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UNIVERSITY OF CALIFORNIA RADIATION LABORATORY

MONTHLY PROGRESS REPORT NO. 56

Covering Contract No. W-7405-Eng-48

December, 1947

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1. 184-inch Cyclotron Program

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The performance of the cyclotron during the month has been very satisfactory with only five per cent of the time required for correcting mechanical or electrical troubles. Seventy-six per cent of the time was spent on operation for research purposes and eleven per cent on the installation of an evacuated tube and associated parts for carrying the deflected deuteron beam through five feet of the neutron shielding wall into a "cave" in which deuteron bombardments can be made without excessive neutron background. Over 10^{-9} amperes is obtained at the end of the tube. This marks the practical completion of the project for deflecting the deuteron beam which has been under way throughout the year.

Deliveries of the remaining concrete blocks to complete the second five feet of shielding wall began during the month. Design and fabrication are proceeding on the proton conversion apparatus.

2. 60-inch Cyclotron Program

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The 60-inch cyclotron operated very satisfactorily during the month of December. Major bombardments for this interval included alpha particle irradiation of plutonium for Professor Seaborg; the deuteron bombardment of lithium, and the deuteron bombardment of iron for the Clinton Laboratories at Oak Ridge; the deuteron bombardment of ionium for the Argonne National Laboratories at Chicago; and the deuteron bombardment of cobalt for Doctor J. H. Lawrence at Berkeley. There were a number of alpha particle bombardments of bismuth for Professor Segre and his colleagues.

A great deal of time, effort, and energy has been spent on the plan and design of certain changes in the instrument in the over-all improved radiation protection program for the operating personnel as well as to increase the efficiency of the cyclotron.

3. Synchrotron Program

During the last test of the outer vacuum chamber ring a vacuum was obtained that was very nearly, but not quite, satisfactory for synchrotron operation. The remaining leaks consisted of some small pores which were not closed by any of the impregnating processes. In order not to delay the progress of the synchrotron it was decided to proceed with the design and ordering of a ceramic or glass doughnut. This design has been completed and requests for quotations are now outstanding. Meanwhile, it is planned to continue assembly of the synchrotron in order that the magnetic tests and time consuming adjustments can be made. If after these are done it appears

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that there is a good chance of obtaining a satisfactory operating vacuum with the existing vacuum chamber, additional effort will be devoted to this work.

If the vacuum, however, continues to be poor at the time the ceramic ring is delivered, the vacuum chamber will be disassembled and the ceramic ring installed. A possible alternative in case the vacuum is nearly satisfactory is the construction of another outer ring using the same methods of fabrication which were used in constructing the inner ring. Our experience with the inner ring indicates that this method of construction produces a satisfactory part and it now appears feasible to construct such a ring in our own shops.

Assembly of the upper yoke is now in progress and all parts for its completion are on hand with the exception of one outer slab which is en route. Six flux bars remain to be shipped and one spare outer slab is expected in February.

4. Linear Accelerator Program

Van de Graaff Generator. During the past month the primary effort has been concerned with increasing the operational reliability of the machine. To this end it has been found necessary to make certain minor mechanical and electrical changes. The machine has been closed and operating for over a week with a total beam sufficient to heat a 1/8 inch thick quartz plate to incandescence over an area of diameter about 3/16 inch. It will hold 4.2 to 4.3 million volt indefinitely with no sparking and with a large beam. After the 90 degree deflecting magnet is installed it will be possible to make a good measurement of the total proton current.

40 Foot Section. In order to improve the reliability and raise the Q of the accelerator, the drift tubes and stems have been electroplated and a new set of spring fingers connecting the drift tube stems to the liner have been made. As a result of these changes it is now possible to operate at will at rf levels satisfactorily high for reliable accelerator operation.

A number of bombardments have been made with the machine among the results of which were an activation curve for the carbon (p,pn) and the observation of the reaction $B^{10}(p,n)C^{10}$.

5. Experimental Physics

Neutron-Proton Scattering -(Cloud Chamber). The number of proton tracks measured was increased to 430; the distribution still favors non-exchange, but not significantly so. The energy distribution of the neutrons, as determined by the knock-on protons, has been measured for 750 cases. The distribution shows a maximum at about 90 Mev as expected, but shows an excess over theory at high energies which is probably experimental, and a greater excess at low energies.

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Considerable work has been done in rechecking measurements and determining sizes of errors. The conclusion reached was that previous results had little significance for scattering angles greater than 60° . It is hoped that changes in technique of measurement will permit accurate measurements to be extended to 80° .

