High Energy Induced Fission of Bismuth and Lead

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A study has been made of the characteristics of the fission induced in bismuth and lead by irradiation with the high energy deuterons 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 fission yield 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. 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 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.

INTRODUCTION

IN two previous communications, the observation of fission of elements in the bismuth region of the periodic table with high energy particles was reported;[1,2] and the characteristics and a theory of the reaction were briefly described.[2] In this paper is presented a more detailed account of the observations which suggested this picture of high energy induced fission. In particular the results of the irradiation of bismuth with 190-Mev deuterons are summarized. The irradiations of bismuth and lead at several energies are included to complete the description.

The distribution of fission products in the fission of bismuth with 190-Mev deuterons was 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, Cb, Mo, Ru, Pd, Ag, Cd, Sb, Te, I, Cs, Ba, Ce, and Eu. The yields of 48 radioactive nuclides from mass numbers 45 to 149 were measured.

PROCEDURE

Bismuth of high purity was cut into 1-mm thick strips which were mounted on the probe to intercept the circulating beam of the 184-inch frequency-modulated cyclotron. Most of the irradiations were for one to three hours with deuteron currents of approximately one microampere. For the measurement of long-lived activities, a similar target was irradiated in this manner intermittently over a period of several weeks. In general, a 10- or 20-mg portion of carrier of the element being investigated was added to an aliquot of the nitric acid solution of the target, the element was separated and purified from all others, and the chemical yield was determined by weighing a compound of the recovered carrier. The chemistry used was based on fission product procedure reports of the Plutonium Project[3] with modifications in many cases because of the presence in large quantities of spallation products covering the region from bismuth to the rare earths. The final precipitates were filtered onto 1-cm^2 area papers, mounted on cardboard, and covered with ~3-mg cellophane for measuring the decay characteristics on ~3-mg mica end-window Geiger-Müller tubes. Wherever sufficient activity was encountered, identification of the radiation by means of absorption methods or with a simple beta-ray spectrometer was carried out in addition. Also, where applicable, identification was made more certain by observation of parent-daughter relationships. The chemistry and isotope identification will be summarized in the appendix.

RESULTS

One of the graphical methods for describing some of the features of the fission process is to plot against the mass number of a fission fragment the incidence with which that mass number appears. This *fission yield curve* of U^{235} with slow neutrons as determined by the exhaustive measurements made by members of the Plutonium Project[4] is reproduced as the familiar double-hump curve in Fig. 1. It should be recalled that the primary fission products in this system have considerable neutron excess and beta-decay toward stability with a general increase in half-life in the members of the decay sequence. For any mass number there is a distribution of *primary* fission products with respect to element but those nearest stability appear in low yield and by inference the probability of producing a stable nuclide as a primary fission product is low. As a consequence the yield of beta-emitting nuclides one or two removed from stability represents, with fair accuracy, the total yields for their respective mass numbers if sufficient time is elapsed to permit accumulation of yield through decay of the shorter lived antecedents. The fission yield curve for uranium with slow neutrons is therefore accurately defined by such measurements.

[1] Perlman, Goeckermann, Templeton, and Howland, Phys. Rev. 72, 352 (1947).
[2] R. H. Goeckermann and I. Perlman, Phys. Rev. 73, 1127 (1948).
[3] National Nuclear Energy Series—Plutonium Project Record Vol. 9B, Chapter 8 (to be issued).
[4] See "Nuclei Formed in Fission," J. Am. Chem. Soc. 68, 2411 (1946).

In Fig. 1 is also shown a curve to represent the fission yield *vs.* mass number dependence for the fission products from bismuth induced by 190-Mev deuterons. In contrast to the curve for uranium with slow neutrons the highest yields appear in the symmetrical cleavage. Without belaboring this point further for the present another important feature should be discussed, namely, that although there is good evidence to believe that this curve represents in essence the mass number distribution, not many of the measured radioactive fission products actually fall on the curve. It will be necessary to anticipate some of the points which will be brought out in the later discussion in order to explain how this fission yield curve was constructed from the measured fission products.

One striking difference between bismuth fission products and those from the slow neutron fission of uranium is that in the case of bismuth the fission products do not necessarily have a high excess of neutrons; in fact, they may lie on the neutron deficient side of stability. Besides the general propensity of the primary fission products to lie close to stability, it was noted that the lighter fission fragments are β^--emitters while those on the high mass side of the distribution are neutron deficient and decay by β^+-emission or electron-capture. In accounting for these and other observations it was found possible to set up conditions which define the most probable primary fission products for each mass number. It is first of all assumed that about 10 neutrons are boiled out of the nucleus before it undergoes fission. While it is recognized that the fission of no single nuclear species describes all of the events, for the sake of simplicity it is assumed that an excited state of Po^{199} is the immediate nucleus which undergoes fission. This would arise from the reaction: $Bi^{209}(d,12n)Po^{199*}$. It is further assumed that the Po^{199} undergoes cleavage without charge redistribution so that each fragment retains the same neutron/proton ratio as the parent nucleus.

One may now attempt to construct a fission product distribution curve using the measured yields of the fission products. One has but to select the various mass numbers and calculate from the neutron/proton ratio for Po^{199} what will be the most probable charge which will appear with that mass number. This has been done in Table I in which the second column lists the most probable primary fission products. In a number of cases the predicted value for Z falls well between two integers and so two elements are listed. It will be noted that up to mass number $\sim$110 the predicted primary fission products are predominantly β^--emitters, in the mass number range 110–125 they are to a large extent stable, while above 125 most are on the neutron deficient side of stability. Turning to Fig. 2 in which the points may be identified from the mass numbers and yields as listed in Table I, the low mass arm of the curve is constructed by drawing a smooth curve through points of maximum yield. When this is done it is found that the points which fall closest to the curve are indeed those which the theory would predict to be the most probable fission products for each mass number. These are shown as solid circles. The high mass arm of the curve may be obtained by reflecting the low mass arm through mass number 99 as the line of symmetry. As predicted from the theory there will be relatively few measurable isotopes which should fall on the curve since most of the primary fission products in this range are stable. In a few cases one would predict a radioactive fission product to be the most probable and here it was found that these particular species did fall on the curve with one apparent exception which will be discussed below. The open circles in Fig. 2 represent yields of fission products predicted to arise in rare modes of fission since they do not fulfill the conditions set for the most probable events. The half-closed circles are points for which the theory predicts sharing of yield in that the most probable value of Z lies between integers.

