. The use of $PuCl_3$ (prepared by action of CCl_4 on $PuO_2 \cdot xH_2O$) in place of PuF_4 gives a high yield of plutonium metal. Hellman, Gerstein, and Frank have conducted further refractory studies. Lanthana and tantalum metals show promise.

Robinson and Jasaitis have studied the chemical properties of plutonium metal, attempting unsuccessfully to prepare plutonium nitride and to amalgamate plutonium with mercury at room temperature. Ghiorso has considered the effect of the gamma- and x-radiation associated with purified plutonium and concludes that the radiation hazard is not serious because of the low energy of the gamma-radiation (0.3 Mev) and the self-absorption by the plutonium.

Helen attended the OCD class and had lunch with Betty Bloechel, a member of her class. In the late afternoon Helen and I played golf at Jackson Park.

Although the main headline reports continued Allied successes in Italy, a smaller banner indicates that two islands (Wakde Islands near New Guinea) have been won by U.S. troops.

Saturday, May 20, 1944

Work began today to isolate microgram amounts of Np^{237} from 64.5 pounds of uranium metal that has been bombarded with high energy neutrons at the Berkeley cyclotron. The initial extraction step is being carried out in the semiworks. The metal will be dissolved in two batches and a lanthanum fluoride precipitation made from Fe^{+2}-reduced solution to which 93^{239} tracer has been added to permit detection of the element in the presence of plutonium and fission-product radiation. The precipitate will be taken over by Magnusson and La Chapelle who are monitoring the semiworks phase.

Cunningham sent A. C. Hyde the detailed procedure for the semiworks to use in extracting the Np^{237} from the Berkeley neutron bombardment of uranium metal.

I summarized for Hamilton, at his suggestion, the three main experiments which we have in mind to do in the future with the Berkeley cyclotron. I pointed out that these experiments could best be done after the cyclotron has undergone the contemplated change so as to produce appreciably higher energy-bombarding particles than at present:

(1) Bombardment of uranium with high-energy helium ions in order to produce 94^{240} via the reaction $U^{238}(\alpha,2n)94^{240}$ and perhaps via the reaction $U^{238}(\alpha,pn)93^{240}$, followed by beta decay. It is important to produce a sufficient amount of this material to test whether it undergoes fission with slow neutrons in view of the recently observed, apparently large, cross section for the n,γ reaction on 94^{239}. (This is important from the standpoint of the operation of the final device made from 94^{239} and from the standpoint of the possibility of "burning" U^{238} on a large scale for power production purposes in chain reacting devices.)

(2) Bombardment of 94^{239} with deuterons to look for the formation of radioactive 95^{240} or 95^{239}. It is important to know the chemistry of element 95 for three reasons: a) If 94^{240} produced at Hanford decays by beta emission to 95^{240} and if 95^{240} has a high spontaneous fission rate or emits long-range alpha particles, it would be necessary to remove it from the 94 before final use. b) Knowing the chemistry of 95 would make it possible to look for this element in nature. c) It would extend our general knowledge of the chemistry in the transuranic region.

(3) Bombardment of 94^{239} with helium ions to produce 95^{241} or 95^{242} by the reactions $94^{239}(\alpha,pn)95^{241}$ and $94^{239}(\alpha,p)95^{242}$. Reasons for producing these isotopes are the same as stated under (2) above. The isotopes 96^{241} or 96^{242} might be produced by the reactions $94^{239}(\alpha,2n)96^{241}$ and $94^{239}(\alpha,n)96^{242}$ or by the beta decay of 95^{241} or 95^{242} (in the improbable event these are beta emitters). The radioactive isotopes of element 96 would be used to investigate the chemical properties of this element in order to extend our overall chemical knowledge of the transuranic region.

I also mentioned as other experiments which might be worthwhile, the deuteron bombardment of U^{235} to produce U^{236} and the deuteron bombardment of U^{238} to produce enough 94^{238} to measure its fission properties. I suggested we might want to continue to make 93^{237} by means of cyclotron neutrons.

Helen and I had dinner at the 51st Street YMCA with the Baumbachs and then went to a movie.

Banner headlines today are again from Italy and the Pacific.

Sunday, May 21, 1944

I played 18 holes of golf at the Evergreen Golf Club (91st Street and Western Avenue) with Luther Arnold and Stan Thompson. Scores were, LA-108, ST-116, GTS-114. We rode out to the course with Arnold, who has a car and is able to procure sufficient gas for the trip. This course is also available to us via three successive streetcars.

Helen was ill part of the day but was able to prepare dinner for Iz Perlman and me.

Crawford returned to Chicago at 2:45 p.m. from his one-week trip to Clinton Laboratories.

Albaugh is leaving at 11:50 p.m. tonight on a trip to Clinton Laboratories to obtain information on the use of scavengers in the Clinton semiworks. Ralph James and Ghiorso are leaving at 11:55 p.m. on a trip to St. Louis to carry out deuteron bombardments of plutonium using the Washington University cyclotron.

Monday, May 22, 1944

Watt suggested, by memo, a consolidation of the monthly CN reports and the reports prepared each month for the Clinton steering committee meetings. He believes that such a consolidation will save time and effort on the part of all concerned, but successful operation is dependent upon meeting the established deadlines.

Today I had a meeting with Underhill, Secretary of the Regents of the University of California, who is in town, to discuss the patent applications of Kennedy, Segrè, and element 94. I found him in complete agreement with the plan we presented to Captain Lavender and which Lavender rejected, including the amounts involved and their distribution. Underhill and I agreed it would be a good idea for him to participate in the next conference with Lavender, providing Kennedy and Segrè approve. Perhaps we can meet in Chicago first and then go on to Washington. Underhill is going to hear from Lavender and will then let me know what the next move should be. I sent a letter to Kennedy today giving him this information.

In a memo to A. C. Hyde on the use of Pb_3O_4 as an oxidizing agent in the Bismuth Phosphate Process, replacing $NaBiO_3$, Dreher proposed that it be tested in as many as four semiworks runs.

Marjorie Bohlman, who has been working for J. B. Miles, transferred to Cunningham's group.

Helen attended the OCD class and had lunch with Wilma. Marilyn Howe and Wilma had dinner with us in our apartment.

U.S. troops are still moving quickly in Italy, according to today's headlines.

Tuesday, May 23, 1944

Baumbach, Davidson, Heath, Orlemann, Simpson, and I met to discuss purification and metal production problems. The items covered are as follows: Various refractories have been tested for production of plutonium metal using barium reduction of plutonium fluoride. Tantalum was unsatisfactory as a refractory, but La_2O_3 yielded material of density 19.6 gm/cm^2 which is to be evaluated metallographically and spectrographically. Everything is in shape for a run on a 30-mg scale except for a satisfactory refractory. Despite its limitations, beryllium oxide probably will be used.

J. Karle now has available all equipment for centrifugal reductions and additional stainless-steel reduction bombs are being made in the shops. Bomb reductions will be tried using pellets of the halides. As for the vapor pressure of plutonium metal, in the initial tests using a tantalum crucible at 1,300°C, considerable difficulties were experienced: a great deal of vaporization of an unknown material occurred, the vacuum was not satisfactory, the threaded lid sintered to the bulk of the crucible, and the thermocouple leads alloyed with the crucible. A 6-mg piece of plutonium metal will continue to be set aside for the vapor pressure work.

Plutonium bromide of satisfactory purity has been made in platinum by the action of HBr on the oxide; it also has been sublimed in platinum in HBr at about 1,000°C. Such material made in platinum will be supplied for plutonium metal production when the metal production group wants it. It has only been possible to synthesize plutonium oxyiodide up to the present. Platinum apparatus for carrying out the reaction of iodine with plutonium metal is being constructed, and apparatus for the synthesis using HI is being prepared.

"Chemical Research — Separations Processes for Plutonium. Process Development and Semiworks Operation," (CN-1584), was issued, covering joint efforts of the Technical and Chemistry Divisions up to April 15. In summary, Hyman, Makins, and Flox report on the removal of bonded aluminum-silicon jackets from slugs using sodium hydroxide with the addition of hydrogen peroxide and sodium nitrate. Tests in the semiworks of the effects on the bismuth phosphate separations process of the insoluble scale remaining after removal of the jackets show waste losses of 7.5% as compared with 2.8% for control runs. Four semiworks runs have been made during the month to provide bismuth phosphate precipitate for concentration studies. About 26 pounds of Clinton pile metal containing

1.3 mg of plutonium were dissolved for each run. The present Clinton process procedure has been followed except that U(IV) was used in all four runs for prereduction. Two lanthanum fluoride-hydrochloric acid concentration runs and one uranous oxalate-hydrochloric acid concentration run have been made during the month on a scale equivalent to about 150 pounds of uranium metal. Margolis, Jordan, and Malekow have studied (1) the distribution of Pu(VI) between 46% uranyl nitrate-1 M nitric acid solution containing 0.02 M sodium dichromate acid and 40% uranyl nitrate-ether solution, and (2) extraction of plutonium from ether solutions with aqueous solutions containing reducing agents. The semi-works continuous countercurrent extraction equipment has been used for four initial ether extraction runs. Results are inconclusive.

An electroscope survey by the Health Division, Room 29, New Chem, occupied by Sedlet, shows that his working area gives the weekly tolerance of radiation in three hours. The acid hood next to the door measures 100 mr in 1½ hours, and it was recommended that his work area be adequately shielded.

The Baumbachs, Edrey Smith, and Wilma had dinner in our home.

Today's headlines are all from Italy reporting a German counter-attack at Turocina, coastal gateway to the Pontine plain (58 miles south-east of Rome).

Wednesday, May 24, 1944

At 8:00 a.m. I held a meeting in my office of the Council of Section C-I, attended by Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Heath, Hindman, Katzin, Manning, Simpson, S. Thompson, Watt, and Willard. I noted that job-rating sheets are due and that Watt, Willard, and Manning will go over them with the group leaders. Various tests of centrifuge tubes to be used with product were discussed in view of the accident on May 2.

Thompson is consolidating his reports so that the Clinton Steering Committee, the Hogness (Chemistry Division) abstract, and the regular CN report will cover essentially the same material. Orlemann is expanding the Thomas meeting report into his regular report. The Cunningham report is to be CN with additional emphasis on basic chemistry. The Council decided against the use of canvas overshoes for wearing in our "clean" building. It was also pointed out that talc should not be used with rubber gloves because of the dust hazard; cornstarch was suggested as a substitute.

James and Ghiorso returned to Chicago at 7:30 a.m. from their trip to St. Louis where they went to bombard plutonium with deuterons. Last Monday they bombarded a 42-microgram sample of plutonium mounted on platinum, a 75-microgram sample mounted on beryllium, and sample 49D Be-4 which was bombarded with deuterons at the Washington University cyclotron on January 29 of this year. The three samples were bombarded

for 10-15 minutes (50 to 150 microampere hours), and alpha counts were started within 6 to 8 minutes after shutdown. The next day, Tuesday, they repeated the deuteron bombardments of plutonium. There was no evidence of growth or decay of alpha particles; they conclude there is no short-lived (between one minute and a few days) long-range alpha activity produced by deuteron bombardment on 94^{239}.

Today James purified the major fraction (16.2 mg plutonium) of the sodium plutonyl acetate that has been irradiated in the Clinton pile for about a month. Part of the purified material (4.43 mg) was ignited to plutonium oxide and placed in a short tube. We plan to place it in the Clinton pile for a long indefinite irradiation (sample designation SA-2). James also prepared another 8.2-mg plutonium oxide sample in a long tube, using some of Dawson's stock plutonium. It will be irradiated for one month at Clinton (sample designation SA-1).

Hogness sent a memo to Allison requesting the following changes and new appointments, at my recommendation, be made in Section C-I: George W. Watt — to Associate Section Chief to replace Willard who will be going to Hanford; Donald G. Pye — to Group Leader of Group 2, to become effective upon Watt's appointment; Roy E. Heath — to Group Leader of Group 5A; and Oliver C. Simpson — to Group Leader of Group 6A, a group splitting off from Group 6 (Metal Production), to be made up of T. E. Phipps, R. Seifert, G. Sears, L. Gilpatrick, and F. Johnson.

I received a teletype from Prescott of the University of California Radiation Laboratory who is visiting Oak Ridge. He indicates he will be in Chicago May 25 and 26, arriving at 7:30 a.m. on the 25th.

Thompson sent me a memo about process improvement. He is of the opinion that within the next two or three months the current problems with respect to the Hanford separations process (the need for better decontamination, satisfactory extraction step carrying, elimination of impurities in isolation step which interfere with precipitation of plutonium peroxide) will be solved. He therefore suggests that we will be turning our attention to finding means of increasing plutonium yields. He then elaborated on the following six points: (1) development of better holding reductants, (2) development of suitable complexing agents for zirconium and columbium fission product activity, (3) use of holdback carriers in the by-product precipitation steps, (4) use of better holding oxidants, (5) development of better scavengers, (6) elimination of steps in the concentration cycles. He says that the possible methods of improving the process as proposed in Items (1) through (6) may improve the yields of decontaminated product as a result of two fundamental approaches. The first approach involves improving the yield in each step of the process. The second approach involves shortening the process so that there are fewer steps with correspondingly decreased occasion for losses.

At 2:00 p.m. Roy Heath left on a trip to New York and Wilmington.

Helen attended the OCD class and worked at the downtown YWCA.

After work, Al Ghiorso and I played 11 holes of golf at Jackson Park (AG-52, GS-49 for nine holes; AG-65, GS-60 for eleven holes). We played until it got dark. The eleventh green is near 67th St. which has a streetcar line, so we returned home via streetcars on 67th St., Stony Island Avenue, and 55th St.

Twin battles for Rome are on is the report in today's paper. Allies are striking from the Anzio beachhead and the main Italian front.

Thursday, May 25, 1944

A delegation of seven visitors from the Montreal group arrived this morning for three days of meetings and discussions with Met Lab personnel to obtain information needed for construction in Canada of a heterogeneous heavy water pile using U.S. uranium and heavy water. The pile will be of moderate power, somewhere between 1,000 and 10,000 kw. The visitors are Cockcroft, Placzek, Volkoff, Pontecorvo, Newell, Ginns, and Halban.

Zay Jeffries transmitted, for my handling, a recent letter he received from D. F. Hewett of the U.S. Geological Survey. Hewett wishes information on analytical procedures for small quantities of thorium.

An alpha ionization survey by the Health Division showed off-scale readings in hoods in Room 27, a badly contaminated bench top in Room 41, and off-scale readings everywhere in hoods in Room 33.

Cunningham wrote a memo to me giving a general picture of the activities of Sub-section III. He reviewed the current status of work on the basic chemistry of the Bismuth Phosphate Process. He said that Howland, Hindman, and Malm have shown that, in process solutions, the only oxidation states of practical importance are states IV and VI; for this reason Cunningham sees no point in further effort to find conditions to stabilize oxidation state III in the Hanford process. On the status of experiments on the mechanism of carrying, he reports the following: Samples of plutonium(III) phosphate have been prepared which are isomorphous with the alpha form of bismuth phosphate. Smith is now attempting to prepare plutonium(III) phosphate which is isomorphous with the beta form of bismuth phosphate. Crystalline samples of plutonium(IV) phosphate have been prepared by Smith. These samples are definitely not isomorphous with either the alpha or beta form of bismuth phosphate, but are isomorphous with thorium phosphate. In order to gain a better understanding of what occurs when bismuth phosphate is precipitated in the presence of large amounts of Pu(IV) (up to 10% of the bismuth), Smith has prepared crystalline bismuth phosphate and plutonium(IV) phosphate and mixed them mechanically in varying ratios. The samples will be examined by Mrs. Mooney before and after digestions. Hopefully, this will show that Pu(IV) phosphate is picked up in the bismuth phosphate and also the approximate maximum amount that can be so incorporated.

Cunningham suggests that the program on basic chemistry of the process be extended to include: (1) A systematic study of interferences with carrying of Pu(IV) by Fe(III), Cr(III), hydrazine, etc. (2) A systematic study of variables such as rate of stirring, digestion, order of addition of reagents, etc., on carrying of Pu(IV).

Among other developments of interest, Hindman has redetermined the value of the Pu(III) = Pu(IV) + e^- couple in 1 M HNO_3 to be -0.916 ± 0.010 v.

A number of new compounds of plutonium have been prepared by the men in Dawson's group in connection with the routine work of recovery and purification. Arrangements have been made to supply samples of these to Zachariasen for examination. In addition, rough solubility and melting point determinations will be run. Among the compounds are plutonyl nitrate, plutonyl chloride, plutonium(IV) sulfate, and plutonium(III) nitrate (the latter prepared by Hindman's group). Isolation of Np^{237} from the 64 pounds of Berkeley neutron-bombarded metal has proceeded smoothly; preliminary indications are that better than 90% extraction was obtained in the extraction step. The lanthanum fluoride precipitate is now being put into solution in Zr(IV) preparatory to oxidation and ether extraction. Hufford has had numerous requests for the preparation of U^{235} and Pu^{239} films. Three of these foils are for an experiment that Herbert Anderson, Cunningham, and I are planning. In the Instrument Group, the multiple neutron chamber, with four chambers, is nearly ready for routine use.

Katzin sent a memo to Moulton concerning an invention of mine, Patent Case No. S-828, MUC-PA-1218, having to do with electrolysis of fused salts containing plutonium. He referred Moulton to reports CN-328, CN-343, CN-427, and my notebook 139, page 1.

Margolis and Blaedel summarized for Dreher the chemical experiments on ether extraction since the 12th of this month. Experiments in the following three categories have been performed: (1) Removal of hydroxylamine and of hydrazine from aqueous solutions by contact with ether. (2) Extraction of Pu(VI) from a composite ether solution from Run 8. (3) Distribution of $Ca(NO_3)_2$-$UO_2(NO_3)_2$ and HNO_3 in water-$Ca(NO_3)_2$-UN-HNO_3-$K_2Cr_2O_7$)-ether systems.

This is the week that merit raises and promotions are being considered at the Met Lab. In my case, Hogness sent the following memo to Stearns:

"In spite of the general order from you to the effect that there will be no salary increases for those who already have salaries in excess of $5,000, I am nevertheless proposing that Mr. Glenn T. Seaborg's salary be increased from $550 to $575 a month. I am sure that you are aware that Mr. Seaborg is by far the most important man in the Chemistry Division and that the discovery of our product and the working out of the Clinton process is very largely due to his efforts; in fact, without Mr. Seaborg I feel that we would have had no project at all. At the present time he is the section chief directing the work of about 100 men, and these men are practically all working on process chemistry for both Y and W.

"I have found Mr. Seaborg an excellent organizer, and he is running his section in an exemplary and eminent manner. If anyone in our section deserves a merit increase, I feel that it is Mr. Seaborg."

At 10:30 a.m. in Room 261, Ryerson Laboratory, the weekly conference on solvent extraction was held and attended by Bird, Buffam, Dreher, D. K. Duffey, A. C. Hyde, Kircher, K. C. Lampert, Maloney, E. H. Shade, and Tepe. Semiworks reported that a 14-hour run of the continuous ether extraction system was completed during the week. Preliminary analytical results indicate that steady state operation occurred for a 6-hour period during the middle of the run. In this period, about 85% plutonium recovery and 20-fold decontamination was realized. Indications are that hydroxylamine is not an entirely satisfactory reducing agent for use in the aqueous extraction column. A new series of runs will be made using another reducing agent to be recommended by Dreher's group.

This afternoon I received a phone call from Underhill in Berkeley bringing me up to date on the patent situation as he found it when he returned to Berkeley this week. The matter of getting a security clearance for Owen, the University's patent attorney, is proceeding satisfactorily. Another exchange of letters between Sproul and Bush, however, is necessary; so it now appears that the best time for our next meeting will be between July 16-19. Apparently the procedure will be as follows. After Owen is cleared to look into the patent applications filed with Baird (the Rad Lab's patent attorney), he will give his opinion to Conard, the attorney for the Regents, and to Underhill. Conard and Underhill will then come to Chicago and meet with Kennedy and me before we all go to Washington. This all assumes we will hear something from Lavender in the meantime and that there exists by then a reason to see him again.

Harrison Brown, visiting from Site X, had dinner with Helen and me in our apartment. Then he accompanied me to the regular meeting of the Purification and Metal Production Sub-section of Section C-I. Others included at the meeting held at 7:45 p.m. in Room 209, Eckhart Hall, were Baumbach, Brody, Cunningham, Davidson, Fried, E. K. Hyde, Jaffey, Katzin, Kohman, Manning, Orlemann, Smith, Jake Warner (purification coordinator), Willard, Seifert, and George A. Cowan (the latter as a visitor). Orlemann and I talked to the group about safety precautions and said that some discussion on this subject will be held at each meeting. I then turned the meeting over to Orlemann. Cowan reviewed recent work on the cupferron method for removing the plutonium from light impurities — recoveries seem satisfactory. Seifert described his work on oxygen analysis by vacuum fusion, a method that uses a hot carbon crucible so that the oxygen in the sample is converted to carbon dioxide and analyzed gasometrically. He believes the apparatus should be in operation in a few weeks.

Hyde announced that the first authentic sample of plutonium bromide was prepared by the reaction of bromine on plutonium metal by Hagemann. It showed an expanded lattice that differs from the x-ray picture obtained from material made by the reaction of HBr on PuO_2. The compound PuOBr was described. It is insoluble in water and soluble in dilute acid. It has a tetragonal structure that is isomorphous with LaOBr, BiOBr, and PuOI.

Some hydrates of plutonium halides were described. Kohman gave an account of the neutron counter now being used. It consists of four boron fluoride chambers imbedded in paraffin. Approximately five percent of neutrons can be counted, but in a 6-mg [plutonium] pellet no neutrons were detected. Larger samples will be utilized in the near future.

During the day Helen attended a housing luncheon at the downtown YWCA and then brought the galley proof from the *Reviews of Modern Physics* for the "Table of Isotopes," which arrived at home in today's mail, to the Met Lab for me to check.

Italy maintains center stage on the front page today. U.S. troops are reported to be only 25 miles from Rome.

Friday, May 26, 1944

I submitted rating sheets recommending that Howland and Jensen each be promoted from Junior Chemist to Associate Chemist. I noted that Howland during the last six weeks has been employed in a semi-administrative capacity in connection with mainline process research and that Jensen is now acting as a group leader.

Brody, Davidson, Fried, Hellman, Manning, Meyer, Orlemann, and Simpson met with me to discuss purification and metal production problems. The following decisions were reached: (1) *Metal production in vacuo:* Equipment is set up and ready to operate based on the use of BeO as a refractory material. Reductions of PuF_3 with barium should proceed immediately in BeO, and CaO and La_2O_3 replacement crucibles should be fabricated as rapidly as possible. (2) *Bomb reductions:* Satisfactory reductions of UF_4 with lithium have been obtained. The 500 mg of plutonium available for metal production will be supplied as PuF_3 by Wednesday. At that time reductions will be carried out on the 30 to 50 mg scale. Bomb reductions of $PuCl_3$ and $PuBr_3$ must await construction of a satisfactory dry box by the Ryerson shop. (3) *Vapor pressure of the metal:* Over the weekend or by Monday, it is expected that the initial run on the 6-mg sample of plutonium metal available will be carried out. It is expected that this initial study may not be satisfactory and additional measurements will be made on metal provided by the metal production group first. Later, the metal will be provided by Simpson's group who will carry out a barium reduction of fluoride within the apparatus to prepare their own plutonium metal, if satisfactory results are not obtained on previously prepared metal. (4) *Fluorides:* The preparation of PuF_3 by electrolytic reductions of Pu(IV) solutions containing HF is to be studied with high priority. (5) *Bromides:* Twenty mg of $PuBr_3$ of purity greater than 93% has been prepared, and its reduction to plutonium metal will be studied by Westrum as soon as possible.

At 4:00 p.m. Albaugh returned from a four-day visit to Clinton Laboratories where he discussed current problems with Balthis, Davies, English, Miller, Olsen, Perlman, Peterson, Sullivan, and others. He

summarized his discussions in a 13-page memo. A primary concern has been to obtain information on decontamination improvement studies underway, particularly the use of cerium-zirconium scavengers.

Helen attended the OCD class. In the evening she and I played cards with the Ghiorsos.

News today states that the final battle for Rome has opened.

Saturday, May 27, 1944

James Barrick is transferring to Clinton next Wednesday on an indefinite loan basis.

I wrote to Kennedy describing Underhill's phone call to me day before yesterday and the tentative plans for all of us to meet on or before July 19 and then go on to Washington together.

I sent a letter to Shane in Berkeley concerning Patent Case No. S-310, "93^{237} as a Source of Power," for which I am the inventor. Lavender has asked whether or not I am willing to assign the entire rights to the Government. I asked Shane if the University thinks it would be all right for me to do so. I mentioned that since 93^{237} doesn't undergo fission with slow neutrons I doubt that it will ever be in the same class as 49, 25, and 23 as a source of power. It is my inclination to go ahead and assign the case to the Government.

I responded to the May 15 letter from D. F. Hewett to Jeffries on determining small quantities of thorium. I said that we have been faced with this problem. I referred him to some procedures described in the literature and mentioned that it is very likely he is already familiar with them.

I sent a letter to E. C. Lingafelter, in response to his of May 16, suggesting that it probably could be arranged for him to come to work for us in the summer although we probably could pay transportation only one way.

Cunningham wrote A. C. Hyde and E. A. Foskett expressing appreciation for their excellent cooperation in the extraction operation in the neptunium recovery from Berkeley-bombarded metal; the operation has given a Np^{237} yield of at least 85%.

I met with A. C. Hyde, Kircher, Thompson, Watt, Willard, and Dreher to decide what to do with the plutonium contained in the Chicago semiworks solutions. We agreed that the amount of plutonium contained in most of the solutions would not justify the effort of recovery. It was decided, however, that it would be advisable to store the plutonium in some partially decontaminated and concentrated form. The concentrate containing the plutonium will be prepared by semiworks personnel, probably

in the form of a LaF_3-BiF_3 precipitate, and delivered to Section C-I for storage. Also raised at this meeting was the concern expressed by Whitaker through Cooper and Kircher about the number of special batches of slugs for Chicago now in storage in the cooling canal at Clinton Labs. My opinion was that no change should be made in the current metal pushing schedule; the backlog is not considered excessive in view of the more rapid use we anticipate in the near future.

Squires and I discussed the sequence of operations and time schedule for the extraction and first decontamination cycles in the Hanford plant. The extraction step will require 15.5 hours, the decontamination by-product precipitation, 15.5 hours without scavengers and 24.0 hours with scavengers, and the plutonium precipitation in the decontamination cycle 18.0 hours. Capacity of the plant is 3 tons of metal dissolved per day.

Roy Heath returned to Chicago at 4:30 p.m. from his trip to New York and Wilmington, Delaware.

Helen and I went to see a double feature at the Frolic Theatre this evening.

The Allies are reported to be only 16 miles from Rome.

Sunday, May 28, 1944

I played 18 holes of golf at Hickory Hills Golf Club outside Chicago (8201 West 95th Street, Palos Park) with Al Ghiorso and Zene Jasaitis. Scores were: AG-133, ZJ-200, GTS-112. Zene lives with his mother in Lockport and is eligible for a gasoline allotment to drive to work. Today we rode with him in his mother's car to the golf course and back.

At 6:00 p.m. John Willard and I left on the C&NW R.R. train "City of Portland" for Pendleton, Oregon. From there we are to travel by automobile to Richland, Washington, where we shall visit the Hanford Engineer Works for the first time. We had a steak dinner in the diner; somehow the operators of the streamliners are able to locate the meat, which adds a special treat to these train trips.

Monday, May 29, 1944

Willard and I are enroute to Pendleton, Oregon. Tonight we had a wonderful roast beef dinner in the diner. This seems to be a tradition for the second night out on the streamliners.

Tuesday, May 30, 1944

At 5:30 a.m. John Willard and I arrived in Pendleton, Oregon. We were met by representatives of the Hanford plant and rode with them by automobile to the plant. After we arrived, we spent the remainder of the day visiting at the plant headquarters where Walter O. Simon, the plant manager, and others described the progress in building the production piles and chemical extraction plants. We checked into the transient quarters in Richland (Fig. 1), the town where the supervisory and professional personnel connected with the Hanford plant are living in specially constructed and quite comfortable homes.

Wednesday, May 31, 1944

Willard and I visited the sites (100 areas) where the plutonium production piles are under construction and the sites (200 areas) where the plutonium chemical extraction plants are under construction (Figs. 2 and 3). It is an awe-inspiring experience to see the thousands of workmen busily engaged in the building of these complicated edifices. These are located in a vast expanse of area (almost 500,000 acres in all) with the piles near the Columbia River for cooling purposes and the extraction plants somewhat removed. To reach these areas we drove over some of the flattest, most lonesome territory I have ever seen.

For lunch we were taken into the Hanford Camp where the construction workers live in rows of barracks, tents, and trailers stretched out in all directions. We ate in the largest mess hall (Fig. 4) I have ever been in — a sea of faces all being well-fed in shifts. We were told that there are some 40,000 residing at Hanford.

After lunch, Simon, who is serving as Hanford's Mayor, took us on a tour of the Hanford Camp. This included a visit to the jail, which was full of the motliest aggregation of tough-looking characters that I have ever seen. We learned that in order to find the large quantity of labor needed, it is necessary to hire "off the street," so to speak, and they have a large turnover with a substantial element of people of dubious character. We learned there was a murder last night and that this is a rather common occurrence.

After continuing our visits at the construction sites, which ran until quite late in the afternoon, we started back to Richland with Simon in his automobile. Being impatient with the secondary roads in the area (actually, there are no other kind), he decided to take a shortcut straight across the desert. In a particularly isolated area the wheels of Walter's car got trapped in loose sand. Fortunately, we discovered a water hole nearby, and Willard and I carried water over to pack down the sand to create a little better traction. However, Walter spun the wheels of his car before we had finished our compacting project, and he impatiently dug the car in deeper and deeper. With a great deal of persuasion and some restraint, John and I finally succeeded in slowing Walter down, and we managed to get free and make our way back to town.

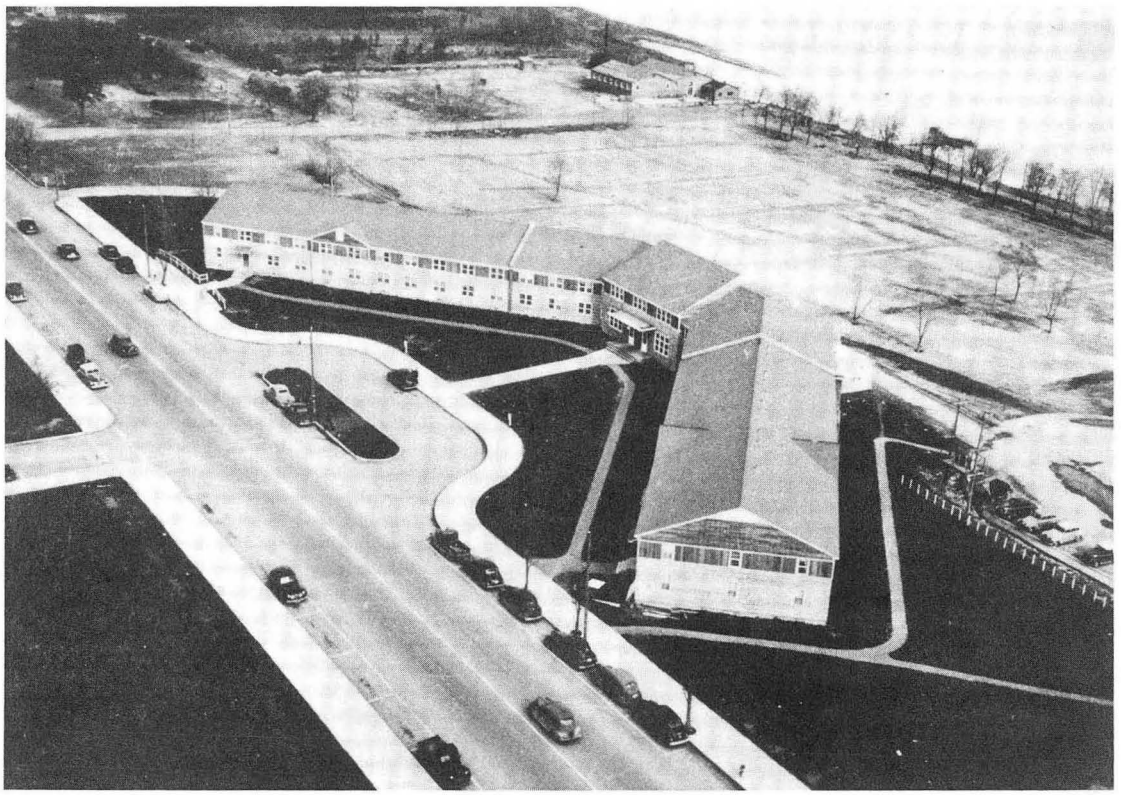

Figure 1. Transient Quarters, Richland. Summer 1944 XBB 7810-13182

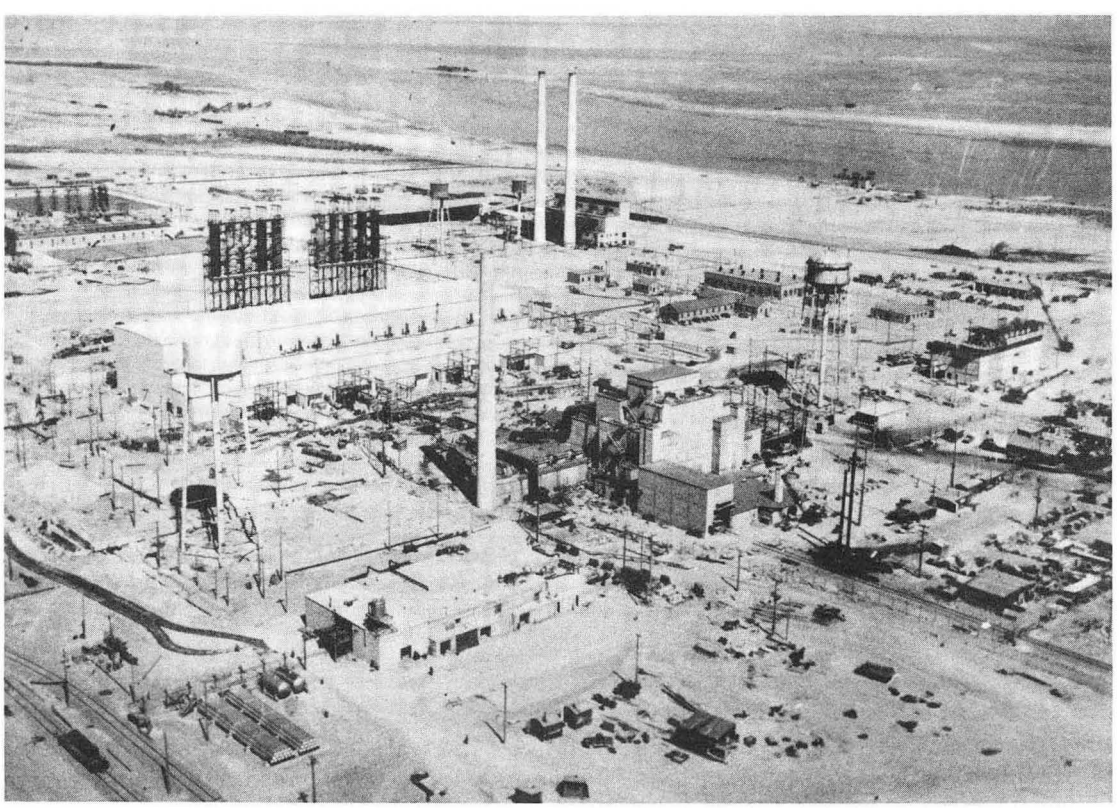

Figure 2. Aerial view of Pile (100B) area. Summer 1944.
XBB 7810-13183

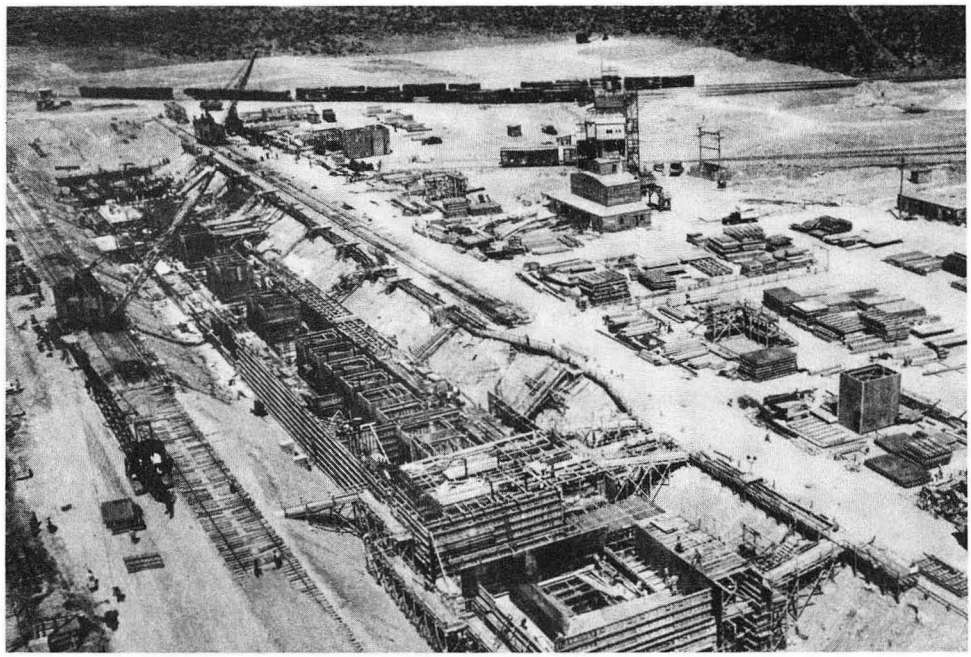

Figure 3. Construction of Extraction and Decontamination Building (221B). Summer 1944.

XBB 7810-13181

Figure 4. Hanford mess hall. Summer 1944.

XBB 793-3587

We arrived back in Richland at about 8:00 p.m. and then went to the nearby town of Pasco to have our dinner.

May ends with headlines reporting Allied gains on the Rome line and with reports of the first tank battle of the War in the Pacific on Biak Island.

JUNE 1944

Thursday, June 1, 1944

After visiting the Hanford Project headquarters in the morning, Willard and I were driven by automobile this afternoon to Pendleton, Oregon. Here we had dinner in a special restaurant that has somehow been able to find the meat to serve steaks. We then boarded the "City of Portland" at about 9:00 p.m. to start our trip back to Chicago.

Friday, June 2, 1944

Enroute back to Chicago on the "City of Portland" with John Willard.

Saturday, June, 3, 1944

We arrived back in Chicago at 12:30 p.m.

In a meeting with Baumbach, Orlemann, and others, we decided to devote the remaining PuF_3 to vibrator bomb reductions and to discontinue the vapor phase reductions "in vitro-vacuo" for the present.

Today's paper indicates that there is a battle raging in the Alban hills near Rome and that U.S. troops have captured two towns.

Ralph James discussed with me his calculations of the change in specific activity from neutron irradiation of Pu^{239} as a function of various assumed half-lives for Pu^{240}. We decided to try to get a factor of 10 more neutrons in a sample of Pu^{239} in order to have a better chance of detecting a change in specific activity. I also discussed with Ralph, Joe Hamilton's suggestion to bombard U^{233} with deuterons to obtain 93^{235} and other isotopes. Since we already have a deuteron-bombarded U^{235}-U^{238} target in which 93^{235} and 93^{236} should be formed by d,n and d,2n reactions on U^{235}, it was agreed that Ralph will isolate the 93 fraction of this target using the bromate technique and look for alpha particles from these isotopes. The target to be used received 2,350 microampere hours at Berkeley during March 3-4, 1944.

Eleanor Lewis began working as a technician for Ghiorso.

I received a letter dated May 31 from Kennedy saying he has heard nothing from Lavender or other parties concerning our patent negotiations. He said that he is sure Segrè and Wahl have no objections to any points I raised in my letters of May 22 and 27.

An updated outline (MUC-GTS-731) of the research program of the Fluoride Chemistry Group (Group 5A) was sent today to Orlemann by Heath. The outline is as follows:

PuF_4. Whereas Site Y has assumed temporarily at least that the high-temperature method of preparing PuF_4 is to be the final method, the following program will be carried out in order to provide the necessary information for larger-scale operation.

(1) Determine the optimum conditions for the production of PuO_2 from $Pu(NO_3)_4$.

(2) Determine the optimum conditions for converting $Pu(NO_3)_4$ directly to PuF_4 with HF.

(3) Determine the optimum conditions for converting PuO_2 to PuF_4 with HF. Draw HF from the liquid phase.

(4) Study purity of PuF_4 prepared by above methods.

(5) Make stability studies on PuF_4.
- (a) Determine at what temperature H_2 reduces PuF_4 to PuF_3.
- (b) Determine at what temperature moist air hydrolyzes or oxidizes PuF_4.
- (c) Determine at what temperature $PuF_4 \rightarrow PuF_3 + F_2$
- (d) Determine the rate at which PuF_4 picks up H_2O when standing in air.
- (e) Determine at what temperature PuF_4 reacts with steam.

(6) Study HF absorption by PuF_4.

(7) Determine the rate of hydrolysis of PuF_4 in aqueous solution at varying temperatures (25°, 50°, 75°, 90°) by titrating free acidity.

(8) Study to some extent the other methods of preparing PuF_4.
- (a) $Pu(OH)_4 + HF\ (aqueous) \rightarrow PuF_4 + H_2O$
- (b) $PuO_4 + 4HF \rightarrow PuF_4 + 2H_2O + O_2$
- (c) $PuO_2 \cdot H_2O \overset{\Delta}{\rightarrow} PuO_2 + H_2O$
- (d) $PuO_2 + 4HF \rightarrow PuF_4 + 2H_2O$

PuF_3. Since experience here at Chicago shows that PuF_3 can be satisfactorily reduced, the following program will be carried out in order to insure that all the necessary control data are available.

(1) Electrolytic reduction of $Pu(NO_3)_4$ solution in the presence of HF is to be investigated as a possible means of preparing PuF_3. In case this system works the reduction would be effected in the same room used for final purification. Samples will be submitted for analysis.

(2) Reduction of $Pu(NO_3)_4$ solutions with SO_2 and the precipitation of PuF_3 by HF will be studied. The variables of the process, temperature of the solution, concentration of ions, time for crystal growth, etc. will be determined. Optimum conditions for the preparation of the PuF_3 will be determined. Samples will be submitted for analysis.

(3) Reduction with hydrogen and the subsequent formation of the

trifluoride will be investigated as above.

(4) The solubility of PuF_3 is to be determined during the course of the above work.

(5) The degree of hydrolysis of PuF_3 is to be determined at 25°, 50°, 75°, and 90°C in water by titrating the acidity.

(6) The temperature at which moist air oxidizes or hydrolyzes PuF_3 will be determined.

HF. The purest possible HF is to be obtained by supplying special HF cylinders to an HF producer. These cylinders will be filled directly from the still with no other handling. Aqueous HF will be made from this anhydrous HF.

Higher Fluoride. Continued attempts will be made to handle this material in glass. It is considered advisable to use 25 mg of plutonium per run. The $HF + Cl_2 + PuF_4$ reaction will be tested to see if this yields a volatile fluoride.

Distribution of Work.

Florin — Higher fluoride, except for $HF + Cl_2$ method. Help on stability studies for PuF_3 and PuF_4.
Heath — Higher fluoride, $HF + Cl_2$ method. Obtain HF pure; help on all other problems.
Meyer — $PuF_4 + PuF_3$
Zvolner — Analyze $PuF_4 + PuF_3$, run aqueous hydrolysis studies on $PuF_4 + PuF_3$. Do general wet preparations for metal production group.

Helen worked at the Met Lab in the morning. We went dancing in the evening.

* * * * *

The following is a day-by-day account of activities that took place at the Met Lab in Chicago while I was on my tour to see the Hanford Engineer Works.

Monday, May 29

My office received a copy of a May 27 letter from Captain Lavender to Kennedy in Los Alamos. Lavender refers to "the decision made by your group not to accept a nominal sum for the assignment of certain inventions to the Government." He went on to say that it is the usual procedure in offering the sale of an invention for the owners to define their invention in a definite form, e.g., a patent application, so that the purchaser may be apprised of all their details. Lavender stated he would therefore await the receipt of Kennedy's definition of these inventions with substantiating evidence. He also indicated that, upon request, arrangements would be made

to clear a patent attorney to assist Kennedy. Copies of this letter were also sent to Segrè, Wahl, and Shane.

Ruth Casler, one of our technicians, is transferring to Section C-IV. Reid Harding, our bench hand, has transferred to the Ryerson-Armory shop group but will remain assigned to New Chem.

Dreher recommended to A. C. Hyde that hydrazine (N_2H_2) be substituted for hydroxylamine (NH_2OH) in future ether extraction runs.

Vernon asked Chapman to make arrangements for shipment of 16 mg of plutonium that I am sending to Clinton for irradiation. (These are the two samples of PuO_2 prepared by Ralph James on May 24.)

Helen attended OCD class, had lunch with Betty Bloechel, and then had the Ghiorsos over for dinner.

Tuesday, May 30

Ralph James prepared a range sample (sample PuAc-N) from the sodium plutonyl acetate irradiated with neutrons at Clinton. Crawford will measure the alpha-particle range in the differential chamber.

Helen worked on the classified "Table of Isotopes" at the Met Lab and went to Marilyn Howe's for dinner.

Wednesday, May 31

At 8:00 a.m. there was a meeting of the Council of my section in my office, attended by Albaugh, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Heath, Hindman, Jensen, Katzin, Manning, Orlemann, Pye, Simpson, S. Thompson, and Watt. Manning, as Associate Section Chief, presided over the meeting. He reported that he and Selwood have decided that no one except members of Section C-I may come through the back entrance of the New Chemistry Building. Visitors will enter the air lock near the main entrance where they will be issued overshoes.

Manning also reported that a Colonel Kenneth D. Nichols of Groves' staff will inspect the New Chemistry Building some time later in the week. He informed the group of a teletype received from Warner about two possible modifications of plutonium metal reported at Los Alamos. Simpson noted that no liquid air was available on Memorial Day. It was decided that some adequate arrangement should be made for keeping a supply on hand at all times.

Ralph James finished his review of the data from the deuteron bombardments at the Washington University cyclotron. He concludes that if the yield of 95^{239} from a plutonium target is comparable with that of

94^{238} from a uranium target and if its alpha particles are of longer range than 94^{239}, then the half-life of 95^{239} must be either less than one minute or greater than one hour. (These poor limits are caused by geometry for counting long-range alpha particles.) Otherwise, the new activity would have been detectable in the recent bombardments.

James also wrote a memo giving the results of some calculations to show the specific activity changes we can expect by bombardment of 94^{239} with neutrons to produce 94^{240}. He shows that for a six weeks' bombardment of 94^{239} at Clinton in the center of the pile, the percent change in specific activity caused by production of 94^{240} would be 0.14 percent if the half-life of 94^{240} is 2,000 years and 3 percent if the half-life is 200 years. His calculations also show that if the half-life of 94^{240} is greater than that of 94^{239} we would have to detect fewer than three alphas from 94^{240} in the presence of 10,000 alphas from 94^{239}.

My office received a summary from J. J. Howland for me of the latest work on the basic chemistry of the extraction process, covering the following topics: [1] Prereduction and extraction. [2] Carrying of Pu(III) and Pu(IV) by bismuth phosphate. [3] Direct proof that plutonium is carried in the IV state in the process. [4] X-ray diffraction studies of plutonium(IV) phosphate. [5] Instability of Pu(III) in the Bismuth Phosphate Process. [6] Solubilities of plutonium in Process solutions.

My office also received my copy of a memo from Warner to Hogness, as a follow-up to the teletype, giving information from Cyril Smith of Los Alamos that Smith considers almost certain evidence of the existence of two modifications of metallic plutonium. A sample of plutonium metal annealed at 185°C or at 400°C and cooled has a density of 15.9 to 16.2 gm/cm^3. The same sample immersed in liquid air increased in density to 16.8 gm/cm^3. Compression in a cold forging die to 200,000 pounds per square inch raises the density to 18.4 gm/cm^3, supposedly due to strain-induced transformations. The cold-worked, high-density form was unchanged by annealing at 111°C, but density and hardness both dropped back to lowest values by annealing at 185°C. All of the measurements were made on the same sample, namely, 630 mg of metal reduced from fluoride with lithium in a beryllium oxide crucible and remelted in a tantalum crucible.

Smith suggested that Baumbach's group check the density of a low density piece of metal after extremely slow cooling and liquid air treatment. He further reported that another sample of metal was remelted in a reduced cerium sulfide crucible with excellent results. The density of the 20 mg-button was 20.0 gm/cm^3.

Hamilton replied to my memo of May 20 about the possible future use of the cyclotron. He would like to obtain sufficient amounts of isolated U^{233} for bombardments with deuterons and helium ions so that several transuranic isotopes might be prepared, e.g., 93^{233}, 93^{234}, 93^{235}, 94^{235}, and 94^{236}. Also, U^{230} could be prepared by deuteron bombardment of ionium, and U^{231} might be made by deuteron bombardment of Pa^{231}. Hamilton is interested in bombarding 93^{237} if and when sufficient quantities are isolated.

Hamilton expressed his regret that the alpha-particle bombardments on our U^{238} and Pu^{239} samples have not yet been accomplished. He promised, however, this will be done a few days before July 15, the date of a scheduled shutdown. The uranium neutron bombardments are proceeding satisfactorily according to Hamilton and, if all goes well, should receive a total of 150,000 microampere-hours before the shutdown. The cyclotron will be down for about three months.

At six o'clock this evening, Simpson, Sears, and Phipps started their first run on measuring the vapor pressure of plutonium metal. They were using a tantalum furnace which contains 5.5 mg of plutonium metal, operating in a vacuum. The experiment is based on the Knudsen effusion principle. Plutonium vapor at a given temperature effuses through a small aperture and is condensed on a collecting plate. The amount effusing in a given time gives a measure of the vapor pressure for that temperature.

Helen attended the OCD class and had lunch with Betty Bloechel. She worked at the Y in the afternoon. Then she had dinner and went to a movie with Frances Chilson.

Thursday, June 1

Colonel Kenneth D. Nichols, who received his Ph.D. in engineering in 1937, is in Chicago to visit the Met Lab. Today he paid an inspection visit to Section C-I.

John Willard's transfer to the duPont payroll is official today although he is not scheduled to report to Hanford until September 4. George Moore and John Blomeke today transferred to Clinton.

Heath is now promoted to group leader for Group 5A (General Methods and Cooperation in Analysis Development) of Section C-I. Orlemann until now has been serving as acting group leader for this group in addition to his regular duties as Assistant Section Chief.

Phipps ended the first vapor pressure measurement run on plutonium metal using the specially designed apparatus constructed with Simpson's help. The thermocouple used gave somewhat erratic readings causing Phipps to report, "The thermocouple is still a mystery wrapped in an enigma inside of a question mark."

A memo to me from Vernon states that the two plutonium samples (sample SA-1 consisting of 8.2 mg plutonium oxide for a one-month irradiation and sample SA-2 of 4.4 mg plutonium oxide for an indefinitely long irradiation) were shipped to Clinton Laboratories on time.

The weekly conference on solvent extraction was held and attended by Buffum, Dreher, Hyde, Kircher, Maloney, E. H. Shade, and Tepe. Kircher referred to a report (AM-8) by C. Kraus of Brown University concerning the equilibrium distribution of UN between diethyl "cellosolve" and water and

suggested considering such a system in the development of a solvent extraction separation process for plutonium. Dreher reported on the effect of calcium nitrate and UN concentration on the equilibrium distribution of UN between the ether and aqueous phases.

A meeting of the Basic Chemistry, Recovery, and Instruments Groups of my section was held at 7:45 p.m. in Room 209, Eckhart Hall. In attendance were Ames, Asprey, Cunningham, Davidson, Dawson, Dreher, Hagemann, Hindman, Howland, Jaffey, Katzin, Kohman, Krueger, Kraus, Malm, McLane, Manning, Nickson, Pye, Peterson, Studier, Thompson, and others. Manning opened the meeting and discussed the adjustment of the airflow in hoods to assure safer conditions. Nickson, from the Health Division, reported data relating to the toxicity of alpha emitters. He referred to a paper published by Rajewsky in *Strahlen Therapie*, 1941, on occupational diseases of miners working in the Joachimsthal uranium ore mines. In 63 post mortems made on men during the period 1929-38, during which time an average of 400 were employed, the causes of death were identified as follows: 45% carcinoma of the lung; 31% tuberculosis; 8% pneumoconiosis; 3% other cancer; and 13% other diseases. As a comparison, the incidence of lung cancer was only about 1% in the general population, with other forms of cancer being a much more important factor than lung carcinoma. Radon alpha particles are assumed to be mainly responsible for the miners' lung cancers. Nickson cited experiments on twelve mice exposed to a radon concentration of about 1×10^{-9} curies/cc over their lifetime. The animals lived only 75% of their normal lifetime and ten of the twelve developed tumors. A control group developed no tumors. Nickson said he believes that the accumulation of about 5 micrograms of plutonium would cause similar effects. If one assumes a plutonium concentration in air of 1×10^{-15} g/cc and 100% retention in the body, the tolerance dose would be exceeded in three years. He pointed out that the tentative measurements of the plutonium concentrations in air in various New Chemistry Building rooms have been betweeen 10^{-16} and 5×10^{-15} g/cc. He also pointed out that rat experiments with plutonium have shown that a considerable amount of plutonium can be absorbed from a site of injection, such as a deep wound. The tone of Nickson's entire talk was that of the need for increased caution in working with plutonium.

Smith reported that a new Pu(III) phosphate was prepared by heating the purple, gelatinous +3 phosphate in a sealed tube with SO_2. The color changed from purple to a light yellow. Smith believes that the new compound is the one which is isomorphous with the beta form of bismuth phosphate. The existence of this new compound was previously predicted by Mooney. Smith cited the cooperative work between the x-ray group and the basic chemistry group on the mechanism of carrying. A prediction by Zachariasen that up to 25 mole percent of PuF_3 could be taken up by SrF_2 was verified. Kraus said that, at present, there is no good evidence for the hypothesis that Pu(IV) forms a nitrate complex. Malm reported on his work, which shows Pu(IV) is carried much better than Pu(III) by bismuth phosphate. It is further reported that a new, probably plutonium(IV), ion was discovered whose absorption spectrum materially differs from the "normal" plutonium(IV) ion. The abnormal form was prepared several times by heating dilute nitric acid solutions of the normal Pu(IV) (0.1 to 0.2 M HNO_3).

Helen worked at the Met Lab in the morning. She had Charlotte Pearson, Adelaide Willard, and Jeanette Sawyer in for lunch and then attended an orientation class in the evening.

Friday, June 2

Ralph James prepared two samples for Jaffey and Crawford to use on the differential range chamber. Sample No. 1 was 20 μg of plutonium from the sodium plutonyl acetate irradiation by neutrons in the Clinton pile. Sample No. 2 was about 5 μg of plutonium from the California deuteron bombardment.

Dreher talked with Thompson today concerning the programs for personnel in the Process Development Group. He proposed that Blaedel and Margolis should work on ether extraction problems and should investigate the possibility of preferentially removing fission products from an ether solution containing Pu(IV) by countercurrent extraction with an aqueous solution. $Ca(NO_3)_2$ as a salting out agent and its effect on the distribution of fission products and Pu(IV) will be studied. Lincoln and Rasmussen will study the parameters affecting plutonium yields when using prereduction with $NaNO_2$ prior to the extraction step of the Bismuth Phosphate Process at Site W concentrations. Rasmussen will devise liter-scale and 100-ml-scale runs in the hot lab with maximum use of remote control. Hot lab construction is not complete and is awaiting revisions in the construction plans being developed by Christy under Maloney. Hyman will work on uranium recovery. Investigations directed toward devising feasible means of concentrating and storing waste solutions of uranium will be conducted. This has importance because of the recent concern about the disposal of waste solutions containing uranium into the city sewer system.

Larson and Winner are currently working in Albaugh's group determining the effectiveness of cerium and zirconium phosphate scavengers on a 100-ml scale. The work should be completed by about June 17, after which time the two men will make liter-scale studies on various means to improve decontamination by the Bismuth Phosphate Process, using Site W product and fission product element concentrations. Following completion of the semiworks investigation of the use of Pb_3O_4 as an oxidizing agent in the Bismuth Phosphate Process, which should be completed in a week or so, Dreher plans to launch the decontamination program. He plans to hold weekly meetings on the Process Development group at 9:00 a.m. each Friday.

The report, "Metallurgical Laboratory Progress Report for April 1944," (MUC-SKA-665) was issued today. Items of special interest to me that are included in the report of the Nuclear Physics Division are:

"The construction of the P-9 reacting pile is essentially complete. All the main items of equipment are now on hand. The metal rods have been jacketed and welded and are now being given the final test. It will be started as soon as sufficient P-9 is on hand.

"The cross section for absorption of thermal neutrons by 49 has been found to be 950×10^{-24} cm^2 for neutrons of 2200 meters per second velocity. Assuming the ratio between the fission cross sections of 49 and 25 to be 1.4 (as recently measured at Y) and assuming further that the capture branching ratio is equal for the two isotopes, one would find a cross section of about 900. There is, therefore, some indication that α may be slightly larger for 49 than it is for 25, although the difference is not outside of experimental error.

"The number of neutrons emitted by 49 and 25 has been compared. The ranges of the two types of neutrons for slowing down in graphite are equal within the experimental error. The number of neutrons emitted by 49 per neutron absorbed seems to be slightly less than the corresponding number for 25. A direct comparison of the ν values for the two isotopes is in progress.

"A second attempt has been made in order to determine whether the activity of a 25 sample irradiated at Clinton increases due to the formation of an alpha-active isotope. No evidence was found, and the result indicates that if 26 is formed, its lifetime must be greater than 6×10^5 years."

The General Physics Division reports, "A considerable number of experiments have been carried out on the measurement of concentration of product dust in the air of the chemical laboratories. Some success has been had with a method of collecting the dust by drawing a known volume of air through a filter paper with subsequent alpha counting. Some interference is found from active deposits with shorter half-lives. The results of radiation distribution are for the most part less than the tentative tolerance level of 5×10^{-6} α/cc sec. Fifty 'Pluto' meters are expected to be delivered early in May. The first model of the 'Sneezy' meter has been forwarded to Y."

It is also reported that intensive work on electronic warning devices for the protection of personnel working with the product has gone on during the month. A standardized design for a meter capable of detecting product spilled on flat surfaces in the Laboratory has been completed and such instruments are in commercial production. The first model of an instrument warning that dangerous concentrations are in the air has been sent to Site Y for trial.

Accomplishments of my section during the month are described in the report as follows: "X-ray diffraction studies show $PuCl_3 \cdot xH_2O$ exists and that anhydrous $PuCl_3$ can be prepared from it. Oxychloride of plutonium will certainly prove to be isomorphous with NdOCl and slow air dehydration of $PuCl_3 \cdot xH_2O$ should give, at least in part, insoluble PuOCl. A sample of bromine-treated metal gave a pattern with UCl_3 type of structure and is probably $PuBr_3$ with about 10% of the bromine atoms isomorphously replaced by chlorine. Trivalent plutonium phosphate is definitely isomorphous with bismuth phosphate and isomorphous substitution would be an obvious carrying mechanism in any proportion. The monoclinic bismuth phosphate which is insoluble will have a counterpart in plutonium as well as cerium. Samples of LaF_3 and mixtures of LaF_3-PuF_3 show conclusively that the carrying mechanism is isomorphous substitution. No information is yet

available on LaF_3 carrying of Pu(IV) as all samples showed reduction of the plutonium to the III state. Attention is called to the fact that SrF_2 should carry up to 25 mole percent of PuF_3. CaF_2 probably will carry appreciable amounts also.

"Effects of deviations from the Hanford flowsheet conditions have been studied in small-scale tests of individual process steps. Two 100-ml scale process test runs have been completed. Both were carried through two $BiPO_4$ cycles and a crossover cycle.

"Product precipitations at high acidities and at Clinton concentrations have been studied. Using Fe(II) as the reducing agent, the experimenters have found that decontamination is considerably improved by raising the HNO_3 concentration from 1 N to 1.5 or 2 N, but at the expense of 3 to 4% of product loss. Using U(IV) as the reducing agent, they observed no improvement in decontamination. At HNO_3 concentrations of about 2 N, product loss was severe. The work on scavenging from dilute acid solutions with $LaPO_4$ shows decontamination is improved by a factor of about 10 as a result of the $LaPO_4$ by-product precipitation.

"The oxidation of plutonium by Pb_3O_4 has been studied in a series of 2-ml scale tests. If conditions of temperature, Pb_3O_4 concentration, and HNO_3 concentration are favorable, plutonium is oxidized quantitatively. It appears that PbO_2 might be a satisfactory oxidizing agent at high temperatures and in the presence of permanganate ion catalyst.

"Study of the influence of temperature of precipitation (over the range 25-80°C) upon the physical characteristics of PuO_4 has shown that 60°C is the optimum temperature and that higher yields (97-99%) may be obtained at this temperature. It has been demonstrated that the entire peroxide isolation process may be operated using filtration (with suction) of product slurries through a medium-porosity sintered glass filter. It has been demonstrated that PuO_4 precipitations under Hanford flowsheet conditions may be carried out in 25-12 stainless steel.

"If one uses Clinton plant solutions after adjusting the plutonium content to Hanford levels, the fluoride concentration procedure has been shown to provide a 99.3% product yield (up to the isolation step).

"Since it appears that the complete removal of the aluminum-silicon bonded jacket will require prolonged treatments with either HNO_3 or NaOH, the rate of dissolution of the uranium metal in these coating removal solutions was investigated. The calculated weight loss for a clean Hanford slug was less than 0.1% to 0.5% per hour, depending on process conditions.

"The distribution of plutonium(VI) between 46% uranyl nitrate-1 M HNO_3 solution containing 0.02 M $Na_2Cr_2O_7$ and 40% uranyl nitrate-ether solution has been more precisely determined. The extraction of plutonium(VI) from 40% uranyl nitrate-ether solutions with 46% uranyl-nitrate-1 M HNO_3 solutions containing either 0.001 M H_2O_2 or 0.1 M $NH_2OH \cdot HCl$ has been found to be complete.

"The behavior of element 93 has been investigated through the extraction step and one decontamination cycle of the $BiPO_4$ process. The average yield of neptunium in the extraction step was about 80% at all concentrations. In all experiments the loss with the by-product precipi-

tate was less than 2% and the plutonium yields were normal. The variation in the product yield now appears due to partial oxidation of the plutonium.

"A purification cycle applicable to plutonium consists of extraction from 10 M NH_4NO_3, 1 M HNO_3 into various organic solvents and re-extraction of the plutonium into 1 M HNO_3. Numerous solvents show the fraction of elements removed is around 10^{-3}.

"Separation factors were determined for the case where plutonium can be extracted, using organic reagents to complex the plutonium. Separation factors varying from 0.005 to 10.0 were obtained and, in general, separation of plutonium from the light element metals appears to be quite limited.

"Techniques for carrying out precipitation of thorium and plutonium chlorides from organic solvents under completely anhydrous conditions have been developed. Pu(IV) chloride solution was reduced with a slight excess of HI to a Pu(III) solution and evaporated to dryness in a stream of HCl at ~60 mm pressure, and 50°C. The solid residue was heated at 100° and 300°C in HCl to give "pure" anhydrous $PuCl_3$.

"Metal with a density of 20.2 has been shown by spectroscopic analysis to be strikingly pure in those substances detectable by the method. The only impurity above the blank level was beryllium, which was present at only 40 ppm.

"X-ray diffraction photographs have not yielded lines for plutonium hydride. The hydride was made from pure hydrogen and metal reduced in thoria and in beryllia. Plutonium hydride has been formed at room temperature on a 250-microgram sample of metal made by barium reduction in beryllia. The formula calculated from this determination was $PuH_{4.0}$. Plutonium metal made by barium reduction of the tetrafluoride in beryllia, of the same high purity mentioned in connection with the density, melted in vacuo on an electrically heated tantalum ribbon as low as 950°C.

"Basic chemistry continued on the following topics: (1) oxidation potentials of plutonium couples, (2) plutonium complex ions, (3) solubility of plutonium phosphates, (4) organic derivatives of plutonium, (5) rates of oxidation and reduction of Pu(IV) and (VI), (6) isolation of neptunium, (7) gas evolution from plutonium solutions, (8) extraction process steps, (9) radiation spectrum of plutonium, (10) plutonium recovery methods, and (11) analytical methods in organic materials.

"Total expenditures for the month of April are reported as $799,925. Personnel employed at month-end are 1,651 (526 academic), a net increase of 66 during the month."

Helen worked on the classified "Table of Isotopes" at the Met Lab. She had lunch at Jeanette Sawyer's, ginger ale at Adelaide Willard's in the afternoon, and ate dinner at the Ghiorsos'.

Sunday, June 4, 1944

Helen, I, Stan Thompson, Zene Jasaitis, Herman Robinson, and his brother-in-law Bob Freeman, played golf at Big Run Golf Club. Bob had a score of 105, Stan had 109, and I had 105. A snapshot was taken of the group (see Fig. 5).

After our golf game we went to the nearby home of Zene's mother, Mrs. Insoda, in Lockport, Illinois, where we had supper and spent the evening. Helen and I recorded a description of our June 6, 1942 wedding onto a phonograph record.

Sometime during last night a leak developed in a five-gallon glass bottle (used by Britain, Fineman, Haeckl, and Leventhal) in Room 34. It is thought that the leak was caused by the etching action of dilute hydrofluoric acid over the last two months. The bottle lost about five to six liters of waste materials containing about 5 to 10 micrograms of plutonium per liter. A guard reported the leakage about 8:30 a.m. this morning. Two men from Room 34 mopped up the solution and scrubbed the floors. After cleanup, measurements made on the floor surfaces, using a "Pluto" portable ionization chamber survey meter, showed normal readings. Nevertheless, the floor surfaces will be thoroughly scrubbed again tomorrow morning.

Monday, June 5, 1944

Two plutonium-239 oxide samples, prepared by Ralph James, are to be placed in the Clinton graphite pile today. Sample SA-1, which contains 8.2 mg PuO_2, will be irradiated for one month. Sample SA-2, containing 4.4 mg PuO_2 will remain in the pile for an indefinitely long period of time.

Steacie, Goldschmidt, Maddock, and Greenwood from the Montreal Group arrived at the Met Lab for a visit and to participate in technical cooperation discussions on P-9 problems. I joined Steacie, Goldschmidt, and Maddock to discuss the chemistry of U^{233} and its separation from Th^{232}. Meetings are scheduled through Thursday of this week.

Dawson sent a memo to Nickson reporting Saturday's waste-bottle leak in Room 34 and stating the steps which have been taken to guard against such an accident in the future. In the future, the 5-gallon bottles for storage of wastes containing dilute HF acid and small concentrations of plutonium will see shorter periods of service and will be kept standing in a large earthenware jar for safety.

Discussions concerning purification and metal production problems were held in my office. Those who came to confer with me were Baumbach, Davidson, Dixon, Heath, Manning, Orlemann, and Simpson. The following investigations were discussed and decisions reached:

Metal production in vacuo: The vapor phase reduction equipment has been constructed and two 20-mg scale barium reductions of PuF_3 have

Figure 5. Helen Seaborg, Glenn Seaborg, Bob Freeman, Zene Jasaitis, and Stan Thompson at Big Run Golf Club, Lockport, Ill. June 4, 1944.

XBB 771-628

been carried out. The resulting plutonium, however, is not in a form directly applicable for further studies. Temporarily at least, it does not seem feasible to push this technique as a means of obtaining larger quantities of useful plutonium metal. Instead, it was decided that the equipment will be used for testing $PuBr_3$, PuOBr, and PuOCl for reduction to metal using the vacuum technique.

Bomb reductions: Promising results on bomb reduction to metal of uranium, lanthanum, and cerium (as stand-ins) were obtained using both static and vibrated bombs. The vibrational agitation was found to give little if any improvement for reductions on a 30 to 100-mg scale. Three static bomb reductions of plutonium on the 30-mg scale were carried out; the first failed, the second had large slag inclusions, and the third resulted in a metal product that was not entirely satisfactory. A reduction, however, on about 35 mg of unpelleted PuF_3 using the centrifugation technique with a 200% excess of lithium yielded a quite promising single metal agglomerate. This latter technique appears to be the most promising technique for the production of large quantities of usable plutonium metal. It was decided that Robinson and Fried should work with Karle to become acquainted with the technique. It was also decided that reductions on the 30 to 100-mg scale will be carried out using our present stock of PuF_3, which amounts to about 375 mg.

Use of existing metal: It was decided that phase studies with plutonium will be conducted using 30 mg of plutonium metal obtained by centrifugal reduction. Details are to be worked out between Baumbach and Orlemann. Plutonium metal from the third static bomb reduction is to be remelted and density measurements made. The material is to be temporarily reserved for vapor pressure work until larger quantities of satisfactory metal are obtained for this work, at which time the bomb reduction material will be transferred to Davidson's group for the iodide preparation work. A 100 to 200 microgram sample of fluoride- and chloride-prepared metal is to be provided to Watters for conducting fluoride and chloride analysis.

Refractories: Beryllium oxide is probably the most satisfactory of the oxides, but present results show that no oxide is thermodynamically stable in contact with molten plutonium. The laboratory at Site Y has found that CeS is not attacked by molten plutonium and ThS is yet to be tested. CeS refractories will be obtained from Eastman. A decision as to the future refractory program will be reached after the existing data have been summarized and we can see how complete the tests of existing refractories have been.

Fluorides: It was decided that the general fluoride program outlined by Heath in his June 3 memo (MUC-GTS-731) is to be followed, subject to any priority-dictated modifications. Synthesis of the higher fluoride using atomic fluorine will be attempted in glass. The preparation in nickel before transferring to glass equipment will also be attempted.

Bromides, chlorides, and sulfides: A commitment to produce 20 mg of PuOBr by June 13, and 20 mg of PuOCl by June 8 was made. Hydrogen reduction of $PuBr_3$, requested last week, was unsuccessful at 800°C. An attempted synthesis of a sulfide by the action of sulfur on the oxide will be made.

Vapor pressure of the metal: The following preliminary values for plutonium vapor pressure have been obtained but may represent minimum values.

Temperature (K°)	*Pressure (in mm of Hg)*
1,256	3×10^{-6}
1,365	4×10^{-5}
1,450	2.9×10^{-4}
1,495	9.6×10^{-4}
1,571	7.1×10^{-3}
1,624	3.2×10^{-2}

A 60-mg high-purity sample is needed to do a complete study.

Solvent extraction work: A summary of all existing pertinent data on various solvents is to be made and completed by June 14. Emphasis is to be placed on the extraction of Pu(VI) by the most promising solvent.

I wrote a note to Emilio Segrè to ask whether he has any comments on the photostatic copy of the "Table of Isotopes" which Kennedy has. We are now reading the proof, and want to return it as soon as possible.

Ralph James began processing the deuteron-bombarded uranium sample to look for new isotopes of neptunium. The sample target received a 2350 microampere-hour bombardment at Berkeley on March 3 and 4.

Run 12 was made with the continuous ether extraction system using 31 pounds of Clinton metal. Analytical results have not been completed. Run 11 was made last week which gives an overall plutonium transfer of 50%.

A 43-page report was sent as a memorandum from me to Hogness (MUC-GTS-735) describing work of the last month on Separation Processes. A summary of the report is the following:

A. The work covered concerning Albaugh's Extraction Decontamination Group is related to four problem assignment areas:

a. Bismuth Phosphate Research — General Problems. The work by Ader, Bartell, and Hoekstra is reported in "Conditions suitable for the quantitative carrying of Hanford concentrations of plutonium when the product precipitate of the $BiPO_4$ cycle is formed by adding Bi(III) after the H_3PO_4." The data lead to the conclusion that the dominant variable controlling the percent of plutonium carried is the concentration of Fe(III) present, although increased time and temperature of digestion appear to promote the carrying of the plutonium to a lesser degree.

b. Bismuth Phosphate Extraction and Decontamination — "W" Concentrations. Sheft's work on "Overall effect of minor process modifications in the decontamination cycle" is reported. In some recent studies certain minor deviations were made from the "W" flowsheet conditions that appear to increase decontamination factors but do not affect plutonium yield significantly.

Five of these variables have been combined into a test process run, through one bismuth phosphate cycle, with the hope that the apparent improvements in decontamination might be accumulative. Data resulting from this test run (which involve studies of the extraction, by-product precipitation, and plutonium precipitation steps) indicate no pronounced improvements in decontamination factors over the control run.

c. Bismuth Phosphate Research — Decontamination.
(1) Greenlee's work, "Lead as a scavenging agent" is reported. Lead sulfate as a scavenger has been tested in a single bismuth phosphate decontamination cycle and compared with similar barium sulfate scavenger and Site W flowsheet control runs. Only slight improvements in both gamma-decontamination factors have been observed.
(2) Work by Gilbreath and Post, "Use of lanthanum phosphate as a rare-earth scavenger at reduced acidities", is reported. It has been thought advisable to develop an alternate method for specific removal of rare-earth fission products from the "W" process solutions should the present use of by-product lanthanum fluoride precipitations not prove entirely adequate under Site W conditions. A single run using YPO_4, as a scavenger has given unsatisfactory results. Lanthanum phosphate scavenger runs were made with solutions of Site X pile metal dissolved and extracted by the Chicago semiworks. In runs where diammonium phosphate has been used to reduce the acidity before lanthanum phosphate precipitation, decontamination factors are increased 10 to 15 times for beta-particle radiation and 5 to 10 times for gamma radiation with respect to the control runs. Plutonium loss is essentially negligible.

d. Solubility of Bismuth Phosphate under Process Conditions.
Peterson's work is reported on the above problem. The solubilities for bismuth phosphate plutonium precipitate and extraction precipitate under process conditions is greater than for pure bismuth phosphate. It appears from the data that the best value for the solubility of product precipitate at 50°C in 5 N HNO_3 is 29.6 g of bismuth phosphate per liter. On the basis of extrapolation to zero time, 24.8 g of bismuth phosphate per liter seems the best value for this solubility of extraction precipitate in 5 N HNO_3 at 50°C.

B. Work in Watt's Concentration-Isolation Group is reported under two problem assignment areas:

a. Isolation at "W" — Peroxide Method; Concentration at "W" — Fluoride Method.
(1) Work by Pye and Walling, "Isolation experiments on metathesis product solutions," is reported. The use of $KOH-K_2CO_3$ mixtures as reagents in the metathesis operation has been shown to be successful (CN-1585). Tests have been run to determine if the peroxide isolation process, which has been previously shown to be satisfactory for simulated plant $La(NO_3)_3$ product solutions,

will be equally satisfactory using actual Clinton plant solutions. Results show that plutonium yields are only slightly less using actual plant solutions.

(2) Work by Walling and Yett is reported in "Test of the Hanford fluoride concentration process employing Clinton plant solution." Clinton solutions, fortified to simulate Site W conditions, have been processed in four parallel experiments — two in stainless steel vessels and two in glass vessels. It is noted that large losses in the lanthanum fluoride by-product precipitates occur in the stainless steel experiments, which is believed due to interference in the complete oxidation of plutonium caused by long hold-up of the HF solutions in the stainless steel vessels. As the stainless steel surface-volume ratio in these tests is far larger than expected at Site W, the conditions were not considered representative of Site W operations.

(3) Walling's work on "Metathesis of lanthanum fluoride from Clinton" is reported. In the experiment, the plutonium peroxide precipitation step does not appear normal as excessive H_2O_2 decomposition occurs together with the formation of a dark brown color. No data, however, were obtained as the tube containing the phosphate precipitate broke during centrifugation.

(4) Work by Goeckermann, Hopkins, Kelley, Pye, Walling, and Yett is reported in "Metathesis and peroxide isolation experiments on Clinton Room D lanthanum fluoride slurry." Low plutonium yield appears to be correlated with the formation of a dark brown color in the supernatant and H_2O_2 decomposition in the plutonium peroxide precipitation. Attempts were made to determine the inhibiting agent or agents. It seems evident that these inhibitors are removable by the addition of an additional lanthanum fluoride cycle with no zirconium added.

(5) Work by Goeckermann and Hopkins is reported in "Influence of various ions on the plutonium peroxide precipitation." A number of cations may be present in unpredictable concentrations at Site W plant solutions, and 1 ml experiments have been conducted to determine possible interferences with the plutonium peroxide precipitation. Low concentrations of either Fe(III) or zirconium have been shown to seriously affect plutonium yields. If both zirconium and Fe(III) are present, however, the combined effect is much greater than the sum of the individual effects. In an extreme case, 0.5 mg/ml of zirconium with 0.01 M Fe(III) gives essentially 0.0% plutonium yield. It is recommended that steps be taken to keep the zirconium concentration at 0.01 mg/ml or less since iron will always be present to some degree.

(6) "The effect on increased H_2SO_4 concentration on plutonium peroxide precipitation" is briefly reported covering work by Goeckermann. Data obtained from tests at Site W concentrations of lanthanum and plutonium are normal and contribute to the data reported earlier in CN-1585.

(7) Hopkins' work is reported in "Operation of the peroxide isolation process in stainless steel equipment." At Site W Building 231 process cell reactors may be made of stainless steel. It is considered desirable to determine if the isolation process will operate satisfactorily in stainless steel at room temperatures, as compared with previous operation at 60°C. Tests, using simulated Site W solution, have been run which indicate that stainless steel in itself causes little effect but that the precipitation and digestion steps should continue to be done at 60° as room temperature digestion of the precipitate does not coagulate the plutonium peroxide sufficiently to give high filtration efficiency.

b. Concentration at "W" — Early Operation. Work by Kelley and Yett is reported concerning this problem assignment. Metal from the initial Site "W" pile will have about 30 g of plutonium per ton of uranium. This is only about 10% of the expected final concentration when the pile is at its full production level. Because of this, experiments have been conducted to determine how adequate the mainline process will be under early operating conditions. Results, although erratic, indicate that satisfactory performance can be expected. It is believed that additional cycles will produce less erratic results as steady state operation is approached.

C. The work reported for Dreher's Process Development Group is related to three problem assignments:

a. Evaluation of Process Modifications. Work by Lincoln, Larson, Rasmussen, Hyman, and Winner, covering experiments on "Carrying of plutonium by bismuth phosphate in the extraction step" is described. Poor yields in the extraction step have been observed which suggest that poor carrying of plutonium by bismuth phosphate may be due to the presence of some Pu(VI). A considerable amount of plutonium may become oxidized between the final addition of formic acid and the precipitation of bismuth phosphate. The use of sodium nitrite and oxalic acid were used as prereduction agents in tests with uranyl nitrate solutions. Results show that a combination of $NaNO_2$ and oxalic acid quickly reduces Pu(VI) quantitatively and that satisfactory plutonium yields can be obtained.

Larson's work is reported on the "Use of Pb_3O_4 as an oxidizing agent in the $BiPO_4$ process." From the experimental results obtained it appears that Pb_3O_4 can be substituted

for $NaBiO_3$ currently being used as the only satisfactory oxidizing agent for use in the Bismuth Phosphate Process. Only minor modifications in the process, with substantially no change in resulting plutonium yield or decontamination factors, should be required. It is recommended that the Chicago semiworks investigate the use of Pb_3O_4.

b. Extraction and Decontamination — Ether Method. Dreher's group reports that the distribution of HNO_3 in a uranyl nitrate-water-ether system is found to be markedly affected by the uranyl nitrate concentration.

c. Hanford Metal Coatings. Dreher's group reports that experiments show that a ratio of 10 mg of aluminum to 1.0 g of uranium does not significantly affect plutonium yields or decontamination factors. Also a ratio of 4 mg of zinc to 1 g of uranium has no significant effect on the process.

D. Hindman's Basic Chemistry of the Separation Processes is reported on the problem assignment, Basic Chemistry of the Extraction Process.

a. Work by Ader, Sheft, Malm, and Howland is reported in "Explanation of extraction step product losses; oxidation and reduction of product before $BiPO_4$ extraction." The results of this work confirm suspicions that partial oxidation of Pu(IV) to Pu(VI) does occur in the extraction step, which causes a decrease in plutonium yields.

b. Work on "Relative carrying of Pu(III) and Pu(IV) by $BiPO_4$: mechanism of carrying" by Bartell and Malm is reported. Pu(IV) phosphate is found to be carried from 1 N HNO_3 solution in a higher yield than is Pu(III) phosphate under identical conditions. The SO_2 required to maintain plutonium in its +3 state may have interfered with the carrying of Pu(III).

c. Work by Malm and Howland on "Direct proof that Pu(IV) is carried in the $BiPO_4$ process" is described. It has been suggested that since plutonium(III) phosphate is isomorphous with bismuth phosphate, Pu(III) would be carried by bismuth phosphate in preference to Pu(IV). Also, a shift of equilibrium might occur which would reduce Pu(IV) to Pu(III) in the process solution from which bismuth phosphate is precipitated. To test this hypothesis, bismuth phosphate was precipitated from synthetic solutions containing known amounts of Pu(IV) and the precipitate analyzed for the presence of Pu(III). No Pu(III) was found. All plutonium in both the extraction and the plutonium precipitation steps was present as Pu(IV), therefore disproving the hypothesis.

d. Smith's work on the "Mechanism of carrying of Pu(IV) by $BiPO_4$, LaF_3, and $U(C_2O_4)_2 \cdot xH_2O$" indicates that Pu(IV) oxalate is isomorphous with U(IV) oxalate and thorium oxalate. The mechanism of carrying Pu(IV) by U(IV) oxalate is undoubtedly that of a mixed crystal formation.

e. Work by Morgan on the "Instability of Pu(III); persistence of Fe(III) in the product precipitation solution" is reported and gives reasonable assurance that Pu(IV) is the only stable state in the plutonium precipitation of the decontamination cycle under the conditions being presently employed.

f. O'Connor"s work on "Solubilities of product in process solutions" is presented. Studies to investigate the solubilities of various forms of plutonium that will be present in the Site W process are required to prevent plutonium precipitation at any point prior to the isolation step.

Helen worked at Crerar Library in the morning and at the Met Lab in the afternoon.

Today's banner headline reads "Yanks Capture Rome." In the Pacific, American bombers have attacked the Kurile Islands for the fifth consecutive day.

Tuesday, June 6, 1944

Today is D-Day. The radio is full of news of the Allied Forces landing on the coast of Normandy where they secured the beachhead.

Kennedy telephoned me from Los Alamos to discuss the May 27 letter he has received from Captain Lavender, who also sent me a copy. Kennedy and I agree that we have two alternatives: (1) Tell Lavender he is just stalling, and we are dropping the whole matter; or (2) Let the University of California lawyers work up the cases and submit them for sale, having first reached agreement with the University on a settlement. I offered to call Underhill in Berkeley, then call Kennedy back.

I attended the Project Council Meeting on Nuclear Physics which began at 10:30 a.m. in Room 209 of Eckhart Hall. Others present were Allison, Anderson, Babcock, Bacher, S. F. Bernstein, Chipman, Compton, Cooper, E. C. Creutz, L. F. Curtiss, S. M. Dancoff, K. K. Darrow, Dempster, Franck, H. H. Goldsmith, Hilberry, Hogness, D. J. Hughes, Jeffries, W. P. Jesse, Larner, A. S. Langsdorf, H. V. Lichtenberger, Morrison, Mulliken, L. W. Nordheim, L. A. Pardue, Peterson, Seitz, L. Seren, F. R. Shonka, Smyth, Snell, Spedding, Stearns, Stern, J. L. Stephenson, Stone, Szilard, E. H. Wakefield, Warren, Way, Weinberg, Wheeler, Whitaker, Wigner, E. O. Wollan, and Zinn.

Wigner introduced the speakers in Fermi's absence. Nordheim summarized the physics work at Clinton, saying among other things that the new pile loading is now completed and the power output has been increased from 1,600 to 1,700 kilowatts. Bacher and Anderson had a discussion about measuring the pronounced resonance in Pu^{239} (at 0.3 ev) recently discovered at Los Alamos. Anderson said that such a measurement of the resonance could be carried out by comparing the absorption and fission cross sections as was

done for U^{235}, but it is a difficult task inasmuch as the grains in the available samples are coarse. Allison turned to me and asked what value we obtained for the half-life of Pu^{240}, considering the negative results we got with our specific activity measurements. I responded that the half-life must surely be greater than 50 years. Zinn reported on the P-9 pile which went critical at Argonne on May 15. Seren gave a brief summary of the Argonne program of measuring the slow neutron activation cross sections involved in n,γ reactions. Of 140 stable isotopes which give rise to radioactive isotopes by the capture of slow neutrons, 110 have now been measured. The meeting recessed at lunch and resumed at 2:00 p.m. Other speakers at the meeting were Weinberg, Morrison, Way, Zinn, Anderson, Bacher, Friedman, Hughes, and Seren.

After adjournment of the foregoing meeting I attended the 3:30 p.m. Project Council Information Meeting on General Physics which also met in Room 209 of Eckhart Hall. Attendees additional to those at the session on Nuclear Physics were R. J. Moon, V. C. Wilson, Mrs. Mooney, F. L. Friedman, Zachariasen, I. Bloch, and J. A. Simpson. Watson introduced the speakers. Creutz described extrusion work and said that pure beryllium has been extruded for the first time. Of 1,000 Clinton slugs tested, 15 leaky ones were found. Zachariasen reported that no samples of plutonium metal have been received for the past two months. Two plutonium compounds were identified as PuOBr and PuOI (originally assumed to be $PuBr_3$ and PuI_3). Zachariasen has also studied $PuCl_3 \cdot 6H_2O$ and $PuCl_3 \cdot H_2O$. Mrs. Mooney has been able to show that bismuth phosphate is isomorphous with strontium and lead chromates. Snell reported that Be^{10} has a half-life greater than 10^6 years. Other speakers were Dempster, Wollan, and Jesse.

A decontamination program meeting, called by Thompson, was held to discuss the proposed semiworks program of scavenger-decontamination investigations. Attending were Thompson, Willard, Kircher, Dreher, and A. C. Hyde. It was proposed that runs be made following the Hanford flow-sheet with the use of scavengers to establish a "base-line": a series of scavenger runs, using Zr^{+4} and Ce^{+4} precipitations will be tried; scavenging at lower acidity will be attempted by making the by-product bismuth phosphate precipitation from 0.1 N instead of 1.0 N HNO_3; a study will be made to complex the peptized precipitates which now cause centrifuge difficulties.

Helen's friend, Joy Townsley, visited us. We had Bert Goldschmidt who is visiting from Canada, to our apartment for dinner. The evening included so much absorbing conversation with Bert about D-Day that identification of today as Helen's and my second wedding anniversary passed almost unnoticed.

Wednesday, June 7, 1944

The Council of Section C-I met in my office at 8:00 a.m. Attending the meeting in addition to me were Albaugh, Baumbach, Davidson, Dawson, Dreher, Ghiorso, Heath, Hindman, Jensen, Katzin, Manning, Orlemann, Pye, Simpson, Thompson, Watt, and Willard. Procedures to be followed by group leaders during vacations were outlined. I recommended that weekly meetings be held in those groups not presently doing so in order that all our men in Section C-I are kept posted on current problems, reports, etc. Safety problems were discussed. Simpson proposed that we replace all soft glass dewars with pyrex dewars. Heath noted that vacuum desiccators should have protective screens around them when being evacuated as protection against breakage. Dawson stated that Coco-Cola bottles should not be brought into the laboratory. I requested that all group leaders have individuals within their groups post their names and telephone numbers on the doors to their rooms, including the name and phone number for the group leader, so that appropriate individuals can be called in case of an accident. Miss Smith will also keep a list of these names and numbers. It was proposed that a general meeting be held near the date for the beginning of operation of our clean air section of New Chemistry Building — a method in which all entrances must be through the air lock in the alley — to inform personnel in both Section C-I and Section C-IV (Ashcraft's) about the general plan of operation.

Spof English arrived in Chicago today from Clinton Laboratories for a three-day visit. He brought with him copies of the revised Hanford flowsheet. In a memorandum I received from Hogness today, he suggests that a conference be held to discuss the flowsheet prior to our departure for Site X tomorrow.

I received a memo from J. C. Warner referring to the forthcoming June meeting with Thomas on the 19th concerning final purification and metallurgy of plutonium. The usual written reports of progress on the problems assigned to my section will not be required. He asks me, however, to present an oral discussion roughly 45 minutes in duration.

Willard sent W. W. Johnson a strong endorsement for hiring of T. O. Jones, an Associate Professor of Chemistry at Haverford College. Willard plans to meet with Jones next Saturday and have him fill out the necessary application forms.

J. Flox sent a memorandum to Nickson describing a method of wiping the cheeks, forehead, and chin of laboratory workers with a piece of filter paper to obtain a good sample from which an indication of plutonium "exposure" can be determined. He said that after the little incident that occurred to Don Stewart of Dawson's group, it seemed that a face count might be better for determining the alpha-particle exposure to an individual rather than using a nose count. As an example, he said he found about 500 alpha counts per minute on a filter paper that had been used to wipe an individual's face.

Recently I heard that the technicians find that the most efficient method of taking nose swabs is to sit on the lap of the laboratory worker. This is particularly efficient in the case of our SED members.

I wrote a letter to Miss Kittredge in the Department of Chemistry at Berkeley to ask her assistance in locating a missing University of California library book. I am surprised that I am charged with only one book. I also told her that I am ordering 800 reprints with covers of my "Table of Isotopes" and charging the order to the Department of Chemistry. I told her that I had discussed this with Professor Latimer.

Compton wrote Allison that Colonel Nichols has approved the extension of our Met Lab contract and that of Clinton Laboratories from July 1 of this year through June 30 of next. Nichols' letter is quoted by Compton as follows:

"The technical program and priorities outlined in these documents are generally approved subject to the following comments and reservations.

"a. Obtaining and training new personnel should be restricted to those required for work of the first and second priorities, viz., support of Hanford '49' production program and cooperation with Site Y toward use of products '25' and '49'.

"b. No new personnel should be obtained for third priority work. Postwar considerations do not fall within the authorized function of the Manhattan District, and it is a matter to be settled by higher authority at the appropriate time.

"c. You are authorized to retain key personnel for possible trouble shooting at Hanford and Y. When the situation regarding the first and second priority work permits, this personnel may be utilized for fundamental research and design, development of basic materials, and investigation of piles suitable for the development of power and of other new possibilities. However, no major construction or detailed engineering design should be done nor commitments made prior to approval of the specific project. In submitting requests for such projects, reasons supporting the work should be given, for it is the plan of this office to approve no new construction or detailed engineering design without the specific authority of the Policy Committee.

"d. All third priority work to be done involving the use of P-9 is subject to arrangements which are now being worked out with the Montreal group. Such arrangements may indicate a decrease in the P-9 work at Chicago during the coming year.

"In general, it should be kept in mind that funds available to the Manhattan District are available only for work pertaining to winning the present war. All long range work and work which would have no application in the present war should be restricted to the utilization of existing equipment and manpower kept available for standby purposes. It is believed that if this policy is followed the third priority work will be a very small percentage of the total work conducted, and the change in emphasis from development to production will result in a gradual decrease in your activities."

Helen and I had dinner at the 51st Street YMCA with the Baumbachs.

A Project Council Policy meeting was held today. In attendance were Allison, Chapman, Chipman, Compton, Cooper, Dempster, Franck, Hilberry, Hogness, Jeffries, Mulliken, A. V. Peterson, Smyth, Spedding, Stearns, Stone, Szilard, Vernon, Warner, C. J. Watson, W. W. Watson, Whitaker, and Wigner. Compton opened the meeting by speaking in general terms about the Laboratory program for the coming year. In doing so he presented the order of priority for future work as received from Colonel Nichols. Of top priority will be work needed to make Hanford go; of second priority will be cooperation with Los Alamos; forward-looking work has the lowest priority — as present men and facilities become available some effort can be put on longer-range problems. Compton said that a general approval of our program has been made with the following reservations: (a) Obtaining and training of new personnel should be restricted to the first two priorities. (b) We are authorized to retain key personnel as standby for Site W and Site Y problems. They may be used on forward-looking problems if these other problems are not pressing. The Policy Committee will be asked to pass on all proposals for major construction or detailed design effort. (c) Arrangements are being made with the Montreal Laboratory for P-9 pile work which may result in the reduction in P-9 activity here in Chicago. It is believed that as the Project changes from development to production, there will be a slow decrease in activity of the Metallurgical Project.

Franck asked if someone is considering the time, distance, and area over which the effects of the by-products from a nuclear detonation would be felt. He suggested that consideration be given to the effects of meteorological conditions. Compton replied that some investigations have already been made and that these problems are the responsibility of Los Alamos. He then requested Allison to discuss these matters at Site Y and report back to him.

Next, Compton asked Hilberry to summarize his visit to the various Hanford areas and said that construction work is going on well, much better than the labor situation might lead one to expect (intra-union struggles of the electrical workers, however, threatens to retard an early completion of the project). Work is progressing smoothly in the 100 Area with lessons learned in putting up 105B showing up with faster construction of 105D, which may be completed within a month. An algae problem has developed, but it is being kept under control by chlorination. Hilberry reviewed all the major elements of the building program and gave the impression that work on them is progressing satisfactorily. As far as the 200 Area is concerned, construction is coming along rapidly, with completion expected within the required time. During the short discussion that followed, Compton mentioned that, at present, visits to the Hanford site are being limited to persons on specific missions.

Cooper described the slug production program for Site W. The first full charge for a Hanford pile containing 1,500 tubes and requiring a minimum of 32,000 slugs must be ready by about July 15 to 30. A maximum full charge will require 48,000 slugs. The second full charge should be ready about August 15 to 30 and the third charge after September 15. The aluminum-silicon bonded slug production operation is proceeding well and the requirements should be met.

Near the close of the meeting, Stone stated that personnel on the Project are taking the hazards of handling plutonium much too lightly. He pointed out that it is difficult to detect assimilation of plutonium after a few days. He said we must take every precaution to reduce the hazard and should confine work with plutonium to a minimum number of locations.

Today's paper has many articles on the Allied landings yesterday (D-Day) in France stretching one hundred miles along the French coast.

Thursday, June 8, 1944

At my suggestion Ralph James calculated the expected changes in specific activity for plutonium samples subjected to neutron bombardment. The calculations show that in order to obtain a 1% or greater increase in specific activity, the half-life of the Pu^{240} we are searching for must be less than about 1,000 years. This probably explains why James has detected no increase in the specific activity of the plutonium sample even though considerable neutron capture occurred. It appears that to effect changes large enough to detect, we will either have to have longer and more intense bombardments or concentrate the 94^{240} by a Szilard-Chalmers mechanism. James claims that if the half-life of Pu^{240} is greater than that of 94^{239}, we will have to detect three alphas from Pu^{240} for every 10,000 alphas from Pu^{239}. This would be a difficult task for the differential range chamber unless the range for the Pu^{240} alphas is at least 1 cm shorter than the range of 94^{239} alphas.

La Chapelle and Magnusson have so far carried the isolation of microgram quantities of Np^{237} (from the 64.5 pound uranium target bombarded with neutrons in the Berkeley cyclotron for 100,000 microampere-hours and received by the Met Lab early last month) through an ether extraction, a lanthanum fluoride precipitation, and three cold bromate cycles. The percent recovery at this point is 57.5%.

I wrote to Joe Hamilton answering questions contained in his letter received May 31. I agreed to the postponement of the alpha-particle bombardments until a few days before shutdown on July 15. I expressed pleasure that the uranium neutron bombardment is progressing satisfactorily. In addition I made the following comments concerning the search for the transuranium isotopes:

I believe that the search for new isotopes of 93 by the deuteron bombardments of U^{233}, U^{234}, and U^{235} would be interesting and worthwhile. All the isotopes of 93 (93^{233}, 93^{234}, 93^{235}, and 93^{236}) which would be produced in such bombardments should be alpha emitters. If one of these isotopes should happen to have a sufficiently short half-life for spontaneous fission and not too short a half-life for alpha emission, it would be an interesting creature from the standpoint which you mentioned in your letter. Of the three isotopes (U^{233}, U^{234}, and U^{235}) the U^{235} should be the most readily available within a short time. The U^{234} might be obtained as an appreciable percentage of some U^{235} fractions. Both of these bombardments could probably be started a good deal sooner than could the bombardment of U^{233}. Actually we have been looking for alpha-emitting 93 isotopes in one of the bombardments of ordinary uranium with deuterons which you did for us some time back. There have been no positive results as yet. Of course, the alpha-particle bombardment of U^{233}, U^{234}, and U^{235} to form the 94 isotopes (94^{235}, 94^{236}, 94^{237}, and 94^{238}) would be equally interesting.

Were you planning to send some more of your purified U^{232}?

Interest in the production of H^3 from lithium in the Hanford pile is increasing. What are your latest ideas on the best way to do this?

Thompson sent me a report of the work done in Separation Processes during the past month. He suggests I may wish to discuss the information at the Clinton Steering Committee meeting that will be held tomorrow at Oak Ridge. Thompson presents the following list of manpower and problem assignments for his sub-section:

Group 1. Extraction-Decontamination (Albaugh, Group Leader). Increasing production at Hanford. Malm, time of digestion and increased H_3PO_4 with 0.6 mg/cc Bi(III) in extraction step. Ader, same with 2.5 mg/cc Bi(III) and 35% UNH. Greenlee, 1.87 mg/cc Bi(III) in 30% UNH — run cycle at 2 N HNO_3 [Malm, Bartell, and Greenlee].

Checking complete process runs using Ce(IV)-Zr(IV) scavenging. Same method as in previous runs. Will avoid recontamination and use clean equipment for each cycle [Hoekstra, Larson, Winner, and Peterson].

Checking low acidity scavenging using both bismuth phosphate and lanthanum phosphate with "W" concentrations of plutonium and fission products [Gilbreath, Post, and Sheft].

Scouting work on HF and $H_2C_2O_4$ complexing to dissolve zirconium and columbium and improve decontamination. Use HBO_3 to destroy HF at proper points if HF carries through to cause trouble. Trying scavengers from 10 M HNO_3-$BiPO_4$ [Thompson and Morgan].

Learning fission-product analysis [Ader and Sedlet].

Group 2. Concentration-Isolation (Pye, Group Leader). Filtration rates (rates of filtration for various filter-cake volumes and thicknesses at different pressures) [Yett].

Working on LaF_3 slurries from Cell 4 to test isolation flowsheet [Walling].

Checking present concentration cycle with lower $H_2C_2O_4$ (specified by flowsheet) and two-shot addition of lanthanum at 50 mg/l for each shot in plutonium precipitation step. Will use 10 N HNO_3 solutions for bismuth phosphate from plant [Kelley].

Working out cleanup cycle in case it is needed at Hanford. What volumes at Hanford concentrations of plutonium are OK by Room D procedure? [Goeckermann].

Basic chemistry-determine mechanism for plutonium peroxide precipitation and mechanism for effect of zirconium [Hopkins].

Finish up $Bi(OH)_3$ work [Haeckl].

Uranous oxalate procedure (procedure for precipitation of plutonium(IV) oxalate from uranyl solutions obtained in first step) [Beard].

Group 3. Process Development (Dreher, Group Leader). Working out optimum conditions for $NaNO_2$ prereduction [Lincoln].

Designing remote control equipment for hot laboratory and working with Lincoln [Rasmussen].

Temporary work on uranium recovery from extraction wastes at request of technical division [Hyman].

Ether extraction decontamination. In particular, studying extent of possible decontamination and $Ca(NO_3)_2$ system of washing [Margolis and Blaedel].

Scheduled to make one-liter scale decontamination runs. Temporarily loaned to Albaugh's group [Winner and Larson].

Sixteen decontamination runs have been completed, of which eight are control runs according to the present Hanford flowsheet. Eight of the runs have used Ce(IV)-Zr(IV) scavenger in the by-product precipitation step. The decontamination factors were low in all cases, presumably due to iron contamination from the stainless steel stirring rods used in the crossover cycle. Low material balances also occurred which have not been explained. It is believed that losses in the extraction waste can now be corrected by the use of $NaNO_2$ for prereduction.

In connection with isolation work, the plutonium peroxide from the last batch of Room D plutonium from Clinton, which first precipitates, goes back into solution, and upon long standing a white crud separates out. After removal of the crud, plutonium peroxide can be reprecipitated from the solution with the addition of more peroxide, resulting in a high yield. The white crud is suspected of containing substances that are responsible for low yields of the plutonium peroxide. A special analysis of these cruds has been arranged with Ashcraft. The presence of zirconium in the Room D plutonium solutions is expected to be partly the cause of the problem.

Katzin received a letter dated June 6 from Perlman containing information on the patent cases involving electrolysis and crystallization

as a means of isolating plutonium. Perlman mentions the dates when he first applied these methods: His first work on crystallization was in Berkeley on April 1, 1942, and on electroplating in Chicago on June 3, 1942.

Dreher attended the weekly conference on solvent extraction held at 10:30 a.m. in Room 261 of Ryerson Hall. Others in attendance were: W. J. Blaedel, G. M. Brown, Buffum, K. C. Lampert, E. H. Shade, and Tepe. Run No. 11 has been completed in the semiworks and gave a plutonium recovery of 51%, with 31% of the plutonium going out in the waste solution. Mechanical difficulties contribute to the low yield. Excessive evaporation in the oxidizing tank required Run No. 12 to be stopped.

The objectives of the continuous solvent extraction program are to accomplish the following goals before July 15 when the present authorization for this project expires: (1) A successful first cycle must be carried out, obtaining at least 95% plutonium recovery and good decontamination. (2) The treatment of a single batch of dissolved uranium metal must be carried through a sufficient number of repeated cycles to demonstrate that the necessary decontamination can be obtained without excessive plutonium loss. (3) Finally, the system must be operated without major chemical or mechanical failure for a period of time sufficiently long to indicate that a plant capable of sustained operation can be designed.

Hydrazine will be used as a reducing agent in Run No. 13, which will be run at one-half full dissolver activity. Separate laboratory-scale studies will be run to determine the rate of plutonium reduction in ether solution by an aqueous solution of hydrazine to indicate whether or not complete plutonium recovery can be expected in the aqueous extraction column without increasing the depth of packing.

Spof English and I, along with others, met with Hogness at 11 a.m. in his office to discuss changes in the Hanford flowsheet which English brought to Chicago with him from Site X.

A summary meeting was held with Montreal Laboratory representatives following a series of conferences that took place while they have been here. The agenda for these conferences covered (1) separation of U^{233}, (2) irradiation of materials at Site X, (3) protection of chemical operations and design of special laboratories, (4) radiation chemistry, (5) chemistry of film formation in cooling water, (6) liaison on analytical work, (7) corrosion chemistry, (8) chemistry of the homogeneous experiment, (9) metallurgy, and (10) canning.

At the summary meeting I was requested to provide (a) the latest type of alpha counter and FP-54 chamber, (b) two cans of thorium carbonate from the material already irradiated at Site X, and (c) U^{232} tracer material, separated or preferably in the form of a target. I was also asked to take up the question of putting repurified thorium compounds into the Clinton pile for neutron irradiation.

Hogness and I left the Chicago Midway Airport at 5:15 p.m. on an American Airlines airplane and flew to Cincinnati, where we boarded a Southern Railway System train to travel to Site X. We plan to stay in Oak Ridge only tomorrow.

The official bulletin of the Allied Expeditionary Force says, "Allied troops have cleared all beaches of the enemy and have in some cases established links with flanking beachheads."

Friday, June 9, 1944

Hogness and I arrived in Knoxville on the Southern Railway at about 5:00 a.m. After breakfast at the Andrew Johnson Hotel, we were driven by auto to Oak Ridge.

At Clinton Laboratories, Hogness and I attended the Chicago-Clinton Steering Committee Meeting to discuss flowsheet revisions to the Site W mainline process. Others present were Doan, Greager, Johnson, Kay, Kirst, and Perlman. At the meeting a memorandum from J. B. Work to W. C. Kay with suggested revisions was reviewed point by point. With regard to the dissolver charge, it was recommended that 3 tons of slugs represent the limit per charge. Some difference in opinion arose with regard to the H_3PO_4 precipitation step conditions. I recommended no change in H_3PO_4 concentration (0.8 N), while Greagor stated that the semiworks is now standardizing on 0.6 N. Their primary reason is that the lower H_3PO_4 concentration will result in a reduced waste disposal problem. The problem was referred to the Separations Division for resolution after more results are available.

With regard to the lanthanum fluoride by-product precipitation, it was recommended for the present that no change be made. It was further recommended that the HF concentration be retained at 1 N and that the $Na_2Cr_2O_7$ be the same concentration as in the bismuth phosphate cycles. Studies are now in progress using Pb_3O_4 as oxidizing agent. It was recommended that the $NaBiO_3$ be omitted from the flowsheet at the point following bismuth phosphate removal. It should be possible to recommend a metathesis procedure in about a month's time. Perlman recommended a straight K_2CO_3 procedure, but some difficulties in the mechanical operation to implement this procedure may be experienced. For the present, it was recommended that the present flowsheet procedures stand as written.

It was decided that the next meeting of the Committee to consider the Hanford flowsheet will be on August 4, 1944. In the course of the discussion of "Chicago-Clinton process problems," agreement was reached on the following points:

Coating removal: No more laboratory work will be done on this problem at Chicago. The work is now in the hands of the Separations Division at Clinton for development. The Separations Division has obtained 100 slugs from Hanford and will get another 100 in the near future for use in checking the Chicago procedure as well as others.

Extraction: Both $NaNO_2$ and $H_2C_2O_4$ are excellent reducing agents in the extraction step, but the $NaNO_2$ possesses the distinct advantage of being considerably more rapid in its action. Both the Chicago and Clinton groups will continue working on the extraction step of the "W" process, with Chicago placing more emphasis on the basic chemistry of the process than on the process variables. In this connection, it is recommended that more work be done in an attempt to obtain information relative to the mechanism of carrying. In addition, all groups will work with higher concentrations of UNH than the present flowsheet demands.

Decontamination: All groups will continue working on scavengers in the two decontamination cycles. It was recommended that Chicago place two or three men on a study of three consecutive standard bismuth phosphate cycles in an attempt to see whether sufficient decontamination can be obtained by such a procedure. This decision is based upon the fact that the Clinton plant has had several months' experience with the standard bismuth phosphate cycle and that we should make the most of this experience. In addition, this program would represent the development of an alternate decontamination process. It was felt by the group as a whole, however, that the major portion of the available manpower working on decontamination should confine their activities to scavengers, since it is our opinion that means will be found for handling them satisfactorily and that the chances for obtaining the necessary decontamination are better than with straight bismuth phosphate cycles. It was recommended that Boyd and Davies, who are engaged in the new decontamination improvement program, should present the results obtained by the groups of their section at the next meeting of the Chicago-Clinton Conference to be held Friday, July 7, 1944.

Concentration and isolation: The work on concentration will continue at Clinton as in the past. In the case of Hanford isolation work, both Chicago and Clinton will continue using as a basis the tentative flowsheet recommended by me. It was felt, however, that the W isolation program at Chicago has reached the point of nearing completion and that the emphasis in the near future should be increased at Clinton, particularly in the Separations Division.

Following the Chicago-Clinton Steering Committee meeting, Hogness and I boarded the Southern Railway train just before midnight for our trip to Cincinnati, where we will take an American Airlines flight to Chicago.

Saturday, June 10, 1944

Hogness and I arrived in Chicago around 3:00 p.m. from Cincinnati, after being held up for several hours by fog at the Cincinnati Airport. Helen told me that she worked at the Met Lab yesterday.

The meeting of the Separation Process Sub-section of Section C-I was held last Thursday at 7:45 p.m. in Room 209 of Eckhart Hall. In attendance were: Ader, Albaugh, Arnold, Bartell, Beard, Dawson, Dreher,

English, Fineman, Gilbreath, Haeckl, Hoekstra, Hopkins, Howland, Hyman, Katzin, Kelley, Larson, Leventhal, Malm, Manning, Margolis, Morgan, Nickson, S. Peterson, Post, Pye, Rasmussen, Sedlet, Templeton, R. Thompson, S. Thompson, Walling, Watt, Winner, and Yett. Dawson opened the meeting by speaking about general safety precautions. He presented eight safety rules to be followed by our section. Watt informed the group that unannounced inspections of the laboratories are planned by the Safety Group.

Nickson announced that in addition to regular hand, nose, and skin (face) counts, the Health Division is initiating an immediate program to run sputum, urine, and feces analyses. He said that the latest values on plutonium tolerance (based on investigations with inactive zirconium) are 5.0 micrograms for the lungs and 4.2 micrograms for the bones, assuming a safe dosage of 0.1 roentgen per day. In the future everyone working with plutonium will be required to take one gram of calcium per day to reduce the amount of plutonium that might deposit in the bones. He referred to animal studies currently being conducted that indicate that 60 to 80% of the plutonium absorbed goes to the bone. About 8% of an injected dose of Pu(III) is adsorbed; 40% of Pu(IV) and 70% of Pu(VI) are absorbed according to preliminary studies. Nickson reviewed the effects of gamma and x-ray radiation on chromosomes. A dosage of 35 r of x-rays can double the mutation rate of fruit flies; an accumulated dose of 500-700 r increases the mutation rate of mice. There is yet no real evidence, however, that the mutation rate in humans is increased from exposure to gamma radiation, and long term case studies are needed.

English talked about isolation process studies at Clinton. He presented the problem of the apparent interference by zirconium, added to complex the fluoride, with the peroxide isolation cycle precipitation; he described runs at Clinton using K_2CO_3-KOH for metathesis. Results have not been entirely satisfactory. English said three two-liter process runs with Hanford fission products present have now been completed using the double bismuth, cerium-zirconium scavenger procedure in the two decontamination cycles and barium-lanthanum scavengers in the crossover cycle. Overall gamma-ray decontamination factors in the three runs were 2×10^7, 1.5×10^7, and 1×10^8 respectively. In the three 100-ml runs using Hanford concentrations of plutonium and fission elements, the gamma-ray decontamination factors through three cycles were 4×10^4, 5×10^4, and 3.5×10^4 (with scavengers).

Howland summarized for Willard yesterday the experiments by Ader, Sheft, Malm, and Howland on the reduction of Pu(VI) to Pu(IV) in the extraction solutions during digestions and bismuth phosphate precipitation. It has been assumed that poor carrying in the extraction step is due to oxidation of plutonium to the VI state during the heating which follows the formic acid prereduction. At Hanford concentrations this effect is much greater than at lower concentrations. As oxide of nitrogen or other formic acid-nitric acid reaction products may reduce Pu(VI) to Pu(IV) in the extraction solution, several 1-ml scale experiments were run in 20% UNH, 0.1 N HNO_3, 1 N H_2SO_4, using 25-microgram amounts of Pu(VI) stock solution for each experiment. The details of four sets of experiments, arranged in order of decreasing amount of reduction, were presented by Howland.

Also yesterday, Kircher sent to my office a copy of a memo to C. M. Cooper about the shipping containers for Clinton metal. The memorandum implies that the current shipping containers are adequate and that radiation levels at the surface do not exceed the specified 50 mr/hr level. The Met Lab still requires a weekly shipment of 24 slugs for use in the semiworks operations here. Four previous batches arrived at Chicago on March 3, April 20, May 6, and June 3, with a calculated content of 37, 37, 42, and 48 micrograms of plutonium per pound. From data obtained from Pardue's group, the radiation dose rates at the surface of the slug containers, as received from Clinton, measured 13, 16, 16, and 10 mr/hr respectively. Cooper has authorized Maloney to procure six more of the standard four-slug shipping containers.

I read Emilio Segrè's prompt reply to the note I wrote him Monday. He offers a few comments on the section of the "Table of Isotopes" with which he is familiar.

Jerome Karle and Isabella Karle submitted their resignations and will be placed on a one-month terminal leave status.

Roy Heath left at 5:00 p.m. on a trip to Cleveland.

T. O. Jones is in Chicago today. He met with John Willard to discuss possible employment and his attempt to obtain a leave of absence from Haverford College where he is an Associate Professor of Chemistry.

Arthur A. Frost, Assistant Professor of Chemistry, Northwestern University, invited me to present one of a series of twelve lectures being planned by Northwestern University under the general title "New Tools in Chemical Research." The topic he wants me to speak on is "Isotopic Tracer Technique." The tentative date for my lecture is December 6 and the compensation will be $25.

Maloney informed Hogness by memo today that Standard Oil has agreed to the loan of Stephen Lawroski of the Esso Laboratories in Elizabeth, New Jersey, to assist in solvent extraction work in my section.

"Metallurgical Laboratory Report for May 1944," (MUC-SKA-719), was prepared by the Laboratory Director's Office. The report states that the outstanding event in the Physics Division for May was 'the demonstration of the chain reaction in a heterogeneous heavy-metal P-9 system. The critical condition was reached on May 15. Some alterations in the design are being made before operation at power takes place.'

Accomplishments of my section were described as follows: "A study has been made of the variables governing the degree of carrying of plutonium ("W" concentration) by the product precipitate of the $BiPO_4$ cycle when the precipitate is formed by adding all, or nearly all, of the Bi(III) subsequent to the addition of H_3PO_4. Concentration of Fe(III) was found to be the dominant variable involved.

"Preliminary tests have been made on the use of $PbSO_4$ as a scavenger in the decontamination cycle. It was found that 1 to 3 mg Pb/ml are almost quantitatively removed from an oxidized solution by 0.1 N H_2SO_4.

"The use of $LaPO_4$ as a scavenger in the by-product step, at HNO_3 concentrations of 0.1 to 0.3 N, has been tested extensively.

"The Ce-Zr scavenger combination has been tested in a number of simultaneously performed process runs. There is every reason to believe that factors approaching 10^7 can be obtained for beta and gamma decontamination factors.

"Work is continuing on a method for decontamination from rare earths.

"Study of the complete concentration-isolation process (proposed for Hanford) through the use of laboratory solutions has shown overall yields of 97 to 98%. Preliminary experiments employing several solutions and slurries from Cell 3 and Cell 4 at Clinton have indicated that good yields may be obtained in the concentration procedure and that good PuO_4 precipitations may be obtained.

"Assuming that the plutonium content of the first pile metal to be processed will be ca 30 g/T of uranium or less, experiments to determine the feasibility of recycling by-product solutions from peroxide precipitations were carried through five cycles and they indicated that the percentage yield based on the initial amount of product in each cycle increases with successive recyclings.

"Work continued on evaluation of process modifications.

"For purification and metal production purposes, a survey of some twelve organic solvents was made to determine Pu(IV) and Pu(VI) distributions and separation factors for light elements. Nitrobenzene showed a separation of about 10^{-3} for those elements tested.

"200 mg of $PuCl_3$ were prepared by the action of CCl_4 on $PuO_2 \cdot xH_2O$ at 750-800°C. The action of HBr on $PuO_2 \cdot xH_2O$ at 750° yields $PuBr_3$ with a crystal structure *not* isomorphous with that of $LaBr_3$. The preparation contains PuOBr in varying amount and also an unidentified acid-insoluble phase. $PuO_4 \cdot xH_2O$ was reacted with PCl_5 liquid at 280° in a capillary bomb. After distilling off the reagent and volatile reaction products by heating to 400°C in vacuo, the product was well crystallized $PuCl_3$. Although x-ray studies gave no evidence of a second phase, there was some water-insoluble material in the $PuCl_3$ so prepared.

"Additional successful preparation of $PuCl_3$ has been made by drying HI-reduced Pu(IV) solutions in HCl and dehydrating the $PuCl_3 \cdot xH_2O$ by slow heating to 300°C in ca 60 mm of HCl.

"Wet preparations of $PuBr_3$ have been carried out by reducing aqueous Pu(IV) with HBr, evaporating to dryness in a stream of HBr at low pressure, and slowly heating in HBr to 300°C. No adequate evaluation of this material is available.

"It was previously reported that the reaction of Br_2 with plutonium metal yielded a hexagonal $PuBr_3$, isomorphous with $LaBr_3$, $CeBr_3$, and UBr_3 (cf CK-1556). Using bromine known to be free of chlorine for the reaction with metal, a $PuBr_3$ has now been obtained which is not hexagonal but is crystallographically identical with the $PuBr_3$ made by the action of HBr on $PuO_2 \cdot xH_2O$ at 750°C.

"Experiments were carried out to determine at what temperature oxygen replaces fluorine in PuF_4. Using platinum equipment and liquid oxygen traps to remove water from the O_2 gas used, it was found that conversion of PuF_4 to PuO_2 occurs at 325-350° but not at 275-300°. A series of experiments was run in which dry oxygen was passed over approximately 1-mg quantities of PuF_3 in platinum equipment for two hours. No conversion of PuF_3 to PuO occurs at 225-250°, partial conversion (15-20%) occurs at 275-300°, and complete conversion is effected at 325-350°.

"For the reduction of PuF_3 in BeO crucibles, samples were prepared by both the wet and dry methods. From the standpoint of purity the wet method is most desirable because it uses readily purified reagents and low temperatures.

"Vickers hardness values are greatly dependent upon the method of preparing and remelting the metal. For example, plutonium remelted in BeO and in TaN-coated tantalum show respective values of 124 and 192.

"Plutonium metal made by barium reduction of the tetrafluoride in beryllia, with no impurities detectable spectrographically, has melted on tantalum in vacuo at 810° ± 25°C. The plutonium metal, when stripped off the tantalum, contained no tantalum detectable by spectrographic examination (under 0.12%).

"The compression of plutonium halide particles into pellet form has continued to show considerable advantage in reductions on a 5 to 8-mg scale. PuF_4 is formed by treatment of dense PuO_2 with HF. Reductions of PuF_4 with calcium have been made in beryllia by vapor phase reaction, but it is almost impossible to remove the excess calcium from the interior of the crucible. Even prolonged heating at 1200°C in vacuo does not allow the calcium to escape from the beryllia. Sodium will not reduce PuF_4 to metal by vapor phase reduction, as was previously demonstrated. Reductions of $PuCl_3$ have been successful for the sublimed crystalline halide made by the action of CCl_4 on oxide. Unpelleted $PuCl_3$ made in the 'wet' way and dried in HCl shows poor yields of metal when reduced with barium. The crystalline $PuCl_3$ is very hygroscopic and must be handled entirely in a dry box. Pellets (5 to 7 mg) of $PuCl_3$ (CCl_4-prepared) have been successfully reduced to plutonium metal in beryllia crucibles by barium, calcium, sodium, and potassium in vapor phase reductions and by sodium and potassium in liquid phase reductions. The yields in these reductions are rather low.

"Work continued on basic chemistry; viz. oxidation potentials of plutonium couples, inorganic derivatives of plutonium, hydrolysis of Pu(IV) and Pu(III), rates of plutonium oxidation and reduction, isolation and study of Np^{237}, assay of "W" plutonium solutions, heavy isotopes by bombardment of Pu^{239}, basic chemistry of the extraction process, carrying of Pu(III) and Pu(IV) by $BiPO_4$, direct proof that product is carried in the IV state in the process, preparation of plutonium compounds for preparation of x-ray diffraction studies, instability of Pu(III) in the process, solubilities of product in process solutions, research on methods for plutonium recovery, chemistry of plutonium as related to its recovery and purification, and routine separation of 93^{239}."

Total expenditures for the month of May were $811,031. Personnel employed at month-end were 1,765 (533 academic), a net increase of 114 during the month.

Helen worked at the Met Lab in the morning and had lunch at Dagmar's. This evening she and I read the proof on my "Table of Isotopes," which will be published in *The Review of Modern Physics*.

The front page of the newspaper is still filled with items about the Allied invasion in France.

Sunday, June 11, 1944

Stan Thompson, Luther Arnold, and I in one threesome, and Helen and the Baumbachs in another threesome played 18 holes of golf at Evergreen Golf Club. Luther shot a 103, Stan shot 109, and I shot 108. The Baumbachs then came home with us to have supper.

The invasion in France continues to go well as forty villages are captured as Americans advance across the Cotentin peninsula.

Monday, June 12, 1944

Roy Heath is still in Cleveland today.

Clark Egan wrote me accepting an offer of employment with my section. He explained that because of report writing obligations he will not be able to report for duty before July 19. Egan, whom I knew at Berkeley before I came to Chicago, has been working for Professor W. F. Giauque at Berkeley under NDRC contract during the last three years.

Bernard M. Abraham is transferring to our section from Section C-III and will work in Davidson's group.

I completed and submitted a Personnel Rating Sheet for Ralph James and recommended that he receive the maximum merit increase of $15.

I telephoned Underhill, as I had told Kennedy I would do, concerning our patent problems. I informed him about Lavender's letter of May 27 to Kennedy. He told me that Owen is cleared to read the patent information dated prior to the contract with the Government. He will call me tomorrow concerning the applicable dates for any Government contracts that might relate to our problem. He is undecided about a meeting with Conard, an attorney for the U.C. Regents, planned around July 19, until the situation becomes more clear. He will write Bush tomorrow to try to clarify Owen's clearance.

Hindman sent me the program of Group 7, Section C-I, now in progress and which will continue into next week. The following information is contained in the memo:

"Kraus is preparing solutions of Pu(III) and Pu(VI) to complete the magnetic susceptibility measurements; Pu(IV) has already been measured.

In addition, he is devoting part time to the problem of determining whether or not the Tyndall effect observed in many stock plutonium solutions really means that an appreciable amount of colloidal phase is present. The immediate measurement planned is a dialysis of a relatively concentrated plutonium nitrate solution. He has also succeeded in preparing in pure form the 'abnormal' green ion [probably of Pu(IV)] and will furnish samples to Howland for certain carrying experiments. It is planned that the conditions for preparation and relative stability of this ion will be investigated further. An additional check on the valence state by titration will also be made.

"McLane is engaged in the wet preparation of what we believe is an anhydrous $PuCl_4$ through the benzoate and ether-HCl salting-out technique. In addition, he is making runs on the transference of Pu(IV) in various nitric acid solutions with the intention of correlating his results with the observed changes in absorption spectrum in changing nitric acid. In addition, McLane has under construction an extraction apparatus designed for ether extraction of Pu(VI) solutions preparatory to the making of pure Pu(VI) solutions for various solubility measurements. It is expected that this equipment will also be useful in the preparation of Pu(III) solutions, in which iodide is used as the reducing agent and the I_2 formed is extracted with CCl_4 or $CHCl_3$.

"O'Connor is extending his solubility measurements to the iodate and bromate of Pu(IV). He is continuing work on the solubilities of PuF_3, PuF_4, and the potassium salt of Pu(IV). His work will be correlated with that of Smith to settle, if possible, the crystal structure of the double salt of plutonium(IV) and lanthanum which to date has not been crystalline or has showed apparently a mixture of PuF_3 and LaF_3. Smith is finishing his titrations to demonstrate that Pu(IV) is co-precipitated with LaF_3. In addition, he is trying to prepare the sub-oxide and higher oxide of plutonium. Additional preparations under way include the $Pu_3(PO_4)_4 \cdot xH_2O$, $BiPO_4$ mixtures intended to shed further light on the mechanism of carrying of Pu(IV) by bismuth phosphate. Smith is also engaged in the preparation of the Fermi samples. Ames is investigating the effect of plutonium concentration on the rate of oxidation of Pu(IV) to Pu(VI). The levels investigated are to include measurements from 25 mg/l to 2500 mg/l. Since the disproportion reaction has been found to have a rather complicated rate function, the measurements are to be extended to include the rate of oxidation in H_2SO_4 with Ce(IV) and in $HClO_4$ with $KMnO_4$.

"Howland now has under investigation the validity of the $KBrO_3$-LaF_3 method of determining the percentage of plutonium in the (IV) and (VI) state. In addition, he is to check the carrying of the abnormal ion of Pu(IV) with bismuth phosphate and lanthanum fluoride. Finally, the disproportionation measurements made by Hindman in 0.5 M and lower HNO_3 concentrations will be extended by Howland to 20% and 40% UNH solutions. La Chapelle and Magnusson are now completing the isolation of 93^{237} from the Berkeley bombardment. This material is to be combined with the remainder of the material now on hand and final isolation carried out under the direction of Cunningham. In addition, isolation of the 93^{237} from the Clinton LaF_3 sample is to be completed."

Helen worked at the Met Lab on the classified version of my "Table of Isotopes" today.

According to today's paper, fighting is raging along a 51-mile front in France.

Tuesday, June 13, 1944

Heath returned from his visit to Cleveland.

Sam Jackson, a technician in Baumbach's group, resigned today.

At 3:45 p.m. Underhill and Conard telephoned me from Berkeley. They tentatively plan to meet with Kennedy and me in Chicago on July 19. Underhill told me that Contract 201 was let on October 1, 1941, was retroactive to April 1, 1941, and covers fast neutron work. Moreover, Bush wrote Sproul on May 14, 1943, admitting that the University of California had supported the key part of the work. Then on May 27, 1943, Bush wrote Sproul that some of the work in the applications as written was done after the Government contracts. For this latter reason Lavender feels that Owen, when cleared, should not see the existing applications in their entirety.

I then called Kennedy at Los Alamos and relayed the substance of my conversation with Underhill and Conard. I suggested that we write to Lavender, saying politely that he is only stalling and suggesting that for applications he use the ones in his office and for evidence he see A-33 (May 29, 1941) and letters of January 21, 1941, and March 7, 1941, to Abelson and Briggs from me.

The Record of Invention and Disclosures No. S-534 for the low-geometry ionization chamber for counting alpha particles developed at Berkeley was sent today by Captain H. E. Metcalf, patent lawyer with the Corps of Engineers, to Kennedy and Wahl at Site Y for them to sign the Record of Invention.

Manning received a memo from Nickson stating that during the week ending last Saturday, Rooms 1, 2, 4, 10, 30, 31, 34, and 35 in the New Chemistry Building were found to have alpha-particle contamination readings greater than 50 units. In particular, Rooms 34 and 35 are badly contaminated.

L. B. Arnold, Chemistry Division's Assistant Director, sent Stone at Clinton a memorandum today concerning eating in hazardous areas. Arnold noted that some chemists in the Clinton 706A building eat their lunches in offices next to laboratories where plutonium is handled. He pointed out that in Chicago no eating at all is allowed in the Chemistry laboratories and requested Stone's comments on the relative merits of our practices as compared with the Clinton practice.

In a memo to Thompson, Albaugh presented the research program of Group 1 for the period June 5 to about June 20. Included in the summary are the following comments: a. *Cerium-zirconium scavenging experiments*

(4 men): Four men are involved in making four runs at Hanford plutonium and fission product levels. Prereduction will be done by means of the $NaNO_2$ method. There is every reason to believe the gamma-ray decontamination factor will exceed 10^5. b. *Low acidity scavenging method* (2 to 3 men): In addition to other studies, a new scavenging method is being tried which has the advantage of requiring only a single digestion period. In this method the 5 N HNO_3 solution is diluted with a sodium acetate-H_3PO_4 solution in a quantity to give a proper process volume after separating off the by-product precipitate at a pH of 0.75 and final adjustment of the acidity to 1 N HNO_3. c. *Scouting work* (2 to 3 men): A variety of scouting tests are being conducted in a search for alternate or improved methods of decontamination. d. *Increase of plutonium production at Site W:* The problem of finding the most efficient means of increasing the initial rate of metal processing at Site W is being attacked in several ways, including a method proposed by Squires which involves extraction from 30% UNH solution with only 1.9 mg Bi(III)/ml, with by-product and product precipitation in a solution 2 N in HNO_3. Since at 2 N HNO_3 considerable plutonium (5 to 15%) may not be carried in the plutonium precipitate, Albaugh proposes to use $(NH_4)_2HPO_4$ instead of H_3PO_4 since it will simultaneously act as a neutralizing as well as a precipitating agent. e. *Training in fission product analysis work* (2 men): Work on the assessment of various scavengers has been handicapped in the past for lack of men trained in the techniques of fission-product analysis. The two men now being trained will be available for this type of work in about one week. f. *Anticipated program after June 20:* If the cerium-zirconium scavenger experiments in progress are successful and provide gamma-ray decontamination factors of 10^5 through the bismuth phosphate cycles and 10^7 through a crossover cycle, the efforts of Group 1 will be applied to developing decontamination procedures that do not use cerium and zirconium but provide equivalent decontamination. Albaugh points out that if a low-acidity by-product separation could be incorporated into the process it might be possible to eliminate the currently troublesome lanthanum fluoride by-product precipitate in the cross-over cycle.

I held a meeting to discuss purification and plutonium metal production problems with Baumbach, Davidson, Jensen, Manning, Meyer, Orlemann, and Simpson. The following decisions were reached: *Metal production in vacuo* — Tests of the reducibility of $PuBr_3$, PuOBr, and PuOCl can be carried out next week. *Bomb reductions* — 100-mg scale centrifugal reductions have been postponed because of the unsatisfactory character of the plutonium metal produced. Modifications to provide thermocouples for better temperature control are being made and studies on PuF_3 reductions on a 30-50 mg scale will be made during next week. *Use of existing metal* — Difficulties in remelting plutonium metal obtained by centrifugal reduction have been experienced. At the present time we have no metal of sufficient quality and amount to run vapor pressure determinations. Los Alamos has been requested to supply us with 50 mg of good metal so that we may conduct these measurements in the near future. *Phase studies with plutonium* — Strong quantitative evidence for the existence of at least two plutonium metal phases has been obtained, and it was decided that dilatometric studies should be prosecuted with the highest possible priority. The possibility of

studying plutonium metal deposited as a thin foil by vacuum sublimation will be discussed with Zachariasen. Samples of plutonium will also be submitted for chloride and fluoride analyses, as previously agreed. *Refractories* — Of the many materials tested so far, CeS, tantalum, columbium, ZrN, and TaN were found to be satisfactory for use with plutonium metal at 1000°C. Also ZrN, TiN, UN, (ThU)S, and carbon will be tested. *Vapor pressure of the metal* — An additional week's work will be needed to get the apparatus in satisfactory condition.

O_2 analysis — Preliminary tests are being conducted using CuO and SiO_2. If these prove satisfactory, the PuO sample which is in the apparatus will be run. *Bromides, chlorides, and iodides* — The preparation of $PuCl_3$ from plutonium peroxide in bomb tube reactions has been found successful using $PCl_5 + Cl_2$, $S_2Cl_2 + Cl_2$, or $CCl_4 + Cl_2$. $PuO_2 \cdot xH_2O$ has been found to react rapidly with $S_2Cl_2 + Cl_2$ to yield $PuCl_3$. The reaction of $PuBr_3$ with liquid Cl_2 has been carried out to yield an unidentified product. The reaction of metal with iodine in a platinum apparatus has again yielded only PuOI. *Fluorides* — Solubility studies of PuF_4 necessary in the wet preparation of PuF_4 have been conducted. *Solvent extraction work* — Dawson's use of methylisobutyl ketone for recovery of product has indicated good separation from light elements but only partial separation from zirconium, bismuth, and lanthanum.

An information meeting of Sub-section II is scheduled for 2:00 p.m., Saturday, June 17, 1944, prior to the Thomas meeting.

Representatives from both the Chemistry and Technical Divisions attended a meeting in my office to discuss the operation of the Chicago semiworks. Manpower requirements were reviewed in light of the number of du Pont employees who have already been transferred from the semiworks and the possibility that additional employees will be transferred to Hanford in future months. It was agreed that we should attempt to supply the replacements and other manpower needs from outside the project. They would be assigned to either the Chemistry or Technical Divisions.

Helen and I had the Thompsons, Drehers, and Mrs. Lundgren (Dagmar's mother) to dinner.

Today the paper indicates that the Allies are pushing more deeply into France. In the Pacific carriers have raided the Mariannas for a second day.

Wednesday, June 14, 1944

I met with the Council of Section C-I in my office at 8:00 a.m. Attending the meeting were Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Heath, Jensen, Katzin, Manning, Orlemann, Pye, Simpson, Thompson, and Watt. Alpha-particle hazards were discussed, and it was decided that smoking in the halls, offices, and laboratories of the clean air section of the New Chemistry Building should be completely forbidden. Beginning today, outside admittance to the clean air section of the New Chemistry Building will be limited to the alley entrance.

Canvas overshoes will be provided in the other air lock between the two parts of the building for personnel who need to go from one part of the building to the other. The Council decided that Coca-cola bottles are not to be brought into the clean air section of the building. A set of containers for safe temporary storage of both recoverable radioactive residues and waste materials should be provided for each laboratory. A record of the approximate amount of active materials in each container should be maintained.

The provisions of the one year Contract of Employment, which individuals have been asked to sign, were discussed. The Transportation and Moving Agreement form received for the signatures of employees was also discussed. The meeting concluded with the reminder to group leaders that before leaving on vacation, they should make sure that the research schedule for their groups be either in my hands or the respective sub-section chiefs.

Cunningham is leaving by train tonight at 11:30 p.m. to travel to Site X. He will return this weekend.

Watt sent a memo to W. W. Johnson enclosing an application from Quentin Van Winkle. Watt emphasized the desirability of securing Van Winkle's services as early as July 1 for work in Group 2 of Section C-I.

In a summary report of the solvent extraction process written by E. H. Shade in the Technical Division, the analytical results of run 12 were reported in terms of constant flow rates. The percent of plutonium overall transfer obtained in the second, third, and fourth hours of the run were 23%, 16%, and 21% respectively.

After work I played 13 holes of golf with Ghiorso and Katzin at Jackson Park. Our scores were AG — 59, LK — 73, GS — 56 for nine holes; AG — 84, LK — 106, GS — 76 for thirteen holes.

Helen worked at the Met Lab today on the classified "Table of Isotopes." She spent the evening with Wilma.

Today's paper indicates that the Germans have counterattacked American troops at Mountebery and Carentan, but the Allies still retain the initiative.

Thursday, June 15, 1944

Roy G. Post's transfer from Section C-I to Technology Section II is effective today.

Ralph James took time out today to get married. He will return to work next Monday.

Dreher attended the weekly solvent extraction meeting at 10:30 a.m. in Room 261 of Ryerson Hall. Others in attendance were Brown, Buffum,

A. C. Hyde, Kircher, Lampert, Maloney, and Tepe. It was noted that work being done by the semiworks will be curtailed because of a shortage of manpower due to personnel changes. Fundamental work on decontamination is to be continued by Dreher's group and the General Engineering Section, but the work in continuous ether extraction semiworks will be held up for four to six weeks. As yet no attempts have been made to operate the equipment by remote control.

I sent a strong recommendation to Stearns that Orlemann's present salary be increased by $25 per month. I pointed out that as Assistant Section Chief he is directly responsible for the work of the five groups concerned with purification and metal production and that he also maintains liaison between our section and the analytical work on purification in Section IV.

A. H. Compton received a letter from Felix Morley, President of Haverford College, expressing some reluctance in providing T. O. Jones with a leave of absence to work at the Met Lab. The decision, however, will be left up to the head of the Haverford Chemistry Department, W. B. Meldrum, and we should receive an answer soon.

A Notice of Visitors was received stating that W. C. Johnson from Clinton and F. H. Spedding from Ames will be in Chicago, June 19, 20 and 21, during and following the times of the Thomas meeting.

In a memo I sent to Captain H. G. Hawkins, Jr., of the Area Engineers Office, I inquired as to the security implications that might be involved should I present a lecture on "Isotope Tracer Techniques" at Northwestern University next December.

Pye sent Thompson the status of the Concentration-Isolation Process. The conclusions that are reached from work on the mainline process during recent months are as follows: (1) The overall concentration-isolation process as used in this laboratory will give good yields on laboratory solutions. (2) The process is also satisfactory in stainless steel on Clinton plant solutions with the exception of the precipitation of the first peroxide in the isolation procedure. (3) It is felt that these experiments present strong evidence that zirconium and iron present in the plant solutions are causing poor yields. However, it is questionable whether these are the only ions that cause poor yields in the first peroxide precipitation since some trouble was experienced with the 1 g lanthanum fluoride slurries obtained from Cell 4 that had never been in contact with zirconium as a reagent. Additional work is therefore necessary. (4) The process will be satisfactory if an additional lanthanum fluoride "clean-up" cycle is incorporated in the flowsheet following the metathesis step.

The proposed program concerning mainline process problems will consist of: (1) studies on the concentration procedure to reduce corrosion of tanks by HF and the use of other metathesis reagents to improve quality of the first plutonium peroxide precipitate; (2) studies on the isolation procedure, including small-scale experiments to secure higher yields of plutonium peroxide in the first precipitation, and studies of product

solution ion equilibrium, the effect of presence of Pu(VI), and plutonium peroxide slurry filtration rates; (3) the development and study of alternate isolation methods should trouble be encountered at Site W; and (4) basic chemistry studies with emphasis on the following investigations: [a] the structure of plutonium peroxide, [b] the exact mechanism of the precipitation reaction, [c] the function of zirconium in preventing precipitation, [d] the behavior of the other valence states with H_2O_2, [e] the volume and nature of gas evolved in the presence of known amounts of iron.

In a memo to A. C. Hyde, Gilbreath describes his investigation of cerium and zirconium scavengers in the Bismuth Phosphate Process. Appreciably increased decontamination factors have been observed in laboratory runs using these scavengers in the oxidation step of the decontamination cycle. In the experiments, following product oxidation and dilution of the oxidized solution to 1.0 N HNO_3, the H_3PO_4 arising from the dissolution of bismuth phosphate is precipitated by the addition of an excess of bismuth. Cerium and zirconium are then added, and their phosphates along with the remaining bismuth are precipitated by the addition of H_3PO_4. Gilbreath recommends that at least two Chicago semiworks runs be made using the one centrifugation procedure, and suggests at least two runs using the "two-shot" method developed at Site X. Gilbreath describes both of these procedures in detail but suggests that when the runs are made that further details be obtained from him or from Dreher.

A meeting on Purification and Metal Production by the sub-section of C-I was held at 7:45 p.m. in Eckhart Hall. Manning and Arnold reviewed the new air filtering system and procedures concerning its use. Manning said our chemists must take it upon themselves to clean up radioactive hot spots, that surveys will be made every other day, and that no active waste material of any kind should be discarded into the wastebaskets but must go in the waste containers which have been recently provided. Reinhardt, speaking on solvent extraction, listed experimental results obtained with 42 different solvents tested with Pu(IV) and Pu(VI). Gilpatrick described the method being used in our laboratory to determine the vapor pressure of plutonium metal. He drew a diagram of the apparatus on the board and illustrated its operating principles. Sears commented on vapor pressure measurements made on plutonium metal; over a temperature range of 1258° to 1678°C, the vapor pressure increased from 3.05×10^{-6} mm Hg to 3.07×10^{-2} mm Hg.

Hellman described refractory studies. Most oxides including BeO, La_2O_3, CaO, and ZrO_2 react with plutonium metal. Of these BeO is the least unsatisfactory. Cerium subsulfide and zirconium nitride show promise. S. Katz summarized experiments during which plutonium metal samples that were subjected to pressures of 4,000 lbs per square inch and 140°C temperature were examined to determine if density changes could be observed. The samples of low density that have been so treated are found to have their densities raised; the samples of high density metal are found to have their densities lowered. The magnitude of these changes is about one density unit. Dilatometric methods of following the suspected phase change are now underway.

Helen worked at the Met Lab today and spent the evening at Wilma's while I was at the meeting.

The war news today indicates that the Germans have hurled four armored divisions into the battle in France, but the Allies are holding firm.

Friday, June 16, 1944

We learned that Iz Perlman from Clinton will be in Chicago next Monday through Wednesday.

D. F. Hewett of the Geological Survey in Washington, D. C., wrote expressing his appreciation for receiving several references to methods for determining thorium in low concentrations. He said the Geological Survey has been working on methods for determining minute quantities of uranium and expects to have the problem solved soon.

Hilberry requested that all visitors to Site X must obtain CEW (Clinton Engineer Works) passes for each trip from his secretary, Dorothy G. Lipps.

I sent a letter to Greager of Clinton Laboratories about the use of sodium nitrite for prereduction in the Bismuth Phosphate Process. I described in detail the procedure developed on the basis of our laboratory and semiworks experience and suggested that he may wish to recommend the use of $NaNO_2$ in a test run in the Clinton plant. I wrote, "The 40% UNH solution from the metal dissolver may be diluted with water containing dissolved $NaNO_2$ to give a final UNH concentration of about 20% and a $NaNO_2$ concentration of approximately 0.1 M. This solution, at a temperature of the order of 35°C, may then be made 1 N in H_2SO_4 and heated to 75°C at which temperature the solution should be held for one hour. At the end of this hour the reduction of Pu(VI) should be complete, and the Bi(III) may be added immediately. The extraction step is then carried out in the usual manner." I also noted that brown fumes are produced when the H_2SO_4 is added but that we do not believe this will be a serious hazard.

Albaugh, Dreher, Howland, Pye, Smith, Thompson, Watt, Willard, and I attended a meeting on Basic Process Chemistry at which our general program for the immediate future was outlined. Because of the need for more detailed information and the fact that Cunningham could not be present because of his trip to Clinton Laboratories, a follow-up meeting is planned for June 20.

Helen worked at the Met Lab and later at the downtown YWCA. Then she and I went to the movie "Broadway Rhythm" in the evening.

The U.S. is still ahead in France, but the top news of the war today shifted to the Pacific where super-fortresses raided Japan and Nimitz invaded two new islands in the Marianas.

Saturday, June 17, 1944

Herbert H. Anderson, whose return to the Met Lab last December was marred by army induction, has returned to Section C-I as a SED man. He is working in Dawson's group.

Cunningham returned from his visit to Clinton Laboratories.

I received a copy of a 17-page write-up prepared by Morgan and Katzin entitled, "Introductory Notes on Fission Product Activity and Decontamination." It will serve as an invaluable source of information for use in the training of our incoming chemists.

An information meeting of Sub-section II was held at 2:00 p.m. in preparation for the forthcoming Thomas meeting. The manpower assignments in Sub-section II were discussed. Orlemann summarized the manpower assignments in Sub-section II as of June 17. These are as follows:

Group 4. Purification and Analysis of Plutonium Compounds (Jensen, Group Leader). Supervision, reports, x-ray, and plutonium analysis records (Jensen). Extraction of inorganic plutonium compounds by various solvents; light element separations by solvent extraction; study of physical properties of solvents for use in continuous extraction devices (Reinhardt, Stein, and Dixon). Development of continuous solvent extraction technique in cooperation with the Technical Division (Brody).

Group 5. Volatility and General Dry Chemistry of Plutonium Compounds (Davidson, Group Leader). Supervision, reports, liquid phase chlorination in bombs (Davidson). Syntheses of plutonium iodides using various iodinating agents; vacuum sublimation of halides and oxy-halides (Hagemann). Reactions of plutonium compounds with HI (J. Katz). Syntheses of $PuCl_3$ by various methods (Abraham). Synthesis of $PuBr_3$ and determination of its properties (Hyde). Analyses of plutonium halides; study of halide hydrates (Wolf).

Group 5A. Chemistry of the Plutonium Fluorides (Heath, Group Leader). Supervision, reports, properties of the lower fluorides (Heath). Chemistry of higher plutonium fluorides (Florin). Study of methods of lower fluoride synthesis; properties of the lower fluoride; preparation of fluoride for metal production (Meyer). Analyses of the fluorides; preparation of fluorides for metal production (Zvolner).

Group 6. Metal Production (Baumbach, Group Leader). Supervision, reports (Baumbach). Density determinations; preparation of samples for analysis; dilatometric studies of the phase transitions (S. Katz). Methods of preparing plutonium by thermal reduction of the halides in vacuo (Westrum). Thermal bomb reduction on the ca. 100-mg scale (Robinson and Fried). Preparation and study of refractories for use with plutonium (Hellman and Gerstein). Melting point determination; preparation of plutonium samples for physical studies; electrolytic preparation of plutonium; phase transition studies (Jasaitis). Hardness determinations; metallographic examinations (Frank).

Group 6A. High Vacuum Techniques (Simpson, Group Leader). Reports, direction (Simpson). Vapor pressure of the metal and halides

(Simpson, Phipps, Sears, Gilpatrick, and Johnson). Oxygen analysis (Seifert).

Helen had lunch at Wilma's and then this sweltering evening we had dinner at the home of Winston and Dorothy Manning (5612 Ingleside Avenue).

Top headlines today say the Allies continue to gain in France and U.S. troops smashed at three of the Bonin Islands.

Sunday, June 18, 1944

Katzin, Albaugh, Arnold, and I played golf at the Evergreen course. Our scores were LK — 167, LA — 96, GS — 111, and FA — 71 (nine holes).

Kennedy dropped by our apartment in the evening for a visit. He arrived in Chicago yesterday from Los Alamos and is staying at the Windermere East Hotel where, after dinner, he ran into Latimer, who has just been in Panama and Washington, D.C. Kennedy told me of the discussions he, Segrè, and Wahl have had with Lavender when the latter visited Los Alamos last Thursday. Lavender explained that any cases prepared in his office could not be used because the U.S. strictly forbids him to help any case "against" the U.S. He wants to hire a lawyer for us to work up the cases to be submitted for sale. He does not like the idea of Owen doing this, pointing out the complexity that Owen cannot work for both the University of California and us. Kennedy told me that Wahl and Segrè will go along with hiring a lawyer, up to $500 each.

Monday, June 19, 1944

Kennedy and I called Lavender in Washington about the meeting Lavender had in Los Alamos last Thursday with Kennedy, Segrè, and Wahl. In view of what Lavender said about being unable to use cases against the Government that he himself has helped to prepare, we agreed to write up new cases.

Asher Margolis is transferring from Dreher's group to the Technology Division to work with A. C. Hyde. Shirley Nyden was hired to work as a technician for Heath.

Iz Perlman, Spedding, and W. C. Johnson arrived today for the Thomas meeting and Information Meeting on Chemistry.

James is back at work after a short honeymoon. In the search for new alpha-emitting isotopes of neptunium, in order to eliminate Pu^{238} he completed eight cold bromate oxidation cycles on the deuteron-bombarded uranium target from the 2,350 microampere-hour bombardment in the Berkeley

cyclotron last March 3 and 4. James finds there is a signficant amount of alpha activity in the "neptunium" fraction but is not sure whether or not it is due to growth or to contamination. He will continue with the bromate oxidation cycles.

La Chapelle and Magnusson completed additional bromate cycles in the isolation of Np^{237} from the 64 pounds of uranium metal that was bombarded with high energy neutrons in the Berkeley cyclotron. On the basis of their observed alpha activity they compute the yield to be 72 micrograms which speaks well for a successful isolation of Np^{237} in pure form.

I attended the monthly meeting with Thomas on "Purification and Metallurgy of 49." During the morning portion of the meeting, which began at 9:30 in Room 209 of Eckhart Hall, the minutes and special assignments resulting from the May meeting were discussed in a preliminary session followed by presentations on the specific subject, "Metal Production and Properties of Metal." Others present at the morning session besides Thomas and me were Ashcraft, Baumbach, Chipman, Compton, Derge, Eastman, Frank, Hilberry, Hogness, Jeffries, Kennedy, Latimer, Spedding, and Warner.

Kennedy reported on the work at Site Y relative to plutonium metal production, analytical data and neutron counts on plutonium metal samples, evidence of allotropic transformation, mechanical properties, and remelting of plutonium. With regard to metal reduction, Kennedy said the stationary bomb method of reducing $PuCl_3$ with calcium has been developed to a point where it now gives good yields of relatively pure metal. Although Site Y has nearly frozen on this method for producing plutonium metal, electrolytic reduction has been successful to a limited extent and produces very malleable, homogeneous metal. Recent work has shown the importance of using plutonium chloride free from oxygen — a chloride which gives excellent results when first prepared, gives inferior results after exposure to oxygen for several days even under "dry" conditions. Kennedy also described work on the reduction of $PuCl_3$ by barium vapor, and of PuO_2 by carbon. Jeffries commented on the PuO_2-carbon reduction work and considered it to be highly significant as it could eliminate the necessity for making halides and would permit the use of graphite as a refractory. The density of lithium-reduced plutonium metal, which is cold-worked and annealed, varies between high and low values of 18.5 and 16 gm/cm^2, respectively. This is considered evidence for allotropic crystal transformation.

Kennedy said expansion versus temperature measurements have shown a transition point for plutonium metal. I expressed some concern that it would be almost impossible to machine plutonium without some transition occurring. Kennedy agreed and said that desired shapes would probably have to be produced by casting, followed by cold forging. The best refractory crucibles tried by Site Y to date for remelting plutonium are the reduced cerium sulfide types, called "brass" crucibles, prepared by Eastman at Berkeley. Uranium nitride is satisfactory for casting if the temperature is kept only slightly above the melting point of the metal for short times. Magnesium oxide also seems satisfactory under these conditions, but purity of the product may not be so good.

I presented the work of the Met Lab Chemistry Division following Kennedy's report. I reviewed our work that shows evidence for allotropic transformations in plutonium metal. Density changes and metallographic and x-ray diffraction evidence were presented; these agree well with Kennedy's data from Site Y. Our work of the last few days on dilatometric studies was presented, giving much more conclusive evidence for the transformation than any of the other studies.

I described our method for determining the vapor pressure of plutonium metal and presented data obtained. Extrapolating these data to 760 mm gives a boiling point of about 2260°K. Hogness emphasized that this value and the values for vapor pressure should be treated as preliminary and minimum values, as larger samples of plutonium are needed for more accurate measurements. Hogness asked to borrow a 50-mg sample of the purest plutonium metal available at Site Y to use with new apparatus being constructed in Section C-I.

I reviewed our metal production work and gave data on our vapor phase reductions of PuF_3 with barium and on our bomb reduction studies that have given metal in good yield for the four reductions we have made of PuF_3 by means of lithium. During the last few days, one approximately 100-mg bomb reduction was successful and yielded about 60 mg of metal.

Reports by Spedding of Ames and Derge of the Met Lab Technical Division on metallographic studies and impurity analyses of uranium and thorium metals completed the planned portion of the morning session. Before adjourning for lunch, Compton described a suggestion made by Szilard for the purification of plutonium metal. This method would use a travelling induction heater to produce a liquid metal zone that could move slowly through the length of a cylinder of metal carrying with it many of the impurities in the metal. After some discussion it was decided that although the idea is an interesting one, it would require an enormous amount of time and work to establish feasibility. As the other present methods seem very promising, it was recommended that no effort be expended on Szilard's suggestion at this time.

The afternoon session, which met at 2:00 p.m. in Room 209 of Eckhart Hall, was on the subject of "Refractories for Use in the Production, Remelting, and Casting of Metal." In attendance were Ashcraft, Baumbach, Chipman, Derge, Eastman, Hilberry, Hogness, Jeffries, W. C. Johnson, Kennedy, Latimer, Norton, Peterson, Spedding, Thomas, Warner, and I. Thomas made the opening remarks and introductions. This was followed by Kennedy's report on Site Y refractories requirements. F. H. Norton, of the Central Refractories Laboratory of M.I.T., reviewed the work of his laboratory, and described his production capabilities for large numbers of crucibles needed at Site Y in the immediate future. Eastman of the Berkeley Project reported on sulfide refractories, in particular, CeS crucibles. He also reported on effects of vacuum fusion on light element impurities and on the analyses of light elements in metals. Spedding of Ames discussed work at Ames on CaO and UN refractories.

I described the refractory work of the Met Lab Chemistry Division, including the remelting studies with 1-mg particles of plutonium metal in oxide, sulfide, and nitride crucibles and test strips. Our results with $CeS_{0.9}$ are very encouraging; they show no evidence of crucible attack by

the metal and the hardness of the metal after remelting is normal (around 150).

In the concluding remarks and summary for the refractory session, the recommendation was made that producers of new or improved refractories should obtain some preliminary results on suitability by sending samples to me for trial on a small scale. Kennedy recommended that our small-scale trials at Chicago be made at several temperatures just above the melting point of the plutonium metal.

I attended the Executive Session, which completed the Thomas meeting. The session began at 4:30 p.m. in Room 209 of Eckhart Hall. Others present at the session were Chipman, Compton, Eastman, Hilberry, Hogness, Jeffries, Kennedy, Latimer, Peterson, Spedding, Thomas, and Warner. As a result of this session, the following decisions affecting the Chicago Chemistry Division were made: a. It is recommended that Site Y lend 50 mg of their purest metal to the Chicago Chemistry Division for use in the determination of the vapor pressure of Pu^{239} as a function of the temperature. We believe the measurements can be completed in ten days after receipt of the metal. It was suggested that Chicago should also determine the dissociation pressure of plutonium carbide. The latter is of importance in connection with the new method of making metal by the reduction of the oxide with carbon. Thomas will forward this recommendation along with a request to Oppenheimer. b. The Chicago Chemistry Division should continue its program on the development of a continuous solvent extraction process for purification of plutonium. This also is related to separation, isolation, and concentration at Hanford. c. The Chicago Chemistry Division should continue with metal production by the small-scale technique as a tool to complete all steps in the process. The study of bomb reduction on a larger scale should not be undertaken at Chicago. Chicago should have the option of receiving its allotments of plutonium as compound from Site X or as metal from Site Y.

Allison responded to Compton's request that Allison discuss the problem of toxic and radiation effects "following operation of one of our final devices" (nuclear detonation) during his visit to Site Y. The necessity for such a consideration was pointed out by Franck at the June 7 meeting. Allison reported that Oppenheimer informed him that the problem is definitely considered to be the responsibility of Site Y and that they are actively engaged in the investigation there.

Orlemann received a copy of Arnold's letter to C. A. Kraus of Brown University inviting him to visit us the early part of the week of July 3.

"Chemistry Division, Summary Report for May, 1944," (CS-1725), was issued. The work of my section was summarized in 19 pages under three main headings: Separation Process Studies, Purification and Metal Production, and Basic Chemistry.

Report CN-1700 by Hogness, Franck, Seaborg, Willard, and Thompson for the month ending June 1, 1944, entitled "Chemical Research — Separation Processes for Plutonium," was issued today. It includes information

contained in my memo (MUC-GTS-735) to Hogness of June 5.

Iz Perlman had dinner with Helen and me in our apartment.

Governor Dewey of New York claims he has enough support to win nomination as the Republican candidate for the presidency.

A meeting of the Project Advisory Board was held this evening in Eckhart Hall. The agenda consisted of three major items. (1) The policy which the Project should adopt toward items in its program concerned chiefly with the provisions of "insurance" for the Hanford operations. (2) Discussion of the policy to be adopted toward future operations of the Clinton Laboratories. (3) A discussion of the joint British, Canadian, and American Project at Montreal.

Tuesday, June 20, 1944

Using the 72 micrograms from the Berkeley cyclotron source and about 35 micrograms from Site X pile material, Magnusson and La Chapelle have continued their purification procedure and have now managed to precipitate about 20 micrograms of Np^{237} as the hydroxide; they dried it under a heat lamp and brought it to orange heat in a platinum crucible using a Bunsen burner. The resulting oxide was transferred to a capillary for x-ray analysis by Zachariasen.

Thompson sent a memo to L. B. Arnold, Jr. about the shortage of glassware for use in developing the Hanford separations Bismuth Phosphate Process and requested an AAA priority, together with expeditious handling, for 45 dozen assorted items.

I attended the Project Council Information Meeting on Chemistry which began at 9:30 a.m. in Room 209 of Eckhart Hall. Others present were Ashcraft, Boyd, Burton, Compton, Connick, Cooper, Dempster, Doan, Eastman, Franck, Greager, Hilberry, Hogness, Hume, Jeffries, W. C. Johnson, Latimer, Lum, McKinney, Mulliken, Perlman, Quill, Spedding, Stearns, Stone, Sugarman, Sutton, Thomas, Vernon, Warner, W. W. Watson, Whitaker, Wigner, and Zachariasen. Following the opening of the meeting by Hilberry and introductions of the Chicago Chemistry Division speakers by Hogness, I presented our Section C-I work on problems for Hanford and Los Alamos and our work on basic chemistry. In discussing Hanford problems, I pointed out that at the time of the last Project Council Chemistry meeting (which was last held in Chicago on May 16) the actual demonstration of a decontamination factor of 10^5 in the canyon and 10^7 through the crossover had rarely been achieved. We now have shown that after two bismuth phosphate cycles, with scavengers, a decontamination factor of 5×10^5 can be obtained. We have also given some effort to developing a process which operates without a scavenger. The abnormally large volumes of solutions with low plutonium concentrations, 30 to 50 grams of product per ton, which will

result from Hanford's early operation, make it necessary to modify the Bismuth Phosphate Process. The modified process will use: (a) higher UNH concentrations in the extraction step, (b) smaller amounts of bismuth ion per unit volume, (c) more than one extraction tank in parallel operation, and (d) higher concentrations of acid through the decontamination step making it possible to combine batches after extraction.

I reported our work on the identification of potential losses in plutonium yield in the extraction step due to the oxidation of Pu(IV) to Pu(VI) by HNO_3. The Pu(VI) thus formed is lost in the extraction step. As an important partial solution to this problem, sodium nitrite has been found to be a good agent for reducing Pu(VI) to Pu(IV) in the prereduction step. I pointed out that Pu(VI) is not the entire reason for losses because there apparently is another form of Pu(IV) ion (possibly resulting from the hydration or hydrolysis of the more ordinary Pu(IV) ion). This newly identified form of Pu(IV) ion is formed slowly in process solutions with high acidity and is not carried well by bismuth phosphate. In an attempt to minimize this problem, we have found that the presence of H_2SO_4 tends to stabilize the ordinary Pu(IV) ion and therefore tends to inhibit the formation of the new form of Pu(IV).

With regard to Los Alamos problems, I described our work related to the determination of plutonium metal density. During the past several months densities of 13, 16.3 ± 0.5, and about 20 gm/cm^3 have been obtained on samples supposed to be plutonium metal. We now believe the material with a density of 13 gm/cm^3 is probably plutonium oxide or plutonium nitride. Some time ago Zachariasen suggested the possibility of two allotropic modifications. This has now been clearly demonstrated to be true at Los Alamos and also in our Chicago Laboratory. The transformation occurs at about 140° C. The low temperature form, called the alpha form, has a complex crystal structure and a density close to 20 gm/cm^3. The high-temperature beta form may have a face-centered cubic structure and appears to have a density close to 16 gm/cm^3. In addition to our heating and quenching experiments which verify the Site Y results, we now have good dilatometric evidence for the transition.

I described the preliminary vapor pressure measurements of plutonium metal, as a function of temperature, over a range of 1258°C to 1648°K, made by Phipps and Simpson. If the logarithm of the vapor pressure data obtained is plotted against the inverse of temperature, the data fall on two straight lines that correspond to heats of vaporization of 120,000 cal/mol at high temperatures and 87,000 cal/mol at low temperature. A boiling point of about 2000°C is obtained by extrapolation of the high temperature line to 760 mm pressure.

I reported our success in producing PuI_3 by the reaction of HI on plutonium metal. X-ray analyses by Zachariasen positively identify the PuI_3 and show that it is isomorphous with $PuBr_3$. The volatility of PuI_3 is not so high as we would like. It seems likely that PuI_4 does not exist. This makes the "hot wire" method of metal production seem less promising.

About our Basic Chemistry work, I reported that during the past month K_2PuF_6 has been prepared and has been found to be isomorphous with the corresponding double salts of Ce(IV) and Th(IV). The previously

described newly identified Pu(IV) ion has been studied with an absorption spectrophotometer and gives no peak at 4,706 Å, the location of the strong peak for ordinary Pu(IV). However, the absorption spectrum shows new peaks at 5,780, 6,180, 7,360, and 8,170 Å.

Burton presented Section C-II work on the chromate problems at Site W, and on the effects of prolonged in-pile exposure on the conductivity and other properties of graphite. The changes in properties observed by Burton's group emphasize the need to proceed with experiments at CP-3 with all possible haste. Burton said a solid moderator used in a pile will "inevitably finally go bad." Franck said Burton's calculations are based on assumptions that are not entirely true and he believes that we would have a warning before any failure. In ensuing discussions among Compton, Burton, and Franck, it was agreed that the statement about the eventual failure of a solid moderator is too strong if the operating time limit were, say, 200 days. All agreed, however, that it would be a fair statement to say: "It is improbable that operation with graphite as a moderator can go for two years" (at planned Hanford levels).

Sugarman described fission product work conducted by his Section C-III. Ultra-fast (high energy neutron) fission product activities in Clinton material are being looked for and studied. The isotopes Ag^{111}, Pd^{112} (with Ag^{112} daughter), and Cd^{115} have been found and identified. Uranium-237 has been found in Clinton material, probably produced from U^{238} by a n,2n reaction. Longer-lived xenon is being looked for. Studies on the fission yields from plutonium were started this month. It was found that the fission yield curve for the light element fission product group is probably shifted because of the higher mass of plutonium than uranium. The Ba^{140}/Sr^{89} ratio from plutonium fission is about 2.5-fold higher than from uranium fission. The shift is about 4 mass units to the heavier element side. This emphasizes the silver, palladium, and cadmium group. The distribution of yields for the heavy fission product elements appears to change very little going from uranium to plutonium.

Ashcraft reported on the work conducted by his Section C-IV on light-element analysis capabilities. The groups under Curtis and Kirk are working on process control methods for Hanford and have prepared volumetric or colorimetric methods for the analysis of some 30 ions that incorporate remote control equipment and which is in the design stage. An approximately 200-mg sample is presently needed to analyze for all the light element impurities at process tolerance limits. Hogness mentioned that the procedure will be to make a "shot-gun test," i.e., overall test for neutron emission from the final plutonium metal; if the product is sufficiently pure, no analyses for individual light elements will be required.

W. C. Johnson introduced the Clinton Chemistry speakers. Perlman reported on "Product Carrying and Decontamination Studies for Hanford." The "abnormal" form of Pu(IV) ion has been found to be very important in interfering with plutonium carrying. The solubility of the "abnormal" ion, as $Pu(IO_3)_4$, is found to be about 100 times greater than $Pu(IO_3)_4$ with "ordinary" Pu(IV). There are also large differences in the precipitation of the peroxides from solutions containing ordinary and those containing "abnormal" Pu(IV) ions. The Clinton work on the identification of the "abnormal" plutonium ion and its relation, along with other factors,

to the possible plutonium loss in the extraction step, verifies our Section C-I work.

In discussing decontamination, Perlman said a process has been designed that will give a decontamination factor of 10^7 and will be compatible with the Hanford process. Four runs using this procedure gave whole process plutonium yields of 85%, 80%, 87%, and 94%, and decontamination factors of 2×10^7, 1.5×10^7, 1×10^8, and greater than 1.5×10^7, respectively. Calculations show that about 3% of the fission product activity at Hanford will be caused by plutonium fission.

Boyd reported on "Adsorption Process and Decontamination Studies;" and Hume reported on "Short-Lived Fission Product, Plutonium Fission Products, and Production of Fission Radio-Lanthanum." Hume described a system for counting uranium foils for alpha particles within a few seconds after exposure to a neutron beam. With this device, no evidence has been found for alpha-emitting fission products with half-lives within the 0.8 to 1.5 seconds range.

Greager introduced the Clinton Semiworks and Separations Development speakers and then reported on "Additional Studies on the Influence of Strike and Digestion Time on Percents of Product and Bismuth in Waste." In several experimental runs it was found that if H_3PO_4 is added suddenly to the solutions containing Bi(III), the plutonium is not carried at all at Hanford levels. I questioned Greager as to the valence state of the plutonium ion and whether or not the observed effect might not be due to the "abnormal" form of Pu(IV). Greager admitted this might be the case. He also reported that preliminary results using $NaNO_2$ in the prereduction step on the semiworks level are very encouraging. Greager said that he does not anticipate any trouble from NO_x fumes as the plant is well equipped with ventilation facilities.

Sutton reported on the following Hanford-related problem assignments conducted by his Separations Development Section: (a) improvement in decontamination, (b) product isolation, (c) improvement in yield, and (d) flocculation studies. Reproducibility on the semiworks scale has been good; gamma decontamination has averaged 1.5×10^5 and the individual runs have not varied by more than a factor of 3 from this average. Refinements in the process have been investigated, and one employing barium sulfate in addition to the cerium-zirconium scavengers has given gamma decontamination factors averaging 2.2×10^6 for four semiworks runs. Plutonium losses were a few percent higher, however, than runs without scavengers.

Sutton, in discussing Hanford product isolation work, said the plutonium concentration at average Hanford levels of operation will be sufficiently high to permit direct precipitation of plutonium peroxide. Previous work at Clinton shows that the impurity level is too high at the end of the standard crossover cycle to allow quantitative direct precipitation to product peroxide when the plutonium concentration is adjusted to Hanford levels. Laboratory experiments conducted during the past month, however, have shown that the impurities interfering with plutonium precipitation can be removed by lanthanum fluoride precipitation purification procedures. The lanthanum fluoride by-product precipitation step has been identified as being the important step in the procedure. Elimination of the impurities (iron, nickel, zirconium, manganese, bismuth, and calcium)

carried by the lanthanum fluoride by-product decreases the plutonium peroxide solubility from 4,850 mg/l to 25 - 50 mg/l.

Eastman and Connick were introduced by Thomas as the speakers from Berkeley. Eastman talked about "Refractories, Remelting, and Electrolysis Experiments," covering essentially the same information presented at yesterday's Thomas meeting. Connick discussed the Berkeley work related to "The Valence States of Plutonium." The +3, +4, and +6 valence states of plutonium are now well-known. Connick reported on recent evidence which supports the existence of a +5 state. A colorless solution is often obtained upon reducing Pu(VI) with SO_2 or hydroxylamine. This liquid does not show absorption spectra peaks for the +3, +4, and +6 states. The results of even more convincing electrolytic reduction experiments were described together with possible change-of-state mechanisms. So far no compounds of Pu(V) have been prepared and identified. With regard to the analysis for plutonium valence states, Connick said the method for Pu(III) and Pu(IV), developed by Don Ames, seems to be the best. This method makes use of the fact that more than 98% of Pu(IV) and less than 1% of Pu(III) are carried by a zirconium phenylarsonate reagent, with an excess of reducing agent present. Less than 0.5% of Pu(VI) is carried by the above reagent. The magnetic moment for PuO_2^{++} has been measured in 0.5 M HCl at Berkeley and is found to be 2.85 Bohr magnetrons.

Spedding summarized the progress at Ames on the properties of uranium compounds. In addition, he reported that Ames has recently found in tracer scale studies that ditertiary butyl-dichloro-disalicylalethylene-diamine is a promising complexing reagent for Pu(IV) and extracts more than 98% of Pu(IV) into $CHCl_3$ from aqueous solutions in the pH range of 3 to 7. Work is continuing using Ferron as a complexing reagent in the absorption process.

Compton introduced Kennedy as the speaker for Los Alamos, who reported on "Progress in Production and Remelting of Metal; Properties of Metal-Allotropy." Yields of 80% or better have been obtained in 16 of the 17 recently completed plutonium reductions on the 0.5 to 1.5 gram scale. The one failure was an electrolytic experiment thought to be caused by excessive oxygen in the plutonium salt. The stationary bomb technique using calcium as the reducing agent with MgO or CaO as the liner is now very satisfactory and has replaced the lithium reductions in BeO crucibles in the centrifuge. For remelting, the CeS ("brass") crucibles made by Eastman at Berkeley are the best refractories tried to date. ThS seems inferior to CeS. So far Kennedy has not determined how much purification can be accomplished by vacuum remelting.

Data on neutron emission rates from plutonium metal products were presented by Kennedy and give an indication of the amount of light element impurities in the metal. A new method for producing metal was proposed, which involves the distillation of plutonium from PuC_x formed in the reaction:

$$PuO + (2+x)C = PuC_x + 2CO \quad .$$

Temperature arrests, dilatometric, and density changes have provided definite evidence for an allotropic transformation in plutonium metal at 136° ± 10°C.

Manning replied to Nickson's memos of June 12 and 13 about contamination in Rooms 1, 2, 4, 10, 11, 30, 31, 34, and 35 of the New Chemistry Building. Manning reported that the rooms have been cleaned up and that, with the exception of Room 10, all have been monitored and found acceptable. Room 10 will be checked within the next 24 hours.

A meeting was held, as a follow-up to the meeting on June 16, on Basic Process Chemistry, attended by Albaugh, Cunningham, Dreher, Goeckermann, Hopkins, Howland, Perlman (from Clinton), Pye, Thompson, and Watt. More detailed plans for work on the basic chemistry work were discussed, particularly for the peroxide problem. The minutes of the meeting present the following information:

a. Mechanism of peroxide precipitation. Man assigned: Hopkins. In order to understand how various ions may affect the precipitation of plutonium peroxide, it is first necessary to know something of the mechanism of the precipitation from a relatively pure plutonium(IV) solution in nitric acid. Recent results of Werner at Clinton indicate that the formula may not be $Pu(O_2)_2 \cdot xH_2O$ as suggested by Howland (CK-1511), who found that a washed peroxide contained six equivalents of reducing power for ceric nitrate per mole of plutonium. Werner found only about 5.4 equivalents of reducing power in very similar experiments. Various methods of removing H_2O_2 from the plutonium peroxide will be tried including drying at about 90°C and washing with both water and organic solvents. Successive water washings will be titrated for H_2O_2 content to determine how soon it approaches a minimum value (the solubility of plutonium peroxide). The formula of the peroxide precipitated under various conditions including from a solution of the abnormal or "green" Pu(IV) will be determined later.

There is no definite proof that the plutonium in the peroxide precipitate is Pu(IV). However, that is the most logical choice when precipitation is made from H_2SO_4 solution in which reduction to Pu(III) should be impossible. Additional information may be obtained by dissolving the precipitate in HCl (this would not favor Pu(IV) in solution as would H_2SO_4 and HNO_3), and examining the solution spectrophotometrically.

It is possible that impurities such as iron and zirconium interfere with the precipitation of the peroxide either by acting upon the plutonium or upon the H_2O_2. Since Fe(III) is known to catalyze the decomposition of H_2O_2, this reaction will be followed to determine whether it occurs rapidly enough to account for the observed increase in solubility of the plutonium peroxide. Iron and zirconium will also be added to uranyl solutions from which the peroxide of uranium is to be precipitated. Since the structure of this compound is much different [a peroxy acid of U(VI)], its precipitation may possibly be unaffected by those impurities if they do not function by lowering the concentration of available H_2O_2. As a more direct approach the absorption spectrum of a zirconium solution will be taken with and without H_2O_2 present.

A search will be made for a suitable complexing agent for Fe(III) that will allow it to be present in larger amounts without increasing the solubility of plutonium peroxide. The extent to which iron is carried by LaF_3 should be determined as it would serve as a guide to how much iron may be encountered in the process solution out of which the first peroxide precipitation will be made.

b. Bromate and dichromate as holding oxidants for LaF_3 assays. Man assigned: Howland. Either 0.03 N $K_2Cr_2O_7$ or 0.02 N $KBrO_3$ have been found by Dreher's group to be satisfactory holding oxidants to allow LaF_3 assays for Pu(IV) in the presence of Pu(VI). However, extraction step studies in which the bromate-lanthanum fluoride assay was used showed variable and quite incomplete reduction by $NaNO_2$. Similar experiments at high product concentration always indicated complete reduction according to the spectrophotometric analysis for Pu(VI). Since the discrepancy may be due to faulty assays, the carrying method will be checked on product solutions of known Pu(IV) and Pu(VI) content determined by the absorption spectrum. The product concentration will be reduced 2500-fold by two successive dilutions into $K_2Cr_2O_7$ or $KBrO_3$ solutions before making the LaF_3 precipitate. In some experiments the lanthanum will be added 30 minutes after the HF; in others HF will be added last.

c. Extent of oxidation and disproportionation at 25° in 40% UNH. Man assigned: Howland. It is possible that in the low acidity solutions of 40% UNH some oxidation or disproportionation of Pu(IV) may occur during the storage period before $BiPO_4$ extraction. Also some of the Pu(IV) might be converted to the abnormal or "green" form. The experiments will be performed at a concentration of about 0.5 g Pu/l in order to allow spectrophotometric analysis. Similar experiments will be carried out in Dreher's group with product concentration 50 mg/l and using the $KBrO_3$-LaF_3 method of assaying for Pu(IV).

d. Rate of oxidation and disproportionation as a function of product concentration. Man assigned: Ames. A comparison of existing studies of oxidation and reduction of plutonium indicates that, in general, when the concentration of plutonium is lower, the reactions seem to proceed to a smaller percentage of completion in the same period of time. The rates of disproportionation of Pu(IV) in HCl solution and of the oxidation of Pu(IV) by $Ce(SO_4)_2$ in H_2SO_4 solution will be determined spectrophotometrically over the range of concentration 0.25 g Pu/l to 2.5 g Pu/l.

e. Solubilities of plutonium(VI) phosphate and of plutonium fluorides at low acidity. Man assigned: O'Connor. The solubility of plutonium(VI) phosphate in 0.6 M H_3PO_4 will be determined at a lower acidity (0.03 N HNO_3) than previously required since this condition might be useful in precipitating scavengers. The solubilities of the plutonium fluorides will be investigated at various acidities including as low as 0.1 N HNO_3 which is being considered for use in the concentration procedure.

f. Occurrence of the abnormal Pu(IV) in the process. Various process solutions will be examined in the spectrophotometer for the presence of the abnormal "green" form of Pu(IV) which is not carryable by $BiPO_4$ and which precipitates slowly with H_2O_2. Since low acidity is known to favor formation of the "green" Pu(IV), the solution from LaF_3 metathesis and the solution of 40% UNH which may be stored before $BiPO_4$ extraction are the most likely points at which the undesirable form might be produced. The UNH solution will be tested under problem 3; the metathesis solution under problem 1.

g. Effect of ions on carrying by $BiPO_4$. Iron(III) above 0.01 N interferes with carrying of Pu(IV) by $BiPO_4$. Whether the interference is through a mechanism of tying up the Pu(IV) in solution or by preferential

building of Fe(III) into the $BiPO_4$ crystal lattice is not known. A study of carrying as a function of Fe(III) concentration and including analysis of the $BiPO_4$ for iron content may be useful. A knowledge of which other ions also interfere is of immediate practical value.

h. Effects of agitation, method of addition of reagents, etc., on carrying by $BiPO_4$. Since the mechanism by which Pu(IV) coseparates with $BiPO_4$ is still unknown, the optimum conditions for carrying have not all been defined. Variables that might be studied include agitation, order of addition of reagents, and time schedule for addition of reagents and for digestion. The time factor might be important since $Pu_3(PO_4)_4$ and $BiPO_4$ precipitate at different rates.

i. Solubility of LaF_3. As a guide for planning of fluoride concentration procedure experiments, the solubility of LaF_3 over a wide range of conditions is required.