The iron core magnet for the new cloud chamber has been completed and tested. It gives a field of 21,000 oersted, uniform to 10%, in the area to be used.

Counter Development. Tests have been made of naphthalene counters following reports of their successful use in the east. The preliminary tests were most encouraging and it appears that these can be developed into a useful research tool.

Delayed Neutron Emission from Light Elements. At the end of the month, observations were made indicative of delayed neutron emission from a number of light elements in the neighborhood of aluminum under high energy deuteron bombardment. Intensive investigations are in progress to confirm the reality of this effect and to determine the mechanism.

Scattering of Deuterons. Exploratory runs have been made using particle sensitive plates and ionization chambers to determine the scattering of deuterons in various materials.

Bismuth Fission Cross Section. The cross section for the fission of Bi has been measured in the 90 Mev neutron beam using standard carbon disks to determine the neutron flux. The result is $\sigma = 0.05 \times 10^{-24} \text{ cm}^2$, or approximately the geometrical cross section of the thorium nucleus.

Neutron Scattering. The data on scattering of 90 Mev neutrons by Pb, Cu, and Al has been completed during this month. In each case the distribution followed that predicted by diffraction from an opaque nucleus out to the vicinity of the first minimum of the diffraction pattern. The minimum was not observed, probably due to contributions of inelastically scattered neutrons, and to some extent due to the wavelength spread in the primary beam. The differential cross section values were higher in the vicinity of 0° than those expected from total cross section measurements. The reasons for this have not yet been clarified.

Neutron-proton Scattering. This month has been spent in studying the instrumentation with which these experiments are done to insure proper plateaus with respect to such variables as voltage, resolving time, counting rate, and background of counts not due to fast protons. Causes of systematic errors have also been investigated. These investigations have not altered the previous conclusions from the experiment.

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6. Theoretical Physics~~RESTRICTED~~

Good progress has been made in the calculations of n-p scattering. Angular dependence curves in good agreement with observation have been obtained using spherically symmetric potentials. Corresponding curves including tensor forces are nearly completed. These are being calculated by a new method, which is much easier than the variation method previously investigated, and which seems to give very reliable results. Some work has been done on the theory of diffraction scattering. An investigation of betatron and synchrotron injection problems has been started. Other programs are continuing.

7. Isotope Research Program~~RESTRICTED~~

A successful deposit of C^{14} was made which yielded observation of the C^{14} lines in the hyperfine spectrum. A separation of isotopes 107 - 111 was performed upon ions from a sample of Ag bombarded with α particles. Identification of certain indium activities was sought, with partial success.

8. ChemistryPart A~~SECRET~~

Radioactive Rare Earth Isotopes. A program is under way to identify the neutron deficient isotopes of the rare earth elements. The activities are produced by spallation reactions on tantalum with high energy particles and of individual rare earths with lower energy projectiles. Several new activities have been found but have not yet been assigned. Of some interest is an 8.4-hour positron-emitter in the thulium fraction from the bombardment of holmium with 40-Mev helium ions. This is the heaviest positron-emitter that has yet been established. A 36-hour K-capturing cerium has been tentatively assigned to Ce^{137} as a result of its formation from 40-Mev deuterons on lanthanum. Since it decayed essentially to background, with no sign of a lanthanum daughter, La^{137} must have a half-life of >100 years or the assignment of Ce^{137} is in error.

Fission of Bismuth with 200-Mev Deuterons. Over 25 fission products have been identified and in most cases yields have been determined. The fission yield curve is characterized by a single peak with the maximum at mass number 97-99. In general the light fragments have neutron excess and the heaviest fragments are neutron deficient. By inference the most prominent primary fission products in an intermediate region are stable. A mechanism which conforms with the observations has been advanced. It is postulated that the nuclei responsible for most of the fissions are light bismuth or polonium isotopes of mass number 197-199, which result from the ejection of 10-14 neutrons from the compound nucleus. In order to account for the neutron-proton distribution in the fission fragments it is necessary to assume that the parent nucleus undergoes cleavage, preserving the same

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neutron/proton ratio in both fragments. By means of this theory it is possible to predict the most abundant primary fission product for each mass number. A fission yield curve has been constructed with these considerations and integration of the curve gives a rough cross section for fission of $0.2 \times 10^{-24} \text{ cm}^2$.