Considering the manifest oversimplification in this model for the fission process the agreement between measured yields and predictions is considered good. Individual points will be discussed below which will serve to indicate that agreement may be better than indicated by a cursory examination of Fig. 2. Furthermore there is a considerable amount of other evidence

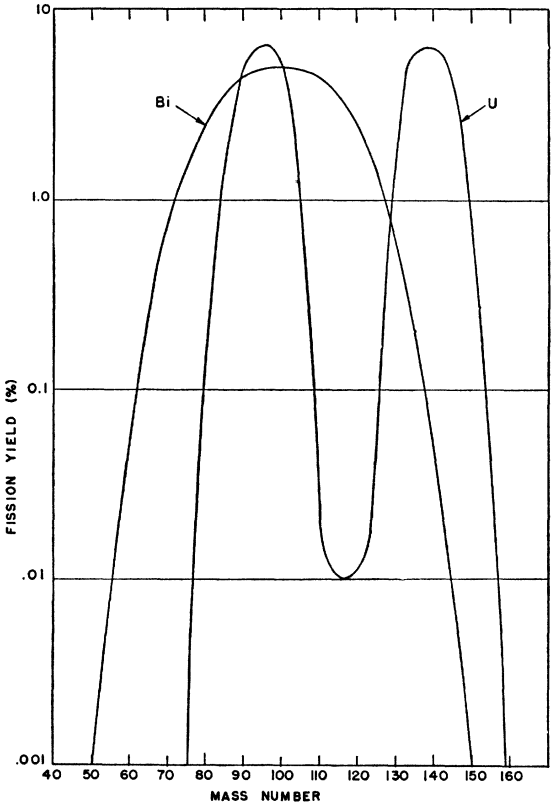

FIG. 1. Fission yield for U^{235} with slow neutrons and bismuth with 190-Mev deuterons.

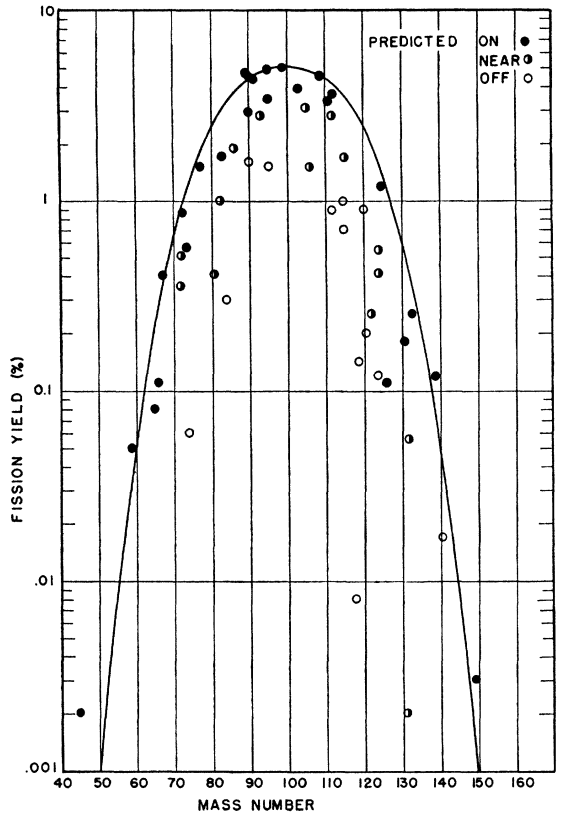

FIG. 2. Yields for products of the irradiation of bismuth with 190-Mev deuterons.

to support the general picture both in terms of the shape and position of the curve and the ability to predict primary fission products.

Fission Yield Data—190-Mev Deuterons on Bismuth

In the second column of Table I is listed the predicted primary fission product for each mass number for which one or more activities have been measured. The designation $A \xrightarrow{\beta^-} B$ means that element A is the predicted primary product but its yield was determined by measuring the daughter activity B much in the same sense that the primary fission products in the slow neutron fission of uranium are not usually measured. For a number of mass numbers more than one measurable nuclide exist and the yields are followed by an asterisk in column 4 for those cases in which the yields were independently determined. Column 5 presents a summation of yields of measured activities and these values should agree with those read off the smooth curve (column 6) only in those instances in which the predicted fission products are those which could be measured. For example, at mass number 72 the predicted primary charge for the fragment lies between 30 and 31 so that one would predict that the 49-hr Zn^{72} and 14-hr Ga^{72} would share the bulk of the yield for this mass number. The yields of the two activities were independently measured as 0.35 and 0.51 percent respectively, the sum of which agrees within experimental error with the value 0.93 percent for mass number 72 as read off the smooth curve. On the other hand for mass number 74 the predicted fission product Ga^{74} was not measured and is an unknown, presumably short-lived, activity. It decays to stable Ge^{74} which blocks the possibility of detecting it through a daughter activity. For mass number 74, the 17.5-day As^{74} activity was measured and since this is two units of atomic number removed from the theoretical primary product it should appear in low yield. As expected its yield is very low, accounting for only 5 percent of the yield taken off the curve for mass number 74. Not all of the data agree so well with the predictions as the examples just cited. However, before ascribing these descrepancies to shortcomings of the theory, one should rule out several factors which produce apparent low yields. The difficulties of absolute measurement of β-emitters and especially of isotopes which decay by electron-capture are well recognized. Of those encountered in these studies, a number do not have well worked out decay schemes and the possible errors lie principally in the direction of indicating low yields. Continued study of radioactive species over the past several years has turned up more and more cases of nuclear isomerism. As a result it is not always possible to ascribe the entire yield of an isotope to the measured species since an undetected isomer may contribute. A well known case of isomerism, namely Cd^{115}, was measured and it was found that the two isomers are formed in comparable yields. Arnold and Sugarman[5] have uncovered some cases of isomerism in uranium fission products which explain previous yield descrepancies among members of a decay chain. Another phenomenon which can give an apparently low fission yield is that of delayed neutron emission. In the case of bismuth fission it would be predicted that such species would lie on the light wing of the distribution where nuclides with considerable neutron excess are expected.