Problems g, h, and i will not be begun at the present time.

Helen had coffee at Wilma's. I ended the day with a migraine headache.

Today the front page of the paper indicates that the Allies are increasing their hold on the Cherbourg Peninsula while Americans destroyed an estimated three hundred Japanese planes.

Wednesday, June 21, 1944

At 8:00 a.m. I held a meeting of the Council of my section, attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Heath, Hindman, Jensen, Katzin, Manning, Simpson, Studier, Thompson, Watt, and Willard. Safety rules and cleaning schedules were discussed. The Council decided the "no smoking" rule is to go into effect immediately. Studier is to prepare a memo concerning cleaning to be circulated to all Section C-I employees. Dawson expressed concern about the high plutonium losses occurring in the section.

A sample of what is thought to be neptunium oxide has been sent by La Chapelle and Magnusson to Zachariasen for x-ray analysis.

Jaffey has observed two distinct alpha-particle plateaus from a plutonium source when counted in his alpha-range differential ionization chamber. The plateau at the higher gain is interpreted as coming from the lower energy back-scattered alpha particles. There seems to be at least a 3% difference in the counting rates for the two plateaus. This agrees with a lower limit of 3% for alpha-particle backscattering suspected earlier.

A memo from Tepe was received by Dreher concerning a request that copies of all meeting notes and other information pertaining to the ether extraction process be sent to Leverett at Clinton. A specific request has been made by Leverett for information on the effect of calcium nitrate on the distribution of uranyl nitrate between water and ether solutions.

The report for the month ending June 1, 1944, (CK-1701), by Hogness, Seaborg, Manning, and Orlemann, entitled: "Chemical Research — Special Chemistry of Plutonium," was issued today. It includes the following information:

a. Purification of Plutonium Compounds Group (Jensen, Group Leader). (1) Work by Jensen and Reinhardt involving tracer-scale experiments on the salting out of plutonium from 10 M NH_4NO_3, 1 M HNO_3 into various organic solvents is reported. Diethyl sulfide, benzyl "cellosolve," and menthone extract useable amounts of Pu(VI).

b. Volatility and General Dry Chemistry Group (Davidson, Group Leader). (1) Five projects under the problem assignment "Preparation and properties of plutonium chloride," are reported, including: Karle's work on the preparation of $PuCl_3$ using CCl_4 vapor; Katz's work on "wet" preparation techniques for $PuCl_3$; Davidson's liquid phase chlorination of plutonium oxide; Hagemann's liquid phase chlorination of $PuCl_3$; and Brody's preliminary study of the hydration of $PuCl_3$ and of the formation of PuOCl. Brody's work resulted in a compound that Zachariasen has determined to be isomorphous with $NdCl_3 \cdot 6H_2O$. The melting point of several hexahydrated samples was observed to be 94°-96°C.

(2) Work by Hagemann and Hyde is reported on the hydrobromination of $PuO_2 \cdot xH_2O$, plutonium oxybromide, and the preparation of pure $PuBr_3$ from the metal.

(3) The work by Hagemann on the preparation and properties of plutonium iodides is described. Plutonium treated with iodine vapor at 400°C in a quartz capillary forms a green PuOI. Plutonium metal treated with liquid I_2 at 250°-300° forms a light brown product consisting mostly of PuOI. The source of oxygen in these two reactions is not understood at present.

c. Fluorine Chemistry Group (Heath, Group Leader). (1) Work by Florin, Meyer, and Heath is reported concerning projects under two problem assignments: "The hydrofluorination of plutonium oxide," and the "Preparation of plutonium halides for metal production." The action of O_2 on PuF_4 has been studied, and it has been determined that in the absence of HF, oxygen completely converts milligram quantities of PuF_3 to a green PuO_2 in two hours at temperatures above 325°C; at 275°-300°C partial conversion occurs; but at 225°-250°C, no conversion was determined.

Satisfactory PuF_3 has been prepared by both the "wet" and dry methods. In the dry method Pu(IV) nitrate is ignited at 700°-800°C yielding dense PuO_2. The PuO_2 is converted to PuF_3 using HF and H_2 at 600°C. The wet method is preferred because lower temperatures are required and reagents can be more easily purified, therefore yielding a higher purity product. In the wet method, a HCl solution of Pu(IV) is reduced with SO_2; PuF_3 is precipitated by adding HF; digestion of the PuF_3 in contact with the mother liquid is used to increase particle size. The resulting PuF_3 has been dehydrated in streams of HF at temperatures ranging from 150° up to 550°C, PuF_3 dried at 300°C having been found to give the best reduction to metal.

(2) Work by Zvolner on the chemical composition of plutonium halides is reported. His work shows that a higher fluoride can be made by the action of F_2 on PuF_4 at 600°C which can be revolatilized in N_2 at 300°C.

d. Production and Properties of Plutonium Metal Group (Baumbach, Group Leader). (1) As plutonium metal samples to be studied have increased in size recently, regular Pyrex capillary tubing, of 0.1 to 1 mm bore, is now being used for liquid displacement measurements in the determination of plutonium metal density. An accuracy of ±0.1 density units can be achieved. Measurements on six plutonium metal samples of various origins have given densities ranging from 13.6 to 19.3 gm/cm^3; values for four of these were between 16.6 and 17.6 gm/cm^3. The 19.3 gm/cm^3 density material was obtained by Westrum who used barium reduction of PuF_3 in La_2O_3 crucibles with 1,200°C firing.

(2) Fried is now using a gold cup to contain plutonium metal samples studied to determine the dissociation pressure/temperature relationship for plutonium hydride. The gold cup prevents the sample from contacting the glass walls of the apparatus. Plutonium metal in the amount of 0.264 mg absorbed 34.6 mm^3 of hydrogen at room temperature, corresponding to a formula of $PuH_{2.8}$ for the hydride.

(3) Frank has continued studies on the hardness of plutonium metal. An Eberbach microhardness tester with an indenting force of 49.5 grams was used. Hardness values made on 17 plutonium samples of various origins range from 57 to 298.

(4) Jasaitis has run a melting point determination on a 6-mg plutonium sample and has obtained a value of 700°C. Although the purity of the melted sample has not yet been ascertained, the true melting point for plutonium is probably lower than the 810°C value reported earlier.

Jasaitis has remelted this same plutonium metal sample on a nitrided tantalum foil at 1,200°C and held it in its molten state for 10 minutes at 800°C. The cooled plutonium button has a very thin black layer of what is thought to be PuN where it contacted the TaN surface. The plutonium metal density changed from 17.2 gm/cm^3 before remelting to 16.7 gm/cm^3 following remelting. Metallographic examinations have not yet been made.

(5) Karle's work shows that PuF_3 is initially obtained from the treatment of PuF_4 with atomic hydrogen, but upon further treatment a material consisting of PuO_2, PuF_3, and probably PuOF is formed.

(6) Work by Hellman, Frank, and Gerstein on tests of various refractories is reported.

(7) S. Katz reports on the spectroscopic analysis by Section C-IV of 15 samples of plutonium metal; no impurities except beryllium were detected.

(8) Karle's work concerning the production of plutonium by thermal bomb reduction is described. Tests of performance are being conducted using 100 mg samples of UF_4.

e. High Vacuum Techniques (Simpson, Group Leader). Simpson is testing the entire "Y" designed micro-oxygen analysis system using CuO samples first, to be followed with PuO and uranium samples.

Helen worked at the Met Lab in the afternoon.

Katzin, Ghiorso, and I played nine holes of golf at Jackson Park after work while it was still light. Our scores were LK — 70, AG — 57, GS — 54.

Al Ghiorso left by train for Site X this evening at 11:30. He plans to return on June 25 at 7:00 p.m.

The Russians have captured Viipuri (a Finnish seaport city), Marshal Stalin announced tonight.

There was a Project Council Policy Meeting at 9:00 a.m. in Room 209 Eckhart Hall, attended by Chapman, Chipman, Compton, C. M. Cooper, Dempster, Doan, Eastman, Franck, Greager, Hilberry, Jeffries, Johnson, T. R. Jones, Latimer, Colonel Lansdale, J. H. McKinley, Mulliken, A. V. Peterson, Spedding, Stearns, Stone, Szilard, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. After opening remarks by Hilberry, Cooper presented a "Summary of Canning Situation." He said as of June 20 about 10,000 usable Al-Si slugs have been produced with a current production rate of 1,500 per day, of which 70% are acceptable (as compared with an acceptance percentage of only 30% just two weeks ago). W. W. Watson was assigned the responsibility to develop a slug "swell" detector with Cooper, Franck, and Wigner as his advisory committee and Creutz as coordinator.

Compton, after having been detained elsewhere during the early portion of the meeting, presented Allison's report of discussions at Site Y with Oppenheimer concerning the question of contamination resulting from military use of plutonium — Oppenheimer and others at Site Y have given this active consideration and will be responsible. Compton brought before the Project Council the problem of maintaining security. He said Chicago's reputation is not the best and introduced Colonel Lansdale, who discussed security in light of accomplishing both scientific as well as war objectives. He said the need to accomplish a job speedily has made security especially difficult to maintain. In general, it has been necessary to make many decisions in favor of speed rather than security. Compton said it is the responsibility of all to do everything possible, especially during the next few months, to maintain the best possible security. He requested each Division head and the Administrative Chief within the next 24 hours to get across to their employees the need for utmost security and to report back to him.

Thursday, June 22, 1944

At 11:00 a.m. we received word from Zachariasen that the structure of the neptunium sample prepared by Magnusson and La Chapelle day before yesterday is definitely the compound NpO_2. This date marks the first positive identification of a neptunium compound. Zachariasen finds the NpO_2 to be isomorphous with PuO_2 and UO_2 with properties intermediate between these two compounds. The density is calculated to be 11.14 gm/cm^3. Zachariasen believes that this isomorphism is evidence of a new rare earth series involving "f" electrons.

At my request, Watt today sent a summary to Hogness of the opinions arrived at during the June 14 meeting of the Council of Section C-I concerning the "Contracts of Employment" and "Transportation and Moving" agreements.

Stephen Lawroski is in Chicago; Hogness, Daniels, and I met with him today. We told him about the problem of purifying a new material from light elements to a high degree and how we would like him to take leave from Esso Laboratories (Standard Oil) in New Jersey to help our war related project. Earlier this month, Standard Oil agreed to the loan of Lawroski to assist us in solvent extraction work if he should wish to make the change.

I attended the meeting of the Basic Chemistry, Recovery, and Instruments Groups of my section which met at 7:30 p.m. in Room 209 of Eckhart Hall. Others in attendance were Ames, Arnold, Cunningham, Davidson, Dawson, Dixon, Dreher, Fields, Florin, Hagemann, Jaffey, Katzin, Kohman, Kraus, La Chapelle, Magnusson, Manning, McLane, Meyer, S. Peterson, Phipps, Scott, Smith, Studier, Thompson, Warner, and Willard. Cunningham spoke a word of caution concerning the handling of plutonium and stressed the need for workers to wear protective clothing. Face shields and rubber gloves should be worn whenever there is danger of fuming, spraying, or explosions; and respirators must be worn whenever dry material is to be handled.

Studier of the Recovery Group reported on important extraction process developments in plutonium recovery, isolation and purification. The old method, which involves successive fluoride cycles followed by peroxide precipitations, has been almost completely abandoned in favor of ether extraction. The procedure now used involves the lanthanum fluoride precipitation of plutonium, dissolving the precipitate with $ZrO(NO_3)_2$, oxidizing to Pu(VI) with $KMnO_4$ (or other suitable oxidizing agent) followed by ether extraction in the presence of 10 M ammonium nitrate and approximately 2-3 M nitric acid. The aqueous phase is frozen, and the ether phase with plutonium is poured onto distilled water to extract the plutonium from the ether. The water phase with plutonium extract is frozen, and the ether is removed for reuse in a second extraction. After two ether extraction cycles, nearly all impurities are removed below limits of detection of the spectrograph. Three extraction cycles with the ether volume twice that of the aqueous solution are sufficient to extract 95% of the plutonium.

Other solvent extraction methods are being considered, such as the use of hexone (methyl isobutyl ketone) for the extraction of Pu(IV).

A rather promising counterflow extraction apparatus is being built by Stewart. In response to my inquiry, Studier said that preliminary experiments have been made on the hexone extraction of solutions of bismuth phosphate that show that 45% of the plutonium is extracted when using equal volumes of hexone and bismuth phosphate solutions. The bismuth tends to be extracted by hexone.

Kohman described his recent studies on the aggregate recoil (ejection of minute particles) of plutonium. A clean collection plate was mounted 2 mm above the surface of a plutonium sample. After two days many alpha-particle counts per minute were measured on the collection plate. In another experiment with a 3 mm spacing, alpha-particle counts of the plutonium collected on the collector were measured three times over a 6-day period and seemed to indicate that the amount of plutonium deposited per unit time decreases with time. If the phenomenon of aggregate recoil is real, it could be a source of contamination and could cause errors in sample counting results due to loss of plutonium.

La Chapelle reported on the isolation of about 80 micrograms of Np^{237} from process solutions of Site X pile slugs and from 64 pounds of specially-irradiated uranium bombarded with neutrons at the Berkeley cyclotron. About 35 micrograms of Np^{237} were recovered from the supernatants from peroxide and iodate precipitations of Site X pile material and about 40 micrograms of the 72 micrograms originally calculated to be present in the Berkeley-bombarded uranium were isolated by means of ether extraction, followed by successive bromate cycles to separate the Np^{237} from the Pu^{239}. The neptunium thus isolated was combined, and two more bromate cycles were carried out. The hydroxide was ignited at 700°C to obtain the oxide. Today Zachariasen identified this oxide as NpO_2.

Ames reported on his investigations concerning the deviation from Beer's law exhibited by Pu(VI). He believes the cause may be instrumental reasons. He finds that a change in nitric acid concentration profoundly alters the amplitudes and positions of absorption peaks. He also believes that changes in the type of acid used can cause similar changes. Hindman made an attempt to explain the mechanism of oxidation that is causing the formation of Pu(VI) during the extraction step in the Bismuth Phosphate Process. The effect of nitrite was discussed. During HNO_3 oxidation, nitrous acid is rapidly formed tending to reverse the reaction. Calculations of the equilibrium constant show that reversal will occur with very small nitrous acid concentrations.

Joy Townsley visited us. Later we had dinner at Morton's Tavern at 54th Street and South Lake Park.

Today's headlines indicate that carrier planes have opened a naval battle midway between Luzon and Saipan and that Allied forces have made further progress in the battle for Cherbourg.

Friday, June 23, 1944

Ralph James completed the twelfth bromate oxidation cycle on the deuteron-bombarded uranium target he started processing on June 6 in a search for new neptunium isotopes. Alpha-particle activity measurements were made on the sample, and it appears that about 200 counts per minute are present due to what may be alpha-particle-emitting Np^{235} or Np^{236} isotopes.

The first photographs, in color and black and white, were taken of the neptunium compound, NpO_2, using the material just isolated by Magnusson and La Chapelle. One of them is shown in Fig. 6.

A. A. Frost of Northwestern University sent me additional information about the lecture on "Isotopic Tracer Techniques" I have been requested to give. The date has now been advanced one day to Tuesday evening, December 5. Frost says he hopes to hear of my acceptance in the near future.

Thompson sent a memo to Albaugh, Dreher, and Pye concerning problem assignments in the separation processes area. He summarized an earlier memorandum by Kirst, of June 16, which contains many suggestions and recommendations concerning studies on the effect of aluminum from Hanford slugs on the extraction process, the storage of metal and crude plutonium solutions, the reduction of cell and equipment decontamination, the loss of plutonium in lanthanum fluoride by-product precipitates due to stainless steel reactions in the presence of HF, and the refinement and improvement of the isolation step. Kirst points out in his memo that no further work is required on compounds suitable for storage and shipment.

Dreher sent a memo to Ashcraft about a method devised by Blaedel of our section and G. W. Cressman of the Engineering Development Group, Technical Division, for determining free nitric acid present in solutions of uranyl nitrate. Details of the procedure, as developed for use in our own process development work, were enclosed with the memo on a separate sheet. Since this method may offer definite advantages in routine analytical work where accuracies need not be greater than 5%, it was suggested that Ashcraft may wish to evaluate the method and, if he finds it satisfactory, possibly authorize it as a standard method.

Helen worked at the Met Lab in the afternoon. Wilma had dinner with us in our apartment this evening since Albert is at Site X.

Encirclement of the fortress of Cherbourg is now almost complete, according to an Allied headquarters bulletin.

Hilberry initiated the first "Director's Meeting" this morning. Persons present were C. M. Cooper, Hilberry, Hogness, L. O. Jacobson, Kimpton, Vernon, W. W. Watson, and Wigner. The major items discussed were security, our relations with the Montreal Laboratory, handling of plutonium, New Chemistry Building machine shop facilities, laboratory coat needs, and completion of

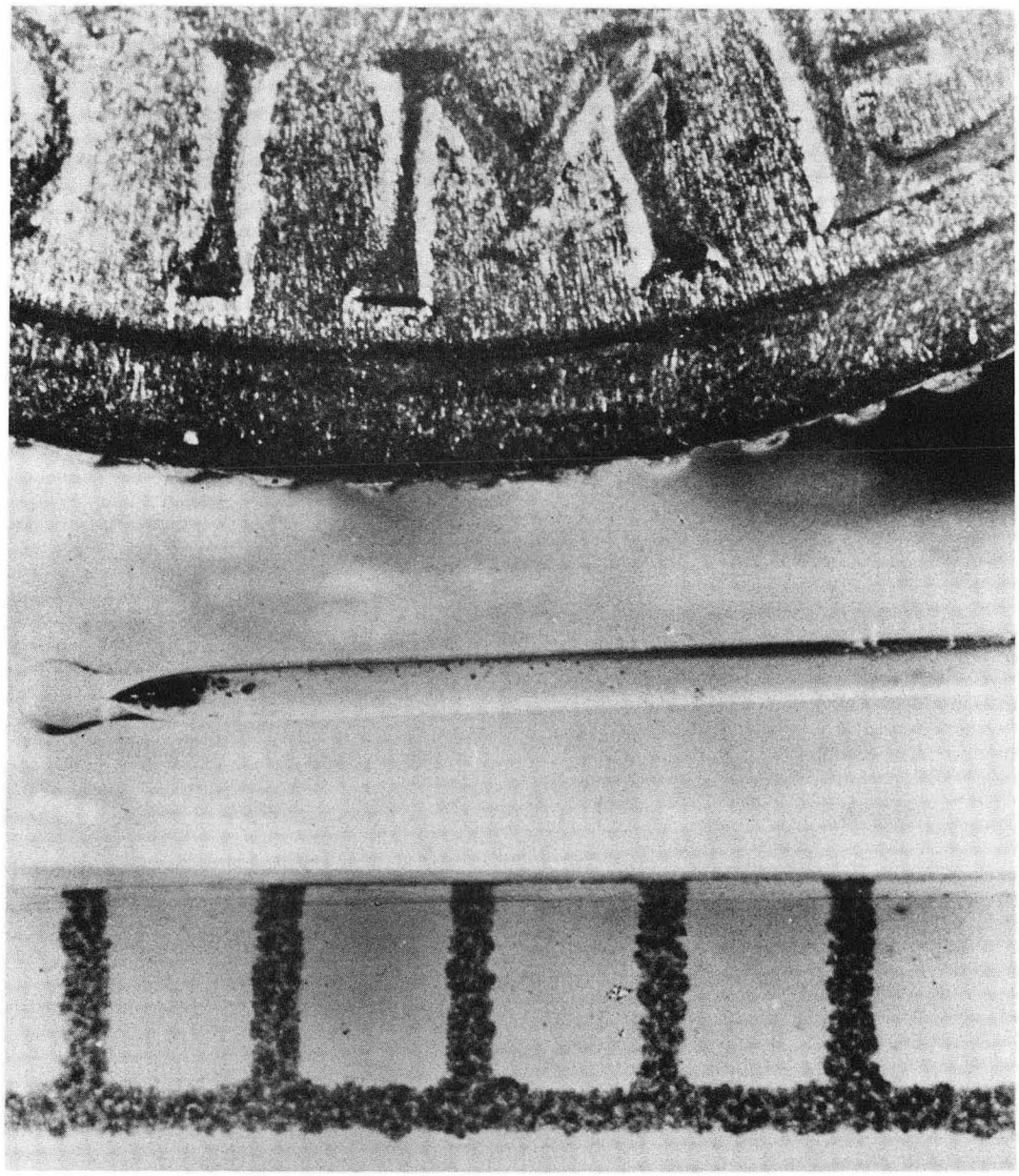

Figure 6. Neptunium oxide, NpO_2, isolated June 20, 1944, appears at the bottom (left end) of the horizontal capillary tube. The sample weighed approximately 10 micrograms. Below is shown a millimeter scale and above a U.S. dime for purposes of comparison.

CHEM 2517

the Drexel House for Health Division use. In a memo to the participants, Vernon questions the desirability of issuing minutes for future "Director's Meetings," except where specific action assignments need to be recorded.

Saturday, June 24, 1944

In the evening Helen and I went to the Tivoli Theater, Cottage Grove at 63rd Street, to see the movies "Cover Girl," featuring Rita Hayworth and Gene Kelley, and "Chip Off the Old Block" with Donald O'Connor, Joel Kupperman, and Peggy Ryan.

The Soviets have announced a big offensive against the Nazi armies on the Eastern front, gaining up to nine and a half miles and liberating more than 150 towns.

Sunday, June 25, 1944

I played 18 holes of gold with Howland, Arnold, and Thompson at the Evergreen Golf Club. Our scores were JH —110, LA —105, ST —115, and GS — 110.

Helen had lunch at Wilma's. Al is scheduled to return from Clinton this evening.

Monday, June 26, 1944

Rexford Hale Bradt transferred from Section C-II to Section C-I today and will work in Albaugh's group. We also hired a new secretary, Kathryn A. Buehler. Laura Boyd, a clerk in Ghiorso's group, transferred to Site B.

In a memo to Allison and Vernon about the design requirement for twelve-foot stainless steel laboratory hoods for Hanford, Willard refers to an earlier memo of May 12, concerning suggested changes to be made in the drawings for a four-foot hood which should also apply to the twelve-foot hood.

Hogness wrote to Moulton about my reluctance to permit the transfer of Joe Katz from his present work in Section C-I to the Patent Office. Hogness informed Moulton that Katz is working on a high priority problem and is the "best man in his group, and, while he doesn't know this himself, he very probably will be made a group leader very shortly." This work is considered so valuable to us that his loss would impair our work. Hogness said he will keep his eyes open for another man with suitable qualifications for the Patent Office position.

Report No. CN-1702 for the month ending June 1, 1944, entitled "Chemical Research — Basic Chemistry of Plutonium," was issued today. It includes the following information of interest:

a. Basic Chemistry Group (J. C. Hindman, Group Leader).

(1) James and my work on heavy isotopes by the bombardment of Pu^{239} is briefly noted in the abstract. Following a six-week neutron irradiation of a Pu^{239} sample in the Clinton pile, James detected no long-range alpha particles. A short bombardment of a second Pu^{239} sample with cyclotron-produced deuterons counted immediately afterwards, did not show the presence of any alpha activity with a half-life between 1 minute and 30 days. Material which has been given a longer deuteron bombardment was repurified and its specific activity determined. It was found to be no different from that of an unbombarded control sample. Various proposed methods of detecting element 95 in bombarded material have thus far failed to show its presence.

(2) Hindman has made oxidation potential measurements of the formal potential (at equimolar concentrations) of the Pu(III) = Pu(IV) couple in 1 M HNO_3, using electrolytic reduction to obtain various ratios of Pu(III)/Pu(IV). The formal oxidation potential is found to be -0.916 ± 0.006 volts at about 25°C.

(3) O'Connor has made solubility measurements which show that Pu(III) and Pu(IV) phosphates become, respectively, at least 3 times and 1.5 - 2.0 times more soluble in 1 M H_2SO_4 when temperature is increased from 25° to 75°C. Further work on the solubility of Pu(IV) phosphate in various concentrations of HNO_3 containing 0.03 M $Fe(NO_3)_3$ shows that the rate of precipitation is decreased by Fe(III), the final equilibrium being reached in 10 days as compared with one hour in the absence of Fe(III). The Pu(IV) phosphate solubilities at equilibrium increase by 8-10 times with 0.03 M $Fe(NO_3)_3$. Iron present in the process solutions would markedly increase the plutonium(IV) phosphate solubility to a point well above process plutonium concentrations. O'Connor's work on the precipitation of plutonium(IV) oxalate shows that as long as 1 M HF is present in the crossover step in the mainline process, precipitation cannot occur at Hanford product concentrations.

(4) O'Connor has determined, with x-ray analysis assistance from Zachariasen and Mooney, that plutonium(IV) oxalate is isomorphous with uranium(IV) oxalate. In concentration procedures using uranous oxalate as a carrier, the mechanism of co-precipitation probably involves isomorphous incorporation of plutonium into the uranous oxalate crystal lattice. Evidence has been obtained which shows that carrying of Pu(IV) by bismuth phosphate does not appear to involve isomorphous incorporation of the plutonium into the bismuth phosphate lattice.

(5) Smith's attempts to prepare a crystalline double salt of lanthanum fluoride and plutonium(IV) fluoride have not been successful.

(6) Kraus has investigated methods for the preparation of a pure plutonium hydroxide from which pure solutions can be made by solution of the hydroxide in an appropriate pure acid. The hydroxide produced still has about 0.1 mole sulfate per mole of plutonium, and thus will require reprecipitation before using for high purity work.

(7) Ames has studied rates of plutonium oxidations and reductions. Sulfuric acid can stabilize Pu(IV) solutions against oxidation by hot nitric acid, and Pu(IV) in 1 M HNO_3 is completely reduced to Pu(III) by SO_2 within one minute at 25°C. When he uses 1 M H_2SO_4 with SO_2, more than 48 hours are required to complete the reduction to Pu(III) at room temperatures.

(8) In studies related to developing methods for the assay of Site W plutonium solutions, Ames finds that Pu(III) in 1 M H_2SO_4 has principal adsorption bands at 560, 600, 664, 810, and 905 millimicrons. He is continuing his study of the variations of optical density of the Pu(III) absorption bands as a function of plutonium concentrations.

b. Recovery and Service Group (L. R. Dawson, Group Leader).

(1) Asprey, Britain, Burr, Fineman, Leventhal, Stewart, Studier, and Wetlaufer have continued work on the recovery of plutonium. During the month a substantial amount of plutonium has been recovered and purified. In most cases extraction with ether has been the final step in the process and has yielded product that is at least 99.5% plutonium. The plutonium has been recovered from such material as spectrographic equipment, washings from a centrifuge cup, concentrated sulfuric and nitric acid solutions, thoria and beryllia crucibles, and solutions containing a variety of other elements and activities.

(2) Asprey, Leventhal, Stewart, Studier, and Dawson have directed their research on methods for plutonium recovery toward improvements in the oxidation step for obtaining Pu(VI) prior to extraction with ether. The oxidation state of plutonium extracted into ether and the partitioning of Pu(IV) between ether and 10 M NH_4NO_3 - 1 M HNO_3 have also been studied. They have used 3 N HNO_3 successfully to oxidize Pu(IV) to Pu(VI) but find it ineffective for oxidation in a solution containing large quantities of zirconyl ion; Pb_3O_4, however, was determined to be a very satisfactory oxidant in the ether extraction work.

(3) Asprey, Stewart, Studier, and Dawson have determined the aqueous solubilities of plutonium(VI) nitrate and plutonium(VI) chloride to be 500 gm of plutonium per liter and 350 gm of plutonium per liter, respectively, at room temperature. Attempts to prepare sulfides of Pu(IV) and Pu(VI) have failed. Treatment of a weakly acidic solution of Pu(VI) with $(NH_4)_2S$ produces a gray-brown precipitate that changes to greenish-brown upon standing over night. After washing, the solid gives a positive test for sulfide ion. At present the compound has not been identified.

(4) Fields is now using the Pu^{239} recovery extraction method for the separation of Np^{239} from active UNH. An initial separation from uranium is made by lanthanum fluoride precipitation. Approximately 6×10^{10} Np^{239} beta-particle disintegrations per minute have been supplied each week for neptunium research.

Helen worked at the Met Lab on my classified "Table of Isotopes" in the morning and shopped downtown in the afternoon.

American troops are in Cherbourg, according to the paper today.

Tuesday, June 27, 1944

Ralph James made alpha-particle absorption measurements on the "93-fraction" and the "94-fraction" resulting from the thirteen bromate cycles he has completed on the deuteron-bombarded uranium sample. He finds evidence in the 93 fraction for the existence of short-range alpha particles, as well as for what are thought to be alpha particles from Pu^{238}. He estimates that 80% of the alpha particles could be from a new neptunium isotope. He was somewhat handicapped in taking the absorption measurements by an erratic instrument performance, probably caused by the high ambient temperature in the counting room, which reached 97°F before he finished with the measurements. He has decided to carry out another bromate cycle to eliminate more of the Pu^{238} from the 93 fraction.

Pye pointed out to Thompson that, according to his calculations, even 50 ppm of trace iron in the KOH used for metathesis causes a marked decrease in the plutonium yield. He suggests that if his calculations are correct, this matter should be investigated as a definite source of trouble.

Thompson wrote a memo to Watt concerning a program initiated by Willard to obtain aluminum-silicon bonded, aluminum-jacketed uranium metal slugs with and without Hanford-level inactive fission product elements being present. These slugs are to be irradiated in the Clinton pile so that tests of the effects of the slug coatings and Site W fission product levels on the Hanford process can be made prior to the beginning of operations at Hanford. In addition, a 10-gram or larger sample of uranium metal containing both plutonium and fission product elements at Hanford concentrations was requested. This piece would be placed in an aluminum-silicon bonded aluminum jacket and, following irradiation in either the Argonne or Clinton pile, would be subjected to a complete process run to determine if there are any unforeseen factors in the process that might give trouble.

Thompson wrote Squires about problems that might arise with his leave of absence from Standard Oil if he were to transfer to Hanford this fall. He requested a contract termination date of May 1, 1945, to phase in better with his plans.

Norman Hilberry described for Captain J. H. McKinley of the Manhattan District the basic considerations upon which recommendations for remodeling of the north wing of the New Chemistry Building were based. He wrote,

"These modifications were projected to meet two objectives; namely, reduction of the health hazard to safe limits on work with the product and its compounds; and provision of filtered air with refiltered air for a few laboratories to reduce or eliminate contamination of the product compounds and the product in research on the purification process.

"To accomplish these objectives, additional hood facilities over laboratory benches have been provided with an air velocity at the openings of 100 linear feet per minute, in accordance with the specifications of the Health Division. Furthermore, a filtered air supply for the entire section of the building has been provided. All laboratories have been dustproofed by sealing cracks in the walls, by removing odd corners and the like where dust might collect, and by painting the walls white and covering the floors with linoleum to make cleaning easier. This section of the building will be separated from the rest of the building and from the outside by pressure airlocks. Locker facilities have been provided where the personnel can leave their outer street clothing and their street shoes when they change to clean shoes for use in the laboratories.

"Facilities have been provided for a group of chemists called Radiation Protection Group whose duties will be to survey all sections of the laboratory for radiation hazards and also to survey personnel."

Helen spent the evening at Wilma's.

Domestic and war news compete on the front page today. The Republican Party convention has convened and U.S. troops are in control of Cherbourg, although pockets of resistance continue.

Wednesday, June 28, 1944

The weekly meeting of the Council of Section C-I took place in my office at 8:00 a.m. In attendance were Albaugh, Cunningham, Davidson, Dreher, Ghiorso, Heath, Hindman, Jensen, Katzin, Manning, Orlemann, Simpson, S. Thompson, Watt, and I. I asked if there were any objections to changing the regularly scheduled Thursday night sub-section meetings back to Wednesday evenings. A canvas of personnel will be made to see if this will meet with general approval. Manning suggested that a qualified person should be put in charge of the cleaning program. This person should effectively coordinate this program with that of the Radiation Protection Group. It was also noted that canvas shoe covers will soon be available for use in the New Chemistry Building. I mentioned again the need to be certain that all secret material is kept locked up and that any misplaced secret reports in our files be searched for and located.

"Chemical Research — Separations Processes for Plutonium. Process Development and Chicago Semi-Works Operation," (CN-1751), for the period ending May 15 was issued today. It was prepared jointly by the Chemistry and Technical Divisions. Activites reported for Section C-I are as follows: Hyman, R. T. Makins (Technical Division), and Winner continued work on the removal of bonded slug jackets and have studied the rate of dissolution of aluminum in nitric acid solutions containing mercuric nitrate and the rate of dissolution of uranium in nitric acid and in sodium hydroxide coating-removal solutions.

Margolis and Blaedel have studied reduction of dichromate and plutonium(VI) during ether extraction and the equilibrium distribution of plutonium(VI) and uranyl nitrate between aqueous and ether layers containing nitric acid and sodium dichromate.

Foster York had dinner with Helen and me. Helen and I then visited Walter and Walborg Eggen in Evanston later in the evening. Ed and Lala Cuyler (a first cousin of mine) were also at the Eggen's. Walborg Eggen, who is Lala's half sister, served coffee, Wheatie cookies, etc., on the back porch.

The Republican convention adopted its platform, and indications are that Thomas Dewey (Governor of New York) will be their candidate for the presidency.

Thursday, June 29, 1944

The results from twenty-one 1,000-liter air sample assays taken by the Radiation Protection Group in ten rooms of the New Chemistry Building gave very low alpha-particle counts, with the exception of a sample collected within the hood in Room 27 where 200 mg of plutonium peroxide precipitate is located and where Goeckermann, Beard, and Hopkins work.

I attended the meeting of the Separation Processes Sub-section of Section C-I at 7:45 p.m. in Room 209 of Eckhart Hall. Others present were Ader, Albaugh, Arnold, Beard, Blaedel, Dreher, Goeckermann, Greenlee, Haeckl, Hoekstra, Hyman, Katzin, Larson, Malm, Manning, Morgan, S. Peterson, Pool, Post, Pye, Rasmussen, Templeton, R. Thompson, S. Thompson, Walling, Watt, Winner, and others. With regard to extraction-decontamination work, Albaugh reported on the use of cerium phosphate-zirconium phosphate dual scavengers to remove columbium and zirconium fission product contaminants, which normally persist through two bismuth phosphate cycles to the extent that the desired decontamination factor of 10^5 cannot be reached. Two series of experiments, using Site W concentrations of plutonium and fission product elements, have been made with reasonable success. Tests on the semiworks scale have been or are being arranged at Clinton and at Chicago.

Stan Thompson reported on work conducted largely at Clinton Labs that is directed toward improving decontamination in the bismuth phosphate-plutonium precipitation steps by complexing certain fission product elements with HF or H_2SiF_6.

With regard to concentration-isolation work, Goeckermann described attempts to improve the isolation yield from Hanford plant process solutions. Chemical and spectrochemical analyses have strongly indicated that the presence of zirconium increases the solubility of plutonium peroxide so that excessive losses occur in the peroxide precipitation. To reduce the inhibiting action of zirconium, HF was added to the $La(NO_3)_3$ product solution to complex the zirconium and thereby reduce its inhibiting action. It was determined that a HF concentration corresponding to a 6 to 1 mole ratio of fluoride to zirconium gives optimum conditions; and plutonium peroxide yields of around 98% are obtained from plant $La(NO_3)_3$ solutions containing as much as 2 g Zr(IV) per liter.

Goeckermann also described the detailed procedures devised for a second method of removing or reducing the effects of the interfering agents in peroxide precipitation. This procedure consists of a lanthanum fluoride "clean-up cycle." Lanthanum fluoride by-product and then potassium plutonium fluoride precipitations are made starting with the $La(NO_3)_3$ plutonium solutions obtained from metathesis and dissolution of the "final" Clinton lanthanum fluoride plutonium precipitate. Solution of this potassium plutonium fluoride by metathesis with $KOH-K_2CO_3$ and the action of HNO_3 allows the precipitation of plutonium peroxide with yields of 98%.

Beard reported on the uranous oxalate concentration and isolation method, devised as an alternate process for possible use at Hanford should the mainline process fail. The principal advantages of the method are that it involves no use of HF or H_2O_2 and requires fewer steps than the mainline procedure. Beard pointed out several apparent disadvantages, however, such as the lower concentration factor, the need to standardize the uranous sulfate solution prior to each use, and the large excess of sulfate needed to complex the uranyl and prevent precipitation of uranyl oxalate in the isolation step.

In discussing process development work, Rasmussen reported on the status of the $NaNO_2$ prereduction studies. Our Chicago experience has shown that oxidation is easier and reduction more difficult at Hanford plutonium concentrations as compared with studies made at tracer concentrations. It is therefore necessary to consider the use of stronger reducing agents than formic acid for the prereduction step. This need seems to be answered by $NaNO_2$, and experimental evidence indicates that the $NaNO_2$ prereduction procedure devised for use at Hanford is capable of reducing Pu(VI) to the extent of 90-95% in one hour.

Blaedel described recent investigations of the ether extraction procedure directed toward increasing efficiency of plutonium recovery and improving decontamination. $Ca(NO_3)_2$ produces a significant reduction in solubility of uranyl nitrate and Pu(VI) in the aqueous phase and is being used to "salt out" these materials from the aqueous phase into the ether phase, thereby increasing the efficiency of product extraction. Operating conditions for a continuous liquid-liquid extractor employing this method were formulated by Blaedel and are as follows:

Raw ether would be introduced at the bottom of the column, the ether extract would be withdrawn from the top of the column, and the aqueous raffinate would be discharged from the bottom of the column.

Further, an aqueous phase containing no $Ca(NO_3)_2$ would be introduced at the top of the column, so that the column would comprise essentially of a liquid-liquid extractor in series with a water scrubber. Sufficient 5 M $Ca(NO_3)_2$ solution to bring the aqueous reflux to 3 M in $Ca(NO_3)_2$ would be added at a point just above the point of introduction of the charge. By suitable control of flow and feed rates, appreciable decontamination could be obtained by virtue of the comparative distribution ratios of fission and product elements (FPE) and plutonyl between ether and water, i.e., a significant portion of the beta and gamma-activity would be removed in the aqueous phase in the scrubber section of the column while substantially all the plutonium and uranyl nitrate would be retained in the ether extract. The operating conditions which have been proposed would, in a highly efficient column, remove all FPE with distribution ratios greater than 3. Decontamination factors of 500 - 5,000 are predicted for such operation. Additional decontamination could be achieved by a second column which could be operated in an analogous manner. It is believed that the most important problem involved in the operation of such a column would be that of heat exchange. Alarming heat effects have been observed in laboratory conditions.

Paul Aebersold, a physicist whom I knew in Berkeley, had dinner with us at home.

The GOP convention has taken the headlines from the war; Dewey and John W. Bricker, Governor of Ohio, will head the Republican ticket.

Friday, June 30, 1944

Stephen Lawroski was hired to work in Section C-I today. He received a Ph.D. degree from Penn State last year and has been employed by the Esso Laboratories of Standard Oil Development Company, Elizabeth, New Jersey.

I sent a memo to Vernon in regard to the future Chicago Chemistry Division program for shipment of slugs from the Clinton pile in light of the planned increase in operating power level of the Clinton pile. I make reference to my January 15 memo of this year which stated our earlier anticipated requirements for Clinton slugs and the desired minimum product concentrations and maximum penetrating leakage of radiation levels. I request that we continue to receive 24 slugs per week that have been sufficiently irradiated and cooled to give about 50 micrograms of plutonium per pound of metal. To continue to receive material of the same quality as we have been receiving will, of course, require either shorter irradiation times or a less favorable position in the higher power pile.

In addition to the above slugs, we have been receiving one "special" slug about every two weeks that has been irradiated for 100 days and cooled for about 45-70 days. These slugs contain about one mg of plutonium per pound. Any higher product concentrations resulting from the increase in pile power will not be objectionable, and I request that we continue to

receive these "special" slugs and that the 100-days irradiation time be maintained. I point out that we are now capable of handling shielded slug containers weighing as much as one ton, but recommend that we continue the use of the present containers, which we believe will provide adequate shielding for future slugs.

Helen worked at the Met Lab in the afternoon. I played 9 holes of golf with Stan Thompson at Jackson Park after work. Our scores were ST — 48, GTS — 47.

Headlines today come from the European fronts. The Russians are advancing and captured two cities; and the British, five miles southwest of Caen, are expecting a big German counterattack. The Germans are drawing reserves from Germany for the first time since D-Day.

Saturday, July 1, 1944

Quentin Van Winkle started work under Dawson in the Recovery Group (Group 8) as a Junior Chemist. He has been a graduate student and research associate at the University Engineering Experiment Station of Ohio State University.

Arthur Baer and Beatrice Foreman have terminated their employment in Section C-I. Baer plans to return to school. Robert Young transferred to the Ryerson shop.

Helen had lunch with Wilma. Frances Chilson spent the night with us.

Today's headline from the Pacific indicates that nearly ten thousand Americans have been killed or wounded or are missing in action from a fortnight of bitter fighting in Saipan.

Sunday, July 2, 1944

Helen, Frances and I went to a golf tournament on the North Side at the Edgewater Golf Club; Harold "Jug" McSpaden tied Ben Hogan by sinking a long putt on the last hole. After this we went to Frances' apartment on the north side for dinner. By this time I had developed a migraine headache.

Monday, July 3, 1944

I sent to Hogness a 35-page summary (MUC-GTS-820) of the work of the past month on separation processes that took place in Section C-I. It will be issued as a CN Report later this month. Investigations reported on are the following:

Extraction-Decontamination (Albaugh, Group Leader). Gilbreath conducted studies that show that an oxidation treatment with dichromate prior to the prereduction step could be a means of eliminating extraction losses that might be caused by the presence in the metal solution of interfering foreign material or the abnormal form of Pu(IV) believed to be a highly polymerized Pu(IV) ion carrying a small overall positive charge. Further tests have shown that such an oxidation treatment does not adversely affect the subsequent prereduction with $NaNO_2$ and extraction operations.

Ader, Bartell, Greenlee, and Malm have studied methods of increasing the rate of metal processing at Hanford during the early stages of operation when it may be desirable to process three or four tons of metal per day, rather than the one ton per day, specified by

the Hanford flowsheet. Possible adjustment of process conditions have been tested with inactive UNH solutions spiked with 80 g of Pu/ton U, a possible early operating level.

Beard, Greenlee, Haeckl, Hoekstra, Larson, S. Peterson, R. Thompson, and Winner have carried out two series of process runs in which cerium-zirconium scavengers were included in both bismuth phosphate cycles. All experiments were on a 100-ml scale and were made with active Clinton metal solution fortified with Hanford concentrations of plutonium and fission product elements. There were contamination difficulties in the first series, but the second series gave gamma-ray and beta-particle decontamination factors of 2.5×10^5 and 5×10^5, respectively, through two bismuth phosphate cycles. Plutonium losses have not been high in the scavenger precipitates.

R. Thompson has studied the use of HF or H_2SiF_6 to complex columbium and zirconium as a means of aiding decontamination from these two elements. In 20-ml scale experiments, decontamination factors through two cycles have been about ten-fold greater than those of the control runs.

Morgan has attempted to influence the state of dispersion of columbium and zirconium as a means of improving decontamination. The addition of sulfate ion, presumed to promote flocculation of columbium and zirconium, did not improve decontamination in one cycle. He also carried out scouting experiments on new decontamination methods that show that $Ba(NO_3)_2$, $Sr(NO_3)_2$, and the organic reagents "Trilon A" and "Trilon B" are ineffectual as specific complexing or scavenging agents.

Concentration-Isolation (Pye, Group Leader). Walling has conducted a preliminary experiment to see whether or not it is feasible to operate the present concentration procedure at low acidities (0.2 N HNO_3 and 0.2 N HF) rather than at the flowsheet values (1 N HNO_3 and 1 N HF). Yields are comparable except for a large unexplained loss (5%) in the bismuth phosphate by-product. Walling has also tested the isolation procedure using lanthanum fluoride slurries from Cell 4 at Clinton. His experiments show that a substance is present in the slurries that inhibits the precipitation of plutonium peroxide. Chemical analyses are in progress to identify the substance.

Goeckermann has studied the effect of the constituents of 25-12 stainless steel on the precipitation of plutonium peroxide and finds that in the absence of zirconium, 97-98% yields may be obtained even in the presence of 0.002 M Fe(III).

Goeckermann, Hopkins, Kelley, and Yett have carried out additional experiments on a lanthanum fluoride slurry obtained at the end of the concentration process in Clinton Room D (Batch 17) that indicate that zirconium and Fe(III) are the principal inhibitors of the precipitation of plutonium peroxide. It is found that fluoride ion will complex zirconium so successfully that good yields can be obtained. An additional method of removing the inhibiting substances in the plutonium peroxide precipitation has been developed by using a lanthanum fluoride cycle at high concentrations of plutonium.

Haeckl has investigated the $Bi(OH)_3$ alternate concentration method using KOH, NaOH, and NH_3 as neutralizing agents to precipitate the $Bi(OH)_3$ from simulated 10 N HNO_3 solutions following the last decontamination

cycle. The procedure is promising, but when tested using solutions from Clinton, bulky precipitates are formed that dissolve with difficulty. No further work is planned for the immediate future.

Beard has continued the work to develop a uranous oxalate alternate concentration-isolation procedure. Experiments show that yields of 94.5% may be obtained using a procedure involving the oxidation of the $U(C_2O_4)_2$ plutonium precipitate with $KMnO_4$ and the subsequent isolation of plutonium by reduction and precipitation of plutonium with $H_2C_2O_4$.

Process Development (Dreher, Group Leader). Lincoln, Winner, Hyman, and Rasmussen have investigated the variables associated with the use of $NaNO_2$ as a prereducing agent in the extraction step. The stability of $NaNO_2$ is not materially affected by variations in the concentration of UNH and of H_2SO_4. However, the rate of decomposition increases markedly with increasing temperature.

Basic Chemistry of the Separation Processes (Hindman, Group Leader). Kraus has studied the behavior, in dilute acids, of a green polymeric plutonium ion believed to be Pu(IV). A brilliantly green ion of plutonium can be prepared by heating the usual Pu(IV). The properties of the "green" ion differ markedly from those of the usual Pu(IV) ion. The green ion has a characteristic absorption spectrum. Precipitates obtained by adding SO_2, $H_2C_2O_4$, KIO_3, or H_3PO_4 to solutions of the ion are brilliantly green. This ion is carried well by lanthanum fluoride but not by bismuth phosphate. It has been shown experimentally that only 0.04 mole of $H_2C_2O_4$ per mole of plutonium is required to precipitate most of the ion from solution, indicating that the ion is highly polymeric.

Howland has tested holding oxidants to allow lanthanum fluoride assays for Pu(IV) in the presence of Pu(VI). The method involves the addition of 0.02 M bromate to the test solution to prevent reduction of Pu(VI) upon the subsequent addition of HF. It appears successful unless the bromate concentration is raised to 0.1 M which brings about slow oxidation.

In a memo to Whitaker, attention Doan, at Clinton Laboratories, I ask approval for Paul Fields to work at Clinton for a week or ten days beginning about July 10. The purpose is to trace, with the help of Np^{239} tracer, the course of the 93^{237} through a plutonium isolation run in Perlman's chemistry section so that we shall be able to ascertain more exactly in which fraction to look for this isotope. We need a larger supply of neptunium in order to investigate its chemical properties mainly for the purpose of devising a better method for the 93-94 separation and for the analytical control work during the early operation at the Hanford Engineering Works. The by-product solutions at Clinton offer a relatively cheap method of obtaining some of the 93^{237} isotope. Perlman prefers that one of our men do the work since it consumes an appreciable amount of time and also demands some rather special experience. I also request that Fields be allowed to place ten to twenty grams of a uranium compound in the Clinton pile for a day's bombardment with neutrons so that he can prepare the Np^{239} tracer.

Helen had coffee at Wilma's. Fred Albaugh came over to lunch. Fred, Helen and I then went to the Edgewater Golf Club where we saw McSpaden beat Hogan in the playoff, 70 to 73.

Today's headlines show that the Russians are within 12½ miles of Minsk.

Tuesday, July 4, 1944

Although this is Independence Day and was celebrated throughout the city, work went on as usual at the Met Lab.

Another technician was hired for Davidson's group, Helen Thomson.

I received a copy of a memo from Hogness to Hilberry concerning Stan Thompson's transfer to Hanford this summer. Hogness explained that Thompson, who is on leave from the Standard Oil Company, would not be able to extend his leave beyond July 1 of next year, if he is still at Hanford because the du Pont Company has a policy of not asking for the loan of employees from other companies. Therefore, Thompson would like to return to the Met Lab on May 1 next year, and would like a statement from the Laboratory that a position will be open for him here on that date. If necessary, he would transfer back to Hanford later.

At 10:30 a.m. I attended the Project Council Information Meeting on Nuclear Physics. Others present were Allison, H. L. Anderson, I. Bloch, L. J. Brown, Captain Chapman, Compton, Cooper, Dancoff, Darrow, Dempster, Doan, Fermi, Goldsmith, J. Hamilton, Hilberry, D. L. Hill, Hogness, Hughes, Jeffries, Jesse, A. Langsdorf, H. V. Lichtenberger, R. J. Moon, Morrison, Mulliken, D. E. Nagle, Nickson, L. A. Ohlinger, M. B. Sampson, R. B. Sawyer, R. Scalettar, Seitz, L. Seren, Shonka, Smyth, Snell, Soodak, Stearns, J. L. Stephenson, Stern, Stone, Szilard, Vernon E. H. Wakefield, Warner, A. Wattenberg, K. Way, Weinberg, Wheeler, Whitaker, Wigner, Wilson, Wollan, Young, Zachariasen, and Zinn. Zinn presented a terminal report on the construction of the P-9 pile at Argonne. Start-up occurred on June 20 when 6½ tons of satisfactory heavy water had been accumulated. The pile is designed for operation at 250 kw but has not yet been run above 190 kw. Preliminary measurements indicate that at 250 kw the neutron flux in the center of the pile will be 9×10^{11} n/cm^2/sec. By way of comparison, the flux at the center of the Site X pile is 4×10^{11} n/cm^2/sec at 1,700 kw.

Later at 2:30 p.m. I attended a Project Council Information Meeting on General Physics in company with the same people who attended the morning session on Nuclear Physics. Edward C. Creutz and Howard R. Kratz of the General Physics Division were additional attendees. Snell reported that the repairs and improvements on the Chicago cyclotron have been completed. Deuteron energy is now 8 Mev. Beam currents of 200 microamperes can be obtained. Joe Hamilton reported that the repairs planned for the Berkeley cyclotron (rewinding of magnet and replacing and narrowing the dees) will increase the deuteron energy from 13-14 Mev to 20 Mev.

Hamilton was followed by Zachariasen, who reported that a sample supplied by Cunningham, believed to be the plutonium oxide, was examined. It exhibited two phases; one of which was that of platinum, caused by the crucible material; the other is believed to be PuO_2. The lattice dimension of the latter is a = 5.39 Å. The lattice dimension of oxides of the four heaviest elements show the following regularity: ThO_2 = 5.59; UO_2 = 5.46; NpO_2 = 5.42; PuO_2 = 5.39. Thus the crystal structure data give unambiguous evidence that the 5f shell is being filled in the elements starting from thorium. In analogy to the early members of the rare earths (lanthanides), these elements may be called thorides. Zachariasen said that he has now definitely identified the hydrate $PuF_4 \cdot 2.5H_2O$. This proves to be isomorphous with uranium fluoride hydrate with this water content. Also he has succeeded in interpreting the complex diffraction patterns of $PuBr_3$ and PuI_3. They are orthorhombic crystals and isomorphous with UI_3, LaI_3, and $NdBr_3$. A sample submitted as $K_2PuF_6 \cdot xH_2O$ was found to consist of K_2PuF_6. The diffraction pattern indicates a rhombohedral cell. This substance is isomorphous with K_2ThF_6 and K_2CeF_6.

I received a copy of a memo from Tepe to Maloney referring to the decision by mutual consent to temporarily curtail the ether extraction investigation in the semiworks group because of the manpower shortage. During this period of inactivity it is planned to gather fundamental data on the double-solvent extraction process that offers the advantage that plutonium and uranium can be separated completely from fission products in a single solvent-extraction operation. Dreher's group will study effects of salts on the equilibrium distribution between ether and aqueous solutions; the General Engineering Section will work on the design, construction, and operation of a small glass continuous-extraction system.

Joe Kennedy telephoned me from Los Angeles about our patent matter. I agreed to write up something on the patents and confirmed that we will meet with Bob Underhill and others as planned on July 19 here in Chicago.

At 6:00 p.m. I called Underhill in Berkeley to confirm our July 19 date for a meeting in Chicago with Kennedy. He and Conard will arrive on the Streamliner at 12:15 p.m., Wednesday, July 19, and will stay over until 6:00 p.m. the next day. I told him we plan to write up the cases in view of our conference with Lavender on June 19.

Helen met Frances and Bob Chilson (Frances's cousin) for lunch. Later she stopped by Wilma's.

The Russians have taken Minsk, and the Americans have made gains in Normandy, according to today's paper.

Wednesday, July 5, 1944

At 8:00 a.m. I held a meeting of the Council of our section, which was attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Heath, Hindman, Jensen, Katzin, Manning, Orlemann, Simpson, S. Thompson, and Watt. I first brought up the question of plutonium losses. I said that it will be necessary to provide Vernon, who has been Assistant Laboratory Director all this year, with an itemization of losses, including an estimate of the number of experiments performed on a given quantity of material.

The question of health protection was discussed. It was pointed out that the present estimate gives one microgram of plutonium as the tolerance dose in the lungs. Ten micrograms is estimated as the lethal dose. It will be necessary in some work, perhaps, to employ special masks having a separate air supply. Desk tops having appreciable contamination are to be painted. More Plutos [portable alpha survey meters] are needed. Canvas shoes should be available at the end of the week and must be put on before entering the air lock. It was decided that no coke or milk vending machine should be put in the locker room of the filtered-air section. Manning is to write a memo asking that the present water dispensers be replaced by a system of bubblers using city water. The sub-section meetings on Thursday evenings will be held on Wednesdays again beginning today.

Stan Thompson presented me with a 10-page list of the problem assignments of his sub-section on separation processes and showing the following group organization: *Group 1. Extraction-Decontamination.* Albaugh (Group Leader), Sedlet, Ader, Malm, Bartell, Greenlee, Hoekstra, S. Peterson, Morgan, R. Thompson, and Bradt. *Group 2. Concentration-Isolation.* Pye (Group Leader), Hopkins, Goeckermann, Beard, Haeckl, Yett, Kelley, and Walling. *Group 3. Process Development.* Dreher (Group Leader), Winner, Larson, Rasmussen, Hyman, Blaedel, Lincoln, and Gilbreath.

Thompson also included a write-up of recent and planned work on prereduction, decontamination, increased production at Hanford (greater throughput and shorter cooling times), concentration and isolation, problems connected with the Hanford slug coating, and the effect of initial storage of UNH solutions at Hanford.

I wrote to Kennedy about our coming meeting in Chicago with Underhill and Conard on July 19, giving him the dates and times they will arrive and depart Chicago.

I sent Allison a review of the chemical specifications for the Hanford 200 Area (Chemical Processing) as stated by W. B. Gideon and W. E. Kirst. I suggested the specification for iron in the potassium hydroxide be lowered from 0.01% to 0.001%.

A notice was issued to all personnel at New Chemistry that a lunchroom has been provided on the first floor of the West Stands at the south end of the main hallway. This will be welcomed by most everybody as personnel have been forbidden since April 1 to eat lunches in the New Chemistry Building.

Organization charts of the technical and administrative organization of the Metallurgical Laboratory were issued. The Technical Chart shows Compton as Director of the Metallurgical Project and the following as Division Directors: Argonne — Fermi; Health — Stone (Watson, Associate); Physics — Hilberry (Acting); Chemistry — Hogness; Technology — C. M. Cooper; and Administrative — Kimpton. Section Chiefs and Group Leaders are not named.

I received a copy of a request from Maloney to Cooper that a chemist be assigned to the General Engineering Section in order to help in developing the double-solvent extraction process. He pointed out that all the Chemistry Division personnel who are working on this problem have recently been assigned to other problems.

Brody, Duffey, Jensen, Maloney, Orlemann, and Tepe met for the weekly conference on solvent extraction for product purification and reviewed the status of the plans to construct an all-glass continuous countercurrent solvent-extraction unit that would reduce the level of light element impurities to a factor of 10 below the established limit. Permission has been obtained to use the northern end of the West Stands squash court, and many of the drawings have been completed. The platform and equipment supports will be completed by next Tuesday.

The evening meeting, changed from Thursday evenings, of the Purification and Metal Production Sub-section (II) of Section C-I met at 7:45 p.m. in Room 209, Eckhart Hall, attended by Abraham, Cowan, Davidson, Hagemann, Hyde, J. Katz, Katzin, Kohman, Kraus, Orlemann, Manning, Sheft, Watters, Weissbourd, and others. Manning opened the meeting with remarks on cleanliness in the laboratory and the frequent security violations of late. George A. Cowan of Section C-IV, speaking for Mark Fred, reviewed the status of the light-element analytical work based on spectroscopy. James Watters reviewed the analytical work of Section C-IV based on chemistry.

Davidson spoke on the halides and oxyhalides of plutonium. Methods of preparation of the trichloride were reviewed. It is interesting to note that no higher chloride of plutonium has been formed with any of the powerful chlorinating agents used under rather severe conditions. This is interpreted as meaning that higher chlorides than $PuCl_3$ are not to be expected. The oxyhalide of plutonium has been prepared and identified; it is insoluble in water, soluble in dilute acid. Methods for production of oxybromide are under investigation. Plutonium oxyiodide is easily made and is obtained whenever the attempt is made to make PuI_3.

Weissbourd discussed neutron counting. Three samples of plutonium metal have been counted, which gave specific neutron emission rates of 10,000 to 23,000 n/m/g. Measurements were also made on three plutonium compounds or solutions. Values for fluorine and water were as expected, but deuterium oxide gave a surprisingly high count of 35,000 n/m/g of plutonium, compared with 7,200 for water. Kohman has suggested an explanation for this based on a fast alpha particle giving rise to a fast deuteron which interacts with another deuteron to produce a neutron plus a He^3 particle.

Helen worked at the YWCA. Ghiorso, Thompson, and I played 13 holes of golf at Jackson Park before dark (AG −50, ST −51, GS −49 for nine; AG −74, ST −73, GS −69 for thirteen). We travelled to and from the course by street car.

"U.S. Takes Saipan Capital" is the banner headline today.

The Project Council Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Compton, C. M. Cooper, Dempster, Doan, Fermi, Hamilton, Hilberry, Hogness, L. O. Jacobson, Jeffries, Mulliken, Peterson, Smyth, Stearns, Szilard, Vernon, Warner, C. J. Watson, Whitaker, Wigner, and Zinn. Walter Zinn was made a member of the Council. Compton said that arrangements are being made for Met Lab observers at HEW. Three are expected, one representing the Project as a whole (Hilberry), one on canning and corrosion (Howe), and one on separations process (not yet selected). Fermi will also be at Hanford during the early phases. First operation is still expected early in August, with the chemical processing plant operation in September.

Compton read a letter from General Groves concerning Project programs and personnel during the coming year. We are instructed to freeze our present level of personnel and to consider possible reductions in personnel that can be carried out without sacrificing effort on work believed to have potential value in the present war. No discussion of reduction in staff should at present be held with members of the Project in less responsible positions. Compton explained that the freeze means that personnel numbers are not to increase, but replacements for losses are in some cases allowable. He asked that consideration be given to reductions in staff as work goes into a stand-by basis — perhaps reductions between the limits of 25% to 75%. Timing will vary, but we might expect the reductions to start about September of this year and be completed by next January. There is no consideration of dropping effort on jobs with war significance. Hogness mentioned the difficulty of keeping strong men under stand-by situations. He also pointed out that the development of solvent extraction processes is one of our most important jobs, but there is some question about it being useful in this war.

Smyth urged that peacetime planning be pressed in order to avoid drifting into an untenable situation when, for instance, Germany collapses. Compton asked that each Division Director prepare a suitable program and personnel plans for longer-term work. He also agreed we must prepare a long-range program insofar as it is feasible to do so.

Compton read another letter from Groves requesting recommendations for a program in connection with the use of U^{233}. Whitaker said that 60 slugs are now out of the pile, containing perhaps 150 milligrams of U^{233}. Two slugs have been sent to Montreal. Fermi said that two or three milligrams of U^{233} are needed to get started and that Site Y has asked how they should go about

obtaining 1-2 mg. Compton said he believes that we should provide Site Y with an estimated delivery date; the U^{233} work is still third priority class but now near the top. He asked that Hogness review the situation in the Chemistry Division and attempt to get a separation process going. If it turns out the Project should extract U^{233} from the full 60 slugs, the logical place would be Clinton Lab with the process development carried out at Chicago. Compton will also investigate the possibility of the Montreal people agreeing to do the extracting.

Thursday, July 6, 1944

At 5:15 p.m. I left Chicago Municipal Airport by American Airlines plane for Cincinnati where I will take a Southern R.R. train to Knoxville in order to attend the monthly Steering Committee Meeting in Oak Ridge.

This morning's paper reports a Soviet drive west from Minsk. Premier Stalin announced the capture of the rail center of Molodeczno. Later, the Soviets reported the taking of Smorgonil, 21 miles farther west on the same railroad.

Friday, July 7, 1944

I arrived in Knoxville in the morning and was taken by automobile to Clinton Laboratories where I attended the first session of the Chicago-Clinton Conference (Steering Committee Meeting) which began at 9:20 a.m. Others present were Boyd, H. S. Brown, Davies, Greager, Hogness, W. C. Johnson, Kay, Kirst, and J. B. Work. Boyd outlined his program on decontamination improvement studies. Davies reviewed the work of his section on adsorption of zirconium on lanthanum fluoride; the use of lanthanum oxalate as a scavenger; and the colloidal properties of columbium, zirconium, and tellurium.

Kay talked about progress at Hanford, emphasizing that the first material to be processed at Hanford will run between 5 and 50 grams of plutonium per ton of uranium. The pile may operate at 30,000 kw in September, 60,000 kw in October, and 120,000 kw in November. Building 200 T (first canyon) could start processing the first output of plutonium on September 15 — Kay thinks it will be a miracle if we make it. The Hanford demonstration runs will be finished at Clinton on September 1, after which Clinton will study means of increasing production.

Kay raised the following problems as the most pressing in Hanford process development: (1) Should there be a decontamination step ahead of extraction? (2) What is the effect on plutonium yield of UNH storage? (3) How will scavengers behave in the plant? (4) Is a lanthanum fluoride by-product necessary in the concentration (crossover) step, and if so, how can the yield of this step be improved? (5) How can one best demonstrate the Hanford concentration and isolation procedures — i.e., how can one eliminate zirconium and iron at Clinton? (6) How can one

increase the plant capacity? (7) What is the minimum aging time for the Hanford slugs before dissolving and how can it be demonstrated?

The first session closed at 12:45 p.m. Then at 2:45 p.m. I attended the second session of the Conference; Boyd and Davies were not present. Greager reported on the work of his Division in relation to the Hanford flowsheet. He emphasized that his schedule is extremely tight and that the possibilities of trying anything not already scheduled are very small. Because of forthcoming transfers of personnel, little if any work can be done after August 15 on the Hanford flowsheet.

I reported on the work of my section in Chicago and, in addition, proposed that two points be added to Kay's seven points: (8) Prereduction, and (9) Problems of handling the coating of uranium slugs. It was decided that there will be Committee meetings at Clinton on August 4 and September 8. The meeting adjourned at 5:15 p.m.

At midnight I caught the Southern R.R. train to Cincinnati.

Saturday, July 8, 1944

On arrival in Cincinnati, I learned that I would be unable to use my American Airlines return ticket — lack of priority prevented getting a seat. So I caught the NYC train to Chicago and arrived at 6:30 p.m.

Yesterday Helen shopped in the afternoon and had dinner at Wilma's.

The bombardment of 10 mg of plutonium (sample 49A) with 32 Mev helium ions is scheduled to begin in the Berkeley 60-inch cyclotron, while a 40 microampere-hour bombardment of natural uranium with helium ions is supposed to end today.

Yesterday afternoon while in the process of converting 350 mg of plutonium from nitrate solution to PuF_3 urgently requested by Baumbach, Meyer had an accident that resulted in about 95 mg of PuF_3 precipitate popping out of a test tube during heating, most of it striking the finger of his glove and the wall of the water bath. He worked during the evening to recover the portion of the plutonium that had fallen into the water bath. Meyer, with the help of Florin and others, spent most of today cleaning up his hood and the laboratory.

Yesterday Manning and Watt, in a memo to the eleven Section C-I Group Leaders, asked each of them to prepare a detailed statement to show how plutonium losses occur in his group and an estimate of the magnitude of the losses incurred in the various types of operation.

A report from the Housekeeping Committee of my section (Irene Corvin, Roy Thompson, Jasaitis, and La Chapelle) classifies the rooms in the filtered-air section of New Chemistry as follows in terms of cleanliness and orderliness: (1) Very good rooms: 38, 39, 40, 25, 26; (2) Good rooms: 10, 12, 22, 41, 42, 43; (3) Fair rooms: 1, 4, 5, 8, 9, 11, 13, 16b, 16c, 21, 23, 27, 29, 30, 31, 33, 36, 37; (4) Not good rooms: 2, 6,

7, 34, 35; (5) Bad rooms: 28.

Zachariasen reports that he has evidence for the existence of two trivalent thorium compounds, ThF_3 and ThOF. He believes the former to be isomorphous with PuF_3 and UF_3 and the latter to be isomorphous with PuOF and LaOF.

Rexford H. Bradt, working in collaboration with Hiskey's section, asked permission to take a sample of fission products solution to the Illinois Institute of Technology Electron Microscope Laboratory next Monday to see whether or not a radioactive sample can be handled by the electron microscope. He believes the electron microscope may be a useful tool in studying peptization of fission products in the Bismuth Phosphate Process.

On my behalf Edrey Smith transmitted to Lois Moquin in Berkeley, a reprint of my article on Artificial Radioactivity, properly addressed and stamped, for mailing to the requestor from Berkeley. For security reasons I cannot let the man know I am in Chicago.

Watt forwarded to J. E. Cole at du Pont a report on filtration and decantation experiments on plutonium peroxide. The experiments were conducted by Yett and Pye, using about 500 mg of plutonium. They find that decantation, filtration, and centrifugation are all feasible methods.

In a memo to Compton, Allison asks his advice about taking Maurice Goldhaber, an associate professor of physics at the University of Illinois, and his wife Trudy, into the Metallurgical Laboratory to write a handbook on experimental nuclear physics. He quotes a letter from Mulliken strongly supporting the idea.

Sunday, July 9, 1944

Al Ghiorso, Stan Thompson, and I travelled by street car to the Evergreen Golf Club to play 18 holes of golf. Ghiorso shot 132, Thompson shot 108, and I shot 113.

Helen spent the day at home.

George Watt and Paul Fields are leaving at 11:50 p.m. on a trip to Clinton Laboratories. Watt will attend a meeting on plutonium isolation on Tuesday. Fields is to conduct an experiment using 93^{239} to trace the course of 93^{237} through an isolation run in Perlman's chemistry section so we can determine which fraction is the best source of this isotope.

Monday, July 10, 1944

The bombardment of ~15 mg of Pu^{239} with 32 Mev helium ions from the Berkeley 60-inch cyclotron ended at 8:00 a.m. with 36.5 microampere-hours [Sample 49αA].

Edrey Smith, my secretary, started her vacation today. She will return in two weeks.

Another laboratory assistant, Inez Jones, was hired.

The fellows in Room 2 are still chuckling about a recent incident. Helen Pellock, a technician, is in the habit of taking off on her bike at lunch time for a swim. On a recent day she changed into her swim suit in the women's room, donned a pair of slacks, and then walked through the lab. As she passed Norm Davidson, who was working with some greasy thing, she got some of the grease on her suit just above the waist. Norm was quite distressed as it appeared to be his fault, so he tried to mollify her by cleaning off the grease. He took a wad of cotton, soaked in acetone, and wiped away the middle section of the rayon suit.

Katzin commenced setting up for the extraction of U^{233} from thorium carbonate cans irradiated in the Clinton pile.

Helen had lunch with Wilma.

Admiral Chester W. Nimitz announced that U.S. forces "have completed the conquest of Saipan."

Tuesday, July 11, 1944

The terminations of Jerome and Isabella Karle are effective today.

The two helium ion-bombarded targets flown in from Berkeley were turned over to Ralph James. These targets are: (1) Sample 49αA — 36.5 microampere-hours of 32 Mev helium ions on ~15 mg plutonium — stopped at 8:00 a.m. yesterday. (This sample had been prepared by James on a gold target on March 24 of this year.) (2) Sample UαA — 40 microampere hours of 32 Mev helium ions on uranium — for which the bombardment ended on Saturday. An alpha-particle absorption curve on the plutonium sample using the nitrogen chamber was started by Ghiorso using the whole target. A very small amount of the plutonium was dissolved by James and five plates of various sizes prepared to look for alpha-particle activity of a range different from that of Pu^{239}.

The 35-page summary of work on the separations processes in Section C-I that I sent to Hogness July 3 was issued today as Report No. CN-1762, "Chemical Research — Separations Processes for Plutonium."

"Chemistry Division — Summary Report for June 1944," (CS-1816),

was issued. It contains information on work by Sections C-I, C-II, C-III, C-IV, and C-VI. The Table of Contents lists the following subjects under my section:

Separations Process Studies

Bismuth Phosphate Research — General Problems
Bismuth Phosphate Research — Decontamination
Concentration-Isolation at "W", Fluoride and Peroxide Methods
Concentration at "W" — Alternate Methods
Extraction and Decontamination — Ether Method
Semiworks Waste Disposal
Evaluation of Process Modification
Hanford Flowsheet

Purification and Metal Production

Survey, Solvent Extraction Method for Plutonium Purification
Purification of Plutonium by Extraction of Organic Complexes
Solvent Extraction Method for 1 to 10 mg Scale
Chemical Composition of Plutonium Halides
Preparation and Properties of Plutonium Chlorides
    Synthesis of $PuCl_3$ and Attempted Synthesis of $PuCl_4$
    Synthesis of PuOCl
Preparation and Properties of Plutonium Bromides
Preparation and Properties of Plutonium Iodides
Study of the Higher Fluorides
Hydrofluorination of Plutonium Oxide
Preparation of Plutonium Halides for Metal Production
Plutonium Density Determination
Hardness of Plutonium Metal
Reduction of Plutonium Compounds to Metal
Electrolytic Metal Production
Refractory Studies
Production of Plutonium by Thermal Bomb Reduction
Micro-Oxygen Analysis Apparatus (after "Y")
Flowsheet for Metal Production (as given in MUC-GTS-766)

Basic Chemistry

Inorganic Derivatives of Plutonium
Hydrolysis of Plutonium(III) and Plutonium(IV)
Isolation and Study of Pure Np^{237}
Preparation and Uniform Deposits of U^{235} and Pu^{239}
Assay of "W" Plutonium Solutions
Heavy Isotopes by Bombardment of Pu^{239}
Recovery of Plutonium
Research on Methods for Plutonium Recovery
Routine Separation of Np^{239}
Neutron Counting with BF_3 Pressure Ion Chambers
Plutonium-Finding Alpha Counter
Investigation of Back-Scattering of Alpha Particles
Geiger-Müller Counter Tube Research
Material for Project Handbook
Search for Aggregate Recoil in Plutonium

Helen had lunch at the Standard Club.

Before dark this evening Stan Thompson and I played nine holes of golf at Jackson Park (ST −50, GS − 49).

Headlines today indicate a good push on both eastern and western fronts in Europe. The Soviets are within 60 miles of the Polish border, and both the British and Americans are making gains in France.

Wednesday, July 12, 1944

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Hindman, Jensen, Katzin, Manning, Orlemann, Pye, Simpson, S. Thompson, and Willard. I brought up a number of items: (a) Security violations must be eliminated. (b) There has been a limited authorization for work along other lines, such as the U^{233} work. In line with this, four periods of the work are now recognized: (1) present, (b) stand-by, (3) transition, and (4) postwar. (c) A report is to be prepared by the sub-section chiefs on what should be done in the period up to October 1 (the stand-by period). This will be due July 24. These reports should include an estimate of the manpower required. (d) Manpower is now frozen. No new men can be added. Orlemann noted that the glass shop is the worst bottleneck in getting our work done.

Continuing with the work on target 49αA (plutonium plus 32 Mev helium ions, Berkeley bombardment), Ghiorso finished the absorption curves for the whole target and a similar curve for a plutonium, unbombarded sample. No new alpha-particle emitter of range larger than that of the unbombarded sample was observed. James then dissolved the plutonium off the gold target with HNO_3 and H_2SO_4. The H_2SO_4 was fumed off, and the sample made up to 2 ml in 1.2 N HNO_3. An alpha assay showed a total of 4.9 mg of plutonium. About 0.4 mg was reserved for a sample for Jaffey in the differential chamber. The remainder was split into two 2.2-mg portions. Duane Hufford will prepare a plate from one portion for Ghiorso to use in the nitrogen chamber. With the other sample, James began the dichromate cycle — we have decided to proceed on the assumption that elements 95 and 96 cannot be oxidized to fluoride-soluble states with dichromate ion. James added $K_2Cr_2O_7$ to oxidize the plutonium and at midnight will put the sample into a 75°C oven to remain overnight.

In a letter to Whitaker, attention Sinclair, at Clinton Labs, I amended our needs for batches of pile-irradiated uranium slugs.

I met with Albaugh, Dreher, A. C. Hyde, Kircher, and Stan Thompson to discuss the present status of the semiworks. It is now agreed that immediate steps should be taken to obtain men to replace those expected to be transferred from the Chicago semiworks to Hanford. Twenty-eight men are now assigned to the semiworks; 12 of them are du Pont employees. It was decided that an effort should be made to secure ten replacements not later than August 15. In view of the manpower freeze, it may be

necessary to obtain these men by transfer from elsewhere on the Project. I expressed my opinion that the need for semiworks operation within the next few months will be concerned with (a) the evaluation of proposed improvements in specific steps of the present Hanford separations process, (b) the study of improved decontamination cycles that might consist of a slightly modified Bismuth Phosphate Process without scavengers, (c) the preparation of active heavy metal solutions for use in extraction and decontamination studies, (d) work on recovery of uranium, (e) the development of alternate separation processes, (f) the development of methods for the extraction of U^{233} and Pa^{233} from thorium, (g) the processing of special bombardments of heavy metal, e.g., for the purpose of extraction of 93^{237}, and (h) other special jobs which cannot be anticipated.

The weekly conference on Solvent Extraction for Product Purification was attended by Brody, Duffey, Jensen, Lawroski, Maloney, Orlemann, and Tepe. It was reported that the platform and equipment supports for the all-glass countercurrent extraction system have been erected at the northern end of the West Stands squash court and all subassemblies for the glass equipment have been completed by the glass blowers. Assembly of the equipment is to start tomorrow and is to be completed by July 21. Operations will start immediately thereafter.

At 6:30 p.m. Watt returned to Chicago from his trip to Clinton Laboratories.

I attended the evening meeting of the Basic Chemistry, Recovery, and Instruments Groups (Sub-section III) of Section C-I at 7:45 p.m. in Room 209, Eckhart Hall. Others present were Crawford, Cunningham, Fineman, Hindman, Hoekstra, Hopkins, Jaffey, Katzin, Kohman, La Chapelle, Manning, McLane, O'Connor, Orlemann, Pye, Scott, Cliff Smith, Stewart, Walsh, and Willard. Stewart reported on the work done by the recovery group on hexone extraction of plutonium(IV). The technique consists of three extractions from 10 M NH_4NO_3 solution with the same sample of hexone (methyl isobutyl ketone). The hexone layer is then washed with H_2O to to remove the plutonium, followed by precipitation as hydroxide, and dissolving in HNO_3 to give stock solution. The best run to date on a sample from Site X gave 99.0% plutonium recovery with all impurities but calcium below the limits of detection, while calcium was present to the extent of 0.014%. Runs on bismuth phosphate samples show that as soon as the bismuth phosphate phase separates, no more plutonium is extracted. I suggested saturating the hexone layer with HNO_3 to oxidize any impurities and also to aid in keeping bismuth phosphate in solution. Stewart stated this had been attempted but that the HNO_3 extracts into the water layer and prevents extraction of plutonium from hexone into water. There was considerable discussion concerning application of this technique to the Bismuth Phosphate Process. I pointed out that as a concentration and isolation step it holds immediate promise but that as an overall operation from UNH solution, it is not practical at this time.

Crawford presented his calculations on the efficiency of a neutron counter filled with heavy water as a moderator as compared with one filled with paraffin. Theoretical models give 6.4% for the paraffin, the latter figure comparing well with the 5% found experimentally. Using a homogeneous mixture of heavy water and BF_3 surrounding the fast neutron

source, he finds that the theoretical efficiency is raised to 16.5%.

McLane reported on experiments on dialysis of plutonium(IV) nitrate solutions. Kraus has previously found evidence that indicates that the green "abnormal" form is colloidal, whereas the brown "normal" form of the nitrate is not. McLane's experiments were carried out in a collodion sac (volume about 700 microliters). With an inside concentration of plutonium of 0.76 gm/l, he obtained 13% and 10% dialyzable at outside plutonium concentrations between 0.16 and 0.17 gm/l.

Helen was home all day.

The headline "Roosevelt Seeks 4th Term" has driven war news from the first position in today's paper.

Thursday, July 13, 1944

James removed sample 49αA from the oven at 9:00 a.m., where it had been allowed to oxidize in dichromate solution overnight at 75°C. Lanthanum fluoride was precipitated. (This precipitate presumably contains the insoluble fluoride of element 95 or 96, plus a small fraction of the plutonium, nearly all of which should remain in the supernatant in the soluble VI oxidation state.) James added H_2SO_4, fumed the precipitate to dryness, and dissolved it in 2 ml of 0.75 N HNO_3. He then took an alpha count on an aliquot of the solution which showed that 6.8% of the plutonium originally present had been carried by the lanthanum fluoride precipitate. James also took a beta-particle count on the aliquot used for the alpha analysis. A second dichromate oxidation was carried out (75°-90°C for 1 hour) and HF added to form a lanthanum fluoride precipitate. The precipitate was dissolved; the alpha-particle activity carried by the precipitate was found to be 420,000 c/m or 0.26% of the original alpha activity. (A beta-particle count on the aliquot used for the alpha analysis gave 0 ± 2 c/m.) Ralph began a third dichromate cycle by adding $K_2Cr_2O_7$ to the solution and putting it into a 75°C oven at 6:00 p.m. to remain overnight.

Sheft began construction of a vacuum apparatus for the preparation of PuI_3 by the wet method.

I sent a detailed statement to Vernon about the distribution of the 269-milligram plutonium loss reported for the month of June. There have been no large individual losses, rather, the total represents many small losses incurred during the month. I pointed out that over 300 experiments with plutonium were performed by members of our section during the period. I mentioned that 269 milligrams of product in the form of metal or salts represents a total volume considerably less than that of a single drop of water. I stated, however, that we are eager to reduce further our losses in any manner that is practicable, and I asked his suggestions for improving our procedures.

"Chemical Research — Basic Chemistry of Plutonium. Report for Month Ending July 1, 1944," (CN-1764), was issued. The following investi-

gations are reported:

Basic Chemistry (Hindman, Group Leader). O'Connor has determined the solubilities at room temperature of the following halide derivatives of Pu(IV): plutonium(IV) fluoride in 0.1 to 2 N HNO_3 containing 0.5 to 2.0 M HF, $K_2PuF_6 \cdot xH_2O$ in 0.5-2.0 M HNO_3 containing 0.5 to 2.0 M HF, plutonium(IV) lanthanum double fluoride in HNO_3-HF solutions, plutonium(IV) iodate in KIO_3-HNO_3 solution, plutonium(IV) bromate in H_2SO_4-$KBrO_3$ solutions.

C. Smith has prepared $K_2PuF_6 \cdot xH_2O$ in crystalline form. Zachariasen finds it to be isomorphous with $K_2CeF_6 \cdot H_2O$ and $K_2ThF_6 \cdot H_2O$. Smith has also attempted preparation of the iodate, oxalate, and phosphate of the "abnormal" Pu(IV) ion. The precipitates obtained with iodate and phosphate are amorphous, and their crystal structure could not be determined by x-ray diffraction study. The oxalate precipitate gives a diffraction pattern which is identical with that for normal Pu(IV) oxalate.

Magnusson has conducted experiments on the oxidation and carrying of tracer amounts of neptunium that demonstrate, contrary to earlier belief, that dichromate-oxidized neptunium is carried by sodium uranyl acetate — provided fluoride ion is present.

Magnusson and La Chapelle have isolated a total of 80 micrograms of Np^{237}. Of this, 40 micrograms were extracted from 64 pounds of Berkeley cyclotron neutron-bombarded uranium, and most of the rest was recovered from peroxide supernatants originating from separation processes at Clinton Laboratories. The neptunium was finally precipitated as the brown hydroxide and ignited to the oxide. The x-ray diffraction pattern shows it to be NpO_2, establishing a IV oxidation state for neptunium.

Recovery Group (Dawson, Group Leader). H. H. Anderson, Asprey, Britain, Burr, Fineman, Leventhal, Stewart, and Studier have continued recovery and purification operations, reclaiming more than twice as much plutonium as during the previous month. Final purification was accomplished by extraction with either ether or hexone (methyl isobutyl ketone).

Asprey, Fineman, Leventhal, Stewart, Studier and Dawson have experimented with hexone as an extraction solvent for plutonium(IV). In contrast to diethyl ether, hexone is not hazardous and seems to be equally efficient. Studies of the efficiency of the hexone in decontamination and concentration, as well as in isolation and purification, are in progress. Stewart (largely) has designed a continuous-batch extractor on the countercurrent principle which was built and used for a few experimental runs that are quite promising.

Instruments (Ghiorso, Group Leader). Scott completed a study of counting yield versus sample thickness for U_3O_8 mounted on platinum. Sample densities range from 0.8 to 0.007 milligrams of U_3O_8 per square centimeter.

Helen took a golf lesson at the YWCA.

Katzin, Thompson, and I played nine holes of golf at Jackson Park before dark. Scores were LK —59, ST —56, GS —55.

The Germans report that U.S. troops in France have opened a new drive near St. Lo.

Friday, July 14, 1944

James removed sample 49αA from the oven at 8:15 a.m. where it had been undergoing the third dichromate oxidation. The sample evaporated almost to dryness during the night, so he reoxidized with dichromate for an hour. A lanthanum fluoride precipitation was made, and the entire precipitate mounted on a platinum counting disc (labeled 49αA#9). It was found to contain 6,200 alpha-particle counts per minute, or about 0.004% of the original alpha-particle activity. During the late morning and early afternoon an alpha-particle absorption curve was taken using the low geometry counter. From the data there appears to be a long-range alpha-particle emitter! To check the curve, absorption data were taken on a pure 94^{239} sample prepared in the same manner as sample 49αA#9. James and I examined these data and the absorption data from sample 49αA#9 and recorded the following observation:

> Plotting and comparing these data shows that a 49 sample 20% larger than 49αA#9 falls to zero c/m much faster than 49αA#9 itself. This definitely shows the presence of a long-range alpha emitting isotope in sample 49αA#9. This is undoubtedly due to a product of the nuclear reaction of alphas on 94^{239} and is probably one of the following: 95^{242} (by α,p reaction) or 96^{242} (by α,n reaction) or 96^{241} (by α,2n reaction). Other isotopes are possible but these seem the most likely. The isotope 95^{241} from α;n,p is fairly possible. It is difficult to estimate the range of the alpha particles from this new isotope very accurately but it would seem to be about 4.65 ±0.15 cm of air.

I sent a memo to Hogness on the work of the past month on purification and metal production, covering the following:

I. Production and Properties of Plutonium Metal

A. Vapor phase reductions. Two 5-mg reductions of PuOCl have been made with barium in an all-beryllia crucible system fired to 1250°C. Metal was not produced, according to Zachariasen's x-ray analysis. Drying "old" PuF_3 by heating in vacuo gave a satisfactory reduction to metal.

B. Bomb reductions. The two bomb reductions of PuF_3 by means of lithium on the 100-mg scale using 1100°C firing for 5 minutes produced fair yields of metal (60% and 80%). One piece (60 mg) had a density of 16.2 gm/cm^3 and a neutron count of 23,000 n/m/g.

C. Density. Zachariasen has examined filings from the 60-mg piece and reports that at room temperature the sample exhibits the complex structure with certain lines differing markedly in intensity from previous samples reduced with barium. He also reports that the sample contained about 20% PuO. His studies using a high temperature camera have not been completed.

D. Vapor pressure of plutonium metal. Two additional runs have been made with the original apparatus containing the 5.6-mg sample of metal used to obtain the preliminary results reported last month. Vapor

pressure experiments with this original sample have been discontinued in favor of using a larger metal sample obtained from Site Y and a new apparatus. Other experiments with, presumably, plutonium oxide indicate that some oxide of plutonium may be vaporizing as such.

II. Purification of Plutonium Compounds

A. Solvent extraction. Tracer experiments have been carried out on the salting-out of plutonium from 10 M NH_4NO_3 into cyclopentanone and dibutoxytetraethylene glycol. Purification of milligram quantities of product by extraction with hexone (methyl isobutyl ketone) and diethyl cellosolve show that the distribution of light elements in the presence of macro amounts of product agrees essentially with those in the absence of product.

B. Continuous extraction studies. The small "continuous batch extractor" using hexone has been modified to increase its efficiency and has been used to purify a 50-mg sample of plutonium of unknown purity with quite satisfactory results from a purity point of view. A larger countercurrent column now under construction (designed by the Technical Division) will be capable of processing two liters of solution per four hours and should extract 99.9% of the plutonium and remove light elements to less than 0.1% of the tolerance limit.

C. Colloidal properties of plutonium(IV) nitrate solutions. Cunningham's group has recently acquired evidence for the existence of a colloidal Pu(IV) compound in HNO_3 solutions. It is important that the existence of this phenomenon be taken into account in considering precipitation and solvent extraction methods for purification since it may be the source of considerable difficulty in using such methods. At the present time the properties of these Pu(IV) solutions are not known insofar as they affect specific precipitation and solvent extraction procedures.

III. Volatility and General Dry Chemistry

A. Hydration of $PuCl_3$ at room temperature in water vapor at controlled pressures has been carried out and 1, 3, and 6 waters of hydration were observed.

B. Sublimed $PuBr_3$ has been prepared from $PuO_2 \cdot xH_2O$ in a quartz apparatus at 800°C by the action of a mixture of bromine and sulfur bromide vapors.

C. PuOI has been prepared by the action of an H_2-HI mixture on $PuO_2 \cdot xH_2O$ at 750°C.

D. PuF_4 is not reduced to PuF_3 by H_2 at temperatures below 250°C but is reduced above this temperature.

E. Plutonium fluosilicate. The precipitate of the reaction of fluosilicic acid and $Pu(NO_3)_4$ has been examined by Zachariasen and shows a diffraction pattern similar to a sample supposed to be UOF_2.

IV. High Vacuum Techniques. Additional tests of the oxygen analysis apparatus have been made. Construction of an effusion apparatus for vapor pressure measurements of plutonium compounds is nearly complete. The target chamber will accommodate 100 targets.

In another memo I sent to Hogness today, I summarize the results of our July 12 meeting on the semiworks and request ten men to replace the du Pont men who are transferring from the semiworks to Hanford.

Watt recommended to Hogness that he obtain from Clinton (a) an aliquot from the series of plant runs in which zirconium will be omitted from the second lanthanum fluoride cycle in Room D — needed because simulation of Hanford process conditions has been difficult because of the high concentration of zirconium present, and (b) a sample of lanthanum fluoride slurry from Room D containing one gram of plutonium for use in Dawson's work on solvent extraction.

I dictated a paper to Nathalie Baumbach, called "Inferences Concerning the Electronic Structure of Plutonium from Chemical and Physical Properties." In this I suggest that the elements heavier than actinium should be placed in the Periodic Table as an "actinide series." It is to be presented at next Monday's meeting with Thomas on the final purification and metallurgy of plutonium.

In the evening Helen and I saw a double feature at the Frolic Theater, "Pardon My Rhythm" with Arthur Lake and "Sailor's Holiday" with Bing Crosby.

Today's paper says that the Soviets are only 30 miles from the Polish border and that the Americans are gaining at St. Lo.

Saturday, July 15, 1944

James made further alpha-particle absorption measurements on sample 49αA#9 in the nitrogen chamber using mica absorbers. The data continue to show the presence of a long-range alpha-particle emitter.

Katzin has worked out the procedure for extracting U^{233} directly from the Clinton pile-irradiated thorium carbonate, without preliminary Pa^{233} separation via MnO_2. Ether extraction will be used. The carbonate will be dissolved, siphoned over in 100-ml portions to the extraction vessel, NH_4NO_3 added, followed by ether extraction.

I received a copy of a memo from Kircher to C. M. Cooper pointing out that unless arrangements are made well in advance for replacement of the du Pont men scheduled to leave, the Chicago semiworks program will have to be abandoned. He told Cooper that we in the Chemistry Division have advised him that our efforts to improve decontamination to the desired levels can only be effective if a semiworks is available for large-scale evaluation runs.

Hogness sent me a copy of his memo to Hilberry requesting two additional grams of plutonium as lanthanum fluoride slurry. One gram is to be used for work on concentration and isolation in accordance with the desire of Greager and Kay. With regard to the second gram, he refers to

the very promising work of the Recovery Group since March 1 using hexone (methyl isobutyl ketone) for extracting plutonium from all kinds of solutions. Hogness states his belief that it can be made into a very practical step for concentration and isolation and refers to Greenewalt's interest and proposal that a flowsheet be drawn up incorporating this step. A gram of plutonium will be used to make a run incorporating this extraction step. He also points out that we have obtained the services of Professor Merrell Fenske of Pennsylvania State University as a consultant and the full-time services of Stephen Lawroski, both of whom are solvent experts.

Katzin outlined for Moulton two possible patent cases deriving from Dawson's work. One is a general case on the solution of lanthanum fluoride by means of zirconyl or ferric ions followed by ether extraction and the second is a specific case using hexone as the solvent.

Helen took a walk with Wilma in the afternoon.

U.S. troops took nine French towns, and the Russians troops captured Pinsk, according to today's newspaper.

Sunday, July 16, 1944

Ghiorso and James took additional alpha absorption measurements on sample 49αA#9. The data again show definitely that element 95 or 96 must be present (see Fig. 7).

While Helen went to the beach at Lake Michigan with Marilyn Howe, I played 18 holes of golf with French Hagemann and Luther Arnold at Hickory Hills Golf Club (8201 West 95th Street, Palos Park). Scores were FH —107, LA —92, and GS —104. After the match Luther had a flying session at Ashburn Flying Field on Cicero Ave. By coordinating our golf matches with Luther's flying sessions (for which he receives an extra gasoline ration), we are able to play at more distant courses.

Monday, July 17, 1944

James and Ghiorso conclude they have about 260 c/m of the new element 95 or 96, based on extrapolation of data collected using the nitrogen chamber. James next initiated an effort to extract more of the new activity from the original plutonium fraction from which sample 49αA#9 was derived. The solution was reduced with SO_2 and HF added to precipitate PuF_3 and any element 95 or 96 present. The precipitate was dissolved and put through a dichromate oxidation. Some La^{+3} was added and precipitated with HF, presumably carrying the element 95 or 96. The precipitate was dissolved and combined with the liquid fraction from the second dichromate cycle carried out on the original target (liquid 49αAII), and both were heated to fumes with H_2SO_4. A precipitate, probably chromic sulfate, formed and was difficult to dissolve.

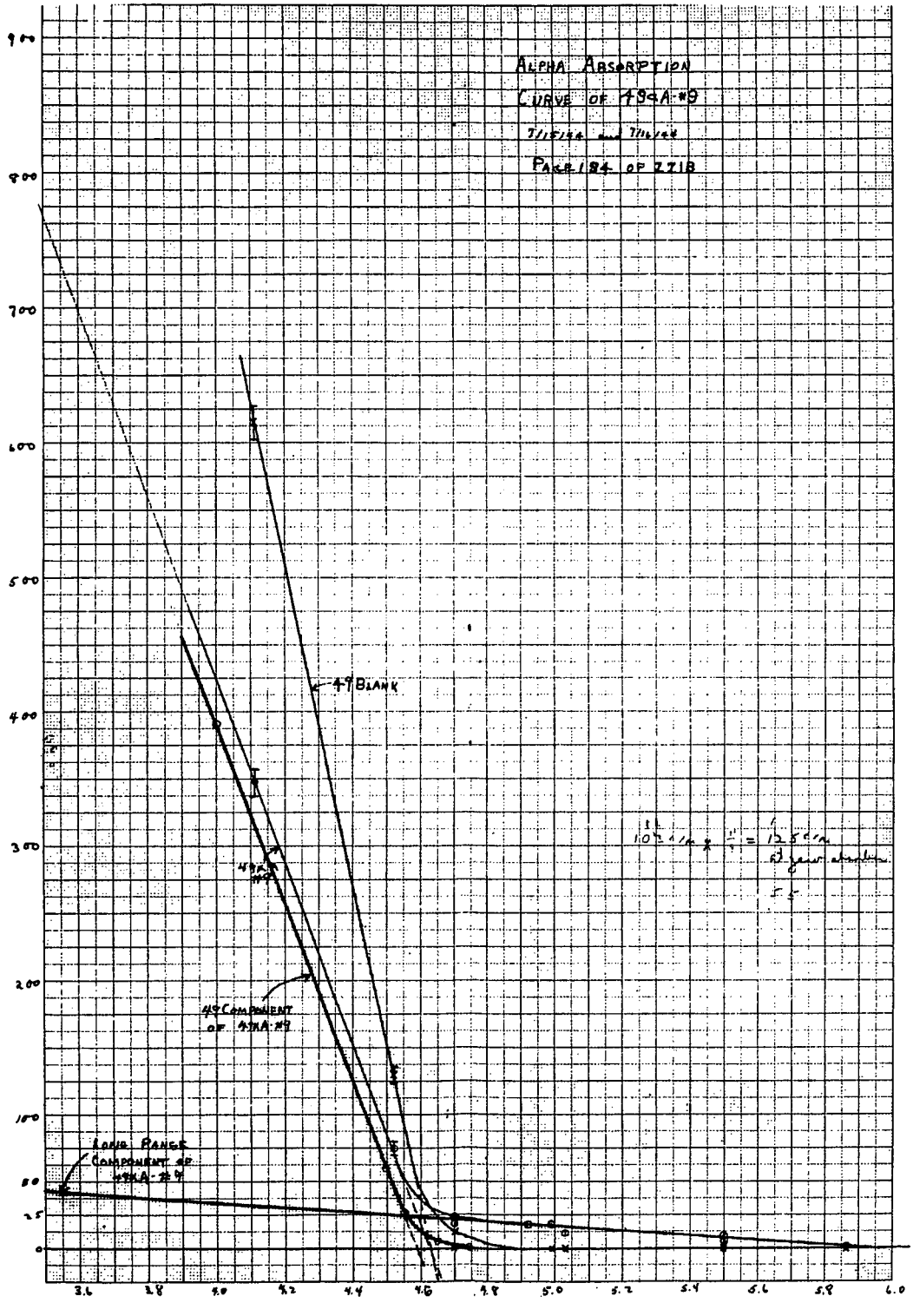

Figure 7. Mica absorption data showing presence of 96^{242} alpha particles in helium-irradiated Pu^{239}. July 15-16, 1944.

XBL 7811-13185

Currently, the code names for special materials that we are using are as follows: U metal — Tuballoy; $Na_2U_2O_7$ — Sodium salt; U_3O_8 — Black; $UO_4 2H_2O$ — Peroxide; UO_3 — Orange; UO_2 — Brown; $UO_2(NO_3)_2 6H_2O$ — Hex; UF_4 — Green; UCl_3 — Trichloride; UCl_4 — Product 516, UCl_5 — Pentachloride; UCl_6 — Hexachloride; UF_6 — PG; UO_2F_2 — WE-202; $U(SO_4)_2$ — Chemical 701; $UO_2(SO_4)$ — Chemical 728. (An asterisk when used after a code name denotes "hot" or irradiated material.)

From 9:30 a.m. to 12:30 p.m. and from 1:30 p.m. to 4:00 p.m. in Room 209, Eckhart Hall, I attended the monthly meeting with C. A. Thomas on purification and metallurgy of 49. Others present at the morning session were Allison, Ashcraft, Chipman, Connick, Cunningham, Derge, Franck, Hogness, Jeffries, W. C. Johnson, Kennedy, Orlemann, A. V. Peterson, C. S. Smith, Spedding, Thomas, Voigt, Warren, and Zachariasen. Smith and Kennedy reported on progress at Site Y. In metal reduction work, the plutonium metal buttons produced usually have densities of 18.9 gm/cm^3 or higher. One large (8 gram) reduction has been tried and resulted in failure because the compound used was PuOCl instead of $PuCl_3$. The material has been reprocessed, and another reduction on this scale will soon be made. Electrolytic reduction has given no new results because the one-gram scale is too small to evaluate the relative advantage as compared with bomb reduction. To date, all attempts at remelting plutonium under fluxes have been unsuccessful. Measurements of dilation vs temperature have fully established a large density change near 135°C; volume increases of 6.7% have been observed.

Connick reviewed the Berkeley work on equilibria between the various valence states of plutonium in aqueous solution. He feels it is certainly established that plutonium exists in aqueous solutions in the valence states of +3, +4, +5, and +6. The equilibrium between these states can be represented by the following equations:

$$\overset{64\%}{3\,Pu(IV)} = \overset{24\%}{2\,Pu(III)} + \overset{12\%}{Pu(VI)} \quad (1)$$

$$Pu(IV) + Pu(V) = Pu(VI) + Pu(III) \quad (2)$$

$$2\,Pu(V) = Pu(IV) + Pu(VI) \quad (3)$$

$$Pu(V) + Pu(III) = 2\,Pu(IV) \quad (4)$$

$$2\,Pu(VI) + Pu(III) = 3\,Pu(V) \quad (5)$$

Equilibria (1) and (2) have been studied. The equilibrium shown for (1) is based on starting with pure Pu(IV) solution in 0.5 M HCl at 25°C. Connick pointed out that a description of the discovery of the +5 oxidation state of plutonium at Berkeley appears in the recently issued Berkeley report CN-1912.

Cunningham spoke on "Ionic Species in Aqueous Solutions and Solubilities of 49 Compounds," first giving the recent evidence that a certain fraction of plutonium in some solutions is non-dialysable through collodion, making it necessary to consider the circumstances under which plutonium exists as a colloidal dispersion. Pu(IV) may be caused to aggregate into colloidal-sized particles from solutions of much lower concentration than

80 gm Pu/l if the acidity is lowered to 0.1-0.3 M. Heating is necessary to bring about the "polymerization" quickly. Precipitation properties of colloidally dispersed plutonium are very different from those of molecularly dispersed plutonium. The polymeric green ion may be converted to normal Pu(IV) ion by heating in H_2SO_4, or by oxidation to Pu(VI) and reduction to Pu(IV), or by reduction to Pu(III) and oxidation to Pu(IV) under circumstances where direct reconversion to the polymer cannot occur. It is not yet certain whether or not the colloidal plutonium obtained in concentrated plutonium nitrate solutions has identical, similar, or very different properties from those of the "polymeric Pu(IV) ion." The unusual precipitation properties of the colloidal green material suggests that the formation of colloidal plutonium would give rise to difficulties in certain of the purification steps. Cunningham then reviewed the species of plutonium ion oxidation states and the nature and number of chemical groups held in stable association with the plutonium in the ion. He also presented the formal potential values for the $Pu(III) = Pu(IV) + e^-$ and $Pu(IV) + 2H_2O = PuO_2^{++} + 4H^+ + 2e^-$ couples. Solubilities of Pu(III), (IV), and (VI) compounds were tabulated.

Orlemann spoke on the distribution of plutonium between aqueous and non-aqueous phases, presenting a table of data on the extraction of plutonium from 10 M NH_3NO_3, 1 M HNO_3 solution into an equal volume of various organic solvents. He also talked about the extraction of "light" elements and about the chemistry of the oxides and halides of plutonium, taking into account the data available from work reported by Berkeley, Chicago, and Site Y.

Zachariasen reviewed the results of x-ray diffraction studies on the following plutonium preparations, giving the certainty of the assigned chemical formula, the crystal system, the number of molecules per unit cell, the lattice constants, the density, and isomorphous compounds: Pu(I), PuO_2, Pu_2O_3, PuO, PuF_3, PuF_4, $PuF_4 \cdot 2.5H_2O$, PuF_3(II)?, PuOF, $PuOF_2 \cdot 2H_2O$, $PuCl_3$, $Pu(Br_{0.8}Cl_{0.2})_3$, $PuBr_3$, PuI_3, PuOCl, PuOBr, PuOI, PuN, $Na(PuO_2)(Ac)_3$, K_2PuF_6, $PuPO_4 \cdot 0\text{-}\frac{1}{2} H_2O$. In the discussion which followed these items were emphasized:

1. Zachariasen still believes ThF_3 exists in spite of the evidence presented by Selwood against the existence of Th(III).

2. From crystal structure considerations, Zachariasen would predict lower volatility for plutonium compounds than for the corresponding uranium compounds.

3. Thus far, $NaPuO_2Ac_3$ is the only Pu(VI) compound examined by x-ray diffraction. It seems highly desirable that other Pu(VI) compounds such as PuO_2F_2 be examined.

4. Pu(III) phosphate is very similar to one form of Bi(IV) phosphate, although Pu(III) compounds are not, in general, similar to Bi(III) compounds. Pu(III) fluoride is very similar to La(III) fluoride. Zachariasen favors the view that carrying of plutonium by bismuth phosphate and by lanthanum fluoride involves Pu(III) by isomorphous substitution. Cunningham and I, however, claim there is plenty of chemical evidence for carrying of plutonium by bismuth phosphate as Pu(IV); we also believe there is much chemical evidence for carrying of plutonium(IV)

by lanthanum fluoride. I suggest carrying proceeds by the mechanism of compound formation between Pu(IV) and the carrier.

5. Zachariasen has no x-ray evidence for the existence of more than two forms of plutonium metal.

6. Zachariasen commented that the possession of the higher density by the low-temperature complex form of the metal is unusual but not impossible.

I gave a summary of my paper, "Inferences Concerning the Electronic Structure of 49 from Chemical and Physical Properties," which I dictated last Friday. I pointed out the probability that some sort of a transition group should begin in the neighborhood of the elements in the seventh period (elements 89-94). After reviewing the chemical and physical properties of neptunium, plutonium, thorium, and uranium, I suggested that:

> An attractive hypothesis is that this rare earth-like series begins with Ac in the same sense that the 'lanthanide' series begins with La. On this basis, it might be termed the 'actinide' series, and the first 5f electron might appear in Th. Thus, the ground state of Th might have the structure $5f^1 6d 7s^2$ beyond the radon core. With an 'actinide' series, U might have the electron configuration $5f^3 6d 7s^2$. It is very interesting to note that Kiess, Humphreys, and Laun in the recent report A-1747, in which they give a preliminary description of the analysis of the spectrum of neutral U atoms, come to the conclusion that the electron configuration of the lowest state of U is $5f^3 6d 7s^2$ (with the term symbol $^5L_6$), which supports the above view.

I also speculated about the chemical properties of the series members which we have not yet had an opportunity to study: $_{91}$Pa, element 96 (possible configuration $5f^7 6d^1 7s^1$) and element 95 (possible configuration $5f^7 7s^2$.

Spedding pointed out that while he agrees with me in the main, he thinks it would be extremely dangerous to depend on the existence of any super rare-earth group occurring, or that if such a group does appear that it would start at the same element for the various valency numbers. In summary, he stated:

> To sum up, due to the fact that the energy difference is rather small between whether an electron goes into the 5f or 6d and that the resonance effects are very large, the latter probably predominates in determining which level occurs lowest. Accordingly, any rule which might be derived for a new rare earth type series starting at actinium, thorium, or uranium would probably have more exceptions than regularities, and these exceptions should become more marked as the atomic number increases since the spectra becomes more complicated. I would therefore predict that it would be difficult to make any simple set of rules as to the valency states of the eka-uranium elements.

At 3:00 p.m. in Room 209, Eckhart Hall, I attended the Executive Session of the Thomas meeting, along with Allison, Chipman, Conant, Connick,

Franck, Hogness, Jeffries, Kennedy, Peterson, C. S. Smith, Spedding, Thomas, and Warner. It was mentioned that the 50-mg sample of metallic plutonium has been received by us as a loan from Site Y for vapor pressure measurements to be made in the near future. We reported good progress on the continuous solvent extraction method as an alternate concentration and isolation procedure for Hanford.

Consideration of problems proposed as a part of the future research program of the Chicago Chemistry Division was postponed awaiting some important decisions to be reached by the Project Council in their meeting to be held on Wednesday.

"Chemical Research — Special Chemistry of Plutonium, Report for Month Ending July 1, 1944," (CK-1763), was issued. The following information of interest is reported.

Purification of Plutonium Compounds (Jensen, Group Leader). Reinhardt and Stein have carried out tracer experiments on the extraction of plutonium from 10 M NH_4NO_3, 1 M HNO_3 into various organic solvents. Brody has tested a continuous batch extractor for the solvent extraction of plutonium using as solvents diethyl ether, methyl isobutyl ketone (hexone), methyl-n-amyl ketone and 2-ethylbutyl cellosolve. Dixon has used uranium as the stand-in for plutonium in a preliminary search for a suitable complexing agent for plutonium purification. Salicylaldehyde ethylene diamine complex of uranium(IV) was prepared. Attempts to extract the quinaldinac acid and the acetylacetone ethylenediamine complex of Pu(IV) from a citrate solution have not been successful.

Volatility and General Dry Chemistry (Davidson, Group Leader). I. Karle and Abraham have successfully prepared $PuCl_3$ from $PuO_2 \cdot xH_2O$ in a flow system using as chlorinating agent a mixture obtained by bubbling Cl_2 through S_2Cl_2. At 800°C this produces rapid conversion of PuO_2 to sublimed $PuCl_3$. Davidson and Hagemann have carried out liquid-phase chlorination with liquid CCl_4 in capillary glass bombs at 280°C to convert vacuum-dried plutonium peroxide to $PuCl_3$. X-ray analysis shows only the diffraction pattern of $PuCl_3$. By a similar technique, liquid S_2Cl_2 at 280°C converts $PuO_2 \cdot xH_2O$ to $PuCl_3$. J. Katz has formed $PuOCl_2$ by heating $PuCl_3 \cdot 6H_2O$ at 400-600°C in a sealed tube in vacuo.

Hagemann and Katz have been unsuccessful in attempts to prepare $PuCl_4$ by (a) treating $PuBr_3$ with liquid Cl_2 in a capillary bomb, (b) evaporation of an HCl solution of plutonium(IV) in a stream of HCl at low pressure, (c) heating in an HCl atmosphere of 200°C. Hyde has worked on the preparation and properties of plutonium bromides. $PuBr_3$ of greater than 95% purity is obtained by the hydrobromination of $PuO_2 \cdot xH_2O$ at 800°C in platinum. PuOBr is formed by heating $PuBr_3$ with calculated amounts of water vapor in a glass tube at 400°C.

Hagemann has synthesized PuI_3 by reacting plutonium metal and HI at 450° in quartz; the identification by Zachariasen is preliminary. The reaction of I_2 gas with plutonium metal in quartz and in platinum at 400°C and the reaction of I_2 liquid with plutonium metal in a pyrex bomb and in a platinum bomb at 250-400°C have yielded only PuOI. Wolf has developed an analytical procedure for iodide in which iodides are determined by conversion to iodine by iodate, distillation into KI, and thiosulfate titration.

Fluoride Chemistry (Heath, Group Leader). Florin has studied the higher fluorides. In a glass system, the reaction $UF_4 + F_2 = UF_6$ goes readily below 400°C. In contrast, PuF_4 remains unchanged even up to 500°C. UF_4 is converted to UF_6 readily by electrically activating fluorine. Conversion of PuF_4 to a volatile product is more difficult. Meyer and Zvolner have prepared PuF_3 on a half-gram scale by the wet method. They have also found that H_2 reduces PuF_4 to PuF_3 at temperatures as low as 250°C. A successful "wet" preparation of the PuF_4 was accomplished by treating Pu(IV) hydroxide with aqueous HF, followed by vacuum drying of the insoluble product, and finally dehydration in an $HF + O_2$ stream at 350°C.

Production and Properties of Plutonium Metal. (Baumbach, Group Leader). S. Katz has conducted experiments that establish the occurrence of a phase transition in plutonium (also observed at Site Y) on the basis of observed changes in density, x-ray diffraction studies, and dilatometer studies. The transition appears to occur at 135°C. Frank has carried out hardness tests on plutonium metal which show metal that is remelted is usually softer than metal obtained directly by reduction. Decreased hardness appears to follow lowering of the density.

Westrum has studied reduction of plutonium compounds to metal. Vapor phase reductions have been carried out successfully on PuF_3 with barium using crucibles of tantalum, nitrided tantalum, La_2O_3, CaF_2, and CaO. Hellman, Gerstein, and Frank have tested remelting of plutonium metal with various refractories and find that cerous sulfide and zirconium nitride appear suitable as refractories at 1,000°C. They find that graphite does not react at 1,000°C but does react at 1,300°C.

Fried and Robinson have carried out four bomb reductions of PuF_3 by means of lithium on the 30-mg scale, patterned after those performed at Site Y. Metal in good yield has been obtained. Phipps, Sears, Gilpatrick, and Simpson, using a tantalum effusion furnace, have obtained the following preliminary vapor pressures of plutonium metal:

Temperature (°K)	1,260	1,500	1,650
Pressure (10^{-4} mm Hg)	0.03	10	300

They think that these preliminary measurements definitely give the correct magnitudes of the vapor pressures and are minimum values. To get more accurate results larger amounts of plutonium will be used. A new apparatus incorporating necessary improvements is being built. These preliminary data lead to a calculated boiling point for plutonium metal of 2,260°K.

Seifert has conducted further tests of the apparatus to be used for the microanalysis for oxygen and finds essentially complete conversion to CO_2.

The "Metallurgical Laboratory Report for June 1944," (MUC-SKA-743), by Allison, with portions signed by Captain J. H. McKinley and W. C. Munnecke was issued. The report contains a summary of work by all Met Lab Divisions, personnel levels, estimated costs for June and budget estimates for July and August 1944, and a summary of outstanding obligations as of June 30, 1944. The overall Met Lab summary is as follows:

During the month of June the study of the heterogeneous heavy metal-light water system proceeded rapidly with the theoretical section of the Nuclear Physics Division at the Metallurgical Laboratory cooperating with experimental physicists at Clinton Laboratories. Lattices were tested that proved to be so close to the chain reacting condition that there is no doubt that with slight and practicable modications, a chain reacting system can be set up. The heterogeneous heavy metal-heavy water pile, which was shown to be chain reacting in May, has been modified during the month so that operation at power in early July is anticipated.

During the month a development of great interest took place in that an alteration of the isotopic constitution of an element by neutron absorption was demonstrated. Samarium, which had been exposed in the Clinton pile, was examined with the mass-spectroscope, and an increase of isotope 150 with a corresponding decrease in 149 was found. This identified 149 as the high cross section neutron absorber in samarium and has important consequences in the pile poisoning problem.

It was shown that beryllium and thorium of commercial purity can be extruded at commercial temperatures and pressures.

In chemistry, the work on the Hanford process continued to be the most important part of the program. Variations of the process that would enable the extraction of two kilograms of product at the earliest possible date were studied.

Considerable effort was placed on the development of a solvent extraction method for removal of the product from lanthanum fluoride in the final stages of the process, and indications are strong that the method will be successful.

In the work on the fundamental chemistry of our products, results on the vapor pressure of the metal were obtained. Enough Np^{237} was accumulated to permit determination of the structure of its oxide by x-ray methods.

During the month the Technical Division of the Laboratory cooperated with the Quality Hardware Company in getting under way the production of unbended slugs for Hanford. A development program on the "long cans," a scheme for installing one single aluminum tube filled with slugs and avoiding welds within the pile, was given high priority and begun during the month.

On June 15 Drexel House was made available to the Health Division for clinical laboratory examinations, thus greatly increasing the facilities for this work.

Animal experimentation on the hazards of inhaling radioactive dusts were a prominent feature of biological research during the month.

The total expenditures for the month of June are $862,254.38 as compared with $811,031.40 for May, an increase of $51,222.98. The net increase in the number of personnel employed on the project was 65 during the month of June.

A breakdown of the academic personnel employed in each Met Lab Division as of June 30, 1944, is reported in the text of the report to

be as follows: Chemistry, 237; Physics, 124, Technology, 172; Health, 69.3; and Argonne Lab, 44; thus making a total of 646.3 people.

Helen worked at the Met Lab today on the secret version of the "Table of Isotopes." I came home from work with a severe migraine headache.

The Allies seem to be making headway on all fronts in Europe, according to today's paper.

There was an evening meeting of the Project Advisory Board in Room 209, Eckhart Hall, called by Compton, preceded by a dinner at 6:00 p.m. The program was scheduled to discuss (a) postwar plans for the Project as a guide for present changes in Project policy and organization; (b) the importance of light water moderated units in the overall Project program and the effect on transfers of associated personnel required if the program is to be pushed.

The meeting, however, concentrated on something that came up which was much more immediate. Robert Oppenheimer, who was attending the Board Meeting from Los Alamos, announced that E. Segrè, O. Chamberlain, and G. W. Farwell have found strong evidence for the existence of the plutonium isotope Pu^{240}, which undergoes decay by spontaneous fission! This was found in the neutron-irradiated Pu^{239} from the Clinton pile that we had purified for them.

It should be noted that this disclosure came as a great shock to everyone. However, over a year ago in my "Report of Month Ending March 15, 1943, Special Chemistry of 94," (CK-514), I had written about the possibility of an n,γ reaction on Pu^{239} giving the isotope Pu^{240}. I had said that a possible spontaneous fission decay of this isotope would seriously impinge upon our ability to use Pu^{239} as intended. Now it was learned that indeed this reaction takes place; and that since the neutron flux in the Hanford piles would be so high, Pu^{240} would be produced in so great a relative abundance that the neutrons resulting from its spontaneous fission would overshadow those from any α,n reactions on impurities that might be present in Pu^{239}.

Because of this new development, Site Y will now have to rethink how it will proceed in the design of a plutonium bomb. Furthermore, it was decided at this Board Meeting to demobilize Thomas's staff which is handling the coordination and general direction of the chemistry, purification, and final metallurgy of Pu^{239} because the planned extreme purification of plutonium would be futile — this could not prevent the emission of the unwanted neutrons. This meant that there would be no further monthly meetings at the Met Lab on this aspect of the chemical program.

Tuesday, July 18, 1944

Paul Fields returned to Chicago at 7:30 a.m. from his trip to Clinton Laboratories where he traced the course of Np^{237} in the concentration operations in Room D and the subsequent isolation step.

Ralph James checked sample 49αA#9 (plutonium plus 32 Mev helium ions, the Berkeley bombardment) for decay of the long-range alpha particles and finds some slight indication that decay occurs.

James summarized for me, in a memo, the work done on heavy isotopes so far and in progress. He reviews the neutron bombardments of Pu^{239} at Clinton (sample 49NA), the deuteron bombardments of Pu^{239} at St. Louis, the deuteron bombardment of Pu^{239} at Berkeley (sample 49DC), the neutron bombardment of uranium at Clinton, the deuteron bombardment of uranium at St. Louis and Berkeley, and the alpha-particle bombardment of Pu^{239} at Berkeley (sample 49αA).

I attended the Project Council Information Meeting on Chemistry at 9:30 a.m. along with Allen, Allison, Apple, Ashcraft, Boyd, Burton, Chapman, Chipman, Compton, Connick, C. M. Cooper, Dempster, Doan, Elliott, Fermi, Franck, Greager, Hilberry, Hogness, Jacobson, Jeffries, W. C. Johnson, Keller, Kennedy, Kirk, McKinney, Miller, Quill, Selwood, Spedding, C. S. Smith, Sugarman, Szilard, Thomas, Turkevich, Vernon, Voigt, Warner, Wensel, Whitaker, Wigner, and Willard.

I reviewed work on Hanford problems, mentioning the very promising alternate method for isolation and concentration being investigated, namely, the extraction of Pu(IV), without the need for an oxidation step, into hexone from an aqueous layer that is 10 N NH_4NO_3. After extraction of product into the hexone, I said it is re-extracted back into 1 N HNO_3. It is expected that, in cooperation with the Technical Division, it will be possible to have the extraction equipment ready for operation by November 1 for use in testing this process. I mentioned the plans to measure the vapor pressure on the 50-mg plutonium metal sample lent by Site Y. I reviewed the studies under way on the "abnormal" form of Pu(IV). I also spoke of the isolation of 70 micrograms of Np^{237} by cold bromate cycles and the identification of the compound NpO_2 by Zachariasen, the first identification of a compound of neptunium. I mentioned that the sulfate and chloride of neptunium have also been prepared.

Sugarman outlined a method for determining the short half-lives of some of the fission gases. These experiments should give data on half-lives of the very early members of the fission chains and on the fission yield in the first part of the chain.

Allison, who was chairman of the meeting, also called on speakers from Berkeley, Oak Ridge, and Los Alamos. There was a hint at the meeting of impending changes in the plutonium purification program which, of course, intrigued me very much.

Helen spent the day at home.

According to the paper today, there is fighting in the streets of St. Lo, although political headlines have the top spot. Mississippi delegates will vote for Senator Byrd of Virginia for President.

Wednesday, July 19, 1944

At 8:00 a.m. I held a meeting of the Council of my section in my office, attended by Albaugh, Baumbach, Davidson, Dawson, Dreher, Ghiorso, Heath, Hindman, Jensen, Katzin, Manning, Pye, S. G. Thompson, Watt, and Willard. Manning pointed out that security violations are becoming too numerous again. Secondly, he noted that the contamination levels in some parts of the lab rooms are too high. I alerted the members of the Council to the impending, but still unannounced, discontinuance of the purification program and predicted that I will be notified of this later in the day. A brief summary of the program for this evening's meeting was given by Thompson. I asked that copies of these programs be sent to me, Hogness, Hyde, Arnold, and Warner.

I suggested to Ralph James that he reexamine the samples from the dichromate oxidations on the deuteron-bombarded sample (sample 49DC, 3 micrograms of plutonium that received 1,931 microampere-hours of deuteron bombardment on the Berkeley cyclotron between March 13 and March 16). I also suggested that we look for positrons in this sample. We decided that sample 49DC-H, the final sample, is the most promising for this reexamination.

Kennedy, who came here from Los Alamos for the Thomas meeting Monday and the Project Council Information meeting yesterday, met with Underhill, Conard, and me on the plutonium patent matter. It was agreed that we shall meet with Lavender in Washington on Friday, September 15. Kennedy will send me money for his round trip and Pullman fare, Chicago to Washington, so that I can get him a reservation. In the meantime, we will write up the cases and submit them to Lavender if Conard gives the OK to this procedure. Conard will get income tax advice on all possibilities and report to us by mail. Underhill sees no reason for University objection to our plan for a 50-50 split between the inventors and the University, with the University receiving its share with "strong recommendation for use in the field of research that led to the invention." Underhill will come to Washington in September with full authority to speak for the Regents.

The weekly conference on solvent extraction for product purification was held attended by Brody, Duffey, Jensen, Lawroski, Maloney, and Orlemann. It was indicated that assembly of the small-scale glass equipment in the squash courts will be completed by this Friday. Mechanical tests will then be run, followed by tests with tracer solutions.

After the Project Policy Meeting this morning, Hogness and Warner came to my office to tell me that the purification program is no longer needed. I was standing in the hall in front of my office as they approached. They said that Compton had agreed I should be given the reason but that I was not to tell others. I said they didn't need to tell me the reason — I assumed the spontaneous fission rate of Pu^{240} has been found to be so high as to overshadow the neutrons from the α,n reaction. I went on the say that, since no one has given me this information, I feel free to pass my interpretation on to my men.

"Chemical Research — Separations Processes for Plutonium," (CN-1787), by Hogness, Seaborg, Watt, C. M. Cooper, Kircher, and Miller, dated June 15, 1944, was issued. The report contains information on process development and Chicago semiworks operation resulting from work in the Chemistry Division (under S. Thompson and Dreher) and the Technical Divisions (under A. C. Hyde, Holt, and Lampert).

The Technical Division portion of the report covers semiworks dissolution, extraction, and decontamination studies, salvaging improperly jacketed "W" slugs, recovery of uranium from semiworks operations, and ether extraction as a portion of a separations process.

The Chemistry Division portion of the report covers work by Hyman, Blaedel, Margolis, Thompson, Dreher, and others and may be summarized as follows:

a. Hanford metal coatings. Studies of the conditions resulting in the formation of gelatinous precipitates in NaOH solutions that are used to dissolve aluminum from the Hanford slugs show that, with the recommended flowsheet procedure employing 15% NaOH - 15% $NaNO_3$, the aluminum concentration can be as high as 100 g/l without the formation of gelatinous precipitates. A wide range of conditions were studied; there is nothing critical about the composition of the solutions with respect to gel formation. These conclusions are based upon the relative viscosities of NaOH-$NaNO_3$ solutions (containing aluminum) at various temperatures as determined by the times required for these solutions to flow through a given orifice.

b. Extraction and decontamination — ether method. In the use of hydroxylamine as a reducing agent in the ether process, it has been found that the hydroxylamine reacts with ether solutions in such a way as to deplete rapidly the concentration of this reducing agent. Studies of the use of hydrazine indicate that it is superior to hydroxylamine in this respect. The use of $Ca(NO_3)_2$ appears to be a desirable salting-out agent for increasing the distribution of uranyl nitrate into ether.

Compton received a memo from Doan about the Clinton Laboratories research and development program after Hanford start-up. Doan made the point that we have at Clinton Laboratories a plant unique in the entire world for the study and further development of nuclear energy phenomena and the chemistry of nuclear-derived products. He urged that in the post-Hanford period a strong physics program be built up and that a moderate amount of work on alternate extraction methods be continued. He also suggested a moderate effort in investigating the possibilities of radioactive warfare using fission products.

Helen stayed at home today, waiting for a business call. In the evening she went to Wilma's where Wilma's sister-in-law, Gen (Genevieve), was visiting.

The top headline today is again political — "Louisiana Swings to Byrd" — even though U.S. troops captured St. Lo.

I attended the evening meeting of the Separation Processes Sub-

section (I) of Section C-I at 7:45 p.m. in Room 209, Eckhart Hall.

The Project Council Policy Meeting met at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Dempster, Doan, Fermi, Franck, Greager, Hilberry, Hogness, Jacobson, Jeffries, Johnson, A. V. Peterson, Spedding, Szilard, Thomas, K. Tracy, Vernon, Warner, Whitaker, Wigner, and Zinn.

Compton took up in more detail the decision reached at the meeting of the Project Advisory Board Monday night, namely, that the need for exceedingly high purity plutonium no longer exists and that the present intensive work on purification can be dropped. Hogness asked if it would be proper to inform others on our staffs that it is the properties of plutonium that have made the change in emphasis necessary; adding that he was "thinking especially of Seaborg." Compton said he believed I should know. The section chiefs at Argonne also should know, he added, but it is highly essential to limit the information to the smallest number of people. Fermi mentioned the implications of such a finding must be known in order to plan properly for the construction of new piles.

Compton stated that there is still an urgent need for chemists on war jobs both within and without our Project. He mentioned that a committee is being formed of people who are not now working on this Project to appraise future potentialities and develop plans which can be acted upon by Congress.

At Compton's request, consultant Zay Jeffries set forth his ideas as to the future of atomic energy by reading a memorandum he has prepared. He likened the nuclear field to the electronics field thirty years ago, predicting a bright future for both. He suggested the word "nucleonics" as a name for the new nuclear field. Among the possible postwar uses he identified were tagged-atom experiments in scientific studies and industrial processes, related need for electronic instrumentation, piles for manufacture of artificial radioactive substances and power. In the latter connection he speculated on the use of piles in making desert or semi-arid land productive. He discussed the possible use of nuclear detonations to relieve geological faults and to form a sea-level opening across the Isthmus of Panama. Jeffries suggested the following objectives:

"(a) The projects relating to 49 and 25 should be prosecuted by the government, no matter when the war ends, to a point sufficient for military appraisal.

(b) The development of the nucleonics industry by private enterprise should be encouraged. The military by-products of the industrial developments should be made available to the government, and the use of government information patents should be made available to industry so far as the military situation may permit.

(c) A suitable agency, with both government and non-government

representatives, should be established to guide and coordinate such nucleonics activities as may affect the military or other interests of the government."

After discussion of Jeffries' presentation, Compton expressed the Council's desire that plans for a permanent organization be pressed as rapidly and as specifically as possible. He asked Jeffries to organize a committee within the Project for preparation of a prospectus.

Thursday, July 20, 1944

Clark J. Egan, who received a Ph.D. degree from Berkeley in 1936, started work in Section C-I. I knew him there as a fellow graduate student. He has been doing research with Professor Giauque at the University of California on an NDRC contract.

Donald Ames has terminated with the Met Lab. After basic training, he will return as a member of the SED.

General Groves is in Chicago today, presumably to discuss the new turn of events — the discontinuance of the plutonium purification program.

Melvin Calvin wrote from Berkeley about two men, Richard Bailes and Lloyd Ferguson, who will be available the end of August when Calvin's contract is completed. Calvin also mentions a solvent extraction method on which he has been working.

I reviewed the personnel chart of my section, which I have been keeping up-to-date since April of this year. With the termination of the plutonium purification program, changes must be made in the organization. At the present time, however, the scientific manpower of Section C-I totals 99 (the highest to date) and is distributed as follows: S. Thompson (Extraction, Decontamination, and Isolation) — 31; Orlemann (Purification and Metal Production) — 31; Cunningham (General Problems and Service) — 32; Seaborg, Willard, Watt, Manning, Katzin — 5. The organization is as follows:

Glenn T. Seaborg — Section Chief
Edrey Smith — Secretary to Section Chief
Kathryn Buehler — General Secretary to Section
John E. Willard — Associate Section Chief (now with du Pont)
Winston M. Manning — Associate Section Chief
George W. Watt — Associate Section Chief
Leonard I. Katzin — Assistant to Section Chief
Irma Saxton — Secretary to Associate Section Chiefs

Sub-section I — Separations Processes

Stanley G. Thompson — Assistant Section Chief
Dorothy Gottlieb — Secretary

Group 1 — *Extraction-Decontamination*

Albaugh, Frederic W. — Group Leader
Ader, Milton — Research Assistant
Bartell, Lawrence S. — Research Assistant
Bradt, Rexford H. — Research Associate
Egan, Clark — Research Associate
Greenlee, Roy W. — Research Assistant
Hoekstra, Henry R. — Research Assistant
Malm, John G. — Research Assistant
Morgan, Leon O. — Research Assistant
Peterson, Sigfred — Research Assistant
Post, Roy G. (on loan to Semiworks) — Research Assistant
Sedlet, Jake — Research Assistant
Thompson, Roy — Research Associate
Summers, Mildred — Technician

Group 2 — *Concentration-Isolation*

Pye, Donald G. — Group Leader
Beard, Walter C. — Research Assistant
Goeckermann, Robert — Research Assistant
Haeckl, Frank W. — Research Assistant
Hopkins, Horace — Research Assistant
Katz, Joseph J. — Research Associate
Kelley, Alec E. — Research Assistant
Walling, Matthew T. — Research Assistant
Yett, Fowler R. — Research Assistant

Group 3 — *Process Development*

Dreher, J. Leonard — Group Leader
Blaedel, Walter — Research Associate
Gilbreath, James R. — Research Assistant
Hyman, Herbert — Research Assistant
Larson, Raymond G. — Research Associate
Lincoln, Dwight C. — Research Assistant
Rasmussen, Robert W. (on loan to Semiworks) — Research Assistant
Winner, Bernard — Research Assistant
Boykin, P. — Technician
Freeman, J. Elaine — Lab Assistant
Parnell, Alice — Librarian

Sub-section II — Purification and Metal Production

Edwin F. Orlemann — Assistant Section Chief
Eda Kelley — Secretary

Group 4 — *Purification and Analysis*

Jensen, Lyle H. — Group Leader
Brody, Bernard B. — Research Assistant
Dixon, Jonathan S. — Research Assistant
Lawroski, S. — Research Associate
Reinhardt, Richard A. — Research Assistant
Stein, Amanda P. — Research Assistant

Group 5 — *Volatility and General Dry Chemistry*

Davidson, Norman R. — Group Leader
Abraham, B. — Research Assistant
Hagemann, French T. — Research Associate
Hyde, Earl — Research Assistant
Sheft, Irving — Research Assistant
Wolf, Michael — Research Assistant
Pellock, Helen — Technician

Group 5A — *Fluoride Chemistry*

Heath, Roy E. — Group Leader
Florin, Alan E. — Research Assistant
Meyer, Fred — Research Associate
Zvolner, Hyman P. — Research Assistant
Nyden, Shirley — Technician

Group 6 — *Metal Production*

Baumbach, Harlan L. — Group Leader
Frank, Ray — Research Assistant
Fried, Sherman — Research Associate
Gerstein, Melvin — Research Assistant
Hellman, Nison N. — Research Assistant
Jasaitis, Zene V. — Research Associate
Katz, Sidney — Research Associate
Robinson, Herman — Research Associate
Westrum, Edgar F. — Research Associate
Watts, Ellen — Technician

Group 6A — *High Vacuum*

Simpson, Oliver C. — Group Leader
Gilpatrick, Louis — Research Assistant
Johnson, Frederick — Research Assistant
Phipps, Thomas E. — Research Associate
Sears, Gerald — Research Associate
Erway, Norman D. — Glassblower

Sub-section III — Basic Chemistry and Service

Burris B. Cunningham — Assistant Section Chief
Marjorie Bohlmann — Secretary

Group 7 — *Basic Chemistry*

Hindman, James C. — Group Leader
Ames, Donald P. — Research Assistant
Howland, Jerome J. — Research Associate
James, Ralph A. — Research Assistant
Kraus, Kurt — Research Associate
La Chapelle, Theodore — Research Assistant
McLane, Keith — Research Assistant
Magnusson, Lawrence — Research Assistant
O'Connor, Paul — Research Assistant
Smith, Clifford — Research Associate
Billington, Hubert — Technician

Group 8 — *Recovery*

Dawson, Lyle R. — Group Leader
Anderson, Herbert H. — Research Associate
Asprey, Larned B. — Research Assistant
Baumbach, Nathalie S. — Research Assistant
Britain, J. W. — Research Assistant
Burr, John G. — Research Assistant
Fields, Paul — Research Assistant
Fineman, Phillip — Research Assistant
Leventhal, Leon — Research Assistant
Stewart, Donald C. — Research Assistant
Studier, Martin H. — Research Assistant
Wetlaufer, Donald B. — Technician

Group 9 — *Instruments and Physical Measurements*

Ghiorso, Albert — Group Leader
Crawford, John A. — Research Assistant
Golden, Lorraine — Research Assistant
Hufford, Duane — Research Assistant
Jaffey, Arthur H. — Research Associate
Kohman, Truman P. — Research Associate
Krueger, Albert C. — Research Associate
Scott, Benjamin F. — Research Assistant
Walsh, Patricia — Research Assistant
Weissbourd, Bernard B. — Research Assistant
Lewis, Eleanor — Technician

Non-Academic Service Group

Freeman, E. M. — Lab Assistant
Gavin, K. — Clerk
Jennings, N. — Lab Assistant
Krinsky, Jerome — Lab Assistant
Parsons, A. — Lab Assistant

Dieners

Cunningham, Ida
Dixon, Doris
Hubbard, Cardea
Johnson, Carrie
Knowles, Martin
Ouderkirk, Floyd
Prothrow, Annie
Schroeder, Emil
Slattery, John
Tadnac, Mike
Walker, L.

Luther Arnold and I played 13 holes of golf at Jackson Park before dark. Scores were LA —43, GS —53 for 9 holes; LA —62, GS —73 for 13 holes.

Helen visited the Dunes area of Lake Michigan south of Chicago today in order to locate a suitable place for us to take a short vacation. This evening she had a golf lesson at the downtown YWCA.

Democratic Convention news crowds today's headlines again. Indications are that there will be a battle for the vice-presidential nomination.

Friday, July 21, 1944

Following my suggestion, Ralph James checked for positrons in the deuteron-bombarded sample 49DC-H (final sample from working up the 3 micrograms of plutonium that had received 1,931 microampere hours of deuterons on the Berkeley cyclotron between March 13 and 16). The sample was counted on the magnetic deflecting G-M counter, and the radiation was shown to be composed entirely of negative particles with no detectable positrons.

Westrum gave to Zachariasen for x-ray examination a sample of plutonium that has been prepared in an attempt to reduce PuF_3 with $CaSi_2$.

I received a copy of a letter from Hogness to Greenewalt, transmitting a report on the hexone solvent extraction process for concentrating and isolating plutonium. He invited Greenewalt's comments and criticisms.

I sent a nine-page outline to Hogness detailing the proposed research program for Section C-I for the period October 1, 1944 to July 1, 1945 (MUC-GTS-875). As a basis for drawing up the program, I assume that full time of 75 men will be available to the section (present personnel totals 99). The work for the sub-divisions is presented as follows:

I. Separations Processes — *19 men*
   1. Hanford support.
   2. Alternate processes (precipitation, solvent extraction).
   3. Separation of plutonium fission products from plutonium.
   4. Recovery of uranium.

II. Properties, Preparation, and Separation of U-233 — *6 men*

III. Basic Chemical Research on Elements of Atomic Number 89 and Higher — *30 men*
   1. Actinium
   2. Thorium
   3. Protactinium
   4. Uranium
   5. Neptunium
   6. Plutonium (Oxidation states, oxidation potentials, complex ions, inorganic compounds, hydrolytic behavior of Pu(III), (IV), (VI), organic compounds of plutonium, rates of oxidation and reduction, specific activity of Pu^{239}, co-precipitation studies, basic dry chemistry, basic chemistry of plutonium metal, service production of plutonium metal.)
   7. Production and study of new, heavy isotopes including elements 95 and 96 (mostly to be produced by bombardment with high-energy particles in the new Berkeley 60-inch cyclotron).

IV. Services — *20 men*
   1. Development and maintenance of instruments, recovery and repurification of plutonium, isolation of natural Pa^{231} for Site Y.

Hogness asked Compton for help in obtaining information on naval power units (e.g., submarines, destroyers, cruisers, and battleships) for the informal Power Committee chaired by Allison. He said that Allison has asked him to obtain such information as it would be useful for those Lab people who are now giving thought and attention to the development of new sources of power, such as nuclear.

Helen worked at the YWCA today. She and I listened to the Democratic National Convention on the radio and then, on the spur of the moment, decided to go by streetcar to Convention Hall at the Coliseum on the south side of Chicago where we saw Harry Truman make his acceptance speech for the Vice Presidential nomination. We managed to just walk in on the Convention and found a place quite close to Truman as he gave his speech. Then, we went to the movie, "Show Business" featuring Eddie Cantor, George Murphy, and Joan Davis, back at the Picadilly Theatre at 51st and Blackstone.

Saturday, July 22, 1944

In view of the termination of the Metal Production program, Melvin Gerstein is accepting a position on another University of Chicago war contract.

During the morning I held a meeting in my office to discuss the status of the solvent concentration and extraction process, attended by Dawson, Lawroski, Maloney, Orlemann, Stewart, and Watt. Maloney reported that a place has been arranged for setting up the extraction columns in the south stairwell at the West Stands. The three-inch pyrex pipe is expected to arrive this week. Stewart reported on the status of laboratory-scale research on the extraction process.

Ralph James made absorption measurements today on the alpha particles from the following chemical fractions that might contain element 95 from (a) plutonium plus deuterons, Berkeley bombardment — sample 49DC-H, (b) plutonium fraction from plutonium plus 32 Mev helium ions, Berkeley bombardment (49αA), (c) 49αA#9, chemical fraction in which element 95 or 96 was identified, (d) 49αA#12, reworked residues to obtain a second 95 or 96 fraction from plutonium-helium ion bombardment from which 49αA#9 was derived. He concludes (1) 49DC-H does not have any long-range alpha particles, (2) the long-range alpha particles (95 or 96) in 49αA#9 are not decaying, (3) there is about the same amount of 95 or 96 in sample 49αA#12 as there is in sample 49αA#9.

Sheft began the experiment to prepare PuI_3 by the wet method, using the apparatus he constructed starting July 13. He will add ammonium iodide to a plutonium(IV) chloride solution to reduce the plutonium to the III state and provide excess ammonium iodide. The solution will be heated in an atmosphere of anhydrous HI at controlled pressures to drive off the water. Then, the ammonium iodide will be sublimed away at 350°C, hopefully leaving a residue of PuI_3. A 3.6 mg amount of plutonium is being used.

I received an outline from Fields and Dawson of a batch hexone solvent extraction method that has been devised for separating 93^{239} from fission products in the bismuth phosphate precipitate of the second decontamination cycle. The method has been worked out at the suggestion of Willard, who feels it is necessary to remove the 93^{239} so that the decontamination factors for the fission products can be determined accurately in succeeding process operations.

I received a letter from H. G. Hawkins, Jr., of the Area Engineer's Office, who is Chief of Intelligence and Security, saying that he has been told by higher authority that there is no objection to my giving a lecture on uses of radioactive tracers at Northwestern University on December 6, under the following conditions: (1) The lecture must be attended by a representative of the Manhattan District (for my own protection) and (2) the Area Engineer should receive copies of any printed announcements.

Helen took a walk with Wilma today.

Sunday, July 23, 1944

I played 18 holes of golf at Cog Hill No. 1 with Helen, Zene Jasaitis, and Herman Robinson. Herman scored 129 while I scored 98 (this is the first time I have succeeded in breaking 100.) Snapshots were taken of Helen hitting the ball and of Zene, Helen, Herman, and me in a group at the golf course (see Figs. 8 and 9). Zene joined us for dinner at Normandy House, 800 N. Tower Court (facing N. Michigan at Chicago Avenue), after which we went to the Chicago Theater on N. State Street and saw the movie, "Going My Way" with Bing Crosby and Rise Stevens, as well as an excellent performance by the Four Ink Spots on the stage.

Lyle Dawson, Steve Lawroski, and J. O. Maloney are leaving for Site X at 11:55 tonight for a conference at Clinton Labs with Squires and others on Tuesday and Wednesday to discuss the prospects of using the hexone solvent extraction process in either the 224 (Concentration) building or 231 (Isolation) building at Hanford.

Monday, July 24, 1944

Charles Kraus of Brown University is visiting the Met Lab today and tomorrow.

I replied to Calvin's letter, explaining that we are no longer adding academic personnel since one phase of our work has been terminated. I suggested that he send me the information about the solvent extraction method via the regular registered secret letter.

Ralph James began purifying sample 49αA#12 to eliminate the

Figure 8. Helen Seaborg at Cog Hill Golf Course. July 23, 1944. Glenn Seaborg in background. XBB 780-14618

Figure 9. Zene Jasaitis, Helen Seaborg, Glenn Seaborg, and Herman Robinson at Cog Hill Golf Course. July 23, 1944. XBB 769-8626

plutonium activity. The plan is to use this sample from the plutonium (plus 32 Mev helium ions, Berkeley bombardment) to study the chemistry of element 95 or 96, while sample 49αA#9 is to be kept for measurement and observation of decay properties.

I received a copy of a memo from Zachariasen to Allison stating that he has identified $PuSi_2$ with certainty in the sample received from Westrum last Friday; the compound is tetragonal with four molecules per unit cell. Preliminary values for the lattice constants are a_1 = 3.96 Å and a_3 = 13.50 Å. The sample was prepared by Westrum in an attempt to reduce PuF_3 with $CaSi_2$.

In a letter to Frost, Department of Chemistry, Northwestern University, I say that I will be able to give the December 6 lecture on uses of radioactive tracers, about which he wrote me in his letters of June 7 and June 22. I explain that for security reasons I will have to be represented as coming from the University of California and no mention should ever be made of my connection with Met Lab.

Helen went downtown in the evening with Wilma while I attended the Chemistry Division seminar.

War news is back in the headlines. U.S. troops are battling their way in Pisa, and the Soviets have entered Lublin, Poland, and are fighting in the streets.

Tuesday, July 25, 1944

Ralph James continued the purification of sample 49αA#12. His first attempt at an oxidation cycle using silver and persulfate to remove plutonium was unsuccessful. In order to start over, he combined the appropriate fractions, reduced the solution, and threw down lanthanum fluoride. He checked both the precipitate and the supernatant for the long-range alpha particles that signify the presence of 95 or 96 and found none, whereupon he recorded in his notebook, "Where the hell are the long-range alphas?" He then recounted all the samples from the persulfate cycle and found the missing alpha particles. He began combining and fuming down all the persulfate samples.

Sheft completed his experiment to prepare PuI_3 by the wet method. Part of the green solid product resulting from the experiment was transferred to an x-ray capillary (in a dry box) and turned over to Zachariasen for analysis.

I received a copy of a 13-page memo from Hogness to Allison (MUC-TRH-138) giving the future plans for the Chemistry Division. He incorporates rather completely what I gave him in my July 21 memo (MUC-GTS-875) but did leave out the manpower estimates I made. In his summary, Hogness indicates that the men held in stand-by condition for Hanford will be occupied for the large part in process improvement and

basic chemistry studies for the Hanford process and in solvent extraction studies. He states that the latter subject will require a semiworks group but that it is not contemplated that such a group will be needed any longer for the precipitation or Hanford process studies as the work in this field can be better carried out at Clinton. Hogness also states his belief that we are equipped with personnel and facilities to carry out laboratory work on the extraction of U^{233}, the development work and large-scale extraction to be done at Clinton. He ends his summary by stating that, with the closing out of most of the work for Site Y, the Chemistry Division will be reorganized into five major groups: (1) Extraction Studies, (2) Basic Chemistry, (3) Radiation Studies, (4) Fission Product Chemistry, and (5) Analytical Services. The first two of these groups will be under my direction; the third, under Milton Burton; the fourth, with Nathan Sugarman; and the fifth, under D. S. McKinney. The present academic personnel in the Chemistry Division now numbers 235, and this will be reduced to 150-160 by about October 1 of this year.

I asked Hogness, by memo, to arrange to get lanthanum fluoride-plutonium slurries from the forthcoming Clinton plant runs in order for us to continue our tests on the Hanford process. I asked that the quantity of the slurry be such as to contain 2 grams of plutonium. We want the materials to test the Hanford peroxide isolation process as well as for work on the alternate isolation process involving solvent extraction.

Kohman sent a memo to Jesse asking whether C. W. Schaeffer of the University of Chicago Chemistry Department can assist in an experiment to test the effectiveness of a carbon coating in reducing the background in neutron counters caused by alpha contamination.

It is now decided that Stan Thompson rather than Len Katzin will go to Montreal to discuss the work there, including the processing of two irradiated thorium carbonate slugs that we sent to Montreal from Clinton early this month.

Ghiorso, Katzin, and I played nine holes of golf before dark at Jackson Park. Scores were LK — 63, AG —55, and GS —51.

Since the Baumbachs, Len Dreher, and Stan Thompson are about to leave Chicago, the UCLA group of chemists had a "reunion-picnic farewell" today (see Fig. 10).

Alice Thompson and her baby Ruth left to visit her parents near San Diego, on their way to their move to Hanford.

Banner headlines today indicate that Lublin has fallen to the Soviets.

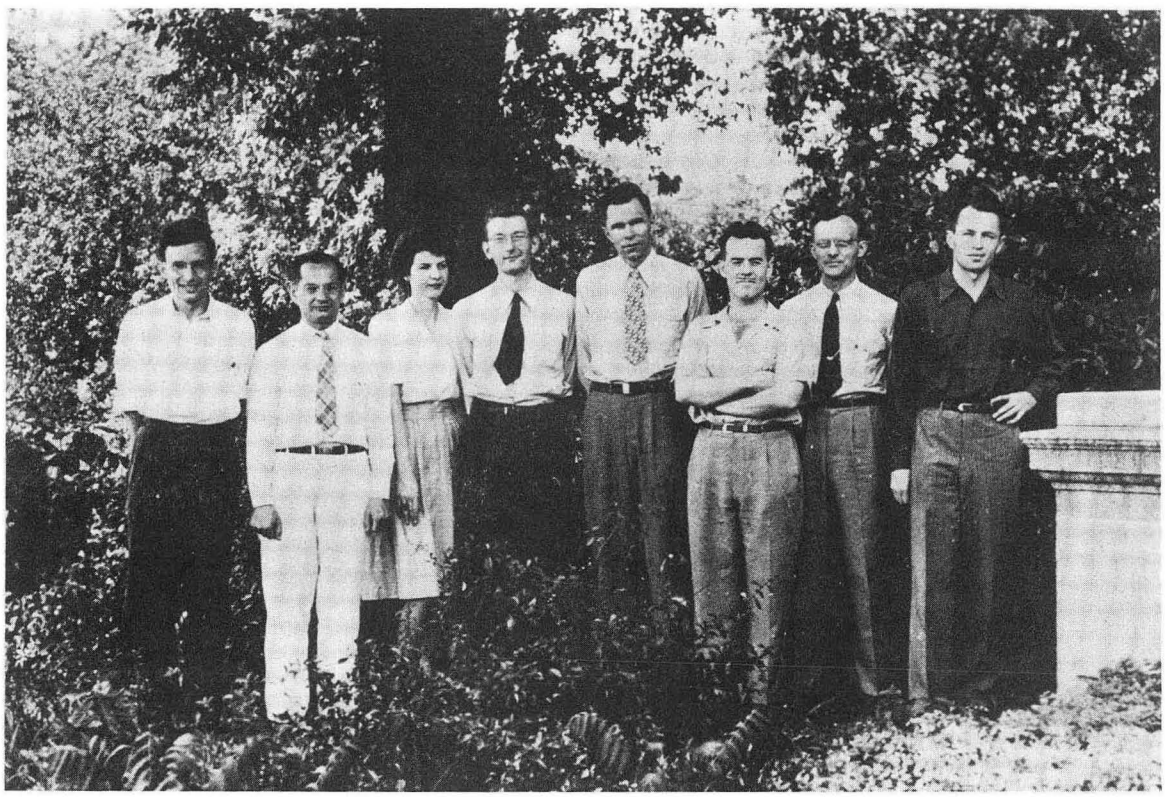

Figure 10. UCLA Group, from left to right, Leonard Katzin, Zene Jasaitis, Nathalie Baumbach, Harlan Baumbach, Glenn Seaborg, Stan Thompson, Leonard Dreher, and Fred Albaugh. July 25, 1944.

XBB 769-8627

Wednesday, July 26, 1944

At 8:00 a.m. I held a meeting of the Council of my section in my office, attended by Albaugh, Cunningham, Davidson, Dreher, Ghiorso, Gilbreath, Hindman, Jensen, Katzin, Kraus, Manning, Nickson, Simpson, S. Thompson, and Watt. I noted that since the purification sub-section is being disbanded, the following reorganization is taking place: (a) To Cunningham's sub-section has been added a basic dry chemistry group with Simpson as group leader and Davidson as assistant group leader. (b) A solvent extraction group has been formed in S. Thompson's sub-section. Lawroski is the group leader. (c) Katzin has been placed in charge of the U^{233} work. His rank is that of Assistant Section Chief, and he will report directly to me. (d) Kraus has been made assistant group leader in the basic chemistry group under Hindman. (e) Gilbreath, J. Katz, and R. Thompson have been made assistant group leaders in those groups now headed by Albaugh, Pye, and Dreher. (f) At the time of Stan Thompson's move to Hanford, Albaugh is to become Assistant Section Chief, replacing Thompson. (g) Orlemann is to take Watt's job as Associate Section Chief. His duties are to begin immediately, although Watt will not leave for Hanford until October.

Nickson reviewed the health hazards we are subjected to in working with plutonium. Animal experimentation supports the original premise that the problem is serious. In brief, about 4 micrograms is a tolerance dose for man, either in the bones or in the lungs. It is desirable to keep hand counts below 0.01 microgram and face and nose counts below 100 c/m. Gloves should be worn when more than 50 micrograms of plutonium are handled.

James completed fuming down all the samples from the persulfate cycle on sample 49αA#12 (plutonium plus 32 Mev helium ions, Berkeley bombardment). He went through a dichromate oxidation cycle to remove plutonium, and he and Krueger began counting the resulting lanthanum fluoride precipitate (sample 49α#17), hopefully containing the non-oxizable 95 or 96, on the low geometry counter and in the nitrogen chamber.

Kohman transmitted to E. H. Wakefield his outline on instrumentation and techniques for use in writing Chapter VII of the Project Handbook.

At 8:00 p.m. Stan Thompson left for Montreal where he will meet with Goldschmidt, Guéron, and others to discuss the work being done at Montreal on the British-Canadian atomic energy project. He will be back next Monday.

British and U.S. troops have launched twin drives in Normandy, according to today's newspaper.

Thursday, July 27, 1944

Clark Egan transferred to Technology to eventually replace A. C. Hyde.

Dawson, Lawroski, and Maloney returned to Chicago at 7:30 a.m.

from a trip to Clinton Laboratories where they met on Tuesday and Wednesday with Doan, Kay, D. M. Smith, Squires, and others to discuss for possible alternative uses in the plutonium concentration or isolation step, the proposed hexone solvent extraction process and the problem of selecting equipment which will fit into either the 224 (Concentration) or 231 (Isolation) building at Hanford.

I sent Allison a description of a method and apparatus for making fission control analyses for plutonium as it leaves Site W; the description was prepared by Kohman. The general method we prescribe is to measure in a parallel-plate ion chamber the fissions of an aliquot of the product solution when exposed to a known neutron flux.

Katzin, Ghiorso, Arnold, and I played 13 holes of golf before dark at Jackson Park. Scores were LK −58, AG −52, LA −46, GS −49 for nine; AG −71, LA −64, GS −67 for thirteen

Helen had an evening golf lesson at the downtown YWCA.

U.S. tanks have taken the highest peak on the island of Tinian in the Marianas is today's report.

Friday, July 28, 1944

Because of the termination of the Metal Production program, Ray D. Frank has resigned. He is going to work for the Aluminum Company of America Research Laboratories but will continue his schooling at Carnegie Tech.

James and Krueger, by extrapolating to zero absorber the absorption curve made from the alpha-particle counts taken with the nitrogen chamber, find a value of 23 counts per minute. This is much less than was expected from sample 49αA#17 (plutonium plus 32 Mev helium ions, Berkeley bombardment). James has therefore gone back to the supernatant from which sample 49αA#17 was precipitated, adding SO_2 to reduce the plutonium (and any element 95 or 96 present), and is leaving it overnight.

I received a copy of a memo from Hogness to Burton stating we have decided to press our solvent extraction work with hexone rather hard in the hopes that we may be able to get a concentration process into Hanford operation before the end of the war. In view of this, Hogness asks that Burton lay out an investigation as to the possible dissocation of hexone by radiation from alpha and beta particles and gamma-rays.

Paul Fields sent me a summary of the results of his visit to Clinton Labs between July 9 and July 18 to trace the distribution of neptunium in the Clinton concentration and isolation procedure. (Although it is known from experiments conducted here that about 85% of the neptunium is discarded in the waste in the first decontamination cycle, a part of the remainder should be available for recovery.) Using Np^{239} added as

a tracer, Fields found that 90% of the remaining neptunium follows the plutonium all the way to the final plutonium nitrate in the concentration and isolation steps in Room D.

Baumbach received a copy of a memo from Zachariasen to Allison giving the results of an examination of a sample Westrum prepared by lithium reduction of PuF_3 in a graphite crucible. Zachariasen found three phases, one of which he believes to be PuC, cubic face-centered with a lattice constant $a = 4.910 \pm 0.005$ Å. The calculated density is 13.99 gm/cm^3. He also found some evidence for Pu_2C_3, but the diffraction lines are weak.

I received a July 27 letter from Mulliken, asking me to comment on Zay Jeffries' memo to Compton on the future of nucleonics (presented at the July 19 Project Council Meeting). Mulliken asked me to jot down my own ideas and speculations and send them to him for transmittal to Jeffries, who has been appointed as chairman of a committee to prepare a prospectus in simple language for reference by the people who are working on plans for support of a postwar program continuing the work of this Project.

I informed Stearns that Baumbach, group leader of Group 6, the Metal Production Group, has completed his assignment in an extremely satisfactory manner. In view of the termination of this program he has decided to return to Paramount Pictures in Hollywood rather than accept another assignment here. His last day of service will be August 15. I asked Stearns to write a letter to Paramount commending Baumbach for his excellent work.

Allison sent a memo to Compton transmitting a review of the Met Lab activities in the four Divisions: Chemistry, Health, Physics, and Technical, as anticipated for a period beginning about October of this year. The summarizing comments are as follows:

"The Hanford studies, which are the items of highest priority, will be almost entirely laboratory-scale problems on process improvement. We anticipate that work on these problems at semiworks scale will be done at Clinton. Alternate processes for Hanford, such as the promising solvent extraction (hexone) method for plutonium recovery, will be carried to the semiworks state at Chicago, however. Certain studies on the effect of pile radiations on materials contemplated for Hanford, such as the current study of dichromate reduction, will also be of highest priority.

"Probably next highest priority will be accorded studies of the basic chemistry of plutonium and to methods of extraction of 23 studied on a laboratory scale. The recent abandonment of attempts to reach high purity in our product has, in my opinion, increased somewhat the interest in 23. This is because we are now committed to only one method of final assembly; and if this should fail, the military usefulness of our product is doubtful. Under these conditions we would immediately give consideration to the conversion of our product to 23. Work on extraction of a few milligrams of 23 to be tested at the Argonne Laboratory will probably carry over into the post-October period.

"In this same priority would be the preparation and chemical isolation (if possible) of Pu^{240} and U^{236}. We now know that the former will be a constituent in our product, and the latter will be produced in our piles and in the final gadget. These atomic species in a pure state should be presented to the physicists for study as soon as possible.

"Work of the third priority will be carried out on the fission products, the chemistry of $_{91}Pa$, $_{93}Np$, and of elements 95 and 96 if they can be produced. Continuation of radiation studies on graphite and pile structural materials will probably be in this category a few months after Hanford start-up."

The review incorporates the Section C-I material I gave to Hogness in a memo last Friday, as modified by Hogness and transmitted to Allison. Allison also incorporates the summary material Hogness has provided for the other sections of the Chemistry Division.

Helen had lunch at Dagmar's. Helen and I had dinner downtown at Gus' Restaurant at 420 N. Dearborn with Zene Jasaitis and the Westrums, after which we went to the Riverview Amusement Park and went on a number of rides.

Both eastern and western fronts in Europe are in the news today. U.S. troops have made new gains in France, and the Soviets have captured six German bases, the fortress cities of Lwow, Bialystok, and Stanislawow.

Saturday, July 29, 1944

Ralph James attempted to further eliminate the plutonium from sample 49αA#17. (This was the sample from the plutonium plus 32 Mev helium ions, Berkeley bombardment, which he and Krueger counted on the 26th and 27th and found to contain less of the long-range alpha activity than was expected.) He went through a dichromate cycle and counted the resulting lanthanum fluoride precipitate (labeled 49αA#21). He now finds that instead of purifying the sample, he contaminated it with additional plutonium.

I held a meeting in my office to discuss the status of the solvent extraction process, attended by Buffum, Dawson, Lawroski, Maloney, Manning, and Watt. Dawson, Maloney, and Lawroski reported on the conferences at Clinton on July 25 and 26 when it was decided that a feasible process is to be developed at Chicago and checked at Clinton before possible introduction as an alternate method at Hanford for use in the concentration step. Further, an investigation will be made of the feasibility of using a batch process that would use the tanks now proposed for installation in the 224 and 231 buildings at Hanford. It was agreed that Dawson will advise Hogness of the quantities of plutonium necessary for carrying out all the experimental work. Someone pointed out that it would be of great help to know what the specifications of purity should be for plutonium in final form before shipment to Site Y. I stated that no such specifications have been settled on yet. It was also reported that study of the process on the small glass columns

is well underway. Most of the equipment is ready for setting up the 3-inch glass columns, and the scaffolding is being erected.

Helen and I went to the Frolic Theater and saw the movies "King Kong" with Fay Wray and Bruce Cabot and "Iceland" with Sonja Henie and Jack Oakie.

The headline today reads "Nazis Fleeing Normandy!" Admiral Chester Nimitz announced that the U.S. flag is again flying over Guam.

Sunday, July 30, 1944

I played 18 holes of golf with Arnold and Hagemann at Hickory Hills (FH −108, LA −106, GS −107). Then, while Arnold took flying lessons, Hagemann and I played an additional seven holes at Walnut Hills Country Club (FH −47, GS −42). Arnold drove us to both golf courses.

Monday, July 31, 1944

Ralph James put sample 49αA#21 through another dichromate cycle to eliminate the plutonium, including the additional plutonium contamination picked up in the previous dichromate cycle (sample 49αA#21 is from the plutonium plus 32 Mev helium ions, Berkeley bombardment). He picked up still more plutonium contamination that he finally eliminated with yet another dichromate cycle. He labeled this sample 49αA#23.

Stan Thompson returned to Chicago at 8:20 a.m. from his trip to Montreal, Canada. He told me that he spent considerable time with Goldschmidt and Guéron discussing the work being carried on at the Montreal laboratory. He described to me the procedure used there to extract the U^{233} from the two irradiated thorium carbonate slugs received from Clinton early in July — an ether extraction method similar to that used by Stoughton at Oak Ridge. They obtained about 8 to 9 mg total uranium which may or may not be fairly pure U^{233} — analyses are not completed.

Although Thompson was not permitted to discuss plutonium work, they did volunteer information about what they have accomplished. Using about 300,000 counts per minute of Pu^{238}, they have carried out much work on the use of bismuth phosphate as a carrier. Their rather ingenious method involves precipitating bismuth phosphate from 20% UNH solutions in the presence of a small amount of zirconium. The extraction precipitate is then dissolved in 1 N HCl, and a zirconium phosphate precipitate containing the plutonium is separated. The zirconium phosphate precipitate is then contacted with 9 N HCl that elutes the plutonium. Goldschmidt has studied the mechanism of carrying by bismuth phosphate and concludes that carrying is probably by the mechanism of adsorption; however, his data are similar to ours, which we have interpreted as not necessarily indicating adsorption.

Thompson is of the opinion that it would aid their microchemical work considerably if our microchemists could spend a few days at the Montreal Laboratory. He also suggested we send them some micro lusteroid cones. He was told of plans for building a pile at Petawawa, which is about 110 miles from Ottawa. This pile will be operated with approximately 13 tons of heavy water and about 8 tons of uranium, the structure will be built of aluminum, and the uranium slugs inserted in the channels so that water can flow between the wall of the channel and the uranium cans. There will be a 1/8" aluminum jacket on the cans. There will be extra holes in the pile for inserting thorium to produce U^{233}, which is central to their program. The quantity of thorium and the form in which it is to be used have not been decided yet, although there is considerable talk about using thorium metal.

I received a nine-page memo from Albaugh summarizing the work of the past months in his sub-section on Separations Processes. His report covers the following: In Prereduction, Dreher has studied percent oxidation and consequent reduced extraction yield as functions of time of storage between dissolver and extraction. Hoekstra is trying hydroxylamine as a prereducing agent. Early operations — metal processing: A procedure for processing four tons of uranium per day has been tested in four 100-ml scale runs. Results are good. Some small-scale tests were made following my suggestion that use of an increased quantity of bismuth in the plutonium extraction precipitation step of the first cycle and precipitation at 2 N HNO_3 would be preferable to the use of a buffering agent. Results are promising, and Dreher is about to make some one-liter scale tests.

Decontamination: A number of decontamination methods have been investigated with the aim of eliminating the need for cerium-zirconium scavengers. Basic chemistry of the process: Howland has followed the oxidation of plutonium (10 times W concentration) in 40% UNH - 0.2 N HNO_3 solutions at 25°C as a function of time and finds no evidence of the abnormal form of Pu(IV) and no disproportionation to form Pu(III). Pu(VI) phosphate is found to have a solubility of at least 370 mg/l in 0.03 N HNO_3 - 0.6 M H_3PO_4 - 0.1 N $KBrO_3$ at 25°C. This indicates it should not precipitate during low acidity by-product precipitations.

Concentration-isolation at Site W — fluoride methods: Modifications in the HEW flowsheet have been tested. The problem of eliminating iron from process solutions, should this become necessary, has been studied. The feasibility of employing a lanthanum fluoride cycle at high concentrations of plutonium has been demonstrated. Experiments have been initiated to determine the structure of plutonium peroxide. Filtration methods of separating the plutonium peroxide precipitate have been investigated. Concentration at Site W — alternate methods: Work on the $U(C_2O_4)_2$ alternate concentration-isolation procedure has shown uniformly good results. Concentration-isolation — hexone extraction method: In the proposed process an aqueous solution is obtained by complexing the lanthanum fluoride plutonium precipitate at the end of the crossover cycle with zirconyl nitrate and making the resulting solution 3 M in NH_4NO_3 and 3 N in HNO_3. The plutonium in this solution is then extracted with hexone and subsequently removed from the hexone with water. The equipment required consists of two columns, one 29 feet and

one 21 feet tall. Both are three inches in diameter and packed with Raschig rings. A still and condenser are also required for final concentration of the plutonium.

Albaugh in his memo gave present work assignments of members of the Separations Process Sub-section, including nine people reassigned from the disbanding Purification and Metal Production Sub-section. Members of the three groups were listed as follows:

Group 1. Albaugh (Leader), Ader, Bartell, Bradt, Greenlee, Hoekstra, Howland, Malm, Morgan, S. Peterson, and Sedlet.

Group 2. Pye (Leader), Beard, Goeckermann, Haeckl, Hopkins, J. Katz, Kelley, Meyer, Walling, Wolf, and Yett.

Group 3. Dreher (Leader), Blaedel, Hyman, Larson, Lincoln, and Winner.

At Hanford the 200 T area (chemical separations area) is being rapidly completed. The final end wall of the canyon is being poured which practically completes the concrete work. It is expected that equipment will begin to be installed this week. In the 224 building (concentration building, analogous to Clinton Room D), equipment is now being installed.

Helen worked at the Met Lab with Truman Kohman on the secret version of the "Table of Isotopes."

The British have launched a new drive in Normandy and are making gains.

Tuesday, August 1, 1944

James put sample 49αA#23 (a sample derived from plutonium bombarded with 32 Mev helium ions at the Berkeley cyclotron) through another dichromate oxidation cycle to further purify it from plutonium. The resulting precipitate was mounted on a platinum disc and labeled sample 49αA#24. The results obtained from counting the sample on a low-geometry chamber gave little evidence of the presence of elements 95 or 96. James concludes that the long-range alpha-particle activity he is looking for must be in the supernatants from the last four dichromate cycles. Therefore, he has combined the supernatants and added SO_2 to reduce the solution.

Zachariasen's x-ray diffraction analysis of the sample Sheft gave him on July 25 reveals no PuI_3 present. The sample is mostly PuOI, with some PuO_2. Sheft is making another attempt with a 3.6-mg sample of plutonium to prepare PuI_3 using the wet method as before.

A request was made today for the transfer of Sidney Katz from our section to Section P-VII under Gurinsky, and Robert Rasmussen's transfer to Technology (A. C. Hyde) is effective today. Aquilla Parsons, one of Kay Gavin's assistants, terminated.

R. H. Bradt of Albaugh's group began his visit today at Clinton Laboratories. He plans to return here on August 7.

James Franck sent a letter to Vannevar Bush, Carnegie Institution in Washington, D.C., pointing out the general feeling that exists throughout the Project concerning the need for a peacetime atomic program that will continue to supply the research necessary for both military and technical applications in the postwar period. Planning is important and may require at least a year's effort after the war ends. Franck points out that one of the major difficulties we face is the irreversible scattering of key men during the present production and trouble-shooting period. Should the war suddenly stop, there is danger of wholesale dissolution of projects and personnel before intelligent long-range plans will have been formulated. Franck proposes that an interim organization under the Government be created to act as a buffer between the wartime military program and a new peacetime organization with long-range goals.

Helen worked in the afternoon with Truman Kohman on the classified version of my "Table of Isotopes."

U.S. troops have made a new landing in Guinea, and other advances are being made in the Pacific, according to today's news. In Europe, U.S. troops have made a combined air and ground "lightning" push in France and captured the port of Avranches.

Wednesday, August 2, 1944

This was Nathalie S. Baumbach's last day of work at the Met Lab. She and Harlan will return to California later this month.

Ralph James completed the search for long-range alpha particles in the supernatants from the last four dichromate cycles which he ran (beginning on July 24) on a portion of the plutonium sample bombarded with 32 Mev helium ions at Berkeley. He concludes that little, if any, of the long-range alpha-particle activity is to be found in the supernatant samples. Thus, his efforts have been unsuccessful in isolating detectable amounts of element 95 and 96 from sample 49αA#12. There is still another 2.2-mg portion of the bombarded plutonium target sample, however, that was made into a plate for Ghiorso to make pulse height measurements in the Frisch nitrogen chamber.

Manning replied to the letter T. O. Jones at Haverford College wrote on July 25 to Willard. Manning explains that he is replying because of Willard's relocation on the Project and says that we are glad to hear that arrangements for his Jones's) leave of absence from Haverford are finally taking shape. Manning expresses our hope that he will be able to join us at the earliest possible time.

After regular hours, about 5:45 p.m., Katzin, Studier, and Hagemann began the job of extracting U^{233} from two cans of thorium carbonate that were irradiated for seven months in the Clinton pile. They fished the first can out of its shipping container and deposited it in the dissolver unit containing nitric acid. Following dissolution, ammonium nitrate will be added to the thorium nitrate solution to a total nitrate normality of 8-9 N. The uranium will then be extracted from this solution using ether extraction procedures.

I attended the meeting of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I, which was held at 7:45 p.m. in Room 209 of Eckhart Hall. Those present included Arnold, Cunningham, Davidson, Dawson, Dixon, Florin, Hagemann, Hindman, Howland, Hufford, Katzin, Kohman, Kraus, Krueger, Manning, McLane, S. Peterson, Clifford Smith, Willard, and others. Manning explained the recent reorganizations which have taken place within the section. The Purification Sub-section has been liquidated. A solvent extraction group, with Lawroski as its group leader, has been organized under Katzin to work on U^{233}.

Smith reported on his recent studies on the mechanism of coprecipitation. Various attempts to prepare $La_2PuF_{10} \cdot xH_2O$, the double sale of plutonium and lanthanum thought to be responsible for the carrying of Pu(IV) with lanthanum fluoride, have all failed. Hufford reported on four methods he has used to prepare thin films of plutonium and uranium. These methods are: (a) slow evaporation and precipitation with tetraethylene glycol present; (b) sublimation of acetyl acetonates; (c) evaporation of an $(NH_4)_2PuF_6$ slurry; and (d) electrodeposition.

McLane summarized his work on the dialysis of Pu(IV) solutions. A reassay of previously reported dialyses on the "green" solutions

indicated that the original analysis had been in error. A new run on a solution containing a mixture of normal and "green" Pu(IV) was carried out; 70% of the material was found to be dialyzable. The dialyzate was found to contain "green" and "brown" material, indicating that the "green" material can have different particle sizes depending on the method of preparation. A brown solution (0.075 g Pu/l) was found to have dialyzed 95% within one hour. Another brown solution containing a small amount of "green" (spectrophotometer) was found to have dialyzed 93% after 72 hours.

Kraus reported on the general problem of the interconversion of the "brown" and "green" forms of Pu(IV). It was found that a "green" solution could very slowly be converted to a "brown" solution on standing at room temperature in 1 M HNO_3. The process of conversion may take several months. The evidence this far thus indicates that the "green" form is composed of polymers probably of different sizes and that the formation of the green form is due to the polymerization of a basic ion. The aging of the various green solutions in 1 M HNO_3 would then correspond to a depolymerization.

Report MCA-1949, "Met Lab Personnel as of July 15, 1944," which was issued today, shows the following persons as the technical directorate of the Metallurgical Project and the Metallurgical Laboratory:

Project Director	Arthur H. Compton	Eck 313
Consultant	Zay Jeffries	Eck 313
Associate Project Director, Research	Norman Hilberry	Eck 316
Associate Project Director, Health	Robert S. Stone	Site X
Special Assistant to Project Director	Arthur J. Dempster	Eck 118
Special Assistant to Project Director (Coordinator of Information)	Robert S. Mulliken	Eck 210
Laboratory Director	Samuel K. Allison	Eck 325
Consultant	Henry D. Smyth	Eck 327
Assistant Laboratory Director (du Pont)	Harcourt C. Vernon	Eck 407
Assistant to Laboratory Director	Lester C. Furney	Eck 407
Assistant to Laboratory Director (Shop Foreman)	Thomas J. O'Donnell	Ry Shop

Director, Division of Chemistry	Thorfin R. Hogness	NC 42
Acting Director, Division of Physics	Norman Hilberry	Eck 331
Acting Director, Division of Health	Leon O. Jacobson	BH
Director, Argonne Laboratory	Enrico Fermi	Eck 304
Director, Division of Technology (du Pont)	Charles M. Cooper	Ry 257

(NOTE: Eck = Eckhart Hall, Ry = Ryerson Laboratory, NC = New Chemistry Building, BH = Billings Hospital.)

Helen went to a luncheon at Isbell's given by Pauline Watt for Dagmar Dreher. Dot Maloney also attended. The Drehers plan to leave Chicago this weekend to go to Hanford.

Helen worked at the Met Lab in the afternoon.

Stan Thompson will leave Chicago on the Pennsylvania Railroad at 11:55 tonight for a trip to Site X. I will see him again Friday when I go there myself. We plan to return to Chicago together around three o'clock Saturday afternoon.

At 11:25 p.m. I left Englewood Station for a one-day visit to the Monsanto Chemical Central Research Laboratory (Monsanto Unit No. 1) in Dayton, Ohio, where I shall meet with Thomas, Warner, Hochwalt, Staniforth, and others concerning the Plutonium Volume which is to be prepared under Thomas's editorship.

Today's newspaper indicates that U.S. troops have smashed into Brittany.

Thursday, August 3, 1944

I arrived in Dayton early this morning and went directly to the Monsanto Chemical Company's Research Laboratory and met with Thomas, Hochwalt, Staniforth, Warner, and others.

Later I boarded a train in Dayton and traveled in the reserved seat section to Cincinnati, then I occupied a lower berth in a Pullman car for the rest of the trip to Knoxville.

Headlines today indicate a big breakthrough in France by British troops, and U.S. troops are also moving well.

Friday, August 4, 1944

I arrived in Knoxville by train early in the morning and was driven to Clinton Laboratories by automobile.

I attended the monthly Chicago-Clinton Chemical Conference (Clinton Steering Committee meeting) at Clinton Laboratories. Others present were Brown, Doan, Greag er, Hogness, W. C. Johnson, Kay, Kirst, Larson, Perlman, Squires, Struthers, Sutton, and S. G. Thompson. Kay presented information on the plutonium losses obtained for recent Clinton process runs. Greagor compared the Clinton semiworks results of the simulated average Clinton process data with the Hanford process data. Losses in the average Clinton runs total 12.5% whereas the Hanford process losses, combining three runs, total 7.33%.

Squires gave the status of the Hanford start-up. The 105B pile area is undergoing a shakedown period which will run through September 1. The chemical areas — 221T canyon, 224T concentration, 241T and 231 isolation areas — will be ready for operation by August 25, September 7-15, September 15, and November 15, respectively. The U-plant area will be ready a few months later. A decision has been made that the fourth canyon, which had been planned for earlier, is not required. It is expected that the cold runs in the T-area will be completed on August 15. This will be followed by dummy runs using 9 tons of rejected uranium slugs including 6 runs with Clinton metal as a plutonium tracer. The dummy and tracer runs should be completed by the first of November, about the time the first 6-ton batch of low plutonium concentration (30 g/ton) slugs, with 25 days cooling, are available from the 105B pile. The processing of this early operations batch will be conducted through December 5. The next batch, which will consist of 40 tons of 70 g/ton slugs, will be processed using two canyons at a rate of 2 tons per day per canyon. After the processing of this intermediate concentration material, the plant will be operated at full schedule and at the planned Hanford concentrations of plutonium.

Larson gave a short statement on the status of plant isolation, and Sutton described his group's work on concentration and isolation. Perlman talked about modifications to the isolation process which includes solubilization with 30% K_2CO_3 and 0.05 M $(NH_4)_2S$. After heating for one hour at 75°C, the FeS and K_2CO_3 are removed as insoluble precipitates. The plutonium in solution is then precipitated with 2 M KOH, the precipitate dissolved in HNO_3 prior to the final product isolation precipitation as the peroxide.

Following the Clinton Steering Committee meeting, I joined Stan Thompson, who arrived in Oak Ridge on Thursday, and Vance Cooper at the Municipal Golf Course in Knoxville, where we played 9 holes of golf. Our scores were ST-53, VC-55, and GS-46. Perlman joined Thompson and me at the railway station, and the three of us then boarded the Pullman section of a Southern Railway train to Cincinnati. From there we will continue to Chicago on the New York Central Railroad.

Saturday, August 5, 1944

Thompson, Perlman, and I arrived in Chicago at 4:45 p.m. The train was nearly two hours late. I went directly to the Jackson Park Golf Course to see Chick Evens beat Harold Mathisen in a sudden death playoff (20th hole) in a city championship match. In spite of my late arrival, I was able to see the last few holes.