The fission products which have been identified are: Ca^{45} , Fe^{59} , Cu^{67} , Zn^{72} , As^{74} , Rb^{86} , Sr^{89} , Y^{90} , Y^{91} , Zr^{95} , Mo^{99} , Ru^{103} , Ru^{106} , Ag^{111} , Pd^{112} , Cd^{115} (44-days), Te^{119} , Sb^{122} , I^{124} , I^{125} , I^{126} , Ba^{133} , Ce^{139} . The yields for several of these isotopes fall below the smooth curve but in almost every case the result is as expected since the calculations show such isotopes to be other than those which should be formed in greatest yield.

Chemistry of Astatine. The chemical studies of astatine have continued with results which may be summarized as follows: ferric or mercuric compounds are sufficient to rapidly oxidize 10^{-12} molar astatine in the zero state. Cold persulfate is a slow oxidizing agent. Astatine strongly oxidized by persulfate failed to deposit at the cathode under the conditions previously described. At the same time deposition at the anode was normal. The critical deposition potential at the anode was $-1.45 \pm .02 \text{ v}$ with respect to a normal H_2 electrode.

Chemistry. Part B.

Synthetic and Experimental Organic Chemistry. High yield, small scale synthetic procedures have been or are being developed for the preparation of the following compounds: 1) formic acid, $\text{HC}^{14}\text{O}_2\text{H}$; 2) ethyl alcohol, $\text{CH}_3\text{C}^{14}\text{H}_2\text{OH}$; 3) ethyl alcohol, $\text{C}^{14}\text{H}_3\text{CH}_2\text{OH}$; 4) ethyl bromide, $\text{CH}_3\text{C}^{14}\text{H}_2\text{Br}$; 5) ethyl bromide, $\text{C}^{14}\text{H}_3\text{CH}_2\text{Br}$; 6) ethyl iodide, $\text{CH}_3\text{C}^{14}\text{H}_2\text{I}$; 7) ethyl iodide, $\text{C}^{14}\text{H}_3\text{CH}_2\text{I}$; 8,9) an improved synthesis of methyl or carboxyl labeled glycine; and 10) lactic acid, $\text{CH}_3\text{C}^{14}\text{H}_2\text{OHC}^{14}\text{O}_2\text{H}$.

Biological Chemistry. The conversion of C^{14} -labeled tyrosine, dihydroxyphenylalanine and tryptophane to melanin has been studied by in vitro incubation experiments with melanoma tissue. All three compounds are converted actively to melanin under these conditions but only tryptophane and tyrosine appear in the melanin when these compounds are administered intravenously to living mice.

Studies are in progress on the metabolism of C^{14} -labeled dibenzanthracene and tyrosine in mice and on the nature of the labeled compounds found in the feces and urine. The rate of metabolic oxidation to carbon dioxide of labeled acetic acid in tumor and non-tumor bearing mice is being determined.

Photosynthetic Chemistry. Work has continued with experiments on the growth and decay of the reducing power of algae with illumination by measurement of up-take of radioactive carbon dioxide in the dark. The distribution of activity in algae under varying conditions of illumination is being determined with particular interest directed toward the plant acids, amino acids and plant sugars.

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A study is in progress on the function of chlorophyll in photosynthesis to determine if chlorophyll acts as a light sensitizer, a hydrogen donor, or both. Deuterium is used as a tool in this work.

Chemistry. Part C.

Subproject 48B

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Metals and High Temperature Thermodynamics. A report has been completed summarizing the thermodynamics of plutonium. The work on high temperature equilibria and absorption spectra of CN has given a value for the heat of formation of this important molecule. Work continues on the carbide refractories and on the liquid metal systems containing uranium. Progress has been made in the research dealing with high temperature molecular species.

Basic Chemistry. Including Chelates. The following research programs are in progress:

The hydrolysis and complexing of Zr(IV) as determined by extraction studies using TTA in benzene.

The rate of exchange of I_2 and IO_3 , using radioactive iodine tracer.

The solvent extraction of U(VI). Equipment is being set up for an H_2O analysis on the organic phase by Fisher reagent.

The complexing of Pu(IV) by TTA, involving a study of the solubility of PuK_4 at high TTA concentrations.

The extraction of La(III) into benzene with TTA. The question of complexing will be studied.

The hydrolysis of U(VI) using benzene TTA extraction equilibria.