The following gives a brief discussion of the yield data according to mass number. The yields of the lightest and heaviest nuclides measured, Ca^{45} and Eu^{149}, were so low that the position of the yield curve as determined by these values has little significance. References to almost all of the activities mentioned will be found in a compilation of nuclear data.[6]

Mass 59.—The predicted primary fission product for this mass number is manganese. Mn^{59} is not known and is probably a short-lived β^--emitter whose yield can be determined through its decay product 46-day Fe^{59}. None of the other chemical fractions would be expected to contain a measurable activity of this mass number.

[5] J. R. Arnold and N. Sugarman, J. Chem. Phys. **15**, 703 (1947).
[6] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).

Mass 65.—No activities corresponding to the mass number range 60–64 were measured. Significantly no appreciable yields of Cu^{64} were found which is in agreement with the prediction that Co^{64} is the primary fission product for this mass number.

The predicted primary fission product for mass number 65 is divided between Co^{65} and Ni^{65}. It would be expected that Co^{65} is sufficiently short-lived to allow the accumulated yield to be measured as Ni^{65}. The next neighboring isobar is stable Cu^{65}. The measured yield for Ni^{65} is about a factor of three below the curve.

Mass 66.—For this mass number nickel would be expected and a new 56-hr. period was found in the nickel fraction and identified as the parent of the 5-min. Cu^{66}. The radiation from Ni^{66} was not itself measured but the yield was determined from the hard β^--particles of Cu^{66}. The yield so determined falls below the curve and the discrepancy is thus far not explained.

Mass 67.—This is another case in which the predicted primary fission product is a previously unknown isotope, Cu^{67}. The copper fraction contained a 56-hr. period with a 0.5-Mev β^--particle which does not have an active zinc daughter. The assignment to Cu^{67} is reasonable in view of its radioactive properties and is in keeping with its preparation from zinc by the γ-p reaction.[7]

Mass 72.—The sharing of this mass number between Zn^{72} and Ga^{72} as predicted has already been mentioned.

Mass 73.—The measured yield of Ga^{73}, which should be the major primary fission product of this mass number, is about a factor of two below the curve.

Mass 74.—The predicted charge for this mass number lies near gallium, element 31. The only activity of mass number 74 measured was As^{74} and as expected the yield fell well below the curve indicating its appearance in only about 5 percent of the events producing this mass number.

Mass 77 (and 76).—The measurement of 40-hr. As^{77} was made at a time in which the 12-hr. Ge^{77} would have decayed into it so the yield as determined is cumulative. The predicted primary product lies between germanium and arsenic and the cumulative yield of As^{77} lies close to the curve. It was possible to resolve a small amount

TABLE I. Fission yields in 190 Mev deuteron fission of bismuth.

Mass number	Predicted primary product	Product measured	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			.06	1.2
		As	.06		
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			0.3	3.4
	Kr(stable)	—			
		Rb	0.3		
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				
106	Rh			1.5	4.8
	Ru	Ru	1.5		
109	Pd	Pd	4.6	4.6	4.6
111	Ag	Ag	3.4	3.4	4.2
112	Ag	Ag	2.8*	3.7	4.1

TABLE I.—(*Continued*).

Mass number	Predicted primary product	Product measured	Measured yield (%)	Total measured yield for mass number (%)	Predicted fission yield (%)
	Cd(stable)	—			
		Pd	.9		
115	In(stable)	—		1.7	3.6
	Cd(2.3 *d.*)	Cd(2.3 *d.*)	1.0		
	Cd(43 *d.*)	Cd(43 *d.*)	.7		
118	Sn(stable)	—		.008	3.0
		Te	.008		
119	Sn(stable)	—		.14	2.7
		Te	.14		
120	Sb(6 *d.*)	Sb(6 *d.*)	.90	.9	2.5
	Sb(17 *m.*)				
	Sn(stable)	—			
121	Sb(stable)	—		.2	2.3
		Te(143 *d.*)	.20		
122	Te(stable)	—		.25	2.0
	Sb	Sb	.25*		
124	Te(stable)	—		.55	1.6
	I	I	.43*		
		Sb(60 *d.*)	.12		
125	I	I	1.2	1.2	1.4
126	I	I	.11*	.11	1.2
131	Cs	Cs	.002*	.18	.54
	Ba	Ba	.18		
132	Ba(stable)	—		.06	.45
	Cs	Cs	.056*		
133	Ba(39 *h.*)	Ba	.25	.25	.35
	$K \xrightarrow{} Ce$				
139	Pr	Ce	.12	.12	.07
140	Pr			.0004	.05
		Ba	.0004		
141	Pr(stable)	—		.017	.03
	Nd				
		Ce	.017		
149	Eu	Eu	.003	.003	.0015

* Independent yields.

[7] R. B. Duffield and J. D. Knight, private communication.

of shorter-lived activity, probably 27-hr. As^{76}. The yield was about ten-fold lower than As^{77} which is to be expected since stable Ge^{76} is the calculated fission product for this mass number.

Mass 81.—The predicted atomic number is close to 34 (selenium) with some of the yield to be shared by stable Br^{81}. The selenium fraction was removed too late to observe the 17-min. Se^{81} ground state but a moderate amount of 58-min. Se^{81m} was measured amounting to 15 percent of the total yield predicted for this mass number.

Mass 82.—This mass number is predicted to be shared between stable Se^{82} and the shielded isotope 34-hr. Br^{82}. The yield of Br^{82} is one-third of the total yield for this mass number as taken off the curve. Some difficulties were experienced with the bromine chemistry so that the actual yield may be higher than that measured.