Helen went to the Crerar Library today and later went shopping. We spent the evening at Luther Arnold's apartment at a "sort-of party."

Today's headlines report an army purge by Hitler. This is in response to the recent assassination plot against Hitler.

Sunday, August 6, 1944

Iz Perlman is in town from Oak Ridge. He ate Sunday breakfast with Helen and me at home and then went with us to play golf at the Evergreen Golf Club (9140 S. Western). I played with Perlman, Al Ghiorso, and Stan Thompson in one foursome, and Helen played with Harlan and Nathalie Baumbach and Zene Jasaitis in another foursome. Perlman shot 105, Ghiorso 113, Thompson 103, and I 100. The Baumbachs left, and the six of us remaining played an additional 9 holes for which we scored: IP-59, AG-55, ST-55, and GS-54. Following the game Helen and I and some of the others had dinner at Jules Restaurant at 814 West 71st Street.

At the Plymouth Congregational Church in Hammond, Indiana, Henry Hoekstra married Marilyn (Jodie) Jordan, a laboratory assistant in Arnold's C-V Section. Jim Gilbreath, also of Section C-I, was best man. The Hoekstras will return August 15 from their honeymoon. Several members of Section C-I attended the wedding (see Figures 11, 12, 13, 14).

Monday, August 7, 1944

The following activities took place at the Met Lab while I was away at Dayton and Oak Ridge last week.

On Thursday Ralph James began a series of dichromate cycles on the 2.2-mg portion of the plutonium plus 32 Mev helium ions, Berkeley bombardment. This is the sample which had been used to prepare a plate for use in the nitrogen chamber. The object is to obtain some additional element 95 or 96 so its chemistry can be studied.

Zachariasen sent a memo to Allison (with a copy to me and many others) on Thursday giving further evidence to confirm his claimed discovery of trivalent thorium. A recently received thorium sample from the Lindsay Light and Chemical Company claimed to be $ThF_4 \cdot xH_2O$ was found upon x-ray diffraction analysis to be nearly pure ThF_3. Zachariasen

Figure 11. Met Lab group at Greyound Bus Depot (Stony Island and 63rd St.) enroute to Hoekstra wedding. Left to right: unidentified, Jake Sedlet, Milton Adler, Ellen Skirmont, Irving Sheft, Don Wetlaufer in white shirt, Mrs. Sheft, Beatrice Foreman, Roy Post, Ray Meschke, Phillip Fineman holding gift , Walter Beard, and Caroline Rose. August 6, 1944.

XBB 7810-13177

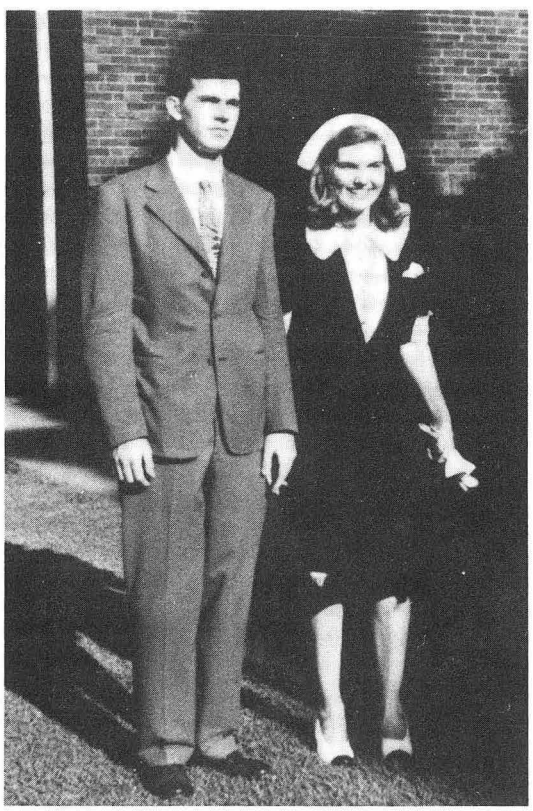

Figure 12. Walter Beard and Caroline Rose at Hoekstra wedding. August 6, 1944.

XBB 780-15276

Figure 13. Hoekstra wedding, August 6, 1944. Marilyn (Jodie) Jordan Hoekstra, Henry Hoekstra, Caroline Rose, and Walter Beard on right.

XBB 7810-13179

Figure 14. Henry and Marilyn Hoekstra. Ray Meschke and Roy Post (extreme right). August 6, 1944.

XBB 7810-13180

remarked, "In view of the fact that the chemistry of thorium has been extensively studied, it is indeed surprising that the trivalency of thorium has not been discovered until now." He said that although the x-ray diffraction identification is absolute in itself, he has submitted an 0.5-g sample of ThF_3 for direct chemical analysis. Zachariasen recommended to Allison that the Met Lab make further studies of thorium chemistry including the preparation of several trivalent thorium compounds. This recommendation is being made over P. W. Selwood's objection to the correctness of Zachariasen's observations. Zachariasen takes strong issue with Selwood's claim that magnetic measurements on the Lindsay sample prove there cannot be over 10% trivalent thorium present.

Last Thursday a telegram was received by Kay Tracy, Compton's secretary, from Compton who is participating in a Project Review colloquium in Los Alamos. The telegram instructed her to give a message to either A. V. Peterson or Nichols that he recommends Farrington Daniels for the post of Associate Director of the Chemistry Division, which will soon be vacated by Selwood. He goes on to say that Daniels would be "suitable to take Hogness' place if another likely change occurs within a few months. Though Chicago Chemistry will be cut by one-third within two months and probably more before Christmas, good direction is required for continued work to be effective. Alternative to Daniels's appointment is closing by October of two-thirds Chicago Chemistry, which I do not recommend because of uncertainties ahead but will do on request."

"Metallurgical Laboratory, Report for July 1944," (MUC-SKA-773), was prepared on Thursday by the Laboratory Director's Office. In the summary it is noted that the total number of persons employed increased by 64 during July although a directive issued on July 14 from the Project Director stated that employment should not exceed that of July 1. Another July event noted was the abandonment of the purification program for plutonium. This has caused a large amount of reorganization in the Chemistry Division and the Metallurgy Section of the Technical Division. Also noted in the summary is that, with the considerable encouragement of the du Pont Company, a solvent extraction tower has been set up to proceed with work on this process for product recovery.

Accomplishments of my section are described as follows: "Because of the difficulties in handling cerium and zirconium scavengers in the plant, four different attempts have been made to eliminate this scavenger step. These are: (1) the use of dilute HF to complex zirconium, (2) and (3) two separate low acidity methods, and (4) the use of three unmodified $BiPO_4$ cycles. All of these are tested on 100 ml runs and some also on the 1-liter scale. Decontamination factors and product losses were satisfactory in all cases. The problem of corrosion in connection with the use of HF has, however, not been solved. During these runs attention was given to the fact that efficiency of centrifugation is necessarily lower in the plant than can be achieved in the laboratory.

"Early operation with the use of four tons of metal per day have been tested with decontamination factors of the order of 10^5. The effect of a storage period of ten days between dissolution and extraction has been shown to be negligible. This is true whether sulfuric acid is present or not.

"The use of sodium nitrite ($NaNO_2$) prereduction has given product losses of 1% as opposed to 4% without this step.

"Satisfactory progress has been made in the filtration of product peroxide, provided the mixture is free from crud and does not contain too many fines. It is estimated that a 250-gram batch may be filtered in 1½ hours on a six-inch filter. A clean-up for crud removal improves precipitation and filtration of peroxide. Various reagents have also been tried for the elimination of iron in this step.

"Two promising new solvents for extraction of Pu(VI) from 10 M NH_4NO_3 are cyclohexane and dibutoxytetraethylene glycol.

"Vapor pressure data on plutonium metal reported last month has been verified as to order of magnitude by two subsequent runs with the original sample.

"The vapor pressure of PuO_2 in the range 1430° to 1640° K is about 100 times smaller than that of the metal according to preliminary results obtained.

"An 18-foot column has been set up for testing the solvent extraction purification process using hexone (methyl isobutyl ketone) on Room D lanthanum fluoride concentrate.

"The abnormal form of Pu(IV) which is not carried by $BiPO_4$ has been shown to be largely colloidal, presumably hydrous oxide or hydroxide.

"The rate of gas evolution from product solutions has been redetermined to be 0.58 ml/g Pu/24 hours. During storage of 40% UNH, slow oxidation occurs, reaching equilibrium at 37% Pu(VI). None of the colloidal abnormal Pu(IV) seems to be formed. A reliable assay method has been developed for the estimation of reduced plutonium in the presence of oxidized plutonium."

"Total expenditures for the month of July were $1,244,264. The number of personnel now employed is 1,988 (823 academic)."

Helen worked at the Met Lab on Thursday. She also took golf lessons at the downtown YWCA.

According to Thursday's paper, U.S. troops which engulfed the Breton peninsula have turned east and are moving towards Paris.

While liquid nitrogen was being poured into a trap in a vacuum system in Room 25 (used by Simpson, Jasaitis, Phipps, Seifert, Johnson, and Erway) Friday morning, a portion of the glass vacuum apparatus broke. The system contained a cold crucible with 46 mg of plutonium. Simpson determined that most of the plutonium remained in the crucible and was recovered. Some of the broken glass had small amounts of plutonium adhering to it from previous experiments. No alpha-particle contamination was detected on the floor or the vacuum rack.

On Friday Sheft completed his second experiment in an attempt to prepare PuI_3. He sent the resulting sample to Zachariasen for x-ray analysis.

A request was made Friday for the transfer of Zvolner to A. C. Hyde's semiworks.

Zachariasen sent a memo to Allison (with a copy to me and others) on Friday concerning the crystal structure of thorium hydride determined by x-ray diffraction analysis. The samples had been supplied by Eastman of Berkeley. The structure for thorium hydride is found to be cubic body-centered with a new and unusual structural configuration. Assuming the formula ThH_3, he calculated the density to be 8.25 gm/cm^3.

The Health Division made 143 alpha-particle surveys in 28 rooms of New Chemistry during the past week. At the end of the week, Rooms 4, 5, 10, 11, 28, 30, 35, 36, and 37 gave spot readings which were off-scale on the "Pluto" portable alpha-particle ionization chamber survey meter. Beta-particle and gamma-ray surveys showed unsatisfactory conditions in Rooms 9, 29, 30, and 33. The most serious of these beta-gamma readings occurred in Rooms 29 and 30.

In Room 29 (Bradt and Ader), a survey of a fume hoood enclosing a cave made of lead bricks showed that a tolerance dose to the face, head, and neck of anyone working at the hood would be delivered in 20 minutes. A recommendation was made to build the cave bricks higher or to split the contained radioactive material among several caves so as to lower the leakage radiation dose rate. In Room 30 (Hoekstra, Malm, and Bartell) an unshielded test tube rack, left on a desk top, gave a tolerance dose of only one-half hour as measured by a Lauritsen quartz fiber electrometer beta-gamma survey meter.

A Saturday morning meeting was held to discuss the present status of the Solvent Extraction Program. In attendance were Albaugh, Arnold, Dawson, Lawroski, Maloney, Manning, Pye, and Tepe. Dawson reported that the hexone batch-extraction procedure handles lanthanum fluoride precipitates from Room D at Clinton very easily on the laboratory scale. Complexing with zirconium is rapidly accomplished. Recovered material, however, does not handle as well. It is suspected that silicate collected from the glass apparatus used in past procedures may be causing some interferences.

Lawroski reported that the small extraction column appears to operate well with test solutions. A run is now being started with lanthanum and zirconium at Hanford process concentrations but with no plutonium present. On balance, work is progressing without serious difficulty. About 90% of the materials for the 3-inch diameter glass column extraction system are now on hand, and the final assembly should be completed about August 15-20. Maloney reported that Dawson's data on the batch run are sufficiently encouraging so that the batch process may be the process for Hanford to test first. It was noted that definite final specifications for the form and purity of the plutonium to be produced at Hanford Engineering Works have not yet been received. This information is needed to complete the development of a solvent extraction procedure to meet Hanford's requirements.

At a Liaison Committee Meeting Saturday in the Health Division (with J. G. Allen, Jacobson, J. Nickson, M. Nickson, Pyle, W. H. Ray,

J. E. Rose, and Wattenberg attending) it was noted that on the basis of past surveys including the past week's 143 alpha-particle surveys and beta-gamma surveys, a pattern seems to be established; certain rooms in New Chem tend to be always contaminated and other rooms tend to be usually relatively uncontaminated. The reason for this is thought to be a function of both the individual chemist using the room and the nature of the work usually carried out in that room.

Of 20 individuals who had valid overexposures for gamma-rays, as recorded on their pocket condenser ionization chambers during the week, only one was from my section in New Chem. However, the West Stands activities involving Albaugh's and Dreher's groups and the group from the Technical Section "scored" a total of 13 out of the week's 20 recorded overexposures. This points out the many contamination and radiation hazard problems currently existing in the West Stand's operations.

Yesterday the Drehers left Chicago. They will take some terminal leave before reporting for duty at Hanford toward the end of this month. Also, Hoekstra began his 10-day vacation. Larson and Howland began vacations today and plan to return on August 14 and August 22, respectively.

Bradt returned today from his visit to Clinton where he has been since August 1. The main purpose for his stay at Clinton was to obtain information from Iz Perlman and his staff on the colloidal Pu(IV) problem and its effects on product recovery and purity. Special emphasis was given to the effects on decontamination of the bismuth phosphate by-product precipitation step. Bradt feels the evidence obtained at Clinton definitely indicates the presence of colloidal systems that can be settled but only in centrifugal fields and do not appear to exhibit a Tyndall light-scattering cone. Upon returning to his laboratory here, Bradt began to devise an independent test for the presence of the colloidal material that he feels is responsible for retaining plutonium in suspension.

Zachariasen's x-ray diffraction analysis of the sample Sheft gave him on Friday identifies the substance as being pure PuOI with no PuI_3 present.

I received a copy of a memo from Selwood to Allison concerning Selwood's controversy with Zachariasen regarding the existence of trivalent thorium. Selwood claims his interpretation of magnetic measurements shows the samples, that Zachariasen claims to be pure ThF_3, cannot contain more than 1% Th^{+3}, if any. Selwood found the sample to be slightly diamagnetic; whereas according to him, it should be highly paramagnetic if it were a Th^{+3} compound.

I received a copy of R. E. Clark's memo to C. M. Cooper requesting approval for a tentative plan, worked out in cooperation with Nickson, for the disposal of active Met Lab waste materials. Storage both above and underground will be at the Argonne Site. Where possible, waste paper containing appreciable amounts of plutonium is to be recovered by the Chemistry Division. At present the Chicago active wastes on hand are

distributed in about 728 gallons of solutions in various forms and in three animal carcasses.

I have a migraine headache this evening.

Tuesday, August 8, 1944

Richard S. Rosenfels and Lyle H. Jensen terminated their employment with my Section C-I. Rosenfels is scheduled to report to Hanford August 23, where he will work with Kirk. Kirk, himself, left Chicago last Saturday to vacation in Berkeley before starting to work at Hanford.

I sent a memo to Allison concerning the work done in my section on the identification and possible recovery of protactinium and ionium from uranium ore residues. I suggested in an earlier memo (MUC-GTS-539 of March 15 of this year) that certain uranium ore residues should be likely sources of protactinium. Colonel Ruhoff supplied me with various promising residue samples. In work done in Katzin's group, sample no. 3, a carbonate precipitate of the uranium extraction process, was dissolved in nitric acid and precipitated with MnO_2 used as a carrier. Sixty percent of the total alpha activity precipitated; 25% appears to be protactinium, and 75% ionium. These values check well with alpha-particle range measurements made on the activity in the manganese dioxide precipitate. Rough calculations indicate that the carbonate precipitate uranium ore residue contains more than one gram of protactinium per metric ton and that it may be possible to recover easily this element in reasonably good yield by the use of the manganese dioxide carrier procedure. The ionium concentration is estimated to be about 10 ppm.

I suggest that, although only preliminary investigations have been made so far involving only one uranium ore residue fraction, should Site Y need a gram or so of protactinium as soon as possible we could start work on a ton or more of the carbonate precipitate residue material immediately. I request five additional kilograms of the same carbonate precipitate sample material received before from Ruhoff, for use in our research program which includes certain necessary investigations on protactinium.

Helen was busy at home all day.

The Soviets continue to move well in Poland, and the Japanese are making a last stand on Guam peak.

Wednesday, August 9, 1944

Florin converted a sample of PuF_4 to a higher fluoride using a procedure that was shown by a previous experiment to convert UF_4 to UF_6. A frost-white product thought to be PuF_6, which condensed on the walls of the trap, was taken into solution by admitting an atmosphere of nitrogen

and washing out the trap with ½ cc of zirconium sulfate in H_2SO_4. This wash solution was further diluted and analyzed for the valence state of the plutonium with a spectrophotometer. The results obtained by Hindman showed the plutonium to be almost entirely hexavalent. An experiment, with the object of subliming another portion of the material into capillaries for x-ray tests by Zachariasen, was unsuccessful because of the accidental introduction of H_2SO_4 into the sample holder.

Katzin and his co-workers have nearly completed the extraction of U^{233} from two cans of Clinton pile-irradiated thorium carbonate. The extraction run was started one week ago and has proceeded very well. Adequate planning and careful handling of materials have kept radiation exposures and contamination problems to a minimum. (This fact was given special notice by John Rose in last Saturday's Health Division Liaison Committee meeting.) It seems likely that at least 5 mg of U^{233} will be obtained from the extraction run.

In view of this success and since we would like to have larger quantities of U^{233} for experimentation, I sent a memo to Hogness requesting four more cans of irradiated thorium carbonate similar to the type previously received from Clinton. In addition I requested the irradiation of five cans of thorium carbonate material that has been completely purified of natural uranium by Katzin. This material is being canned and should be ready for shipment to Clinton within a few days. I asked Hogness to arrange for these cans to be irradiated for a period of several months in a high flux position within the Clinton pile. When processed, this material should provide us with U^{233} that is very free from natural uranium impurities.

I received a copy of Selwood's memo to Allison in which Selwood argues against my contention that 5f electrons exist in plutonium. He claims the magnetic studies on plutonium compounds by Cunningham's subsection are very incomplete and point more toward a "d" transition group than a rare earth series. He urges that magnetic studies be run on plutonium metal and estimates that about 100 mg of plutonium would be required for these studies. Selwood also notes that "thorium gives no evidence whatever for f electrons because its only known valence state (+4) is diamagnetic."

Hopkins (Figure 15), of Pye's group, has been wearing fitted filters in his nose for the past few weeks because of the hay-fever season. Out of curiosity, he saved the filters for the past two weeks, dissolved them, and prepared samples for counting. The filters for the week before last gave 32,000 alpha-particle counts per minute while last week's filters gave 5,000 counts. The highest daily face counts for these periods are 298 and 12, respectively. As the nose filter counts seem unusually high, they may have resulted from contamination acquired in handling. Hopkins will continue wearing the hay-fever filters for the next several weeks and will take special care to avoid their becoming contaminated. The used filters will be analyzed by Gardner of the Health Division in a contamination-free area.

Figure 15. Horace Hopkins. 1944.

XBB 780-14619

The Met Lab received a Clinton Laboratories progress report on "Decontamination in the Hanford Process Using Cerium-Zirconium-Phosphate Scavengers" (CN-1861).

I attended the evening meeting of the Extraction and Separation Processes Sub-section of my section at 7:45 in Room 209, Eckhart Hall. Others present were Ader, Albaugh, Blaedel, Bradt, Dawson, Gilbreath, Greenlee, J. J. Katz, Katzin, Manning, Meyer, Morgan, S. Peterson, Pye, Sheft, Studier, Templeton, R. Thompson, S. Thompson, Walling, Watt, Willard, Winner, and others. Stan Thompson summarized the status of tests at Clinton and Chicago that indicate that the Hanford process may work satisfactorily (but there is room for much improvement). About an 80% yield is expected on the basis of Clinton plant tests. The extraction step shows about 0.1% to 0.2% loss. The ability to achieve adequate decontamination seems assured. The work of the sub-section is now devoted to improvement of the Hanford process which includes reducing losses, such as losses in the bismuth phosphate by-product precipitation step, and trouble shooting problems that may arise, such as the poor results which have been encountered in the unmodified crossover cycle and the reduced yields in the isolation step due to iron interferences.

Bradt described work done to prove the existence of non-colloidal systems in the solutions involved in the first bismuth phsophate by-product precipitation. A slurry consisting of a bismuth phosphate by-product precipitate and the supernatant from which it was precipitated was divided into several equal fractions, and the precipitate in the individual fractions allowed to settle for times varying from 1 minute to 60 minutes. A portion of each supernatant was decanted off, and the liquid with suspended solids counted. The remaining portions of each fraction were centrifuged; the resulting solid and clear liquid phases were then counted. The distributions of activities in all samples, including the initial precipitates, were evaluated for a correlation between the weight of suspended matter and retained activity. It was concluded that since the activity retained by a given weight of suspended matter shows no variation with the total weight of material remaining in suspension, a colloidal phase must be present that contains activity. This agrees with conclusions reached on the basis of adsorption experiments on such solutions at Clinton employing Amberlite resins and the Chicago experiments involving the use of dialysis.

Pye described work on the development of a modified crossover cycle. The work was initiated because of the high plutonium losses experienced in the lanthanum fluoride precipitation step and the reduced yields in the isolation step due to iron carried through or introduced by the normal crossover cycle. The modified crossover cycle removes lanthanum fluoride containing plutonium directly from the 10 M HNO_3 solution of the bismuth phosphate containing plutonium from the last bismuth phosphate cycle. The lanthanum fluoride containing plutonium is precipitated by addition of HF to the solution to which La(III) has been added. Yields of 96.8% and 98% were obtained with 1 M and 2 M HF, respectively. There was no indication of the presence of iron in the subsequent plutonium peroxide precipitation step. Compared with the regular crossover cycle, this modified procedure accomplishes a marked reduction in process steps and operating time — and eliminates the loss

in the lanthanum fluoride by-product precipitation by simply eliminating the lanthanum fluoride by-product precipitation. The modified crossover is quite applicable for the early operations at Hanford.

Gilbreath described recent liter-scale process runs to determine if sufficient decontamination can be obtained in the unmodified and reduced acidity Bismuth Phosphate Process without the need to employ scavengers. The results obtained indicate that, although the unmodified Bismuth Phosphate Process gives higher decontamination than originally expected, the reduced acidity Bismuth Phosphate Process may yield the desired 10^5 decontamination factor through only two cycles. In either case, if necessary, scavengers may be sidestepped at Hanford by the use of three Bismuth Phosphate cycles.

Helen went to St. Joseph, Michigan, and then to Michigan City, Indiana, to look for a place to take our vacation next week. She found Michigan City to be suitable for our needs.

American troops are closing in on Le Mans in France. In the sports pages came the announcement that Lynn (Pappy) Waldorf, football coach at Northwestern University, will be the head coach of the 1944 College All-Stars.

An Executive Session of the Project Council was held this morning, attended by Allison, Bartky, Chapman, Compton, C. M. Cooper, Dempster, Doan, Franck, Hamilton, Jeffries, Mulliken, A. V. Peterson, Pratt, Spedding, Stearns, Stone, Szilard, Tracy, Vernon, Warner, C. Watson, W. Watson, Whitaker, Wigner, and Zinn. Compton in his "State of the Nation" opening gave some impressions gained during his recent trip to Hanford and Los Alamos.

Compton then said that Groves would like a reviewing committee survey of new ideas for improved piles. These survey results might determine the future course of our Project. Compton asked if ideas could be ready by October 1. Spedding suggested there should be an adequate group of individuals whose primary effort would be to develop a clear picture of the future of nucleonics. Doan added that the picture of future possibilities would not be complete unless all fields of technical knowledge are covered, and Compton replied that the committee under Jeffries is preparing to develop such a picture.

Mulliken was asked to make a statement on Lab reports. He said that although the written material developed by the Project is voluminous, it is not in shape to be convenient or even suitable for reference. He proposed the preparation of about a dozen books to cover the Project work in a thorough manner. He cited Wheeler's textbook and the Project Handbook as examples of what is wanted. Compton remarked that Mulliken as head of the Information Department will be reponsible for organizing and carrying through on this matter.

Allison stated that beryllium, bismuth, and thorium are materials which will be important to our future. Whitaker said

he would like some thorium metal to replace the thorium carbonate in the Clinton pile. Szilard added that if poisoning of the Hanford piles were done with about one ton of thorium metal, this would be an excellent source for larger amounts of U^{233}. Allison proposed to authorize Spedding to produce a definite amount of thorium.

Whitaker announced that with the installation of two new fans for the air-cooled graphite pile at Clinton, the power level has been increased to three times the original design power level. One fan, however, was seriously damaged last night, and the production power level is now down by about 25% to 30%. The results in the Clinton extraction plant indicate the Hanford plants will operate with yields above 80%.

W. Watson reported on the Canadian project's all-purpose light water-cooled pile that is to be constructed on the Ottawa River about 110 miles upstream from Ottawa (near a small village called Chalk River). The design power level will be 10,000 kw. Zinn said that the heavy water pile at Argonne is now operating 12-24 hours every day.

Hamilton reported on the University of California cyclotron and said it should be back in operation about October 10. The meeting adjourned at 11:45 a.m.

Thursday, August 10, 1944

Further chemical purification of James' sample 49αA#9 was completed to the extent that essentially all Pu^{239} has been removed.

The Director of the Montreal Laboratory, John Cockcroft, arrived in Chicago by airplane from Montreal this afternoon. He will visit the heavy water pile at Argonne and may spend some time at the Met Lab tomorrow.

I received a copy of a memo from Zachariasen to Allison stating that the previous claims for the existence of trivalent thorium, based on Zachariasen's x-ray diffraction analyses of thorium samples from the Lindsay Light and Chemical Company, have been found to be in error. The chemical analyses by Clifford Smith in Cunningham's group showed the Lindsay samples to consist of as much as 40-50% lead and caused Zachariasen to make a false evaluation of his x-ray data. The solid solution $(Th_{1/2}^{+4}\ Pb_{1/2}^{+2})F_3$ is thought to be the form in which the lead is present in the thorium fluoride sample. Zachariasen points out that $(Th_{1/2}^{+4}\ Pb_{1/2}^{+2})F_3$ has the lanthanum fluoride type of structure and that, if lanthanum fluoride is precipitated from a Pu^{+4} solution containing divalent ions, it may be expected that solid solutions will be formed. This has a direct bearing on the lanthanum fluoride precipitation in the crossover step since it may be anticipated that lanthanum fluoride will carry the compound $(Pu_{1/2}^{+4}\ Sr_{1/2}^{+2})F_3$ in solid solution and prevent satisfactory separation of plutonium from strontium.

Watt sent an outline to Willard of an initial research program for the Hanford Isolation Process Group (Building 231). The program includes a description of plant assistance and process improvement procedures.

Beard of Pye's group was found by the Health Division monitors to have an unusually high face count of 374 and a nose count of 642 alpha-particle counts per minute as the result of working with a solution containing about 400 mg of plutonium. He has not been wearing a respirator during his experiments nor has he checked himself for alpha-particle contamination during the day. In the future, greater caution must be exhibited.

Helen worked at the Met Lab today.

"Yanks Capture St. Malo!" reads today's headlines.

Friday, August 11, 1944

The Analytical Section (Section C-IV) has agreed to release Ralph Seifert for transfer to Simpson's group. Seifert has been working with Simpson on loan from Section C-IV. Manning sent a memo to W. W. Johnson officially requesting the transfer which is to be effective August 14.

Heath sent me a trip report covering the visits he and Arnold made to du Pont in Wilmington and to the SAM Laboratories at Columbia University. These visits were planned to see if their methods could be applicable to our plutonium purification program at a time before this program was discontinued. The process for making uranium metal was reviewed with Holbrook, Downing, and D. M. Smith at du Pont, with special emphasis on the impurities existing at various points in the process. Samples of UO_3, UO_2, UF_4, and uranium metal, obtained through Captain McKinley, were spectrographically analyzed by the Met Lab Analytical Division in order to determine more precisely what impurities are present at each step of the operation. These samples all originated from the same UO_3 starting material. The purity for the redistilled HF used by du Pont was also obtained from Downing.

At the SAM Laboratories, the methods used to prepare milligram quantities of UF_6 were discussed. A platinum liner is used there to hold the U_3O_8 which is converted with F_2 to UF_6 at 370°C. Florin has checked this method and finds it to be satisfactory and gives conversions of approximately 100%.

I received a telegram from Kennedy at Site Y stating that the copies of patent write-ups that I sent him earlier are being returned by air mail, special delivery, and should arrive here tomorrow.

Willard sent a letter to Squires, now at Site X, (with copies to Simon, Dawson, Perlman, and me) giving rough calculations of the extent to which Np^{239} activity may interfere with control laboratory measurements

of the decontamination of long-lived fission products. A tabulation of data is given for 30-day and 100-day pile operation and for various times of cooling. He concludes that in order to measure a decontamination factor of 10^5 with regard to true fission product activity at times less than 35 days following 30-day operation at 1000 kw/T of uranium, the control analysis must include a procedure for separating out the Np^{239}. A factor of 10^7 without neptunium separation can be measured if 50 days cooling is allowed to permit decay of the neptunium. For 100-day pile operation at 2.5×10^3 kw/ton of uranium, 30 days cooling is required to determine a decontamination factor of 10^5 and a minimum of 45 days is required to determine 10^7 unless neptunium separations are made. Willard points out that Dawson's group in my section has found it possible to remove neptunium from process solutions by a factor of 10^3 using solvent extraction with hexone.

Flox reported to Nickson that a sputum sample from Fowler Yett, obtained from phlegm coughed up yesterday morning upon arising, was analyzed and gave 2,640 alpha-particle counts per minute, or nearly 0.04 microgram of plutonium.

In a memo to Hogness, Nickson states that the storage of active materials in the West Stands, Room 216, used by Walling and Blaedel, is entirely unsatisfactory. Some bottles give radiation level readings as high as 4 R/hr. Consultation with Rose was suggested to correct the condition. In the interim, he requested that personnel traffic in and out of the room be kept to an absolute minimum.

Helen worked at the Met Lab on the classified "Table of Isotopes." She ate lunch at the Baumbachs'. Stan Thompson had an early dinner with us at home. Then, Helen, Stan, and I took the South Shore electric train to Michigan City, Indiana and checked into the Hotel Spaulding. Stan will return to Chicago in time to go to work Tuesday morning, but Helen and I will vacation here.

"B-29's Bomb Japan Again!" and "Super Forts Attack Nagasaki and Oil Base on Sumatra" put the focus of today's headlines on the war in the Pacific.

Saturday, August 12, 1944

After breakfast at Hotel Spaulding, Helen, Stan, and I took a taxi to the Michigan City Golf Club and played 18 holes of golf. Stan's score was 108 while my score was 106. We had lunch at the club at the end of nine holes. Temperatures reached the mid-90's today.

Stan, Helen, and I again spent the night at the Hotel Spaulding.

Sunday, August 13, 1944

Stan, Helen, and I ate breakfast in the hotel and again taxied to the Michigan City Golf Club and played 18 holes of golf. Stan scored 101, and I shot 108. Lunch was again at the club at the end of nine holes.

Today was another day of heat; temperatures reaching about 97°.

Monday, August 14, 1944

Stan, Helen, and I had breakfast together, taxied to the Pottawattomie Country Club, also in Michigan City. We played the 9-hole course twice for a total of 18 holes of golf. Stan's score was 107 and mine was 103. We had our usual lunch break at the club at the end of nine holes. Later in the afternoon Stan returned to Chicago.

Temperatures climbed above the mid-90's again today.

Tuesday, August 15, 1944

After breakfast at the drug store across the street from our hotel, Helen and I taxied to the Michigan City Golf Club where we played 18 holes of golf with lunch after the ninth hole. Later, Helen and I played tennis.

Today is another hot day.

In a small item in today's paper, note is taken of the death of Lt. Joseph P. Kennedy, Jr., son of the former U.S. Ambassador to Great Britain. Lt. Kennedy was killed in action.

Wednesday, August 16, 1944

Helen and I had breakfast at the drugstore across the street from our Michigan City hotel. Al Ghiorso arrived from Chicago, and the three of us taxied to the Pottawattomie Country Club and played 18 holes of golf. Al shot 121, and I shot 102. After eighteen holes, we had lunch at the club. Al and I then played an additional 9 holes (AG-53, GS-50). Al spent the night at our hotel.

The temperature reached nearly 97° again today but dropped this afternoon; perhaps the heat wave is over.

Thursday, August 17, 1944

We again ate breakfast at the drugstore across from our hotel, then Al, Helen, and I taxied to the Michigan City Golf Club and played 27 holes of golf. Our scores for the first 18 holes were AG-97 and GS-107, and for the last 9 holes were AG-47 and GS-56. We ate lunch at the club. Afterward, Ghiorso returned to Chicago.

Friday, August 18, 1944

Helen and I ate breakfast at the drugstore, then taxied to the Pottawattomie Country Club to play 18 holes of golf. Following the game we had lunch at the Club, checked out of the hotel, and returned to Chicago on the South Shore Railroad electric train.

The Soviets have reached the German border.

Saturday, August 19, 1944

Many Section C-I-related activities took place during my week's vacation in Michigan City. A day-by-day account is as follows:

* * * * * *

Saturday, August 12

Robert L. Patton resigned today for health reasons; he will return to Cornell.

Katzin, Studier, and Hagemann completed the extraction and purification of U^{233} from the two cans of thorium carbonate that were irradiated for about seven months at Clinton. Preliminary indications are that about 6 mg of U^{233} have been isolated from the two cans.

Kennedy wrote me from Los Alamos of the changes he has made in our patent information and claims. He suggested that I telephone him if I am not in agreement.

Bradt wrote up a trip report to Thompson about his visit to Clinton during August 1 through 6. Bradt covered much of the information in this report, which relates to the colloidal problem encountered in the recovery of plutonium, in his last Wednesday evening's presentation (August 9) during the Extraction and Separation Processes Sub-section meeting. Bradt believes Boyd's group at Clinton can make significant contributions to the solution of this problem.

Willard outlined in a nine-page memorandum to Squires a tentative start-up program of work for the Process Chemistry groups at Hanford. Six groups are planned with the following functional titles: Group I: Process Improvement Scouting, Group II: General Process Development, Group III: Special Process Development, Group IV: Fission Products and Radiochemistry, Group V: Basic Chemistry and Microchemistry, and Group VI: Isolation Process Chemistry.

In his introductory remarks he stated that the Process Chemistry Section will work in close cooperation with the Semiworks Section. He went on to say that the somewhat detailed program of work he suggests will be subject to revision at the time work starts at Hanford since some of the problems that are now of greatest importance will have been solved and since urgent new problems will have arisen. For convenience this tentative program is presented though the assignment of certain problems to certain groups at this time is, in some cases, rather arbitrary. The program incorporates many ideas suggested by S. Thompson and reflects discussions with Watt, Kohman, and Howland. A memorandum from Watt covering, in somewhat more detail, the problems of the isolation group was attached to the memorandum. Willard said that further suggestions have been requested from Sullivan, M. D. Peterson, and Olsen at Clinton and will be also be obtained from other members of all the groups.

Hopkins wore hay fever nose filters at work throughout this week and took every precaution not to contaminate them through handling or other means. The filters were dissolved with nitric acid, and the sample plated out and counted. The plutonium alpha-particle count was 150 ±5 c/m, considerably less than the 12,000 c/m and 5,000 c/m values reported from Hopkins' nose filters of previous weeks.

The weekly meeting to discuss the status of the development of Solvent Extraction for product isolation was held in my office in the morning. The meeting was attended by Arnold, Dawson, Lawroski, Maloney, Manning, Orlemann, Pye, Tepe, and Watt. Dawson reported on batch extraction studies that simulated Site W conditions as closely as possible. Only 0.1 Site W plutonium concentrations, however, were used on material from Room D in Clinton because of the hazards associated with handling the higher concentrations under present laboratory conditions. Analyses of the two runs showed the resulting plutonium contents of 99.44% and 99.60%, respectively, with the principal impurities being zirconium, silicon, and a trace of lanthanum. Studies are now being run using containers made from the 25-12 columbium stainless steel used at Hanford.

Lawroski reported that one run was made in the small diameter column, but operating problems were uncovered for which special parts are required in order to correct. Distribution coefficient studies using radioactive zirconium are in progress. Early results indicate that the zirconium distribution coefficient is about 60 in favor of the ammonium nitrate solution compared with hexone. With a plutonium distribution coefficient of one-fourth, a separation coefficient of plutonium from zirconium of about 250 is obtainable. Maloney and Tepe discussed the 3-inch diameter column and said that some difficulties have been encountered in obtaining the remaining materials for the system. They hope to have the procurement problems solved in the next few days.

Monday, August 14

Ralph Seifert's transfer from Section C-IV to C-I was approved, and Seifert became an official member of my section. Seifert received his Ph.D. from the University of Illinois in 1937.

Haeckl began his vacation and will return to work on August 26.

James completed the isolation of the elements 95 and 96 fraction from the 2.2-mg sample of plutonium target material from the plutonium plus 32 Mev helium ion bombardment at Berkeley. He started working up this sample on August 3 and since that date, has carried out four dichromate and two silver persulfate oxidation cycles (the latter demonstrating that the new element is not oxidized by silver persulfate). The final sample (49αA#31) appears to be almost free of 94^{239} and, on the regular 52% geometry counter, gives 110 alpha-particle counts per minute.

The deuteron bombardment of a large (200 mg) plutonium sample began today at the Washington University cyclotron in St. Louis. (The bombardment is scheduled to end in about two weeks with approximately 50,000 microampere-hours.) The purpose is to attempt to form element 95 by the d,n or d,2n reactions, or possibly to form 94^{240} by the d,p reaction.

L. B. Arnold sent me a copy of his memo to Bartky requesting 40 millicuries of 12-day barium and 2 millicuries of 65-day zirconium for our use in determining the distribution coefficients for zirconium and lanthanum in the solvent extraction process being studied for possible use as the concentration step at the Hanford Engineer Works. Immediate shipment of the barium by truck is requested in order to arrive before the end of next week. If this is impossible, it is requested that 5 mc of barium be brought up from Clinton by courier to be here by Monday if possible. The zirconium should be ready to ship within about 10 days, but courier service was not requested for this material.

The Chemistry Division "Summary Report for July 1944," (CS-1960), prepared by the Chemistry Division Director's Office, was issued today. The work of my section is summarized in 11 pages under the headings Separation Process Studies, Purification and Metal Production, and Basic Chemistry. The information in this report has been covered in more detail in other reports.

Tuesday, August 15

A revised organization chart and several recommended organizational changes for my section were sent by Manning to Hogness. The number of sub-sections is reduced to two — namely, the Separation Processes Sub-section and the Basic Chemistry and Services Sub-section. The group doing work on U^{233} is not included in either of the sub-sections. The Separation Processes Sub-section now includes a fourth group on solvent extraction. The Basic Chemistry Sub-section includes a new group, Basic Dry Chemistry,

in addition to the other three groups.

The memorandum suggests that the following personnel changes be made, effective immediately: Orlemann — to Associate Section Chief from Assistant Section Chief (to replace Watt who is leaving for Hanford about October 10); Katzin — to Assistant Section Chief from Assistant to Section Chief; R. Thompson — to Assistant Group Leader (to replace Albaugh as Group Leader about September 24); Albaugh — to Assistant Section Chief from Group Leader when S. Thompson leaves for Hanford about September 24; J. J. Katz — to Assistant Group Leader (to replace Pye as Group Leader when Pye leaves for Hanford about October 21); Lawroski — to Group Leader (replacing Jensen, who has already left); Davidson — to Assistant Group Leader; and Kraus — to Assistant Group Leader.

The staff of Section C-I is listed in the following manner:

Glenn T. Seaborg, Section Chief; Edrey Smith, Secretary to Seaborg; Kathryn Buehler, Stenographer; Winston M. Manning, Associate Section Chief; Edwin F. Orlemann, Associate Section Chief; George W. Watt, Associate Section Chief (leaving for Hanford about October 10); John E. Willard (now on du Pont payroll but still helping part-time in the capacity of Associate Section Chief); Irma Saxton, Secretary to Manning, Watt, and Willard; Eda Kelley, Secretary to Orlemann. *Sub-section I — Separation Processes:* Stanley G. Thompson, Assistant Section Chief in charge of Sub-section I (leaving for Hanford about September 24, when he will be replaced by F. W. Albaugh); Dorothy Gottlieb, Secretary to Thompson; *Group 1 — Extraction-Decontamination:* F. W. Albaugh, Group Leader; R. C. Thompson, Assistant Group Leader (to replace Albaugh about September 24). *Group 2 — Concentration-Isolation:* D. G. Pye, Group Leader (leaving for Hanford about October 21); J. J. Katz, Assistant Group Leader (to replace Pye about October 21). *Group 3 — Process Development:* J. R. Gilbreath, Group Leader. *Group 4 — Solvent Extraction:* S. Lawroski, Group Leader. *Sub-section II — Basic Chemistry and Service:* Burris B. Cunningham, Assistant Section Chief in charge of Sub-section II. Marjorie Bohlman, Secretary to Cunningham; *Group 5 — Basic Dry Chemistry:* O. C. Simpson, Group Leader; N. R. Davidson, Assistant Group Leader. *Group 6 — Basic Wet Chemistry:* J. C. Hindman, Group Leader; K. A. Kraus, Assistant Group Leader. *Group 7 — Recovery:* L. R. Dawson, Group Leader. *Group 8 — Instruments and Physical Measurements:* A. Ghiorso, Group Leader. *Group 9 — 23 Work:* L. I. Katzin, Assistant Section Chief.

James, studying the chemistry of element 95 or 96, completed an experiment Tuesday that showed that the element is precipitated with $La_2(C_2O_4)$ in a neutral solution with an excess of oxalate present. He made the studies using a portion of sample 49αA#31 from the plutonium plus 32 Mev helium ions, Berkeley bombardment.

The law firm of McLaren, Goode, and Company sent Underhill an opinion about the tax consequences of the alternate ways we have for disposing of our patent position. The facts in the case, as understood by McLaren, Goode, and Co., are as follows:

"Four individuals (referred to hereinafter as taxpayers) made a basic discovery which can be patented. Patent applications were made

during 1942, but in view of the importance of the discovery to the war effort, a secrecy order was issued by the Army or Navy under which the patent application has not been delivered to the Patent Office. The taxpayers and the University is entitled to a one-half interest in this discovery. The Government insists upon the purchase of the discovery, and our opinion is requested upon the possible alternative plans of disposal.

"Four alternative plans are reviewed:

"a. Direct Sale. The question as to whether the profits resulting from a direct sale to the Government would be considered as long-term capital gain or as ordinary income was considered. The law firm's opinion was that a strong case can be made for capital gain and that chances of winning in litigation are good should the Treasury Department take a contrary view. In spite of this, the law firm considers it advisable to combine, if possible, the capital gain feature with some sort of an installment sale.

"b. Installment Sale. A sale could be made where the taxpayers receive a relatively nominal cash payment plus later payments equal to a specified percentage of the income derived from the assets (as, for example, might be the case with an oil well asset) or from sales proceeds if the assets were sold.

"c. Trust Arrangement with University. The taxpayers might create a trust with the University to which they would transfer their interests under provisions whereby the proceeds from the sale to to Government would be held for the benefit of the University, but the income from the trust would be payable to the grantors and their families during their lifetime.

"d. Annuity Arrangement. The taxpayers would transfer their interest in consideration for some sort of an annuity payment by the University. The law firm feels that any attempt to establish arrangements where the taxpayers have assured rights to future payments other than by sale would be dangerous.

"The law firm concludes that the direct sale of the taxpayers' interests without an installment sale is the most practical answer to our patent problem, unless it can be maneuvered into an involuntary conversion."

Compton wrote to James B. Conant in Washington about the transfer of nucleonics to postwar conditions. (In a footnote, Compton explains that the word "nucleonics" has recently become widely used throughout the Project to designate the field of research, development, and application of atomic nuclear reactions. It, according to Compton, comes from the word "nucleon" which is the name for the elemental particles in the atomic nucleus.)

Compton believes the most important applications of nucleonics will be for some time in the area of military weapons; however, other applications will grow rapidly such as radioactive material for medicine, agriculture, and scientific purposes, and the use of atomic power especially for naval vessels and perhaps for heating of cities. He suggests that the U.S. Engineers may be the best group to continue its Project

management role into the postwar period. But, in order to provide for coordination of the various aspects of nucleonic work throughout the nation, he suggests that a Committee (or an individual) reporting directly to the President, be given the responsibility for overall supervision. The Committee would function in a way similar to the Military Policy Committee and would see that appropriate controls are put into effect for regulating nucleonic work.

Compton suggests an immediate program to encourage universities, industries, and private laboratories to undertake their own activities in nucleonic research. This would include the transfer of technical information from the Project to the public making available special materials. Authorization to use the tools built under government contract for the production of tracers and other useful materials would greatly aid medical and scientific developments. Public education on the significance of nucleonics is urgently needed in order to prepare for the postwar adjustment.

Regarding the future of the Metallurgical Project, Compton suggests a sharp distinction be made between continuing military work including continued support of the programs at Sites W and Y and the development of nuclear weapons and non-military work such as nuclear physics and properties of neutrons; fundamental chemistry of products and by-products; long-time instrument developments; the preparation, properties, and application of radioisotope tracer materials; scientific, medical, and industrial applications of radioactive materials. Compton says it would appear appropriate to encourage the University of Chicago to see that (non-military) scientific use be made of the tools now in its charge. Compton concludes his letter by saying that the problems associated with the interim period and postwar adjustments, especially as applicable to the Metallurgical Project, will be the main subject for discussion at the August 21 Advisory Committee Meeting.

Wednesday, August 16

"Chemical Research — Separation Processes for Plutonium, Report for Monday Ending August 1, 1944," (CN-1946), included the following information on extraction-decontamination, concentration-isolation, process development, and basic chemistry of the extraction process:

I. Extraction-Decontamination (Albaugh, Group Leader). Bartell and Greenlee have conducted four 100-ml scale runs to study the special conditions required in the Hanford mainline process in order to process uranium metal slugs at a four-ton per day rate. The conditions require that (a) extractions be made from 28% UNH, using only 1.9 mg bismuth/ml; (b) twice the normal quantity of extraction precipitate be processed as a single batch in the first bismuth phosphate decontamination cycle; and (c) excessive volumes of process solutions be avoided by the addition of a buffering agent, sodium acetate, or other means. Two of the runs using zirconium-cerium scavengers have given good decontamination factors; two runs made without scavengers have given rather poor decontamination results. Considerable plutonium losses occur in the extraction step.

Several experiments conducted to determine the influence of variations in experimental conditions upon decontamination achieved in the bismuth phosphate process may be reported as follows:

Sedlet conducted four single bismuth process runs to determine the effectiveness of bismuth phosphate scavenging and percent plutonium loss as a function of increased pH. The use of acidities considerably lower than the 1 N HNO_3 called for in the Hanford flowsheet has the advantage that solubilities of the rare earth phosphates are significantly reduced. Within the range investigated (1.0 N to 0.063 N), decontamination shows a steady improvement with decrease in acidity. Unfortunately, the percent plutonium loss in the by-product precipitation step increases from 0.85% to 7.1% with decrease in acidity. The reason for this is not clearly understood at present.

Malm and Morgan have conducted four 100-ml scale process runs with single-bismuth, low acidity by-product precipitation. The results obtained compare favorably with results using cerium-zirconium scavengers. The experiments, however, verify the steady increase in plutonium loss with decreased acidity. Morgan, R. C. Thompson, Ader, and Bartell have run four 100-ml scale "double-bismuth" low acidity by-product precipitation runs and two control runs through two bismuth phosphate decontamination cycles. The "double-bismuth" procedure involves both a bismuth strike and a phosphate strike to enhance precipitation.

R. C. Thompson and Morgan have conducted a series of eight 100-ml scale process runs through two decontamination cycles to determine the effects of combining in single runs the decontamination procedures that are known to be individually beneficial. Two of the runs combine double-bismuth, low acidity by-product separations with cerium-zirconium scavengers; two combine double-bismuth, low acidity by-product separations with 0.05 M HF included in the plutonium precipitation steps; two are control runs using the double-bismuth procedure and low-acidity; and the remaining two are normal flowsheet runs. The average of beta-particle and gamma-ray decontamination factors through two cycles for all combination runs exceed 5×10^5. The gamma-ray decontamination factor for the low-acidity-HF runs are as high as 5.2×10^6. Except for greater than 20% losses in the extraction step, plutonium losses are normal for all runs.

R. C. Thompson and Morgan have also performed two process runs to study the possibility of using three bismuth phosphate decontamination cycles without cerium-zirconium scavengers in preference to two bismuth phosphate cycles with cerium-zirconium scavengers. If successful, the three-cycle procedure could eliminate the difficulties involved in handling the cerium-zirconium scavenger precipitates and the additional time required for their use. Beta-particle and gamma-ray decontamination factors considerably better than 10^5 have been obtained for three cycles and the gamma-ray factor through the crossover is approximately 10^7. Three-cycle runs on the 1-liter scale were made later, in comparison with other runs by Hyman, Larson, Rasmussen, and Winner of Dreher's Process Development Group. Their results verify that the above decontamination factors can be obtained. S. Peterson has run four 25-ml scale process runs to evaluate the beneficial effects to gamma-ray decontamination of HF added in the plutonium product precipitation steps in different

concentrations, viz: no HF (control) 0.01 M, 0.03 M, and 0.05 M. The gamma-decontamination factors show a steady improvement with increased HF concentration. The beta-particle decontamination and plutonium loss are not significantly affected.

II. Concentration-Isolation (Pye, Group Leader). Experiments have been conducted to test modifications in the Hanford flowsheet concerning the lanthanum fluoride methods for plutonium concentration and isolation. These include: experiments by Kelley related to slight variations in the Hanford concentration procedure; the development by Goeckermann of a lanthanum fluoride cycle for use at high plutonium concentrations; the studies by Goeckermann, Hopkins, J. J. Katz, Kelley, Walling, and Yett on ways to eliminate interferences caused by iron in process solutions; studies on the constitution of plutonium peroxide by Hopkins which suggest the average empirical formula $PuO_{3.35}$; and the work by Yett on the separation of plutonium peroxide from the supernatant in the isolation procedure.

The results obtained by Beard on the use of $U(C_2O_4)_2$ as a carrier in an alternate concentration-isolation procedure are somewhat discouraging. Although it is thought the procedure could ultimately be made successful, a recommendation is made to discontinue the work in favor of more promising alternate procedures. Good yields were obtained with $U(C_2O_4)_2$ through the concentration procedure, but the yields in the isolation step were discouraging and range from zero to 70%.

Very promising results have been obtained by Haeckl as a continuation of Malm's earlier work on the alternate method involving the precipitation of lanthanum fluoride directly from bismuth phosphate solutions at high acidity. The advantages to be gained by this procedure are the complete elimination of an oxidation-reduction cycle and two by-product precipitation steps. In addition, the exposure period for equipment in the Site W plant to solutions containing HF would be cut in half. The results of two 50-ml scale experiments on synthetic bismuth phosphate solutions, using a 4:1 ratio of lanthanum to plutonium and a HF strike, gave overall yields of 97% and 98% through the first plutonium peroxide precipitation. Although highly encouraging, the value of this alternate method for plant use will depend upon the ability to obtain adequately high decontamination factors in the decontamination cycle, as only one additional decontamination step will be available in the "crossover."

III. Process Development (Dreher, Group Leader). Hyman, Lincoln, and Rasmussen have studied the effect of storage up to 12 days of uranyl nitrate solutions, containing Hanford concentrations of plutonium and inactive fission product elements, on plutonium yields in the mainline process. They find that length of storage does not appear to affect plutonium yields in the extraction step of the bismuth phosphate process.

IV. Two investigations are covered under the topic Basic Chemistry of the Separations Processes (Hindman, Group Leader). Both of these (work by Howland on "Effect on Pu(IV) of Storage in 40% UNH-0.2 N HNO_3," and work by O'Connor on "Solubility of Plutonium Compounds in Process Solutions") are also included in CN-1948, "Chemical Research — Basic Chemistry of Plutonium, Report for Month Ending August 1, 1944," to be issued later this month.

The Council of Section C-I met in my office at 8:00 a.m. Attending were Albaugh, Arnold, Dawson, Gilbreath, J. J. Katz, Katzin, Lawroski, Manning, Nickson, Pye, Simpson, R. Thompson, Watt, and Westrum. Arnold announced that a memorandum is being prepared giving instructions and procedures to be followed by chemists in case of product spills. A proposal was made that a working limit of eight hours daily be established and applied to individuals who are required to work with large amounts of product. In certain cases, shift work may be required. The desirability of a general limitation on night work was discussed. It was generally agreed that no one should work alone in any area of the building at night. Those people who need to do night work should make arrangements for at least one other person to be present or in a room close by. Voluntary compliance will be tried with more rigorous enforcement at a later time, if required.

Dawson expressed concern for the high alpha-particle nose counts in his group and made a plea for more space and the possible use of nose filters for the men to reduce the hazard of plutonium in the air. It is now thought that the biological half-life of plutonium retention in bone is probably three or four years. The results of face, hand, and nose alpha-particle counts are now being posted by the radiation protection group.

A committee consisting of Dawson, J. J. Katz, and Katzin was appointed to survey the present laboratory practices of handling plutonium. The results of this committee's findings together with their recommendations will be discussed at the Section C-I Council's next meeting. It was noted that not all hoods in the filtered air section are working properly. Arnold and Manning will discuss this with Captain Ware.

Harlan Baumbach terminated his work with the Met Lab today and will return to his former employer, Paramount Pictures, Hollywood, California. He and Nathalie, who quit two weeks ago, plan to drive to California on the 28th.

Katzin calculated, using alpha-particle counts by Hufford, that the specific activity of the U^{233} extracted from the irradiated thorium carbonate is 1.98×10^{4} disintegrations per minute per microgram. This figure, however, is subject to revision, as the extent of natural uranium impurity is not yet known.

Pye outlined a program for the work of his group during August 16-24, the time he will be on vacation. The work falls logically under two problem areas relating to concentration and to isolation. *Concentration:* J. J. Katz, in addition to serving as acting group leader, will continue the search for complexing agents applicable to iron for use either in the lanthanum fluoride plutonium precipitation step or in the metathesis step. The study will also include methods of dissolving the lanthanum fluoride precipitate so that reprecipitation will result in a lower amount of iron carried. Kelley will make radioiron tracer studies of the more promising complexing agents for iron as related to the Site W flowsheet. Wolf will continue development of a satisfactory colorimetric technique for iron analysis that will be correlated with the radioiron tracer measurements.

Meyer will investigate plutonium losses in the by-product precipitation step as a function of plutonium concentration using Site W conditions.

Isolation: Goeckermann and Hopkins are determining optimum conditions for plutonium peroxide precipitation at room temperature. The feasibility of using 0.5 N H_2SO_4-0.5 N HNO_3 for the dissolution of metathesized lanthanum fluoride precipitate is being investigated. Lanthanum fluoride slurry from Room D at Clinton will be used to run experiments to test metathesis and isolation under flowsheet and modified flowsheet conditions, and to determine the effects of adding a known concentration of iron. In addition, Hopkins will begin studies to determine factors involved in the loss of weight of the plutonium peroxide caused by thermal decomposition. Beard and Yett will work on the development of new acid media for use in the peroxide precipitation, including additional work on HCl, HCl mixtures with H_2SO_4, and scouting for other suitable acids.

A Solvent Extraction and Decontamination Meeting was held in my office to discuss the solvent extraction approach to the separation of plutonium from uranium and fission products. Those in attendance were Albaugh, Blaedel, Egan, Gilbreath, Orlemann, S. Thompson, Walling, and Watt. It was generally agreed that greater emphasis should be placed on decontamination since any scheme resulting in efficient decontamination will result in at least a partial separation of plutonium from uranium. Several conclusions were reached at this meeting. Based on past laboratory work, ether seems to be a good solvent from the standpoint of separation and decontamination, but is considered to be an almost impossible substance to use safely on an engineering scale for a continuous extraction process. Because of this several other approaches might be considered, including the following:

a) It might be possible to find a safer substance that has the solvent properties of ether. Of all the substances which have been studied so far, ether is unique in that it has a high extractive capacity for Pu(VI) and a low one for Pu(IV). It is this property that makes it particularly adaptable to the extraction-decontamination problem. Of the many substances so far investigated, most of them have capacities for Pu(IV) and Pu(VI) of the same order of magnitude when NH_4NO_3 is used as a salting-out agent. A few that might resemble ether are yet not investigated.

b) If it were possible to work with Pu(III), the extraction-decontamination objectives could be obtained with any of a great number of solvents already studied for Pu(III) is insoluble in almost all organic liquids, while Pu(IV) and Pu(VI) are soluble in many.

c) The use of $Ca(NO_3)_2$ or $Al(NO_3)_3$ instead of NH_4NO_3 might lead to a greater difference in extractive capacities for Pu(IV) and Pu(VI) in some solvents than the work using NH_4NO_3 indicates. Kraus' work on the salting of $UO_2(NO_3)_2$ into various solvents shows that for comparable concentrations, $Ca(NO_3)_2$ is more efficient (by a factor of 10-100) than NH_4NO_3. There is a chance that such a specificity in regard to salting-out agents might exist for either Pu(IV) or Pu(VI), but not both.

d) Of the investigated solvents some might have a lower extractive capacity for all the fission product elements than for plutonium. With such a solvent complete separation of plutonium from the fission product elements would be theoretically possible in a single column and the decontamination obtainable would be limited only by mechanical difficulties.

e) The difference in extractive capacities for Pu(IV) and Pu(VI) might be increased for some solvents by working at temperatures other than 25°C, the temperature at which available data were taken.

A telegram arrived from Hamilton stating that the uranium sample received 40,000 microampere-hours of Be + D bombardment (neutrons) in the Berkeley Radiation Laboratory 60-inch cyclotron.

Allison sent a memo to Franck referring to statements made at the June 20 Project Information Meeting that, (a) a pile with solid moderator will inevitably go bad, and (b) it is improbable that operation with graphite as a moderator can go on for two years. G. D. Graves of du Pont has questioned these statements and pointed out to Allison at a recent meeting that such statements, if left to stand on the record, could seriously embarrass the Project unless they are extremely well-founded in fact. Allison suggested that if similar considerations come up at the Project Information Meeting on August 22 that the record should be made clear as to exactly how large an extrapolation from our current limited knowledge is involved.

Thursday, August 17

Watt requested of Hogness that arrangements be made to obtain a batch of lanthanum fluoride-plutonium precipitate slurry from Clinton for use in studies of the peroxide isolation process and the alternate isolation process involving extraction with hexone. The material should be the product of one entire plant run — through extraction, decontamination, and crossover — but definitely not material which has gone through Room D. The best possible simulation of Site W conditions can be achieved by adding some additional plutonium.

In a memo to Mulliken concerning the future use of plutonium and the fission by-products of the Plutonium Project, Hogness says he has asked the Chemistry Division sections chiefs to come up with as many suggestions for future use as possible. Ideas from Burton's section are enclosed and include: (a) the use of radioisotopes for tracers in the study of metallographic and metallurgical problems, (b) the use of radioactive carbon in the study and analysis of polymers, (c) mineral exploration, (d) applications to biology and medicine, and (e) the use of radioactive materials for cutting down on static, such as might be useful in an explosives plant, or plastic film manufacturing plant.

Leo Szilard wrote to Vannevar Bush in Washington, enclosing the text of a personal letter he wishes to send to Lord Cherwell of the British Cabinet in England, whom he knows well. The letter is for the

purpose of bringing to the very influential Cherwell's attention some of the possible ways Germany might produce atomic weapons. Szilard says in his letter to Bush,

> The possible use of atomic bombs by the Germans is perhaps the only factor that may reverse the present favorable trend of the war in Europe. It would therefore seem very important to find the industrial installations in Germany which are likely to be used for this purpose and to destroy them. I believe that the British efforts in this direction would be greatly intensified if if Cherwell took a personal interest in them.

Szilard's letter to Lord Cherwell is six pages long. In one paragraph he states,

> Private communications originating from Switzerland which reached me two years ago indicated that the Germans knew by the middle of 1942 how to make a chain reaction go and that Heisenberg, who was in charge of that work had some conspicuous successes along this or the other parallel line of work during that year and consequently was put in full charge of all the work (made director of the Kaiser-Wilhelm Institute for Physics) late in 1942. This was about the time of Stalingrad after which the Germans must have realized that they may have to win this war by other than ordinary methods. Unless I completely misjudge the psychology of the Germans, they must have gone full scale into this work soon after Stalingrad at the latest.

Further on in his letter Szilard remarks,

> I am rather convinced that a properly organized effort of the British Intelligence will lead to the discovery of industrial installations for the manufacture of the relevant materials in Germany but when the location of the German factories is discovered, it may be found that we would have to pay an exceedingly high price for their destruction by large-scale parachute invasion or other such methods. Some of us might think that these factories have to be destroyed practically at any price but in order that you should be in a position to advise the War Cabinet how far to go in this respect you ought to have first-hand knowledge of the action radius within which life will be destroyed if a small atomic bomb is detonated above a city. Clearly you are the only member of the Cabinet who can have convictions based on his own computations rather than on official 'reports' or other forms of 'hearsay'. If you check the calculations of others or make calculations of your own you will have a firm conviction of your own and will be able to arrive at a balanced recommendation and also be able to assist the War Cabinet in reaching what may be a very difficult decision.

Friday, August 18

Continuing the study of the chemical properties of element 95 or 96, James completed experiments that show that the new element cannot be

precipitated with zirconyl phenyl arsonate either under reducing (SO_2) or oxidizing (after prolonged treatment with $Cr_2O_7^{=}$) conditions. For these experiments James used the material isolated as sample 49αA#31, from the second 2.2-mg portion of the plutonium target which he had worked up between August 3 and August 14.

Kohman prepared a handwritten letter for me concerning his explanation of the mesothorium problem. Rose of the Health Division has objected to the way the solutions are stored, handled, and shipped. He said that a shielded hood will be required for storage and handling the mesothorium material as well as our radium solutions. (Kohman and I had already discussed the shipment problem with Rose as it pertained to our plans to ship three to four millicuries of mesothorium to Fulbright at St. Louis. This matter was mentioned by Rose in the Health Division's Liaison Committee meeting on August 5.) He also mentions that the sample he has to send to Fulbright in St. Louis has developed a leak and must be recovered and another sample sent to Fulbright. Kohman referred the problem concerning the need for handling and storage facilities to Manning. We must solve this problem if we are to retain custody of this material and prepare additional samples for other users.

In a memo to the Area Engineer, Compton refers to the telegraphic message he sent on August 3 while in New Mexico recommending that Daniels be appointed to replace Selwood in the Chemistry Division in order to effect stronger direction during the reduction-in-force period ahead. The Area Engineer's subsequent refusal to act upon Compton's recommendation is taken by Compton to imply that the Area Engineer was disappointed with the Chemistry Division's plans for reducing its staff. On July 28 Allison suggested to Compton that the reduction was to be from 235 chemists on August 1 to about 155 chemists on October 1. The overall Met Lab reduction for the same period was to be from 688 to about 588 employees with a gradual reduction thereafter. Recently Nichols and Groves indicated that only those men who are required in winning this war should be retained. On the basis of this interpretation, the Chemistry Division may require a reduction to about 80 chemists, the minimum number required for reasonable support of the Hanford program. This would leave no place for fundamental scientific work. Compton says that although such a drastic reduction could be made, it would be done against his and the recommendation of others concerning what is required for the nation's welfare. A continuing "nucleonics" research and development effort is thought to be required. Compton stresses that "such studies looking toward stable peace should now be of hardly lower priority than those directed toward winning this war."

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Dawson prepared an outline for me this morning of the Solvent Extraction and Recovery Group work for the week of August 21 to 26, the week Dawson will be on vacation. Asprey will be in charge of the recovery and purification work and of the distribution of purified plutonium. Stewart will have supervision over the solvent extraction program which will include the following work for the week: (a) batch extraction studies using Clinton Room D lanthanum fluoride precipitates, (b) purification of radioactive lanthanum and zirconium, and (c) the determination

of the distribution coefficients for lanthanum and zirconium between salt solutions and hexone and between hexone and water, using radioactive materials.

I wrote a letter to Underhill (with a copy to Kennedy) enclosing two copies each of our two plutonium patent cases. I inform Underhill that as soon as I receive his telegram, I plan to send two copies of each case to Lavender. One of these cases, "Methods for Preparation of a Radioactive Isotope," with Seaborg, Wahl, Kennedy as the inventors, is the basic chemical case on plutonium and includes the claim, "A composition of matter which consists substantially of plutonium."

Westrum wrote to Joe Hamilton stating that the nine spectroscopic electrodes furnished by Hamilton have been loaded with plutonium as he requested. A total of 2.87 mg of plutonium, in the form of small spherical pieces or particles packed in spectroscopically pure graphite, have been loaded into the nine electrodes.

Franck answered Allison's letter of last Wednesday about the statements made at the Project Information Meeting of June 20 regarding the expected short operating life of a graphite-moderated pile. Franck points out that the statements made at the meeting should not embarrass the Project as Graves from du Pont implies they might, in that it was foreseen by Wigner right from the Project's beginning that a risk would be involved in the use of graphite as a moderator. So far the experience with the Clinton pile shows that the risks are not so great that a pile cannot be run long enough to turn out amounts of plutonium exceedingly useful for the war effort. Franck points out, however, that it is absolutely necessary to monitor the change in properties of the graphite in piles at Hanford during their operation. It is impossible to predict the degree of deterioration of pile graphite based only on an equivalent of three days of operation of the Clinton pile at Hanford radiation levels.

Katzin and I played nine holes of golf at the Evergreen Golf Club. I shot 54.

Helen worked at home today.

American troops are getting closer to Paris; the Germans say our troops are only twelve miles from the city.

Sunday, August 20, 1944

Stan Thompson and I played 18 holes of golf at the Cog Hill Golf Course No. 2. Our scores were ST-100 and GS-92.

Orlemann, Watt, and R. Thompson will leave at 11:55 p.m. on the Penn and Southern R.R. for Site X where they will attend conferences on Hanford separation processes. They all plan to return by train to Chicago on August 24.

Helen worked at home today on my classifed "Table of Isotopes."

Monday, August 21, 1944

Continuing the study of the chemical properties of element 95 or 96, James dissolved sample 49αA#9 (original sample used to identify the existence of element 95 or 96) in order to be able to use the material for experimentation. He carried out a bismuth phosphate precipitation on the solution in order to test whether or not the new element 95 or 96 is carried. The results show that element 95 or 96 is not carried by bismuth phosphate under normal conditions.

I read a memo from Hogness about an announcement made by a commentator last Tuesday at 7:00 p.m. over Chicago radio station WGN. The commentator said it could now be made public that the United States is working on a secret weapon. Although Columbia University was mentioned, no tie-up between the University of Chicago and the weapon was made. Hogness says the announcement was a mistake and was due to a leak in security. No one should take the unfortunate announcement as signalling a lessening in security restrictions. I am requested to make all employees in my section aware of this fact and that if any changes in regulations occur, they will be announced only through official channels.

Thompson prepared a summary, dated August 21, "Sub-section I — Separation Processes," for use in the presentation I will give at the Project Council Information (Chemistry) meeting tomorrow. Work for Groups 1, 2, and 3 is summarized as follows:

Group 1. Extraction-Decontamination

A. Prereduction-extraction

1. *Storage tests.* Solutions of 33% UNH-2 N H_2SO_4 containing Hanford concentrations of plutonium and fission products were stored at 30°C in stainless steel with various reducing agents. After eleven days of storage with 0.2 M $H_2C_2O_4$, or 0.1 M NH_2OH, or 0.1 M $NaNO_2$ + 0.02 M $H_2C_2O_4$, less than 1% oxidation occurred. Storage under the same conditions using 0.1 M $NaNO_2$ resulted in about 10% oxidation.

2. *NH_2OH as a prereduction agent.* With inactive UNH fortified with Site W concentrations of plutonium and fission products, plutonium losses in the bismuth phosphate extraction step averaged 2-3% after prereduction with 0.05 M NH_2OH. Losses increased to 4-5% when the NH_2OH concentration was increased to 0.1 M.

3. *Mechanism of the effect of N_2H_4.* In the presence of H_2SO_4 at 75°C, 0.1 M N_2H_4 reduces part of UO_2^{++} to U^{+4}. The mechanism of the effect of N_2H_4 on carrying might be due to one or a combination of three factors: interference by U^{+4} which is stabilized by N_2H_4; the presence of Pu^{+3} which might not be carried well in the presence of UO_2^{++}; or complexing of Pu(III) or Pu(IV) by N_2H_4 or its decomposition products.

B. Methods of increasing production

In earlier work, satisfactory yields were obtained from 28% inactive UNH solutions with 1.9 - 2.2 mg bismuth/cc. However, when active slugs were used, losses up to 22% were observed.

By reducing the HNO_3 acidity to 1 N through the addition of sodium acetate, decontamination factors of 1.1×10^4 for gamma-rays and 4×10^4 for beta particles were obtained for two cycles without scavengers. A plutonium yield of 74.5% was obtained of which 19.8% was lost in the extraction step. In runs with cerium-zirconium scavengers, decontamination factors of 1.2×10^6 for gamma-rays and 3.3×10^5 for beta particles was obtained after two cycles. The yield (overall) was 78.8% of which 16.8% was lost in the extraction step.

C. Decontamination Studies

1. *Low acidity by-product precipitation.* With 0.1 N HNO_3 in the by-product precipitation step, gamma-ray decontamination factors of 10^5 are obtained with 0.1-liter runs through two cycles. An acidity of 0.1 N HNO_3 is considered optimum. A factor of two or three additional improvement, however, may be possible if present studies of holdback carriers permit going to 0.05 N HNO_3 or lower acidities.

2. *Effect of HF.* The effects of various concentrations of HF in the plutonium precipitation step on decontamination have been studied. Gamma decontamination factors steadily improve with increase in HF concentration up to 0.05 M. Beta decontamination and plutonium yield are not affected.

Group 2. Concentration-Isolation

Results of experiments show that in the presence of 0.05 M iron, about 17 times the average Clinton iron content in the flowsheet $La(NO_3)_3$ solution, satisfactory plutonium precipitation yields can be obtained at Hanford using 0.5 N HNO_3 - 0.5 N H_2SO_4 with a 15-minute addition of H_2O_2 and a temperature of 40°C for one hour. (The Hanford flowsheet calls for a 60°C temperature.) These conditions are now being tested for the first time on plant solutions to verify the procedure and to determine maximum iron tolerances. If these tests are successful, it will probably not be necessary to make an iron separation at Hanford.

Although a large number of complexing agents for iron have been investigated for use in the $La(NO_3)_3$ solution or in the $KOH-K_2CO_3$ metathesis solution, none has been found entirely satisfactory to date. One promising method of iron removal is presently being tested, however, that involves the reprecipitation of lanthanum fluoride at the end of the crossover cycle, then dissolving of this precipitate in 20% of the original volume of 10 N HNO_3, and reprecipitation by making the solution 1 N in HF. Experiments carried out with radioiron tracer show that less than 1% of the iron originally present in the solution before lanthanum fluoride-plutonium precipitation in the crossover cycle is carried by the lanthanum fluoride precipitate before reprecipitation.

A procedure involving the precipitation of lanthanum fluoride from 10 N HNO_3 solutions of bismuth phosphate has been developed wherein satisfactory carrying of plutonium at Hanford concentrations by the lanthanum fluoride is obtained when the lanthanum/plutonium ratio is 4:1 or greater. This procedure shows promise for early operation use at Hanford where the plutonium concentrations will be about 30 grams per ton.

Group 3. Process Development

Data obtained by the semiworks group confirm the previous 0.1-liter and 1.0-liter-scale unmodified Bismuth Phosphate Process runs without the use of scavengers. It now seems fairly definite that the unmodified Bismuth Phosphate Process can be used at Hanford without scavengers and that adequate decontamination can be obtained — if not in two bismuth phosphate cycles, definitely in three cycles. Although the low acidity process gives a definite improvement in fission product decontamination over the unmodified process, plutonium losses are slightly higher.

Katzin summarized for me the work his special group has completed on the extraction of U^{233} from the two cans of thorium "carbonate" that were irradiated for about seven months in the Clinton pile. Each can contained about 164 grams of thorium and a small undetermined amount of natural uranium. The initial extraction was quite successful. Approximately 6.5 mg of U^{233} were extracted based on the half-life 1.2×10^5 years for U^{233}. Initial measurements (subject to correction) give a specific activity of 2×10^7 d/m per mg, which corresponds to a half-life of 1.7×10^5 years. Gofman, Stoughton, and I estimated the half-life of U^{233} to be 1.2×10^5 years in our original work on U^{233}. Katzin reports the range of U^{233} alpha particles, based on absorber foil measurements, to be 3.35 - 3.40 cm in standard air. A more precise determination, however, is expected using Jaffey's differential alpha-particle range chamber. Preliminary calculations using data from thermal neutron fission cross section measurements give a U^{233}/U^{235} cross section ratio of about 1.3, based on the value 1.2×10^5 years as the half-life for U^{233}. The data are consistent with an isotopic purity of 70% for the U^{233}. A more accurate value for the ratio of neutron fission cross sections, U^{233}/U^{235}, will be obtainable when a more precise half-life value determination is made following a mass spectrographic analysis of the isotopic composition of the sample.

Flox reported to Nickson that the samples of sputum and pieces of Kleenex used by Yett to blow his nose last Monday, give an alpha-particle count of 30 c/m and 50 c/m, respectively.

The Project Publication Advisory Committee held its initial meeting this afternoon and unanimously agreed to R. S. Mulliken's recommended plan for publication of a Proceedings of the Metallurgical Project. Mulliken sent a memorandum to Compton reporting the committee's action and requesting that Compton bring the matter to the attention of the members of the Project Advisory Committee this evening to obtain their consensus. He also included a tentative outline (MUC-RSM-190) of the contents. Thirteen sections are proposed. Two sections of primary interest to me are Section X, "Chemistry and Metallurgy of Transuranic Elements," consisting of three volumes, and Section XII, "Separation Processes," consisting of four volumes. The descriptive note attached to the Section X entry states, "Note that a survey volume on this subject is now under preparation, at the request of L. R. Groves, under the editorship of C. A. Thomas and J. C. Warner, with a special editorial board. The volumes now contemplated should be properly coordinated with this and, in general, with the work of the Los Alamos Project." The Committee also approved

the outline for the Thomas volume on "The Chemistry, Purification, and Metallurgy of Plutonium."

Helen worked at home today on the classified version of the "Table of Isotopes." She also took care of Kristine, the Ghiorsos's baby daughter (see Figures 16 and 17).

Industrial areas of five Japanese cities on Kyushu Island were bombed by U.S. B-29 superfortresses yesterday according to today's headlines.

The Metallurgical Project Advisory Committee met this evening for a six o'clock dinner session at the Quadrangle Club. This was immediately followed by a classified session in Eckhart Hall, Room 209. A major topic discussed was the interim program that the Metallurgical Project should follow. The need for the initiation of research in the fundamental sciences using project information, supported by private institutions (but under Manhattan District control regarding security) was discussed. Mulliken's plans for the publication of a "Proceedings of the Metallurgical Project" were discussed.

Tuesday, August 22, 1944

Hyman wrote Gilbreath about the semiworks operation. He says that some difficulties have been encountered in the semiworks runs on the low acidity decontamination procedure. These have been determined to be of an operational nature and not inherent in the low acidity procedure nor caused by the larger-scale operations. The plutonium yields and decontamination factors obtained in a low acidity and a control run have not been so high as should be ultimately obtainable. Hyman considers the low values are due to recontamination and other operational problems, and he suggests that further studies be made in the semiworks after the present equipment changeover has been completed.

Hogness reiterates in a memorandum to Compton his need for a scientific assistant to fill the position of Associate Director of the Chemistry Division when Selwood leaves. Many of the best administrative people who were in the Chemistry Division have been taken by Clinton, Los Alamos, and Hanford. Hogness says with the exception of me, those remaining in the Division are out of the question for consideration in filling the position. In my case, Hogness writes, "Seaborg would be a logical candidate, but he is so efficient in his present position that the Division would suffer if he were to be taken away from his present work." Because of these facts, Hogness believes that an outsider must be considered for the job, and again he strongly recommends Farrington Daniels.

Flox informed Arnold about a plutonium hazard revealed by exhaust-air studies conducted by Schoenberg on two International centrifuges. One centrifuge, which was badly contaminated in a previous spill, deposited hazardous amounts of plutonium on the filter paper of air sampling equip-

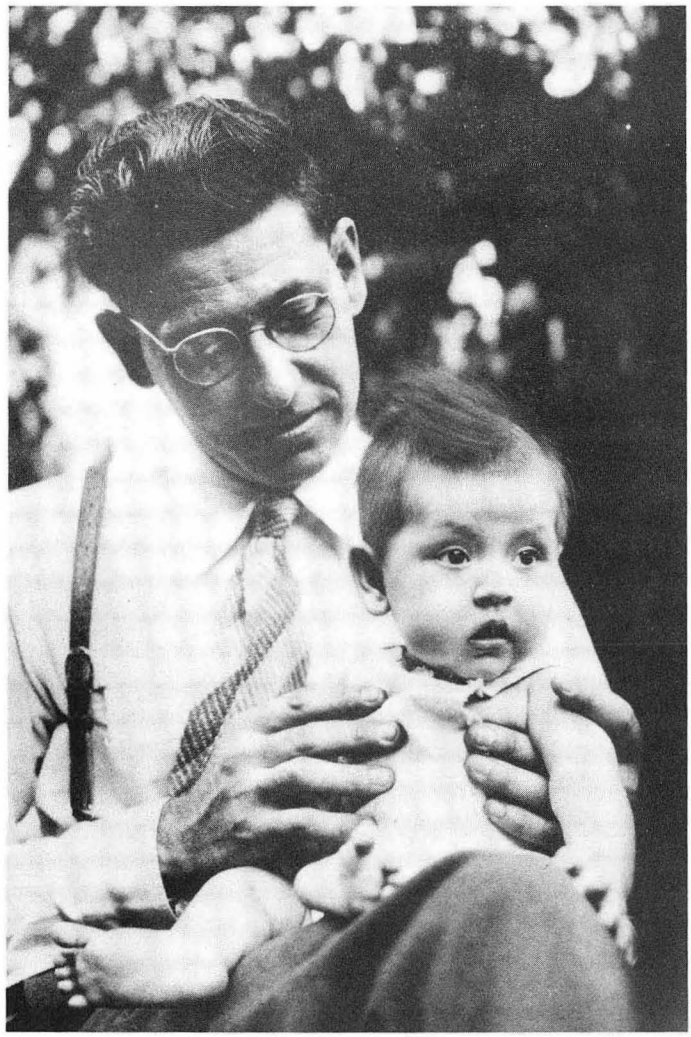

Figure 16. Al and Kristine Ghiorso. August 1944.
XBB 7810-13184

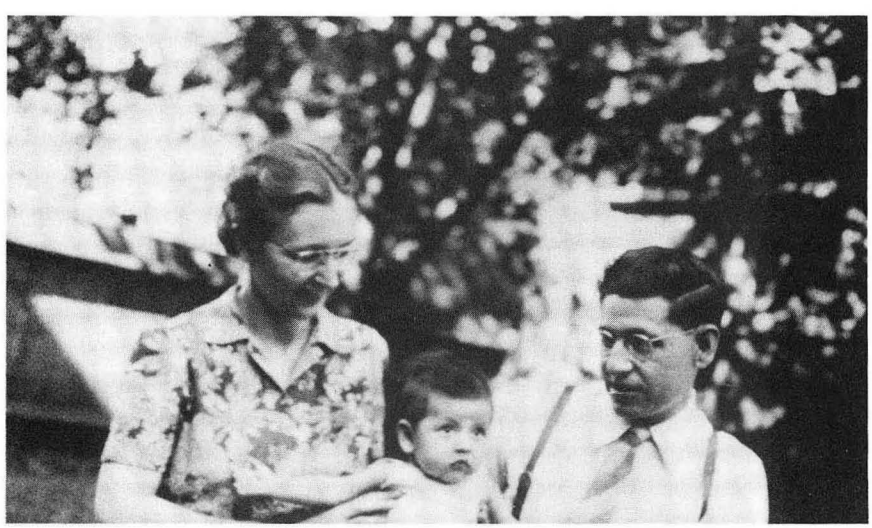

Figure 17. Wilma, Kristine, and Al Ghiorso. August 1944.
XBB 7810-13185

ment mounted near the hinge of the lid. The alpha-particle counts of plutonium collected in a similar way from a clean centrifuge were low, even though a 250-mg plutonium precipitate had been centrifuged for one hour. Thus it appears that a contaminated centrifuge is the source of the hazard. More tests will be run, and all centrifuges will be inspected on a weekly basis hereafter.

I attended the Project Council Information Meeting (Chemistry) in Room 209, Eckhart Hall, this morning. Others in attendance were Ashcraft, Bowman, Boyd, Brown, Burton, Cannon, Chipman, Compton, Conant, C. M. Cooper, Coryell, Dempster, Doan, English, Franck, Greager, Hogness, Howe, Huffman, W. C. Johnson, McKinney, Manning, Mulliken, Quill, Rabinowitch, Rollefson, Selwood, C. S. Smith, Smyth, Spedding, Squires, Stearns, Sugarman, Sutton, Szilard, Turkevich, Vernon, Warner, Whitaker, Wigner, Willard, and Zinn. Stearns opened the meeting, and Hogness introduced the Chicago Chemistry Division speakers. Turkevich reported the work for Sugarman's section (C-III). Fission-product krypton and xenon gases have been studied by catching decay products on charged wires. To date, Kr^{91}, Kr^{92}, Kr^{95}, Xe^{133}, Xe^{139}, Xe^{140}, and Xe^{141} have been detected. The longest-lived rare gas fission product found so far is Xe^{133} with a 5.3-day half-life. Measurements on fission recoil ranges were discussed. Burton reviewed the work of his section (C-II) which includes work at the Argonne pile on the Hanford chromate problem, and the graphite radiation effects problem. In the latter case, recent repetition of the Fulbright graphite resistance vs exposure experiment conducted in vacuo gives no concave nose on the curve, as previously reported. The total exposure of graphite samples has now reached about four days of equivalent Hanford pile exposure. This fact emphasizes the impossibility of obtaining long-range data on graphite before the Hanford pile startup.

Following an introduction by Hogness, I presented information on Section C-I studies associated with Hanford problems and basic chemistry. I pointed out that most of the remaining purification people are now associated with the basic chemistry work. I presented a summary of the work (Thompson's Sub-section I) on the problem involving the effect of iron in the isolation step. This information was contained in Thompson's memo to me yesterday.

I outlined our progress in solvent extraction procedures and described the hexone-water system being used with our one-inch column. Extractions using one-gram amounts of plutonium in lanthanum fluoride slurry from Clinton have given yields of about 98-99%. An attempt has been made to simulate in the laboratory the Hanford plant conditions for carrying out centrifugation for decontamination using no scavengers. A centrifugation of one G was used, i.e., the solutions stood for one hour on the laboratory bench after which the supernatants were siphoned off. Gamma-ray decontamination factors of about 2.9×10^4 were obtained at the end of two cycles; 1.4×10^6 was obtained after three cycles.

In the semiworks runs, using the unmodified Bismuth Phosphate Process but without scavengers, decontamination factors of 1.6×10^4 were obtained through two cycles; a third cycle with 1 G centrifugation gave 6×10^5.

I reported the results of our studies on the green "abnormal" Pu(IV) and its colloidal nature. Our work suggests that "abnormal"

Pu(IV) is a polymer but seems to have no bad effect on the process if precautions are taken. The results of our studies on Np^{237} were presented. I reported that, based on direct measurement, Np^{237} is present in the pile to the extent of approximately 0.33% of the Pu^{239} present. This checks with work by Turkevich. In the chemical plant, Np^{237} is lost in the initial plutonium-carrying step because of the presence of small concentrations of iron that causes the neptunium to stay in the supernatant liquid. If no iron is present, neptunium is carried.

I reported that work on crucibles is progressing and that plutonium metal does not seem to attack CeS or ThS. Davidson's vapor phase studies to produce $PuBr_3$ from the action of HBr on PuO_2 were described. I briefly mentioned the work Cunningham's sub-section is doing on the behavior of PuF_6, Phipps and Simpson's plutonium vapor pressure studies, and Zachariasen's identification of PuS_2 and $(NH_4)_2Pu(NO_3)_6$. The successful separation of over 6 mg of U^{233} from Clinton-irradiated thorium, described in Katzin's memo to me yesterday, was reported. I said that the question of the half-life for U^{233} is still not settled but may be as high as 1.7×10^5 years instead of our earlier determination of 1.2×10^5 years. W. C. Johnson said that initial analyses show that there is about 0.05 ppm natural uranium impurity in the starting thorium "carbonate" leading to about 10% natural uranium in the U^{233}. (If this is correct, the half-life may be about 1.53×10^5 years.)

I announced the results of the helium ion bombardment of 10 mg of Pu^{239} in the 60-inch Berkeley cyclotron. A new alpha-particle activity has been found in the reaction with a range of 4.7 cm in air as compared with plutonium-239's 3.65-cm range. About 100 c/m of the product has been isolated, and chemical studies show the new activity to be fluoride-insoluble under all conditions. As the new activity cannot be oxidized to a fluoride-soluble state, it is possible to separate it from plutonium in a fluoride precipitation step in which the oxidized plutonium is left in solution. The new activity is carried by lanthanum oxalate from excess alkaline oxalate; it is not carried by zirconium phenyl arsonate; it probably does not have a valence of four under these conditions; in addition, it is not carried by bismuth phosphate. After considering these facts, we can eliminate elements, 94, 93, 92, 91, and 90 as possibilities. The new alpha-particle activity is therefore probably due to element 95 or 96.

After I completed my presentation, Stearns called upon W. C. Johnson to introduce the Clinton chemistry speakers. English spoke first. He stated that four sets of problems have been studied at Oak Ridge: (a) the effect of zirconium in the lanthanum fluoride concentration cycle in Room D; (b) the interference of zirconium in the plutonium peroxide precipitation step; (c) reactions of plutonium in alkaline solution; and (d) decontamination studies. With regard to the studies on plutonium in alkaline solution, English said that, on the basis of analyses made by spectrophotometric methods, Pu(VI) is reduced to Pu(IV) with peroxide and sulfite in solutions containing 45% K_2CO_3 and 0.05 M plutonium; Pu(IV) is oxidized to Pu(VI) with hypochlorite; the effect of sulfite is anomalous in that absorption peaks are low and may indicate the formation of Pu(III). I pointed out that the solubility of Pu(III) may be such that 45% K_2CO_3 is needed to give appreciable solubility, whereas Pu(IV) and Pu(VI) may be considerably more soluble in 45% K_2CO_3, or that

a lower pH will give the same solubility for Pu(IV) and Pu(VI) as a higher pH might give for Pu(III).

Coryell presented work done at Clinton on I^{131} production from the decay of Te^{131}. The half-life of I^{129} should be obtainable after the eight-day I^{131} has decayed out of the sample. Coryell said work on the tritium problem is progressing and that 10% yields of tritium have been obtained in recent experiments. About half of Coryell's laboratory space and 25 men are now employed on the barium-separation project. Radiobarium is being distributed to various Project groups for experimentation; Site Y is especially interested in acquiring appreciable quantities of Ba^{140}. Strontium-90 has been separated and transferred to the Health Group.

Boyd reviewed his section's work at Clinton on decontamination studies which includes: (a) carrying mechanisms, (b) precipitation processes, (c) colloid studies, and (d) plant orientation studies. An interesting explanation of the carrying of lanthanum by bismuth phosphate, on the basis of co-precipitation and surface effects, was presented.

Sutton presented the Clinton semiworks and process development work. Intensive process development work on the bismuth phosphate separation process, started 15 months ago, has resulted in the successful demonstration of the process on the Clinton plant scale of operations. Attention is now being directed toward expected problems at Hanford that could result from differences in plutonium and by-product concentrations and effects of the aluminum-silicon protective coatings on the Hanford-type slugs. The objectives will be to obtain 10^5 gamma-ray decontamination at the end of two cycles and 10^7 decontamination at the end of the crossover cycle and as high a plutonium yield as possible. Isolation procedures are also being worked on. Sutton hopes to attain these objectives within the next month. He presented a set of data summarizing his expectations about the performance of the bismuth phosphate process at Hanford. Overall plutonium yields at the end of the plutonium isolation step should be 92% at the highest and 80% at the lowest.

Spedding reported on the work at Ames relating to the production of cesium and thorium metal. I asked how much thorium metal he is planning to make. He replied that it could be produced on a large scale, but the quantity must be decided by Washington. C. S. Smith from Los Alamos reported that several 10-gm samples of plutonium have been made which have a density of about 19.9 gm/cm^3. Since the alpha form of the metal is very ductile, the beta form must be used for fabrication of component parts above 150°C. The coefficient of expansion for alpha plutonium has been measured to be 63.5×10^{-6} per degree C (average of measurements by two methods). This is a large value compared with other metals and is about twice the coefficient of expansion for the beta-form of plutonium. Smith also reported that the change in purity specifications now makes plutonium fluoride the better choice for reduction by metallic sodium than plutonium chloride.

Helen worked at home today on the classified "Table of Isotopes."

Allied troops are battling inside Toulon, and U.S. troops are enveloping Paris, according to today's newspaper.

Wednesday, August 23, 1944

Beard reported to Pye the results of experiments he has run to test the feasibility of dissolving the products of metathesis in, and precipitating plutonium peroxide from, acid solutions other than HNO_3. Trichloroacetic acid is one of the acids tried. Although the lanthanum and plutonium hydroxides failed to dissolve in trichloroacetic acid, there was evidence that at least one of the two hydroxides was converted into a new compound, probably plutonium trichloroacetate. Subsequent experiments show that the lanthanum hydroxide readily dissolves in the acid and that the plutonium hydroxide is converted into an insoluble, pink compound. This compound is found to be quite soluble in acetone and can be precipitated from acetone using 30% H_2O_2 to form plutonium peroxide with only a 2.4% loss in plutonium. Two additional runs have been successfully made to study the process. As an explosive compound might be formed with acetone in the presence of hydrogen peroxide, however, the isolation of plutonium peroxide using this process does not look practical. Several other solubility studies have been conducted.

I attended the 7:45 p.m. meeting of the Extraction and Separation Processes Sub-section I of my section. Others in attendance were Ader, Albaugh, Beard, Blaedel, Brody, Egan, Fineman, Gilbreath, Hindman, Hoekstra, Hopkins, J. J. Katz, Katzin, Kraus, Larson, Lawroski, Leventhal, Manning, Margolis, Meyer, Morgan, S. Peterson, Sheft, Stewart, S. Thompson, Walling, and others. Hopkins reported on the improvement in yield in plutonium peroxide precipitations from $La(NO_3)_3$ solutions containing iron. This information was also covered in the Group 2 portion of Thompson's memo to me of August 21. Brody discussed the hexone extraction-purification process devised as an auxiliary plutonium concentration and isolation procedure for possible use at Hanford. The results obtained with the synthetic solution, containing zirconium, lanthanum, 3 M HNO_3, and no plutonium, were encouraging with regard to the separation of lanthanum and zirconium. It is evident that the desired plutonium purity of 99% should be easily obtainable. Further work is planned using radioactive lanthanum and zirconium tracers and tracer amounts of plutonium.

S. Thompson reviewed the developmental history of the mainline process from its birth in 1942 to the present. Although the details of the process have changed considerably in the last year, the process remains basically the same as when first proposed. The principal problems still remaining are: (a) elimination or alleviation of the effects of iron in the isolation steps; (b) improvement of prereduction step; (c) reduction of bismuth phosphate by-product loss in the presence of scavengers.

Helen worked at the Crerar Library on the classified "Table of Isotopes."

"Yank Tanks Drive 65 Miles" is today's banner headline; U.S. troops pushed past Paris.

The Project Council Policy Meeting was held today in Room 209 of Eckhart Hall. The meeting was begun at 9:00 a.m. and was attended by W. Bartky, Chapman, Chipman, Compton, C. M. Cooper,

Dempster, Doan, E. T. Filbey, Franck, Greager, Hogness, Howe, L. O. Jacobson, Jeffries, W. C. Johnson, Leverett, Mulliken, A. V. Peterson, Smyth, Spedding, Stearns, Szilard, K. K. Tracy, Vernon, Warner, C. J. Watson, Whitaker, Wigner, and Zinn.

Compton reviewed recent suggestions he made to Conant that the transfer of nucleonics to the postwar period should be accomplished with a change in organization to provide for a "coordinating board," reporting directly to the President, under which would be an organization for military applications and an organization for scientific, industrial, and medical developments and applications. Compton said in his declaration that the war is not over until "there exists a firm international control over the production of nucleonic weapons." During the present interim period our major objectives are to provide backup for Sites W and Y, produce U^{235} and Pu^{239}, improve production methods, and look at possible military applications of fission products. As we approach the postwar period, we need to start encouraging private research and the exchange of technical information. Public education concerning the value of nucleonics is urgently needed. Smyth will prepare information for public release when this becomes possible.

Compton then summarized his memorandum to Conant by expanding on the outline:

A. Projects of Probable Military Value before 1946
   1. Improvement of pile and its production level.
   2. Improvement of chemical separation process.
   3. Support of Site Y program.
   4. Services (technical and health).

B. Projects of Possible Military Use before 1948
   1. Pile studies.
   2. Improved chemical separation studies.
   3. Tubealloy (uranium) supply.
   4. Medical and biological studies.

C. Projects of Scientific, Medical, or Industrial Value
   1. Neutron physics.
   2. Isotope studies.
   3. Fundamental chemistry.
   4. Hot laboratory studies.
   5. Biological.

The heading, "Fundamental Chemistry," was further divided into five parts with suggested laboratories for each specific research [e.g., C for Chicago and X for Oak Ridge]. The parts are (a) of heavy elements over No. 87 [C, X]; (b) of fission products and rare earths [Ames, C, X]; (c) search for elements 95 and 96 [X]; (d) nature of coprecipitation and other processes [C, X]; (e) uses of tracers in chemistry (microtechniques, Szilard-Chalmers reactions [C], chemical reactions induced by ionization [X]). Of particular significance is Compton's suggestion that the search for elements 95 and 96 take place at Clinton Laboratories and not at the Met Lab here in Chicago.

According to Compton, the Manhattan District feels that the

personnel level for the Project has reached its maximum level and that no additional men should be hired except for highest priority work.

Howe reported on Hanford Engineer Works progress and said that, in general, everything is progressing favorably. The 105 B pile is essentially completed and should start up before September 1. The 105 D pile is nearly ready to be loaded with graphite. Pile 105 F is much less advanced. Enough slugs are on hand for the first loading. Spedding said he is planning to produce about 500 pounds of thorium metal in the next six weeks and would like the production order confirmed. Smyth stated that he believes it is perhaps as important to get good project record reports written as it is to do further research work. It will be a big job. Peterson remarked that Mulliken has asked for a staff of 12 to 18 scientists to prepare such reports.

Thursday, August 24, 1944

Sheft is attempting a high-temperature reaction to prepare PuI_3 from plutonium carbide using Fried's vacuum sublimation furnace. The sample, a mixture of carbides from Westrum's reduction of PuF_3 (sample 188) in a graphite crucible using lithium, was loaded into a platinum "boat," inserted into the furnace, and the system was evacuated, heated to 1300°C, and left to pump overnight.

E. O. Lawrence telephoned me from Berkeley about the patent matter. He is concerned that we would consider receiving personal profits. Lawrence would prefer that we make a free gift to the University, with the University, in turn, making a free gift to the Government. Lawrence has already called Segrè and indicated he would also call Kennedy. He will recommend to the Regents at their meeting tomorrow that we make such a gift to the University and the University give the cases to the Government. I was non-committal in my response.

I received a copy of Warner's outline for the Project volume on "The Chemistry, Purification and Metallurgy of Plutonium," which was approved by the Project Publication Advisory Committee on August 21.

Helen worked at home and later practiced golf at Jackson Park.

Rumania has accepted peace terms of the Allies, according to today's news.

Friday, August 25, 1944

Orlemann, Watt, and Roy Thompson returned to Chicago this afternoon from their visit to Clinton.

Sheft admitted HI into the sublimation furnace containing the plutonium carbide sample. The heater filament burned out in about two minutes. A brown material was deposited on the cold finger and other cold surfaces. After pumping for several hours, nearly all the iodine was removed and a brown ring was left on the cold finger, which had by now warmed up to room temperature. An attempt was made to obtain a large enough sample to fill an x-ray capillary, but there was not enough material present in the brown ring.

Approximately 50 mg of plutonium in an HCl solution was spilled on the floor in Room 35 of the New Chem Building today. This room is used by Stewart, Asprey, and Anderson. Most of the material was blotted up with Kleenex for recovery; however, the floor could not be cleaned with repeated washings and several reagents. Asprey obtained permission from Captain Ware to cut away the piece of linoleum involved. About 5 mg of plutonium may still be in the linoleum piece, and an attempt will be made to recover it.

Kathleen Gavin and Alan Florin are being married today in her home at 908 W. 51st Place.

Helen and I went to the Tam O'Shanter Golf Tournament with Zene Jasaitis and Stan Thompson. Following the tournament we all had dinner at Gus's Good Food Restaurant at 420 N. Dearborn.

A Free French Radio broadcast today says that U.S. troops have entered Paris.

Saturday, August 26, 1944

Magnusson today completed an attempt to prepare sodium neptunyl acetate. He freed 50 micrograms of Np^{237} from plutonium by repeated bromate cycles and reduced the amount of lanthanum carrier to 50 micrograms, ending with a volume of 7 microliters. After a final extended oxidation with $KBrO_3$, a solution of $NaNO_3$-NaAc was added, whereupon a crystalline precipitate formed which appeared colorless to faint pink under incandescent light and green in daylight. The precipitate was sealed off in a capillary and given to Zachariasen for x-ray diffraction analysis.

The results of a mass spectrographic analysis on the U^{233} isolated from the two cans of irradiated thorium carbonate by Katzin was reported today by Dempster; he finds an isotopic purity of 95.5 ±1.5%. Using this information, Katzin has recalculated the specific activity of U to be 2.1×10^4 dis/min/microgram and the half-life to be 1.58×10^5 years.

Continuing the study of the chemical properties of elements 95 or 96, James completed an experiment which demonstrates that the new element is not carried by lead sulfate from acid solution.

Later, James left on a trip to St. Louis to bring back the 200-mg plutonium sample that has been undergoing deuteron bombardment at the Washington University cyclotron since August 14.

Report CN-2080, "Technical Division — Weekly Report for August 19 to August 26, 1944," was issued and contains the following information on the solvent extraction program. The principal effort of Dawson's group during the week has been devoted to carrying out additional laboratory batch extraction experiments and measuring the equilibrium distribution coefficient for lanthanum using a radioactive tracer. The present feeling with respect to a batch process is unfavorable because recent evaluations by Dawson's and Lawroski's groups indicate that the lanthanum and zirconium distribution coefficients are considerably lower than originally reported. This would necessitate carrying out at least two batch extraction cycles in order to obtain the required purification.

A second run in the 19-mm glass continuous countercurrent extraction system has been made by Lawroski's group with approximately the same operating conditions as for Run No. 1. The main objective of this run was to attempt to check the favorable results previously obtained. The feed solution contained all substances in Hanford concentrations except for plutonium, which was not present. No mechanical problems were encountered. Satisfactory progress was made on the construction of the 3-inch continuous countercurrent extraction system located in the center stairwell at West Stands. Initial operation will begin in early September.

This evening Helen and I played cards with the Ghiorsos.

Rumania has declared war against Germany. The Free French radio has announced that General Charles de Gaulle is in Paris.

Sunday, August 27, 1944

The deuteron bombardment of 200 mg of plutonium at the St. Louis cyclotron is scheduled to end at 6:00 p.m. It was started on August 14, 1944. Ralph James is in St. Louis to handle the sample.

Ghiorso, Lawroski, Foster York, and I played 18 holes of golf on the Evanston Community Course (par 68). Our scores were AG-84, SL-91, FY-80, GS-82. We had planned to go to the Tam O'Shanter Open, but it was rained out. Helen and I visited the Foster Yorks and stayed overnight.

Monday, August 28, 1944

Donald Ames returned to work in my section as an SED man after being away on basic training with the Army.

Zachariasen reports that the preparation Magnusson gave him yesterday shows an x-ray pattern identical to the patterns for sodium uranyl acetate and sodium plutonyl acetate. Hence it may be said that the compound is sodium neptunyl acetate, and the existence of a VI oxidation state for neptunium is established.

A meeting was held in West Stands to discuss and make final decisions concerning the health protection measures to be taken to insure that the operating personnel of the 3-inch solvent extraction system, located in the center stairwell at West Stands, will not be subjected to avoidable health hazards. In attendance were Ware, Tepe, Nickson, Rose, Duffey, and Buffum. Six preventative radiation safety measures were decided upon.

Helen and I went to the Tam O'Shanter Golf Tournament with Foster York. Byron Nelson won the tournament over Ed Dudley 280 to 285. After the tournament we went to dinner at Tiffin's (in the market called Stop and Shop, 16 W. Washington Street).

On the western front U.S. troops have reached the Marne. On the eastern front Soviet troops are within 75 miles of Rumania's capital. The Rumanians are trying to protect their oil fields.

Tuesday, August 29, 1944

Simpson had the first meeting of his Group 6A today at 8:30 a.m. Robinson described his apparatus for the measurement of vapor pressure of plutonium compounds. Westrum recounted his unsuccessful attempts to produce plutonium carbide by the reaction of PuO_2 with carbon and PuO_2 plus carbon tetrachloride. Hellman described his experiments on the reactions of Ca_2Si with PuO_2 and PuF_3. Fried told about his sublimation studies whereby PuF_4 is reduced to PuF_3 at high temperatures (900-1000°C). Sheft described the production of $PuBr_3$ from PuO_2 plus HBr at 700°C. The attempts to produce PuI_3 by heating plutonium metal with HI have not been successful to date. The current attempts to use HI with plutonium oxalate, or PuO_2 plus carbon, were mentioned. Finally, Sears reviewed his work on the vapor pressure of plutonium halides.

Joe Kennedy called me from Los Alamos. Lawrence called him last Thursday, and again on Friday, about the patent matter. In the Friday call, Lawrence told Kennedy he does not think it appropriate for us to sell our patent rights. He was planning to meet with the University Regents to urge the course of free gifts of the cases from us to the University and from the University to the Government. Lawrence has also talked with Vannevar Bush, who mentioned the possibility of later rewards for the inventors.

I read a copy of R. E. Clark's memo to C. M. Cooper concerning the disposal of active wastes at Chicago. The memo supplements a previous memo of August 5 and deals with procedures for both temporary and final waste disposal as well as the recovery of materials from wastes.

Ralph James returned today from his trip to St. Louis with the 200-mg plutonium sample that has received 47,842 microampere-hours of deuteron bombardment at the Washington University cyclotron. Upon returning to the lab, James began the search for element 95 by dissolving part of the irradiated plutonium material off the target sample. The dissolved

sample (labeled sample 49DD) was found to contain about 90 mg of the original 200 mg of plutonium. A disc containing one mg was prepared for Ghiorso to use for range measurements. The remainder of the solution was heated with dichromate (to oxidize the plutonium) and a lanthanum fluoride precipitation made (to carry down any element 95 present). The supernatant liquid was labeled sample 49DD-1.

"Chemical Research — Basic Chemistry of Plutonium, Report for Month Ending August 1, 1944," (CN-1948), includes the following information:

I. Basic Chemistry Group (Hindman, Group Leader). O'Connor has completed measurements on a number of plutonium compounds under the exact conditions now expected in the bismuth phosphate precipitation process at Hanford. The solubilities obtained in simulated Hanford mainline process solutions for Pu(VI) fluoride show that no precipitation of product without carrier will occur in the Hanford operations. Cliff Smith, in his studies of the oxidation state of plutonium in co-precipitation reactions, has obtained results which show that when lanthanum fluoride is precipitated in the presence of Pu(IV), the plutonium in the precipitate is Pu(IV) and not Pu(III) as had been proposed by some. McLane has clearly demonstrated, in his collodion membrane dialysis experiment, that the green "abnormal" Pu(IV) is a colloidal material. A solution prepared by Kraus, which he tentatively called "a green polymeric Pu(IV) ion," is found to be only 15-16% dialyzable.

Kraus and Hindman have studied changes that occur in solutions of "abnormal" Pu(IV) in 1 M HNO_3 on standing. It has been previously observed that the green color of some solutions of Pu(IV) in HNO_3 allowed to stand in the laboratory, changes from greenish-brown to ochre with time. These visible changes are accompanied with changes in chemical properties and in absorption spectra. The precipitation properties for plutonium, in the form of the freshly formed nondialyzable Pu(IV), are greatly altered so that stoichiometrically minute amounts of IO_3^-, $C_2O_4^=$, etc., cause almost complete precipitation.

Magnusson and La Chapelle have isolated an additional 33 micrograms of Np^{237} from Clinton material. Minute quantities of a yellow crystalline neptunium chloride and a pale green crystalline neptunium sulfate have been prepared. An x-ray diffraction pattern for the neptunium sulfate sample was obtained by Zachariasen; however, the crystal structure has not yet been determined. There is an insufficient amount of the neptunium chloride for x-ray analysis. Howland has conducted studies that prove the validity of the lanthanum fluoride assay for tracer amounts of Pu(IV) in the presence of trace amounts of Pu(VI). The lanthanum fluoride assay technique gives results that compare favorably with results obtained spectrophotometrically. Howland also conducted storage studies and finds that when Pu(IV) is stored in 40% UNH-0.2 N HNO_3 at room temperature, an equilibrium mixture of 36% Pu(VI) and 64% Pu(IV) occurs in 71 hours. No formation of the polymerized "green form" of Pu(IV) has been observed in nine days of storage. Clifford Smith has determined that the rate of gas evolution from a solution of Pu(IV) nitrate in about 6 M nitric acid (DNO_3 in D_2O) is about 0.58 ml per gram of plutonium per 24 hours.

II. Recovery and Service Group (Dawson, Group Leader). Asprey purified a relatively large batch of plutonium from Clinton by dissolving

in HNO_3, making the resulting solution 5 M in NH_4NO_3 and 3 M HNO_3, and extracting the plutonium(IV) from this solution with methyl isobutyl ketone followed by shaking with distilled water to remove the plutonium from the ketone. Except for 140 ppm of calcium, no other cation impurities were detected upon spectrographic analysis.

H. H. Anderson, Asprey, Britain, Burr, Fineman, Leventhal, and Stewart have all participated in work related to plutonium recovery. Since the initiation of the program on March 1 of this year, the group has made progress in its effort to develop solvent extraction methods for the concentration, isolation, and purification of plutonium. At first diethyl ether was used exclusively. Methyl isobutyl ketone, now being used as the extractant, is much less hazardous and satisfactory yields and plutonium purity can be readily obtained with it. The success that has been achieved for the solvent extraction method on a laboratory-recovery scale makes it a logical candidate for the mainline concentration and isolation process at Hanford. A development program to this end has recently been initiated in cooperation with the General Engineering Station.

Fields has spent approximately one-third of the month at Clinton Laboratories where he has separated Np^{239} from UNH which was irradiated in the Clinton pile. The isolated material was added to plutonium solutions in the Clinton mainline process to serve as a tracer for the Np^{237} isotope. Britain is considering ways to separate about 99.9% of the Np^{239} from fission products in the bismuth phosphate cake solution in order to ensure that the Np^{239} does not interfere with accurate measurements of decontamination factors for fission products in the succeeding steps of the Hanford mainline process. Solvent extraction methods for accomplishing this degree of separation appear promising.

Stewart, Fields, and Fineman have done some preliminary work on plutonium decontamination that indicates extraction with methyl isobutyl ketone from a solution of bismuth phosphate from the extraction step may produce decontamination factors of from 20 to 200 for combined beta particles and gamma-ray activity. Plutonium has been extracted from the supernatant solution resulting from a bismuth phosphate by-product precipitation. In both cases $(NH_4)_2SO_4$ was added to the solution being extracted.

Katzin and Van Winkle have worked on uranium ore processing and the analysis of ore samples for protactinium. Preliminary results indicate that the acid soluble-carbonate insoluble fraction resulting from uranium ore processing contains about 1.22 ppm of protactinium and about 10 ppm of ionium (Th^{230}). Katzin is currently preparing to extract U^{233} from thorium irradiated with neutrons at Clinton using solvent extraction.

Helen and I had a farewell dinner with the Roy Heaths at Nielson's (on Western Avenue). Stan Thompson spent his first night with us in our apartment. Since Alice is vacationing with her parents in California, Stan will stay with us until he leaves for Hanford around September 26.

U.S. troops have advanced to Chateau-Thierry. German troops are pulling out of Italy and falling back toward the Alps and Brenner Pass.

Wednesday, August 30, 1944

Today is the last day at the Met Lab for Roy E. Heath. His termination is effective September 5 when he will return to the Wyandotte Chemical Company.

James continued working up the large sample of plutonium that has been bombarded with deuterons in the St. Louis cyclotron. He carried out another dichromate cycle on the "95 fraction."

I received a memo from Hogness stating that all recommendations for work to be done in the 100 areas at Site W must be transmitted through Allison. All work desired in the 200 areas must be recommended through Whitaker. He also asks for recommendations of experiments we might conduct at Site W.

A meeting was held to discuss progress and plan future work on the development of the solvent extraction process for the concentration-isolation of product. In attendance were Maloney, Dawson, Lawroski, Stewart, and Tepe. The results of work completed to date on the determination of equilibrium distribution coefficients for lanthanum and zirconium between hexone and aqueous salt solutions were reviewed. The reason for the discrepancy between the results based on spectrographic analysis and those determined using radioactive tracers has not yet been discovered. Also, it is not known for certain which values are correct. The general feeling favors those determined using radioactive tracers. For solutions having about the concentrations of lanthanum and zirconium that would be suitable for feed to a continuous countercurrent extraction system (approximately 6 gm/l lanthanum and 19 gm/l zirconium), the best estimate at this time is that the zirconium distribution coefficient is 80 to 100. The value of the lanthanum distribution coefficient is apparently not less than 1000. These values are lower by a factor of about 100 than the values originally reported and based on spectrographic analysis.

The advisability of doing additional work on the determination of distribution coefficients was discussed. Lawroski feels strongly that the correct values of distribution coefficients must be established as soon as possible. This is particularly true if a sound basis is to be established from which to project future work on the development of solvent extraction methods for separating product from uranium and fission products and for performing other operations in the separation plant.

Dawson said that experiments conducted by his group indicate that a batch extraction process probably cannot be successfully carried out using solutions sufficiently concentrated (25 gm/l of lanthanum and 50 gm/l zirconium) to permit the use of the small equipment planned for installation in the 231 building at Hanford. Such solutions have been found to form stable gels when repeatedly contacted with hexone in laboratory-batch extraction experiments. The cause for this gel formation is not known. The degree of agitation and other operating variables are apparently contributing factors.

A decision was made that basic chemical investigations in the immediate future will be directed toward (1) establishing definitely the

values of the distribution coefficients for zirconium and lanthanum for solution concentrations corresponding to those which might be used in batch or continuous extraction processes, and (2) determining the conditions for carrying out a batch process necessary in order to obtain the required yield and purity to avoid physical difficulties such as gel formation. Dawson, Lawroski, and Tepe agreed to cooperate in preparing, by the 10th of next month, a progress report covering all work to date on the solvent extraction concentration-isolation process development.

I attended the meeting of the Basic Chemistry, Recovery, and Instruments Groups of my section which met at 7:45 p.m. in Room 209 of Eckhart Hall. Others present were Ader, Beard, Cunningham, Davidson, Dawson, Dixon, Gilbreath, Hindman, Hoekstra, Hopkins, Howland, James, Katzin, Kraus, Krueger, Magnusson, Malm, Manning, Morgan, O'Connor, Orlemann, Sig Peterson, Sheft, Simpson, Clifford Smith, Roy Thompson, Watt, and others. I called on Cunningham to preside as usual. James reported on the isolation of the alpha-particle emitting isotope with approximately 200 c/m which was produced in a 40 microampere bombardment a month and a half ago of a 10-mg Pu^{239} target with 32 Mev alpha particles in the Berkeley cyclotron. The isotope was isolated by oxidizing with dichromate in nitric acid and carrying out lanthanum fluoride precipitations of the resulting solutions. The lanthanum fluoride precipitate, which carried the isotope, was dissolved by fuming in sulfuric acid; and several additional lanthanum fluoride precipitations were carried out. The new activity detected has a range about 1 cm longer than alpha particles emitted by Pu^{239}. The following chemical properties for the new isotope have been found to date:

1) The isotope cannot be oxidized to a fluoride soluble state by $Cr_2O_7^{--}$ in nitric acid.
2) It cannot be oxidized by $Ag^{++}—S_2O_8^{--}$ to a fluoride soluble state.
3) It can be precipitated by lanthanum oxalate in neutral solution in the presence of excess oxalate.
4) It cannot be precipitated with zirconyl phenyl arsonate either under reducing (SO_2) or oxidizing (after prolonged treatment with $Cr_2O_7^{--}$) conditions.
5) It cannot be carried by bismuth phosphate under normal conditions (1 M HNO_3, 0.1 M H_3PO_4).
6) It cannot be carried by lead sulfate from acid solution.

On the basis of these experiments, either element 95 or 96 is the probable cause for the activity, with element 96 being the more probable. James presented the following data that shows that elements 82 through 94 cannot be the source of the new activity:

Element	*Reason for elimination*
Pb	would be carried by lead sulfate
Bi	would be carried by bismuth phosphate
Po	would be carried by bismuth phosphate
85	does not behave like halogen
Rn	would be volatile

87	is not an alkali since it has an insoluble fluoride and oxalate
Ra	would be carried by lead sulfate
Ac	would be carried by bismuth phosphate
Th	would be carried by zirconyl phenyl arsonate and bismuth phosphate and would not be carried by lanthanum oxalate in the presence of excess $C_2O_4^{--}$
Pa	would be carried by bismuth phosphate and zirconyl phenyl arsonate
U	would be oxidized to fluoride soluble state
93	would be oxidized to fluoride soluble state
94	would be oxidized to fluoride soluble state

The half-life of the activity has been estimated to be greater than two months. Either element 95 or 96 could be produced from Pu by one or more of the following reactions: α,n; α,2n; α,p; or α,pn to yield 96^{242}, 96^{241}, 95^{242}, or 95^{241} respectively. Analysis of the daughter products of the activity may lead to positive identification.

During the discussion period I referred to the information obtained concerning carrying properties and pointed out this evidence does not lead to any unique oxidation state for the new isotope. I summarized this evidence as follows:

Carrying Evidence	*Possible oxidation states*
lanthanum fluoride	2,3,4,5
$La(C_2O_4)_2$	2,3,5
Not by zirconyl phenyl arsonate	2,3
Not by bismuth phosphate	2
Not by lead sulfate	3,4,5

I also pointed out that element 95 would be predicted to have a very stable +3 state if a new "rare earth series" starts with actinium. Actually, the expected stability of the +3 state was used in devising the method of isolation being used by James.

Hindman reported on his work related to nitrate complexes of Pu(IV). He has concluded from experiments that two types of nitrate complexes can exist. The first nitrate complex is formed nearly completely when the HNO_3 concentration reaches 2 M and seems to consist of one mole of NO_3^- per mole of Pu(IV). The second is formed rapidly when the HNO_3 concentration exceeds 4 M and seems to involve five nitrate ions in the following reaction: $Pu(NO_3)^{+3} + 5NO_3^- \rightarrow Pu(NO_3)_6^{=}$. The complex ion $Pu(NO_3)_6^{=}$ is brilliantly green and can also be formed with calcium nitrate as well as with HNO_3. The absorption spectrum of hexone-extracted Pu(IV) nitrate suggests that a similar complex ion is present in that solution.

McLane reported on Dixon's and his experiments on electrical transfer. The results showed that Pu(IV) becomes negatively charged beyond 4 M HNO_4, is appreciably negatively charged in 0.1 M $SO_4^{=}$, becomes negatively charged in 10 M HCl, but is not negatively charged in 10 M $HClO_4$. It was also found that Pu(IV) in process solutions at the extraction step is largely negatively charged.

Manning presented the recommendations of the safety committee appointed to investigate procedures and rules for handling plutonium materials. The committee consists of Asprey, Hopkins, J. J. Katz, Katzin, and Westrum. A set of ten mandatory safety rules was given and discussed. These rules are as follows:

1. All plutonium handling and manipulations *must* be done in the hoods or approved transfer boxes. Plutonium should be outside the hood or box only when being carried in a safely covered container between hoods or hoods and transfer box.

2. All containers with plutonium must be so labelled in conspicuous fashion.

3. Plutonium must be stored only in approved locations. All transfers of possession of plutonium must be from person to person, not from person to location.

4. All hoods and working surfaces must be covered with stainless steel tray, glass plate, or other approved impervious material.

5. Rubber gloves must be worn at all times when handling plutonium except when forceps are to be used in the counting rooms.

6. Respirators, face shields, or assault masks must be worn whenever handling or transferring exposed plutonium or plutonium solution.

7. All centrifuges used for plutonium must be in hoods, cubicles, or protected in other approved fashion. No centrifuge tube shall be filled to more than within a radius distance of its top.

8. All glassware, equipment, or tools contaminated with plutonium shall be subject to the same rules as product. Equipment which has been in hoods or transfer boxes shall be considered contaminated until proved otherwise.

9. Fume hoods and dust hoods are not to be open more than 25% of capacity except when necessary for actual manipulations. (For dust hoods this means only two of the four hinged sections of the lowest level.)

10. Laboratory coats must be worn at all times in laboratory where plutonium is present.

Helen worked at home today on my classified "Table of Isotopes."

U.S. troops rolled on in France, and U.S. bombers raided Japanese bases over a 4000 mile front, according to today's paper.

Thursday, August 31, 1944

One of the three Associate Directors of the Chemistry Division (along with Selwood and Franck), Warren C. Johnson, transferred from the Met Lab payroll to the Clinton Laboratories payroll as a Principal Chemist. Johnson has been on the Met Lab payroll since his "temporary" assignment to Clinton Laboratories on January 26 of this year. He and his family returned to Oak Ridge from Chicago last Saturday by automobile.

I read a copy of a memo from Zachariasen to Allison about the 50-microgram sample of supposedly sodium neptunyl acetate submitted to Zachariasen for analysis by La Chapelle and Magnusson. A satisfactory x-ray diffraction pattern has been obtained and gives conclusive proof that the sample is sodium neptunyl acetate. The crystal structure is cubic; the density is calculated to be 2.556 gm/cm^3. The compound is isomorphous with corresponding compounds of uranium and plutonium. The dioxide of neptunium was identified some time ago. Thus, both +4 and +6 oxidation states have been established for neptunium.

D. F. Hewett, Staff Geologist for the Department of Interior Geological Survey, wrote me about two methods developed by the Geological Survey for determining small amounts of uranium and thorium in a wide variety of rocks. Hewett asked for my comments on the methods.

Flox reported to Nickson that the sputum sample brought in by Yett last week gave only 2 c/m and Hopkins' nose filters for the week ending August 19 gave 60 c/m. Beard's clothing was surveyed with a Pluto alpha ionization chamber. His trousers gave no reading; the waist and lap of his lab coat gave readings ranging from 10 to 30 c/m; his shoes had a slight amount of contamination and gave a reading of 15 c/m for both soles and the upper part of his left shoe.

Abstracts for the work of Sub-sections I and II and of the "23 group" (Group 9) were prepared and submitted to me by the group leaders (MUC-GTS-971). The abstracts contain a listing of problem area assignments, the individuals who are working on the problems together with the approximate time spent by each person on the problem, references to technical notebooks applicable to each problem area and a brief description of each problem area, and the current status of developments.

"Chemical Research — Separations Processes for Plutonium, Process Development and Chicago Semiworks Operation, July 15, 1944," (CN-1953), was issued. The following investigations are reported:

A preliminary study on the application of an ether extraction process for the recovery of uranium from the extraction waste solutions obtained in the Bismuth Phosphate Process is being made by Hyman. Blaedel and Margolis have studied extraction and decontamination by the ether method, evaluating the effect of an increase in the concentration of $Ca(NO_3)_2$ on the distribution of Pu(VI).

The Technical Division has carried out semiworks runs which demonstrate successful application of $NaNO_2$ as a prereduction agent in the extraction step. Decontamination in the Bismuth Phosphate Process

has been studied. Three ether extraction runs have been made to evaluate further the use of hydrazine as a reducing agent in the aqueous phase.

Helen worked at home on my classified "Table of Isotopes." After work, Thompson, Katzin, and I played nine holes of golf at Jackson Park. Our scores were ST-53, LK-60, and GS-46.

U.S. forces are ten miles past Reims; and the Soviets have captured the Ploesti oil fields and are 17 miles from Bucharest in Rumania according to the news today.

Friday, September 1, 1944

Joe Katz started his vacation today.

This is Clifford Smith's last day of service in my section. His former employer, the Owens-Illinois Pacific Coast Company in San Francisco, will not extend his leave of absence for another period; he is returning there this month.

Hogness issued a summary of manpower distribution (MUC-TRH-167) in the Chemistry Division as of September 1, 1944, which shows a total of 87 men for my section. The Hanford and Los Alamos distribution of effort is as follows:

Thompson (Hanford work)		*Number of Men*
Albaugh,	Extraction and decontamination	12
Pye,	Concentration and isolation	9
Gilbreath,	Process development	7
Lawroski,	Solvent extraction	4
Cunningham (Los Alamos work)		
Simpson,	High vacuum work	13
Hindman,	Basic chemistry	10
Dawson,	Recovery	10
Ghiorso,	Instruments & physical measurements	10
Katzin,	23 work	6

Helen worked at home on my "Table of Isotopes."

Roy Thompson, Fred Albaugh, Stan Thompson, and I played nine holes of golf at Jackson Park before dark. Scores were RT-67, FA-71, ST-51, GS-44. We travelled to Jackson Park by street car getting off on Stony Island Avenue at a point opposite the first tee. As usual when we stop after nine holes, we caught a streetcar on 67th Street to go home.

The top headlines today all contain news from the European front. The Soviets have entered Bucharest. French forces are only 60 miles from Spain, and U.S. troops have crossed the Meuse River in the Argonne Forest.

Saturday, September 2, 1944

La Chapelle and Magnusson, using the 19-microgram sample of sodium neptunyl acetate analyzed by Zachariasen, began efforts to make a direct determination of the specific activity and half-life of Np^{237}. In 1942, Wahl and I reported (in our paper on "Properties of 93^{237}," Report A-151 dated April 13, 1942) a value of 3×10^6 years for the half-life of Np^{237}, obtained by measuring the ratio of 7-day U^{237} activity to that of the daughter, Np^{237}. The plan now is to further purify the 19-microgram sample and determine the specific activity by direct weighing and radioactive assay.

I received a memo from Allison stating the "hurry-up" program for initial production at Hanford has been dropped; he was referring to the projected recovery of 2 kg of plutonium at the earliest possible moment and the processing of as much as four tons of uranium per day.

Paul Aebersold, an old friend from Berkeley, visited me. He is now with the Manhattan District Office in Oak Ridge. He told me that the following early contracts of mine have not been officially settled because of the lack of final reports: NDCrc-197, G. N. Lewis and G. T. Seaborg, 5-1-41 to 12-1-41; NDCrc-201, E. O. Lawrence and G. T. Seaborg, 4-1-41 to 10-1-41; OEMsr-206, G. T. Seaborg, 8-1-41 to 7-31-42. He asked that I prepare a brief final report of the results of each investigation and agreed to send me a confirming letter. I invited him to come to our apartment this evening and to see Helen.

Frances Chilson who had dinner with us in our apartment was here when Paul dropped in.

Banner headline in today's paper reads "Yanks Near German Border." British troops are making progress in Italy at a good rate also.

Sunday, September 3, 1944

Helen and I played 18 holes of golf with Stan Thompson and Zene Jasaitis at Wilmette Country Club (Lake Avenue at Harris Road, Wilmette, Illinois). Stan scored 105 while I scored 103. Zene drove us to Wilmette and back in his mother's car.

Monday, September 4, 1944

English at Clinton Labs wrote that three samples of bismuth phosphate are coming to us by truck. He wants Zachariasen to determine the relative percentages of alpha and beta forms of the bismuth phosphate.

The principal members of my staff and I received copies of a letter from Arnold to Allison saying that all concerned have been asked not to approach members of the Chemistry Division about positions elsewhere on the Project (such as Hanford), without first receiving the permission of Hogness. Similarly, members of the Chemistry Division should obtain Hogness' permission before undertaking discussion with representatives of related projects.

In a memo to Hogness, Watt requests a lanthanum fluoride precipitate containing one gram of plutonium from Room D at Clinton Labs. The material is needed to evaluate changes we are making in the peroxide isolation process in an effort to increase the tolerance for iron.

Allison sent a letter to Oppenheimer asking if the increase in purity that would result from the use of a solvent extraction process in

the last stages of Hanford extraction (from 99% to 99.7-99.9%) would be of appreciable benefit to the Site Y operation.

Joe Kennedy called me from Los Alamos. I told him I have sent the written-up patent cases to Lavender, but I have heard nothing from Underhill concerning a change in plans at Berkeley. I also told Kennedy that I will phone Underhill if I don't hear anything by Thursday.

The report, "Chemical Research — Separations Processes for Plutonium, Process Development and Chicago Semiworks Operation," dated August 15, 1944, was issued today. It contains an account of the following research: Blaedel and Lincoln have investigated the effect of slugbonding scale on the extraction and first decontamination cycle of the Bismuth Phosphate Process. It is found that the bonding scale from the dissolution of one-dip aluminum-silicon bonded Hanford slugs contains less than 1% of the total alpha- and beta-particle and gamma-ray activity when filtered from active UNH solutions. The scale does not appreciably affect either the decontamination or product recovery at Hanford plutonium and fission product element concentrations. The relative rates of solution of the bonding scale in HF and HNO_3 have been determined.

Larson and Lincoln have studied the effects that the possible Hanford process modification, which involves a third bismuth phosphate decontamination cycle, will have using material that has been extracted and run through two cycles by the Chicago semiworks. Six runs were made on material that, at the end of two cycles, gave an average decontamination factor of 1.4×10^4, resulting in an average factor of 1.0×10^6 after the third bismuth phosphate cycle. The Technical Division reports that it appears that excessive volumes of ether may be necessary to recover substantial quantities of UNH from semiworks waste solutions using $Al(NO_3)_3$ as a salting-out agent.

Helen saw her missionary friend Esther Bacon at the railroad station for a little while as she was passing through Chicago today. I had a migraine headache this evening.

War summaries in today's paper indicate progress on all fronts, both in Europe and the Pacific.

Tuesday, September 5, 1944

Stearns sent a memo to all Division Directors giving the procedures on communication with members of the Montreal Laboratory. Telephone conversations are monitored at the border and must be limited to non-technical matters. Telegrams between the two laboratories are prohibited. All letters to the Montreal Lab must be over the joint signature of the author and Stearns.

Helen took a walk with Wilma. In the evening Helen and I went with Stan Thompson and had malted milks.

The top headline in today's paper reads "Allies Capture Antwerp!" British troops have been moving quickly and captured that city yesterday.

Wednesday, September 6, 1944

Iz Perlman informed me that he is accepting a long-term assignment at Hanford (while remaining on the Clinton payroll) and that his family will be joining him as soon as he has a house at Richland, which should be next month. It appears that the major work of Greenewalt's Technical Division at HEW will be completed in the not-so-distant future so plans for Perlman's proposed transfer to the Division have been changed. Perlman will join a group headed by Hilberry (who is the Associate Project Director); his primary responsibility will be to act as an observer of all the work on the chemical extraction process in order to aid the overall planning and direction of this program.

Thomas O. Jones started work as my assistant at $4980 per year. He is on leave of absence from Haverford College where he is an associate professor in the Department of Chemistry. He received his Ph.D. in organic chemistry from the University of Wisconsin in 1937.

L. O. (Tom) Morgan, who works in Albaugh's group, today joined James and Ghiorso in our program of search for new elements. Morgan and his wife, Betty, live in an apartment at 6007 Woodlawn Avenue (see Figs. 18 and 19).

James completed six dichromate oxidation cycles on sample 49DD (large plutonium plus deuterons, St. Louis bombardment) in order to separate the plutonium from any element 95 formed in the bombardment. He started this processing on August 29. His final precipitate, mounted on a counting disc as sample 49DD-10, was given to Ghiorso and Krueger. A range measurement in counter N-1 showed the presence of perhaps three different ranges in terms of absorber thickness: (1) 4.5 mg mica, (2) 5.0 mg mica (maybe Pu^{239}), (3) 5.4 mg mica. The sample was turned over for further processing to Tom Morgan.

James next tried the "HNO_3 approach" on the plutonium fraction. He combined the supernatants (49DD-1 and 49DD-3) from the first two dichromate oxidation cycles that contained most of the original plutonium dissolved off the target. The solution was reduced with SO_2, and the plutonium (and presumably any element 95 present) was precipitated as PuF_4. After dissolving the precipitate in nitric acid, he tried an electrolytic oxidation without success. He then precipitated plutonium hydroxide, dissolved it in nitric acid, and heated it overnight at 95°C.

I attended the evening meeting of the Separations Process Subsection of Section C-I at 7:45 p.m. in Room 209, Eckhart Hall. Hagemann told us about the extraction of U^{233} from the third and fourth cans of thorium carbonate irradiated by neutrons for several months in the Clinton pile. About 8.5 mg of U^{233} were obtained. Specific activity determinations carried out by Hufford by counting a weighed sample of oxide gave 23,000

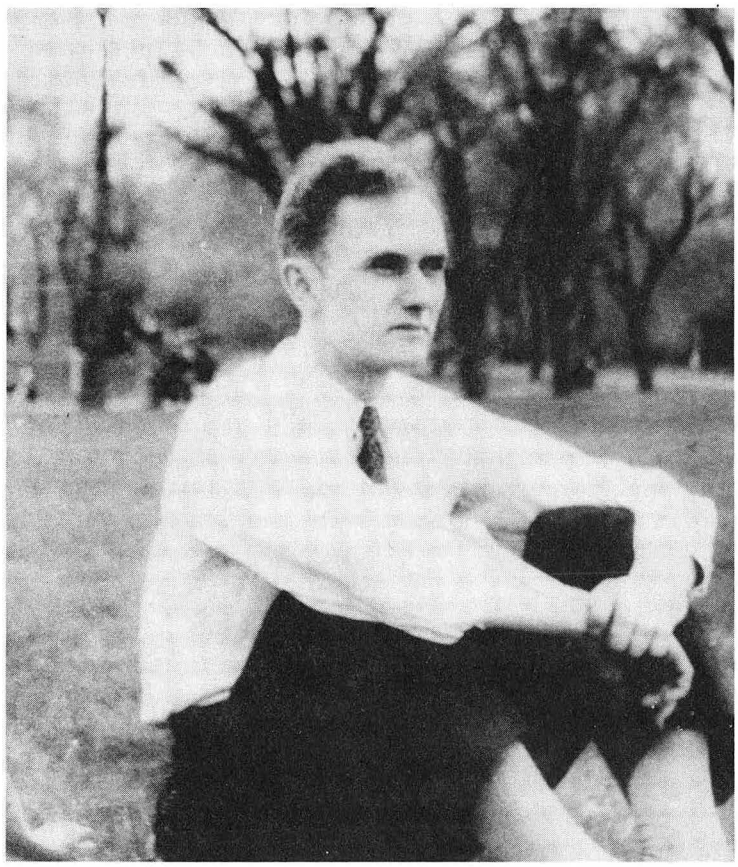

Figure 18. L. O. (Tom) Morgan, 1944.
XBB 792-2374

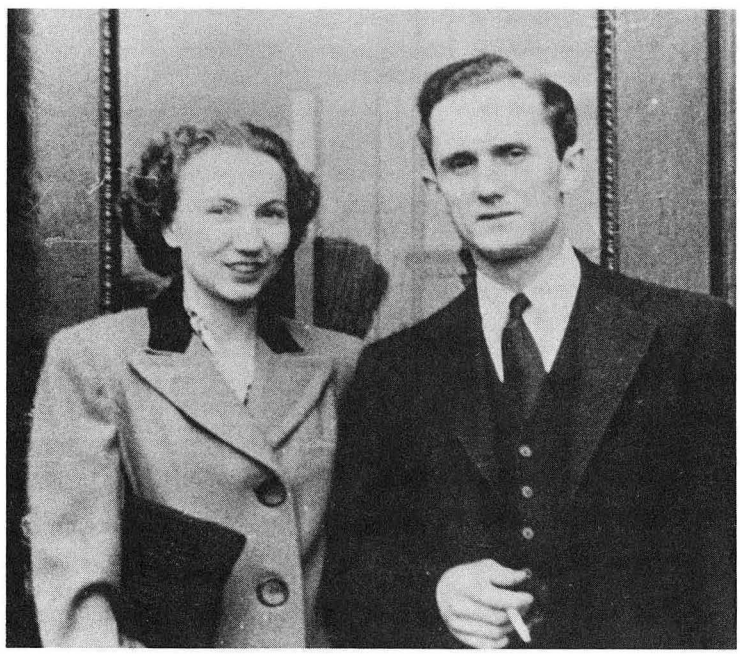

Figure 19. Betty and Tom Morgan in front of their apartment building at 6007 Woodlawn Avenue, 1944.
XBB 769-8633

d/min/microgram as the best value for pure U^{233}. I mentioned that the Montreal group has been obtaining much lower values than this — perhaps impurities still present in their U^{233} account for these low values.

Hellman gave an account of the extraction of uranium and thorium into ether from solutions of varying NH_4NO_3, $Th(NO_3)_4$, and HNO_3 concentrations. Hyde reviewed the extraction of uranium and thorium by hexone; he has found that thorium decay products are extracted by hexone to a greater extent than by ether. Reinhardt spoke on the effect of lanthanum, calcium, and ammonium nitrates as salting agents for extraction of plutonium into ether and hexone. He has found that their effectiveness in both solvents is in the order named. Van Winkle talked about the work being carried out on isolation of natural Pa^{231} and Io^{230} from residues obtained from commercial uranium production. He gave a summary of the existing knowledge of protactinium and ionium chemistry. Separation and isolation of protactinium and ionium by means of UF_4, which carries ionium but not protactinium, will be the next step in the investigations.

Helen had lunch with Wilma.

Russia has declared war on Bulgaria, according to today's paper. The Soviet Union has repeatedly warned that small nation that her belated attempt at strict neutrality is insufficient at this late date.

Thursday, September 7, 1944

Continuing the processing of the main plutonium fractions (samples 49DD-1 and 49DD-3), James precipitated the plutonium as the fluoride from the nitric acid solution that had been heated overnight at 95°C. The precipitate was metathesized with NH_4OH, dried under vacuum, and turned over to Morgan for further processing as sample 49DD-18. James then left on vacation for two weeks.

The Health Division completed the weekly radiation survey in rooms occupied by members of Section C-I. Thirty-six rooms were surveyed for alpha contamination, and the following were found to have off-scale readings with "Pluto": Room 2 (hood used by Davidson); Room 4 (Florin); Room 10 (Hindman, Kraus); Room 11 (Ames, Dixon, McLane, O'Connor, Howland); Room 27 (Beard, Goeckermann, Hopkins); Room 30 (Bartell, Hoekstra, Malm); Room 34 (Britain, Fineman, Haeckl, Leventhal); Room 35 (Anderson, Asprey, Stewart); and Room 36 (Kelley, Yett). The beta-gamma surveys showed that the following rooms are contaminated: Rooms 12 and 13 (James, La Chapelle, Magnusson); Room 29 (Bradt, Ader); Room 31 (Greenlee, S. Peterson).

I received a letter from Frost at Northwestern University informing me that lecture number 5, my talk on "Isotopic Tracer Techniques," is now scheduled for Tuesday evening, October 24. The lecture series, entitled "New Tools in Chemical Research," is to be given in the Law School Building, Chicago campus, 357 E. Chicago Avenue, at a cost of $15 for the series of twelve lectures. Lectures are from 7:00 to 9:00 p.m.

I played in the Met Lab Golf Tournament with "BP" at Jackson Park and won, 10 and 8 (I shot 53 for ten holes, "BP" shot 78).

Helen worked at home on my classified "Table of Isotopes."

U.S. troops are fighting German troops in the Moselle Valley, and Russian troops have driven to Yugoslavia, according to today's paper.

Friday, September 8, 1944

Hyman started his vacation. He will return on September 19.

Morgan took over on sample 49DD-10 prepared by James (this was the final sample from six dichromate cycles on 49DD — the 200 mg of plutonium plus deuterons, St. Louis bombardment). Morgan carried through another dichromate oxidation cycle. The precipitate was mounted (49DD-4), and an extensive series of range measurements began today.

Sheft completed an attempt to prepare PuI_3 by a high temperature method according to the reaction: $PuO_2 + 2C + 3HI \rightarrow PuI_3 + 2CO + 3/2\ H_2$. He has heated to 1500°C a mixture of plutonium oxalate (4 mg of plutonium) and lampblack in a platinum microcrucible in an atmosphere of hydrogen iodide. A small amount of a light-green sublimate collected above the crucible, but there was not enough for an x-ray analysis.

Aebersold wrote from Oak Ridge to explain the kind of final report needed in order to settle officially the three early contracts of mine he discussed with me here last Saturday.

We have been so successful in the development of our solvent extraction process for use as an alternate method at Hanford in the concentration-isolation steps that we have decided to try to develop an alternate method (to the Bismuth Phosphate Process) for use in extraction and decontamination. Blaedel has been leading an effort on this during the last three weeks. Today he sent Burton a memo giving a schematic outline of such a continuous solvent extraction-decontamination process. He indicates the possible solvents we might use (hexone, ethyl-butyl cellosolve, diethyl cellosolve, nitroethane, and dibutyl carbitol) and asks Burton to speculate on the effects of radiation on these solvents to help us eliminate some of them from consideration.

I called Underhill, reaching him in Davis, to discuss what the University of California is doing with respect to the patent matter as a result of Lawrence's intervention. Underhill reported (1) Sproul has wired Bush to see why Lavender is reluctant to have him, Underhill, participate in our forthcoming September 15 meeting in Washington; (2) Lawrence is advocating that we give the patent applications to the University and that the University give them to the Government; (3) he has not seen Sproul to learn the University's position and will phone me as soon as he does so.

Helen and I had dinner at Morton's Tavern on Lake Park Avenue with Stan Thompson. During our dinner conversation Stan and I agreed that Fred Albaugh and Edrey Smith would make a fine couple if we could somehow encourage Fred to date her.

The Allies have invaded Yugoslavia from the Adriatic Sea. In the Pacific carrier planes have attacked the Palau Islands, on the path to the Philippines.

Saturday, September 9, 1944

I received a copy of a September 8 memo from Dempster to Stearns giving the results of a mass spectrographic assay to determine the ratio of the isotope 233 to the isotope 238 in a sample of the 7.2 mg of uranium isolated from the thorium carbonate irradiated by neutrons for several months in the Clinton pile. His data indicate the percentage of U^{233} to be 85 ±1.5% of the total uranium.

I also received a copy of the September 8 memo Arnold wrote to Bartky about our desire for the entire slurry from one or two lanthanum fluoride plant runs at Clinton for use in further work on the peroxide isolation process and the alternate hexone extraction isolation process. The material wanted is the lanthanum fluoride-plutonium precipitate after the first lanthanum fluoride by-product precipitate. Arnold asks that shipment be made on the truck leaving Clinton next Thursday.

Gilbreath recommended to Egan that several semiworks runs employing tracer quantities of plutonium be performed in order to determine the feasibility of reprecipitating lanthanum fluoride as a means of eliminating iron in the separations process isolation step where it interferes with the plutonium peroxide precipitation. It has been found recently that lanthanum fluoride carries less than 1% of the iron from a solution of 1.0 M in Fe(III). Laboratory experiments have indicated that if this lanthanum fluoride is dissolved in 10 N HNO_3 and reprecipitated by the addition of HF, the iron content is again reduced by a factor of 100, thus bringing it below the tolerance limit.

Hindman sent H. L. Anderson the results of an analysis for D^+ and NO_3^- in the solution of plutonium in DNO_3 and D_2O that we prepared for Anderson's use in the measurement of neutron properties of Pu^{239}.

I called Kennedy at Los Alamos and told him of yesterday's conversation with Underhill. I told him that Underhill would call me back after he has talked to Sproul and learned the University of California's position. I promised to call Kennedy again after I hear from Underhill.

The Health Division completed the weekly radiation surveys of the West Stands and find unsatisfactorily high beta-gamma readings in Rooms 217, 216 (Blaedel and Walling) and 212 (Larson, Hyman, Lincoln, and Winner).

Today's banner headline reads "Liege Captured by Yanks."

Sunday, September 10, 1944

Lawroski, Arnold, Thompson, and I played golf at Cog Hill No. 2 (SL and LA won "low ball plus low total" match 6 and 4. Our individual scores were SL-97, LA-94, ST-103, GS-99). This served as my match with Stan in the Met Lab Golf Tournament, which I won 3 and 2. As we do when Luther Arnold plays with us, we rode in his car. We stopped on the way home at Ashburn Flying Field on Cicero Avenue where Luther takes flying lessons; this earns gasoline ration points for him.

The Vance Coopers, on their way from Oak Ridge to Hanford, had lunch with Helen in our apartment. The Coopers, Fred Albaugh, and Stan Thompson (who is still staying in our apartment with us) had dinner with us at home. Fred told us that he became engaged to Edrey Smith last night. Stan and I were amazed and delighted at how soon our plans for Fred and Edrey had reached fruition.

During the evening I received a call from Underhill on the patent matter. He told me that Lawrence has won his battle with Sproul and the Regents. Sproul has consulted a list of "big shots," including Regent Moffitt and Governor Warren; apparently they agree with Lawrence. The University will enter into no sale, but requests that we give the patent rights to them so they may give the rights to the U.S. I indicated to Underhill that I think this is unfair to the inventors but am ready to go along with it. I called Kennedy afterwards, reaching him at the movie theater in Los Alamos. I told him what I learned from Sproul through Underhill and said that I feel I must go along with their wishes. Kennedy indicated that he is inclined to hold out for a cash sale.

Monday, September 11, 1944

This is the last day at the Met Lab for Louis O. Gilpatrick who is going to the Tennessee Eastman Corporation at Site X.

I had to speak to Eda Kelley, Ed Orlemann's secretary, about the fact that she refuses to wear her identification badge. H. W. Hoyt, Director of the Security Division, wrote a memo to me saying that she consistently refuses to wear the badge and that she has told the guards she only wears it "once in a while." When I spoke to her, she assured me she will obey all rules with respect to the wearing of her badge and the security regulations in general.

Watt wrote to Squires at Hanford in accordance with Squires' request to be kept informed of the progress of our work on the peroxide isolation process. Watt explains that the Concentration-Isolation Group has been working almost exclusively on the problem of devising (a) a satisfactory method for removal of iron prior to metathesis, and (b)

process modifications which will permit isolation of plutonium from process solutions that contain relatively high concentrations of iron. He describes the recently developed procedure to eliminate iron by reprecipitation of the lanthanum fluoride-plutonium precipitate at the end of the "crossover," and mentions the procedure that is now being evaluated in the Chicago semiworks from the standpoint of operational feasibility. Watt also describes proposed modifications to the peroxide isolation process that improve the filtering characteristics of the first and second plutonium peroxide precipitations.

Albaugh sent a memo to Maloney, commenting on Maloney's report "Status of Separations Processes." He makes numerous suggestions of ways to improve the description of the present status on all the steps in the Bismuth Phosphate Process.

Kennedy called me from Los Alamos to report that he has discussed with Segrè and Wahl the University of California's request that we give the patent rights to the University and, thence, to the Government. Segrè feels he should get something from the University (professorship in physics at Berkeley or Los Angeles); otherwise he will join Kennedy and Wahl in considering a hold-out for case sale. Kennedy asked me to cancel my plans to meet him in Washington, D.C. this Friday.

Helen worked at home on my classified "Table of Isotopes.

Reports today say that U.S. troops are firing across the German border.

Tuesday, September 12, 1944

September 21 will be the termination date for Gerald W. Sears; however, today is his last day of work here. Sears will work for the Tennessee Eastman Corporation at Site X. Donald Wetlaufer, a technician in Dawson's group, also resigned. He intends to return to school.

I received a memo from Blaedel and Walling proposing a solvent extraction method for the extraction, decontamination, and partial concentration of plutonium from dissolver solution using procedures which make use of Pu(III) and a higher oxidation state of plutonium (MUC-GTS-996). They outline the proposed method as follows:

"Dissolver solution, which has been brought up to a high concentration in NH_4NO_3, enters column A and flows countercurrent to a rising non-aqueous solvent stream. The amount of solvent used will be sufficient to extract all the $UO_2(NO_3)_2$ and the Pu(IV,VI). This solvent solution is then scrubbed with a volume of aqueous solution in the top part of the column such that all fission product elements (FPE[G]) having distribution ratios (water/solvent) greater than those of Pu(IV,VI) and $UO_2(NO_3)_2$ are stripped out of the solvent solution. (Those fission product elements (FPE[L]) having distribution ratios less than those of Pu(IV,VI) and $UO_2(NO_3)_2$ cannot be removed without losing plutonium and $UO_2(NO_3)_2$ to

the aqueous waste solution.) The solvent solution then enters column B and rises countercurrent to a falling aqueous stream containing NH_4NO_3 and a reducing agent. The reducing agent reduces plutonium to the III state, which has a very high distribution ratio (water/solvent), but does not appreciably affect $UO_2(NO_3)_2$. By a proper choice of volumes and conditions, it should be possible to retain $UO_2(NO_3)_2$ and the remaining fission product elements in the solvent stream and to extract the plutonium into the aqueous stream."

Blaedel and Walling have conducted fifty batch extractions to determine the feasibility of the various operations and have concluded the system is chemically feasible using hexone as the solvent and $NH_2OH \cdot H_2SO_4$ or $N_2H_4 \cdot H_2SO_4$ as reducing agents.

Albaugh sent a 14-page memo to Thompson giving the status of separations process research, covering:

1. *Extraction-Decontamination.* Plutonium reprecipitation runs, studies on preextraction process solutions, spectrophotometric studies, carrying studies on preextraction process solutions, storage prereduction studies.

2. *Concentration-Isolation.* Lanthanum fluoride reprecipitation studies (using 10 N HNO_3 to dissolve the lanthanum fluoride precipitate), isolation studies (to develop conditions for obtaining a filterable precipitate at 25°C).

3. *Process Development.* One-liter scale runs to test the low acidity decontamination procedure — low decontamination factors have been obtained.

Helen worked at home on the classified "Table of Isotopes."

Two-inch headlines today read "U.S. Invades Germany!"

Wednesday, September 13, 1944

Today the first uranium slugs are being loaded into the first production reactor at Hanford — the 100B pile. Fermi is there to oversee and direct the operation.

Fred Meyer's termination is effective today. He is going to the University of Kansas City in Missouri.

Underhill in Berkeley and I talked by telephone about the patent matter. He informed me the University of California patent committee consists of Charles Lipman, Bill Young (UCLA), Walter Porter, E. O. Lawrence, etc., who are not unanimously in favor of the Lawrence plan. The Regents will take formal action on our patent matter at a meeting on September 26. Following our conversation, I called Kennedy at Los Alamos to give him this new information. I also mentioned that I am mailing him Conard's document on how to assign the patent rights to the University and two documents on tax matters.

I attended an evening meeting of the Basic Chemistry, Recovery, and Instrument Groups of my section at 7:45 p.m. in Room 209, Eckhart Hall. Others present were Abraham, Ames, Ashcraft, Cunningham, Davidson, Dawson, Dixon, Fields, Florin, Fried, Gilbreath, Hindman, Howland, Hufford, Jaffey, Jones, Kohman, Krueger, La Chapelle, Magnusson, Manning, McLane, Orlemann, S. Peterson, Phipps, Reinhardt, Sheft, S. Thompson, Walsh, Warner, Westrum, and others. Sheft reported on the preparation of two compounds (which he believes are PuOBr and $PuBr_3$) in the presence of water vapor, using PuO_2 as the starting material. The apparatus involves a quartz reaction vessel containing the PuO_2 that is heated to 750°C, over which is passed a mixture of HBr and H_2 gases.

Magnusson reported on the chemistry of Np^{237} and the preparation of pure sodium neptunyl acetate. The preparation was submitted to Zachariasen for x-ray analysis and found to be isomorphous with the corresponding plutonium and uranium salts. The lattice constants *a* and density are given below:

	a (Å)	*density* (gm/cm^3)
$Na(UO_2)Ac_3$	10.671	2.552
$Na(NpO_2)Ac_3$	10.659	2.556
$Na(PuO_2)Ac_3$	10.643	2.578

The sodium neptunyl acetate was further purified and used to prepare NpO_2 samples for determining the specific activity and the half-life. Preliminary values have been obtained and give 1500 disintegrations per minute per microgram, corresponding to a half-life of 2.25×10^6 years. The values may be in error due to the absorption of water vapor during weighing, and further experiments are in progress. I suggested the specific activity results might also be high because of the contamination with plutonium; this could be checked by range measurements of the alpha particles. Cunningham and Jaffey suggested that the geometry factor might be higher for neptunium than for plutonium due to the greater back-scattering of the shorter-range alpha particles.

Jaffey gave rules pertaining to the use of the counters and reported on the recent work of the instruments group. He pointed to the use of the differential-range chamber for making very accurate measurements of unknown ranges by reference to a standard such as polonium. Jaffey also described techniques for making range measurements using varying absorbing thicknesses of, e.g., aluminum, between the sample and counting chamber.

At 11:30 p.m. I left Chicago (Englewood Station) with S. K. Allison and Stan Thompson on the Penn R.R. to Cincinnati, where we will continue on the L&N R.R. to Site X. The purpose of the trip is to attend conferences at Clinton Laboratories and Y-12 where I will meet Orlemann. I have been asked to make an analysis of the processes used for the chemical recovery of the enriched U^{235} produced by the electromagnetic process because they are experiencing large losses of U^{235}.

Battleships have joined in the bombardment of Palau in the south Pacific.

Thursday, September 14, 1944

Allison, Stan Thompson, and I arrived in Oak Ridge this afternoon. I am staying at the Guest House.

Two hundred Japanese planes were destroyed in a raid on the central Philippines.

Friday, September 15, 1944

At Site X. I again spent the night at the Oak Ridge Guest House.

Saturday, September 16, 1944

In Oak Ridge. Orlemann and I met with some of the people stationed in the Y-12 area where Lawrence has his calutrons set up. These were H. J. Emeleus, a British chemist; Clarence Larson; and F. A. Jenkins from Berkeley. We discussed chemical procedures and organizational arrangements needed to carry out the project to recover chemically the U^{235} enriched by the electromagnetic process. I indicated that the complexities of their job are comparable with those we face on the Metallurgical Project and that, on this basis, it seemed to me that they have less than a sufficient number of chemists for it. I suggested three main places in which increases in the quantity and quality of chemical personnel are needed:

1. The staff that handles the control analyses should be greatly expanded — perhaps, by a factor of five. I also recommended that analytical development work be done in the Analytical Division rather than in the Chemical Development Division in order that there be close association between the development men and those handling the routine control work.

2. A process development group should be formed to test the yield of the process, step by step, all the way through. This group might consist of at least 20 men, including a substantial portion of competent Ph.D.'s.

3. The operators of the chemical production process should include more graduate chemical engineers and industrial chemists. I suggested that the running of the Kraus reactors, the dissolving of the product from the M, L, and E containers, etc., should be supervised by trained chemists, at least until running smoothly.

I also discussed the relative merits, from their standpoint, of the ether process and the uranous oxalate process. I indicated that the ether process has the advantage of having a start toward being in production whereas the uranous oxalate process has the advantage of being inherently simpler. I pointed out they may be underestimating the problem of working in a HCl system, citing our own experience of having the chemical engineers reject an HCl-based process that was simpler than the one put into operation.

Other items discussed were the best way to dissolve uranium carbide; test for uranium at 1 mg/gallon; and the RSW&P (refining sample wash and purification) operation, directed by George De Croes up to the hydrofluorination step, where H. A. Young, on leave from the University of California at Davis, takes over.

I spent the night at the Andrew Johnson Hotel in Knoxville.

Sunday, September 17, 1944

In Knoxville. In the afternoon, Stan Thompson and I boarded the L&N train to Cincinnati where we will continue on the Penn R.R. to Chicago.

Monday, September 18, 1944

Stan Thompson and I arrived in Chicago at 7:30 a.m.

I had breakfast with Helen. She told me that on Thursday she went downtown, met Frances Chilson for lunch, shopped in the afternoon, and met Wilma for dinner. She and Wilma then went to the Harris Theater to see Ruth Gordon in the play, "Over Twenty-One." On Friday she had lunch with Wilma. Then on Saturday, Frances Chilson came over to our apartment and spent the night. The next morning they had breakfast together.

* * * * *

Last Thursday Egan received a memo from Nickson giving the results of the beta-gamma contamination survey of the semiworks for the week ending September 9. Unsatisfactory conditions were found in Rooms 8, 9, and 10.

The same day my office received a memo to me from Zachariasen giving the following results of x-ray examination of three bismuth phosphate samples, E, D, and O, from Site X. Extraction precipitation E: more than 95% of this sample is alpha-phase bismuth phosphate; less than 5%, if any, is beta phase. Decontamination precipitation D: 95% alpha-phase; 5% beta phase. By-product precipitation O: more than 95% alpha-phase; less than 5%, if any, beta-phase.

Also on Thursday, Allison sent a memo to Mulliken of the chronology of the Project. Thirty-six highlights are identified from January 1939 (chemical evidence for fission by Hahn and Straussman announced by Bohr) to September 12, 1944 (loading of metal into first production pile at Hanford). Other highlights include:

March 28, 1941. California group demonstrates that 49 is fissionable at thermal energies.

February-March, 1944. Milligram and gram amounts of 49 from Site X are available for experimentation.

July, 1944. Spontaneous fission of plutonium-240 discovered at Site Y, and purification program abandoned for plutonium. Ceiling placed on Met Lab personnel.

Friday Fineman started his vacation. He will return on September 28 (see Fig. 20).

La Chapelle and Magnusson completed on Friday their determination of the specific activity of Np^{237} by direct weighing and radioactive assay. They started this work on September 2, when they began to purify further the 19-microgram preparation that Zachariasen had identified as sodium neptunyl acetate. The purification was aimed at removal of lanthanum or platinum contamination and consisted of successive hydroxide and sodium neptunyl acetate precipitations, followed by two more hydroxide precipitations. The final precipitate was dissolved, and two portions of the solution containing about 3.8 micrograms of neptunium each were placed on platinum plates and ignited to constant weight. The samples were weighed with a Kirk-Craig microbalance, and the plates were counted in the 50% geometry alpha-particle counter. The data correspond to a specific activity of 1,517 disintegrations per minute per microgram and a half-life of 2.21×10^6 years. The value is considered accurate to within 5%.

Friday Westrum prepared nine additional plutonium-loaded electrodes and submitted them for delivery to Hamilton in Berkeley. A total of 3.12 milligrams of metal was used in the loading.

The weekly radiation survey of rooms occupied by my section was completed. Unsatisfactory conditions exist in the following rooms: New Chemistry Room 2 (Abraham, Sheft, Pellock), 4 (Florin), 6 (Wolf, Hagemann, Hellman, Studier, Hyde), 9 (Billington), 10 (Hindman, Kraus), 11 (Ames, Dixon, Howland, McLane, O'Connor), 27 (Beard, Goeckermann, Hopkins), 29 (Bradt, Ader), 30 (Hoekstra, Malm, Bartell), 33 (R. Thompson, Pye), 34 (Britain, Fineman, Haeckl, Leventhal), 35 (Stewart, Asprey, Anderson), 36 (Kelley, Yett), 41 (Fried, Westrum, Robinson), West Stands (WS) Rooms 216 and 217 (Blaedel, Walling), 212 (Larson, Hyman, Lincoln, Winner), 26, 23.

Last Saturday Joe Katz returned from his vacation which began September 1 while Bartell started his. Bartell will return to work Monday, September 25.

Watt wrote me on Saturday concerning Squires' request that an effort be made to develop a batch extraction process involving hexone for the concentration-isolation steps of the Hanford process. Squires has also suggested that the batch process should be compatible with the equipment that is to be provided for the process cells in the 231 Building. Watt points out that because of the uncertainty as to what is desired,

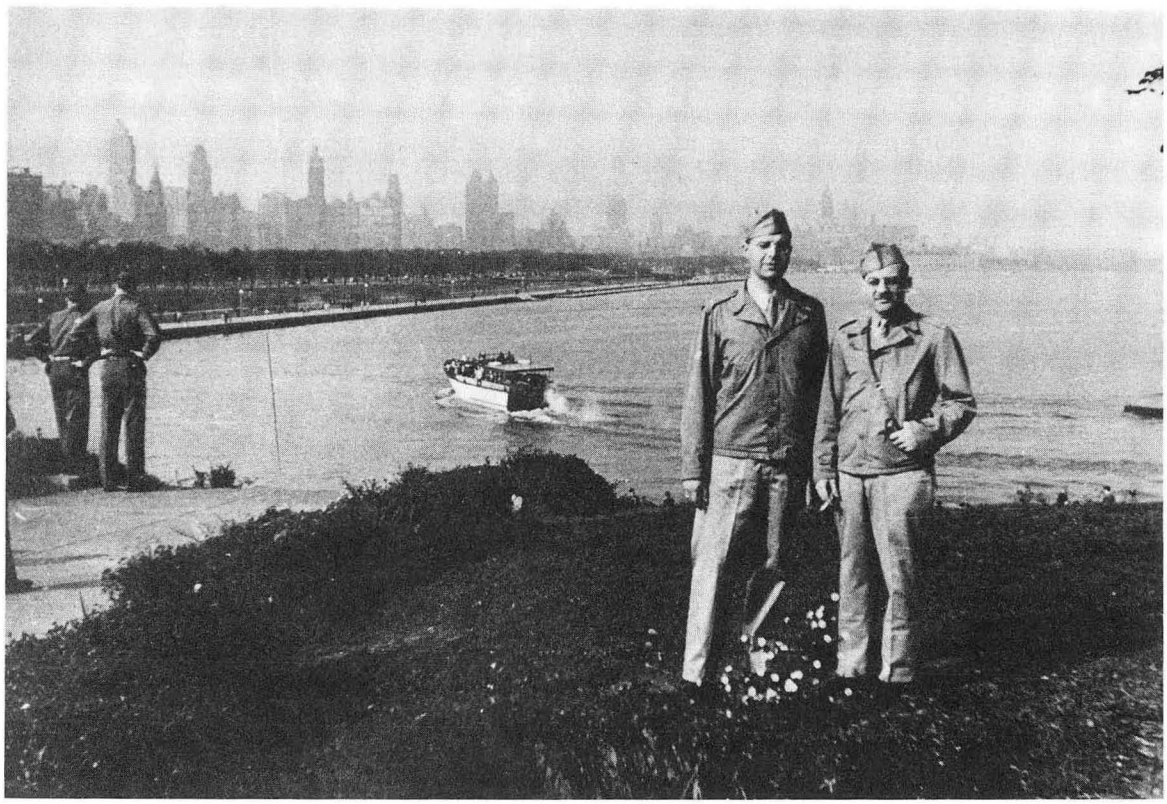

Figure 20. Phillip Fineman and B. R. Wendrow near Grant Park, Summer 1944. Our SED men, who have been in civilian clothes since their arrival at the Met Lab, returned to army uniforms in July.

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the work on batch extraction at Chicago has employed zirconyl-complexed lanthanum fluoride rather than the nitrate solutions to be delivered to the 231 Building; the results have not been encouraging. Watt suggests that considerable emphasis be placed upon batch extraction studies and that, in view of the extensive facilities being provided in the 231 Building, consideration be given to operating the two processes in parallel in order to make an early selection of the best available method.

Report CS-2135, "Chemistry Division Summary Report for August 1944," was issued last Saturday. The Section C-I portion of the report covers separation process studies, basic chemistry, and U^{233} work. In addition to the description of work done in my Section C-I (which I have covered already) it contains the following summary of investigations.

Burton reports on the radiation effects work of Section C-II, which is primarily related to effects on Hanford graphite and waters; and the radiation-enhanced corrosion of tuballoy. In addition Burton reports on preliminary experiments recently completed concerning the effects of high alpha-particle irradiations of concentrated hexone. Exposures equivalent to 200 days of Hanford operation were given to hexone containing feed solution by adding a polonium solution. With up to 125 days of equivalent Hanford operations the irradiated hexone solution behaved approximately as an unirradiated control solution, but thereafter the viscosity increased more rapidly and the solution took on a deep yellow color. The control remained colorless. A control and polonium-containing sample kept at 5°C for about the same time, however, showed no relative change in color. A tentative conclusion is that the G value (100 electron-volt yield) is considerably greater than 1.0 at the higher temperature under the oxidizing conditions studied.

Sugarman reports on Section C-III work. Several new fission products and their yields, including isotopes of europium, germanium, arsenic, columbium, zirconium, lanthanum-praseodymium, krypton, xenon, strontium-yttrium, are reported. The collection of information pertinent to the Bismuth Phosphate Process presented by Sugarman includes work on the preparation of more than 60 samples of phosphate crystals for x-ray analysis. A summary of studies conducted by Section C-III on long-lived beta- and gamma-activities is also presented.

McKinney reports on Section C-IV work involving analytical services. Over 560 samples were received by the section for analysis during August. Arnold gives the status of Pure and Rare Chemicals work for Section C-VI with emphasis on the procurement of rare earth salts, rubidium nitrate, thorium nitride, and rhenium metal.

Kurt Kraus left Chicago Saturday afternoon via NYC R.R. on a trip to Clinton Labs. He will return sometime next week.

Alec Kelley started his vacation today. He will return September 25.

Mary Sue Lytle was hired as a technician for Katzin.

I received a memo from Davidson giving a summary of existing data on the thermodynamic properties of oxides and fluorides of uranium and

plutonium in order to consider more precisely the question whether it is possible to oxidize or convert PuF_3 or PuF_4 with oxygen to PuO_2, the replacement of fluorine by oxygen being a rather unusual reaction. He also records some estimates of the probable values of the heats and free energies of several reactions involving the plutonium fluorides. Davidson concludes that his calculations throw sufficient doubt on the experimental results on the oxidation or conversion of UF_4, PuF_3, and PuF_4 to make it necessary to reexamine these questions with improved experiments.

Westrum informed Hamilton by letter that the additional nine electrodes were loaded with plutonium and submitted for delivery on Friday. A total of 3.12 mg of plutonium was used in the loading, prepared in the same manner as were the original electrodes on August 18 — metal in the form of small spheres located at a distance of between 0.080 inch and 0.100 inch from the end of the electrode which were individually sealed in evacuated glass tubes.

Allied airborne troops landed in Holland at the rear of Nazi lines, according to today's paper.

Tuesday, September 19, 1944

Hyman returned from his vacation that began September 8.

At 9:30 a.m. I attended a Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Allison, H. L. Anderson, Arnold, Bartky, Burton, Chipman, Connick, C. M. Cooper, Daniels, Davies, Dempster, Doan, Eastman, Franck, Graves, Hogness, Huffman, Jacobson, Jeffries, W. C. Johnson, Leverett, Levy, Manning, McKinney, Mulliken, Nickson, Perlman, Rabinowitch, Spedding, Stearns, Sugarman, Sutton, Szilard, Turkevich, Vernon, Watters, Warner, Wigner, Zachariasen, and Zinn. Manning presented the report for Section C-I. In reference to work on the mainline separations process, he described the investigation of the influence of hydrazine on the extraction step and the unsatisfactory results of the last two low-acidity bismuth phosphate decontamination runs. He mentioned it would be useful to be able to obtain Np^{237} with a better yield than before and went on to suggest that if U(IV) were used as the reductant instead of Fe(II) in the decontamination process, this could lead to a better neptunium carry-through with the plutonium — perhaps only a 20% loss of neptunium instead of the usual 70-80%. Reprecipitation of lanthanum fluoride as a means of eliminating iron in the concentration-isolation step was stated to look promising.

On the subject of solvent extraction for concentration-isolation, Manning mentioned the good results being obtained with the small hexone column originally designed for purification which has now been converted for use with zirconium-complexed lanthanum fluoride precipitates. He mentioned that hexone has been examined also for use in extraction and decontamination, utilizing the III state of plutonium for separation of

plutonium from uranium — again results are promising.

On the subject of basic chemistry, he discussed the formation of negative plutonium ions in HCl, the vapor pressure studies of PuF_3 and $PuCl_3$, and the new half-life determination of Np^{237}. He summarized recent work on U^{233} extracted from irradiated thorium carbonate, giving the latest value for the half-life of U^{233} as 1.38×10^5 years, in good agreement with the earlier value by Seaborg and co-workers of 1.4×10^5 years (corrected for counter geometry factor of 0.52 rather than 0.45). He mentioned that the divergent results in Montreal concerning the half-life were cleared up when a uranium sample received from Montreal was found to be strongly contaminated.

Burton reported work of Section C-II on the problem of sudden release of energy accumulated in the pile of graphite by displacement of carbon atoms in the crystal lattice. The possibility of annealing was mentioned. Sugarman reported on fission product work in Section C-III and indicated that the distribution curve of fission products for the heavy group from neutron-irradiated plutonium appears to be identical to that from U^{235} except for distinctly higher yields at the extreme heavy-weight end of the curve.

Sutton presented the last report to the Council from the Clinton Separations Development Division. He said that all steps in the separation process at Hanford are in a satisfactory state of development and are believed to be fully adequate for satisfactory operation. Other Clinton Laboratory speakers were Levy, Perlman, and Davies. Eastman and Connick reported for Berkeley, and Spedding reported for Ames.

"Chemical Research — Separation Processes for Plutonium, Report for Month Ending September 1, 1944," (CN-2085), was issued today. It summarizes the following information:

Extraction-Decontamination (Albaugh, Group Leader). S. Peterson, Howland, and Malm have made further tests of methods of increasing the metal processing rate at Hanford during early operation when plutonium content of the irradiated slugs will be low. They find that high plutonium losses can be avoided when extracting from 30% UNH solutions if the total acidity does not exceed 1.1 N. They also find that the plutonium precipitation in the first bismuth phosphate cycle can be carried out successfully from 2 N rather than 1 N HNO_3 if the Bi(III) used is increased by a factor of 1.5.

Morgan has made four process runs to test the effect of phosphate and oxalate on decontamination in the plutonium precipitation step; it has been noted that these ions are somewhat effective in preventing coagulation and precipitation of zirconium and columbium phosphates. He finds that variation of H_3PO_4 concentration affects decontamination only slightly. Inclusion of 0.15 M $H_2C_2O_4$ in the plutonium precipitation step improves gamma decontamination by a factor of 8. Morgan, Hoekstra, Ader, Malm, Larson, and Lincoln have worked on prereduction studies. Although the present nitrite prereduction procedure appears satisfactory, there is uncertainty as to the mechanism of the beneficial effect. Accordingly, a rather comprehensive study has been planned, and data are being accumulated. A spectrophotometric study of the effect of various

reducing agents in 22.5% UNH solution has been started; the possibility of effecting prereduction by introduction of a suitable reducing agent prior to the 10-day storage of the 40% UNH solution is being investigated; the inhibitory effect of hydrazine on plutonium carrying in the extraction step has been confirmed.

Concentration-Isolation (Pye, Group Leader). Haeckl, Kelley, and Yett have studied the co-precipitation of iron with lanthanum fluoride in order to determine how much iron may be expected to be carried into the isolation step where it is known to affect plutonium peroxide precipitation. Kelley has investigated methods for eliminating iron from metathesis and isolation process solutions. Sorbitol was found capable of complexing iron in the presence of plutonium. J. Katz and Walling have examined the possibility of eliminating iron during the lanthanum fluoride-plutonium precipitation. Complexing agents have been found ineffective, but solution and reprecipitation of lanthanum fluoride using 10 N HNO_3 shows promise and is being further investigated.

Goeckermann and Yett have studied the HEW flowsheet peroxide isolation procedure using Clinton plant material from runs that do not contain zirconium and hence are more nearly comparable with Hanford material. In two runs, plutonium yields of 96.4% and 96.8% were obtained. Goeckermann, Hopkins, and Yett have studied the following variables in the peroxide isolation process: H_2SO_4-HNO_3 ratios, iron concentration, temperature, time of addition of H_2O_2, and time of digestion.

Goeckermann and Hopkins have tested the modified isolation process described in the HEW flowsheet of June 21 with Clinton material. Satisfactory yields have been obtained. Beard has investigated the possibility of using acid media other than HNO_3 in the plutonium peroxide isolation process. Hydrochloric acid appears to be satisfactory; sulfuric and benzenesulfonic acids fail to dissolve the metathesis plutonium completely. Haeckl has studied the precipitation of lanthanum fluoride from bismuth phosphate solutions 10 N in HNO_3 using Clinton material fortified to Hanford concentrations of plutonium; yields were satisfactory.

Process Development (Gilbreath, Group Leader). Hyman, Blaedel, Rasmussen, S. Peterson, Sedlet, and Ader have studied the influence of centrifuging efficiency on decontamination achieved by the low-acidity Bismuth Phosphate Process (by-product bismuth phosphate precipitation from 0.1 N HNO_3 instead of 1.0 N). Four one-liter scale runs have been carried out, two at low acidity and two employing the unmodified process. The low-acidity runs give better decontamination at the end of two cycles. The efficiency of centrifugation is found to influence decontamination appreciably. Plutonium losses in the low-acidity runs average 3.5%.

A 46-page report, dated September 1, 1944, "Chemical Research — Special Chemistry of Plutonium, Status of Purification Program at Termination," (CK-2089), was issued. Summaries of the following investigations are reported.

Purification of plutonium compounds by precipitation and solvent extraction (Jensen and Dixon). Results on purification by precipitations, using thorium as a stand-in for plutonium, indicate that usually the first

precipitation removes the largest fraction of the impurities and successive precipitations succeedingly smaller fractions of the impurities. Certain elements, such as phosphorus and silicon are very persistent. Elements such as iron and potassium (third-period elements) are, as a rule, more difficult to remove than are first-period elements (beryllium and lithium, for example). This is due in part to the fact that iron and potassium are more polarizable and therefore more strongly adsorbed. In general, inorganic precipitants are more effective than organic precipitants.

Plutonium(IV) and (VI) nitrate can be extracted into various organic solvents; however, there are few fundamental data that lead to an understanding of the nature of the basic factors involved in these extractions. The extraction is markedly dependent upon the salt concentration in the aqueous phase; the salt fulfills two roles; first, two increase the concentration of undissociated plutonium nitrate (the plutonium is extracted as a neutral molecule), and second, to "salt out" the plutonium nitrate by decreasing its solubility in the aqueous phase. With regard to the extraction of light elements, there is little tendency for them to be extractable under conditions where Pu(IV) and (VI) can be extracted as nitrates.

Preliminary studies on the possible use of organic reagents for the purification of plutonium were reported by Dean and Orlemann as early as July 1943. Since that time considerable work has been accomplished and summarized in Reports CK-932, 1221, 1371, 1512, and 1763. Several types of compounds have been used in an attempt to obtain the ideal organic reagent for plutonium, i.e., one that is specific for the plutonium ion and forms a compound soluble in an organic solvent immiscible with aqueous solutions.

Oxides of plutonium (Davidson). The formula PuO_2 was conclusively proved by the x-ray work of Zachariasen and Mooney (CK-1096, 1119). No other oxide of plutonium is definitely known to exist; however, there is strong evidence for the existence of PuO and Pu_2O_3. Zachariasen is of the opinion that one of the phases obtained by treatment of PuO_2 with atomic hydrogen is Pu_2O_3 (CK-1518). The compound PuO is believed to be the substance formed during halide reductions when the crucible system is incompletely degassed or when water or oxide is present in the halide preparation. The compound PuO_4 was unsuccessfully looked for on a tracer scale; plutonium peroxide has been shown by Howland to be $PuO_4 \cdot xH_2O$ containing tetravalent plutonium and two peroxide groups per molecule (CK-1511).

Chemistry and purification of the fluorides of plutonium (Heath). Efforts to prepare the higher fluoride of plutonium have been unsuccessful to date, although there is evidence for the existence of a fluoride higher than PuF_4. It is concluded that the higher fluoride is probably not stable.

Chemistry and purification of the chlorides, bromides, and iodides of plutonium (Davidson). Preparation and properties of $PuCl_4$ are described. As yet, no anhydrous $PuCl_4$ of proven identity has been prepared. Attempted "wet" preparations of $PuCl_4$ have given rise to phases with unidentified crystal structures both at Chicago and Los Alamos (CK-1763). Preparation and properties of $PuBr_3$ and PuOBr are described. The crystal structure of these two compounds is described in CN-1813 and CN-1833, respectively.

The preparation and properties of PuI_3 and PuOI are also described. The tracer dry chemistry of plutonium (studies of volatility) is reviewed; it is concluded that these experiments have yielded no reliable information as to the formulas of the halides of plutonium, although they may have been of some value in indicating what are useful halogenating agents and providing some clues as to the volatility of plutonium halides.

The oxidation states of plutonium in solid compounds are considered. It is concluded that the high "electrophilic" higher oxidation states must be stabilized by coordination to the anions of strongly electronegative atoms such as $O^=$ and F^-. At present no simple oxide of the VI or V state is known. Plutonium(VI) or plutonium(V) exists as a fluoride, the "higher fluoride of plutonium." Plutonium(IV) exists as a fluoride and oxide and probably not as any other halide (although oxyhalides may be stable). Plutonium(III) exists as any of the halides or oxyhalides and probably as the oxide.

