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CHARACTERISTICS OF BISMUTH FISSION WITH HIGH ENERGY PARTICLES

Robert H. Goeckermann

December 14, 1948

Berkeley, California

Thesis of
ROBERT H. GOECKERMANN

to be submitted to the
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Berkeley, California

Characteristics of Bismuth Fission with High Energy Particles

Robert H. Goeckermann

Radiation Laboratory, University of California
Berkeley, California

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ABSTRACT

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A study has been made of the characteristics of the fission induced in bismuth by irradiation with the high energy deuterons and alphas from the Berkeley 184-inch frequency-modulated cyclotron, using radiochemical methods for the determination of yields of the fission products. The fission yield curve resulting from 190 Mev deuteron fission of bismuth was found to be a single symmetrical peak with a maximum of 5.0 percent at a mass number of approximately 100. The ratio of neutrons to protons was observed to be nearly constant for all nuclides formed in relatively good yield. A survey of the effect of varying the bombardment energy indicated that the fission remains predominantly symmetrical, although the mechanism and the height of the maximum change. The excitation function of the Mo^{99} yield in two lead samples enriched, respectively, in Pb^{204} and Pb^{208} showed that the fission cross section is larger and decreases less rapidly as the deuteron energy decreases in the lead sample which has a higher proportion of the light isotopes. A survey of the fissionability of bismuth, lead, thallium, platinum, and tantalum irradiated with 380 Mev alphas showed that the fission cross section decreased by a factor of approximately 1000 from bismuth to tantalum. A mechanism has been postulated for the nuclear reaction which involves prior neutron evaporation from the initial highly excited nucleus followed by fission with unchanged charge distribution.

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CHARACTERISTICS OF BISMUTH FISSION WITH HIGH ENERGY PARTICLES

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Section I

Introduction

Interest in the mechanism of nuclear fission and the fissionability of a nuclide began with the observation of fission of U^{235} induced by absorption of a slow neutron. Neutrons of a few Mev energy have been found sufficient to induce fission in thorium and all higher elements. The heaviest stable elements, terminating in Bi^{209} , have been irradiated previously with 14 Mev neutrons, 20 Mev deuterons, 40 Mev helium ions, and 100 Mev bremsstrahlung with no fission detected and reported at these or lower energies. The 184-inch frequency-modulated cyclotron made available at Berkeley 90 Mev neutrons, 190 Mev deuterons, and 380 Mev helium ions. Fission of elements over the range from tantalum to bismuth induced by one or more of these projectiles was first observed by Templeton, Howland, Perlman, and Goeckermann, by chemical identification of radioactive fission products, soon after the cyclotron began operating¹. This thesis describes work which followed the discovery of the fission of these elements. It is concerned principally with the fission of bismuth with 190 Mev deuterons and shows that the characteristics and apparent mechanism of the reaction differs markedly from fission induced by low energy particles or quanta². Physical studies^{3,4} of the fission fragment energies in irradiations with the 90 Mev neutrons support the general conclusions presented in Section VII.

The yields of nuclides resulting from 190 Mev deuteron fission of bismuth were determined by studying the activities of 27 elements from

$Z = 20$ to $Z = 63$ ---- Ca, Cr, Fe, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Nb, Mo, Ru, Pd, Ag, Cd, Sb, Te, I, Cs, Ba, Ce, and Eu. The yields of fifty radioactive nuclides from mass number 45 to 149 were measured and used to determine the distribution of fission yield as a function of mass number. Bismuth and two lead samples of different isotopic composition were bombarded at several energies to further elucidate the nature of the fission process.

Section II

Experimental Procedures

Since the main investigation was that of the fission of bismuth with 190 Mev deuterons and the techniques used in other phases of the work were similar, the experimental procedures will be discussed in detail for this bismuth fission and any important differences in the other studies will be mentioned in the section devoted to that study.

Bombardment Technique

The circulating beam of the 184-inch frequency-modulated cyclotron was used almost exclusively because its intensity is much greater than the deflected beams. The average beam is of the order of one microampere, but neither it nor its day-to-day variations were measured or known accurately. Therefore it was necessary to use an internal monitor--in this case 67 hr β^- -emitting Mo^{99} --in each bombardment and measure all yields relative to it. The absolute yield of Mo^{99} was later determined using a deflected collimated measured beam ($3.0 \times 10^{-5} \mu\text{a}$) of 190 Mev deuterons.

Bismuth strips $0.1 \times 1 \times 3$ cm were machined from an ingot obtained from the American Smelting and Refining Company. Spectrographic analyses showed the absence of any impurity that might interfere with the measurements. Because the circulating beam is subject to radial oscillations of ~ 0.2 cm effective amplitude and a vertical spread of ~ 1 cm at 81" radius (190 Mev deuterons), the bismuth strips were mounted on a water-cooled copper block so as to expose to the beam an area at least 0.3×3 cm, which was assumed to intercept the entire beam. In the case of two targets bombarded intermittently over a period of several weeks to study the long-lived fission products, the bismuth was actually melted into a slot cut in the leading edge of the copper block. One to three-hour bombardments were used in the study of the short-lived activities. Only a few elements were studied in any one of the short bombardments.

Chemical Procedures

The irradiated portion of the bismuth was dissolved in hot $6N$ HNO_3 and stored in a closed calibrated glass container. Aliquots of this solution were used for the study of the activities of a particular element. Liter amounts of carrier solutions had been prepared, by weighing out some suitable compound to contain 10 mg/ml of the element. (A few of these carrier solutions have been standardized and found to be accurate within 10%.) One or two ml of this solution of an element was added to the aliquot of the bismuth solution as carrier for the radioactive isotopes present, precautions to insure exchange of active and inactive atoms taken in some cases, and the element purified from all others which would interfere in the radioactivity measurements. Radioactive isotopes of most of the elements up to astatine are probably produced to some extent but the predominant fission products are in the calcium(20) to europium(63) region and the predominant spallation products are in the hafnium(72) to astatine(85) region. From the determined fission cross section it is estimated that the yield of spallation products is several times as great as the yield of fission products. Part or all of the purified element was then precipitated as a weighable compound, filtered through a Hirsch funnel on a tared ~ 1 cm diameter filter paper and the chemical yield determined. The filter paper is washed and dried like the precipitate it will carry prior to taring. Most of the chemistry used was adapted from fission product procedure reports to appear as part of the National Nuclear Energy Series⁵. The procedures are outlined here primarily because no compilation of them is yet generally available. The amounts of scavengers used are only approximate. All precipitates were separated by centrifugation except the final one in each case.

(1) Calcium (adaptation)

- (a) Add 20 mg Ca and 10 mg each of Ru, Cb, Fe, and La. Make 0.5N in HCl and precipitate with H_2S .
- (b) Add 10 mg Ru and Bi to supernatant and repeat H_2S precipitation.
- (c) Boil out H_2S and precipitate with NH_3 .
- (d) Add 10 mg Fe and Y to supernatant and repeat NH_3 precipitation.
- (e) Boil down to 5 ml, add 10 mg Ba and Sr, and precipitate with cold fuming HNO_3 .
- (f) Add more Sr and Ba carriers to supernatant and repeat precipitation three times.
- (g) Add Sr alone and repeat two times more.
- (h) Boil down to ~ 5 ml, add 5 ml sat $(NH_4)_2C_2O_4$, and make basic with NH_3 .
- (i) Dissolve CaC_2O_4 in HNO_3 , destroy $C_2O_4^{2-}$ with $KClO_3$, and make basic with NH_3 . Add Fe, La, and Y.
- (j) Add more Fe, La, and Y to supernatant and repeat precipitation.
- (k) Repeat step (e).
- (l) Boil supernatant down to ~ 5 ml, make basic with NH_3 , heat, add 5 ml sat $(NH_4)_2C_2O_4$ slowly. Stir 2 min., filter, wash three times with 5 ml portions of H_2O , three times with 5 ml 95% C_2H_5OH , three times with 5 ml ether. Dry in vacuum dessicator--2 min. evac., release, 5 min. evacuation. Weigh as CaC_2O_4 (31.97 mg per 10 mg Ca).

The chemical yield was $\sim 20\%$.

(2) Chromium (new).

- (a) Add 10 mg Cr as $Cr_2O_7^{2-}$. Add HCl and HCOOH to reduce to Cr^{+3} . Precipitate Bi_2S_3 and Sb_2S_3 scavenger from 1N HCl solution.
- (b) Precipitate $Cr(OH)_3$ with $K_2CO_3 + H_2S$, by long boiling.
- (c) Oxidize in 10 ml 0.5N NaOH with H_2O_2 , scavenge with $Fe(OH)_3$, and destroy H_2O_2 .

- (d) Make slightly acid with $6N H_2SO_4$ and wash with ether.
- (e) Add H_2O_2 and extract H_3CrO_8 into ether.
- (f) Reoxidize and re-extract aqueous phase.
- (g) Evaporate ether over an H_2O layer and repeat H_3CrO_8 extraction.
- (h) Evaporate ether over H_2O , oxidize with H_2O_2 in alkaline solution, scavenge with $Fe(OH)_3$.
- (i) Acidify with $6N HNO_3$, add 1 ml $6N CH_3COOH$ and 2 ml $6N CH_3COONH_4$, heat, add several ml 50 g/l Ba solution. Filter, wash with hot H_2O and C_2H_5OH , dry 10 min. at $110^\circ C$. Weigh as $BaCrO_4$ (48.73 mg per 10 mg Cr).

The chemical yield was $\sim 50\%$.