Mass 83.—The yield of the predicted product 24-hr. Br^{83} is almost a factor of two below the curve but, as mentioned, some bromine may have been lost in isolating the bromine fraction.

Mass 84.—The most probable nuclides for this mass number are stable Kr^{84} and 30-min. Br^{84} which was not measured. The rubidium fraction showed a 40-day period which is probably that for which positrons were observed and assigned to Rb^{84} by Barber.[8] The predominant mode of decay appears to be by electron-capture and the yield as estimated from the 12-kev x-rays was only about 8 percent of that for this mass number taken from the curve. The low yield would be expected since Br^{84} and Kr^{84} should be the most abundant fission products.

Mass 86.—This mass number should be shared between stable Kr^{86} and 20-day Rb^{86}. The measured yield of Rb^{86} is about one-half that taken from the curve for this mass number. Positrons in low abundance were observed to decay with a 20-day half-life which indicates some positron branching in the decay of Rb^{86}.

Mass 89.—The predicted product, Sr^{89}, was found in slightly higher yield than the point on the smooth curve.

Mass 90.—The yield of the predicted primary product 25-yr. Sr^{90} fell about 30 percent below the curve and the independent yield of 62-hr. Y^{90} made up the difference.

Mass 91.—The measured yield of 57-day Y^{91} also included that from the decay of 10-hr. Sr^{91} and these are the two nuclides which should appear with this mass number. The cumulative yield of Y^{91} fell on the curve.

Mass 93.—The predicted nuclides are 10-hr. Y^{93} and the unobservable long-lived Zr^{93}. The measured yield of Y^{93} accounted for over half of that expected for this mass number.

Mass 95.—The principal primary product is predicted to be 65-day Zr^{95} with a small contribution by 33-day Cb^{95}. The total measured yield lies on the curve with the Zr^{95} accounting for 70 percent of the yield. The columbium fraction was removed too late to observe any Cb^{96}.

Mass 99.—Mo^{99}, predicted to be the primary product for this mass number, represents the highest yield of any fission product and lies at approximately the point of symmetry in the cleavage.

Mass 103.—The yield of 42-day Ru^{103} should include that of the unknown Tc^{103} and these two nuclides are predicted to share mass number 103 as primary fission products. The yield so determined falls about 20 percent below the curve.

Mass 105.—This mass number should be shared between 4.5-hr. Ru^{105} and 36.5-hr. Rh^{105}. Rhodium could not be isolated and the yield of Ru^{105} accounted for about 60 percent of the expected total yield.

Mass 106.—The principal primary fission product should be Rh^{106} with a moderate contribution by 1.0-yr. Ru^{106}. The yield of Ru^{106} amounted to about 30 percent of the total expected for this mass number while Rh^{106} could not be independently measured.

Mass 109.—The 13-hr. Pd^{109} is predicted to be the primary fission product and its yield helps define the yield curve in this region.

Mass 111 (and 110).—Mass 110 is expected to be shared between stable Pd^{110} and the isomers 24-sec. Ag^{110} and 225-day Ag^{110}. Some long-lived silver was observed but a reliable yield could not be calculated. Ag^{111} is expected to be the principal fission product for mass 111 and its measured yield is about 80 percent of the value from the curve.

Mass 112.—For this mass number 3.2-hr. Ag^{112} is predicted to account for the major amount for the yield with moderate contribution by stable Cd^{112}. It was found that Ag^{112} did account for most of the yield but that the 21-hr. Pd^{112} contributed significantly.

Mass 115.—The 2.3-day and 43-day isomers of Cd^{115} should share the mass number yield with In^{115}. The sum of the yields for the cadmium isomers is about half the total yield for this mass number. A point of importance is that the two Cd^{115} isomers are formed in almost equal yield. The ratio of 2.3-day Cd^{115}/43-day Cd^{115} is 14 in the slow neutron fission of uranium which is presumably the ratio which ensues from the β^--decay of Ag^{115}. The interpretation is that in the fission of bismuth, Cd^{115} is formed as a primary fission product as the theory demands.

Mass 118 and 119.—The predicted major element for both of these mass numbers is tin, element 50. In the case of 118 the favored value of Z is slightly below 50 and for 119 it is slightly above. The only activities measured which are attributed to these mass numbers are two isotopes of tellurium, element 52. One would predict low yields for both, with Te^{118} being more rare than Te^{119}. The yields of Te^{118} and Te^{119} are respectively 0.3 percent and 5 percent of the expected total yields for the mass numbers.

[8] W. C. Barber, Phys. Rev. **72**, 1157 (1947).

Mass 120.—The predicted primary fission product is the shielded nuclide Sb^{120} (of which two isomers apparently exist) with some contribution by stable Sn^{120}. Only the 6.0-day isomer of Sb^{120} could be measured, and its yield was estimated from the x-ray counting rate as about 40 percent of that read from the smooth curve.

Mass 121.—Stable Sb^{121} should be the primary product with slight contribution by Te^{121}. The yield of the 143-day isomer of Te^{121} was measured as about 10 percent that expected for the mass number.

Mass 122.—This mass number should be shared between stable Te^{122} and shielded 2.8-day Sb^{122}. The yield of Sb^{122} was somewhat lower than expected amounting to about one-tenth that for the mass number.

Mass 124.—This represents an interesting case in that the principal predicted product, stable Te^{124} lies between a recognized β^--emitter and β^+-emitter. The prediction actually would indicate appreciable contribution by 4.0-day I^{124} and very little by 60-day Sb^{124}. The measured yield for I^{124} is about 25 percent the expected yield for the mass number while Sb^{124} contributes about 7 percent.

Mass 125.—The 56-day iodine which has been assigned to I^{125} was obtained in good yield. There is the difficulty in this case, as well as others in which yields are determined by counting x-rays, in estimating the x-ray counting efficiencies. If the counting efficiency is assumed to be 0.5 percent then I^{125} falls near the curve as would be predicted.