Production and properties of plutonium metal (Baumbach). The historical development of techniques for the production of plutonium metal on a microscale is described (work of Kirk and Rosenfels). Reduction products of PuF_4, PuF_3, UF_4, and ThF_4 with sodium have never been identified as metal. The reductions of 1-50 micrograms of plutonium compounds using BeO as the refractory material for crucible construction are described (work of Baumbach). The successful reduction of 0.5 mg samples of PuF_4 and PuF_3 is described (work of Fried and Dirksen). Reductions on a 10-mg scale of PuF_3 by Westrum is noted, as is Westrum's reduction of $PuCl_3$ to plutonium metal. The larger scale reductions of PuF_3 by Karle, Fried, and Robinson on the 30-100 mg scale are described. All attempts to electrolyze plutonium metal from aqueous solutions have failed (work of Zvolner). Electrolysis of fused salt baths in an ultramicro scale (using uranium as a stand-in) have not been promising due to volatilization of the salts.

Properties of plutonium metal. Using the capillary displacement method, Kirk determined the density of plutonium metal to be 15.5 gm/cm^3 (CK-1145) with samples weighing from 4 to 28 micrograms that were prepared by the action of barium on PuF_4 in ThO_2. Later S. Fried obtained a value of 16.2 gm/cm^3 on similarly prepared metal and values of 19.5 to 20.2 gm/cm^3 on plutonium metal formed by reduction of PuF_3 by barium in BeO (CK-1512).

S. Katz has made density determinations on variously prepared samples of plutonium metal (CK-1512, CK-1701), obtaining values from 13 to 19.3 gm/cm^3. Further experiments by Katz and Jasaitis have confirmed the existence of two forms of the metal with a low transition temperature. S. Katz has designed a microdilatometer and obtained on heating, a transition at 140°C that involves an increase in volume of about 10% without disintegration of the metal piece. On cooling, the transition to the denser form requires several hours at room temperature. The values for the densities of the two forms of the plutonium metal are estimated to be 16.4 ± 0.5 gm/cm^3 and 19.5 ± 0.5 gm/cm^3 in view of present evidence.

The hardness of plutonium metal samples as found by R. Frank varies from 70 to 300 Vickers (CK-1512, CK-1701, CK-1763), and hardness values for various alloys have been determined (CK-1701). Plutonium metal in its soft, lower density form (beta form) has a hardness about like that of copper, while the hardness of its higher density form (alpha

form) resembles that of uranium. The melting point of plutonium has proved difficult to determine on a microgram scale. Jasaitis and Robinson have finally refined their experiments to indicate a melting point under 810°C on tantalum (CK-1586, CK-1701). Results in another laboratory using a "thermal arrest" method on relatively large quantities have since shown the true melting point to be about 630°C.

Plutonium metal is, when freshly prepared, a white, bright, soft metal that tarnishes quite readily in air with continued oxide formation. The metal is pyrophoric when finely divided and burns with a bright white light. The metal does not react appreciably with water or alkaline solutions but is rapidly soluble in dilute hydrochloric acid, liberating hydrogen, and giving a bright blue solution of $PuCl_3$. The metal does not appear to form a mercury amalgam (CK-1586). Plutonium metal does not appreciably alloy with tantalum, columbium, or tungsten at 1000°C, but alloys are formed with platinum, molybdenum, and zirconium (CK-1701).

Vapor pressure of plutonium metal and plutonium compounds (Simpson). The following vapor pressure tables are included.

Vapor pressure of plutonium metal in tantalum:

T°K	p (mm of Hg)
1800	1×10^{-2}
1700	5×10^{-3}
1600	2×10^{-3}
1500	5×10^{-4}

Vapor pressure of PuO_2:

2000°K	1.1×10^{-3}
1900	1.8×10^{-4}
1800	2.4×10^{-5}
1700	2.5×10^{-6}
1600	2.0×10^{-7}
1500	1.2×10^{-8}

Vapor pressure of PuF_3:

1250°K	2.8×10^{-5}
1316	1.9×10^{-4}
1390	1.3×10^{-3}
1470	9.0×10^{-3}
1562	5.4×10^{-2}

Neutron measurements (Kohman, Weissbourd). The development of the multiple ion chamber neutron counter is described. It is shown capable of detecting with 95% certainty the emission of about 20 neutrons per minute from a source in a total counting period of 12 hours. Results of neutron measurements on four samples of plutonium metal are given.

Helen took care of Kristine, the Ghiorso baby, in the morning.

War news on all fronts is favorable to the Allies except near Chungking where the Japanese are closing the 110-mile gap to divide China.

Wednesday, September 20, 1944

I sent a memo to Hogness requesting that four more cans of the irradiated thorium carbonate be shipped to us from Clinton as soon as possible. The U^{233} to be extracted from it is needed mainly for our own experiments; the previously extracted U^{233} is needed for use at Site Y and by Anderson at Argonne.

At 3:30 p.m. Stan Thompson left for New York City and New Haven, Connecticut. He will be attending a conference in New York tomorrow and Friday to decide what to do about recovering uranium from the process waste solutions. At New Haven he will visit Yale University to observe the ether extraction equipment being used to recover uranium from ores. He will be back in Chicago this Sunday.

I attended an evening meeting of the Extraction and Separation Processes Sub-section of Section C-I at 7:45 in Room 209, Eckhart Hall. Others present were Albaugh, Arnold, Beard, Blaedel, Daniels, Egan, Gilbreath, Goeckermann, Hindman, Hoekstra, Hyman, Katz, Katzin, Larson, Lawroski, Manning, Orlemann, S. Peterson, Post, Pye, Sheft, Stewart, R. Thompson, Walling, Winner, and others. Hoekstra presented extensive data on prereduction studies aimed at determining the effect of hydrazine upon product carrying; Clinton has found the hydrazine is formed in the dissolution of uranium slugs. Evidence was presented which indicates that the poor carrying observed in the presence of N_2H_4 is at least partially due to reduction to Pu(III).

Experiments employing hydroxylamine as a reducing agent have been performed and show that the carrying of plutonium by bismuth phosphate is adequate. Goeckermann presented methods of improving the yields in plutonium peroxide precipitation from $La(NO_3)_3$ solutions containing iron. He has found that increasing the HNO_3 concentration from 0.5 M to 2 M markedly increases the tolerable limit for Fe^{+3}; the revised procedure also shortens the operating time for precipitation and digestion from two hours to one hour.

Blaedel summarized current work on the solvent extraction and decontamination process, in particular, the consideration of schemes utilizing the selectivity exhibited by many solvents for either Pu(IV) nitrate or Pu(VI) nitrate as compared with Pu(III) nitrate; this process replaces the ether solvent extraction process which has been abandoned because of its hazardous nature. Batch extraction experiments on a laboratory scale indicate the chemical feasibility of a process employing the Pu(IV)-Pu(III) nitrate system. Several reducing agents have been found capable of producing and maintaining a non-extractable valence state of plutonium. Hexone has been used as the solvent in most of the laboratory determinations, but other possibilities, notably the polyethers (cellosolves, dibutyl carbitol, etc.) are receiving consideration at present.

War news is pushed out of the top headline today because Governor Thomas Dewey, presidential nominee, was in a train wreck. He was, however, only "shaken up," according to the newspapers.

Thursday, September 21, 1944

In a memo to Stearns, Dempster and Ralph Lapp state that two samples of U^{233} from Montreal have been analyzed by means of the mass spectrograph and found to contain 79% and 72.8% U^{233} respectively.

Report CN-2088, "Chemical Research — Basic Chemistry of Plutonium, Report for Month Ending September 1, 1944," was issued. It contains the following information of interest:

Basic Chemistry Group (Hindman, Group Leader). McLane and Dixon, studying plutonium complexes, have used electrical transference methods to determine the sign of the charge of Pu(IV) in solution. Evidence for complex anions of Pu(IV) has been obtained in solutions of 10 M HNO_3, 10 M HCl, 0.1 M H_2SO_4, 1 M H_2SO_4, 1.1 M HAc-0.06 M NaAc-1.1 M $NaNO_3$, 0.1 M HNO_3-0.1 M $H_2C_2O_4$, 20% UNH-0.5 M H_2SO_4-0.1 M H_3PO_4, 20% UNH-0.5 M H_2SO_4-0.6 M H_3PO_4.

Hindman has studied the complex ions of plutonium(IV) and nitrate and finds that the optical density of certain portions of the absorption spectrum of Pu(IV) in 0.1-2.1 M HNO_3 has a linear dependence on the first-power function of the NO_3^- concentration. The visual appearance of Pu(IV) solutions in this range of HNO_3 concentration is brown. Plutonium(IV) in more concentrated solutions (ca 8 M) appears green. This color has been attributed to the formation of $Pu(NO_3)_6^{=}$. The absorption spectrum of Pu(IV) in strong nitrate solutions [$Pu(NO_3)_6^{=}$] is markedly similar to the absorption spectrum of Pu(IV) extracted into hexone from strong nitrate solutions, suggesting that the $Pu(NO_3)_6^{=}$ group enters hexone perhaps in the form of some compound such as $(NH_4)_2Pu(NO_3)_6$ or $H_2Pu(NO_3)_6$.

O'Connor has compared the hydrolytic behavior of Pu(VI) and U(VI) by potentiometric titration of these ions with NaOH. Titration of Pu(VI) does not result in the precipitation of an insoluble compound, as is the case with U(VI). The Pu(VI) appears to react rapidly with two moles of OH^- and very slowly with additional OH^-. Hindman and Ames have studied the completeness of Pu(VI) reduction by NO^- in various solutions. The reduction of 0.05 M Pu(VI) in 20% UNH-0.2 M HNO_3-0.5 M H_2SO_4-0.1 M $NaNO_2$ is found to be 99% complete in 20 minutes at room temperature.

Magnusson and La Chapelle, carrying out a general investigation of the basic chemistry of neptunium, have established the +6 oxidation state for that element by the following experimental procedure:

By repeated bromate cycles, 50 micrograms of uranium-free Np^{237} were freed from plutonium and the amount of lanthanum carrier reduced to 50 micrograms. The fluoride precipitate from the last bromate cycle was dissolved by heating to fuming with sulfuric acid. The solution after diluting to 100 microliters was 1 N in H_2SO_4. This solution was transferred to a capillary cone and made alkaline with ammonia gas. A white, flocculent precipitate was obtained which was washed twice with 50 microliter portions of water. The precipitate was partially dried by warming it under an aspirator vacuum for several hours. The precipitate was dissolved in 4.7 microliters of 2 M H_2SO_4 and 2.3 microliters of 0.45 M in $KBrO_3$ to give a colorless solution. The solution was heated to 90°C for two hours in order to oxidize completely the neptunium, the volume being maintained at 7 microliters by the addition of water. Immediately upon heating, the

solution became pale green in color, the intensity of which was somewhat greater than that of a solution of $UO_2(NO_3)_2 \cdot 6H_2O$ of comparable concentration. The solution does not exhibit fluorescence. When tested with an ultraviolet source it gives mainly the 3650 A line of mercury. Seven microliters of 4 M $NaNO_3$-4 M NaAc were added to the cooled solution to give a concentration of 4 M Na^+ and 2 M Ac^-. A crystalline precipitate was formed within the solution which when centrifuged appeared colorless to faint pink under intense incandescent light and green in daylight. The precipitate was not fluorescent when tested with the source previously mentioned. The solubility of the precipitate in 4 M Na^+ at the end of 24 hours equilibration at room temperature appears to be approximately 0.23 g Np/l. (Based on a specific activity of 500 alpha-particle counts per minute per microgram of Np^{237} with the "50%" geometry counters.)

X-ray analysis of the compound by Dr. Zachariasen shows it to be isomorphous with sodium uranyl acetate and sodium plutonyl acetate. The lattice constants for the three compounds are:

a for uranium compound = 10.671 Å

a for neptunium compound = 10.659 ± 0.002 Å

a for plutonium compound = 10.643 Å

Helen played nine holes of golf with the senior Luther Arnold at Jackson Park today.

According to today's paper, British troops are waging a battle for a big Rhine bridge near Nijmagen, Holland.

There was a Project Council Policy Meeting at 9:00 a.m. in Room 209, Eckhart Hall, attended by Allison, Bartky, Chapman, Chipman, C. M. Cooper, Daniels, Dempster, Doan, Eastman, Hogness, Jacobson, Jeffries, Johnson, Leverett, Mulliken, Spedding, Stearns, Stone, Szilard, Vernon, Warner, Wigner, and Zinn. Allison led the meeting, as Compton could not be present.

Hogness reviewed the cooperative effort between the Chemistry Division and the Technical Division on solvent extraction for use in the concentration-isolation step with particular reference to hexone that has shown excellent solvent properties without the explosion hazard of ethyl ether. The use of hexone has been well-demonstrated on a tracer scale, and full-scale equipment in glass is ready for operation at the West Stands, also on a tracer scale. There will be a cooperative effort between Chicago and Clinton in order to carry out full-scale tests at Clinton. Warner asked if this would save work at Site Y, and Hogness replied that if the indicated purities are obtained, it should make unnecessary their purification step and save considerable effort.

There was a discussion of cooperative efforts with Evergreen (Montreal Area) and the limitations on the furnishing of information on pile design, etc. Allison announced that the Chicago cyclotron is about to shut down. Hogness mentioned the Y-12

need for men and that Ashcraft has gone to head the chemical program and wants to take Orlemann. Allison thought that Potratz could be made available from Site Y. Cooper suggested that a group be organized during the next six months to supply tracers, techniques, and equipment. This would be used initially for the Project, but later wherever needed. This is in line with the emphasis on tracer applications contained in many of the suggestions prepared for the Jeffries committee on postwar uses of nucleonics.

Friday, September 22, 1944

I received a copy of a memo from Zachariasen to Allison confirming the formula $UO_2HPO_4 \cdot 4H_2O$ for samples of uranyl phosphate submitted some time ago by Clifford Smith of Cunningham's group and by Leader at Site X.

Last Monday Magnusson made the remainder of his $Np(NO_3)_4$ solution ammoniacal and obtained a dark yellow-brown amorphous precipitate, which by 10 a.m. Tuesday changed to yellow-green crystals. Today the crystals of ammonium dineptunate were definitely green. Magnusson made four exposures of the sample with Kodachrome B using a Zeiss microscope and a Zeiss Ikon camera attachment.

Helen visited Mrs. Metcalf who lives in the apartment on the floor above us. Metcalf works in the patent department and is involved with many patent applications evolving from Section C-I.

Today's reports indicate that German troops were going to blow up the Nijmagen bridge across the Rhine, but American Air Force troops were able to dismantle the wiring and save the bridge.

Saturday, September 23, 1944

Theocian Carter, a technician in Orlemann's group, resigned in order to accept another position. Elaine Freeman, who is returning to school, also resigned.

Kurt Kraus returned at 2:30 p.m. from his trip to Clinton Labs which began on September 16.

I wrote to Curtiss at the Bureau of Standards in Washington to inform him that we will try to participate in the program of comparative measurements of the radium content of sludges and ores. This recently has been the subject of correspondence between Allison and the Bureau. I pointed out we are very pressed at the moment and are not certain we will be able to analyze all six samples that he has sent us. Also, I asked for his suggestions as to assay methods we might use.

Manning sent E. R. Russell a list of all Section C-I personnel. This will be helpful in the collection of urine specimens as it shows each person's room number and the type of material he handles, e.g., plutonium, U^{233}, fission product, or non-radioactive (see Figs. 21 and 22).

Helen had coffee with Wilma and her mother, Mrs. Belt.

The top headline today indicates that Russian troops have captured Tallinn, the capital of Estonia.

Sunday, September 24, 1944

Helen and I played golf at Gleneagles Golf Club, Course No. 2. Luther Arnold, Sr., Luther Arnold, Jr., Steve Lawroski, and I were in one foursome (scores: LA,Sr.-109, LA,Jr.-101, SL-96, GS-96), and Zene Jasaitis Herman Robinson, and Helen were in a threesome. A snapshot was taken of Steve, Helen, Zene, Herman, and me, which is shown in Fig. 23. We had lunch after nine holes. I had a nine and Steve a three on the ninth hole; so I was four down at the end of nine. I pulled even with Steve at the end of 17, then we both took seven on the par five 18th to come in with a tie.

Stan Thompson returned at noon from his trip to New York City and New Haven, Connecticut. He has been gone since last Wednesday.

Monday, September 25, 1944

Saturday, another laboratory romance resulted in the marriage of Sigfred Peterson and Helen Pellock.

Bartell returned from his vacation which started on Saturday, September 16.

After extensive range measurements on sample 49DD-4 (the counting disc prepared by James and Morgan from repeated dichromate oxidation cycles on the 200-mg plutonium plus deuterons, St. Louis bombardment), Morgan has decided to attempt to separate the possible components of the sample or identify the activity by chemical means. Therefore, he has dissolved sample 49DD-4 and carried out chemical manipulations on the solution which yielded the following mounted samples:

49DD-14: Lanthanum fluoride precipitation made after oxidizing the solution with silver plus persulfate (50 alpha-particle c/m).

49DD-17: Precipitate (Ag_2SO_4 or Ag_2SO_3) which formed on SO_2 reduction of supernatant from Sample 49DD-14 (38 alpha-particle c/m).

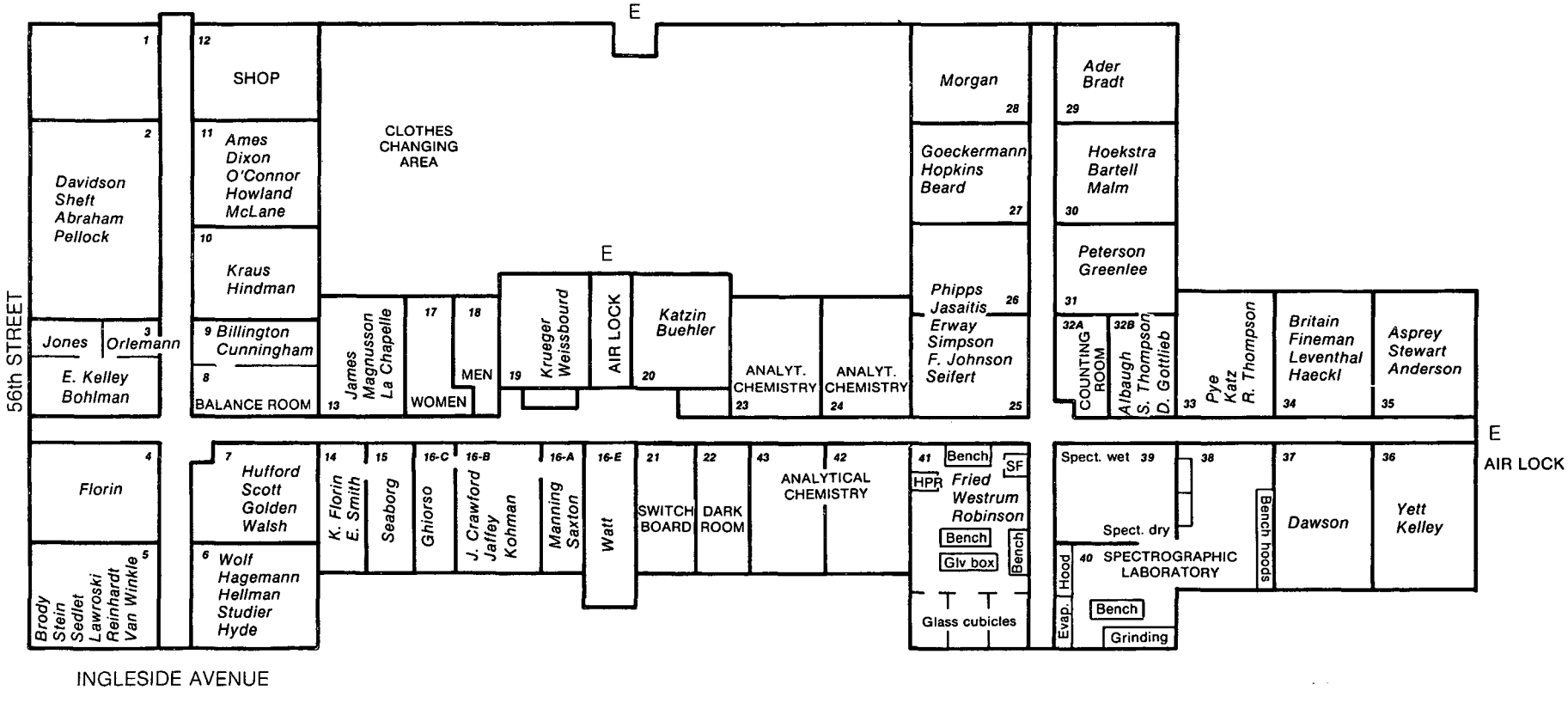

Figure 21. Room assignments in New Chem. September 1944.

XBL 792-592

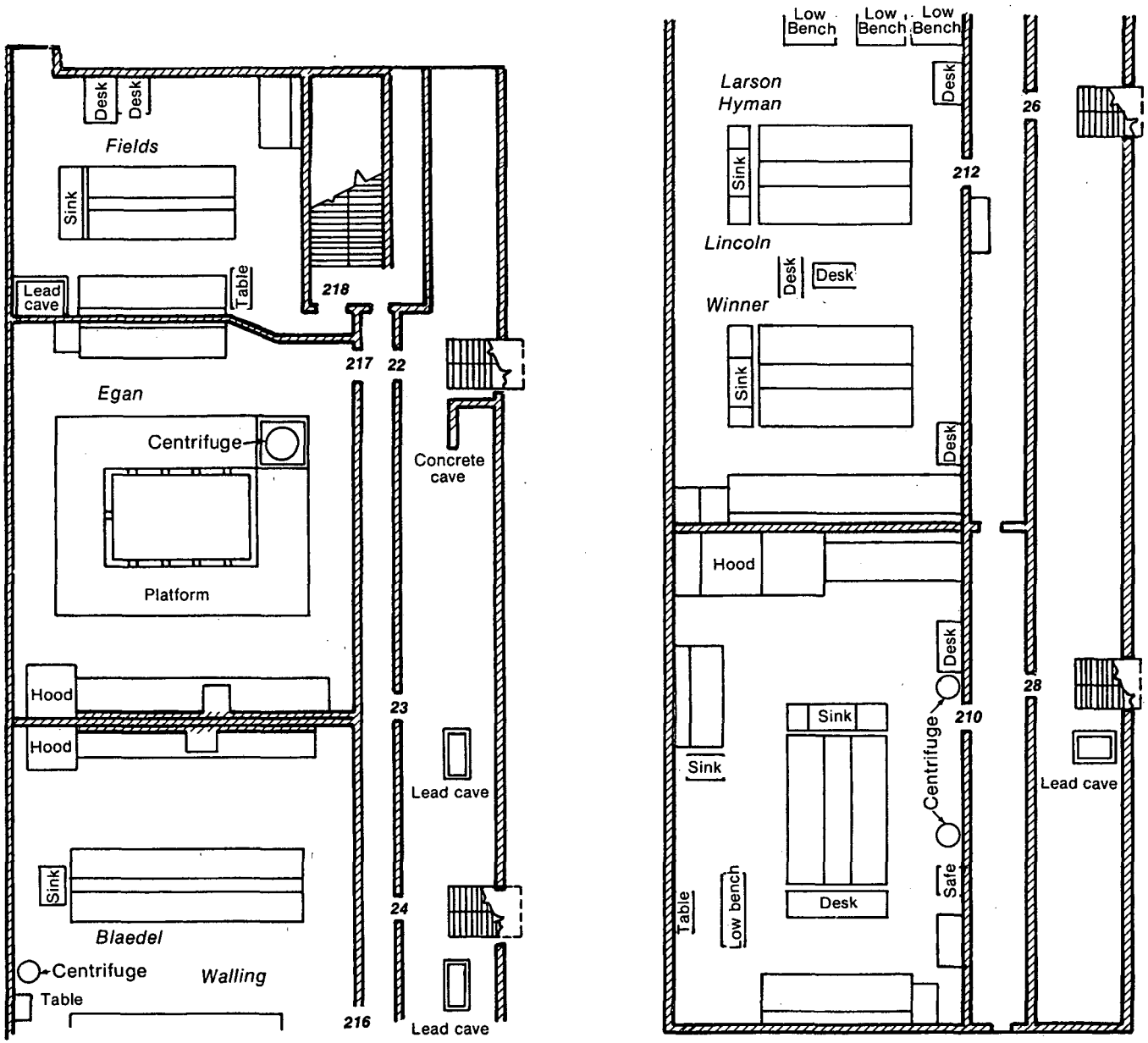

Figure 22. Partial room diagram and room assignments of second floor, West Stands. September 1944

XBL 793-896

Figure 23. Stephen Lawroski, Glenn Seaborg, Helen Seaborg, Zene Jasaitis, and Herman Robinson at Gleneagles Golf Club. September 24, 1944.

CBB 792-2377

49DD-15: Lanthanum fluoride precipitation made from supernatant of 49DD-17 (96 alpha-particle c/m).

49DD-16: Supernatant from 49DD-15 precipitation, evaporated to dryness (no activity).

I summarize in a memo to F. A. Jenkins in the U.S. Engineer's Office in Oak Ridge, the suggestions I made during our visit to Y-12 on September 16 concerning the organizational arrangements needed to carry out their project to recover chemically the uranium-235 enriched by the electromagnetic process.

I also mention my subsequent discussion with Ernest Lawrence about using, as a source of the needed manpower, the men now engaged in purifying and converting their final U^{235} product to the fluoride before delivery to Los Alamos — I suggest they explore the possibility of doing this production work on the final product elsewhere. I close by indicating we are looking forward to receiving their flowsheet to see if we can make further suggestions more specifically on their chemical processes.

Helen had Wilma and her mother, Mrs. Belt, over to lunch.

Japanese reports say that 200 U.S. planes raided several central Philippine cities, including Legaspi and Cebu.

Tuesday, September 26, 1944

This was the last day at the Met Lab for Stan Thompson who is transferring to Hanford. He is going by way of southern California to pick up his wife Alice and little daughter Ruth Ann and will include a visit to his mother's home on Dorothy Street in South Gate.

John Burr resigned to accept a position as a chemistry instructor at Northwestern University.

I received a memo from Stan Thompson about his conferences in New York City last Thursday and Friday. The meetings were held at the offices of the Manhattan District Engineer and were attended by Brown, Doan, Kridel, Leverett, Maloney, Murphy, Ruhoff, Tepe, Vance, Winters, and others. The purpose was to discuss recovery of uranium from the process waste solutions. The Army considers uranium recovery a B priority problem; A. V. Peterson will be sending a letter to Compton covering the distribution of effort and a suggested program for Chicago and Clinton. Thompson indicated that we are in a position to do general scouting work on uranium recovery and would like to do so. Last Friday afternoon, Thompson, Maloney, and Tepe visited Yale University and observed the ether extraction equipment. The design of the glass system appears very simple and working well. It is being used entirely for the recovery of uranium from ores.

I sent Captain Chapman in the Chicago Area Engineers Office some secret material that I want to have today's courier take to Montreal for my use during my trip there starting tonight.

Watt wrote to R. F. Peterson, attention F. S. Chambers, in Wilmington, responding to the concern Chambers has expressed over the possible explosion hazard involved in adding sodium nitrite to solutions containing ammonium salts; this concern has arisen as a result of Watt's September 11 letter to Squires in which he proposes that solutions to be processed in the 231 Building (isolation) at Hanford be made 0.25 M in ammonium sulfate before peroxide precipitation. Watt wrote it seems highly improbable that any difficulty should arise in such dilute aqueous solutions. He also questions whether sodium nitrite is the best reagent to use in destroying excess hydrogen peroxide prior to recycling peroxide supernatants. Finally, he points out that if there is a potential hazard, sulfate salts other than ammonium sulfate could be used.

The Health Division completed the weekly radiation surveys of the New Chem, filtered air section. The following rooms were found to have high alpha or beta-gamma contamination: Room 5 (Lawroski, Brody, Reinhardt, Stein); 6 (Wolf, Hagemann, Hellman, Studier, Hyde); 10 (Hindman, Kraus); 11 (Ames, Dixon, Howland, McLane, O'Connor); 13 (James, La Chapelle, Magnusson); 22, 23, 27 (Beard, Goeckermann, Hopkins); 30 (Bartell, Malm, Hoekstra); 31 (Greenlee, S. Peterson); 33 (Pye, R. Thompson); 34 (Britain, Fineman, Haeckl, Leventhal); 36 (Kelley, Yett); and 41 (Fried, Westrum, Robinson).

Today Helen began taking an afternoon chemistry course at the YMCA College in downtown Chicago.

Again the only discouraging war news for the Allies comes from Chungking, where reports are that two Japanese columns are marching to flank Kweilin.

At 8:30 p.m. I left Chicago on a N.Y.C. train to Toronto on my way to visit the Montreal Laboratory of the Canadian Project. I expect to be back in Chicago by next Sunday.

Wednesday, September 27, 1944

I arrived in Toronto and then continued on the N.Y.C. and Canadian Pacific R.R. to Montreal.

Thursday, September 28, 1944

In Montreal. W. W. Watson, the Metallurgical Project representative stationed in Montreal, and I visited the Montreal Project. Most of my discussions were with the chemists — Paneth, E. W. R. Steacie, Bert

Goldschmidt, and J. Guéron were present in practically all the discussion. The more important topics I covered with them today are:

- Thorium metal vs. carbonate for irradiation (Sutton and Bewick).
- Separation of U^{233} or Pa^{233} by adsorption (L. G. Cook, W. E. Grummitt, and G. Wilkinson).
- Solvent extraction (Reid, L. Yaffe, Fitch, Russell, A. G. W. Cameron, and Mungen).
- Organic complexes (A. G. Maddock, A. C. English, and Musgrave).
- Fluorine method (Maddock).
- Purification of U^{233} (Yaffe, English, and Cruikshank).

In the afternoon I gave a talk on the hazards connected with the alpha-particle radiation from U^{233} and the gamma-radiation from Pa^{233} and made a few suggestions on how these might be minimized. In the same talk I discussed the latest measurements in Chicago on the specific activity, half-life, alpha-particle range, etc., of U^{233}. Also, I gave some speculations as to a probable decay chain for U^{233} (the 4n + 1 series) and also some speculations on the electronic structure of elements 89-95, inclusive (i.e., my actinide concept).

Friday, September 29, 1944

In Montreal. I held further discussions with members of the Montreal Project, covering the following:

- Extraction of protactinium from ores (H. G. Heal, Cook, and F. Morgan).
- Fission products of U^{233} (Yaffe, Wilkinson, and Grummitt).
- Barium branching ratio in U^{233} fission (Yaffe, Wilkinson, and Grummitt).
- Rare gas experiments — measurement of helium, krypton, and xenon (W. J. Arrol).
- Fission chambers (Maddock, Miller, and B. Pontecorvo).

At 4:30 p.m. Watson and I met to review decisions with Arrol, Goldschmidt, Guéron, Huffman, Maddock, Paneth, and Steacie. The more significant items discussed were:

Metal versus carbonate. I indicated that, for the immediate task of making one gram of U^{233} per day with material around the outside of the pile, everything is in favor of thorium carbonate.

Extraction of Pa^{233} versus U^{233}. It was agreed the product should be separated as U^{233} — no point in separating as Pa^{233}.

Elimination of protactinium by adsorption or precipitation. Direct separation by adsorption is not practical. The precipitation method involving MnO_2 is fairly certain to work.

Solvent extraction. This is clearly the best method for extracting U^{233} from thorium carbonate. Ether is probably the best solvent so far developed, but work on other solvents should proceed.

Purification of U^{233}. The details of how to purify U^{233} extremely well were discussed. As soon as there is a sample of quite pure U^{233}, the Montreal group wants to measure the decay period. As soon as there is a sample absolutely free from beta particles, the group will launch an investigation to find the disintegration products and look for half-life periods and radiations of these products.

Extraction of protactinium from ore and determination of valences. Montreal chemists have been working with the most insoluble residues of Canadian uranium ore in search of protactinium. I suggested the more soluble carbonate fraction might better be used. I pointed out it will be especially interesting to look for an oxidation state of 4 and possibly state 3 in the protactinium.

Fission products of U^{233}. Montreal chemists would like to have the 10 mg of the U^{233} they sent to Anderson at Chicago retained in Chicago and irradiated by neutrons in one of the piles for at least a month. Then it should be sent to Montreal for a study of the fission products of U^{233}. I agreed to prepare the sample for irradiation in a convenient form.

Fission chambers. The Montreal group has measured the fission cross section of U^{233} by using UF_6 and BF_3 gases in their fission chambers. The spontaneous fission rate of U^{233} has been measured by Pontecorvo at equal to or less than 3 neutrons per minute per gram, this value obtained even with U^{238} impurity. A discussion was held on the difference between the Montreal and Chicago measurements of the fission cross section of U^{235}. The Montreal value is 4.6×10^{-24} cm^2 and the Chicago value is 3.9×10^{-24} cm^2. There seems to be no explanation for the discrepancy.

In a general discussion it was decided that for the present there should be no division of work between Montreal and Chicago. Montreal will carry on investigations of all problems connected with U^{233} production while we at Chicago will consider only those problems which interest us.

Saturday, September 30, 1944

I visited Watson's office and prepared notes on my discussions with members of the Montreal Project on Thursday and Friday.

I boarded a train for Toronto and Chicago.

Sunday, October 1, 1944

I arrived back in Chicago at 8:00 a.m., via the Canadian Pacific and New York Central Railroad, from my trip to Montreal.

Helen and I played 18 holes of golf at the Gleneagles Golf Club No. 1 with Edrey Smith (my secretary), Zene Jasaitis, Herman Robinson, and Bob Freeman (Robinson's brother-in-law). Our scores were ZJ-150, HR-125, BF-95, and GS-97. Helen and Edrey played in a twosome. Photographs were taken of the group (see Figs. 24 and 25).

I was told that Joe Katz and Sonia Weiner, Thorfin Hogness' secretary, are being married today in Grand Rapids, Michigan. He will be back to work tomorrow.

Later Helen brought me up-to-date on her activities while I was away. Last Wednesday she played 18 holes of golf at Jackson Park with two men and had a very good first two holes. She then had dinner at Wilma's. The next day she went to her afternoon chemistry class at YMCA College downtown. On Friday Helen worked at the Met Lab with Kohman on the secret version of the "Table of Isotopes." Wilma came for coffee in the afternoon. Wilma, with her mother Mrs. Belt, again came over for coffee yesterday afternoon.

Baseball has taken the top headline from war news today. The St. Louis Browns have won the American League pennant.

Monday, October 2, 1944

Last Wednesday morning, a few minutes after midnight, marked the first power run of a Hanford chain-reacting pile. Two weeks earlier, Fermi had brought the production pile (100B) into criticality for the first time but without water cooling. During this midnight run of about two hours, the power level exceeded anything that had ever been attained in a pile before, either at Oak Ridge or Argonne. But following the two-hour run, to everyone's consternation, the power level began to decline slowly and steadily of its own account. By Wednesday evening the chain-reaction ceased completely. Then early the next morning the chain-reaction started up spontaneously, and by evening the power was up to its former high level. Soon the pile began shutting itself down again. Many hypotheses were advanced, the most acceptable being that the pile had generated some kind of poison to reduce the reactivity. If it were indeed a poison, then it would have to have a half-life of around 9.7 hours to fit the pile's behavior pattern. The fission product Xe^{135}, with a half-life of 9.4 hours, was thus suspected. Zinn at Argonne and Doan at Clinton Laboratories were immediately notified and asked to run experiments to determine if a Xe^{135} effect on the operation of the piles there could be detected. Zinn soon confirmed that the production of Xe^{135} in the chain-reaction does have a poisoning effect. Now the question arises, must the Hanford piles be modified if they are to produce Pu^{239} on a grand scale?

Figure 24. Helen Seaborg and Edrey Smith at Gleneagles Golf Club. October 1, 1944. XBB 792-2375

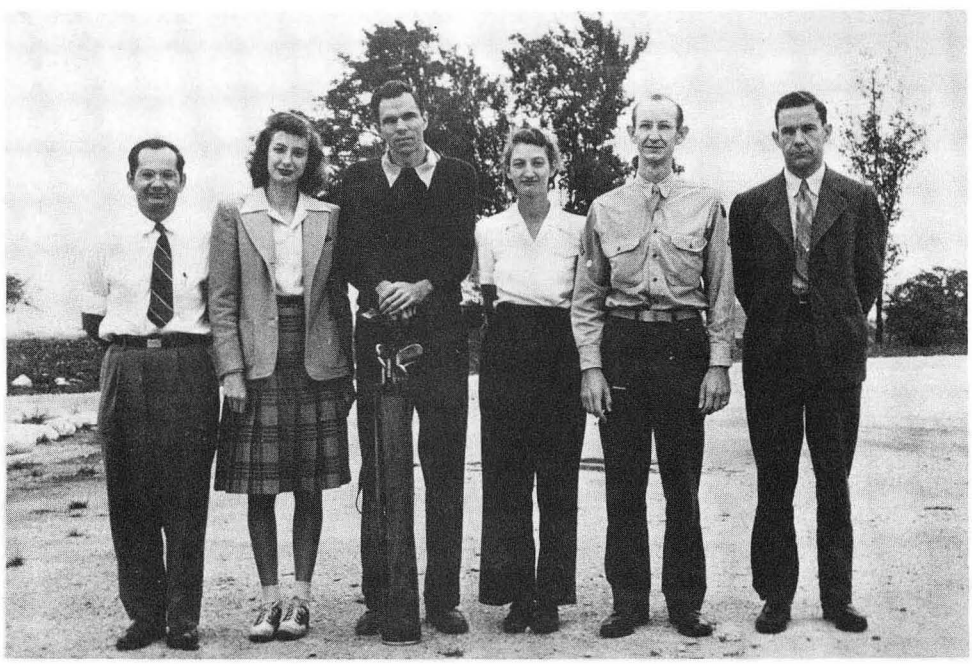

Figure 25. Zene Jasaitis, Edrey Smith, Glenn Seaborg, Helen Seaborg, Bob Freeman, and Herman Robinson at Gleneagles Golf Club. October 1, 1944. XBB 792-2371

Last Wednesday, the day I arrived in Montreal, Morgan completed a series of HNO_3 oxidation cycles and dichromate oxidation cycles on the main plutonium fractions (sample 49DD-1 and 49DD-3) from the dissolved target from the 200 mg plutonium plus deuterons, St. Louis cyclotron bombardment. His final supernatant and lanthanum fluoride precipitate both showed alpha-particle activity with the range of 94^{239} (200 c/m and 50 c/m, respectively). He has concluded that there is no element 95 isotope that has a half-life of the order of magnitude necessary for detection which can be oxidized by dichromate, but not nitric acid, under the conditions used. We have decided therefore, that the best prospects for finding element 95 seem to be in working with the fraction unoxidized by either dichromate or Ag^{++}. This would be sample 49DD-14 which was derived from the first series of dichromate oxidation cycles on the original solution of the plutonium cyclotron target, followed by a silver plus persulfate oxidation on Monday.

Wednesday Hogness made a request to Bartky for another batch from Clinton Laboratories of the lanthanum fluoride slurry containing one gram of plutonium. He mentioned that the slurry we now have will be used to make one long continuous run in the 3-inch solvent extraction column as soon as data on spiked artificial slurry are obtained. The requested additional batch will be used for a second run. The container for the slurry will be sent from Chicago by truck and will arrive in Oak Ridge some time before October 18.

Hilberry wrote to W. O. Simon at the Hanford Engineer Works on Wednesday about schedules for tentative visits there by Metallurgical Project personnel for the months of October and November. I was cited in this paragraph of his letter.

"Leaving Chicago November 4th and arriving in Richland November 6th, T. Hogness, Warren Johnson, and G. Seaborg. The length of this trip has been left indeterminate since its duration will probably depend on the nature of the discussions between the chemical staff here and those Metallurgical representatives. It would probably be of approximately one week's duration. The purpose of this visit is to coordinate the programs of the chemical laboratories of the Project with the needs of the plant as determined after a month of plant operation."

As Hilberry wrote, the schedule is tentative; I am not sure that I shall be able to get away at the time he specifies.

Last Wednesday the evening meeting of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I at 7:45 in Room 209, Eckhart Hall, was attended by Ames, Arnold, Brody, Cunningham, Daniels, Dawson, Dixon, Fried, Hagemann, Hindman, Jaffey, Jones, Katzin, Kohman, Kraus, Krueger, La Chapelle, Manning, McLane, O'Connor, S. Peterson, Simpson, R. Thompson, Warner, Weissbourd, and Westrum. Weissbourd reported on the progress of the work to determine the rate of spontaneous fission of Pu^{240}. The problem is difficult because only one or two fissions per week or month are observed to occur. This makes it necessary for the counter to have a zero background and requires that it be perfectly shielded against electrical disturbances and free from microphonics. The assembly was checked for background for 200 hours and

found to give no counts. A sample believed to contain Pu^{240} is now being counted.

O'Connor discussed the titration of U(VI) and Pu(VI) with sodium hydroxide. A study of the absorption spectra of the plutonium nitrate solution suggests that Pu(VI) passes through three ionic species [such as PuO_2^{++}, PuO_2OH^+, $Pu(OH)_6$] as the pH of the solution is raised. Dixon mentioned the electrical transference work which McLane and he have done since the latter's report. Thirteen experiments have been carried out on Pu(III) chloride, Pu(IV) chloride, Pu(IV) phosphate, Pu(IV) fluoride, and Pu(III) sulfate.

La Chapelle reported on further work on the specific activity of Np^{237}, giving the following values:

(a) 791 c/m/microgram of neptunium
(b) 1,517 dis/m/microgram of neptunium
(c) 2.21×10^6 years half-life

Range measurements show no trace of plutonium. The measurement is accurate only to 5%.

The growth of Pa^{233} has been followed in a precipitate of sodium neptunyl acetate and is found to follow the theoretical curve given by the equation $A = A_0 + A_S(1 - e^{-\lambda t})$, in which A represents counts per minute at time t, A_0 at time zero, and A_S at equilibrium. La Chapelle added that attempts to prepare the +3 state of neptunium by reduction with $NaNO_2$, SO_2, and NH_2OH, have resulted in no characteristic color change. Manning asked if compounds other than the dioxide and sodium neptunyl acetate have been subjected to x-ray analysis, and La Chapelle indicated that a compound, as yet unidentified, of the type $(NH_4)_2Np_2O_7$, has been examined.

Thursday was the last day at the Met Lab for George W. Watt. His transfer to Site W is effective October 12. Watt will direct the initial plutonium isolation operation in the 231 Building. Fineman returned from his vacation which started on September 15.

On Thursday, James completed the counting of various fractions of the plutonium plus deuterons, St. Louis bombardment, that Morgan worked on during James's vacation from September 8 to September 20. On this day also James began leaching additional plutonium from the target from that same bombardment.

Manning sent Allison a memo on Friday concurring in a revised specification for iron in potassium carbonate to be used in the Hanford process.

The Metallurgical Laboratory Report for August 1944 was issued last Friday and contains the following information of interest. The regular program studies of the Hanford process were continued by the Chemistry Division. It has been found that iron, which exerts a redissolving effect on plutonium peroxide, may be removed below a troublesome concentration by two prior precipitations of the plutonium with lanthanum fluoride. Work is proceeding on handling various isolation solutions.

The solvent extraction method for plutonium recovery made rapid advances during the month of August, and installation at Clinton was recommended. Experimental operation of the solvent used, hexone (methyl-isobutylketone), seems to extract plutonium as a hexanitrate complex. Hydrazine is being used for the removal of plutonium from the hexone, and the reduction proceeds quite satisfactorily. There is some expectation that the effect of radiation may limit the life of the hexone to about 125 days.

Extraction decontamination studies during the month continue to show satisfactory results at 1-G centrifugation (standing only) for three unmodified bismuth phosphate cycles or for two bismuth phosphate cycles plus one scavenging cycle. Studies on a four-ton batch show that a certain critical condition exists between the acidity and the plutonium losses in the special process developed for early operation at Hanford. This process is still regarded as feasible, even though extra care is required to control the acidity.

Approximately 8 mg of U^{233} have been recovered from thorium carbonate irradiated about seven months in the Clinton pile. The U^{233} was found to be 85% pure upon analysis with a mass spectrograph. A corrected half-life determination on the U^{233} sample gives a figure of 1.38×10^5 years. A corrected cross section for U^{233} as compared with U^{235} (23:25) gives a ratio of 0.96 for thermal (paraffin-moderated) neutron fission. The +6 oxidation state of neptunium has been identified by means of sodium neptunyl acetate.

Health protection services were strengthened during August. Radiation surveys in New Chemistry indicated that several rooms frequently exceed 100 mr per eight-hour day. Levels of radiation in the work areas of the West Stands semiworks have consistently been above 100 mr per eight-hour day. It has been found necessary to set up detailed and routine film procedures in order to have records for both health and legal purposes. Of 140 pocket ionization chamber readings obtained during the month with readings that exceeded 100 mr, 80 readings were considered valid. Most of these high readings came from personnel in West Stands and, in particular, Room 217 of West Stands.

Studies have been initiated to investigate the uptake, absorption, and transport of fission product materials in soils from Clinton and Hanford sites. This work is being conducted upon a subcontract with the Radiation Laboratory at Berkeley. A program to determine the biological effects of radiation from various radioactive substances is continuing. Progress has been made in die-casting jackets around heavy metal slugs, and the production of a large number of slugs for test purposes appears imminent. Cycling tests of unbonded Hanford slugs show that the thermal conduct between slug and jacket remains good even with four 300°C temperature cycling. This indicates that full Hanford power operation is feasible.

The total technical staff of August 31 as compared with July 31 and the ceiling figure is as follows:

Technical	*As of 7-31-44*	*As of 8-31-44*	*Ceiling*
Project Director's Office	2	2	
Laboratory Director's Office	3	3	
Chemistry	312	275	
Health	145	157	
N.R.C.C.	2	2	
Physics	149	146	
Technology	197	165	
Information	11	10	
Patent	6	6	
TOTAL	823	766	916

The total expenditures for the month of August were $903,344 as compared with $1,244,264 for July. This is a decrease of $340,920.

An outline of the organization for my section, as of the beginning of October is as follows:

Section C-I. G. T. Seaborg, Section Chief; W. M. Manning, Associate Section Chief; E. F. Orlemann, Associate Section Chief (replacing G. W. Watt who left for Hanford 9-28-44).

Sub-section I. (primarily W work). F. W. Albaugh, Assistant Section Chief (replacing S. G. Thompson who left for Hanford on 9-26-44).

Group 1: Extraction and Decontamination. R. C. Thompson, Group Leader (replacing Albaugh who became Assistant Section Chief).

Group 2: Concentration and Isolation. D. G. Pye, Group Leader (J. J. Katz, Assistant Group Leader, to replace Pye about October 21).

Group 3: Process Development. J. R. Gilbreath, Group Leader.

Group 4: Solvent Extraction. S. Lawroski, Group Leader; (Semiworks: C. J. Egan).

Sub-section II. B. B. Cunningham, Assistant Section Chief.

Group 5: Basic Dry Chemistry. O. C. Simpson, Group Leader.

Group 6: Basic Chemistry. J. C. Hindman, Group Leader.

Group 7: Recovery. L. R. Dawson, Group Leader.

Group 8: Instruments and Physical Measurements. Albert Ghiorso, Group Leader.

"23 Group", *Group 9: Uranium-233 Separation.* L. I. Katzin, Assistant Section Chief in charge of Group 9's U-233 work.

As of October 1, 1944, the Metallurgical Laboratory's total technical manpower level is 690. The Chemistry Division has a total of 221 technical employees of which 158 are classed as academic. These latter 158 employees are distributed approximately as follows: Director's Office — 3; Burton's Section C-III — 23; Sugarman's Section C-III — 18; Section C-IV — 29; Section C-VI — 1; Section C-I — 84 (or about 53% of the entire Chemistry Division).

An alphabetical listing of these 84 Section C-I workers (as of October 1), together with their titles, groups, and room numbers, is as follows:

"Academic" Employee	*Title*	*Group*	*Room No.*
Abraham, Bernard M.		5	2
Ader, Milton		1	29
Albaugh, Frederic W.	Asst. Section Chief	Sub-section I	32
Ames, Donald P.		6	11
Anderson, Herbert H.		7	35
Asprey, Larned B.		7	35
Bartell, Lawrence S.		1	30
Beard, Walter C., Jr.		2	27
Blaedel, Walter J.		3	WS-216
Bradt, Rexford H.		1	29
Britain, J. W.		7	34
Brody, Bernard B.		4	5
Crawford, John A.		8	16B
Cunningham, Burris B.	Asst. Section Chief	Sub-section II	9
Davidson, Norman R.	Asst. Group Leader	5	2
Dawson, Lyle R.	Group Leader	7	37
Dixon, Jonathan S.		6	11
Egan, Clark J.	In charge of Semi-works group	3	WS-217
Erway, Norman D.		5	25
Fields, Paul R.		7	WS-212
Fineman, Phillip		7	34
Florin, Alan E.		6	4
Fried, Sherman		5	41
Ghiorso, Albert	Group Leader	8	16B
Gilbreath, James R.	Group Leader	3	WS-214
Goeckermann, Robert H.		2	27
Golden, Lorraine		8	7
Greenlee, Roy W.		1	31
Haeckl, Frank W.		7	34
Hagemann, French T.		9	6
Hellman, Nison N.		9	6
Hindman, James C.	Group Leader	6	10
Hoekstra, Henry R.		1	30
Hopkins, Horace H., Jr.		2	27
Howland, Jerome J.		1	11
Hufford, Duane L.		8	7
Hyde, Earl K.		9	6
Hyman, Herbert H.		3	WS-212
Jaffey, Arthur H.		8	16B

"Academic" Employee	*Title*	*Group*	*Room No.*
James, Ralph A.		6	13
Jasaitis, Zene V.		5	25
Johnson, Frederick D.		5	25
Jones, Thomas Oswell			3
Katz, Joseph J.	Asst. Group Leader	2	33
Katzin, Leonard I.	Asst. Section Chief	9	20
Kelley, Alec E.		2	36
Kohman, Truman P.		8	16B
Kraus, Kurt A.	Asst. Group Leader	6	10
Krueger, Albert C.		8	19
La Chapelle, Theodore J., Jr.		6	13
Larson, Raymond G.		3	WS-212
Lawroski, Stephen	Group Leader	4	5
Leventhal, Leon		7	34
Lincoln, Dwight C.		3	WS-212
Magnusson, Lawrence		6	13
Malm, John G.		1	30
Manning, Winston M.	Assoc. Section Chief	Section C-I	16A
Morgan, Leon O.		1	28
McLane, C. Keith		6	11
O'Connor, Paul R.		6	11
Orlemann, Edwin F.	Assoc. Section Chief	Section C-I	3
Peterson, Sigfred		1	31
Phipps, Thomas E.		5	25
Pye, Donald G.	Group Leader	2	33
Reinhardt, Richard A.		4	5
Robinson, Herman P.		5	41
Scott, Benjamin F.		8	7
Seaborg, Glenn T.	Section Chief	Section C-I	15
Sedlet, Jacob		1	5
Seifert, Ralph L.		5	25
Sheft, Irving		5	2
Simpson, Oliver C.	Group Leader	5	25
Stein, A. Pauline		4	5
Stewart, Donald C.		7	35
Studier, Martin H.		9	6
Thompson, Roy C., Jr.	Group Leader	1	33
Van Winkle, Quentin		9	5
Walling, Mathew T., Jr.		3	WS-216
Walsh, Patricia D.		8	7
Weissbourd, Bernard B.		8	19
Westrum, Edgar S., Jr.		5	41
Winner, Bernard M.		3	WS-212
Wolf, Michael J.		1	6
Yett, Fowler		2	36

A report entitled, "Magnetic Susceptibilities of Plutonium in Solution in Its Various Oxidation States and of Certain of the Heavy Metal Sulfides Used as Refractories," dated October 1, 1944, describes measurements at Berkeley by Melvin Calvin on solutions containing Pu(III), Pu(IV), Pu(V), and Pu(VI) ions. The measurements of magnetic susceptibil-

ities were made by the Gouy method using a Sartorius microbalance mounted over a large magnet capable of generating fields of the order of 30,000 gauss through a volume of 5 cm^3. The simplest qualitative explanation of the results would be a rare earth series with the electronic configurations Pu(VI) $5f^2$, Pu(V) $5f^3$, Pu(IV) $5f^4$, Pu(III) $5f^5$. From the fact that metallic gadolinium with $4f^7$ becomes ferromagnetic just below room temperature, it may be expected that plutonium metal will have a fairly high number of 5f electrons, perhaps, 5, 6, 7, or 8, and might become ferromagnetic at temperatures not far below room temperature.

In order to obtain a more sensitive method for determining plutonium in the urine of employees and thereby providing a more accurate picture of plutonium body burdens, the Health Division has begun to develop and use cupferron-chloroform extraction as well as bismuth phosphate precipitation and other carrier techniques. Ion exchange resins, such as IR-4, will also be tried. The effect of lanthanum fluoride precipitation on 6 N HCl elutriants from the IR-4 resin will be studied. Techniques for determining the plutonium concentration in feces will also be investigated.

Albaugh wrote a memo to me concerning the separation of neptunium from process solutions. Two 100-ml scale runs have been made to trace the distribution of neptunium in process solutions. The overall yield of neptunium through one bismuth phosphate cycle and a lanthanum fluoride crossover was about 48% in both runs. As the material balance was somewhat low, this value is considered to be a minimum value.

I received a letter from A. A. Frost of Northwestern University concerning the lecture I will give on October 24 on "Isotopic Tracer Technique." Frost requests that I arrange to break my lecture into two parts with a discussion period following each part. This will also provide an opportunity for an intermission for those who may need it during the two-hour lecture period.

L. F. Curtiss of the National Bureau of Standards, Washington, D.C., replied to my letter of September 23 about our participation in a program of comparative measurements for the radium content of radium ores and sludges. I asked Curtiss what method of analysis we should use in the comparison study. Curtiss replied that he assumed we already were set up for analytical procedures for radon but since we are not, to return the six samples to NBS.

Farrington Daniels sent Mulliken suggestions for several possible applications of "nucleonics," including high-temperature radiation chemistry, process radiation, radioluminescence, and weather modification applications. He suggests the development of a gas-cooled pile operating at temperatures up to 2000°C to obtain good thermodynamic efficiency. The high temperature and heat produced might be used to replace electric furnaces and might find other applications for producing chemicals such as calcium carbide, nitric oxide, phosphorus, and possibly even alcohols from carbon monoxide to provide automobile fuel. The intense radiation available in a pile would make possible the consideration of photochemical reactions on a large scale — such as the production of resinous materials from petroleum products and possibly even carbohydrates.

The ionization from pile-produced materials might be used with an electrostatic precipitator to remove dust and smoke. Daniels also suggests that intense radiation in the form of radioactive gases or particulates could be dispersed in the air to dispel fog or possibly even to induce rainfall.

Helen and I had dinner at Morton's Tavern with Muriel and Ted Mayer who are on their way to Spokane. Muriel is one of Helen's girlhood friends.

American troops launched a new attack and gained two miles in the Rhine push, according to today's newspapers.

Tuesday, October 3, 1944

This is the last day in Chicago for Dwight C. Lincoln, who is transferring to Hanford. His termination is effective Friday. Arthur LeFeuvre has transferred from Section C-I to C-V.

Manning asked Walter Bartky to modify our request to Lum of Monsanto for 10 millicuries of polonium to assure that the polonium will be free of RaD, RaE, and solid materials. The polonium provided should be in the form of a solution suitable for electrolytic deposition and of a type of preparation that contains tellurium rather than bismuth as an impurity. Manning also requested that a description of Monsanto's recommended method of polonium electrodeposition be obtained from Lum.

Although I was sick with a cold and a migraine headache, I attended the Project Council Information Meeting on Health, which met in Room 209, Eckhart Hall. Also in attendance were Allison, Anderson, E. S. G. Barron, Cole, Compton, Curtis, Dempster, Feld, Franck, Fulbright, Hamilton, Hill, Hogness, Jeffries, Langsdorf, Lichtenberger, Manning, McKinley, Miller, Nickson, Roberts, Spedding, Stearns, Stone, Tannenbaum, Vernon, Wakefield, S. Warren, C. J. Watson, W. W. Watson, Wattenberg, Whitaker, and Wirth. Following introductory remarks by Allison and Stone, Barron talked about the effects of x-rays and uranium ingestion on animals and their proteins. Ionizing radiation affects enzymatic systems which at certain doses cause diarrhea and, at larger doses, shock and death. Membrane permeability is affected probably because of the effects of free oxygen, produced by the ionizing radiation, on the phosphorylating mechanism. Studies that have been run on uranium and enzyme solutions in test tubes clearly demonstrate the inhibiting effect of small concentrations of uranium on enzymatic systems. A concentration of 2.6×10^{-4} gram atoms of uranium per mole of enzyme inhibits the action of urea by 50%. The reaction is found to be reversible with citrate.

Cole presented a description of tracer experiments on iodine inhalation that show only slight holdup in the lungs, small thyroid uptake, and high urine excretion. Fission product and plutonium inhalation studies will begin soon. Cole finds that Sr^{89} ingested in rats gives an LD-50 in 30 days at a concentration of 5-8 microcuries per gram of body weight.

Although elimination from the intestines is rapid at low doses, higher doses cause an increased percentage absorption from the intestines and a markedly increased deposition in the bone. Studies on the effects of Na^{24} in water on fish (as may be of concern at Site W) show that fish concentrate Na^{24} to the extent of 25 times the concentration found in their water environment. Cole said that gills show the highest concentrations. The effects of x-ray exposure show a chronic dose of 25 r/day is lethal to rats in 7-10 months. Six out of ten rats develop mammary tumors before death. At half this chronic dose (12.5 r/day), only one rat died at 7 months. At 10 months, eight out of ten rats are alive; but four of them have developed mammary tumors. (The malignancy of these tumors has not yet been determined.) In a control group only one out of 20 rats has developed mammary tumors. Cole said that mouse survival data following high energy photon exposure will be completed soon, hopefully before the cyclotron is shut down. Preliminary data show that 10 out of 16 mice have developed mammary tumors at 25 r/day for seven months. Stone said that an exposure of 8 r/day produces no more tumors than normal in "C_3H" mice (studied at Clinton). Stafford Warren commented that the significance of these types of experiments can be proved conclusively only by using a very large population of animals (under various exposure and environmental conditions).

Nickson reported on the health protection work at the Met Lab. The contamination problem in New Chem is still with us and remains essentially unchanged from the conditions reported a month ago. The West Stands contamination problem has resulted in closing the semiworks area and room 217 for routine use. Room 217 is used by Egan and others of the C-I group assigned to the semiworks in West Stands. Cooperation from the Chemistry and Technical Divisions has been good, and decontamination procedures are nearly completed. A hot-lab facility in Room B-2 of the Annex should help the New Chem problem. Installation of more remote control equipment will improve the West Stands situation in the future. Personnel monitoring has shown that the majority of overexposures are occurring in West Stands and in New Chem.

Hamilton reported on active smoke inhalation studies as related to oxides produced from $PuCl_4$, $Pu(NO_3)_4$, and plutonium metal dust. Excretion from the lungs takes place at a rapid rate with 89% of the original amount being eliminated in about two months. Studies at Berkeley on the uptake of Hanford wastes in soils have shown that both Clinton and Hanford soils are good both from adsorption capacity and moisture capacity. Under reasonable conditions, 93% of the waste activity is co-precipitated with the soils. (Hamilton said that if a pit were dug at Hanford 300 feet long by 30 feet deep and filled with 1,000,000 gallons of liquid material, the liquid material would stay in the vicinity of the pit indefinitely.)

Tannenbaum reported that work at Michael Reese Hospital shows that an injection of 1.0 mg of uranium compound is the LD-50 for mice in a few weeks period of time. Wirth of Clinton announced that film badge readings are now being made on persons with pocket chamber readings greater than 100 mr. This reduces the number of valid high readings to five or six a day. Studies run on samples of algae collected in the Clinton Laboratories settling pond show that algae may concentrate activities which have settled out and at a later time float to the surface

where the algae can agglomerate to produce radiation hazards exceeding in some cases 100 mr/hr. Precautions are being take to prevent this algae material from entering the Clinch River. However, definite radiation levels have already been detected at White Oak Dam.

Wirth also mentioned that the "hot run for Y" (to produce a batch of Ba^{140}) has been completed with only three persons being somewhat over-exposed. Curtis reported on Cohn's resin adsorption column work that has been successfully used after ether extraction to obtain a large number of fission products in carrier-free form for health studies.

In addition to this meeting, I attended the Project Council Information Meeting on Physics which met in Room 209, Eckhart Hall. Zinn described the 105B Hanford pile start-up problems. The initial 9 Mw start-up power level at 0.00088 excess $\underline{k}$ fell off to 3.7 Mw because of poisoning by a 9.4-hour Xe^{135} (the daughter of 6.6-hour I^{135}). With no slug or pile modifications, this poisoning effect will limit the full operating power of the Hanford pile to 216 Mw. A modification providing 1.85% excess $\underline{k}$ would completely override the poisoning effect by destroying the Xe^{135} by neutron absorption and should thus permit pile operation at any power level. The poisoning effect has not yet been oberved at Clinton It may be caused by resonance in the thermal neutron energy region.

H. L. Anderson reported on experiments concerning the thermal neutron (2200 meters per second) absorption cross section for plutonium in plutonium nitrate dissolved in deuterium nitrate, heavy water solutions, that was determined to be 1150 barns ± 3%. This result is in fair agreement with the value of 1050 barns obtained at Los Alamos. The fission cross section is 773 barns (alpha = 0.49). The resonance cross section is found to peak at 6730 ±240 barns. Anderson also gave values of 650 barns and 535 barns, respectively, for the thermal neutron absorption and fission cross sections of U^{233}. The cross section for U^{233} is higher than the cross section for U^{235}, which in turn is higher than for Pu^{239}, over the neutron energy range from 0.5 to 2900 ev.

"Chemical Research — Extraction and Properties of U^{233}, Report for Period Ending September 10, 1944," (CS-2165), was issued today. A summary of the contents is as follows. Studier, Hagemann, and Katzin have extracted 7.2 mg of U^{233} from two cans of thorium carbonate irradiated seven months at Clinton. The extracted material is 85% pure U^{233} with the rest being natural uranium. Four cans are undergoing extraction by the same ether-extraction method used for the first two cans.

Katzin, in cooperation with Dempster, Lapp, Crawford, Hufford, Jaffey, and Weissbourd, has determined that the specific activity of pure U^{233} is close to 23,300 disintegrations per minute per microgram, corresponding to a half-life of 1.46×10^5 years. The range of alpha particles for U^{233} has been determined as 3.38 ±0.01 cm in air. The fission cross section for cadmium-absorbable neutrons slowed in paraffin is 1.1 times that of U^{235}.

Hellman and Malm have conducted a study of ether extraction of uranium from thorium to determine the optimum conditions for the extraction and to obtain data for possible concentration procedures. A study of the distribution of thorium nitrate, uranyl nitrate, and nitric acid between

aqueous solutions and diethyl ether has shown the extraction of the uranium into the ether to be a function solely of the total nitrate concentration. Hyde has tested the usefulness of hexone for the extraction of uranium from solutions of varying concentrations of thorium nitrate, ammonium nitrate, and nitric acid. It is shown to be comparable to ether in effectiveness.

Van Winkle, Sedlet, and Katzin have tested three ore samples for protactinium. The material used was the carbonate precipitate produced during the extraction of uranium from its ores. The protactinium content ranges from about 0.4 to 1.2 parts per million of the dry precipitate, and the ionium makes up from 4 to 20 parts per million.

Katzin talked to members of the semiworks group and others today at 4:00 p.m. on "some interesting aspects of the Project."

Helen attended her chemistry class at YMCA College today.

Again war news on all fronts is favorable to the Allies except that the Japanese have landed on the China coast, periling Foochow.

Wednesday, October 4, 1944

A spill occurred today in Room 212 of West Stands, used by Larson, Hyman, Winner, and Fields. About 6 mg of plutonium in about 9 liters of solution contaminated about a 100 square foot area of the concrete floor. After initial cleanup the area has been temporarily covered with corrugated fiber board to permit use of the room. The Health Division was notified, and surveys will be made to determine what decontamination procedures may be required.

"Chemical Research — Separation Processes for Plutonium, Chemistry Division and Process Development and Chicago Semiworks Operation, Technical Division, September 15, 1944," (CN-2162), was issued. The joint report summarizes work on Hanford metal coatings, decontamination in the Bismuth Phosphate Process, and recovery of uranium from semiworks operations. Hyman, working on the removal of Hanford metal coatings reports that for all three coatings studied the most satisfactory method for removal involves treatment with a 15% NaOH solution containing from 10 to 20% $NaNO_3$. The Technical Division personnel have conducted three runs with the low-acidity modification of the Bismuth Phosphate Process; decontamination factors are about the same as in the unmodified process, but this could be due to operational difficulties. A uranium recovery process has been tested which involves precipitation of uranyl peroxide, dissolution in HNO_3, and extraction with ether or hexone using aluminum nitrate as the salting out agent.

I went home for lunch today. Later Helen had Wilma and Wilma's mother over for coffee.

The Project Council Policy Meeting for October was held today at 9:00 a.m. in Room 209, Eckhart Hall. In attendance were Allison, Bartky, C. M. Cooper, Daniels, Dempster, Franck, Hamilton, Hogness, Howe, Jeffries, Mulliken, A. V. Peterson, Spedding, Stearns, Stone, Szilard, Vernon, C. J. Watson, W. W. Watson, Whitaker, Wigner, Wirth, and Zinn. Allison announced that Compton flew to Hanford yesterday and that up until yesterday afternoon the Hanford 105B pile has operated twelve hours at 10 Mw. The xenon-poisoning phenomenon was discussed. Wigner estimated that the maximum power that could be attained at full Hanford pile loading (2000 tubes) for 1.2%, 1.8%, and 2.25% excess $\underline{k}$ would be 200 Mw, 400 Mw, and 800 Mw, respectively. It is possible that additional excess $\underline{k}$ can be had by making changes in slug canning, removing thimbles, etc.

Hogness said that Site Y would like the top people in Sugarman's Section III, but Allison recommended maintaining the present status until the pile poisoning problem has been solved. Allison said the Met Lab has lost about 175 technical people over the past two months with Chemistry being the big loser. Clinton has lost about 150 technical employees, primarily to Hanford.

Questions concerning cooperation with England ("Evergreen") were raised by Allison, in particular, whether or not the U.S.A. should give England information on the xenon poisoning phenomenon. W. W. Watson agreed there are problems and cited our specific directions to exchange all information on U^{233} chemistry but none on Pu^{239} chemistry, even though the "Evergreen" people obviously understand the relation of the U^{233} to the Pu^{239} chemistry very well. He said all evidence points to England pushing vigorously ahead after the war. Stearns noted that if England does emphasize nucleonics after the war and we do not, the U.S.A. will be at a serious disadvantage. He raised the question of whether we should wait for requests or should volunteer information to England. Both Peterson and Zinn spoke out in favor of volunteering needed information. Zinn said England will have the finest research pile in the world within a few years and we should keep on a good cooperative basis in order to obtain information on their results. Cooper suggested that a copy of the directive covering cooperation with "Evergreen" be inserted in the minutes of the Project Council Policy Meeting minutes. However, because of the wide circulation of the minutes, it was later decided to issue the directive, including further restrictions by McKinley, on a smaller selected distribution list.

Thursday, October 5, 1944

Walter Beard started his vacation today.

I wrote to Curtiss of the National Bureau of Standards stating that, in view of Curtiss' letter to me of September 29, we are returning the six radium ore samples. On another matter, I told Curtiss that we are

doing some work on a differential range selector and requested the circuit constant information for circuits described in reports A-1722 and A-1965 by Davis and Curtiss.

Kohman wrote to W. O. Simon, attention Willard, concerning heavy isotope problems at Hanford. In the 12-page memo Kohman lists all the isotopes in the heavy element region that can conceivably be produced in the Hanford piles in appreciable yields by known nuclear reactions. He discusses their possible importance in connection with the Hanford process and points out that many of these reactions are reviewed in my earlier reports, CK-514, MUC-GTS-690, and MUC-GTS-875, and in Weinberg's report, CP-1592. The probable production rates and effects of the following heavy isotopes are considered in Kohman's memo: thorium, uranium, and protactinium isotopes (easily separated and should have little effect); plutonium isotopes with mass numbers 238, 240, 241, and 242; neptunium with masses 236, 237, 238, 239, 240, 241, and 242; element 95 isotopes 238, 239, 240, 241, and 242; element 96 and higher — he states that it is rather improbable that two or more successive beta decays (beginning with a plutonium isotope with neutron capture possibly interposed) required to produce isotopes of 96 or higher, will occur to give yields of heavy isotopes that would affect Hanford operations.

Duffey sent Maloney and Tepe a suggested procedure for preparing the feed and "reflux" solutions for the 3-inch solvent extraction equipment under construction in the center stairwell of West Stands. The procedure has been reviewed with the Health Division and with Dawson and Stewart of Section C-I.

Helen went to her chemistry class at the YMCA College today. During the evening, Steve Lawroski dropped by to see us in our apartment.

The World Series is a local contest between the Browns and the Cardinals, both of St. Louis. According to today's front page, the Browns beat the Cardinals in the opener yesterday with a score of 2 to 1.

Friday, October 6, 1944

I read a copy of a letter from Zachariasen to Crandall of the Berkeley group concerning x-ray diffraction analyses of three samples of different kinds of thorium oxalates that Crandall sent him.

In a memo to the file I summarize the situation with respect to the disposition of J. G. Hamilton's RaD-Po solution. Hamilton claims he originally gave us 90-100 millicuries of RaD-Po material, but Livingston of Burton's group estimates that it was about 30 millicuries. The residues from Getz's operations on Hamilton's original material are in various containers in Room 212 of Kent Hall. Livingston has Getz's notebook and the key to this room.

Again baseball pushed war news from the headlines. The Cards beat the Browns 3 to 2 in the eleventh inning to tie the series in yesterday's game.

Saturday, October 7, 1944

Today was Henry R. Hoekstra's last day of service at the Met Lab. He is transferring to Hanford.

Health protection surveys were completed today on all the rooms in New Chem under the control of Section C-I. Slight to moderate spots of contamination were found on some of the hoods, benches, instruments, or floors from readings with Pluto (alpha-particle contamination) in Rooms 1, 2, 11, 27, 28, 33, and 34, and gamma-ray contamination was found in Rooms 4, 6, 28, and 31. These detected spots have been or are to be cleaned up. Improvements over the survey made a week ago were cited for Room 10 (Hindman and Kraus) and Room 30 (Bartell, Hoekstra, and Malm), but still more decontamination work is necessary. Hot solutions stored in a cabinet in Room 6 (Hagemann, Hellman, Studier, Hyde, and Wolf) gave "above tolerance" readings and required additional shielding. Dust and fume hoods in Room 27 (Beard, Goeckermann, Hopkins, and Shirley Nyden) and Room 36 (occupied by Kelley and Yett) gave off-scale Pluto readings on some surfaces and equipment within them and are to be cleaned. Off-scale plutonium readings of hot spots or working surfaces were detected in Rooms 33 (R. Thompson, Pye, and Katz) and Room 35 (Anderson, Asprey, and Stewart). A slight spill occurred today in Room 33 but has been adequately cleaned up following the survey.

Health protection surveys for the past week have also been completed for the rooms in West Stands under the control of my section. Room 217 (used by Egan and his semiworks group) was found to be adequately decontaminated with the exception of a sink and two electrical outlets which are to be cleaned or replaced.

I received a copy of Allison's memo to Captain McKinley requesting a ruling on what information can be given to D. F. Hewett, Staff Geologist, U.S. Geological Survey, concerning a survey USGS wishes to make of the heavy metal content in the rocks of the earth's crust. I had earlier informed Allison that we have information on this subject that should be of considerable help. Allison also requested information about who would be sponsoring the survey. It was suggested that if the survey is to be conducted under the auspices of the Manhattan District, time could be saved if Hewett were permitted to make a short visit to the Met Lab to receive the information.

Mulliken, as Chairman of the Project Publication Advisory Committee, has prepared a summary memo report (MUC-RSM-215) entitled, "Publication of the War-time Record as a Foundation for the Postwar Development of Nucleonics." The Project Publications Advisory Committee, appointed by Compton in August, will review the publication standards and advise on the preparation and publication of the proposed 10-15 volumes of survey articles and 25-30 volumes of collected research papers. The volumes should average perhaps 400 pages each and may be published later as Metallurgical Project Proceedings or Memoirs.

Helen worked on the classified "Table of Isotopes" at Crerar Library. She had lunch with Frances Chilson.

Again war news is all favorable to the Allies except in China where the Japanese are reported to be in the suburbs of Foochow. And in baseball news, the Cardinals beat the Browns 5 to 1 in the fourth game of the World Series today.

Sunday, October 8, 1944

I played golf in partly cloudy weather at Jackson Park with Helen, Katzin, and Hagemann. I shot a 92 for the 18-hole course.

The headlines today state that U.S. armored divisions "scored a major breakthrough north of Aachen today and moved unchecked across the open plain for Cologne." In political news we read "Dewey Again Hurls 'Red' Cry at FDR." The Cardinals blanked the Browns in the fifth game of the World Series today with a score of 2 to 0 and are now leading in the series three games to two.

Monday, October 9, 1944

Frederick D. Johnson transferred from Section C-I to the Physics Division.

The Personnel Office recorded a name change today. Dorothy Gottlieb, one of our secretaries (until recently with Stan Thompson, now with Fred Albaugh), has married Simon Black, who works on another war project here at the University of Chicago.

I received a memo from Albaugh concerning modification of the Bismuth Phosphate Process to recover 93 with the plutonium. He refers to data in a previous memo (MUC-GTS-1026) concerning the recovery of 93 using $H_2C_2O_4$ as the reducing agent which led to incomplete reduction of Pu(VI). Albaugh's group has since studied the efficiency of reduction on the decontamination cycle using both $H_2C_2O_4$ and Mn(II)-$H_2C_2O_4$ reducing reagents. They obtain a plutonium loss of 62.8% plutonium in the waste when 0.05 M $H_2C_2O_4$ is used and only 0.2% loss when Mn(II)-$H_2C_2O_4$ is used. Plutonium and 93 losses were measured when a run through a complete decontamination cycle was carried out with both 93 and 94 present. High losses for 93 occur, however, in the extraction step and are caused by iron contamination.

I wrote to Goldschmidt in Canada announcing that the report on our recent U^{233} work has been issued under Report No. CC-2165. I make several comments on the reported work, in addition to the points I made in my talk during my visit to the Montreal Project at the end of last month. I quote our more accurate value for the half-life of U^{233}, 1.46×10^5 years, based on our specific activity of 23,300 d/min/μg. I mention the as-yet unidentified insoluble residue we have observed when dissolving thorium metal in hydrochloric acid in light of similar experiments performed at Montreal and described by Goldschmidt during my visit there.

I also say much less Pa^{231}, extracted from uranium ores, is to be found in the soluble carbonate fraction than is reported in CC-2165. The problem is complicated by the presence of polonium in this fraction. I said it is now not at all certain that the soluble carbonate fraction will be a good source for Pa^{231}, and we are continuing to investigate other fractions from uranium ore residues.

Helen worked at the Met Lab on the secret version of the "Table of Isotopes."

The Cardinals won the World Series, defeating the Browns 3 to 1 in the sixth game.

Tuesday, October 10, 1944

Manning received memos from Nickson summarizing the health protection surveys made last week in New Chem and West Stands. Concern was expressed with procedures now being used by Egan's semiworks group in West Stands to dispose of active wastes. Nickson points out the problem of potential contamination of areas outside the Project that could result from the discarding of plutonium and by-product solutions down the sink drains in Room 217 (used by Egan's group) and also in Room 216 (occupied by Blaedel and Walling). Nickson suggests that the problem of waste disposal and the need for additional disposal facilities should be discussed with Egan and Gilbreath.

Charles Cooper presented a discussion on "New Goals for the Metallurgical Project" at the meeting of the Tolman Postwar Policy Committee today. Cooper points to the possibility that our job may soon, if not already, be completed with regard to assuring successful W operation. He asks, "with this prospect before us, is it not our immediate and major responsibility to obtain agreement upon a continuing course of research which must be pressed if the United States is to maintain the lead in the field which we presume it now possesses?" Cooper summarizes the current Project status as follows:

> By some two years of great effort under emergency conditions we have arrived at a situation which may be summarized thus. A process has been developed which very certainly will produce quantities of 49 adequate for experimental purposes and probably sufficient to meet situations immediately foreseen. The present process is very complicated and unduly expensive, both as to manpower requirements and the materials which are involved. The present approach is not adaptable to generation of power nor is it designed to manufacture or recover effectively possible by-products of operation. The product will be contaminated with undesired isotopes which could possibly be eliminated if other methods, as yet only sketchily developed, were to be employed. We cannot be entirely certain until final trials have been made that the product will be as useful as anticipated, and it may well develop that other elements or isotopes will turn out in the end to be much more useful than the one we have prepared to manufacture.

In order that our country may be as well prepared as possible, it would appear from the above that research should be pressed along the following lines.

(1) All possible effort should be made to discover what material and isotope is most promising for our present ultimate use.

(2) As soon as point (1) can be settled, the development of adequate manufacturing facilities for the chosen substances should proceed.

(3) While the developments indicated under (1) and (2) above are in progress, studies aimed at improving the country's position with respect to production of the present isotope and studies aimed at the production of power and of reaction by-products should be vigorously proposed.

Cooper points out that the development of usable electrical power from nuclear reactions "poses a problem much more difficult technically than the one which we have had to solve in connection with our present manufacturing objectives." Many of the problem areas are identified such as high temperatures, corrosion resistance, the need for new materials and alloys for fuel fabrication, structural purposes, and shielding. A wide variety of engineering, scientific, and manufacturing organizations will be required. The Met Lab has many of the facilities and scientific and engineering personnel required, but Cooper points out it is not possible to attract or even hold able persons in the Project because of the future being so uncertain. He says unless the Project "can be given a greater degree of certainty by a new directive indicating clearly new objectives, it will hardly be possible to maintain even our present working organization much beyond the first of the year."

Helen went to chemistry class at the YMCA College.

"Yanks Pin Nazis in Aachen" reads today's top headline.

Wednesday, October 11, 1944

General Groves and his secretary, Jean O'Leary, are in Chicago today. Groves is conferring with individuals and groups in the District Area office and the Met Lab.

I met formally with Miles Leverett of the Technical Division at Clinton, together with Maloney and Miller of the Met Lab Technical Division and Albaugh, Manning, and Orlemann of my section, to discuss and correlate the work on uranium recovery at Clinton and the work to be undertaken here. On Monday Colonel Nichols sent a letter to Compton requesting comments as to how a program of recovery of uranium from plant wastes could best be carried out at Clinton and at the Met Lab. Following our meeting I prepared a statement for Hogness for use by Allison in answering Nichols' request. This statement summarizes today's meeting with Leverett as follows:

"It was tentatively decided that (1) the Technical Division here would not participate in the program until a promising process is uncovered by the chemists; (2) the efforts at Clinton for the present will be directed principally toward recovery of metal now accumulating in waste storage at Clinton; and (3) the work at Chicago will be directed entirely toward a solution of the Hanford problem where the decontamination which must be achieved will be a hundred-fold greater than at Clinton. In work on the Hanford problem, first emphasis will be placed on methods for recovery from neutralized wastes rather than directly from extraction supernatant since it is unlikely that any process can be ready for Hanford before the accumulation of waste metal becomes large.

"Mr. Leverett stated that some work on uranium recovery is now under way at Clinton. In our section here at Chicago one man is now working on the problem, and we plan to assign two additional men to this work in the near future. Discussion of the work indicated that there is now no overlapping between the work here and at Clinton. It was agreed that close contact should be maintained between the two groups.

"Aside from the primary overall problem of economically achieving sufficient decontamination, the most immediate problem appears to be to find ways of overcoming the interference of sulfate and phosphate ions which are present in the neutralized waste solutions and which complex the uranium. It is probable that a solvent extraction step will be an important part of any process which is worked out. It is our plan in part of the work at Chicago to attempt to separate sulfate and phosphate from the uranium by precipitation either of the uranium or of the interfering ions prior to solvent extraction. 'Scavenger' precipitation and solvent extraction may contribute toward the relatively high decontamination factor which will be required for the Hanford metal. According to Mr. Leverett, present efforts at Clinton are being directed principally toward direct solvent extraction including the use of high concentrations of salting out agents such as iron salts (which complex phosphate) to overcome the complexing of uranium by sulfate and phosphate. This procedure without any precipitation step would seem to be more feasible for Clinton metal, where less decontamination is needed, than for Hanford metal."

Truman P. Kohman is spending some of his terminal leave from the Met Lab taking care of personal affairs prior to leaving for Hanford next week.

Mulliken, as Chairman of the Project Publications Advisory Committee, has prepared another memo report (MUC-RSM-221) on the "Current Status of Tentative Plans for Metallurgical Project Record," giving the general purposes and plans, scope and contents, and organization of the work for the Project Record. The major portion of the final record should be completed by the date of termination of the Project, and the entire work should be completed by a few months thereafter or around January 1, 1947. It is estimated that about 40 man-years of technical grade effort will be required during 1946 to complete the task of writing and compiling the 40 volumes of 400 pages each now called for in the Tentative Outline for the Project Record. Of this the Met Lab Chemistry Division contribution called for will be about seven technical man-years. Final editing will be done by the Project Information Division.

At 7:45 p.m. I attended the meeting of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I, in Room 209, Eckhart Hall. Also attending were Cunningham, Daniels, Davidson, Dawson, Dixon, Fields, Florin, Fried, Ghiorso, Hagemann, Howland, Hufford, Jaffey, James, Jasaitis, Jones, La Chapelle, Lawroski, Magnusson, Malm, Manning, O'Connor, Orlemann, S. Peterson, Phipps, Stewart, Studier, Westrum, Kraus, and others. Westrum reported on the preparation of plutonium nitride using either the reaction of plutonium metal with NH_3, or $PuCl_3$ with NH_3 at 1000°C (it was identified as PuN by x-ray diffraction).

James reported his attempts to isolate and identify heavy isotopes from a target which was bombarded at Berkeley with 16 Mev deuterons some time ago. Isotopes 93^{236} and 93^{235} were looked for as the reaction products of deuterons on U^{235}. To separate 93 from 94, nearly 25 bromate cycles were carried out. After the twentieth cycle, the remaining 200 alpha-particle counts per minute (of the original 6×10^6 c/m behaved more like 93 than 94. Range measurements, using microabsorbers with the nitrogen counter, showed that the range of these alpha particles indicates an isolated material different from 93^{237} (range 3.22 cm), 94^{238} (range 3.97 cm), and 94^{239} (range 3.69 cm). Although the number of counts is low, the range data seem to indicate the presence of two new isotopes. James hopes to identify these isotopes by examining the daughter protactinium isotopes which should grow from them. At the present, the conclusions are very tentative.

Hindman summarized the spectrographic and potentiometric information on plutonium thus far determined through our work. Spectra of Pu(III) in $HClO_4$, HNO_3, HCl, and H_2SO_4 are available; there is some evidence for complexes with chloride and sulfate ions. Spectra are now available on Pu(VI) in perchlorate, nitrate, sulfate, and chloride solutions. A number of complex ions of Pu(IV) are formed with chloride, nitrate, and sulfate ions of various concentrations. Spectra of Pu(VI) in 1 M HNO_3 and 1 M $HClO_4$ have been taken. The Pu(VI) ion is complexed in 1 M HCl and 1 M H_2SO_4. Diagrams were presented by Hindman of the potentials of the various couples in 1 M HCl (in the presence of 1 M NaCl), and in 1 M HNO_3.

Helen worked at the Met Lab on the secret version of the "Table of Isotopes."

From Pacific Fleet headquarters come reports that American carriers destroyed 89 Japanese planes.

Thursday, October 12, 1944

Fowler R. Yett is our latest transferee to Hanford. He is expected to report October 28.

Following through on his decision of September 25 that sample 49DD-14 (derived from the first series of dichromate oxidation cycles, plus one silver plus persulfate oxidation cycle on the original leaching of plutonium from the 200 mg plutonium plus deuterons bombardment at the St. Louis

cyclotron) offers the best prospects for isolating element 95, Morgan dissolved the sample from its counting plate and carried out another silver plus persulfate oxidation. The subsequent lanthanum fluoride precipitate from this oxidized solution (labeled 49DD-29) gave 88 c/m.

I read a copy of a memo from Duffey to Burton which included a copy of a reply from D. C. Lewis of Carbide and Carbon Chemicals Corporation, to Duffey's letter of last Friday. In the letter Lewis states that George Jones of the Bureau of Mines advises that the explosive limits of hexone in air are 1.35% and 7.6% by volume, determined at 20°C and 50°C, respectively. Lewis had no information on the reaction of hexone with nitric acid. Duffey wrote in his memo to Burton that a Mr. White of C&CCC has suggested Shell Oil Company as another source of information on hexone characteristics in various environments, and Duffey has now written to Shell.

I wrote to Whitaker of Clinton about the possibility of separating Np^{237} from Clinton process solutions. Several milligrams of Np^{237} are potentially available in each daily batch in the separations plant but are now lost. I include information on our simple modification in the Clinton separation process which would materially improve the yield of Np^{237} without interfering with the yield of Pu^{239}, namely, our method of using oxalic acid plus Mn(II) instead of ferrous ammonium sulfate as reducing agent just prior to the product precipitation step in the bismuth phosphate decontamination cycle. I suggest that the modification be introduced into the Clinton procedure in two of the sixteen half-ton "production batches" to be run during the interval October 20 to November 10. I suggest that we send a man or two to Clinton to help separate the Np^{237} from the Pu^{239} in connection with the peroxide isolation step.

In a letter to A. H. Voigt of Iowa State College, Albaugh requests the name and address of the commercial supplier or the procedure for synthesizing N-N-bis-(2-hydroxy)-ethylene-diamine for use in the analysis of Pu(III) and Pu(IV) states as described in the Ames report CN-1777. Albaugh also enclosed a copy of a September 11 memo from Blaedel to Walling (MUC-GTS-996) about the status of solvent extraction-decontamination as of September 7, 1944.

The Chemistry Division issued its "Summary Report for September, 1944" (CS-2229). As usual, contributions from the following sections are included. Section C-I — Plutonium Chemistry (G. T. Seaborg, Section Chief); Section C-II — Radiation Effects (Milton Burton, Section Chief); Section C-III — Fission Product Chemistry (N. Sugarman, Section Chief); Section C-IV — Analytical (D. S. McKinney, Section Chief); and Section C-VI — Pure and Rare Chemicals (L. B. Arnold, Jr., in charge). The work of my section falls into three parts: Separation Process Studies, Basic Chemistry, and Uranium-233 work. (Our work in each of these areas is described in greater detail in other reports.) This report briefly summarizes investigation in these three fields as follows:

Separation Process Studies

Bismuth Phosphate Research — General Problems. Experiments on extraction from 28% UNH at Hanford concentrations of plutonium. Study of

by-product precipitations at low acidities — poor decontamination factors obtained. Study of sedimentation characteristics of the by-product precipitate formed in process solution. Preliminary experiments on carrying of U(IV) on bismuth phosphate from H_2SO_4 solutions — very little is carried. Carrying of Pu(III) and Pu(IV) on zirconium phenyl arsonate — Pu(IV) carries to extent of 93-95%, while Pu(III) carries about 5-10% using tracer quantities where it is hard to maintain a mixture of the two valence states. (Bartell, Bradt, Greenlee, Howland, Larson, S. Peterson, Winner)

Pre-extraction Treatment of Process Solutions. Spectrophotometric studies show that 0.001 M N_2H_2 reduces Hanford concentrations of plutonium to the +3 state, and under these conditions about 50% carrying is obtained from UNH solution. (Ader, Bartell, Hoekstra, Malm)

Neptunium-Plutonium Separation. Neptunium in tracer amounts is shown to be oxidized by 16 N HNO_3 in one hour at 95°C. (R. Thompson)

Alternate Precipitation Methods for Extraction-Decontamination of Plutonium. In four runs, successive plutonium reprecipitations have been made following the extraction precipitation with varying amounts of H_2SiF_6, Fe(II), HF, H_2CrO_4. (Greenlee and S. Peterson)

Isolation at Hanford — Peroxide Method. Process has been modified to give granular filterable precipitates in the presence of high quantities of iron, the modification including increasing acidity to 2.0 N HNO_3. (Beard, Goeckermann, Hopkins)

Concentration at Hanford — Fluoride Method. Removal of iron from process solutions by dissolution of the lanthanum fluoride precipitate in 10 N HNO_3 and reprecipitation is demonstrated. Behavior of zirconium in the concentration procedure has been studied with 65-day zirconium. (J. Katz, A. Kelley, Malm, Walling, Wolf, Yett)

Concentration at Hanford — Early Operation. The feasibility has been demonstrated of precipitating lanthanum fluoride directly from 10 N HNO_3 solutions of bismuth phosphate at Hanford concentrations of plutonium. (Haeckl)

Metal Coatings Removal. Hyman's research, showing that the most satisfactory method for removing the various coatings from slugs proposed for Hanford involves treatment with a 15% NaOH solution containing from 10% to 20% $NaNO_3$, is reported.

Evaluation of Process Modifications. The low acidity Bismuth Phosphate Process has been further investigated with two-liter scale runs — results are poorer than in previous runs. Two 100-ml scale runs investigating the distribution of neptunium in the Bismuth Phosphate Process show that the use of uranous sulfate or oxalic acid as reducing agent instead of ferrous iron in the product precipitation step reduces the loss of neptunium from 75% to 30%. (Ader, Larson, Lincoln, Winner)

Solvent extraction and Decontamination Methods for Plutonium. Preliminary experiments have shown it may be feasible to have a solvent extraction, decontamination, and concentration method for plutonium involving variation of the oxidation state of plutonium between +3 and either +4 or +6 to allow alternate transfers of plutonium between aqueous and non-aqueous phases. Hexone seems to be a satisfactory solvent. (Blaedel and Walling)

Recovery of Uranium from Process Waste Solutions. Previous work is surveyed, and a few scouting experiments are reported. (Hyman)

Hexone Extraction for Isolation-Development. A series of runs has been made with hexone and feed solution containing tracer plutonium in the 19-mm continuous countercurrent solvent extraction column. Overall plutonium recovery was in excess of 99.5%. There have been no operating difficulties. Construction of the 3-inch full plant size extraction unit of pyrex pipe has been completed, and tests will be initiated in the near future. Plans are to construct later a unit at Clinton of similar size but made of stainless steel. (Brody, Lawroski, Reinhardt)

Solvent Extraction — Fundamental Research. Distributions of iron, lanthanum, and zirconium between Hanford feed solutions and hexone have been measured. (Lawroski, Reinhardt, Stein)

Basic Chemistry

Vapor Pressure of Plutonium Compounds.

$PuCl_3$: new measurements give

$$\log P_{liquid} = 9.233 - 12{,}550/T$$

$$\log P_{solid} = 12.503 - 15{,}840/T$$

Heat of vaporization (liquid) = 58,800 cal/mole

Heat of vaporization (solid) = 72,500 cal/mole

Pressure, P, at melting point = 5.75×10^{-4} mm of Hg

Entropy of fusion = 13.6 entropy units

Melting point = 733°C

ΔH fusion = 13,700 cal/mole

The apparatus used for $PuCl_3$ is being modified for measurements on $PuBr_3$. (Johnson, Sears, Seifert, Simpson)

Preparation and Properties of Plutonium Compounds by Dry Reactions. PuN has been prepared. PuO_2 has been decomposed on heated tantalum in vacuum to give a phase identified by Zachariasen to be Pu_2O_3. The previous identification by Zachariasen of PuS is now considered uncertain. (Westrum and Abraham)

Preparation and Properties of Plutonium Halides. $PuCl_3$ has been successfully prepared from PuO_2 in a vapor phase of H_2, HCl, and H_2O at 650°C. Previously it was reported that under these conditions only PuOCl is obtained. The compound is found to be stable and non-volatile in high vacuum at 1200°C. (Sheft, Fried, Robinson, Abraham)

Refractory Studies. Further metal reduction studies have been conducted in Berkeley sulfide crucibles, but interpretations are not complete. (Westrum, Hellman)

Transference Measurements. The work of last month has been extended. New data indicate Pu(IV) in 2-4 M HCl is positively charged — transformation to negatively charged complex ions occur at 4-10 M acid. Absorption spectrum measurements indicate three ionic species may be formed in these HCl solutions. (Dixon, McLane, Ames, Hindman)

Hydrolytic Behavior of Plutonium. Spectrophotometric changes accompanying hydrolysis and "polymerization" of Pu(IV) are being studied in detail. (Kraus)

Mechanism of Co-precipitation. Experiments are in progress on the mechanism of co-separation of Pu(III) and (IV) with bismuth phosphate. (Howland)

Isolation and Study of 93^{237}. The specific activity and half-life of 93^{237} have been determined by weighing NpO_2 and radioactive assay of the weighed sample. The specific activity has been found to be 1517 disintegrations/minute/microgram, and the calculated half-life is 2.21×10^6 years. (La Chapelle, Magnusson)

Chemistry of Uranium. X-ray analyses of precipitates found in titrations of UO_2^{++} with alkali indicate that more than one solid phase may have been formed. (O'Connor)

Heavy Isotopes by Bombardment of Pu^{239}. A larger target than those previously used has been bombarded with deuterons at St. Louis. Chemical separations are in progress. (James, Florin)

Routine Separation of 93^{239}. Continued at ten-day intervals by Fields.

Hexone Extraction for Isolation Research. Influence of acid, salt, and plutonium concentrations on distribution ratios of plutonium between column feed and solvent have been determined. Laboratory-scale batch extractions from zirconyl-complexed lanthanum fluoride have been made. (Britain, Fineman, Fields, Stewart, Dawson)

Neutron Counting with BF_3 *Pressure Ion Chamber.* Attempts to coat ion chamber surface with carbon black to reduce alpha contamination have failed. (Weissbourd)

Alpha-Particle Range Measuring Instruments. Continued use is being made of the alpha-particle range measuring instruments in connection with the search for new heavy isotopes. (Weissbourd, Jaffey, Crawford, Hufford, Walsh, Kohman) (a) A low geometry aluminum foil range chamber is being used for routine measurements. (b) A low geometry aluminum foil range chamber with magnetic field has been constructed. (c) Two 50% geometry mica absorption nitrogen-filled range chambers are being used extensively in these investigations because of [1] their successful operation and [2] their high resolving power for a mixture of alpha-emitting species. (d) The differential range chamber is being used to investigate the alpha-particle radiation from U^{233}. The range of the U^{233} alpha particles has been accurately determined as 3.38 cm by John Crawford. (e) A differential pulse height selector has been constructed for use with the existing spherical range chamber, and is being tested.

Plutonium-Finding Alpha Counter. An improved flat-type proportional counter has been designated and will be built by the shop. The instrument is being tested. (Scott)

Nitrogen High-Resolving Power Alpha-Particle Counter. Another circuit is being built. Two new chambers have been designed. (Krueger, Kohman, Jaffey)

Measurement of Fission Properties of Pu^{240}. Measurements are underway by Weissbourd.

Work of U^{233} Group

Extraction of U^{233}. Four more cans of carbonate have been extracted (in addition to the first two). There are approximately 4 mg of U^{233} per can. (Hagemann, Studier)

Survey of Solvents for Extraction of U^{233}. (Hyde, Wolf)

Methods of Concentration of U^{233} Following Extraction. (Hellman and Wolf)

Determination of Physical Constants for U^{233}. The latest values of the specific activity and corresponding half-life of pure U^{233} are 23,300 disintegrations per minute per microgram and 1.46×10^5 years. The range of the alpha particles is 3.38 ± 0.01 cm at standard conditions. The fission cross section for neutrons slowed by paraffin and absorbable by cadmium is 1.1 times that for U^{235}. Checks on material from the Montreal Laboratory corroborate the half-life determination. (Instruments Group)

Methods of Dissolving Thorium Metal. (Katzin)

Analysis of Ore Samples for Protactinium. Isolation of Milligram Amounts of Pa^{231} and Ionium. First steps toward isolation have been taken. (Van Winkle, Sedlet)

I played in the final of the Met Lab Golf Tournament against R. Parker at Jackson Park. Al Ghiorso played along with us. Parker beat me match play with a long birdie putt on the long fourteenth hole (560 yards par 5), 5 and 4. I shot 90, Parker 75, and Ghiorso 102 for 18 holes. I am being awarded a wallet engraved with the designation "runner-up" in recognition of second place in this laboratory-wide golf tournament.

Helen went to chemistry class at the YMCA College.

According to today's paper Soviet troops have captured the second largest city in Hungary — Cluj.

Friday, October 13, 1944

Kohman sent a memo to W. P. Jesse of the General Physics Division about the status of the "carbon coating problem" for lining ion chambers with carbon films to eliminate alpha-particle counts from contaminant wall materials. Since Kohman is leaving the Met Lab, he suggests that Jesse may wish to authorize Weissbourd in Ghiorso's group to continue these experiments.

Kohman replied to a letter that J. H. Lum, Central Research Department, Monsanto Chemical Company, wrote to Hogness requesting a new design 3.5-inch diameter neutron ion chamber for testing. Arrangements have been made through Manning to send to Lum one of the new chambers together with a paraffin-filled moderator can.

Technical Council Members received a memo from Stearns enclosing a list of rules for the interchange of information between the Evergreen Area (the Canadian-British group at Montreal) and the U.S.A. Project groups. Three of these rules of concern to me are the following: (a) "Information connected with the transformation of thorium to element 23 and the separation and measurement of the physical and chemical properties of thorium and 23 will be freely interchanged." (b) "Information necessary for the guarding of the health of the operators at the Montreal Plant will be exchanged. Information concerning the toxic effects of either 49 or fission products will not be included in this exchange." and (c) "Information concerning the chemistry of element 49, the method of separating 49 (including all engineering details), and the purification together with the chemistry of the fission products will not be transmitted until further instructions are issued."

"Yanks Win Aachen Suburb" reads the top headline of today's newspaper.

Saturday, October 14, 1944

Truman P. Kohman terminated from the Met Lab today and will report to Hanford on Wednesday. He will work in Building 3706.

Kathryn Buehler, who is going to return to school, and Ellen Watts also terminated today.

Health protection surveys were completed for the past week. Air samples taken throughout the week in nine rooms used by Section C-I were all below "tolerance levels." Alpha- and beta-particle, gamma-ray contamination, and radiation level surveys were also made and showed low levels of alpha-particle contamination in Rooms 10, 11, 12, 13, and 33, and gamma-ray contamination in Rooms 6, 12, 13, and 34. Because of contamination of some of the benches and floors, moderately high alpha-particle radiation levels were found on some work benches, hoods, and lab equipment in Rooms 27 (used by Beard, Hopkins, and Goeckermann), 35 (used by Stewart, Asprey, and Anderson), and 36 (used by Kelley and Yett). It was requested by the Health Division that the contamination in these rooms be cleaned up at once.

I replied to Hogness's request for a report on the recent informal discussions with Leverett of Clinton Labs concerning the uranium waste recovery problem. My memo contained the statement I had prepared following the meeting with Leverett last Wednesday.

Jaffey wrote an informal letter to Willard at Hanford enclosing a copy of two articles published in the *Review of Scientific Instruments* on the use of the FP-54 electrometer amplifier. He also sent instructions on the setting up and use of the FP-54 circuit and ionization chambers.

Hogness requested that Bartky obtain about 200 mg of plutonium metal from Los Alamos to be used for future vapor pressure measurements

and basic chemistry work on plutonium and its compounds. Since the Met Lab no longer works on plutonium metal production problems, it has been agreed that any plutonium metal we may need will be supplied by Site Y provided we return an equivalent amount of plutonium in the form of a useable compound.

Helen spent the day at home.

Banner headlines today read "Formosa Toll: 396 Planes" and "Win Riga; Reach Belgrade"; a U.S. carrier force attacked Formosa, destroying 396 aircraft. Soviet troops captured Riga on the Baltic and in the south have penetrated to Belgrade, the capital of Yugoslavia.

Sunday, October 15, 1944

In 52° temperature Helen and I played 18 holes of golf with Fred Albaugh and Edrey Smith at the Pipe O'Peace Golf Course located at Halsted and 131st Streets. Fred shot 136 and I shot 95. We all had dinner at the home of Edrey's parents, the Royal B. Smiths, who live at 9514 South Damon Street. After dinner the four of us played miniature golf at Beverly Hills Putting Range located at 95th and Claremont Streets. Our scores were ES-60, HS-53, FA-48, and GS-40.

Today's headlines read "Hungary Deserts Hitler" and "The Hungarian government has accepted Allied armistice terms and Hungary will become a co-belligerent in the war against Germany." Other news states that British troops have occupied Athens. Yesterday's sports news reports that Ohio State beat Wisconsin, 20-7.

Monday, October 16, 1944

Walter Beard returned to work this morning after a vacation of a week and a half.

L. F. Curtiss of the National Bureau of Standards sent me the information I requested in my letter of October 5 about the circuit constants and characteristics for an ionization chamber amplifier.

I received a copy of a memo from Zachariasen to Allison giving his analysis of two samples, submitted a few days ago by Katzin, consisting of residues resulting from Katzin's attempts to dissolve thorium metal (obtained from Ames Laboratory) in HCl. The x-ray diffraction patterns show the presence of about 25% thorium nitride and 75% ThO_2. It is suggested that the ThN_x could have resulted from making or remelting the thorium metal in nitride crucibles at Ames.

I sent an outline for my October 24 evening lecture to A. A. Frost of Northwestern University.

I replied to the letter I received from Paul Aebersold (with the Manhattan District U.S. Engineers Office in Oak Ridge) on September 7 and enclose copies of three final reports, MUC-GTS-1066, MUC-GTS-1067, and MUC-GTS-1068, which Aebersold requested. These three reports, I inform him, summarize work under the three contracts, NDCrc-197, NDCrc-201, and OEMsr-206, respectively, and account for all of the technical reports he listed in his letter with the following exceptions: the work described in Report A-44 was not done in connection with any of the three contracts as far as I know; work described in A-174 was performed in connection with the big Berkeley Project that had its inception about January 1942; work reported in A-135, A-136, and A-152 was largely my graduate student research and not under the auspices of any contract; and we have not been able to account for A-173 as we have no record of this report. The contents of these reports are as follows: Report MUC-GTS-1066 briefly describes work conducted under Contract NDCrc-197 relating to the search in nature for elements 94 and 93. The experiments and results are covered in more detail in Reports A-146 and CN-246. Report MUC-GTS-1067 describes work on the continued study of the fission properties of Pu^{239}. It includes work on the demonstration of the slow neutron fission of Pu^{239} (reported in more detail in Report A-33) which was largely completed before Contract NDCrc-201 went into effect; work conducted under this contract relates to the measurements of the fast neutron fission cross section of Pu^{239} (reported in Report A-22) and of the spontaneous fission of Pu^{239} (reported in Report A-68).

Report MUC-GTS-1068 reviews the investigations conducted under Contract OEMsr-206 related to measurement of the fission properties of U^{233}, Np^{237}, and Np^{239}. The discovery of U^{233} and measurements of its radioactive properties and its slow neutron fission are described in Reports A-153 and A-192. The discovery of Np^{237} and the demonstration that it does not undergo fission with slow neutrons is covered in Report A-151. Contemplated work on the fission properties of Np^{239} was never done. The work on the preparation of U^{234} through the extraction of UX_1 (reported in A-172), the search for spontaneous fission in U^{234} (reported in A-268), and the measurement of the fast neutron fission cross sections of Pu^{239} and U^{233} (reported in A-269) were also done under Contract OEMsr-206.

Katzin, Hindman, Davidson, Cunningham, and I attended a meeting to outline a program of research on protactinium: 5 mg are being purchased for special studies. X-ray diffraction and spectrographic methods for determining the purity of the purchased protactinium material were discussed. It was concluded that peroxide or oxalate precipitation may be required to separate the protactinium from zirconium and tantalum, the most probable impurities. Separation from thorium can be achieved by precipitating the thorium with HF in the absence of K^+.

It was recommended and approved that the immediate research program objectives should be: (a) to attempt the preparation of oxidation states other than Pa(V) using both wet [Hindman] and dry [Davidson, Westrum, Fried] reactions; (b) to attempt to identify the ion species in 0.5 M H^+ [Hindman] by studying hydrolytic behavior of $PaCl_5$ (to be prepared by Davidson), or by preparing and determining the structure of

salts precipitated or crystallized from 0.5 M H^+; (c) to accurately determine the specific activity and half-life of protactinium [Magnusson and La Chapelle]; (d) to determine the structure of a limited number of typical protactinium compounds [Davidson, Westrum, Fried, Hindman]; (e) to conduct a limited survey of complex ion formation using the transference method [McLane); and (f) to make a selected number of solubility determinations of typical compounds such as K_2PaF_7, protactinium oxalate, and protactinium peroxide [Hindman, O'Connor]. Care will be taken to avoid contamination of plutonium with protactinium and vice-versa. Protactinium recovery methods were discussed, but no final decisions were reached except that all agreed an adequate recovery scheme must be included in our protactinium program.

Nickson requested information from Stafford Warren about the protective measures that may be in use at other Manhattan District projects for handling polonium in order that adequate health protection procedures might be established at the Met Lab for this material.

Helen worked on the classified "Table of Isotopes" at the Met Lab.

Hungary has asked the Allies for an armistice, according to today's newspaper.

Tuesday, October 17, 1944

Since last Thursday Morgan has carried out a further silver ion plus persulfate ion oxidation cycle on sample 49DD-29 (derived from a series of dichromate and silver plus persulfate oxidation cycles on the original leaching of plutonium from the 200-mg plutonium plus deuterons, St. Louis cyclotron bombardment). He then made a lanthanum fluoride precipitation from the oxidized solution and mounted it for counting as sample 49DD-31. A range determination (Fig. 26) was made by the mica absorption technique that showed 30 c/m of alpha particles, all having a 4.05 cm air equivalent range. (The range for Pu^{239} alpha particles is 3.7 cm.) Morgan today recorded the following observation in his notebook (No. 727B): "Among the heavy isotopes (Z = 80-94) Ac, Th, and Pa might follow the observed chemistry. This activity would have to be a new isotope of these elements, however, since none of the known ones would have this range, or behave in this way (growth, decay). Element 95 would also be expected to have this chem(istry)." This seems to be very good evidence that we have synthesized and chemically identified an isotope of element 95. The hypothesis that element 95 should be difficult or impossible to oxidize to a higher (fluoride-soluble) oxidation state seems to be borne out.

I attended the 9:30 a.m. Project Council Information Meeting (Chemistry) in Room 209, Eckhart Hall. Also attending were Aebersold, Allison, Arnold, Burton, Chipman, Compton, C. M. Cooper, Daniels, Davies, Dempster, Doan, Elliott, English, Franck, Greager, Huffman, Hogness, Jacobson, Jones, W. C. Johnson, Manning, McKinney, Mulliken, Newton,

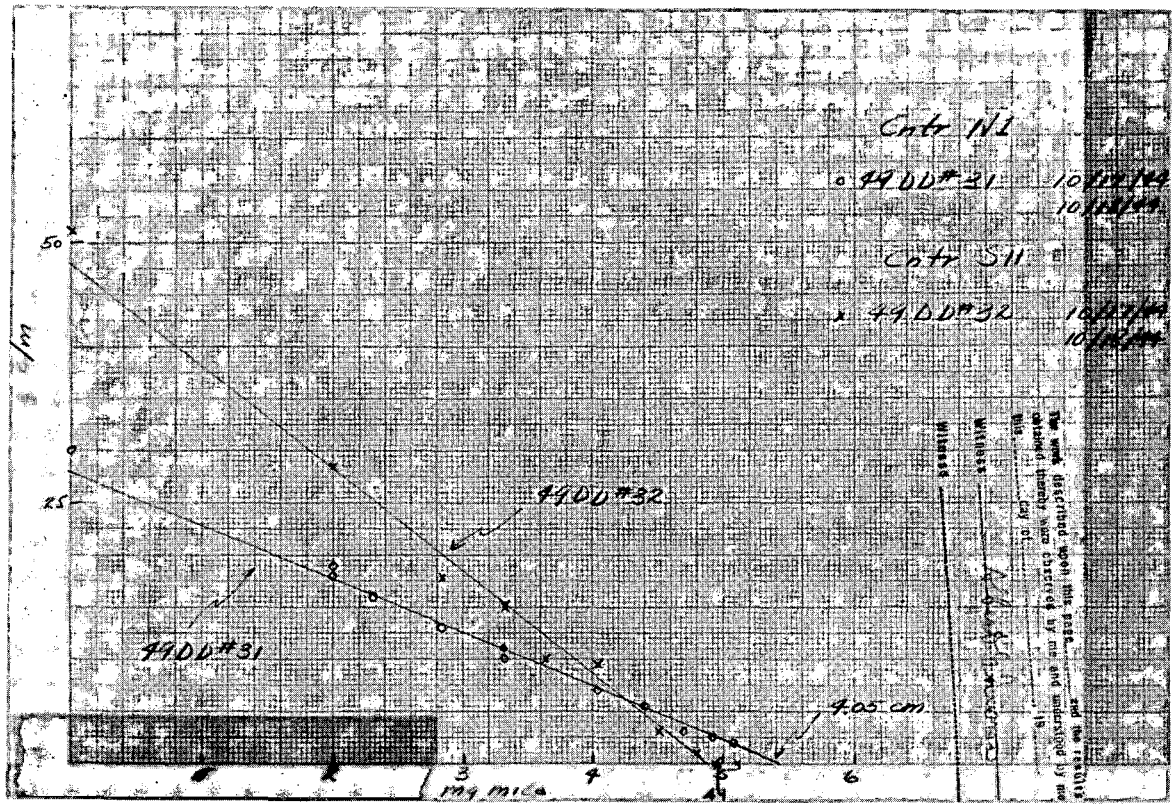

Figure 26. Mica absorption data showing presence of 95^{241} alpha particles in deuteron-irradiated plutonium, October 17, 1944. 49DD#31 range curve for 95^{241}; 49DD#32, comparison range curve for Pu^{239} fraction.

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Nickson, Quill, Spedding, Stearns, Stone, Sugarman, Turkevich, Vernon, Wakefield, Warf, Warner, Watters, Whitaker, Wigner, and Zachariasen. Following opening remarks by Allison, Hogness introduced the Met Lab speakers. My coverage of the Section C-I program included our work on Hanford problems under Albaugh and on basic chemistry under Cunningham. I indicated that preextraction treatment is considered the main problem in connection with the Hanford work. The carrying of Pu(III) by bismuth phosphate is not so good as the carrying of Pu(IV). The carrying is not too good in the presence of UNH. Hydrazine (N_2H_2), probably formed during solution of the metal, seems to reduce plutonium to Pu(III). The effect of N_2H_2 causes a denser bismuth phosphate precipitate and an increased solubility of bismuth phosphate and also of plutonium phosphate. Weak concentrations of N_2H_4 (0.0005 M) can reduce Pu(IV) to Pu(III) and the Pu(III) formed does not easily re-oxidize. It may be necessary to oxidize the hydrazine with $NaNO_2$ or $KMnO_4$.

Another Hanford related problem, I said, concerns procedures for increasing overall the rate of plutonium recovery. Plans are designed to process four tons of uranium per day with about 250 grams of plutonium per ton. I also described studies with a 28% UNH solution in the Bismuth Phosphate Process. The extraction is followed by the decontamination cycles using 2 N HNO_3 in by-product and plutonium precipitation steps with cerium-zirconium scavengers and twice the normal quantity of bismuth in the plutonium precipitation steps. The use of 0.005 M $KMnO_4$ as oxidizing agent and 0.1 M NH_2OH as reducing agent in the decontamination cycles can reduce plutonium losses.

I reviewed our work directed toward determining the fate of neptunium in the Bismuth Phosphate Process. Although it is desirable to remove or lose neptunium in the regular Hanford process, should a decision be made to recover neptunium at a later date, we have determined that the neptunium is probably lost in the first plutonium precipitation step. This is most likely caused by the interference of iron in the form of the $FeSO_4$ used as a reducing agent. I mentioned our conclusion that carrying of neptunium can be increased by substituting $H_2C_2O_4$ and Mn(II) for the $FeSO_4$. I described the lanthanum fluoride precipitation for iron decontamination and the high acidity (2 N HNO_3) peroxide isolation procedure.

Next, I described our solvent extraction work on the decontamination and recovery of uranium from the Hanford waste solutions. Then I said several 8-hour solvent extraction runs have been made with feed solutions containing Hanford concentrations of zirconium and lanthanum and 1/1000 Hanford plutonium concentration. They demonstrate that satisfactory recovery and purification can be consistently obtained. A 25-hour run has been made to minimize the effects of operating holdup and minor variations in flow rates and to test the system under conditions more closely approaching the conditions of true continuous operation. Factors yet to be contended with include the determination of whether or not evaporation of the product solutions can be made without inducing corrosion by-products, and the determination of the effects of large amounts of NH_4NO_3. Batch extraction results are relatively poor, and I described the problems we have encountered due to the polymerized "green ion" of Pu(IV) that is not very soluble in hexone.

I reported the studies conducted under our Basic Chemistry Subsection problem assignments show that in the presence of SO_4^{--} ions the polymerized "green ion" does not easily form; however, SO_4^{--} adversely affects extraction with hexone. I presented the results of our transference studies on the complexing of plutonium with various ions and stated that Th^{+4} may have a polymerizable form as well as U^{+4}.

I mentioned that a value of -0.9660 ±0.011 volt at 25°C has been obtained for the potential of the Pu(III)-Pu(IV) couple (independent of H^+ concentration in 1 M Cl^-). A cubic face-centered structure for plutonium nitride has been determined (a = 5.64Å) and is similar to ThO_2 (a = 5.586Å). The vapor pressure of $PuBr_3$ has been measured, and the heat of vaporization found to be about 60,000 cal/mole. I reported on the properties of the "plutonium mirror," a thin film of about 200 atom layers of plutonium that was accidentally deposited on the glass walls of our plutonium vapor pressure experiment apparatus when a leak occurred. The film of plutonium metal seems to exhibit pyrophoric properties and does not oxidize rapidly.

I presented the status of our U^{233}, protactinium, thorium, and neptunium work and future plans. The growth of Pa^{233} by alpha-particle decay from Np^{237} has been observed and is being followed, I said. The U^{233}-Th^{229} decay chain is being followed. I reported that the structure of NpO_2 has been identified with Zachariasen's help and that neptunium in the +6 state is being studied in the form of $NaNpAc_3$. Np(III) cannot be extracted with hexone and in this respect is analogous to Pu(III). I said that intensive work has been started on the use of dibutyl carbitol for the extraction of thorium and uranium. A black residue remains after dissolving thorium metal (from Ames) in HCl. X-ray analysis shows the residue to consist of about two-thirds ThO_2 and the remainder thorium nitrate.

I also discussed our work on the use of $Al(NO_3)_3$ to increase the amount of thorium that can be extracted by ether. Allison asked me if it would be possible to exploit the use of $Al(NO_3)_3$ as a salting-out agent for uranium. I said it would be possible if the ranges of concentration are carefully controlled; I drew curves to illustrate this point. Compton then asked me about a previous statement I had made to the effect that three to four times the uranium metal could be dissolved in one day at Hanford than previously planned. He asked how long it would be before this procedure could be used at Hanford and inquired if the plutonium losses might be higher. I replied that the procedure appears practical for use at Hanford but should first be tested under semiworks conditions to evaluate safety and operational factors. I said that although plutonium losses may be somewhat higher on a percentage basis, the overall losses should not be excessive (and probably permissible in view of the increased production rate benefits to be gained).

Next, Hogness introduced Burton, who spoke about Section C-II work which includes the graphite radiation damage problem and the effects of the Hanford process radiation environmental conditions on organic solvents and on Hanford process radiation environmental conditions on Hanford cooling water.

Sugarman, representing Section C-III, talked about the radioactive

poisoning effect discovered in the Hanford pile when the pile was first brought up to a power level of 9 Mw. The poisoning effect is caused by short-lived Te^{135} which decays to 6.6-hour iodine and eventually into 9.4-hour Xe^{135}; the latter probably has a high thermal neutron absorption cross section. Xenon-135 normally decays to a 20-30-year cesium that becomes stable barium. The yield of this fission chain is reported to be about 4.3%.

Johnson introduced the speakers from Clinton Laboratories. Elliott spoke about the xenon poisoning problem also. His measurement on the half-life of I^{135} gives 6.9 hours instead of 6.6 hours. The fission yield was found to be 5.7% instead of 4.3% for the chain beginning with tellurium. Sugarman suggested the resulting difference may be due to a difference in counting techniques. Problems concerning the accuracy of the physicists' neutron flux values were also discussed. Elliott also described experiments with neutron-bombarded LiF to produce tritium. So far only helium has been found.

English described three problems being studied, two concerning the Hanford process and one concerning U^{233}. The two problems in the extraction step are to study losses caused by complex formation and to study the action of interfering agents such as hydrazine. Nitrite ($NaNO_2$) is not a good reducing agent for the Hanford process especially if much Pu(VI) is present in the process solution. Oxalic acid seems fairly good. English said $NaNO_2$ seems to kill the effects of hydrazine: this is probably due to the $NaNO_2$ acting as an oxidizing agent. The work at Clinton on U^{233} is progressing. English reported results of the dissolution of thorium metal with HNO_3 and with HF in concentrated HNO_3. Some residue remains and probably contains silicon. I cautioned English that when one uses HF, a complex will be formed that will interfere with the extraction process. English said HCl will also dissolve the thorium but this is undesirable and causes increased corrosion problems. With regard to the residue, Zachariasen asked Spedding how the thorium was melted. Spedding said beryllium oxide crucibles were used (not nitride as Zachariasen had earlier suspected).

Davies described recent work at Clinton on three problems: (1) behavior of Fe(III) in separation processes; (2) solution of uranyl phosphate; and (3) colloid factors in the separation process. He referred to our Chicago work on bismuth phosphate precipitated over a several days period from solutions with known concentrations of iron. One-third of the precipitated material was alpha phase bismuth phosphate, but two-thirds was a new phase, the structure of which has not as yet been analyzed and reported.

Initial studies at Clinton on the solubility of uranyl phosphate have been completed. It was observed that the effect of H_3PO_4 upon the solubility of uranyl phosphate in HNO_3, at low concentrations of about 0.2 M per liter, is first to reduce solubility due to the common ion effect and then to increase solubility in a way almost proportional to H_3PO_4 concentrations above about 0.2 M per liter. This increase is caused by an increase in the phosphate complex formation. The Clinton program to study the colloidal problems that might arise in the Hanford separations process has nearly been completed. It has been determined that Pu(IV) will remain ionic in all fractions examined.

Doan said that the report on the separation processes usually given by Sutton will not be given because Sutton and Greager are not now at Clinton. He announced that a new technical division is being organized under Leverett. Struthers presented the Clinton Analytical Division report. Proportional counters are now being used to count alpha-particle samples instead of the 50% and low geometry parallel plate pulse ionization chambers. A thin film of collodion is routinely being placed over alpha-particle samples to prevent materials from flaking off. For low-level gamma measurements the ionization chamber measurements using a Lindemann electrometer are found to give identical decontamination factor results as have been previously obtained using Geiger tube counters.

Spedding, introduced by Allison, presented the work at Ames on metallurgy of beryllium and thorium. He introduced Amos Newton, who reported some initial tracer studies on neptunium complex formation. Phenylarsonic acid has been used to precipitate Th(IV) and Zr(IV) and is a good indicator of the IV state. He said the data presented indicate that the oxidation stages of neptunium can be either II and III, or IV and V. The data also suggest that Np(V) may be fluoride insoluble but phenylarsonate soluble. I observed in the closing discussion that the behavior of Np(III) seems to be as interpolated between U(III) and Pu(III). There were no speakers from Berkeley or Los Alamos at this meeting.

Helen went to chemistry class at the YMCA College.

Wednesday, October 18, 1944

An 8:00 a.m. meeting of the Council of Section C-I in my office was attended by Albaugh, Cunningham, Davidson, Dawson, Ghiorso, Gilbreath, Hindman, Jones, J. J. Katz, Katzin, Lawroski, Manning, Orlemann, Simpson, R. Thompson, and me. I read a memo received from Vernon about security regulations. The responsibility of group leaders in enforcing security was again emphasized. There was some discussion of a talk made yesterday by Compton.

All group leaders were requested to complete a current inventory of all radioactive isotopes and present the list to Manning. A central repository for radioactive materials other than plutonium is being considered and should have considerable utility. The protactinium work, which will be started after receipt of the 5-mg sample, is to be kept entirely separate from the rest of New Chem laboratories working with plutonium in order to avoid contamination of the protactinium with plutonium. Both isotopes have about the same energies and half-lives. It was decided Room 1 will be used to carry out the protactinium work.

Manning received a report from Nickson about the results of air velocity measurements made in Section C-I hoods by Rose's group last week. In general, all hoods were found to have acceptable air flow rates with the exception of two hoods in Room 1, now being generally used for materials that are not highly active. It was recommended that corrective action be taken before active materials above a tracer level are handled in these hoods.

I attended the Section C-I meeting of the Extraction groups at 7:45 p.m. in Room 209, Eckhart Hall. The material covered was similar to that in my presentation to the Project Council Information Meeting in Chemistry yesterday.

Helen worked on the secret version of the "Table of Isotopes" at the Met Lab.

War news the past couple of days has focused on the Pacific. Today's headline reads "B-29's Hit Formosa Again."

The Project Council Policy Meeting was held at 9:00 a.m. in Ryerson Hall. Attending were Allison, Bartky, Chipman, Compton, C. M. Cooper, Daniels, Dempster, Doan, Franck, Hogness, Huffman, Jacobson, Johnson, McKinley, Mulliken, Major Murphy, Spedding, Stearns, Stone, Szilard, Tracy, Vernon, Warner, Whitaker, and Wigner. Compton cited priority needs for physicists at Los Alamos and for no less than 500 chemists at Y-12 (the electromagnetic process area at Oak Ridge); some 25-50 of these should be research chemists. Hogness suggested the need at Y-12 is not well defined and may be more an organizational problem than a need for more trained chemists. Compton said the eventual need is very real and that Y-12 requires the kind of Chemistry Division backing that was given to Hanford. He suggested that where chemists are not now backing up Hanford, the needs of Y-12 should take precedence. Whitaker said Clinton experimental physicists have been approached and are generally willing to work wherever the Project needs are most acute. Allison feels that the Instrument Section should not be weakened but rather strengthened. He said highly skilled shop men are needed also.

Wigner said that the immediate progress at Los Alamos is not currently limited by a lack of understanding about the theoretical physics of the bomb. Compton remarked that since the loss of a large fraction of our Met Lab experimental physicists to Site Y will so greatly weaken our ability to go forward with new pile studies, effective utilization of theoretical physicists in Chicago may not be possible. It would therefore seem desirable to transfer the theoretical physicists to Site Y also. Franck and Mulliken felt differently about the matter. Franck pointed out that Chicago needs a strong theoretical group for consultation, and Mulliken said such a transfer would seriously hurt the work on the preparation of Project records.

Compton reviewed the situation at Hanford and said that a week-long operation of the 105B pile at about 30 Mw showed no new surprises. Xenon production in the pile is still the best explanation as to the cause of the poisoning effect.

Whitaker said the large new fan at Clinton has been repaired and the X-10 pile is operating at 4 Mw. He said careful measurements were made of about 7% of the center pile materials irradiated for two months. This material was pushed out and replaced with fresh material. An increase in reactivity was observed that

corresponded closely to the amount of xenon calculated to be present in the old materials which had been pushed out.

Chipman reported on the MIT work and said that precision casting has been transferred to Site Y. MIT's major emphasis is on ceramics. According to Spedding nearly 30% of the Ames effort is on Y-12 problems. The thorium and the beryllium metal production work is progressing favorably. Huffman said Evergreen pile drawings are being studied in Chicago for advice or recommendations.

Stone said that he has been instructed to limit biology research plans in the Division of Health to one year and that the building program he hoped to initiate has been cancelled. Some plutonium has been detected in the urine of workers, but so far, there are no indications of overdoses. He said that the Project still has no good way to determine the concentration of plutonium product in the air. In Compton's concluding remarks he mentioned that the University of Chicago intends to continue a program of nucleonics into the peacetime years, regardless of Government backing, but the magnitude of that program cannot be determined at present.

Thursday, October 19, 1944

Manning transmitted to W. Johnson of Personnel, letters from Cunningham and himself to Dean Paul P. Boyd of the University of Kentucky for Johnson's approval and dispatch. These are letters of recommendation for Lyle R. Dawson, who is being considered for a position in the Department of Chemistry at the University of Kentucky.

Hogness sent a memo to Allison quoting my summary of the meeting of October 11 with Leverett of Clinton and the arrangements we agreed to concerning the uranium recovery work at Clinton and at Chicago. I was requested to supply this information in response to a recent inquiry from Colonel Nichols to Compton concerning our plans for uranium recovery. Hogness said that it appears that an informal agreement has been worked out but that he does not know if Allison would wish to call a meeting with Clinton Laboratories' higher management to set up a more formal understanding of program responsibilities.

Report CN-2159, "Chemical Research, Basic Chemistry of Plutonium — Report for Month Ending October 1, 1944," was issued. It contains the following information on work conducted primarily on Los Alamos problems by three of the four groups in Cunningham's Sub-section II:

a. *Basic Dry Chemistry (O. C. Simpson, Group Leader of Group 5; N. R. Davidson, Assistant Group Leader).* Westrum's work on nine projects, related to the preparation and properties of plutonium compounds by dry reactions and the reduction of plutonium compounds to metal, is described. An attempted vapor phase reduction of PuF_3 with $CaSi_2$ at 1550°C to obtain plutonium has been unsuccessful. The compound $PuSi_2$ is produced even when PuF_3 and $CaSi_2$ are present in the exact stoichiometric proportions for the

attempted production of the metal. Ignition of $PuSi_2$ in air at 700°C produces PuO_2. Westrum has been able to produce PuC by the calcium vapor reduction of PuF_3 in a graphite crucible. He finds that PuO_2 does not react with CH_4 vapor at 1500°C to give a carbide. Attempts to form a plutonium carbide by heating a mixture of carbon and PuO_2 to 1500°C have also failed. When PuO_2 is heated to 1800°C on a tantalum filament in a vacuum, the PuO_2 is converted into what is believed to be Pu_2O_3. Vapor phase reductions of PuO_2 with sodium, potassium, lithium, calcium, magnesium, and barium have been attempted by Westrum. Only lithium and barium react partially with the PuO_2 to produce some "Pu_2O_3."

Westrum and Abraham have prepared PuN from the reaction of plutonium metal on a platinum filament at 1000°C in the presence of NH_3. The reaction is observed to proceed at a rather slow rate. PuN may also be produced by reacting $PuCl_3$ with NH_3 at 800-900°C. X-ray crystallographic evidence suggests that the metallic-appearing golden bronze-colored material obtained by the vapor phase reduction at 1250°C of PuF_3 with calcium in a BaS crucible is impure PuS (or possibly BaO). No detectable change in weight of the sample occurs upon air ignition, and the resulting black oxidized material gives only a PuO_2 x-ray pattern upon analysis. This strongly suggests that the golden bronze-colored material is PuS rather than BaO, but further work is required to be entirely certain of this conclusion. Westrum has studied the vapor phase reduction, on a 5-mg scale, of PuOCl with barium at 1250°C, which yields primarily PuO with a low yield of plutonium metal. He has also studied the reduction of PuF_3 with lithium vapor at 1200°C. In one run he finds that plutonium metal is produced that is unusually pyrophoric. The other runs produce globules of plutonium that do not spontaneously ignite in air. He has attempted to produce ThF_3 and UF_3 by treating ThF_4 and UF_4 with sodium vapor at 750°C. No reaction takes place. An attempt has also been made to produce ThF_3 using stoichiometric proportions of ThF_4 and thorium fired in vacuo at 700°C for one hour. X-ray analysis indicates that no reaction has occurred. Westrum states that ThF_3 is "elusive and perhaps mythical."

The problem of the preparation of plutonium halides has been studied further during the month of September. Florin has conducted an experiment in which about 1 mg of PuF_4 was placed on a nickel filament and heated to about 800°C in the presence of fluorine gas. A "higher plutonium fluoride," supposedly PuF_6, was volatilized off the filament and directly onto a glass surface cooled to liquid oxygen temperature. The white condensate, which accumulated on the cooled surface, sublimed away when the surface was allowed to warm to room temperature. The filament current was then shut off, and the source of liquid oxygen removed. The material condensed in a liquid nitrogen cooled glass "U" tube leading to the vacuum pump. Within a short time a liquid phase appeared, which probably indicated a reaction of the "PuF_6" with the glass surface. After nitrogen was admitted to the system, a solution of zirconyl sulfate was pipetted into the "U" tube, and a clear solution of all the plutonium was obtained. Spectrophotometric analysis of this zirconyl wash showed the plutonium present to be almost entirely in the Pu(VI) state. Further work is needed, however, to identify definitely the substance that condensed directly on to the liquid-oxygen cooled surface.

Fried has studied the vacuum decomposition of PuF_4 and was able to produce rapidly what appeared to be PuF_3 by heating PuF_4 in a platinum crucible at 900°C in vacuo. The product was shown to be pure PuF_3 by Zachariasen. Robinson has succeeded in melting PuF_3 on an electrically heated tantalum filament in an argon atmosphere at temperatures somewhat below 1435°C. High accuracy was not possible in the measurement because of the formation of PuOF from trace amounts of oxygen in the system. $PuBr_3$ has been synthesized by Hyde as a sublimate by reacting $PuO_2 \cdot xH_2O$ with mixed sulfur and bromine vapors at 800°C in a quartz reactor.

Several attempts have been made by Abraham, Davidson, Sheft, and Hagemann to synthesize and isolate PuI_3 in a pure form starting with plutonium compounds such as PuO_2 as well as $PuCl_4$ in solution (which should be more easily obtainable starting materials than plutonium metal used previously to synthesize PuI_3 successfully). The following reactions were tried, but all were considered to be unsuccessful:

(1) $PuO_2 + HI \xrightarrow{750°C} PuOI$

(2) PuI_3 (solution) + HI (at 60 mm) + $NH_4I \xrightarrow[\text{then to } 350°C]{\text{to dryness at } 60°C} PuOI$

(3) $PuO_2 + C + HI$ (at 60 mm) $\xrightarrow{1000\text{-}1400°C}$ No volatile reaction product

(4) $PuO_2 + AlI_3$ (vapor) $\xrightarrow{800°C}$ No reaction

(5) $PuO_2 + AlI_3$ (liquid) $\xrightarrow{450°C}$ Green reaction product, partially volatile at 800°C but not identified

Attempts have also been made to determine the equilibrium constants for the reactions:

(1) $PuO_2 + \frac{1}{2}H_2 + HX \longrightarrow PuOX + H_2O$

(2) $PuOX + 2HX \longrightarrow PuX_3 + H_2O$

Hyde and Sheft have studied the vapor phase hydrolysis of $PuBr_3$ and PuOBr. Five experiments on the hydrobromination of PuO_2 using five sets of reaction gas mixtures of adjusted partial pressures of H_2, H_2O, and HBr have been run at set temperatures between 750°C and 800°C. The reaction rates have been determined, and the products have been identified by Zachariasen using x-ray analysis. The equilibrium constant for reaction (1) is determined to be between 9.1 and 0.13; the equilibrium constant for reaction (2) is between 1.3×10^{-4} and 5×10^{-4}.

Abraham has studied the vapor phase hydrolysis of $PuCl_3$. The results were not so reproducible as those obtained by Hyde and Sheft using bromides. In this study, adjusted known mixtures of H_2, HCl, and H_2O vapors were passed over the plutonium compound in a platinum boat in a quartz tube. A sample of $PuO_2 \cdot xH_2O$ was converted to $PuCl_3$ in four hours with a vapor composition of 570 mm HCl, 190 mm H_2, and 4 mm H_2O at a temperature of 675°C. The compound PuOCl resulted from the treatment of $PuCl_3$ with two different sets of vapor mixtures, indicating that under

the given conditions equilibrium was probably not reached during the four hour reaction time.

b. *Basic Chemistry (J. C. Hindman, Group Leader for Group 6).* Dixon and McLane's studies to determine the sign of ionic charge of plutonium in various complexing anion solutions is described; they report that Pu(III) is largely positively charged in 1 M and 10 M HCl. The ion Pu(IV) changes sign — is positive at 4 M and negative at 10 M HCl. The specific activity and half-life of Np^{237} determined by La Chapelle and Magnusson are reported as 1517 d/m/μg, and 2.21×10^6 years, respectively. R. Thompson describes his work on the oxidation of neptunium to a fluoride-soluble state with nitric acid. Fields reports that both $NaNO_2$ and NH_2OH can reduce neptunium to a fluoride-insoluble state and on the distribution of reduced neptunium between aqueous solutions and hexone. The presence of NH_2OH in the aqueous phase affects the distribution coefficient quite differently than does the presence of SO_2 or $NaNO_2$. A greater fraction of neptunium remains in the aqueous phase when NH_2OH is present.

c. *Recovery (L. R. Dawson, Group Leader for Group 7).* Dawson, Britain, Fineman, Fields, and Stewart have conducted detailed investigations of the factors affecting the distribution of plutonium and other elements between hexone and aqueous phases of various compositions. The change in distribution coefficient for plutonium as a function of plutonium concentration has been studied. The influences of free HF, of HNO_3 and NH_4NO_3 concentration, and of the distribution of HNO_3 between the two phases on the distribution coefficients have been investigated. The distribution of Zr(IV) and La(III) between hexone and aqueous solutions has been studied. A detailed report on the hexone extraction work is being prepared and will be issued as a joint report of the Chemistry Division and the Technical Division late this month or early November.

Helen went to her chemistry class at the YMCA College, and in the evening she and I went to the movies.

War news fades today because a hurricane hit Key West.

Friday, October 20, 1944

Stan Thompson is expected to arrive at Hanford today.

James began a series of nitric acid oxidation cycles on sample 49DD-N1, the second leaching of plutonium (the first was on September 28) from the 200 mg of plutonium plus deuterons, St. Louis cyclotron bombardment.

The Health Group completed its weekly surveys of rooms under the control of my section. Air surveys for alpha-particle activity have been taken on a daily basis. In no instance did the amount of activity found approach or exceed tolerance levels. High alpha-particle contamination in spots on floors or benches, or contaminated equipment and hoods, were

found in Rooms 1, 4, 5, 22, 27, 30, 33, and 36. Low to moderate contamination was found in Rooms 10, 29, 36, and 41. Room 28 was not monitored, at the request of Morgan, its occupant. The weekly survey of the West Stands semiworks showed widespread gamma-ray contamination in Room 217. Readings as high as 0.4 r/hr were obtained through the open door to the hot lab. Individuals spend little time exposed in this area during runs. Room 216 does not show any serious health hazards. Measurements with Pluto show that the cleanup of a spill in Room 212 has been quite thorough, but further alpha-particle decontamination is still desirable.

A survey was made of all 57 centrifuges in New Chem. The result indicates a marked improvement in the care with which centrifuges are now being used to avoid contamination. The highest reading, about 300 alpha particle c/m, was obtained on a clinical centrifuge in Room 35, occupied by Stewart, Anderson, and Asprey.

I received a phone call from Kennedy and Wahl in Los Alamos. We compared notes and find there is evidence that Lavender is working up many small cases to try to cover the subject matter of our own Case 52. We agreed to watch this closely. All three of us are tempted to go ahead with filing on our own. I agreed to consider Wahl's proposal that the four of us give Case 61 (fissionability of plutonium) to the University of California and sell Case 52 (discovery of plutonium and its chemistry) to the U.S. Under this plan Segrè could not be criticized by the University of California since he is not an inventor on Case 52; I would be acceding to the majority vote of Art and Joe (who are not members of the University of California professorial staff). I agreed to call Kennedy back with my answer.

The Patent Office of the Met Lab prepared a patent disclosure, OSRD Case No. S-2268, relating to a "method of separating plutonium from uranium and fission products by the use of alternate carriers both of which carry plutonium." I received a copy for review and find it to be acceptable except for what I feel is an unnecessary limitation to the repetitive use of the "first carrier" as applied to the alternating use of different carriers for plutonium.

An organization chart issued today for the four Divisions of the Metallurgical Laboratory (Chemistry, Physics, Technology, and Health) gives the title for Section C-I as the "Separations Studies and Basic Chemistry of the Heavy Elements Section," with the following administrative officers: Section Chief, G. T. Seaborg; Associate Section Chiefs, W. M. Manning and E. F. Orlemann; Assistant Section Chiefs, B. B. Cunningham, L. I. Katzin, and F. W. Albaugh.

The complete organization chart for the four Divisions, listing those positions above the group level, is as follows:

Chemistry

Director	T. R. Hogness
Associate Directors	J. Franck Farrington Daniels

Assistant Director	L. B. Arnold, Jr.

Sections

C-I Separations Studies and Basic Chemistry of the Heavy Elements

Section Chief	G. T. Seaborg
Associate Section Chiefs	W. M. Manning
	E. F. Orlemann
Assistant Section Chiefs	B. B. Cunningham
	L. I. Katzin
	F. W. Albaugh

C-II Radiation Studies

Section Chief	M. Burton
Associate Section Chief	A. O. Allen

C-III Chemistry of the Fission Products

Section Chief	N. Sugarman
Associate Section Chief	A. Turkevich

C-IV Analytical Services

Section Chief	D. S. McKinney
Associate Section Chief	J. I. Watters
Assistant Section Chiefs	M. S. Fred
	C. R. Schwob

Physics

Director	J. C. Stearns
Associate Director	E. P. Wigner

Sections

P-I Instruments

Section Chief	W. P. Jesse

P-II (Dissolved)

P-III (Dissolved)

P-IV Crystal Structure

Section Chief	W. H. Zachariasen

P-V Mass Spectroscopy

Section Chief	A. J. Dempster

P-VI Materials and Methods

Section Chief	E. Creutz
Assistant Section Chief	H. R. Kratz

P-VII Theoretical

Section Chief	Gale Young

P-VIII Properties of Solids

Section Chief	Frederick Seitz

P-IX Engineering

Section Chief	L. A. Ohlinger

Technology

Director	C. M. Cooper
Associate Director	A. B. Greninger
Assistant Directors	E. W. Brugmann
	L. A. Ohlinger (on loan from Physics)

Sections

T-1 Engineering Development

Section Chief	A. C. Miller
Assistant Section Chiefs	G. M. Brown
	R. N. Lyon

T-II Coatings

Section Chief	J. H. Chapin

T-III Corrosion

Section Chief	E. W. Brugmann

T-IV Metallurgy and Metallography

Section Chief	F. Foote

T-V General Engineering

Section Chief	J. O. Maloney
Associate Section Chief	W. O. Christy

T-VI Optics

Section Chief	G. S. Monk
Associate Section Chief	W. H. McCorkle

Health

Director	R. S. Stone
Assistant Director	L. O. Jacobson

Sections

H-I Clinical Medicine and Medical Research

Section Chief	L. O. Jacobson

H-II Biological Research

Section Chief	K. S. Cole
Associate Section Chief	C. L. Prosser

H-III Medical Industrial Hazards and Health Physics

Section Chief	J. J. Nickson
Associate Section Chief	J. E. Rose

Advisory Committee

William Bloom	Alexander Brunschwig
W. R. Harrison	P. Hodges
C. J. Watson	Sewell Wright

Allison replied to Colonel Nichols' October 9 letter to Compton in which he inquired about how a program to devise a means of recovery of uranium from Hanford wastes could best be carried out at Clinton Laboratories and the Metallurgical Laboratory. Allison quotes my report on the Section C-I and Technical Division meeting with Leverett which I

sent to Hogness last Saturday. Allison says he talked with Doan yesterday and it was agreed that the major division of program responsibilities which Leverett and I arrived at on October 11 are acceptable to both laboratories.

Helen worked at the Met Lab.

Saturday, October 21, 1944

Donald C. Pye, who just returned from Clinton where he went October 13 as an observer of their plutonium isolation procedures, terminated his work at the Met Lab and will transfer to Hanford on October 29.

Helen went to the opera "Die Walkure" with Frances Chilson. Frances then spent the night with us.

American troops have driven deeper into Leyte according to today's newspaper.

Sunday, October 22, 1944

Morgan, still attempting to verify the nature of the 4.05-cm range alpha-particle activity of sample 49DD-31 (derived from a series of dichromate and silver plus persulfate ion oxidation cycles on the original leaching of plutonium from the 200 mg plutonium plus deuterons, St. Louis cyclotron bombardment), carried out chemical manipulations which rule out thorium, polonium, bismuth, and lead, leaving as possibilities actinium, protactinium, and element 95. The sample, labeled 49DD-35, still contains 20 c/m of this alpha-particle activity at this time.

I played golf with Jerry Howland, French Hagemann, and Luther Arnold at Navajo Field's (123rd Street and Ridgeland Avenue) 18-hole golf course. The temperature was 58°. Our scores were JH-106, FH-108, LA-103, and GS-105.

Frances Chilson again stayed with us.

The paper today reports FDR attacking Dewey and the GOP for isolationism. In war news the capital of Leyte in the Philippines is reported captured. Charles A. Lindbergh flew a fighter against the Japanese; he is supposed to be teaching pilots how to conserve gasoline. Lindbergh is unofficially credited with shooting down a Japanese fighter.

Monday, October 23, 1944

I read a memo Pye wrote to me reporting on his recent trip to Clinton. He states that the amount of material (slugs) being processed in the canyons per run has recently been increased from one-third ton to one-half ton. Processing, using two decontamination cycles and a cross-over cycle prior to isolation, continues to be satisfactory. Eleven runs have been made involving recycling experiments in order to build up the plutonium concentration in the lanthanum fluoride-plutonium solution to a level where the Hanford isolation procedure could be evaluated on a plant scale. The recycling procedure used was to metathesize and dissolve the combined lanthanum fluoride-plutonium precipitates from two runs; precipitate the plutonium as the peroxide; dissolve this in Room D; and with the supernatant added, recycle this entire solution to the lanthanum fluoride-plutonium precipitation step as the lanthanum precipitating agent in 204 Building. The Hanford isolation equipment has been "copied" at Clinton, and two isolation operations have been carried out successfully in 204 Building under conditions which closely approximate Hanford conditions.

Pye reports that Stoughton's group is investigating methods for dissolving thorium and ThO_2 and has determined that a solution of 8 N HNO_3 and 0.3 to 0.5 N HF [or 0.005 N $(NH_4)_2SiF_6$] offers promise. Clinton Laboratories' future plans include the recovery of all U^{233} from the remaining cans of irradiated $Th(CO_3)_2$, the development of a method for obtaining irradiated materials in the future on a continuing basis, and the development of procedures for isolating very pure U^{233}. The isolation procedure contemplated for the latter will consist of immediate ether extraction of the irradiated thorium carbonate to remove uranium and other impurities. The aqueous phase solution will be then aged to permit the growth of U^{233} from its parent Pa^{233}; the U^{233} will then be removed in a pure state by a second ether extraction. A continuous ether extraction system is planned for eventual use.

I wrote to Segrè in response to a letter of his to Allison (which was referred to me in Allison's absence) concerning our work on Np^{237}. In my letter I say: "Our total present supply of 37 amounts to about 25 micrograms spread throughout the laboratory. The work on 37 described in the reports was all done on the ultra-micro scale with microgram amounts. The total supply in the laboratory up until now has never exceeded some 60 to 70 micrograms. However, I have recently recommended a change in the process conditions at Clinton which should result in a substantial improvement of the yield of 37 from the plant. This change will be tried out in one or two daily batches about a month from now. With good luck this may make it possible to supply you with a hundred micrograms sometime thereafter."

Allison sent a memo to Colonel Metcalf of the Manhatten District's Patent Office at the Met Lab about the Met Lab patent situation and the acceptability of Fermi's signature as "sole inventor" on several specific patent claims. Allison says in his letter:

"It seems to me that the fact that American patent law requires that a patent application be signed by a person who claims that, to the

best of his knowledge, he is the sole inventor, is unfortunate. The great majority of patentable ideas developed in the Laboratory arose from discussions between members of the group, which makes it uncertain who should claim to be sole inventor.

"Since this provision is an unalterable factor of the patent situation, we must, in many cases, consider the signature of an individual on a patent as a formality required by Law. Under these conditions, I am sure that Dr. Fermi should sign the patents and that he would not be subjected to criticism by other members of the Laboratory for doing so."

Compton officially requested Oppenheimer the transfer of 200 mg of metallic Pu^{239} to be used in our further work on compounds, basic chemistry, and vapor pressure of plutonium.

Helen worked at the Met Lab all day and then spent the evening at home.

Again come reports of U.S. troops in Leyte; they have taken more towns.

Tuesday, October 24, 1944

Egan and Gilbreath received a notice from Nickson stating that eating of lunches in offices in West Stands should be strongly discouraged. He recommends the use of the lunchroom on the first floor.

I responded to a letter from Catherine Sands, Secretary of the Counsel and Guidance Center, Institute of International Education in New York City, saying that I would be happy to meet with Mario Eduardo Báncora at any time during the week from November 6-11, 1944. I suggested he call me at home phone number, Fairfax-2338. Báncora is from Argentina and is scheduled to work in the Physics Department at Berkeley.

This evening, starting at 7:00 p.m., I presented a two-hour lecture on "Isotopic Tracer Techniques" as the fifth lecture in a 12-lecture series, on the general theme of "New Tools for Chemical Research," sponsored by the Northwestern University at its downtown University College. I lectured in Lincoln Hall of the Law School Building, 367 East Chicago Avenue. A Manhattan District security man attended in order to monitor what I said. The University of California was listed as my university affiliation on the program in order to avoid focussing attention on the Metallurgical Project at the University of Chicago. I received a $25 honorarium for the lecture.

Helen went to her chemistry class at the YMCA College.

Stalin announced that the Soviets have driven into East Prussia.

Wednesday, October 25, 1944

The Council of Section C-I met with me in my office at 8:00 a.m. In attendance were Albaugh, Cunningham, Davidson, Dawson, Ghiorso, Gilbreath, Hindman, Jones, J. J. Katz, Katzin, Lawroski, Manning, Orlemann, Simpson, and R. Thompson. Manning brought up the health protection subject relating to the changing of shoes before entering the filtered air section of New Chem. As a result of group discussion, we decided that the rules concerning shoe changes should be maintained and that the shoes should again be marked to show clearly the area in which they are to be used.

It was generally agreed that a cleansing of all centrifuges be performed on a regular routine basis by the chemists who use the centrifuges. Several individuals aired complaints concerning the performance of the Health Physics group. They feel that the surveys are not adequate and that the distinction is not being made clear between serious health hazards and tolerable conditions. They also believe that nose and face counts should not be discontinued. It was also suggested that more "Pluto" alpha particle ionization chamber survey meters are needed.

I phoned Kennedy in Los Alamos. I told him I will watch Lavender's new, small patent cases and that I would be tempted to file at once, if we knew how. I also said I am agreeable to Wahl's plan to give Case 61 to the University and sell Case 52 to the U.S. government.

I received a copy of Duffey's memo to Maloney and Tepe enclosing a procedure, which has been reviewed and approved by Dawson and Nickson, for the disposal of waste solutions from the 3-inch extraction apparatus under construction in the center stairwell of West Stands. All precautions are being taken to avoid hazards to operators and possible contamination of surrounding areas.

"Chemical Research — Separation Processes for Plutonium, Report for the Month Ending October 1, 1944," (CN-2158), was issued today. This is an 89-page report, which contains the following information on work conducted on Hanford problems last month by the four groups in Albaugh's Sub-section I:

a. *Extraction-Decontamination* (R. C. Thompson, Group Leader of Group I). The work of Greenlee, Bartell, Winner, and Larson on by-product precipitation at low acidity in relation to the Bismuth Phosphate Process research problem assignment is reported. Experiments on extraction from 28% UNH have been extended to solutions containing Hanford concentrations of plutonium. These studies show that extractions with yields of 96-98% can be made from 28% UNH at a total acidity of 0.8 N and a Bi(III) concentration of 1.9 mg/ml. By-product precipitations at low acidities, using newly prepared Clinton slug solution with added Hanford concentrations of fission-product elements, have given very poor decontamination factors (up to 10 for the step) compared with factors (as high as 500) obtained previously with other slug solutions. Variations in holding oxidant and the use of "holdback carriers" have failed to increase the decontamination factor. The difficulty is thought to be due to a different colloidal state of the fission-product elements (FPE) in the new slug solutions.

The FPE were, for the first time, added as a clear solution, and the new slug solution was completely free of haze.

Bradt's work is reported on the centrifugation and 1-G settling characteristics of the by-product precipitate that will form in Hanford process solutions. Centrifugation is found to be about six times as effective as settling in the removal of gamma-ray contamination. Most of the centrifugable beta-particle activity is found to settle out in a few minutes without high-G centrifugation. The amount of activity that remains in suspension after settling is not proportional to the total weight of the precipitated solid material.

R. Thompson's work on the oxidation of neptunium by HNO_3 is reported. Neptunium is oxidized when heated at 95°C in 16 N HNO_3 for one hour, and then heated for an additional 48 hours at 90°C in the solution diluted to 0.7 N HNO_3. (These conditions nearly completely oxidize plutonium also.) A lanthanum fluoride precipitate, resulting from adding lanthanum and making the solution 1 N in HF, brings down only 8.9% of the neptunium activity present in the original solution sample.

Four 75 ml-scale process runs by S. Peterson and Greenlee, have been made at Hanford concentrations of plutonium and inactive FPE (1) to confirm decontamination results obtained by Bohlmann's group at Clinton involving the repeated reprecipitation of the bismuth phosphate from reduced plutonium solutions (10^4 through five reprecipitations), and (2) to study the effect of complexing agents other than H_2SiF_6. All runs have been made in the presence of stainless steel. The use of $HF-H_3PO_4$ and $H_2C_2O_4$ as complexing agents give high plutonium losses. The overall gamma-ray decontamination factor achieved with HF is inferior to that obtained with H_2SiF_6; the use of H_2SiF_6 as a complexing agent gives a factor of 6.1×10^4 after the fifth reprecipitation and very low plutonium losses.

Hoekstra, Morgan, Ader, Malm, Larson, and Bartell have studied the preextraction treatment of process solutions involving "prereduction" with $NaNO_2$. The exact nature of the beneficial effect of the $NaNO_2$ treatment has never been fully understood. It was thought earlier that the prereducing agent was necessary to reduce any Pu(VI) to Pu(IV). When the $NaNO_2$ prereduction step is eliminated by using Clinton solutions containing essentially no Pu(VI), however, it is found that large extraction losses (15-25%) still occur. This strongly indicates that $NaNO_2$ must play yet another role. Furthermore, laboratory tests have often shown that excellent carrying from $UNH-H_2SO_4$ solutions can be achieved even though a major fraction of the plutonium is in the Pu(VI) state at the beginning of the precipitation.

A number of experiments following the fate of Pu(III), Pu(IV), and Pu(VI) under various conditions have been run. Experiments in the well-known role of N_2H_4 in reducing plutonium to its +3 state are described in detail. At Hanford concentrations of plutonium the amount of N_2H_4 thought to be present in Clinton pile metal will seriously interfere with plutonium carrying in the extraction step unless measures are taken to destroy it. Sodium nitrite has seemed to serve a role of destroying the N_2H_4 in Clinton plant-scale runs, but erratic extraction-loss results

have been obtained on laboratory scale experiments in which pretreatment with N_2H_4, followed by $NaNO_2$, is employed.

b. *Concentration-Isolation* (D. G. Pye, Group Leader of Group 2). Kelley, Malm, and Yett have continued their studies on the fluoride method of concentration for application at Hanford. The procedure for the elimination of iron by a lanthanum fluoride-plutonium reprecipitation has been tested on a 135-ml scale in glass equipment. Iron concentrations were found to be reduced to below the present safe limit of 0.002 M. A large scale 530-ml run using stainless steel equipment has resulted in a 98.5% plutonium yield and a reduction of iron concentration although, understandably, not so great as is obtained in the glass equipment. In additional experiments it has been determined that Clinton plant lanthanum fluoride precipitate would dissolve completely in about 10 minutes at room temperature. Plutonium yields on two of these runs have averaged 99.7%. It is noted that some zirconium (known to be present in the lanthanum fluoride) persists in the lanthanum nitrate-plutonium solution. This implies that the reprecipitation process is not very effective for zirconium decontamination. To study the elimination of zirconium in the concentration procedure, Walling has used 65-day radio-zirconium tracer. Starting with 0.005 M zirconium in the 10 N HNO_3 solution of bismuth phosphate at the end of the last decontamination cycle (more than to be expected at the Hanford plant), he finds that 0.001 M zirconium is present in the lanthanum nitrate-plutonium solution at the end of the concentration procedure. This concentration of zirconium would cause only a slight increase in solubility in the first peroxide precipitation.

Meyer has studied plutonium losses in the lanthanum fluoride by-product precipitate as a function of plutonium concentration. Although the percentage of plutonium losses decreases with an increase in plutonium concentration over the range of one-half Clinton to two times Hanford concentrations, the absolute losses remain essentially constant and in no case are the percentage losses excessive (>2%).

Experiments concerning isolation at Hanford using the peroxide method are reported. Beard, Goeckermann, and Hopkins have studied the development of a high acidity plutonium peroxide isolation procedure that will permit improved filterability of the plutonium peroxide precipitates in the presence of high iron concentrations (0.01-0.05 M) in the first peroxide precipitation. The high acidity procedure has been evaluated using lanthanum fluoride slurry (from Clinton, Room D) and found to be entirely satisfactory (97.7% yield). Zirconium contamination is the major cause of the small loss. The settling characteristics of precipitates formed by the high acidity procedure have been studied to determine the feasibility of supernatant removal by decantation. The results of the experiments indicate that overall yields should be greater than 95% if care is taken to prevent mechanical loss of the precipitates during decantation.

Hopkins has run preliminary spectrophotometric studies that indicate that Pu(IV) may be the form of plutonium in plutonium peroxide in HCl solutions. Previous investigation also points to this conclusion. Haeckl has explored alternate methods for achieving necessary plutonium concentrations during the early Hanford operations when the plutonium concentration in process solutions is expected to be only about 10% of

the anticipated final Hanford value. The feasibility of using high acidity (10 N HNO_3) solutions of second bismuth phosphate product precipitates to obtain good carrying of plutonium by lanthanum fluoride has been studied. Material balances have been poor, but losses in the lanthanum fluoride precipitation step are not excessive.

c. *Process Development* (J. R. Gilbreath, Group Leader of Group 3). The decontamination achieved by the low-acidity Bismuth Phosphate Process has been further studied by Ader, Hyman, Larson, Lincoln, and Winner. Data obtained indicate that the low acidity process is much more sensitive to the colloidal state of dispersion of the FPE than is the cerium-zirconium scavenger process. Lincoln has conducted experiments to determine the distribution of neptunium in the Bismuth Phosphate Process. From results of two 100-ml scale runs he finds that the overall yield of neptunium through an extraction step, one bismuth phosphate cycle, and the cross-over cycle is 48%. Uranous sulfate (0.01 M) or oxalic acid (0.05 M) are used (1 hour at 75°C) instead of ferrous iron in the plutonium precipitation step.

Solvent extraction and decontamination studies have been continued during September. Blaedel and Walling have studied the solvent extraction scheme based on the trivalent state of plutonium. The process can be applied directly to plutonium solutions, and alternations in the oxidation state of plutonium between +3 and either +4 or +6 allow alternate transfer of plutonium between aqueous and non-aqueous phases for as many cycles as may be required to attain required decontamination from fission products and uranium. On the basis of preliminary experiments, it has been found that extraction of Pu(IV,VI) and UN from dissolver solutions into hexone is feasible. A hexone insoluble state of plutonium, presumably Pu(III), can be formed at 25°C on treatment with various reducing agents and may be retained for reasonable periods of time in the hexone-H_2O-NH_4NO_3-HNO_3 system. Blaedel and Walling find that hydroquinone in a concentration of 0.05 M (in the hexone phase) is a satisfactory reducing agent.

d. *Solvent Extraction Methods* (L. R. Dawson and S. Lawroski, Group Leaders of Group 4). The solvent extraction method proposed for concentration and isolation of plutonium consists of the following steps: (1) dissolution of the lanthanum fluoride-plutonium precipitate in $ZrO(NO_3)_2$ and the addition of NH_4NO_3 and HNO_3 to this "feed" solution to secure optimum conditions for extraction; (2) extraction of the plutonium, as a nitrate, from the feed into hexone; (3) removal of the plutonium from the hexone phase (which contains HNO_3 at this point) into a "recovery" water phase. Various factors involved in the application of this method to batch operation and to continuous countercurrent column operation are reviewed in this report.

Stewart, Britain, Fineman, Asprey, and Leventhal have studied various factors concerning the hexone extraction procedure. They have determined that when a zirconium/lanthanum ratio of 3 is used, HF remaining in the washed lanthanum fluoride precipitate in concentrations of 0.1-0.5 N will not interfer with subsequent lanthanum fluoride dissolution. Distribution coefficients have been studied using both spectrochemical and radiochemical methods of analysis. The effects of centrifuge lubricating oil on the dissolution of the plutonium lanthanum fluoride, the extraction of plutonium from the standard feed solution, and the reextraction of

plutonium into the recovery water, in all cases, is found to be small.

Batch extraction with hexone has been studied, but a selection of a recommended procedure and optimum set of conditions is not considered possible on the basis of the present limited work. The distribution coefficients of zirconium, lanthanum, and iron between feed solution and hexone have been determined by spectrochemical, colorimetric, and radiochemical methods. Radiochemical distribution coefficients for the three elements are 50, 1500, and 10,000, respectively, in favor of the feed solution.

Brody, Fields, Fineman, Lawroski, Reinhardt, Stein, and Stewart have conducted a series of four runs to obtain preliminary data on the continuous countercurrent solvent (hexone) extraction method for plutonium isolation for possible application at Hanford. The apparatus used is described in this report together with a brief account of the operating procedures. The results of these four runs indicate that approximately a 100,000-fold reduction in zirconium concentration and more than a 15,000-fold decrease in lanthanum is attained between the feed solution and the aqueous column drain.

A total of six 8-hour runs has been made each using about two liters of feed solution containing about 6.2 g of lanthanum and 18.6 g of zirconium per liter and about 1 mg of plutonium per liter (about Hanford concentration for lanthanum and zirconium and about 1/1000 Hanford level for plutonium concentration). The results of these runs give an overall plutonium recovery of about 99.5%. The purity of the plutonium is about 70% which should be equivalent to about 99.96% purity at Hanford plutonium concentrations. No operating difficulties have been experienced. The 3-inch full plant size extraction unit of pyrex has been completed and tests should begin soon.

Vernon sent a memo to all Division Directors and Administrative Officers stating that effective next Wednesday, Lester C. Furney (Eckhart Hall) will take over the handling of all the Project's "special products" including Products Nos. 9, 23, 25, and 49." Vernon pointed out that it is necessary to maintain clear records of the amounts of special products received and their disposition.

During the late afternoon, I played nine holes of golf (Nos. 1-7, 13, and 16) at Jackson Park with Leonard Katzin, Luther Arnold, and Steve Lawroski. Our scores were LK-49, LA-47, SL-49, and GS-50.

I attended the 7:45 p.m. meeting of the Basic Chemistry and Instruments groups (Sub-section II) of Section C-I in Room 209, Eckhart Hall. Also in attendance were Arnold, Asprey, Brody, Cunningham, Daniels, Davidson, Dawson, Dixon, Egan, Fried, Gilbreath, Hagemann, Hindman, Howland, E. K. Hyde, Jones, J. J. Katz, Katzin, Kraus, La Chapelle, Lawroski, Manning, Orlemann, S. Peterson, Robinson, Seifert, Stewart, Studier, R. C. Thompson, Warner, Westrum, and others. Seifert presented data on recent vapor pressure measurements of Pu, PuO_2, Pu_2O_3, PuF_3, $PuCl_3$, and $PuBr_3$. A diffusion method is used and employs a tantalum crucible with a small orifice in the lid. The crucible is heated by induction in high vacuum to provide for saturation pressure within the crucible. The vapor

escaping through the orifice is passed through a collimating slit and condensed on one of a series of sample collector plates cooled with liquid air. The analysis of the diffusion data over temperature ranges of interest gives extrapolated boiling points of 3090°C, 2070°C, 1710°C, and 1490°C for Pu, PuF_3, $PuCl_3$, and $PuBr_3$, respectively. Melting points (phase transitions), indicated by abrupt changes in slopes of the plotted vapor-pressure data as a function of inverse temperature, are found to be 1145°C, 742°C, and 654°C for PuF_3, $PuCl_3$, and $PuBr_3$, respectively. The PuO_2 data yield two different lines that are thought to be due to the formation of Pu_2O_3 from PuO_2 at elevated temperatures in the presence of tantalum. The possible presence of a more volatile impurity in the halide samples may explain why lower melting points are observed for $PuCl_3$ and $PuBr_3$ than have been obtained by Robinson using direct melting techniques. Robinson reported that the melting points for sublimed $PuCl_3$ and $PuBr_3$ determined directly are 760-765°C, and 681°C, respectively. These temperatures compare well with data from Site Y, which are 760°C and 685°C, respectively.

Asprey commented on recent difficulties in the solvent extraction work. It is found that although extraction of zirconyl complex solutions gives good yields, this method does not give a very pure plutonium solution. Attempts at re-extraction into this organic phase gave very low yields (85%) at first thought to be due to the presence of some Pu(III). Oxidation with $KMnO_4$ gave some improvement, but subsequent analysis by spectrophotometry identified the non-extractible material to be almost entirely polymerized Pu(IV). Heating the solutions containing this form of Pu(IV) in about 5 M HNO_3 for some time appears to be a satisfactory method for increasing the extractibility and thereby permits purification of the plutonium solution together with high yield.

Kraus reviewed his work on the behavior of Pu(IV) in solutions of low acidity. Kraus has slowed down the rates of polymerization and disproportionation by properly adjusting the acidity and by reducing the Pu(IV) concentration. This has permitted him to observe and follow the changes that occur with time and at various acidities, using a spectrophotometer. An attempt has been made to identify some monomeric basic Pu(IV) ions by extrapolating the data back to time of mixing. From the data obtained, it appears that two hydroxide ions are being picked up by a hydrated ion in solutions of given acidities. About 50% of the Pu(IV) is hydrolyzed around pH 1.5. Kraus observes that the polymerization, which occurs at room temperature around pH 1 and 2, continues over long periods of time as evidenced by the progressive changes of the absorption spectra of the polymeric solutions with time.

Today's headlines indicate that U.S. planes have attacked the Japanese fleet.

Helen worked at the Met Lab today.

Thursday, October 26, 1944

In a memo to Dan Koshland at Clinton, Cunningham refers to discussions he had during Koshland's last visit to the Met Lab concerning the nature of the soluble and insoluble end products of Pu(VI). Cunningham told Koshland that we now have excellent x-ray diffraction data on three of the compounds discussed resulting from the reaction of Pu(VI) and KOH. These patterns differ from the products resulting from the reaction of U(VI) with NaOH, KOH, and NH_4OH but may be similar to the product resulting from the precipitation of Np(VI) with NH_4OH. Cunningham asks Koshland for the details of the methods used at Clinton to prepare reaction products of Pu(VI) and bases. In return, Cunningham promises to keep Koshland informed of additional information obtained on our continuing experimental work relating to the same problem.

Zachariasen sent W. Rubinson the x-ray diffraction data on beta and alpha form in 14 bismuth phosphate samples and the results on five lanthanum phosphate samples. Zachariasen then sent Allison the x-ray diffraction results and lattice dimensions of samples received from G. R. Leader at Site X containing a new phase of uranyl phosphate.

Helen went to chemistry class at the YMCA College.

"Jap Fleet Defeated" reads today's headlines.

Friday, October 27, 1944

Morgan carried out a perchlorate oxidation yesterday on sample 49DD-35 (derived from a series of dichromate and silver plus persulfate ion oxidation cycles on the original leaching of plutonium from the 200 mg plutonium plus deuterium, St. Louis cyclotron bombardment). Zirconium iodate was precipitated from the oxidized solution (labelled 49DD-36), the supernatant was reduced with SO_2, and lanthanum fluoride was precipitated. The zirconium iodate was found to contain only three alpha-particle counts per minute, and none was found in the lanthanum fluoride precipitate from the reduced solution. This was out of 20 c/m in sample 49DD-35. Morgan carried out a second SO_2 reduction and lanthanum fluoride precipitation and again obtained only three alpha-particle c/m. Today Morgan carried out a third lanthanum fluoride and PbS precipitation and found that neither contained any alpha-particle activity. For some reason, the alpha-particle activity is remaining in the supernatant or has somehow been lost. This is a mystery.

The Health Physics Group completed its survey of the semiworks for the past week. Some contaminated spots and equipment were found in Rooms 23, 212, 216, and 217. Gilbreath's group is attempting to clean up.

An "Informal HEW Summary Report for the Week of October 9 to 15, 1944," M-CS-(HEW)-2270, by Hilberry, Howe, and I. Perlman was issued. The report contains the following information of interest: Howe reports

that the run on the 105-B pile started earlier this month and ended at a power level of 30 Mw. Loading of the 105-B pile up to 1300 tubes (completed October 15 at 1:45 a.m.) has permitted an increase in power to 50 Mw.

Perlman reports on progress in testing the chemical extraction equipment in 221-T building in the 200 Area and on experiments being conducted in the Separations Process Laboratory in the 300 Area (the area closest to Richland). He reports that reaction rate in the dissolver, involving the removal of the aluminum slug jackets with $NaOH-NaNO_3$, is controlled by cooling and not by the slow addition of NaOH which seems to produce insoluble aluminate precipitates. Perlman says that attention is being given to the radio-iodine stack gas problem resulting from the metal dissolving process. Also, high on the priority list of problems is the development of an improved decontamination scheme to permit the elimination of one cycle and to make it possible to process three batches of slugs in one canyon each 24 hours.

I received a copy of the Clinton Group Leaders' report for the meeting of October 21. It was attended by Bohlmann, Borkowski, English, Gevantman, Koshland, and Stoughton. Koshland reports on dehydration studies on the white precipitate formed by neutralizing a HNO_3 solution of Pu(VI) with KOH. The precipitate coloration changes with time from white to greenish to yellow-green as the temperature is increased from 70°C to 500°C. It is thought that the formula changes from $K_2Pu_2O_7 \cdot 6H_2O$ to $K_2Pu_2O_7$, based on the specific activity and the dehydration of the analogous uranium compound. Koshland finds that the NaOH titration of HNO_3 solutions of Pu(VI) does not give precipitates similar to those obtained with KOH.

Helen worked at the Met Lab. In the evening, she and I took Edrey Smith and Fred Albaugh dancing at the Rio Cabana in honor of their engagement. In a way I feel I am partly responsible for the coming marriage. A year ago, September 22, when Albaugh had quite made up his mind not to join the Met Lab, I wrote a confidential letter to him, perhaps revealing more about our work than I should have, asking him to change his mind, which he subsequently did. It was here, of course, that he met my secretary, Edrey.

U.S. victory in the Pacific is growing, according to today's paper.

Saturday, October 28, 1944

This is the last day at the Met Lab for Frank Haeckl whose termination will be effective Thursday. He has accepted a position with the Research Department of Standard Oil Company of Whiting, Indiana, where he will work in his preferred field of organic chemistry.

The Health Department surveys for alpha-particle contamination of rooms in New Chem under the control of Section C-I were completed for the week ending October 28. Mild contamination was spotted in Rooms 2, 13, and 35. Moderate contamination was found in Rooms 33 and 34.

Moderate to high contamination was found in Rooms 4, 10, 27, 30, 31 and 36. In addition gamma-ray contamination was found in Rooms 7 and 19. A somewhat different system for checking and reporting contaminations is being devised by Jean Wallace. If successful, the unsatisfactory conditions will be checked within 24 hours so that expeditious corrective action can be taken.

A meeting on Solvent Extraction and Isolation was held in New Chem with Blaedel, Britain, Dawson, Fields, Lawroski, Manning, Stein, and Stewart attending. Stewart reported that he is still waiting for the results of the analyses on samples submitted to the Analytical Section relative to the batch extraction run a week or so ago. This run used the equivalent of an eight-gallon batch at Hanford and ended up with approximately 40 gallons of Pu(IV) solution. In a current experiment, H_2O_2 is being used to reduce the plutonium to Pu(III) in the aqueous phase. A 1.5 ratio of final aqueous to hexone volumes has been achieved. In this modification of the method plutonium is being extracted into the hexone in the Pu(VI) state and re-extracted into the aqueous phase as Pu(III). Work is underway to work out a batch extraction procedure which will end up with a relatively small amount of Pu(III) solution.

Dawson and Britain described their plans to precipitate Pu(IV) oxalate from the dilute final aqueous solution without preliminary evaporation. They hope the solubility will be as low as 50 mg/l in 2 to 4 N HNO_3. Britain will study the precipitation of oxalates from process solutions. Lawroski reported that data have been obtained on three column runs which employed reused hexone for all three runs. Although some decomposition is evidenced by the increased yellow color, there is no observed decrease in selectivity, and in each case only about 0.15% of the product is lost in the cycle. Purity assays have not yet been made. Lawroski said that assembly of the large 3-inch column should be completed within a very few days.

Blaedel described experiments which indicate that the presence of stainless steel may decrease the stability of solvents. For example, the insertion of stainless steel rods in ethyl-butyl cellosolve causes marked gas evolution. Lawroski asked whether this gas evolution might represent ebullition rather than decomposition. Fields suggested samples of gas should be analyzed to settle this point.

A summary was made of the problems to be pushed in the immediate future. These problems are as follows: (a) batch extraction of lanthanum nitrate-product solution employing the III state in the reextraction step in an effort to achieve minimum volume, (b) study of precipitation of plutonium(IV) oxalate from process solutions, (c) experimental tests of evaporation, (d) modification of experimental column operation to minimize final volume and hence decreasing the amount of evaporation required, and (e) test operation of 3-inch steel column.

Helen worked at the Met Lab in the afternoon. We had Steve Lawroski over for dinner and then the three of us went to Soldier Field (which was packed full) where we saw President Franklin D. Roosevelt ride around the field in an open car and heard him make a campaign speech.

Reports still come from the Pacific indicating advances in the Philippines.

Sunday, October 29, 1944

I played golf, but Helen stayed home all day except for a walk with Wilma.

The paper today reports that General Stilwell has been recalled after a fight with Chiang. He has been relieved of command of the China-Burma-India theater forces.

Monday, October 30, 1944

I sent Hogness my recommendations concerning the transfer of a small semiworks group from the Technical Division to the Chemistry Division, a reorganizational arrangement proposed by C. M. Cooper in face of the impending major changes in the Technical Division. I recommended the transfer of the following individuals: C. J. Egan (Group Leader), I. J. Schaffner (SED Operations Chief, ordering, etc.), A. J. Margolis (Analysis), J. Schraidt (SED Operator), W. C. Giegold (SED Operator), R. W. Rasmussen (Operator), B. R. Wendrow (SED Operator) [see Fig. 27], H. E. Flotow (Analysis), and H. Hasenfus (SED Maintenance). I also requested that R. G. Post, who has been on loan to the semiworks group from the Chemistry Division, be transferred back to the Chemistry Division.

Adele Koskosky transferred from Property to work as a Technician in Dawson's recovery program.

In an attempt to locate the missing alpha-particle activity from sample 49DD-35 supernatant (derived from 200 mg plutonium plus deuterons, St. Louis cyclotron bombardment), Morgan fumed the supernatant to dryness, redissolved it, and precipitated lanthanum fluoride. The precipitate was found to contain 13 alpha-particle counts per minute. Morgan concludes that this is the total alpha-particle activity left in the solution. It is obviously very difficult to draw any conclusions from experiments with such small amounts of the alpha-particle activity that was found to have an intriguing air equivalent range of 4.05 cm in our experiments on October 17.