(3) Iron (adaptation)

- (a) Extract 15 ml of $9N HCl$ containing 10 mg Fe^{+3} with 15 ml isopropyl ether. Wash ether twelve times with 10 ml portions of $7.5 N HCl$. Extract Fe^{+3} into 10 ml H_2O and evaporate dissolved ether.
- (b) Scavenge with Sb_2S_3 from HCl solution--repeat.
- (c) Scavenge with TeS by H_2S precipitation.
- (d) Precipitate $Fe(OH)_3$ with NH_3 and dissolve in HCl .
- (e) Fume twice with $HClO_4$ after adding Ru.
- (f) Precipitate $Fe(OH)_3$ with NH_3 --repeat.
- (g) Scavenge with $BaSO_4$.
- (h) Scavenge with AgI (excess Ag).
- (i) Scavenge with CB_2O_5 by adding HNO_3 and $KClO_3$ to oxalate solution.
- (j) Scavenge with Zr phenylarsonate.
- (k) Ether extract Fe^{+3} again, precipitate as $Fe(OH)_3$ with NH_3 , filter and ignite to Fe_2O_3 .

(4) Nickel (new)

- (a) Add 20 mg Ni, make neutral, add 3-5 ml 1% dimethylglyoxime in

alcohol. Wash.

- (b) Dissolve Ni dimethylglyoxime in 1 ml concentrated HNO_3 , dilute, and scavenge twice with CuS .
- (c) Scavenge three times with PdS .
- (d) Scavenge with Sb_2S_3 .
- (e) Boil out H_2S , make basic with NH_3 , scavenge twice with $\text{Fe}(\text{OH})_3$.
- (f) Make 0.5N in HCl and scavenge with Pd dimethylglyoxime.
- (g) Neutralize supernatant. Wash Ni dimethylglyoxime.
- (h) Dissolve in concentrated HNO_3 , make basic with NH_3 and scavenge with AgI .
- (i) Neutralize supernatant and precipitate Ni dimethylglyoxime.
- (j) Dissolve in concentrated HNO_3 , reprecipitate Ni dimethylglyoxime, filter, wash with H_2O and $\text{C}_2\text{H}_5\text{OH}$, dry 10 min. at 110°C . Weigh as Ni dimethylglyoxime (29.44 mg per 10 mg Ni).

The chemical yield was $\sim 60\%$.

(5) Copper (new).

- (a) Add 10 mg Cu , make basic with NH_3 , centrifuge out $\text{Bi}(\text{OH})_3$ and scavenge with $\text{Fe}(\text{OH})_3$ and BaCO_3 (add K_2CO_3).
- (b) Make 0.5N in HCl . Bubble in SO_2 , add KCNS until CuCNS precipitates.
- (c) Dissolve CuCNS in HNO_3 and make neutral. Add KCN and scavenge with CdS .
- (d) Acidify and boil off HCN , add Sr and Y , precipitate CuS from 3N HCl .
- (e) Dissolve in HNO_3 , add HCl and destroy HNO_3 . Dilute to 0.5N HCl and scavenge twice with AgCl .
- (f) Precipitate CuCNS from supernatant, filter, wash with H_2O and $\text{C}_2\text{H}_5\text{OH}$, dry 15 min. at 110°C . Weigh as CuCNS (19.15 mg per 10 mg Cu).

(6) Zinc (adaptation).

(a) Add 10 mg Zn, 1-2 ml saturated $\text{H}_2\text{C}_2\text{O}_4$, and make 1N in HNO_3 .

Cool in ice bath, add 3 ml Zn reagent*, stir 3-5 min. (scratch glass to begin precipitation).

(b) Dissolve $\text{ZnHg}(\text{SCN})_4$ in 5 ml 2N HNO_3 and dilute to 1N . Add 1 ml saturated $\text{H}_2\text{C}_2\text{O}_4$ and 3 ml Zn reagent.

(c) Repeat (b).

(d) Dissolve precipitate in 2N HNO_3 , dilute to 1N , and precipitate HgS . Neutralize 0.3N HNO_3 with NaOH and precipitate Bi_2S_3 (5 mg Bi) three times.

(e) Make 1M in $\text{CH}_3\text{COONH}_4$ and precipitate ZnS .

(f) Dissolve precipitate in 5 ml concentrated HBr , evaporate to dryness several times, take up in 10 ml 0.5N NaOH , add 1 ml 2M Na_2CO_3 . Precipitate $\text{Fe}(\text{OH})_3$ (2 mg) and BaCO_3 (5 mg) three times from hot solution.

(g) Add HCl to make 1N and 1 ml saturated $\text{H}_2\text{C}_2\text{O}_4$. Add 3 ml Zn reagent to cold solution, stir 3 min., filter, wash three times with 5 ml absolute $\text{C}_2\text{H}_5\text{OH}$ and three times with 5 ml ether, dry 7 min. in a vacuum dessicator. Weigh as $\text{ZnHg}(\text{SCN})_4$ (76.2 mg per 10 mg Zn).

The chemical yield was $\sim 80\%$.

(7) Gallium (adaptation).

(a) Add 10 mg Ga and 2 mg Mo, destroy HNO_3 and make 6N in HCl . Extract with an equal volume of ether, wash with $1/3$ volume 6N HCl .

Re-extract Ga into $2/3$ volume of H_2O .

(b) Boil out ether. Make 0.3N in HCl and add 1 ml alcohol solution of α -benzoin oxime to precipitate Mo.

* 39 g KSCN dissolved in 200 ml H_2O . Stir in 27 g HgCl_2 while diluting to 1 liter.

- (c) Precipitate Bi_2S_3 (5 mg) three times.
- (d) Boil out H_2S . Make 1N in NaOH , 0.2M in Na_2CO_3 , and precipitate $\text{Fe}(\text{OH})_3$ and BaCO_3 scavengers three times.
- (e) Make solution 6N in HCl and extract with 1-1/2 volumes ether. Wash ether with 6N HCl and re-extract Ga^{+3} into H_2O .
- (f) Boil out ether. Evaporate to dryness with 5 ml concentrated HBr three times. Take up residue in 10 ml 6N HCl and repeat ether extraction.
- (g) Dilute H_2O layer to 35 ml and heat to 60°C . Add 3 ml of 8-hydroxyquinoline solution (50 g/l in 2M CH_3COOH) and 6M $\text{CH}_3\text{COONH}_4$ dropwise until permanent yellow precipitate forms, then add 1 ml excess. Add aerosol and filter, wash with 10 ml warm H_2O , twice with 5 ml cold H_2O , dry 15 min. at 110°C . Weigh as Ga 8-hydroxyquinolate (72 mg per 10 mg Ga).

The chemical yield was $\sim 80\%$. Gallium ferrocyanide precipitations were included in the original procedure but were tested and discarded as unsatisfactory.

(8) Arsenic (adaptation).

- (a) Add 20 mg As and 10 mg Ge, boil nearly to dryness, take up in 15 ml 6N HCl , add crystal of NH_4I , and saturate with H_2S at ice temperature. Wash with H_2S -saturated 6N H_2SO_4 .
- (b) Dissolve precipitate in 1 ml concentrated NH_4OH and dilute to 10 ml (discard any residue). Transfer to a glass still and add 10 mg each Te, Sb, and Sn. Add 10 ml concentrated HCl and distill all but 5 ml in a Cl_2 stream into 5 ml H_2O in an ice bath. Add 5 ml concentrated HCl and 10 mg Ge to residue and repeat distillation.
- (c) To residue from GeCl_4 distillation, pass in HCl gas to remove Cl_2 , add 1 ml saturated CuCl in concentrated HCl , and distill over 5 ml in

an HCl stream into 10 ml concentrated HCl in an ice bath. Add

CuCl-HCl to residue and distill further.

(d) Pass H_2S into distillate and centrifuge out As_2S_3 . Dissolve in 1 ml concentrated NH_4OH , add 10 mg Te, Sb, and Sn and repeat distillation.

(e) Precipitate As_2S_3 again, dissolve in 1 ml concentrated NH_4OH , add 10 ml concentrated HCl, saturate with H_2S , filter, wash with H_2O , $\text{C}_2\text{H}_5\text{OH}$, and ether, dry 10 min. at 110°C . Weigh as As_2S_3 (16.42 mg per 10 mg As).

The chemical yield was $\sim 50\%$.

(9) Selenium (adaptation).

(a) Add 10 mg Se and Te, 10 ml concentrated HBr, and 0.5 ml liquid Br_2 in a glass still. Distill in air stream to 3 ml residue, into 5 ml saturated Br_2 water in an ice bath.

(b) Keep at ice temperature and reduce to Se^0 (red) with SO_2 or $\text{NH}_2\text{OH}\cdot\text{HCl}$. Add aerosol and centrifuge.

(c) Dissolve Se^0 in few drops concentrated HNO_3 , add 10 ml concentrated HCl and reduce with SO_2 in an ice bath. Centrifuge with aerosol.

(d) Repeat SeBr_4 distillation and Se^0 precipitations as often as necessary for desired purity.

(e) Precipitate Se^0 , filter, wash three times with 5 ml H_2O , three times with 5 ml $\text{C}_2\text{H}_5\text{OH}$, three times with 5 ml ether, dry 10 min. at 110° . Weigh as Se^0 .

The chemical yield was $\sim 90\%$.

(10) Bromine (adaptation).

(a) Add 10 mg Br^- and I^- , make 1N in HNO_3 , add a few drops of 0.1M NaNO_2 , and extract I_2 into an equal volume of CCl_4 .

(b) Add equal volume CCl_4 to aqueous phase, and add 0.1M KMnO_4 .

dropwise until color persists. Make five extractions of Br_2 .

(c) Combine CCl_4 and wash with 1N HNO_3 containing KMnO_4 , add 10 ml H_2O and 0.1N NaHSO_3 dropwise until both layers are colorless. Wash aqueous layer with CCl_4 .

(d) Make 1N in HNO_3 , add 10 mg I^- , warm a few min. to oxidize SO_3^{2-} , add a few drops of 0.1M NaNO_2 , and extract I_2 five times with 10 ml CCl_4 .

(e) Repeat KMnO_4 oxidation and Br_2 extraction and reduction.

(f) Repeat I_2 and Br_2 extraction cycles as needed for purity.