Mass 126.—According to prediction the shielded isotope 13.0-day I^{126} should account for most of this mass number. The yield as determined is a factor of 10 below the curve and this is the farthest deviation from prediction that is known. It is worth examining this activity to see if it actually decays largely by electron-capture.

Mass 131 and 132.—The determination of yields of the predicted activities for mass number 131(Cs^{131} and Ba^{131}) was unsatisfactory because of difficulties with cesium chemistry and in the estimation of counting efficiencies of both activities. Nevertheless the yields as estimated accounted for one-third of the total for this mass number and, significantly, 8.0-day I^{131} which is two units of Z removed from the predicted value, could not be detected at all.

Mass number 132 is predicted to fall principally on stable Ba^{132} with moderate contribution by 7-day Cs^{132}. The yield of the latter was estimated at over 10 percent of the total expected for mass 132.

Mass 133.—The predicted nuclide is Ba^{133} of which two isomers are known. Only the 39-hr. Ba^{133m} could be measured and its yield alone could account for almost all that was expected for this mass number.

Mass 139.—The position of the fission yield curve becomes rather indeterminate in this range so it is difficult to say whether or not measured yields conform with the theory. For mass 139 the predicted primary fission products are Pr^{139} (unknown) and 140-day Ce^{139}. On the assumption that Pr^{139} would have decayed into Ce^{139}, the yield of the latter determines the yield for both. This measured yield fell somewhat above the smooth curve.

Mass 140.—The predicted product Pr^{140} could not be measured and its decay product is stable. A careful search was made for 12-day Ba^{140} by identifying its 40-hr. La^{140} daughter. Since barium is three units of Z removed from praseodymium one would expect a very low yield, and it was found to occur in less than 1 percent of the expected number of events giving this mass number.

Mass 141.—The major predicted primary fission product is stable Pr^{141} with some contribution by Nd^{141}. The only species of this mass number measured was 28-day Ce^{141} whose yield was about half that given by the curve. A small displacement of the curve in this region makes great differences in yield so that measurements here can only show whether the predicted nuclear types are appearing.

Cross Section for Bismuth Fission

In the short bombardments comprising this study the yield of Mo^{99} was determined in each case to serve as a standard for comparison between bombardments. These irradiations were carried out with probe targets in the circulating beam in which it is not possible to measure accurately the beam current which strikes the target. The fission yields shown in Table I and Fig. 1 were determined by integrating the smooth fission yield curve and by this means Mo^{99} was found to occur in 5.0 percent of the fissions.

A single experiment was carried out in this laboratory in which the 190-Mev deuteron beam was deflected outside the vacuum chamber and a well collimated beam of 3.0×10^{-5} microamperes was measured. A bismuth foil 1-mm thickness was irradiated and the Mo^{99} measured. The absolute value for the Mo^{99} yield along with its fission yield gave a fission cross section of 0.20×10^{-24} cm^2. The uncertainties of this cross section are largely that of estimating the shape of the fission yield curve and the value is probably accurate within 20 percent.

Fission of Separated Lead Isotopes

In order to explain the distribution of the fission products of bismuth it was necessary to postulate the emission of some 10 or 12 neutrons so that the actual nuclei undergoing fission lie close to Po^{199}. It is perhaps significant that the fissionability parameter, Z^2/A, for this nucleus lies in the same range as the naturally occurring uranium isotopes. One would expect that the fission of lead would be governed by a mechanism similar to that of bismuth and that the lighter isotopes of lead would undergo fission more readily than the heavier isotopes. Thus Pb^{204} should not require the loss of so

many neutrons as Pb^{208} before fission begins to compete with other modes of energy dissipation. As will be discussed, it is not to be expected that there exists a sharp energy threshold for fission so the effect to be looked for is a difference in yield at relatively low deuteron energies.

Two samples of isotopically enriched lead were irradiated with deuterons from 30 to 100 Mev and the yields of Mo^{99} compared. In both samples the yields decreased with decreasing deuteron energy; the sample enriched in Pb^{204} showed the higher cross section at all energies and the difference became greater at lower energies. The results are shown in Fig. 3 in which the relative cross sections between the two samples are indicated by the factors between them. The absolute cross sections plotted as ordinates should not be considered accurate because of the uncertainty in estimating the deuteron beam. In Fig. 3 are also shown the compositions of the two lead samples. In order to make certain that the observed differences of Mo^{99} yields were proportional to fission yields and did not merely reflect a shifting of the fission product distribution, two other fission products were measured representing points on both arms of the distribution curve. These nuclides Cu^{67} and Ba^{133} could only be measured with 50, 75 and 100 Mev deuterons because of low yields. Within the limits of error both of these showed parallel changes with that noted for Mo^{99}.

It should be mentioned that the yields shown in Fig. 3 are for the lead compositions indicated. The yield differences between pure Pb^{204} and Pb^{208} should be even greater. Also, in the case of Pb^{208} with 30 Mev deuterons the yield was so low that it could have resulted from 0.1 p.p.m. uranium in the lead, an amount which could conceivably have been present.

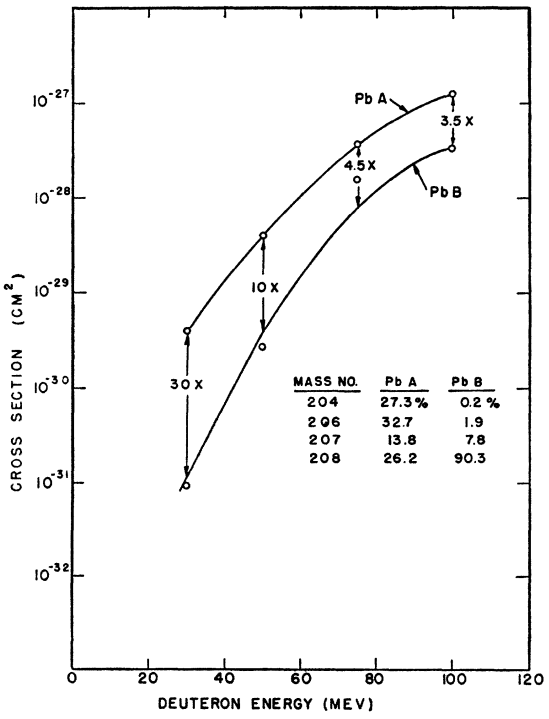

FIG. 3. Yields of Mo^{99} in the fission of partially separated lead isotopes.