Cunningham sent a letter through Whitaker to Spof English at Clinton in which he says:

"It has been found here that the polymerized or colloidal form of Pu(IV) is not extracted by hexone and, if present in a plant solution, would prevent satisfactory concentration and isolation by the hexone method. Laboratory observations show that there is considerable danger of polymer formation when a plutonium(IV) hydroxide is dissolved in acid. We are interested in knowing if polymer formation occurs to any appreciable

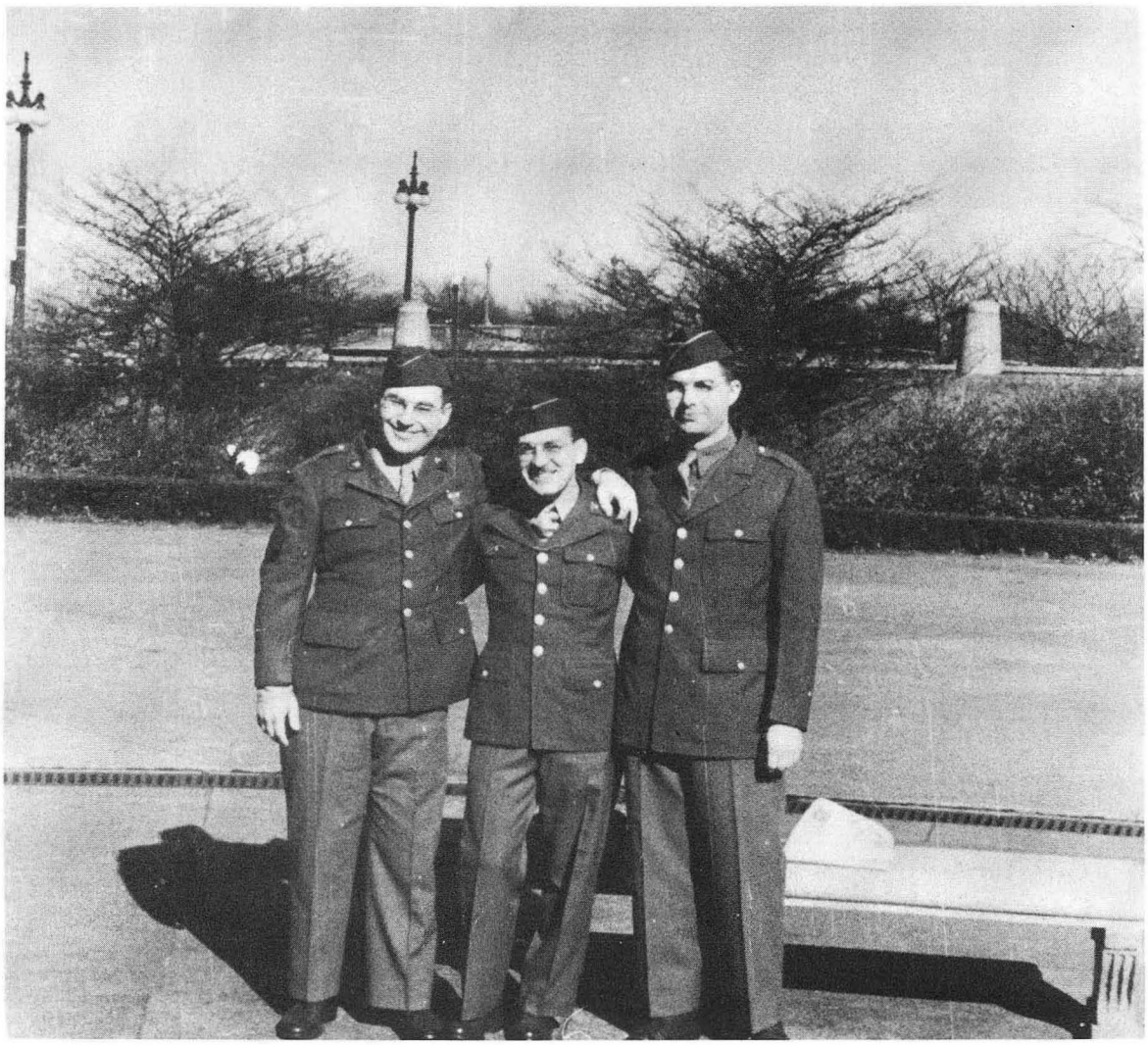


Figure 27. Leon Leventhal, B. R. Wendrow, and Phillip Fineman at Jackson Park. October 1944.

XBB 780-13176

extent under the conditions of solution of the mixed hydroxides of La(III) and Pu(IV) following metathesis, as carried out at Clinton. Would it be possible to have acid solutions of the mixed hydroxides examined spectrophotometrically for polymerized Pu(IV)? Presumably it would be necessary to have solutions from four to five typical plant runs examined in order to evaluate normal variations in the amount of polymer formation.

"The presence of the polymer is easily detected by its marked apparent absorption in the near ultraviolet. However, the details of the polymer absorption spectrum are available to you in the curves which were obtained by Werner. Significantly positive results would bear further investigation since the apparent absorption of the polymer in the near ultraviolet may in fact be due to scattering of light of the shorter wave lengths and might be given by any material in the colloidal dispersion.

"If it is not feasible to arrange to have the examination of the solutions done at Clinton, perhaps arrangements could be made to have samples sent here."

In a memo to Vernon, Hogness states that the request for alpha-particle standards from the Area Engineer is already being acted upon by my section and that I will send the samples directly to the Area Engineer's office, notifying Hogness, who in turn will notify Vernon, when the materials have been shipped.

Helen worked at the Met Lab.

Again news from the Pacific! The U.S. has bombed three more Japanese cruisers.

Tuesday, October 31, 1944

Jaffey received a letter from Willard at Hanford thanking us for the information and reprints on the FP-54 electrometer circuit. Willard says that Truman Kohman will probably ask us for information concerning the Curtiss-Davis type circuit on which Jaffey and Weissbourd are working. He concludes his letter by saying, "Most of the men are here now, and things are well under way."

I sent a memo to H. H. Fussler of the Met Lab Information Department concerning the terminal report on the purification program. The text of my letter in its entirety is as follows:

"I am writing to you with respect to the terminal report on the purification program which is being prepared under the supervision of the Editorial Advisory Board headed by Mr. C. A. Thomas and Mr. J. C. Warner. This report is being prepared at the request of General Groves and is concerned with a complete review of the chemistry, purification, and metallurgy of plutonium. I think that it is important that we have an adequate number of copies of this report available on the various

parts of the Project. In addition to the more specialized discussion of purification and metal production, this report will contain a great deal of material which will be of value to everyone concerned with investigations involving the chemical properties of plutonium. The report will contain the best discussion of the fundamental chemistry of plutonium which has appeared to date. In addition there are valuable review sections on the crystal chemistry of plutonium, the methods used for its recovery prior to reuse, and special analytical methods. This report will appear a good deal sooner than any of the sections in Mr. Mulliken's proposed Proceedings of the Metallurgical Project which might eventually cover the same ground.

"I should think that a distribution list similar to and at least as large as that which was used for Mr. Warner's minutes of the monthly Thomas meetings on purification is indicated. In addition, there should be an adequate number of copies in the main library and in the libraries of the sites connected with the Project."

Dawson pointed out to K. S. Cole that it is not considered feasible to recover 1 or 2 mg amounts of plutonium associated with large amounts of animal tissue or solutions of one liter or more containing appreciable quantities of organic matter. He suggests that these materials be turned over to the Safety Section for appropriate burial.

In another memo Dawson explains to Sergeant R. Bidlach that one pair of shoes, size 10½D, and one pair of trousers 38" waist, 30" length (army issue), belonging to Leon Leventhal (SED) had to be destroyed because of damage beyond repair in chemical operations. Dawson did not explain that the clothing had been contaminated from a radioactive spill nor did he mention that Leventhal had to walk across campus attired only in a lab coat.

Arnold commented on Stone's suggestion for the use of a secondary container within hoods to hold glass containers. Such use would help prevent accidental spills should the glass break or overturn. Arnold said he will give a copy of Stone's letter to Katzin, who is the chairman of the committee on "Product Hazards."

I learned that Lorraine Golden and John Crawford went to City Hall today and were married (see Fig. 28).

Helen went to chemistry class at the YMCA College today.

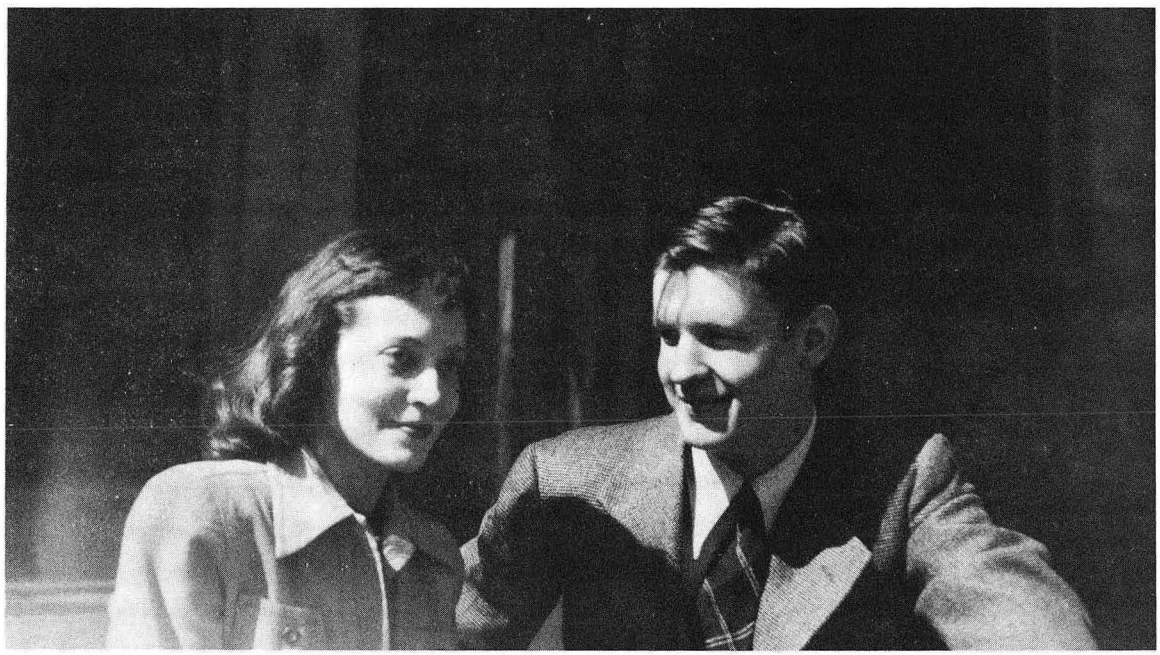


Figure 28. Lorraine and John Crawford. October 1944.

XBB 780-15277

Wednesday, November 1, 1944

The Council meeting of Section C-I at 8:00 a.m. in my office was attended by Albaugh, Cunningham, Davidson, Dawson, Ghiorso, Gilbreath, Hindman, J. J. Katz, Katzin, Lawroski, Manning, Simpson, R. C. Thompson, and me. I announced that members of this section now are invited to attend meetings of other sections just as other section members are invited to ours. The Monday night Chemistry Division meetings came up again for discussion. It was agreed unanimously that such meetings would benefit by having better speakers. On the whole, the group feels that a biweekly meeting with the speaker delivering a 40-minute talk on a general topic would have the greatest value. With regard to Section C-I meetings, it was decided that Katzin's group will talk at the next extraction meeting. It was also announced that the Technical Division is to be completely reorganized leaving only a small group; the semiworks may be transferred to our section.

Manning talked about the disposal of dilute plutonium wastes. Solutions containing less than 0.1 mg per liter, and considered unrecoverable, may be poured down the drain under Dawson's or Gilbreath's supervision; no more than 10 mg may be disposed of in the sink without specific authorization. More concentrated waste solution shall be buried at Argonne. According to new regulations a guard is compulsory for the transport between laboratories of even small amounts of plutonium (such as are present in x-ray capillaries).

I reviewed recent results on the physical constants of U^{233}. It appears that the fission cross section is about the same as for U^{235} (550 barns); the total neutron absorption is only around 620 barns, giving a favorably small value for the parameter alpha (0.12). The number of neutrons per fission is 1.07 times the U^{235} value. The number of neutrons given off per neutron absorbed is 1.13 times the U^{235} value. These values are within the range to enable U^{233} to be made from thorium by a chain reaction on the U^{233} (i.e., to make breeding possible) — extremely important because it may make it possible to be independent of uranium once a supply of U^{233} for starting purposes is on hand.

A meeting of the Division Directors took place today. Some of the topics that were considered are the following: (1) Dr. Jacobson stated that nose and face counts are not now being made on a routine basis in the Chemistry Division because results show that they are not necessary. They are still available on a voluntary basis. (2) Dr. Jacobson agreed to the advisability of a shower cabinet in the Chemistry Building in order that the employees may wash after getting product on their skin. (3) Allison states that Compton has issued a directive requesting that the separation of U^{233} from 40 slugs of thorium carbonate, irradiated at Site X, be done entirely at the Metallurgical Laboratory. Provision for doing this should be made. The remaining slugs on hand at Site X will be irradiated, and fifteen will be required for shipment to Montreal. Allison requested that 25 mg of U^{233} be set aside for the use of Professor

Koch at the University of Illinois. (4) Compton has issued a directive that Site X take the lead on the development of methods for the recovery of uranium from Hanford waste solutions. (5) A request has been made that the working day end at 5:00 p.m. instead of 5:30 for all personnel, with a one-half hour lunch period. Kimpton is reviewing this problem. (6) Cooper stated that the Metallurgical Division which will replace the present Technical Division will be under A. B. Greninger as director, with three sections — Metallurgical Section headed by Frank Foote, Corrosion Section headed by E. W. Brugmann, and Fabrication Section headed by J. H. Chapin.

Hogness issued a summary of the manpower distribution in the Chemistry Division (148 total). My section employs 73 men. The three research sub-sections or groups of Section C-I are divided in the following way.

		Number of Men	
		Sept.	*Oct.*
Albaugh (Hanford work, 26 men)	R. Thompson, Extraction and decontamination	11	10
	Pye, Concentration and isolation	8	5
	Gilbreath, Process development	7	6
	Lawroski, Solvent extraction	4	4
Cunningham (Los Alamos work and basic chemistry, 38 men)	Simpson, High vacuum work	11	10
	Hindman, Basic chemistry	10	10
	Dawson, Recovery	9	8
	Ghiorso, Instruments & physical measurements	10	9
	Katzin, U^{233} work	6	6

Helen worked at the Met Lab on the classified "Table of Isotopes."

The important right column of the paper today deals with Japanese losses in the Philippine sea battle; those losses approach 35,000 men.

Thursday, November 2, 1944

Helen went to her chemistry class at YMCA College.

War summaries in today's paper include news of a Japanese broadcast saying that B-29's raided Tokyo and news that there are three British drives on Walcheren Island at the entrance of the Schelde estuary in Holland. This latter is an assault to knock out the last German batteries barring Antwerp to Allied shipping.

Friday, November 3, 1944

I sent a memo to Hogness indicating that the following men of the operating crews of the Technical Division (solvent extraction and semiworks) are being proposed by Miller and Maloney for transfer to the Chemistry Division: Solvent extraction Operating Crew: I. J. Schaffner (Squad Leader), H. E. Fritz (SED, Analyst), H. Zvolner (SED, Analyst), G. W. Cressman (SED, Operator), E. W. Quinn (SED, Operator), W. Simon (Operator), K. P. Moseley (SED, Operator), and J. H. Schraidt (SED, Operator). I said that we have not yet decided whether these men should be transferred to the Chemistry Division or lent from the Technical Division — I mentioned that Hogness has already earmarked Cressman for the Analytical Section. The Semiworks Operating Crew consists of C. J. Egan (Group Leader), A. J. Margolis (SED), W. C. Giegold (SED), R. W. Rasmussen, B. R. Wendrow (SED), H. E. Flotow, H. Hasenfus (SED), B. L. Vondra, H. Delaney, and R. Van Winkle. In addition, R. G. Post will transfer back to our Division — he may go into the solvent extraction operating crew or into research on solvent extraction.

I read a copy of a letter from Warren Johnson to Fussler urging that the forthcoming publication on the chemistry of plutonium (L. I. Katzin, editor) receive at least a limited distribution. He gave as the reason that it would enable those working on basic chemistry of plutonium and on certain phases of the separations process to carry out their work in an efficient manner. If the proposed volume is not made available, he is certain there will be considerable danger of unnecessary repetition of work already carried out.

"Metallurgical Laboratory. Report for September 1944," (MUC-SKA-835), was issued by the Laboratory Director's Office. In the summary it is noted that: "The Chemistry Division continued its program of studies directly concerned with Hanford operation, concentrating during the month on the effects of iron in the separation process, problems arising from the formation of hydrazine in the metal dissolution step, and the 'colloidal' state of Pu(IV) in some of the process solutions. Among the most important problems of lower priority continued during the month were: (1) the hexone product extraction process, (2) separation, analysis, and nuclear properties of 23. The health protection service at the Laboratory is continually being enlarged and intensified."

Accomplishments of Section C-I during September are described as follows: "The solvent extraction alternative isolation process has passed successfully from the laboratory stage to semiworks, and plans are well under way for pilot-plant scale. The solvent hexone (methylisobutylketone) is used in a continuous process to extract the plutonium from a solution of zirconium complexed lanthanum fluoride after the crossover step. Product recoveries are exceeding 99%, and purities are expected to exceed 99.9% for W concentrations.

"The difficulties arising from high concentration of iron in the peroxide isolation process may be minimized or eliminated by precipitation from 2 N instead of 1 N HNO_3 at room temperatures. Under these conditions, high yields are obtained even in the present 0.01 molal iron, which is considerably above the anticipated Hanford level. In addition to this,

experiments have shown that the iron concentration may be reduced by a factor of 100 in the present equipment by utilizing a reprecipitation of lanthanum fluoride before isolation.

"The presence of hydrazine, formed in the metal dissolution step, may interfere in the Product extraction by reduction of the Product to the plus III state with consequent poor carrying and adversely alter the physical properties of $BiPO_4$ precipitate. The destruction of hydrazine by $NaNO_2$ gives erratic results and present experiments suggest treatment with 0.003 $KMnO_4$.

"Experiments on migration in an electrical field give clear support for the existence of complex ion formation for Pu(IV) in 7 molal HCl. Evidence for this formation has not been found in acid concentrations less than 4 molal. Oxidation and reduction potentials in the presence of complex ions have been measured in order to obtain information pertinent to the plant procedures. Stable complexes have been found with sulfate, phosphate, fluoride, nitrate, oxalate, and acetate using spectrophotometric observation, electrode measurements, and solubility determinations.

"The hydrolysis of plutonium solutions in various states of oxidation has been measured to obtain information required for storing solutions.

"Resolution of precipitated plutonium hydroxide is successful in concentrated acid but unsuccessful in dilute acid. In any case, once the precipitate has dried, resolution is extremely difficult.

"Irregularities recently obtained in the vapor pressure measurements of plutonium oxide have been explained by the establishment of dissociation of PuO_2 to O_2 and Pu_2O_3.

"The 23 separated from the thorium carbonate from the X pile by extraction with ether has shown 85% purity with respect to U^{238}. The half-life has been set at 1.4×10^5 years, the alpha-particle range as 3.38 cm in air. Samples have been sent to both Y and Argonne for additional work."

Total expenditures for the month of September were $802,387 (not including expenditures for the insurance collateral and employee benefit fund totaling $4,500,000). Personnel employed at the end of the month were 1050 (690 academic), a net decrease of 33 during the month.

Helen worked at the Met Lab on the secret version of the "Table of Isotopes." In the evening, we had ice cream at the Ghiorsos'.

War news today is all from Europe. The top headline indicates that 208 Nazi planes were destroyed in a great air battle over Merseburg, Germany.

Saturday, November 4, 1944

Mary Sue Lytle, a technician in Katzin's group, resigned to take a job in occupational therapy.

I received a memo from C. Schwob of the Analytical Section concerning the interference of iron with the precipitation of plutonium by H_2O_2. He suggests on the basis of his own early work, that H_2O_2 decomposition can be inhibited by amyl alcohol, HCN, and possibly other substances.

I sent a letter to Hilberry at Richland, Washington, attention Perlman, regarding the importance of irradiating samples of Pu^{239} and Np^{237} in the Hanford piles at high power levels. I suggest that we receive 25-mg samples of Pu^{239} from each of the four foils now in the 105B pile. I point out that the work we have already done on the irradiation of plutonium with neutrons at Clinton indicates that the maximum change in alpha-particle specific activity that could be possible in Hanford-produced plutonium amounts to a factor of five greater than that of pure Pu^{239} and that the upper limits for the changes in beta-particle and gamma-ray activity are factors of ten and twenty, respectively. These limits are still in the range corresponding to potential trouble in the separations process. I mention that a sample we have irradiated much more strongly at Clinton will be examined within a few weeks and that we shall advise him of the new limits when they are obtained.

I emphasize that it will not be adequate merely to establish whether or not the specific alpha-particle activity of Hanford-produced plutonium is different from that of pure Pu^{239} and mention the possibility of interference by elements other than plutonium including those of greater atomic number. With regard to the latter, I indicate that we have some good ideas and perhaps even experimental results on chemical properties.

I express the belief that more plutonium should be put into the Hanford pile for irradiation and mention that, in addition to the necessity for such irradiations to investigate possible effects on the separation process, there is the general importance of investigating the whole region of heavy isotopes — possible production of U^{236}, possible production of Pu^{239} with various concentrations of Pu^{240}, and the possible production of isotopes of atomic number higher than that of element 94 — all have obvious importance from the standpoint of the objectives of the DSM project as a whole.

With regard to Np^{237} irradiation, I point out these could lead to (1) the production of U^{236}, (2) an evaluation of K-electron-capturing Np^{236} as a much needed, better tracer isotope for the study of neptunium chemistry, and (3) an estimate of the amount of highly alpha-particle active [50 year] Pu^{238} that will form in the piles.

Steve Lawroski dropped in to visit Helen and me during the evening.

American troops are closing in on the escape port of the Japanese on the island of Leyte, according to reports in today's newspaper.

Sunday, November 5, 1944

Helen, Zene Jasaitis, Herman Robinson, Steve Lawroski, and I

went to Wrigley Field to see the Chicago Bears-Green Bay Packers football game. The Bears won 21-0.

Major headlines in today's paper deal with the presidential election. The newspaper and the Gallup poll predict that the election will be close.

Monday, November 6, 1944

About mid-morning I was visited in my office by W. P. Grove and F. A. Paneth from the Montreal Project, who are here on a three-day visit. They were accompanied by Captain Chapman of the Chicago Area Office. We first held a general discussion on the radiation hazards connected with U^{233} work. In answer to Grove's question, I gave the opinion that the tolerance level for U^{233} in the lungs is something like 10 to 50 micrograms. I showed them a copy of our "mandatory product safety rules" that apply to any alpha-particle emitter. We also discussed precautions in handling beta- and gamma-activity, and I described our "hot laboratories. I mentioned our system of continuous cleaning of laboratory desks, equipment, floors, walls, and ceilings and our system of changing to special laboratory shoes upon entering the building. We discussed particularly our rule of handling plutonium only when working with gloves and our rule that plutonium in any appreciable amount at all can be handled only in hoods under forced draft. After this discussion, we went through Rooms 5 and 6 where the U^{233} work is done and Rooms 34, 35, and 37 where the plutonium recovery work and other work is done. After lunch I again met in my office with the visitors and Captain Chapman. Also present was Dr. Nickson who described in detail our radiation monitoring system.

A memorandum on secrecy (MUC-TRH-188) was sent to Compton, signed by 19 people who hold the rank of Division Director or above — leaders of the Met Lab, Clinton Labs, and the Ames, and California Projects. The memorandum gives three reasons for lifting to a certain extent, the Project secrecy restrictions: (1) If the enemy has the weapon and uses it first, the lack of forewarning could cause panic among our Allies. (2) Our unexpected use of the weapon could have dangerous postwar effects stemming from the sudden, unanticipated conclusion of the war and the lack of postwar plans which take account of the existence of this new weapon. (3) The enemy may be able to create division among the Allies by pointing to the existence of big production plants in which are fabricated weapons which they can contend are not designed for the present war.

The writers/cosigners advocate that a general statement be made to the public concerning the existence of a new weapon and stating that work is going on in this country for manufacturing it and that it is bound to affect the relations of the nations in the future. They further advocate that, since these considerations may transcend those of the military, active steps should be taken to have them laid before the highest authorities of the land and before all those who will advise on the specific conditions of postwar control.

Helen worked at the Met Lab on the secret version of the "Table of Isotopes."

The banner headline today reads "Singapore Bombed by B-29's. Superforts bombed Singapore naval installations and the second biggest airplane fuel source in the Far East located on Sumatra.

Tuesday, November 7, 1944

James completed a series of nitric acid oxidation cycles on sample 49DD-N1, the second leaching of plutonium (on September 28) from the 200 mg of plutonium plus deuterons, St. Louis cyclotron bombardment. He mounted the final sample, 49DD-N3, for counting and obtained 4149 alpha-particle counts per minute. He dissolved off the precipitate and carried out a dichromate oxidation.

At 9:30 a.m. I attended a Project Council Information Meeting on Health in Room 209, Eckhart Hall. Others present were Allison, Arnold, Bartky, Borst, Cole, Curtis, Dempster, Ferry, Franck, Friedell, Goldsmith, Hamilton, Jacobson, Langsdorf, Lichtenberger, Morgan, Mulliken, Nickson, Nordheim, Pardue, Rabinowitch, Rose, Shonka, Spedding, Szilard, Tannenbaum, Vernon, Wakefield, C. J. Watson, Wattenberg, Way, Whitaker, Wigner, and Zinn. Much of the meeting was devoted to health hazards from uranium and radioactive fission products and methods for detection of the presence in the human body. Nickson reported on the routine surveys being made of the various areas of the laboratory and indicated their frequency is approaching the projected level. There is a shortage of alpha-particle hand counters, and "Plutos" are hard to obtain. Hamilton from Berkeley talked about animal studies using plutonium. For human inhalation of plutonium it is felt that tolerance is 25 micrograms, based on 10% permanent retention (2.5 micrograms) in the lungs. The effective half-life for retention of plutonium in the skeleton is greater than two years.

At 2:00 p.m. I attended the Project Council Information Meeting on Physics at which Zinn presented the following nuclear constants for U^{233}, comparing them with the constants for Pu^{239} and U^{235}:

	49	*23*	*25*	*Ratio 23/25*
σ_{abs}	1150	620 ±20	640 ±13	
ν	2.94	2.60 ±0.13	2.41 ±0.10	1.079
σ_f	820	550 ±20	548 ± 10	1.004
α	0.47	0.11 ±0.05	0.17 ±0.03	
η	2.05	2.4	2.14	
σ_c	330	62 ±28	92 ±17	

During the afternoon, Grove of the Montreal Project, accompanied by Captain Chapman, again visited me. I took Grove over to see the "hot laboratory" in the West Stands. We then returned to New Chem where we

met Burton, Paneth, and V. J. Houska. All of us then went together to see Sugarman's "hot laboratory" and to visit a number of Burton's laboratories.

Helen went to her chemistry class at YMCA College.

Paneth and Grove had dinner with us in our apartment. After dinner Steve Lawroski, Leonard Katzin, and the Ghiorsos dropped in, and we all listened to the Presidential election returns on the radio — President Roosevelt is the victor over Thomas Dewey and is now reelected for a fourth term.

Wednesday, November 8, 1944

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Cunningham, Davidson, Dawson, Ghiorso, Gilbreath, Hindman, Katzin, Lawroski, Manning, Simpson, and R. C. Thompson. I asked that renewed emphasis be placed on conforming to security regulations. In connection with the proposed schedule for locking of the back entrance to the filtered air section at 10 p.m., it was noted that there has been little night work of late and that the men do not appear to be getting here until 8:30 in the morning; the rear entrance will open at 6:15 a.m. The sink on the west wall of Room 4 has been designated as the one for disposal of dilute plutonium waste solution, under Dawson's direction.

With regard to U^{233} it was stated that, in order to evaluate Herbert Anderson's data obtained at Argonne, the specific activity of the samples he used must be determined accurately; this will require about 800 micrograms of oxide. It was mentioned that Site Y has detected Pu^{240} by mass spectrographic analysis — the sample used had about 5 parts of Pu^{240} per 10,000 parts of Pu^{239} present.

The pile at Site W has been operating at 90,000 kw for the past two weeks. It appears likely that the pile cannot be operated above 225,000 kw at the present potential loading capacity. The present plan is to run until the concentration of plutonium approaches 30 g/ton and then commence extraction of some of the plutonium.

I mentioned Hamilton's latest results in Berkeley on the retention of plutonium in lung tissue. Of material inhaled, 10 percent is retained in the lungs indefinitely; tolerance is set at 2.5 micrograms. Skeletal tolerance is placed at one microgram, with the half-life of plutonium in the skeleton at somewhat greater than two years. It is very important that plutonium be kept off skin both because the presence of minute breaks in the skin allow access of plutonium to the bloodstream, and because of the possibility of introducing appreciable amounts of plutonium into the blood by means of wounds.

Morgan ran alpha-particle range curves (mica absorbers) on sample 49DD-83. This sample gives 13 alpha-particle counts per minute, all that remains from the chemical manipulations carried out since August 29 on

the 200 mg of plutonium plus deuterons, cyclotron bombardment. The present curves indicate an alpha particle of shorter range than 4.05 cm. Morgan thinks it most likely that the shorter range is caused by a thick sample. In the chemical procedures on this sample last month Morgan identified a long range alpha-particle activity as originating from an unknown isotope, possibly from element 95.

Late in the morning I met with Paneth of the Montreal Project in Arnold's office. His visit this morning was primarily a courtesy call to thank us for the consideration which we have shown him; however, I did provide him with some references in connection with his work on meteorites.

Report CN-2287, "Chemical Research — Separation Processes for Plutonium, Process Development and Chicago Semi-works Operation," dated October 15, was issued today. This joint report by the Chemistry Division (Albaugh and Gilbreath) and the Technical Division (Egan, Margolis, Schaffner, and Wendrow), contains the following information of interest. Section C-I reports that Hyman has completed preliminary investigations of various methods for recovering uranium in a decontaminated form from the carbonate-complexed metal waste solution. The decontamination can be achieved with solvent extraction, but the uranium concentration in nitric acid solution must be high before the process is economically practical. The uranium can be concentrated by precipitation and the precipitate dissolved in a minimum quantity of HNO_3 before proceeding with solvent extraction. An overall gamma-ray decontamination factor of 10^4 has been demonstrated in a three-step process which includes (a) batch adsorption of fission product activity from the carbonate solution using titrated silica gel, (b) the precipitation of uranium as sodium uranyl acetate, and (c) the dissolution of this precipitate in HNO_3 followed by three cycles of solvent extraction with hexone as the solvent and aluminum nitrate as the salting out agent. The recovery of uranium through one solvent extraction cycle is 94%.

The Technical Division reports on investigations on the semiworks scale of the reprecipitation of lanthanum fluoride as a means of reducing the iron concentration at the end of the crossover cycle in the Bismuth Phosphate Process. Nine 100-liter scale experiments were conducted with tracer concentrations of plutonium. The iron decontamination factors Section C-I obtained in the laboratory experiments could not be duplicated because of excessive corrosion of the stainless steel piping by the fluoride solutions. Direct solvent extraction of uranium from an active bismuth phosphate extraction effluent solution with dibutyl carbitol and re-extraction with water has, through three extraction cycles (using aluminum nitrate as the salting-out agent), resulted in gamma-ray decontamination factors of 4.7×10^3 and 6.1×10^3 for two duplicate runs. Overall uranium recoveries appear to be greater than 90% and possibly as high as 98%, based on rough calorimetric observations.

In the evening I attended a 7:45 meeting of the Basic Chemistry, Recovery, and Instrument Groups of Section C-I. Others present were Abraham, Ames, Arnold, Bartell, Cunningham, Davidson, Dawson, Dixon, Florin, Fried, Ghiorso, Hagemann, Howland, Jaffey, Jasaitis, J. J. Katz,

Katzin, Kraus, Larson, Lawroski, Magnusson, Manning, McKinney, Niedrach, O'Connor, Orlemann, S. Peterson, Robinson, Seifert, Sheel, Sheft, Simpson, Studier, Templeton, Van Winkle, Winner, and others (see Fig. 28). Ames reviewed the evidence for various sulfate complexes of Pu(IV). Spectrophotometric examinations of Pu(IV)-HCl solutions have been carried out after addition of various small amounts of H_2SO_4. The data suggest the existence of the two complexes $PuSO_4^{+2}$ and $Pu_2(SO_4)_3^{+2}$, in which the latter complex seems to be completely formed in 0.2 M H_2SO_4. (These complexes are in addition to previously reported negatively charged sulfate complexes in solutions of higher sulfate concentration.)

Fried discussed recent work on reactions of fluorides of uranium and plutonium. Using specially dried oxygen he has been able to demonstrate that, at 800°C, the reaction $2UF_4 + O_2 = UO_2F_2 + UF_6$ takes place. Four to five milligrams of PuF_3 have been treated similarly, yielding a mixture of 80% PuO_2 and 20% PuF_4 at 600°C. PuF_4 does not react at 300°C with oxygen but yields 10% PuO_2 at 600°C. I called attention to the possible importance of the reaction of UF_4 for producing UF_6 without the use of free fluorine.

Jasaitis reported on vapor pressure measurements on plutonium metal carried out in a tantalum crucible containing an accurately measured pinhole under conditions that should supply saturation pressure within the crucible. Difficulty has been encountered because of the formation of a plutonium mirror on the surrounding glass tube below the lower edge of the crucible (reflectivity measured as similar to that of iron), believed to have resulted from seepage of the metal through small pores of the tantalum container. Vapor pressures measured in mm of Hg range from 8×10^{-5} at 1150°C to 1.37×10^{-2} at 1450°C. By extrapolation, a vapor pressure of 760 mm of Hg would occur at 2845°C. Simpson reported some data on counting plutonium samples on plates prepared by vaporization of $PuBr_3$ at a known temperature with the amount deposited varied by changing exposure time. He finds that the counts per minute are not proportional to the amount deposited. Results indicate a loss in counting efficiency with increase of the amount of sample beyond losses due to coincidence and self-absorption. A check with the nitrogen counter has indicated that an unexpected resolution loss cannot account for the observation. Simpson said it is not impossible that the results may have been caused by depletion of material in the crucible since a large number of experiments had preceded these determinations.

During the discussion of unexplained counting effects, Jasaitis described an experiment, the results of which are also difficult to explain. In an attempt to determine the activity distribution of plates prepared by high-vacuum techniques, a narrow (1 mm) slit was moved across the plate; the plate counted with the slit in various positions. It was found that when the slit was close to the edge of the disc a considerable count was observed which did not change very much when the slit was moved closer to the center of the plate. Near the center a large increase in count was then observed caused by exposure of the deposited material.

Scott reported on methods to determine the amount of U^{235} in enriched uranium samples based on analysis for the beta-particle emitting daughter isotopes: (24.6 hours) UY from U^{235}, (24.5 days) UX_1 and (1.1 minute) UX_2 from U^{238}.

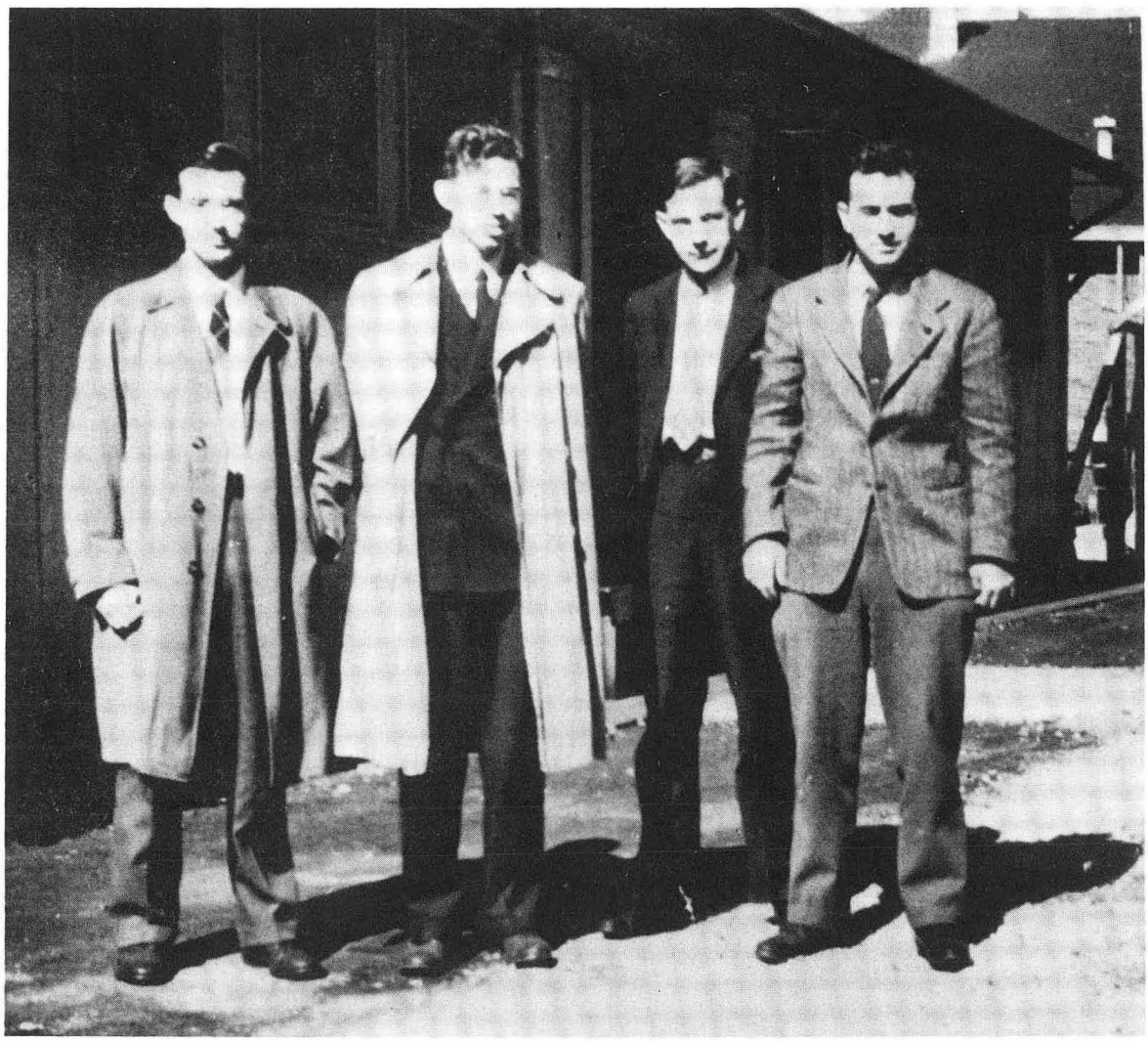


Figure 29. E. K. Hyde, D. H. Templeton, H. P. Robinson, and S. Fried in alley behind New Chem. Autumn 1944.

XBB 769-863

Helen worked at the Met Lab on the secret version of the "Table of Isotopes."

Election returns from yesterday pushed the war out of the headlines today. The voter turnout was large and much more in President Roosevelt's favor than the polls had predicted.

The Project Council Policy Meeting at 9:30 a.m. in Room 209, Eckhart Hall, was attended by Allison, Bartky, Dempster, Franck, Greninger, Hamilton, Jacobson, McKinley, Mulliken, Spedding, Stone, Vernon, W. W. Watson, Whitaker, Wigner, and Zinn. Allison outlined C. M. Cooper's proposed reorganization of the Met Lab which primarily affects the Technical Division. All du Pont men will be leaving within the next two months, with the possible exception of R. H. Jebens who may join the Met Lab staff. There was a discussion of Cooper's proposal to organize an Engineering Division with Cooper remaining as director until early next year, with the possibility of bringing in top engineering talent from M.I.T. or elsewhere — there are to be preliminary discussions with M.I.T. next week. Zinn objected to bringing in men who know nothing of the Project. Allison agreed to get in touch with Compton and point out that there should be a meeting in Chicago before Cooper discusses the subject with M.I.T.

Allison reported that two directives have been received by Compton. First, responsibility for waste recovery of uranium has been assigned to Clinton and responsibility for recovery of U^{233} has been assigned to the Met Lab. The second directive is that Allison's responsibilities at Chicago will end in one week on the 15th and Stearns will take over. Allison is to go to Los Alamos.

Thursday, November 9, 1944

After completing the dichromate oxidation cycle begun day before yesterday on sample 49DDN-3 (derived from the second leaching of plutonium from the 200 mg of plutonium plus deuterons, St. Louis cyclotron bombardment), James carried out an alpha-particle absorption curve on the resulting sample 49DD-N4. He finds that "very long range (emanation?) was present along with other activities and about 30 c/m of Pu^{239} — total of 70 c/m." The supernatant from sample 49DD-N4 was put through an SO_2 reduction, La^{+3} and HF added, and the resulting precipitate mounted as sample 49DD-39J, giving 3490 alpha-particle counts per minute.

I received a memo from Simpson, Seifert, and Erway concerning tests of standard alpha-particle counters to investigate statistical results of Bennett and Johnston that indicate a possible disproportionality in the rate of counting with amount of active material on the counting plates. To prepare suitable plates, $PuBr_3$ has been vaporized onto a series of plates for different known times at a constant temperature. The counting data obtained from the plates give evidence of a loss

of counting efficiency with increase in the amount of the sample that is beyond that caused by self-absorption and coincidences.

Cunningham wrote to English at Clinton Laboratories, stating that the products mentioned in his letter of October 26 to Koshland have been identified by Zachariasen as either $K_2Pu_2O_7$ or K_2PuO_4.

Nickson recommended to Arnold alterations or replacement of laboratory benches or flooring in West Stands Rooms 217, 216 (Blaedel and Walling), 223, 212 (Larson, Hyman, Lincoln, Winner, and Fields) as a means of eliminating persistent contamination with radioactive materials.

Helen went to her chemistry class at YMCA College.

An interesting item on the front page of the newspaper today features an increase in the price of milk from eleven to twelve cents a quart.

Friday, November 10, 1944

I attended a meeting in Room 104, New Chem Annex, to discuss the cooperative effort between Clinton Labs and the Met Lab on solvent extraction. Others present were Arnold, W. C. Johnson, Lawroski, Maloney, and Hogness. It was decided that Clinton Labs would direct their efforts chiefly toward the uranium waste recovery program and that the Met Lab would work on the general problem of extracting Pu^{239} from product solutions. It was recognized that the solvent extraction process might be applied after any one of the steps in the Hanford Bismuth Phosphate Process and might even replace the whole process itself. Therefore, plutonium distribution and decontamination studies were to be made in solutions of (a) lanthanum fluoride-plutonium precipitates, (b) bismuth phosphate precipitates, and (c) in UNH solutions.

At Clinton, sample SA-2, containing 4.4 mg of PuO_2, was removed from the pile. It has undergone neutron irradiation since June 5 of this year and has received 8,475,774 kwh (about 300,000 kw days) of irradiation. Also at Clinton, 15 cans of $ThOCO_3$, specially purified from uranium, went into the pile in a position where there is 1.4 times the average flux.

I received a copy of a memorandum from Daniels to Captain Chapman in the Chicago Area Engineer's office, explaining the importance of U^{233} as a fissionable material and identifying the research programs and cooperative activities of the Met Lab, Clinton Labs, Ames, and Evergreen (Montreal Project). He mentioned the decision to center thorium and U^{233} research at Chicago and indicated that 5.2% of the Chemistry Division's effort is being devoted to U^{233}. This information has been requested by the Area Engineer's office.

Report CS-2316, "Chemistry Division: Summary Report for October 1944," was issued. The material in the report for Section C-I under the general title "Plutonium Chemistry" has been covered in my accounts of

various meetings of Section C-I, memoranda, and reports and will not be repeated here.

Burton's Section C-II report on Radiation Effects contains information on the corrosion of stainless steel by barium chloride, the analysis of helium gas, the effects of neutrons on graphite, the effects of radiation on solid compounds and on water and aqueous systems, and a section describing Burton's work on the effects of radiation on separation processes. This latter section includes the effects of 1.0 Mev electrons at 10 microamperes for 30 minutes on pure hexone, and 1.2 Mev electrons at 2 microamperes for one hour on hexone in equilibrium with blank feed solution. The pure hexone remains colorless and yields only about 0.5% polymer with the 30-minute exposure at 1 Mev. The hexone in equilibrium with feed solution becomes greenish-yellow when bombarded for an hour at 1.2 Mev. Tests to determine the amounts of nitration and decomposition of the hexone are in progress.

Sugarman's Section C-III report on Fission Product Chemistry covers the pile poisoning problem, studies on fission activities and their yields, absorption of active gases such as xenon, studies of fission chains with short-lived members such as Sr^{91}-Y^{91}-Zr^{91}, the production of U^{237} and Np^{237} in the CP-3 pile, the collection of information pertinent to the Bismuth Phosphate Process, and other items. McKinney's Section C-IV report contains an accounting of the number of samples received and analyzed under its Analytical Services work. The Chemistry Division's report is concluded by a short report on Arnold's Section C-VI Pure and Rare Chemicals activities.

Mario Eduardo Báncora of Argentina (working for a year in the Physics Department at Berkeley) had lunch with Helen and me in our apartment. Helen then worked at the Met Lab in the afternoon on the secret version of the "Table of Isotopes."

Top war news in today's paper indicates that the U.S. 3rd Army broke across the Moselle River at two points north of Metz, threatening to outflank this mightiest fortress left to the Germans west of the Rhine.

Saturday, November 11, 1944

Moulton sent me two memos on patent matters. One memo concerns Case S-2268, the case being set up dealing with a "Method of Separating Plutonium from Irradiated Uranium and Fission Products — Alternate Carriers, but without oxidation and reduction cycles being required"; he asks that I review the material and supply the information needed for completion of a Record of Invention. The other memo deals with Case S-1112, "Electrodeposition of Plutonium from Fused Salt Solutions." Moulton said it would appear from references in notebooks 139 and 846 that this work was done with uranium as a stand-in for plutonium. He asks that if there has been any of this work in which electrodeposition of plutonium occurred, I should supply the necessary notebook references.

Hyman submitted to me, at my suggestion, a summary of work to date

on uranium recovery, in view of the directive to discontinue such work at the Met Lab. He describes the most promising procedure as involving three steps: (1) an adsorption-type scavenger precipitate from the carbonate solution of uranium (the type of solution in which the uranium waste from the extraction step might be stored at Hanford), (2) precipitation of the uranium with sodium hydroxide or acetic acid, and (3) dissolution of the precipitate in nitric acid and extraction with a solvent such as dibutyl carbitol or hexone.

I sent Whitaker at Clinton Labs a description of the arrangements made with W. Q. Smith, subject to Whitaker's approval, for two special plant runs at Clinton for recovery of 93^{237}; the runs to be made starting next Thursday. The special runs will require only two changes in the present plant operational procedure. One is the use of a new preextraction treatment: $KMnO_4$ oxidation followed by $H_2C_2O_4$-Mn(II) reduction. This, in laboratory tests, gives losses of neptunium under 15% as compared with losses of 90% and 35% using the regular $NaNO_2$ or $NaNO_2$-$H_2C_2O_4$ preextraction treatment. The second change is the substitution of U(IV) as the reducing agent in the cycle in place of Fe(II); this is expected to reduce neptunium losses from 75% to 20%. I indicate that probably J. J. Katz, A. E. Kelley, and L. B. Magnusson will come to Clinton to isolate the neptunium from the lanthanum fluoride slurry at the end of the first concentration cycle, the composition of which will be 50 grams of lanthanum and 3 grams of plutonium. Smith is to advise us by phone on Monday, a week from now, as to the date on which he wants the men to arrive.

Helen worked on the secret version of the "Table of Isotopes" at the Met Lab.

American forces sank seven Japanese destroyers at the port of Leyte, although the Japanese did succeed in landing some reinforcements at Ormoc.

Sunday, November 12, 1944

I played 15 holes of golf at Jackson Park with Katzin, Arnold, and Lawroski (LK-54, LA-46, SL-49, GS-49 for 9; LK-87, LA-72 for 14; SL-78, GS-82 for 15).

Eight-column headlines in this morning's paper read," Fliers Sink 6 More Jap Warships" and "Patton Smashes Ahead 6 Miles."

Monday, November 13, 1944

Edwin F. Orlemann terminated work at the Met Lab today. He is going to Y-12 in a position of responsibility to help them solve their difficulties in the recovery of enriched U^{235} in the electromagnetic process.

James completed dissolving sample 49DD-N4 and precipitated lead sulfate from the resulting solution (49DD-N4 is derived from the second leaching of plutonium from the 200 mg of plutonium plus deuterons, St. Louis cyclotron bombardment). The precipitate was mounted as sample 49DD-N5 and gives 32 alpha-particle counts per minute. A zirconium arsenate precipitation was carried out on the supernatent from 49DD-N5, mounted as sample 49DD-N6, and gave about 140 alpha-particle counts per minute. A lanthanum fluoride precipitation was carried out on the supernatant from the zirconium arsenate precipitation, mounted as sample 49DD-N7, and gave 15 alpha-particle counts per minute. The remaining supernatant was evaporated to dryness and labeled 49DD-40J.

Margolis sent a memo to G. M. Brown and A. C. Miller about recovery methods for the uranium from semiworks bismuth phosphate extract effluent solutions as an alternative to the present method of disposing of these solutions (including their uranium content) to the sewer. Laboratory tests indicate that the most successful method is one of direct extraction wherein aluminum nitrate is used as a salting-out agent, and the dibutyl carbitol is used as the solvent.

Today's war summaries indicate that U.S. armored divisions slashed at supply lines southeast of Metz, and in the Pacific Americans attacked 45,000 Japanese troops who have been holding the Ormoc line on Leyte.

Tuesday, November 14, 1944

I read a copy of a memo from J. B. Tepe to D. K. Duffey on operation of the 3-inch solvent extraction column to investigate an alternative plutonium isolation procedure for use at Hanford. Duffey is asked to assume responsibility for the remaining construction, testing, and operation of the continuous countercurrent column (located in the center stairwell at West Stands) in place of Buffum who is leaving the Project this Saturday. Tepe identified three phases for the experimental program:

1. Shakedown runs using Hanford concentrations of zirconium and lanthanum but no plutonium.
2. Runs as above, but with tracer plutonium added until good material balances are obtained.
3. One or two runs containing Hanford concentrations of zirconium and lanthanum and about 1/100 Hanford concentration of plutonium (2 g plutonium/50 gal of feed solution). These runs will demonstrate as far as is possible at Chicago the feasibility of the solvent extraction isolation process.

In a memo to Compton concerning Cooper's plan for reorganizing the Technical Division, Allison says he has signed a directive establishing a Metallurgical Division, subject to Stearns' approval, but he indicates there is considerable uncertainty about Cooper's other proposal to

establish a new Engineering Division. Allison expects formal installation of such a division will be delayed until these points have been more fully discussed.

Helen went to her chemistry class at the YMCA College.

Today's banner headline reads "Six-Ton Bombs Sink Tirpitz." The 41,000-ton German battleship was sunk by the British in Tromsø fjord along the north coast of Norway.

Wednesday, November 15, 1944

Stearns has replaced Allison as Director of the Met Lab; Allison is going to Los Alamos.

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Cunningham, Davidson, Dawson, Egan, Ghiorso, Gilbreath, Hindman, J. J. Katz, Katzin, Kraus, Lawroski, Manning, Simpson, and R. C. Thompson. I mentioned that abstracts for the Project Council Chemistry Information Meeting of next Tuesday are due this Saturday. Abstracts for the December meeting are due on Saturday, December 16. I announced that our personnel records as to performance and efficiency have not been adequate. On the secretarial situation, I urged that the work be organized in such a way that the secretaries would not be without work for extended periods of time and very much rushed at other times.

Cunningham proposed another meeting (of Section C-I Council members) in which research rather than administrative matters would be considered; tentative plans were made for such a meeting of Basic Chemistry Group Leaders and a few others. The tentative schedule and speakers for the revised biweekly Monday night Chemistry Division meetings was reviewed. I am scheduled to speak on "The Heavy Elements" at the first two meetings on November 20 and December 4. Weinberg will speak on "Elements of Pile Calculations" on December 18. Other speakers and topics were suggested without assigning dates. On personnel allocations, Davidson and Katz are to maintain liaison with Wigner and his group on conversion piles because of the importance of the chemistry problems involved. Blaedel and Westrum are to help me assimilate information in connection with the continuing chemical problems of Y-12. I announced that the semiworks is now under the jurisdiction of our section.

T. O. Jones has been delegated responsibility for maintaining an overall record of the amounts and disposition of the various isotopes which we now have in our Section C-I laboratories or will have in the future. It is important that no material be shipped from the Laboratory without a record as to where it is being sent and for what purpose. It was suggested that a place be designated in the laboratory notebooks for such a record. A very accurate record will also have to be kept of the history of the samples after they are received in the laboratory. The importance of this cannot be overemphasized, since the slightest carelessness may lead to disaster. Katzin concluded the meeting by mentioning the

health group's concern about absorption of plutonium through the skin via an open wound or abraded skin. He said that the health group is very much worried about absoprtion of plutonium through the skin. In the event that plutonium should come in contact with abraded skin or an open wound, the following procedure is to be followed: (1) flush with water and soap, (2) use no antiseptics, (3) if there is any suspicion of plutonium having been introduced, the matter must be discussed with Dr. Allen at once.

Effective today, the semiworks is under the jurisdiction of Section C-I and will operate as a group in Albaugh's sub-section under the supervision of C. J. Egan. I worked out the details of this in a meeting with Maloney and A. C. Miller yesterday. The other men transferring to Section C-I (or returning) are A. J. Margolis, W. C. Giegold, R. W. Rasmussen, B. R. Wendrow, H. E. Flotow, H. Hasenfus, K. P. Moseley, and R. Van Winkle. A number of other members of the semiworks (I. J. Schaffner, H. E. Fritz, H. Zvolner, J. H. Schraidt, G. W. Cressman, E. W. Quinn, W. Simon, E. A. Hausman) are being transferred to the General Engineering Section as an operating crew for the solvent extraction columns. They will be on loan to this section under the supervision of Lawroski and Albaugh after the first of the year.

I received a copy of a memo from Arnold to Gore referring to his November 1 letter requesting installation of floor covering for West Stands Rooms 212 (Larson, Hyman, Lincoln, Winner, and Fields), 216 (Blaedel and Walling), the Storage Room 23 and the hallway between the rooms. He asks further, that Rooms 24 and 26, which are storage rooms should also have their floors covered, as well as Room 22 which, after November 15, will be transferred from the Technical Division to the Chemistry Division.

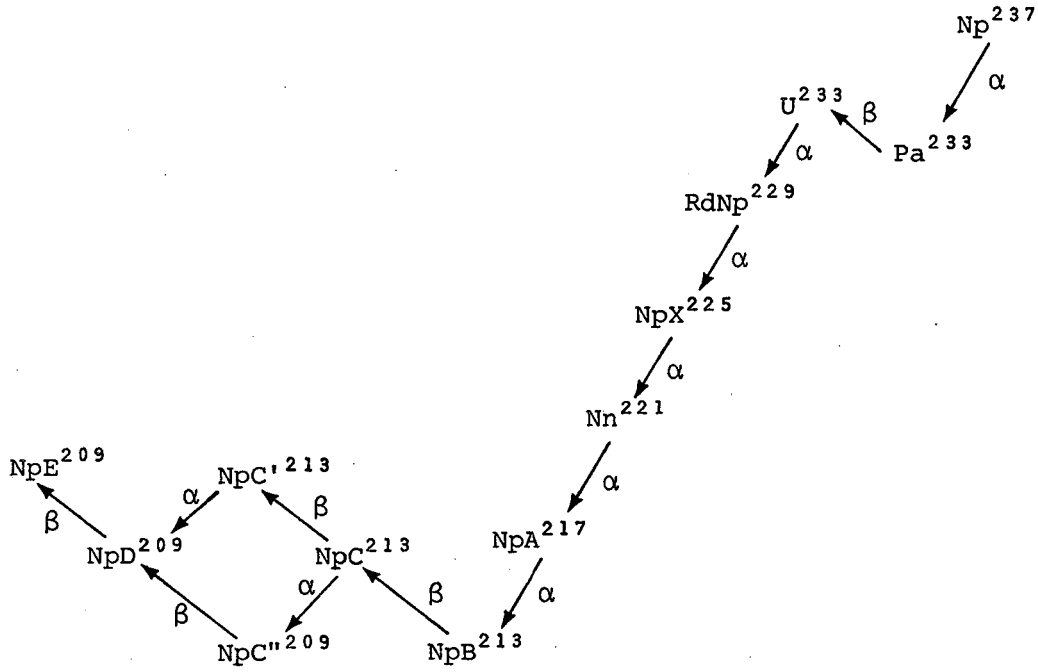
Katzin and Davidson held their first meeting with Wigner, Weinberg, Ohlinger, and Young for the purpose of becoming oriented in the chemical problems involved in the design of conversion piles (U^{233}-fueled, D_2O-moderated piles for the conversion of thorium to U^{233}).

I attended an evening meeting of the Separation Process Sub-section of my section. Studier talked about gamma- and x-ray activity found to be associated with pure U^{233} analogous to that previously reported to be associated with 94^{239}. Aluminum and lead absorption curves have been run on a 16-mg sample purified by ether extraction and by $BaSO_4$ and lanthanum fluoride precipitations. Gamma-rays have been detected at 420, 230, and 150 kev energy, also x-rays of 87, 19, and 14 kev. The presence of electrons, probably arising from internal conversion of the gamma-rays, was demonstrated by magnetic-deflection counting techniques. I offered a probable relationship between the energies of the radiations and the quantum levels in the Th^{229} daughter atoms which are involved.

Hyde reported some exploratory investigations of extraction of uranium and thorium by cellosolves, esters, ketones, and diethyl ether (in connection with U^{233} extraction studies). Hellman spoke on the effect of thorium, aluminum, lanthanum, copper, manganese, and ammonium nitrates as salting agents upon the extraction of uranium and thorium

into ethyl ether. There is a pronounced increase in uranium extraction at comparatively low aluminum nitrate concentrations, indicating its value for preliminary extraction. He said that NH_4NO_3 is indicated as a salting agent for final purification where the amount of thorium extracted is an important factor.

Katzin reported on the probable decay chain for the "neptunium series," of which U^{233} is the third member. Based on preliminary experiments and by analogy with the known series, he postulates the following decay chain:



Katzin then presented experimental evidence in support of that part of the proposed decay chain following U^{233} and indicated the possibility of branching decay leading to isotopes of elements 85 and 87.

Today's newspaper indicates that American troops were within two miles of Metz yesterday.

Thursday, November 16, 1944

I read a copy of a November 15 memo from Zachariasen to Stearns reporting on a sample submitted by Davidson (labeled PuO_2-H_2S) in which Davidson reacted PuO_2 with H_2S. The sample consists of a single phase and the intensity distribution in the x-ray diffraction pattern shows the phase to be Pu_3S_4 and isomorphous with the corresponding cerium compound. Some metal sites may be vacant, however, so the actual composition of the sample may be anywhere in the range Pu_2S_4-Pu_2S_3.

Zachariasen points out that more than six months ago, (CK-1518), he reported the discovery of the very close similarity between plutonium and rare earth elements. He adds that the results given in this and another memorandum (MUC-FWHZ-79) demonstrate this similarity in a most convincing way.

Melvin Calvin wrote from Berkeley that he and his wife, Gen, will come to Chicago on the Streamliner Sunday noon and are planning to stay at the Palmer House. He recalled his earlier letter to me about a liquid-liquid extraction process and said that the method seems to work very well for him. He will bring a sample of the key compound [trifluoro-acetylacetone] so we can work on the process.

Helen attended her chemistry class today.

Today's top headline says that U.S. troops are in the suburbs of Metz. In the north British troops are within 37 miles of the key industrial center of Duisberg.

Friday, November 17, 1944

A request was made by Ghiorso for the services of John M. Dorsey who is working with Wakefield. Horace Hopkins is now a member of the SED.

"Chemical Research — Separation Processes for Plutonium. Report for Month Ending November 1, 1944," (CN-2288), was issued. It contains an account of the following investigations.

Extraction-Decontamination (R. Thompson, Group Leader). *Methods of increasing the rate of metal processing at Hanford.* S. Peterson, Bartell, and Howland have continued work on a method for processing up to four tons of uranium per day in one Hanford canyon. It does not appear possible to obtain consistently satisfactory yields from active solutions at UNH concentrations above 25%-27%. Two "four-ton" runs have been completed through one decontamination cycle from 28% UNH solution and excessive losses have been encountered in the plutonium precipitation step. *Effect of hydrazine in process metal solutions.* Ader, Malm, Bartell, and Greenlee have extended studies to N_2H_2 concentrations as low as 0.0002 M and have found as much as 65% Pu(IV) reduced to Pu(III) and extraction losses of 17%. Satisfactory extraction yields have been obtained after destruction of N_2H_4 by either prolonged heating, $NaNO_2$ treatment, or oxidation with $KMnO_4$ or $K_2Cr_2O_7$, followed by prereduction. *Prereduction studies.* S. Peterson, Ader, and Hoekstra show that in 28% UNH, the maximum concentration of $H_2C_2O_4$ which can be used for prereduction without precipitating $UO_2C_2O_4$ is 0.04 M. Spectrophotometric study of the effect of $NaNO_2$ on Pu(IV) at room temperature over a period of 12 days has indicated no reduction to Pu(III).

Concentration-Isolation (J. Katz, Group Leader). *Investigation of the HEW flowsheet procedure of August 26, 1944, on a laboratory scale.* Kelley has investigated a number of minor changes in the new flowsheet

consisting of the use of different concentrations of HF and $H_2C_2O_4 \cdot 2H_2O$, the use of $KMnO_4$ as a re-oxidant, a 1 N HNO_3 wash of the lanthanum fluoride precipitate, and a double metathesis. Results show that the variations do not yield significantly different results than the old flowsheet, except for the use of the 1 N HNO_3 wash that gives rise to considerable product losses. *Elimination of iron in the metathesis step.* J. Katz finds that the use of sorbitol to eliminate iron is of doubtful utility as it leads to high plutonium losses through the formation of alkali-soluble plutonium-sorbital complexes. *The effect of titanium and cerium in the high acidity peroxide isolation procedure.* Goeckermann finds that these elements interfere with the precipitation of the peroxide. It is unlikely that these elements will be encountered in the chemical processing at Hanford. *The effect of zirconium in the high acidity peroxide isolation procedure.* Beard has carried out additional studies that show that if as much as 1 mg/ml of zirconium is present during the first precipitation, losses in the second precipitation will be excessive. *Carrying of sulfate by plutonium peroxide precipitates.* Hopkins has determined that the plutonium/sulfate ratio in the plutonium peroxide precipitate is roughly 6:1 when the high acidity process is used. *Tests of the high acidity and the flowsheet isolation procedures in Clinton plant material.* Goeckermann and Hopkins have tested both procedures using the best attainable Clinton stand-in for the Hanford lanthanum fluoride slurry. Results have been satisfactory.

Process Development (Gilbreath, Group Leader). *Investigation of possible solvents for the extraction and decontamination of plutonium utilizing the III state of plutonium.* Blaedel and Walling have tested this solvent extraction procedure on a batch-scale using various solvents with hydroquinone as the reducing agent. The solvents which work satisfactorily are hexone, dibutyl carbitol, 2-ethylbutyl cellosolve, diethyl cellosolve, ethylbutyl cellosolve, methyl isobutyl carbinol, and diethyl ether. Unsatisfactory results are obtained with dibutyl carbitol, nitromethane, 2-ethyl hexoic acid, and methyl isobutyl carbinol acetate.

Concentration and Isolation of Plutonium by Solvent Extraction (Dawson and Lawroski, Group Leaders). *Factors that interfere with hexone extraction.* Asprey, Britain, Stewart, and Dawson have conducted a series of investigations to determine why, on occasion, a considerable part of the plutonium fails to extract. It is concluded that in those cases in which extraction is from zirconium-complexed plutonium solutions, the poor results are caused by reduction of the plutonium to the III state due to substances produced when hexone is allowed to stand in contact with HNO_3 for prolonged periods. In those cases in which plutonium is prepared for extraction by precipitation of $Pu(OH)_4$ and dissolution in HNO_3, either polymerization of Pu(IV) or reduction of Pu(IV) to Pu(III) could be responsible. *Batch extraction.* Britain, Fineman, Fields, Stewart, and Dawson have extracted plutonium by batch operations from a solution prepared by dissolving a Room D, lanthanum fluoride-plutonium precipitate with zirconyl nitrate. Two extractions have been made using three cycles in the first extraction and two cycles in the second. An overall yield of 96.8% with 97.5% purity has been obtained which indicates that batch extraction from a zirconyl solution is not as effective as the use of extraction columns. *Preliminary investigation of a continuous countercurrent solvent extraction method for plutonium isolation.* Brody,

Fields, Fineman, Lawroski, Reinhardt, Stein, and Stewart, using the 19-mm ID column system, have successfully reused hexone three consecutive times. Nitric acid assays have been made to determine the distribution of acid in the various effluent streams; it may be necessary to presaturate partially the solvent with HNO_3 to prevent precipitation. The solvent leaving the aqueous column is found to be practically free of acid. Preliminary laboratory investigation has been started on the evaporation of the aqueous column product solution for concentration purposes. The Henry's Law "k" value for hexone appears to be sufficiently high to insure substantially complete removal of this component by atmospheric distillation.

War summaries in today's paper say that six Allied armies strike at the Germans; that Americans seize Japanese lookout islands of Guinea; that Soviet troops are driving northward through Hungary, flanking Budapest.

Saturday, November 18, 1944

The Personnel Office was notified by Irma Saxton, Manning's secretary, of her resignation from the Met Lab effective at once. She wrote from the Southern Pacific Hospital in San Francisco where she is recuperating from head injuries suffered in the wreck of the Challenger on November 8 while on vacation. Johnson, Director of Personnel, replied today by asking her to reconsider and offered to hold the position open for her for any responsible length of time. He mentioned that clerical workers will very likely in the future be paid time and a half for hours over forty which, on the basis of a 44-hour week, would amount to an $8.00 a month increase for her.

Fried and Davidson sent me a memo concerning the reaction of UF_4 with oxygen at elevated temperatures to yield $UF_6 + UO_2F_2$, the yield of UF_6 being 40% of the theoretical or greater. They summarize their experiments and calculations of the free energy equations for the reactions involved. Their conclusions are (1) It is possible to prepare UF_6 from UF_4 in fair yield by the action of O_2 at 800°C. (2) The failure to obtain a quantitative yield of the residue of UO_2F_2 is caused by the volatilization of UO_2F_2 under the conditions of the experiment. (3) It is common analytical practice to convert UF_4 to U_3O_8 by ignition in air. Their results demonstrate that this conversion must be caused by moisture in the air, and they suggest that caution must be observed with respect to free access of moist air, etc., so that some of the uranium is not lost by volatilization as UF_6 or UO_2F_2.

I received summaries of the work of my sub-sections for presentation at the Project Council Information Meeting on Chemistry, scheduled for next Tuesday, as follows:

a. Katzin reports that cans nos. 7-10 of thorium carbonate have been extracted, yielding about 4.5 mg of U^{233} per can. He also describes studies of salting-out action of various nitrates on thorium and uranium, solvents for uranium-thorium separation, the radiations from U^{233} and

the determination of the decay chain of U^{233}.

b. Albaugh reports on work of Sub-section I on separation processes, describing experiments on extraction from 25%-30% UNH, carrying of Pu(III) and Pu(IV) on zirconium phenyl arsonate, preextraction treatment, procedures for isolation of neptunium at Clinton, uranium recovery from process waste solutions, solvent extraction and decontamination methods, and concentration-isolation studies in the continuous extractions columns using hexone.

c. Simpson, Davidson, and Hindman report on the work of Sub-section II, covering vapor pressure of plutonium metal; preparation of samples for alpha-particle range measurements by volatilizing PuF_3 onto polished quartz plates; vapor pressure of uranium metal; preparation of plutonium sulfides; reductions of PuO_2; unsuccessful attempts to prepare a higher nitride of plutonium by heating in a slow stream of NH_3 at 600°C; melting points of $PuBr_3$ and $PuCl_3$; reactions of UF_4, PuF_4, and PuF_3 with oxygen; reaction of UF_4 with silicon; complex ions of plutonium and potentials of plutonium couples; hydrolytic behavior of plutonium; and the chemistry of neptunium.

Dr. Nickson of the Health Division has ordered 24 Army assault masks to be used in the New Chem filtered-air section as a safeguard against breathing plutonium. Twelve are to be worn by the men in Section C-I at their discretion. The other twelve will be kept by the Health Physics Group working in the New Chem filtered-air section for use in emergencies and as replacements.

The Jeffries Committee, appointed by Compton last July to prepare a report on the future prospects of atomic energy, submitted to Compton its 65-page report, "Prospectus on Nucleonics." Members of the committee are Fermi, Franck, Hogness, Jeffries (Chairman), Mulliken (Secretary), R. Stone, and C. A. Thomas. The report begins with a review of the history of nuclear science and the principles of chain reacting piles. With regard to the future of nucleonics, it explores the question of whether there is enough uranium to supply the power needs of the U.S. by nuclear reactors, points to the use of thorium as a supplementary source of power from fission, and describes the possibility of converting hydrogen into helium to give a virtually unlimited source of energy. The report identifies the possibility of nuclear generators as sources of power in deserts, polar regions, on ships, or even in excursions into space. It suggests the production of new heavy elements — new neptunium isotopes or elements beyond plutonium — that may be suitable for use in piles or other nuclear power devices. The report looks forward to the production of radioactive isotopes in piles in large quantities for use in tracer studies and for medical purposes. With regard to the nearer future of nucleonics the report explores: Physics. Nucleonics to be the liveliest part of theoretical and experimental physics with applications of radiation and radioactive tracers to other branches of physics. Chemistry. Study of transuranic elements will be pursued; use of radiation in chemical and physico-chemical processes (replacing catalysts or heat); use of tracers in reaction kinetics — C^{14} for investigating the mechanism of organic reactions; study of fluid flow. Biology and Medicine. Radioactive iodine in thyroid,

tracer elements in studying basic problems of animal and plant metabolism — study of photosynthesis, use of radioisotopes in diagnosis. Metallurgy. Use of tracers to study diffusion, inclusions, distribution of minor constituents. Engineering and Construction. Problems of wear and lubrication (tracers); leak testing with radioactive gases; tracers for tracing flow in sewers or dams; embodying radioactive materials in paints and floor covering to eliminate static; radioactive sources as substitutes for industrial and clinical x-ray machines; illumination by mixing radioactive materials with luminescent material. Agriculture. Study of poisons and micronutrients. Power. Limited amount of available uranium precludes a widespread use of pile power for energy production in competition with coal, oil, or falling water. However, the (by-product) heat from piles built for plutonium production can be utilized for central heating of large areas, thus freeing oil for premium uses. On the other hand, should not conclude that the cost of pile power is necessarily going to be high as compared with other power sources. Pile power may also be used for transportation "while pile-powered interplanetary ships still belong to the realm of scientific day-dreaming, pile-powered battleships or submarines have been considered as likely applications of nuclear power in the relatively near future." Explosives. Changing direction of sea currents, destroying hurricanes, removing danger of earthquakes or volcanic eruptions, blasting of waterways, e.g., Panama Canal. Impact of Nucleonics on International Relations. Danger of a blitzkreig. Need for central authority to exercise control over nuclear power — necessity for all nations to make every effort to cooperate now in setting up an international administration with police powers which can control the means of nuclear warfare. Control of Materials. Should be possible to give considerable scope to free enterprise and still have the government hold a tight rein on the important factors. Postwar Organization of Nucleonics in America. Nation must maintain its leading position in nuclear research and nuclear industry. "There should be government supported nucleonics laboratories having ample facilities for both fundamental and applied research." Without a healthy development of a nuclear industry, nucleonics research will be insufficient to guarantee the leading position of this country. The report concludes with a statement of broad objectives: to stimulate widespread research in nucleonics in the United States, to encourage the development of a free nucleonics industry in the United States, to coordinate the governmental activities in nucleonics with the scientific and industrial developments in this field in such a way as to insure maximum national security, to strive for the establishment of an efficient international supervision over all military aspects of nucleonics.

Suggestions for the accomplishment of these objectives are given as follows: (a) The projects relating to plutonium, U^{235}, and perhaps U^{233} should be prosecuted by the Government, no matter when the war ends, to a point sufficient for military appraisal. (b) The development of the nucleonics industry by private enterprise should be encouraged. The military by-products of the industrial developments should be made available to the government, and the use of government information and patents should be made available to industry so far as the military situation may permit. (c) Scientific education and research should be encouraged in existing university laboratories, and new research laboratories for nucleonics with special facilities should be created at

universities. (d) A suitable agency, with both government and non-government representatives, should be established to guide and coordinate such nucleonics activities as may affect the military or other interests of the nation. (e) Enlightenment of public opinion on the scope and significance of nucleonics should start as soon as possible to bring about realization of the dangers for world security caused by the new scientific and technical developments, and to prepare for decisions which will have to be taken to meet this danger. (f) Cooperation with friendly nations in all these problems — particularly the last named one — should be given serious and prompt attention.

The top war news story in today's paper indicates that U.S. troops are fourteen miles inside the borders of Germany.

Sunday, November 19, 1944

Genevieve and Melvin Calvin arrived on the Streamliner at noon. Helen and I greeted them; they visited us in our apartment, and we then spent much of the day together.

The cost of living has increased 29% since January, 1941. The War Labor Board is going to be backing wage increases, and Senator Claude Pepper is backing a 65-cent minimum wage, according to this morning's Chicago *Sun*.

Monday, November 20, 1944

The "Fermi Special Sample" of plutonium will be discharged from the B pile at Site W sometime today. It will have received about 12 W days of irradiation at three to four times the average Hanford pile reaction flux.

There was a small meeting in my office at which Melvin Calvin described a process for separating plutonium from uranium, fission products, and inactive impurities. The process depends upon the use of trifluoroacetylacetone that forms a chelate complex with Pu(IV). This complex is soluble in benzene and other organic solvents but largely insoluble in aqueous solutions of low acidity. Calvin has carried out a limited number of batch extraction experiments wherein more than 95% of the plutonium was extracted with a decontamination factor of about 50. Five repetitions should give complete decontamination.

Jaffey sent a memo under my name to Hilberry, attention Perlman, at Hanford, transmitting circuit diagrams of the preamplifier and amplifier for the fission counter as requested.

A 4.4 mg sample of neutron-irradiated Pu^{239} oxide arrived from Clinton yesterday. This is sample SA-2 which was prepared by Ralph James on May 24 and irradiated in the Clinton pile from June 5 to November 10,

receiving 8,475,774 kwh, or about 350,000 kw-days of irradiation. It has been designated sample 49NC by James.

In the evening I was the speaker at the first of the revised biweekly Chemistry Division Monday meetings, held at 7:45 p.m. in Room 251, Ryerson Hall. In the first of two talks on "The Heavy Elements," I discussed the nuclear properties of elements from atomic number 80 to 96, with emphasis on elements 89 to 96. Aspects covered were characteristics of heavy nuclei, energetics of radioactivity, systematics of type of radiation and half-life, neptunium series, neutron fission in the "Big Three" (U^{233}, U^{235}, and Pu^{239}), and prospects for useful nuclei other than the "Big Three." The talk included a description of our recent work on the decay of the 4n + 1 series (see Fig. 29). This also includes the 4n, 4n + 2, and 4n + 3 decay series and suggests the nomenclature of classical radioactivity for the new 4n + 1 series. I included a description of the recent observation by James and Ghiorso of an isotope of element 96 produced by helium ion bombardment of Pu^{239} in the Berkeley 60-inch cyclotron. I said this isotope, which has a half-life of a few months, decays by the emission of alpha particles with range 4.6 cm in air, and exhibits chemistry characteristics of the +3 oxidation state of a rare earth, may be 96^{242} produced by the reaction $Pu^{239}(\alpha,n)96^{242}$. I also predicted that there is a good chance that Pu^{241}, produced by successive neutron capture in Pu^{239} [i.e., by $Pu^{239}(n,\gamma)Pu^{240}$ and $Pu^{240}(n,\gamma)Pu^{241}$] may be a beta emitter, leading to the possibility of observing the daughter isotope, 95^{241}.

Sports news appeared on the front page of today's paper as the Green Bay Packers gained the Western Division championship of the National Football League on Sunday.

Tuesday, November 21, 1944

At 9:30 a.m. I attended a Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Adamson, Allen, Arnold, Burton, Calvin, Capt. Chapman, Compton, Daniels, Dempster, Doan, English, Franck, Glendenin, Hogness, Huffman, Jeffries, W. C. Johnson, Capt. Karl, Latimer, Manning, McKinney, Nickson, Pratt, Rabinowitch, Smyth, Spedding, Stearns, Stone, Sugarman, Szilard, Tepe, Turkevich, Wakefield, Warner, Watters, Whitaker, Wigner, Zachariasen, and Zinn. Compton opened the meeting with the announcement that upon a suggestion of Allison a group of senior officers is to be formed to preside at the Information Meetings. He then called upon the Clinton Chemistry group to present the first reports.

English, representing Clinton Chemistry Section C-I, reviewed basic plutonium chemistry work and mentioned the evidence for tetravalent plutonium in plutonium peroxide. He also described the studies of potassium plutonates. In order to account for the comparatively low percentage of plutonium found by analysis, one has to assume the presence of 5 or 6 molecules of water. To check this, drying experiments have been made. The increase in plutonium content was such as to indicate reduction of plutonium in addition to dehydration. English also covered

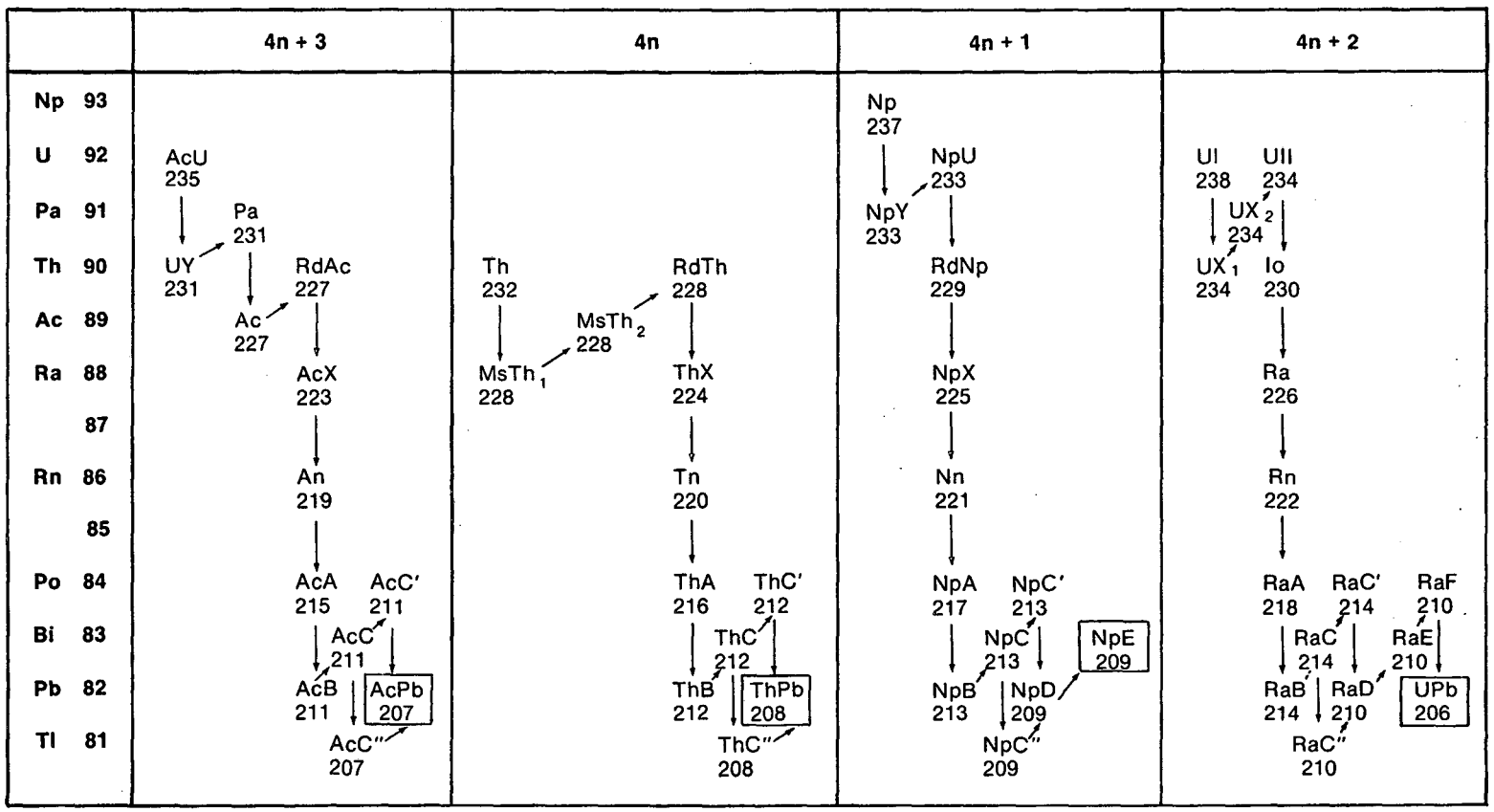


Figure 30. The four radioactive decay series. November 1944

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work on process studies — decontamination and extraction. English was followed by Glendenin (Section C-II) and Adamson (Section C-III) of the Clinton Chemistry Group.

Compton next called on Latimer to take over the chair. Latimer invited reports from the Chicago Chemistry Division. Sugarman (Section C-III) and Burton (Section C-II) were the first two speakers. Sugarman reported that Turkevich, using the Argonne P9 pile, has determined the cross section of the reaction $U^{236}(n,\gamma)U^{237}$ to be less than 150 barns. I followed them by reporting on the work of three sub-sections or groups in my section — (1) Hanford problems [Albaugh], Basic Chemistry [Cunningham], and the Uranium-233 group [Katzin].

Hanford Problems. The work on separation of neptunium was described, how to get rid of the element more completely than is achieved in the present procedure or how to keep it in the plutonium fraction as fully as possible — whichever may be desired. Increasing the yield of neptunium by substituting oxalate + Mn(II) for $NaNO_2$ in the prereduction step and by substituting uranous for ferrous salt as reducing agent in the decontamination cycles was described. I explained that precipitation of peroxide carries the neptunium well and that separation from plutonium can be achieved by dissolving the peroxide in 1 N sulfuric acid containing NaOCl and precipitating the plutonium as K_2PuF_6.

I reviewed solvent extraction for isolation, mentioning that 14 new runs have been made with a 19-mm column in the study of the hexone process. In runs lasting 8 to 24 hours with 1.5 to 6.5 liters of feed solution, the recovery of plutonium is 99.5%, with the plutonium being 99.9% pure. I noted that the large glass column (3 inch diameter × 28 feet high) is completed and ready for operation. I mentioned that a complete decontamination process by solvent extraction is also under consideration. One may, for example, use UNH solution with NH_4NO_3 addition directly for the treatment with hexone. The solvent extracts uranium(VI) and Pu(IV) or (VI). The latter can be reduced to Pu(III) by hydroquinone and extracted with water.

Basic Chemistry. I stated that we have determined the melting point of $PuCl_3$ ($750 \pm 5°C$) (in good agreement with Los Alamos values). The oxide $PuO_2 \cdot xH_2O$ is found to react with H_2S, giving Pu_2O_2S after one-half hour at 1300°C and $Pu_2S_3 + Pu_3S_4$ after two hours at about 1400°C; these reactions are analogous to those observed at Berkeley with cerium.

I described the study of the reduction of PuO_2 with hydrogen, which requires the presence of platinum. Probably Pu_2O_3 is the first product which by dismutation gives PuO_2 and plutonium metal. The metal alloys itself with the platinum, which explains the influence of the latter on the reaction. Zachariasen interrupted my talk to mention that the actual composition of samples obtained so far is not exactly Pu_2O_3, being between it and PuO_2. He agreed with me that Pu_2O_3 probably exists, but has not yet been obtained in a pure state.

I mentioned our further studies of the reaction between PuF_3 and oxygen and our interesting method of producing UF_6 by the reaction of UF_4 with oxygen. I noted the preparation of neptunium peroxide, Np(IV) hydroxide, and an oxidized neptunium hydroxide formed by the action of 0.1 M $HBrO_3$ in 1 M H_2SO_4.

Uranium-233 Studies. Finally, I reported on our systematic study of the influence of salting-out agents on thorium and uranium solutions. I indicated that best results are obtained with calcium nitrate, ammonium nitrate (the old stand-by), and aluminum nitrate. I also mentioned our survey of solvents.

I described the gamma-rays and x-rays we have found for U^{233}. (Here I showed the slide [Fig. 29] of the four radioactive decay series that I had presented last night at the first biweekly Chemistry Division meeting.) I stated that: "The new radioactive disintegration chain of which U^{233} is a member, has now been almost completely identified. It starts with Np^{237}, which is transformed by alpha emission into Pa^{233} which in turn gives U^{233} by beta emission. U^{233} is converted by alpha emission into Th^{229} which has a half life of 10^3-10^4 years. From the growth of activity of isolated Th^{229}, which has now been followed for several weeks, it is clear that an element with a lifetime of at least several months occurs in the disintegration chain below Th^{229}. The alpha activity of Th^{229} contains at least five different groups, which is consistent with the assumption that the stable final product is Bi^{209}. The daughter element of Th^{229} is Ra^{225}. The latter emits alpha particles (perhaps also betas) and gives a short-lived emanation (86^{221}). In the figure, the 'neptunium series,' which may also be called '(4n + 1)-series' is represented tentatively alongside the three old established series, the uranium-(4n + 2), thorium-(4n), and actinium-(4n + 3) series."

Jeffries took over the chair from Latimer so that Latimer could present the report of the California chemistry group. He reviewed the thermodynamics of dismutation of plutonium and studies on plutonium peroxide. Calvin, the other California speaker, presented data on the magnetic susceptibility of Pu(III), (IV), and (VI) which indicate that the valence electrons of these oxidation states are 5f electrons. He described plutonium extraction by chelation using trifluoroacetylacetone which forms a complex with plutonium containing four molecules of the chelating agent around each atom. Benzene has been used as the organic solvent, an accidental choice. In a few preliminary experiments, batch extraction has given 95% extraction of plutonium with a decontamination factor of about 50. I pointed out that the chelation process can be used to determine tetravalent plutonium, which alone is extracted in the presence of plutonium ions of other valences. The meeting ended with Spedding reporting on activities of the Ames Project, emphasizing their recent work on producing metallic thorium and beryllium in quantity and Th-Be alloys.

James began dissolving sample 49-NC consisting of 4.4 mg of PuO_2 that has received about 350 megawatt days of neutron irradiation in the Clinton pile as sample SA-2.

I received a letter from Kennedy at Site Y concerning our patent problem. He says that Segrè and he have been following the earlier suggestion that the two patent cases be filed at once by us pending final arrangements as to their disposition. They find it unncessary to hire a lawyer merely to get the cases on file since the cases will not even be considered by the Patent Office until security restrictions are lifted. He expects soon to have the cases in a form suitable for submission and

asks if I am in agreement with this procedure (Wahl agrees). The Patent Office fee for the claims for the two cases will total $104, and Kennedy suggests dividing this expense among ourselves.

Cunningham answered E. R. Russell's request for a list of names of members of Sub-section II of Chemistry Section C-I who are exposed to active materials, the types of material handled, and the room location of the various individuals. The names are as follows:

Group 5
O. C. Simpson, Group Leader
N. R. Davidson, Assistant Group Leader

Individual	*Material handled*	*Room Number*
B. M. Abraham	Pu^{239}	2
N. R. Davidson	Pu^{239}	2
N. D. Erway	Pu^{239}	25,26
S. M. Fried	Pu^{239}	41
Z. Jasaitis	Pu^{239}	25,26
H. Peterson, Mrs.	Pu^{239}	2
T. E. Phipps	Pu^{239}	25,26
H. Robinson	Pu^{239}	41
I. Sheft	Pu^{239}	2
O. C. Simpson	Pu^{239}	25,26
R. L. Seifert	Pu^{239}	25,26
H. Thomson, Mrs.	Pu^{239}	41
E. F. Westrum, Jr.	Pu^{239}	41

Group 6
J. C. Hindman, Group Leader
K. A. Kraus, Assistant Group Leader

Individual	*Material handled*	*Room Number*
D. Ames	Pu^{239}	10
H. Billington	Pu^{239}	10
J. Dixon	Pu^{239}	11
A. Florin	Pu^{239} and fission activity (β and γ)	4
J. C. Hindman	Pu^{239}	10
J. Howland	Pu^{239}, U^{238}	11
R. James	Pu^{239} and fission activity (β and γ)	4,13
K. A. Kraus	Pu^{239}	10
T. La Chapelle	Np^{239}, Np^{237}	13
K. McLane	Pu^{239}	11
L. Magnusson	Np^{237}, Np^{239}	13
P. O'Connor	Pu^{239}	11

Group 7
L. R. Dawson, Group Leader
D. C. Stewart, Assistant Group Leader

Individual	*Material handled*	*Room number*
H. H. Anderson	Pu^{239} and fission activity (β and γ)	35
L. B. Asprey	Pu^{239} and fission activity (β and γ)	35
J. W. Britain	Pu^{239} and fission activity (β and γ)	34
L. R. Dawson	Pu^{239}	37
P. Fields*	Pu^{239} and fission activity (β and γ)	212 West Stands
P. Fineman	Pu^{239} and fission activity (β and γ)	34
L. Leventhal	Pu^{239} and fission activity (β and γ)	34
D. C. Stewart	Pu^{239} and fission activity (β and γ)	35

Group 8
A. Ghiorso, Group Leader
A. H. Jaffey, Assistant Group Leader

Individual	*Material handled*	*Room number*
J. Crawford	Pu^{239}, U^{233}, Po	16B
A. Ghiorso	Pu^{239}, fission activity (β and γ), U^{238}	16C
D. Hufford	Pu^{238}, Pu^{239}, U^{233} U^{235}, Cb^{93}, S^{35}, P^{32}	7
A. H. Jaffey	Pu^{239}, Cb^{93}	16B
E. Lewis	Cb^{93}	
B. Scott	Pu^{239}, U^{238}, U^{235}, Po	7
B. Weissbourd	Pu^{239}, Pu^{238}, U^{233}	19

*Unusually heavy exposure to β and γ radiation.

Helen attended her chemistry class at YMCA College.

Today's double headline reads "Belfort and Metz Seized; Reach Rhine at 3 Points."

Wednesday, November 22, 1944

Jane Horwich has been hired as a secretary to replace Kathryn Buehler. She has a B.A. from the University of Chicago.

Edrey Smith, my secretary began a two-week vacation today. She will spend part of the time preparing for her wedding and then will honeymoon before returning to work December 5.

At 8:00 a.m. I held a meeting in my office of the Council of my section, attended by Cunningham, Dawson, Egan, Ghiorso, Gilbreath, Jaffey, J. J. Katz, Katzin, Kraus, Lawroski, Manning, Schaffner, Simpson, Stewart, and R. C. Thompson. I gave a resume of yesterday's Project Council Meeting covering the following topics: *Absorption cross section of* U^{236}*:* The cross section for the formation of U^{237} from U^{236} appears to be less than 135 barns. *Poison chain:* The cross section for Xe^{135} is of the order of 3 megabarns. The fission yield according to Sugarman is about 5%. The 27-year cesium is no longer considered a part of the chain. A long-lived noble gas activity of half-life greater than 2.5 years has been detected, presumably a krypton isotope. *Peroxide:* Clinton has determined the formula to be Pu_2O_7 or $Pu_2O_6 \cdot SO_4$ if precipitated from sulfate solution. Berkeley has advanced the formula $PuO_{3+}(NO_3)_{\frac{1}{2}} \cdot xH_2O$. *Potassium plutonates:* Clinton has prepared a series of compounds varying in formula from $K_2Pu_2O_7$ to K_2PuO_4 with varying amounts of H_2O. *Disproportionation of Pu(IV):* It is presumed to take place according to the equation: $3Pu(IV) + 2H_2O = 2Pu(III) + PuO_2^{++} + 4H^+$. This disproportionation is forced to the right with increasing temperature and to the left with increasing hydrogen ion concentration. The delta H for this reaction is 43,000 calories and delta S is equal to +134 E.U. *Trifluoroacetylacetone as an extraction procedure for plutonium:* Calvin's work was described. Gilbreath is to check the decontamination factors that may be achieved. *Magnetic susceptibility measurements of Pu(III), (IV), and (VI) in* HNO_3 *solution:* Calvin's preliminary experiments give results that are consistent with the magnetic moment arising from magnetic spin and orbit contribution. The results are consistent with 5f electrons and not with 6d electrons.

I attended an evening meeting of the Basic Chemistry, Recovery, and Instrument Groups of Section C-I, at 7:45 in Room 209, Eckhart Hall. Present were Ames, Arnold, Bartell, Cunningham, Daniels, Dawson, Davidson, Dixon, Fried, Hagemann, Howland, Hufford, Hyde, Jaffey, Katzin, Kraus, Krueger, Lawroski, Magnusson, Manning, McKinney, McLane, Norton, O'Connor, S. Peterson, Phipps, Robinson, Sheft, Simpson, Stewart, Van Winkle, Warner, Westrum, and others. Jaffey reported on the problem of standardizing absorbers that are used for range measurements of alpha-particle radiation. Since it is difficult to find a monochromatic long-range alpha-particle radiation, it has been decided to use a soft beta-particle radiation and thus obtain a set of values for the absorbers relative to each other. McLane summarized the existing data on transference experiments which have been collected by him and Dixon.

Kraus reported on a number of experiments and the rate of depolymerization of polymeric Pu(IV) solution. Heated polymeric solutions depolymerize about ten times more slowly than unheated solutions. In 6 M HNO_3, 50% depolymerization requires 800 minutes for a polymer which has been heated and 80 minutes for a polymer which has not. Both heated and unheated polymeric solutions can be rapidly oxidized in 1 M HNO_3 by Ce^{+4} at room temperature. Davidson described the synthesis of Pu_2O_2S and Pu_2S_3-Pu_3S_4 by treatment of PuO_2 in a heated graphite crucible with H_2S.

Plans for a meeting of the "Big Three" appear to have hit a snag so that early 1945 is the first possible date for such a conference, according to today's paper.

There was a Project Council Policy meeting at 9:00 a.m. in Room 209, Eckhart Hall, attended by Bartky, Chapman, Chipman, Compton, C. M. Cooper, Daniels, Dempster, Doan, Franck, Greninger, Hilberry, Hogness, Howe, Huffman, Jacobson, Jeffries, W. Johnson, Latimer, Leverett, Mulliken, Pratt, Smyth, Spedding, Stearns, Stone, Szilard, Tracy, Vernon, Warner, Whitaker, Wigner, and Zinn. Compton pointed out that the laboratories of the Metallurgical Project are entering into a new phase. The contribution to Site W is now almost complete. For Site Y we can still be of some help, but we have no major responsibilities. Hence, the main part of the task has been completed. We all appreciate, however, that there is before us still a large effort to provide for the future strength of the nation in this field.

Compton asked that those present consider whether the work from here on should continue to be organized around the immediate future or whether some other objective should be followed. In the discussion of possible objectives that followed, Jeffries pointed out that the objective of Chicago has been achieved only in part since plutonium does not have all the desirable properties that had been anticipated. The same apparently holds for U^{235}. Perhaps, then, the only way to provide adequate insurance that the objective really be achieved would be to be ready to convert Pu^{239} over to U^{233}. This could provide a clear-cut objective.

Compton summarized the discussion, stating that (a) The primary Pu^{239} objective has reached the point where we should advise the Army that our part of the 49 task is largely complete. (b) There is general agreement that a large amount of work in utilization of nuclear energy is still before us, such as hexone extraction process development and conversion of 49 and 25 to 23. A discussion of pressing the work on U^{233} followed, and Compton indicated that agreement by the Army to proceed with moving and setting equipment for a metal fabrication laboratory would have to wait until after his meeting with Colonel Nichols this week.

On the matter of Project reports, Jeffries indicated his committee's report on future possibilities in the field of nucleonics, submitted in draft form to Compton last Saturday, will be ready for issuance on a limited distribution basis within the next few days. Mulliken reviewed the plans for the writing of a series of survey volumes for publication and distribution within the Project. He asked for cooperation of Lab and Division directors in making available people for writing and compiling. It was agreed by all present that this work is a proper part of the completion of Project responsibility for 49.

Thursday, November 23, 1944

I phoned Hilberry at Hanford to discuss procedures to follow in obtaining 25 mg of a plutonium sample — one of the "Farmer's Specials" — being irradiated at Hanford. [Farmer: code name for Fermi.] The sample

was removed from the pile this week and will be ready for shipment in 2-3 weeks. There are also four samples of more than 60%-enriched U^{235} in the pile, but there has been no discussion yet [at Hanford] about our receiving part of one or more of these.

Katzin and Davidson attended the second meeting on the design of the homogeneous U^{233} pile, conferring with Wigner, Young, Weinberg, and at times, Ohlinger and Szilard. Problems discussed included handling and cooling the thorium absorbing blanket, production of water vapor and gases in the reacting unit, mechanism of ridding of fission product gases, cooling of the reacting solution, effect of heat and energy release on the materials, e.g., solvents used for processing the highly concentrated fuel solution, without preliminary cooling.

At 1:15 p.m. Katz, La Chapelle, Magnusson, and Beard left on a trip to Clinton Labs to work on the recovery of neptunium from two special plant runs made under conditions which increased the yield of neptunium. They should be back December 6.

Helen and I had Thanksgiving dinner with Melvin and Gen Calvin. We spent part of the day in our apartment. He told me about sources of supply for trifluoroacetylacetone — it, or the intermediate ester, can be obtained from Columbia-Oregon Chemicals, Inc., Columbia, South Carolina, or from the du Pont Company, Jackson Laboratories.

Today's headline reads "Three Nazi Citadels Fall." On this Thanksgiving Day the stage is being set for the crossing of the Rhine.

Friday, November 24, 1944

I wrote to A. H. Dahl at Y-12 in Oak Ridge about investigations by B. F. Scott and A. H. Jaffey in Section C-I on samples of enriched uranium, showing that it is possible to determine the isotopic composition (U^{235} to U^{238} ratio), as well as the absolute U^{235} content by beta-particle counting of the UY and UX daughter activities, combined with chemical separation in some variations of the method. The letter was five pages long with six figures attached.

I answered Kennedy's letter which I received last Tuesday. I told him Foster York has spoken to Lavender by phone and that, in the conversation, Lavender said he is waiting for our definitions before he can act. York has recommended that one of us write to Lavender and point out to him again the obvious fact that we consider the disclosure and claims in the cases we sent him to be our definitions. York suspects Lavender has not had time to look at the cases and has left it to an assistant who has bungled it. York also said that Lavender has asked the University of California to submit whatever data they have in regard to their claim, if any, to the title to these inventions.

I told Kennedy that I agree to submission of the cases as proposed in his November 17 letter without waiting for the above-promised action by Lavender since we have waited so long and patiently to no avail. I

mentioned that I do not have a set of the photostatic copies of the notebook data to go with my copy of Case 52 and suggested that for completeness perhaps I should have such a set.

In a memo to Spedding at Ames, Daniels asks for 50 grams each of uranium monocarbide and uranium dicarbide in the form of pellets. The samples are to be irradiated in the pile and then heated to 2000°C.

The Metallurgical Laboratory "Report for October 1944," (MUC-SKA-847), was issued by Allison's office. The summary section highlights the Hanford pile poisoning problem, the reorganization of the Technical Division because of du Pont's withdrawing its men, and the decision to transfer a large fraction of the remaining physicists at the Met Lab to Site Y. The Cyclotron Section has been closed down, and Dr. Creutz's section of the Physics Division is almost completely disbanded. Allison is to be transferred to Los Alamos, and Stearns will take over as the new Laboratory Director here.

The portion of the Chemistry Division's section of the report pertaining to Section C-I is as follows:

"The isolation of product by hexone extraction continues to be very promising. Several additional tests were made on the small column, including one 24-hour run, and the same high recovery (99%) and purity (99.9% for W concentrations) were obtained. The large 3-inch column, 28 feet high, has been completed and its mechanical features are being tested.

"Some of the secondary problems in connection with hexone extraction are now being studied, such as the possibility of reduction of Pu to the III state by material formed from the action of the nitric acid on the hexone, and the accumulation of impurities in the concentrated final solution after evaporation. In order to obtain the 30% Pu nitrate in nitric acid ready for shipment, it is necessary to concentrate the final aqueous solution by 50- or 100-fold, and this naturally leads to a large increase in the concentration of ammonium salts and other impurities.

"It is realized that isolation of product will be going on at W long before the continuous hexone-extraction column can be tested at X, and so the use of hexone in batch extraction has been studied. In case there is trouble from Fe or some unforeseen cause, it appears likely that batch extraction of $La(NO_3)_3$-product solution with hexone can be successfully substituted for the peroxide precipitations without appreciable alteration of the existing equipment.

"The hydrazine difficulty in the extraction step of the $BiPO_4$ process can be solved by adding an excess of $KMnO_4$ to oxidize the hydrazine and Pu(III) and then reducing any Pu(VI) which may be formed to the IV state with oxalate and Mn^{++}.

"The formation and destruction of Pu(IV) polymer in dilute acid solutions has been studied in order to understand the factors involved in the large-scale recovery of product. These studies have practical application to the hexone concentration-isolation process, since the polymer is not extracted by hexone. Whenever the Pu hydroxide is dissolved in acid there is some formation of the polymer; this is probably connected

with the behavior of Pu(IV) in the dilute acid solution which surrounds the precipitate as the concentrated acid becomes neutralized.

"The formation of complex ions between sulfate ion and Pu(IV) has been studied further. Stability constants for three different complex ions are being determined. The coprecipitation of Pu(III) with $BiPO_4$ from perchloric acid is poor in the presence of hydrazine, but the addition of sulfate ion restores the ability of $BiPO_4$ to carry down the Pu. These studies are designed to give further information concerning the proper conditions for the precipitation of Pu with $BiPO_4$.

"Accurate vapor pressure measurements have now been completed on Pu metal, PuO_2, Pu_2O_3, PuF_3, $PuCl_3$, and $PuBr_3$. The apparatus is arranged with magnetic shifters and sealed-off compartments so that twenty and more single determinations can be made on the material vaporized at high temperatures out of the tantalum crucible, using only a single evacuation. The melting points have been determined directly and checked with calculations from the vapor pressure data. These data are of value in the production of metallic Pu.

"The study of the formation of certain inorganic compounds of Pu at high temperatures and in the vapor phase is proceeding in order to obtain information of use in preparing the metal Pu.

"The supply of several milligrams of pure U^{233}, which has been made available for the first time, has opened up a whole new field of exploration in radioactivity. It is now clear that U^{233} starts a decay chain leading to a whole set of new isotopes of the heavy elements which have been missing gaps in the isotopic chart. It is now possible with pile-produced U^{233} to fill in these gaps and work has been started on determining the lives and properties of these new isotopes.

"The sample of U^{233} purified in the Chemistry Division has been used at the Argonne Laboratory for the determination of the cross section for neutrons and the fission yield. Fission chains can be produced from U^{233} as well as from U^{235} or Pu^{239}, but the details will be given in reports from Argonne. Solvent extraction methods are being perfected for the extraction and purification of U^{233} from irradiated thorium."

The report also includes the following information. The Chemistry Division began October with 221 technical employees. Of these, 158 were academic grade employees. During October the Division lost 11 academic employees and gained one, resulting in a total of 148 academic employees at the end of October. Overall, the Division had 191 technical employees at the end of October, reflecting a loss of 30 technical persons.

The total Met Lab employment as of October 31, 1944, is 1676 as compared with 1740 as of September 30, 1944. Of these numbers, the total technical employment was 655 for October 31, 1944 as compared with 690 for September 30, 1944. The distribution of technical employees between Divisions and Offices is as follows:

	As of 9-30-44	*As of 10-31-44*	*Ceiling*
Project Director's Office	2	2	
Laboratory Director's Office	3	3	
Chemistry	221	191	
Health	167	173	
N.R.C.C.	2	2	
Physics	138	129	
Technology	141	140	
Information	10	10	
Patent	6	5	
Total	690	655	916

The total Met Lab expenditures for the month of October are $1,085,634.40 as compared with $802,387.28 for September, an increase of $283,247.12 accounted for by a large payment to du Pont for loaned employees' services and an increase in disbursements for the regular payroll.

In the evening I attended the Smith-Albaugh wedding rehearsal in Bond Chapel at the University of Chicago. I am to be Fred's best man. After the rehearsal we all went to the home of Edrey's parents, Mr. and Mrs. Royal Smith, at 9514 S. Damen Avenue for refreshments.

French tanks have smashed into Strasbourg, according to today's headlines.

Saturday, November 25, 1944

Milton Ader, who is being drafted, terminated at the Met Lab today. He is expected to return shortly as a member of the SED. Eda Kelley, one of our secretaries, resigned today because of ill health.

I had a conference with Dancoff and Koch regarding the preparation for them of a sample of Pu^{239} for neutron measurements. The sample is desired by December 16 and is to consist of 100 mg of plutonium distributed over an area of 200 cm^2 on an aluminum plate to come to us from our shop in Ryerson by December 9. The plate will be 13" × 13" with a plated area in the center of about 6½" × 6½".

Initial operation of the 3-inch diameter glass pipe continuous hexone extraction columns took place today.

I took part, as best man, in the wedding of Edrey Smith and Fred Albaugh, held at 3:00 p.m. in Bond Chapel of the University of Chicago, immediately followed by a reception at the Windermere East Hotel. Others in the wedding party were Edrey's parents; Margaret Smith (Edrey's sister, maid of honor); Helen Fenn Ritter (Edrey's cousin, bridesmaid); Robert Harrison (Edrey's cousin, usher); and Tom Morgan (usher). Mildred Hartough was the soloist (see Figs. 31 and 32).

Figure 31. Smith-Albaugh wedding party. Helen Fenn Ritter, Robert Harrison, Tom Morgan, Edrey Smith Albaugh, Fred Albaugh, Margaret Smith, and Glenn Seaborg. November 25, 1944.

XBB 7810-13186

Figure 32. Fred and Edrey Albaugh. November 25, 1944.

XBB 7810-13187

Fires are burning in Tokyo after an attack by B-29 bombers, according to this morning's paper.

Sunday, November 26, 1944

The front page of the Chicago *Sun* today features an article headlined "U.S. Assails Nazi Murder of Civilians." The U.S. government is charging that the Germans have "deliberately and systematically murdered millions of innocent civilians — Jews and Christians alike — all over Europe."

Kenesaw Mountain Landis, popular Commissioner of baseball, died.

Ohio State University won the Big Ten title by defeating the University of Michigan, 18-14, yesterday — an undefeated and untied season.

Monday, November 27, 1944

James completed a nitric acid oxidation cycle on sample 49NC (4.4 mg of PuO_2 that has received about 350 megawatt-days of neutron irradiation in the Clinton pile). The lanthanum fluoride precipitate has been mounted as sample 49NC-2 and is found to contain 50.36 micrograms of plutonium; it is also very active with respect to beta-particle and gamma-radiation. The supernatant, containing the bulk of the plutonium (oxidized), has been turned over to Paul O'Connor.

I received a handwritten letter from Allison in Los Alamos thanking me for my signature on the round-robin letter given him just before he left the Met Lab. He expressed the hope that he soon can get back to a small-scale job that needs less psychology and more physics.

Manning received a letter from T. O. Jones, who is recovering from a recent intestinal operation in Oshkosh, Wisconsin, saying that he is not yet able to come back to work. He has a lot of pain for which he is taking morphine, and his doctor wants him to stay on longer. He hopes to be back by December 2 or 3.

I sent Stearns the text of a proposed letter for him to send to Compton requesting that we be given about 25 mg of plutonium from the first of the "Farmer's specials," as discussed by Fermi, Hilberry, and Perlman; also that we receive 25 mg samples from each of the three Farmer's specials following the first one. I explained to Stearns that the preparation of such a draft letter was suggested by Hilberry.

Dempster and I talked by phone about the preparation of plutonium samples to be used for mass spectrometer identification of Pu^{240} and also the preparation of U^{233} samples to be used to determine their isotopic composition.

Daniels sent a memo to Stearns about the Chemistry Division progress outline for November, summarizing work in Section C-I on (1) modifying the Hanford Process to permit handling more metal with the present equipment, (2) best method for insuring that plutonium is in the IV state in the extraction step, (3) complex ions of Pu(IV) and Pu(VI), (4) chemistry of neptunium, (5) hexone for solvent extraction and decontamination, (6) California work on trifluoroacetylacetone, (7) plans to make alpha-particle range and specific activity measurements on a sample of Pu^{239} exposed in the Clinton pile for several months in order to ascertain if the presence of Pu^{240} to the extent of 2% or 3% in the Hanford plutonium will interfere with the radioactive assay of plutonium, (8) preparation of UF_6 by reacting UF_4 with O_2, (9) isolation of 18 more mg of U^{233} from irradiated thorium, (10) radiations from U^{233} and its decay chain.

The memo ends with an overall summary of work in the five sections, Sections I, II, III, IV, and VI. It reads as follows: "Procedures have been developed for handling four times as much uranium metal as planned in the original flowsheet. This change is possible when the plutonium content of the uranium is low. A sample of Pu^{240} has been prepared and its radioactivity constants are being measured in order to determine the necessary corrections in the assay of Pu^{239}. Methods are being worked out for the removal of neptunium so that the time of 'cooling' can be reduced. The formation of complex ions as well as other properties connected with the different valence states of plutonium has been studied in order to determine optimum conditions for precipitation and recovery. Hexone is still regarded as the best solvent for extraction. Plutonium can be obtained in concentrated form by precipitation as oxalate. A simple method has been found for making UF_6 which may be important for large-scale production.

"Hexone extraction of U^{233} from thorium is satisfactory in the presence of an excess of thorium and aluminum salts. Experiments are under way to find the most suitable ketone. The new neptunium decay series is being filled in with new isotopes which have become available as a by-product of the pile. For the first time a decrease in elastic modulus has been observed in graphite which has been subjected to long exposure in the pile. It is not known yet whether this phenomenon, connected with the Wigner effect, is to be interpreted as favorable or unfavorable. The latest measurements on the properties of xenon connected with the poisoning of the pile are: yield — 5.6 percent, cross section for slow neutrons — 3.1 million barns, and half-life — 9.2 hours. New developments in analytical techniques include the use of porous graphite electrodes for spectrographic analysis which act as wicks to feed in solution at the proper rate, the application of the polarograph to the determination of plutonium in uranium solutions, and the simplification of analysis of zirconium and iron by extraction with organic solvents."

The top headline today reads "Sink 16 Jap Ships Off Luzon"; this was a carrier-borne strike.

Tuesday, November 28, 1944

The discharge of the first uranium slugs from the Hanford pile was completed today. The plutonium content is low because the uranium did not receive full production-level irradiation.

I sent a note to Lois Moquin in Berkeley enclosing some letters for her to put into Department of Chemistry envelopes and mail, a procedure that seems to be necessary for security reasons. I also asked her to send me a list of the addresses of people on the Project there for Helen and me to use in sending Christmas cards.

Report M-CN-2408, "Product Hazard Control Regulations and General Safety Precautions," was issued today by a Product Hazard Control Committee consisting of L. I. Katzin (Chairman), G. Cowan, J. Katz, R. Livingston, and J. Rose. The following set of mandatory rules was included:

1. All product handling and manipulations must be done in the hoods, or, in the case of dry product compounds, approved transfer boxes. Product should be outside the hood or box only when being carried in a safe covered container between hoods or hoods and transfer boxes.

2. All containers with product must be so labelled in conspicuous fashion.

3. Product must be stored only in approved locations. All transfers of possession of product must be from person to person, not from person to location.

4. All hoods, floors, and other working surfaces must be covered with stainless steel trays, uncracked glass plate, and/or other approved impervious material.

5. Rubber gloves must be worn at all times when handling or transferring exposed product or product solution, except when handling ignited counting plates.

7. All containers with 0.1 mg or more of product must be protected by a secondary safety container. Where the primary container is of unstable configuration (for example, graduated cylinders) the secondary container should be such as to increase the mechanical stability.

8. All centrifuges used for product must be in hoods, cubicles, or protected in other approved fashion. No centrifuge shall be opened while in motion. No centrifuge tube shall be filled to more than within a radius distance of its top; lesser filling is desirable. Balancing of centrifuge tubes shall be done in the hoods (refer to Rules Nos. 1 and 7 in particular).

9. All glassware, equipment, or tools contaminated with product shall be subject to the same rules as product. Equipment which has been in hoods or transfer boxes shall be considered to be contaminated until proved otherwise.

10. All discarded glassware used with product should be washed immediately with concentrated nitric acid by the chemist, and well flushed with tap water before being turned over to the glasswashing personnel.

11. Fume hoods and dust hoods are not be open more than 25% of capacity except when necessary for actual manipulations. (For dust hoods this means only two of the four hinged sections at the lowest level.)

12. Laboratory coats must be worn at all times in a laboratory where product is present. For personnel subjected routinely to large amounts of product, this should be replaced by special coveralls. Special shoes should only be worn in the laboratory and should be replaced by clean ones at least once a week, and more often if much product is handled in the particular laboratory. If contamination is suspected, the coats should be monitored by the Health Physics Group.

13. All digestions at boiling temperatures or where gas is evolved should be conducted in an approved type of system designed to confine any spray produced. Similar precautions should be observed in stirring operations. Evaporations should also be conducted in a spray-tight system (some form of retort is recommended).

14. Respirators should be handled on the outside surface only, to avoid contaminating the interior. Wash them periodically — preferably at the end of each day's use. A hot 20% sodium tartrate rinse is desirable. Respirators should be stored only in a covered container, such as the original carton, after wrapping in Kleenex or clean paper towels.

15. Centrifuges should be wiped out with a damp or oiled cloth after each day's use.

16. Any skin breaks or wounds, however trivial, caused by objects which might be contaminated, or which penetrate through possibly contaminated rubber gloves or other similar objects, should receive only a flushing with water before being brought to the attention of the group leader and the medical staff. Do not use antiseptics!

17. No eating or smoking is to be allowed in the laboratories of the Chemistry Division in which active materials are handled, and no pipetting is to be done by mouth.

Report CK-2240, "Chemical Research — Chemistry of Plutonium; Survey of the Chemistry of Plutonium," by L. I. Katzin, a 43-page report dated October 1, 1944, was issued today. The report contains a thorough review of the history and current knowledge of plutonium and its chemistry. The introduction to the report, reproduced here in its entirety, is as follows:

"Element 94 was discovered late in 1940 by Seaborg, McMillan, Wahl, and Kennedy[1] in uranium bombarded with deuterons in the Berkeley 60-inch cyclotron. The isotope found was later shown to be 94^{238}.

"Early in 1941 the isotope of major interest, 94^{239}, was discovered by Seaborg, Segrè, Kennedy, and Lawrence[2] in uranium bombarded with neutrons formed in the Berkeley cyclotron. The element was given the name plutonium, symbol Pu,[3] to follow uranium and neptunium in the same

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[1] CN-1488, "Radioactive Element 94"
[2] A-33, "Properties of 94^{239}."
[3] A-135, Seaborg and Wahl, "Chemical Properties of Elements 94 and 93."

order as the corresponding planets.

"The investigation of the chemical and physical properties of plutonium has been in progress continuously since the element's discovery. During the first year and a half the researches, carried on mainly at Berkeley, were done with isotope Pu^{238}, using the tracer technique. The first pure plutonium in the form of compounds of the isotope Pu^{239}, was isolated in August and September of 1942 by Cunningham and co-workers, who experimented on the ultra-micro scale with amounts of Pu^{239} ranging from 1 to 200 micrograms.

"These experiments, on the so-called 'microgram scale,' differed in principle from the experiments with 'tracer' techniques in that the work was done with pure plutonium compounds at the concentrations normally used in investigating chemical reactions. This was accomplished in spite of the minute amounts of material available by working with extremely small volumes, usually of the order of one to several thousandths of a milliliter, with specially constructed small capillary containers, burettes, and pipettes, together with extremely sensitive ultramicro balances and especially adapted microscopes. Using these techniques the principal oxidation states of plutonium were established and the solubilities of the important compounds of plutonium in these oxidation states were first measured. The presently accepted half-life of Pu^{239} was first accurately determined by this technique. Metallic plutonium was produced, and the methods for production of metallic plutonium were studied in some detail using a few micrograms of material. These experiments extended over a period of about a year and a half, and the total amount of plutonium in the laboratory at any time during the period never exceeded a few hundred micrograms.

"Work continued on approximately this scale of operations until about January of 1944, at which time milligram amounts of Pu^{239} became available.

"This survey is based upon work which was started with trace scale experiments and confirmed and extended by experiments with ponderable amounts of material. The major portion of the information in this survey is based upon the latest experiments with these larger amounts of plutonium. Only where time has not allowed confirmatory experiments is information based solely on tracer scale work given.

"The style chosen for this presentation of the chemical and physical information on plutonium is that of an essay survey without reference to original sources of information. A large portion of the information has been gathered from the written reports from the several laboratories of the Project but a certain amount of unpublished observation has been obtained through conversation, from memoranda of limited circulation, and other such sources available in the Chicago Laboratories of the Metallurgical Project. An especial indebtedness is acknowledged to the members of Section C-I who have assisted the writer in many ways. This survey is in a sense complementary to the detailed listing of data and sources of information given in Report LA-30 and its periodical supplements, issued from Los Alamos."

The problem of plutonium separation is well summarized in the introduction to the Separation Processes section, which is as follows:

"As material comes to the Hanford extraction plant, following pile irradiation and 'cooling,' it consists of cylinders of uranium metal encased in aluminum. In each metric ton of uranium metal there are expected to be about 250 grams each of plutonium and fission products. The radioactivity of the fission products, after 100 days of irradiation at 2500 kw per ton and 40 days 'cooling,' is about 3.5×10^6 curies per ton of uranium. After 60 days cooling the activity is about 2.5×10^5 curies, and further cooling has less and less effect. The extraction problem thus becomes a double one — chemical extraction of 250 grams of plutonium from 1000 kg of uranium, and separation of plutonium from fission activity. Accepting the value of 10 millicuries as the maximum amount of beta and gamma radiation that may be worked with directly by a chemist, with some shielding, 'decontamination' from fission activity must be accomplished by a factor of $2.5\text{-}3.5 \times 10^7$ for material cooled 40-60 days. Furthermore, this extraction and decontamination must be performed entirely by remote control, behind biologically adequate shielding (e.g., 8 feet or more of concrete).

"Methods of extraction. Several methods of separation of the product plutonium from uranium suggest themselves. One method is that of carrying the traces of plutonium from a solution of the uranium metal by means of a precipitate which will not carry uranium simultaneously. A number of possibilities suggest themselves here — any of several phosphates listed earlier, possibly arsenates, fluorides, and other precipitates that will carry the known insoluble salts of plutonium without at the same time precipitating appreciable amounts of uranium. Even such a salt as uranous oxalate might serve as a carrier for this first bulk separation from uranium. Following this extraction precipitation, similar procedures may be used to concentrate plutonium with respect to carrier until the point is reached where the plutonium compounds may be precipitated directly without carrier.

"A second type of procedure is really a relative of the first type. It makes use of a 'specific' adsorbent, perhaps arranged in the form of an adsorption column, to remove the plutonium from the solution of uranium with which it is in contact. This is in a sense carrying of an insoluble compound. The plutonium can then be eluted from the adsorbent with a much smaller volume of solution, resulting in a simultaneous concentration and separation from uranium. This procedure can be repeated, or coupled with a carrier procedure in some fashion, until the desired degree of concentration and purification is obtained, allowing direct separation of plutonium. Several materials can be used successfully for the adsorbent, among which may be mentioned silicious materials, a synthetic titania-mica zeolite, and a synthetic exchange resin (Amberlite IR-1). This by no means exhausts the list.

"Still a third type of procedure is applicable — immiscible solvent extraction. Plutonyl nitrate, like uranyl nitrate, is soluble in solvents such as ether when the nitrate ion concentration is sufficiently high, and procedures have been worked out in which separation from uranium and subsequent concentration result. Such a process has many points in its favor and can be made quite efficient. Methyl isobutyl-ketone and other solvents will extract plutonium(IV) nitrate also.

"In addition to these types of procedures, in themselves subject

to many variations and combinations, chemical ingenuity can suggest more. An example might be the isolation by means of a volatile higher fluorine, analogous to UF_6, if such could be demonstrated.

"Methods of decontamination. It is seen from the above that the chemical separation and concentration of plutonium *per se* is a far from impractical or even difficult procedure. The difficulty comes in combining this goal with simultaneous removal of fission product radioactivities by a factor of 10^7-10^8, with a process reliable and simple enough to operate by remote control. Of the three main types of procedure outlined above, each is potentially capable of eventually giving the desired results, given favorable conditions and sufficient time. One reason is the possession by plutonium of oxidation states with different types of ion, charge, and chemical behavior. With the proper solvent and extraction procedure, for example, it is possible to separate Pu(VI) from the bulk of fission elements by extraction from high nitrate concentrations which 'salt out' plutonyl nitrate even with trace concentrations. It is then possible to separate the plutonium from elements which may have been soluble in the organic solvent used by reducing it to the tetravalent state which does not extract with the solvent, and removing the offending contaminant by extraction. Similarly, with the adsorption column procedures, it is found that whereas Pu(IV) may absorb well, Pu(VI), or complexed Pu(IV), will not adsorb. Combining this with the possibilities of preferential adsorption and elution, separation from fission products may be achieved. Likewise, in the procedures using carriers, alternation of carriers, and use of the several oxidation states, allows adequate separation from fission elements. The use of more than one oxidation state to achieve the decontamination effect is so generally used it is known by the name of oxidation-reduction cycle."

In addition to the introduction, Katzin's report has three main sections: Nuclear-Physical Properties, Fundamental Chemistry of Plutonium, and Separation Processes.

Within the Nuclear-Physical Properties section, the methods of producing plutonium isotopes in particle accelerators and by fission are reviewed. The radiations emitted by disintegrating plutonium isotopes are reviewed. Alpha particles from Pu^{238} and Pu^{239} have measured energies of 5.5 Mev and 5.15 Mev respectively. Highly purified Pu^{239} has been observed to have soft gamma- (or x-ray) radiations with energies of about 0.4 Mev, 65 kev, and two or more additional quantized radiations with lower energies. The half-life of Pu^{239} has been determined with the low-geometry counter to be 24,000 years, with an estimated probable error of 2%. (Earlier measurements on sub-visible amounts of Pu^{239} by Wahl and me used a comparison of the rate of beta-particle decay of the parent U^{239} material with known half-life, to the product Pu^{239} alpha decay after complete decay of the U^{239}. The results of these measurements give an average half-life of 22,500 years, which is quite close to the more accurate current value of 24,000 years.)

Under the section concerned with Fundamental Chemistry of Plutonium, element 94's position in the periodic system, as a member of my suggested 'actinide' series of rare earth-like transition elements, and its oxidation states, oxidation-reduction potentials, known compounds, complexes, and trace chemistry are reviewed.

The Hanford Separation Process (as of this September is reviewed by Katzin and includes information on preparation for extraction, decontamination cycles, crossover or concentration cycle, and isolation procedure. The "Hanford Separation Process Flowsheet for the Bismuth Phosphate Process" is included as the last three pages of the report.

Helen went to her chemistry class today.

Because of ailing health, Cordell Hull has resigned as Secretary of State, and President Roosevelt has appointed Edward Stettinius to replace him. Roosevelt also nominated Major General Patrick H. Hurley to be the U.S. Ambassador to China.

Wednesday, November 29, 1944

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Cunningham, Davidson, Dawson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Katzin, Kraus, Lawroski, Manning, Schaffner, Simpson, Stewart, and R. C. Thompson. I listed the quantities of plutonium possessed by Section C-I. Of the 10,180 milligrams in the section, 7,840 milligrams are in stock solutions, in recovery processes, or contained in lanthanum fluoride slurries. The remaining 2,340 milligrams are distributed among Hindman (300), Simpson (500), Davidson (220), Thompson (180), Katz (400), Lawroski (20), James (340), Ghiorso (45), and Gilbreath (335). In a review of procurement priorities, I stated that the Project has an AA-1 blanket priority, but special directives and AAA priorities can be secured only under strict conditions and require the signature of a Laboratory Director as well as the Division Director.

I brought up questions on certain of the minor safety rules involving the filtered air section that are being disobeyed. After a discussion we decided that people will be allowed to go from the filtered air section (1) to the Annex for such activities as getting cokes and using the library, and (2) to the machine shop, in each case without the use of overshoes or changing shoes. Manning read portions from a health group memo involving accidents in handling plutonium. Apparently, contact with plutonium is very dangerous when lesions of the skin are present since absorption may be very rapid and efficient. I mentioned that Room 28 (formerly occupied by Morgan) is to be completely cleaned and repainted to remove all plutonium contamination; it is to be reserved for work on materials other than plutonium.

Katzin gave a brief review of the work on the homogeneous U^{233} pile, which is to consist of about a cubic meter of heavy water with 7 or 8 kilograms of U^{233} in solution. Since the pile is to be run at 100,000 kw, the cooling problem is severe, and since the power per unit volume is much higher than any other pile, the problem of poison by fission products is a much more serious one. Apparently, the fission products will have to be removed almost daily. I reported on the progress of the 93^{237} extraction at Clinton (J. Katz, Magnusson, La Chapelle, and Beard are at Clinton for this purpose). The plutonium extraction and decontamination of the regular run was not much affected by the process

changes made to get a good neptunium yield. It is known (by use of 93^{239} tracer) that the lanthanum fluoride slurries contain at least 25% of the original neptunium. I related to the group an anecdote which took place when the men arrived in Knoxville. Magnusson and La Chapelle could not locate Joe Katz or Malm, and Magnusson reported to the police that the two men were missing. Eventually the mystery was solved. Katz's and Malm's section of the train arrived in Knoxville ahead of the section on which La Chapelle and Magnusson were riding.

Morgan began looking for 93^{236} in sample 49DD (200 mg of plutonium plus deuterons, St. Louis cyclotron bombardment) with the idea that it could be the source of some of the activities found in this bombardment which have been identified as daughters of Th^{228}. The mechanism postulated is this:

$$Pu^{239}(d,n)95^{240} \xrightarrow{\alpha} 93^{236} \xrightarrow{\alpha} 91^{232} \xrightarrow{\beta} 92^{232} \xrightarrow{\alpha} Th^{228} \xrightarrow{\alpha} \text{etc.}$$

Morgan will use fraction 49DD-15, which is derived from a supernatant resulting from chemical manipulations he carried out September 25 on sample 49DD-4 which, in turn, is the result of a series of dichromate oxidation cycles on target material 49DD.

In response to Moulton's memo early this month about Case S-1112, "The Electrodeposition of Plutonium from Fused Salt Solution," I indicated, by memo, that all our work on this problem has been done with uranium as a stand-in and plutonium has not been used.

There was a Laboratory Steering Committee meeting at 9:00 a.m., attended by Bartky, Compton, C. M. Cooper, Dempster, Gore, Greninger, Hogness, Jacobson, Karl, Kimpton, McKinley, Mulliken, Munnecke, Stearns, Vernon, Wigner, and Zinn. Stearns asked Compton to read a letter about the future program here at the Laboratory. Compton first read a letter notifying the District Engineer that our work for Site W should be considered complete as of June 30 of next year, requesting authorization to terminate such work as of that date, and pointing out that the nation's welfare requires continuance of work in this field.

Compton then read a letter addressed from him to Stearns, which states that he is transmitting a copy of the letter to the District Engineer and expressing the expectation that arrangements will be made to continue essential work after the June 30 date. The letter went on to identify the following tasks of the Met Lab between now and this coming June 30.

"1. Continue the research and other services that are agreed upon between this office and the District as essential.

2. Prepare recommendations, for me to transmit to the District Engineer, regarding work in the general field covered by your present research which for the winning of this war, for the continued safety of the nation, and for the national welfare, should be carried on after July 1, 1945, for the duration of the war.

3. Prepare technical reports on the results of work done

by the Metallurgical Laboratory to be ready for transmittal to the Area Engineer by June 30, 1945.

"Among the continuing essential tasks of your laboratory are:

a. Long cartridge development.

b. Neutron tests on graphite and metal as requested by the Area Engineer.

c. Effects of radiation on graphite and other materials.

d. Maintenance of chemical studies related to the W process until this is operating smoothly.

e. Active studies until February, 1945, in biology, pile design, neutron physics, new chemical processes, metallurgy, etc., needed as a basis for recommendations for future work.

f. Service, medical care, physics information, chemical analysis, engineering help, etc., as required within your laboratory and as requested by associated projects as part of the essential tasks.

g. Development and production of such instruments as are required at W and associated DSM projects and for essential studies within your laboratory.

h. Limited work in pile design, neutron physics, fundamental chemistry of our special materials and products, and metallurgy of possible pile materials. The level of this work should be carefully considered by you in consultation with the Area Engineer and myself. It should support rather than interfere with preparation of the final reports, and should not be the basis for securing new contracts, extensive new equipment or additional personnel. It should, however, be continued to June 30, 1945, in order to establish a suitable foundation for the work that will be carried on after July 1, 1945."

Compton stated that the most urgent present need is to get our technical information into good shape. He indicated that it appears likely that the U.S. Engineers will have to continue to take responsibility for the duration of the war and that the Met Lab and associated organizations will have to carry on also.

During Compton's temporary absence from the meeting, Stearns explained that the present recommendation of Compton is a way to get action toward ultimate authorization of work beyond our present directive.

Thursday, November 30, 1944

James completed three dichromate oxidation cycles, removing plutonium from the rare earth fluoride precipitated in each cycle, on

sample 49NC-2, which he started working up on November 21 (derived from 4.4 mg of PuO_2 that received about 350 Mwd of neutron irradiation in the Clinton pile). The resulting sample labeled 49NC-3 has too much beta-particle activity to count, but the alpha-particle activity is estimated as 16,000 c/m. James dissolved the sample and continued with further dichromate cycles to remove the plutonium still present. The aim is to concentrate any alpha-particle activity due to an isotope of an element, like that with the atomic number 95, that cannot be oxidized with dichromate.

I supplied Moulton with the details he requested in his memo concerning Case S-2268, "Method of Separating Plutonium from Irradiated Uranium and Fission Products — Alternate Carriers, But Without Oxidation and Reduction Cycles Being Required in the Process."

Our report of abstracts of work in Section C-I, MUC-GTS-1154, was finished today and submitted for inclusion in the Chemistry Division Summary Report for November 1944 to be issued as soon as it can be assembled. The abstracts show problem assignments, individuals, notebook numbers, and describe accomplishments during the month of November.

"Metallurgical Laboratory, Report for November 1944," (MUC-JCS-93), is being prepared by the Laboratory Director's Office. In the summary it states:

"It is assumed that by June 30, 1945, the purpose for which this Laboratory was established — namely, that of aiding the Hanford Engineering Works to establish production — will have been fulfilled. The work from now until June 30, will be divided into three parts:

1. Research and development work required by Hanford or Y, and that necessary for the completion of the final report of the Metallurgical Laboratory. Such work, unless prior approval by the District to the contrary is given, is limited to the following conditions: (a) must be done with existing personnel; (b) must require no increase in the number of sub-contracts; (c) must not require the purchase of extensive equipment.

2. From now until February 1 a large part of the time of the personnel of this laboratory will be spent in preparing a report which will outline future work in the field of Nucleonics which, it is believed, should be carried out for the continued safety of the nation.

3. From February 1 until June 30, most of the effort will be spent in preparing final reports which will make available to the U.S. Engineers the results of the work which has been accomplished at the Metallurgical Laboratory.

"During all this time a competent staff of personnel must be maintained which is qualified and willing at a moment's notice to devote its entire attention to any unforeseen problems which may arise at the Hanford Engineer Works or Y. To maintain such a staff intact with a sufficient degree of morale, it is necessary that they work on some forward-thinking problem. Most promising of the problems at the present time seems to be in the field of (1) 'converter' piles and 'breeder' piles; (2) chemical procedures for handling metal much faster than planned in the original flowsheet; (3) the continued investigations of

the effect of radiation on graphite."

Accomplishments of Section C-I during the month were summarized as in the memo Daniels sent to Stearns on Monday.

War summaries, with good and bad news, in today's paper come from five fronts as follows: B-29's raid Tokyo for the third time in a week; five German towns fall to U.S. troops; Soviets cross the Danube River on a 93-mile front; Japanese bombs damage U.S. warships; Japanese drive on Kweiyang, a center highway in China.

Friday, December 1, 1944

My unclassified "Table of Isotopes" was published in *Reviews of Modern Physics*, Vol. 16, No. 1 (1944); this was prepared, based on the "Table of Isotopes" published in my article "Artificial Radioactivity" (*Chemical Reviews*, Vol. 27, No. 1 [August 1940]), by adding all the more recent relevant information from the published literature with the help of Helen. Now this has been expanded to include secret Project information and brought up-to-date. Truman Kohman, with the help of Helen, worked until September 15 of this year adding all the relevant Project information. After his departure to Hanford, Helen and I finished the work on this classified "Table of Isotopes," bringing it up-to-date using both secret Project and published literature; it is now ready for publication in the Metallurgical Project literature.

Mary Williams was hired today at $30 per week as a secretary for Katzin and Jones. At one time she worked for A. J. Dempster.

I talked by telephone with Zinn and John J. Goett about the irradiation of a sample in the heavy water pile at Argonne some day next week. The sample is to be irradiated approximately one hour at highest power (250 kw) and be returned to New Chem immediately after irradiation.

Hogness issued a summary of manpower distribution in the Chemistry Division which shows that there are 157 people in the Division as of December 1, including 80 in my section. The groups under Albaugh, Cunningham, and Katzin are divided as follows:

		Number of Men	
		Oct.	*Nov.*
Albaugh (Site W work, 32 men)	R. Thompson, extraction and decontamination	10	9
	Katz, concentration and isolation	5	3
	Gilbreath, process development	8	6
	Egan, semiworks	0	9
	Lawroski, solvent extraction	4	4
Cunningham, (Site Y work, 39 men)	Simpson, high vacuum work	10	10
	Hindman, basic chemistry	10	10
	Dawson, recovery	8	8
	Ghiorso, instruments and physical measurements	9	10
	Katzin, 23 work	6	6

American troops have reached the Roer River on a 20-mile front, according to today's newspaper.

Saturday, December 2, 1944

The record of alpha-particle hand counts for the week ending today shows James having twelve instances of high alpha- or beta-particle hand counts during that period. The Health Division reports he has considerable difficulty in decontaminating his hands.

Alec E. Kelley terminated today; he expects to return December 31 as a member of the SED.

U.S. troops have reached the Saar River along a nine-mile front. The Germans blew up the Merzig Bridge just as U.S. troops came into view.

Sunday, December 3, 1944

At Clinton, sample SA-1, containing 8.2 mg of PuO_2, was removed from the pile. It has been undergoing irradiation since June 5 of this year and has received 10,233,244 kwh (about 426,000 kw days) of irradiation.

Also at Clinton, 15 cans of $ThOCO_3$, specially purified from uranium, went into the pile in a position giving 1.4 times the average flux.

According to this morning's Chicago *Sun*, U.S. troops have stormed two Roer bastions, and German resistance on the west bank of the Roer River in northern Rhineland is nearly crushed.

Monday, December 4, 1944

The first production run of irradiated Hanford uranium slugs is scheduled to be received in the separation plant's underwater storage area today.

I received a teletype from Allison in Los Alamos asking for 20 to 30 micrograms of 37 (Np^{237}) of low and known natural uranium contamination for use in measuring spontaneous fission. He also asks if we have some 48 for similar experiments with fairly well known 49 contamination.

I immediately replied to Allison and indicated our current supply of 37 is smaller than 20 micrograms but that more should be available in two to three weeks from the special 37 runs at Site X. We will then grant his request if possible. I asked if this request includes Segrè's 100 microgram request. With regard to the 48, I said there seems to be 10 million dis/m of material containing 48; as much as one-half of the alpha activity may be 49. The sample is in 3 ml solution with about 2.5 mg of lanthanum, and we are willing to ship it if he wants it.

English and Katz at Clinton Labs held a phone conversation with me about the progress of the special 37 runs. The neptunium yield has been 25-30% and currently is in solution with 4.5 g of plutonium. With

regard to future 37 runs, there is only 30-day irradiated uranium on the schedule so we would have to decontaminate from the 39. We might get some 40-day irradiated material near the end of December, but we would have to furnish men for the Np^{237}-Pu^{239} separation and would have to work fast before the Clinton chemical separation plant is closed down on January 1. They told me that 40 irradiated thorium carbonate cans left Clinton for Chicago last Wednesday. Only the 15 new thorium carbonate cans (specially purified from natural uranium) that went in on Sunday are now in the pile, and they are presently scheduled to go to Montreal. No new cans have been put in yet for our Section C-I. The new sample of 40-50 mg of plutonium for pile irradiation should be identified by a MP-95 form and sent to the attention of Stoughton with complete information on the sample.

Compton asked Stearns for the following information to aid in deciding whether or not to authorize an immediate program for the production of 23 for possible use in the present war: "(1) A feasible time schedule for the production of 23 in quantities of 400 grams per day, assuming the availability of the required amounts of 25 or 49, (2) Additions to the research staff required for such a vast project, (3) Assuming as an alternative that the program is to be carried out, but not for immediate application in this war, what work should be done by the Met Lab before June 30, 1946." Compton mentioned that he has received a memo from Clinton Labs with regard to future developments which includes a proposal for construction of a small experimental 23 "producer" at Clinton as a part of a larger program toward 23 production.

At 7:45 p.m. I gave my second talk on "The Heavy Elements" at the new biweekly Monday evening Chemistry Division Meeting in Room 251, Ryerson Laboratory. After recapitulating my November 20 talk on nuclear properties, I completed that aspect by discussing the work of James, Morgan, and Ghiorso on our possible observation of an isotope of 96 from Pu^{239} plus helium ions, possible isotopes of 95 from Pu^{239} from deuterons, and our proposed look for 95^{241} from Pu^{241} beta decay or, later, 95^{243} from Pu^{243} beta decay (the Pu^{241} and Pu^{243} to come from intense neutron irradiation of Pu^{239}). I mentioned the U^{232} found in pile-produced U^{233} in the work of Ghiorso, Katzin, Studier, and Hagemann. I then turned to the chemical properties, covering the topics of electronic structure, "actinide" hypothesis (the evidence for a 5f actinide series starting at actinium), oxidation-reduction potentials, the work on neptunium and protactinium, and the general importance of studying the whole group of heavy elements as a means of predicting the behavior of new transplutonium elements and new oxidation states of known heavy elements. As examples I predicted a +3 state for neptunium, +4 state for protactinium, and +2 state for element 95. I emphasized that element 96 should have a very stable +3 state.

U.S. troops have crossed the Saar River, and superforts have bombed Tokyo for the fourth time.

Tuesday, December 5, 1944

The first uranium slugs are to be charged into the second Hanford plutonium manufacturing pile, the D pile, today.

My secretary, now Edrey Smith Albaugh, returned to work today.

At 10:30 a.m. I attended the Project Council Information Meeting on Physics. Others present were Borst, H. Brown, Burton, Compton, C. M. Cooper, Dancoff, Daniels, Darrow, Dempster, Estermann, Franck, Friedman, Goldsmith, Greninger, Hamilton, Hill, Jesse, Langsdorf, Lapp, Lewis, Lichtenberger, Manning, Maurer, Moon, Mulliken, Nordheim, Ohlinger, Rabinowitch, Seitz, Shonka, Simpson, Spedding, Stearns, Stephenson, Stern, Sugarman, Szilard, Wakefield, Watson, Wattenberg, Way, Weinberg, Whitaker, Wigner, Young, Zachariasen, and Zinn. Some of the high points are as follows. Weinberg presented the results of calculations on the properties of a pile run at high temperature (2000°C) as envisioned in the "pebble" pile suggested by Daniels. The advantage to be gained by such a pile is the elimination of the 40 difficulty (production by neutron capture in 49) by the continuous distillation of plutonium from the uranium carbide spheres. His calculations indicate the prospects for such a pile are poor because of the loss in K at the elevated temperatures. Young reported on his study of a design of a U^{233} "breeder," i.e., a pile that would operate on U^{233} and breed more U^{233} in a surrounding thorium shield than is consumed in the pile. He concludes that it is a promising possibility but that some types of losses may have been overlooked. There are many technical problems that have not been investigated. Nordheim reported on a small pile designed for Site X, which would be a 23 breeder or converter. He visualizes a pile using uranium enriched by a factor of 12.5, moderated by heavy water with a graphite reflector, and surrounded by thorium.

Franck and Dancoff reported on experiments at Urbana by Groetzinger, Kruger, et al. on the possible existence of a new kind of sub-atomic particle. Bombardments of 20 targets with 10 Mev deuterons have yielded in all cases, very penetrating radiations which pass through lead and water and give off beta particles at the end of their paths. These experiments may point to a "neutretto," which is a hypothetical particle with zero charge and a mass between that of the electron and neutron.

At the end of the meeting Compton gave the following information he has just received from Los Alamos: (a) neutrons per fission for U^{233} = 2.45 ± 5%, which is equal to the value previously obtained for U^{235}; (b) the relative abundance of 94^{240} to 94^{239} as measured in metal obtained from Site X is determined by mass spectrograph to be 3.4×10^{-4}.

W. C. Fernelius and R. A. Staniforth, who are visiting here from Mound Laboratory in Dayton, conferred with me about the chemistry and properties of polonium. They have had a wealth of experience in polonium chemistry at Mound Laboratory and are interested in polonium fluoride as a neutron source and asked if $Po(CF_3)_2$ would do as well if volatile. I told them I think so.

York, Metcalf, and I had a conference with Lavender, who is in town, about Patent Cases 52 and 61. I learned that (1) Lavender lost our cases in his file in Washington and did not know he had them until told by Metcalf the other day. (2) Lavender has had no word from the University of California and wants to go ahead without them. (3) He wants to work up the complete 52 and 61 cases, using data and calculations from before and after April 1, 1941, after Metcalf has reviewed all the data including the two Wahl notebooks involved. The cases would be filed within 30 days after Metcalf gets the two notebooks. (4) He will talk terms with us (the inventors — Kennedy, Segrè, Wahl, and me) shortly after the cases are filed. I agreed to call Kennedy, present these points to him for discussion with Segrè and Wahl, and ask them to call back tomorrow with their position.

I tried to call Kennedy in Los Alamos but he was tied up in meetings. I was able to reach Wahl, however; so I gave him the information to pass on to Kennedy and Segrè. I also asked him to mark the pertinent sections in his notebooks (he said that several notebooks would be involved). I mentioned to him I would be seeing Lavender at 2:00 p.m. tomorrow (he will leave for his train at 3:00 p.m.) to tell him their decision. I then suggested they call me in the morning at 11:00 a.m., Chicago time. I mentioned that, if desired, I could arrange a subsequent three-way phone conversation (Kennedy, Lavender, and me) at 2:00 p.m. if they want it.

Helen attended her chemistry class at YMCA College today.

U.S. troops are within six and a half miles of Saarbrücken, the Saar basin's greatest industrial city.

Wednesday, December 6, 1944

At 7:30 a.m., Katz, Beard, La Chapelle, and Magnusson returned to Chicago from their trip to Clinton Labs made for the purpose of isolating 93^{237} from two special Clinton chemical plant runs made under conditions chosen to maximize the yield of 93^{237}. They left here on the trip to Clinton on November 23.

The 8:00 a.m. Council meeting of our section in my office was attended by Albaugh, Davidson, Dawson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Katzin, Lawroski, Manning, Schaffner, Simpson, Stewart, Roy Thompson, and me. I announced that Section C-I has been the worst section on security violations. The situation is considered so bad that I asked the Assistant Section Chiefs to check on violations.

On the subject of materials, I mentioned we have a pound of depleted uranium in which there is a 25-fold depletion of U^{235}. We also have about 20 mg of ionium (20% ionium, 80% thorium mixture) from Berkeley that may contain as much as one mg of protactinium; it was originally obtained from a process for isolating UX_1 from tons of uranium to make 10 micrograms of U^{234}. It has been decided that six cans of neutron-

irradiated extra pure thorium (free of uranium) will be brought up from Clinton on January 1. Forty cans of ordinary thorium (irradiated) have come from Clinton for extraction — about 4 mg of U^{233} per can.

I brought up the subject of working hours of the Met Lab academic personnel. The Army has been examining attendance records and says that few people are working the established 48-hour week. Only a few arrive at 8:30; by 9:00 a small number more have come in. On the other hand, the signature sheets show that relatively few check out after 6:00 p.m. or come back to work at night. This laxity probably will result in a very strong request for our men to go to other sites where manpower is badly needed. I also requested that no newspapers and magazines be left lying about because the Army objects to the reading of these on working time.

Manning and I reported on the Physics Information Meeting which we attended yesterday. I mentioned that Hamilton reports that gut absorption of plutonium is quite inefficient and hence not too dangerous, but plutonium in the bloodstream or lungs is still believed to be dangerous.

James completed three additional dichromate cycles on sample 49NC-2, making a total of seven such cycles on material from the 4.4 mg sample of PuO_2 that has received about 350 megawatt days of neutron irradiation in the Clinton pile. A range curve was run on the resulting sample 49NC-7 that contains 150 alpha-particle counts per minute. The curve indicates a component of range longer than the Pu^{239} and one of shorter range. James plans to try to decontaminate further this alpha-particle activity by various chemical means so that more accurate range curves can be taken.

At 11:30 a.m. I received a phone call from Joe Kennedy in Los Alamos in response to the message I gave Wahl yesterday about my discussion with Lavender. Kennedy outlined their position as being: (1) They intend to continue their preparations for filing the cases themselves and will file after 30 days unless some other agreement is reached with Lavender. (2) They have no objection to Lavender reworking the cases but insist on an agreement as to disposition of the cases before filing. As to the disposition, they wish to handle both cases simultaneously and to give Case 61 to the University of California for a gift to the U.S. Government and to sell Case 52 to the U.S. Government for $1 million, our old price. Segrè is to be a silent partner (non-inventor) in Case 52. This plan of action was voted for by Kennedy and Wahl and against by me. Kennedy asked me to transmit this information to Lavender and urge him to call Los Alamos at 2:00 p.m. today in a four-way conversation to discuss this.

At 2:00 p.m. Lavender and I phoned Los Alamos and talked with Kennedy and Wahl together. Lavender asked for 30 days after Metcalf receives Wahl's notebook. There was agreement on the other points made by Kennedy in this morning's phone conversation. Lavender said that after the 30 days he would be quite agreeable to our filing for protection of the cases. After the phone call Lavender asked me to obtain a letter from Underhill saying that Underhill would assign Case 61 to the Government if we assign it to the University of California.

Organization charts of the scientific divisions of the Met Lab have been prepared, showing personnel at the assistant section chief level and above. They are:

Chemistry Division

Director	T. R. Hogness
Associate Directors	J. Franck
	Farrington Daniels
Assistant Director	L. B. Arnold, Jr.

Sections

C-I: Separation Studies and the Basic Chemistry of the Heavy Elements

Section Chief	G. T. Seaborg
Associate Section Chief	W. M. Manning
Assistant Section Chiefs	B. B. Cunningham,
	L. I. Katzin
	F. W. Albaugh

C-II: Radiation Studies

Section Chief	M. Burton
Associate Section Chief	A. O. Allen

C-III: Chemistry of the Fission Products

Section Chief	N. Sugarman
Associate Section Chief	A. Turkevich

C-IV: Analytical Services

Section Chief	D. S. McKinney
Associate Section Chief	J. I. Watters
Assistant Section Chiefs	M. S. Fred
	C. R. Schwob

Health Division

Director	R. S. Stone
Associate Director	L. O. Jacobson

Sections

H-I: Clinical Medicine and Medical Research

Section Chief	L. O. Jacobson

H-II: Biological Research

Section Chief	K. S. Cole
Associate Section Chief	C. L. Prosser

H-III: Medical Industrial Hazards and Health Physics

Advisory Committee

William Bloom
Alexander Brunschwig
W. R. Harrison
Paul Hodges
C. J. Watson
Sewell Wright

Physics Division

Director	A. J. Dempster
Associate Director	E. P. Wigner

Sections

P-I: Instrument	
Section Chief	W. P. Jesse
P-IV: Crystal Structure	
Section Chief	W. H. Zachariasen
P-V: Mass Spectroscopy	
Section Chief	A. J. Dempster
P-VI: Materials and Methods (Temporary)	
Section Chief	R. Schlegel
P-VII: Theoretical	
Section Chief	G. Young
P-VIII: Properties of Solids	
Section Chief	F. Seitz
P-IX: Engineering (Temporary)	
Section Chief	L. A. Ohlinger

Technology Division

Director	C. M. Cooper
Assistant Director	L. A. Ohlinger (on loan from Physics)

Sections

T-I: Engineering Development	
Section Chief	A. C. Miller
Assistant Section Chief	R. N. Lyon
T-V: General Engineering	
Section Chief	J. O. Maloney
T-VI: Optics	
Section Chief	G. S. Monk
Associate Section Chief	W. H. McCorkle

Metallurgy Division

M-I: Metallurgy	
Section Chief	F. Foote
M-II: Corrosion	
Section Chief	E. W. Brugmann
M-III: Fabrication	
Section Chief	J. H. Chapin

The following is a summary of the personnel status of Section C-I:

G. T. Seaborg — Section Chief
E. Albaugh — Secretary

W. M. Manning — Associate Section Chief
J. Horwich — Secretary

T. O. Jones — Assistant to Section Chief
M. Williams— Secretary

Sub-section I

F. W. Albaugh — Assistant Section Chief
D. Black — Secretary to F. Albaugh

Group 1: Extraction-Decontamination

R. C. Thompson — Group Leader (AC)
M. Ader (JC)
L. S. Bartell (RA)
R. H. Bradt (C)
R. W. Greenlee (RA)
J. G. Malm (RA)
L. O. Morgan (JC)
S. Peterson (JC)
J. Sedlet (RA)
J. Krinsky (Tech)
M. Summers (Tech)

Group 2: Concentration-Isolation

J. J. Katz — Group Leader (AC)
W. C. Beard (RA)
H. Hopkins (RA)
S. Nyden (Tech)

Group 3: Process Development (cooperation with Semiworks)

J. R. Gilbreath — Group Leader (JC)
W. J. Blaedel (AC)
R. G. Larson (AC)
H. Hyman (RA)
A. Margolis (AE)
R. Post (RA)
M. T. Walling (RA)
B. Winner (RA)
P. Boykin (Tech)

Group 3A: Semiworks

C. J. Egan — Group Leader (AC)
H. Flotow (RA)
W. C. Giegold (RA) [SED]
H. Hasenfus (RA) [SED]
K. P. Moseley (RA) [SED]
R. Rasmussen (JC)
R. Van Winkle (JC)
B. Wendrow (RA) [SED]

Group 4: Solvent Extraction

S. Lawroski — Group Leader (AC)
B. B. Brody (RA)
R. Reinhardt (AC)
A. Stein (RA)
I. J. Schaffner — Squad Leader [transfer to C-I pending]
J. H. Schraidt (RA) [SED] [transfer to C-I pending]
W. Simon (AC) [transfer to C-I pending]
E. Hausman (RA)
E. Quinn (RA) [SED]
H. Fritz (RA) [SED]
H. Zvolner (AC)

Sub-section II — Basic Chemistry and Service

B. B. Cunningham — Assistant Section Chief (C)
M. Bohlman — Secretary to B. B. Cunningham

Group 5: Basic Dry Chemistry

O. C. Simpson — Group Leader (SC)
N. R. Davidson — Assistant Group Leader (AC)
S. Fried (AC)
Z. Jasaitis (AC)
T. E. Phipps (SC)
H. P. Robinson (AC)
R. Seifert (C)
E. F. Westrum (AC)
B. Abraham (JC)
I. Sheft (JC)
H. Peterson (Tech)
H. Thomson (Tech)
N. Erway (Glassblower)

Group 6: Basic Wet Chemistry

J. C. Hindman — Group Leader (AC)
K. A. Kraus — Assistant Group Leader
D. Ames (RA)
J. S. Dixon (JC)
A. E. Florin (JC)
R. A. James (JC)
T. La Chapelle (JC)
K. McLane (RA)
L. Magnusson (JC)
P. O'Connor (JC)
J. J. Howland (AC)
H. Billington (Tech)

Group 7: Recovery

L. R. Dawson — Group Leader
D. C. Stewart — Assistant Group Leader [SED]
H. Anderson (AC) [SED]
L. B. Asprey (RA) [SED]
J. W. Britain (RA) [SED]
P. R. Fields (JC)
P. Fineman (RA) [SED]
L. Leventhal (RA) [SED]
A. Koskosky (Tech)

Group 8: Instruments and Physical Measurements

A. Ghiorso — Group Leader
A. H. Jaffey — Assistant Group Leader (AC)
A. C. Krueger (AC)
J. A. Crawford (JP)
B. F. Scott (JC)
B. B. Weissbourd (JC)
L. Crawford (RA)
D. Hufford (RA)
P. Walsh (RA)
J. M. Dorsey (RA)
[transfer to C-I pending]
E. Lewis (Tech)

Group 9: U-233 Group

L. I. Katzin — Assistant Section Chief
M. Williams — Secretary
F. T. Hagemann (AC)
N. N. Hellman (AC)
M. Studier (AC)
M. Wolf (AC)
E. Hyde (JC)
Q. Van Winkle (JC)
S. Shupp (Tech)

Non-Academic Service Group

K. Florin — Group Supervisor
E. M. Freeman — Lab Assistant
N. Jennings — Lab Assistant
J. Krinsky — Technician (part time)

Dieners (Laboratory Assistants)

J. Slattery — Supervisor

A. Barbic	A. L. Patterson
K. Gnjec	D. Pierce
C. Johnson	J. Prince
I. Jones	E. Schroeder
E. Little	M. Tadinac
L. Mabins	J. Tookes
A. Martin	I. M. Wallace
F. Ouderkirk	H. Calloway

(SC — Senior Chemist; C — Chemist; AC — Associate Chemist;
JC — Junior Chemist; RA — Research Assistant; Tech — Technician)

The top headline in today's paper reads "Meeting in Atlanta Prison!" About 25 long-term prisoners in the Atlanta federal prison are protesting their incarceration in the same building with German saboteurs and spies.

The Project Council Policy Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall, attended by Bartky, Chapman, Compton, C. M. Cooper, Daniels, Dempster, Franck, Greninger, Hamilton, Hogness, Jacobson, Karl, Mulliken, Spedding, Stearns, Stone, C. J. Watson, W. W. Watson, Whitaker, Wigner, Wirth, and Zinn. Compton announced that as a result of his communications with Colonel Nichols regarding the near completion of the Metallurgical Project tasks for Site W, it is expected that the present contract will be closed as of June 30, but that decisions will be reached soon enough to provide for continuance under suitable auspices. Between now and June 30 the tasks of all laboratories in the Project are: (1) continue essential tasks as agreed between Project and Army, (2) prepare recommendations for work after June 30 needed for national welfare, (3) prepare technical reports to be ready for transmittal by June 30, 1945.

The members discussed reducing the frequency of Council meetings and decided to change from the present semimonthly arrangement to a monthly schedule with Policy and Information meetings to occur on the third week of each month beginning next month. All information and policy sessions are to be held on Tuesday and Wednesday. The usual Chemistry-Technology Information Meeting will be held Tuesday, December 19, as will the usual policy meeting the next day.

Thursday, December 7, 1944

In a teletype to Kennedy at Los Alamos today, I say that Lavender wants a letter from Underhill at the University of California promising that the University will assign Case 61 free to the Government if the plan we discussed goes through (our giving it to the University). I say I shall contact Underhill unless he, Kennedy, teletypes me otherwise.

In response to his request, I sent Hogness a list of 10 people from my section who could be released for transfer to other parts of the DSM Project: Flotow, Giegold, Hasenfus, Moseley, Rasmussen, R. Van Winkle, Wendrow (see Fig. 33), Zvolner, Margolis, and Stein. In addition, I ask that he consider Howland as one available for transfer, but I state, however, that I believe he should be given more choice in regard to a place of transfer than the other 10 men in view of his greater maturity and much higher level of skill.

Manning asked Arnold to obtain from Stoughton eight aluminum cans to be used for thorium samples to be sent to Clinton for irradiation.

I read a copy of a memo from R. E. Clark to Maloney (Technology Division) giving the results of a comparative evaluation of peroxide precipitation and hexone extraction as a means of carrying out the isolation step at Hanford. He concludes the two methods offer about equal advantages and, since the equipment for the peroxide method has already been installed, it seems questionable that the installation of solvent extraction equipment can be justified at this time. He recommends, however, further research on the hexone extraction cycles for more steps in the Hanford separation process, such as concentration, isolation, and possibly decontamination. This would result in the elimination of a large number of operations.

Warner, Hogness, Daniels, and I met to discuss preparation of terminal and summary reports. It was agreed that journal type articles should be written rather than terminal reports and would be authored by the men involved in the investigations — these articles will be due June 30 and will be published in special volumes. The following responsibility assignments were agreed on for the accompanying summary volumes, also due June 30: Volume 18 — Chemistry of the Transuranium Elements (Editor, Seaborg); Volume 19 — Bismuth Phosphate Process (Editor, Perlman; Editorial Committee, S. G. Thompson, English and Albaugh); and Volume 20 — Alternate Processes (Editor, Manning; Editorial Committee, English, Eastman, Albaugh, Thompson, Brown). These same men will also edit the volumes of journal articles.

Helen went to her YMCA college chemistry class.

Fred and Edrey Albaugh, Herman Robinson, Helen and I had dinner with Zene Jasaitis, Gene Gullahorn (Zene's sister) and her daughter Barbara at the home of Zene's mother, Mrs. Vincenta Insoda in Lockport, Illinois (see Figs. 34 and 35).

On the European war fronts, U.S. troops have captured two-thirds of Sarreguemines, and the Soviets have driven within 35 miles of the Austrian border.

Figure 33. B. R. Wendrow in front of the Mira-Mar Hotel, 6218 Woodlawn. December 1944.

XBB 780-13178

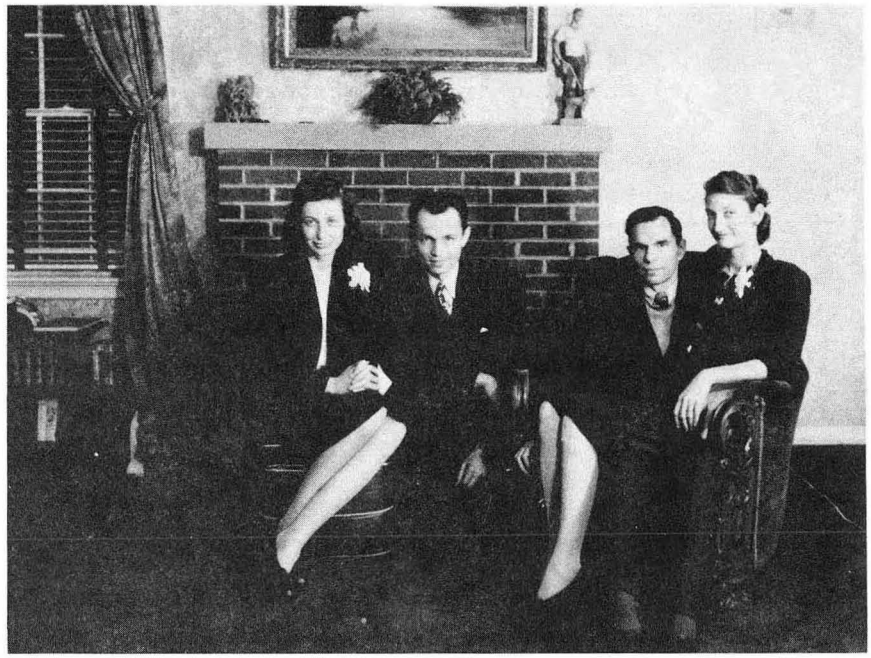

Figure 34. Edrey and Fred Albaugh, Glenn and Helen Seaborg at Mrs. Insoda's home, Lockport, Ill. December 7, 1944.

XBB 7810-13188

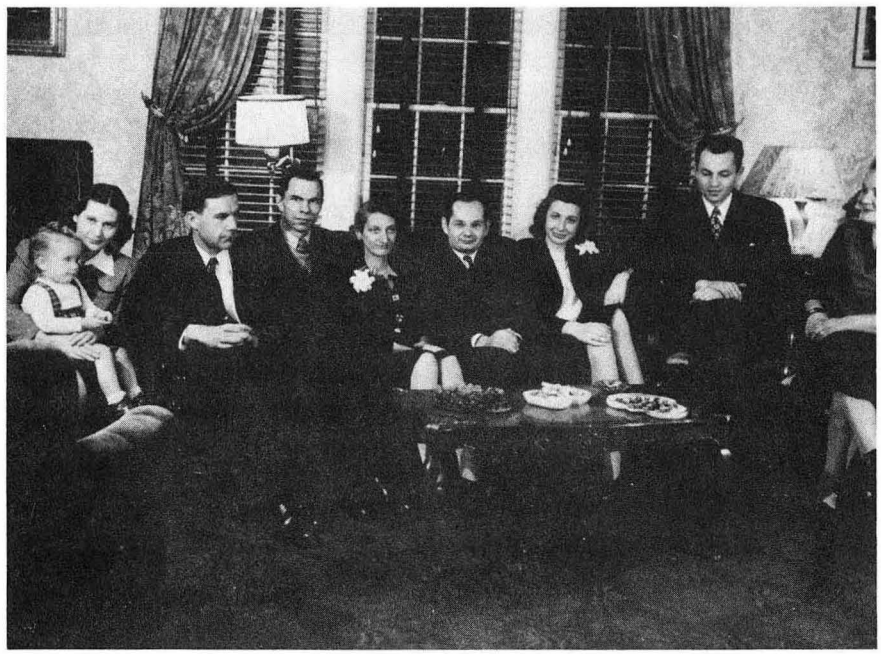

Figure 35. Barbara (baby) and Gene Gullahorn, Herman Robinson, Glenn and Helen Seaborg, Zene Jasaitis, Edrey and Fred Albaugh, and Vincenta Insoda — guests at Mrs. Insoda's after dinner. December 7, 1944.

XBB 7810-13189

Friday, December 8, 1944

At Clinton today, 14 additional cans of $ThOCO_3$, specially purified from uranium, go into the pile in a position giving 0.3-0.4 times the average flux.

I phoned Pan Jenkins at Y-12 to ask for a flowsheet of the chemical separation process used at Y-12. He said he will see that it is sent soon. He informed me he is returning to Berkeley next week (but stopping here in Chicago next Thursday enroute) and will stay there from now on. He is beginning to think in terms of long-range work.

Manning asked Monk for the assistance of his section in having a number of metals evaporated on glass discs in connection with studies on the back-scattering of alpha particles and its effect on counting yields.

Jaffey sent Kohman at Hanford a copy of the present differential pulse height selector circuit. He told Kohman that we are still using the spherical chamber, the big chamber that just came from the shop. Jaffey also informed Kohman that we find the use of negative high voltage very important, as well as putting the sample on the high voltage electrode.

"Chemical Research — Basic Chemistry of Plutonium. Report for Month Ending November 1, 1944," (CN-2289), was issued. Some of the highlights of the report are as follows.

Basic Chemistry Group (Hindman, Group Leader). *Formal oxidation potentials of plutonium couples in various acids.* Hindman has continued the earlier work (as reported in CK-1371, CK-1587, and CN-1702) on the potential of plutonium couples. Careful measurements of the potential of the Pu(III)-Pu(IV) couple in mixed NaCl-HCl solutions show that the potential has no (H^+) dependence. The conclusion is reached that the ion in a Pu(IV) solution more than 0.3 M in H^+ is largely Pu^{+4} (hydrated). Electromotive force measurements on solutions containing plutonium in disproportionating equilibrium indicate that the relative complexing power of three acids (HCl, $HClO_4$, and HNO_3) at 1 M concentration does not differ greatly. The dependence of the Pu(III)-Pu(VI) and Pu(IV)-Pu(VI) potentials on the (H^+) is probably according to a fourth power of the (H^+), which would correspond to the formation of the PuO_2^{++} ion in solution. *Study of a sulfate complex of Pu(IV).* Ames has obtained data that show there are one or more positively charged Pu(IV) sulfate complexes in addition to various negatively charged complex ions of plutonium(IV) and sulfate indicated by both transference and spectrophotometric observations.

Transference measurements on Pu(III), (IV), and (VI). K. McLane shows (1) the existence of a Pu(VI) sulfate complex anion in 1 M H_2SO_4; (2) transformation from a positively-charged Pu(VI) ion to a negatively-charged complex in 6-7 M HCl; and (3) the probable existence of a complex of Pu(III) and sulfate.

Hydrolytic behavior of plutonium. Kraus has found (a) the hydrated Pu(IV) ion undergoes rapid hydrolysis to give probable monomeric ions around pH 1.0 and is about 50% hydrolyzed around pH 1.5; (b) the monomeric hydrolysis products undergo rapid polymerizations or disproportionation in the cold; (c) the polymers that are formed in the cold in the pH region

1 - 1.5 undergo an aging process. Depending on the pH of the solutions, heating of cold, polymerized solutions causes a considerable increase in particle size of the polymers.

Mechanism of co-precipitation. Howland has carried out two experiments, one in the presence of $SO_4^=$ and the other in its absence, that indicate that co-precipitation of Pu(III) with bismuth phosphate is much better in the presence of sulfate. A considerable quantity of $SO_4^=$ has been found in the bismuth phosphate precipitate. These observations suggest that $SO_4^=$ may play an important role in the mechanism of co-precipitation of Pu(III) with alpha phase bismuth phosphate.

Isolation and study of 93^{237}. Magnusson has measured the counting yield of Np^{237} in the "50% geometry" alpha-particle counter by comparison of the counting rate in the 50% geometry counter with the disintegration rate of the sample determined in a low-geometry vacuum counter. He finds the counting yield to be 52.9 (±0.5, nine-tenths error) percent. This gives a calculated half-life of 2.25×10^6 years. The earlier value, based on a 52% counting yield, is 2.21×10^6 years.

U.S. troops have invaded Ormoc Bay on Leyte Island. A Reuters report says that 4,000 Japanese troops and 62 planes were destroyed. Japanese, on the other hand, made their strongest counter-raid on our B-29 base on Saipan.

Saturday, December 9, 1944

I wrote to Underhill in Berkeley stating that it seems probable the inventors (Kennedy, Segrè, Wahl, and I) will want to assign Case 61 to the University of California without remuneration provided the University will, in turn, assign it to the Government without any compensation. I told him of Lavender's request that he receive from Underhill a letter stating that the University will make such an assignment in the event the inventors do go through with this plan.

In a letter to Whitaker in Oak Ridge, attention Sinclair, I cancelled our continuing orders for irradiated uranium slugs in view of the cancellation of the Chicago semiworks program.

Cunningham suggested to Manning a method for crediting individuals for work done on the Project by including in projected Project publications, a summary of review nature with name-references to the original work of the many individuals who have been associated with the Project.

The Health Division issued to Section Chiefs a procedure for caring for wounds possibly contaminated with plutonium. The procedure specifies that the injured area be washed with large amounts of running water, immediately after which the individual is to go to Drexel House, or if it is closed, to the Hospital Admitting Office and have Dr. Allen or Dr. Nickson called.

Albaugh, in a memo to me, comments on each step in the Hanford

separation process flowsheet and gives his recommendations for my use in the forthcoming meeting at Hanford to consider the final specifications for the Bismuth Phosphate Process. In general the process appears to be in good shape, but he does recommend a number of changes, including (1) The addition of H_2SO_4 to the 40% UNH metal solution to prevent oxidation during storage, thus eliminating the need for a prereduction treatment or any additional N_2H_4 destruction treatment. (2) In the reduction steps, use a shorter time than one hour for reduction with Fe^{+2}. (3) In the plutonium precipitation step, add $(NH_4)_2SiF_6$ as a means of lowering plutonium losses. (4) In the isolation step, use 2 N HNO_3 rather than 1 N HNO_3 to improve the quality of the peroxide precipitate. In the realm of speculation he mentions a number of other possible improvements of the Bismuth Phosphate Process and also describes the considerable amount of scouting work done on the development of an all-solvent extraction process for extraction, decontamination, and concentration of plutonium and that it may well be the ultimate separations process.

A special health physics survey was made of about thirty-five frequently used classified documents in my office to determine whether or not they are contaminated with radioactive substances. The entire outer surface of each report was examined, as well as three or more randomly selected pages. No radioactive contamination could be detected with either the GM counter or Pluto.

Spof English at Site X and I talked by phone about the arrangements for another set of special Np^{237} runs. Four runs are planned, the first two to go into the dissolver on about December 22 and to be ready for processing by Section C-I on January 1. Two men will have to go to Clinton Labs on Sunday, December 17, in order to be there in time to prepare and add the Np^{239} tracer on the 23rd. The third and fourth runs will go into the dissolver by about December 25 and the Np^{239} tracer is to be added by our men on the 26th. Arrangements should be made for two or more of our men to arrive on December 30 to help the first two with the processing.

According to today's paper, Britain and the U.S. are having problems over the question of intervention in internal strife in European countries. Churchill is fostering an intervention policy, and the U.S. State Department has publicly avowed a hands-off policy.

Sunday, December 10, 1944

At 6:00 p.m. I left Chicago for Pendleton, Oregon, on the City of Portland in order to attend meetings at Hanford to consider the final specifications for the Bismuth Phosphate Process.

According to this morning's Chicago *Sun*, snow is hampering the U.S. armies all along the Rhine front. Here in Chicago it started snowing about 8:30 this morning and has continued all day.

Monday, December 11, 1944

Enroute to Pendleton, Oregon, on the City of Portland.

France and the Soviet Union have signed a mutual assistance pact and a treaty of alliance.

Tuesday, December 12, 1944

At 9:45 a.m. I arrived in Pendleton, Oregon. I was met and then continued by automobile to the Hanford Engineer Works. There has been much construction at Hanford Village since my visit last May. During the afternoon I made a tour of the pile and chemical separations areas. The one pile now operating is running at a power level of 125,000 kw but is expected to go to its rated level of 250,000 kw when all 2,000 tubes have been filled. Each pile has 200 tons of metal instead of the originally planned 100 tons. It will take 200 days operation at 250,000 kw to attain 250 grams of plutonium per ton of metal.

One canyon is finished and is running with tracer now. The canyon is a tremendous construction 860 feet long. Looking down at the control boards the length of the canyon, one sees nothing but 860 feet of valves, meters, indicators, controls — a fantastic sight. Everything is handled completely by remote control. The insides of the cells cannot be seen except by means of a thoroughly shielded periscope handled by an operator from an overhead crane. Each cell has three staggered seven-foot concrete block covers (15 tons each) which can be removed by an operator in a thoroughly shielded overhead cab. The periscope enables the operator to use remote control wrenches and other tools to replace defective equipment.

Slugs coming out of the pile glow red from the photons shooting out. (Their temperature is, of course, much below red heat.) They are dropped into a deep water channel and caught in big buckets which carry them into large lead coffins, after which they are hauled out into the desert five or ten miles to cool.

The piles are in the 100 area and the extraction canyons in the 200 area. Both run on a 24-hour schedule. Extraction and decontamination takes place in Bldg. 221B. Concentration (the crossover step) takes place in Bldg. 224B, and isolation of the product occurs in Bldg. 231. The Control Labs (Bldg. 222B) are also in this area. The 300 area is near Richland — about 20 or 25 miles away from the 100 and 200 areas. This area contains the research building (No. 3706) where John Willard and his group work: the test pile, the canning building, the cold semiworks, etc. (see Figs. 36-47).

I had dinner at the Perlmans' and then spent the night at the Richland transient quarters.

Figure 36. Aerial view of Richland, late 1944.

XBB 7812-15937

Figure 37. View of Hanford barracks, 1944.

XBB 672-658

Figure 38. Store at Hanford, late 1944.

XBB 7812-15927

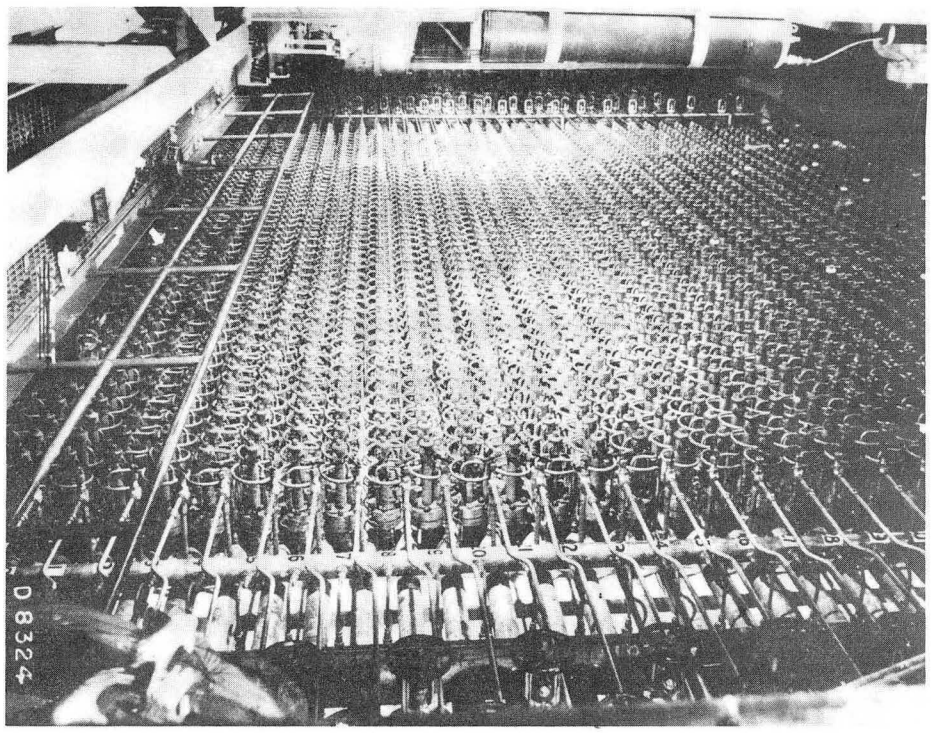

Figure 39. Front face of "Pile" showing "Pig-tails" — 100 Area at Hanford, late 1944.

XBB 7812-15930

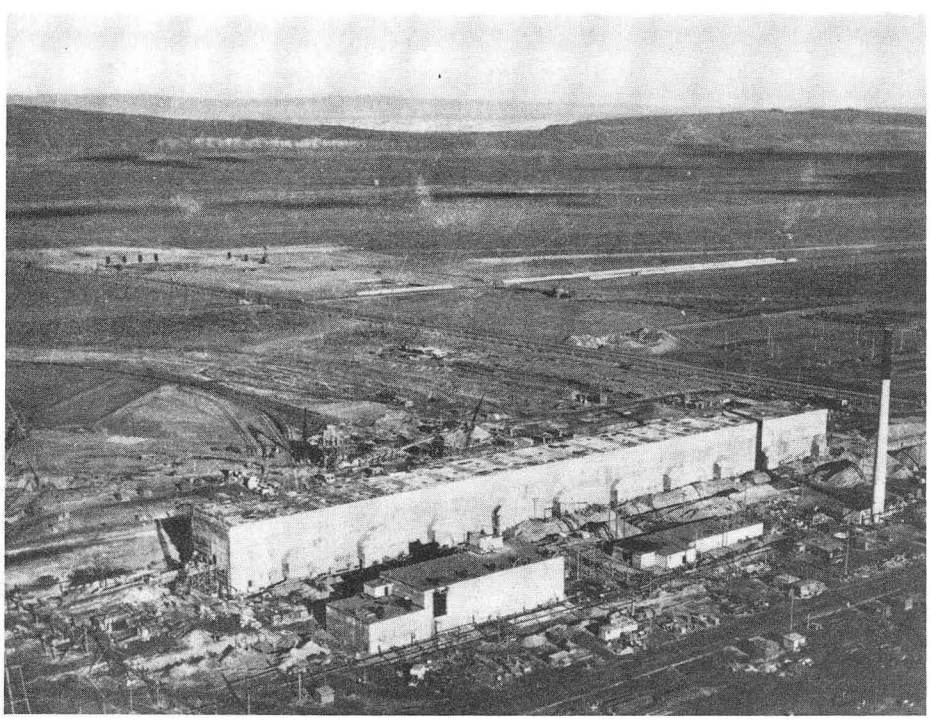

Figure 40. Construction of Extraction and Decontamination Building (221B) at Hanford, late 1944.