(g) Make last aqueous solution of Br^- 1N in HNO_3 , heat, add 2 ml 0.1N AgNO_3 , stir, digest a few min., filter, wash three times with 5 ml H_2O , three times with 5 ml $\text{C}_2\text{H}_5\text{OH}$, three times with 5 ml ether, dry 10 min. at 110°C . Weigh as AgBr (23.5 mg per 10 mg Br).

The chemical yield was $\sim 60\%$. To prevent possible losses of the trace Br during dissolving of the bismuth, one might dissolve the target in 6N HNO_3 with the carrier Br^- present in a glass still and distill Br_2 or HBr directly from this solution into H_2O in an ice bath.

(11) Rubidium-Cesium (new). Rubidium and cesium were purified together and then separated.

(a) Add 20 mg Rb and Cs and 5 ml concentrated HClO_4 . Evaporate to fuming, cool, transfer to centrifuge tube with 15 ml absolute $\text{C}_2\text{H}_5\text{OH}$. Cool in ice bath 10 min. with stirring. Wash precipitate twice with 10 ml absolute alcohol.

(b) Dissolve in dilute HCl and scavenge with Te , Ru , Sn , Sb , and Ag sulfides (2 mg each).

(c) Scavenge with NH_3 precipitation of La , Ce , Y , Zr , and Cb (2 mg each).

(d) Scavenge with Sr and Ba carbonates (4 mg).

- (e) Acidify with HCl, repeat (b).
- (f) Repeat (c).
- (g) Scavenge with $\text{La}(\text{OH})_3$.
- (h) Scavenge with Sr and Ba carbonates and Y, Zr, and Nb hydroxides.
- (i) Acidify with HCl, evaporate to dryness, bake off NH_4Cl (few drops HNO_3 help).
- (j) Dissolve in a minimum of H_2O , transfer to cone and centrifuge out any residue, evaporate to dryness. Add 6 ml H_2PtCl_6 and 6 ml $\text{C}_2\text{H}_5\text{OH}$, wash Rb and Cs chloroplatinates with $\text{C}_2\text{H}_5\text{OH}$.
- (k) Dissolve in HCl and repeat Te sulfide scavenging.
- (l) Repeat NH_3 precipitation of La and Fe hydroxides.
- (m) Repeat Sr and Ba carbonate scavenging.
- (n) Acidify with HNO_3 , add HCl, evaporate to dryness, bake off ammonium salts.
- (o) Cs - Take up residue in 20 ml 6N HCl, add 1 ml silicotungstic acid solution and digest. (Save this supernatant for Nb fraction). Wash twice with 5 ml 6N HCl (discard). Dissolve Cs silicotungstate in dilute NaOH, add 20 ml 6N HCl and discard yellow precipitate. Add 1 ml Cs reagent, digest, filter, wash with 6N HCl, three times with 5 ml acetone, dry 10 min. at 110°C . Weigh as $\text{Cs}_8\text{SiW}_{12}\text{O}_{42}$ (37.3 mg per 10 mg Cs)--standardize the Cs carrier solution by the same procedure. The chemical yield was $\sim 40\%$.
- (p) Rb - Precipitate a Cs silicotungstate scavenger from the supernatant saved. Evaporate until crystallization begins, add H_2PtCl_6 and $\text{C}_2\text{H}_5\text{OH}$, filter, wash with $\text{C}_2\text{H}_5\text{OH}$, dry 10 min. at 110° . Weigh as Rb_2PtCl_6 (33.9 mg per 10 mg Rb). The chemical yield was $\sim 30\%$.
- Sodium 6-chloro-5-nitrotoluenesulfonate is a good specific precipitant for Rb but was not obtainable in time to be used.

(12) Strontium - Barium (adaptation). Strontium and barium were partly purified together and then separated.

- (a) Add 10 mg Sr and Ba, 30 ml fuming HNO_3 , digest cold 1-2 min.
- (b) Dissolve precipitate in 2 ml H_2O and reprecipitate with 15 ml fuming HNO_3 .
- (c) Dissolve precipitate in 5-10 H_2O , add 5 mg Fe^{+3} , and precipitate $\text{Fe}(\text{OH})_3$ with $\text{NH}_3(\text{CO}_3^- \text{ free!})$. Repeat $\text{Fe}(\text{OH})_3$ scavenging.
- (d) Neutralize supernatant with 6N HNO_3 , add 1 ml 6M CH_3COOH and 2 ml 6M $\text{CH}_3\text{COONH}_4$. Heat to boiling and add 1 ml 1.5M Na_2CrO_4 dropwise with stirring. Digest one min. (Save the supernatant for Sr fraction).
- (e) Ba - Wash BaCrO_4 precipitate with 10 ml hot H_2O . Dissolve in 1 ml 6N HCl , add Sr, and reprecipitate BaCrO_4 . Redissolve, add 15 ml ether - HCl reagent (5 parts concentrated HCl to 1 part ether), digest cold 2 min., wash with 5 ml absolute alcohol containing a few drops of HCl .

Dissolve BaCl_2 in 1 ml H_2O , make just basic with NH_3 and scavenge with $\text{Fe}(\text{OH})_3$. Add 15 ml ether - HCl and reprecipitate BaCl_2 . Repeat if necessary. Filter last BaCl_2 precipitate, wash three times with 5 ml ether, dry in vacuum dessicator - 2 min. evacuation, release, 5 min. evacuation. Weigh as $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ (17.78 mg per 10 mg Ba).

The chemical yield was $\sim 50\text{-}80\%$.

- (f) Sr - Precipitate BaCrO_4 scavenger from the supernatant saved. Add 2 ml concentrated NH_4OH , heat, add 5 ml saturated $(\text{NH}_4)_2\text{C}_2\text{O}_4$ slowly. Stir 2 min., filter, wash three times with 5 ml H_2O , three times with 5 ml $\text{C}_2\text{H}_5\text{OH}$, three times with 5 ml ether, dry like BaCl_2 . Weigh as $\text{SrC}_2\text{O}_4 \cdot \text{H}_2\text{O}$ (22.10 mg per 10 mg Sr). The chemical yield was $\sim 80\%$.

(13) Yttrium (adaptation).

- (a) Add 10 mg Y, La, Ce, and Zr, make 2N in HCl , and add 2 ml concen-

trated HF. Wash precipitate with 10 ml 1N HCl containing HF.

(b) Dissolve in 1 ml saturated H_3BO_3 and 8 ml concentrated HNO_3 . Add 0.2 g $KBrO_3$ and 20 ml 0.35 M HIO_3 , digest 3 min. in ice bath. Centrifuge $Ce(IO_3)_4$.

(c) Add second portion $KBrO_3$, 10 mg Ce and Zr, and digest 5 min. in ice bath.

(d) Add 9 ml 19M NaOH to supernatant. Wash precipitate with H_2O .

(e) Dissolve precipitate in 1 ml concentrated HCl, dilute to 8 ml, reduce with SO_2 . Precipitate hydroxides with NH_3 and wash.

(f) Dissolve in HCl and scavenge with Bi_2S_3 twice.

(g) Boil out H_2S , make 2N in H^+ and add 2 ml concentrated HF. Wash.

(h) Metathesize with NaOH and wash. Dissolve in HCl and evaporate to dryness. Add 3 ml 50% K_2CO_3 and heat to dissolve precipitate. Add 20 ml H_2O and heat until gelatinous precipitate becomes crystalline, then 2 min. longer. Centrifuge La group precipitate.

(i) Acidify supernatant with HCl, precipitate $Y(OH)_3$ with NH_3 , and wash twice with H_2O . (May repeat La group separation.)

(j) Dissolve in 1 ml 6N HCl, add 15 ml H_2O , boil, add 15 ml saturated $H_2C_2O_4$, digest 10 min. in ice bath, filter, wash with 3 ml H_2O , three times with 5 ml C_2H_5OH , three times with 5 ml ether, dry in vacuum dessicator - 2 min. evacuation, release, 2 min. evacuation, release, 2 min evacuation. Weigh as $Y_2(C_2O_4)_3 \cdot 10H_2O$ (35.0 mg per 10 mg Y).

The chemical yield was $\sim 70\%$. Sometimes no separation from Ce or La group activities was made.

(14) Zirconium (adaptation).

(a) Add 10 mg Zr, make sample up to ~ 5 ml of 5N HNO_3 . Add 2 ml concentrated HF and 10 mg La. Repeat LaF_3 precipitation.

(b) Add 50 mg Ba to supernatant, Wash.

- (c) Dissolve BaZrF_6 in 2 ml 5% H_3BO_3 , add 1 ml concentrated HNO_3 and 5 ml H_2O , repeat LaF_3 precipitation twice.
- (d) Dissolve with 2 ml H_3BO_3 , 2 ml concentrated HCl , and 5 ml H_2O . Add 1 drop concentrated H_2SO_4 and centrifuge out BaSO_4 .
- (e) Dilute supernatant to 20 ml and add 2 ml 6% cupferron in an ice bath, filter, wash with cold 1N HCl containing cupferron, ignite in a porcelain crucible. Weigh as ZrO_2 (13.51 mg per 10 mg Zr).

The chemical yield was $\sim 60\%$.

(15) Columbium (adaptation).