DISCUSSION

In developing the mechanism for the fission of bismuth it is convenient to refer to the process as "high energy fission" since it is probable that a similar mechanism is at play in the fission of all heavy elements with high energy particles. It is also profitable to contrast this process with that observed for nuclei which undergo fission with slow neutrons and to correlate this information with fragmentary data obtained at intermediate energies. A number of aspects of the high energy reaction demand examination and these include the excitation function, mass and charge distribution between the fragments, neutron emission and competitive reactions.

Excitation Function

It is not possible to define a threshold energy for the fission of bismuth as the true excitation energy or threshold is undoubtedly considerably lower than that at which an appreciable cross section is observed. Since the question of levels of excitation with high energy particles is itself complicated by nuclear transparency[9] it may be well first of all to consider this point. With deuteron energies up to at least 50 Mev the interaction is probably accurately described by compound nucleus formation. Even at 190 Mev, the probability for complete transparency is low so that events in which some nuclear excitation occurs approximate the geometric cross section. It has been calculated[10] that collisions with 190-Mev deuterons in which less than 100 Mev is transferred occur in only about 25 percent of the cases. Since the proposed mechanism to account for bismuth fission with 190-Mev deuterons postulates the evaporation of about 12 neutrons it is probable that these 25 percent of the events cannot produce the fissions observed. (This contradiction in terms in view of the observed fission of bismuth at 50 Mev is only apparent, since for the present we are discussing reactions which occur with cross section of 0.2 barn while the cross section for 50-Mev neutrons is less than 10^{-3} barn.[11]) Of the remaining 75 percent of the events let us assume that there is a broad maximum probability for excitation at about 150 Mev. The cross section for such excitations is then about 2 barns which is approximately ten times the observed fission cross section. Since we assume that any excited nucleus which is degraded to Po^{199} has a high probability of undergoing fission, it follows that about 90 percent of the 2 barns is taken up by reactions in which some charged particles are

[9] R. Serber, Phys. Rev. **72**, 1114 (1947).
[10] L. W. Baumhoff, private communication.
[11] E. L. Kelly and Clyde Wiegand, Phys. Rev. **73**, 1135 (1948).

emitted or which fall short of the necessary neutron emission by the emission of high energy neutrons. It may be pointed out that on the present theory it is not possible to account for the formation in good yield of such shielded nuclides as Sb^{120} and Br^{82} and of the Cd^{115} isomers in the observed ratio if as many as two protons are lost giving Pb^{199} instead of Po^{199} as the nucleus responsible for most of the fissions.

With regard to a threshold energy for bismuth fission it has already been mentioned that any observed threshold will probably be that at which fission can compete appreciably with other forms of energy dissipation. The liquid drop model which leads to the deformation or fissionability parameter Z^2/A predicts an increased tendency for fission for decrease in A. For $Z=84$, the fission threshold is calculated to be ~ 12 Mev at $A=210$ and ~ 6 Mev for $A=200$.[12] We know from observations in the vicinity of uranium that when the excitation energy is around the binding energy of a neutron, i.e., ~ 6 Mev, that fission is capable of using up a considerable portion of the geometric cross section (see, for example, Klema[13]). From the results of the present study it is to be inferred that if the excitation energy for fission is about twice this value, excitation near or even considerably above the threshold results in a very poor competition by fission and instead neutron emission is favored. It is only when the threshold for fission is decreased through the ejection of neutrons that the rate for fission increases. Further emission of neutrons only serves to produce a nucleus which is even more susceptible to fission. From this picture one would expect to observe fission of bismuth with increasing yields as the projectile energy increases. There is good evidence that this is the case. Figure 3 shows the effect for lead isotopes up to 100 Mev, and Kelly and Wiegand[11] have counted fissions of bismuth with neutrons in which an increase by a factor of about fifty was observed between 50 and 85 Mev. In a series of experiments not recorded above, the yields from bismuth fission were measured for Cu^{67}, Mo^{99} and Ba^{133} with 100-Mev deuterons, 190-Mev deuterons, and 380-Mev alpha-particles. There was a progressive increase in yield for all three nuclei with increase in projectile energy. That the fission product distribution also broadens is evidenced in that, whereas Mo^{99} yield increased about threefold between the 100 Mev and the 380 Mev irradiations, the yields of Cu^{67} and Ba^{133} increased tenfold. In summary, it will be expected that the distribution of fission products of bismuth as shown in Table I and Fig. 2 is that for reactions produced by sufficient excitation to result in high fission cross section, in this case about one-tenth the geometric cross section. If it were possible to measure the rare events which occur at low excitation energy, the distribution could be quite different.

[12] S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).
[13] E. D. Klema, Phys. Rev. **72**, 88 (1947).

Charge Distribution

The ratio of charge to mass in the asymmetrical cleavages from slow neutrons on U^{235} and Pu^{239} can be described by the postulate that the beta-decay chains from the two fission fragments are of equal length.[14] This demands a redistribution of charge in the fission process and probably permits the maximum amount of energy to be directed toward the separation of the fission fragments. This demand for energy economy in order to allow maximum kinetic energy for the slow neutron process is in sharp distinction with the observations for high energy fission. One of the requirements to explain the types of fission products in the high energy reaction is that there be no redistribution of charge, that is, both fragments bear the same neutron to proton ratio as the parent nucleus. Obviously this mechanism does not in general produce maximum kinetic energy of the fission fragments in the asymmetrical cleavages since it would be more economical to form two stable products instead of one which is β^--unstable and the other β^+-unstable. These observations indicate that the high energy fission process takes place from a slightly supra-threshold energy state in which the further energy which would accure from rearrangement of neutrons and protons is not important, and that by the same token the reaction is faster than slow neutron fission in which such rearrangements do take place.