XBB 7812-15934

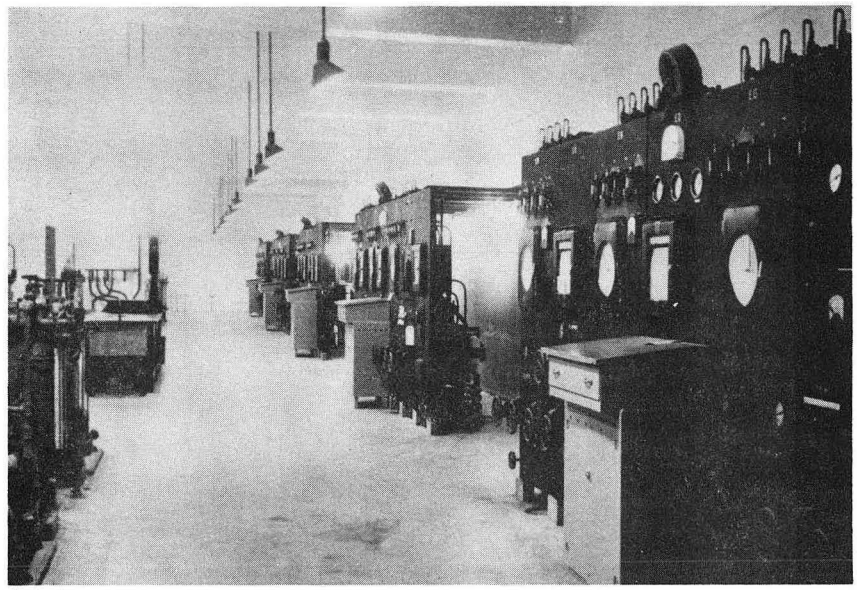

Figure 41. Control Gallery, Extraction and Decontamination Building (221B), late 1944.

XBB 7812-15936

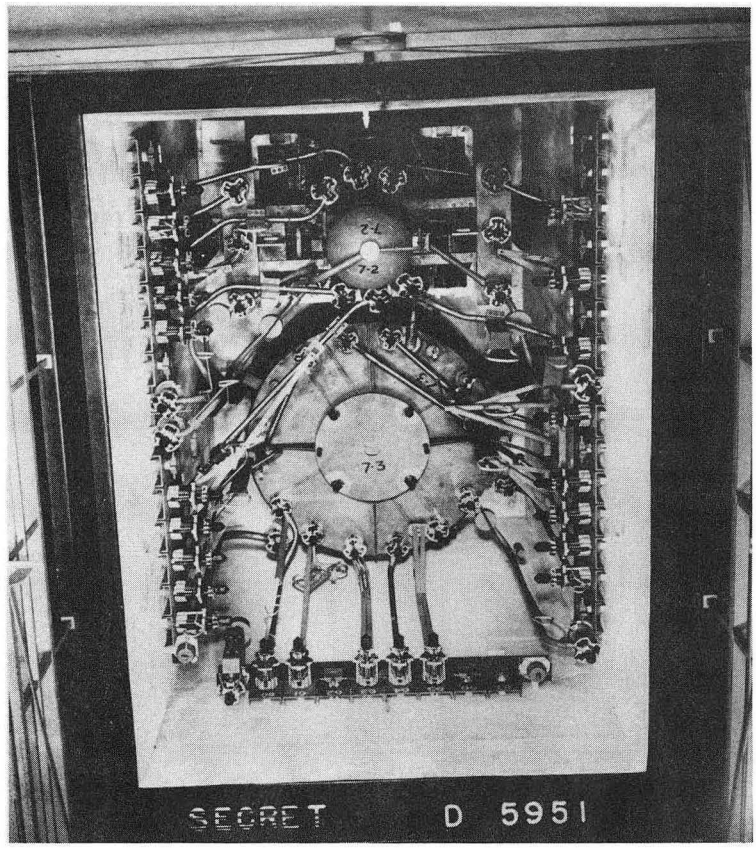

Figure 42. A cell in the Extraction and Decontamination Building (221B) at Hanford, late 1944.

XBB 7812-15933

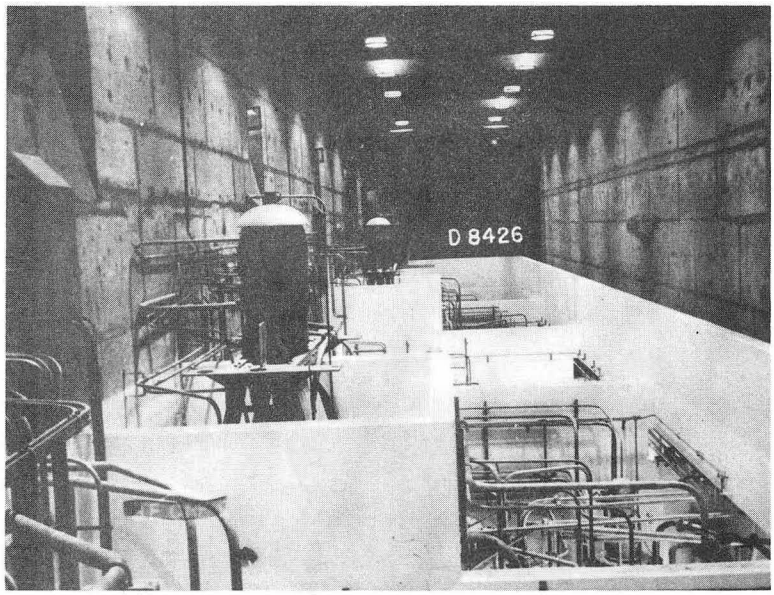

Figure 43. Equipment in the Concentration or Crossover Building (224B) at Hanford, late 1944.

XBB 7812-15932

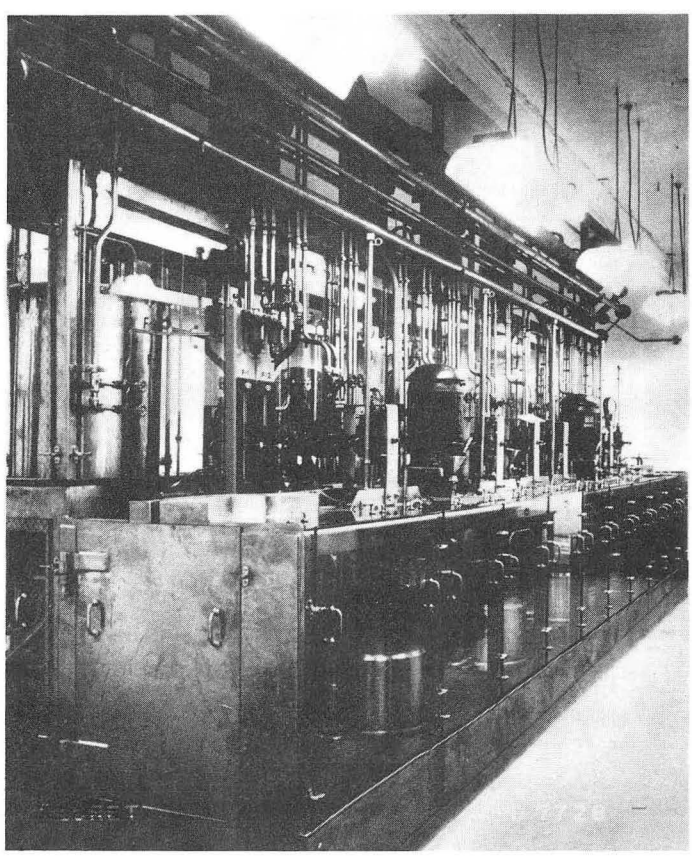

Figure 44. Equipment in Isolation Building (231) at Hanford, late 1944.

XBB 780-15929

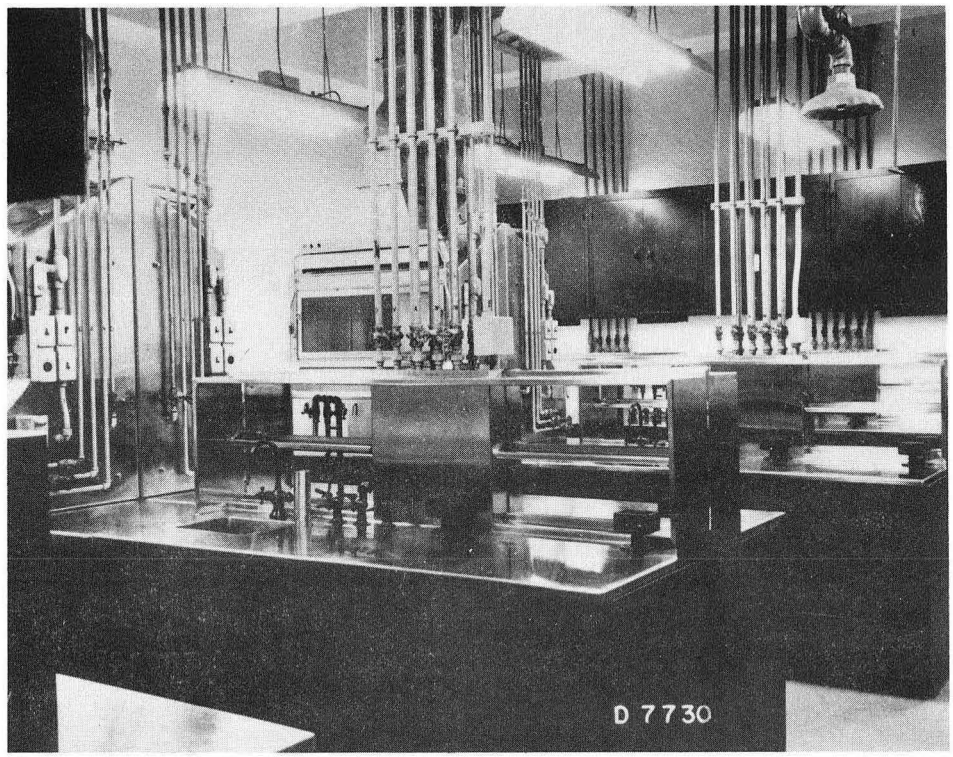

Figure 45. Laboratory in the Control Laboratory (222B) at Hanford, late 1944.

XBB 7812-15928

Figure 46. Laboratory (3706) — 300 Area at Hanford, late 1944.

XBB 7812-15935

Figure 47. Single shell waste tanks at Hanford, late 1944.

XBB 7812-15931

Wednesday, December 13, 1944

I attended the opening meeting of a three-day meeting, held in the conference room of the general technical (research) building (no. 3706) in the 300 Area to consider the final Operating Standards for the Bismuth Phosphate Process at the Hanford Engineer Works. We are examining in detail the individual documents describing the Operating Standards for each step of the Process. These details include the amounts of materials, concentrations, temperatures, time of mixing, time of settling and centrifugation of precipitates, and so forth, together with the references to Project literature that form the bases for these specifications.

Present at the meeting are the top operating and relevent scientific personnel of the Hanford Engineer Works and other corresponding people from the Metallurgical Laboratory, and so forth. The group thus includes L. Squires, W. O. Simon, W. G. Kay, F. A. Otto, H. Worthington, O. Greager, J. E. Willard, S. G. Thompson, G. W. Watt, L. C. Peery, J. B. Work, N. M. Hilberry, I. Perlman, T. R. Hogness, and me. We are considering individual Operating Standards documents covering the extraction step; the first oxidized decontamination step in which bismuth, ceric, and zirconium phosphates are precipiated; an alternate first oxidized decontamination step in which only bismuth phosphate is precipitated; the first reduced decontamination step in which bismuth phosphate is precipitated carrying the plutonium; an alternate first reduced decontamination step in which ammonium fluosilicate is present to act as a complexing agent; the second oxidized decontamination step in which bismuth, ceric, and zirconium phosphates are precipitated; an alternate second oxidized decontamination step in which only bismuth phosphate precipitate is precipitated; the second reduced decontamination step in which plutonium is carried with bismuth phosphate; an alternate second reduced decontamination step in which ammonium fluosilicate is present to act as a complexing agent; the lanthanum oxidized crossover step in which by-product lanthanum fluoride is precipitated; the lanthanum reduced crossover step in which plutonium is carried with the lanthanum fluoride precipitate; the metathesis step in which the lanthanum fluoride and its incorporated plutonium fluoride are metathesized with a mixture of KOH and K_2CO_3 to produce the hydroxides; and an alternate metathesis step in which ammonium sulfide is used to remove iron. J. B. Work has been responsible for the preparation of all of these Operating Standards which apply to all the steps in the Bismuth Phosphate Process that will be carried out in Buildings 221 and 224. In addition, we have before us the Operating Standards for the first plutonium peroxide precipitation, the second plutonium peroxide precipitation (that is, its reprecipitation), and the preparation of the final plutonium nitrate solution in a form suitable for shipment to Los Alamos. These Operating Standards for the operations to take place in Building 231 were all prepared by G. W. Watt.

We spent the day working our way through these documents. I am being asked to sign individually in each case as we finish discussing them. My signature constitutes a final sign-off and perhaps a sort of guarantee that these processes will work.

After a full day I returned to Richland and spent the night at the Richland transient quarters.

More than 1,300 German prisoners of war went on a sitdown strike yesterday at Fort Sheridan, following removal of one of their comrades as a group leader. This is the first serious difficulty the post has encountered since the arrival of prisoners.

Thursday, December 14, 1944

We are continuing today, in the conference room of the research building (no. 3706) in the 300 Area, our rather arduous examination of the documents covering the individual Operating Standards of the Bismuth Phosphate Process. It is again a matter of proceeding systematically through the documents, discussing each step in detail, making suggestions and revisions, and my signing the documents as we finish them.

I again spent the night at the Richland transient quarters.

Friday, December 15, 1944

I attended the concluding meeting to consider the final specifications for the Bismuth Phosphate Process. We made our choices among the alternate possibilities in the decontamination cycles. The final Process, as authorized for operation, contains two decontamination cycles, in which the first precipitation of bismuth phosphate from the oxidized solution will include ceric and zirconium phosphates while the second oxidized cycle will consist only of bismuth phosphate; the ammonium fluosilicate will be used during both of the reduced precipitation steps in which plutonium is carried by bismuth phosphate.

I am spending my last night at the Richland transient quarters before returning to Chicago.

Lupe Velez, noted movie actress, committed suicide.

Saturday, December 16, 1944

In Richland. I caught the Northern Pacific R.R. North Coast Limited for Chicago, boarding in Pasco, Washington. W. Q. Smith of Clinton Laboratories is sharing a bedroom with me. Hogness is on the same train.

Sunday, December 17, 1944

Enroute to Chicago on the North Coast Limited.

The Green Bay Packers won the National Football League championship by defeating the New York Giants, 14-7.

Monday, December 18, 1944

I arrived back in Chicago at 9:00 p.m. Helen told me she attended her chemistry class at YMCA College last Tuesday and Thursday.

Tuesday, December 19, 1944

While I was away from the Lab last week and yesterday, the following events took place.

Monday, December 11, 1944

Kennedy wrote to me that five of Wahl's old notebooks are being sent to me by a messenger who will follow his letter by a few days. He suggested it would be worth pointing out to Metcalf that though the notes extend past April 1, 1941, no work described in them is covered by any Government contract, except for the preparation of Samples B and F, which hardly can be described as research.

My office received a teletype addressed to me from Allison, saying that the 20 micrograms of Np^{237} requested in his December 4 teletype is included in Segrè's request and that they could use the 20 micrograms as soon as available and then 80 micrograms later on when more is produced. He also expressed interest in our 48 sample but left it up to me to decide whether his program or ours could make better use of it. He asked my advice on whether it is all right to attempt to decontaminate U^{233} enough so that beta-particle emission would offer data on the presence of U^{238} equal in accuracy to the mass spectrograph; he expressed the opinion that Dempster's results should be reexamined.

Tuesday, December 12, 1944

Keith McLane was recommended for promotion from Research Assistant to Junior Chemist by Hindman and Manning.

James, continuing the purification of sample 49NC-7 (derived from 4.4 mg PuO_2 that received about 350 Mwd of neutron irradiation in the Clinton pile), completed the series of four silver plus persulfate oxidation cycles which he began on December 8. The resulting sample 49NC-14 gives 115 alpha-particle counts per minute. James also investigated other fractions, i.e., 49NC-12, 13, and 14; these would contain any activity oxidizable by $S_2O_8^{=}$ and reduced by SO_2. He carried out a dichromate oxidation cycle and determined that there is no alpha activity (or at least less than 10 c/m) that is not oxidized by dichromate but is oxidized by persulfate.

Stone urged Compton to continue the Health Division's research activities as an essential and yet incompleted part of the Metallurgical Project's program. He pointed out that we are just beginning to understand the hazards of plutonium and the levels at which damage may occur, and we are just on the threshold of finding out what various fission products may do when in the body in amounts not sufficient to cause acute trouble.

Wednesday, December 13

The Council of my section met at 8:00 a.m. in my office; the meeting was attended by Albaugh, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Katzin, Kraus, Lawroski, Manning, Nickson, Schaffner, Simpson, Stewart, and Roy Thompson. Manning presided. Nickson reported, for practically the whole meeting, on the present knowledge of the pharmacology of plutonium. Most of the data presented has been obtained by Hamilton from rat experiments and described in Report CN-2383. Massive amounts of radium and plutonium were injected intravenously in rats to determine the amounts required for acute death. Plutonium was found to be slighly less toxic than radium. Because deaths are attributable to liver damage, the significance of the results is therefore somewhat obscured. Absorption from the gut is less than 0.05%. Daily urinary excretion is about 0.01%.

Nickson summarized the present status: There is considerable holdup of plutonium in the alveoli of the lungs (when plutonium is injected into the lungs) and such amounts as leave the lungs by absorption are deposited in the bone. The excretion rate is small, and the half-life of plutonium in the body may be a matter of years. No promising therapeutic measures have been developed.

One of our laboratory assistants, Inez Jones, was discharged today.

Sample SA-1 arrived from Clinton. It consists of 8.2 mg of PuO_2 (7.23 mg of plutonium) (prepared by Ralph James on May 24 of this year) which has been irradiated in the Clinton pile from June 5 to December 3, receiving a total of 10,233,244 kwh or about 426,400 kwd of irradiation. The sample arrived last Saturday but was not noticed in the office until today, December 13. James and Morgan began their chemical operations on the sample.

The evening meeting of the Separation Processes Groups of Section C-I began at 7:45 p.m. in Room 209, Eckhart Hall, and was attended by Abraham, Albaugh, Ames, Arnold, Beard, Blaedel, Daniels, Davidson, Egan, Fields, Gilbreath, Hagemann, Hufford, E. K. Hyde, Hyman, J. J. Katz, Katzin, Kraus, Larson, Lawroski, Manning, Margolis, McKinney, S. Peterson, Phipps, Quinn, Schaffner, Sedlet, Sheft, Simpson, Watters, Westrum, Winner, and others. Katz reported on the isolation of Np^{237} at Clinton from the two plant runs processed by the modified procedure that was devised to enable recovery of Np^{237}. Results show that about 20% of the Np^{237} was carried through to the lanthanum fluoride-plutonium precipitation step. This precipitate was metathesized with $KOH-K_2CO_3$ and dissolved in HNO_3. A peroxide precipitate was removed from this solution by the high acidity procedure and dissolved in H_2SO_4. The separation of the plutonium from the neptunium was effected by oxidizing with NaOCl to convert the neptunium to an oxidized fluoride-soluble state, leaving the plutonium in a fluoride-insoluble state. The plutonium was precipitated as K_2PuF_6, the supernatant was reduced with SO_2, and the neptunium carried out of the solution with lanthanum fluoride. The overall yield of Np^{237} in the isolation step was greater than 90%.

Greenlee reported on efforts to develop an analytical method for

Wednesday, December 13 (cont.)

Pu(III) in which the efficient carrying of Pu(IV) by zirconium phenylarsonate and slight carrying of Pu(III) by the same precipitant would be utilized to determine the Pu(III) content of process solutions. Results are promising, and it appears that the method should be accurate to within about 3%.

Hyman reported on test runs to determine the behavior of Np^{237} in the Bismuth Phosphate Process. He found that decontamination with respect to 93 is obtained only in plutonium precipitations; by-product precipitations, with or without scavengers, carry no 93. The modified procedure that has been devised for recovery of 93 employs an $H_2C_2O_4$-Mn^{++} prereduction, probably following a MnO_4^- preoxidation, and U(IV) as the reducing agent in the plutonium precipitation steps. With this modified procedure, neptunium yields of 60-98% have been obtained in the extraction step and yields of 60% to 90% in plutonium precipitation steps. Thus, it appears it should be possible to obtain overall neptunium yields of from 30-80%.

Blaedel reported on investigations to determine the feasibility of using trifluoroacetylacetone as a specific complexing agent for Pu(IV) in the extraction and decontamination of plutonium; the studies being made as a result of a verbal presentation by Calvin of data obtained at Berkeley. The results showed efficient extraction of plutonium but unsatisfactory decontamination of gamma-ray and beta-particle activity. It appears that decontamination can only be achieved by very high extract to raffinate ratios, i.e., extracting the plutonium into very small volumes of solution.

Thursday, December 14

Edrey S. Albaugh teletyped Kennedy at Los Alamos asking whether he plans to be at the Chicago meeting next week.

Gilbreath and Morgan were both recommended for promotion from Junior Chemist to Associate Chemist by Albaugh and Manning.

Manning, Simpson, Phipps, and Davidson were invited by Farrington Daniels to attend a symposium on the "Metallurgy of Tuballoy and Its Alloys," which is being held today and tomorrow.

At an afternoon meeting of Division Directors in Eckhart Hall, Stearns and Wigner reported that a letter has been received from Compton asking if it is possible to make plans for converting 100 grams of Pu^{239} into at least 70 grams of U^{233} per day. Wigner outlined the sandwich pile which his Physics Division considers to be the most practical for quick results. (Plutonium is sandwiched between thin aluminum sheets folded in the form of a bellows and placed in a cylindrical tank two feet in diameter and eight feet high. Ordinary water is forced through the bellows. Surrounding the plutonium cylinder and the water cooling is a concentric cylinder of thorium metal, oxide or oxide slurry.) The plutonium must be decontaminated from fission products by a factor of 10^7 once every ten days

to remove neutron poisons and to make it possible to refabricate the plutonium; high yields of plutonium (99.9%) are required to make the desired yields of U^{233} possible. A report to Compton is to be written by January 6. Daniels was asked by Wigner to bring back a report from the Chemistry Division on the feasibility of obtaining high efficiencies (yields of the order of 99.9%) in purification and decontamination by the solvent extraction method.

Report CN-2414, "Chemical Research — Separations Processes for Plutonium, Chemistry Division," a joint report of the Chemistry Division and Technical Division, was issued. The report, dated November 15, 1944, contains an account of the following research.

Uranium recovery from process waste solutions. Hyman has conducted experiments to determine the effect of varying conditions on the gamma-ray decontamination factor attained by treatment of the recovery solution with titanated silica gel and $CaCO_3$ scavengers, followed by precipitation of uranium with acetic acid. Batch-wise extraction of the HNO_3 solution of this precipitate into dibutyl carbitol, using $Al(NO_3)_3$ as the salting-out agent and subsequent extraction into water, leads to an overall gamma-ray decontamination factor exceeding 10^3.

The Technical Division has carried out two 75-liter scale runs to test the behavior of the lanthanum fluoride reprecipitation procedure for the removal of iron from process solutions. In these runs, the reprecipitation has been from a solution 10 M in HNO_3 and 1 M in HF. Plutonium losses in the reprecipitation have been inconsistent, 33% and 9%, as compared with an average loss of 8% in previous runs in which the lanthanum fluoride was reprecipitated from a solution 1 M in HNO_3 and 0.57 M in HF.

Art Wahl's notebooks nos. 11, 12, 13, 14, and 16 were delivered to my office by the Chicago Area Engineer's Office. They have been sent from Los Alamos by Kennedy for review by Lt. Col. Metcalf in connection with Patent Cases 52 and 61. The notebooks were turned over to Metcalf today, along with my notebook no. 1.

Leon Leventhal, a SED man, is transferring to Los Alamos today. Eugene A. Hausman, another SED man who has been working in the Technical Division, is transferring to Section C-I and will continue to work with Lawroski. Nellie Jennings, a laboratory assistant since April 1943, was discharged because of poor work and attendance.

Katzin wrote a summary of the U^{233} work for my use at the Project Council Chemistry Information Meeting on Tuesday. Topics covered are (1) Additional information has been obtained on salting-out action of various nitrates on thorium and uranium. Data obtained are in harmony with the interpretation that high extraction of thorium into ether results in "salting back" of the uranium into the aqueous phase. The desirable combination of high salting of uranium and low salting of thorium into ether is shown most outstandingly by calcium nitrate, by ammonium nitrate at high concentrations, and by aluminum nitrate at low concentrations.

Thursday, December 14 (cont.)

(2) Tests on solvents for use in the uranium-thorium separation have been extended to the extraction on protactinium. It has been found that diisopropyl ketone will extract protactinium to the extent of 50%. (3) Calculations and analysis of data obtained on the 4n + 1 decay chain indicate that as little as 10^{-8} parts of U^{232} formed by the n,2n reaction on U^{233} or Pa^{233} may produce sufficient amounts of radio-thorium and the subsequent members of the thorium decay chain to complicate the experiments on the 4n + 1 series. So far, it has been learned that RdNp(Th^{229}) probably has a half-life of several thousand years and NpX (Ra^{225}) has a half-life of about one month. (4) If a radium-thorium separation is made and the radium fraction set aside for several weeks, the ThX and its daughter activities decay out leaving the presumably pure neptunium series members. Experiments on material that has been handled in this fashion have confirmed the presence of Pb^{209} (NpD) with the known half-life of 3.3 hours. Isolation of the bismuth fraction from the aged NpX has given an alpha activity of approximately 45 minutes half-life. Although no accurate alpha-particle range curve is available from which to determine the branching ratio, there are definitely two differing ranges of alpha particles detectable, one of which, no doubt, corresponds to NpC and possesses a very long range. Unsuccessful attempts to isolate members of the series between NpX and NpC, correlated with the rate of growth of NpC and NpD, indicate that all other half-lives involved are probably shorter than ten minutes. No decision can be made yet concerning possible branching at NpX and other points along the chain.

Friday, December 15

Mulliken issued an outline of the status as of December 15 of the series of volumes that will constitute the Metallurgical Project Record. The titles of the 24 volumes are indicated. The transuranium elements will be covered in volume 16.

Alice Parnell, a stenographer who used to work for Beaton and Dreher, resigned today because of home responsibilities. A request was made to formalize the transfer of John H. Schraidt and Wilbur Simon from Maloney's section to Lawroski's group. W. C. Giegold is being transferred from Clark Egan's Semiworks group to Hanford.

Edrey, my secretary, received a teletyped reply from Kennedy in Los Alamos saying that he will not be at the Chicago Project Council Chemistry Information Meeting scheduled for Tuesday.

In the afternoon Manning, Katzin, and Davidson met with Daniels to discuss the feasibility of obtaining high efficiencies in purification and decontamination operations required in connection with the converter pile being designed for the conversion of 100 grams of plutonium per day to U^{233} with a yield of at least 70 grams. It was concluded by everyone that at the present state of solvent extraction development the best efficiency would be about 95% for a decontamination factor of 10^2 making it necessary to fabricate the plutonium by remote control, and that the required yield of 99.9% and 10^7 decontamination is impossible.

Monday, December 18

B. Wendrow and H. Hasenfus, two of the men in Egan's Semiworks Group, were transferred today to Oak Ridge.

At Hanford, the first tracer run was to be started through the 221 building in the separations plant using material containing irradiated Clinton slugs that were dissolved on December 6.

James and Morgan have spent most of the past week in the chemical operations begun last Wednesday to remove the 93 and 95 fractions from sample 49ND, the 7.23 mg Pu^{239} (8.2 mg PuO_2) that has received about 426 Mwd of neutron irradiation in the Clinton pile. At this point they have three fractions: (1) the 95 fraction, 49ND-14, containing 38 micrograms; (2) the 93 fraction, 49ND-6; (3) the 94 fraction, 49ND-13, containing 5.83 milligrams of plutonium, turned over to O'Connor.

Albaugh wrote a summary of work on Separation Processes in Section C-I for my use at Tuesday's Project Council Chemistry Information Meeting. The topics covered are (1) *Increased production by the proposed "four ton/day" process.* The process has been tested in two runs using Clinton metal solution fortified with plutonium and inactive FPE. Losses have been quite low except for 7% losses in the extraction step in both runs and an unexplained loss of 12% in the second cycle by-product step in one run. Radioactive decontamination has been quite satisfactory. (2) *Analysis for Pu(III) and Pu(IV) by precipitation of zirconium phenylarsonate.* (3) *Preextraction treatment.* The efficiency of $KMnO_4$ or $K_2Cr_2O_7$ in destroying N_2H_2 in process solutions has been further demonstrated. (4) *Isolation of Np^{237} at Clinton.* Preliminary indications are that about 2.3 mg of neptunium have been recovered from the two modified plant runs. (5) *The use of trifluoroacetylacetone (TFA) in extraction-decontamination.* A two-cycle batch extraction of dissolver solution has been made under conditions outlined by Calvin. TFA was found capable of giving excellent separation of plutonium from uranium and also high concentration factors for plutonium. The decontamination factors, however, are small, being 10-100. (6) *Hydroquinone stability.* Hydroquinone is the most suitable reducing agent yet found for the solvent extraction-decontamination process. Studies in progress indicate that there is a need to increase its stability before testing under continuous counter-current conditions. (7) *Isolation of 93 from Clinton plant solutions.* Using Clinton concentration of plutonium and tracer concentrations of Np^{239}, the men have carried two 100-ml scale experiments through the extraction step, one bismuth phosphate cycle and the crossover, using $H_2C_2O_4$-Mn(II) and U(IV) as reducing agents in place of $NaNO_2$. A 30% loss of neptunium occurs in the extraction step. There is also an unexplained 20% loss in the lanthanum fluoride by-product precipitation in the crossover step. (8) *Decontamination at Hanford with respect to 93^{239}.* Three 100-ml experiments using Hanford flowsheet conditions have been carried out to test the efficiency of $(NH_4)_2SiF_6$ in decontaminating Np^{239}. The overall decontamination factors through the crossover are 770 for the run without $(NH_4)_2SiF_6$ and 232 for the two runs that include it. (9) *Semiworks study of decontamination in the Bismuth Phosphate Process.* Seven 100-liter scale process runs have been completed through

Monday, December 18 (cont.)

an extraction step and two decontamination cycles. Four runs have followed the Hanford flowsheet except for the omission of scavengers (baseline) and three others have been of the low acidity type. The low acidity runs give better gamma-ray and beta-particle decontamination but a lower plutonium yield (61% as compared with 80% for the baseline runs). (10) *Concentration-isolation in the continuous extraction columns using hexone.* A series of runs with blank feed solution have been made in the three-inch diameter column. Operation has been generally satisfactory and indicates that greater than 99.9% plutonium purity can be attained. (11) *Fundamental research on utilization of hexone for decontamination and concentration-isolation.* Preliminary experiments have been performed on the distribution coefficients for decontamination starting with dissolved bismuth phosphate extraction precipitate as feed solution. Various salting out agents have been tested, indicating that $Mn(NO_3)_3$ and $Al(NO_3)_3$ give the highest distribution coefficients for plutonium (alpha particles), while NH_4NO_3 and $Ca(NO_3)_2$ give the highest values for sources of beta particles and gamma-rays.

At 2:00 p.m. there was a meeting in Daniels' office to discuss the problems of the converter pile. Those present were C. M. Cooper, Daniels, Davidson, English, Katzin, Maloney, Manning, and Tepe. All agreed that the efficiency in chemical processing of plutonium that Wigner hopes for, namely 99.9%, is impossible if decontamination is to be 10^7. If decontamination of 100-fold is permitted, requiring remote control for plutonium sandwich refabrication, a yield of 95% might be hoped for. Other types of piles were discussed with English advocating the homogeneous heavy water pile, Cooper the homogeneous pile using circulating water with plutonium nitrate in solution, and Daniels the high temperature pebble pile using bismuth as coolant.

Report CS-2456, "Chemistry Division Summary Report for November 1944," was issued. It includes abstracts of our work in Section C-I, submitted by us to the Director's Office as Report MUC-GTS-1154.

The biweekly Monday evening Chemistry Division Meeting was held at 7:45 p.m. in Room 251, Ryerson Laboratory. Weinberg spoke on "Elements of Pile Calculations."

* * * * *

At 9:30 a.m. I attended the Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Arnold, Bartky, Boyd, Burton, Cohn, Compton, Connick, Coryell, Daniels, Dempster, Doan, English, Fred, Hilberry, Huffman, Jeffries, W. Johnson, Manning, Mulliken, Nickson, Rabinowitch, Spedding, Stearns, Stone, Sugarman, Szilard, Turkevich, Wakefield, Watters, Whitaker, Wigner, Zachariasen, and Zinn. I was first on the program and reported on investigations of separation processes for Hanford, on basic chemistry, and U^{233} as follows:

(1) Separation processes — Albaugh's people have tested the entire Hanford flowsheet with 10% losses. Most of the loss occurs in the extrac-

tion step, but it is not yet certain whether the loss is caused by plutonium being in the III or the VI valence state. I reported on the decontamination of neptunium and the semiworks experiments on the low acidity process. I pointed out this is the last report from the semiworks, which is closing out.

(2) Basic chemistry — (a) Cunningham, Davidson, and Hindman's groups have studied various plutonium reactions, e.g., reactions of oxygen with PuF_3 and PuF_4, formation of plutonium carbide by reduction of PuO_2 with carbon, the reaction of PuO_2 with ammonia and hydrogen in graphite crucibles (which gives a mixture of Pu_2O_3 and Pu_4O_7 as identified by Zachariasen). (b) The oxidation-reduction potential of the pair U^{+4}-U^{+6} has been redetermined at a lower value. (c) Two runs of half-ton batches at Clinton have recovered about 2 mg of Np^{237}, some of which has been sent to Site Y and some of which will go to English at Clinton. We will use some to study the basic chemistry of neptunium and a small amount will be used for measuring the cross section of the reaction $93^{237}(n,\gamma)93^{238}$. (If the neutron absorption cross section of 93^{237} is of the order of 200 barns, the specific alpha activity of the plutonium produced at Hanford will be larger by 3% than was expected because of the formation of Pu^{238}.) (d) The decay of Pu^{238} has now been followed for about a year, and its half-life is estimated at 50 years. A search has been instituted for the plutonium isotope, Pu^{240}. No positive results are yet available, but we have not yet investigated the alpha-particle activity of the recently obtained material containing as much as 5 grams of plutonium per ton. We have also looked for beta-particle activity as evidence of the isotope Pu^{241}, but no beta-particle activity has been found in the material containing 5 g plutonium per ton. The isotope Pu^{241} could also be converted, by alpha-particle emission, into U^{237} — a known 7-day beta-particle emitting uranium isotope. We have found some evidence of a growth of such alpha-particle activity in samples suspected to contain Pu^{241}, but results are as yet very tentative.

(3) U^{233} program — Katzin's group has made preparations for the extraction of 37 cans of irradiated thorium carbonate shipped from Clinton, and 28 mg of U^{233} have so far been extracted. We have redetermined the half-life of U^{233}, based on the new isotopic composition of 96% U^{233} and 4% U^{238} measured by Dempster (the old measurement was 86% U^{233}). The new value is 1.6×10^5 years. I presented the latest information on the neptunium decay series.

I was followed by other speakers. Topics covered by them which are of interest to us are: Sugarman spoke on the I^{135}-chain yield, neutron absorption cross section of Xe^{135}, and identification of 25-year cesium. Watters of Section C-IV described rare earth determination in thorium and uranium, oxygen determination in thorium, polarographic determination of plutonium, and referred to a thesis on the polarography of uranium which describes U^{+5}. Connick from Berkeley reviewed the work with TFA and on plutonium phosphate and peroxide complexes. Spedding of Ames Laboratory showed a successful casting of a 60-pound, 6-inch diameter thorium metal hemisphere.

Cohn (Section IV, Clinton Laboratories) described the preparation of high-activity carrier-free fission products. Boyd (Section III, Clinton Laboratories) presented more theory on the adsorption process.

Coryell (Section II, Clinton Laboratories) reviewed the large-scale production of Ba^{140}, work on the I^{135} chain, and the preparation of milliliter amounts of tritium. English (Section I, Clinton Laboratories) described recent Clinton plant runs using $(NH_4)_2SiF_6$, reprecipitation of bismuth phosphate as a means of improving decontamination, studies of the high solubility of plutonium in the peroxide precipitation step and studies on the use of plutonium sulfate in homogeneous piles.

I sent a memo to Colonel H. E. Metcalf noting that six personal research notebooks (five of Wahl's and one of mine) have been turned over to him for his custody and for use, on a confidential basis, as supporting evidence for the definitions of joint inventions by Kennedy, Segrè, Wahl, and me. I said we are looking forward to reaching an agreement with him on this matter on or before January 15. I sent copies of the letter to Lavender, Kennedy, Wahl, and Segrè.

James is continuing the purification of sample 49NC-14 (derived from 4.4 mg PuO_2 that had received about 350 Mwd of neutron irradiation in the Clinton pile). It still shows about 120 alpha-particle counts per minute not oxidizable by either dichromate or persulfate. During the past week James has carried out a persulfate oxidation followed by a zirconium iodate precipitate which carried 80% of the alpha activity. This precipitation resulted in sample 49NC-18.

I reviewed a memo (MUC-GTS-1201) prepared for Daniels on the pile proposed by Wigner for converting Pu^{239} to U^{233}. In this memo I note that the losses envisioned by Wigner in the operation of the pile are such that the 30% operating margin is essentially completely used up in the physical losses caused by neutron absorption by fission products, materials of construction, etc. A complete 100% yield is necessary for all chemical procedures to stay within the required 70% efficiency. I include the following table, based on the assumptions that the converter will run on a cycle in which either one-tenth or one-twentieth of the material must be processed daily to eliminate neutron-absorbing fission products:

Chemical yield (%)	Production loss (grams)	Overall efficiency (in percent, 10-day cycle)	Overall efficiency (in percent, 20-day cycle)
100	0	70	70
99.9	2.8	68.2	69.0
99.5	14	61.4	65.4
99.0	28	54.6	61.4
98	56	44.9	54.6
95	140	29.2	41.0
90	280	18.4	29.2

I point out that an efficiency of 99.9% for a decontamination by a factor of some 10^7 followed by refabrication of an aluminum sandwich is totally impossible by any known process and probably also by any conceivable process. It is the consensus that a process giving a 95% yield is an optimistic possibility which means an efficiency of not better than 41% for a 20-day cycle. This might involve a combination of precipitation and solvent extraction procedures.

I suggest that if the decontamination requirement can be lowered to a factor of 100 (presuming it is feasible to have remote-control refabrication of the fuel), a chemical yield of 98% is possible by a solvent extraction procedure alone. I also mention, as an alternative possibility, a homogeneous reactor with ordinary water that would require a minimum of decontamination (only to remove fission product poisons) and would avoid the hold-up due to fuel fabrication operations; however, little is known about this approach, and hence this could require the most research work.

With regard to the processing of the thorium blanket in Wigner's converter pile, I indicate the problems seem straightforward, although there would be a delay in harvesting the uranium because of the need to await the decay of the 27.4-day protactinium precursor. I suggest the use of thorium metal as the best practical form.

I mention some of the heavy isotope problems that will arise in the operation of a converter unit. Pu^{240} will be formed at the rate of 40 g/day if the rate of fission of Pu^{239} is 100 g/day. If Pu^{240} has a low fission cross section (and a low capture cross section), the average cross section of the plutonium in the pile will decrease at a rate of 15 barns per day. If Pu^{240} has a capture cross section of 10 barns, there may be 10 g of Pu^{241} in a unit after 100 days operation.

In summary, I say we favor the heterogeneous arrangement with refabrication by remote control, in which case an "individual" chemical cycle yield of 98% may be possible, corresponding to an overall yield of some 55% U^{233} on a 20-day cycle. Thorium metal seems to be the best material for the thorium absorption blanket.

I indicate this summary was prepared at my direction by Katzin and Davidson.

"Chemical Research — Basic Chemistry of Plutonium. Report for Month Ending December 1, 1944, Section C-I, Sub-Section II, B. B. Cunningham, Assistant Section Chief," (CN-2431), was issued. The following investigations are reported:

Basic Dry Chemistry Group (Davidson, Group Leader). *Reductions of PuO_2* (Westrum). The previously reported reduction in vacuo of PuO_2 on tantalum at 1650°-1800°C has been repeated on iridium under similar conditions. The reduced phase has the composition 20% "Pu_2O_3-Pu_4O_7" - 80% PuO_2 (identification by Zachariasen). PuO_2 on platinum at 1200°C in the presence of anhydrous H_2 at one atmosphere reacts with the platinum, but the oxide phase remains PuO_2. It is concluded that "Pu_2O_3-Pu_4O_7" in contact with excess platinum decomposes to give PuO_2 and a "Pu-Pt alloy." Davidson has obtained Pu_2O_2S by heating PuO_2 for 3/4 hour in a graphite crucible at 1225°-1300°C in an atmosphere of H_2S; also Pu_2S_3-Pu_3S_4 by

heating PuO_2 for two hours at 1340°-1400°C. Melting points of $PuBr_3$ and $PuCl_3$ have been determined by Robinson to be 681°C and 760-765°C, respectively.

Isolation and study of 93^{237}. La Chapelle has prepared an insoluble, colorless peroxide from a reduced neptunium solution. Magnusson has studied separation of plutonium and neptunium by a series of lanthanum fluoride oxidation-reduction cycles using hypochlorous acid in 0.5 M H_2SO_4 as the oxidizing agent. The neptunium has been found to be 97% oxidized within 20 minutes, whereas 97% of the plutonium is in the reduced state.

Recovery group (Dawson, Group Leader). Research on methods for recovery of plutonium. Asprey, Anderson, and Dawson have adopted several modifications in recovery procedures, including (1) use of smaller quantities of lanthanum and a greater number of precipitations in carrying plutonium with lanthanum fluoride, (2) hexone extraction in two cycles instead of three to reduce the amount of impurities carried over, (3) the use of ferric or aluminum ions as complexing agents, and (4) removal of plutonium from hexone by a dilute aqueous solution of H_2O_2 or hydrazine sulfate.

Helen went to her chemistry class at YMCA College today.

U.S. planes again raided the Nagoya airplane factory in Japan.

Wednesday, December 20, 1944

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, J. J. Katz, Katzin, Kraus, Lawroski, Manning, Schaffner, Simpson, and Stewart. I again brought up the matter of security violations which seem to have become very serious in the last few weeks and consist mostly of unlocked files and notebooks left out of them. The possibility of levying $1.00 fines was discussed. I then gave a report on my visit to Hanford last week.

There was a discussion of the program for publishing the Project literature. Jake Warner will assist in directing the writing program. The probable plan is to have two types of writing, one to consist of 24 survey volumes, each with a separate editor. The second part will consist of volumes each with a number of research papers written in journal style by the men who have done the actual work. The program is to be finished by June 30 next year and has highest priority — we are expected to spend 30% of our time in writing. Our primary interest is in Volumes 16 to 19. I have been suggested as editor for Volume 16, which will cover the chemistry of the transuranium elements. Volume 17 will cover the bismuth phosphate process — Perlman has been suggested as editor. Volume 18 will cover alternate chemical separation processes and Volume 19 will cover U^{233}. I spent the last part of the meeting giving a report on the Project Council Chemistry Information Meeting of yesterday.

Manning delivered my memo (MUC-GTS-1201) completed yesterday to Daniels the first thing this morning for Daniels to pass on to Wigner.

Zene V. Jasaitis terminated work at the Met Lab. His leave of absence has expired, and he is returning to Shell Oil Company and Shell Development Company in Los Angeles (Wilmington).

Daniels sent Wigner a memo about his conception of a high temperature bismuth-cooled converter pile to be fueled with pellets of plutonium oxide spaced in a lattice composed of pellets of beryllium oxide. The U^{233} is formed in an outer shell through which thorium oxide powder is passed slowly.

At 4:00 p.m. I attended a meeting on conversion piles under Wigner's chairmanship. Compton was also present. At Daniels' request, I presented technical details of the chemistry involved in the processing of fuel from Wigner's sandwich pile. Wigner was disappointed in the low yields, but we indicated such yields would be inevitable. C. M. Cooper described a heat exchanger which he has proposed for homogeneous water piles. Greninger outlined his idea of sandwich fabrication. Daniels discussed the high temperature bismuth-cooled pile.

At the conclusion of the meeting, there seemed to be the general opinion that there is probably a good chance that a converter pile can be made which will be certain to operate with an efficiency of at least 50% for the conversion of Pu^{239} to U^{233}. All effort must be directed toward trying to make an attractive and practical case for this pile to be presented by January 6.

The evening meeting of the Basic Chemistry, Recovery, and Instrument Groups of my section, at 7:45 p.m. in Room 209, Eckhart Hall, was attended by Arnold, Brody, Cunningham, Daniels, Dixon, Ghiorso, Hagemann, Hufford, Jaffey, Katzin, Krueger, Larson, Manning, McLane, Morgan, Nickson, S. Peterson, Phipps, Sedlet, Sheft, and me. I turned the meeting over to Cunningham to preside as usual. Jaffey gave the first of a series of lectures to be presented at these meetings to acquaint chemists with the essentials of nuclear physics. His topic was the concept of nuclear cross sections. Near the end of Jaffey's lecture Katzin asked whether the unit of cross section referred to is a barn or a barn door, to which I replied that the unit is called a barn because it is as large as a barn door in comparison with actual cross sectional areas of nuclei. The term was originated by Fermi. I also called attention to the fact that the series of lectures on the fundamentals of nuclear physics begun by Jaffey will include concepts with which any chemist having worked on the Project should be thoroughly familiar.

Nickson next reported on the physiological research on rats and rabbits done thus far with plutonium on a tracer scale by Hamilton at Berkeley and by Cole at Chicago, including some late, hence yet tentative, work done at Chicago on acute lethal dose and chronic lethal dose levels. Tracer work has been carried out with all three valence states of plutonium, but the method of preparation of the tetravalent form is likely to produce the colloidal form so these results are doubtful. Results of intramuscular injections show similar distribution after 64 days of all three states,

with 3 to 4.3% in the bone, 0.1 to 1% in the lungs, and 0.35 to 0.58% in the liver. Subcutaneous injections show similar results. This indicates that plutonium exists mainly as the IV oxidation state in the body. Intravenous injection shows the liver retains (after 64 days) 38% of the III, 73% of the IV (high due to colloid form?), and 29% of the VI. For pulmonary administration (solution introduced directly into lungs) the distribution after 64 days between the lungs [L] and bone [B] is III (L25%, B66%), IV (L66%, B24%), VI (L6%, B88%). Gastric injection (i.e., through the digestive tract) leads to less than 0.05% absorption. The plutonium/radium toxic amount ratio of 50/1 estimated earlier may be lowered as a result of tentative data on an acute lethal dose level (which, of course, may not actually be applicable to the chronic case).

Dr. Copp has suggested the use of a low calcium, high parathormone diet in cases of plutonium ingestion. This is followed by a high calcium, high vitamin D diet which sheaths the plutonium in the bone with calcium, thus protecting the vulnerable marrow.

Nickson described the bone structure calling attention, in particular, to the spicules, which are bony partitions extending through the marrow. Whereas radium largely deposits on the bone itself, plutonium deposits equally well on the spicules where its damage to the bone marrow is greatest. Nickson concluded by saying that in view of these preliminary results the present tolerance limits for activity in air and environment have been set, conservatively, low enough to take care of any lowering of tolerance that may come in the future — the present tolerance in air is 5×10^{-16} g/cc. Air samples taken so far show that the highest level in New Chem is one-third tolerance. In the West Stands, the highest is two-thirds tolerance. The only reading above tolerance was obtained on the roof in the exhaust from the hoods of Room 34 (Britain, Fineman, Haeckl, and Leventhal), New Chem, where the plutonium level is 10 times tolerance. Nickson said that plans are under way to secure some 50-year Pu^{238} in order to study acute lethal dosage levels without chemical toxicity. I suggested that such a target be worked up in Section C-I.

Report CN-2432, "Chemical Research — Separation Processes for Plutonium. (Section C-I, Sub-section I, Albaugh, Assistant Section Chief.) Report for Month Ending December 1, 1944," was issued. A summary of the research reported is as follows:

Extraction-Decontamination (Roy Thompson, Group Leader). *Methods of increasing the rate of metal processing at Hanford.* S. Peterson concludes, on the basis of a total of 95 small-scale extraction experiments, that satisfactory extractions can be made from 28% or even 30% UNH if conditions are carefully controlled. Losses are minimized by increasing the effective length of time during which precipitation occurs. *Effect of N_2H_4 in process metal solutions.* Malm, Bartell, Ader, and R. Thompson have extended previous studies on destruction of N_2H_4 by an oxidation treatment with either $KMnO_4$ or $K_2Cr_2O_7$ at concentrations from 0.005 N to 0.02 N with extraction losses usually of less than 1%. Destruction of N_2H_4 by heating has been rechecked and extraction losses have been reduced to normal after only 5-8 hours heating even though considerable Pu(III) still remains. *Differentiation of Pu(III) and Pu(IV) by zirconium phenylarsonate.* Greenlee and Bartell have studied the use of zirconium

phenylarsonate as a selective carrier for Pu(IV) in the presence of Pu(III) which is not carried. This carrier has been effective at Hanford concentrations of plutonium in 2.5 N HCl, but is less satisfactory in UNH-HNO_3 solutions.

Concentration-Isolation (J. Katz, Group Leader). The behavior of neptunium in the concentration-isolation process has been studied from the point of view of developing a method for separating small amounts of neptunium from large amounts of plutonium. In the peroxide precipitation any procedure which gives high yields of plutonium peroxide also results in at least 95% neptunium carrying. A procedure has been developed which involves dissolving the peroxide precipitate in H_2SO_4 and adding NaOCl to oxidize the neptunium while leaving plutonium in the IV state. The plutonium is removed from the solution as K_2PuF_6 by addition of KF and HF. This procedure removes 98% of the plutonium and leaves more than 90% of the neptunium in the supernatant. The procedure for isolation of neptunium is being tested in two plant runs at Clinton. Preliminary investigations also have been made on a solvent extraction method for separating neptunium from plutonium that depends on a preferential reduction of plutonium to nonextractable Pu(III), while maintaining neptunium in the IV state which is extractable with hexone. Hydrazine nitrate has been tested successfully as the reducing agent.

Process Development (Gilbreath, Group Leader). *Distribution of neptunium in the Bismuth Phosphate Process.* Larson and Hyman have made two 100-ml scale runs and find that when either $H_2C_2O_4$-Mn(II) or U(IV) is used as the reducing agent instead of Fe(II) in the plutonium precipitation step in the decontamination cycle, the loss of neptunium in the waste solution is less than 5% and plutonium yields are normal. Prereduction with NO_2^- in the extraction precipitation step gives a 50% loss of neptunium. *Choice of a solvent for the extraction, decontamination, and concentration of plutonium.* Blaedel, Walling, and Post have investigated the following solvents: hexone, dibutyl carbitol, 2-ethyl butyl cellosolve, diethyl cellosolve, dibutyl cellosolve, ethyl butyl cellosolve, nitromethane, methyl isobutyl carbinol, methyl isobutyl carbinol acetate, 2-ethyl hexoic acid, and diethyl ether. Although no solvent fulfills all of the requirements completely, hexone appears to be the most promising.

Concentration and Isolation of Plutonium by Solvent Extraction (Dawson and Lawroski, Group Leaders). *Hexone extraction for isolation — Research.* Asprey, Britain, Stewart, and Dawson have completed a series of experiments designed to develop an alternate isolation process using batch extraction methods in which hexone is employed to remove plutonium from the dissolved metathesis product. Three cycles using a volume of hexone amounting to 1.25 times that of the aqueous phase have been employed. Yields of 97.5 to 99% of the plutonium which are better than 99.7% pure may be obtained with excellent separation from lanthanum iron, and manganese. *Hexone extraction for isolation — Development.* Brody, Dawson, Fineman, Lawroski, Reinhardt, Stein, and Stewart have obtained the following results from preliminary investigation of a continuous countercurrent solvent extraction method for plutonium isolation: Completed analyses of samples from solvent extraction runs employing thrice re-used (without purification) hexone indicates purities and recoveries of approximately 99.9% and 99.5%, respectively, demonstrating that no noticeable loss in

selectivity or extractability occurs even though the solvent is slightly decomposed as evidenced by the intensification of yellow color. Continued work on the evaporation of aqueous column plutonium solutions for concentration purposes has indicated the method to be feasible from the chemical standpoint.

The following conclusions can be drawn from the development studies now substantially completed on the 19-mm experimental continuous countercurrent solvent extraction column.

1. Continuous countercurrent solvent extraction with hexone can be successfully employed for the isolation and purification of $Pu(NO_3)_4$ starting with a feed prepared by complexing with zirconyl nitrate the lanthanum fluoride-plutonium precipitate after the crossover cycle.

2. The following solutions can be satisfactorily used in the extraction process:

   a. Reflux or scrubbing solution — 3 N in HNO_3 and 5 N in NH_4NO_3.
   b. Feed solution — 3 N in HNO_3, 5 N in NH_4NO_3, 0.2 g La(III)/l and 18.6 g Zr/l.
   c. Wash solution — distilled water.
   d. Solvent — commercial grade hexone which may be, though not necessarily, partially pre-saturated with HNO_3 or HNO_3-NH_4NO_3 solution.

3. Recovery of approximately 99.5% of the plutonium with 99.9% purity.

4. Solvent may be used up to at least three times without noticeable loss in extractability, selectivity, and ease of column operation.

5. At least a 2-fold decrease in volume from the original feed solution (50 gallons) can probably be obtained in the solvent extraction process, the remaining concentration to be effected either by evaporation or oxalate precipitation.

6. Satisfactory techniques for following column operation and maintaining process control have been developed.

The German army has launched an all-out counter-offensive in Belgium.

The Project Council Policy Meeting was held at 9:30 a.m. in Room 209, Eckhart Hall, attended by Bartky, Compton, C. M. Cooper, Daniels, Dempster, Doan, Greninger, Hilberry, Huffman, Jeffries, W. C. Johnson, Karl, McKinley, Mulliken, Spedding, Stearns, Stone, Szilard, Tracy, Whitaker, Wigner, and Zinn. The schedule for future Council meetings was discussed and it was agreed to open each morning session at 8:30 and extend it until 12:30 p.m. Afternoon sessions, if necessary, will begin at 2:00 p.m. On this basis the Council Information meetings for January 16 and 17 will be January 16, a.m. — Physics, p.m. — Health and Metallurgy; January 17, a.m. — Chemistry, p.m. — Technology/Clinton, to be followed by a Policy Meeting at 3:05 p.m.

Compton said that he recently discussed the U^{233} program with Tolman and Conant and concluded that studies looking toward its production should probably be the major effort of the Project, although not with the expectation of use in this war. The development of nuclear power is longer term, not for this war; hence plans can be delayed for a few months and need not be ready by February 1, 1945. Compton stated that it is of the greatest importance that we emphasize our need for fundamental scientific information in our recommendations to the Army. He believes a general feeling exists throughout the U.S. that war research has greatly advanced our knowledge of fundamentals, whereas the actual case is quite different since most war developments have been in the application of existing knowledge. Our duty is to correct this impression. Wigner complained about the lack of liaison with Site W and Y that has developed. Greninger agreed that information from Site Y has dropped off since Cyril Smith ceased his visits to Chicago. Compton agreed to have John Wheeler attend the February Physics Information meeting to help bring us up-to-date on Site W activities.

Hilberry reported on Hanford activities, saying that the D pile started operating last Sunday evening and that the 30 g/ton material probably will be dissolved on Christmas morning at the 200 west chemical separation plant. Whitaker reported on Clinton activities, indicating that the pile area ceased operation for production of 49 last month and the chemical separation plant will close down about January 1, 1945, with the last shipment of product a few days later.

Thursday, December 21, 1944

Today is the last at the Met Lab for Lyle R. Dawson. His termination is effective December 31. He will go to the University of Kentucky as head of the Department of Chemistry.

Manning received a memo from Nickson complaining that Room 36 (Kelley and Yett) has not yet been decontaminated from alpha-particle activity although readings were taken and reported on a few weeks ago. No one occupied the room for awhile after this report but we are now beginning to use it again. The Health Division therefore requests that the areas of high reading on Pluto be decontaminated before routine work recommences in this room.

The isolation and purificiation of the 25 mg of U^{233} for Los Alamos is now complete. The material has been shipped.

Today in my notebook, no. 139, I recorded the following conception related to a "Redox Process" for separating and decontaminating plutonium:

Solvent extraction methods should be very good for separating the fission products and other isotopes from plutonium which has

been used for running a chain reaction. A number of solvents, e.g., ethers, ketone, esters, etc., should be good for this purpose; solvents in which the +4 or +6 state of plutonium is soluble are the type which should be best. The mixture of plutonium and fission products in aqueous solution, possibly in the presence of NO_3^- ions from agents such as NH_4NO_3, $Ca(NO_3)_2$, $Al(NO_3)_3$, etc., would be contacted with the solvent, either batchwise or in a column or columns, and the plutonium would go largely into the solvent while the fission products would remain largely in the aqueous phase. It might be well to use the principle of changing the oxidation state of the plutonium and contact the plutonium-fission product solutions with successive solvents (the same or different ones) in an arrangement which makes use of the difference in the solubilities in the solvents of the plutonium in different oxidation states. For example, Pu^{+4} (and/or Pu^{+6}) may be soluble in the solvent (relative to the aqueous phase) while Pu^{+3} may be insoluble in the solvent (relative to the aqueous phase) so that successive uses of the solvent or solvents while changing from Pu^{+3} to Pu^{+4} (and/or Pu^{+6}) might result in very efficient decontamination of Pu from the fission products. It should, of course, be possible to use essentially the same types of methods to decontaminate U^{233} and U^{235} and mixtures of these isotopes with each other and/or with Pu^{239} (in the presence or absence of U^{238}) from the fission products and from other isotopes formed when these (U^{233}, U^{235}, etc.) are used for running chain reactions.

Daniels sent a memo to Wigner stating that I am preparing a more complete report on the research on solvent extraction required in order to meet the new drastic requirements for recovery and decontamination in connection with the "sandwich" converter pile. Daniels quotes significant parts from my memo of December 19, MUC-GTS-1201, as an interim report.

In addition I conferred with Wigner about converter piles and explained again the difficulty of achieving complete recovery of plutonium and high decontamination. Wigner indicated he will think about the matter.

Helen as usual went to her chemistry class today.

The German counter-offensive on the Western front continues to mount in intensity, according to today's newspaper.

Friday, December 22, 1944

Magnusson and La Chapelle have been studying the chemistry of neptunium using part of the supply obtained from the special chemical plant extractions at Clinton. Today Magnusson made the first absorption spectrum measurements on neptunium(VI) in 1 M $HClO_4$.

All this week Morgan has been working simultaneously on the uranium fraction of sample 49DD (49DD-11), the 200 mg plutonium plus

deuterons, St. Louis cyclotron bombardment, and the 95 fraction of sample 49ND (49ND-14), the original sample SA-1 consisting of 8.2 mg of PuO_2 which received 10,233,244 kwh of neutron irradiation in the Clinton pile. He has carried out several $TiCl_3$-HNO_3 cycles on the 49DD-11 sample to purify it from plutonium contamination and to look for evidence of U^{232}. His final sample, 49DD-11.26, shows alpha-particle activity with only the Pu^{239} range. Morgan is not convinced that the chemistry was adequate for the isolation of the uranium and hopes the $TiCl_3$ reduction can be checked with known amounts of uranium. In the meantime, Morgan has put sample 49DD-14 through a number of dichromate oxidation cycles (current 95 fraction labeled 49ND-14.14).

At Jesse's request, I sent him a list of five documents (C reports and MUC's) containing information on transuranium elements and related topics.

Manning transmitted to Maloney a rough draft of a memo on batch solvent extraction.

Stearns asked Compton by memo for 5 grams of plutonium to be used by the Metallurgical Division in determining metallurgical properties.

I spent most of the day conferring with Foster York who is going through Cases 52 and 61, claim by claim, and looking for notebook support. With regard to Case 61 (my draft MUC-GTS-941), I interpreted for him the neutron-induced fission counting data obtained prior to April 1, 1941, and which is recorded in my black notebook, Volume 1. These data are as follows: 75 mc of 93^{239} corresponding to 0.25 microgram of 94^{239}, the latter giving a slow neutron fission counting rate of about one count per minute per microampere of deuterons on beryllium at the Berkeley 37-inch cyclotron; 50 alpha counts per minute of uranium standard corresponding to 0.5 microgram of U^{235}, the latter giving a slow neutron fission counting rate of 11 counts per minute per microampere. These data give the result that the slow neutron fission cross section of 94 is one-fifth that of U^{235}. (This work, on sample A, was very rough.) (The matter of the April 1, 1941, date came up, and I pointed out to York again that this is a fictitious date as far as this Case 61 is concerned.)

We discussed the fission counting data of about May 18, 1941, on sample B. In this measurement the weight was 0.5 microgram of 94^{239} (changed from 0.25 microgram due to recalculations, etc.) giving 80 slow neutron fission counts per minute and 1.46 micrograms of U^{235} giving 140 slow neutron fission counts per minute with 9 microamperes of deuterons on beryllium in each case; these data lead to a cross section of 94^{239} which is 1.7 times that of U^{235}. York pointed out we have no notebook entries showing how to apply these results to the development of a chain reaction; also, we have no notebook entries which support Claims 5-9, probably Claim 10, Claims 11-14, probably Claim 15, Claims 15-18, probably Claim 19, and Claims 20-43.

In regard to my draft of Case 52 (MUC-GTS-942), we went through Wahl's notebooks (Volumes 11, 12, 13, 14, and 16) rather carefully. York wanted to know who, besides the inventors, could identify some of these notebooks. I told him that probably Friedlander, English, and/or Gofman

could do so. York pointed out that the letters of January and March, 1941, to Briggs constitute perhaps our best corroborative evidence since these describe the results of experiments whose entries can be found in the notebooks. They give good support for the claims on oxidation-reduction cycles and for the claims to certain compounds of plutonium, e.g., fluoride, iodate, and peroxide. The question of the April 1 date came up again. I pointed out that this date has no meaning also in regard to this case. A date which might have some meaning is January 1, 1942, when Wahl and I both went on the Berkeley Project payroll. The best disclosure for the oxidation-reduction method of getting rid of the fission products is the July, 1941, experiment, i.e., the one in which Sample F was prepared. York pointed out that the generic claim to the oxidation-reduction cycle (which of course is the most important one of all) might have to rest on the many persulfate experiments plus one experiment with cold permanganate, since these are the only ones done before January 1, 1942. Claims like No. 24 are not specifically disclosed, says York, even in the July experiment (i.e., the precipitation of fission products from a solution containing oxidized 49 was not performed in the presence of all the fission products). The same goes for a claim like No. 28. Likewise, York could not find the notebook references to cover the electrolysis of plutonium; the carrying by sodium uranyl acetate; the carrying by thorium oxalate, rare earth oxalate, thorium fluoride, or rare earth iodate, all of which are touched upon in the claims. In this connection I raised the question of how much obvious deduction one can use in drawing up claims. I think that there comes a point when it is just a waste of time to do experiments just in order to have notebook entries.

The claims in Case 61 (as listed in our draft MUC-GTS-941) with inventors Segrè, Kennedy, and Seaborg are as follows:

1. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with neutrons.

2. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with thermal neutrons.

3. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with neutrons produced in the fission reaction.

4. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with fission neutrons which have been slowed to thermal energies.

5. The method of producing energy and radioactive fission products which consists of reacting mixtures of neutron-fissionable isotopes, including Pu^{239}, with neutrons produced in the fission of Pu^{239}.

6. The method of producing energy and radioactive fission products which consists of reacting mixtures of thermal neutron-fissionable isotopes, including Pu^{239}, with neutrons produced in the fission of Pu^{239}.

7. The method of producing energy and radioactive fission products which consists of increasing the mass of Pu^{239} in a composition of matter containing Pu^{239} from an amount below the critical up to at least the critical mass.

8. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with neutrons, the Pu^{239} being mixed with non-fissionable isotopes of such an amount so that the amount of capture by neutrons by Pu^{239} is greater than the amount of capture of neutrons in processes not leading to fission.

9. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with neutrons, the Pu^{239} being mixed with non-fissionable isotopes of such an amount so that the amount of capture by neutrons by Pu^{239} is greater than the amount of capture of neutrons in processes not leading to fission, and with the Pu^{239} being present in such an amount and concentration that a nuclear self-sustaining chain reaction takes place.

10. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with neutrons, the Pu^{239} being present in a composition which consists of Pu^{239} and material which is especially efficient for slowing neutrons.

11. The method of producing energy and radioactive fission products which consists of reacting Pu^{239} with neutrons, the Pu^{239} being present in a composition which consists of Pu^{239} and material which is especially efficient for slowing neutrons, and the Pu^{239} being present in such an amount and concentration such that a nuclear self-sustaining chain reaction takes place.

12. The method of producing energy and radioactive fission products whereby in a mass containing Pu^{239} and undergoing a nuclear self-sustaining chain reaction, a reflecting material is used to reflect back into the mass a substantial proportion of the neutrons emanating from the mass.

13. The isotope Pu^{239} as a fissionable isotope in a composition for producing a self-sustaining nuclear fission chain reaction.

14. The isotope Pu^{239} as a fissionable isotope in a composition for producing a self-sustaining nuclear fission chain reaction, in a mixture consisting of a fissionable isotope and material particularly capable of slowing neutrons.

15. A composition of matter consisting of Pu^{239} and material which is particularly efficient for slowing neutrons.

16. A composition of matter consisting of Pu^{239} and material which is particularly efficient for slowing neutrons, the Pu^{239} being present in such a concentration that the composition is capable of undergoing a self-sustaining nuclear fission chain reaction when the amount is above the amount of the critical mass.

17. A composition of matter consisting of a mixture of Pu^{239} and another isotope which is fissionable with thermal neutrons and material which is particularly efficient for slowing neutrons to thermal energies, the concentration of the thermal neutron-fissionable isotopes being such that the said fissionable isotopes will undergo a self-sustaining nuclear fission chain reaction when the amount is above that of the critical mass.

18. A composition of matter consisting of Pu^{239} and neutron-

slowing nuclei, the neutron-slowing nuclei having a capture cross section for neutrons of less than about 10^{-24} sq.cm., a scattering cross section of greater than about 10^{-24} sq.cm., and a mass of less than 30 mass units.

19. A composition of matter consisting of Pu^{239} and atoms selected from the group of light atoms composed of hydrogen, deuterium, helium, beryllium, carbon, oxygen, hydrogen, and fluorine.

20. A composition of matter consisting mostly of Pu^{239} and water.

21. A composition of matter consisting mostly of Pu^{239} and heavy water (D_2O).

22. A composition of matter consisting mostly of Pu^{239} and beryllium.

23. A composition of matter consisting mostly of Pu^{239} and carbon.

24. A composition of matter consisting mostly of Pu^{239} and water, the Pu^{239} having a concentration between about 10^{-2} gm per cc and 10 gm per cc.

25. A composition of matter consisting mostly of Pu^{239} and water, the Pu^{239} being present at a concentration of approximately 0.05 gm per cc.

26. A composition of matter consisting mostly of Pu^{239} and heavy water (D_2O), the Pu^{239} being present at a concentration between about 10^{-4} gm per cc and 1 gm per cc.

27. A composition of matter consisting mostly of Pu^{239} and heavy water (D_2O), the Pu^{239} being present at a concentration of about 0.01 gm per cc.

28. A composition of matter consisting mostly of Pu^{239} and carbon, the Pu^{239} being present at a concentration between about 10^{-4} per cc and 1 gm per cc.

29. A device for the production of energy containing a mass consisting of Pu^{239} distributed in neutron-slowing material, the amount and concentration of Pu^{239} being such that a self-sustaining nuclear chain reaction can take place.

30. A device for the production of energy containing a mass of Pu^{239} distributed in neutron-slowing material, the amount and concentration of the Pu^{239} being such that a self-sustaining nuclear chain reaction which can be controlled by neutron-absorbing control rods will take place.

31. A device for the production of energy containing a mass of Pu^{239} distributed in neutron-slowing material, the amount and concentration of Pu^{239} being such that a self-sustaining nuclear chain reaction which can be controlled by means of the temperature change which occurs in the mass will take place.

32. A device for the production of energy containing a mass of Pu^{239} and another slow neutron fissionable isotope distributed in a neutron-slowing material, the amount and concentration of the Pu^{239} and other slow neutron-fissionable material being such that a self-sustaining nuclear chain reaction will take place.

33. A device for the production of energy containing a mass of

Pu^{239} distributed in a neutron-slowing material and also containing a means for reflecting neutrons, arising in this mass, back into the said mass.

34. A composition of matter which consists of Pu^{239} and an isotope which is capable of transformation as the result of neutron absorption to an isotope of higher mass number, which in turn decays to an isotope which is slow neutron-fissionable.

35. A device for the production of energy containing a mass of Pu^{239} distributed in a neutron-slowing material, the amount and concentration of Pu^{239} being such that a self-sustaining nuclear chain reaction will take place, and this mass being surrounded by a composition containing Th^{232} for the purpose of absorbing in the Th^{232} some of the excess neutrons which escape from the mass.

36. A device for the production of energy containing a mass of Pu^{239} distributed in a neutron-slowing material, the amount and concentration of Pu^{239} being such that a self-sustaining nuclear chain reaction will take place, and this mass being surrounded by a composition containing the natural mixture of uranium isotopes for the purpose of absorbing in the U^{238} some of the excess neutrons which escape from the mass.

37. A composition of matter consisting of Pu^{239} and U^{238} and material which is particularly efficient for slowing down neutrons.

38. A composition of matter consisting of Pu^{239} and the natural mixture of uranium isotopes and material which is particularly efficient for slowing down neutrons.

39. A composition of matter consisting of Pu^{239} and Th^{232} and material which is particularly efficient for slowing down neutrons.

40. A composition of matter consisting of Pu^{239} and U^{233} and neutron-slowing material.

41. A composition of matter consisting of Pu^{239} and U^{235} and neutron-slowing material.

42. A composition of matter consisting of Pu^{239}, U^{233}, and U^{235} and neutron-slowing material.

43. A composition of matter consisting of Pu^{239} and another slow neutron-fissionable isotope and neutron-slowing material.

44. The features of novelty herein disclosed and/or described.

Similarly, the claims in Case 52 (as listed in our draft MUC-GTS-942), with inventors Seaborg, Wahl, and Kennedy, are as follows:

1. A composition of matter which consists substantially of plutonium.

2. A composition of matter which consists substantially of an isotope of plutonium.

3. A composition of matter which consists largely of a compound of Pu.

4. A composition of matter which consists largely of a compound of an isotope of plutonium.

5. A composition of matter which consists substantially of Pu and which is largely free from fission products.

6. A composition of matter which consists substantially of an isotope of Pu and which is largely free from fission products.

7. A composition of matter which consists substantially of a compound of Pu and which is largely free from fission products.

8. A composition of matter which consists substantially of a compound of an isotope of Pu and which is largely free from fission products.

9. A composition of matter consisting of a compound of Pu.

10. A composition of matter consisting of a compound of an isotope of Pu.

11. A composition of matter consisting of a substantial proportion of a compound of Pu.

12. A composition of matter consisting of a substantial proportion of a compound of an isotope of Pu.

13. A composition of matter consisting predominantly of Pu^{239} and U^{238}.

14. A composition of matter containing Pu^{239} and the natural mixture of uranium isotopes.

15. A composition of matter containing Pu^{239} and Th^{232}.

16. A composition of matter consisting substantially of Pu^{239} and U^{233}.

17. A composition of matter consisting substantially of Pu^{239} and U^{235}.

18. A composition of matter consisting substantially of Pu^{239}, U^{233}, and U^{235}.

19. A composition of matter consisting substantially of a lower fluoride of plutonium.

20. A composition of matter consisting substantially of a lower oxalate of plutonium.

21. A composition of matter consisting substantially of a lower iodate of plutonium.

22. A composition of matter consisting substantially of a higher fluoride of plutonium.

23. A composition of matter consisting substantially of a higher iodate of plutonium.

24. A composition of matter consisting substantially of a peroxide of plutonium.

25. In a method for the separation of Pu from fission products, the step of oxidizing the Pu from a lower oxidation state or states in which it is substantially carried by a substance of the rare earth fluoride type to a higher oxidation state or states where it is not significantly carried by a substance of the rare earth fluoride type.

26. In a method for the separation of Pu from fission products, the step of reducing the Pu from a higher oxidation state or states in which it is not significantly carried by a substance of the rare earth fluoride type to a lower oxidation state or states in which it is substantially carried by a substance of the rare earth fluoride type.

27. In a method for the separation of Pu from fission products, the step of reducing the Pu from a higher oxidation state or states in which it is not significantly carried by a substance of the rare earth fluoride type to a lower oxidation state or states in which it is substantially carried by a substance of the rare earth fluoride type.

28. In a method for the separation of Pu from fission products, the step of co-precipitating the Pu, from a solution containing Pu in its lower oxidation state or states and fission products, with a carrier precipitate of the rare earth fluoride type.

29. In a method for the separation of Pu from fission products, the step of co-precipitating the Pu, from a solution containing Pu in its lower oxidation state or states and fission products, with a carrier precipitate of lanthanum fluoride.

30. In a method for the separation of Pu from fission products, the step of co-precipitating the Pu, from a solution containing Pu in its lower oxidation state or states and fission products, with a carrier precipitate of cerous fluoride.

31. In a method for the separation of Pu from fission products, the step of co-precipitating some of the fission products from a solution containing Pu in its highest oxidation state or states and fission products, with a carrier of the rare earth fluoride type, substantially all the Pu remaining in solution.

32. In a method for the separation of Pu from fission products, the cycle which consists of co-precipitating the Pu, from a solution containing Pu in its lower oxidation state or states and fission products, with a carrier of the rare earth fluoride type; dissolving the rare earth and plutonium fluorides, and oxidizing the Pu to its highest oxidation state or states; co-precipitating some of the fission products with a carrier of the rare earth fluoride type, and finally reducing the Pu to its lower oxidation state or states.

33. In a method for the separation of Pu from fission products, the step of co-precipitating Pu with a carrier of the rare earth or thorium oxalate type.

34. In a method for the separation of Pu from fission products, the step of co-precipitating the Pu with a carrier of the thorium or rare earth iodate type.

35. In a method for the separation of Pu from fission products, the step of co-precipitating from a solution of Pu in its higher oxidation state or states and fission products, part of the fission products with a carrier of the thorium or rare earth iodate type.

36. In a method for the separation of Pu from fission products, the step of co-precipitating the Pu with a carrier of thorium peroxide.

37. In a method for concentrating Pu from a solution containing Pu and foreign material, the step of precipitating the Pu as its insoluble peroxide.

38. In a method for separating Pu from element 93, the step of treating a solution of the Pu and element 93 with an oxidizing agent which will oxidize the element 93 to an oxidation state such that the element 93 is not co-precipitated with a rare earth fluoride, and at the same time leaving the Pu in the lower oxidation state such that the Pu is carried by a rare earth fluoride.

39. In a method for separating Pu from element 93, the step of treating a solution of the Pu and element 93 with the oxidizing agent bromate ion (BrO_3^-), which will oxidize the element 93 to an oxidation state such that the element 93 is not co-precipitated with a rare earth fluoride, and at the same time leaving the Pu in the lower oxidation state such that the Pu is carried by a rare earth fluoride.

40. In a method for separating Pu from element 93, the steps of treating a solution of the Pu and element 93 with an oxidizing agent which will oxidize the element 93 to an oxidation state such that the element 93 is not co-precipitated with rare earth fluoride, and at the same time leaving the Pu in the lower oxidation state such that the Pu is carried by rare earth fluoride, and then co-precipitating the Pu with a rare earth fluoride.

41. In a method for the separation of Pu from uranium, the step of co-precipitating from a solution of Pu, the Pu with a rare earth fluoride.

42. In a method for the separation of Pu from uranium, the step of co-precipitating from a solution of Pu, the Pu with thorium iodate.

43. In a method for the separation of Pu from fission products, the step of oxidizing the Pu by means of the oxidizing agent dichromate ion from a lower oxidation state or states in which it is carried substantially by a rare earth fluoride to a higher oxidation state or states in which it is not significantly carried by rare earth fluoride.

44. In a method for the separation of Pu from fission products, the step of oxidizing the Pu by means of the oxidizing agent permanganate ion from a lower oxidation state or states in which it is carried substantially by a rare earth fluoride to a higher oxidation state or states in which it is not significantly carried by rare earth fluoride.

45. In a method for the separation of Pu from fission products, the step of oxidizing the Pu by the oxidizing agent peroxydisulfate and silver ion from a lower oxidation state or states in which it is carried substantially by a rare earth fluoride to a higher oxidation state or states in which it is not significantly carried by rare earth fluoride.

46. In a method for the separation of Pu from fission products, the step of reducing the Pu by the reducing agent sulfur dioxide from a higher oxidation state or states in which it is not significantly carried by a rare earth fluoride to a lower oxidation state or states where it is substantially carried by a rare earth fluoride.

47. The features of novelty herein disclosed and/or described.

The German winter offensive into Belgium and Luxembourg is still moving, but the Soviets, having encircled Budapest, are seeking the most direct invasion highway to Vienna.

Saturday, December 23, 1944

At Clinton Labs, slugs are being charged to the dissolver for the first two of four additional processing runs (294, 295, 296, 297) under special conditions to provide recovery of Np^{237}. The process modifications consist of the use of MnO_4^- and $H_2C_2O_4$-Mn(II), in the extraction step and 0.01 M U(IV) in place of Fe(II) in the Bismuth Phosphate Cycle.

War summaries come from six fronts according to today's paper. On the Western front U.S. troops are checking the Germans forty miles inside Belgium. U.S. troops have also slowed the German drive beyond Luxembourg. The Soviets have opened a big drive in Latvia. U.S. troops are driving for the Japanese escape port of Ormoc on Leyte. Nagoya, Japan's big aircraft center was bombed again. And at Chungking, China, the Japanese have rushed in more troops to hold that rail city.

Sunday, December 24, 1944

This morning's Chicago *Sun* eight-column headline reads "Allied Planes Batter Nazis; General Situation Better." Clearing skies over Europe enabled Allied planes to concentrate on German communication and supply lines that have been feeding the German drive into Belgium and Luxembourg.

Monday, December 25, 1944

Christmas Day. Helen and I spent a quiet day at home.

U.S. troops are gaining again in Luxembourg, and Washington announced the second bombing this month of Iwo Island, midway between Japan and Saipan.

The Chicago *Sun* reported this morning that Glenn Miller's plane has been missing since December 15. Allied headquarters in Paris made this announcement about the well-known orchestra leader's plane on December 24.

Tuesday, December 26, 1944

Irwin Schaffner and Wilbur Simon officially transferred into Section C-I from Tepe's group today. They will work with Lawroski. Today is the last day at the Met Lab for Robert W. Rasmussen.

The first neutron-irradiated uranium from a Hanford reactor 105B pile) was charged today to the dissolver in the 221T bismuth phosphate separation plant for the isolation of plutonium.

Magnusson, continuing his work of last Friday, made the first absorption spectrum measurements on Np(IV), in a 0.5 M H_2SO_4 solution today.

I wrote to Allison at Los Alamos describing the following samples we have sent him. (1) 100 micrograms of 93^{237}. This material may contain about 0.05% Pu^{239} by weight. It came from Clinton uranium at a level of 5 grams Pu^{239} per ton. (2) 25 milligrams of U^{233}. The actual amount is more than this because the new measurement by Dempster indicates that the isotopic composition is 96% U^{233}/4% U^{238} rather than 88% U^{233}/12% U^{238} on which the original quantity was calculated. I point out the difficulties in his suggestion that it might be feasible to measure the amount of U^{238} in our U^{233} by counting the UX beta particles. It would not be possible to do this directly, without a chemical separation, because of the x-rays and gamma-rays arising from the disintegration of the U^{233} itself; it might be barely feasible to look for UX in the separated UX_1 fraction containing Th^{229}, although the Ra^{225} daughter of Th^{229} gives rise to the whole remainder of the 4n + 1 series and makes the measurement of UX_1 difficult. (3) 10^7 disintegrations per minute of a mixture which seems to be 50-50 by weight of Pu^{238} and Pu^{239} on the basis of alpha-particle range measurements; the Pu^{239} probably was introduced into this deuteron-produced Pu^{239} by contamination in the laboratory.

I also comment on possible sources of gamma-emitters with energies above the threshold for the γ,n reaction in beryllium and deuterium, which he asked about in a December 15 letter to Compton. Finally, I ask when we might expect to receive the 25 mg of plutonium from each of Fermi's special samples that have been irradiated at Hanford and which I understand were requested in a letter from Compton to Oppenheimer.

A bulletin from the Pacific today announces the close of the Leyte Island campaign.

Wednesday, December 27, 1944

At Clinton Labs, irradiated slugs are scheduled today to be charged to the dissolver for the third of four special runs to be made to recover Np^{237} for use at Met Lab.

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, J. J. Katz, Katzin, Lawroski, Manning, and Simpson. I suggested that plutonium from various sources be kept separate from each other and that some convenient nomenclature be adopted to identify them, proposing that the "grams Pu/ton uranium" symbol, "gt", be used as a unit of the amount or irradiation and, hence, proportion of Pu^{240}. Manning and Katz talked about the telephone situation. Phones with less than 10

calls per week probably will be removed. Installation of a pay phone in New Chem is suggested since it is now impossible to make an outside call except on the Project phone. Davidson announced that Robinson and Westrum's calorimeter is almost completed and should be available for measurements in about two weeks.

Ghiorso said that the pulse analyzer circuit is being worked on and should be ready for preliminary testing today — the circuit will enable us to measure amounts of many different alpha-particle ranges present in a given sample. He said we will be getting a new gamma-ray ionization chamber in which the gamma-ray intensity can be measured by putting a test tube of solution into a hole in the chamber and reading the results on a meter — a very convenient tool for use in decontamination studies. I suggested securing a proportional counter and a gamma-ray ionization chamber for use in the West Stands.

Simpson stated that he is about ready to volatilize onto quartz discs the PuF_3 made by Florin from the plutonium which was bombarded at Site X for a long period of time. The resulting thin samples are to be used as sources in the differential range chamber for determining the presence of Pu^{240} and Pu^{238}.

I mentioned that a sample of Pu^{239} bombarded for a long time in the Site X pile has been studied for a long-lived beta-particle emitting 94^{241}. The plutonium was decontaminated, and the sample was examined for beta-particle activity above the normal plutonium emission. About twice the normal emission was found, but this is not sufficient to suggest the presence of a 94 beta emitter. An experiment to be done very soon is the irradiation of some plutonium (5 g plutonium/ton U, i.e., 5 gt material) from Site X at the Argonne P-9 pile in order to look for a possible short-lived beta-particle-emitting Pu^{241}.

Katzin mentioned the problems in separating the uranium, thorium, and radium in the 4n + 1 series. Thorium-229 unfortunately is carried on $BaSO_4$ to approximately 30%, so $BaSO_4$ is not too good to use to precipitate the radium out first. I suggested precipitating the thorium first, perhaps with zirconium iodate or ceric iodate, following which the radium could be precipitated with barium sulfate.

Magnusson and La Chapelle have found that the reduction of aqueous Np(VI) by one equivalent of stannous ion leads to the hitherto unobserved oxidation state Np(V). Today Magnusson made absorption spectrum measurements on Np(V) in 0.5 M H_2SO_4 solution.

Daniels sent a memo to Stearns concerning the importance of retaining SED men in the Chemistry Division, several of whom have already been taken in the first draft of twenty SED men. He identified the SED men we wish to retain as follows:

Extraction-Decontamination: Ader, Kelley, Ames, Post, and Hopkins (it was indicated that Post, Kelley, and Ader were drafted from the laboratory and are now in the Army with the expectation that they will be assigned back here). These men are highly trained in radiochemistry, and will be needed for any extraction-decontamination research for piles converting

plutonium to U^{233}. Recovery: Fineman, Anderson, Britain, Asprey, and Stewart. These men have a high degree of skill, are needed for the important ongoing program of recovering, purifying, and preparing special forms of plutonium. Solvent Extraction: Hausman and Schraidt. Shotgun Tests: Brownell, Lewis, and Cressman (not C-I men).

Dempster sent a two-page memo to Compton stressing the importance to the future welfare of the nation of putting as much scientific effort as possible into extending our fundamental knowledge of all aspects of nuclear phenomena.

I talked with Perlman at Site W by phone about samples that we can send for neutron irradiation in a Hanford pile. It would be possible for us to irradiate a milligram of Np^{237} in a platinum crucible (like those we use at Site X) in a special aluminum can. With regard to Pu^{239} samples, we would be limited to 5 milligrams per can, which is the maximum they can handle in unloading.

Iz asked if Cunningham and English can postpone our order on behalf of Site W to Camenson (of Berkeley) for micro equipment. I told him we already have put 2-3 days work into this (information from Manning); I told him, however, we could wait 30 days. I learned the special Fermi samples (neutron-irradiated plutonium) left Site W (for Site Y) about a week ago.

I attended the evening meeting of the Separation Process Subsection of Section C-I at 7:45 in Room 209, Eckhart Hall. Others present were Albaugh, Arnold, Blaedel, Bradt, Cunningham, Daniels, Egan, Ghiorso, Gilbreath, Hagemann, Hyde, Hyman, Jaffey, Jones, Katzin, Kraus, Larson, Manning, Morgan, S. Peterson, Sheft, Walling, Winner, Wolf, and others. Davidson discussed the breeder and converter piles currently being proposed. The structure which has been proposed for the so-called "breeder pile" is of the homogeneous type. A solution containing 3.5 kg U^{233} in 400 liters of D_2O (or possibly H_2O) would be continuously recycled through a chamber surrounded by a thorium reflector or blanket (to produce more U^{233} from Th^{232}). Davidson pointed out that this type of structure has some advantages from a chemical standpoint. While it would be necessary to decontaminate the circulating U^{233} solution either at intervals or continuously, the fact that the fission products would be formed in a solution might simplify the chemical processing problem.

Davidson mentioned that more serious consideration has been given to the possibility of using the fission of Pu^{239} as the source of neutrons to form U^{233} from Th^{232} — this is a converter pile. The homogeneous type of pile has been considered for this scheme also. Davidson described the alternative sandwich-type converter pile devised by Wigner. He detailed the chemical and other problems in the use of the sandwich pile as described in my memo to Daniels of December 19. He also stated that difficulties could arise from the production of U^{232} in the U^{233}.

Albaugh supplemented Davidson's remarks with information on the plutonium decontamination problems involved in recovering 98% of the plutonium with a decontamination factor of 10^7, covering the following: (1) The high activity levels associated with only a two-day cooling period

may have a serious effect on the stability of the solvent. (2) New and unfamiliar fission products probably would be encountered as a result of the short cooling period and the fact that the fission products would be from plutonium rather than from U^{235}. (3) Possible formation of colloidal fission products. (4) The consequence of inopportune formation of Pu(III) or the abnormal Pu(IV). (5) Losses that would occur in the concentration of the decontaminated plutonium solution.

The Germans are still advancing toward the Meuse River, but the Soviets have completed their encirclement of Budapest, according to today's newspaper.

Thursday, December 28, 1944

Milton Ader, now a member of SED, returned to the Met Lab today. He will work with Lawroski. J. J. Katz, Beard, Hopkins, Malm, and La Chapelle will be at Clinton Labs for the next two weeks, where they will recover Np^{237} from the four special Clinton plant runs.

Fried submitted to Zachariasen a sample hoped to be neptunium fluoride. It weighs about 25 micrograms and was prepared by $HF + H_2$ treatment of neptunium oxide at 500°C.

I completed a ten-page report (MUC-GTS-1218) entitled "Conversion of Pu^{239} to U^{233}" and sent it to Hogness. It gives a more detailed presentation of the chemical problems involved in the pile for converting Pu^{239} to U^{233} than covered in my earlier memo to Daniels (MUC-GTS-1201 of December 12, 1944).

I begin by revealing the complications that will be introduced by the nuclear side-reactions in relation to the problem of final chemical purification of U^{233}. I point out that if care is not taken to control small amounts of heavy isotope impurities, the final material may have the same limitations as the material (plutonium) whose deficiencies are being remedied. At the time the material is worked up the thorium will contain elements 90, 91, 92, and in some cases, 93 and 94. In addition to U^{233}, there will be produced as relatively stable long-lived isotopes, Pa^{231}, U^{232}, U^{234}, and, in the case where 94 is present, U^{235}, Pu^{239}, Pu^{240}, and Pu^{241}. Of particular concern is the presence of U^{232} that has four short-lived (hence long-range) alpha-particle decay products, which means that the problem of purifying the material from light element impurities assumes importance again as it originally did in the case of Pu^{239}. There is also the yet unanswered question of spontaneous fission in U^{232} or one of its daughters. All of these factors make it seem extremely important that the factor of possible fast neutron reaction (the source of U^{232}) in the thorium mass be minimized, including the possible necessity of processing the thorium at shorter intervals than other considerations might dictate.

In the area of chemical considerations, I point out that should a pile containing thorium enriched with uranium be adopted, the chemists would be confronted with the very interesting, although somewhat staggering,

problem of devising a means for separating elements 90, 91, 92, and 94 from each other and from fission products with a decontamination factor of 10^7 — with all these separations done rapidly and with high yields.

I then review the problems of chemical decontamination, assuming the sandwich pile proposed by Wigner, with a 20-day operating cycle. I urge that for great simplification of chemical processing, a decontamination factor of only 10^2 (rather than 10^7) be required, which would certainly justify the expenditure of a great deal of effort to develop a remote control process for sandwich fabrication. As a basis for estimating the manpower requirements for development of a solvent extraction process that would give 98% plutonium recovery and 10^7 decontamination, I examine the following factors that would require laboratory investigation: (1) Development of a method for complete dissolution of the irradiated plutonium sandwich. (2) Selection of a solvent that would be stable at the high radiation levels encountered. (3) Determination of distribution coefficients for plutonium and fission product activities. (5) Study of feed and subsequent process solutions for undesirable valence or state of aggregation of plutonium. (6) Development of a method for final concentration of plutonium solutions. (7) Determination of optimum recycle operations to obtain maximum plutonium recovery. I also review the many possibilities for and the magnitude of plutonium losses.

On the basis of a 50-50 probability of success I estimate that it would require over 100 men and at least 6 months to develop a process that would give 98% recovery with 10^7 decontamination. I bring up the simpler problem of 10^2 decontamination, suggesting that a yield of 99% rather than 98% might be attained. In conclusion I briefly cover the problem of U^{233} separation from thorium and indicate the problem is much simpler; we have had the benefit of experience in carrying out the separation on a laboratory scale in Katzin's group.

I attended a meeting of the Chemistry Division Directorate and Section Chiefs in Hogness' office, along with Hogness, Arnold, Burton, Franck, Sugarman, Daniels, and Watters (who has replaced D. S. McKinney as Chief of Section C-IV). The main topic of discussion was reports. The report on conversion of 49 to 23 is due Saturday, December 30. The schedule for the report on the peacetime program after July 1 next year, calls for the outline due by Thursday, January 4, and the report itself by Thursday, January 11. The regular monthly abstracts are to be submitted on Monday, January 1, or Tuesday, January 2. Section Chiefs are to meet with Research Associates at 10:00 a.m. next Tuesday; all of them will be asked for suggestions to incorporate in the peacetime program report.

I prepared a listing of night and Sunday telephone numbers for key people in the Chemistry Division and for contacts at Sites X, Y, W, Berkeley, Columbia, Montreal, SAM Laboratory, Colonel Warren's group, and Chicago Intelligence. There are almost 50 numbers on my list.

Daniels sent a memorandum to Stearns giving the Chemistry Division Progress Outline for December (MUC-FD-39). The work of my section is described as follows:

"The three-inch column for product isolation by solvent extraction

has been operated using all the elements to be expected in plant operation except product. This large column has operated just as satisfactorily as the small column. It is planned now to carry out tests in which plutonium is included along with all the other elements. The plutonium will be introduced up to 1/100 W concentrations, this being the limit deemed practical from the point of view of available material and health hazards. This three-inch column represents full-scale capacity and if solvent extraction were introduced for the isolation step at W the columns would be no larger. It may be concluded then that the step-up in solvent extraction to full capacity has been accomplished.

"Several additional tests have been carried out on trifluoroacetonylacetone which has been recommended as a specific solvent for Pu(IV). The practical results have not been as favorable as had been hoped for. The decontamination falls off sharply after the first cycle, and zirconium follows along with plutonium. The tests are being continued in the hope that more favorable conditions will be found. Trifluoroacetonylacetone will probably prove very useful in analyzing for plutonium in the IV state.

"The special operation at Clinton designed to obtain a quantity of Np^{237} has been completed without interfering with plutonium production, and the material has been isolated giving 2.3 mg of pure Np^{237}. This is forty times as much as was ever available heretofore. Np^{237} is an alpha-emitter with a half-life of 2.2 million years. It falls in the same decay series as U^{233}, decaying to 91^{233}, which in turn decays into U^{233} by beta emission with 27-day half-life. This new sample of neptunium will be used to learn more about the chemistry of neptunium, thereby making possible improvements in the separation of Np^{239} from plutonium in the Hanford process. Previous work on the chemistry of neptunium has been handicapped by the fact that experiments could only be done on the tracer scale, using Np^{239} which has a short half-life of two days. The new sample of long-life neptunium has already yielded the very interesting and scarcely predictable result that neptunium has a relatively stable plus five valence state. It also has permitted a determination of the absorption spectrum of neptunium in different states, starting with Np(VI). It has been established that Np(VI) is reduced by one equivalent of stannous ion to give complete conversion to Np(V). The further reduction to a lower state is accomplished with some difficulty. A comparison of the newly determined neptunium spectrum with the spectrum of plutonium and uranium gives interesting information regarding the position of these elements in the periodic table.

"Further work has been carried out on the separation of neptunium from plutonium using Np^{239} as a tracer in laboratory experiments designed to study the decontamination of plutonium with respect to neptunium in the Hanford process. An overall decontamination factor (D.F.) of 770 has been obtained through the crossover using $(NH_4)_2SiF_6$. When the $(NH_4)_2SiF_6$ was not used, a D.F. of 230 was obtained. If the D.F. of 770 can be increased by a factor of ten or a hundred-fold, it will then be possible to reduce materially the cooling time — perhaps to 20 or 30 days.

"The oxidation of UF_4 by oxygen at high temperatures to form UF_6 was reported last month. It has been found that PuF_4 does not undergo a similar oxidation with oxygen at one atmosphere at 600°C. PuF_3 is oxidized readily at 600°C by oxygen to give PuF_4 and PuO_2.

"The oxidation-reduction potential of the U(IV)/U(VI) couple has been found to be -0.55 in 1 M HCl, and -0.62 in 1 M H_2SO_4. This value is in disagreement with the value of -0.4 recorded in the literature. An accurate knowledge of the electrode potential is important in separation processes in order to determine conditions under which plutonium will be reduced without a corresponding reduction of U.

"An additional 25 mg of U^{233} has been extracted from eight cans of thorium carbonate irradiated in the pile at X. This U^{233} has been purified and shipped to Y. A continuous extraction process is being devised which should make it possible to work up the remaining 29 cans containing U^{233} within the next few weeks. It is expected that the remaining cans will yield about 130 mg of U^{233}, but after these have been worked up there is no further source of U^{233} in sight at the present time. These experiments on the production of U^{233} constitute important pioneering work for any later development of conversion piles.

"A fundamental study of the new neptunium decay chain was reported last month. As a result of these studies it has been found that some U^{232} is formed in the pile along with the U^{233} as a by-product according to one or more of the following reactions:

$$U^{233}(n,2n) \rightarrow U^{232}$$

$$Pa^{233}(n,2n) \rightarrow Pa^{232} \rightarrow U^{232}$$

$$Th^{232}(n,2n) \rightarrow Th^{231} \rightarrow Pa^{231}(n,\gamma)Pa^{232} \rightarrow U^{232}$$

U^{232} has a short half-life (approximately 50 years) giving off alpha rays and producing decay elements which fit into the natural thorium decay series. This series includes several short-lived alpha-emitters. Among these alpha-emitters is ThC' which gives off alpha-rays of exceedingly high energy. This fact introduces complications, in that the tolerance limits for some of the lighter elements may have to be made more exacting for the ultimate use. U^{233} appears to be better than Pu^{239} and purification from light elements would seem to be less exacting by a factor of ten or so. However, this advantage may be completely nullified and, in fact, the situation may be worse if appreciable amounts of U^{232} are present in the U^{233}. The possibility of high spontaneous fission rates for U^{232} and its decay products must also be taken into account. These possibilities must be considered in designing piles for converting plutonium into U^{233}. It seems likely that the formation of U^{232} can be sufficiently minimized by placing the thorium only at the outside of the pile where there are no fast neutrons and by reducing the length of exposure before the U^{233} is extracted from the thorium."

Daniels sent a memo to Captain McKinley, describing for the possible benefit of other laboratories on the Project, the method of preparing UF_6 from UF_4 and O_2 as developed by Fried and Davidson.

Today's banner headline reads "Report Nazi Retreat." U.S. troops are launching new attacks on the Western front.

Friday, December 29, 1944

Since December 19, James has carried out additional purification operations on sample 49NC-18, derived from 4.4 mg PuO_2 that received 350 Mwd of neutron irradiation in the Clinton pile. A by-product zirconium phenylarsonate precipitation was carried out from SO_2-reduced solution. The precipitate was converted to lanthanum fluoride, yielding sample 49NC-20. The supernatant from the arsonate precipitation was subjected to a dichromate oxidation cycle, ending with sample 49NC-21. Alpha-particle absorption counts on the two samples are being taken in an attempt to identify element 95 or 96.

Shirley Nyden and Adele Koskosky, two technicians, and Irma Saxton, a secretary, terminated today. Nyden intends to return to school.

German armies are pulling back in Belgium; according to Berlin reports the Germans are withdrawing "according to plan."

Saturday, December 30, 1944

I read a copy of a memo from Zachariasen to Stearns regarding the identification and crystal structure of neptunium fluoride. He has examined the 25-microgram sample submitted by Fried on Thursday and obtained a fairly good x-ray diffraction pattern. The sample contains 30-50% platinum. The remaining portion consists of NpF_3. It is hexagonal with lattice dimensions of $a_1 = 4.108 \pm 0.001A$, $a_3 = 7.273 \pm 0.004A$. Neptunium fluoride is isomorphous with LaF_3-SmF_3, UF_3, and PuF_3. Zachariasen points out that six months ago he reported the identification and crystal structure of NpO_2 and of $Na(NpO_2)Ac_3$, and is interested to note that each of the three neptunium compounds that has now been identified corresponds to a different valence state; namely, +6, +4, and +3.

Abstracts of the work of Sub-section I (MUC-GTS-1221, Issue 1), Group 9 (MUC-GTS-1221, Issue 2), and Sub-section II (MUC-GTS-1221, Issue 3), are now prepared. The abstracts show problem assignments, individuals, notebook numbers, and describe accomplishments during the month of December.

"Metallurgical Laboratory, Report for December 1944," (MUC-JCS-112) was prepared by the Laboratory Director's Office. The summary contains the following information pertaining to Section C-I.

"In the Chemistry Division, the column for product isolation by solvent extraction has been successfully operated, and this method should prove to be of great value if a 49 to 23 conversion program is undertaken. An additional 25 mg of U^{233} have been prepared and shipped.

"The main efforts of the Laboratory have been toward the preparation of (1) a report to the Project Office on the feasibility of a conversion of 49 into 23 as a war-time insurance measure; (2) the preparation of a report to the Project Office making recommendations for a research and development program for the Laboratory from June 30, 1945, to the end

of the war; and (3) the preparation of final reports, looking toward the termination of the present contract as of June 30, 1945." All information on Section C-I in the Chemistry Division portion of the report is derived from Report MUC-FD-39, which was submitted to Stearns on Thursday. Total expenditures for the month of December were $861,462. Personnel employed at the end of the month were 1,522 (581 technical), a net decrease of 44 during the month.

U.S. bombers have ripped Iwo Island for the 21st day in a row, according to this morning's front page.

Sunday, December 31, 1944

Top war news today comes from the Western front where German forces have launched a two-day attack against the U.S. third army near Bastogne. This army force, led by Lt. General George S. Patton, has reportedly advanced five miles northwest of Bastogne, where Patton has just rescued the 101st Air-Borne Division, which had been surrounded during the Battle of the Bulge.

Morton Downey, on an entertainment tour of the European theater, is in a Paris hospital with a throat infection. Downey was to sing in a Christmas show in Paris with the orchestra leader Glenn Miller, who has been missing on a flight from London since December 15.

On the home front, the Army has seized the offices of Montgomery Ward Company to administer the firm following a continuing dispute between the government and the company.

Monday, January 1, 1945

The six cans of thorium carbonate, specially purified from uranium, were removed from the Clinton pile for shipment to Katzin here at the Met Lab.

Hogness issued a summary of the manpower distribution in the Chemistry Division as of January 1, 1945. It shows the following for my section accounted for out of a total of 77 men:

		Number of Men Nov.	Dec.
Albaugh (Site W work, 30 men)	Thompson, Extraction and decontamination	9	7
	Katz, Concentration and isolation	3	3
	Gilbreath, Process development	6	8
	Egan, Semiworks	9	0*
	Lawroski, Solvent extraction	4	11
Cunningham (Site Y work, 37 men)	Simpson, High vacuum work	10	9
	Hindman, Basic chemistry	10	11
	Stewart, Recovery	8	6
	Ghiorso, Instruments and physical measurements	10	10
	Katzin, 23 work	6	7

*Egan transferred to Lawroski, Group 4

The new year starts with uniformly good news for the Allies on all war fronts. In Europe U.S. troops have retaken Rochefort in Belgium and Barga in Italy. Here in Chicago the new year is ushered in with subzero temperatures and bitter winds.

Tuesday, January 2, 1945

James completed taking data for alpha-particle absorption curves begun Friday on sample 49NC-21 derived from 4.4 mg PuO_2 that has received 350 Mwd of neutron irradiation in the Clinton pile. The curves show an alpha-particle range of 4.43 ± 0.10 cm of air corresponding to an energy of 5.81 Mev. James concludes in his notebook no. 1117B:

> There is no known activity with even approximately this energy except some which would have been observed to decay and some which would have grown very long-range alpha-emitting daughters. This activity is then new and probably an isotope of 95. Most likely 95^{241} by $94^{239}(n,\gamma)94^{240}$ [then] $94^{240}(n,\gamma)94^{241} \xrightarrow{\beta^-}$ $95^{241} \xrightarrow{\alpha}$.

In running the curves James also noted a possible shorter-range component. [Subsequently established to be due to 95^{241} with an alpha range of 4.0 cm; the longer-range activity ascribed by James to be due to 95^{241} was later shown to be 96^{242}.]

I received a letter dated December 28, 1944, from Aebersold in Oak Ridge asking for a copy of MUC-GTS-1138 for reproduction and limited distribution. This was a November 25, 1944, letter I wrote to A. H. Dahl concerning beta counting methods for determining the isotopic composition of U^{235}-U^{238} samples.

At 10:00 a.m. there was a meeting of Section Chiefs with Research Associates to solicit ideas for the peacetime program of the Met Lab after July 1 of this year.

I wrote to Kennedy reviewing in detail my day-long session last month (December 22) with York on Patent Cases 52 and 61. With regard to York's question on who besides the inventors could identify Wahl's notebooks 11, 12, 13, 14, and 16, I suggested that Art might give this some thought, mentioning Friedlander, English, and/or Gofman as possibilities. I also suggested it might be possible to establish witnesses who know about Kennedy's building of the ionization chamber and linear amplifier referred to in the notebooks, and which were built about January 1941.

I explained to Kennedy that I am writing all this to him in an attempt to point out the degree to which the lawyers are going into these cases and what is of interest to them. Also, in the event he wants to make some changes in the cases to cover the points brought out by York, it occurs to me that he might want the information in time to do this before January 15.

Last Sunday at Clinton Labs, slugs were charged to the dissolver for the last of four special runs to recover Np^{237} for use at Met Lab.

Helen officially resigned from the Information Division of the Met Lab today. Now that she has helped me finish my classified "Table of Isotopes," she no longer needs to make use of Lab facilities. She then attended her chemistry class at YMCA college.

U.S. troops have advanced six miles at the Bastogne salient in Belgium.

Cold weather continued today, with a high of 5°F in the city.

Wednesday, January 3, 1945

At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Jaffey, Jones, Katzin, Kraus, Lawroski, Manning, Schaffner, Simpson, Stewart, and Roy Thompson. I reviewed the activities of the past few weeks concerning the calculations on technical extraction problems involved in the use of converter piles. I mentioned that the problems are

more complicated than at Hanford since the necessary decontamination factor will be about 10^8 (at Site W it is 10^7) and about 2 kg of plutonium per day must be handled as compared with 1/4 kg per day per canyon at Site W. I also mentioned the high yields (98% to 99%) required because of the repetitive nature of the decontamination process and the fact that only the solvent extraction process can give such yields. A further problem, not solvable by chemical processes, I went on, is that for every 140 grams of Pu^{239} present there are 40 g of Pu^{240} formed in addition to 100 g of fission products. If Pu^{240} will not fission with slow neutrons, the result will be that in successive decontaminations the dead isotope will be carried along, taking up valuable space, and contributing nothing. It may even cause considerable trouble by the reaction $94^{240}(n,\gamma)94^{241}$. This, in effect, would be a completely unremovable poison although its effect may be somewhat mitigated since 94^{241} is probably slow neutron-fissionable.

On the matter of plans for the future I stated it is necessary for us to send in a memorandum on our views as to the direction the research of the Project should take after July 1. I read a number of the suggestions received from members of Section C-I, most of them centering around fundamental chemical and physical studies and problems associated with converter and breeder piles.

Roscoe Van Winkle of the semiworks group terminated today.

I read a copy of a memo from Zachariasen to Stearns reporting on his successful identification and determination of the crystal structure for NpF_4. A few days ago Zachariasen determined the crystal structure for NpF_3. The sample was returned to Davidson's group where Fried and Florin tried to oxidize it to the +4 state and then resubmitted it. Zachariasen has made a positive identification of NpF_4; this is monoclinic and isomorphous with green uranium tetrafluoride and the tetrafluorides of thorium, cerium, plutonium, hafnium, and zirconium.

Manning received a report from Nickson that there is activity as high as 250 mr/hr on a set of steps and the adjacent main corridor in the West Stands, probably caused by active material leaking from a vent pipe on a dissolver in the semiworks, Room 9. Nickson recommends that this area be decontaminated to less than 5 mr/hr and that the problem at the vent pipe be remedied.

Stearns sent a list to the telephone operators of the personnel (only Section Chiefs and above) able to place calls to Site W and Site X without further authorization (53 persons are mentioned).

I had a meeting with H. W. Koch, a research physicist at the University of Illinois, and Dancoff regarding preparation of samples for neutron measurements by Koch using the University of Illinois cyclotron as the neutron source. The program calls for Cunningham to prepare the following samples for the measurements: (1) a $6\frac{1}{2}" \times 6\frac{1}{2}"$ ordinary uranium, 100 mg, on an aluminum plate with a cellulose acetate cover by January 10; (2) same type of plate with depleted U^{238} by January 17; (3) same type of plate with U^{233} by February 5 (Zinn

may use the U^{233} first); (4) same type of plate with Pu^{239} (after the U^{233} plate, unless we cannot get the U^{233} extracted by January 31). Koch will be here to watch the preparation of the U^{233} or Pu^{239} plate about February 1-5. We will send a man to the University of Illinois with the U^{233} plate to show their physicists how to handle it for the neutron cross section measurements.

I attended the evening meeting of the Basic Chemistry, Recovery and Instruments Groups at 7:45 p.m. in Room 209, Eckhart Hall. Present were Abraham, Ader, Ames, Asprey, Bartell, Cunningham, Daniels, Davidson, Dixon, Fried, Ghiorso, Gilbreath, Hagemann, Hellman, Hindman, Hufford, Hyde, Hyman, Jaffey, Katzin, Lawroski, Magnusson, McLane, Morgan, S. Peterson, Phipps, Schwob, Scott, Sedlet, Simpson, Stewart, R. C. Thompson, Van Winkle, Watters, Wolf, and others. As usual I turned the meeting over to Cunningham. Magnusson reported on studies of absorption spectra, made using one mg of pure Np^{237} in various solutions; three oxidation states (IV, V, and VI) were studied. Spectrophotometric titrations show each of these states to differ from the adjoining state by a one electron change. The colors of the VI state are yellow green in H_2SO_4 and HCl and colorless to paint pink in $HClO_4$. The color of the V state is bluish green, while the IV state is colorless in the concentration used. The potentials of the IV/V and the V/VI couples have been estimated by potentiometric titration. A stannous reduction of Np(VI) to Np(V) gives the following potentiometric value for E_f: (HCl)Np(V) = Np(VI) + e, $E_f = -1.10 \pm 0.01$ v. A potential measurement in partially reduced (with SO_2) Np(V) solution gives: Np(IV) = Np(V) + e, E_f = about -0.56 v. I mentioned that the Np(IV)/(VI) potential would then fall between the IV/VI potentials of uranium and plutonium and asked if the equilibrium for disproportionation of Np(V) has been calculated. I was told no but that undoubtedly little, if any, disproportionation would occur. All attempts to prepare a III state have been unsuccessful — indications are that the III/IV potential must be greater than +0.1 v. I remarked that the III/IV potential is still possibly between the corresponding potentials for uranium and plutonium.

I asked if a brief statement on the dry chemistry of neptunium could be made, and Fried replied that the tri- and tetrafluorides of neptunium have been prepared in the same way as the corresponding plutonium compounds. I said that since UF_3 could be prepared similarly, but with more drastic reduction treatment, the III/IV couple of neptunium could be estimated to be near but slightly below that of uranium or just below about +0.4 v. Davidson suggested that the conditions used were not drastic enough to produce UF_3, to which I responded that the Np(III)/(IV) couple might be guessed at around +0.2 v.

In answer to a suggestion by Watters I agreed that polarographic measurements would be valuable and said that more Np^{237} will undoubtedly be available in the future for use for this purpose. I mentioned several nuclear physical properties of Np^{237} that should now be determined, including the presence of Np^{236} in the Np^{237} and the determination of the cross section for the reaction $Np^{237}(n,\gamma)Np^{238}$. This would enable one to calculate how much Pu^{238} is produced in the Hanford pile by beta decay of Np^{238}.

Dixon reported on his work to prepare an insoluble Pu(VI) compound;

the work was done in an attempt to prepare a pure Pu(VI) solution. He has investigated plutonates of calcium, barium, and magnesium, looking first at the calcium compound [solution 0.1 M in $Ca(NO_3)_2$, 5 M in NH_4OH with 0.46 g/l Pu(VI)] which shows a solubility of 338 mg/l after waiting 10 minutes and 536 mg/l after 42 hours. I called attention to the markedly different behavior from ammonium diuranate, the solubility of which is 1 mg/l or less. Barium plutonate [solution 0.1 M in $Ba(NO_3)_2$, 5 M in NH_4OH with 0.46 g/l Pu(VI)] shows a solubility of 112 mg/l after two hours; making this solution 1.0 M in NaOH caused the solubility to decrease with time — 25.7 mg/l after 14 hours. Dixon said the barium plutonate was examined by Zachariasen and found to have a different type of x-ray pattern from a barium monouranate sample prepared by fusion. Magnesium plutonate has a solubility of 63.6 mg/l after 150 minutes.

Abraham reported on studies to determine the equilibrium constant for the reaction: $PuO_2 + \frac{1}{2} H_2 + HCl = PuOCl + H_2O$. Measurements were carried out by placing a measured quantity of H_2O vapor in contact with PuOCl in a system of known volume and measuring the total pressure as a function of temperature with a Bourdon gauge. On the first cycle, the reaction proved reversible, giving the equation $\Delta F(kcal) = -8.5 + 9.5$ T/1000 over the temperature range 500° to 650°C. On subsequent cycles the pressures were not reproducible. The following reaction is now being studied: $4PuF_3 + O_2 = 3PuF_4 + PuO_2$. This reaction was found to be reversible by Fried who first studied it. Current results confirm Fried's observation.

I had two conferences with Lt. Roland Anderson and Col. Metcalf in Metcalf's office. The first conference was from 10:40 a.m. until 2:15 p.m. Anderson presented the plan that Kennedy, Segrè, Wahl, and I would hold title to all Case S-52 work done up to date in exchange for giving the Government an exclusive, royalty-free license for governmental applications. Lavender's office will do all the work of preparing an omnibus Case 52 with our cooperation. We would give Case S-61 to the Government.

At about noon Anderson and I telephoned Kennedy at Los Alamos and presented the plan; I indicated I favor the proposition. Kennedy objected to the indefinite breadth of the proposed Government license, and Anderson responded that similar language was used in other cases. Kennedy said he will discuss the matter with Segrè and Wahl and call us back after 3:30 our time.

At 3:30 p.m. I returned to Metcalf's office. Kennedy called at 3:40 p.m. and told Anderson and me that Segrè, Wahl, and he are against the plan, mostly because they do not understand "Governmental applications" and do not believe there would be any "non-Governmental applications" left for the inventors. Anderson offered to discuss the matter further if Kennedy does come to Chicago about Friday, January 12, as planned. Anderson did point out, however, that if his proposal is rejected, he will need an extension of our deadline from January 15 to February 20. (On December 6, 1944, Lavender asked for 30 days after receipt of Wahl's notebooks by Metcalf to prepare an agreement regarding disposition of the cases; if nothing acceptable could be worked out, the inventors would file the case themselves.) Kennedy brought up the point that for months we have been trying to get Captain Lavender to evaluate the case in terms

of its dollar value. After the phone call Anderson expressed the belief that to have York prepare an opinion might be damaging to both sides if copies come to light during hearings. He left at 4:30 p.m. with the statement that he has no alternative but to have York prepare the opinion. He also indicated that a cash settlement seems out of the question.

Nazis are again attacking U.S. troops in two Saar areas, but U.S. troops are advancing in the Bastogne area in Belgium.

It is warming up; temperatures reached 23°F at 8:00 a.m. but went down again to 16°F by 6:00 p.m.

Thursday, January 4, 1945

At Clinton Labs, Katz, Beard, Hopkins, La Chapelle, and Malm are scheduled to receive today the LaF_3 slurry from the first two of four special runs to recover Np^{237}.

In a memorandum (MUC-GTS-1232) to Hogness I outline those problems which in the opinion of my section should be made part of a general research program pertaining to atomic power. For the heavy elements program (atomic numbers 81 to 96) I propose the following:

I. *Preparation and basic study of properties of elements involved in atomic power.*

A. Heavy elements (atomic numbers 81-96).

1. Preparation.

a. Natural sources.

1) Chemical separation of elements.

2) Separation of individual isotopes.

b. Artificial means.

1) Isolation of individual isotopes from decay chains of artificially prepared isotopes, e.g., U^{233} decay products.

2) Specific nuclear reactions.

a) New isotopes from bombardment of Pu^{239} (and Pu^{240}) with deuterons and α-particles.

b) New isotopes such as Pu^{241}, Pu^{242}, Pu^{243}, 95^{241} and/or 95^{243} and others from intensely neutron-irradiated plutonium; also e.g., $Pu^{239}(n,\gamma)U^{236}$.

c) New isotopes from bombardment of Np^{237} with neutrons and alpha particles.

d) Same as (c) except U^{233} to be bombarded.

e) Same as (c) except enriched U^{235} and depleted U^{238} to be bombarded [e.g., prepare Pu^{240} from $U^{238}(\alpha,2n)Pu^{240}$].

f) Prepare pure Pu^{240} via $U^{239}(n,\gamma)U^{240} \xrightarrow{\beta}$ in pile.

g) Production of Pu^{239} free of Pu^{240} from mixture of Pu^{239}-Pu^{240}, using continuous extraction (e.g., UF_6) on blanket of natural uranium or depleted U^{238}.

I. A. 1. b. 2) h) Search for other heavy products (e.g., protactinium, thorium) in chain-reacting units.

2. Determination of nuclear properties of individual isotopes.
   a. Fission with neutrons.
      1) Determine whether slow neutrons produce fission (e.g., Pu^{240}, Pu^{241}, Th^{229}).
      2) In case of non-fission with slow neutrons, determine threshold energy for fast neutron fission.
   b. Neutron capture and fission cross sections, etc.
   c. Spontaneous fission half-life.
   d. Radioactive decay constants and other nuclear properties.
3. Basic chemical and physical-chemical properties of elements 89 to 96.
   a. Atomic structure (including study to correlate properties and determine electronic structure of the elements in the "actinide" series).
      1) Magnetic moments.
      2) Spectra [emission, absorption (in solution), Raman].
      3) Correlation of chemical properties.
   b. X-ray studies (crystallographic).
   c. General chemistry.
      1) Metallurgy and properties of metals.
      2) Oxidation states and potentials of couples.
      3) Preparation and properties of inorganic derivatives.
      4) Preparation and properties of organic derivatives.
      5) Reaction rate studies.
      6) Solubility studies.
      7) Hydrolytic behavior.
      8) Complex ion studies.
      9) Transference behavior.
      10) Coprecipitation behavior.
   d. Volatility and dry chemistry.
   e. Thermodynamic properties including calorimetry and colligative properties.
   f. Analytical techniques (cf. Section V on instrumentation).

B. Fission Product elements.
1. Preparation
2. Nuclear properties of individual isotopes (especially those properties of importance in pile design and operation; e.g., neutron capture cross section).
3. Chemical properties of less familiar fission elements — zirconium, columbium, molybdenum, ruthenium, rhodium). [This information will be of value in developing improved decontamination procedures, particularly in connection with conversion and breeder piles in which high concentrations of fission elements will be reached. The development of rapid methods for preparation of individual short-lived species will be facilitated.]
4. Analytical methods.
5. Development of use as tracers in general chemical research and industry.

I. C. Other elements of importance, e.g., in structural materials (including use of nuclear reactions and separation procedures for preparing pure isotopes desirable because of special nuclear properties).

II. *Separation processes work.*

A. Basic precipitation processes data.
1. Investigation of carriers for plutonium and other elements of atomic numbers exceeding ca. 82.
2. Chemistry of plutonium at pH values 0.5.
3. Search for specific complexing agents for plutonium (through entire range of pH values).
4. Colloidal FPE in Bismuth Phosphate Process and under altered conditions.

B. Alternate precipitation processes and major revisions of Bismuth Phosphate Process (success here would be based upon results of A, 1, 2, 3, and 4).

C. Methods for recovery and separation of FPE from process wastes.

D. Continuation of uranium work.

E. Methods for decontamination of 2-year-cooled W metal, at a 500 g Pu/ton level, etc.

F. Continuation of work on substituting solvent extraction for all or most of present W process.

G. Stabilities of solvents and other chemicals at very high radiation levels.

H. Decontamination of highly active plutonium solutions (from conversion or breeder piles) by solvent extraction.

I. Basic solvent extraction data.
1. Basic data for engineering design of columns for aqueous-organic extraction.
2. Formulae of extractable compounds of elements above atomic number 85 and general enlargement of the chemistry of metal compounds in organic media.
3. Separation of elements or isotopes on the basis of differential rates of diffusion through liquid-liquid interfaces (non-equilibrium extraction).
4. Development of simplified techniques for measurement of radiation activities through cylinder walls.
5. Determination of rates of exchange between active tracers and the inactive species.

J. Investigation of the use of adsorption columns for separation of heavy isotopes from each other and from fission products.

K. Separation studies involved in the U^{233} problem (see Section III).

III. *U^{233} problem.* (Most of the material listed here is covered by various categories already included in the outline. The material is gathered together to emphasize the magnitude of work related to this problem.)

A. Separation studies.

III. A. 1. Continued study of solvent separation from thorium, protactinium, and uranium from fission products.
2. Investigation of possible complexing agents to help in separations.
3. Investigation of solvents for sulfate and possibly chloride, as well as nitrate, looking towards use in breeder.
4. Continue work on theory of salting-out action in solvent extraction of inorganic salts, for use in predicting behavior, agents, etc.
5. Concentration procedures in uranium extraction for
   a. Straight solvent.
   b. Complex in solvent.
6. Precipitation procedures to separate
   a. Uranium from protactinium and thorium.
   b. Uranium from protactinium alone.
   c. Protactinium from thorium and/or uranium.
7. Development of selective carriers for uranium that do not carry thorium or protactinium.
8. Work on ThF_4-UF_6 methods for continuous removal of product.
9. Decontamination of highly active U^{233} solutions (from breeder piles).
10. Study types of fission products removable as volatile fluorides in terms of poisons and competitors for neutrons.

B. Complete the characterization of the U^{233} decay chain (4n + 1 series), including radioactive constants of daughter isotopes.

C. Nuclear properties.
1. U^{233}.
2. U^{233} decay products.
3. By-products which may be formed during production of U^{233}, e.g., U^{232} and its decay products.

D. Basic chemistry of thorium, protactinium, and uranium in relation to
1. Decontamination.
2. Purification
   a. For satisfactory use in pile.
   b. For final purification of U^{233} for use as weapons.

IV. *Chemical problems associated with various types of piles.*

A. Physics and chemistry of slurries.

B. Work on chemistry of uranium and fission products in liquid bismuth system.

C. Study of interference by polonium in bismuth system.

D. Preparation of deuterated organic solvents, and study of the effect of free-radical formation under irradiation on H-D exchange where the molecule is incompletely deuterated.

E. Problems associated with piles which would permit rapid continuous (or semi-continuous) removal of product or intermediates to avoid undesirable secondary reactions (e.g., rapid removal of Pu^{239} or Np^{239} to avoid formation of Pu^{240}).

F. Corrosion problems.

IV. G. Chemical effects of radiation.
1. Problems related to pile operation and separation processes.
2. General applications to chemical research and industry.

V. *Development and improvement of electronic instruments.*

A. Absolute beta- and gamma-ray counters.

B. Fission counting methods for assay of fissionable isotopes.

C. Automatic recording devices for recording counts during counting.

D. High geometry alpha-ray spectrograph.

E. Theoretical aspects of alpha pulse chambers and amplifiers.

F. Research on gamma-ray and neutron absorbers.

G. Fundamental investigation of precision counting methods (including study of back-scattering of alpha and beta particles).

H. New instruments (e.g., pulse selector circuits).

VI. *General research related to atomic power.*

A. Search for neutrino effects: recoil effects, reversal of beta emission.

B. Further development of special apparatus and methods, e.g., remote control devices, more powerful cyclotrons, etc.

C. Power pile development, e.g.,
1. Direct production of electrical power using thermocouples in pile.
2. Cooling heavy-water moderated pile by using vapor at critical temperature to operate turbine.

D. Further development of natural sources of heavy elements.
1. Intensive geological prospecting program for natural sources of elements important to atomic power, including transuranic elements.
2. Improved methods for extraction from low-grade ores.

J. C. Warner, Hogness, Daniels, and I had an evening conference about editors for the Proceedings of the Metallurgical Laboratory volumes. The suggestions for editors are:

Volume 19, Production and Separation of U^{233}. Seaborg, Editor; Katzin and Stoughton, Associate Editors. Editorial Board: Seaborg, Katzin, Stoughton, a Berkeley representative, and perhaps B. Goldschmidt as consultant.

Volume 18, Separation Processes for Product-Alternate Processes. H. S. Brown, Editor; Manning, Associate Editor. Editorial Board: Boyd (adsorption), Lawroski (solvent extraction), English (wet fluoride), H. S. Brown (dry fluoride), Berkeley representative (thorium oxalate and sodium uranyl acetate), Manning (miscellaneous), Willard (to be added later).

Volume 17, Bismuth Phosphate Process. Perlman, Editor; Thompson, English, and Albaugh, Associate Editors. Editorial Board: Perlman, Thompson, Albaugh, English, Warren Johnson, W. Q. Smith, Willard (to

be added later).

My suggestions for the Editorial Board on the volume on Chemistry and Metallurgy of Transuranium Elements (Volume 16) to be edited by me are Manning, Associate Editor; Editorial Board: Cunningham, Connick, Kennedy, Wahl, Perlman, and H. S. Brown.

Helen again went to YMCA college today for her chemistry class.

The House of Representatives voted to continue the Martin Dies House Un-American Activities Committee, a stunning blow to the administration.

Friday, January 5, 1945

After extensive absences Eleanor Lewis, a technician for Ghiorso, terminated today.

Morgan is finding evidence for long-range alpha particles from sample 49ND-14.50, derived from the "95 fraction" of sample 49D, 8.2 mg of PuO_2 which received 10,233,244 kwh of neutron irradiation in the Clinton pile (irradiation sample SA-1). Since December 22 he has carried out the following sequence of chemical manipulations in arriving at sample 49ND-14.50:

a. Two dichromate oxidation cycles (retaining the rare earth fluoride precipitate in each case).
b. Three zirconium iodate by-product precipitation cycles.
c. A bismuth phosphate precipitation followed by dissolution and lanthanum fluoride precipitation to give sample 49ND-14.50.

He is continuing to work up the supernatant (sample 49ND-14.47) from bismuth phosphate precipitation in order to increase the amount of element 95 available for measurement.

I wrote A. F. Voigt to thank him for his new value for the half-life of Tl^{206} I received today. I explained that it is too late to include it in the new secret table which should be issued next week.

I prepared and sent to Perlman at Hanford a summary of the hypochlorite method for separating 94 from 93. I pointed out that the method has been found effective only at tracer concentrations of 93 although 94 concentrations may be as high as 0.02 M. I indicated that for separating 93 in greater than tracer concentrations, the bromate oxidation-lanthanum fluoride cycle has been found more effective. I outlined the steps in the separation.

Headline war news today is from the Pacific, where it was announced that American planes sank or damaged 35 ships off Luzon, main island of the Philippines, on January 2 and 3.

The high today was 14°F at 4:00 a.m.; midday temperature was 7°F.

Saturday, January 6, 1945

James completed further chemical processing of sample 49NC-21 derived from the 4.4 mg PuO_2 that received 350 Mwd of neutron irradiation in the Clinton pile. Last Tuesday he found that this fraction contains alpha particles with a range of 4.43 cm of air and also a possible shorter range alpha particle. Since then James has shown that 23% of the alpha-particle activity and 70% of the beta-particle activity of sample 49NC-21 is carried by zirconium iodate from a solution oxidized with silver and persulfate. This indicates some evidence for a III oxidation state (poor carrying by zirconium iodate which carries the IV oxidation state).

Aebersold in the Oak Ridge Engineers' Office wrote asking that the appropriate person in my section furnish him with information on the proper treatment of tank BF_3 for best results in a BF_3 counter; he also wants several liters of specially purified BF_3 for another laboratory.

I summarized for Compton by memo the present situation about our supply of 93^{237}. I point out that we have recovered about 2 mg from two special Clinton chemical plant runs using a modified procedure during November (an overall yield of 20%) of which a little over a milligram is in fairly available condition and in use in basic chemical and nuclear studies. I mention that four more such special plant runs are underway at Clinton, that a group of Chicago chemists (Katz, Beard, Hopkins, La Chapelle, and Malm) is at Clinton to handle the isolation end of those runs, and that we hope to recover at least 4 mg. While we have plans that could use more than 5 mg of 93^{237} for our chemical and nuclear program, we would certainly release as much for other work as is consistent with the relative importance of the program. In case it should be necessary for us to release some of our supply, I urged strongly that the question of a special run or two at the Hanford plant be investigated. Even assuming the yield will decrease by another factor of five (a somewhat pessimistic assumption) because of the longer process at Hanford, I estimate we would recover about 10 mg of 93^{237} per one ton chemical plant run on 100 g/t material.

I read a copy of a memo from Zachariasen to Stearns giving the identification and crystal structure of NpOS. Fried submitted a sample prepared by treating NpO_2 with H_2S to Zachariasen who made a positive identification: NpOS is tetragonal, with two molecules per cell, lattice dimensions are $a_1 = 3.817$, $a_3 = 6.641$, density = 9.71. The NpOS is isomorphous with ThOS and UOS. An analogous plutonium compound has not yet been observed.

In another memo to Stearns, Zachariasen states that on the basis of crystal chemistry considerations, it is now possible to foretell a great deal about neptunium compounds of the lower valence states. As an illustration, he listed figures for the following compounds that he considers could undoubtedly be prepared: $NpF_4 \cdot 2.5H_2O$, $NpCl_3$, $NpBr_3$, NpI_3, NpOF, NpOCl, NpOBr, NpOI, NpO, NpC, NpN, and $NpSi_2$. Zachariasen also mentions that in terms of crystal chemistry there is a great deal of missing information about the +6 state of the heavy

elements. He suggests it is highly likely that organized studies of the +6 compounds will give valuable results supplementing the type of information obtained from solution studies.

Stearns, Compton, and I had a telephone conversation about the schedule for Pu^{239} transfers in and out of Section C-I. The revised schedule agreed to is that we will return to the Army Office here about 2.8 grams of Clinton material on or about January 20; we will receive no more plutonium from Clinton but will receive about 2 grams at an early date from Hanford stock manufacture at a high concentration level (such as 30 g per ton). It is not clear whether or not we shall be required later to replace the Hanford material with 2 grams from our stock.

Budd Gore, who is now Chief Administrative Officer of the Met Lab, issued an organization chart of the Scientific Divisions of the Met Lab as of January 6, 1945. The chart is as follows:

Chemistry

Director	T. R. Hogness
Associate Directors	J. Franck
	Farrington Daniels
Assistant Director	L. B. Arnold, Jr.

Sections

C-I: Separation Studies and Basic Chemistry of the Heavy Elements

Section Chief	G. T. Seaborg
Assistant to Section Chief	T. O. Jones
Associate Section Chief	W. M. Manning
Assistant Section Chiefs	B. B. Cunningham
	L. I. Katzin
	F. W. Albaugh

C-II: Radiation Studies

Section Chief	M. Burton
Associate Section Chief	A. O. Allen

C-III: Chemistry of Fission Products

Section Chief	N. Sugarman
Associate Section Chief	A. Turkevich

C-IV: Analytical Services

Section Chief	J. I. Watters
Associate Section Chief	M. S. Fred
Assistant Section Chief	C. R. Schwob

Health

Director	R. S. Stone
Associate Director	L. O. Jacobson

Sections

H-I: Clinical Medicine and Medical Research

Section Chief	L. O. Jacobson

H-II: Biological Research

Section Chief	K. S. Cole
Associate Section Chief	C. L. Prosser

H-III: Medical Industrial Hazards and Health Physics

Section Chief	J. J. Nickson
Associate Section Chief	J. E. Rose

Advisory Committee

W. Bloom
A. Brunschwig
W. R. Harrison
P. Hodges
C. J. Watson
S. Wright

Physics

Director	A. J. Dempster
Associate Director	E. P. Wigner

Sections

P-I: Instrument
Section Chief — W. P. Jesse

P-IV: Crystal Structure
Section Chief — A. H. Zachariasen

P-V: Mass Spectroscopy
Section Chief — A. J. Dempster

P-VII: Theoretical
Section Chief — G. Young

P-VIII: Properties of Solids
Section Chief — Frederick Seitz

P-IX: Engineering Physics
Section Chief — L. A. Ohlinger

Technology

Director — C. M. Cooper

Sections

T-VI: Optics

Section Chief	G. S. Monk
Associate Section Chief	W. H. McCorkle

Metallurgy

Director — A. B. Greninger

Sections

M-I: Metallurgy
Section Chief — F. Foote

M-II: Corrosion
Section Chief — E. W. Brugmann

M-III: Fabrication
Section Chief — J. H. Chapin

American troops landed unopposed on Marinduque, a small island near Luzon, giving American control of the Sibuyan Sea, off the southwest coast of Luzon.

Sunday, January 7, 1945

Chicago's temperature has not been above the freezing point so far this year, although today's high of 28°F is the highest temperature we've had.

Monday, January 8, 1945

Today is the last day at the Met Lab for Howard E. Flotow; his termination is effective on January 13. Flotow will join the Fercleve Corporation in Oak Ridge.

Gilbreath is on his way to Clinton Labs to replace Katz as leader of the neptunium recovery group.

Morgan and James prepared a plutonium sample for Clinton irradiation — 111 milligrams in the form of oxide using material obtained from Asprey which has been purified with two wet fluoride cycles, each of which included a barium sulfate by-product precipitate. The sample is labeled 606 and was dispatched today.

I proposed to Art Jaffey that with the stock of Np^{237} now available it should be possible to measure the cross section of the reaction $Np^{237}(n,\gamma)Np^{238}$. He will undertake this experiment using the P-9 pile at Argonne as the source of neutrons. Magnusson has prepared a 100-microgram sample of Np^{237} in the form of an oxide and has sealed it in a small quartz tube. It will be mounted in a graphite tube for insertion in the P-9 pile together with some gold monitors to be used for the determination of the neutron flux.

I informed Daniels of the revised schedule for Pu^{239} transfers agreed upon in my telephone conversation Saturday with Compton and Stearns.

U.S. troops seized control of a main German supply line in Belgium.

The high today was 26°F from noon until 3:00 p.m.

Tuesday, January 9, 1945

Jaffey's Np^{237} sample prepared yesterday was placed in the center thimble of the P-9 pile at Argonne today and bombarded for 10.2 hours with the pile running at 281 kw.

Katzin, T. O. Jones, and I talked by phone with English, Katz, and Stoughton at Site X about the status of the four special Clinton plant runs for the recovery of Np^{237}. Katz told us that the yield of Np^{237} in the first lanthanum slurry from Room D was only 10% (compared with 20% for the previous series of runs) and that the yields of the second slurry may be low. He said the batches of Np^{237} will be shipped to us at the Met Lab as they become available. The extraction losses of plutonium have been from 12% to about 20% in the last three runs; English said the three runs where losses were encountered had $KMnO_4$ pretreatment — two at room temperature and one at 50°C (one of the room-temperature pretreatments should have been at 75°C). All reductions were at 75°C.

I also told Stoughton about the plan for him to be an associate editor of the Proceedings of the Metallurgical Laboratory volume on "Production and Separation of U^{233}" and suggested that he bring information on the samples (of thorium carbonate being irradiated in the Clinton reactor) next week. Stoughton continued the conversation with James and Katzin.

Kennedy sent me a teletype saying he expects to be in Chicago from noon next Monday until Tuesday afternoon.

Report CS-2540, "Chemistry Division Summary Report for December 1944," was issued. The first 15 pages are devoted to activities of Section C-I.

Helen went to her chemistry class today.

It is colder again today with a high of 6°F at 2:00 p.m.

Wednesday, January 10, 1945

Marjorie Bohlman, Cunningham's secretary, has left Section C-I and has been replaced by Ruth Rogers, formerly with Kidd and Maloney.

At 8:30 a.m. (a new meeting time) I held a meeting of the Council of Section C-I in my office. It was attended by Albaugh, Cunningham, Davidson, Egan, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I discussed the schedule for plutonium transfer (my phone call Saturday with Stearns and Compton). Jones has been made coordinator of all special products, special materials, and rare chemicals. I noticed that, due to his Army status, Stewart has not been officially recognized as a group leader by the Project and his signature is not acknowledged on passes and other official requests. To avoid any trouble, the practice will be to have his initials on a given pass or request; another group leader will sign it to make it official.

I mentioned that the Np^{237} extraction runs underway at Clinton are not proceeding as well as we had expected. The yield is probably going to be only about 10%, caused perhaps by other changes in the process which were made at the same time. I mentioned my request to

Compton that Np^{237} extraction runs be made at Site W.

I next commented on the improved secretarial situation, a result of our new secretarial assignments: Jane Horwich — Manning's work and report abstracting and indexing; Ruth Rogers (replacing Marjorie Bohlman) — Cunningham and all members of his subsection; Mary Williams — Katzin and Jones and also general secretary; Dorothy Black — Albaugh and all members of his subsection; Mildred Bolden — West Stands people. All large typing jobs are to clear through Edrey Albaugh; Edrey and Kay Florin will do less typing because of the large amount of other work they have to do.

I announced there will be a meeting of the editorial advisory board on the two Thomas volumes, "The Chemistry, Purification, and Metallurgy of Plutonium" at 10:30 a.m. next Monday to consider the extensive changes suggested in the volumes. I gave the names on the tentative editorial boards which have been suggested for the Proceedings of the Metallurgical Laboratory, Volumes 16, 17, 18, and 19 (as worked out at my January 4 meeting with Warner, Hogness, and Daniels).

Jaffey received the neutron-bombarded Np^{237} plus gold monitors from Argonne and made measurements on the gold in order to determine the neutron flux. He found a value for nv of 7.46×10^{11}.

I met with Captain Lavender and Metcalf in Metcalf's office. We covered the following items:

1. Vannevar Bush has sent a letter to the three inventors (Kennedy, Wahl, and me) describing the advantages of a single application covering all the work (before and after the contract period) as against two applications (one by us, one by Government).

2. If we do not accept Lavender's proposition, i.e., if we decide to go ahead with the two cases (ours and the Government's), a letter will issue from Lavender's office about January 15, suggesting we file our case. We can get Anderson to come here next Tuesday, by phoning him on Monday, to discuss this situation when Kennedy is in Chicago. Lavender's office will evaluate the value of our case after seeing what claims we get allowed by the Patent Office.

3. Lavender thinks it imperative that we get an attorney, but through the Security Office, not his office.

4. We are also the inventors on the Government's case and representatives of the two cases should get together in discussions to prevent interference proceedings in the Department of Justice; attorneys can collaborate, but otherwise we cannot use Lavender's attorney.

5. Our offer to sell our case to the Government for $1 million cash is turned down, and no substitute offer is made. We should file our patent case, and an evaluation will be made after the claims are determined by the Patent Office.

6. For foreign patents we would each file if there are two cases, or together if we accept Lavender's proposition.

7. The contract date is important — the wording of the April 1, 1941, contract is very general.

8. Lavender will be at Site Y from the morning of January 22 to the morning of January 23 at which time he could confer with Kennedy and Wahl on this matter.

Allison sent me a teletype from Site Y saying they have packed the irradiated sample from Hanford [plutonium from the Fermi special sample] in an evacuated glass bulb placed inside a brass capsule. The contents are estimated at 25 mg. The package is leaving from Site Y on the Santa Fe California Limited day after tomorrow.

I wrote to Aebersold in the U.S. Engineer's Office in Oak Ridge, enclosing two copies of my memo of November 24 to Dahl about our beta-counting method of determining the isotopic compositions of enriched uranium (MUC-GTS-1138), as requested in his letter of December 28. In answer to his January 4 letter I indicated that Kohman, who knows most about the use of purified BF_3 in neutron counters, has been transferred to Site W. I gave him the address at which Kohman can be reached. I also referred Aebersold to James H. Lum at Dayton, who is now familiar with Kohman's technique.

Along with the other Section Chiefs, I sent a memorandum to Hogness suggesting a program for future research and development in the field of atomic power. This memorandum (MUC-GTS-1267) expands upon and treats in more detail (30 pages) the problems I outlined in my earlier memo (MUC-GTS-1232) to him last Thursday. In my proposed program to study the nuclear and chemical properties of the heaviest elements, I include reference to elements 95 and 96 in view of our increasing evidence for the discovery of isotopes of these elements.

In a memo to G. R. Gibson in Personnel I recommend a merit salary increase for Edrey Albaugh of about $1.50 a week, accompanied by a promotion to the status of Secretary A, if possible.

Katzin summarized, by memo, information on U^{233} for my use at the Project Council Information Meeting on Chemistry next Tuesday. He reports that 25 mg of U^{233} have been extracted from eight cans of irradiated thorium carbonate, purified and sent to Site Y. A continuous batch extractor, semi-automatic in operation, was used. There was some difficulty with the appearance in the ether of an organic material which held uranium in the ether phase.

The U^{233} on hand emits about one alpha particle in two thousand that is from U^{232}, corresponding to about one part of U^{232} in 10^7 of U^{233}. The yield seems high in comparison with the expected n,2n yield in uranium and should be investigated because of the possible bearing on the purity requirements of U^{233}. The previously reported data on the new specific activity of U^{233} based on the new determination of the isotopic ratio are restated. Studies on extraction of thorium and uranium by a series of esters are summarized, showing the required large extraction of uranium and low extraction of thorium. A detailed study of the protactinium extraction by diisopropyl ketone shows a rather different mechanism for extraction of the nitrate than in the case of uranium.

Preliminary tests have been made on the use of sodium diethyldithio-

carbonate in amyl acetate as a reagent for uranium extraction as recommended by the Evergreen (Canadian) group. Katzin mentions that a program has been initiated on the study of slurries of thorium oxide in anticipation of their possible use in U^{233} production units.

American forces have landed on Luzon, with relatively light losses.

Colder again! Today's high was 14°F from 2:00 p.m. to 5:00 p.m.

Thursday, January 11, 1945

Katz returned to Chicago from Clinton Laboratories where he has been heading the Np^{237} recovery group. He has been replaced at Clinton by Gilbreath.

I received a letter dated January 8 addressed to Kennedy, Wahl, and me from Vannevar Bush regarding our Patent Case 52, with the following content:

> I have discussed with Captain Lavender the purchase by the Government of rights under certain chemical discoveries that have been the subject of negotiations and I have preliminarily considered the details of the discoveries in their relation to the work undertaken by the University of California under contracts, and also as to the parts of the discoveries that were made before the contracts were in existence as well as during the periods of the contracts.
>
> It appears to me that the extent of the rights to which you are entitled and the rights to which the Government is entitled are so interwoven that it is very difficult to separate them. As you know a heavy responsibility is placed upon Government officials in authorizing the expenditure of Government funds of the amount that you have mentioned and I feel that the extent of the rights to discoveries made before the contracts and during the contracts periods should really be determined by formal Patent Office proceedings.
>
> You, of course, will appreciate that the filing of a single application for patent to cover all of the involved subject matter, arising both before and during the contract periods, would secure a much firmer patent position than would result from filing applications piecemeal by you and the Government.
>
> For this reason I have approved in general the proposal made by Captain Lavender to you that a single application be prepared covering all the work done on this particular subject with the understanding, to be confirmed in writing, that the title to the application covering the entire subject will be in you, subject to a non-exclusive license in favor of the Government for all Governmental purposes.
>
> It is not my desire to influence your decision in this

matter as I feel that you should have a free hand in disposing of any rights to your discoveries made before any Government contracts. However, I do wish to express my opinion that both you and the Government would benefit through mutual cooperation in the filing and prosecution of a single application.

Helen, as usual, attended her chemistry class at YMCA college.

U.S. troops are gaining in their drive toward Manila. In Europe the Soviets have won nearly all of Budapest.

Temperatures reached 29°F by 4:00 p.m. this afternoon.

Friday, January 12, 1945

The "Fermi special" sample (25 mg plutonium irradiated in the Hanford pile) is scheduled to leave Site Y for Chicago on the California Limited, leaving Lamy at 9:45 p.m. It should arrive in Chicago Sunday morning at 7:30.

U.S. troops have driven for Clark Field in Luzon, and the Allies have regained sixteen towns in Belgium.

Temperatures climbed above freezing, to almost 40°F, for the first time this year.

Saturday, January 13, 1945

This is the last day at the Met Lab for Hymen Zvolner. His termination will be effective January 24.

Jaffey has been making decay measurements since November 1943, on four samples of cyclotron-produced 94^{238} (from deuterons on uranium) using non-decaying 94^{239} as a standard. Today he made a calculation of the half-life of 94^{238} using principally the earliest data taken in November and December 1943 (average date, December 3, 1943) and the most recent data taken last month and this month (average date, January 1, 1945). He obtained a half-life for 94^{238} of 66 years with an estimated error of ±15%.

I sent Hogness the information prepared with Davidson's help on the value of further heat capacity measurements in answer to a request of Captain Chapman for justification of such work. I pointed out that heat capacity measurements (and therefore entropy and heat content determinations) for compounds of uranium and other interesting elements provide us with data for making accurate calculations of the possible equilibria in the chemical reactions of importance for the technology of these elements. The data also serve as a basis for the estimation of the thermodynamic quantities for analogous compounds of these elements which are available only in small amounts.

I suggested the following as a partial program of heat capacity measurements: (a) high temperature heat capacities of UCl_3, UCl_4, and UF_4; (b) heat capacities from 15°K (or lower) to high temperatures for UCl_5, UCl_6, UO_2F_2, UF_3, UBr_3, UBr_4, UI_3, and UI_4; (c) heat capacities of refractory substances; (d) heat capacities from 15°K to high temperatures of the rare earth halides, $CeCl_3$, CeF_3, $NdCl_3$, $NdBr_3$, NdI_3, as stand-in values for plutonium compounds.

Manning and I sent Warner the changes and corrections suggested by members of Section C-I to monograph MUC-JCW-223 on the plutonium purification program (the Thomas volume).

Albaugh wrote a summary for me of the work on the separation processes for my use at the January 16 Project Council Information Meeting on Chemistry. The topics covered are: (1) *Extraction-Decontamination.* The high uranium (28% to 30% UNH) extraction step appears to work best at the 0.3 to 0.4 N acidity. Further modifications have been made in the bismuth strike. Hanford preextraction storage conditions have been simulated by adding various amounts of hydrazine to air-sparged metal solutions, some of which were stored at 75°C and some of which were stored at room temperature. Extractions performed on the solutions show low plutonium losses despite the presence of a large amount of Pu(III) in some cases. Other prereduction experiments show that if H_2SO_4 is added to the dissolver before removal of the uranium heel and the 40% UNH solution is stored at an elevated temperature, no hydrazine interference will be encountered and there will be no need for a prereduction treatment. (2) *Concentration-Isolation. Recovery of neptunium from plant runs 294-7.* In these runs, as in the previous runs 266-7, the Clinton plant procedure has been modified by use of MnO_4^- and $H_2C_2O_4$-Mn(II) in the extraction step and 0.01 M U(IV) in place of Fe(II) in the bismuth phosphate cycle. The slurry from runs 294-5 was delivered to Katz and co-workers on January 4. From the amount of Np^{239} present after the first peroxide precipitation, it appears that 10% of the Np^{237} present in the metal has come through the plant. No information is yet available on the slurry from runs 296-7. (3) *Process Development.* Results are reported on (a) hydroquinone stability, (b) choice of salting-out agent, (c) extraction of plutonium by hexone from aqueous solutions containing anions other than nitrate, (d) use of dichromate to increase the hexone extractibility of plutonium, (e) oxidation of various solvents by dichromate, (f) decontamination with respect to neptunium — the most promising way to increase this factor appears to be reprecipitation of bismuth phosphate-plutonium extraction precipitates in the presence of Fe(III). (4) *Solvent Extraction Methods.* Excellent results have been obtained with the three-inch diameter plant-size column in the first continuous hexone solvent extraction runs made with plutonium for the development of an alternative isolation procedure. During the first run a total of 50 gallons of feed solution containing 212 mg of plutonium have been processed. It is found that the maximum throughput is 4 gallons of feed per hour as compared with the original design figure of 6 gallons per hour. Observations on the hexone solvent extraction method for decontamination and isolation of plutonium, starting with dissolved bismuth phosphate extraction precipitates, show good yield of plutonium and good decontamination from fission products in one decontamination cycle, but the decontamination falls off in a second cycle.

I read Zachariasen's memo to Stearns reporting that a sample of UO_3 submitted by Davidson and Fried is crystalline with a hexagonal pattern. All previous samples submitted to Zachariasen have been found to be amorphous; in order to prepare the crystalline form Davidson and Fried heated the sample to 500°C for 8 hours in oxygen at 20 atmospheres.

U.S. troops are moving in the Pacific, pushing twelve miles inland on Luxon, seizing the only railroad from Ligayen Gulf to Manila. Germany has fallen back and abandoned nearly one hundred square miles of Belgium and Luxembourg to the Allies.

Sunday, January 14, 1945

The high today was 30°F at 3:00 a.m. but fell to 26°F by 7:00 p.m.; it is still quite cold.

Monday, January 15, 1945

Compton wrote to Stearns to thank him for sending the report on the feasibility of the 23 converter pile. The matter has been placed in the hands of the Army with the statement that further active steps along this line will await a request from them. Compton also mentioned that in discussing the problem with Colonel Nichols, the impression has been gained that it is unlikely that we shall receive a request to proceed further with the 23 converter pile. Nichols has expressed himself, however, as actively interested in a research and development program that would lead to the more efficient use of uranium and thorium in producing a useful product.

At 10:30 a.m. Kennedy and I attended an Editorial Advisory Board meeting on the Thomas volume covering the plutonium purification program.

The "Fermi special sample" arrived in Chicago yesterday on the California Limited. The 25 mg Pu^{239} sample (labeled CW-1, but given the designation 49NE by us) was irradiated for 11.7 W days at 2.8 times the pile average neutron flux, discharged from the 105B pile at 2:00 p.m. on November 20 and subsequently sent to Site Y. James received the sample and immediately began to dissolve it.

Joe Katz's transfer to the Information Division is effective today. I have reluctantly agreed that his talents can be better used to help edit the Proceedings of the Metallurgical Laboratory. His group will be dissolved and the members reassigned to other groups. Benjamin Struminski, an SED man who has a B.S. in chemical engineering from City College of New York, is now working with Lawroski.

In the afternoon I went over the patent matter with Kennedy who is in town for the Editorial Advisory Board meeting and the Project Council Information meeting. I indicated that I favor Anderson's proposal and

trust York and Anderson when they say the title as offered would be valuable in covering all commercial applications. I gave Kennedy information from Anderson that indicates that patent people are getting tough about contract dates. I also raised these points about Case 52: (a) Was our July 1941 experiment (in preparation of sample F) done under the NDCrc-201 contract with the University of California which was signed June 14, 1941, but made retroactive to April 1, 1941? (b) What would be the position of Pu^{238} in the patent cases? (Kennedy feels the broader claims certainly cover it.) (c) Our narrowed claims might dominate the present crossover step. (d) York is surprised at how far Anderson's proposal goes, and says this would be the only privately owned case in the whole U.S. governmental patent operation.

We called Metcalf and asked him to have Anderson come to Chicago and meet with us on Thursday to discuss those matters.

Late in the afternoon Kennedy met with Cunningham, Perlman, and me to discuss his possible participation in the preparation of Plutonium Project Record, Volume 16A, "The Chemistry and Metallurgy of the Transuranium Elements." Kennedy thinks that it may be decided not to include Los Alamos work in this.

The British have gained sixteen miles in a drive toward Mandalay, an area of the war about which we receive little information.

Temperatures remained in the 20's all day.

Tuesday, January 16, 1945

Lawrence S. Bartell is terminating work at the Met Lab to enter military service. Mrs. Howlett of Personnel informed him that it is now impossible to secure a SED assignment for him.

Gilbreath, Beard, Hopkins, La Chapelle, and Malm arrived back in Chicago from their trip to Clinton Labs where they have recovered Np^{237} from four special Clinton plant runs. They left Chicago the last week of December except for Gilbreath who went down on January 8 to replace Katz who originally headed the group.

At 8:30 a.m., the new meeting time, I attended the Project Council Information Meeting on Chemistry. Others present were Arnold, Bartky, Borst, Boyd, Brown, Burton, Captain Chapman, Cohn, Compton, C. M. Cooper, Coryell, Daniels, Dempster, Doan, English, Fermi, Franck, Fred, Fussler, Greninger, Hamilton, Howe, Jeffries, W. C. Johnson, Keller, Latimer, Leverett, Manning, Mulliken, Perlman, Rabinowitch, C. Smith, Spedding, Stearns, Stern, Stone, Stoughton, Sugarman, Szilard, Turkevich, Warf, Watters, Wigner, Wollan, Zachariasen, and Zinn. Johnson opened the meeting.

When it came time for me to speak I reported on the following investigations conducted in Section C-I: 1. *Separation Problems.* (a) Preextraction treatment to eliminate hydrazine. (b) Elimination or preservation of neptunium — 4 milligrams are expected to be produced from the runs at Clinton. (c) Solvent extraction as a substitute isolation

procedure. A Hanford-size glass column is now in operation at the West Stands. It is 35 feet high. The feed solution (50 gallons) enters 11 feet under the top of the column and is 3 M in HNO_3 and 5 M in NH_4NO_3. One hundred gallons of hexone enters the bottom of the column while 38 gallons of reflux solution (3 M in HNO_3 and 5 M in NH_4NO_3) enter from the top. One run has been made with 1 mg plutonium per liter and one with 6 mg plutonium per liter. From the top of the first column the plutonium hexone solution is carried to the bottom of a second column which is 24 feet long and 3 inches in diameter, where it is washed by water coming from the top of the column. The columns have functioned perfectly in the first two runs, with yields of 99.5 and 99.7%, respectively. The plutonium is >99.9% pure as far as contamination with lanthanum and zirconium is concerned. The process as developed is intended as a substitute for the present process following lanthanum fluoride precipitation. It is being studied, however, as an overall process or for application at intermediate stages to substitute for more of the present process.

2. *Basic Chemistry of Neptunium.* Through the use of 1 mg of Np^{237} we have been able to characterize three valency states of neptunium — IV, V, and VI. No Np(III) compound has yet been obtained in aqueous solution. The following potentials have been observed in preliminary investigations: Np(V)-Np(VI) = -1.1 v (1 M HCl or H_2SO_4); Np(VI) is yellowish green (pink in perchloric acid), Np(V) is bluish green and remarkably stable, and Np(IV) compounds are colorless; Np(IV)-Np(V) = -0.6 to -0.7 v (1 M HCl); Np(IV)-Np(V) = -0.9 (1 M H_2SO_4). In dry reactions NpF_3 and NpF_4 have been obtained and identified by Zachariasen.

3. *Miscellaneous Problems.* (a) Search for heavy plutonium isotopes. We have found some alpha particles with a range of about 4.1 cm in Clinton plutonium and have attributed them tentatively to Pu^{238}, although the range is not exactly as expected. The value of the cross section for neutron capture by Np^{237} is estimated to be 100 ± 25 barns on the basis of the yield of 93^{238} and the known flux during the bombardment carried out at the P-9 pile at Argonne. (b) Some work has also been done on the U^{232}/U^{233} ratio, protactinium extraction, and theoretical considerations on breeder piles. In conclusion I mentioned that the extraction column I described is now in operation, and I extended an invitation to those present to view it at the West Stands.

Items of interest reported by others were: Warf of Ames reported on the purity of thorium metal and nitrates produces at Ames. The more highly purified thorium assays at a purity of 98-99%.

Burton talked about the Wigner effect in graphite. At Site W there is enough energy stored in the pile graphite to cause a 100°C rise if it were suddenly released. There is apparently less worry about this aspect than the possibility of the graphite crumbling because of disintegration of its crystal structure.

Sugarman reported on the neutron absorption cross section of Xe^{135} — 3.3 megabarns.

Stoughton reported that Clinton Laboratory work suggests the formula of plutonium peroxide is Pu_2O_7. The ratio Pu/O has been measured at 3.3. If precipitated in the presence of H_2SO_4, the ratio SO_4/O is found to be 1/2.75.

Boyd described his work on the mechanism of bismuth phosphate precipitations. The size of the crystals depends on the competition between the rates of formation of nuclei and the growth of the larger crystals.

Latimer talked about the solubility of plutonium phosphate. The probable formula is $Pu_2H(PO_4)_3$. The solubility decreases with increasing phosphate concentration but comes to a minimum at 2 M phosphate, after which the solubility increases, probably due to complex formation. The plutonium in plutonium peroxide seems to be in the tetravalent state and the composition of the peroxide when precipitated from H_2SO_4 solution is very complex.

James completed the dissolution of the 25 mg Hanford-irradiated plutonium (Fermi special sample), labeling the sample 49NE. His assay shows a total of 24 mg of plutonium present. He gave 2.4 mg of the sample to Albaugh for tests of separation processes for converter piles by Hopkins (precipitation process) and Hyman (solvent extraction). The remaining 21.6 mg were precipitated as the hydroxide, dissolved in nitric acid, and oxidized overnight at 90-100°C.

Helen went to her chemistry class today.

The Soviets have taken Kielce, a key city in Poland, according to today's news.

Temperatures still in the 20's!

Wednesday, January 17, 1945

In the morning I attended the Project Council Information Meeting on Physics.

From 12:00 to 3:00 p.m. Kennedy and I met with Anderson, York, and Metcalf in Metcalf's office to discuss Cases 52 and 61. Anderson assured us that under Lavender's proposition, any use of our patent for profit, except by the Government or by a Government licensee solely to supply the Government, would constitute infringement of our patent by such a user. No other deals just like ours are being made. The other kinds of deals are (a) Government take all, used in ordinary cases; (b) contractor retain non-exclusive license, in cases where contractor had prior interest; (c) contractor retain sole license — except Government license (very similar to our deal), in cases where contractor brought to the Government prior work useful to the contract.

Anderson prepared an informal memo describing his proposal. Omitted from the memo is the matter of foreign rights. The Government proposal insists on rights to a non-exclusive license for our Government and the government of the nation of patent; this is the essential part of the proposition. Our government, under the proposal, will prosecute applications in foreign countries where security permits.

The memo provided for the signatures of Anderson, Kennedy, and

me to signify acceptance of the proposal. Anderson and I signed the memo, and Kennedy took a copy to discuss with Segrè and Wahl.

War news remains good for the Allies according to the war summaries in today's paper: British attack Nazis north of Aachen; Soviets win Radom in new Polish drive; U.S. troops advance 32 miles in Luzon.

Temperatures were down to 10°F at 6:00 a.m. but climbed to 28° at 3:00 p.m.

The Project Council Policy Meeting was held at 3:10 p.m. in Room 209, Eckhart Hall, attended by Bartky, Captain Chapman, Chipman, Compton, C. M. Cooper, Daniels, Dempster, Doan, Franck, Hamilton, Hilberry, Howe, Huffman, Jacobson, W. C. Johnson, Captain Karl, Latimer, Leverett, Captain McKinley, Mulliken, Perlman, Smyth, Spedding, Stearns, Stone, Szilard, Tracy, Wigner, and Zinn.

Compton announced that all program recommendations for next year have been submitted to him. He finds that the proposals fall into two categories: (1) essential products and services required by, e.g., Sites Y and W, and (2) research and development. With regard to the latter, Compton said that he was interested to find that three different laboratories have proposed breeder piles for U^{233}. He stated that while we have not been authorized to carry out the program activities as proposed, the indications are that the level of support will be relatively constant and we will have a wide range for making decisions about their priority.

There was a discussion of Compton's proposal that the research and development budget be set at 50% of the essential products and services budget. No conclusions were reached. The readjustment to a research laboratory and the need for a scientific advisory committee to stimulate and correlate fundamental research in the fields of nuclear physics and chemistry were also discussed. Compton asked Smyth and Franck to get together and formulate a formal statement on the need for a review group. That statement will be referred to General Groves.

Compton reported on a discussion with Nichols at Clinton on the possibilities of a U^{233} converter, based on the report prepared early this month which describes the feasibility of such a converter. Compton said that he told Nichols we prefer the breeder, whereupon Nichols said he thinks it is more important that we go ahead on the breeder than on the converter and to lay our plans along this line.

Compton reported that Site Y has produced plutonium metal with a density of 19.4 gm/cm^3. This is significant for weapons construction purposes. The meeting concluded with technical reports by Howe on the 100 and 300 areas at Hanford and by Perlman on the 200 area.

Thursday, January 18, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I announced that the Chemistry Division is retrenching on the number of telephones in the building, and three or four are being moved from our section. The bases for removal are (1) phones on which too few calls are made, and (2) phones on which too many non-Project calls are made. Manning urged that personal calls be cut to a minimum. I also mentioned we still have not rid ourselves of security violations. Last week some individuals in Room 3A (Jones and Eda Kelley), Room 7 (Hufford, Scott, Pat Walsh, and Lorraine Crawford), and 212B in the West Stands (Larson, Hyman, Lincoln, Winner, and Fields), were guilty of security violations, such as leaving reports out and not locking their files.

On the matter of meetings, I mentioned there will be a Section C-I Thursday morning meeting regularly at 8:30 to coordinate the work of the "heavy isotope boys." The time for the meetings of the Council of Section C-I will be shifted to Wednesdays at 8:30 a.m. The Council Information Meeting schedule has been revised so that everyone (Physics, Chemistry, Health, and Technology) is in town at the same time, i.e. all these meetings will be held during the same weeks.

I reported on yesterday's Project Council Information Meeting on Physics. I gave a full account of the meeting. Among other things, Borst reported on an attempt to make U^{236} by long neutron bombardment of enriched U^{235} at Clinton. The material bombarded was UF_6; the U^{236} formed was somewhat concentrated by a Szilard-Chambers reaction, the U^{236} forming UF_4 which deposited on the container walls. Alpha-range measurements by Curtiss at the Bureau of Standards indicate a small lump on the short-range side corresponding to a concentration of 2-3%. A preliminary estimate of the half-life is 10^7-10^8 years.

Also at the Physics Meeting, Stone made an announcement of a very serious matter. The toxicity of plutonium is now determined to be twenty times higher than we have thought. Weight for weight it is as toxic as radium since its physiology is such that it deposits in all the wrong places. It is very serious to get plutonium in cuts. If a vessel containing plutonium breaks and cuts the skin, great care should be taken to wash immediately, to suck out the material if possible, and then hurry to a doctor to have that tissue cut out as soon as possible. The Health people are most afraid of absorption through cuts, secondly of absorption through the lungs. Absorption through ingestion is not so bad. Stone remarked about an acute experiment on a dog in which 1/3 μg of plutonium per gram weight of dog was injected. The dog died in 12 days. Although such large doses are not likely in the laboratory, the chronic effect with much smaller amounts is very serious, because of deposition and strong retention in very critical regions of the body.

There was not enough time to discuss the Chemistry Information meeting.

I sent Hogness a summary of the reasons why we think it is

important to irradiate Pu^{239} and Np^{237} very soon in one of the piles at Hanford. In discussing the Pu^{239} irradiation, I indicated we are interested in (a) the extent of production of Pu^{238} that could affect Hanford Pu^{239} assays, (b) high concentrations of Pu^{240} whose nuclear characteristics will have an important bearing on plutonium operation in converter and breeder piles or its use in weapons, (c) formation of a beta-emitting Pu^{241} giving rise to 95^{241} whose properties are of importance in pile operation, (d) formation of U^{236} by the n,α reaction with Pu^{239}, (e) formation of a plutonium-fission product mixture for testing separation processes.

With regard to neutron irradiation of Np^{237}, I mentioned the formation of pure Pu^{238} and also some Np^{236} whose probable gamma-rays may cause trouble in measurement of decontamination factors at Hanford. I added that the irradiation of U^{235} at Hanford would produce U^{236} in concentrations sufficient for a study of its nuclear properties.

The report "Suggested Research Program in Nucleonics," (MUC-FD-56), was submitted by the Chemistry Division along with a transmittal memo from Daniels to Stearns (MUC-FD-55). It incorporates most of the information given in my January 10 memo to Hogness outlining those problems we believe should be made part of a general research program on atomic power, as well as material furnished by Sugarman, Burton, and Watters. The transmittal memo highlights a number of important points including the significance of fundamental research, the setting of sufficiently high goals, changes in organization of research program in the Chemistry Division and international considerations.

Helen again went to her chemistry class at YMCA college today.

The news today reports that Warsaw has fallen to the Soviets.

Temperatures climbed to 36°F at 4:00 p.m. today!

Friday, January 19, 1945

President Sproul of the University of California responded to my December 9 letter to Underhill asking him to inform Lavender that the University will give Case 61 to the Government if we inventors decide to go through with our plan to assign the case to U.C. without renumeration. Underhill has asked Sproul for advice as to the answer he should make to Lavender. Sproul is now asking me what I think the Regents should do. He writes that should I and the other inventors wish either or both cases assigned without renumeration, he has no doubt the Regents will comply, stating, "for they have no interest at any time except to help you protect your rights, especially in support of the types of research in which you are interested."

Allison sent me a teletype asking that the next sample of Np^{237} be shipped in dilute nitric acid solution. He also asked my opinion on the practicality of reducing 50 mg of irradiated Pu^{239} to metal.

I received a copy of a memo from Zachariasen to Stearns reporting

on the identification and crystal structure of $NpCl_4$ from a sample of sublimed material prepared by Fried. $NpCl_4$ is tetragonal with the UCl_4-type of structure.

I wrote to Kennedy at Site Y and transmitted a copy of Sproul's letter. I explain that I do not feel I can answer it until I learn which alternative has been chosen for Case 52. I also ask his advice on the type of letter I should write to Sproul.

In a memo to Brewer in Berkeley, Davidson asks for samples of reduced sulfides to be used to study the temperature coefficients of the electrical resistance.

Tom Jones sent a justification to Stearns for the purchase of 28 registers to be used with the alpha pulse analyzer apparatus that Ghiorso is developing to detect individual alpha emitters in mixtures of alpha-particle emitters. This will be a powerful device and will be of tremendous help in our unravelling the alpha-particle emitters that we are finding and assigning to elements 95 and 96.

In a memo to Warner, I propose that the Editorial Committee for Volume 19A of the Metallurgical Project Record, "The Production and Separation of U^{233}," consist of Warner, Katzin, Rollefson, Spedding, Stoughton, and Seaborg (Chairman). I ask that Bertrand Goldschmidt be appointed consultant to the Editorial Committee. I also give the tentative Table of Contents of the Volume and authors of each of the eight chapters.

The C-I recovery group has completed the preparation of 2.8 grams of Clinton plutonium for transfer back to the Army office. In turn, we are to receive about 2 grams of 30 gt plutonium from Hanford stock, as arranged in my telephone conversation on January 6 with Compton and Stearns.

We sent 97.2 mg of plutonium to Clinton for a month's bombardment in the pile at 1.4 times the average flux. The sample is in the form of PuF_4 in a quartz tube suspended in graphite inside a regular aluminum capsule. Its purpose is to help to elucidate the alpha emitters that James and Morgan are finding as products of our neutron bombardments of Pu^{239}.

War summaries in this morning's paper read: London — Russians reach border of German Silesia. Western front — British take four towns in Holland drive. Philippines — U.S. troops capture town near Tarlac.

Saturday, January 20, 1945

The health physics surveys for Section C-I for this week show a contaminated centrifuge in Room 35 (Stewart, Asprey, and Anderson) and high radiation levels in Room 13 (James, La Chapelle, and Magnusson), Room 4 (James, Morgan, and Florin), and Room 20 (Katzin). Six individuals are listed as having high alpha-particle hand counts, two from Stewart's group and four from Hindman's group.

Selma Shupp, a technician for Katzin, terminated today.

Headlines today indicate the capture of Lodz and Krakow, and the Soviet army is within 238 miles of Berlin.

Sunday, January 21, 1945

The temperatures are still hovering around freezing, going to a high of 34°F.

Monday, January 22, 1945

Morgan, James, and I conferred on the status of the processing of our neutron-irradiated plutonium samples for the separation of elements 95 and 96. We computed the amount of radiation received by the two Clinton-irradiated samples (49NC and 49ND) and the Hanford sample (49NE) and found evidence that the reaction producing the new alpha-particle activity is of second order.

Today Marny Potter was hired as a technician for Katzin at $28 per week.

I wired a reply to Allison's teletype of last Friday, stating that we can meet his request on the Np^{237} and that we hope to send him by February 1 either 1 or 2 mg, depending upon the yield. I also said we will do the reduction for him (of 50 mg irradiated plutonium) and that it would make it possible for us to avoid a tricky remelt if he can use the material as 5 mg pellets.

Cunningham wrote a summary for me of the work on basic chemistry (which I presented at the Project Council Information Meeting on Chemistry last Tuesday). The memo covers (1) the search for new heavy isotopes and (2) the chemistry of neptunium. Experiments have been carried out on 5 gt Clinton plutonium, on Pu^{239} irradiated for six months in the Clinton pile (sample 49NC), and on 6 gt plutonium irradiated at Argonne for a few hours. Lead and aluminum absorption curves carried out on the Clinton-irradiated plutonium after decontamination from extraneous beta particle and gamma-ray activity do not show any significant differences from similar curves on reference plutonium. The experiments are considered to put an upper limit on the half-life of a beta-decaying Pu^{241} of not more than a few hours. No satisfactory evidence (growth of beta-particle activity in decontaminated material) for the formation of U^{237} (alpha-particle decay of Pu^{241}) has been obtained from these samples. Range analyses by Crawford have failed to reveal an alpha-particle emitting isotope having a range shorter than that of Pu^{239}.

Evidence has been obtained from range measurements for the presence of Pu^{238} (identification not certain) in about 1 gt Clinton Pu and in the Pu^{239} irradiated at Clinton and Argonne. The ratio of 48 activity to 49 activity is about 1 to 10^4.

The results of the Np^{237} bombardment at Argonne are summarized. Under wet chemistry, the work on the oxidation states, potentials, absorption spectra, and compounds of neptunium are summarized; also, the dry reactions of neptunium are described.

At 5:00 p.m. Allison called to tell me we are to receive 105 mg more of the first Fermi Special (Hanford neutron-irradiated plutonium). Thus we shall have temporarily a total of 130 mg. We will retain our original 25 mg. We should reduce 85 mg to metal (after decontaminating by at least 10^3) and send the metal to Site Y. The remaining portion of 20 mg is to be converted into some compound and decontaminated as much as possible for use in gamma-ray measurements at Site Y — treatment with organic matter is to be avoided.

U.S. troops are 65 miles from Manila, and German troops are retreating before British and U.S. troops along most of the Western front, according to this morning's paper.

Tuesday, January 23, 1945

James and Morgan, since a week ago Sunday, have carried sample 49NE (derived from 25 mg of Hanford neutron-irradiated plutonium — the Fermi special sample) through six oxidation cycles (four silver plus persulfate and two dichromate) in order to isolate the element 95-96 fraction. The resulting solution (49NE33) was divided and one-half (49NE33a3) prepared for counting and given to Ghiorso today for fission measurements.

Today's paper reports that the Soviets are only 165 miles from Berlin.

Wednesday, January 24, 1945

At Clinton Labs, the 97.2 mg plutonium sample in the form of PuF_4 (49NF1) is scheduled to be charged into the pile.

At 8:30 a.m. I held a meeting of the Council of Section C-I. It was attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I announced that security violations have come up to a new high this last week: Room 2 (Abraham and Sheft) — file unlocked; Room 10 (Hindman and Kraus) — file unlocked; Room 11 — classified material of Dixon's impounded; 16E (formerly Watt) — safe unlocked; West Stands — key to file impounded.

There has been a change in monthly reports and abstracts. Hereafter, instead of a special monthly abstract to Daniels and Hogness, it will be adequate to send copies of the memos written by the Sub-section Chiefs to me for the monthly Chemistry Information meeting: the dittos

of the monthly reports covering work up to the 15th of the month are due in Daniels' office the 23rd. To enable us to make the changeover, the monthly report due January 23 is being skipped. The problem assignments and man-hour report will be reported to Daniels in an MUC-GTS-memo from each Sub-section Chief. I mentioned two reports of general interest: (1) Diffusion Theory of Neutrons (CP-2475) by Darrow, (2) Principles of Carrying (CC-2516) by Khlopin, translated from the Russian by Rabinowitch.

I then explained the standardization of code nomenclature for heavy isotopes: only two numbers will be used for isotopes whose atomic numbers are 90 or over. For elements between 80 and 89, the same system will be used except that the two numbers will be preceded by an 8.

I next announced the following reshuffling of men: Greenlee from Roy Thompson's group to Hindman's; Hyman from Gilbreath's group to Lawroski's; Hopkins from Katz (group dissolved) to Roy Thompson; Beard from Katz (group dissolved) to Gilbreath. There probably will be a change in the secretaries' time schedules in the very near future. Three possibilities have been suggested: (1) that they work from 8:30 to 5:00 with a half-hour lunch, (2) from 8:24 to 5:06 with a 42-minute lunch period, or (3) from 8:30 to 5:12 with a 42-minute lunch period. There was general agreement that a half-hour lunch period is inadequate, but also a general feeling that while the reasons for such a change may apply to Armory personnel, it would interfere somewhat with the work in the laboratories. The question as to whether this schedule will apply for women technical help will be determined very soon.

Manning reported on the Project Council Information Meeting on Chemistry held Tuesday of last week.

Gilbreath sent me the processing procedures followed on Clinton Plant Runs 294, 295, 296, and 297, carried out under special conditions to maximize recovery of Np^{237}. Each run was carried out on a 2/3-ton scale with material of 8.3 to 11.1 gt plutonium. The slugs were charged to the dissolver between December 23 and 31. The amount of neptunium recovered has not yet been determined, but high plutonium yields were obtained. A flowsheet is attached to the memo.