- (a) Add equal volume of concentrated HNO_3 to sample, 1 ml 6N HCl , 1 ml saturated $\text{H}_2\text{C}_2\text{O}_4$, 20 mg Cb, and 10 mg Zr. Heat and add 0.5 g KBrO_3 in small portions. Digest 5 min. and centrifuge Cb_2O_5 . Wash precipitate hot with 3 ml 6N HNO_3 , 2 ml 6N NH_4OH , and 5 ml H_2O .
- (b) Dissolve in 1-2 ml saturated $\text{H}_2\text{C}_2\text{O}_4$ + 10 drops 6N HCl . Make up to 20 ml of 6N HCl , 0.05M H_2SO_3 , and add 10 ml CHCl_3 . Cool in ice bath, add 2 ml fresh 6% cupferron and shake well. Extract a second time with more CHCl_3 and cupferron. Wash CHCl_3 with 20 ml cold 6N HCl and 0.05M H_2SO_3 containing more cupferron.
- (c) Boil with 10 ml concentrated HNO_3 , adding KBrO_3 until CHCl_3 is gone, solution is pale yellow and Cb_2O_5 has precipitated.
- (d) Transfer to lusteroid with dil. HNO_3 . Dissolve by adding 1 ml concentrated HF . Add 2 ml 6N HNO_3 , 10 mg Zr, and 5 ml H_2O . Add 50 mg Ba dropwise and centrifuge BaZrF_6 .
- (e) Add 6 ml concentrated NH_4OH to supernatant (to pH . 8-10). Centrifuge Cb_2O_5 , and wash with 3 ml 6N NH_4OH , 1 ml 6N HNO_3 , and 5 ml H_2O . Use a second wash to transfer precipitate back to glass.
- (f) Dissolve hot in 2 ml saturated $\text{H}_2\text{C}_2\text{O}_4$ plus 10 drops 6N HNO_3 . Add 3 ml H_2O , 5 ml concentrated HNO_3 , heat, add 0.5 g KBrO_3 slowly,

digest 5 min., centrifuge. Stir precipitate up with 3 ml 6N HNO_3 , 2 ml 6N NH_4OH , and 5 ml H_2O , heat, filter, ignite in a crucible 15 min. Weigh as Cb_2O_5 (14.30 mg per 10 mg Cb).

The chemical yield was $\sim 50\%$.

(16) Molybdenum (adaptation).

(a) Add 10 mg Mo and concentrated HCl , destroy HNO_3 . Make up to 5 ml 6N HCl , add one drop Br_2 water, extract twice with 15 ml ether. Combine ether layers and wash twice with 2 ml 6N HCl . Evaporate ether over 5 ml H_2O .

(b) Boil out ether, dilute to 20 ml, add 5 mg Fe^{+3} and precipitate with NH_3 .

(c) Add 6 ml concentrated HNO_3 and 1 ml saturated $\text{H}_2\text{C}_2\text{O}_4$ to supernatant, cool, add 5 ml 2% solution of α -benzoin oxime in alcohol, digest 2 min., add aerosol and centrifuge 5 min. (If the Mo oxime does not settle well, dilute the solution with $\text{C}_2\text{H}_5\text{OH}$). Wash twice with 20 ml 1N HNO_3 .

(d) Add 2 ml concentrated HNO_3 and 1 ml concentrated HClO_4 and boil carefully to fuming. Cool, dilute to 15 ml with H_2O , add 5 mg Fe^{+3} , and precipitate with NH_3 .

(e) Add 1 drop methyl orange to supernatant and make just acid with 6N H_2SO_4 . Buffer with 1 ml 10% CH_3COONa , boil, add 10 drops 1M AgNO_3 , cool slowly with stirring, filter, wash five times with 5 ml 0.03 M AgNO_3 , three times with 5 ml $\text{C}_2\text{H}_5\text{OH}$, dry 15 min. at 110°C . Weigh as Ag_2MoO_4 (39.16 mg per 10 mg Mo).

The chemical yield was $\sim 50\text{-}80\%$. More cycles of ether extraction and oxime precipitation may be added if needed for purity.

(17) Ruthenium (adaptation).

(a) Add 10 mg Ru and Os, boil 6N HNO_3 solution to volatilize OsO_4 .

Transfer to glass still, add 10 mg I^- , 0.5 g $NaBiO_3$, 1 ml concentrated H_3PO_4 , and 10 ml 70% $HClO_4$. Boil in air stream without bumping and distill over RuO_4 into 12 ml 6N NaOH in an ice bath. Distill until 1-2 ml of $HClO_4$ have distilled (2-3 min. after fuming begins).

(b) To distillate, add 3 ml C_2H_5OH and boil 1-2 min. until Ru oxide is coagulated. Wash with 10 ml H_2O and 1 ml 6N NaOH and boil.

(c) Distill RuO_4 again, if necessary, and reprecipitate oxide.

(d) Dissolve precipitate in 2 ml hot 6N HCl and dilute to 12 ml.

Add ~ 0.2 g Mg chips or coarse powder slowly (add aerosol) until solution passes through blue stage and Ru^0 appears. Boil gently until coagulated. Add 5 ml concentrated HCl to remove excess Mg and boil, filter, wash three times with 5 ml hot H_2O , three times with 5 ml C_2H_5OH , three times with 5 ml ether, dry 10 min. at $110^\circ C$. Weigh as Ru^0 .

The chemical yield was $\sim 60\%$.

(18) Palladium (adaptation).

(a) Add 10 mg Pd^{+2} , make up to 20 ml 0.5N HCl, add 5 ml dimethylglyoxime solution (1% in alcohol). Wash with dilute HCl.

(b) Dissolve precipitate in 1 ml concentrated HNO_3 , dilute with 10 ml H_2O , add 5 mg Fe^{+3} and make basic with NH_3 .

(c) Add 10 mg Ag to supernatant and enough I^- to precipitate all the Ag. Repeat AgI scavenging.

(d) Make supernatant 0.5 N in HCl and centrifuge out any AgCl. Add 5 ml dimethylglyoxime. Wash.

(e) Repeat purification cycle if needed for higher purity.

(f) Filter last Pd dimethylglyoxime, wash with H_2O and C_2H_5OH , dry 10 min. at $110^\circ C$. Weigh as Pd dimethylglyoxime (20.70 mg per 10 mg Pd).

The chemical yield was $\sim 60\%$.

(19) Silver (adaptation).

- (a) Add 10 mg Ag, aerosol, dilute to 20 ml, and precipitate AgCl with 1 ml 6N HCl. Wash with 10 ml H₂O.
- (b) Dissolve AgCl in 2 ml 6N NH₄OH, dilute to 10 ml, and add 5 mg Fe⁺³.
- (c) Saturate supernatant with H₂S in the cold, wash Ag₂S.
- (d) Dissolve Ag₂S in 1 ml concentrated HNO₃, dilute to 20 ml, and precipitate AgCl with 1 ml 6N HCl. Wash with H₂O.
- (e) Repeat (b).
- (f) Repeat (c).
- (g) Dissolve Ag₂S in 1 ml concentrated HNO₃, dilute to 10 ml, add 5 mg Fe⁺³, and make basic with NH₃.
- (h) Add 5 drops 6N HCl to supernatant, aerosol, make 1N in HNO₃, heat, filter, wash three times with 5 ml H₂O, three times with 5 ml C₂H₅OH, dry 10 min. at 110° C. Weigh as AgCl (13.28 mg per 10 mg Ag).

The chemical yield was ~ 80%.

(20) Cadmium (adaptation).

- (a) Add 10 mg Cd and make basic with NH₃, centrifuge out Bi(OH)₃.
- (b) Add 10 ml concentrated HCl to supernatant and evaporate to dryness. Take up in 20 ml 0.2N HCl and saturate with H₂S. Wash CdS.
- (c) Dissolve CdS in 1 ml 6N HCl, boil out H₂S, dilute to 10 ml, add 5 mg Fe⁺³ and 6N NH₄OH dropwise until Fe(OH)₃ precipitation begins. Redissolve in 1-2 drops HCl, heat, add 4-5 drops 6N CH₃COONH₄, centrifuge out basic ferric acetate precipitate.
- (d) Add 10-15 drops 6N HCl to supernatant and precipitate CdS.
- (e) Dissolve CdS in HCl and expel H₂S, add 1N and precipitate with NH₃.
- (f) Re-acidify and precipitate CdS.
- (g) Dissolve CdS in 2 ml 6N HCl, dilute to 10 ml, add 10 mg Pd⁺², heat,

and saturate with H_2S .

(h) Add Sb^{+3} to supernatant and scavenge with Sb_2S_3 .

(i) To supernatant add excess NH_4OH and saturate with H_2S .

(j) Dissolve CdS in a few drops of HCl , expel H_2S , dilute to 15 ml, add 2 ml $3M$ HCl , boil, add 2 ml $1M$ $(NH_4)_2HPO_4$, and digest hot 15 min. Filter, wash with H_2O , dry 10 min. at $110^\circ C$. Weigh as $CdNH_4PO_4 \cdot H_2O$ (21.66 mg per 10 mg Cd).

The chemical yield was $\sim 70\%$.

(21) Antimony. No satisfactory procedure was found but the following three were tried:

(a) Precipitations of Sb_2O_5 with fuming HNO_3 , distillations of $SbCl_3$ from H_2SO_4 in an HCl stream, precipitations of Sb_2S_3 from hot $3N$ HCl ; separations of $AsCl_3$ by distillation, and As_2S_3 precipitation.

(Adaptation).

(b) Precipitations of Sb_2O_5 , electrolysis of SbH_3 from $6N$ H_2SO_4 with a Pb cathode⁶, reaction of SbH_3 with $AgNO_3$ solution to precipitate $SbAg_3$, reaction of Ag_3Sb with HCl to precipitate Ag and $AgCl$ to give $SbCl_3$, scavenging with As_2S_3 , precipitations of Sb_2S_3 from hot $3N$ HCl . (New).

(c) Precipitations of Sb_2O_5 and Sb_2S_3 ; scavenging with As_2S_3 , MoS_3 , Te^0 , $Fe(OH)_3$ from KOH solution, and Bi_2S_3 from K_2S solution. (New).

(22) Tellurium. (adaptation).

(a) Add 10 mg Te and Se and concentrated HBr , boil nearly to dryness a few times. Take up residue in 10 ml concentrated HCl , add Se , precipitate Se^0 with SO_2 in an ice bath.

(b) Dilute to $3N$ HCl , heat, and precipitate Te^0 with SO_2 , centrifuge with aerosol. Wash with HCl .

(c) Dissolve Te^0 in a few drops HNO_3 , evaporate excess acid, dilute

to 10 ml, add 6N NaOH dropwise until H_2TeO_3 precipitate forms and then redissolves, 10 drops excess. Scavenge with 1-2 mg $\text{Fe}(\text{OH})_3$.