9. Medical Physics

Part A. Project 48A - I

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Tracer studies are now under way with carrier-free preparations of the radioactive isotopes of zirconium, technetium, silver, cadmium, indium, tin, antimony, element 61, actinium, uranium. The most significant observations to be made during the month are as follows. Technetium has been observed to be excreted, following intramuscular administration, with an extraordinary degree of rapidity, there being approximately 95 percent eliminated within 24 hours and the kidneys acting as the principle channel of elimination. Cadmium is observed to be eliminated very slowly, principally by way of the digestive tract. A significant degree of selected localization has been observed in the liver, kidney, and pancreas. Approximately 0.2 percent has been noted to be absorbed following oral administration by

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stomach tube. Actinium, as predicted, follows a metabolic pattern quite similar to that observed for the lanthanide rare earths and americium and curium.

The radioautographic studies with zirconium, columbium, cerium, element 61, plutonium, americium and curium are being continued.

The study of the possible methods of the treatment of plutonium poisoning is also continuing.

Methods for the radio-chemical isolation of carrier-free molybdenum are being investigated and a small amount of carrier-free calcium has been prepared for the purpose of comparing, on a rather precise quantitative basis, the possible differences between the metabolism of this element and strontium. A preparation of radio-europium, while not carrier-free but of a high specific activity, has been ordered from the Clinton Laboratories at Oak Ridge and negotiations are in progress for the acquisition of a carrier-free preparation of the 47 hour radio-samarium that arises from fission.

Medical Physics

Part B. Project 48A - II

The first biological experiment with the direct beam of the 184-inch cyclotron was performed in an effort to determine the lethal effect on mice. It was designed to demonstrate the time of death with a wide range of dosage. Within ten days after bombardment all animals in the groups that received 1000 rep and 3000 rep died while no animals in the groups receiving 100 rep and 330 rep have yet succumbed. The white blood cell count of all groups showed a very marked drop in the first twenty-four hours, but the red blood cell count was not affected in any group. Observations were also made of the weight variations with time after exposure.

An unexpected result from an experiment conducted sixteen months ago was revealed when some mice were sacrificed which had received intravenously 0.5 mg of colloidal UO_2 enriched 87 percent in U^{235} . Subsequently one-half of the animals had been bombarded with neutrons from the 60-inch cyclotron. At the time of autopsy seven out of nine bombarded mice showed tumorous growths in liver and kidney while none of the animals that were not bombarded showed such changes.

Rates of metabolism to CO_2 of administered radioactive carbon labeled acetate, glycine, glucose, and tyrosine have been studied. Following intravenous administration each substance has its characteristic rate of oxidation. The CO_2 production has a rapid phase, which is believed to coincide with oxidation of the substance administered, and a slow phase resulting from the oxidation of other substances that have been derived from the labeled compound administered. The time constants of oxidation are indicative of the rate of utilization of the pool of substances which are in metabolic equilibrium with the labeled substance. The rates of oxidation appear to be characteristic for each of the above mentioned substances and there is no

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obvious or significant differences in the metabolism of these substances in normal and neoplastic animals.

10. Health Physics

Radiation Measurements. The concrete enclosure at the deuteron beam exit tube was carefully surveyed immediately after its erection. The shielding is adequate except for regions which look down into the enclosure at close range. Only the crane operator is apt to be in such locations and he is not allowed to operate the crane during external use of the beam.

Thorough surveys of the linear accelerator radiation field have been made during its increased operation this month. Locations of certain offices, facilities and work areas must be changed when more continuous operation begins. Crew members and experimenters are equipped with pocket dosimeters, film badges, and pocket electroscopes.

Monitoring Storage, and Disposal. During the month routine monitoring was carried out as usual. The use of slow neutron dosimeters has been extended to cover both the 60-inch and 184-inch cyclotron crews and associated personnel. Two hundred and three film badges were processed and recorded none of which showed an over tolerance to exposure. (The analysis of routine air sampling and of hand counting revealed no over tolerance contaminations.) A number of routine collections of radioactive waste was made and disposal of the material cared for. The storage tubes for storing radioactive materials were put into place in the pit in the chemistry annex and are now ready for use.

Research and Development. The lead glass viewer for the beta-gamma dry box is now ready to be mounted. The mechanism for the manipulators is complete while the shell design is near completion. Other items upon which research and development work is being done include the preparation of platinum capsules for pile bombardments, further work on the Conway spark box and air filters, and the erection of the spinner column that was received from Oak Ridge.

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APPROXIMATE DISTRIBUTION OF EFFORT

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