At 7:45 p.m. I attended an evening meeting on Protactinium Chemistry of Sub-section 2 and Group 9 of Section C-I. Others present were Ader, Asprey, Bradt, Cunningham, Daniels, Davidson, Dixon, Egan, Fields, Fineman, Florin, Fried, Ghiorso, Gilbreath, Greenlee, Hagemann, Hellman, Hindman, Howland, Hyde, Hyman, Jaffey, James, Jones, Katzin, Kraus, Krueger, La Chapelle, Larson, Lawroski, Manning, McLane, Morgan, S. Peterson, Post, Robinson, Schaffner, Sedlet, Sheft, Stewart, Studier, Roy Thompson, Van Winkle, Westrum, Wolf, and others. I introduced Wolf who discussed the testing of a number of solvents for their ability to extract protactinium. He has investigated those solvents which Hyde had previously tested for effectiveness in extracting uranium and had found to pick up little or no thorium. The solvents tested were various ketones, esters and ethers, as well as n-heptanol. He has found that diisopropyl ketone and n-heptanol show the most promise and has worked out tentative optimum conditions for the separation of protactinium from thorium using diisopropyl ketone. I pointed out that evidently solvent extraction from a mixture of protactinium

and uranium nitrates with diisopropyl ketone will result in obtaining both elements in the same fraction. Katzin suggested the use of diethyl ether to perform an initial separation of the uranium from the protactinium. Lawroski suggested the possibility of finding a mixed solvent which will extract protactinium and leave both uranium and thorium in the aqueous phase. I cited the development by the British of a mixed solvent consisting of xylene and a lower ketone for the extraction of uranium.

I introduced Kraus who reported on the isolation of Pa^{231} from uranium ore residues. By working up 6 kg of the carbonate precipitate from the second step in treatment of pitchblende ore, he and his co-workers have isolated 0.5 mg of Pa^{231}. After the protactinium was extracted, MnO_2 was precipitated to eliminate polonium. The protactinium fraction was worked up for protactinium by a zirconium iodate concentration procedure followed by solvent extraction of protactinium into diisopropyl ketone. It is planned to look for the +4 and +3 protactinium ions in aqueous solution using polarographic and spectrophotometric techniques and to study the absorption spectrum of the +5 ions to compare it with that of UO_2^{++}.

I pointed out that the British have reported two alpha-particle components in Pa^{231}. Ghiorso added that he now has an alpha-particle pulse analyzer in operation that will make it possible to check the British findings.

Next I called on Davidson who discussed plans for the preparation of the following dry compounds of protactinium: Pa_2O_5, a lower oxide of protactinium by reduction in H_2; $PaCl_5$, a lower chloride of protactinium; and possibly PaF_5. Reduction of Pa^{+5} to metal and a determination of its crystal structure by x-ray diffraction is planned. I mentioned that the comparison of properties of Pa^{+5} with those of Np^{+5} and Ta^{+5} will be of interest; also that the n,γ cross section of Pa^{231} may be important since we find U^{232} in our U^{233}.

Daniels said that Pa^{231} would be formed in a converter pile by an n,2n reaction on Th^{232}. I estimated that perhaps 0.1% of the neutrons in the center of the pile would be used up by an n,2n reaction. Near the periphery, where the neutrons are slow, the n,2n reaction would not occur. This would still be a better source of Pa^{231} than our present one.

Thursday, January 25, 1945

C. M. Cooper terminated work at the Met Lab as Director of the Technical Division to return to the du Pont Company.

At 8:30 a.m. I held the first meeting of our newly organized Heavy Isotopes Group in my office, attended by Cunningham, Florin, Ghiorso, Hagemann, Hindman, Jaffey, James, Jones, Katzin, Kraus, Larson, Manning, Morgan, O'Connor, Studier, and Van Winkle. I stated that the following program of research is underway or planned: (1) Chemistry of neptunium — Magnusson and La Chapelle; (2) Chemistry of protactinium — Van Winkle Larson, and Katzin; (3) Chemistry of actinium — McLane and Hindman;

(4) Chemistry of 95, 96 — James and Morgan; (5) 4n+ 1 series — Studier, Hagemann, and Katzin; (6) Transplutonium isotope studies — Morgan, James Florin, and O'Connor; and (7) Physical measurements — Ghiorso, Jaffey, Crawford, and Weissbourd.

Katzin talked about the status of the search for U^{232} in U^{233} produced by the neutron bombardment of pure thorium carbonate. Hagemann described the work on the 4n+ 1 series, and Katzin gave the current situation with respect to the search for "neptunium emanation (Nn)."

It was decided that at the next meeting Ghiorso or Jaffey will discuss standard beta-particle counting geometry. It was also decided that for the next meeting we will schedule talks on (a) the search for x-rays from our 5 mg sample of U^{233}, (b) the search for x-rays from Pa^{231}, (c) absorption curves derived from counts on Pa^{233} beta particles, gamma-rays, and electrons, (d) Katzin's work on the 4n+ 1 series, (e) Crawford's work on Hanford neutron-irradiated plutonium.

Transfers of materials arranged at this meeting were (a) Kraus to give 5 micrograms of Pa^{231} to Ghiorso for alpha particle energy measurements on his pulse analyzer, (b) Katzin to give purified U^{233} to Jaffey and Crawford to look for U^{232} on their differential alpha-particle range chamber, (c) Hindman to give 0.4 mg of Np^{237} to Florin to look for 93^{236}, (d) a sample of Hanford neutron-irradiated plutonium prepared by O'Connor, Florin, and Simpson to be given to Jaffey and Crawford to look for 94^{240} using their differential alpha-particle energy chamber.

I attended a Chemistry Division Section Chief's meeting at which all Section Chiefs and Associate Section Chiefs, along with Franck and Daniels, were present. William M. Branch, who is associated with Stearns in the Laboratory administration's work, attended the meeting for the first hour in order to get acquainted with us.

It was announced that Hogness has gone to Washington on a special assignment and will be gone for at least a month. The new proposed schedule for non-academic personnel — 8:30 a.m. to 5:12 p.m., with 42 minutes for lunch — was discussed. There was considerable objection to these hours on the ground that the secretaries do much work between 5 p.m. and 5:30 p.m.

There is to be a change in the general policies of the research program for the next few months. It seems unlikely the program for the sandwich-type converter pile will be approved. Although there is much pressure to release men (especially those from du Pont) from our Project to work on weapons that can be used immediately, such as rockets, the Army and advisers who are familiar with our Project are much interested in new kinds of chain-reacting piles. We can now undertake our own responsibility with a considerable change of emphasis. We must stay within the present personnel and budget, but we can carry out those lines of research that we think are most important regardless of our connection with Hanford. These lines of research will include greater emphasis on 23 work, on fundamental problems, and on new types of piles.

Memorandums were written today by Franck and Daniels to Stearns concerning the organization and morale of the Met Lab. The memos were

written in response to a memo from Maloney criticizing the administration and accomplishments of the Met Lab.

Franck defends the organization of the Lab and, with respect to the Chemistry Division, states

> It was a hectic time for the Chemical Division, and the writer believes that the Project can be proud of the achievements of the academic chemists. All the qualities of our product had to be studied, first with tracer amounts and later with micrograms of the material; the fission products and their chemistry had to be developed; and radiation chemistry had to be built up from scratch. All that had to be done by men who had, with the exception of a few leaders, no experience in the field of radioactivity. That was no minor task. It was well done, even if friction here and there was unavoidable. So far as cooperation is concerned, it was helpful that chemists are more accustomed to teamwork than physicists are.

Daniels likewise supports the Lab, saying as a newcomer he found the Chemistry Division extremely well organized. He states further

> The existence and operation of the structure at X and W and the development in two years of the chemical process, starting with only tracer quantities of a new element, is proof that the job was well done. Undoubtedly, mistakes were made and some unnecessary and expensive experimetns were carried out, but time was short and answers to questions could not be found in books. The record speaks for itself.

Today there was a spill involving beta-particle and gamma-ray activity in Room 7, occupied by Hufford, Scott, Patricia Walsh, and Lorraine Crawford. The highest reading was 9 mr soft radiation at 3 inches distance. The activity was spread over a wide area.

I wrote to Allison at Site Y indicating that we may be able to obtain as much as 5-6 mg of Np^{237} from the second set of special Clinton plant runs and accordingly plan to send him 2 mg on or shortly after February 1.

I then reviewed the results of our studies on the first 24-mg sample of Hanford neutron-irradiated plutonium (Los Alamos No. CWIA, Met Lab sample 49NE). I said we have found some very interesting new alpha-particle radioactivity that we have already observed in some Pu^{239} irradiated with neutrons in the Clinton pile, but the amount was so small that we wanted to check it in this Hanford-irradiated plutonium. This alpha activity (about 25,000 alpha-particle c/m) follows the rare earths in chemistry, and we have not succeeded in oxidizing it to a state or states where its fluoride is soluble; this is just the sort of chemical behavior that I have predicted for the transplutonium elements. It has been at most only partially separated from the rare earths (and actinium), but it is chemically separable from all the rest of the 94 elements.

I said we are inclined to believe that this alpha-particle activity is due to element 95 and/or 96. A very attractive possibility is that we

have found 95^{241} formed from the beta-particle decay of short-lived Pu^{241} which comes from the reaction $94^{240}(n,\gamma)94^{241}$. We find an alpha-particle range of about 4.0 cm, which might correspond to that of 95^{241}. There seems also to be a second alpha-particle component of range about 4.7 cm which could be 96^{242} (i.e., the previously observed isotope of several months half-life) from beta-decaying 95^{242} following the reaction $95^{241}(n,\gamma)95^{242}$. There is, in fact, some direct chemical evidence for this view in that the 4.0 cm alpha-particle activity seems to be at least partially separable from the 4.7 cm activity by chemistry based on our predicted chemical behavior for elements 95 and 96.

Reports in today's paper say that U.S. troops have encountered the first Japanese opposition of any consequence in their drive to Clark Field on Luzon.

Friday, January 26, 1945

Kennedy replied to my letter of last Friday. He encloses a copy of his January 22 letter to Lavender and suggests that I notify Sproul of the arrangements we are making. In the January 22 letter to Lavender, Kennedy refers to Kennedy's and my conference on Cases S-52 and S-61 January 17 in Chicago with Anderson, Metcalf, and York. He states he has discussed Lavender's proposal with Segrè and Wahl and that we four inventors are now agreeable to the proposition as covered in Anderson's informal memorandum prepared at the conclusion of our January 17 conference. Kennedy goes on to restate some matters not clearly included in the Anderson memorandum: (1) The possible option for the Government to buy title (Item 3 of memo) is not considered desirable, and this portion of the proposal is rejected. (2) As regards Item 5 of the proposal, it is the wish of the inventors to assign Case S-61 to the Government either directly or through the University of California. (3) With respect to patent applications in foreign countries it is understood that title to such foreign patents when issued would rest with the inventors, just as in the case of the U.S. patent, and that the Government of this country and also the Government of the country of patent issue would be entitled to a non-exclusive license for governmental purposes. Kennedy closes his letter to Lavender by saying that he is authorized by Segrè, Wahl, and me to accept the proposal as described.

I read a copy of a January 25 memo from Zachariasen to Stearns about the identification and crystal structure of $NpCl_3$. The sample was prepared by Fried by hydrogen reduction of a sample previously identified by Zachariasen as $NpCl_4$. Zachariasen finds the crystal structure of $NpCl_3$ to be hexagonal with a UCl_3- type of structure.

Report CN-2495, "Section C-I, Sub-section 2 — Chemical Research — Basic Chemistry of Plutonium (Cunningham, Assistant Section Chief), Report for Month Ending January 1, 1945," was issued. It contains the following summary of investigations.

Basic Dry Chemistry Group (Davidson, Group Leader). *Vacuum deposition of PuF_4.* Fried and Davidson have found that PuF_4 samples

(ca 5 mg) in degassed CaF_2 or BeO crucibles heated to 1000°C in vacuo for five minutes decompose to PuF_3. This confirms previous results on PuF_4 samples in platinum crucibles at 900°C. It is believed that at 900°C or higher the reaction $PuF_4 = PuF_3 + \frac{1}{2} F_2$ takes place at a low pressure of F_2 gas.

Reaction of UF_4 with dry O_2. Fried and Davidson have found that at 800°C this reaction produces UF_6 plus UO_2F_2, representing a new method of synthesis of UF_6.

Testing of refractories for use in remelting plutonium metal. Hellman has tested seven refractory sulfides that were prepared by the Berkeley group to check their suitability for use in the vacuum remelting of plutonium metal. All refractories tested (thorium, cerium, and barium sulfides at 1000°C) seem to be suitable, with the $Th_{0.32}Ce_{0.65}S$ refractory appearing as the most satisfactory.

Testing of refractories for use in the production of plutonium metal by calcium reduction of the trifluoride. Westrum has found that the following sulfide refractories prepared by the Berkeley group are suitable: $ThS_{1.77}$, $ThS_{1.5}$, ThS, $ThUS_2$, $Th_{0.32}Ce_{0.65}S$, $ThCeS_2$. The plutonium metal produced (7-mg scale) is of good quality.

Electrical transference behavior of plutonium and neptunium. McLane summarized the electrical transference work done on plutonium and neptunium. From this work it is concluded that Pu(IV) and Pu(VI) form complex anions more readily than does Pu(III). Pu(IV) and Pu(VI) change direction of migration in similar ranges of concentration of HCl, HNO_3, and H_2SO_4. The electrical migration behavior of oxidized neptunium is similar to that of Pu(VI). In general, a tendency to form stable complex anions of Pu(IV) [and also of Pu(VI) and Pu(III) which were investigated] is shown by sulfate, oxalate, and acetate, less stable complex anions are formed by nitrate and chloride, while perchlorate apparently forms no complex ions with plutonium. In addition, some evidence is presented for the existence of positively-charged complex ions of Pu(IV) with fluoride and perhaps with phosphate.

Formal potential of the U(IV)-U(VI) couple. Howland has found the formal potential of this couple to be -0.54 v in 1 M HCl and -0.60 v in 1 M H_2SO_4, although in 1 M H_2SO_4 the value in the literature is reported to be -0.41 v.

Recovery Group (Dawson, Group Leader). *Research on methods for plutonium recovery.* Stewart, Britain, and Dawson have found that Pu(IV) is slowly reduced to Pu(III) in dilute HNO_3 solutions saturated with hexone (about 80% reduction in 4 or 5 weeks) when the starting Pu(IV) solution is obtained by hexone extraction of monomeric plutonium from an aqueous solution containing large amounts of the polymeric form.

Effect of sulfate on the distribution of Pu(IV) between aqueous solution and hexone. Stewart, Britain, and Dawson have found that the distribution coefficient of Pu(IV) between hexone and 3 M HNO_3 is changed more than 50-fold in favor of the aqueous phase if this phase is made 1 M in H_2SO_4.

U.S. troops have captured Clark Field in Luzon, according to this morning's paper.

Saturday, January 27, 1945

I informed Perlman at Site W by letter that I am sending drawings of a proposed solution container which might be used to ship dissolver solution from Hanford, should we decide to request it. I gave him the results of our measurement of the cross section for the reaction $Np^{237}(n,\gamma)Np^{238}$ (undertaken with the view to finding how much the Np^{238} decay product, Pu^{238}, will increase the specific alpha activity of Hanford plutonium). The preliminary value for slow neutrons appears to be about 100 barns corresponding to an increase of about 1% in the alpha-particle activity of plutonium manufactured at the 250 gt level.

I also elaborated on an earlier discussion about the new alpha activity we have found in both Clinton- and Hanford-irradiated plutonium, pointing out the possible presence of 95^{241} and 96^{242} in the material. I mentioned the conflicting estimates of the neutron flux to which the Hanford neutron-irradiated material was exposed. Perlman gave us the figure of 3 to 4 times the average flux, whereas Burton gave us the figure 2¼ times pile average. (A knowledge of the flux is necessary for a calculation of the yield which is important with respect to our interpretation of the whole effect.)

Manning sent a memo to Daniels discussing our need for remote control equipment to permit handling of small quantities of highly radioactive material at a distance of 12 to 18 inches. He refers to the work done in Chicago by Kirk and Curtis in developing such equipment for use at Hanford and suggests someone familiar with our problems visit Hanford and learn which types of their equipment would be of greatest use in our work.

According to the health physics survey of Section C-I for this week, there is a contaminated centrifuge in Room 35 (Stewart, Asprey, Anderson) and high radiation levels in Room 4 (James, Morgan, Florin), Room 13 (James, La Chapelle, Magnusson), Room 19 (Krueger, Weissbourd), Room 34 (Britain, Fineman, Haeckl), and Room 35 (Stewart). Six individuals have high alpha hand counts — four from Hindman's group, one from Stewart's group, and one from R. Thompson's group. Nine individuals have high beta-particle hand counts, four in Hindman's group, three in Lawroski's group, one from R. Thompson's group, and one from Ghiorso's group.

Wigner sent the following memo to Compton, with copies to the Reading File:

> Bits of news are reaching us occasionally about rumors at W concerning the validity of the advice which this Laboratory gave the Company operating W. The present letter is prompted by our hearing two bits of news of this kind within two weeks. Rumors of the kind referred to unintentionally impair not only the reputation of the Laboratory but also that of the persons involved. It puts us into an awkward position since we follow secrecy regulations and hence cannot contradict these rumors in the majority of cases.
>
> The first rumor we heard about within two weeks was that, had the Company followed our advice in connection with the number

of tubes at W, the production rate would have been even lower than it is now. It is true that our original report (January 1943) called for 1700 tubes while the piles actually built contain 2004 tubes. This gives an effect of 0.2% in the multiplication constant. However, the Company intended to operate, and did operate for a long time against our oral advice, with 1500 tubes. This gave a loss of 0.1% in the multiplication constant as compared even with our original report. Furthermore, our original report called for much longer slugs which would have given not 0.2% but 0.7% rise in the multiplication constant. As late as May 1944, MUC-GY-9 and CP-1729 called attention to the waste in the multiplication constant occasioned by the unnecessarily thick end caps. It is clear, therefore, that the rumor referred to is unfounded.

The second rumor which we heard within two weeks claimed that, had the Company followed our advice concerning the number of safety rods, it would have been very difficult to operate the pile. The exact opposite is true. The blueprints originally submitted by the Company specified 19 safety rods and this number was increased at our repeated insistence to 29 rods. Fortunately, we heard this rumor from somebody whom we know to be cleared to receive information on this point and we could set it right.

It is suggested that rumors of the above kind be officially discouraged as long as the present secrecy restrictions prevent us from answering them.

Report CN-2496, "Chemistry Division, Section C-I, Sub-section I, Report for Month Ending January 1, 1945," was issued. It contains the following information.

Extraction-Decontamination (R. Thompson, Group Leader). *Methods of increasing the rate of metal processing.* S. Peterson and Greenlee have made two process runs employing a procedure that increases by a factor of four the rate of plutonium throughput in the mainline process without increasing present process volumes at any critical point. Decontamination is completely satisfactory but 7% plutonium losses occur in the extraction step. Further work on this step at UNH concentrations of 28-30% and 1.9 gram bismuth/liter has indicated that plutonium losses decrease somewhat as the plutonium concentration is lowered. Lowering the acidity below 0.8 N and modifying the precipitation procedure slightly seems to show promise of decreasing losses.

Effect of storage of process metal solutions. Bartell, Malm, and Ader have conducted two series of experiments attempting to simulate flowsheet conditions of metal dissolving and subsequent storage. When H_2SO_4 is added before removal of the solution from the metal heel, no Pu(VI) is formed during storage. When H_2SO_4 is not added, 50-75% Pu(VI) is formed. No Pu(III) is found. Addition of 0.001 M N_2H_4 to the 40% metal solution results in Pu(III) formation.

Destruction of N_2H_4 in process metal solution. R. Thompson and Bartell have performed experiments that indicate that $KMnO_4$ but not $Na_2Cr_2O_7$ at room temperature will destroy N_2H_2 in process metal solution. The

presence of H_2SO_4 seems to speed the oxidation of Pu(III) to Pu(IV).

Concentration-Isolation (Katz, Group Leader). *Recovery of Np^{237} in plant runs.* Beard, Hopkins, Katz, and Kelley have found in laboratory experiments that neptunium follows plutonium very closely through the metathesis and peroxide isolation steps. Neptunium losses are less than 1% in metathesis and less than 5% in peroxide precipitations. Neptunium has been separated from plutonium by dissolving the peroxide precipitate in H_2SO_4, oxidizing the neptunium with NaOCl, removing the plutonium as K_2PuF_6, reducing the supernatant with SO_2, and finally removing neptunium with lanthanum fluoride. Overall neptunium losses in this procedure are about 10%. Beard, Katz, Magnusson, and La Chapelle have used this special isolation procedure to recover 2.3 mg of Np^{237} from two Clinton pilot plant runs (half ton of uranium each) modified to assure a considerable yield in the lanthanum fluoride slurry from Room D.

Process Development (Gilbreath, Group Leader). *Effect on decontamination of the presence of 0.05 M HF during the plutonium precipitation step.* Winner and Larson completed two one-liter scale runs (inactive FPE and plutonium concentration at 25 mg/l) in which 0.05 M HF was present in the bismuth phosphate-plutonium precipitation step. They find about a two-fold increase in gamma decontamination at the end of the crossover cycle, in comparison with control runs.

Decontamination with respect to neptunium in the Bismuth Phosphate Process. Hyman and Larson have found in three 100-ml-scale runs that omission of scavengers in the second cycle and the addition of $(NH_4)SiF_6$ to the plutonium precipitation step reduce the neptunium decontamination (removal) factor from 1000 to 230.

Solvent extraction and decontamination. Blaedel and Walling, studying the possibilities of a solvent extraction process for the extraction of Pu(IV-VI) and U(VI) from dissolver solution, have investigated the stability of a number of solvents toward dichromate and find that hexone is highly stable. Means for increasing the stability of hydroquinone in hexone-H_2O-UNH-HNO_3 systems to a point where it would be satisfactory as a reducing agent to produce plutonium(III) have been investigated. It is found that anthranilic acid and hydrazine prolong the life of hydroquinone. Pretreatment of hexone with acid dichromate also greatly increases the life of hydroquinone. Preliminary experiments indicate that aluminum and calcium nitrates are more efficient salting-out agents for plutonium and UNH than is NH_4NO_3. Aluminum nitrate, however, hastens the decomposition of hydroquinone in the second column.

Use of trifluoroacetylacetone (TFA) in extraction-decontamination. Blaedel, Margolis, and Sedlet have found TFA capable of giving excellent separation of plutonium from uranium and also appreciable concentration. However, decontamination from fission products appears to be rather poor.

Semiworks (Egan, Group Leader). The low acidity decontamination procedure has been studied by Flotow, Giegold, Hasenfus, Moseley, Rasmussen, Van Winkle, and Wendrow in nine 100-liter-scale process runs completed through an extraction step and two decontamination cycles. Four of the runs have followed the flowsheet except for omission of scavengers and five are of the low acidity type. Better decontamination is obtained in the low acidity runs, but at the expense of a plutonium yield of 60% (85% accounted for).

Solvent Extraction (Lawroski, Group Leader). Hausman, Schaffner, Schraidt, Simon, and Zvolner have carried out runs employing blank feed solutions and hexone, which indicate satisfactory mechanical operation of the new 3" × 35' glass countercurrent extraction column.

Solvent extraction for decontamination and isolation — fundamental research. Brody, Reinhardt, and Stein have begun laboratory measurements of alpha- and beta-particle and gamma-ray distribution coefficients in order to investigate the possibility of utilizing hexone solvent extraction for decontamination, purification, and concentration of plutonium. As the first step, distribution coefficient studies have been made on feed solutions prepared from active bismuth phosphate extraction precipitates. Compounds $Mn(NO_3)_2$ and $Al(NO_3)_3$ appear to be better salting-out agents than $Ca(NO_3)_2$ and NH_4NO_3, giving alpha particle distribution coefficients (hexone/aqueous) of 20 to 100, beta particle coefficients of 1/3 to 1/300 and gamma-ray coefficients of 1/3 to 1/200.

War news was pushed out of the top headline in today's paper with the rejection by the Senate of President Roosevelt's nomination of Henry Wallace, former Vice President of the United States, as Secretary of Commerce.

Sunday, January 28, 1945

Temperatures hovered in the teens today, finally reaching 20°F from 2:00 p.m. to 5:00 p.m., when they returned to the teens again.

Monday, January 29, 1945

Jones sent a memo to L. C. Furney arranging for the transfer of 2 mg Np^{237} as the nitrate in nitric acid solution (recovered from Clinton runs 294-297) to Site Y by courier. This is the material requested by Allison; these advance arrangements are being made so that the courier will be ready to go as soon as the sample is ready.

The Soviets captured Memel, driving the Germans completely out of Lithuania. U.S. troops opened a new drive on the Siegfried Line.

Tuesday, January 30, 1945

Morgan recorded the following summary of the chemical properties for the alpha emitters obtained from 49ND (8.2 mg of PuO_2 which received 10,223,244 kwh of neutron irradiation in the Clinton pile):

1. Carried 90% on lanthanum fluoride.
2. $K_2Cr_2O_7$ (0.1 M — 100°C — 2 hrs) does not produce a state which is not carried by lanthanum fluoride.
3. Ag^{++} and $(NH_4)_2S_2O_8$ (excess ½-2 hrs) does not produce a

state not carried by the lanthanum fluoride.

4. Zirconium iodate carries 20 to 30% of total alpha particles from oxidized solution.
   a. $K_2Cr_2O_7$ (0.1 N — 100°C — 2 hrs).
   b. Ag^{++} and $S_2O_8^{=}$ (excess — room temperature — ½ to 2 hrs). Note: recycled precipitates yield 50% in the zirconium iodate after long argentic oxidations.
5. Bismuth phosphate carried a part of the total activity, the yield varying with varying conditions.
   a. 20 to 30% from 1 N NO_3^-, 1 N H^+, 0.1 M H_3PO_4.
   b. 50% from 1 N NO_3^-, 1 N $SO_4^{=}$, 1 N H^+, 0.6 M H_3PO_4.
6. Carried 90% by $La_2(C_2O_4)_3$ from excess oxalate and basic oxalate.

Yesterday Ghiorso returned to James sample 49NE33a3 which was given to him for fission measurements last Tuesday. Ghiorso has not found any fission properties of note in this sample. (This sample is a portion of the 95-96 fraction isolated from Hanford neutron-irradiated plutonium — the Fermi special sample). Today James carried out a dichromate oxidation cycle on the sample, remounted it, and returned it to Ghiorso as sample 49NE33a3.1.

Jones requested that Furney arrange for the transfer of 1 mg U^{233} from Katzin to Dempster for mass spectrographic analysis. He mentioned that the sample is approximately 97% isotopically pure.

Jesse, John Simpson, Collins, and I had a discussion in my office concerning the detection of thorium in the presence of uranium in ores. My suggestion was to dissolve the ore in HNO_3, then bubble H_2S through the solution, precipitating as sulfides with lead carrier ThB, ThC, polonium, RaE, and RaD. After 30 hours the first two thorium beta-decay activities which serve as an indicator for the thorium will die out, and beta-particle counts on the uranium decay products will continue. I indicated I did not think much of alpha-particle range measurements because low intensity and self-absorption will defeat the measurement.

San Fernando, "Gateway to Manila," has been captured by the U.S., according to today's newspaper.

It is still cold, with a high of 13°F.

Wednesday, January 31, 1945

Although their terminations will not be effective until some time in February, this is the last day of service for Asher Margolis, A. Pauline Stein, and Lorraine Golden Crawford.

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Kraus, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I described the radioactivity storage facilities being

arranged by Tom Jones. Room 13 has a large cabinet to be used for storage of our supplies of heavy isotopes such as 48, 37, RaD solutions, pure RdTh, actinium, pure radium, and protactinium. Room 4 will have a lead vault for all hot samples. Room 28 will be used for very pure and quite inactive samples.

Stewart complained that the splitting of samples makes the product inventory difficult to maintain. I stated that from now on the man who originally gets the sample or his group leader will be responsible for keeping track of it, no matter how it is split up and transferred to other groups. I also reiterated the need to keep plutonium of different gt levels separate — all product received before October 1944 will be considered to be approximately 2 gt. Gilbreath announced that the semi-works equipment will have to be taken apart soon and suggested that Kraus plan to use the ether extraction equipment for working up his large batch of concentrate (to get Pa^{231} within the next few weeks).

I sent Kennedy a rough draft of my answer to Sproul's letter of January 16, explaining that I want his advice on whether or not he approves of the letter in general and whether or not copies should be sent to Lavender and Bush. My draft letter to Sproul describes the plan evolved in discussion with Lavender and his people for the inventors (Segrè, Kennedy, Wahl, and me) to retain title to Case 52 while the inventors and the University of California assign title to Case 61, without remuneration, to the U.S. Government.

Cunningham sent a memo to Group Leaders of Section C-I describing the accounting system to be used in keeping track of special samples. As I stated in the Council meeting this morning, the basic feature of the system is that the Group Leader or person first receiving the sample is responsible for maintaining a record of all subsequent transfers of the sample. Tom Jones will have the responsibility for seeing that these transfer records are properly prepared and maintained.

At 11:45 a.m. Hamilton called me from Berkeley with the interesting news that the cyclotron is now working with deuterons of an observed energy of 20 Mev (as determined by measuring the range in air) and alpha particles of 40 Mev (up from 16 Mev and 32 Mev). I told him what we would prefer: first, alpha particles on uranium (200 microampere hours); second, alpha particles on 49 (200 microampere-hours); next, deuterons on 49 (possibly using material of high 40 content) and on uranium. I also indicated we want some of the thorium bombarded by deuterons that he is now running.

During this month of January the monograph, "The Chemistry, Purification, and Metallurgy of Plutonium," (MUC-JCW-223), Charles A. Thomas, Editor, and John C. Warner, Assistant Editor, was issued. The material presented comes from the program of research work that had for its principal object the preparation of plutonium in a very high state of purity. In the latter part of July 1944 the extremely high purity specifications of plutonium were removed. The program was therefore terminated and a directive given that a final report be prepared so that work on other matters could be resumed with a minimum loss of time. The Editorial Board for this project is listed as John Chipman, Ermon D. Eastman, Thorfin R. Hogness, Joseph W. Kennedy, Glenn T. Seaborg, Cyril S. Smith,

and Frank H. Spedding. This edition of the monograph appears as two volumes, totaling 539 pages.

The frontispiece for the monograph is a six-times enlargement of a photo showing a ten-gram regulus of pure plutonium as it comes from the reduction bomb. Chapter headings are as follows: I. Introduction. II. Present Process for the Production of Pure Plutonium. III. Fundamental Chemistry of Plutonium. IV. The Crystal Structure of Plutonium and Its Compounds. V. Production of Pure Compounds of Plutonium. VI. Refractories. VII. Production of Plutonium Metal. VIII. Fabrication of Plutonium Metal. IX. Properties of Plutonium Metal. X. Recovery of Plutonium for Re-Use. XI. Special Analytical Methods. XII. Health Hazards in the Handling of Plutonium.

The volume concludes with an alchemical symbol for plutonium designed by Thomas (see Fig. 48).

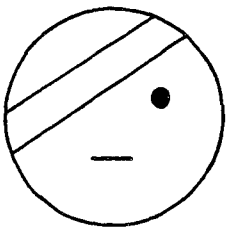

Fig. 48. Alchemical symbol for plutonium designed by Charles A. Thomas.

Katzin, Albaugh, and Cunningham have sent memos to Daniels giving the problem abstracts for the month of January 1945 for Group 9, Sub-section 1 and Sub-section 2, respectively. These include for each problem number a brief one-sentence description of results this month, the names of the people involved, the numbers of the notebooks in which the work is recorded, and in some cases, the numbers of the reports in which the work is described.

"Metallurgy Laboratory, Report for January 1945" was prepared by the Laboratory Director's office. The summary includes the following items of interest:

"A report appraising the feasibility of converting 49 into 23 was completed and transmitted to the Project Director, who in turn presented the report to Col. Nichols. The Division Directors have prepared and submitted outlines recommending plans for the work of the Laboratory beginning July 1, 1945. The final reports will be submitted during the month of February.

"A metal column is being fabricated for the solvent extraction of Product. The results with the 3-inch glass column have proved entirely satisfactory and resulted in the effective separation of metal of very high purity.

"Mr. W. M. Branch has been promoted to the position of Assistant Laboratory Director. The following responsibilities have been assigned

to him: administering of sub-contracts; supervising of the handling and accounting of Special Materials, Product, and previous metals. A procedure for handling and accounting for precious metals is being established. This, as indicated above, will be handled in much the same manner and in the same office as the Product and Special Materials."

The activities of Section C-I, described in the body of the report, covered the period through January 15 and a more complete description of these will be included in the Chemistry Division Progress Report for January.

I attended a 7:45 p.m. meeting in Room 209, Eckhart Hall, of the Basic Chemistry, Recovery, and Instrument Groups (Sub-section 2) of Section C-I. Others present were Abraham, Ader, Ames, Asprey, Beard, Blaedel, Bradt, Brody, Cowan, Cunningham, Daniels, Davidson, Dixon, Fields, Fineman, Fred, Fried, Ghiorso, Gilbreath, Greenlee, Hagemann, Hellman, Hindman, Howland, Hufford, Hyde, Hyman, Jaffey, James, Jones, Katzin, Krueger, La Chapelle, Larson, Lawroski, Magnusson, Manning, McLane, Morgan, O'Connor, S. Peterson, Phipps, Post, Robinson, Schwob, Sheft, Simpson, Stewart, Studier, Templeton, R. Thompson, Walling, Westrum, Winner, and others. I opened the meeting and then, as usual, turned it over to Cunningham who introduced Morgan. Morgan reported on our exciting evidence for the isotopes 95^{241} and 96^{242}. He reported on the work done on the products from four bombardments:

1. Deuteron bombardment of Pu^{239} at the St. Louis cyclotron last fall (49DD).
2. Neutron bombardment of Pu^{239} at Clinton last November and December (49NC).
3. Same as item no. 2, except different amount of Pu^{239} and irradiation, done last December and this month (49ND).
4. Neutron bombardment of Pu^{239} at Hanford this month (49NE).

Morgan explained that, on the assumption that the best place to search for further isotopes might be the rare earth fractions from the chemical procedures performed on the various samples, the first Clinton sample (49NC) was carried through several successive lanthanum fluoride by-product precipitations from dichromate-oxidized solutions. Some 250 alpha c/m (52% geometry) survived which seemed to have a range greater than that of the Pu^{239} alpha particles (first observed December 1, 1944). Bombardments no. 3 and 4 were later carried through similar processes, and residual alpha counts were found to be 1100/m in no. 3 and 27,800/m in no. 4. The following chemical properties of the alpha emitter surviving this treatment have been found:

1. It is carried by lanthanum fluoride.
2. It is not oxidizable to a fluoride-soluble state by dichromate or argentic ions.
3. 20-30% is carried by bismuth phosphate from 1 N HNO_3, 50% is carried from 1 N sulfate, 1 N nitrate, and 1 N total acid solution.
4. 90-100% is carried by lanthanum oxalate from excess oxalate (5% oxalic acid). It is carried from basic solution containing an excess of ammonium oxalate.
5. 20-30% is carried by zirconium iodate from a solution oxidized with either dichromate or silver and persulfate.

In a second cycle on the dissolved zirconium iodate precipitates, 50-70% is carried by zirconium iodate from argentic solution, giving evidence that some fractionation is accomplished by zirconium iodate carrying under oxidizing conditions, perhaps due to carrying of some of the material in the +4 oxidation state.

A range curve (I) obtained with mica absorbers is shown in Fig. 49; the Pu^{239} curve is given for comparison (II).

The curve I indicates the presence of two isotopes. An oxidation number of +3 seems likely for both isotopes, while in addition, one of the isotopes may have a +4 state as evidenced by fractionation by zirconium iodate. On the basis of the properties observed, the following nuclear reactions for formation of these isotopes seem plausible: $94^{239}(n,\gamma)94^{240}$ followed by $94^{240}(n,\gamma)94^{241}$. The 94^{241} apparently decays by beta-particle emission with a short half-life to 95^{241} (which is an alpha-particle emitter with range 4.0 cm) followed by $95^{241}(n,\gamma)95^{242}$. The 95^{242} decays by beta-particle emission of shorter half-life to 96^{242}, an alpha-particle emitter with a 4.8 cm range. Morgan recalled that in the deuteron bombardment of Pu^{239} last fall (49DD) an alpha emitter of range 4.0 cm and with essentially the same chemical properties was found which probably was due to 95^{241} possibly formed in the reaction $Pu^{240}(d,n)95^{241}$. He also recalled that the 4.8 cm alpha-particle emitter found by James last summer must be due to 96^{242} produced in the reaction $Pu^{239}(\alpha,n)$. Morgan then presented data indicating that the reaction producing the alpha-particle activity in the neutron bombardments of plutonium is second order. For example, the ratio of alpha-particle activity found per mg 94^{239} bombarded at Hanford and at Clinton is 7.3. The ratio of total neutron flux in the bombardments in this case is 2.8. The square of 2.8 is about 7.8, in reasonably good agreement with 7.3. Morgan also gave estimates of from 300 to 100 barns for the slow neutron capture cross section for the formation of 95^{242}, based on the half-life energy relationships of Geiger and Nuttal (who estimate the half-life of 95^{241} as 100 to 300 years) and the measured half-life of 6 months for 96^{242} (as measured by James as a result of its production from the helium ion bombardment of Pu^{239} last summer).

Manning asked about the cross section for radiative capture of neutrons by 94^{240}. I answered that the slow neutron capture cross section of 94^{240} on this picture would be lower than that of 94^{239}, the former being about 5-10 barns for a half-life of some 500 years for 95^{241}, and the latter, about 400 barns. A lower limit may be placed on the half-life of 95^{241} because of the fact that it was not found as the product of an α,n reaction on U^{238}.

Davidson asked if it is proposed to prove definitely the identity of these elements by identifying the decay products. Morgan affirmed that it is and said that attempts also will be made to prepare pure 95 and 96 solutions and look for a +2 valence state in 95, using conditions similar to those used for obtaining a +2 state of europium. I suggested it might be necessary to use conditions as drastic as those necessary to obtain the +2 state of samarium. Morgan then presented my table showing the similarities between the actinide series and the rare earths in regard to oxidation states emphasizing the higher oxidation states of

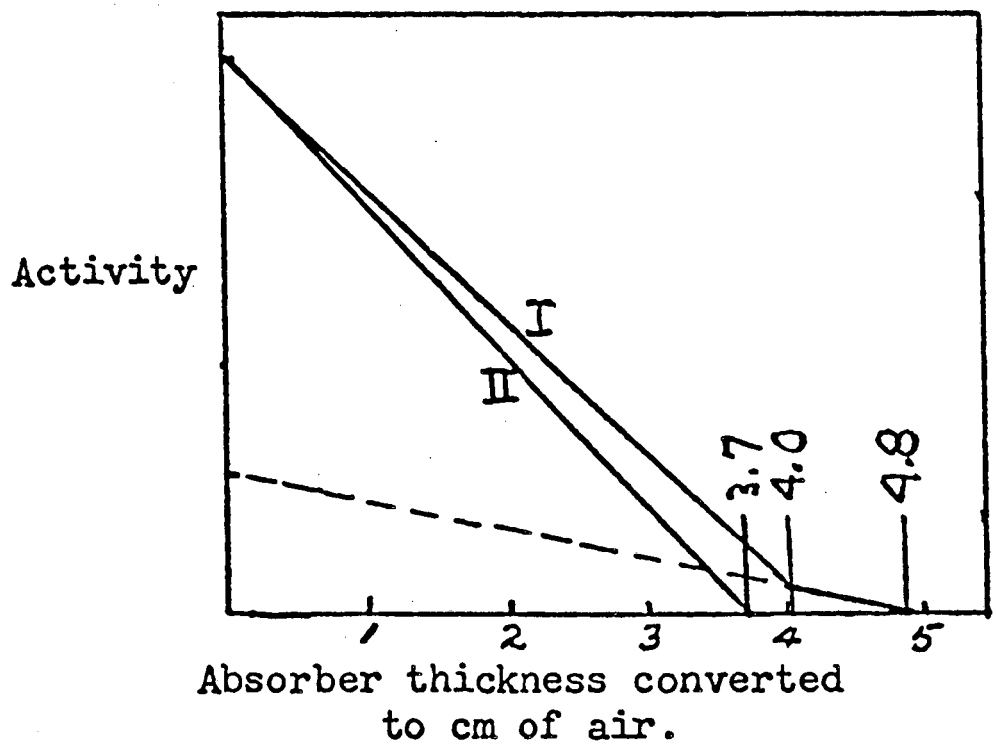

Figure 49. Range curve (I) with mica absorbers; Pu^{239} curve (II) for comparison.

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the actinides and predicting a +2 state for element 95 and +3 and +4 states for element 97.

Comparison Between Oxidation States of Rare Earth and Actinide Series Elements

RE Element	La	Ce	Pr	Nd	61	Sm	Eu	Td	Tb
Oxidation states			5						
		4	(4)						4
	3	3	3	3	(3)	3	3	3	3
						2	2		
Actinide element	Ac	Th	Pa	U	Np	Pu	95	96	97
Oxidation states				6	6	6			
			5	5	5	5			
		4		4	4	4	4		(4)
	3			3	3	3	3	3	(3)
							(2)		

Underlined states are obtained with difficulty, while those in parentheses are extremely doubtful or not yet demonstrated, as in the case of the predicted states of 97.

Cunningham introduced Magnusson who spoke on the radiation characteristics of 93^{238}. Lead and aluminum absorption curves were taken on 93^{238} produced by neutron bombardment of 100 micrograms of pure 93^{237} in the Argonne heavy water pile. The curves indicate there exist two beta-particle components; the one of lower energy corresponding to 0.31 Mev and that of higher energy to 1.35 Mev. No good energy measurements have been obtained on the gamma-ray. The sample was examined by Engelkemeier who looked for coincidence counts and found that the low energy beta particle coincides with the gamma-ray. There was no coincidence between the gamma-ray and the high-energy beta particles. From these data it has been deduced that 93^{238} decays by two paths to 94^{238}: (1) the low energy beta particle is emitted followed by a gamma-ray, or (2) the high energy beta particle is emitted directly to the ground state of 94^{238}. The fact that no fission activity is noticed may be taken as an indication that 93^{237} is incapable of undergoing fission with neutrons at thermal energies. I added that the estimated fission threshold energy for 93^{237} is above the thermal energy, being about 0.4 Mev.

Cunningham introduced Jaffey who spoke on the calculation of the cross section for the reaction $93^{237}(n,\gamma)93^{238}$. Jaffey stated the reason for determining the cross section for this reaction is to learn how much 94^{238} is produced by beta-particle decay of the 93^{238} in Hanford piles that are known to produce one part of 93^{237} to each 300 parts of 94^{239}. A knowledge of the amount of 94^{238} is essential for a radioactive assay of plutonium. The results obtained indicate a cross section of 108 barns, using the physicists' measurement of the neutron flux of 78 barns using the chemists' flux value based on counting an aliquot of the dissolved gold foil used as a monitor. It is believed the physicists' value for the flux

is low. Jaffey also reported on his measurement of the range in air of the 94^{238} (daughter of the 93^{238} produced in the Argonne bombardment) alpha particles; he finds 4.0 cm for the 94^{238} component extending beyond the 93^{237} (3.2 cm) and 94^{239} (3.7 cm) alpha particles in the sample.

Jaffey also presented an account of the experiment, extending over a year [experiment began November 25, 1943] to determine the half-life of 94^{238} by counting the decay of a sample of 94^{238} containing 800 c/m. From these counts a value of 66 years has been obtained for the half-life. This relatively high value (compared with the previously reported value of 50 years) may be due to some 94^{239} in the sample.

U.S. forces have made a new landing on Luzon, says today's paper.

Hitler made a speech last night on the twelfth anniversary of his assumption of power. He spoke twenty minutes and called on his people to stand firm despite the blows they are receiving.

January ended on a cold note weatherwise, with a high of 15°F today, consistent with most of the month!

Thursday, February 1, 1945

At 8:30 a.m. I held the second meeting of the Heavy Isotopes group in my office. It was attended by Cunningham, Ghiorso, Hagemann, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Magnusson, Manning, McLane, Morgan, O'Connor, Studier, and Van Winkle. Ghiorso described the differential alpha energy chamber which can be used on 94^{240}, $94^{238}/94^{239}$ mixtures, and U^{232}/U^{233} mixtures; the accompanying pulse selector circuit was explained. He also described his fission counting techniques. Jaffey discussed backscattering of beta particles and the scattering of gamma-rays. It was decided that Jaffey will arrange with the shop to build ten sets of aluminum and lead absorbers.

The following new activities are scheduled: (1) Hindman is to give Ghiorso 0.4 mg Np^{237} (purified by $NaNpO_2Ac_3$ precipitation) by Saturday. (2) A sample of Hanford neutron-bombarded plutonium (49NE) after preparation by O'Connor, Florin, and Simpson (to put it into a suitable thin layer) is to be given to Crawford and Jaffey to look for Pu^{240}. The next Argonne pile irradiation of Np^{237} to measure the n,γ cross section will take place around February 15. Activities to be considered for scheduling at next week's meeting are: (1) Search for characteristic x-rays from a 50-mg sample of U^{233}. (2) Search for characteristic x-rays from Pa^{234} (and Pa^{233}). (3) Beta, gamma, and conversion electron absorption measurements on Pa^{233}. (4) Katzin's next run on the decay products of U^{233} (the 4n+ 1 decay series).

At 12 noon a special courier left for Site Y on the Santa Fe Chief with the 2.0 mg Np^{237} sample for Allison. The material is in the +4 state in nitric acid solution. I wrote Allison a letter, notifying him of this shipment and describing the purity and other characteristics of the sample in some detail.

I received a memo from Mulliken suggesting that, in connection with the setting of editorial standards for the Metallurgical Project Record, there is an opportunity to take a few steps forward in simplifying and improving nomenclature in the field of nucleonics. He asked my reaction to the following proposals.

1. *Names of radioactive families.* Thorium or (4n + 0) family becomes "One or Tetron-one or Polytetron-one"; uranium or (4n + 2) becomes "Two or Tetron-two or Polytetron-two"; actinium or (4n + 3) becomes "Three or Tetron-three or Polytetron-three."

2. *Symbols for isotopes.* Abandon the historical designations for natural radioisotopes such as RaAc, ThC", U_{II}, in favor of the more logical type of symbolism used throughout the rest of the periodic system.

3. *Names of particles emitted in radioactive transformations.* Change negative beta particle to "negative beton," positive beta

to "positive beton," alpha particle to "alphons" and gamma-ray to "gamma photon," and introduce "radio-neutrons" and "radio-proton when these are emitted in radioactive decay.

My first impression to most of these suggestions is not very favorable.

Jones sent Furney the adjusted figure for the U^{233} inventory (193.6 milligrams, raised from 175.9 milligrams), based on the redetermination of the half-life from 1.46×10^5 years to 1.62×10^5 years.

Compton today sent to Colonel K. D. Nichols the statement (MUC-AC-2562) on the future of the Metallurgical Project that we have helped him put together. The target date of February 1 for completion of this document has thus been met. Such a statement was requested by Nichols in response to a recent letter from Compton advising him that the main assignment of the Metallurgical Project is nearing completion. The statement consists of three parts: (a) The Task of the Metallurgical Project (8 pages), (b) Proposed Budget for Fiscal Year July 1, 1945 through June 30, 1946, (c) Memos on Research (remarks by Dempster dated 12/20/44 entitled "Dangers of exclusive emphasis on winning the present war"; memo to Tolman from Fermi dated 10/30/44 entitled "Anticipated Developments of Uranium Project of Probable National Value"; and letter to Bush from Franck dated 8/1/44 proposing that an interim organization be created during the transition to a more permanent, peacetime organization.) The first of these, (a), includes a summary of the material that the Section Chiefs and Division Directors furnished to Compton.

The proposed program for the Metallurgical Project, 1945-46, is outlined in broader form in the Proposed Budget part of the memorandum, (b), as follows:

I. Fundamental Research

1. Basic laws of nuclear structure and mass-energy transformations.
2. Nuclear properties and the physics, chemistry, and metallurgy of fissionable isotopes, fission product isotopes, and artificial radioactive isotopes.
3. Nuclear properties and the physics, chemistry, and metallurgy of structural materials.
4. Interaction between radiation and matter and its effects on materials and on physical and chemical processes.
5. Fundamental studies in heat transfer, energy removal, energy utilization, and associated fields.
6. Basic investigation of the detection of radiation and of elementary particles.
7. Studies of the mechanisms of toxic poisoning of biological systems.
8. Investigation of the mechanisms of radiation damage to biological systems.
9. Use of radioactive materials for physical, chemical, metallurgical, and biological "tracer" research.
10. Fundamental chemical studies in connection with chemical processes.

II. Applied Research

1. Construction and operation of high-neutron and radiation-flux density piles of enriched "breeder" type with its necessary chemical plant.
2. Process design of new units and their associated chemical plants, particularly process design of "profit operation" "breeder" plants.
3. Consultation and process studies on power piles.
4. Evaluation of present Project information with respect to new military possibilities.
5. Evaluation of industrial possibilities.
6. Determination of tolerance dosage of toxic and radioactive materials.
7. Determination of tolerance dosage for "fast" and "slow" neutron, beta, gamma, and neutrino irradiations.
8. Development of tests for incipient toxicological or radioactive materials damage.
9. Development of tests for incipient "radiation" damage.
10. Development of prophylactic and curative procedures.

III. Consultation and Service Outside Metallurgical Project

In the transmittal memorandum of five pages, Compton identifies the future task as consisting of two parts. The major part is that of scientific research and development of new possibilities in the field covered by our Project. The other is that of supplying essential products and services to Sites W and Y and other parts of the D.S.M. project. Compton goes on to make the following recommendations:

1. That some industry be found to take over the production of radioisotopes and other operations of a production character now being carried out at Clinton Laboratories.
2. The strength of the research and development of new possibilities in atomic nuclear processes be gradually increased by bringing in a few strong research leaders and replacing the poorer men at lower levels with more promising men.
3. Concentrate the research and development in the Chicago area, using Clinton Labs only for production of special materials. (It is not expected, however, that research in this field will remain centralized — as the situation becomes more normal the various universities will handle independently most of the fundamental research.)
4. The work now in hand as prime contracts at Ames, M.I.T., Carnegie Institute, Berkeley, Battelle, and Notre Dame be continued at about the present level.
5. The total annual budget be set at $17.9M, broken down as follows:

Clinton Lab	$ 3.0M
Prime contractors	1.5M
Chicago Laboratories,	
campus	7.8M
present Argonne	1.1M
new Argonne	4.5M

Daniels issued a summary of the manpower distribution in the Chemistry Division which shows the following for my section (78 men):

		Number of Men Dec.	Jan.
Albaugh, Site W work (29 men)	Thompson, extraction and decontamination	7	6
	Katz, concentration & isolation	3	0
	Gilbreath, process development	8	8
	Lawroski, solvent extraction	11	14
Cunningham, Site Y work (38 men)	Simpson, high vacuum work	6	9
	Hindman, basic chemistry	11	12
	Stewart, recovery	6	6
	Ghiorso, instruments and physical measurements	10	10
	Katzin, 23 work	8	8

The detailed organization of Section C-I as of today is the following:

G. T. Seaborg, Section Chief
Edrey Albaugh, Secretary to Seaborg
W. M. Manning, Associate Section Chief
Jane Horwich, Secretary
T. O. Jones, Assistant to Section Chief
Mary Williams, Secretary

Sub-section I — Separation Processes

F. W. Albaugh, Assistant Section Chief (C)
Dorothy Black, Secretary

Group 1 — Extraction-Decontamination

Roy C. Thompson, Jr., Group Leader (AC)
Bradt, Rexford H. (C)
Hopkins, Horace H., Jr. (RA) [SED]
Malm, John G. (JC)
Morgan, Leon O. (AC)
Peterson, Sigfred (JC)
Krinsky, Jerome (Tech)
Summers, Mildred (Tech)

Group 2 — Concentration-Isolation (discontinued)

Group 3 — Process Development

James R. Gilbreath, Group Leader (AC)
Bolden, Mildred, Secretary
Beard, Walter C. (RA)
Blaedel, Walter J. (AC)
Post, Roy G. (RA) [SED]
Sedlet, Jake (RA)
Walling, M. T., Jr. (JC)
Winner, Bernard M. (RA)
Boykin, Pearline (Tech)

Group 4 — Solvent Extraction

Stephen Lawroski, Group Leader (C)
Egan, Clark J., Squad Leader (C)
Schaffner, Irwin J. Assistant Squad Leader (AC)
Simon, Wilbur O. (AC)
Ader, Milton (JC) [SED]
Bernstein, George J. (RA) [SED] (temporary)
Brody, Bernard B. (JC)
Hausman, Eugene A. (RA) [SED]
Hyman, Herbert H. (AC)
Kelley, Alec E. (RA) [SED]
Reinhardt, Richard A. (JC)
Schraidt, John J. (RA) [SED]
Struminski, Benjamin (RA) [SED] (temporary)
Dorcy, Dan J., draftsman
Betty Murray, typist

Sub-section II — Basic Chemistry and Services

Burris B. Cunningham, Assistant Section Chief (C)
Ruth Rogers, Secretary

Group 5 — Basic Dry Chemistry

Oliver C. Simpson, Group Leader (SC)
Davidson, Norman R., Assistant Group Leader (AC)
Fried, Sherman M. (AC)
Phipps, Thomas E. (SC)
Robinson, Herman P. (AC)
Seifert, Ralph L. (C)
Westrum, Edgar F. (AC)
Sheft, Irving (JC)
Peterson, Helen (Tech)
Thomson, Helen (Tech)
Erway, Norman (glassblower)

Group 6 — Basic Wet Chemistry

James C. Hindman, Group Leader (AC)
Ames, Donald P. (RA) [SED]
Dixon, Jonathan S. (JC)
Florin, Alan E. (JC)
Greenlee, Roy W. (JC)
Howland, Jerome J. (AC)
James, Ralph A. (JC)
La Chapelle, Theodore J. (JC)
McLane, C. Keith (JC)
Magnusson, Lawrence B. (JC)
O'Connor, Paul R. (JC)
Billington, Hubert (Tech)

Group 7 — Recovery

Donald C. Stewart, Group Leader (AC) [SED]
Anderson, Herbert H. (AC) [SED]
Asprey, Larned B. (RA) [SED]
Britain, J. W. (RA) [SED]
Fields, Paul R. (JC)
Fineman, Phillip (RA) [SED]
Giacchetti, Olga (Tech)

Group 8 — Instruments and Physical Measurements

Albert Ghiorso, Group Leader (AC)
Jaffey, Arthur H., Assistant Group Leader (AC)
Crawford, John A. (JP)
Dorsey, John M. (RA)
Hufford, Duane L. (RA)
Krueger, Albert C. (AC)
Scott, Benjamin F. (JC)
Walsh, Patricia D. (RA)
Weissbourd, Bernard B. (JC)

Group 9 — 23 Work

Leonard I. Katzin, Assistant Section Chief (C)
Hagemann, French T. (AC)
Hellman, Nison N. (AC)
Hyde, Earl K. (JC)
Larson, Raymond G. (AC)
Studier, Martin H. (AC)
Van Winkle, Quentin (JC)
Wolf, Michael J. (AC)
Potter, Marny S. (Tech)

Non-Academic Service Group

Florin, Kathleen, Group Supervisor and Secretary
Freeman, Elsie Mae, Laboratory Assistant
Porter, Lillie May, Laboratory Assistant

Dieners (all classified as laboratory assistants)

Slattery, John, Supervisor
Barbic, Anna
Calloway, Helena
Douglas, Lester
Little, Edgar
Lucas, Estelle B.
Mabins, Lucy
Martin, Alice
Patterson, Annie Lee
Prince, John
Schroeder, Emil
Smith, Byron
Stull, Vernon
Sykes, Franklin L.
Tadinac, Mike
Tookes, Jessie
Wallace, Ida Mae

(C — chemist; AC — associate chemist; JC — junior chemist;
SC — senior chemist; RA — research assistant;
JP — junior physicist)

In summary, this breaks down as follows: GTS + WMM + TOJ = 3; FWA + 6 + 7 + 11 = 25; BBC + 9 + 11 + 6 + 9 = 36; LIK + 7 = 8; total = 72 (plus two temporary men in Group 4). Secretaries = 7;

Laboratory Assistants, Technicians, etc. = 14; Cleaners (Laboratory Assistants) = 17; total = 38.

Academic employees = 72; Non-academic employees = 38; Total = 110.

"Metallurgical Laboratory, Report for January 1945," (MUC-JCS-156) has been prepared by the Laboratory Director's office. The summary includes the following:

"A report appraising the feasibility of converting 49 into 23 was completed and transmitted to the Project Director, who in turn presented the report to Col. Nichols. The Division Directors have prepared and submitted outlines recommending plans for the work of the Laboratory beginning July 1, 1945. The final reports will be submitted during the month of February.

As usual, Helen attended her chemistry class today.

German radio indicates tonight that the Soviet army is only 45 miles from Berlin; the Soviets say they are 63 miles from Berlin. U.S. forces of the 8th Army in the Philippines have captured the naval based of Olongapo on Luzon, while the 6th Army is within 25 miles of Manila.

February is starting with continued cold weather.

Friday, February 2, 1945

Helen Pellock Peterson, a technician in Davidson's group, terminated today.

I read a copy of a letter dated January 30 from Lavender to Kennedy. Lavender expresses his gratification at receiving Kennedy's letter of January 22, advising acceptance of the patent proposition as set forth in the memorandum prepared by Anderson at our January 17 conference in Chicago. He goes on to say that the preparation of a formal agreement will be undertaken shortly. Lavender refers to an agreement between the inventors of Patent application S-52X and Segrè and says it would be of assistance to have a copy of that agreement. He indicates that Kennedy's understanding that the Government will prepare, file, and prosecute the case without expense to the inventors is correct. Lavender also indicates that he believes it will be acceptable for us to reject the option to the Government to buy title. He confirms Kennedy's understanding that the non-exclusive license for Governmental purpose includes foreign governments.

With regard to Case S-61X, Lavender believes the simplest method would be for the inventors to assign the invention directly to the Government and for the Government to secure the acceptance and approval of the University; however, he wishes to await the letter from the University of California that I advised will be forthcoming.

U.S. Rangers and Philippine guerrillas freed over 500 prisoners, mainly Americans, on Luzon, according to this morning's paper.

It is still cold, with a high of 22°F.

Saturday, February 3, 1945

Ghiorso returned to James today sample 49NE-33a3.1, a portion of the 95-96 fraction isolated from Hanford neutron-irradiated plutonium that was given to him for fission measurements on Tuesday. James began an experiment to determine if fractionation of the sample into 95 and 96 components can be accomplished by oxalate precipitations.

The Health Physics surveys in Section C-I for this week show a contaminated centrifuge in Room 37 (Stewart) and high radiation levels in Room 4 (Florin, James, and Morgan), Room 13 (James, La Chapelle, and Magnusson), Room 31 (Greenlee and Peterson), and Room 36. Two individuals are reported to have high alpha-particle hand counts, one in Katzin's group and one in Hindman's group. In addition, sixteen individuals have high beta-particle hand counts, four in R. Thompson's group, eleven in Hindman's group, and one in Lawroski's group.

Blaedel and Gilbreath sent me a procedure for the determination of free nitric acid in the presence of aluminum nitrate and uranyl nitrate.

In response to Farrington Daniels' request, Davidson and Katzin sent him their personal views on what they consider to be the major initial chemical research problems to be solved in the design of homogeneous, "circulated," D_2O moderated, converter or breeder piles. The problems that they identify as being common to a majority of designs and types of structures are: (1) recovery, decontamination, and recycling of reacting material with extreme efficiency, (2) problems of the absorbing blanket, (3) general mechanical problems of homogeneous structures, and (4) general nuclear problems.

According to war summaries in today's paper, U.S. troops have made good gains on Luzon; U.S. troops are driving to the Rhine on a 30-mile front; and the Soviets announced they are within 51 miles of Berlin.

Sunday, February 4, 1945

The temperature reached 31°F today — the highest so far for February.

Monday, February 5, 1945

The first delivery of plutonium to Los Alamos from Hanford was made last Friday.

Today the second Hanford plutonium manufacturing pile (Pile D) reached its rated power level.

I received a letter from Saul Winstein at UCLA informing me he has learned from Professor Francis A. Blacet that Jack Roof (a mutual

friend when we were undergraduates at UCLA) is at Edgewood Arsenal and may get away about April 1. Winstein says that Jack was approached previously by other branches of the Manhattan Project on the possibility of employment but each time replied that he was tied up for awhile. Winstein goes on to say that Blacet thinks Jack is inclined to try some such work instead of going back to Oregon State immediately where he is an assistant professor of chemistry.

I also received a letter from Ray Stoughton written February 3, saying that the 5 grams of natural uranium carbide (8 pellets) were shipped to me January 25. They were irradiated in the Clinton Pile from January 13 to January 23. The total exposure during that period was 29,500 kw-days, and the samples received 1.4 times the average neutron flux. He added that either he or Spof English will be seeing me about the middle of the month.

Daniels conferred with Stearns and Compton today in order to present his case for high temperature piles. Stearns and Compton both expressed keen interest and encouraged Daniels to have the Chemistry Division spend an appreciable part of its effort on research connected with new piles as long as these activities do not interfere seriously with the program of fundamental research and writing (for the Metallurgical Project Record) now underway.

In the course of discussions immediately preceding the conference Stearns informed Daniels that Hilberry, in looking over the future Project program, has come to the conclusion that there are too many chemists in it. Hilberry also told Stearns that Compton has approved the curtailment of the program as suggested by Hogness. This would reduce the Chemistry Division from 250 to 125 people early in 1945, and it is anticipated that normal resignations will bring the number down to about 100 people by the first of July. Daniels was warned to prepare evidence for a continuing need for a large staff if such is believed necessary.

The banner headline today reads "Yanks Enter Manila." U.S. troops entered the city in a wide encircling movement under cover of darkness.

Tuesday, February 6, 1945

Report CN-2511, "Progress Report. Solvent Extraction Process for Concentration and Isolation of Product. December 30, 1944," was issued. It was written by Lawroski, Dawson, and Tepe and contains the following information of interest. The report describes the progress made during the period from the initiation of work in July 1944 to January 1945 on the development of the hexone extraction process for concentrating and isolating plutonium. An extensive program of engineering development work was undertaken by the General Engineering section and Lawroski's group after the chemical feasibility of the process had been indicated by laboratory studies. Two continuous countercurrent extraction systems were constructed and operated. The hexone extraction column for the first and smaller system was about 1 inch in diameter and 16 feet tall. Fourteen runs were made in which 99.7% yield and 99.9% purity of plutonium

were consistently obtained. These figures are based on 250 mg of plutonium rather than on the tracer quantities employed in the tests. A larger system with a hexone extraction column 3 inches in diameter and 35 feet high was then constructed. Four runs have been made at this time using no plutonium. The transfer of lanthanum and zirconium obtained was the same as that for the one-inch column. These results are highly satisfactory and indicate both the mechanical and chemical feasibility of the process.

It is believed that the substitution of a solvent extraction operation for the present isolation step will improve the overall yield by at least 1% and that the use of such a solvent extraction isolation step will permit considerable reduction in labor costs.

The present consensus as to the most operable solvent extraction isolation process is as follows. A solution of plutonium nitrate is first prepared by complexing the lanthanum fluoride product precipitate from the crossover step using a solution of zirconyl nitrate. Ammonium nitrate is next added to the zirconium-complexed plutonium solution. This so-called "salting-out" agent is added to displace the equilibrium distribution of plutonium in favor of the solvent phase. Practically complete separation of plutonium from the principal impurities, lanthanum and zirconium, is effected by continuous countercurrent extraction with hexone in a packed column having both stripping and enriching sections. The plutonium is quantitatively extracted from the aqueous phase by the hexone stream which flows upward. A small fraction of the lanthanum and zirconium are extracted along with the plutonium. These impurities are stripped from the hexone phase in the upper section of the same column by countercurrent washing with an aqueous solution of ammonium nitrate and nitric acid. The solvent that passes from the top of the column carries practically all of the plutonium, some nitric acid, and essentially no lanthanum or zirconium.

A second extraction operation is necessary to recover the plutonium as an aqueous solution that can be subjected to further purification or metal production operations. This is accomplished by washing the hexone solution countercurrently with pure water in the aqueous extraction column. The final step in the process is a plutonium oxalate precipitation designed to effect the bulk reduction that is necessary prior to shipment. A flow diagram for the foregoing process is attached to the report.

In a memo to Jaffey, Crawford evaluated an earlier proposal to measure the half-life of U^{232} indirectly by observing the rate of emission of alpha particles from such a sample over an extended period; the problem is complicated by the presence of the thoron daughter which would diffuse out of the sample and emit alpha particles under a geometry greater than the rest of the emitters in the sample. Crawford presents calculations that show that the method proposed to slow the diffusion of the thoron from the sample until it decays cannot work reliably.

Katz, Gilbreath, Albaugh, and I met with Nickson to discuss a memo from Stone indicating that Katz and his group of chemists seriously contaminated Room 54 of the 706A building at Clinton when they were there from December 29 to January 15 recovering Np^{237} from the lanthanum fluoride slurries from four special Clinton Plant runs. It was the feeling that a

misunderstanding exists, as the records of room surveys for alpha-particle contamination before, during, and after the visit do not differ substantially, especially when one takes into account the fact that the later surveys are expressed in dis/m, whereas the earlier survey results are in c/m at 50% geometry.

Albaugh wrote a memo to me about the possibility of separating plutonium and fission product elements (FPE) by a flotation process based on a suggestion from Daniels. Albaugh points out that most industrial experience has been with froth flotation which, due to radiation hazards, would be inadvisable for plutonium and fission products. The other type of flotation which involves orientation of a solid particle at a liquid-liquid interface, known as agglomerate flotation or "solvent flotation," is more suitable for our purposes, but the lack of information makes an initial evaluation more difficult. Free energy considerations suggest solvent flotation should be feasible. In order for the highly polar plutonium or FPE particles to be absorbed at an interface, their surfaces must first be rendered relatively non-polar by adsorption on a collector layer; for the systems with which we are concerned, the collector would probably be (1) a soap if operating at a high pH, or (2) a salt of a substituted quaternary amine or a sulfated agent if operating in acid solutions. Albaugh points out that the most favorable situation for the separation of plutonium from FPE by flotation would be to have the plutonium entirely in solution and the FPE entirely as solid phases, or vice versa. This situation, however, is not a requirement as one solid phase can be floated and another rejected by the use of a selective collector or solvent.

Albaugh suggests that two or three man-months of work should suffice to determine whether or not such a plan has any real possibilities. He also believes that useful preliminary experiments along the line of solution vs. solid phases would be to determine the degree of separation of plutonium and FPE obtained by electrodialysis of acetate, oxalate, citrate, and carbonate-complexed solutions.

Helen went to her chemistry class at YMCA college today.

Over 1350 more U.S. soldiers were freed on Luzon; in addition, 3700 civilian prisoners were freed from an interment camp. Tales of brutal treatment from the Japanese and calculated starvation are being revealed.

Wednesday, February 7, 1945

At 8:30 a.m. I held a meeting of the Council of Section C-I in my office. It was attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Kraus, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I began by pointing out the security violations for the last few weeks have hit a new high (a total of 81 altogether). For the week ending last Saturday there were five violations, including Room 7 (Hufford, Scott, and Patricia Walsh), Room 13 (James, La Chapelle, and Magnusson), Room 15 (Seaborg), Room 16B (Ghiorso, Jaffey, and Crawford), and the West Stands.

I announced that Koshland will be in town this Friday, Stan Thompson will be here the week after next, and Perlman and English are coming for this month's Chemistry Information Meeting. I also mentioned that the names of some of us on the Project are considered classified and aliases have been given to be used when necessary, especially in long distance telephone calls and teletypes. For example, my alias from now on will be G. T. Sutton. I then read a summary of a memo from Daniels about such items as the need of the Area Office for an attendance record before that office will recommend deferments; the fact that a non-academic work schedule of 8:30 a.m. to 5:12 p.m. has been adopted as a compromise to be used at all sites; the security hazard of giving our personal correspondence address as the Metallurgical Laboratory; and the extent of interest in having public pay phones installed at the Laboratory. Manning asked those present about the need for Plutos and portable Geiger counters; it was decided that one Pluto instrument per room would be desirable, as well as a total of six or seven Geiger counters.

In view of the increasing emphasis in our section on converter pile problems involving 23, I then asked Katzin to describe some of the latest problems that have been considered. (Katzin and Davidson have been keeping in touch with the physicists, especially Wigner, on these matters.) Katzin made the following observations: (1) At the present time the sandwich structure is not being considered very much because it is less efficient than other types. The heavy-water homogeneous pile is being more thoroughly considered. (2) In the heavy-water pile, the evolution of D_2 and O_2 gas is a serious problem at the minimum planned power level of 100,000 kw. If nitrate is used, the problem of nitrate stability enters. (3) Solvent extraction seems to be the best process for removal of fission product poisons. (4) Other problems include the possibility of insoluble compounds settling out as a result of the 100 grams per day production of fission products and the possibility that, in the nitrate solution, the plutonium may polymerize and not solvent-extract.

Katzin also mentioned that other, non-homogeneous, types of structures are being considered. They involve running a pile at higher neutron temperatures to avoid too high a loss of neutrons in the materials of construction.

I received a teletype from Allison about the Pu^{238} sample we sent him last December. Measurements indicate one-half the total alpha-particle count is caused by Pu^{239}, whereas my December 26 letter is interpreted by Allison as stating that the total sample is one-half Pu^{239} by weight. He asks which of these is correct, and also, whether or not we are able to estimate the Pu^{240} content of the plutonium that contaminates the sample. He cannot be certain until he has answers to these questions, but indications are that Pu^{238} fissions spontaneously.

Daniels sent me comments on Albaugh's memo on the feasibility of separating plutonium and fission products by flotation methods. It is Daniels' opinion that a research program is well worth two or three man-months, and if encouraging results are obtained, the program could be properly expanded. He observes that Albaugh with his special experience along these lines is ideally suited to direct research on flotation processes. He agrees with Albaugh that froth flotation cannot be used

because of the hazards involved and goes on to suggest four lines of attack on the problem. He mentions his awareness of our manpower difficulties and says it would be proper to shift some of the men now working on the Hanford processes to this new program.

Stone sent a supporting memo to Stearns and Whitaker for someone from Section C-I to visit Hanford to see the small-scale remote control apparatus developed by Kirk and Curtis for handling plutonium. Manning suggested such a visit in a memo on January 27.

I conferred with Paul Aebersold who is in town. Among other things he told me about some uranium (perhaps one gram) rich in U^{234} (ratio of U^{234}/U^{235} of about 1/10) made in a special run at Y-12. Arrangements have been made with Murphy and Doan to give some to Clinton Labs. Aebersold will let us know whether or not he can arrange to send us some. He also said they can make about 80 grams of material especially poor in U^{238} and may do so. He told me of the formation of a committee on uranium analysis: Rodden, Bassett, Aebersold, Watters, a Y-12 man, and a K-25 man. For techniques of spreading uranium uniformly on plates, he referred me to a report written within the past two months by R. C. Lilly of the Berkeley Rad Lab.

According to this morning's paper, the squeeze on Germany by the U.S. and British on the west and the Soviets on the east is getting tighter every day.

Thursday, February 8, 1945

At 8:30 a.m. the Heavy Isotopes Group met in my office and the meeting was attended by Florin, Ghiorso, Hagemann, Hindman, Jaffey, James, Jones, Katzin, Larson, Manning, McLane, Morgan, O'Connor, Studier, and me. La Chapelle and Magnusson came in later. Studier gave a tentative flow-sheet for treating 26 mg of U^{233} (which has been decaying since November 18) in order to isolate the various daughter fractions for the purpose of studying the decay schemes. There was then a general discussion of the problem. O'Connor describes his and other proposed work on the sample of Hanford neutron-irradiated plutonium from Site Y sent to us for decontamination.

The following activities are scheduled: (1) Make blanks [48 and 49] for Crawford. (2) The run on $93^{237}(n,\gamma)93^{238}$ is to occur about February 15. (3) Hindman is to provide Jaffey with a sample of ionium plus Pu^{239} for use in testing his alpha-particle range chamber. (4) Hindman is to provide Ghiorso with a sample of Io + 49 + 48 + Po to use to test his alpha-particle analyzer. It was stressed that copies of all curves obtained are to be provided for the file.

Activities to be considered or scheduled at next week's meeting are: (1) Look for characteristic x-rays from the 26 and 50 mg U^{233} samples. (2) Ghiorso is to look at the 6 mg Np^{237} sample for characteristic x-rays. (3) Ghiorso and Van Winkle will discuss the Pa^{231} sample being prepared for Site Y.

Mullikan wrote to Roger Williams in Wilmington to explain that he is going to inform those at Site W to whom we have sent their Met Lab or Clinton Labs notebooks, that the laboratory notebooks are considered to be on loan to them and that they will be receiving a request that all notebooks are to be returned to the Met Lab or Clinton Labs by a certain date.

Helen attended her chemistry class today.

President Roosevelt, Prime Minister Churchill, and Premier Stalin are meeting in the Black Sea area and have reached agreement on joint military operations to conclude the war against Germany.

Temperatures have still not climbed to freezing this month!

Friday, February 9, 1945

Jaffey and Magnusson have completed their beta-particle counting of the Np^{238} produced in the sample of Np^{237} that was bombarded with neutrons for 10.2 hours on January 9. Using the data for the neutron flux, time of bombardment, amount of Np^{237} (correcting for a small amount of Pu^{239} impurity), they obtained a value of 126 barns for the cross section of the $Np^{237}(n,\gamma)Np^{238}$ reaction. This value can be improved by measuring the neutron flux more accurately and by determining the decay scheme of Np^{238} to allow a more accurate translation of beta-particle counting rate to the corresponding absolute disintegration rate.

Koshland arrived today on a visit from Clinton.

Report CS-1951, "Chemistry Division Summary Report for January 1945" was issued. Essentially all of Section C-I information in this report appears in the summaries of work prepared by Katzin, Albaugh, and Cunningham for my use in connection with the January 16 Project Council Information Meeting on Chemistry.

The temperature reached 41°F today — the high for this month!

Saturday, February 10, 1945

James completed two oxalate precipitation cycles begun last Saturday on sample 49NE-33a3.1, a portion of the 95-96 fraction isolated from Hanford-irradiated plutonium. He ran range curves on 49NE-33.8a (second oxalate precipitate) and 49NE-33.5a2 (first oxalate supernatant). Both samples show about the same proportion of long-range alpha-particle activity, and this indicates that very little fractionation of the 95 and 96 has occurred.

At the request of T. O. Jones, a radiation survey was made of Room 212, Kent Chemical Laboratory, by the Health Division. The room was found to be contaminated above the tolerable level.

Dawson, Stewart, and Britain, in describing experiments carried out over the last several weeks, sent me flowsheets for batch (as opposed to continuous) extraction processes employing hexone for removal of plutonium from (a) the dissolved metathesis product at Hanford, and (b) the zirconyl complexed crossover precipitate at Hanford. They conclude that both of these batch processes can be adapted to operations in the equipment already in the Hanford 224 building. Interest in such batch extraction processes was expressed by Squires and Watt as early as last September.

I wrote to Connick in Berkeley to invite him to a meeting of the Editorial Committee of Volume 16A, "The Chemistry and Metallurgy of Transuranium Elements" at 4 p.m., Monday, February 19. This will fit in with his visit to the Met Lab to report at the Chemistry Information meeting. We plan to discuss the detailed organization of Volume 16A, the Survey Volume, and begin the organization of Volume 16B, the Collected Papers Volume. I indicate that we would like to assign to him the responsibility for Chapter IV of Volume 16A, "Oxidation States, Oxidation-Reduction Reactions, Equilibria, and Potentials"; our Section C-I will handle Chapters V, "Ionic and Molecular Species in Solution and Their Equilibria," and VI, "Preparation and Properties of Compounds."

Katzin sent George Cowan at Los Alamos the details on the use of diethyldithiocarbonate to separate uranium and thorium.

"Big Battle in South Manila" is today's banner headline. There is fighting for the Manila docks.

Sunday, February 11, 1945

The D pile at Hanford, which reached its rated power level last Monday and had its power reduced slightly for a few days in order that certain adjustments could be made, is returning to full power today.

Monday, February 12, 1945

Morgan carried out tests yesterday to determine if fractionation of 95 and 96 can be accomplished by zirconium iodate precipitations from argentic-oxidized solution. He used the supernatants from the oxalate precipitations carried out by James from February 3 to February 10 on the 95-96 fraction isolated from Hanford neutron-irradiated plutonium. Morgan combined the two supernatants (49NE-33.5 and 49NE-33.7), precipitated $Y_2(C_2O_4)_3$ by acidifying, dissolved the precipitate, oxidized with silver plus persulfate, precipitated zirconium iodate, metathesized the precipitate with NaOH, and then ran range curves on the various fractions. He found no fractionation of the alpha-particle activities. As in previous experiments the zirconium iodate carried about 30% of the alpha-particle activity.

Yesterday Westrum prepared 60.5 mg of plutonium metal for shipment to Site Y using less than 69.5 mg of trifluoride equivalent. The starting material was part of the Fermi special sample (CW1) of 120 gt material sent from Site Y in the form of +6 nitrate.

I received a memo from Stearns dated February 10 asking me to meet with him in his office at 4:00 p.m. on the 20th to discuss alpha-particle contamination and high alpha air counts discovered in the hot lab at Clinton Labs, alleged to be left by Katz and others from Section C-I during the working up of the product from the special neptunium runs. Others to be present at the meeting are Doan, Daniels, and Stone.

Canadian troops have been moving rapidly the past few days and are beyond Kleve — a German fortress city — according to today's paper.

Tuesday, February 13, 1945

Marian Pinckard was hired to work as a technician in Katzin's U^{233} group.

I talked by phone with Monk about preparing for us surfaces of various elements (aluminum, cerium, antimony, selenium, and bismuth) by the evaporation technique for Jaffey to use in performing back-scattering experiments. These elements were chosen to give us a reasonable spread of points on a back-scattering vs. atomic weight plot. I explained that some substitution of elements is possible if there is experimental difficulty in preparing films from a particular element. I also suggested that since some regions contain no hard metals susceptible to the optical polishing technique, the evaporation technique seems most likely to give good results. Monk indicated that he was willing to do the work, and I told him that I would have Jaffey describe what we want done in detail.

D. P. McDougall of the Bruceton Laboratories (under the auspices of NDRC) visited the Met Lab this afternoon and interviewed Beard, Brody, Dixon, Hyde, Winner, and Howland for possible positions at Bruceton where they would be trained in rockets and high explosive techniques, after which some of them might be sent to Site Y. After the interviews McDougall indicated to Daniels that we might hear further from him about Howland and Winner. The other men he interviewed are too young for the supervisory positions involved.

Helen again went to her chemistry class.

The top headline in today's paper deals with the results of the Big Three (Roosevelt, Churchill, and Stalin) Conference at Yalta. A bulletin was issued covering nine subjects.

Wednesday, February 14, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Asprey, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, and R. Thompson. Security violations were again on the agenda. I remarked that for the last week their frequency has kept up its recent terrific pace. Rooms involved are 3B (Jones), 7 (Hufford, Scott, and Pat Walsh), 12, 14 (Edrey Albaugh and Kay Florin), 27 (Beard and Hopkins), 29 (Bradt), 32A, 32B, 36. I asked what could be done to stop such violations. It was finally decided that security officers would be appointed for the section to check up on all rooms between 6:30 and 7:30 p.m. These officers will be selected from a list of serious security violators (since they know best about the business) and will remain on the job until another serious violator is caught by the security officer. I cautioned the men to be careful about telling Army visitors secret information as there are restrictions about what such visitors are supposed to know.

Jones brought up the subject of safeguarding plutonium. Some of the people high in the security hierarchy are insisting that plutonium must be locked up at all times. These people are not cleared to know what our product is, nor how much we have, nor the nature of its use so it is difficult to explain to them the problems encountered in such a procedure. The Army men at the Armory who do understand some of the difficulties, however, are under pressure to do something about increasing the safeguarding. Jones presented some suggestions which have been made. It was decided that the best solution to the problem would be to (1) have special badges or special colors on the badges for those people who work in the filtered air section and (2) have two extra guards here and at the West Stands at night to check on everybody to see that plutonium is not being stolen. Manning then announced that Kay Florin will make an inventory of the plutonium in our section.

I received a letter from Kennedy who returned the rough draft of my letter to Sproul and indicated that he, Segrè, and Wahl approve of it as written. He is in favor of sending copies of the letter to Lavender and Bush. Kennedy also transmitted for my comment a draft of an answer to the letter dated January 30 that he received from Lavender.

In a memo to Monk, I confirmed our telephone conversation of yesterday and sent him Jaffey's description of the work we would like him to do for us in connection with the preparation of surfaces for back-scattering experiments.

Zinn called me from Argonne about the mounting of a plutonium sample for fission measurements at the Argonne pile. The sample is to be in the form of a 1.5 mg film, 2 cm in diameter. Any compound and any content of Pu^{240} is acceptable although low Pu^{240} content will be best. Zinn will send us the backing plate and holder tomorrow — we suggested 5 mil platinum for the backing plate. Goett will come in from Argonne to help put on the Zapon coating. For neutron measurements on U^{233} Zinn wants a 60-mg U^{233} sample in solution form. He is making a quartz cell and will let us know the concentration desired.

I talked by phone with Hamilton in Berkeley about the following: (1) He now has reached 5 microampere-hours of alpha-particle bombardment on our uranium target. The yield at the increased alpha-particle energy is up by a factor of 500 for the $Bi^{209}(\alpha,2n)85$ reaction. The runs were made at 0.1 microampere. (2) He finds some polonium, perhaps from the reaction $Bi^{209}(\alpha,p2n)Po^{210}$ — the yield is the same order of magnitude as the 85^{211} or maybe, alternatively, some D^2 is present in the beam. (3) He finds no fission in the bismuth + alpha-particle bombardment. (4) There was no D^2 present in the alpha-particle bombardment of our targets. (5) He wants us to send him more (6-12) uranium disks of 3/16 inch thickness — good clean stuff — a 5% molybdenum alloy is best from the standpoint of corrosion resistance.

At 7:45 p.m. I attended the meeting of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I in Room 209, Eckhart Hall. Others present were Abraham, Asprey, Bradt, Crawford, Cunningham, Daniels, Davidson, Dixon, Dorsey, Egan, Florin, Fred, Fried, Ghiorso, Gilbreath, Hagemann, Hellman, Hindman, Howland, Hufford, Hyde, Jaffey, Jones, Katzin, Krueger, La Chapelle, Lawroski, Magnusson, Manning, McLane, Nickson, O'Connor, S. Peterson, Phipps, Post, Robinson, Sedlet, Sheft, Simpson, Studier, Templeton, Van Winkle, Walling, Westrum, Winner, and Wolf. Continuing the series of lectures to acquaint chemists in Section C-I with the essentials of nuclear physics, Crawford talked about alpha, beta, and gamma radiation and how their interaction with matter (ionization, scattering, back-scattering, and secondary radiation) affects the instruments used to measure the radiation.

Ghiorso reported on the Geiger counter radiation characteristics of some of the Np^{237} produced at Clinton (corresponding to approximately 7 gt level of Pu^{239}). The sample used was not specifically purified from its daughter Pa^{233}. Absorption curves taken through lead and aluminum indicate there are soft (L) x-rays associated with the Np^{237}. It is not possible, however, to conclude with certainty whether the x-rays are due to Np^{237} itself or to Np^{236} that might be present (also isomeric Np^{238} or Np^{240} are possibilities).

I remarked that the limit of 50 years has been set for electron capture decay of Np^{235} produced by a d,2n reaction on U^{235} and the limit of 500 years was set for Np^{236} produced by the d,n reaction. Ghiorso said that this would strengthen the evidence against Np^{236} as the source of x-rays in his work. He went on to say that the number of x-rays produced per alpha particle is 10 times as great for Np^{237} than for U^{233} or Pu^{239}. I commented that if a counting efficiency of 10% is accepted for (L) x-rays, then one x-ray is emitted for each 10 alpha particles. This might indicate that several energy levels of the Pa^{233} atom result from the alpha decay of Np^{237}.

Howland presented data on the absorption spectrum of U(III). The solutions used were made by dissolving solid UCl_3 in HCl (Charles Kraus at Brown University provided the UCl_3). The UCl_3 has a 'half-life' in solution [for oxidation to U(IV)] of from 5 to 20 minutes. I asked if there is any correspondence between the U(III) peak and the Np(IV) and Pu(V) peaks. Howland said that there is, but Hindman offered the opinion that it probably doesn't mean anything. I then asked whether or not the

spectra of Np(V) and Pu(VI) have been compared. Hindman replied that these spectra are not similar. I pointed out that the chance for the study of isoelectronic configurations is greater in the case of the actinide series than in the rare earth series.

Later in the discussion I asked if the strong and sharp peaks may be taken as evidence for the filling of the 5f electron shell since such sharp peaks are found in the case of the rare earths where the 4f shell is being completed. Hindman said that he believes that it could although other transition elements such as iron, cobalt, and permanganate manganese show peaks of similar intensity of absorption of which the half-intensity width is of the same order of magnitude as in the case of the rare earths and the actinide elements. I asked about the extinction coefficient for the permanganate. Hindman replied that this is not in the dipole radiation class (its $\varepsilon = 2000$). He commented that all that sharpness of peaks means is that electronic transitions are protected from perturbation by the surrounding ion atmosphere. In some cases, e.g., the cobalt halides, this is accomplished by complex formation. Basically isoelectronic configurations such as U(IV), Np(V), and Pu(VI) might be expected to be altered by the presence of covalent oxygen.

At the conclusion of the meeting Daniels announced a seminar for Monday in Room 2 of Rosenwald, at which Coryell will speak on fission product activities.

Today is Valentine's Day, and I gave Helen a box of candy.

U.S. troops have captured Nichols Air Field and Cavite naval base in Manila Bay, according to this morning's newspaper.

Thursday, February 15, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, which was attended by Cunningham, Florin, Ghiorso, Hagemann, Hindman, Jaffey, James, Katzin, La Chapelle, Larson, Magnusson, Manning, McLane, Morgan, O'Connor, Studier, and Van Winkle. I covered the following items: (1) I told of Hamilton's phone call yesterday about cyclotron operations. (2) I asked people to write their data in my notebook in order to help fill in my summary table on carrying. (3) I asked for copies of absorption curves for the files, specifying the data desired.

We discussed the progress of the work on the 20 mg Site Y sample (part of the larger Fermi special plutonium sample CW1 that had been bombarded at Site W and put through a decontamination cycle at Site Y — total sample 90 mg). I indicated we would put at least one more man, in addition to O'Connor, to work on a small fraction (perhaps 1 mg) by a different method.

The following activities were scheduled: (1) Hufford, Seifert, and Florin will make samples of 48 and 49 for Crawford to use as blanks. (2) The neutron irradiation of Np^{237} at Argonne will take place next Monday. (3) Morgan and James will make twelve 3/16 inch thick uranium disks for Hamilton.

Activities to be considered or scheduled at next week's meeting are: (1) Ghiorso and Van Winkle will review the work on Pa^{231}. (2) Ghiorso to look for x-rays in 26 mg and 50 mg samples of U^{233}. (3) Ghiorso to look for x-rays in 6 mg of Np^{237}.

Yesterday and today Morgan carried out tests on aliquots of a 95-96 fraction (sample 49NE-33.30) derived from Hanford-irradiated plutonium (Fermi special sample) that show: (1) There is essentially no fractionation of 95 and 96 by zirconium iodate precipitation from Fe^{+2}-reduced solution. Sixty percent of the total alpha-particle activity is carried. (2) Under strong reducing conditions, i.e., Jones reductor — zinc amalgam, 95-96 is not reduced to the +2 oxidation state. Europium is reduced more than 50%. (3) The 95-96 fraction is not carried on Bi_2S_3 at 0.25 N HCl, giving additional proof of the unique chemical behavior of 95 and 96. Bismuth, lead, and polonium are known to carry on Bi_2S_3 under these conditions.

Connick wrote from Berkeley that he expects to arrive in Chicago either Sunday or Monday at noon and so will be able to attend the 4:00 p.m. meeting on Monday of the Editorial Committee of Volume 16A, "The Chemistry and Metallurgy of Transuranium Elements."

I sent a letter to President Sproul of the University of California about Cases 61 and 52. In answer to his letter which I received January 19, I tell him of the arrangements being made between the inventors and the Government. I indicate the different handling of the two cases: (a) The inventors (Kennedy, Wahl, and I) will retain title to the inventions related to Case 52, with the Government receiving a non-exclusive license for governmental purposes. (b) The inventors (Segrè, Kennedy, and I) will assign all rights to Case 61 to the Government, either directly or through the University of California.

I explain that it is the wish of the inventors that the University of California will agree to the foregoing proposal and that he, Sproul, will decide in conversations or correspondence with Bush, the appropriate mechanism for making the gift of our rights (i.e., those of the inventors and the University) in connection with Case 61 to the United States Government. Copies of the letter were sent to Bush, Lavender, and my co-inventors.

I also wrote to Kennedy returning his draft letter to Lavender with my suggestions. I suggest that perhaps he and Art should give some thought to the possibility of sending more notebooks, if there are any, which might be used by the attorneys here to make an even stronger Case 52. I remind him that, according to the agreements, any work upon which the three of us can be considered to have collaborated may be used. I point out that it may even be possible to control the whole present production process under the broad claims.

In a letter to Latimer in Berkeley, I enclose, as a matter of interest, copies of my correspondence with the University concerning Patent Cases 52 and 61 (my letter to Underhill, December 12, 1944; Sproul's letter to me, January 16, 1945; and my letter to Sproul, February 15, 1945).

This was the last day at the Met Lab for Kurt Kraus who is transferring to the Clinton Laboratories; he will report there on February 17.

Albaugh and Cunningham sent Daniels the Problem Abstracts for their Sub-sections I and II of Section C-I for the period February 1 through 15.

A new organization chart of the Scientific Divisions of the Met Lab was issued today. It is essentially the same as the one issued January 6 with the exception of Charles M. Cooper who terminated his work at the Met Lab on January 25. The Technical Division has been disbanded.

Helen attended chemistry class at YMCA College today.

Japanese troops remaining in Manila are still resisting, fighting stubbornly in the dock area, says this morning's paper.

Friday, February 16, 1945

The third Hanford plutonium manufacturing pile (Pile F) was charged with uranium for the first time yesterday.

I received from Albaugh a summary of the work of Sub-section I for my use at the Project Council Chemistry Information meeting next Wednesday. The topics covered are: (1) *Increased production.* Laboratory development of a method for processing four tons of uranium per day in a single Hanford canyon has been completed. (2) *Neptunium-plutonium separation.* In bismuth phosphate extractions preceded by an $H_2C_2O_4$-Mn(II) prereduction about half the neptunium is carried; if nitrite or no prereduction is employed, less than 10% of the neptunium is carried. (3) *Recovery of Np^{237} in plant runs.* In four plant runs (two-thirds ton uranium each) the bismuth phosphate process was modified to produce and isolate 8 mg of neptunium via the lanthanum fluoride slurry from Room D at Clinton Laboratories. (4) *Use of trifluoroacetylacetone for analyses of Pu(III)-(IV).* This analytical method was shown to be effective with synthetic UNH solutions of known plutonium(III)/(IV) ratios. (5) *Overall solvent extraction "Redox" Process.* The effect of the composition of the solution on the distribution ratios of plutonium, U(VI), and HNO_3 has been determined for systems containing hexone, H_2O, HNO_3, UN, $K_2Cr_2O_7$, and NH_4NO_3 [or $Al(NO_3)_3$]. (6) *Overall solvent extraction using trifluoroacetylacetone.* A series of successive extractions from aqueous dissolver solution gave more encouraging results than previously found although decomposition of TFA and non-reproducibility of distribution ratios were noted. (7) *Solvent extraction for decontamination, concentration and isolation.* Multi-stage experiments measuring distribution coefficients between hexone and salted solutions of dissolved bismuth phosphate extraction precipitates showed very favorable plutonium extractability, the values being of the order of 20 to 1 in favor of hexone. (8) *Hexone extraction for isolation-development.* Two runs, processing 1.14 and 2.53 g of plutonium in feed volumes of 50 gallons each, were completed successfully in the 35-foot long, 3-inch diameter glass column built to test hexone extraction on a

plant-scale basis. Converter Pile Decontamination Problems: A. *Decontamination of plutonium by solvent extraction.* When hexone is used as a solvent and the feed solution contains 0.8 g plutonium per liter (irradiated at Site W), $Al(NO_3)_3$, and HNO_3, it is found that approximately 1% of the total activity still accompanies the product after four simulated stages in the first column (hexone extraction column) and five stages in the second column (wash column). At least two species of activity appear to be present in the final solution, the predominating one being ruthenium. B. *Decontamination of plutonium by precipitation.* Plutonium compounds including the phosphate, oxalate, iodate, K_2PuF_6, peroxide, and phenylarsonate have been precipitated from solutions containing active FPE and approximately 1 g plutonium per liter. Decontamination factors for gamma-rays varied from 3 to 6. Only the iodate and K_2PuF_6 have solubilities sufficiently low to recommend their use at this plutonium concentration. A lanthanum fluoride by-product precipitate from oxidized solution followed by K_2PuF_6 precipitation gave a decontamination factor for gamma-rays of 83 and a 95% yield of plutonium. A silica gel scavenging followed by peroxide precipitation resulted in a decontamination factor for gamma-rays of 70 and a 84% yield of plutonium.

Nickson sent Stone a memo about the contamination of Clinton Laboratories Room 54, Bldg. 706A, allegedly by Katz and other Section C-I chemists when they were recovering Np^{237} from special Clinton plant runs in the first half of January. The job involved the handling of 17 grams of plutonium. Nickson is of the opinion that the charges are unfounded, that our men were well aware of the possibility of damage by plutonium, and that their techniques of handling plutonium are not inferior to those in groups elsewhere.

Daniels sent a "Memo to File" about his discussions with Stearns on February 5 about the proposed size of the chemistry staff for the future program in nucleonics. Previous discussions with Hogness indicated plans to reduce the size of the total chemistry staff from about 250 last summer to 125 the first part of the year with the thought that an additional 25 would leave in the normal course of events before July 1. When Daniels questioned Stearns recently in regard to our obligation to give physical chemists to Bruceton, Stearns indicated that Daniels should not worry about further reductions for the present and, if we need the men here, we should keep them. Stearns then stated that he conferred with Hilberry who is thinking in terms of 130 chemists for the program after July 1. Daniels concludes from these conferences that no further pressure will be applied to reduce the staff in the Chemistry Division during the next few months but in view of the probable return of some men from Sites X and W, it is advisable to accept any voluntary resignations from the Division.

The Soviet Army has smashed the Oder River line, and Canadians and British have gained on the drive on the Rhine.

Saturday, February 17, 1945

James began experiments to test elements 95 and 96 in a regular separation of rare earths to see which ones they most resemble, inasmuch

as the facts point more and more toward only a +3 state for both 95 and 96. As a source of 95-96 he used a portion of sample 49NE-33.21 derived from the Hanford neutron-irradiated plutonium (Fermi special sample), adding it to a mixture of equal concentrations of Ce^{+3}, indium, lanthanum, yttrium, samarium, praseodymium, europium, scandium, and gadolinium. He plans to follow the procedure of Noyes and Bray's book, *A System of Qualitative Analysis for the Rare Elements*, to separate these elements into a number of fractions.

Katzin summarized for me, by memo, the work of Group 9 for my use at the Project Council Chemistry Information Meeting next Wednesday. He gives the following information: About 600 micrograms of Pa^{231} have been isolated from 6 kg of carbonate residues obtained from uranium extraction procedures. About 150 micrograms of this have been purified and are being used for basic chemical experiments. A larger-scale isolation from 15 kg of carbonate residues, rather richer in protactinium, has been started. A 36-inch countercurrent extraction column employing ether has been set in operation to separate U^{233} from irradiated thorium. The column handles a can of thorium carbonate (200 g) in five hours. About 15 cans have so far been extracted. Reinvestigation of the usefulness of calcium and magnesium nitrates as salting agents has indicated that these two salts are probably the best yet found for uranium-thorium separation by ether procedures.

Manning and I conferred with Daniels, Watters, and Hilberry about the Army's desire to have the Met Lab make independent analyses of the Hanford plutonium as a control. It was agreed we would cooperate in carrying out whatever tests might be necessary. We may be called upon to determine plutonium content by alpha-particle counting, to make spectrograms as a test for impurities, and to prepare 10 mg of material in a form suitable for analysis by Dempster on the mass spectrograph. We also agreed that samples of 50 mg would be ample for all these tests. Since the production of the material at Hanford is top secret, great precautions will have to be taken to prevent any leakage of information concerning the number of samples or the amount of material which they represent. T. O. Jones will take charge of storing, accounting, and administration of the tests.

We also discussed with Hilberry the necessity, for our interests and for scientific purposes, of receiving half-gram samples of plutonium at various power levels during the course of operation of the pile. We explained that we desire only representative concentrations, not samples from every batch, and when the pile gets to full operating level the program can be ended.

T. O. Jones sent Furney a description of the system he has for keeping an inventory on the Hanford plutonium samples to be received from the Army (Special Project Office Product). This is to be kept separate from the Metallurgical Project plutonium.

U.S. troops have captured the Bataan Peninsula, according to this morning's newspaper.

Sunday, February 18, 1945

The Berkeley bombardment with 40 Mev alpha particles on a natural uranium target (sample TαB) began. The bombardment will terminate next weekend. The purpose of this bombardment is to study the reaction $U^{238}(\alpha,2n)Pu^{240}$.

Cold weather has returned with a high of 21°F!

Monday, February 19, 1945

Stan Thompson and Iz Perlman are here from Hanford in order to attend the Project Council Information Meeting on Chemistry and other meetings. They will also confer with members of Section C-I.

The second irradiation of Np^{237} was carried out in the Argonne heavy water pile by Jaffey in order to produce Np^{238}. Jaffey will study its radiation characteristics and determine the n,γ cross section of Np^{237}. The 100-microgram sample of Np^{237} was prepared by La Chapelle. The neutron irradiation took place from 11:11 a.m. until 9:15 p.m. with a gold monitor used to determine the neutron flux.

Cunningham wrote a summary of the work of Sub-section II of Section C-I for my use at the Chemistry Information Meeting the day after tomorrow. It contains information on the following topics:

1. Search for new isotopes. Regarding the search for new isotopes he writes, "Some very interesting new alpha radioactivity has been found, both in plutonium irradiated with neutrons in the Clinton pile and plutonium irradiated with neutrons in the Hanford pile. This alpha activity exhibits just the sort of chemical behavior which has been predicted for the trans-plutonium elements. For example, it is carried by rare earth fluorides, and it has not yet been possible to oxidize it to a state or states where its fluoride is soluble. It seems to be chemically separable from all the rest of the 94 elements, and the present best interpretation is that it is due to elements 95 and/or 96. The alpha activity is composed of two components, one of range 4.0 cm and the other of range 4.7 cm. A very attractive possibility is that the 4.0 cm alpha activity corresponds to 95^{241}, formed from the beta decay of 94^{241} which comes from the reaction $94^{240}(n,\gamma)94^{241}$, and the 4.7 cm alpha activity corresponds to 96^{242} (cf. reports CS-2124 and CS-2135) from the beta-decaying 95^{242} following the reaction $95^{241}(n,\gamma)95^{242}$. The ratio of the yields in the Hanford as compared with the Clinton bombardment seems to be proportional to the second power of the total neutron irradiation, as would be expected on the basis of these particular isotopic assignments, although the accuracy of the estimation of the neutron fluxes is not sufficient to make this at all certain."

Cunningham continues on to say that a sample of Clinton plutonium was bombarded with slow neutrons in the Hanford pile to produce the equivalent of about 100 gt plutonium. An analysis of data taken in the differential range chamber shows a long-range plutonium alpha-particle

activity at a level only twice that encountered in 2-5 gt plutonium. It is concluded that this activity is Pu^{238} and cannot be due to Pu^{240}.

A reevaluation of the data on the neutron bombardment of Np^{237} reported last month gives 95 barns ±20% as the most probable value for the n,γ cross section. The radiation from Np^{238} is found to consist of about 50% 0.35 Mev beta particles and about 50% 1.35 Mev beta particles and 1.0 Mev gamma-radiation. The gamma-radiation originates from the transition to the lower ground state of the product nucleus following the emission of the 0.35 Mev beta particles. Soft x-rays have been found associated with completely decontaminated Np^{237} — about one per ten alpha particles. If the x-rays originate from Np^{236} (formed by an n,2n reaction on Np^{237}) then the half-life for Np^{236} would have to be about one year, which seems improbable on the basis of other evidence.

A search for U^{237} in 10 gt Clinton plutonium gave uncertain results with the measurement of less than 60 beta-particle disintegrations per minute that could be ascribed to U^{237} (which might result from the alpha decay of Pu^{241}). The alpha-particle energy analysis apparatus has been undergoing extensive tests with samples containing several different alpha ranges. It is now possible to distinguish the presence of Th^{230}, U^{233}, Pa^{231}, Pu^{239}, Po^{210}, and Pu^{238} in a mixed sample.

Next Cunningham reviewed investigations on his sub-section in basic chemistry.

2. Chemistry of neptunium. Polarographic measurements by J. Watters place the formal potential for the couple:

$$Np(III) = Np(IV) + e^-$$

at approximately +0.1 v or less in 0.25 M H_2SO_4. The reduction appears to take place irreversibly. A reevaluation of data from work reported last month gives -1.144 v as the most probable value of the potential of the Np(V)-Np(VI) couple in 1 M HCl, and -1.13 v for the potential in 0.5 M H_2SO_4. It is believed that reversible behavior is exhibited by the couple in both solutions.

The preparation of $NpCl_4$ starting with Np(IV) oxalate was accomplished by heating the sample to 500° in the presence of CCl_4 vapor while the preparation of $NpCl_3$ has been accomplished by reduction of $NpCl_4$ with hydrogen.

3. Chemistry of uranium. The absorption spectrum of U(III) in 1 M HCl has been determined on the Beckman quartz spectrophotometer.

4. Chemistry of plutonium. $PuCl_3 + H_2S$ at 800-900°C gives Pu_2S_3, but the structure is not the same as that of cubic Pu_2S_3 obtained by the reaction of H_2S on PuO_2. From a study of the absorption spectrum of Pu(IV) in 1 M HNO_3 containing various amounts of HF, it has been found that the value of K for the reaction:

$$PuF^{+++} \rightarrow Pu^{++} + F^-$$

is approximately 10^{-7}. Preliminary evidence for the formation of PuF_2^{++} and PuF_3^+ in the presence of higher concentrations of fluoride has been obtained.

Daniels sent a "Memo to File" concerning discussions with Szilard on February 12, 14, and 17 on the formation of a small group of physicists to try out in the laboratory radically new ideas, such as the separation of uranium isotopes by the use of solvent extraction through the use of uranium(VI) and uranium(IV) valence states. Szilard is going to discuss the matter with Wigner and then confer with Daniels who afterwards may take it up with Stearns.

At 4:00 p.m. I attended a meeting of the Editorial Committee of Volume 16A, "The Chemistry and Metallurgy of Transuranium Elements," along with Connick from Berkeley, Perlman from Hanford, Brown from Clinton, and Cunningham from our section.

Today I calculated a possible half-life of 100 years for 95^{241} based on an estimated cross section and yield data from the St. Louis deuteron bombardment of plutonium (containing 0.02% Pu^{240}). Then using yield data for the production of 95^{241} and 96^{242} in the neutron bombardment of plutonium at Hanford and values of 100 years and 4 months for the half-lives of 95^{241} and 96^{242}, respectively, I calculated a cross section of 2 barns for the reaction $94^{240}(n,\gamma)94^{241}$ (assuming 94^{241} is a short-lived beta-particle emitter) and 200 barns for the reaction $95^{241}(n,\gamma)95^{242}$ (assuming 94^{242} is a short-lived beta-particle emitter).

In the evening there was a Chemistry Division Seminar in Room 2 of Rosenwald. Coryell spoke on fission product activities.

The war in Europe continues to roll on both fronts with Germany being embattled from both directions.

Tuesday, February 20, 1945

At 8:30 a.m. I attended the Project Council Physics Information Meeting. Items of interest reported were: (1) Harold Lichtenberger of Argonne talked about the development of a velocity selector for neutrons consisting of a rotor with alternate layers of cadmium and aluminum. The device rotates and allows intermittent passage of neutrons and is suitable for work with neutrons of energies from 0.005 to 0.25 electron volts. (2) Zinn described his crystal spectrometer that handles neutrons of higher energies. Using it, he has determined the values of the resonance band for plutonium as 0.28 electron volts and 2500 barns, values somewhat lower than anticipated. (3) Seymour Bernstein of Clinton reported on the measurement of the energy of delayed neutrons from fission by a cloud chamber method. (4) Nordheim talked about the changes in reactivity with temperature to be expected in an enriched homogeneous pile. (5) Jesse described new instruments being devised, including a supersensitive Pluto which will detect 3000 alpha-particle counts per minute. (6) Zachariasen reported that the precipitate obtained from alkaline solutions by English at Clinton does not show any evidence for the purported Pu(V). Results on plutonium peroxide studies are still negative. The purported NpF_5 submitted by our section for examination did not contain crystalline Np(V). (7) Dempster said he has reanalyzed the sample of U^{233} submitted by Katzin

and finds it to contain 95.8% ± 0.4% U^{233}. (8) Evidence has been obtained at Site Y that neutrons are emitted following fission but within 10^{-9} second after fission. (9) Weinberg reported on breeder piles operating at higher neutron energies at which the capture-to-fission ratio would vary from that observed with thermal neutrons. (10) Seitz talked about the graphite stored-energy problem. There appears to be some danger of an explosion due to local heating in the pile that would bring about a rapid release of the stored energy, but there seems to be no danger from this source until the stored energy in the graphite reaches a value of 50 calories per gram. The graphite in the Hanford pile has now reached the 30 calorie per gram level and is still rising. It is hoped that it will level off because of self-healing before it reaches the 50-calorie level; if not, it perhaps will be desirable to attempt to heal the pile by controlled heating.

Gilbreath wrote a memo to me about the extent of contamination that occurred in Clinton Labs Rooms 54-56 of the 706A Building, during the Np^{237} recovery operations carried out there by our Section C-I chemists during the first half of January. He reviews the readings of the surveys and concludes that while they were higher during the Chicago work at Clinton, it must be remembered that at least four times as much plutonium was being handled then as had been handled in any previous work. Even so, the value was only about 25% of tolerance. He asserts that, during the operation procedures, the greatest possible precautions were taken and that the individual health precautions observed (gloves, face masks, and respirators) were more extensive than those of the regular Clinton personnel.

Gilbreath also mentions that the Clinton health group should have mentioned any situations considered hazardous at the time they occurred. Finally he points out that the man in charge of the room, Gevantman, had said that the Chicago group had discharged its duties as far as his laboratory was concerned and that the Clinton group would take care of final cleaning.

Manning and I sent a request to Daniels urging that a way be found to permit two of our seven office workers, Kay Florin and Dorothy Black, to work on an 8:30 a.m. to 5:30 p.m. schedule with one hour for lunch rather than the prescribed 8:30 to 5:12 schedule so that we will have some secretarial help after 5:12 p.m. The Time Schedule Committee has so far refused to grant an exception on the basis that it would lead to a flood of such requests which they could then not refuse.

At 4:00 p.m. I along with Doan, Daniels, Stone, and others attended a meeting in Stearns' office to discuss the matter of contamination of the room at Clinton during our Np^{237} recovery operation. It was generally agreed that if all the facts had been known no fuss would have been made. As it is, however, our honor has been impugned. Stone offered to write a letter distributing the blame for the observed high readings between the Clinton and the Chicago workers, but it was decided that such a letter definitely would not improve the situation. A letter of some sort, however, will be written to the Central File disagreeing with the Clinton accusation.

The top headline today reads, "Big Battle Raging on Iwo," only 750 miles south of Tokyo.

Wednesday, February 21, 1945

General Groves is in Chicago to discuss with Compton next year's research program as proposed for the Metallurgical Project.

At 8:30 a.m. I attended the Project Council Information Meeting on Chemistry along with Aebersold, Allen, Arnold, Boyd, Brown, Burton, Capt. Chapman, Compton, Cohn, Connick, Cooper, Coryell, Daniels, Dempster, Daane, Estermann, Franck, Fred, Hamilton, Howe, Huffman, Jesse, W. C. Johnson, Jeffries, Leverett, Manning, Mullikan, Nickson, Perlman, Rabinowitch, Shapiro, Sinclair, Stern, Stearns, Stone, Spedding, Sugarman, Szilard, S. Thompson, Turkevich, Voigt, Wakefield, Watson, Watters, Whitaker, Wigner, Young, and Zachariasen. When it came my time to speak, I reported first on the separations work. I said that flow-sheets are now available for the increased production at Hanford, that new data have been obtained on the separation of neptunium in the process, that Calvin's TFA reagent is very good for analysis of Pu(III) and (IV) mixtures, that several alternative solvent extraction processes have been studied including the "redox" process in which the plutonium is alternately oxidized and reduced, that the TFA process in recent tests has given much better decontamination than previously observed, and that the combined bismuth phosphate-hexone process and the direct decontamination by the action of hexone on the plutonium-fission product mixture shows promise.

I also reported that, in basic chemistry, new values have been obtained for the potential of neptunium couples and it has been confirmed that Np(V) is remarkably stable, that new neptunium compounds obtained are $NpCl_4$, $NpCl_3$, and (to which Zachariasen disagreed) a Np(V) fluoride whose formula may be analogous to $(NH_4)_2PaF_7$. I reported on the work on heavy isotopes, citing the correct value for the n,γ cross section of Np^{237} as 100 barns $\pm$20%, thus permitting us to estimate more definitely a figure of 1% as the increase in alpha-particle activity of the Hanford Pu^{239} by contamination with Pu^{238}. I reviewed the Geiger counter studies of the radiation from Np^{238} and the search for an alpha-particle decay of plutonium that could be attributed to the transformation of Pu^{241} to U^{237} (the 7-day beta-particle emitter).

I also described our finding alpha particles with ranges of 4.0 and 4.7 cm from Clinton and Hanford neutron-irradiated plutonium. Our interpretation is that these activities are attributable to 95^{241} and 96^{242}, respectively; I gave estimates of the cross sections involved in the various reactions.

In discussing the U^{233} work, I mentioned that 600 micrograms of Pa^{231} have been isolated and 150 micrograms purified and used for study of the basic chemistry of protactinium. The continuous and automatic countercurrent extraction column for the ether extraction of U^{233} from irradiated thorium was described, as was the work on magnesium and calcium nitrates as salting-out agents for the separation of uranium and thorium.

Items of interest presented by other speakers were: (1) Connick gave results of recent TFA studies at Berkeley — zirconium, columbium, and ruthenium are poorly decontaminated by its use. (2) Coryell described the large-scale production of radioactive barium (Ba^{140}) and lanthanum

(La^{140}) at Clinton. (3) Spedding reviewed the thorium production at Ames. (4) Shapiro of Clinton Labs reported on the production of tritium to the extent of 1.5 cc of 12.5% pure material. (5) Leverett reported that the Ba^{140} production building at Clinton is now 75% completed. (6) Burton talked about the gas evolution to be expected in conversion pile solutions — in uranyl solutions 2.7 water molecules will be destroyed per 100 ev. (7) Sugarman told of efforts to be made to obtain the neutron absorption cross section of U^{236} by irradiation at Argonne of Clinton neutron-bombarded uranium. (8) Watters has obtained evidence for the existence in solution of Np(III) from polarographic measurements. The potential for the Np(III)-Np(IV) couple is less than +0.1 volt.

Yesterday and today Morgan carried out a search for a Np^{238} daughter in an aliquot of a 95-96 fraction of sample 49NE-33.9 derived from Hanford neutron-irradiated plutonium (Fermi special sample). The purpose is to establish whether the 4.75 cm range alpha particles might be due to 95^{242}. A Np^{237} tracer was added to the sample, and five bromate oxidation cycles were carried out to isolate the neptunium fraction. He finds no evidence for the Np^{238} two-day half-life and concludes that 4.75 cm alpha-particle activity is not 95^{242} and that most probably 95^{242} is a beta-particle emitter of short half-life leading to 96^{242}.

At 7:45 p.m. I attended the meeting of the Separation Processes Sub-section of Section C-I in Room 209 of Eckhart Hall. Others present were Ader, Albaugh, Arnold, Asprey, Beard, Bradt, Cressman, Daniels, Davidson, Egan, Fields, Fineman, Florin, Gilbreath, Greenlee, Hagemann, Hopkins, Howland, Hyde, Hyman, James, Katzin, Kelley, Larson, Lawroski, Malm, Manning, E. Morgan, L. Morgan, Perlman, S. Peterson, Russell, Schaffner, Schraidt, Sedlet, Sheft, Templeton, R. Thompson, S. Thompson, Walling, Westrum, Winner, and others. I turned the meeting over to Albaugh who called first on Malm. Malm said that the results of laboratory investigations indicate that the Bismuth Phosphate Process may be modified to permit recovery of neptunium without adversely affecting plutonium recovery or decontamination. The major losses of neptunium occur in the extraction and plutonium precipitation steps, loss in extraction resulting from the stable +5 oxidation state which is not carried by bismuth phosphate. In an effort to improve neptunium carrying, various prereduction schemes have been attempted to reduce neptunium to a carryable +4 state. It is found that a prereduction treatment with $H_2C_2O_4$-Mn^{++} followed by addition of either fluosilicate or fluoride results in 80 to 98% carrying of neptunium. It is also found possible to achieve good carrying under these conditions even if $NaNO_2$ is retained in the prereduction step. Investigations of the plutonium precipitation step have centered largely around the effect of $SiF_6^{=}$ on neptunium carrying, which has been shown to increase carrying to 74-83%. These results suggest that the Bismuth Phosphate Process could be altered to permit the recovery of 50% of the neptunium without adversely affecting either the plutonium recovery or the decontamination.

Hyman reported on some preliminary laboratory work to determine the feasibility of employing a solvent extraction scheme to decontaminate the plutonium which would be used as the neutron source in the converter pile. The method tested was one analogous to the procedure used in the hexone concentration-isolation process and uses $Al(NO_3)_3$ as the salting agent. The procedure uses neutron-irradiated plutonium and consists of

a batch-wise simulation of the countercurrent extraction cycle. The gamma-ray activity that follows the plutonium through two hexone extractions and two water extractions appears to be due entirely to ruthenium.

Last night and early this morning Magnusson prepared samples of the neutron-irradiated Np^{237} containing Np^{238} (irradiated on Monday in the Argonne heavy water pile). Magnusson prepared some samples without any decontamination from fission products and some samples with one oxidation-reduction cycle of decontamination. Jaffey will use these samples to measure the decay of Np^{238}, to make beta particle and gamma-ray absorption measurements on Np^{238} and to measure the growth of Pu^{238} daughter alpha particles. Jaffey immediately began to measure the growth of alpha particles in the samples prepared for this purpose.

Battles are raging on Iwo and in Manila. The Germans have rejected the Soviets demand that they surrender Breslau, an important rail city on the Oder River.

Thursday, February 22, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Stan Thompson (still here for his visit), Cunningham, Ghiorso, Hagemann, Hindman, James, Jones, Katzin, La Chapelle, Larson, Manning, McLane, Morgan, O'Connor, Studier, Van Winkle, and later Florin and Jaffey. O'Connor told of progress made on the 20-mg Los Alamos sample [neutron-bombarded plutonium from Hanford that was sent here as Pu(VI) after being put through one decontamination cycle at Los Alamos] being decontaminated. He has 18.5 mg of it which gives a gamma-ray count slightly above a pure laboratory sample, using a 200 mg/cm^2 lead absorber. Greenlee will further purify a standard lab sample for comparison. Decontamination work will continue until the lowest level of gamma-ray activity is reached.

Van Winkle reported that the extraction of Pa^{231} (0.5 - 1 mg) is nearly finished; Larson has started a new extraction on 15 kg of carbonate residue.

The following activities were scheduled at this meeting: (1) Hufford-Florin-Seifert to make blanks (48 and 49) for Crawford; (2) Ghiorso to look for x-rays in a 6-mg Np^{237} sample that La Chapelle is to prepare by Monday; and (3) Florin will work up the uranium target bombarded with alpha particles at Berkeley.

Activities to be considered or scheduled at next week's meeting are: (1) the search for x-rays, by Ghiorso, in about 120 mg of U^{233}; (2) the next extraction of decay products from U^{233} (the neptunium series); (3) discussion of the results of the Berkeley uranium plus alpha-particle bombardment; and (4) Jaffey to report on the subject of Geiger counter coincidence corrections.

At Clinton Labs today, the 97.2 mg plutonium sample is to be discharged from the pile after receiving 58,000 kw days of irradiation

at 1.4 times the average flux. It was put into the pile almost a month ago on January 24.

U.S. marines are fighting for the second air field on Iwo; they already control one-third of the island and its most important air field.

Friday, February 23, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, changed from Wednesday when the Project Council Chemistry Information Meeting was held. It was attended by Albaugh, Asprey, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, and Roy Thompson. I remarked that writing on the Project Record is getting underway and that all those present at this meeting may expect writing assignments with tough time schedules in the near future. I noted that security violations (reported by regular guards) have been practically non-existent since institution of our section's own security patrol system. I said that John Rose of the Health Division reports that 56th Street outside Room 4 (Florin, James, and Morgan) exhibits considerable radioactivity. The condition represents no hazard but is interesting. It was brought to everyone's attention that Hamilton in Berkeley is looking for gram quantities of spectrographically pure bismuth, lead, and thallium to be used in looking for fission by the higher energy alpha particles (40 Mev) now available in the 60-inch cyclotron.

Another topic mentioned was that the Separation Processes Subsection has suggested that Site W extraction waste might be processed for neptunium recovery before routine daily production gets underway at Hanford. It could be looked upon by Hanford as simply a practice run.

Manning and I reported on the Physics and Chemistry Project Council Information Meetings we attended on Tuesday and Wednesday. During the review of what was said on cross section measurements, I interpolated some predictions concerning the n,γ cross sections of the heavy elements and suggested the following classification:

Few barns (one to ten)		*Many barns (~100)*	
(even-even)		*(even-odd)*	*(odd-odd)*
Io^{230} (predicted)		U^{233}	Pa^{231} (predicted)
U^{236}		U^{235}	Np^{237}
Th^{232}		Pu^{239}	95^{241} (perhaps)
U^{238}			
Pu^{240} (perhaps)			

I reported on the results of last Tuesday's meeting about the "Clinton accusation" and said that because of the attention focused by this accusation upon our laboratory techniques for handling radioactivity, we will be watched very closely by the Health Division and it behooves us to be particularly careful in our chemical operations and to clean up all contaminated surfaces and areas promptly.

It was stated that it has been found that plutonium-containing spray is much more dangerous upon inhalation than is an insoluble plutonium dust. The spray is almost quantitatively retained in the body, whereas the dust is, to a large measure, eliminated. It was also mentioned that Luther Arnold is disturbed by the fact that no favorable housekeeping committee reports have reached his desk for several months. We agreed that some sort of housekeeping inspection is desirable and that the old system has not proved very satisfactory.

A committee composed of Arnold, Manning, Watters, Nickson, and Cole has been formed to study the problem of obtaining remote control equipment needed in the future work of the Chemistry Division; it is hoped that some representative of the Chemistry Division will be able to go to Hanford to study the equipment there.

I announced that, in the future, Section C-I Council meetings will be held on Friday instead of Wednesday in those weeks in which the Project Information meetings are held.

James completed his experiments begun last Saturday to determine which of the rare earths are resembled most by elements 95 and 96. His findings are: (1) The scandium fraction contains less than 1% of the total alpha-particle activity. (2) The indium fraction contains 10% of the total alpha-particle activity. (3) The cerium group precipitate contains about 30% of the total alpha-particle activity. (4) The remaining 40% is retained in the soluble fraction containing the yttrium-group elements.

I talked by phone with English and Stoughton in Oak Ridge. English said that in connection with the problem of the polymeric form of plutonium(IV) in the isolation step at Site W, it turns out that the acidity must be greater than 0.5 N or one can have higher acidity and/or heating, e.g., heating with 1 N acidity is very good.

Stoughton told me that our sample SA-1 (49ND) and SA-2 (49ND) were in about equivalent positions in the Clinton pile — about 1.4 times the average flux. For our fast neutron irradiation we can use either a sample tube (low flux), which can be irradiated right away, or a special slug (high flux), which takes longer to go in and out of the pile; the net result is about equal. He suggests that Jones ask for the fast neutron irradiation (about 20 days) in terms of total energy. We then discussed the position in the pile of the irradiated samples.

War news was pushed from the top headline today because two congressmen had a fist fight on the floor of the House of Representatives. Congressman Rankin (Democrat of Mississippi) attacked Congressman Hook (Democrat of Michigan) after Hook called Rankin a liar and Rankin said Hook was mixed up with the Communist party.

Saturday, February 24, 1945

The sample of 97.2 mg of plutonium that has been irradiated in the Clinton pile for Roy Thompson's group was returned to the Met Lab today.

It has been irradiated from January 24 to February 22 and has received 58,000 kw-days at 1.4 times the average neutron flux.

Upon completing a further experiment on the tracer chemistry of elements 95 and 96, James finds that these two elements are not carried by thorium peroxide from 0.1 N HCl.

I sent a memo to Stearns about the possibility of Hanford undertaking two special jobs for us. The first is a special HEW plant run for the recovery of Np^{237}. Our proposal is that a special run be made using the extraction waste solution. The method we have devised is exceedingly simple and consists of throwing down a second bismuth phosphate precipitate from the extraction waste after treating the waste with a reducing agent, probably hydrazine. I said it can be looked upon as a practice run from the standpoint of the HEW management before the regular daily production batches start. This run would yield 15-25 mg of neptunium. Allison, Oppenheimer, Teller, and Zinn have all expressed a need for this isotope, a very convenient and efficient detector of fast neutrons because of its almost unique fission properties.

The second special job I requested is that we be provided with 50 to 100 ml samples of the HEW 40% UNH dissolver solution; the material is needed for study of decontamination methods, especially solvent extraction, and for the investigation and identification of new heavy isotopes and fission products. Shipping containers for the solution have been designed with the advice and help of Perlman.

Stearns relayed to Compton, Daniels' explanation of why the Chemistry Division cannot lend three grams of 94^{239} to Wigner and Zinn. Of the 7.5 grams in the Chemistry Division, a portion is in the form of slurries and most of the remainder is divided among seven different group leaders. There are 1.9 grams of pure material in "the bank," but one gram is 2 gt material and should not be mixed with other material.

Report CC-2636, "Chemical Research — Extraction and Properties of U^{233}, Report for Period Ending December 15, 1944," was issued and contained the following information:

Radiation spectrum of pure U^{233}. Studier has taken aluminum and lead absorption curves on a 16-mg sample of U^{233}. These indicate the presence of gamma-rays of 130, 230, and 420 kev energy and an abundance of from one to ten per million alpha particles. Lower energy radiations (90, 19, 14, and 8 kev) range in abundance from about two in 10^5 alpha particles to about one in ten alpha particles. Magnet bending experiments have demonstrated the presence of conversion electrons.

New half-life determination for U^{233}. Hyde, Hagemann, and Katzin have carried out a new half-life determination in which the specific activity was determined by an isotopic dilution alpha-particle counting method. The isotopic ratio of the uranium sample was determined mass spectrographically by a revised method in which the isotopic ratio (U^{233} to U^{238}) was brought to a value near unity. The isotopic purity of U^{233}, according to these latest measurements is about 97%, rather than 87.5%. The alpha-particle counting results indicate a longer half-life (than hitherto assumed), namely, 1.62×10^5 years.

Determination of the decay chain of U^{233}. Ghiorso, Hagemann, Katzin, and Studier have tentatively established the following identifications of members of the 4n+1 series: RdNp(Th^{229}), an alpha emitter of probably several thousand years half-life; NpX(Ra^{225}), an alpha emitter of about one month half-life; NpC(Bi^{213}), an alpha emitter of about 44-minute half-life; NpD(Pb^{209}), a beta emitter of 3.3-hour half-life, an activity which was previously known. When a sample of Ra^{225}(NpX) is allowed to stand in an alpha counter for several hours and then removed, the counter shows signs of contamination. This is undoubtedly due to active deposit from neptunium(Em^{221}). Attempts to determine the half-life of the emanation by following its growth in freshly purified NpX indicate that its half-life must be quite short. It is not possible to state whether the emanation is alpha or beta active. Attempts to isolate polonium or element 85 from solutions of NpX have been unsuccessful, indicating that any isotopes of these elements that are members of the chain are quite short-lived. The half-lives between NpX and NpC seem considerably shorter than ten minutes.

Extraction of U^{233}. Hagemann, Hellman, and Studier have found an unidentified organic compound that is either an impurity or some reaction product of diethyl ether which complexes uranium and prevents its transfer from ether to an aqueous phase.

Extraction solvents for uranium. Hyde and Wolf have investigated the usefulness of a number of solvents other than ether and hexone for extraction of uranium from thorium. Protactinium extraction has also been tested. In general, the cellosolve derivatives extract undesirably large amounts of thorium together with uranium. The ketones as a class show promise. Of the esters, ethyl acetate shows the most desirable characteristic of high uranium extraction and low thorium and protactinium extraction.

Extraction of HNO_3 and thorium by ether from thorium nitrate and ammonium nitrate solutions. Wolf and Hellman have studied these extractions as a function of total nitrate, thorium, and nitric acid concentration.

Influence of various nitrates on the extraction of uranium and thorium by ether. Hellman has found the most effective salting-out agent for uranium is aluminum nitrate. Salts, such as the nitrates of aluminum, lanthanum, magnesium, copper, lithium, and nitric acid, which are capable of affecting salting of thorium as well as uranium into ether, produce markedly greater uranium extraction if thorium is absent. Other salts that do not give appreciable thorium extraction show only minor differences in uranium extraction in the presence or absence of thorium.

Analysis of ore samples for protactinium. Q. Van Winkle and Sedlet have found that protactinium contained in the carbonate fractions from the uranium ore extraction process is in the acid insoluble silicious fraction of these residues.

The top headline today indicates there is a big drive on in Europe. U.S. troops have crossed the Roer River and are driving toward Cologne.

Sunday, February 25, 1945

The third Hanford plutonium manufacturing pile (Pile F) was placed in operation today.

Monday, February 26, 1945

At Berkeley, the 40 Mev alpha-particle bombardment of a uranium target, labeled TαB, ended today with 140 microampere hours. It started on the 18th of this month.

I received a letter written February 24 from Stoughton about his participation in the preparation of Project Volume 19A, "The Production and Separation of U^{233}," and the corresponding collected papers volume (19B). He suggests an additional topic for Volume 19B to the nine already decided upon, "Production of (small amounts of) U^{233} by separation of Pa^{233} from thorium after a short neutron bombardment of the latter, followed by decay of the Pa^{233} to U^{233}." He also indicates that in view of his past and current work he could be co-author of Volume 19B, chapters (1) "Discovery of U^{233}," (4) "Extraction Theory," (7) "Chemistry of Protactinium," and (10) the additional topic he has suggested.

In a memo to Daniels, Albaugh gave Research Problem Abstracts for the period February 1-15, for Sub-Section I, Section C-I.

Jones teletyped Stoughton asking that our sample for fast neutron irradiation be left in tube 23 in the Clinton pile for an indefinite period. He also said that whenever it is possible we will ask for fast neutron irradiations in energy units.

Edrey Smith Albaugh wrote to Truman Kohman at Hanford trying to help him identify and locate some of the figures attached to a document he had prepared on proposed modifications of BF_3 counters for operation at high pressures. She mentioned that Fred is quite fed up with Chicago's crime. He has had his overcoat stolen and his pocket picked, both within a month and a half.

Jesse talked with me about methods of separating thorium and uranium in connection with some investigations of radioactivity he is making. I suggested dissolving the sample and making an ether separation which would remove the uranium but not the thorium.

The U.S. army has captured Dueren and is only fifteen miles from Cologne, according to this morning's newspaper.

Tuesday, February 27, 1945

General Groves is in Chicago again.

The rare earth fraction from the remainder of the CW1-plutonium sample has been received from Site Y and turned over to Morgan. This corresponds to the 90 mg of semi-purified plutonium solution sent to us for further purification and production of metal to be returned to Los Alamos. Sample CW1 is the first "Fermi Special Sample" of plutonium irradiated in the Hanford reactor. Morgan designated the sample as 49NE-7 and began chemical operations to isolate the 95-96 fraction.

I received a teletype from Perlman at Hanford asking that we send Willard 2 mc of S^{35} for standardizing thin mica absorbers. He also asks for the latest figure for the specific activity of Pu^{239} — he and his co-workers are using 137,000 d/m per microgram.

I had a phone conversation with Stoughton at Site X about the $ThOCO_3$ cans in the Clinton pile. He said 15 cans went into a good position (1.4 times average flux) on November 10 for the Canadians, another 15 cans in a similarly good position on December 3, and 14 cans in a poor position (0.3 to 0.4 times average flux) on December 8. Only these 44 cans out of 80 passed the heating test at 230°C needed for the good position. Stoughton thinks only half of our present 125 cans will past the test. He will make this test on all our cans in order to have some ready (at least 15 are necessary for a whole replacement) in case the Canadians call for five cans to get Pa^{233} (as Watson told Katzin). I told Stoughton about the Canadians' primary purpose to get Pa^{233} [preferred material would be freshly discharged after three months irradiation] and that we might get some of their more strongly irradiated cans provided we can keep them supplied with Pa^{233}.

Manning received a memo dated February 26 from J. A. O'Hearn, Finance Director, complaining that Kay Florin is punching her time card incorrectly, making the numbers illegible.

Arnold asked Wilkinson by memo that the decontamination squad be assigned responsibility for decontaminating Room 212 in Kent Chemical Laboratory so it can be returned in proper condition to the University. The room was used by my section one and a half or two years ago.

Mulliken issued a "Preliminary Outline of Metallurgical Project Record," listing the titles and editorial committee for each of the 20 technical volumes and the four supplemental volumes: "Project Handbook, Project History, Patents Abstract, and the Index and List of C Reports." I am listed as being on the editorial committee for Volumes 9A and 9B, "Fission Products and Radiochemistry," (editors Coryell and Sugarman) and as the co-editor (with Manning) of Volumes 14A and 14B, "Chemistry of Transuranium Elements," and (with Katzin and Stoughton) Volumes 17A and 17B, "Production and Separation of U^{233}." The number designation of these and the other volumes have been undergoing change, but have now, presumably, assumed their final form.

The Allies are moving on all fronts in Europe, and U.S. heavy bombers and fighters battered Berlin in a daylight raid yesterday, reported this morning's paper.

Wednesday, February 28, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. I announced that our security patrol has cut down the violations to one last week and one this week; general satisfaction was expressed with the new patrol method.

There was a discussion brought up by Albaugh about maintenance of the Geiger and alpha-particle counters. Of late they have not been available for use because they are not in working order. Jaffey asked early notification to his group when a counter is going bad. He pointed out that it would be desirable if everybody learned enough about the instruments to detect early malfunction; such knowledge would also help the individual to assess the reliability of his data. I mentioned there is one problem we have not considered and that is, suppose men leave here and we write them recommendations for work in radiochemistry. They will probably be expected to know a fair amount about instruments, yet most of them do not. After discussion it was decided there would be three meetings next Wednesday, each attended by one-third of the section for demonstrations by the instrument people.

Jones announced that badges will be marked with a scarlet streak to identify those who can enter the filtered air section during the day. A second scarlet streak will show those who can enter at night as well. Manning told us that two hot labs are being constructed, one for the heavy isotope boys in Room 4 and another in one of Albaugh's rooms. I mentioned having written last Saturday to Stearns requesting some dissolver solution from Hanford and a Np^{237} run on waste solution.

I received a copy of a letter dated February 24 from Kennedy at Site Y to Lavender, referring to the agreement between the inventors involved in Case 52 and Segrè. Kennedy points out that there is nothing formal or even in writing about this agreement, but that several months ago the four persons involved as inventors on Cases 52 and 61 reached an informal agreement to act together in any disposition that might be reached.

I wrote Whitaker about the irradiation of thorium for Evergreen and the Metallurgical Project, passing on the information from Watson that the Canadians, around the middle of March, intend to ask for four or five of the thorium cans that have been in the Clinton pile since last November or December. I point out that their interest is primarily in Pa^{233} and hence they would prefer freshly-discharged material with three months of irradiation. I suggested that since we have a large number of needs for U^{233}, the remaining 10 or 11 cans from the particular stringer should be processed either here or at Clinton, and new cans be put in the pile later to supply the Evergreen people with an additional source of Pa^{233} and to keep up a continuous manufacture of U^{233} for the Metallurgical Project.

I teletyped Perlman at Site W in answer to his teletype of yesterday saying that the disintegration rate he is using for plutonium (137,000 d/m per microgram) is about our best estimate. I indicate we will send Willard the S^{35} he desires.

In a memo to Daniels, Katzin gives the Program Abstracts for February for Group 9, Section C-I.

Manning sent a memo to Daniels listing the activities of Section C-I in two categories — those directly related to the development of new piles and those that are indirectly related to new piles. Under the first category are shown (1) Study of the chemical and nuclear properties of heavy isotopes [7 men]. (2) U^{233} and related problems [5 men]. (3) Extraction of protactinium from ores and study of its chemistry [2 men]. (4) Decontamination of plutonium from its fission products [2 men]. (5) Volatilization at high temperature of plutonium and fission products from uranium carbide [1 man]. (6) Colloidal behavior of plutonium and its fission products [1 man]. In the second category are (1) Solvent extraction methods for separation and isolation of plutonium from uranium and its fission products [17 men]. (2) Basic chemistry of plutonium [8 men]. (3) Basic chemistry of uranium [1 man]. (4) Maintenance of electronic instruments [5 men]. (5) Recovery of laboratory plutonium for re-use [5 men]. (6) Chemistry of neptunium [4 men]. (7) Chemistry of actinium [1 man].

In a memo to Stearns, Daniels discusses the desirability of a closer relationship among the programs of different divisions, particularly of the Physics and Chemistry Divisions. He also suggests a group or committee be established to give advice to the divisions in the planning of their research programs. He encourages finding a way to permit free and vigorous development of new ideas to cut across divisional lines.

"Metallurgical Laboratory, Report for February 1945," (MUC-JCS-209) was issued. The summary section includes the statement, "Most of the research and development work of the Laboratory is exploratory work in connection with breeder piles. Consideration is being given to the homogeneous thermal breeder and the heterogeneous resonance and high energy breeder type of pile." The activities of Section C-I described in the report are excerpts of Reports CN-2688, "Chemical Research — Separation Processes for Plutonium," and CN-2689, "Chemical Research — Basic Chemistry of Plutonium," which will be issued sometime next week.

Compton received a registered letter from General Groves in Washington. I think it was somewhat of a blow to Compton, for this is what Groves wrote:

> I have given careful consideration to your proposed research program for the year 1946 which we discussed during my last visit to Chicago.
>
> Since the basic consideration for any work performed under the direction of the Manhattan District must be winning the present war, it is necessary that the efforts of the District not be diverted in any way to post-war problems. The Military Policy Committee has concurred in these views. The Committee is also of the opinion it should not assume responsibility for the post-war period.
>
> I am in complete agreement with the recommendation that some commercial firm be found to take over the responsibility

for the operation of the Clinton Laboratories. As you point out, they must be continued in operation for the production of vital materials and to carry on research development essential to the solution of recurring problems.

The work at the University of Chicago and at the Argonne must be restricted to three items:

a. The solution of problems involved in the operation of the Hanford Engineer Works.
b. The carrying of such work as may be desirable for the benefit of the Los Alamos project.
c. Such research as is necessary to determine the value of thorium including the possibilities of its use practically.

The work under paragraph c above must be confined to research and not extended to extensive developmental engineering until our knowledge of the problems involved is much greater than it is now or can be in the months to come.

At 7:45 p.m. I attended a meeting in Room 209, Eckhart Hall, of the Basic Chemistry, Recovery, and Instruments Group of Section C-I. Others present were Abraham, Ader, Albaugh, Asprey, Beard, Blaedel, Crawford, Cunningham, Davidson, Dixon, Dorsey, Florin, Fried, Ghiorso, Gilbreath, Greenlee, Hagemann, Hellman, Hindman, Hufford, Hyde, Hyman, Jaffey, Jones, Katzin, Krueger, La Chapelle, Larson, Lawroski, Magnusson, Manning, McLane, Morgan, Nickson, O'Connor, S. Peterson, Phipps, Post, Robinson, Sheft, Simpson, Stewart, Studier, R. Thompson, Van Winkle, Weissbourd, Westrum, Winner, Wolf, and others. As usual I opened the meeting by turning it over to Cunningham who called on the various speakers. Weissbourd spoke on the pulse height analyzer used to determine energies of alpha particles and to detect and determine quantities of isotopes. He described the 24-channel analyzer which has been constructed. This uses an ionization chamber with a collimator and absorber (to shorten the range) and hence has a low geometry factor (about 0.1 to 0.5%); it is arranged so that register no. 1 counts all pulses, register no. 2 counts all pulses except those falling in the interval corresponding to the difference in the grid bias of channel no. 1 and channel no. 2, etc. In the discussion that followed I commented that it would be interesting to run Np^{237} since it might be expected that alpha particles of two different, but close together, ranges would be found if the x-rays and gamma-rays discovered by Ghiorso are connected with the decay of Np^{237}. I added that such a measurement would be a good test of the resolution of the instrument. Ghiorso said that the only curve on Np^{237} so far obtained with the instrument shows a broader peak than on most isotopes and indicates that such a resolution of the two energy components should be possible.

Crawford reported on range measurements with a differential range chamber on neutron-irradiated samples of plutonium, opening with a description of the differential range chamber. Data on early Clinton material (below 1 gt) indicate that it consists of less than 2 parts in 10^8 of Pu^{238}. The 2 gt material now available shows the presence of 12 parts in 10^8 of Pu^{238}. Another sample of 2 gt material shows no increase

in the Pu^{238} hump when very strongly bombarded in the graphite of the Clinton pile, indicating this isotope is not produced by reaction of slow neutrons on Pu^{239}. Measurements on another 2 gt sample that received an additional 120 gt of bombardment in the graphite at Site W show some increase but not very much. This could be accounted for by traces of Np^{237} remaining in the Pu^{239}. I said it could also be caused by a fast neutron reaction on Pu^{239}. I mentioned that if the Hanford sample had been bombarded inside a slug, the Pu^{238} yield would probably be 25-50 times greater than when the sample was bombarded in its actual position in the graphite. A bombardment of a plutonium sample placed next to a slug at Clinton is planned.

Crawford said there is no difference in the alpha-particle range curves between the 2 gt material and the 120 gt material ascribable to Pu^{240}. He also gave values for the half-life of Pu^{240} for various values of range as obtained from the 4n Geiger-Nuttal curve that show the half-life going from 27×10^6 years for a range of 3.0 cm to 3350 years for a range of 3.5 cm.

Cunningham commented that he and Florin have estimated the half-life of Pu^{240} at greater than 3000 years since the specific activity of the plutonium fluoride produced from this 120 gt sample does not differ by more than 5% from that of cyclotron-produced, or low gt pile-produced material. In answer to my question, he emphasized that this is a lower limit.

O'Connor spoke on the preparation of the Los Alamos sample, for which there were two requirements: first, production of the metal [Westrum] and second, complete decontamination. This is the 120 gt material (first Fermi special sample CW1), part of which was used by Crawford for his alpha-particle energy measurements. The sample came in two sections: 25 mg (James' sample 49NE), from which James and Morgan removed the rare earths, and 90 mg which was sent from Los Alamos as the +6 nitrate. Of the 90 mg sample, 70 mg were given to Florin for preparation of the trifluoride to be used in metal production by Westrum.

O'Connor then described the purification of the 17.2 mg sample received from James and Morgan: two lanthanum fluoride cycles, five peroxide precipitations, one hexone extraction, one ether extraction, and one iodate precipitation. The final product was 16 mg of plutonium (out of 17.2 mg received by him). At this point the sample was combined with the other fractions, and the entire sample put through three peroxide precipitations, one iodate precipitation, and two hexone extractions, reducing the beta particle and gamma-ray activity to essentially basic plutonium levels.

In connection with a discussion of decontamination procedures, I observed that when decontamination is the object, the use of hexone extraction just because of the high recovery which can be experienced is like looking under the street lamp for a dime that was lost way out in the dark because there is more light under the lamp. I added that ether extraction is very specific since only such ions as uranyl, neptunyl, plutonyl, and possibly ruthenium are extracted.

O'Connor then gave the following as the procedure through which the remaining 120 gt material was run: hexone extraction, sodium plutonyl

acetate precipitation, lanthanum fluoride cycle, neptunium separation through hypochlorite oxidation. This represents the present state of affairs. Complete gamma-ray absorption curves are proposed to see whether or not there exists a gamma-ray attributable to Pu^{240}.

Westrum described the preparation of plutonium metal from the remainder of the sample. Starting material was 69.5 mg of plutonium prepared as the trifluoride by Florin. This was reduced in eight runs of 8-12 mg per run, using beryllia crucibles and barium as the reductant. The result was 60.4 mg of metal (88% yield) with each reduction yielding one large piece of metal. I commended Westrum for this piece of work, pointing out it was a job they were afraid to tackle at Site Y. I added that the metal will be used at Site Y to determine its Pu^{240} content by mass spectrographic examination; its Pu^{240} content has been estimated at 0.7% on the basis of spontaneous fission counts.

In answer to Post's question as to how much Pu^{240} could be tolerated in purified plutonium for Los Alamos, I said they have no choice in the matter; whatever is produced must be tolerated. I added that on the basis of the original purification program only about 0.001% could have been tolerated on account of its neutron emission.

February ends with the war progressing well on all fronts for the Allies.

Thursday, March 1, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Cunningham, Florin, Ghiorso, Hagemann, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Manning, McLane, Morgan, Studier, Van Winkle, and later O'Connor. Jaffey talked about coincidence corrections. Then Florin reported on his proposed plan of treatment of the $U + He^4$ Berkeley target TαB. (The target received 140 microampere-hours of 40 Mev alpha particles, 85% between February 18 and February 26.) One purpose of the bombardment is to look for Pu^{240} produced by the reaction $U^{238}(\alpha,2n)Pu^{240}$. To minimize the amount of isotopes produced by the reaction of low energy alpha particles, the surface layer will be milled off. The shavings will be dissolved in nitric acid, and the 93-94 and rare earth fractions will be separated from the uranium by lanthanum fluoride precipitation from SO_2-reduced solutions. After oxidation the rare earth fraction will be removed, reprecipitated, and given to James and Morgan to look for 95 and 96. The supernatant will be put through three oxidation-reduction cycles (using silver-persulfate-permanganate, and SO_2). Then the 93 will be removed by bromate oxidation cycles and a final lanthanum fluoride precipitation made to carry the purified 94.

The following activities are scheduled: (a) the next run on 23 decay products will be made Monday, March 12; (b) a check run will be made for U^{233} gamma-rays and x-rays on 60 mg of U^{233}; (c) La Chapelle will prepare a sample of 100 micrograms of Np^{237} for Zinn; (d) a run will be made on the $Np^{237}n,\gamma$ reaction with cadmium on Thursday, March 8 or Monday, March 12.

Activities scheduled for next week's meeting: (a) Florin to describe progress on the TαB target ($U + He^4$-Berkeley); (b) Ghiorso, O'Connor, and others to give the latest information on x- and gamma-rays of Pu^{239}, U^{233}, and Np^{237}; (c) James will talk on x-rays of heavy elements. For later scheduling: study of n,γ reaction on ionium and Pa^{231}.

I read a copy of a memo from Zachariasen to Stearns giving the results of analysis of a sample submitted by Fried that shows the presence of UCl_6, "with the six chlorines around each uranium forming an octahedron." He indicates there is little hope of establishing the exact chlorine positions unless crystals of UCl_6 can be investigated.

In a teletype to Perlman at Site W, I give results of analyses by Zachariasen of six samples he submitted for analysis: Sample 1 is not characterized, but it is not lanthanum fluoride, $La(OH)_3$, or $KLaF_4$. Samples 2, 3, 5, and 6 are crystalline lanthanum fluoride, and sample 4 is crystalline $La(OH)_3$. I say this looks favorable and ask if he wishes any additional information.

Jones sent transfer forms to Furney for 61.5 mg of U^{233} as oxide to Site Y. Jones indicates that the material is ready for shipment.

In a memo to Kohman at Site W, Jaffey sent data on the effect of

self-absorption on the counting of alpha particles.

The summary of the manpower distribution for the Chemistry Division that Hogness issued today shows the following for my section:

		No. of Men Jan.	No. of Men Feb.
Albaugh (Separation Processes, 27 men)	R. Thompson, extraction and decontamination	6	6
	Gilbreath, process development	8	7
	Lawroski, solvent extraction	14	13
Cunningham (Basic Chemistry, 36 men)	Simpson, high vacuum work	9	9
	Hindman, basic chemistry	12	11
	Stewart, recovery	6	6
	Ghiorso, instruments and physical measurements	10	9
Katzin (23 work, 8 men)	23 work	7	7
Administration	Seaborg, Manning, Jones, Albaugh, Cunningham, Katzin	6	8
		78	74

In today's paper war summaries say that U.S. troops have routed the Germans near Cologne and that the Japanese are still fighting savagely on Iwo Island.

Friday, March 2, 1945

General Groves and Conant are in Chicago.

Wigner commented, in a memo to me dated February 28, on data we have given him on the solubility of plutonium peroxide. He notes the greater solubility of plutonium peroxide in nitric acid as compared with sulfuric acid and asks about the possible use of small quantities of zirconium to stabilize the plutonium solution.

I received a March 1 memo from Dempster giving the result of the mass spectrometer comparison of the abundance of U^{238} in the 23 sample kept for this purpose. He finds a value for U^{233} of 95.8 ± 0.5 percent. This is in agreement with two earlier approximate values.

Hamilton sent me a teletype from Berkeley to notify us that the target from the uranium plus 40 Mev helium ion bombardment [Met Lab sample TαB] has left Berkeley for Chicago by train with a courier. He gives the bombardment details (140 microampere-hours, with 85% received between February 18 and February 26) and mentions that by measuring the alpha-particle beam at the end of the bombardment he deduces that the extent of deuteron contamination to the target is less than 5% of the alpha-particle beam.

Today is Helen's birthday.

U.S. troops have captured the Ruhr city of Muenchen-Gladbach, largest German center to fall on either European front.

Saturday, March 3, 1945

Betty Mokstad was hired today to work as a technician for Davidson.

I received a letter dated February 27 from Perlman at Site W giving me the results of measurements made by Willard's group on the hard gamma-radiation associated with plutonium, using 25 gt Hanford material and 4 gt Clinton material. He says that measurements by Kohman and Sullivan indicate no difference between the two samples.

I read a copy of a March 2 memo from Daniels to Wigner summarizing the present research program in the Chemistry Division related to homogeneous water piles, listing the following investigations and manpower involved: (1) Chemical and nuclear properties of heavy isotopes — 7 men. (2) U^{233} — 4 men. (3) Decontamination of plutonium in absence of uranium — 3 men. (4) Colloidal behavior — 2 men. (5) Fission products — 4 men. (6) Decomposition of aqueous solutions by radiation — 6 men. (7) Solvent extraction applied to high energy piles — 4 men. (8) Protactinium — 2 men. Total, 32 men.

In a memo to Stearns I ask him to procure for us about one gram of Y-12 beta product, i.e., material that contains about 75% of U^{235} by weight, indicating we want the material for a number of purposes: (1) to extend our study of the method of analysis for U^{235} involving the counting of UY beta particles; (2) for bombardment with deuterons and alpha particles to find new isotopes of elements 93 and 94, especially the former for use as a tracer for element 93, and possibly to make pure U^{236} via Np^{236}; (3) to study in detail the alpha-particle radiation and possibly gamma- and x-radiation from the isotope U^{235} itself; and (4) for bombardment with pile neutrons to form U^{236} for the purpose of studying its radiation properties and neutron absorption properties. I also take the occasion to mention our interest in a special Y-12 beta process run that will result in a sample with a much higher proportion of the U^{234} isotope than is the case for their usual product. We would use this product in order to do, with isotope U^{234}, experiments similar to those listed for isotope U^{235}.

I sent a memo to Wigner agreeing with the memo I received from him yesterday about the relative solubility of plutonium peroxide in HNO_3 and H_2SO_4. In response to his questions, I also mention the tendency of zirconium ion towards hydrolysis. This could interfere with its usefulness in stabilizing plutonium solutions with respect to precipitations of plutonium peroxide.

Radiation surveys taken in Section C-I by the Health Division for the week ending today show 13 high beta-particle hand counts, of which nine are in Roy Thompson's group, two in Lawroski's group, and one

in Hindman's group. A general radiation survey completed today of rooms formerly used in Jones and Kent show potential health hazards in Kent 101, 104, 109, 112, and 301; Jones 401, 403, 404, and the attic.

Report CN-2689, "Chemical Research — Basic Chemistry of Plutonium. Report for Period January 1 - February 15, 1945," was issued. It summarizes our investigations in Section C-I as follows:

Basic Dry Chemistry Group (Davidson, Assistant Group Leader). *Preparation of NpF_3 and NpF_4.* Fried, Florin, and Davidson have prepared NpF_3 by reacting NpO_2 with H_2 and HF at 500°C and NpF_4 by reacting the NpF_3 with O_2 and HF at 500°C. The NpF_3 compound is black, and the NpF_4 is light green. *Preparation of $NpCl_4$ and $NpCl_3$.* Fried and Davidson have prepared $NpCl_4$ by the reaction of CCl_4 vapor upon Np(IV) oxalate at 500°C, the $NpCl_4$ forming as a yellow sublimate. Hydrogen reduction of $NpCl_4$ at 450°C gives $NpCl_3$. *Preparation of NpOS.* Fried and Davidson have reacted NpO_2 with an H_2S-CS_2 mixture at 1000°C for two hours and obtained NpOS. It is not possible, however, to prepare NpS_2 or Np_2S_3 by prolonged treatment under these conditions. *Attempted preparation of a higher oxide of neptunium.* Fried and Davidson have found that ignition of Np(IV) in 28 atmospheres of O_2 at 400°C gives rise to NpO_2 and not to a higher oxide.

Micromanipulation of solids. Fried and Davidson describe some of the techniques with particular attention to the preparation of samples for x-ray analysis.

Survey of dry chemistry of uranium, neptunium, and plutonium. Davidson summarizes all available information as to the existence and stability of uranium, neptunium, and plutonium compounds of various oxidation states.

Preparation of Pu_2S_3 by treatment of $PuCl_3$ with H_2S. Abraham and Davidson have carried out this reaction successfully at 800°-900°C. *Equilibrium in the vapor phase hydrolysis of PuOCl.* Abraham and Davidson have measured the equilibrium constants in the temperature range of 500°-650°C for the reaction $PuO_2 + \frac{1}{2} H_2 + 3HCl = PuCl_3 + 2H_2O$. Analysis of the data gives the free energy equation delta F(kcal) = -8.5 + 9.5 T/1000.

Basic Wet Chemistry (Hindman, Group Leader). *Fluoride complex of Pu(IV).* McLane reports that from a study of the absorption spectra of Pu(IV) in 1 M HNO_3 containing various amounts of HF, it is found that the value of the dissociation constant for the reaction $PuF^{+++} = Pu^{++} + F^-$ is about 10^{-7}. Preliminary evidence has been obtained for the formation of PuF_2^{++} and PuF_3^{+} in the presence of higher concentration of fluoride. *Solubilities of some compounds of Pu(VI).* Dixon has made a series of solubility measurements on Pu(VI) compounds of the following alkaline earths precipitated from basic solution: calcium plutonate in ammonia; barium plutonate in ammonia, in NaOH-NH_3 mixtures and in NaOH; and magnesium plutonate in ammonia.

Search for U^{237} in Clinton plutonium. James has sought evidence for the presence of Pu^{241} in 100 mg of 10 gt Clinton-produced plutonium on the basis that it might decay by alpha-particle emission to give 6.8-day beta-particle emitting U^{237}. The material was separated into a plutonium fraction and a uranium fraction (examined for the beta-particle

emitter). The two basic steps in the fractionation are (1) precipitation of the plutonium as the fluoride, first by itself and later with lanthanum fluoride, while keeping the uranium as soluble UO^{++}, (2) precipitation of the uranium with lanthanum fluoride after reduction to U(IV) by Ti(III) in H_2SO_4. Step 1 was repeated seven times on the uranium fraction in order to achieve complete separation of the plutonium from the uranium fraction. At the end of these operations, the uranium fraction contained less than 20 beta-particle counts per minute and shows no evidence of decay. When one assumes the slow neutron cross section for the reaction $Pu^{239}(n,\gamma)Pu^{240}$ to be 400 barns, separation of the uranium fraction has shown that the slow neutron cross section for the reaction $Pu^{240}(n,\gamma)Pu^{241}$ is of the order of 200 barns or less if Pu^{241} decays by alpha-particle emission to U^{237} with a half-life of 10^6 years. It is considered probable that Pu^{241} decays chiefly by beta, rather than alpha-particle emission.

Recovery Group (Stewart, Group Leader). *Recovery of plutonium.* Anderson, Asprey, Britain, Fields, Fineman, and Stewart have recovered the plutonium from the large volumes of aqueous solution resulting from the three solvent extraction runs on the three-inch column. The method consists of co-precipitation with lanthanum hydroxide, dissolution in nitric acid, salting-out with $Al(NO_3)_3$, and batch extraction with hexone.

Research methods of plutonium recovery. Asprey, Anderson, and Stewart have carried out preliminary successful tests of extracting plutonium by hexone from approximately 10 M HCl containing bismuth phosphate. The use of bismuth phosphate instead of lanthanum fluoride as the standard carrier in recovery work would permit the elimination of waxed beakers, pipettes, etc. *Routine separation of 93^{239}. Preparation from depleted oxide.* Fields has investigated the use of U^{235}-depleted uranium for the preparation of Np^{239}. He finds that the Np^{239} from the neutron-irradiated depleted oxide has one-tenth the fission product activity of that from naturally occurring uranium.

This morning's paper, under a March 2nd dateline, says U.S. troops were shelling Dusseldorf and have seized almost all of the vital Ruhr basin west of the Rhine River.

Sunday, March 4, 1945

Under the March 4th dateline an interesting item appears in the paper saying that Suribachi volcano on the southern tip of Iwo Island "acted up a bit today." As a result U.S. marines are having hot rations for the first time in several days. GI's shoved their cans of rations into cracks in the earth, and the rations were hot in fifteen minutes.

Monday, March 5, 1945

Betty Murray transferred from the position of typist to the position of technician for Lawroski. She will receive an increase of $0.75 per week.

James began an experiment today to check the yield (and power dependence of yield) of 95 and 96 using a sample (49NF) weaker than 49NC, 49ND, and 49NE. As starting material he chose 30 mg of the 97.2 mg plutonium sample that was bombarded by Roy Thompson's group in the Clinton pile from January 24 to February 22 this year, receiving a total of 58,000 kw-days at 1.4 times the average flux.

The target from the uranium plus 40 Mev helium ions Berkeley bombardment (Met Lab sample TαB) arrived in Chicago by courier.

In a letter to General Groves, Compton gives his own attitude regarding the political use of our new developments for the safety and welfare of the nation. He states that "our greatest concern is lest our major allies may, in the postwar period, proceed secretly with extensive military preparations" and urges the securing of "advantageous agreements at this time when we are entering upon a period of dominant strength." Otherwise, he believes, there could easily result an international race to a disastrous end.

Compton sent another letter to General Groves for the attention of the Secretary of War, wherein he stresses the importance of research in nuclear science as essential to the Nation's continued safety. He also asserts that if no funds to support longer-range atomic problems are available before the close of this fiscal year, irreparable damage to the atomic research program will result from the dispersal of the scientific staff.

Under yesterday's dateline comes news through London that the Soviet army has reached the Baltic coast near Kalberg, Poland.

Tuesday, March 6, 1945

At Berkeley the 40 Mev alpha-particle bombardment of 100 mg of Pu^{239} is scheduled to begin today. (The Met Lab sample designation is 49αB.)

Report CS-2741, "Chemistry Division Summary Report for February 1945," was issued. Essentially all Section C-I information in this report appears in the summaries of work prepared by Albaugh (February 19), Cunningham (February 19), and Katzin (February 17) for my use in connection with the February 21 Project Council Information Meeting on Chemistry.

James transmitted to me tables of x-ray lines emitted by elements 92 to 96 and their absorption coefficients. The tables are part of the calculations made with the object of identification of these elements by critical absorption of their x-rays. They will later be incorporated into a complete report.

I wrote to Allison at Los Alamos telling him we will soon send the second fraction of sample CW1B which was to be completely decontaminated (work done by O'Connor). The sample contains about 14.3 mg of plutonium

in the form of solid, moist peroxide. I give him our values for the energies of the hard radiation from plutonium and ask to be informed of values to be obtained in experiments at Site Y with this material. I say our values are 0.40 Mev (3.04 gm/cm^2 lead), 0.175 Mev (0.46 gm/cm^2 lead), and 0.1 or 0.6 Mev (0.12 gm/cm^2 lead).

I acknowledge receipt of the rare earth fraction from the remainder of sample CW1 and say we have found about 200,000 c/m of the alpha-particle activity in it which I told him about previously. I say we would also like to receive the rare earth fraction of CW2 in addition to the 25 mg we are scheduled to receive. I indicate that our yield of metal in the other fraction of sample CW1B that we already have sent him was rather high and amounted to about 88%. I then ask for his best values for the spontaneous fission rates of Pu^{240} and Pu^{238}.

I say we have sent 61.5 mg of isotope U^{233} that is 95.5% U^{233} and 4.5% U^{238} (thus 2.8 mg U^{238}). I discuss our plans to take another more careful look at the possibility that the Th^{229} daughter of U^{233} undergoes decay by beta-particle branching; at that time we should have more to say about his interest in the possibility of analysis for U^{233} content in these samples by means of beta-particle counting. I suggest we may have a better solution to this problem through some Clinton neutron-bombarded thorium carbonate that was completely purified of its natural uranium impurity before bombardment. We are now ready to extract the U^{233} and will make an accurate determination of its half-life.

I quote our measurement of a thermal neutron cross section of 100 barns for the n,γ reaction on Np^{237} and mention that the result is somewhat uncertain until we learn more about the decay scheme of Np^{238} which is turning out to be quite complicated. I mention our plans to measure the thermal neutron cross section for the n,γ reactions on Pa^{231} and ionium. I suggest that we could probably determine the cross section of the reaction $U^{236}(n,\gamma)U^{237}$ by measuring the amount of Np^{237} formed in a strongly irradiated sample of highly enriched U^{235}. I ask whether or not Fermi is irradiating some samples of 75% U^{235} along with his plutonium samples and offer to make the measurement if we could have about 25 mg of such a sample. I then say we would also have a look at it for U^{236} alpha particles and for any other unusual alpha-particle emitters, such as rare earths. This would be a useful complement to the similar work we are doing on the neutron-irradiated plutonium samples. I also observe that the 93 fraction from the main portion of such irradiated U^{235} would be worthwhile looking into for Np^{237}.

In a letter to Perlman at Site W, I compare the results of measurements of hard radiations from plutonium as reported in his letter to me that I received on Saturday with measurements made by Ghiorso on 1 gt Clinton plutonium and on Fermi's Special No. 1 (roughly equivalent to about 100 gt material). Ghiorso's results on both samples agree quite well with those obtained by Kohman and Sullivan.

I wrote to Dr. Nickson about the unusually high body contamination of Roy Thompson, his lab coats and shirts, reported for the period February 26 to March 5. I explain that the contamination occurred in connection with work being done upon a sample containing 100 mg of plutonium bombarded for 20 days in the Clinton pile (30 mg-portion used

by James as sample 49NF). Because of the presence of fluoride ions, it was necessary to evaporate in open platinum dishes covered with watch glasses; moreover, adequate remote control tongs, etc., were not available. I point out that the total counts accumulated upon the person and clothing of Thompson did not exceed 100,000 counts, considerably less than one one-millionth of the total activity of the sample. Contamination was confined largely to the area about the wrist (i.e., the area between the gloves and lab coat sleeves), the situation being aggravated by the shrunken state of laboratory coats and the unusual length of Thompson's arms. In conclusion, I observed that this experience has demonstrated anew the need for a hot laboratory designed to facilitate operations such as these.

Hufford described by memo for Jaffey the extraction of S^{35} for Kohman from concentrated NH_4Cl and KCl solutions.

Report CN-2688, "Chemical Research — Separation Processes for Plutonium. Report for Period January 1 - February 15, 1945" (Seaborg, Section Chief; Manning, Associate Section Chief; Albaugh, Assistant Section Chief) was issued. It contains the following report of our investigations.

Extraction-Decontamination (R. Thompson, Group Leader). *Methods of increasing the rate of metal processing.* S. Peterson and Winner have completed development of a method for processing four times the normal quantity of uranium per day in a single canyon at Hanford. Two 1500-ml scale runs have been made using optimum conditions to test the overall process. Decontamination and plutonium losses are found to be completely satisfactory.

Effect of storage on process metal solutions. Bartell finds that Pu(III) persists in preextraction solutions stored with 0.001 M N_2H_4 for 20 days at room temperature even after adding phosphoric acid and heating. Partial reduction of Pu(VI) may be brought about by prolonged heating at 75°C with H_2SO_4.

The influence of hydrazine in bismuth phosphate extraction. Ader, Bartell, Greenlee, Hoekstra, Malm, Morgan, and R. Thompson have prepared a summary report, including data from CN-2085, -2288, -2432, and -2496, on the work done on the hydrazine problem. It is concluded that the inhibitory effect of N_2H_2 upon carrying by bismuth phosphate in the extraction step is caused in large measure, if not entirely, by its action in reducing plutonium to the III state. By far the most promising method of N_2H_2 destruction is that involving prolonged heating, a procedure particularly applicable to plant operations since the 40% UNH solution is to be stored for from 5 to 10 days and will probably become very hot because of energy release from radioactive decay.

Decontamination of plutonium by precipitation (converter pile). Hopkins has conducted scouting tests on a 50 to 100 microliter scale using Hanford neutron-bombarded plutonium to study one-step precipitation of plutonium compounds from solutions of plutonium (ca 1 g/l) and fission products of plutonium. Only moderate decontamination has been achieved. The combination of a scavenger precipitation, such as lanthanum fluoride from oxidized solution, followed by the precipitation of a plutonium compound, has given decontamination factors of about 100.

Concentration-Isolation (J. Katz, Group Leader). *Recovery of neptunium in four plant runs.* Beard, Hopkins, Malm, La Chapelle, J. Katz, and Gilbreath have carried out the separation and isolation of 8 mg Np^{237} from the final plant lanthanum fluoride slurry from four Clinton runs using a modified process to assure a considerable yield of neptunium. The lanthanum fluoride was metathesized and dissolved in acid, a peroxide was precipitated and dissolved in HNO_3, and the resulting solution was treated with NaOCl to oxidize the neptunium. The plutonium was removed as K_2PuF_6, the supernatant was reduced with SO_2, and the neptunium was carried down with lanthanum fluoride. At least 90% of the neptunium present in the lanthanum fluoride slurry was recovered. On the basis of the theoretical ratio of neptunium to plutonium formed in the pile, this represents about a 16% recovery.

Process Development (Gilbreath, Group Leader). *Decontamination with respect to neptunium in the Bismuth Phosphate Process.* Hyman has found that decontamination from neptunium in the extraction step is not appreciably affected by variation in the nitrite concentration. Storage of the 40% UNH solution with H_2SO_4 at 75°C appears to increase neptunium decontamination slightly. An overall decontamination factor of 7000 is obtained after three reprecipitations of the bismuth phosphate extraction precipitate in the presence of 0.04 Fe(III).

Overall solvent extraction — "Redox" process. Blaedel, Post, and Walling have determined the effect of the composition of the solution on the distribution ratios of plutonium(IV), uranium(VI), and HNO_3 for systems containing hexone, H_2O, HNO_3, UN, $K_2Cr_2O_7$, and NH_4NO_3 [or $Al(NO_3)_3$]. In concentrated UN systems the distribution ratios for both plutonium and uranium vary from 1/2 to 2 for HNO_3 and NH_4NO_3 concentrations in the aqueous phase or from 0 to 2 M and 0 to 8 M, respectively; in dilute UN systems these distribution ratios are generally higher and are more sensitive to HNO_3 and NH_4NO_3 concentrations. The distribution ratio for HNO_3 is not greatly dependent on UN or HNO_3 concentrations in 4-8 M NH_4NO_3 solutions but is greatly dependent on them in the absence of NH_4NO_3. The plutonium distribution ratio in the above systems is essentially independent of the plutonium concentration in the aqueous phase in the range 1.0 mg/l to 1000 mg/l.

Solvent Extraction (Lawroski, Group Leader). *Hexone extraction for isolation.* Bernstein, Egan, Hausman, Schaffner, Schraidt, Simon, and Struminski have successfully completed three runs in the 3-inch diameter glass columns in which 0.212, 1.14, and 2.53 grams of plutonium were processed in feed volumes of 50 gallons. The results indicate that a 99.7% yield of plutonium with a purity of 99.9% or better can be consistently obtained (the throughput capacity of the equipment indicates that the contemplated daily Hanford plant volumes of feed solutions could be processed in 12 hours).

Solvent extraction (for decontamination, concentration, and isolation) of a solution of the bismuth phosphate extraction precipitate. Ader, Brody, Kelley, Reinhardt, and Stein have carried out multi-stage distribution coefficient experiments with dissolved bismuth phosphate extraction precipitates that show very favorable plutonium extraction by hexone and good retention of FPE (fission product elements) in the aqueous phase for the first stage of extraction. In subsequent stages

the coefficients for FPE decrease rapidly. This indicates marked preferential solubility of the hexone for certain species of FPE. Fission product analyses indicate zirconium and ruthenium to be the principal offending constituents. On the basis of preliminary experiments with tracer zirconium, it is believed that satisfactory elimination of this constituent can be effected in the first column. The removal of ruthenium appears, at present, to be a more difficult task and may require a special procedure such as volatilization from the initial feed solution after oxidation.

Decontamination of plutonium by solvent extraction (converter pile). Hyman has carried out batch experiments on irradiated plutonium (44 Mwd/ton uranium) using hexone as a solvent and extracting from a feed solution containing 0.8 g Pu/l, FPE from fission, $Al(NO_3)_3$, and HNO_3. Approximately 1% of the total activity is found to accompany the plutonium through a cycle involving extractions into hexone, four washes with salt solution, reextraction into water, and five washes with solvent. A volatile gamma-ray emitter with absorption characteristics resembling ruthenium and a non-volatile beta-particle emitter appear to be present in the final solution.

In today's war summaries come reports that U.S. troops have seized one-fifth of Cologne and that Soviet troops are shelling Steltin suburbs in Poland. Navy Secretary Forrestal announced that 2,050 marines have been killed on Iwo Jima.

Wednesday, March 7, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I opened the meeting with a reminder of the three instruments meetings scheduled for later today. I announced that Zinn, Dancoff, et al., have found eta for U^{233} (neutrons emitted in fission per neutron absorbed) to be greater than 2.3 and the n,γ cross section is probably less than 10 barns.

Manning, Katzin, Albaugh, Ghiorso, Davidson, and I discussed possible locations for the new hot labs. I then summarized the situation by saying it would be desirable to remove the present fluorinating cubicle from Room 4 and build a small hot lab in there; one cubicle in Room 41 will be used as the fluorinating room, Room 28 will be Albaugh's hot lab, and Room 36 or 33 will be made into a cold lab, replacing the present function of Room 28. Room 218 in the West Stands is to be used for chemical work for Ghiorso's group, such as Scott's UY work.

I then indicated we still have some time to talk about our work and called on Simpson to describe the electronic devices that he and the others in his group have built to prevent temperature fluctuations in their furnace. He mentioned a recent run on the oxidation of $PuBr_3$ by adding O_2 in small steps and measuring the vapor pressure; he expects to be able to determine whether PuOBr is formed and to determine its vapor

pressure. Simpson mentioned that in vaporizing PuF_3 onto sample plates for the Instrument Group, he found that a tantalum crucible can be saturated with only 30 micrograms. This led me to observe that it looks as though we will have high efficiency in case we ever have to prepare plates of precious samples such as some small ones containing large amounts of 94^{240}.

In a discussion of the possibility that the calorimeter recently built by Westrum and Robinson could be used for radioactive measurements, Manning mentioned a recent finding at Los Alamos that 80 grams of plutonium metal in one lump is warm to the touch.

I received a copy of a memo from Zachariasen to Mulliken suggesting that neither the term "thoride series" or "actinide series" be adopted for general use in the Project publications. He argues that it is impossible to state at this time that the term "thoride series" may not become justifiable since, so far, valence state observations do not extend beyond plutonium. At the same time, he considers as inappropriate my term "actinide series" (I first proposed this at the July 17 meeting with C. A. Thomas on "Final Purification and Metallurgy of Product," and stated it on page 55 of the report of that meeting, CK-1968), for thorium is certainly not actinium-like. He does concede, however, that if there are nine elements following plutonium, all of which are trivalent (and not all of them tetravalent), there would be some justification for the use of "actinides" even though there is a gap in the series at thorium.

In a teletype to Allison at Site Y, I advise him that as soon as we can get a courier we will send him the second fraction of CW1B (completely decontaminated plutonium peroxide) — about 14 milligrams.

I sent a memo to Natasha Goldowski in the Physics Division commenting on her memo to Elmer Brugmann about the corrosion resistance of metals in uranyl systems that might be used in a homogeneous, heavy water pile.

Captain Chapman requested of the District Engineer, Oak Ridge, six grams of U^{235} containing not more than 25% U^{238}. He indicates that one gram is desired for the purposes I listed in my memo to Stearns last Saturday. The other five grams are desired for cross section measurements and other neutron work by the physicists, including use as a comparison standard for measuring neutrons emitted per thermal neutron absorbed by the isotope U^{233}.

During the morning I attended a meeting on the Project Handbook — Volume 21 of the Project Record.

There were three scheduled meetings in the afternoon at which everybody in Section C-I received instruction in the use of counters. At 2:00 p.m. Albaugh's sub-section met; at 3:00 p.m. Cunningham's sub-section (except Stewart's group) met; and at 4:00 p.m. Katzin's and Stewart's groups met.

At 7:45 p.m. I attended a meeting in Room 209, Eckhart Hall,

of the Separation Processes Sub-section. Others present were Ader, Albaugh, Beard, Daniels, Egan, Fineman, Gilbreath, Greenlee, Hagemann, Hindman, Hopkins, Hyman, Kelley, Larson, Lawroski, Manning, Morgan, S. Peterson, Schaffner, Schraidt, Studier, Thompson, Walling, Winner, and others. I turned the meeting over to Albaugh who first called on Schaffner. Schaffner reported on the cleaning and remodeling underway of the three-inch columns and auxiliary equipment. The column packing has become dirty through deposition of silt from feed solutions and hexone-miscible pipe dope from the solvent lines. Certain modifications, designed to facilitate operation, have been made in the equipment during this period. Following completion of this reconstruction program, further tests of the hexone concentration-isolation process will be made to attempt to obtain increased concentration of plutonium and to secure more exact engineering data. After these tests are completed, it is planned to use the same equipment in tests of the bismuth phosphate-hexone extraction-decontamination process.

Kelley reviewed recent work on the bismuth phosphate-hexone extraction-decontamination process; recent work has been devoted largely to the determination of optimum salt and nitric acid concentrations in the feed and reflux solutions.

Greenlee spoke about attempts to devise an analytical scheme, based upon selective extraction of Pu(IV) by benzene-TFA, that would be capable of distinguishing between Pu(IV) and Pu(III) in dissolver solutions. Extractions of synthetic solutions show less than 95% extraction of plutonium from spectroscopically pure Pu(IV) solutions and 5% or greater extraction of plutonium from spectroscopically pure Pu(III) solutions. It appears probable that the solutions did nevertheless contain mixtures of the two valence states accounting for the less than 95% extraction in the first case and greater than 5% extraction in the second. Tests on mixtures of known composition of Pu(IV) and Pu(III) also show some discrepancies.

Gilbreath reported on the TFA extraction-decontamination process involving extraction of plutonium from dissolver solutions into a benzene solution of TFA and reextraction of plutonium into a 2 N HNO_3 solution. Earlier experiments by Blaedel and Margolis show excellent plutonium extraction but poor decontamination. Subsequent batch experiments, designed to simulate a continuous countercurrent cycle (by means of successive scrubs on the extract phases), give improved decontamination factors. There is some evidence of the formation of a reaction product between TFA and HNO_3 that inhibits the chelation of Pu(IV) by TFA. It is planned to continue the decontamination studies, perhaps evaluating the effect of $SO_4^{=}$.

"Yanks Take Cologne" reads today's banner headline. Cologne is the fourth largest city in Germany and the largest German city to fall to the Allies in either World War.

Thursday, March 8, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Florin, Ghiorso, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Magnusson, Manning, McLane, Morgan, O'Connor, Studier, Van Winkle, and later Cunningham. James described his compilation of information on energies, Auger electron yield, etc., and L- and K- x-rays of heavy elements; and Florin talked about his progress on working up the uranium plus He^4 target (sample TαB, Berkeley bombardment). Work on x-rays of U^{233}, Pu^{239}, and Np^{237} was discussed.

The following activities are scheduled: (a) Studier-Ghiorso run on x- and gamma-rays from 60 mg sample of U^{233}, (b) a run about Monday, March 19, on U^{233} decay products, (c) La Chapelle to prepare a 100 microgram sample of Np^{237} for Zinn, (d) the $Np^{237}(n,\gamma)$ run with cadmium cut-off will be made next Monday.

I received a memo from Johnson in Personnel transmitting a copy of a March 5 letter from the Standard Oil Company of California extending Stan Thompson's leave of absence from March 1 to July 1 of this year.