(d) Make supernatant 3N in HCl and precipitate Te^0 .

(e) Repeat Se^0 , Te^0 , and $\text{Fe}(\text{OH})_3$ precipitations several times.

(f) Filter last Te^0 precipitate, wash three times with 5 ml H_2O , three times with 5 ml $\text{C}_2\text{H}_5\text{OH}$, three times with 5 ml ether, dry 10 min. at 110°C . Weigh as Te^0 .

The chemical yield was $\sim 60\%$.

(23) Iodine (First steps are new).

(a) Place target in glass still, add 10 mg I^- and 10 ml 6N HNO_3 , warm to dissolve. When entirely dissolved, distill over the I_2 and a few ml HNO_3 into H_2O in an ice bath.

(b) Reduce I_2 with a minimum of NaHSO_3 in the cold, adjust, to 0.5N HNO_3 , add a few drops 0.1M NaNO_2 , and extract I_2 into an equal volume of CCl_4 . Extract aqueous with second portion CCl_4 . Wash CCl_4 with dilute HNO_3 .

(c) Shake CCl_4 with 10 ml H_2O , adding NaHSO_3 until both phases colorless. Wash aqueous with CCl_4 .

(d) Repeat extraction cycle several times.

(e) Make last aqueous I^- solution 0.5N in HNO_3 , heat to boiling, add 2 ml 0.1N AgNO_3 , digest, filter, wash three times with H_2O , three times with 5 ml $\text{C}_2\text{H}_5\text{OH}$, three times with 5 ml ether, dry 10 min. at 110°C . Weigh as AgI (18.50 mg per 10 mg I).

The chemical yield was $\sim 40\%$. Active iodine formed as periodate may not exchange with the carrier by this method.

(24) Cesium.

See rubidium procedure.

(25) Barium.

See strontium procedure.

(26) Cerium-Europium (adaptation). Cerium and europium were purified together and then separated from the other rare earths.

- (a) Add 20 mg Ce, Eu, and Y. Make $2N$ in HCl with Zr, Cb, and Sr present and precipitate RE fluorides.
- (b) Dissolve and reprecipitate RE fluorides from $2N$ HNO_3 with Zr and Cb present.
- (c) Scavenge twice with Bi_2S_3 and RuS from $0.5N$ HCl.
- (d) Precipitate RE hydroxides twice with NH_3 ($CO_3^{=}$ free) and Sr present.
- (e) Precipitate RE fluorides from $2N$ HCl twice with Zr and Cb present.
- (f) Scavenge with Bi_2S_3 and RuS from $0.3N$ HCl.
- (g) Precipitate RE hydroxides with NH_3 three times with Sr present.
- (h) Precipitate RE oxalates from dilute HCl.
- (i) Precipitate RE fluorides from HNO_3 .
- (j) Repeat (g).
- (k) Displace air with N_2 , reduce with Zn amalgam, precipitate $Y(OH)_3$ and $Ce(OH)_3$ with NH_3 . Dissolve and reduce again, reprecipitate hydroxides.
- (l) Eu - Oxidize supernatants from hydroxide precipitations with ozone, precipitate $Eu(OH)_3$ with NH_3 . Dissolve in HCl, add Ce, reduce to Eu^{+2} and precipitate $Ce(OH)_3$. Repeat separation cycle three times. Precipitate $Eu_2(C_2O_4)_3$ like $Y_2(C_2O_4)_3$ and treat similarly. Weigh as $Eu_2(C_2O_4)_3 \cdot 10H_2O$ (24.6 mg per 10 mg Eu). The chemical yield was $\sim 15\%$.
- (m) Ce - Precipitate $Ce(IO_3)_4$ twice, then $Ce_3(PO_4)_4$ three times. Precipitate CeF_3 from HCl, metathesize to $Ce(OH)_3$, dissolve in HCl, precipitate $Ce(OH)_3$ with NH_3 . Dissolve in HCl. Weigh as $Ce_2(C_2O_4)_3 \cdot 10H_2O$ (25.84 mg per 10 mg Ce). The chemical yield was $\sim 30\%$.

Isotope Identification

The filter paper discs, on which the final precipitates were spread over $\sim 1 \text{ cm}^2$, were mounted in the center of rectangular cardboard holders and covered with $\sim 2.5 \text{ mg/cm}^2$ cellophane to hold the sample in place. The samples were counted on a $\sim 3 \text{ mg/cm}^2$ mica end-window Geiger Müller tube mounted on a lucite base, having five sets of slots for samples, inside an aluminum-lined lead shield. Particles which enter the tube are assumed to count with 100% efficiency. Quanta counting efficiencies, however, were assumed to be as follows: 13 Kev - 1.0%, 18 Kev - 0.8%, 26 Kev - 0.6%, 0.3 - .5 Mev - 0.5% and 0.5 Mev - 1% per Mev. A natural uranium standard was counted occasionally to correct for any systematic variation in tube or circuit operation. All counting rates were corrected for coincidences and background of the tube. Absorption curves of the particles emitted, for identification and extrapolation to zero absorber, were taken in beryllium so that absorption of x-rays present and bremsstrahlung production would be minimized. Absorption curves of the x-rays and gammas were obtained by placing the aluminum or lead absorbers on top of a slab of beryllium thick enough to stop all the particles and then covering with another slab of beryllium to stop the Compton and photo-electrons. Decay curves of the various particle and quantum components of the radiation were obtained by counting through beryllium absorbers and beryllium-lead-beryllium sandwiches similarly. No back-scattering corrections were made, but the correction to zero absorber included one-half the weight of the precipitate in addition to the second-shelf cellophane + air + window thickness of $\sim 8 \text{ mg/cm}^2$.

A simple form of 180° β -ray spectrometer was used frequently to determine the sign (positron or negatron), energy, and nature (β^+ or e^-) of the particles as well as follow the decay of a small number of positrons in the

presence of an overwhelming number of negatrons. Yield values were obtained from this instrument by counting a natural uranium standard on it and on the standard counter. In addition to these counting techniques, separations of daughter activities which had grown into purified chemical fractions were sometimes used for identification or yield measurements.

A summary of the nuclides identified in 190 Mev deuteron fission of bismuth follows. The properties of these nuclides are not listed here since they appear in the Table of Isotopes⁷. Yields were calculated from β^- 's emitted by the nuclide unless otherwise indicated. No mention of an isotope of observable half-life in an element studied means that isotope was not looked for.

(1) Calcium. Particles, which had a rate of 30 counts/min. initially in a 5% fraction of a 10 uah bombardment, were observed to decay with ~ 155 -day half-life in a highly purified calcium fraction and were assumed to be from Ca^{45} .

(2) Chromium. No activity was observed in the sample but short-lived ($T_{1/2} < \text{one week}$) activities would have been missed - Cr^{51} must have a very low yield.

(3) Iron. A 46-day β^- -emitter of ~ 0.36 Mev was observed and identified as Fe^{59} .

(4) Nickel. The decay curve resolved into a 3-hour and a 57-hour component. The 3-hour period was taken to be Ni^{65} . The 57-hour period was shown to be Ni^{66} by separation of its 5-minute Cu^{66} daughter. Ni^{66} is a new isotope first assigned in these studies.

(5) Copper. The main component of the decay curve was a 57-hour 0.5 Mev β^- -emitter assigned to Cu^{67} . Cu^{67} is a new isotope assigned in these studies on the basis of its half-life and lack of an active zinc daughter.

- (6) Zinc. The decay curve showed the growth of a 14-hour daughter and then a 49-hour decay. Separation of Ga^{72} identified the isotope as Zn^{72} (yield calculated from β^- of Ga^{72} daughter.)
- (7) Gallium. The decay curve resolved into a 4.9-hour and a 14-hour component. The 5-hour period was identified as Ga^{73} , and the 14-hour as Ga^{72} .
- (8) Arsenic. Components of 40 hours and 19 days were resolved from the decay curves. The 40-hour β^- had an energy of ~ 0.6 Mev and was identified as As^{77} . The 19-day activity, which was $\sim 50\%$ positrons, was identified as the shielded As^{74} . (Yield calculated from β^+ and β^- emitted.)
- (9) Selenium. The activity observed two hours after the midpoint of the bombardment decayed to background with a 57-minute period and was identified as the 58-minute I.T. Se^{81m} . (Yield calculated from β^- of Se^{81} daughter.) Se^{75} could not be found, even in a long bombardment.
- (10) Bromine. The activity observed three hours after the midpoint of the bombardment resolved into 2.7 and 35-hour periods. The 2.7-hour period was identified as Br^{83} , and the 35-hour as the shielded Br^{82} .
- (11) Rubidium. A 21-day 2.0 Mev β^- , 23-day 1.5 Mev β^+ , 40-day 12 Kev x-ray and 40-day 0.6 Mev γ were observed. The 20-day periods were associated with shielded Rb^{86} . (Yield calculated from β^- and β^+ emitter.) The 40-day periods were associated with shielded Rb^{84} . (Yield calculated from K x-rays emitted.)
- (12) Strontium. The activity observed a month after the bombardment consisted of a 55-day 1.6 Mev β^- which was identified as Sr^{89} . The presence of 30-year Sr^{90} was detected by separation of its 62-hour Y^{90} daughter.

(13) Yttrium. Components of 10-hour, 62-hour, and 57-day periods were resolved from activity separated soon after the bombardment. The 10-hour period was identified as Y^{93} , and the 62-hour as Y^{90} . The yield of 57-day Y^{91} was measured in an yttrium fraction separated three weeks after bombardment. The rare-earth activities present in the yttrium fraction did not interfere because their yields are much smaller.

(14) Zirconium. The 68-day 0.4 Mev β^- observed was identified as Zr^{95} .

(15) Columbium. The activity consisted of a soft β^- and γ decaying with a 35-day half-life which was identified as the lower state of Cb^{95} . (Yield calculated from γ emitted.)