There is some evidence that this same mechanism is at play in the high energy fission of uranium. Here it is not so easy to observe the effect as in the case of bismuth in which positron emitters are actually produced, but measurements in this laboratory show the ratio of Cd^{115} isomers to be different from that produced in slow neutron fission and the 28.7-hr. barium tentatively assigned to Ba^{135m} is found in good yield.[15]

Mass Distribution

One of the outstanding features of slow neutron fission is the low probability for symmetrical cleavage. In the high energy reaction the most favored cleavage is symmetrical and this apparently applies to uranium[16] as well as to bismuth. That the fission of bismuth with high energy particles has symmetrical cleavage as the most prominent mode is evidenced not only by the identification of fission products. Jungerman and Wright have measured the fission pulses from neutrons on bismuth and found the most prominent mode that in which the fragments have equal energy.[17]

It would be consistent with the other observations on high energy fission to correlate the preponderance of symmetrical cleavages with the rapid reaction rate and the unimportance of small economies of energy.

[14] Coryell, Glendenin, and Edwards, Phys. Rev. **75**, 337 (1949).
[15] O'Connor, Seaborg, and Folger, private communication.
[16] P. R. O'Connor and G. T. Seaborg, Phys. Rev. **74**, 1189 (1948).
[17] J. Jungerman and S. C. Wright, Phys. Rev. **75**, 1470 (1949).

Conversely one might associate asymmetrical fission with a slower reaction rate and with the necessity for putting as much energy as possible into kinetic energy of the fragments. In the case of slow neutron fission the ranges and energy distribution of the fission fragments have been measured[18–20] but there is no evidence that the highly asymmetric cleavages produce greatest total fission product energies. On the contrary the greatest energies appear to result from nearly equal mass distribution. As a result it is probably not possible to state with certainty the reasons for asymmetric fission. What appears to be fairly certain is that as the nuclear excitation becomes greater than the fission threshold the tendency toward splitting into equal fragments increases. Besides the present studies this is evidenced by the shallower valley of the distribution curve for Pu^{239} as compared with U^{235},[21,22] presumably due to lower fission threshold with increase in Z, and the similar effect when Th^{232} is irradiated with fast neutrons[23] and with charged particles.[24]

Neutron Emission

The distribution of the fission fragments in the case of 190-Mev deuterons on bismuth indicates that about 10 neutrons are lost in the reaction. It is most reasonable to assume that these neutrons precede the fission reaction because of the expected faster rate of neutron emission from a nucleus in which the barrier toward fission demands excitation energy several times that of neutron binding energies. However one might expect essentially the same distribution of fission products if fission resulted in highly excited fragments which subsequently boiled off nucleons.

The measurements of Jungerman and Wright[17] of the energy of the fission fragments of bismuth indicate that the fragments come apart with kinetic energy within a few Mev of predictions on the basis of fission with near threshold energy. This is in agreement with deductions made from the present findings, namely, that almost all of the nuclear excitation imparted by the high energy projectile is used in evaporating neutrons.

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[19] Katcoff, Miskel and Stanley, Phys. Rev. **74**, 631 (1948).
[20] D. C. Brunton and G. C. Hanna, Phys. Rev. **75**, 990 (1949).
[21] J. L. Fowler and L. Rosen, Phys. Rev. **72**, 926 (1947).
[22] Steinberg, Seiler, Goldstein, and Dudley, U. S. AEC Report MDDC-1632 (Jan. 1948).
[23] A. Turkevich, U. S. AEC Report MDDC-1568 (Dec. 1947).
[24] A. S. Newton, Phys. Rev. **75**, 17 (1949).

APPENDIX

For each element investigated, the essentials of the chemical procedure and isotope identification used are summarized. The chemistry steps listed were usually repeated several times in cycles. In some cases the amount of activity encountered is mentioned without reference to irradiation conditions solely for the purpose of indicating the limitations in accuracy of identification and yield determinations.

Calcium—Precipitated as CaC_2O_4; separations of sulfides insoluble in 0.5 N HCl, hydroxides insoluble in NH_3, and $Ba(NO_3)_2$ and $Sr(NO_3)_2$ with fuming HNO_3.

Particles, which had a rate of 30 counts per minute initially, were observed to decay with ~155-day half-life and were assumed to be from Ca^{45}.

Chromium—Precipitated as $Cr(OH)_3$ and extracted into ether as H_3CrO_8; separations of Bi_2S_3 and Sb_2S_3 in 1 N HCl, $Fe(OH)_3$ from H_2O_2-NaOH solution.

No activity was observed in the sample but short-lived ($T_{\frac{1}{2}}$ < one week) activities would have been missed.

Iron—Extracted into isopropyl ether as $FeCl_3$ and precipitated as $Fe(OH)_3$ with NH_3; separations of acid-insoluble sulfides, $BaSO_4$, AgI, Zr phenylarsonate, and Ru by distillation.

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

Nickel—Precipitated with dimethylglyoxime in neutral solution; separation of acid-insoluble sulfides, $Fe(OH)_3$ with NH_3, Pd dimethylglyoxime from 0.5 N acid, and AgI in ammoniacal solution.

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.

Copper—Precipitated as CuCNS and CuS; separations of $Fe(OH)_3$ plus $BaCO_3$ with NH_3, CdS in cyanide solution, and AgCl.

The main component of the decay curve was a 57-hour ~0.5 Mev β^--emitter assigned to Cu^{67}.

Zinc—Precipitated as $ZnHg(CNS)_4$ and ZnS; separations of Bi_2S_3 and $Fe(OH)_3$ plus $BaCO_3$ from NaOH solutions.

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}.

Gallium—Extracted into ether as $GaCl_3$; separations of Mo α-benzoin oxime, Bi_2S_3, and $Fe(OH)_3$ plus $BaCO_3$ from NaOH solution.

The decay curve resolved into a 4.9-hour and a 14-hour component. The 5-hour period was identified as Ga^{73}; the 14-hour period was identified as Ga^{72}.