I wrote to Lois Moquin in Berkeley telling her that my name has been changed from Edrey Smith to G. T. Sutton; this name should be used in all telegrams and telephone calls. Correspondence to my secretary should be sent to Edrey Albaugh rather than Edrey Smith because of Edrey's marriage.

Jones sent a memo to Furney to arrange for the transfer of 50 micrograms of Np^{237} to Argonne for 6 hours of neutron irradiation in the pile next Monday.

In a memo to Albaugh, Blaedel and Gilbreath sent the results of computations to determine whether or not batch extraction experiments on decontamination can be extrapolated to predict the results of continuous countercurrent column operation. They have found a method of approximation for doing this that is conservative and acceptable.

According to today's paper the Germans say that the Soviet army has begun its long expected general assault on Berlin from its Oder River bridgeheads, thirty to forty miles to the east.

Friday, March 9, 1945

The third plutonium production pile (F) at Hanford reached its rated power yesterday.

I wrote to Stan Thompson in order to inform him of the extension of his leave from Standard Oil.

James completed isolation of the 95 fraction from sample 49NF (Clinton neutron-bombarded plutonium) which he began last Monday. He finds a total of 55 alpha-particle counts per minute in the final rare

earth fraction along with about 23 millicuries of beta-particle activity (presumably due to rare earth fission products). He next began working up a sample obtained from Florin from the target of uranium (Met Lab sample TαB) which received 140 microampere hours of 40 Mev alpha particles in the Berkeley cyclotron between February 18 and 26. The sample James received is the lanthanum fluoride precipitate that Florin made from the oxidized solution of the target and should contain any element 95 or 96 formed during the bombardment. He designated the sample TAB-J.

Compton received a March 6 letter from Latimer indicating he will be very glad to receive the 1 mg of U^{233} that I told Connick we could transfer to the Berkeley project.

In a letter to Kohman at Site W, Jaffey advises him that we are sending him somewhat more than 6 millicuries of S^{35} formed by bombardment of KCl at Clinton.

The U.S. army has crossed the Rhine at Remagen and is firmly entrenched on the eastern side.

Saturday, March 10, 1945

Virginia Towle was hired to work as a technician for Ghiorso.

I wrote to Spedding in Ames, explaining that Phipps and Simpson believe they can measure the melting point of uranium metal with an accuracy of a degree or so using the apparatus they have developed. I ask for his opinion as to the desirability of obtaining a more accurate value for the melting point of uranium and whether or not he can furnish us with about a cubic inch of high purity metal for this purpose.

Dempster wrote a memo to Stearns about some aspects of the future program of the Met Lab that should be pursued because of their obvious military importance. Stating that they represent examples of the content of such a program he lists the following:

(1) What are the possibilities of a small country developing breeder piles that could make that country an international menace in 25 years from the present time?

(2) What are the possibilities of small piles operating in various neutron velocity ranges as power sources suitable for special purposes?

(3) Is there any possibility of finding new methods of enriching naturally occurring isotopes that could be developed and used secretly by a foreign power or private organization?

(4) What methods can be used to detect unauthorized use or international traffic in special materials or secret activities in various parts of the world leading to the accumulation of "products" in the hands of aggressor nations?

Radiation surveys in Section C-I by the Health Division for the week ending March 10 show high radiation readings in Room 13 (James, La Chapelle, and Magnusson), Room 33 (R. Thompson), and Room 36.

More than 300 superforts bombed Tokyo in a surprise raid before daylight today, the heaviest raid yet made.

The first meeting of the Laboratory Steering Committee took place in the afternoon. Those present included Bartky, Compton, Daniels, Dempster, Franck, Greninger, Hilberry, Stearns, Stone, Szilard, Wigner, and Zinn. At the meeting Wigner summarized the present knowledge concerning the various breeder piles. Szilard spoke about his concern that the first bomb that we detonate may start a race in atomic armaments between us and other nations, in particular, Russia. He believes that for the safety of this nation it is necessary for us, within the next couple of years, to produce ten times as much fissionable material as is now planned (i.e., ten tons instead of one ton) in order to insure an agreement with Russia to prevent its future use in war.

Sunday, March 11, 1945

Under a March 11th dateline the paper says the U.S. army has captured eight towns east of the Rhine River.

Monday, March 12, 1945

Fifty micrograms of Np^{237} were given a 6-hour irradiation in the Argonne heavy water pile with epithermal neutrons (cadmium cut-off). The irradiation ended at about 4:00 p.m., and the sample was immediately returned to New Chem where Jaffey began a study of the transmutation products.

Tom Jones sent a memo to J. A. Wilkinson in Administration suggesting the steps necessary to restore Room 212 in Kent Chemical Laboratory to the condition in which it was found when borrowed by the Met Lab about two and a half years ago. This is the room in which Getz and later Livingston (of Burton's section) attempted to recover Po^{210} from a concentrate of 80 millicuries of RaD-Po originally obtained from Hamilton of Berkeley. Recently, after a survey by the Health group, an attempt to clean up the room was made by a decontamination squad under the direction of Gilbreath. Jones's suggestions include replacement of a number of permanent fixtures that had to be removed because of contamination during this clean up, i.e., replacement of lead top on hood base, part of linoleum flooring, stone sink, and top of laboratory work bench.

Szilard prepared a draft memorandum (MUC-LS-61) on the importance

of the U.S. drawing up a program for the production of ten tons of "heavy elements" within the next few years (ten times the planned amount), in order to convince the Russians that we are so advanced in the field of atomic weapons that no other nation has a chance to catch up to us. Szilard feels that such evidence of superiority is our best chance to convince the Russians that they should agree to a system of joint control by the U.S., Great Britain, and the Soviet Union on the manufacture of "heavy elements" everywhere in the world.

Three hundred B-29's have raided the airplane plant at Nagoya.

Tuesday, March 13, 1945

In a memo to Stearns, Daniels comments on the Steering Committee meeting of last Saturday afternoon. He urges that we begin thinking in terms of piles of a million kw in spite of the serious engineering difficulties involved. He suggests that a series of lectures on piles to which the section chiefs and a few others are invited will be helpful. He agrees with Szilard on the importance of making available, within two years, ten times as much fissionable material as has been contemplated. He then urges an actual demonstration of a high-temperature pile for the generation of power as a means of increasing our prestige with the public and our support by the government.

In a bulletin from Manila comes news of the capture of the city of Zamboanga and of San Roque airdrome, a major field on southwestern Mindanao in the Philippines.

Wednesday, March 14, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. I was happy to report that last week there were no security violations in the whole Division. I announced there will be a meeting at 4:00 this afternoon with Drs. Stone and Russell on the problem of plutonium analysis in medical diagnosis work — they need our advice on the chemical procedures, and therefore we will have some of the people from our section there.

I also said that we have been turned down by the authorities at Hanford on our requests for a special Np^{237} run and the shipping to us of dissolver solution — they do not want to confuse their operators after training them in a definite routine. I indicated we may, however, be able to irradiate our one mg of Np^{237} in the Site W pile, where it would not involve any special procedures. The purpose of the irradiation will be to make some very pure 94^{238}, possibly more than 97% pure by weight, and to see what else might be formed in the process.

I mentioned the stringent time schedules of 6 to 8 weeks on the

Metallurgical Project Record Volumes and said that there will be a meeting to discuss Volume 14 next Monday afternoon. Perlman, English, Hamilton, Eastman, and H. L. Anderson will be here next week for the general meetings.

Manning discussed the hot lab situation which he is about to present to Stearns. He asked about the possibility of getting along with only one hot lab in case we have to compromise. I expressed doubt on the basis that Morgan, James, O'Connor, and Florin usually cannot tolerate the presence of even small amounts of uranium and, in working with other alpha-particle activities, they cannot allow the presence of any 49. I added that some of Albaugh's men probably will be working in their hot lab all the time and this will cause some crowding; we will certainly need separate containers or hoods for different kinds of work. We also discussed the possibility of reducing cost by less lead shielding and by the substitution of concrete for lead.

I asked for a report on the present situation on Pa^{231}. Katzin responded that we have 1 mg in the laboratory in a few hundred milliliters of solution and three additional batches containing 1 mg each that have gone through the first step of the semiworks isolation procedure.

I read a copy of a memo dated March 13 from Daniels to Jones expressing appreciation for the thorough manner in which he handled the unpleasant job of decontaminating Room 212 in the Kent Chemical Laboratory.

In a memo to Wahl at Site Y, I ask whether he should send me some more of his notebooks to be used by the attorneys here to strengthen Case 52. I point out that I wrote to Kennedy about a month ago about this matter but received no reply.

Hilberry asked Stearns to arrange the transfer of the one mg of U^{233} to Latimer in Berkeley, provided it is possible from Stearns' point of view.

At 7:45 p.m. I attended the meeting in Room 209, Eckhart Hall, of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I. Others present were Abraham, Ames, Arnold, Asprey, Cressman, Hagemann, Hellman, Hindman, Howland, Hyde, Jaffey, James, Jones, Katzin, Krueger, La Chapelle, Larson, Lawroski, Magnusson, Manning, McLane, Morgan, O'Connor, S. Peterson, Phipps, Reinhardt, Robinson, Schwob, Schaffner, Simpson, Stewart, Studier, R. Thompson, Tomkins, Van Winkle, Westrum, Wolf, and others. I opened the meeting, announced that Katzin's group will take over our section meeting next Wednesday, March 21, and then I turned the meeting over to Cunningham.

Davidson continued the series of lectures to acquaint section personnel with nuclear physics, talking on the neutron crystal spectrometer. He identified three devices in current use for selecting neutrons of a given velocity for study: (1) modulated cyclotron beam; (2) velocity selector which utilizes a chopper opaque to neutrons, and (3) the neutron crystal spectrometer which has been developed during the last year and a half at Argonne and Site X. The principle of operation of the spectrometer based on the wave nature of any particle in motion was described. The operation of the instrument has been found most satisfactory.

Davidson indicated that from thermal energies to about 0.3 ev the velocity selector permits the most precise measurements. The crystal spectrometer is useful at energies in the range 1½ to 2 ev; the accuracy obtained is no greater than with the modulated cyclotron beam. He described a proposed velocity selector for high energy neutrons which would consist of a rotating cylinder of boron carbide with a helical slot cut in its surface through which neutrons of a specified velocity could pass longitudinally.

Ghiorso reported on the study of x-rays from Np^{237} samples prepared from the Clinton material by La Chapelle and Magnusson, designated as samples 37A (prepared from a run corresponding to the production of 5 gt Pu^{239}), 37B (from 6.9 gt material), and 37Z (from 1.5 gt material). Both x-rays and negative particles are found to be present. Analysis of the gamma-ray absorption curve shows photons of energies of 0.3 Mev, 85 or 150 kev, 30 kev, and 15 kev. Excellent agreement is found between the ratio of x-rays and negative particles to alpha particles and the bombardment time, indicating that both the particles and x-rays are due to an additional isotope or isotopes produced in the bombardment. At least tentatively the observed x-radiation is assigned to Np^{236}, this isotope presumably decaying by K-electron capture. A neptunium isotope of similar properties is observed by James and Florin in their bombardment of U^{235} with deuterons in the Berkeley 60-inch cyclotron. Hence Np^{236} may be formed in these experiments by the two reactions: $Np^{237}(n,2n)Np^{236}$ and $U^{235}(d,n)Np^{236}$. The isotopic assignment of the negative particles cannot yet be made; they must be due to some isotope of neptunium.

Ghiorso next reviewed the calculations that Jaffey, James, and I have made of the counting yields for x-rays based on absorption of the photons in the argon gas of the counter itself. The calculations confirm that L x-rays are counted more efficiently than K x-rays.

In a discussion of the wall effect that followed, I indicated this effect would predominate in counting all photons of energy over 100 kev in argon and all over a few hundred kev in xenon. We then discussed at some length other possible sources of the photons and negative particles found in Np^{237}.

Hindman reported on further spectrophotometric studies of the nitrate complexes of Pu(IV) which were performed in mixed nitric-perchloric acid solutions since perchlorate does not complex Pu(IV) to a measurable extent in these concentrations. In pure perchloric acid an absorption band is found at 471 millimicrons. This diminishes as nitrate concentration is increased, and a new band appears at 476 millimicrons. Analysis of the data indicates the formation of a complex involving one nitrate ion per atom of plutonium as well as complexes involving more than one nitrate ion per plutonium atom.

War summaries today include news from many fronts: Western — U.S. troops are pouring across the Rhine on at least one pontoon bridge they constructed. London — RAF bombers attacked Barmen in the Ruhr. Eastern — Soviets are 33 miles from Berlin, according to Nazi reports. Manila — Americans capture four villages in Mindanao. Guam — superforts blast Osaka, second largest city in Japan.

Thursday, March 15, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Florin, Ghiorso, Hindman, James, Katzin, Larson, Manning, McLane, Morgan, O'Connor, Van Winkle, and later Magnusson. Florin described his progress in working up sample TαB (uranium plus 40 Mev alpha particles, Berkeley cyclotron bombardment), and Studier described his work on x- and gamma-rays of U^{233}.

Activities schedules were (a) further work by Studier-Ghiorso on U^{233} x-rays; (b) a run on U^{233} decay products about Monday, March 26; (c) James to work on the plutonium plus alpha particles Berkeley target [sample 49αB] to be received next week); (d) a practice run next Wednesday to look for 94^{241}.

I received a teletype from Allison referring to my letter to him of March 6 about our metal preparation for Los Alamos of plutonium from sample CW1B. He states that their mass spectrographic study shows 0.65% Pu^{240}. Lead was also found, and he asks if I could study our optical spectrographic plates for lead lines and send him the results by teletype. He also includes the interesting statement that the mass number 241 was seen. Variation of the furnace temperature made the intensity relative to 40 and 49 vary rapidly so they know it is not due to a plutonium isotope. Allison asked if it is possible that this is due to my 51. No line was found for mass number 242 but this mass number is obscured by the presence of a line due to PbCl.

I responded to the March 7 letter of Captain Barnowsky, Corps of Engineers, and gave the new assignments of enlisted men Eugene Hausman, Benjamin Struminski, and Alec Kelley (all three assigned to work on the large solvent extraction column in Lawroski's group) and Horace Hopkins (assigned to Roy Thompson's group).

Allison then phoned me from Site Y to discuss the results of their analysis of the sample of CW1B metal that we processed for them. They found the sample clean — only lead was detected. In their mass spectrographic analysis they find a line at mass 241, not due to Pu^{241} because it varies in intensity from zero at low temperature to as high as the line due to Pu^{240} at high temperature. Teller thinks it due to 96^{241}. I asked him if he would write to Stearns in support of a voltage regulator for Simpson and our hot lab; he will think it over. He said they probably will not back our request for a special Np^{237} run at Hanford.

Marny Potter, a technician who works for Katzin, terminated today.

Compton sent a memo dated March 13 to Szilard stating that he talked to Bush Monday morning. Bush anticipates prompt action with regard to the appointment by the Secretary of War of the committee to consider the policy to be adopted regarding longer-range developments in our field. The committee will be responsible for recommendations about production of fissionable material as well as research. Compton suggests that Szilard be prepared to present his ideas to the committee.

The Soviet army has opened an all-out drive on Koenigsberg. From the Pacific come reports that Japanese resistance has stiffened on Mindanao.

Friday, March 16, 1945

James completed isolation (begun March 9) of the 95 fraction from sample TαB-J (rare earth fraction from Berkeley alpha-particle bombardment of uranium target) and finds 30 c/m of 95^{241}. He next began working up a 250-mg sample of 27 gt Hanford plutonium to look for 95^{241}, proceeding on the assumption that 94^{241} might be a long-lived (about 100 years half-life) beta-particle emitter decaying to 95^{241}. The sample that James obtained from Stewart was decontaminated and purified at Hanford and allowed to stand for 45 days. This sample is designated 51A.

I received a March 16 memo from R. Thompson describing the process modifications necessary to permit recovery of 50 to 90% of the neptunium along with the plutonium at Hanford. Recognizing that any deviation from normal procedures involves some risk, he also describes a procedure developed for recovering neptunium from the UNH wastes from the bismuth phosphate extraction precipitate. The procedure differs only in minor details from the regular bismuth phosphate procedure and could be handled without difficulty in spare Hanford equipment. Experiments performed on a 25-ml scale starting with 125 gt levels of plutonium and Np^{237} together with Np^{239} tracer have given 80 to 90% neptunium yields. Thompson also suggests the possibility of recovering relatively large quantities of Np^{237} by processing a large number of extraction wastes with the same bismuth, building up the concentration of neptunium in successive runs until the neptunium from perhaps 25 or 50 runs is accumulated. The plutonium recovered in such a procedure might well amount to 50 grams and would of itself perhaps justify the effort required.

Albaugh and Roy Thompson described for me by memo the Hanford process modifications that would permit increasing the uranium metal throughput by a factor of four; this limit is imposed by the total dissolving capacity of a canyon. Concentration has been achieved by three methods: (1) by decreasing the bismuth concentration in the extraction step from 2.5 mg/ml to 1.9 mg/ml permitting a smaller amount of HNO_3 in dissolving the extraction precipitate, (2) by separating the extraction precipitate from 28% UNH rather than the flowsheet specified 20% UNH, and (3) by partial neutralization of the solution of the extraction precipitate. These considerations apply to Hanford operations at a 250 gt level. The procedure has been tested on a 1500-ml scale.

Spedding answered my letter of March 10, in which I asked his opinion about the desirability of Simpson and Phipps using their apparatus to obtain an accurate value for the melting point of uranium and whether he could spare a high purity sample of uranium for this purpose. According to Spedding, in the progress report of the National Bureau of Standards for December 1944, Cleaves and Dahl report that the melting point of uranium has a value of 1132° ± 1°C. He said that he will supply us with

some pure Ames biscuit metal if we decide to pursue the matter. Because we reported a measurement of 1130° ± 10°C in November, we shall expend no further effort on this problem.

Estimates of the U.S. losses on Iwo Island are now 4,000 — the costliest invasion of the Pacific War.

Saturday, March 17, 1945

At Berkeley, the 40 Mev alpha-particle bombardment of 100 mg of Pu^{239} is ending today with a total of 53.1 microampere-hours (49αB). The bombardment started on March 6.

Morgan completed isolating the 95-96 fraction from sample 49NEY (the rare earth fraction received from Site Y of the entire Fermi special sample CW1). His data show 250,000 c/m of which 195,000 c/m are 95^{241} and 55,000 are 96^{242}. He mounted the final precipitate on a platinum disk and set it aside for fission experiments, alpha-particle range measurements, and to let 94^{238} possibly grow in from decay of 96^{242}.

I responded to Perlman's request for our results of process runs through two bismuth phosphate cycles on a 25-ml scale to test decontamination with respect to neptunium. I point out that following present Hanford flowsheet procedures, we obtained a decontamination factor of 259. A parallel run omitting $(NH_4)_2SiF_6$ in the plutonium precipitatation steps resulted in a decontamination factor of 1.1×10^4. I add that processing the waste from the extraction step of these runs for Np^{237} using our special procedure gave 82% recovery.

Zachariasen sent a memo to Stearns identifying as $NaPu_2OF_5$ the plutonium phase accidentally prepared by our section early in November 1943 when we were attempting to produce plutonium metal. According to Zachariasen, our chemists did not demonstrate subsequent interest in the problem of identification so he solved it himself by making the isomorphous chemical preparations $NaLa_2OF_5$, KLa_2OF_5, and $KU_2O_3F_3$.

Requests for deferment of the following members of my section were forwarded to the Engineer's Office in Oak Ridge by the Chicago Area Engineer's Office: Blaedel, Fields, Florin, Hufford, Hyde, Hyman, La Chapelle, Magnusson, S. Peterson, Schaffner, R. Thompson, Van Winkle, and Walling.

Daniels' office prepared schedules for a tentative lecture program consisting of a Saturday afternoon program open to Group Leaders and the Monday evening chemistry seminar open to academic personnel, both biweekly. It is as follows:

Saturday Afternoon Programs
open to Group Leaders

March 17	Fundamental Constants for Pile Design	W. H. Zinn
March 24	Present Files — Argonne, Clinton, Hanford	A. M. Weinberg

April 7	Engineering Problems	L. A. Ohlinger
April 21	Chemical Problems	G. T. Seaborg or W. M. Manning
April 28	The Heavy Water Breeder Pile	E. P. Wigner or H. S. Brown
May 12	The 1 to 100 ev Breeder Pile	E. P. Wigner
May 26	The High Temperature Breeder Pile	
June 9	The Fast Fission Breeder Pile	L. Szilard
June 23	Laboratory Program after July 1	J. C. Stearns
July 7	Nucleonics in the Stars	W. Bartky

Monday Evening Chemistry Seminars
open to Academic Personnel

March 19	Chemical Separations at Hanford	I. Perlman
April 2	Solvent Extraction Process	S. Lawroski and W. J. Blaedel
April 16	Carrying Chemical Compounds and Isomorphism	G. E. Boyd
May 7	Power Piles	G. Young
May 21	Uranium and Thorium	Z. Jeffries
June 11	Principles of Radiation Chemistry	J. Franck
June 18	Radiation Chemistry	M. Burton
July 2	The Chemistry of 49	B. B. Cunningham
July 16	The 23 Problem	L. I. Katzin
August 6	Tracers in Chemistry and Biology	W. M. Manning
August 20	Future Application of Nucleonics	L. Szilard
Sept. 3	Future Application of Nucleonics	T. R. Hogness

Radiation surveys by the Health Division in Section C-I for the week ending today show high radiation levels for the Room 30 centrifuge (Bartell and Malm), and for Room 2 (Abraham and Sheft), Room 10 (Hindman), Room 13 (Magnusson, La Chapelle, and James), Room 27 (Beard and Hopkins), Room 31 (Greenlee and S. Peterson), Room 33 (Thompson), Room 36 and Room 37.

American superforts bombed Kobe in the heaviest assault yet made on any Japanese city.

Sunday, March 18, 1945

Helen and I played 18 holes of golf at Evergreen (91st Street and Western Avenue) with Al Ghiorso and Tom and Betty Morgan. Al shot 121, Tom shot 135, and I shot 119.

A bulletin datelined the 18th says a U.S. armored division has entered Kreuznach, 21 miles southwest of the Rhine city of Mainz.

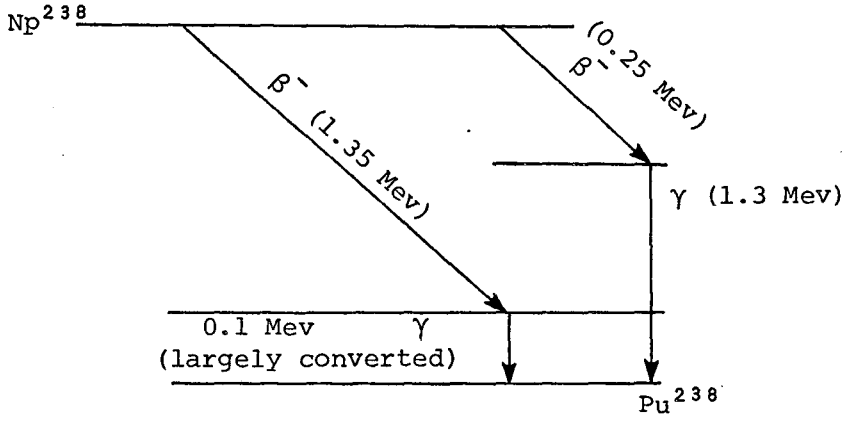
Monday, March 19, 1945

Perlman is in town from Hanford in order to attend the Project Council Information meetings, to talk at the Chemistry Division meeting tonight, and to confer with our chemists. Eastman is here from Berkeley, and English arrived from Clinton Laboratories.

I read a copy of a memo from Nickson to Daniels about our plans to install hot laboratory facilities in Rooms 4 and 27, New Chemistry Building. The Health Division feels this problem is urgent and recommends that adequate facilities be completed without delay.

Albaugh wrote a summary of the work of Sub-Section I, Section C-I, for my use at next Wednesday's Project Council Chemistry Information meeting. The topics covered are (1) *Methods for increasing the rate of metal processing.* (2) *Decontamination from neptunium.* (3) *Recovery of neptunium in process runs.* The supernatants from the extraction steps in the neptunium decontamination runs were processed for neptunium recovery, and it was indicated that 82% recovery could be expected through the cross-over if $H_2C_2O_4$-Mn(II)-$(NH_4)_2SiF_6$ prereduction is combined with use of Fe(II)-$(NH_4)_2SiF_6$ (0.20 M) in the decontamination cycles. (4) *Analysis of plant dissolver solution for* Np^{237}. The yield of Np^{237} in the Clinton pile is about 0.3% of the Pu^{239} yield on the basis of analysis of Clinton Run 297 dissolver solution. There may be considerable error as the results depend on a range curve taken on 20 alpha-particle c/m. (5) *Use of TFA for analysis of plutonium(III)-(IV) mixtures.* An accuracy of 5% is indicated. (6) *Overall solvent extraction — "Redox Process."* It has been shown that plutonium and uranium may be efficiently extracted from aqueous and hexone process solutions and that efficient separation from uranium may be expected in the reduction column. The uranium in the hexone effluent from the reduction column is decontaminated from beta-particle and gamma-ray activity by a factor of 10^3-10^4. Successive batch extractions to simulate operation of scrubber sections in the oxidation and reduction columns have yielded minimum decontamination factors of approximately 10^6 for beta-particle and gamma-ray activities. (7) *Solvent extraction of a solution of the bismuth phosphate extraction precipitate.* Improved decontamination through at least two stages of hexone extraction has been demonstrated by experiments in which the starting feed was previously oxidized with $NaBiO_3$ and air sparged to volatilize the ruthenium. (8) *Hexone extraction for isolation-development.* The 3-inch diameter solvent extraction column has been reassembled after cleaning and modification. (9) *Converter pile decontamination problems in solvent extraction.* Poor decontamination of zirconium results unless the concentration of salting agent is kept low. Batch equilibrium experiments with ruthenium tracer, hexone, and aqueous solutions showed distribution ratios for ruthenium (aqueous/hexone) as high as 70-90 for reduced ruthenium and as low as 1-2 for oxidized ruthenium.

Cunningham wrote a summary for me of the work of Sub-section II, Section C-I, again for my use at Wednesday's Project Council Information meeting. He covers the following: (1) *Isotopes in irradiated samples.* A product with alpha-particle activity found in plutonium irradiated with neutrons that may be due to the isotopes 95^{241} and 96^{242} exhibits chemical behavior as though the elements are almost exclusively in the +3 oxidation state; the yields seem proportional to the second power of the total neutron irradiation. A target of metallic natural uranium was bombarded with 40 Mev helium ions in the newly rebuilt 60-inch cyclotron at Berkeley for the primary purpose of studying the radioactive and fission properties of 94^{240} which might be formed by the reaction $U^{238}(\alpha,2n)94^{240}$. Although no alpha radioactivity which might be attributed to 94^{240} has yet been found, there are some other interesting activities in the plutonium fraction (alpha particles with range 3.68 cm – perhaps Pu^{239} from α,3n reaction; alpha particles with range 4.0 cm – presumably 94^{238} from deuteron contamination in the alpha-particle beam; alpha particles of range about 4.3 cm or greater plus x-rays – perhaps these two activities are due to 94^{237} and/or 94^{236} formed by α,2n and/or α,3n reactions on the isotope U^{235}). The rare earth chemical fraction that might contain the isotope 95^{241} from $U^{238}(\alpha,n)94^{241}$ followed by beta-particle decay, contains about 10 alpha-particle counts per minute. The neptunium fraction has also been isolated and is being studied. (2) *Radiation spectra of some neptunium isotopes.* It now appears there are uranium L x-rays and negative particles of 0.2-0.3 Mev associated with neutron-irradiated, completely decontaminated Np^{237}; their intensities seem to vary approximately with the first power of the total neutron irradiation. If the x-rays originate from Np^{236}, yield considerations indicate a half-life of one-half year. Further work on another sample believed to contain Np^{236} formed by the reaction $U^{235}(d,n)Np^{236}$ seems to indicate a decay of its L x-rays with a half-life of about one-half year. (3) *Decay scheme for Np^{238}.* The following decay scheme is proposed:



(4) *Half-life of Pu^{238}.* From the number of Pu^{238} alpha-particle disintegrations from a known number of pure Np^{238} disintegrations, the half-life of Pu^{238} has been determined as 56 ± 10 years. The purity of a Pu^{238} sample that has been followed for decay for over a year has been determined by range analysis in the alpha-particle pulse analyzer apparatus. The results show that not more than 5% of the alpha particles are due to Pu^{239} and indicate good accuracy for the observed half-life value (66 years)

determined in this way. (5) *Geiger counter efficiency for x-rays.* Calculations have been made that confirm and reduce to a quantitative basis previous indications that L x-rays are counted more efficiently than K x-rays in the argon-filled Geiger tube. The relative counting efficiency is the same in xenon-filled counters with the overall efficiency being at least three times as high. (6) *Chemistry of plutonium.* The reaction $PuBr_3 + O_2$ (tantalum crucible) → $PuOBr + TaBr_5$ has been studied in the vapor pressure apparatus; incomplete evidence suggests that the vapor pressure of PuOBr is about midway between that of $PuBr_3$ and PuO_2. (7) *Chemistry of neptunium.* The compound $NpCl_4$ (orange) has been prepared by treating NpO_2 at 500°C with a mixture obtained by bubbling Cl_2 through CCl_4. The formal potential of the Np(V)-Np(VI) couple in 1 M HCl has been redetermined as -1.117 v. Quantitative observations on the oxidation of Np(IV) to Np(VI) by bromate show that the reaction is complex and that the rate is affected by factors not yet understood. (8) *Chemistry of uranium.* At 1900°C in high vacuum UOS reacts with carbon to form US + CO. The formal potential of the U(III)-U(IV) + e couple has been measured directly in 1 M HCl to be $+0.633 \pm 0.002$ v. (9) *Chemistry of thorium.* At 1900°C in high vacuum, ThOS reacts with carbon to form ThS plus CO. (10) *Recovery of plutonium.* Hexone extraction of 10 M HCl solutions of plutonium containing bismuth phosphate show good plutonium recovery, but much bismuth is extracted and decontamination is poor. Peroxide precipitation has been found to be as efficient as oxalate precipitation in the final concentration step of the recommended hexone extraction process.

Nickson informed Wilkinson by memo that Gardner has been asked to decontaminate the rooms in Kent and Jones Laboratories formerly used by the Project. They were shown to be contaminated in a radiation survey made between February 24 and March 3. The rooms involved are Kent 101, 104, 109, 112, and 301; Jones 401, 403, 404, and the attic.

In the afternoon I attended a meeting of the editorial committee of Volume 14 of the Project Record, *Chemistry of the Transuranium Elements.* Editorial Committee: Seaborg (editor), Manning (associate editor), Connick, Brown, Cunningham, Perlman, and Zachariasen.

At 7:45 p.m. in Ryerson Laboratory, Room 251, there was a Chemistry Division Seminar at which Perlman, as the first speaker in the series, presented a lecture on "Chemical Separation at Hanford."

Nagoya, Japan's third largest city, has been hit by superforts for the second time in a week according to today's paper.

Tuesday, March 20, 1945

At 8:30 a.m. in Room 209, Eckhart Hall, I attended the Project Council Information Meeting on Physics. Items of interest reported were: (1) L. G. Lewis in Dempster's group spoke about plans relative to fission product mass assignment work. His group's instruments are now good enough so that the dispersion between U^{235} and U^{238} is 5 mm, making it possible to determine mass defects in this region. (2) Gale Young spoke

on the problem of turbulence in the design of liquid metal (e.g., bismuth) cooled piles. (3) Kay Way discussed the reasons for wide variation in the kinetic energy for the same mass ratio of fission fragments. (4) Wattenberg spoke on the efficiency of γ,n sources — Hughes has found Wattenberg's sources are superior to Ra-Be sources in providing mono-energetic neutrons. (5) Dancoff reported measurements of eta of U^{233} by comparison with U^{235}; a value of 2.48 ± 7% was obtained. Langsdorf plans to determine eta of U^{233} by a method similar to the danger coefficient method. (6) Snell of Clinton talked about photoneutron sources. He reported on Borst's run on the growth of the reactivity of the Clinton pile after shutdown. This fits Xe^{135} decay completely with no evidence for Wheeler's postulated second activity. Snell also spoke on Borst's crystal spectrometer. (7) Pardue reported on his use of the crystal spectrometer to measure the resonance neutron absorption of Pu^{239}. It was found to be 4000 barns at 0.3 ev.

George Bernstein was transferred by the army to Site Y.

James completed three dichromate cycles and one silver-plus-persulfate oxidation cycle on sample 51A (250 mg of Hanford decontaminated and purified 27 gt plutonium) to separate the element 95 fraction (he began working up this sample on March 16). The final solution was analyzed and showed 60% of the total alpha-particle count — about 12,000 to 15,000 c/m — to consist of 4.0-4.1 cm range 95^{241} alpha particles. This shows that 95^{241} is a long-lived beta-particle emitter because this plutonium was purified from 95, 45 days ago; we estimate the half-life of the 94^{241} to be in the range of 10 to 100 years.

Katzin wrote a summary of the work of Group 9, Section C-I, for my use at the Project Council Chemistry Information Meeting tomorrow. He states that: (1) Extractions are in progress on 60 kg of carbonate residues which may yield as much as 5 mg of Pa^{231}. (2) The 36-inch extraction column was used to complete the extraction of 23 cans of irradiated thorium "carbonate." The product was specially purified and a portion used by Zinn for measurements of fission constants which indicate great promise for U^{233} "breeding." (3) Electron microscope studies have been made of thoria particles in connection with slurry studies. (4) A flowsheet has been drawn up for solvent separation of uranium, protactinium, and thorium; it may prove possible to use a single solvent, diisopropyl ketone. (5) Thermodynamic data are being used to develop a theory for the mechanism of immiscible solvent extraction of uranium from nitrate solutions.

Manning received a memo from Nickson written March 19 noting that levels of beta-particle and gamma-ray activity handled in the filtered air section of New Chem have increased markedly in the past two months. Numerous overexposures to personnel occurred during this period; approximately 60% of the total number of Project personnel overexposures involve members of Section C-I. Nickson concludes that the personnel of Section C-I are not being adequately protected from exposure when handling material of high activity. It is his feeling that additional facilities for working with active material are necessary.

Jaffey prepared a "Suggested Outline on Some Alpha Radiation Measurements." It is divided into four sections: I. Introduction. II. Pulse ion chamber as assay instrument. III. Alpha particle ranges. IV. Some fundamental absolute measurements on half-lives of alpha particle emitters.

At 4:00 p.m. I attended a meeting in Room 209, Eckhart Hall, of the Volume Editors for the Metallurgical Project Record.

American forces have landed on Panay, a large island in the Philippines. This is the 25th Philippine island invaded by U.S. troops and the 7th one of major importance.

A special meeting of the Project Policy Council was held in Room 209, Eckhart Hall, from 5:05 p.m. to 6:30 p.m., attended by Chapman, Chipman, Compton, Daniels, Dempster, Doan, Eastman, Greninger, Hamilton, Harrell, Hilberry, Howe, W. C. Johnson, McKinley, Mulliken, Smyth, Spedding, Stearns, Stone, Tracy, W. Watson, Whitaker, Wigner, Wirth, Zinn, and later Franck. Compton explained that they were called together because of developments with regard to the future programs that have come up in the last few weeks. He then proceeded to read a memorandum (MUC-AC-2633) he has written to the Directors of the Argonne, Clinton, and Metallurgical Laboratories.

The memo states that the entire Metallurgical Project faces heavy cutbacks with the Laboratory at Chicago due for the greatest loss — a cut of 80% by July 1 of this year. Compton stated that the general order of preference in keeping staff is (1) retain men for certain top priority jobs at Site Y; (2) Met Lab will have first right to retain their own men; (3) Clinton Labs, with only a slightly curtailed budget, will be next in line for taking Met Lab personnel; (4) men remaining available after Clinton and Met Lab needs have been taken care of can be transferred to other DSM projects. After that they will be available to other war projects.

Compton said that he is not too unhappy at this way of doing things as a strong group will be maintained at Clinton until the close of the war, Chicago will be able to retain certain key personnel, and Argonne can be kept operating. He explained that the Military Policy Committee and the OSRD have taken the position that the future is the responsibility of some other group. The War Department has been requested by Bush and Groves to set up a committee to consider our program, but this committee will not be able to take any action which will change the position we are now in.

There was a discussion of the situation with respect to the chemists. Johnson mentioned there are 35 chemists at Hanford and about 90% of them want to come back to the Project. Compton said a request came in today from Site Y for four chemists — it is clear that Chicago chemistry will have to be sharply curtailed and Clinton's will certainly need to be reshuffled. Whitaker said they particularly want chemists from Chicago to replace du Pont

people. Wigner inquired when he and the others could discuss with people what Compton told them and Compton replied, "In the morning. I would say a certain number should be informed this week if we want to keep them."

Wednesday, March 21, 1945

At 8:30 a.m. in Room 209, Eckhart Hall, I attended the Project Council Chemistry Information Meeting. Others present were Aebersold, Allen, Anderson, Bowman, G. Brown, H. Brown, Burton, Cannon, Chapman, Compton, Coryell, Daniels, Davies, Dempster, Eastman, English, Franck, Fred, Hamilton, Huffman, W. Johnson, Manning, Mulliken, Newton, Perlman, Rabinowitch, Robinson, Smyth, Spedding, Stearns, Sugarman, Wakefield, W. W. Watson, Wattenberg, Watters, Way, Whitaker, Wilhelm, Young, Zachariasen, Zinn, and Zirkle. I was first on the program and reported first on the separation work. I indicated that work connected with the Hanford process is now completed, including the development of the process to take care of four tons per day per canyon instead of one ton. I described our studies of neptunium decontamination through the Hanford flowsheet and mentioned that we had been turned down by Hanford on our recommendation to recover Np^{237} from the first extraction waste. I reported that the study of the "redox" process is giving satisfactory results and that the 3-inch diameter column is back in operation.

On basic chemistry, I mentioned the volatility measurements on PuOBr and the new determinations of the potentials of Np(V)-Np(VI) and U(III)-U(IV) in 1 M HCl. I sketched the decay scheme of Np^{238} as suggested by our experiments. I reported that the 66-year half-life for Pu^{238} is based on observation of the rate of decay of an old sample. I reviewed our work on the radiation spectra of several neptunium isotopes and the evidence for a half-life of Np^{236} of one-half year.

I reported that the analysis of the transformations in transplutonium elements confirms our picture of last month:

$$94^{240}(n,\gamma)94^{241} \xrightarrow{\beta} 95^{241}$$

$$95^{241}(n,\gamma)95^{242} \xrightarrow{\beta} 96^{242}$$

I stated that the isotope 95^{241} probably is responsible for the 4.0 cm alpha particles and the isotope 96^{242} for the 4.7 cm alpha particles. The ratio of the 4.0 cm alpha particles to fission products varies with irradiation. This is in agreement with the assumption that these alpha particles are not due to a fission product. In short plutonium bombardments at Clinton, the yield increases approximately with the second power of irradiation in agreement with the above-given scheme. I said that evidence indicates that 94^{241} is a comparatively long-lived nucleus. The isotope 95^{241} can be prepared free from 96^{242} by letting 95^{241} grow in decontaminated plutonium irradiated by a high-density flux of neutrons (e.g., at Hanford). We have now found about 15,000 alpha-particle counts per minute of pure 95^{241} (containing no 96^{242}) in six-weeks old regular decontaminated Hanford product. The chemical work continues to indicate that elements 95 and 96 are exclusively trivalent.

At this point Zachariasen observed that the valencies of plutonium did not turn out exactly as predicted on the basis of first observation. I disagreed with the statement. Zachariasen replied that trifluoride was not predicted and that it is better not to have preconceived ideas about valency. I referred to our attempts over the past months to chemically separate 96^{242} from 95^{241} without success. This confirms the close chemical similarity of the two elements.

I described the results of our work on the uranium metal target from the 40 Mev alpha-particle bombardment at Berkeley (sample TαB) and the lack of alpha particles attributable to Pu^{240}. I mentioned our plans to look for neutron fission in the plutonium fraction that could be attributable to Pu^{240}. This might be achieved by bombarding the sample with neutrons to look for fission pulses, which would then be compared in amount with a standard sample of Pu^{239}. I gave the status of extraction of U^{233} from irradiated thorium and mentioned the 60 mg we turned over to Zinn for the cross section determinations reported by Dancoff at yesterday's Physics meeting.

Items of interest presented by other speakers are: (1) Perlman reported that the results of the chemical process at Hanford are very successful with production yields equaling or exceeding those obtained in small-scale experiments. Overall yield at present is 90% with prospects for improvement by another 2%. Total decontamination is 8×10^7; this is well in excess of the value of 10^7 that was the original aim. (2) Sugarman is still working on the cross section determination for U^{236}. He gave tentative tables on some new fission isotopes, giving half-lives, chain assignments, and number of known decays in the chain. (3) Burton reported on some of the radiation problems in milking La^{140} from the very intense Ba^{140} prepared for Site Y. He spoke on the graphite studies. The resistance change has leveled off at Site W, but the energy storage is still increasing. The side holes have 43 cal/gram and the papoose holes 54 cal/cm. (4) Cannon talked about tritium production from neutron irradiation of lithium. The ratio of tritium to hydrogen is 1/1.3 and is still increasing but very slowly. Production rate is 4 ml of tritium per month. (5) English gave a report on the problem of the composition of converter and breeder homogeneous pile solutions. The requirements are minimum neutron absorption, stability of solution, and reasonable solubility of plutonium. (6) Davies spoke about the correlation of theories of co-precipitation with results on various substances. (7) Eastman, reporting for Berkeley, told of a new program of working on the metallurgy of metal on a 200-gram scale with cerium as a stand-in. He said that Connick and McVey found that an investigation of the reaction of H_2O_2 with plutonium shows it to reduce Pu(VI) to Pu(V), then to Pu(III), and then it oxidizes this to Pu(IV). He also reported that Calvin has shown that when he uses TFA for extracting plutonium from Site X slug material, with oxidations and reductions utilizing the +4 and the +3 states, 80% plutonium is recovered with decontamination factors of 3.8×10^5 for beta particles and 3.7×10^4 for gamma-rays. (8) Newton from Ames discussed the use of the feron complex as an analytical tool for plutonium and uranium (colorimetry).

Albaugh sent a memo to Daniels giving the problem abstracts for the period February 15 to March 15 for Sub-section I, Section C-I.

At 7:45 p.m. in Room 209, Eckhart Hall, I attended a meeting of the U^{233} Group (Group 9). Others present were Albaugh, Ames, Asprey, Blaedel, Bradt, Cunningham, Davidson, Dixon, Fineman, Fields, Ghiorso, Gilbreath, Greenlee, Hagemann, Hausman, Hellman, Hindman, Hyde, Hyman, Jaffey, Jones, Katzin, Krueger, La Chapelle, Larson, Manning, Morgan, Phipps, Post, Reinhardt, Robinson, Sedlet, Seifert, Schaffner, Sheft, Simpson, Stewart, Studier, R. Thompson, Van Winkle, Westrum, Winner, Wolf, and others. I announced the meeting tonight will be taken over by the U^{233} group. I then called on Katzin to conduct the meeting. He began by reviewing the methods for obtaining cross sections and fission constants for fissionable isotopes, illustrating the discussion by describing the measurements made on U^{233}, including (a) the experimental determination of the U^{233} absorption cross section by the use of matched absorption cells, one containing pure DNO_3 and the other a solution of U^{233} in DNO_3, and (b) the measurement of the fission cross section by comparing the number of fissions obtained from a known weight of U^{233} (thin film) placed in a neutron beam with the number of fissions observed for a known weight of U^{235} placed in the same beam. When one uses the results of these two measurements, a value for eta of 2.49 for U^{233} is obtained.

Studier described some results on the measurement of the electromagnetic radiations of U^{233} using material that was purified by six successive ether extractions, one peroxide precipitation, and one uranyl acetate precipitation. The purified material amounted to 125 mg of which 60 mg were used for the measurements. Absorption curves were run on a modified Geiger counter, using copper, aluminum, and lead absorbers; these suggest 250, 60, 17, 12, and 8 kev components.

Hagemann described the construction and operation of the one-inch diameter, sixty-inch long ether extraction column being used for the isolation of U^{233} from dissolved, irradiated thorium carbonate slugs. Hellman presented a thermodynamic evaluation of the extraction of uranium into ether.

A bulletin from Pacific Fleet headquarters tells of a battle in Japan's inland seas. American losses are reported to be light without the loss of a single ship.

The Project Council Policy Meeting was held in Room 209, Eckhart Hall from 3:20 to 5:30 p.m. It was attended by Compton, Chapman, Chipman, Daniels, Dempster, Eastman, Franck, Greninger, Hamilton, Harrell, Hilberry, Howe, Huffman, W. C. Johnson, Mulliken, Smyth, Spedding, Stearns, Stone, Tracy, Watson, Wigner, Wirth, and Zinn.

Compton reported on some recent developments in Washington. He reviewed his recommendations of two months ago for continuation of support for current work and for forward-looking research. This program was taken to members of the Military Policy Committee who decided they are not in a position to act on these recommendations because it is not their responsibility to do anything about work that is expected to go on after the war. The Committee did recommend to the Secretary of War that a new committee be appointed to consider our program. Discussion about the proposed committee

has gone on, but nothing definite has been decided.

There was a discussion of the desirability of Compton or someone else going to Bush to ask that he convey the matter to the President, who would then delegate someone with authority to act on the long-range program. Compton stated he has already called Bush, who said he is doing all he can; if nothing happens later on, he [Compton] feels justified in going directly to someone with the power to act. Compton appointed a committee of Zinn, Daniels, and Stearns (as Chairman) to handle matters of allocation of tasks and personnel concerning Met Lab personnel. Stearns indicated they have already started with the chemistry group by getting a preliminary list of personnel to carry on the program after July 1. He has the names and salaries of others for future reference in transfers.

There was a brief discussion of the possible future role of the University in a permanent installation. The meeting concluded with a discussion of retaining the services of some as consultants or through subcontracts and the feasibility of continuing some of the fundamental research at other universities.

Thursday, March 22, 1945

At 7:30 a.m. I held a meeting in my office of the Heavy Isotopes Group. This was attended by Cunningham, Florin, Ghiorso, Hagemann, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Manning, McLane, Morgan, O'Connor, Studier, Van Winkle, and Weissbourd. Florin and I described the results on the uranium plus 40 Mev alpha-particle Berkeley bombardment (sample TαB). O'Connor described a practice run to look for short-lived 94^{241}.

The following activities were scheduled: (a) a run on U^{233} decay products next Monday, (b) further work on gamma-rays of U^{233} by Ghiorso and Studier, (c) a run on $Pa^{241}(n,\gamma)Pa^{232}$ on March 29 or April 2, (d) work on the plutonium plus 40 Mev alpha Berkeley target [sample 49αB] by James, (e) a run by O'Connor the week of April 2 to look for short-lived Pu^{241} by neutron bombardment of high-Pu^{240} material.

James completed working up the residues from 51A (250 mg of Hanford decontaminated and purified 27 gt plutonium) and computed that the total amount of 95^{241} in the 250 mg sample is 15,000 c/m, assuming an 80% yield.

I sent Daniels a list of the personnel and their salaries whom I consider essential for the continuation of our program after July 1, 1945: James ($3180), Morgan ($3360), Florin ($3060), O'Connor ($3000), Magnusson ($3600), Scott ($3000), Cunningham ($5000), Hyde ($3360), S. Thompson (at Site W, $5100), S. Peterson ($3300), R. Thompson ($3780), Westrum ($3660), Simpson ($6800), Ghiorso ($4380), Jaffey ($4260), Kohman (at Site W, ~$4200), Robinson ($4500), Hindman ($4260), Goeckermann (at Site W, ~$3000), Hyman ($3522), Katzin ($4260), Studier ($3060),

Van Winkle ($3000), Gilbreath ($3720), Albaugh ($5000), Blaedel ($4260), Dreher (at Site W, ~$4200), Seaborg ($7200), Manning ($5400), and Perlman ($5400). In addition, six technicians (tentatively, Billington, Erway, Walsh, Giacchetti, and two others), three secretaries, and one clerk. I also include group assignments.

In a memo to Stearns, Daniels states that he thinks it is very unwise for the safety of our nation to curtail in any way the research program directed toward future piles. He also questions what is to be gained by transferring 30 or 40 trained chemists from Chicago to Clinton when all the facilities and leadership are already available here. He urges that a small section be organized to do research on high-temperature oxide pebble piles; if this is not possible he proposes that research on high-temperature piles be undertaken at the University of Wisconsin with University funds; he gives good reasons why the facilities should be located there.

Hawkins wrote to J. O. Pyle, Director of our Security Division, about the possibility that on V-E Day, celebration or civil commotion may endanger the security of the Metallurgical Laboratory. Additional guards are to be on duty at the Laboratory. He recommends that the guards be instructed that "any member of the Metallurgical Laboratory who becomes objectionable while at work, or who comes to work in an unfit condition," should be removed or prevented access to the premises. He expects that undesirable conduct may arise due to indulgence in alcohol.

English, who is visiting from Clinton Labs, and I conferred about the data from Stoughton on our sample irradiations. All samples that have been irradiated since Stoughton took over the supervision are receiving in the range of 65-90% of maximum flux; the maximum is 2.3 times the average flux. Shapiro has found that the maximum neutron flux is 3 to 4×10^5 neutron/sec per watt [determined by gold monitors and by the reaction Pu(n,f)Ba]. For the production of U^{233} and the uranium carbide samples, the flux is 80% of the maximum 3×10^5 nv/watt; this will apply to most future samples. For relative monitoring, silver wire is used (1 inch long, 10 mil diameter).

U.S. troops have entered Ludwigshafen, great German chemical producing center on the Rhine, according to this morning's paper.

Friday, March 23, 1945

At 8:30 a.m. I held a special meeting in my office of the Council of Section C-I which was attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. I opened the meeting by announcing the night number of the telephone in my office has been changed to BUtterfield 1405. I explained this is a prelude to what I am going to say next, which is worse. I then announced that the Met Lab staff is to be cut by 80% on July 1. I said that the War Department is afraid to authorize any more money for any program not directly involved in fabricating the final device. Although they feel the work for a future

program should go on, at least to some extent, they do not wish to stick their necks out to keep this project going. I added that quite probably the War Department will be sorry for this at some future date but, at present, the decision has been made; we will have to adjust to it.

I indicated the Met Lab will have only about 70 academic people left according to present plans; this section will be cut a little less than most of the others, perhaps only 70%. I explained that, in general, our present type of work will no longer be continued. There will be no more work in support of Hanford. Anything we do will be in support of Site Y, and will probably be concerned primarily with Heavy Isotopes.

I described the future lineup of men. There will be four in the Instruments Group and three in the Recovery Group. We would like to keep one or two men on Dry Chemistry and also some on special separation work such as protactinium extraction but these have not been okayed yet The choice of men to be kept has been based primarily on two considerations: (1) those who have had the most experience on heavy isotope work, and (2) other things being equal, the tendency has been to keep the younger men, simply because the limitation of the budget requires we have cheaper men if the same output can be secured. The whole Chemistry Division will have about eight people as non-academic help, including technicians and secretaries.

I mentioned the necessity for much writing between now and July 1 and told the group leaders it is their responsibility to see that the men who are leaving get their papers written so they receive credit for the work.

I outlined the tentative program from now until July 1 for the various groups, mentioning that Albaugh's sub-section will try during the next few months to salvage the redox process by demonstrating it on a large-scale column and that the Instruments Group will build all conceivable useful instruments to use after July 1. Robinson is to start work with the Instruments people immediately. Hyde will be shifted to protactinium work.

I explained that the final decisions as to who will be asked to stay have not yet been made because we have asked for more men than they said we could have; we may be able to inform the men today or tomorrow.

Shifting to a scientific subject, I said it has been found that 95^{241} grows from 94^{241} present in all Hanford-produced plutonium; Pu^{241} seems to be a long-lived beta-particle emitter. The experimental work was done on 250 mg Pu^{239} made at Site W at a 27 gt level (sample 51A) and 120 mg of Pu^{239} bombarded at Site W for a time corresponding to 125 gt bombardment (Fermi Special CW1; Met Lab 49NE and 49NEY). I presented a table based on the data from these experiments, showing sets of possible values for the half-lives of 95^{241} and 94^{241} and for the neutron absorption cross sections of 94^{241} and 94^{240}.

James did no work at the Lab today; his son Marshall Ralph was born.

Cunningham and Katzin sent Daniels the problem abstracts for February 15 through March 15, for Sub-section II and Group 9, respectively.

I received a letter from Lavender enclosing a draft agreement giving the Government (1) a non-exclusive and royalty-free license to the inventions covered by Case 52, (2) all rights to inventions covered by Case 61, and (3) retaining for the inventors title to Case 52. A copy has also been sent to Kennedy at Site Y. Lavender mentioned that President Sproul has not yet advised Bush of the University's approval to the arrangement in connection with the two cases.

Manning received a March 22 memo from Nickson who has noticed a recent considerable increase in the contamination in the filtered air section, particularly in Rooms 2 (Abraham and Sheft), 4 (Florin, James, and Morgan), 27, 31 (S. Peterson), and 36. He suggests that Manning go over the surveys with Jean Wallace of the Health Division and perhaps discuss the situation with the group leaders involved. Also he recalls that the sink in Room 4 is supposed to be used as a disposal sink for all very active material and notes that almost every room at the present time has at least one sink which shows above tolerance readings. He asks to discuss this question.

Stearns summarized for Compton the reorganization of the Met Lab agreed upon today; the research work for the Chicago and Argonne branches of the Met Lab will be grouped under the headings (1) nuclear and pile research in physics at Argonne [40 academic personnel] (2) pile chemistry and metallurgy for Clinton [40 academic personnel] (3) chemistry of heavy elements at Chicago [25 academic personnel], (4) miscellaneous — x-ray, mass spectroscopy, etc. The first three groups would be headed by Zinn, Howe, and Seaborg, respectively. This represents a huge reduction from the 72 scientists presently in my section. Radiation chemistry will be transferred to Clinton Laboratories. Dempster's group and the optical section will be much reduced. Daniels will also oversee group 3, Wigner group 1, Daniels and Wigner group 2, and Ralph Lapp will be placed in charge of service units.

Captain Chapman sent a memo to the District Engineer, Oak Ridge, in support of a request for 200 milligrams of U^{235} to be used by Zinn and my group.

In Washington there is grave concern over the Soviet announcement of the termination of a 20-year nonaggression pact between Turkey and the Soviet Union.

Saturday, March 24, 1945

According to the schedule of the 60-inch cyclotron at Berkeley, the 20 Mev deuteron bombardment of 100 mg of plutonium begins today. The sample was mounted by Morgan on a finely grooved platinum target as PuF_4 and ignited. [The bombardment is to end as of Monday, amounting to about 900 microampere-hours. The Met Lab designation is sample 49DE.]

The alpha-particle bombarded 100 mg Pu^{239} target was received from Berkeley today. James designated the target 49αB and began to dissolve off the plutonium in preparation for separating the transplutonium fraction in a search for new isotopes of elements 96 or 95.

Beatrice Greenberg was hired today to work as a typist.

I received a letter from Wahl at Site Y saying, with reference to my letter of March 14, he doubts that he has additional notebooks of interest. Notebook no. 15 contains only his big kick data and the data in notebook no. 17 does not start until July 1942.

In an informal memo to me Westrum details the procedures followed in preparing the plutonium metal for Site Y using the portion of sample CW1 furnished by Allison. He prepared the memo for my use in responding to queries from Allison.

Manning wrote to Spedding at Ames about furnishing Spedding with one pound of depleted uranium in the form of trioxide. Spedding will convert it to metal for us. Foote will then fabricate three targets from the metal. The isotopic ratio, $U^{238}:U^{235}$, is about 3500 to 1. Manning also sent a memo to Furney to arrange for the transfer of the material.

"3rd Army Over Rhine!" reads today's banner headline. U.S. troops have crossed the Rhine without firing a shot and have established a bridgehead on the short road to Berlin!

Sunday, March 25, 1945

General Groves made a brief stop at the Lab today between train connections. He, Captain Chapman, Captain McKinley, Daniels, and I had a fifteen minute conference in my office. Groves wanted to know why we have not shipped 100 mg, rather than 60 mg of U^{233}, to Site Y. We explained that this was all we were asked to ship. We pointed out that we could get a larger supply and would plan to do so if he desires. Groves explained that he had to know the value of eta with high precision in order to make a proper decision. Zinn has reported it as being between 2.5 and 2.3, but he needs a more precise value. We explained that one difficulty is that we do not have any highly enriched U^{235} with which to make a direct comparison. He suggested that we prepare larger amounts of U^{233} for use at Argonne or Los Alamos to measure its fission properties in order to check the work of Zinn. He also asked us to find out from Zinn the best accuracy attainable at Argonne and to give him an answer by telephone tomorrow morning.

In the afternoon I took the street car to the Evergreen Golf Club to join Tom Morgan, Winston Manning, and Steve Lawroski who had completed 9 holes of golf. I played with them for the last 9 holes. Scores were TM-63, WM-76, SL-58, and GS-62.

Monday, March 26, 1945

At Berkeley the bombardment of 100 mg of plutonium with 20 Mev deuterons is scheduled to end today with 900 microampere-hours. The bombardment started last Saturday.

Early in the morning Daniels and I conferred with Zinn about what can be done to meet General Groves' request for a more accurate value of eta for U^{233}. I said we could prepare within a week, 55 additional milligrams from several laboratory sources. Furthermore, the total present stock could be increased by working up the 15 cans of thorium carbonate now in the Clinton pile, each of which contains 5 mg; this could be done within two weeks after they are delivered to us. For accurate measurements, in addition to lumping all the U^{233} together, it will also be necessary to make available to Argonne a minimum of 200 mg of highest purity U^{235}. We then went to McKinley's office where Zinn talked with Groves by telephone. Groves seems to think it would be best to have the work done at Argonne. He suggests it would be advisable for Zinn and another person, probably me, to go to Site Y for a conference regarding the best means of obtaining quickly an accurate value of eta.

In a front page bulletin in today's paper are reports of complaints of German prisoners. They complain the Allies are not moving fast enough so that the war will end!

Tuesday, March 27, 1945

Zinn, Daniels, and I sent a confirming memo to McKinley about yesterday's discussions with McKinley and with General Groves about the steps to be taken to make more precise measurements of the nuclear constants of U^{233}.

In a memo to Seren at Site Y, I transmitted comments from W. H Sullivan about inaccurate data regarding ruthenium radioisotopes which appear in report CP-2376, "Slow Neutron Activation Cross Sections," November 21, 1944, by L. Seren, H. N. Friedlander, and S. H. Turkel.

Manning sent a memo to Mulliken enclosing, in compliance with a request from Warner, "Evaluation of Separation Processes for Plutonium." This 11-page document gives a historical account of the processes that have been examined (volatility methods, adsorption methods, solvent extraction processes, precipitation processes, and combination processes). He indicates why the first three of these were not chosen, then describes the chosen process (bismuth phosphate plus lanthanum fluoride crossover plus peroxide isolation) in some detail.

In another bulletin, today's paper says Allied tanks are in the Neckar Valley which runs through Heidelberg and into the Rhine at Mannheim.

Wednesday, March 28, 1945

At Berkeley, the helium bombardment of a natural uranium metal target with 40 Mev helium ions is scheduled to begin. The Met Lab designation is sample TαC.

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I mentioned we have shown Stearns the solvent extraction column; he is much impressed and will feature it in his report to the Army this month. I added that in the Redox process, which will be tested in this column, one ends up with three chemical solutions: (1) decontaminated uranium, (2) concentrated fission product solution, and (3) product Pu^{239} solution. Stewart warned that people should get solutions in right away for recovery by his group or they will run the risk of recovering it themselves — we do not know how long we can hold on to our SED personnel.

On the matter of manpower distribution, I said that the personnel shifts I gave last week will take place as planned except that a demand from Groves for U^{233} will shift Hagemann from writing reports to extracting U^{233} from 15 more thorium cans. I mentioned that it will probably be about two weeks before final decisions can be made on just what will happen to the Project. I added that Warren Johnson will be here from Oak Ridge next week to interview those people who might be available to go to Clinton; nothing has been heard from Los Alamos or Y-12.

For the remainder of the meeting, Manning and I reviewed the items of interest brought out at the Project Council Information Meetings on Physics and Chemistry, held last Tuesday and Wednesday.

I read a copy of a memo from Daniels to Stearns reporting that, in accordance with the desire to reduce the SED personnel as much as possible, the 19 SED men now in the Chemistry Division will soon be reduced to 15 men, whom we hope to keep until July 1. After that we hope to be able to keep four or five men who are the most highly trained. The 15 men are highly trained in plutonium chemistry, and many are involved in service tasks. In addition, two men (H. Brownell and Lewis) will be retrained for the "shotgun" tests.

Stearns sent a memo to Compton and Hilberry reporting further on what has been done to date in planning the reorganization of the Met Lab: Howe is appointed Associate Laboratory Director until next July 1. The other positions will be as already indicated. Zinn will continue as supervisor of the pile and nuclear physics at Argonne, Seaborg will head chemistry of plutonium and essential processes, and Daniels will be in charge of pile chemistry and metallurgy and also will be Seaborg's supervisor. With regard to his own position, Stearns says he would rather see Compton or Hogness take over the Laboratory directorship and indicates his willingness to terminate his present duties before July 1 or to continue them temporarily beyond July 1. Stearns also refers to the fact that Daniels, Howe, he, and I are considering what to do with the solvent extraction process. He says he is convinced that

six men should be retained on the process until October 1 of this year, by which time a complete solution to the problem will be obtained.

At 7:45 p.m. I attended a meeting in Room 209, Eckhart Hall, of the Basic Chemistry, Recovery, and Instruments Groups of Section C-I. Others present were Abraham, Ames, Asprey, Cunningham, Daniels, Davidson, Dixon, Egan, Florin, Fried, Ghiorso, Gilbreath, Greenlee, Hagemann, Hindman, Howland, Hufford, Hyde, Jaffey, James, Jones, Katzin, Krueger, La Chapelle, Larson, Lawroski, Magnusson, Malm, Manning, McLane, Morgan, Niedrach, O'Connor, Phipps, Reinhardt, Robinson, Schwob, Sedlet, Sheel, Simpson, Stewart, Templeton, R. Thompson, Van Winkle, Westrum, Wolf, and others. After I opened the meeting and turned it over to Cunningham, James spoke on elements 95 and 96. He began with a reference to Morgan's discussion of this subject at the January 31 group meeting, where it was postulated that 95^{241} and 96^{242} result from short-lived beta-particle decay of Pu^{241} and 95^{242}, respectively. The most significant experiment since that time is that 95^{241} (as identified by a 4.0 cm alpha particle) has been separated from 250 mg of 27 gt Hanford plutonium that had been decontaminated and purified at Hanford (sample 51A). The pure material was allowed to stand for 45 days after which 15,000 alpha-particle counts per minute of 95^{241} were separated. James stated that this experiment gives conclusive evidence that Pu^{241} is a long-lived beta-particle emitter whose radiation is too soft to be detected with the ordinary Geiger counter. James then proceeded to give some values for the n,γ cross sections of Pu^{240} and 95^{241} for assumed values of the half-lives of Pu^{241} and 95^{241}. He continued by saying that very little is known of the chemistry of elements 95 and 96, other than that they are very similar to the rare earths and there is no conclusive evidence for more than one oxidation state (+3). There is, however, some evidence that 95 has a +4 state. James gave the following data on three plutonium samples from which 95 has been extracted and also data on a neutron-bombarded uranium sample:

Sample	*gt Level*	*95 alpha count/mc of beta activity in the rare earth fraction*
49NE	120	610
49ND	25	333
49	5	24
U	~3	1.25

Cunningham introduced Florin who spoke on recent studies of Pu^{240} using sample TαB, the natural uranium target which was bombarded with 40 Mev helium ions at Berkeley. He outlined the steps involved in the separation procedure, mentioning that the first lanthanum fluoride by-product precipitation from oxidized solution was given to James and Morgan who found very little 95 and at first took this as an indication that the reaction $U^{238}(\alpha,n)Pu^{241}$ has a low yield; this is not the case if Pu^{241} is long-lived as we now know it is. Florin interpreted the results of the absorption curves for the neptunium and plutonium fractions as indicating that 4.5% of the activity of the sample is due to Np^{238} and that some of the Pu^{239} formed is produced by the α,3n reaction on U^{238}.

On this basis perhaps one could also expect an appreciable yield of Pu^{236} from the α,3n reaction on the less abundant isotope U^{235}.

Davidson commented on the fact that not much Pu^{238} is formed by the α,n reaction on U^{235}, to which I remarked that study of the amount of Pu^{238} formed by an α,n reaction on U^{235} in bombardment of highly enriched U^{235} would provide a means for determining the yield of the α,n reaction; however, bombardment of Pu^{239} with alpha particles is being done and a determination of the 96^{242} will give the same information.

Cunningham introduced Ghiorso who spoke on some recent measurements on Pu^{240}. He continued discussion of the reactions occurring when uranium is bombarded with high energy helium ions and gave the differential and integral curves Weissbourd obtained in the alpha pulse analyzer apparatus from the TαB plutonium fraction prepared by Florin. Ranges found were 3.7 cm (ascribed to Pu^{239} or Pu^{240}), 4.1 cm (ascribed to Pu^{238}) and 4.4 cm with a four-month half-life (a range hitherto unknown and tentatively assigned to Pu^{237} although Pu^{236} is certainly a possibility). I said that the reason the Pu^{237} hypothesis is favored is that the α,2n reaction should be more prolific than the α,3n. I added that since Pu^{241} is probably long-lived, its radiation is too soft to measure readily. If the amount of Pu^{241} in this sample could be detected, the half-life of 95^{241} could be estimated. Ghiorso also gave the Geiger counter aluminum and lead absorption curves on the sample.

Ghiorso talked about his slow neutron fission measurements on the plutonium fraction at Argonne, which indicate that of the 1140 c/m with range near to that of Pu^{239} only 400 c/m are found to be fissionable like Pu^{239}. If these other 700 c/m are to be ascribed to Pu^{240}, it must be assumed not to undergo slow neutron fission, or to undergo it with a smaller than normal cross section, or to have a short half-life so that it is present in very small concentration.

Daniels asked how long it would take to determine for sure whether or not Pu^{240} is fissionable with slow neutrons. In response I indicated that a new uranium alpha-particle bombardment now in progress at Berkeley should provide the key to this question. The results of the bombardment should be available in six to eight weeks.

Following Daniels' comment on the tremendous importance of knowledge of the fissionability of Pu^{240} to the design and successful operation of breeder piles, I predicted that Pu^{241} will undergo a slow neutron fission and pointed out also that in the series of analogous nuclei, 91^{231}, 93^{237}, 95^{241}, it is known that the first two do not undergo slow neutron fission. In the case of 95^{241}, however, though the even number of neutrons in the nucleus is unfavorable toward fissionability, the high charge-to-mass ratio might be sufficient to cause this nucleus to undergo slow neutron fission. I emphasized that 94^{241} is analogous to 92^{235} and is again therefore likely to undergo slow neutron fission.

I also commented on Pu^{242}, saying that though it would probably not be slow neutron fissionable, it would be the longest-lived isotope of plutonium (maybe 10^6-10^7 year alpha emitter) unless its half-life for spontaneous fission is shorter than this. Pu^{243} is certainly a beta-particle emitter, though Pu^{244} may be either an alpha- or a beta-particle emitter. The isotope 96^{243} may be a beta-particle emitter, though normally

this type of decay would not be expected until 96^{245} or higher is reached. In either case, an isotope of element 96 should be found in a high intensity pile after a year or so of operation. Pu^{242} will also be formed in a pile after a period of operation. I also said decay of 95^{240} and 95^{239} by K-capture is to be expected. These isotopes should have been produced in a target from a Pu^{239}-deuteron bombardment to be worked up by Morgan. They will not be detected, however, if their half-lives are too short or too long.

Ghiorso suggested that as these "new babies" are uncovered, I should provide cigars for the section. Davidson said that a big celebration should follow the identification of element 100. I said that I would gladly sponsor such a celebration. Katzin remarked about the probable difficulty of separating the last three or four members of the actinide series. I countered with a statement that element 97 should follow cerium quantitatively — i.e., it should have +3 and +4 oxidation states. Katzin asked whether or not europium would carry element 98, to which I replied that element 98 probably would not be chemically analogous to europium and I tabulated the rare earth and actinide series as follows:

La	Ce	Pr	Nd	61	Sm	Eu	Gd	Tb
Ac	Th	Pa	U	Np	Pu	95	96	97

I added that the resemblance between elements in the same vertical column is most marked with the general exception that the members of the actinide series, due to larger ionic radius, are more easily oxidized to higher oxidation states than the corresponding rare earths.

Yesterday's paper said that the Japanese claimed the U.S. had not been able to land even one soldier on the Ryukyu Islands. Today's paper says the Japanese claim U.S. forces have invaded the Kerama Islands in the Ryukyu chain.

Thursday, March 29, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Cunningham, Florin, Hagemann, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Magnusson, Manning, Morgan, O'Connor, Scott, Studier, and Van Winkle. I talked on some general considerations of the half-lives, etc., of the heavy isotopes, the importance of plotting decay curves immediately, and on the use of symbols instead of code. Hindman talked on Greenlee's and Ames' determination of the specific activity of plutonium samples of different gt levels. Studier discussed U^{233} gamma- and x-rays.

The following activities are scheduled: (1) Jaffey and Q. Van Winkle to make runs on U^{233} decay products and on $Pa^{231}(n,\gamma)Pa^{232}$ this week. (2) James to start chemical operations on sample 49αB [100 mg plutonium plus 40 Mev alpha particles Berkeley target]. (3) O'Connor to experiment tomorrow to look for 94^{241}. (4) Hindman and his people to continue the work on the specific activity of 2 gt plutonium, 25-45 gt plutonium, and sample 49NE [plutonium from Fermi Special sample CW1] to

look for evidence for Pu^{240} alpha particles. (5) Morgan to work on the 49 + 20 Mev deuterons Berkeley target [to be received tomorrow — Met Lab sample 49DE]. (6) Hyde to prepare to work on the thorium plus deuteron target to be ready when it arrives.

I received a five-page memo from Fried and Davidson giving the results of ignition of U_3O_8 in oxygen to prepare UO_3. With Zachariasen's help, they have shown three phases of UO_3 to exist — the hexagonal phase I, a red phase of unknown structure (phase II), and a bright yellow phase (phase III). The stability of the phases appears to increase in going from phase I to phase III.

I read a copy of a memo from Zachariasen to H. H. Anderson giving the results of examination of samples of double fluorides of Pu(IV) with cesium, ammonium, rubidium, potassium, sodium, lithium, and hydrogen. He could not comment on the cesium sample but indicated that the samples with ammonium, rubidium, potassium, sodium, and lithium all contain as a major component a phase isomorphous with the compound believed to be K_2PuF_6. He said the hydrogen-plutonium fluoride sample contains only one phase, probably KPu_2F_9.

Zinn called me about a teletype he received from Site Y, stating the half-life of U^{233} to be 1.67×10^5 years. The 61-mg sample of U^{233} will be returned here from Site Y tomorrow morning. Fermi made his measurements at Site Y on pressed U^{233} oxide powder in the form of a plaque. He determined the neutron absorption cross section to be 560 barns (compared with 513) and the fission cross section to be 475 barns.

Daniels, Hilberry, and I conferred about arrangements to get the 15 cans of irradiated thorium carbonate from Clinton so we can start as soon as possible to get the additional U^{233} to meet General Groves demand for a more accurate neutron absorption cross section. Since Captain McKinley has not yet made arrangements to get the 15 cans of neutron-irradiated thorium carbonate from Clinton, Hilberry phoned Whitaker and asked that the cans be shipped at once. We have started to assemble all of the U^{233} available in our section.

"Allies Flank Ruhr!" reads today's two-inch headline: British tanks loaded with British and U.S. infantry broke out north of the Ruhr Valley in a sweep eastward through collapsed German resistance.

Friday, March 30, 1945

The 100 mg plutonium plus 20 Mev deuterons target arrived from Berkeley. It received 900 microampere-hours from March 24 to March 26. The sample was turned over to Morgan who observed that some of the plutonium jarred loose in transit. An autoradiograph of the target indicates that the deuteron beam irradiated about 80% of the total plutonium. Morgan dissolved off part of the central portion of the target and began a series of oxidation cycles to separate the 95-96 fraction from the plutonium. The sample is designated 49DE-1.

James finally succeeded in dissolving all the plutonium from target 49αB (100 mg Pu^{239} plus 40 Mev alpha-particle Berkeley bombardment). He began oxidation cycles to separate the 95-96 fraction from the plutonium.

In a memo to Stearns, I reiterated our interest in having a special run made at Hanford to recover Np^{237} from the bismuth phosphate extraction supernatant. I reviewed the needs for this material, in addition to the borderline requirements of Site Y, mentioning that Zinn would find some of it useful as a threshold energy detector of neutrons (via induced fission), as would Clinton Laboratories; in addition Np^{237} can serve as a source material for preparing pure Pu^{238} (a component of Hanford plutonium that should be investigated); also, chemists throughout the Project need the long-lived Np^{237} for the study of chemical properties; finally, this isotope is important from the standpoint of broadening our understanding of the properties of heavy isotopes in general. I told Stearns I understood there will still be time, provided the run is authorized by April 15, to have the run take place before the regular daily production batches start at the second chemical extraction plant, Plant B.

I wrote to Allison, referring to two letters he wrote March 21, one of which commented on our proposal to obtain Np^{237} from a special Hanford chemical plant run. I quoted the information by Westrum on how he prepared the metal for Y from the portion of Fermi Special Sample CW1 which Allison had sent us. I expressed our pleasure at how well the values of Segrè and Deutsch on the x-ray and gamma-radiations from Pu^{239} check our values and mentioned the improved counting yield we are getting in such studies by using Geiger tubes filled with xenon. I then quoted our latest data on the electromagnetic radiations from U^{233}.

In addition I gave him additional details of my proposal to measure the cross section of $U^{236}(n,\gamma)U^{237}$ by measuring the amount of Np^{237} formed. We would look directly for Np^{237} in the second of Fermi's U^{235} samples from the Hanford pile, formed in the set of reactions: $U^{235}(n,\gamma)U^{236}$, $U^{236}(n,\gamma)U^{237} \rightarrow Np^{237}$.

I commented on Site Y finding a spot at mass number 241 in the mass spectrographic study of the metal from Sample CW1, suggesting the possibility that there might have been time for sufficient 95^{241} to grow into the sample from the Pu^{241} present in it. I said that we believe that Pu^{241} is long-lived since we have found the 4-cm alpha particle activity in completely decontaminated plutonium, and that we are looking for very soft beta particles that might be due to Pu^{241}.

I reviewed our results of measurements on the uranium plus 40 Mev helium ions Berkeley bombardment and the possibility that part of the 3.7 cm plutonium alpha-particle activity found may be due to a non-fissioning Pu^{240}. I said we were attempting to determine the amount of Pu^{240} by looking for spontaneous fission counts and asked him what chance there is that Pu^{241} is responsible for part or all of the spontaneous fission.

Manning sent D. L. Collins in the Area Engineer's Office the reasons why it would be impossible to replace Albaugh, Cunningham, Jaffey, Katzin, Lawroski, Robinson, and Seifert for present and future work in Section C-I.

J. H. Lum of Monsanto Chemical Company in Dayton, wrote to Daniels saying that the Mound Laboratory can use 19 additional chemists of varying degrees of experience; they are desired by May 1 if possible.

Daniels called me about a call from Tom Jones that the 15 thorium carbonate cans pushed from the pile this morning are exceedlingly hot and he wondered if they could be handled by the technicians for immediate shipment to us. I assured Daniels that the material will cool very quickly. I also conferred with Dr. Rose in the Health Division and convinced him that we are prepared to handle the hot material as soon as it gets here.

I received a copy of a memo dated March 28 from Perlman to the file, giving the maximum decontamination of 25 gt plutonium that can be obtained considering irradiated uranium that has cooled for 30-60 days. The limit is set by the radiations from plutonium itself. The maximum beta-particle and gamma-ray decontamination factors are 2.2×10^7 and 7.1×10^8, respectively. He points out that in the February series of Site W runs the present separation process has delivered material that is essentially completely decontaminated.

More two-inch high headlines in today's paper read "Yanks Cut Off Ruhr." Soviet troops are reported to be inside Austria in a drive toward Vienna.

Saturday, March 31, 1945

I read a copy of a memo from Daniels to Nickson stressing the importance of constructing the new hot laboratory in the filtered air section of the New Chemistry Building in order to handle safely the increasing radiation intensity of our samples.

Tom Jones sent a formal request to Furney about the administrative arrangements for the transfer of the 15 cans of irradiated thorium carbonate from Clinton to the Met Lab. He also suggests that fresh thorium carbonate cans, now at Clinton, be put in the pile in place of the cans removed.

More banner headlines! Today's reads "40,000 in Ruhr Trap!" A captured German officer is quoted as saying there is no organized German defense between the Ruhr and Berlin.

Easter Sunday, April 1, 1945

Fred Albaugh, Roy Thompson, and I played 18 holes of golf at Evergreen Golf Club. Scores were FA-138, RT-139, GS-113.

Monday, April 2, 1945

Herman Robinson started his vacation. He will return April 14.

Kohman wrote to me from Hanford asking about the propects of rejoining my section in Chicago. He said that Iz Perlman and Stearns have brought the news of the coming reorganization of the Met Lab, and it appears that there will be a reorganization of his section at HEW at about the same time.

I prepared an organization chart of Section C-I as of April 1, which appears as follows:

Glenn T. Seaborg — Section Chief
Edrey Albaugh — Secretary to Seaborg

Winston W. Manning — Associate Section Chief
Jane Horwich — Secretary to Manning

T. O. Jones — Assistant to Section Chief
Mary Williams — Secretary
Beatrice Greenberg — Typist

Sub-section I
Separation Processes

Frederic W. Albaugh — Assistant Section Chief
Dorothy Black — Secretary to Albaugh

Group 3 — Process Development

James R. Gilbreath — Group Leader
Walter J. Blaedel — Research Associate
Rexford Bradt — Research Associate
Herbert Hyman — Research Associate
Horace Hopkins — Research Assistant [SED]
Roy Post — Research Assistant [SED]
Jacob Sedlet — Research Assistant
Mathew T. Walling — Research Assistant
Bernard Winner — Research Assistant
Mildred Bolden — Secretary
Pearline Boykin — Technician

Group 4 — Solvent Extraction

Stephen Lawroski — Group Leader
Clark J. Egan — Assistant Group Leader
Irwin J. Schaffner — Research Associate

Wilbur O. Simon — Research Associate
Milton Ader — Research Associate
Eugene A. Hausman* — Research Assistant [SED]
Alec Kelley — Research Assistant [SED]
Richard Reinhardt — Research Assistant
John H. Schraidt — Research Assistant [SED]
Dan J. Dorcy — Draftsman
Olga Giacchetti* — Technician
Betty Murray* — Technician
Jerome Krinsky** — Technician
Mildred Summers — Technician

Sub-section II
Basic Chemistry and Service

Burris B. Cunningham — Assistant Section Chief
Ruth P. Rogers — Secretary

Group 5 — Basic Dry Chemistry

Oliver C. Simpson — Group Leader
Norman R. Davidson — Assistant Group Leader
Bernard Abraham — Research Associate
Sherman Fried — Research Associate
Thomas E. Phipps — Research Associate
Ralph Seifert — Research Associate
Edgar Westrum — Research Associate
Irving Sheft — Research Assistant
Norman Erway — Glassblower
Helen Thomson — Technician

Group 6 — Basic Wet Chemistry

James C. Hindman — Group Leader
Jerome J. Howland — Research Associate
Leon O. Morgan — Research Associate
Donald Ames — Research Assistant [SED]
Jonathan Dixon — Research Assistant
Alan E. Florin — Research Assistant
Roy W. Greenlee — Research Assistant
Ralph A. James — Research Assistant
Theodore J. La Chapelle — Research Assistant
Lawrence B. Magnusson — Research Assistant
C. Keith McLane — Research Assistant
Paul R. O'Connor — Research Assistant
Sigfred Peterson — Research Assistant
Betty Mokstad — Technician

Group 7 — Recovery

Donald C. Stewart — Group Leader [SED]
Herbert H. Anderson — Research Associate [SED]
Larned B. Asprey — Research Assistant [SED]
J. W. Britain — Research Assistant [SED]
Paul Fields — Research Assistant

*Half-time
**Part-time

Phillip Fineman — Research Assistant [SED]
Olga Giacchetti — Technician (half-time)

Group 8 — Instruments and Physical Measurements

Albert Ghiorso — Group Leader
Arthur H. Jaffey — Research Associate
Albert C. Krueger — Research Associate
Herman Robinson — Research Assistant
Walter C. Beard — Research Assistant
John A. Crawford — Research Assistant
John Dorsey — Research Assistant
Duane Hufford — Research Assistant
Benjamin Scott — Research Assistant
Patricia Walsh — Research Assistant
Bernard Weissbourd — Research Assistant
H. Billington — Technician
Virginia Towle — Technician

Group 9 — 23 Work

Leonard I. Katzin — Assistant Section Chief
French T. Hagemann — Research Associate
Nison N. Hellman — Research Associate
Raymond G. Larson — Research Associate
Martin H. Studier — Research Associate
Michael Wolf — Research Associate
Earl K. Hyde — Research Associate
Quentin Van Winkle — Research Assistant
Marian Pinckard — Technician

No group assignment — Protactinium Work

Roy C. Thompson — Research Associate
Eugene Hausman — Research Assistant [SED] (half-time)
Bernard B. Brody — Research Assistant
John G. Malm — Research Assistant

Non-academic Service Group

Kathleen Florin — Supervisor
Elsie Mae Freeman — Laboratory Assistant
Lillie May Porter — Laboratory Assistant

Total number in Section, 4/1/45: 95

Sub-section I	25.5
Sub-section II	62.5
G.T.S. and others	7
	95
Academic employees	73
Non-academic employees	22
	95

The discontinuance of Group 1 (Extraction-Decontamination) is reflected by its absence from this list.

Hogness issued a summary of the manpower distribution in the Chemistry Division. It shows the following for my section:

		Number of Men	
		Feb.	Mar.
Albaugh (Separation Processes, 27 men)	Thompson, extraction and decontamination	6	6
	Gilbreath, process development	7	7
	Lawroski, solvent extraction	13	12
Cunningham (Basic Chemistry, 36 men)	Simpson, high vacuum work	9	9
	Hindman, basic chemistry	11	11
	Stewart, recovery	6	6
	Ghiorso, instruments & physical measurements	9	9
Katzin (23 Work, 8 men)	23 work	7	7
Administration	Seaborg, Manning, Jones, Albaugh, Cunningham, and Katzin	6	6
		74	73

In the late afternoon, Daniels, Arnold, and I met with Kennedy for a few minutes. He is here from Site Y to interview chemists for jobs there. Tomorrow Kennedy and I will confer about Patent Cases 52 and 61.

Word has reached us that construction of the Hanford Engineer Works is completed and it is now engaged in total operation. Yields of plutonium in the March runs in the 200-T Area continue to be very satisfactory. The losses in the canyon (221 Building) average about 3%, in the crossover cycle (224 Building), about 4%, with an overall decontamination factor of about 3.4×10^7. The plutonium peroxide isolation procedure in the 231 Building has operated very satisfactorily.

There was a Chemistry Division seminar at 7:45 p.m. in Room 251, Ryerson Laboratory. Lawroski and Blaedel spoke on the solvent extraction process.

U.S. troops have won a toehold on Okinawa Island in the Ryukyus.

Reports indicate that this has been the warmest March in local weather bureau history. Now heavy frosts are predicted and are causing worry about ruining crops that have made an early start.

Tuesday, April 3, 1945

Fifteen cans of irradiated thorium carbonate were received from Clinton Labs to be processed here on an urgent basis in order to recover the U^{233} for the measurements General Groves wants.

The Area Engineer's Office sent a memo to the District Engineer in Oak Ridge, requesting continued deferment for Robinson, Seifert, Jaffey, Lawroski, Katzin, Cunningham, and Albaugh.

Kennedy and I conferred about Lavender's proposal that there should be an agreement among Segrè, Wahl, Kennedy, and me about the sharing of rights in Cases 52 and 61. I took the occasion to give Kennedy a letter that I have already prepared for him but had not mailed, suggesting that if he, Segrè, and Wahl will give me their comments on the Lavender draft agreement I will use them as the basis for an answer to Lavender. In the letter, I add that the Patent Office here is preparing a very broad Case S-52. It looks quite good and may cover so much ground that it will have to be split into a number of smaller cases. Because of this broad coverage I repeat my earlier request for additional notebooks, including Wahl's notebook no. 17. We discussed some key points and agreed that I will prepare a draft agreement and send it to him at Los Alamos.

I spent considerable time planning the personnel cutback for this coming summer, reviewing the list of my section members and considering a possible reorganization involving transfer of some of my men into another section. The continuing Section I — headed by me and assisted by Manning and Cunningham — would be comprised of the heavy isotopes men (11 men), Ghiorso's and Katzin's operations (4 men each) plus Simpson and Westrum, and 9 SED men. A new Section II would include Lawroski's solvent extraction group (6 men) and a high temperature pile group (5 men) which Gilbreath, Walling, and Crawford would join. I also made separate lists of the single and married people at Site Y and at Mound Laboratory — and I marked the lists with possible transfer locations: Sites Y, X, Mound, and Y-12.

As usual Helen attended her chemistry class at YMCA College this afternoon.

U.S. troops have advanced half-way across Okinawa according to today's paper. British and Canadian troops are closing in on German troops in Holland.

Wednesday, April 4, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I. It was attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey (30 minutes late), Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I opened the meeting by announcing that it will be possible for those leaving July 1 to take vacations with pay after July 1. Since maximum work can be obtained under such an arrangement, everyone who is leaving should be urged to take a terminal vacation. I brought up the question of working space for S. Peterson and McLane for their actinium work. We decided that they should work in Room 31, leaving Rooms 1 and 12 for protactinium work.

I then introduced for general discussion a suggestion made by D. N. Hume of Clinton Laboratories, who has proposed the terms "radiometric analysis" and "radiochemical assay" to distinguish between simply "weighing" the amount of a given radioactive element by counting its disintegrations

versus determining the percentage of a given isotope present in a mixture by determining the amount of a particular type of radiation in the sample. There was general agreement that the terms are useful. A lengthy discussion of the distinction between "analysis" and "assay" was finally summarized by Jaffey with the statement, "What the hell," and agreement reached that the terms are nearly synonymous.

With respect to the future personnel situation, I could only report that matters are still uncertain. I did say that it is hoped there will be as many as 105 scientific personnel on the Project after July 1. In this case there will probably be two chemistry sections and a physics section, with the first section under me working primarily on heavy isotopes (25 men) and the second section (40 people/metallurgists) devoted primarily to new pile developments. A physics section at Argonne under Zinn might number perhaps 40 people. I also mentioned that Kennedy and Dunlap from Site Y and Brown and Johnson from Site X have been here this week seeing what they can "pick up from the local disintegration."

I wrote to Kohman at Site W, saying that we are including him among those few for whom we are able to make places here at the Met Lab. I added that we are also making a place for Stan Thompson and will have a place for Goeckermann if he wants it. I mentioned that we are giving as careful attention as possible to such former Met Lab men as Hoekstra, Pye, Lincoln, and Yett.

I sent a letter to Kennedy at Site Y enclosing a draft of a letter to Lavender describing the arrangement with the Government that we desire concerning our Patent Cases S-52 and S-61, which I prepared on the basis of my discussion with him yesterday.

At 7:45 p.m. I attended a meeting in Room 209, Eckhart Hall, of the Separation Processes Sub-section of Section C-I. Others present were Ader, Albaugh, Asprey, Beard, Blaedel, Daniels, Egan, Fields, Fineman, Gilbreath, Hopkins, Hyman, Kelley, Lawroski, Manning, S. Peterson, Phipps, Post, Schaffner, Sedlet, Sheft, Stewart, Walling, Winner, and others. Blaedel gave an outline of the Redox process under the operating conditions recommended for the semiworks tests of the process, pointing out that the conditions were chosen solely on the basis of batch data and cannot be regarded as final. The batch data indicate that it should be possible to obtain plutonium yields of greater than 99%, decontamination of 10^6, and uranium recovery yields of 99%.

Walling described the batch experimental procedures used to estimate the decontamination that could be obtained; the experimental effort was directed largely toward simulating the operation of the scrub sections as these play the major role in decontamination. Decontamination was reported in terms of "minimum decontamination factor" (MDF) which is the product of the actual total decontamination and the fraction of the plutonium recovered; on this basis the MDF, through three solvent extraction columns, is 8×10^5 for beta particles and 1×10^6 for gamma-rays.

Post reported on uranium and hexone recovery in the Redox process. Preliminary experiments indicate that the uranium can be removed from the hexone effluent by washing countercurrently with either water or dilute

HNO_3, whereas the hexone can be purified by a steam distillation.

Today's paper reports that the Japanese are still strangely backward in defending the important island of Okinawa.

Thursday, April 5, 1945

E. O. Lawrence is visiting the Met Lab.

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Cunningham, Florin, Ghiorso, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Magnusson, Manning, McLane, Morgan, O'Connor, S. Peterson, Scott, Studier, R. C. Thompson, Van Winkle, and Weissbourd. McLane described his actinium carrying experiments — isolation of $MsTh_2$ for use as a tracer in carrying by bismuth phosphate and $La_2(C_2O_4)_3$, etc. Morgan reported his work on the plutonium plus deuterons target (sample 49DE); he has found a count of a few hundred alpha particles per minute and maybe some x-rays in the rare earth fraction. O'Connor described a practice bombardment of Pu^{240} plus neutrons at Argonne to look for Pu^{241}. Using the ether column for rapid plutonium separation, he obtained a decontamination factor of 150 and fair recovery in about one-half hour. He found a 12- to 20-hour activity — perhaps it is due to ruthenium. James described his work on the target from the Berkeley bombardment of plutonium plus helium ions (sample 49αB). He finds about 70,000 alpha-particle c/m and also x-rays in the rare earth fraction.

The following projects are scheduled: (a) a run on U^{233} decay products, (b) Hindman to work on specific activity of 2 gt, 45 gt, and sample NE [part of Fermi Special sample CW1, 100-120 gt equivalent] plutonium samples, (c) Van Winkle and Jaffey to work on the thorium plus deuterons target, (d) O'Connor to carry out the Pu^{240} plus neutrons irradiation at Argonne with 50 mg of 45 gt material on Wednesday, April 11. At the next meeting Jaffey will talk on the $Pa^{231}(n,\gamma)Pa^{232}$ run.

Range curves taken on a sample of the rare-earth fraction separated by James from sample 49αB (100 mg plutonium plus 40 Mev helium ions Berkeley bombardment) show alpha particles of two ranges: 4.02 cm and 5.07 cm. The 4.02-cm range James ascribes to 95^{241}. The 5.07-cm range he ascribes to 96^{241} or 96^{242}. He suggests that this last isotope may or may not be the same one we found in sample 49ND (Clinton pile irradiation of plutonium) and sample 49NE (Hanford pile irradiation of plutonium), which we are quite sure is 96^{242}. At that time a range of 4.85 cm was assigned to this activity, but it may actually be identical with the one just measured. The value 5.07 cm is believed to be more accurate.

Two lists of Met Lab personnel were drawn up. One shows all the academic and technical personnel and identifies possible retention at Met Lab or sites to which they may be transferred (Argonne, Clinton Labs, Los Alamos, Mound, and others). The second is a list of academic personnel available for transfer, showing salaries, family status, and the sites to which they may be referred for transfer.

Helen went to her chemistry class at the YMCA College.

One of the most severe spring storms in years deposited as much as 17 inches of snow on Nebraska, Iowa, Minnesota, and northwestern Wisconsin today.

Friday, April 6, 1945

More alpha-particle counts were taken on the rare-earth fraction separated by James from sample 49αB (100 mg plutonium plus 40 Mev helium ions, Berkeley bombardment). The alpha-particle activity appears to be decaying with a half-life of about one month. Weissbourd finds that the peak in the sample is distinctly different from the one produced by neutron bombardment. James concludes they are different isotopes and suggests that the longer-range activity is either 96^{241}, produced by an α,2n reaction, or 96^{240}, produced by an α,3n reaction. Another possibility, James thinks, is that the 4.85-cm-range activity observed in neutron-bombarded plutonium is not element 96 but an alpha-emitting 95^{242}.

Morgan completed the isolation of a few hundred alpha-particle c/m in the rare earth (element 95) fraction from sample 49DE-1 (100 mg plutonium plus 20 Mev deuteron Berkeley bombardment) which he began working up on March 30. He made measurements of the electromagnetic radiations from the sample and resolved them into three components of energies about 400 kev, 50 kev, and 30 kev. He considers that these radiations are either secondary x-rays or soft fission gamma-rays, although the 50 kev component could be a gamma-ray from a rare earth.

Captain Chapman in the Area Engineer's Office called to inform me that the given number of grams in our U^{235} sample is in the form of UO_3 (74.5% uranium). The sample is 82.8% U^{235}. (Hence there are about 400 mg of U^{235} according to my calculations.) Chapman asked that future requests for Berkeley bombardments be cleared through the Area office.

Morning and afternoon discussions were held between the Laboratory Office and the Project Office to arrive at a reasonable allocation of the $1 million available to finance the physics and chemistry programs for the year starting July 1. The afternoon session was attended by Compton, Bartky, Wilkinson, Zinn, Stearns, Hilberry, Branch, Daniels, and Howe; and the work to be done in each unit in the Laboratory was discussed. The make-up of the Chemistry Division is now pictured by Daniels as follows:

Section I — Heavy Elements

Group 1 — Heavy elements	12.75	man-years
Group 2 — Instruments	4	man-years
Group 3 — 23 work	4	man-years
Group 4 — Dry preparations	2	man-years

Section II — Pile Chemistry

Group 1 — Fission products	5	man-years
Group 2 — High temperature piles	4.5	man-years

Group 3 — Analytical chemistry	8	man-years
Group 4 — Solvent extraction	1.75	man-years
(to be closed out October 1, 1945)		
Group 5 — Radiation chemistry	4	man-years

After some discussion of the problems to be studied in this Division, Compton felt that no activity could be reduced without impairing some function needed under the directive with the possible exception of those connected with radiation chemistry. Here it might be possible to reduce the group to one man backed up by a consultant.

The Chicago area came through the first spring frost unscathed. We had only a few flakes of snow here.

Saturday, April 7, 1945

I received a teletype from Perlman at Site W saying that our sample W-3 was found to be mainly 36 gt material. It could not have had more than 3% of 25 gt material rather than the 7% figure that was given to him. He will be able to give me more precise specifications later on. Pushing date was December 20 of last year, the last phosphate by-product precipitation was February 13-23, and the last peroxide precipitation was March 4. He asks that I tell Stearns and Hilberry that these are his unofficial estimates.

I wrote to Stan Thompson at Hanford saying that I hope he can rejoin my chemistry section here at the Met Lab as early as next month. I suggest steps that might secure his release from Hanford. In the letter I also discuss future possibilities for Leonard Dreher and Vance Cooper, as well as Hoekstra, Pye, Yett, and Lincoln.

In a memo to Nichols, Compton asks that a supply of U^{235} and Pu^{239} be maintained at Chicago in order not to interfere with production by frequent requests for small quantities. He proposes that 50 grams of U^{235} of about 80% purity be delivered by April 16 and that 25 grams of Pu^{239} be delivered on April 15, with another 40 grams on August 15. Compton states that the present supply of Pu^{239} is 8.6 grams at the Met Lab, 17 grams at Clinton Labs, and less than one gram at other Project laboratories. Also, the entire Metallurgical Project has only one gram of 80% U^{235}.

The Metallurgical Laboratory Report for March 1945 (MUC-JCS-254) has been issued by the Laboratory Director's office. The report describes the reorganization plans to allow a reduction in personnel between now and July 1. It is planned to group the activities of the Laboratory around the work at Argonne, which will be supervised by Zinn; a section that will make studies in the fields of chemistry and metallurgy in connection with future piles, which will be headed by Daniels; and another section that will deal with chemistry and processes basic to the successful operation of the recovery and purification of plutonium, which will be under my direction. It is anticipated that about 100 scientists will comprise the three sections.

The report states that J. P. Howe has been appointed Associate Laboratory Director to assist in the reorganization of the Laboratory. On the scientific side, it states in the summary that the solvent extraction process is giving very satisfactory results and the opinion is expressed that if this investigation can be continued until October, a better plutonium separation and decontamination process will result.

Radiation surveys by the Health Division for the week ending April 7 show unsatisfactory contamination levels in Room 31 (S. Peterson and McLane) and Room 37 (Stewart's group).

Late in the afternoon, Foster York and I played nine holes of golf at the Evergreen Golf Club (scores were FY-56, GS-57). Foster drove us to the course in his car.

Japanese forces are fighting back in Okinawa and have opened a delayed air attack on the U.S. Okinawa fleet.

Sunday, April 8, 1945

I played golf with Arnold, Lawroski, Katzin, Egan, and Morgan at Evergreen. It was a very complicated match — Seaborg, Lawroski, and Egan vs. Arnold, Morgan, and Katzin. Scores for 18 holes were LA-101, SL-124, and GS-113; additional 9 holes, LK-58, TM-65, SL-47, and GS-57.

Monday, April 9, 1945

Perlman sent a teletype to correct the information about sample W-3 in his teletype of last Saturday: 30% is 25 gt level according to latest information. Pushing date of the material was November 23; its processing history is very complex, but the final peroxide precipitation was on March 4. He will have more accurate case histories on these and other samples in about a week.

Stearns sent a memo to W. M. Branch, Budd Gore, J. P. Howe, and Wayne Johnson about the reorganization of the Laboratory. It states that selection of personnel for retention is to be completed by this Saturday.

Daniels had a conference with me and other members of our section to go over the schedule for the production of U^{233}. Including the hoped-for 70 mg in the material now being recovered from the 15 cans of irradiated thorium carbonate received from Clinton last Tuesday, we expect we can complete the delivery of 200 mg this coming Thursday. This is ahead of the tight schedule promised General Groves two weeks ago.

The war in the European theater is moving very fast. U.S. army tanks are 128 miles from Berlin. The Soviet army has surrounded Vienna.

Tuesday, April 10, 1945

I read a copy of an April 9 memo from Daniels to Hogness giving the schedule for release of the SED men as approved by Stearns. The schedule calls for retention of the following men until after October 1 for work on the solvent extraction columns: Schraidt, Hopkins, Post, Ader, Hausman, and Kelley. Ames, Anderson, and Fineman are to be released after July. Men to be retained permanently in order of indispensability to the Project are Stewart, Asprey, Schraidt, Hopkins, Post, Ader, Britain, Cressman, and Lewis (last two retained for the "shotgun" tests).

Report CN-2766, Chemical Research — Separation Processes for Plutonium (Seaborg, Section Chief; Manning, Associate Section Chief; Albaugh, Assistant Section Chief), was issued. It contains the following account of investigations for the month of March 1945:

Extraction-Decontamination (R. Thompson, Group Leader). Peterson has investigated two additional points in this study: (1) The possibility of uranyl phosphate precipitation in the relatively low acidity specified for the extraction precipitate, and (2) The relative merits of $NaNO_2$ prereduction and $H_2C_2O_4$Mn(II) prereduction.

Methods for recovering neptunium in the Bismuth Phosphate Process: Peterson and Malm find that the best prereduction treatment for complete carrying of both plutonium and neptunium in the extraction step is $H_2C_2O_4$(0.03 M)-Mn(II)(0.01 M)-$(NH_4)_2SiF_6$ (0.05 M).

Decontamination with respect to neptunium in the Bismuth Phosphate Process. Malm finds that, contrary to previous reports, storage of 40% UNH preextraction solutions does not have an effect upon the carrying of neptunium in the extraction precipitate.

Process Development (Gilbreath, Group Leader). *Analysis of plant dissolver solution for Np^{237}.* Beard finds that the Np^{237} yield in the Clinton pile is about 0.3% of the Pu^{239} yield, although there may be considerable error in the determination.

The use of TFA for analysis of mixtures of Pu(III) and Pu(IV). Greenlee and Winner find that the TFA method can determine the percent of the total plutonium present in a given valence state with an accuracy of about 5%.

Solvent extraction and decontamination — "Redox" process. Blaedel, Post, Walling, and Sedlet have shown by means of successive batch extractions that, in the presence of $K_2Cr_2O_7$, the plutonium in an aqueous phase (with or without UNH) can be extracted completely into hexone. It is also demonstrated that the plutonium in a hexone phase 0.05 M in hydroquinone may be removed completely from this hexone by repeated washing with an aqueous solution 8 M in NH_4NO_3 and 0.05 M in hydrazine. The extraction of uranium from the aqueous phase into hexone is not adversely affected by the presence of hydroquinone, anthranilic acid, and hydrazine. In further experiments on the use of TFA in the extraction and decontamination of plutonium, a gamma decontamination factor of 10^4 has been obtained.

Solvent Extraction (Lawroski, Group Leader). *Hexone extraction for isolation-development* (Egan, Schaffner, G. Bernstein, Hausman, Schraidt,

Simon, and Struminski). The 3-inch I.D. countercurrent solvent extraction column has been dismantled, cleaned, and reassembled with incorporation of a number of changes in its design and that of its accessory equipment. It is planned to use the equipment to study methods of reducing the amount of wash water for re-extraction of plutonium. It is also planned to study the more promising decontamination processes now being investigated in the laboratory.

Solvent extraction for decontamination and isolation — fundamental research (Ader, Brody, Hyman, Kelley, and Reinhardt). In studies of the solvent extraction of a solution of the bismuth phosphate extraction precipitate, improved decontamination has been demonstrated by experiments in which the starting feed was previously oxidized with $NaBiO_3$ and air-sparged to volatilize the ruthenium.

Decontamination of plutonium by solvent extraction (converter piles). Hyman finds that poor decontamination with respect to zirconium may result unless the concentration of salting agent is kept reasonably low.

Report CS-2793, Chemistry Division Summary Report for March 1945, was issued. The Section C-I information in this report appears in the summaries of work prepared by Albaugh (March 19), Cunningham (March 19), and Katzin (March 20) for my use at the March Project Council Information Meeting on Chemistry (March 21).

Helen attended her chemistry class today.

War summaries today read: Western front — Seven Allied armies roll ahead. London — Soviets seize Koenigsberg; 27,000 Nazis surrender. Guam — Heavy Japanese fire holds up Yanks in Okinawa. Manila — Flyers sink Japanese destroyer in China Sea.

Wednesday, April 11, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. I opened the meeting by giving a list of reports and notebooks we cannot trace. I then announced that we will have to turn our entire stock of U^{233} into various forms for Zinn to enable him to determine *eta* for General Groves before April 26, the date of the San Francisco conference. On the future of the Plutonium Project research program, I emphasized the importance of this determination by stating that everyone in Section C-I must be available for helping on it. Since a portion might be in metallic form, Florin will practice the HF dry fluoride method for making the UF_4 needed for metal production. I added that the possibility of making wet UF_4 should not be overlooked as there is the possibility that the dry method may cause some loss of material. This, of course, is something that cannot be allowed to happen. I suggested Howland might be a good man for making the wet UF_4. I said that Fried and Westrum would work on the actual production of the metal. Depleted U^{238} oxide is to be used for practice runs since if we do get metal from these runs, Zinn probably can use it as a blank.

I next brought up the operation of our section after July 1, indicating it will probably be as one big group. With regard to reporting I said that, besides the usual monthly abstracts, I favor reporting results in the Metallurgical Project Record when the work is done and not try to put out a monthly report.

I raised the matter of security violations which are back again and learned that they usually occur when there are no patrollers to make the rounds. I said there definitely has to be someone patrolling all the time. During the discussion which followed, the best suggestion came from Albaugh, whose idea is to single out the group responsible for the violation and have them patrol until they find a violation in another group.

I mentioned the problem of supplies after July 1 when our budget will be so small that we shall not be able to order much. I suggested that nobody turn in any equipment to Surplus Property without the permission of Manning, Jones, or me, and further, that a surplus list be made up within Section C-I to be circulated among those who will remain.

In the matter of operations after July 1, I said that, in general, the men who remain will continue with what they have been doing with the supervisors probably taking over some of the problems they have been supervising up to now or perhaps some other problems. I suggested the following:

Cunningham: (1) Cooperate with Morgan and James in isolating 95 and 96 on a microgram scale. (2) Continue specific activity determinations of Pu^{239}-Pu^{240} mixtures. (3) Make specific activity determinations on 95^{241}. (4) Cooperate in the isolation of actinium (Ac^{227}) on the ultra-microscale.

Katzin: (1) Isolation of pure Th^{229} and weighing [determination of half-life]. (2) Cooperation in the measurement of the slow neutron fission cross section of Th^{229}. (3) Determine the n,γ cross section of Pa^{233}. This is a toughy. (4) Isolation of Pa^{231} and chemistry of protactinium. (5) Continue work on neptunium (4n + 1) decay series. (6) Investigate transmutation produced upon bombarding U^{233} with alpha particles and deuterons.

Hindman: (1) Specific activity determinations (same as for Cunningham above). (2) Spectrophotometric work with the heavy elements. (3) Concentrate on chemistry of neptunium.

R. C. Thompson: Isolation and chemistry of protactinium.

Gilbreath will be on solvent extraction until October 1 and then may be assigned to Daniels' high-temperature pile work. *Simpson, Ghiorso, Stewart,* and *Jaffey* will do about the same as they are doing now.

I urged that all who are leaving be asked to take their vacations starting on June 30, as we need everyone until that date. I said I would like to talk to anyone who insists on taking his vacation before that time.

Fifty mg of 45 gt plutonium were irradiated in the Argonne P-9 pile for a study of the reaction $Pu^{240}(n,\gamma)Pu^{241}$ being carried out by O'Connor.

Last week Daniels received an amusing memorandum from Wayne Johnson, Personnel Director, demanding an explanation of why Edrey Albaugh worked 0.4 hour overtime on March 28 and March 3 (in violation of Illinois law). Yesterday Daniels replied, at my suggestion, that she did so of her own volition for the good of the Project.

Hogness has returned from his special assignment which took him to Washington, London, and Paris. He has been away since January 22.

At 7:45 p.m. I attended the meeting in Room 209, Eckhart Hall of the Basic Chemistry, Recovery, and Instruments Groups of my section. Others present were Ader, Albaugh, Asprey, Blaedel, Cunningham, Davidson, Dixon, Erway, Fineman, Fred, Fried, Ghiorso, Hindman, Howland, Jaffey, Jones, Krueger, La Chapelle, Larson, Lawroski, Magnusson, Manning, McLane, Phipps, Reinhardt, Schwob, Scott, Seifert, Simpson, Stewart, Templeton, R. C. Thompson, Van Winkle, Westrum, and Wolf. As usual I turned the meeting over to Cunningham.

The first speaker was McLane, who outlined the methods used to separate actinium tracer (Ac^{228}, half-life 6.13 h) which occurs with other members of the thorium series. The tracer was then used to search for a selective carrier for actinium that might be used to concentrate larger amounts of Ac^{227}. Bismuth phosphate, iodate, and oxychloride; zirconium phosphate and cerium iodate fail to carry the tracer, but lanthanum oxalate shows promise.

La Chapelle reported the recent progress in neptunium chemistry. This includes the absorption spectrum of Np(III) and the measurement of the formal potential of the Np(III)-Np(IV) couple in 1 M HCl solution (-0.138 v). Following the presentation, I asked for the values of the (III)-(IV) potentials for plutonium and uranium in 1 M HCl. Hindman and Howland replied that they are -0.966 v for plutonium and +0.633 for uranium; this places the neptunium potential very nearly centered between the two. A remark by Davidson that it is interesting that neptunium shows no tendency to favor the odd oxidation state since it is an odd numbered element, led to a discussion of the significance of that general rule.

Van Winkle made a partial report on work he and Jaffey have done on an absorption curve on a very pure gamma-ray from Pa^{232} formed from neutron capture by Pa^{231}. Ad-libbing until Jaffey arrived, I reviewed the purpose of the measurement of the cross section for the reaction $Pa^{231}(n,\gamma)Pa^{232}$. The thorium scheme [$Th^{232}(n,2n)Th^{231} \xrightarrow{\beta^-} Pa^{231}$] is analogous to the $U^{238}(n,2n)$ scheme, even to close agreement in half-lives and energies. It is Jaffey's problem to establish even further analogy by measuring the neutron absorption cross section on Pa^{231} which differs in mass from Np^{237} by exactly six.

Jaffey apologized for being late, explaining that he lost a factor of 100 in a calculation he was making for this meeting and had to redo all of them. He presented the equations and calculation used to arrive at the neutron cross section for Pa^{231} of 290 barns. He said that, as in the case of Np^{237}, this cross section might account for all the U^{232} formed, which experimentally is found to have about the same activity ratio to U^{233} as the Pu^{238} has to the Pu^{239}.

The headline in today's paper reads, "Yanks 110 Mis. from Berlin; Seize 285 Nazi Diplomats." The diplomats to which the paper refers are members of the German foreign ministry staff.

Thursday, April 12, 1945

At Site W, the second Fermi special sample of plutonium is scheduled to be removed from the pile today after 27,800 megawatt-days in a 250-ton pile.

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Crawford, Cunningham, Ghiorso, Hindman, Jaffey, James, Jones, Katzin, Larson, Manning, McLane, Morgan, O'Connor, S. Peterson, Scott, R. C. Thompson, Van Winkle, and Weissbourd. Manning told the group about the hot lab plans and the present situation. We decided to have one hot lab in Room 28 and make Room 33 into the "clean room". Ghiorso described work on soft beta particles from what may be Pu^{241} in the uranium plus 40 Mev helium ions target (sample TαB). Hindman reported on his specific activity determinations on several samples containing Pu^{240}.

Morgan described his measurements on the rare earth fraction from the plutonium plus 20 Mev deuteron target (sample 49DE). He has found x-rays of 20-30 kev, 50-60 kev, and 300 kev. Approximately the same x-rays were found in sample TDC from the uranium plus deuterons target. There is a growth of alpha particles in the rare earth fraction of 49DE, having many different ranges. There is a possibility of fission involving the emission of C^{12} and production of 89^{228}, 89^{227}, 89^{226}, and 89^{225}.

I received a memo from Stearns stating that he has been unable to do anything to further the cause of the special Hanford run for the recovery of Np^{237}.

Hilberry sent a letter to W. B. Harrell, the University of Chicago Business Manager, giving the proposed budget for the Met Lab for the period July 1, 1945 to June 30, 1946. Requested is $6,900,000 of which $3,629,000 is for salaries and wages. The amount proposed for Physics-Chemistry research is $486,000.

Perlman called me from Site W. He will arrive next Thursday morning and stay until 6:00 p.m., Sunday, April 22. He said there will be a meeting with du Pont people next Monday, Tuesday, or Wednesday at the Met Lab at which I can request that Stan Thompson be allowed to return immediately to the Met Lab — in any case everyone will be available for release from Site W by July 1. Perlman promised to request the prints Daniels asked for. A 1 ml sample of dissolver solution perhaps could be sent in their own container. Some might spill; however, this may be better since it avoids a transfer of solution. He told me that the Np^{237} recovery run is being reconsidered.

Perlman commented that Dreher is willing to accept a position at

Site W but has not received an offer yet. Willard and Watt want to come back to the Met Lab.

Perlman then asked if Hoekstra is slated for Clinton Laboratories — I replied I did not know. He said that Kohman has attracted some attention by showing my April 4 letter which discussed Pye, Lincoln, and Yett as well as Hoekstra.

We completed the isolation of the 200 mg of U^{233} today and turned it over to Zinn for measurement of *eta* at the request of General Groves.

As usual Helen attended her chemistry class at YMCA College.

President Roosevelt died at Hot Springs, Georgia, at 3:35 this afternoon. I first learned of this tragic event from Howard Lange who came by to tell me after hearing the news on his radio. All of our people are in a state of shock. The major radio stations turned their attention to this news for the remainder of the afternoon and throughout the evening. President Harry S. Truman was sworn into office about six o'clock (EST) this evening.

Friday, April 13, 1945

I received a letter dated April 10 from Willard at Site W stating that the du Pont Company probably will be willing to release the major portion of Willard's crew by the first of July. He said that he assumes I will have a voice in some of the manpower decisions and so is giving me his personal preferences for work after leaving here. His priorities are: (1) work in my section, (2) work elsewhere on the Chicago project, and (3) he is averse to going to Site X unless some special need or opportunity should arise. He wants to remain on the Project at least until the war is over and there is good reason to return to the University of Wisconsin.

I wrote to Dreher at Site W citing the prospects for his returning to Chicago. I explained that I have saved a place for him pending information as to what he intends to do about the offer from the du Pont Company to stay at Site W which I heard he is to receive and which he may be interested in accepting. I told him to let me know as soon as possible if the offer falls through. I also told him of the meeting here next week involving du Pont and Chicago representatives, at which I hope to learn more about where he stands as far as a company offer is concerned.

In a memo to Compton on the significance of power piles in the research program, Daniels gives the opinion that it is unnecessary to regard power piles as limited to the distant future and that it will be possible to run a battleship or light a city with power generated by a pile within a year or two, or as soon as fifty or more kilograms of plutonium become available as a starting material.

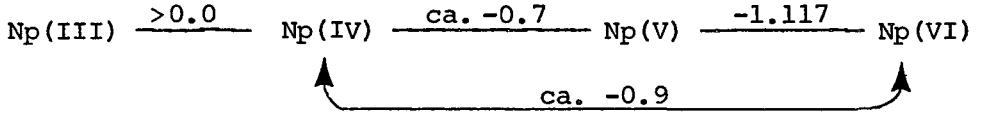
Gore issued an organization chart of the scientific divisions of the Met Lab as of April 13.

Perlman called me again from Site W with the good news that there will be a special Np^{237} recovery run starting this afternoon. The F-10-P dissolver solution will be sent, perhaps 10 days from now. They could ship the 1 cc sample of F-10-P solution in their regular "door stop" container if they decide not to use our container.

Soon after I talked with Perlman, I received confirmation that we will receive the 1 cc of dissolver solution. Stearns phoned me to say that according to a letter he received today from Tilley, HEW will take a 1 cc sample from the first regular run following a receipt of our container which was sent via the Area Engineer offices here to Site W. The container will be returned via the Area Engineer offices. In the future each such sample must be requested separately.

Report CN-2767, "Chemical Research — Basic Chemistry of Plutonium," was issued for the month of March 1945. It contains summaries of the following investigations.

Basic Wet Chemistry Group (Hindman, Group Leader). *Chemistry of neptunium.* Magnusson, Hindman, and La Chapelle, using the isotope Np^{237}, have demonstrated the oxidation states IV, V, and VI for neptunium in aqueous solution by means of spectrophotometric observation and titration with various oxidizing and reducing agents; polarographic evidence has been obtained for the III state in solution (Watters). The characteristics of the absorption spectra of the first three states have been determined. The potential scheme for the various oxidation states in 1 M HCl has been measured:



Noteworthy is the stability of Np(V) with respect to disproportionation into Np(IV) and Np(VI), a notable difference from Pu(V). La Chapelle has remeasured the mean range of Np^{237} alpha particles as 3.27 ± 0.02 cm of air at 15°C and 760 mg Hg pressure. He used a very thin sample of Np^{237} containing less than 0.005% Pu^{239} by weight. Mica absorbers were used and a comparison made with the 3.69 cm range of Pu^{239} alpha particles. The 3.27 cm range corresponds to an energy of 4.77 Mev for the Np^{237} alpha particles.

Recovery Group (Stewart, Group Leader). *Hexone extraction of dissolved bismuth phosphate precipitate.* Britain and Fineman find the method of hexone extraction unsatisfactory for recovery purposes.

Butyl phosphate as a solvent for recovery purposes. Anderson and Asprey find this solvent less selective in its action than hexone. Asprey has started a search for a plutonium-organo compound which can be specifically extracted by one of the ordinary solvents.

Preformed lanthanum fluoride as a carrier for plutonium. Stewart finds that slurries of freshly formed lanthanum fluoride do not adsorb tracer quantities of plutonium from solution — its successful use would have eliminated the etching of glassware. Similarly preformed bismuth phosphate is also unsatisfactory.

Preparation of plutonium compounds. Anderson has undertaken a program of studying pure plutonium compounds in an attempt to explain occasional puzzling behavior of the element as met in recovery work. A number of double salts of the empirical formula M_2PuF_6 have been prepared and examined by Zachariasen.

Hexone extraction for isolation-research. Final concentration of plutonium. Stewart has tested a peroxide precipitation method as a final concentration step in the hexone extraction process and finds it to be satisfactory.

Instruments and Physical Measurements Group (Ghiorso, Group Leader). *Detection of Pu^{238} in various Pu^{239} samples and search for Pu^{240}.* Crawford, using the differential range chamber, has detected small amounts of Pu^{238} in samples of Clinton plutonium of various gt levels. Comparison of the concentration of Pu^{238} in these samples with that in plutonium irradiated at Hanford with slow neutrons shows that this concentration is proportional to the gt level and is not appreciably increased by the Hanford neutron irradiations. Thus the Pu^{238} is produced by a second order reaction involving fast neutrons; the following mechanism is proposed:

$$U^{238}(n,2n)U^{237} \xrightarrow{\beta^-} Np^{237} \quad \text{and} \quad Np^{237}(n,\gamma)Np^{238} \xrightarrow{\beta^-} Pu^{238} .$$

The weight fraction of Pu^{238} is 14×10^{-8} in 2 gt plutonium. O'Connor and Florin have shown that the specific alpha-particle activity of plutonium irradiated with neutrons at Hanford (sample 49NE), with an estimated Pu^{240} content of 0.7%, is the same as low gt plutonium within 5%. This means that the half-life of Pu^{240} must be greater than 3,400 years. Measurements by Crawford have set limits on the intensity of any alpha particles due to Pu^{240} which might have different energies from those of Pu^{239} (3.6 to 3.7 cm range in air); if Pu^{240} decays by the emission by such alpha particles, its half-life can be orders of magnitude greater than 3,400 years.

Radiations from Np^{238}. Jaffey and Magnusson have found about equal numbers of two beta particles having energies of 0.25 Mev and 1.35 Mev. Coincident with the softer beta particle is a 1.3 Mev gamma-ray. There is also evidence of large numbers of a highly converted soft gamma ray of perhaps 0.1 Mev energy following the emission of the 1.35 beta particles.

Half-life of Pu^{238}. Jaffey and Magnusson have measured the growth of Pu^{238} alpha-particle activity from the beta particle decay of Np^{238} and find that the indicated half-life of the daughter Pu^{238} is 56 ± 10 years, in agreement with a direct decay measurement on Pu^{238} of 66 ± 10 years.

The newspapers this morning are all full of the news of President Roosevelt's death yesterday, and the radio stations are continuing with their special programs devoted to his memory.

Saturday, April 14, 1945

At Site W yesterday, the first active metal was charged to the dissolver in the B chemical plant.

Kohman wrote to me from Site W making a definite commitment to rejoin me here in Chicago if he is offered a job.

I replied to the letter from Willard that I received yesterday and summarized the future job situation here. I said I do not know what the chances are of his returning to work in my section, although I would certainly like to have him. I mentioned that perhaps there is a better possibility in the other section of the Chemistry Division that may be headed by Daniels and about which I think Daniels has written him. I then ran through the situation with respect to the other Chicago men now at Site W. (1) We have places for Thompson and Kohman, and probably Goeckermann (because of his relatively small salary and uniformly high recommendations). (2) Indications are that Dreher will stay with du Pont. (3) I have heard reports that Watt is to return to the University of Texas early this summer. (4) Hoekstra, Pye, Yett, and Lincoln are being kept in mind during the manpower discussions going on here.

The simple funeral service for President Roosevelt took place this afternoon at 4:00 p.m. (EST) in the East Room of tne White House.

Sunday, April 15, 1945

I played golf with Steve Lawroski, Winston Manning, and Tom Morgan at Cog Hill Course No. 2. Lawroski and Morgan won our "low ball and low total" match, 8 and 6. Our scores were TM-117, WM-127, SL-103, and GS-102. I sank a 15-foot putt on the 18th green to beat Steve by one stroke.

President Roosevelt was buried this morning in the family garden at his home in Hyde Park, New York.

Monday, April 16, 1945

In Berkeley yesterday, the 40 Mev helium ion bombardment of a natural uranium metal target [Met Lab sample TαC] ended with 162 micro-ampere-hours. The bombardment began on March 28.

Herman Robinson returned to work after a vacation of two weeks.

Edrey Albaugh has decided to quit working. This has left me with the task of finding a replacement secretary. I have asked Ruth Rogers, the very able secretary who has been working with Burris Cunningham, to become my secretary.

I received a letter from Melvin Calvin summarizing the status of

extraction work of his group in view of the fact that there appears to be no one in Berkeley coming to the April Project Council Chemistry Information Meeting day after tomorrow. He told me that Crandall and Thomas have just completed the first TFA run on dissolver solution and obtained a decontamination factor of 2.5×10^6 in two cycles. He also gave me estimates of the costs of manufacture of the diketones as $5 per pound on a large scale. He estimates the amount of TFA required to batch-process a ton of uranium would be 25,000 pounds, not counting any recovery.

Katzin wrote a summary of the work of Group 9, Section C-I, for my use at the Project Council Chemistry Information Meeting. It contains the following information:

The 36-inch counterflow extraction column has been rebuilt to give three times as great flow rates and used to process 20 additional cans of material which were irradiated at Site X at the 4000 kw level for several months. In addition, six cans of material from an earlier irradiation have been extracted, all within about one week. The activity level of each "hot" can was some 40-50 curies of Pa^{233} beta-particle activity, with approximately half that amount of gamma-ray activity.

The U^{233} obtained from the series of extractions, together with other material on hand, was prepared for measurements of physical constants of U^{233} by Zinn. The work included purification and determination of accurate specific activities, as well as making fission foils, making oxide powder for powder experiments and loading the cells and making solutions in "deuterium nitrate" for experiments with the velocity selector apparatus. Similar preparations were made of highly enriched U^{235} and highly depleted U^{238}.

Solvents other than heptyl alcohol found suitable for protactinium extraction are normal octyl alcohol and 2-ethyl hexyl alcohol. Experiments show that the slow neutron capture cross section for Pa^{231} is very high, around 290 barns, indicating that thorium used for production of U^{233} must be very free of Pa^{231} in order to avoid the formation of excessive amounts of U^{232}. It reaffirms the possibility of forming U^{232} in even pure thorium, if there are sufficient fast neutrons present to produce Pa^{231} via the n,2n reaction on thorium.

Albaugh's summary of the work of Sub-section 1 of Section C-I for my use at the Project Council Chemistry Information Meeting covers these topics: (1) TFA has been found unsuitable for analyses of Pu(III)-Pu(IV) in the presence of phosphate. (2) Progress has been made in four aspects of research on the Redox solvent extraction process: (a) Pu(VI) is highly extractable by hexone, whereas Pu(IV) is only moderately so; (b) the most difficult fission products to remove are zirconium, ruthenium, and cerium — neptunium appears to follow plutonium almost quantitatively; (c) beta-particle radiation at five times the expected level produces no adverse effect on the process; (d) tests show that the solutions involved in the Redox process are well adapted to countercurrent extraction. (3) Tests of the newly assembled 3-inch I.D. column show it is easily and almost automatically controlled. The apparatus has been readied for preliminary testing of the Redox process with inactive UNH solutions. (4) Research on the bismuth phosphate extraction-solvent extraction process has included measurement of the distribution ratio for ruthenium between

hexone and aqueous solution. Multi-stage laboratory experiments in which large volumes of active bismuth phosphate feed solutions have been batch extracted give evidence that a decontamination factor of at least 10^7 could be obtained by the use of efficient countercurrent extraction columns in the solvent extraction step.

I read a copy of a memo from Zachariasen to Stearns about a set of uranium sulfide samples, with a sulfur to uranium ratio ranging from 1.6 to 1.2, sent by Brewer from Berkeley. Zachariasen's x-ray analysis shows only two phases, US and U_2S_3. He said that the x-ray patterns are very complex. With the help of these data he can now identify a neptunium sulfide he received previously from Fried as Np_2S_3, isomorphous with U_2S_3 and Th_2S_3.

A list of Met Lab personnel was prepared showing possible retention at Met Lab or sites of transfer. It is an up-to-date version of the one prepared April 5.

At 10:00 a.m. I attended a meeting in Stearns' office to discuss the Hanford manpower situation. Others present were Daniels, Doan, Hilberry, Hogness, Howe, Warren Johnson, Wayne Johnson, Stearns, Whitaker, and Tilley (of du Pont). I reiterated my hope that Stan Thompson be allowed to rejoin my section as soon as possible.

I received a letter from Stan Thompson, dated April 13, in response to my letter of April 7. He wants to know more about the conditions for his return to the Met Lab. I replied immediately to tell him about my meeting on the Hanford manpower situation this morning during which the possibility of his release from Hanford as early as next month was discussed. In the letter I also discuss tentative plans for him to go to graduate school in order to obtain his PhD degree. I also mention the interest the du Pont Company has in retaining the services of Leonard Dreher and my conversation with Tilley urging the Company to clarify Vance Cooper's status.

At 7:45 p.m. there was a Chemistry Seminar in Room 251, Ryerson Laboratory. Boyd spoke on carrying by various chemical compounds and isomorphism.

The Soviets are reported by the Nazis to be 24 miles from Berlin. U.S. troops are 53 miles southwest of Berlin.

Tuesday, April 17, 1945

At 8:30 a.m. I attended the Project Council Information Meeting on Physics. Items of interest reported were: (1) Wollan of Clinton stated that the neutron scattering cross section of Th^{232} has an upper limit of 2.7 to 3.0 barns. New results on the energy of delayed neutrons from fission now agree with the Chicago values. The flux of the Clinton pile is being determined by gold foil monitors. Carbon-14 is now being manufactured by the $N^{14}(n,p)C^{14}$ reaction using NH_4NO_3. (2) Borst of Clinton has investigated the gamma-ray spectrum of 14-hour Ga^{72} and As^{76}.

He also gave some results from neutron crystal spectrometer measurements on highly enriched U^{235} from the beta process at Y-12 and on the elements mercury, europium, and gadolinium. (3) Nordheim spoke on design problems of the 1000 kw small pile which will have five times the thermal flux of a Hanford pile. (4) Hughes of Argonne reported new measurements of delayed neutrons from fission, determination of neutron energies from the γ,n reactions, cross section for the reaction $Cl^{35}(n,p)S^{35}$, and a new cross section for the reaction $Li^{7}(n,\gamma)Li^{8}$. (5) Zinn reported on the continued work on cross sections at the resonance peaks of some of the rare earths. He discussed the measurements on the value *eta* for U using the 200 mg our group has isolated. A value of 2.35 for *eta* was reported to Groves. (6) J. Simpson described a portable alpha-particle counter with varying size probes that will count from a few counts to 30,000 c/m. (7) Zachariasen spoke about the effect of irradiation on Hanford pile graphite. (8) Lewis discussed fission product analysis by means of the mass spectrometer. This offers a powerful means of making mass assignments. (9) Weinberg reported on calculations related to plutonium piles that use neutrons of intermediate energy. He believes *eta* becomes 2.7 at just about 0.4 ev. Under such conditions, breeder operation is possible. (10) Friedman discussed the problem of U^{234} accumulation in U^{233} breeder piles.

James completed the extraction of additional 95^{241} from sample 51A (250 mg of 27 gt Hanford-purified plutonium). He finds that in 30 days 10,000 alpha-particle c/m have grown in. This is the same growth rate observed from the previous extraction from this same plutonium and indicates that the half-life of 94^{241} is greater than (say) two years and confirms that it decays by beta-particle emission to 95^{241}, an alpha emitter of 4.0 cm range.

Cunningham summarized the work of Sub-section 2, Section C-I, for my use at tomorrow's Project Council Information Meeting on chemistry. The memo covers the following subjects:

Heavy isotopes work. Alpha-particle activities in alpha-particle irradiated plutonium, including a 5-cm range alpha particle activity (sample 49αB). Search for plutonium isotopes in alpha-particle bombarded uranium (sample TαB) showing evidence for Pu^{240} because of lack of slow neutron fissionability; measurement of very soft negative particles presumably from Pu^{241}. Study of alpha particle activities in rare earth fraction from deuteron-bombarded plutonium (sample 49DE). The n,γ cross section for Pa^{231}. Unsuccessful search for short-lived beta-emitting isotope of Pu in neutron irradiated samples of Pu of different gt levels.

Chemistry of actinium. Carrying experiments with tracer quantities of Ac^{228} ($MsTh_2$). *Chemistry of neptunium.* Potential of Np(III)-(IV) couple. Absorption spectrum of Np(III). *Chemistry of plutonium.* Equilibrium constants for reaction: $PuOBr^{+} + 2HBr \rightarrow PuBr_3 + H_2O$ at 540-640°C.

Helen, as usual, attended her chemistry class today.

A bulletin on today's front page says that U.S. and Russian planes met and exchanged greetings over the central German battle zone today.

Wednesday, April 18, 1945

At 8:30 a.m. in Room 209, Eckhart Hall, I attended the Project Council Information Meeting on Chemistry. Others present were Aebersold, Allen, Bartky, Bowman, G. Brown, H. Brown, Burton, Cohn, Daniels, Davies, Dempster, Elliott, English, Estermann, Fermi, Fred, Hilberry, Hogness, Howe, Huffman, Hume, W. C. Johnson, Keller, Lyon, Manning, Mulliken, Nickson, Rabinowitch, Rubinson, Spedding, Stearns, W. W. Watson, Wattenberg, Watters, Whitaker, Wilhelm, Young, Zachariasen, and Zinn. Spedding was the first speaker and talked about the need for pure rare earths. I followed him and reported for the Berkeley Project, basing my presentation on the information given in the letter I received from Calvin the day before yesterday. After the Clinton Chemistry Groups and Clinton Technical Division reported, I presented the work of our section, reporting first on the separation studies which I indicated were now concerned mostly with solvent extraction. I stated that the so-called Redox Process appears to be quite successful. I referred to Burton's studies which have shown no radiation effects on the hexone-water system at 5 times Hanford intensities. I mentioned that neptunium is found to follow the plutonium in the Redox Process and that the limiting fission product elements are cerium, zirconium, and ruthenium.

On the subject of basic work, I reported on our continuing work on the long-range alpha-particle activities attributable to transplutonium isotopes (95^{241} and 96^{242}) in neutron-irradiated plutonium samples. I discussed the possibility that the newly found 5-cm range alpha-particle activity found in plutonium plus 40 Mev alphas (sample 49αB) may be due to 96^{241}, 96^{240}, or 95^{240}. I also covered our work on the plutonium chemical fraction from uranium bombarded with 40 Mev alphas (sample TαB) including our evidence that Pu^{240} does not fission with slow neutrons and that Pu^{241} decays by the emission of soft (30-40 kev) beta particles. I described our specific activity determination on a sample containing 0.7% Pu^{240} that indicates that the half-life of Pu^{240} is shorter than that of Pu^{239}, not longer, and may be of the order of 7000-8000 years.

I mentioned our unsuccessful search for short-lived beta-particle emitting isotopes of plutonium, the Pa^{231} cross section work, oxidation-reduction potentials of Np III/IV and V/VI, plutonium chemistry studies of PuOBr and our chemical extractions that have provided 200 mg of U^{233} for Zinn's "thick solution" experiments and will provide an additional 100 mg so that Langsdorf will have 300 mg for his pile poisoning experiments to determine directly *eta*.

Items of interest presented by other speakers were as follows: (1) Spedding of Ames and Davies of Clinton reported on rare earth separations using adsorption on IR-1 resin (based on Waldo Cohn's early work). (2) Hume, reporting for Coryell, said that the last shipment of radioactive barium (Ba^{240}) to Site Y measured 560 curies. (3) Elliott reported on the stabilized palladium catalyst that reduces gas evolution in the homogeneous pile. (4) English spoke on the solubility of uranium and plutonium salts for homogeneous piles. Stoughton's calculations on the Pa^{233} n,γ cross section were also reviewed. (5) Lyon described the work on the uranium recovery program which has now been closed down. (6) Boyd reported on the results of some Szilard-Chalmers reactions for concentrating Sb^{124} and U^{237}. (7) Rubinson reported on studies of the mass number 141 and 143

chains. (8) Burton discussed the graphite problem in operating piles. It has been found that an energy storage amounting to 80 cal/g could be removed by heating to 1000°C, whereas the total energy storage was 102 cal/g. He said Zachariasen has found a 1% increase in lattice spacing of irradiated graphite.

Manning sent Nickson a description of the steps taken to correct the contamination found in the radiation surveys for the week ending April 7.

At 7:45 p.m. in Room 209, Eckhart Hall, I attended a meeting of Sub-section 1 of our section: The meeting was devoted to a description of the high temperature pile proposed by Daniels and intended primarily as a power source. Daniels described the structure to us as a perforated cylindrical refractory block, about 5'7" in diameter and 5'7" high, fabricated out of a homogeneous mixture of 99.6% BeO and 0.4% UO_2 (the latter being the oxide of U^{233}) surrounded by a thorium blanket. The best coolants would appear to be steam or bismuth. The unit would require 30 kg of U^{233} to start and would operate at 100,000 kw and at a temperature perhaps as high as 2000°C and burn some 100 gm of U^{233} per day.

According to Daniels the experimental work will have as its first objective the accumulation of data that will permit the choice of a suitable coolant and structural material — the erosion of BeO and UO_2 by steam, bismuth, and other possible coolants will be investigated and the structural stability and characteristics of BeO and other possible moderator-refractories will be studied at elevated temperatures.

War news was pushed from the top headline today by the verdict of a jury that Charlie Chaplin is the father of Joan Berry's baby. This has been a bitter civil suit.

At 2:10 p.m. there was a Project Council Policy Meeting, attended by Captain Chapman, J. Chipman, A. H. Compton, C. M. Cooper, A. J. Dempster, J. Franck, A. B. Greninger, J. G. Hamilton, W. B. Harrell, N. Hilberry, J. P. Howe, J. R. Huffman, L. O. Jacobson, W. W. Johnson, Captain McKinley, R. S. Mulliken, Major Murphy, H. D. Smyth, F. H. Spedding, J. C. Stearns, R. S. Stone, K. Tracy, C. J. Watson, W. W. Watson, M. D. Whitaker, J. E. Wirth, W. H. Zinn, and W. Bartky (late). Compton reviewed the "State of the Nation," stating that a provisional understanding has been reached with the Army regarding the financial level at which research will continue on the Project. Biology and health studies are based upon the present level until January 1 of next year; in other parts of the research an attempt has been made to maintain a skeleton crew. With regard to the latter, Compton said that the problem of maintaining manpower for studies on future piles has become so acute that he put up to the Advisory Committee last Sunday the question as to whether or not we should close down our research entirely as of January 1 of next year — the unanimous conclusion was that the scope of the research should not be further reduced. Hence, Clinton Labs, Met Lab, and associated laboratories will have their own programs on a somewhat reduced budget — expenditures for research will drop

from $13 million this year to $6 million next year. With regard to the Committee to review the future of the Project, Compton reported progress in forming the committee has been slow and he suspects Roosevelt's death has delayed the matter.

In answer to a question from Franck, Compton stated there is a possibility of transferring responsibility for the Clinton Labs from the University to a commercial company. Since the University would then be responsible for the Met Lab, a steering committee would be set up made up of representatives of the industrial firm, the Directors of the Laboratories, and Compton as Chairman. The Committee would have the responsibility for allocating tasks just as the Project Office now has.

Compton then introduced Major Murphy as the new X-10 Research Director, who said his duties will be to supervise the Met Lab, the Clinton Labs, and other prime contracts in their research tasks. The Chicago Area Engineer will be responsible to Murphy, who will be located in Clinton.

Compton outlined for purposes of discussion the type of information the new policy committee (to consider the future of the Project) might want, grouping topics under three major headings: Scope of Development Program, Organization of Development Program, and Correlation of Program with National Policy. The ensuing discussion was principally on regional laboratories. Smyth wants the laboratories run in sufficiently close cooperation with universities so that interchange of information will be possible through such mediums as lectures for students. W. Watson was of the opinion that postwar work could not be done without cooperation between the universities and the Government laboratories, with perhaps all the non-secret work being carried on by universities. Daniels suggested subsidizing such non-secret work as is now done in agricultural schools in universities. Harrell mentioned that if regional laboratories are formed they should be near a university with a permanent set-up for administering such things.

At the end of the meeting, Compton read his letter of resignation addressed to Hutchins, [giving his decision to accept the Chancellorship of Washington University in St. Louis], asking that those present keep the information confidential until a public statement is made within the next few days. He said he will maintain his office here without university ties as long as necessary to put the Project on a permanent basis. After that he will continue to serve in the desired advisory capacity from St. Louis.

Thursday, April 19, 1945

Perlman arrived in Chicago this morning from Site W. He will be visiting the Met Lab for the next two or three days. In my first discussion with him I learned that the second "Farmer" (Fermi) sample came out of the

Hanford pile last Thursday at 27,800-megawatt days for a 250-ton pile. The sample occupied a position in the pile equivalent to that of Farmer's No. 1. He also told me that sample W-4 was probably pushed out of the pile on January 18 and processed in March. It is approximately 60 gt material mixed with about 20% 36 gt material. We also discussed his future plans. He wants to return to the Met Lab to rejoin my section. We decided to request his transfer to be effective as soon as he can be spared at Hanford.

Cunningham sent Daniels the Problem Abstracts for March 15 through April 15 for Sub-section 2, Section C-I.

Helen attended her chemistry class, and then we had a quiet dinner ending with a delicious cake to celebrate my birthday.

American troops are pushing into Czechoslovakia. The Soviets are smashing toward Berlin with a reputed 2,500,000 men.

Friday, April 20, 1945

At 8:30 a.m. I held a meeting in my office of the Council of our section, attended by Albaugh, Cunningham, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. The meeting started with reports by Manning and me on the Project Council Information Meetings on Physics (April 17) and Chemistry (April 18). As an inside I mentioned some of W. J. Arrol's work in Canada wherein the gas resulting from the dissolution of an activated slug was examined in a mass spectrograph. The Canadian group has found a xenon isotope of mass 136 which is particularly interesting as this chain is hitherto unreported in fission work.

I talked about the redistribution of space in Section C-I: Room 12 will be made into a hot lab. Room 30, and eventually Room 29, will be turned over to protactinium work, while actinium work will be handled in Room 31. Room 33 (Florin and O'Connor) will be used for low Pu^{239} and high alpha-particle activities, whereas Room 28 will be reserved for low alpha-particle activities.

S. W. Scott of the Met Lab Patent Office came by my office to give me a document entitled "Divisible Subject Matter of Case S-52;" this lists subjects that might be patentable in separate individual cases.

Katzin sent a memo to Daniels giving the Problem Abstracts, March 16 through April 15 for Group 9, Section C-I.

Today's war summaries come from widespread fronts: Western front — U.S. 7th army drives toward Munich. London — Reds battle toward town 10 miles from Berlin. Guam — Americans face hardest fight on Okinawa. Rome — U.S. 5th army seven miles from Bologna.

Saturday, April 21, 1945

In a memo to Daniels, Manning objected to the hiring practice of other D.S.M. projects, who are pressing us to release many of our academic employees before July 1, 1945. He asserts that any releases from our section before July 1 will result in (1) reducing the research program below the minimum required level, and (2) failure to complete the writing assignments. He points out that Site Y, which has the highest priorities, has agreed to wait until July 1 for release for our personnel.

I spoke to Daniels to ask him to request the transfer of Perlman back to my section from Hanford.

In the afternoon there was a Group Leaders' meeting on New Piles in Room 251, Ryerson Laboratory, at which Katzin and Blaedel spoke on the use of solvent extraction to purify the fissionable fuel material from poisonous fission products on a repetitive basis in high intensity piles. Katzin reviewed the problems of purification of the pile and blanket materials in U^{233} converter or breeder piles, and Blaedel described the purification of plutonium fuels from homogeneous or heterogeneous pile systems. Attendees at the meeting were Adams, Bane, Blaedel, Burton, Cowan, Cunningham, Dancoff, Daniels, Dempster, Egan, Fred, Freedman, Gilbreath, Goldberger, Hemmendinger, Jesse, Karush, Katzin, Lawroski, Manning, Martin, Maurer, Mulliken, Novick, Rabinowitch, Rubinson, Safford, Schweinler, Shonka, J. Simpson, Jr., O. Simpson, Soodak, Stearns, Stephenson, R. Thompson, Tomkins, Warner, Watters, Way, Weinberg, G. Young, and H. Young.

In the afternoon I decided to skip the Katzin-Blaedel talks and stole out to play 18 holes of golf at Evergreen with Perlman and York.

The U.S. and Soviet armies are only 58 miles apart, according to today's paper.

Sunday, April 22, 1945

I played 18 holes of golf with Hagemann, Perlman, and Lawroski at Evergreen. Hagemann and Lawroski won our "low ball and low total" match, 5 and 4. Scores were FH-114, SL-108, GS-107, and IP-128.

Perlman boarded the train at 6:00 p.m. for Site W.

Monday, April 23, 1945

I received a copy of Saturday's memo from Daniels to Stearns requesting that Perlman be transferred from Hanford to the Met Lab Chemistry Division on July 1 or sooner in order to rejoin me if he can be spared from his present duties.

"Reds 6 Miles from Berlin" reads today's banner headline. A bulletin from Stockholm datelined April 21, says Russians are fighting in the center of Berlin.

Tuesday, April 24, 1945

The target from the natural uranium plus 40 Mev helium ions Berkeley bombardment arrived at the Met Lab. It received a total of 162-microampere hours between March 28 and April 15, and was turned over to James and Florin designated as sample TαC.

Albaugh sent Daniels the Problem Abstracts for the period March 15 to April 15 for sub-section 1, Section C-I.

Report CF-2914, "Chemical Research – Heavy Isotopes," (Seaborg, Section Chief; Manning, Associate Section Chief; Cunningham, Assistant Section Chief), April 24, 1945, was issued. This is a new category of report and includes the following topics:

Range of Np^{237} alpha particles. La Chapelle has determined the mean range to be 3.27 ± 0.02 cm of air at 15°C and 760 mm using some very pure Np^{237} which contained less than 0.005% Pu^{239} by weight. Previous measurements by Ghiorso and Jaffey are based on relatively thick samples containing traces of Pu^{239}.

Yield of Np^{237} in the X Pile. Beard has found the yield to be 0.3% that of Pu^{239}, but the figure is uncertain by a factor of two.

Examination of pile plutonium for Pu^{238} and Pu^{240}. Crawford, using the differential range chamber, has detected a small amount of Pu^{238} in Pu^{239} which underwent various degrees of bombardment. Pu^{240} was not detected. Specific activity measurements by O'Connor on plutonium containing 0.7% Pu^{240} indicate that the half-life of Pu^{240} is greater than 3400 years.

Cross section of reaction $Np^{237}(n,\gamma)Np^{238}$. Jaffey and Magnusson have bombarded Np^{237} in the Argonne pile and investigated the radiations from Np^{238}. The cross section for the reaction was determined to be 112 ± 25 barns. The growth of alpha particles in the sample indicates a half-life of Pu^{238} of 56 ± 10 years, in good agreement with the direct decay measurement of 66 ± 10 years.

Radiations from Np^{238}. Jaffey and Magnusson have found that Np^{238} emits about equal numbers of two beta particles having energies of 0.25 Mev and 1.35 Mev, respectively. Coincident with the softer beta particle is a 1.3 Mev gamma-ray. There is also evidence of large numbers of very soft conversion electrons and soft x-rays.

As usual, Helen attended her chemistry class today.

The link-up between Soviet and U.S. troops in Germany is near. The Soviets have seized half of Berlin, according to today's paper.

Wednesday, April 25, 1945

At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. Cunningham described plans for handling the new Np^{237} batch from Site W that is coming in soon. This was isolated in a special run (so-called "I.P. run" for Iz Perlman) from the supernatant solution left after the regular extraction step precipitation of bismuth phosphate carrying Pu^{239}. The batch will consist of 8 gallons of solution containing 3.7 g/l of $La(NO_3)_3$ and 1 to 2 M HNO_3. It corresponds to plutonium produced at a 100 gt level and contains something like 1 g plutonium and 25 to 100 mg Np^{237} (maximum possible yield of Np^{237} would be 300 mg). Fields will batch extract with hexone in two runs and concentrate the material by precipitation as $La(OH)_3$, after which it will be turned over to La Chapelle and Magnusson.

Lawroski mentioned the construction in the West Stands of new columns out of 1-inch glass pipe — most of the necessary parts have been ordered.

I commented that the Army will get us 5 mg of protactinium at $100 per mg. This is pretty cheap. It should arrive in two days, and Katzin will have two days to analyze it. Katzin said that most of one barrel of our own protactinium material has been through the semiworks and is now being worked up in the lab. This should yield 4 to 6 milligrams. Roy Thompson stated that the semiworks should finish the two remaining batches in less than two months.

I asked how the writing is coming and was greeted with dead silence. I pointed out that strong pressure is being brought to release men before July 1 and we want to be sure their reports are written up before they leave.

I received a teletype from R. H. Dunlap at Site Y, in response to my inquiry to Kennedy about the type of work Brody, Reinhardt, McLane, and Dixon would do if they transfer to Site Y, stating that research-type work is being planned for Brody, Reinhardt, and McLane in accordance with job classfication No. 4. Job classification No. 3 is scheduled for Dixon. Dunlap went on to suggest that the men make their decision on the basis of personal preference since the importance to the Project of employment at Monsanto or Site Y is about equal.

Perlman is back at Hanford. He called to say the following: (1) No word on anybody relative to the employment situation has been received at Site W, e.g., on Thompson. (2) Word did come through, however, on the changed status of Vance Cooper, Lindner, and Ballou; they are available, but no substitutes for them have been proposed yet. (3) Squires will be in Chicago Friday noon to Saturday night, at the Palmer House. (4) The Np^{237} solution will arrive tomorrow. Trouble was encountered in the extraction step. Due to dilution the acidity was down and lots of UO_2HPO_4 came through by jetting. Only 40% of the bismuth at one point came through. (5) The information on sample no. W-4 left Site W on April 18, addressed to Chapman, attention Stearns. (6) Sample no. W-1 is 24.1 gt (short ton) and W-3 is 70% 40 gt and 30% 24.1 gt (not definite).

(7) Willard checks out 95^{241} yield on 60 gt plutonium, using our assumption that it is produced in a second order reaction. For separation they use the Ce(IV) and IO_3^- as oxidizing agents and rare earth fluoride as carrier.

At 7:45 p.m. in Room 209, Eckhart Hall, I attended a meeting of the Basic Chemistry, Recovery, and Instruments Groups. Others present were Ader, Beard, Blaedel, Brody, Cunningham, Davidson, Dixon, Erway, Fineman, Florin, Fred, Greenlee, Hellman, Hindman, Hopkins, Howland, Hyman, Jaffey, James, Jones, Krueger, La Chapelle, Lawroski, Magnusson, Malm, Manning, McLane, Morgan, Niedrach, O'Connor, S. Peterson, Phipps, Reinhardt, Robinson, Sheft, Simpson, Stewart, Templeton, R. Thompson, Van Winkle, Westrum, Winner, and others. I opened the meeting, stating it would be on the subject of heavy isotopes, and then turned the meeting over to Cunningham. Hindman spoke first on the specific activity determinations of plutonium for various bombardment levels that were made to obtain a value for the half-life of Pu^{240} and a better value for the specific activity of Pu^{239}. The work was carried out by Greenlee, Dixon, and Ames. Values obtained for the half-life of Pu^{240} were 6800 and 6600 years in two determinations. I pointed out that if the results are accurate the data indicate more Pu^{240} is formed in the slug than in the bombardment of pure plutonium, an effect which could be caused by a higher yield with resonance neutrons. I suggested that measurements on 100 gt Hanford material would be interesting since, if this effect is real, over 1% Pu^{240} by weight should be found in the product. Hindman then discussed the second part of the problem, which was to obtain a better value for the specific activity of 2 gt plutonium — values obtained were 71,000 and 70,400 alpha-particle counts per minute per microgram at 52% geometry.

James spoke next on the separation of element 95 from previously purified plutonium. The results, he said, could be used to set a minimum value on the half-life of Pu^{241} of two years. I suggested this was a mighty brave estimate, whereupon James revised his minimum to one year. James and I discussed the Hanford results, communicated to me by Perlman today, on 95 extraction from a plutonium sample of a 2.5-fold higher bombardment level than our plutonium sample. This confirms our own findings. The yield presumably was higher by a factor of $(2.5)^2$, which is in agreement for the second order we have postulated for the reaction by which this isotope is formed.

James then compared the result of the first Berkeley run on plutonium plus 30 Mev alpha particles with the second run on plutonium plus 40 Mev alpha particles: the first yielded an alpha-particle activity of 4.8-cm range and a 6 to 8 month half-life; the second an alpha-particle activity of 5.0-cm range and a 1 month half-life. The possible mechanisms for this one-month alpha-particle emission were listed as: 96^{241}, 96^{240}, 96^{240} decaying by K electron capture to 95^{240}, and 95^{242} decaying by K electron capture to 94^{242}. The 96^{241} would be formed by an α,2n reaction, the 96^{240} by an α,3n reaction and the 95^{242} by an α,p reaction. I brought up the possibility of direct formation of 95^{240} by an α,p2n reaction. The 95^{240} in turn would decay by negative beta-particle emission with a short half-life to 96^{240}, which might be the one month alpha-particle emitter. I indicated all these possibilities are about equal in probability and that a choice must be deferred until more work is done.

Morgan next spoke on asymmetric fission, his conjectures based on the results of three deuteron bombardments — two on plutonium and one on uranium — which have shown evidence for the presence of Ac^{227} or $MsTh^{228}(Ac^{228})$. He postulated reactions of the type $Pu^{239} + {}_1H^2 \rightarrow Ac^{228} + n + {}_6C^{12}$.

Following Morgan's talk, at Cunningham's request, I pinch-hit for Ghiorso, who is at Argonne making measurements on the slow neutron fission cross section of Am^{241}. I gave the following isotopic assignments of the various radiations found on the examination of the plutonium chemical fraction of the uranium alpha particle and uranium deuteron bombarded targets worked up by Florin and given to Ghiorso and Weissbourd. In the case of the uranium alpha particle bombarded target I made the following assignments:

Radiation	*Source*
4.1 cm α-particles	Pu^{238}
3.7 cm α-particles	Pu^{239}
3.7 cm α-particles	Pu^{240}
x-rays	Pu^{237} (or Pu^{236})
30-40 kev β-particles	Pu^{241}
4.3 to 4.4 cm α-particles	Pu^{236} (or Pu^{237})

In the case of the uranium-deuteron target, the plutonium chemical fraction is shown to contain the following activities:

Radiation	*Source*
4.1 cm α-particles	Pu^{238}
3.7 cm α-particles	Pu^{239}
4.3 to 4.4 cm α-particles	Pu^{236}

I gave the following table (Fig. 50) showing regularities among the radiations of the heavy isotopes in order to explain why Np^{236} could be expected to be a beta-particle emitter.

Following my explanation of the table, Simpson asked whether or not the activity of U^{236} has been characterized. I said it should be an alpha-particle emitter and, according to the above generalizations, it must be beta stable and is not likely to decay by K-electron capture or positron emission. I predicted a half-life of $10 - 100 \times 10^6$ years for this isotope. Simpson then asked whether 95^{240} was expected to decay by alpha emission, to which I replied that it might decay by K-electron capture.

Concluding my presentation I observed that many isotopes known to be alpha-particle emitters will be found in the beta-particle region of such a table (especially in the region of atomic numbers 81-89). For these isotopes, though the half-life for alpha-particle decay is shorter than that for beta-particle decay, it may be predicted that beta-particle emission will be found. Beta-particle decay has been shown for RaA and ThA. A branch decay has recently been shown for actinium, i.e., 1% decay by alpha-particle emission.

Even Atomic Numbers

	96 a	96 b	94	92	90 ------- 82	80	78	76	74	72	70	68
1	(←96^{249})	(←96^{247})	(←Pu^{243})	←U^{239}	←Th^{233}	←Pb^{209}	←Hg^{205}	←Pt^{199}	←Os^{193}	←W^{187}	←Hf^{181}	←Yb^{177}	←Er^{171}
2	($\underline{96}^{248}$)	($\underline{96}^{246}$)	($\underline{Pu}^{242}$)	$\underline{U}^{238}$	$\underline{Th}^{232}$------$\underline{Pb}^{208}$	$\underline{Hg}^{204}$	$\underline{Pt}^{198}$	$\underline{Os}^{192}$	$\underline{W}^{186}$	$\underline{Hf}^{180}$	$\underline{Yb}^{176}$	$\underline{Er}^{170}$
3	(←96^{247})	(←96^{245})	←Pu^{241}	←U^{237}	←Th^{231}	$\underline{Pb}^{207}$	←Hg^{203}	←Pt^{197}	←Os^{191}	←W^{185}	$\underline{Hf}^{179}$	←Yb^{175}	←Er^{169}
4	($\underline{96}^{246}$)	($\underline{96}^{244}$)	$\underline{Pu}^{240}$	$\underline{U}^{236}$	$\underline{Th}^{230}$	$\underline{Pb}^{206}$	$\underline{Hg}^{202}$	$\underline{Pt}^{196}$	$\underline{Os}^{190}$	$\underline{W}^{184}$	$\underline{Hf}^{178}$	$\underline{Yb}^{174}$	$\underline{Er}^{168}$
5	($\underline{96}^{245}$)	($\underline{96}^{243}$)	$\underline{Pu}^{239}$	$\underline{U}^{235}$	$\underline{Th}^{229}$	($\underline{Pb}^{205}$)	$\underline{Hg}^{201}$	$\underline{Pt}^{195}$	$\underline{Os}^{189}$	$\underline{W}^{183}$	$\underline{Hf}^{177}$	$\underline{Yb}^{173}$	$\underline{Er}^{167}$
6	($\underline{96}^{244}$)	$\underline{96}^{242}$	$\underline{Pu}^{238}$	$\underline{U}^{234}$	$\underline{Th}^{228}$	$\underline{Pb}^{204}$	$\underline{Hg}^{200}$	$\underline{Pt}^{194}$	$\underline{Os}^{188}$	$\underline{W}^{182}$	$\underline{Hf}^{176}$	$\underline{Yb}^{172}$	$\underline{Er}^{166}$
7	($\underline{96}^{243}$)	($\underline{96}^{241}\rightarrow$ 95^{241})	($\underline{Pu}^{237}\rightarrow$ Np^{237})	$\underline{U}^{233}$	$\underline{Th}^{227}$	$\underline{Pb}^{203}\rightarrow$	$\underline{Hg}^{199}\rightarrow$	(Pt$^{193}\rightarrow$)	$Os^{187}\rightarrow$	(W$^{181}\rightarrow$)	(Hf$^{175}\rightarrow$)	$\underline{Yb}^{171}$	$\underline{Er}^{165}\rightarrow$
8		($\underline{96}^{240}$)	($\underline{Pu}^{236}$)	$\underline{U}^{232}$	($\underline{Th}^{226}$)		$\underline{Hg}^{198}$	$\underline{Pt}^{192}$	$\underline{Os}^{186}$	$\underline{W}^{180}$	$\underline{Hf}^{174}$	$\underline{Yb}^{170}$	$\underline{Er}^{164}$
9				($U^{231}\rightarrow$ Pa^{231})	($\underline{Th}^{225}\rightarrow$)		$Hg^{197}\rightarrow$		(Os$^{185}\rightarrow$)			(Yb$^{169}\rightarrow$)	(Er$^{163}\rightarrow$)
10				($\underline{U}^{230}$)			$\underline{Hg}^{196}$		$\underline{Os}^{184}$			$\underline{Yb}^{168}$	$\underline{Er}^{162}$

Odd Atomic Numbers

	95 a	95 b	93	91 ---------- 83	81	79	77	75	73	71
1	(←95^{244})	←95^{242}	←Np^{238}	←Pa^{232}	←Bi^{210}	←Tl^{206}	←Au^{198}	←Ir^{194}	←Re^{188}	←Ta^{182}	←Lu^{176}
2	($\underline{95}^{243}$)	$\underline{95}^{241}$	$\underline{Np}^{237}$	$\underline{Pa}^{231}$	$\underline{Bi}^{209}$	$\underline{Tl}^{205}$	$\underline{Au}^{197}$	$\underline{Ir}^{193}$	$\underline{Re}^{187}$	$\underline{Ta}^{181}$	$\underline{Lu}^{175}$
3	←95^{242}	(←95^{240})	(←Np^{236})	(←Pa^{230})	(←Bi^{208})	←Tl^{204}	←Au^{196}	←Ir^{192}	←Re^{186}	←Ta^{180}	
4	$\underline{95}^{241}$	($95^{239}\rightarrow$)	($\underline{Np}^{235}$ U^{235})	($\underline{Pa}^{229}$ Th^{229})	$\underline{Bi}^{207}$	$\underline{Tl}^{203}$	(Au$^{195}\rightarrow$ Pt^{195})	$\underline{Ir}^{191}$	$\underline{Re}^{185}$		
5	($95^{240}\rightarrow$)					$Tl^{202}\rightarrow$			$Re^{184}\rightarrow$		

Figure 50. Regularities and predictions of radioactivities.

President Truman spoke on the radio today opening the San Francisco Security Conference.

Thursday, April 26, 1945

At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Cunningham, Florin, Ghiorso, Hagemann, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Magnusson, Manning, McLane, Morgan (later), O'Connor, S. Peterson, Scott, R. C. Thompson, Van Winkle, and Weissbourd. After I had discussed some general matters, James and Florin described their plans to handle the new uranium plus 40 Mev helium ions target (sample TαC) which was received from Berkeley a couple days ago.

Hindman reviewed his run on the specific activity of 90 gt and 35 gt plutonium which is nearly finished. Hagemann described his work on the U^{233} decay products. There is evidence that the 30-day half-life alpha-particle emitter is due to actinium, which is followed by two alpha particles with a 5-minute half-life, then by a 40-minute half-life, then by a 40-minute alpha particle. The 5-minute alpha-particle half-life seems to be due to an emanation. Peterson described his work on the chemical fractionation of actinium and rare earths.

The following activities are scheduled: (a) Florin and James to work on the uranium plus helium ion target, (b) Jaffey and Hyde to work next week on the cross section for the reaction $Io^{230}(n,\gamma)UY^{231}$.

The Np^{237} solution from the special run at Hanford on April 13 arrived today.

Ghiorso, using the Argonne pile yesterday, measured the slow neutron fission cross section of 95^{241} using about 75,000 c/m (50% geometry). His results indicate that it probably does not fission with slow neutrons if its half-life is much greater than 5 years.

James and Florin began work on sample TαC, the uranium plus 40 Mev helium ions Berkeley bombardment of March 28 to April 15. Four 0.002" layers of uranium metal were machined off the target and labeled TαC I, TαC II, etc. The plan is to treat each of these layers individually, chemically separating it into plutonium, rare earth, and neptunium fractions, as was done with the previous uranium plus 40 Mev helium ions target TαB. I wrote to Hamilton in Berkeley asking him to send the complete details on this bombardment.

Stearns sent authorization to Hogness for the transfer of 2 mg of 50% U^{233} to the Area Engineer for shipment to Y-12.

I was the speaker at the Physics Division meeting held at 7:45 p.m. in Room 209, Eckhart Hall. My topic was "Heavy Isotopes" and I reviewed the work done on bombardments of U + neutrons, U + deuterons, U + helium ions, Pu + neutrons, Pu + deuterons, and Pu + helium ions. I also talked about predictions and future work, mentioning (1) we expect to look for possible

alpha-particle branching of Pu^{241} to yield U^{237}; (2) bombardment of high-241 plutonium in the pile to look for Pu^{242} and Pu^{243}; (3) search for other isotopes of element 95, for instance 95^{240} and 95^{239}; (4) look in pile neutron-irradiated plutonium for 96^{243}, 96^{244}, and 96^{245} — the latter may be a beta-particle emitter yielding 97^{245}; (5) investigate new isotopes of other elements lower in atomic number, for instance Pa^{230} (to be formed by Io^{230} plus deuterons); (6) bombardments in the Berkeley cyclotron with particles heavier than helium ions; for instance, $U^{238} + Be^{9} = 96^{245} + 2n$.

Helen as usual went to her chemistry class today.

Berlin is surrounded by Soviet troops, and U.S. troops will soon link up as Soviet troops have swarmed across the Elbe.

Friday, April 27, 1945

La Chapelle and Magnusson began isolating the Np^{237} from the special run (sample 37W) at Hanford.

I sent a memo to Hogness stating that my U^{233} group has been operating almost exclusively as a service group since resumption of the U^{233} program last year. I point out the desirability of our keeping, over a period of time, a substantial fraction of the U^{233} supply to enable us to undertake a program of chemical research which has barely gotten underway.

In a memo to Stearns I requested additional samples of Hanford plutonium in view of the interesting results obtained from our recent investigations of heavy isotopes in such Hanford product. I pressed a request for a few grams of material manufactured at a concentration level of about 100 gt and asked him to anticipate a further request in a few months for Hanford plutonium of a higher concentration level, say, around 200 gt. I mentioned the possibility that there may be enough plutonium in the recently received Np^{237} solution from Hanford to fill the first request; in which case we would need authorization to retain it.

German Hamburg radio accounced that Herman Goering resigned as head of the air force because of "acute" heart illness.

Saturday, April 28, 1945

Squires is in town, and I had a long talk with him. Among other things, I discussed Vance Cooper's case (Vance has been asked to stay at Hanford for the duration, but du Pont has no long-term interest in him because of the manner in which he left the Seneca Works — he came to the Met Lab at my request before he was officially released). Squires seems very interested in retaining Cooper's services. I also pressed my desire to have Perlman, Stan Thompson, Kohman, and Willard return to my chemistry section at the Met Lab as soon as they can be spared at Hanford. Lom made no promises.

Perlman called me from Site W to give me the following information: (1) The plant operating people believe that our conditions for precipitating bismuth phosphate from the extraction supernatant to recover Np^{237} lead to an unstable solution, i.e., to a precipitation of uranyl phosphate. They suggest lowering the ammonium salt concentration to 0.02 M from 0.05 M. Their semiworks will make dummy runs. We should check the yield of Np^{237} at 0.02 M. In S. Thompson's lab experiments a precipitate of uranyl phosphate came down even without dilution. (2) There is no more news on the manpower situation. (3) We should receive a 1-ml sample (of uranium dissolver solution), about 100 gt, in a few days — in their container, not ours — Perlman will send me a sketch of it. If their container leaks, they will use ours.

Kennedy called me from the Stevens Hotel in downtown Chicago. He read me a draft of the agreement between inventors that he, Wahl, and Segrè have prepared. Significant points are: right of vote is not inheritable; tangible benefits are inheritable; any inventor can request a vote by mail to the others. I told him about cases like S-1702 (neptunium electrolysis). He told me of the changes that he, Wahl, and Segrè desire in the draft letter to Lavender I sent him on April 4.

Kennedy also said that Site Y is shipping us the element 95 fraction from 150 grams of Pu^{239} which has been processed by ether extraction of dissolved $Pu_2(C_2O_4)_3$ followed by reduction of the H_2O layer with SO_2 and production of plutonium metal by reduction. It should contain 1-5 grams of Pu^{239}.

I sent a memo to Captain Lavender in Washington about the draft agreement he has prepared covering the rights of the Government to the inventions described in Patent Cases 52 and 61 (received by me March 23). I state that we four inventors agree in the main with the ideas presented in the draft agreement; however, I suggest some modifications we would like to make. I note that neither his office nor Dr. Bush's has received the approval from the University of California requested from them in my letter of February 15 to President Sproul. I said I will wait a little longer before writing to Sproul again as I know he is very busy and away from Berkeley much of the time.

In the afternoon I talked at the Saturday meeting of Group Leaders on New Piles in Room 251, Ryerson Laboratory, on the effect of heavy istopes on the operation and products of converter and breeder piles. I reviewed what we have learned from examination of Clinton and Hanford plutonium, plutonium which was re-irradiated at Hanford, and the Berkeley bombardments of uranium with 40 Mev helium ions (to produce Pu^{240} and Pu^{241}). I then applied these ideas to a large-scale converter pile operating on 50 kg of Pu^{239}, indicating that after 30% of the Pu^{239} has undergone reaction one obtains about 10% by weight of Pu^{240} and about 1.0% of Pu^{241}. I stated that if this latter decays to 95^{241} at the rate of about 1% per month, about one gram of 95^{241} will be produced per month.

In the course of my remarks I discussed our experiments on the (small or zero) slow neutron fission cross sections of Pu^{240} and 95^{241}, predicted that Pu^{241} should be fissionable with slow neutrons, and emphasized the importance of these isotopes to the breeder reactor

program. I predicted that Pu^{242} formed by neutron absorption in Pu^{241} will decay by alpha-particle emission, that the alpha-particle emitters 96^{243}, 96^{244}, 96^{245}, 96^{246}, 96^{247} will be produced by successive n,γ reactions, and that beta-particle decay of 96^{245} or 96^{247} might lead to 97^{245} or 97^{247}.

American and Soviet troops met on a girder of a blown-up bridge over the Elbe River at Torgau, 28 miles northeast of Leipzig, on April 25. The news came in a delayed release according to this morning's newspaper.

Sunday, April 29, 1945

Hagemann, Lawroski, Morgan, and I played 18 holes of golf at Evergreen. Hagemann and Lawroski beat Morgan and me in our "low ball plus low total" match, 7 and 4. Scores were FH-112, SL-109, GS-109, and TM-121.

Mussolini has been killed in Milan, the birthplace of Italian fascism.

Monday, April 30, 1945

Hitler committed suicide today.

In a memo dated April 28 Hogness gave me the following information on a revision in Hanford personnel received by the Laboratory Director's office: (1) Lindner will not be available to us. He is to be employed at Hanford for the duration. (2) The following men will be available sometime during the summer [no information on definite time]: Ballou, Fries, Gaarder, V. Cooper, Goeckermann, Hill, Korler, Kohman, Lincoln, Miller, Stehney, S. G. Thompson, Watt, Werner, Willard, and Yett.

I read a copy of an April 28 memo to Stearns from Perlman giving the packing details of the Hanford (pre-extraction) uranium solution we should receive on May 1. He also states that: (1) The sample was taken from Run No. B-5-04-D-7, which material was pushed from the pile on March 15. (2) It is the same as the plant sample, No. 8-1-M, and is from the point in the process where the sulfuric acid has been added and the UNH is in the range 30-35%. (3) The product level in this batch is about 125 gt.

I wrote to Kennedy at Site Y enclosing a copy of my April 28 letter to Lavender. I also enclosed for his comment, a copy of the Patent Office's proposed Divisible Subject Matter of Case S-52 (i.e., proposed individual subjects that might be patentable) that Scott of the Met Lab Patent Office delivered to me on April 20. I also told him that I have gone ahead and signed the Case S-1702 by Seaborg and Wahl, the Electrolysis Case which might come under the subject matter of Case S-52.

Stoughton, who is in Chicago, and I conferred about the thorium carbonate cans. I learned that: (1) Only 3 out of 25 of our thorium carbonate cans passed the heat test of four days at 230°C. All 125 cans passed for 5-6 days at 175°C. These three plus three others went into the "Montreal" stringer (best position: center) about a month ago and are probably still in. (2) About 100 cans went in last week in a position about one-third of the maximum flux — a nearly average position. (3) The maximum flux is 1.5 times the average (according to the chemists), or 2.4 times the average (according to the physicists).

I talked with Perlman at Site W by phone and told him about the latest status of Stan Thompson based on my conference with Lom Squires on Saturday and the memo I received from Hogness today — i.e., it does not appear likely that Stan will be released in the near future. We also discussed the reluctance of Hanford to release John Willard at this time. Iz asked me to send him copies of the Sugarman memorandum. He told me that Thompson recommends 0.02 M fluosilicate and 1.2 N H_2SO_4 with no sodium salt or straight acid for the Np^{237} recovery runs. This procedure is being tested in the cold semiworks.

"The Metallurgical Laboratory, Report for April 1945," (MUC-JCS-305) was prepared by the Laboratory Director's office. The summary section of the report includes two items of particular interest to me.

"It is anticipated that the scientific program of the Laboratory will be carried on with three main divisions — one, the Service and Development work which includes the sections in Optics, Electronics, and Shop; two, the Health Division, the program of which will not be altered before January 1, 1946; and three, the Division of Research and Development in the physical sciences which will include sections in fundamental chemistry, pile chemistry, pile physics, x-ray, mass spectroscopy, and metallurgy.

"Mr. Lapp has been appointed Assistant Laboratory Director and placed in charge of the Service and Development Division. He has been assigned as a liaison man between Site Y and the Metallurgical Laboratory in order to expedite a special job being done by the Metallurgical Laboratory for Site Y."

Captain J. H. McKinley, Area Engineer's Office, issued the following instructions for SED men today:

> 1. All enlisted men are advised that upon announcement of the capitulation of Germany, no celebration will be held at this installation and no time off will be granted therefore.
>
> 2. You are further advised that higher authority has directed that upon announcement of the capitulation of Germany, all military personnel are to remain off the streets of towns and cities, and out of public places to the extent possible. However all are expected to be on the job as usual. These restrictions to go into effect upon announcement of the capitulation of Germany and to remain in effect until contramanded by orders. Orders lifting restrictions will be issued immediately where circumstances warrant such action. This is necessary to avoid being involved

unwillingly or otherwise in any situation which might result from celebrations which may occur among the civilian population.

3. All are cautioned that you are expected to set an example of sobriety and industry and to foster a "stay-on-the-job" attitude. Any willful deviation from the policy as set forth above will result in disciplinary action.

Figure 51 shows the location of the people in New Chemistry as of this date. The members of Group 3 (Process Development and Group 4 (Solvent Extraction) are located generally in the West Stands. Figure 52 shows a view of the main corridor of New Chem.

It is difficult to keep abreast of the news from the European theater of operations at this point. But, Munich has fallen, taken by U.S. troops with almost no opposition as they rolled into the birthplace of Naziism.

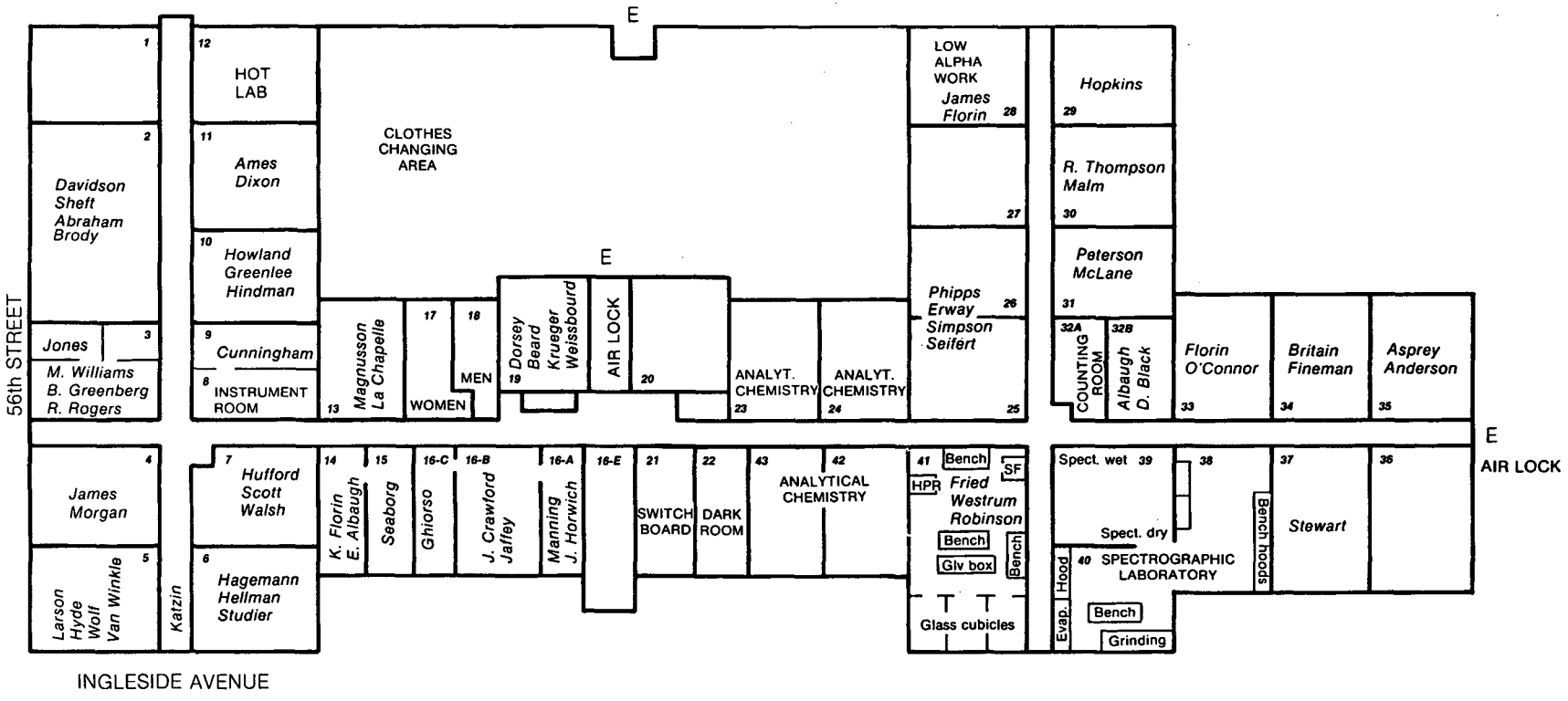


Figure 51. Room assignments in New Chem. April 1945.

Figure 52. South end main corridor of New Chem Building. XBB 795-5922

INDEX

Page numbers in italics indicate illustrations.

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