(16) Molybdenum. The activity observed a few hours after bombardment consisted of a 67-hour 1.2 Mev β^- identified as Mo^{99} .

(17) Ruthenium. The decay curve of activity from a short bombardment resolved into a 4.5-hour, a 36-hour, and a long-lived component. The 4.5 and 36-hour periods were identified as Ru^{105} and its Rh^{105} daughter. In a ruthenium fraction from a long bombardment a 42-day 0.2 Mev β^- and a much longer-lived very hard β^- were observed. The 42-day period was identified as Ru^{103} . The long period is probably the 1-year Ru^{106} detected by counting the 4 Mev β^- of its 30-second Rh^{106} daughter.

(18) Palladium. The decay curve resolved into a 13-hour and a 21-hour component. The 13-hour period was identified as Pd^{109} by separation of its 40-second Ag^{109m} daughter. The 21-hour period was identified as Pd^{112} by separation of its 3.2-hour Ag^{112} daughter. (Yield calculated from β^- of Ag^{112} daughter.).

(19) Silver. The decay curve consisted of a 3.5-hour component identified as Ag^{112} , and a 7.5-day 1.0 Mev β^- identified as Ag^{111} . The 225-day isomer of Ag^{110} was also observed.

(20) Cadmium. The decay curve consisted of a 2.3-day and a 43-day component, and a 4.5-hour indium daughter was separated. The activities were identified as the two isomers of Cd^{115} .

(21) Antimony. An antimony fraction from a short bombardment contained a 2.7-day 1.7 Mev β^- which was taken to be Sb^{122} . A 6-day x-ray activity observed in another bombardment was tentatively identified as the 6-day isomer of Sb^{120} . A 60-day β^- was also observed and identified as the lower isomer of Sb^{124} .

(22) Tellurium. Observation of a 6-day β^+ activity suggested the presence of the 6-day K-capturing Te^{118} which was confirmed by separation of its 3-minute β^+ -emitting Sb^{118} daughter. (Yield calculated from β^+ of Sb^{118} daughter.) Te^{119} was detected by separating the 40-hour x-ray activity of its Sb^{119} daughter. (Yield calculated from K x-rays of Sb^{119} daughter.) The 130-day activity observed consisted of 0.2 and 1.2 Mev electrons, 27 Kev x-rays, and 185 and 650 Kev γ . This activity was identified as the 143-day isomer of Te^{121} and its daughters. (Yield calculated from γ emitted.)

(23) Iodine. Positrons, x-rays, and γ 's of 4.5-day half-life were observed and identified as I^{124} . (Yield calculated from K x-rays and β^+ emitted.) A 13-day 1.0 Mev β^- was identified as I^{126} . The third component of the decay curve was a 55-day x-ray emitter identified as I^{125} .

(24) Cesium. The fraction contained a 6-day 0.65 Mev γ , 10.5-day x-ray, 19-day 1.9 Mev β^- , and a small amount of $\sim$ 30-day and $\sim$ 300-day tails. The 6-day activity was identified as Cs^{132} . (Yield calculated

from γ emitted.) The 10.5-day activity was identified as Cs^{131} .

(Yield calculated from K x-rays emitted.) The other activities could not be identified with known cesium isotopes and may have been rubidium impurity.

(25) Barium. A 38-hour 280 Kev e^- activity was identified as the 38-hour I.T. Ba^{133m} . (Yield calculated from 0.3 Mev e^- emitted.)

A 40-hour lanthanum activity was separated from the barium in very small amount, showing the presence of Ba^{140} . The presence of Ba^{131} was shown by growth in the x-ray decay curve and the separation of Cs^{131} daughter. (Yield calculated from K x-rays emitted.)

(26) Cerium. A ~ 30 -day ~ 0.6 Mev β^- activity was identified as Ce^{141} . The other component of the decay curve was a ~ 140 day activity of soft electrons and x-rays which was identified as Ce^{139} . (Yield calculated from K x-rays emitted.)

(27) Europium. An initial activity of 500 particles, 32 γ , and 5 x-ray counts/minute decayed with a 21 day half-life and was tentatively identified as Eu^{149} . (Yield averaged from calculation on e^- , γ , and x-rays.)

Section III

190 Mev Deuteron Fission of Bismuth

The measured yield of a radioactive nuclide produced in a nuclear reaction such as fission may be one of two types usually called cumulative and independent yields. A cumulative yield includes the amount of the nuclide formed by β^- decay of its smaller atomic number isobars if it is neutron-excess, or K-capture and β^+ decay of its larger Z isobars if it is neutron-deficient, in addition to the amount formed directly by the nuclear reaction. Most of the yields measured are of this type. An independent yield value, which is the amount of the nuclide formed in the nuclear

reaction, can be obtained either when a nuclide is bracketed by stable isobars (i.e., shielded) or when its parent activity has an observable half-life and yield and a correction is made for growth of the nuclide in question. Thirteen independent yields, including 8 shielded nuclides, were measured and are marked with an asterisk in Table I.

Initially, all yields were expressed relative to the yield of Mo^{99} .

When considerations of the mechanism of the reaction made it possible to draw a smooth curve for the fission yield as a function of mass number, the area under this curve was set equal to 200% (since binary fission seems to predominate greatly) and the yields expressed in percent of the total fissions. (Mo^{99} at the peak of the curve has a fission yield of 5.0%). Finally, a measurement of the absolute yield of Mo^{99} , in a bombardment with a known deuteron beam gave a value for the fission cross section--

$$\sigma_f = 0.2 \times 10^{-24} \text{ cm}^2, \text{ with 190 Mev deuterons.}$$

In order to present the data most intelligibly, it is necessary to discuss briefly the proposed mechanism for the reaction. When the yields of the fission products are plotted as a function of their mass numbers, a single-peaked symmetrical distribution curve centered at a mass of approximately 100 is indicated. Moreover, cumulative and independent yield values show that the ratio of neutrons to protons (or Z/A) is nearly constant for all nuclides formed in relatively good yield and that this value of Z/A is that for a polonium isotope of mass about 200. Careful analysis of the data led to the conclusion that the reaction could be approximated best by Po^{199} fissioning with unchanged charge distribution, after the evaporation of about a dozen neutrons from the initial highly excited nucleus. On this basis it is possible to predict which element (or elements) is the most probable primary fission product (i.e., initial fragment) for each mass number, as indicated in Table I. The designation $A \beta^- \text{ (or } K) B$

in this table means that the first member is wholly or partly predicted, but its yield is measured as the daughter activity. In Fig. 1, the yields of all nuclides and the sums of all isobars measured were plotted against their mass number. The points are designated as "on" if they include the yield of the most probable primary fission product, "near" if they include the yield of one of two predicted equally, and "off" if they do not include any probable primary fragment. The smooth curve was then drawn on the basis of the weighted data, and values read off this curve were used in the "predicted fission yield" column of Table I.

A number of examples may be cited to illustrate the agreement of data with theory. Zinc and gallium are predicted for mass 72, and the sum of the cumulative yield of Zn^{72} plus the independent yield of Ga^{72} agrees with the yield predicted for that mass. These yields are shown as the half-shaded circles in Fig. 1 with the sum as the solid circle. The independent yield of Br^{82} , a shielded isotope, is one-third the total yield of that mass, for which the prediction is selenium and bromine. Se^{82} is stable and therefore not observed. The cumulative yield of Sr^{90} and the independent yield of Y^{90} add to give a point on the curve, as do the yields of Zr^{95} and Cb^{95} . The approximately equal yields of the 2.3 day and 43 day isomers of Cd^{115} , like the ratio resulting from the (n,p) reaction, in contrast to the fourteen-to-one ratio of yields in slow neutron fission of uranium where they are produced by decay of Ag^{115} , indicate predominant formation of this nuclide as a primary fragment⁸. The yield of mass number 115 is predicted to be shared between cadmium and stable indium, and one-half is observed as cadmium. For mass number 124, the predicted elements are tellurium and iodine; and independent yields of the shielded nuclides Sb^{124} and I^{124} bracketing stable Te^{124} are only a fraction of the total predicted yield. It is of interest to note that the Ba^{140} yield is less

than one percent of the total yield of that mass, for which Pr^{140} is the predicted nuclide, and that neutron-deficient $\text{Ba}^{133\text{m}}$ is the major activity in the barium fraction as predicted.

A few points predicted to be on the curve are low. One of the largest discrepancies occurs in the case of I^{126} --the good yield of I^{125} makes incomplete exchange of tracer and carrier an unlikely explanation, but Friedlander has suggested that an undetected K-capture branching may be responsible⁹. The low independent yield of Cs^{131} is as yet unexplained, but the possible experimental error is large in this case.