Arsenic—Precipitated with H_2S from concentrated HCl and dissolved in NH_3 solution, distilled as $AsCl_3$; separation of $GeCl_4$ by distillation.

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 ~50 percent positrons, was identified as the shielded As^{74}. These was evidence of the hard positrons of As^{76} in the absorption curve.

Selenium—Distilled as $SeBr_4$, and precipitated as Se° with SO_2 in cold concentrated HCl.

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 isomer of Se^{81}.

Bromine—Oxidized to Br_2 and extracted into CCl_4 after $NaNO_2$ oxidation of I^- to I_2 and prior extraction, extraction of Br_2 into CCl_4 with $NaHSO_3$ solution, precipitation of AgBr.

The activity observed three hours after the midpoint of the bombardment resolved into 2.7-hour and 35-hour periods. The 2.7-hour period was identified as Br^{83}; the 35-hour period was identified as the shielded Br^{82}.

Rubidium—Precipitated as $RbClO_4$, Rb_2PtCl_6; separation of insoluble sulfides, hydroxides, carbonates, and Cs silicotungstate.

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} and the ~40-day periods were associated with shielded Rb^{84}.

Strontium—Precipitated as $Sr(NO_3)_2$ with fuming HNO_3, and SrC_2O_4; separations of $Fe(OH)_3$ and $BaCrO_4$.

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 Sr^{90} was detected by separation of its 62-hour Y daughter.

Yttrium—Precipitated as YF_3, $Y(OH)_3$, and $Y_2(C_2O_4)_3$; separations of Bi_2S_3.

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}. The 62-hour period was identified as Y^{90}. The yield of 57-day Y^{91} was measured in a Y fraction separated three weeks after bombardment. The rare earth activities present in the Y fraction did not interfere because their yields are much less.

Zirconium—Precipitated as $BaZrF_6$, and Zr cupferrate in 1 *M* HCl; separations of LaF_3 and $BaSO_4$.

The 68-day ~0.4 Mev β^- observed was identified as Zr^{95}.

Columbium—Precipitated as Cb_2O_5 from a high acidity solution and from NH_4F solution; separations of $BaZrF_6$, Bi_2S_3, and Te°.

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}.

Molybdenum—Extracted into ether from 6*N* HCl after Br_2 oxidation, precipitated as α-benzoin oxime from 3*N* HCl after Br_2 oxidation, precipitated as α-benzoin oxime from 3*N* HNO_3 containing $H_2C_2O_4$, and as Ag_2MoO_4; separations of $Fe(OH)_3$.

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

Ruthenium—Distilled from fuming $HClO_4$ containing H_3PO_4, precipitated as a lower oxide by ethyl alcohol in NaOH solution, and as Ru° by Mg in HCl; separations of OsO_4 by distillations from HNO_3.

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-hour and 36-hour periods were identified as Ru^{105} and Rh^{105} daughter. In a Ru 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^{105} daughter.

Palladium—Precipitated as dimethylglyoxime from 0.5*N* HCl; separations of $Fe(OH)_3$, AgI, and AgCl.

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^{109} daughter.

Silver—Precipitated as AgCl and Ag_2S; separations of $Fe(OH)_3$.

The decay curve consisted of a 3.5-hour component identified as Ag^{112}, and a 7.5-day 1.0 Mev component identified as Ag^{111}. The 225-day isomer of Ag^{110} was also observed in small amounts. 5.3-hr. Ag^{113} would not have been resolved.

Cadmium—Precipitated as CdS from 0.2 *N* HCl and from NH_3 solution, and as $CdNH_4PO_4$; separations of basic ferric acetate, $In(OH)_3$, PdS, and Sb_2S_3.

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

Antimony—Precipitated as Sb_2O_3, and as Sb_2S_3 from hot 3 *N* HCl; separations of As_2S_3, MoS_3, Te°, $Fe(OH)_3$ from KOH solution, and Bi_2S_3 from K_2S solution.

An Sb 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 Sb^{124}.

Tellurium—Precipitated as Te° by SO_2 in 3 *N* HCl; separations of Se°, $SeBr_4$ by distillation, and $Fe(OH)_3$ from NaOH solution.

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

Iodine—Distilled as I_2 from HNO_3, extracted as I_2 into CCl_4 after $NaNO_2$ oxidation, and precipitated as AgI.

Positrons, x-rays and γ's of 4.5-day half-life were observed and identified as I^{124}. 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}.

Cesium—Precipitated as $CsClO_4$, Cs_2PtCl_6, and Cs silicotungstate from 6*N* HCl; separations of insoluble sulfides, hydroxides, and carbonates.

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 ~30-day and ~300-day tails. The 6-day activity was identified as Cs^{132}; the 10.5-day activity was identified as Cs^{131} and the other activities could not be identified as Cs isotopes.

Barium—Precipitated as $Ba(NO_3)_2$ with fuming HNO_3, $BaCrO_4$, and $BaCl_2$ with ether—HCl mixture; separations of $Fe(OH)_3$.

A 38-hour 280 kev e^- activity was identified as the 38-hour isomer of Ba^{133}. A 40-hour La activity was separated from the Ba, showing the presence of Ba^{140} in very low yield. The presence of Ba^{131} was shown by growth in the x-ray decay curve and the separation of Cs^{131} daughter.

Cerium—Precipitated as CeF_3, $Ce(OH)_3$ with carbonate-free ammonia, $Ce_2(C_2O_4)_3$, $Ce(IO_3)_4$, and $Ce_3(PO_4)_4$; separations of Bi_2S_3.

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}.

Europium—Precipitated as EuF_3, $Eu(OH)_3$ with carbonate-free ammonia, and $Eu_2(C_2O_4)_3$; separations of Bi_2S_3, and $Ce(OH)_3$ after zinc amalgam reduction of Eu.

An initial activity of 500 particles, 32 γ, and 5 x-ray counts per minute decayed with a 21-day half-life and was tentatively identified as Eu^{149}.