Table I

Fission Yields in 190 Mev Deuteron Fission of Bismuth

Mass Number	Predicted Primary FP	FP Meas- ured	Measured Yield (%)	Total Measured Yield for Mass Number(%)	Predicted Fission Yield(%)
45	K $\xrightarrow{\beta^-}$ Ca	Ca	0.002	0.002	0.0002
59	Mn $\xrightarrow{\beta^-}$ Fe	Fe	.05	.05	.04
65	Co $\xrightarrow{\beta^-}$ Ni	Ni	.08	.08	.23
66	Ni	Ni	.11	.11	.32
67	Ni $\xrightarrow{\beta^-}$ Cu	Cu	.40	.40	.40
72	Zn	Zn	.35	.86	.93
	Ga	Ga	.51*		
73	Ga	Ga	.55	.55	1.1
74	Ga	As	.06*	.06	1.2
77	Ge $\xrightarrow{\beta^-}$ As	As	1.5	1.5	1.9
81	Se			.41	2.8
	Se ^m	Se ^m	.41		
82	Br	Br	1.0*	1.0	3.0
	Se(stable)	-			
83	Br	Br	1.7	1.7	3.2
84	Br			1.3	3.4
	Kr(stable)	-			
		Rb	1.3*		
86	Kr(stable)	-		1.9	3.8
	Rb	Rb	1.9*		
89	Rb $\xrightarrow{\beta^-}$ Sr	Sr	4.7	4.7	4.4
90	Sr	Sr	2.9	4.5	4.5
		Y	1.6*		
91	Sr $\xrightarrow{\beta^-}$ Y	Y	4.4	4.4	4.6
93	Y	Y	2.8	2.8	4.8
	Zr				
95	Zr	Zr	3.4	4.9	4.9
		Cb	1.5*		
99	Mo	Mo	5.0	5.0	5.0
103	Tc $\xrightarrow{\beta^-}$ Ru	Ru	3.9	3.9	4.95
105	Ru	Ru	3.1	3.1	4.9
	Rh				

*Independent yields

Table I (Cont'd)

Mass Number	Predicted Primary FP	FP Measured	Measured Yield (%)	Total Measured Yield for Mass Number (%)	Predicted Fission Yield(%)
106	Rh Ru	Ru	1.5	1.5	4.8
109	Pd	Pd	4.6	4.6	4.6
111	Ag	Ag	3.4	3.4	4.2
112	Ag Cd(stable)	Ag - Pd	2.8* .9	3.7	4.1
115	In(stable) Cd(2.3 d.) Cd(43 d.)	- Cd(2.3 d.) Cd(43 d.)	 1.0 .7	1.7	3.6
118	Sn(stable)	- Te	 .008	.008	3.0
119	Sn(stable)	- Te	 .14	.14	2.7
120	Sb(6 d.) Sb(17 m.) Sn (stable)	Sb (6 d.) -	.90 	.9	2.5
121	Sb(stable)	- Te (143 d.)	 .20	.2	2.3
122	Te (stable) Sb	- Sb	 .25*	.25	2.0
124	Te (stable), I	- I Sb (60 d.)	 .43* .12	.55	1.6
125	I	I	1.2	1.2	1.4
126	I	I	.11*	.11	1.2
131	Cs Ba	Cs Ba	.002* .18	.18	.54
132	Ba (stable) Cs	- Cs	 .056*	.06	.45
133	Ba	Ba	.25	.25	.35
139	Pr $\xrightarrow{K}$ Ce	Ce	.12	.12	.07
140	Pr	Ba	.0004	.0004	.05
141	Pr (stable) Nd	- Ce	 .017	.017	.03
149	Eu	Eu	.003	.003	.0015

*Independent yields

Fig. 1. Plot of fission yield as a function of mass number for products
of the irradiation of bismuth with 190 Mev deuterons.

(To be used in conjunction with Table I to locate particular points.)

FISSION OF BISMUTH WITH 190 MEV DEUTERONS

Section IV

Fission at Other Energies

A survey was made of the change in the distribution of mass versus yield of the fission products with variation of the energy of bombardment. The yield curve was studied by measuring the yields of neutron-excess Cu^{67} and Mo^{99} and neutron-deficient $\text{Ba}^{133\text{m}}$, the first two of which lie on and the last very near the 190 Mev deuteron curve. In Table II, the yields of Cu^{67} and $\text{Ba}^{133\text{m}}$ are given relative to the yield of Mo^{99} taken as one in each experiment, since Mo^{99} lies at the peak of the 190 Mev deuteron curve.

Table II
Relative Yields of Cu^{67} , Mo^{99} , and $\text{Ba}^{133\text{m}}$
in Irradiations at Several Energies

Energy (Mev)	Projectile Target		Relative Yields		
			Cu^{67}	Mo^{99}	$\text{Ba}^{133\text{m}}$
380	α	Bi	0.12	1	0.050
190	d	Bi	0.080	1	0.050
100	d	Bi	0.038	1	0.012
100	d	Pb	0.046	1	0.025
75	d	Pb	0.016	1	0.007
50	d	Pb	0.004	1	0.008

It appears that any shifts of the yield versus mass curve are less important than a tendency for the half-width to decrease as the projectile energy decreases; i.e., the probability for symmetrical fission increases as the excitation energy decreases. This is shown graphically in Fig. 2. As Fig. 2 also shows, the fission cross section decreases as the excitation energy decreases as the excitation energy decreases and is approximately three times as large with 380 Mev α 's and one-half as large with 100 Mev d's as with 190 Mev d's. The excitation function of the fission reaction is

discussed more extensively in Section V.

The ratio of the yield of Br^{83} to the yield of shielded Br^{82} was measured in a series of irradiations of bismuth with deuterons of 50 to 190 Mev energy. The yield of Br^{83} is cumulative and a good measure of the total yield of mass numbers in this region.

Table III

$\text{Br}^{83}/\text{Br}^{82}$ Yield Ratio in Bismuth Fission

as a Function of Deuteron Energy

Energy (Mev)	$\text{Br}^{83}/\text{Br}^{82}$ Yield Ratio
190	1.7
150	3.0
90	7.8
70	20.
50	$\geq 100.$

It appears that the mechanism of the reaction changes in such a way as the excitation energy decreases that the probability of formation of Br^{82} as a primary product decreases very rapidly.

Fig. 2. Plot of the absolute yields of Cu^{67} , Mo^{99} , and $\text{Ba}^{133\text{m}}$ produced by irradiation of bismuth with 100 Mev and 190 Mev deuterons and 380-Mev alphas.

BISMUTH FISSION YIELDS

Section V

Isotopic Lead Fission

If the prior emission of neutrons to attain a nucleus like Po^{200} , whose Z^2/A value is comparable to that of U^{236} , is necessary before fission can compete significantly with other reactions in bismuth region elements, then a difference in the fission cross section as a function of bombardment energy might be expected between Pb^{204} and Pb^{208} . Since the pure isotopes were not available, two lead samples enriched, respectively, in these isotopes were bombarded together at several deuteron energies. The yield of fission was measured by obtaining the Mo^{99} yield at all energies and the Cu^{67} and $\text{Ba}^{133\text{m}}$ yields also in the 50, 75, and 100 Mev deuteron bombardments.

The technique was as follows: 10 mg of Cu, Mo, and Ba were added to the 6N HNO_3 solution of the target, Mo α -benzoin oxime precipitated from 3N HNO_3 solution, PbO_2 deposited electrolytically from 6N HNO_3 after the alcohol was boiled off, the supernatant fumed down with a few ml of 36N H_2SO_4 and diluted to precipitate BaSO_4 , and CuCNS precipitated from the supernatant. The Mo, Ba, and Cu fractions were then purified by the usual procedures. The PbO_2 was dissolved in HNO_3 and H_2O_2 , and PbCO_3 precipitated, dried, placed in a pure carbon boat, and reduced to metal by heating in a H_2 stream inside a quartz tube. (For the low energy bombardment the lead was purified from Mo carrier by treating a 6N HCl solution of it with ether.) The lead pellets were rolled out into foils a few mils thick for re-bombardment.

The variations in the vertical location and spread of the deuteron beam are so great when intercepted at the lower energies (i.e., smaller radius) that any absolute yield calculations are very inaccurate, especially in this case since the lead foils were small in area. Furthermore, since the

fission yield versus mass curve changes shape as the energy changes, as discussed in Section IV, so that Mo^{99} does not have a 5.0% fission yield, the fission cross section cannot readily be obtained from the Mo^{99} yield measurements. However, the relative yields of Cu^{67} , Mo^{99} , and Ba^{133m} in two isotopic lead samples showed the curve was similar in both at the same deuteron energy, and therefore the ratio of the Mo^{99} yields in both samples measures the relative amounts of fission produced at each energy. In Fig. 3, the data are summarized along with the isotopic composition of the lead samples. The fission cross section is smaller and decreases more rapidly as the deuteron energy decreases in PbB, which has a smaller proportion of the light isotopes than PbA, as might be expected on the basis of the reactions represented by:

It must be remembered that the cross sections as shown are for the lead of composition shown. If given in terms of isotopic cross sections of Pb^{204} and Pb^{208} respectively, the differences would be even greater than those shown.

Fig. 3. Plot of the absolute yields of Mo^{99} in the fission of two lead samples of different isotopic composition as a function of deuteron energy.

YIELD OF Mo^{99} IN FISSION OF ISOTOPIC LEAD

MODEL

DATE

Section VI

Fission of Elements from Tantalum to Bismuth

One of the first experiments performed in the study of high energy fission was to make a survey of fissionability as a function of atomic number with 380 Mev alphas. Bismuth, lead, thallium, platinum, and tantalum foils were bombarded, and several neutron-excess nuclides observed in each case. The relative yields of fission product activity are given in Fig. 4, in which the dashed curve has no theoretical significance. In the case of fission with 90 Mev neutrons, the yield seems to decrease less rapidly as Z decreases (e.g., the yield in Pt is five times as great relative to the bismuth yield as with 380 Mev α 's)³. If the mechanism of prior neutron evaporation to attain a critical Z^2/A is correct for tantalum also, we would predict that its fission products would be predominantly neutron-deficient. Therefore the nuclides studied in this experiment would be a poor measure of the amount of fission occurring, which might be considerably greater relative to bismuth than indicated in Fig. 4.

Fig. 4. Plot of relative yields of fission product activity, produced by irradiation with 380 Mev alphas, as a function of atomic number of the target.

FISSION WITH 400 MEV He^{+2}

Section VII

Theory of Fission Process

The very extensive investigation of the fission of the heavies elements (i.e., Th, U, etc.) is used as a basis in this discussion. The fission which can be induced in these elements with a total excitation energy of 6-8 Mev has a number of special characteristics^{10,11}. The most probable division is highly asymmetric, with a ratio of masses of the two fragments of ~ 1.5 , despite the fact that experiment and theory show that the maximum kinetic energy, ~ 180 Mev, is produced in a nearly symmetric splitting^{12,13,14}. A fission into two given fragments varies in kinetic energy release over a 30 Mev range¹². There appears to be a tendency for the fission to become somewhat more symmetric as the atomic number increases¹⁵. The primary fission products have 3-5 excess neutrons on the average and β^- decay to stability. Since the n/p ratio of the stable nuclides increases with increasing Z, this means that there has been a redistribution of charge in the fissioning nucleus so that neither fragment has the n/p ratio of the parent nucleus. The total energy released in decay is ~ 30 Mev. Fission competes very well with other reactions, for example, even in the case of irradiation of thorium with 40 Mev alphas for which σ_f is approximately one-fourth of the geometric cross-section²³. The fission yield curve for U^{235} irradiated with slow neutrons is shown in Fig. 5 along with the curve of the 190 Mev deuteron fission of Bi^{209} .

Nuclides in the bismuth region are still energetically unstable with respect to fission--e.g., lead fission releases 120 Mev kinetic energy--but a critical deformation is necessary before fission proceeds spontaneously¹⁴. The liquid-drop model of the nucleus leads to the parameter Z^2/A as the criterion for the threshold excitation energy associated with this critical deformation. For $Z = 84$, the fission threshold is calculated to be ~ 12 Mev for $A = 210$, and ~ 6 Mev for $A = 200$ (i.e., the latter comparable to the thresholds in the uranium region)¹⁶. The liquid drop model does not

lead, as yet, to the prediction of favored asymmetry in low excitation fission of the heavy elements. J. Frenkel has postulated a quantum mechanical tunnel effect to account for the observed asymmetry¹⁷, but J. A. Wheeler does not believe his treatment is valid¹⁸.

Four interrelated aspects of fission need to be examined in detail in developing a mechanism for high energy fission: the excitation function, mass distribution, charge distribution, and sequence of fission and neutron evaporation. The problem of relating these aspects to the excitation energy of the nucleus is complicated by the fact that the transparency of nuclear matter to high energy projectile nucleons means that a wide range of excitation energies is produced by a projectile of given energy¹⁹. In the case of bismuth irradiated with deuterons, compound nucleus formation is probably a very good approximation at least up to 50 Mev. Even at 190 Mev, the probability for complete transparency is negligible so that the inelastic collision cross section equals the geometric cross section.

It has been calculated that the cross section with 190 Mev deuterons for excitations of less than 100 Mev is one-fourth and the cross section for excitations of 100 to 200 Mev is three-fourths of the geometric cross section, with a broad maximum probability for excitations of ~ 150 Mev²⁰. The transparency effect decreases as the deuteron energy decreases in the intermediate region, and therefore identification of the excitation energy with the deuteron energy becomes a better approximation.

When transparency is taken into account, the data on neutron, deuteron, and alpha fission of bismuth indicate that the fission cross section is very small below 50 Mev excitation energy and rises approximately linearly from 50 to 350 Mev. Thus we are observing a relatively pure high energy reaction in the fission of bismuth with 190 Mev deuterons. In contrast, the Np^{237} fission cross section becomes quite large within one Mev of the threshold and remains nearly constant for higher energies.²⁸ Probably the U^{238} fission cross section, which equals approximately 2 barns with 380 Mev alphas, behaves similarly

~~threshold and remains nearly constant up to 350 Mev,~~ and therefore the products of 380 Mev alpha bombardment of uranium are the average of many effects²¹.

The fission of bismuth appears to be predominantly symmetrical at all excitation energies where the yield is appreciable. In the case of the heavy elements, an excitation three to four Mev above the threshold makes symmetrical division about one-tenth as probable as asymmetric²², while a 40 Mev excitation increases the ratio to one-half²³. In the latter case, if any neutron evaporation precedes the fission, the residual excitation of the fissioning nucleus may be considerably less than 40 Mev, of course. The intermediate region has not been studied, but the combined effects in a 380 Mev alpha bombardment of uranium give a single peak for the mass distribution curve²¹.

C. D. Coryell has recently proposed a method of organizing the independent fission yield data to give a more quantitative test for the charge distribution mechanism²⁴. We need to assume that the nuclear charge on a fragment of any particular mass number resulting from a given fission mechanism can be expressed as a probability distribution curve symmetrical about the most probable charge for that mass, Z_p , chosen on the basis of the mechanism--i.e., a plot of the ratio of the yield of a particular isobar to the total yield of that mass versus the difference in charge units between the charge on that isobar and Z_p for that mass, $Z - Z_p$, should be a smooth symmetrical peak with a maximum at $Z - Z_p$ equal to zero. When the data from slow neutron fission of U^{235} and Pu^{239} are so treated, a smooth curve is obtained only when Z_p is chosen by the postulate of equal chain lengths--i.e., that $Z_p - Z_A$, the Bohr-Wheeler maximum stability charge, for each of the two fragments resulting from any fission is the same. The equal chain length postulate does require a redistribution of charge in

the fissioning nucleus and releases close to the maximum energy for a fission into any two given fragments¹⁴. When the same system of organization is applied to the 190 Mev bismuth fission data, the results are shown in Fig. 6 where Z_p is chosen on the basis of Po^{199} splitting with unchanged charge distribution (i.e., $Z_p = 84/199$ A). The charge distribution curve from U^{235} fission is shown for comparison although one would not necessarily expect that the shape of this curve would be the same for all fission mechanisms. The bismuth data are not accurate enough to define any curve but, with the exception of three low points which may be bad experimentally, support our hypothesis of the mechanism. If we assume for the moment that Po^{199} has been substantiated as representing the fissioning nuclei, application of the equal chain length postulate to it predicts predominantly stable products and is contradicted by the observation of division into a neutron-excess and a neutron-deficient fragment--e.g., $84Po^{199} \longrightarrow 56Ba^{133} + 28Ni^{66}$.

If we consider the case of bismuth fission following very high excitation resulting from 190 Mev deuteron irradiation, the problem of dissipation of this energy arises. The kinetic energies of the fission fragments are within a few Mev of those predicted on the basis of fission with near threshold excitation. The mass distribution curve indicates the sum of the two final fragments is ~ 200 mass units. Moreover, if the compound nucleus Po^{211} were fissioning, application of either the unchanged charge distribution or the equal chain lengths postulate to it lead to predictions of entirely neutron-excess fission products in disagreement with the data. If fission preceded the dissipation of much of this excitation energy, the fragments would have this excitation in addition to that normally resulting from the distortion of division and might be expected to behave like a corresponding compound nucleus studied in spallation research. Because of a Coulombic barrier effect rising rapidly with Z , neutron or charged particle

evaporation are of comparable probability for nuclides in the copper region²⁵ but predominant neutron evaporation is already favored in the antimony region²⁶. Therefore, qualitatively one might expect the same kind of relation between beta instability type and mass number of the fission products as observed. The model of prior neutron evaporation (highly favored by high Z) followed by fission with unchanged charge distribution leads to such good agreement between predicted and observed yields that we believe it is correct, however.

We may now try to explain the results observed in irradiation of bismuth with 190 Mev deuterons. The yields observed are a combination of the reactions induced by the range of excitations produced by this projectile. The $\text{Br}^{83}/\text{Br}^{82}$ ratios are sufficient to show that the nuclear reaction mechanism is considerably different for low excitations, but the small probability for such excitations and the small probability that fission will occur in these cases makes the contribution to the observed yields very small. With high excitations, the first step is the evaporation of a number of neutrons which occurs in a time of the order of 10^{-20} seconds. The binding energy of each successive neutron evaporated increases in an unknown manner, although its kinetic energy probably remains of the order of one Mev, and therefore possibly the half-life for evaporation increases somewhat.

Studies of the (α, xn) reaction, which essentially requires compound nucleus formation and therefore a knowledge of the actual excitation energy, indicate that the evaporation of the order of 10 neutrons has a maximum probability at about 150 Mev excitation²⁷. As neutron evaporation proceeds and the residual excitation energy decreases, Z^2/A increases and the fission threshold decreases. With Po^{211} , an excitation energy a few Mev above the fission threshold is also probably sufficient to evaporate up to two neutrons and fission cannot be detected in competition with the other nuclear reac-

tions possible. If 12 neutrons were first evaporated from Po^{211} to reach Po^{199} , however, the fission threshold is probably considerably smaller than the energy necessary to evaporate another neutron. The symmetry and unchanged charge distribution, which is energetically wasteful, suggest that fission occurs here at a higher rate than in slow neutron fission. It is still questionable whether neutron evaporation continues until it is energetically impossible and then fission competes with gamma emission, or whether fission actually competes significantly with neutron evaporation while the residual excitation is still sufficient for the latter to occur. The increase in fission cross section in the 380 Mev alpha bombardment indicates the fission probability is the same or increases for still higher excitation energies. Of course, the evaporation of charged particles decreases very rapidly the probability of fission ever competing with other reactions. It is felt that the preceding analysis gives a reasonable explanation for the observed fission cross section in 190 Mev deuteron bombardment of bismuth of one-tenth of the geometric cross section.

It is tempting to try to generalize the proposed mechanism for high energy fission. The data on 380 Mev alpha bombardment of uranium suggest but do not prove that prior neutron evaporation has taken place in this case also, but nothing has been deduced about the charge distribution mechanism. At the other extreme, this mechanism predicts that the products of 380 Mev alpha fission of Ta^{181} will be predominantly neutron-deficient. A study of the fission of bismuth with 380 Mev alphas should indicate whether fission competes well with further neutron evaporation when 10 to 12 neutrons have been evaporated but a high excitation energy still remains.

It is probable that further refinement of the theory outlined here can be made. It is felt, however, that refinement of the experimental data should precede such speculation.

Fig. 5. Comparison of the fission yield curves for products of the irradiation of U^{235} with slow neutrons and bismuth with 190 Mev deuterons.

BISMUTH AND URANIUM FISSION

Fig. 6. Plot of the ratio of the yield of a nuclide to the total yield of that mass number versus the difference between the nuclear charge of the nuclide and the most probable charge for that mass number for products of 190 Mev deuteron fission of bismuth. The charge distribution curve for slow neutron fission of U^{235} is also shown.

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