$g(\theta)$ probably consists partly of very-low-energy secondary particles and partly of nuclear scattering events.

Since most of these showers were generated in lead, the secondary particles were probably formed in both multiple and plural production processes. It is therefore difficult to make a unique comparison with either theory. The plural production[3] theory predicts values of $s<1.5$; the multiple production theories[2] predicts $s\sim1.5$ to 2 (Fermi's statistical model) or higher values (Lewis, Oppenheimer, and Wouthuysen). From this fact it may be inferred[4] that a theory such as Fermi's will account for the momentum distribution of at least the higher-energy secondary particles.

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High-Energy Fission of Heavy Elements. Nuclear Charge Dependence*

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Radiochemical studies of the fission of heavy elements ($_{67}Ho^{165}$ to $_{90}Th^{232}$) with 450-Mev protons have been performed. Radioactive nuclides varying in mass number from 59 to 115 were isolated from the various targets and, from their measured cross sections, the cross section *vs* mass number dependence was determined on the assumption of a yield *vs* charge distribution curve constant with mass number of the fission product and atomic number of the target nucleus. The integrated fission cross sections, in barns, were calculated to be 0.67, 0.21, 0.061, 0.019, 0.0050, and ~0.002 for thorium, bismuth, gold, rhenium, tantalum, and holmium, respectively. Anomalies in the cross sections and most probable charge values for the heavier nuclides isolated from holmium suggest the existence of another competing process, labelled "fragmentation," along with spallation and fission. The importance of this process in heavy element bombardment is discussed.

I. INTRODUCTION

MANY investigations of the fission of heavy elements, induced by particles and x-rays, have been reported.[1] Using radiochemical methods, Perlman and co-workers[2] showed that fission occurs in the elements tantalum to bismuth with high-energy neutrons, deuterons, and alpha particles. From the studies of Kelly and Wiegand[3] and Jungerman[4] on the cross section of various elements for fission as a function of the energy of the bombarding particle, it is seen that the fissionability increases markedly as the nuclear charge increases. Also, for a given element, except the very fissionable ones like uranium and thorium, the cross section increases substantially as the energy of the bombarding particle increases from 50 Mev to 350 Mev.

A detailed radiochemical investigation of the fission of bismuth with 190-Mev deuterons was made by Goeckermann and Perlman.[5] They showed that the fission-yield curve is a symmetrical single-humped curve, and that the primary fission products could be considered as arising from a fissioning nucleus formed by evaporation of twelve neutrons after the excitation of the target nucleus by the bombarding projectile. The isolated fission products were shown to be neutron-excessive for low mass and neutron-deficient for high mass, as expected from a fission process occurring with a constant neutron to proton ratio. Recently, Biller[6] studied the fission of bismuth with 340-Mev protons and interpreted the results as arising from a distribution of fissioning nuclei.

The work reported in this paper compares the fission process in some of the elements from holmium to thorium. The bombarding particles were 450-Mev protons from the University of Chicago synchrocyclotron. The radiochemical method of isolating the radioactive species, ascertaining their identity, and measuring their radioactivity was used to determine the various parameters of the fission process such as cross section, neutron to proton ratio of the fissioning nucleus, the variation in yield with charge, etc. As the charge of the target nucleus decreases, there is observed a decrease in fission cross section, a decrease in the mass number of the most probable fission products, and a decrease in the neutron to proton ratio of the primary fragments formed in high yield. In contrast, the excitation energy for the fission process remains fairly constant as the nuclear charge is varied.

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[6] W. F. Biller, University of California Radiation Laboratory Report, UCRL 2067, December, 1952 (unpublished).

II. EXPERIMENTAL TECHNIQUES

A. Irradiations

The irradiations were made with the internal 450-Mev proton beam of the University of Chicago synchrocyclotron by placing foils or powder packages of the targets on the 2-in. probe. Foil samples of thorium, bismuth, gold, and tantalum, and powdered samples of rhenium and holmium oxide in aluminum wrapping were used. The foils or powder samples were sandwiched by 1-mil aluminum foils which acted as beam monitors. The target face, measuring 1.9 cm×1.0 cm was placed tangentially along the beam with the protons traversing the longer dimension. This length corresponds to a target thickness of about 20 g/cm^2. The irradiation times varied from 20 min to 2 hr.

B. Preparation of Target Solutions

The target foil or powdered compound was dissolved in the appropriate acid and diluted to volume. Care was taken in the solution of each target to ensure the retention of the radioactivity. Cases in which extra care was needed are noted.

(1) Copper may be lost from target solutions where fluoride is present such as the case of tantalum dissolved in a mixture of conc HNO_3 and HF or thorium dissolved in conc HNO_3 and a crystal of KF. In these cases the targets were dissolved quickly, aliquots were added to copper carrier, and the mixture boiled to expedite interchange.

(2) Bromine may be lost as Br_2 from the solution by volatilization when the target is dissolved in an oxidizing medium. The bromine may be retained by dissolving in a closed system and sweeping the gases through a solution of NaOH containing bromide carrier. It was shown that about 60 percent of the bromine activity was found in the NaOH solution when targets were dissolved in aqua regia or conc HNO_3.

(3) Silver in tracer quantity may be lost by adsorption on walls, dust, etc., when a target is dissolved in a solution containing chloride ion. Irradiated gold foils dissolved in aqua regia gave erratic results for silver. More consistent results were achieved by dissolving the target in aqua regia, and diluting to volume with conc HCl.

In general, the targets were dissolved soon after the irradiations and all aliquots of a target solution were added to the appropriate carriers and shaken before the chemical separations were started.

The 1-mil aluminum monitors were dissolved separately in HCl and diluted to 25 ml with water. Several 0.1-ml aliquots were taken from each solution, evaporated on 1-mil aluminum over an area of 2 cm^2, and mounted on a cardboard card, similar to the fission-product samples.

C. Chemical Procedures, Sample Preparation, and Counting Techniques

The chemistry used in isolating the samples was adapted mainly from procedures of the Plutonium Project[7] and Meinke.[8] For each element, an aliquot of the active solution in either nitric or hydrochloric acid was added to 10 to 20 mg of carrier and the appropriate chemistry was performed. The essentials of the chemical operations are given in the Appendix.

Final precipitates of all samples were transferred by suction to Whatman No. 42 paper over 2-cm^2 area using a chimney-Hirsch funnel apparatus. The precipitates were mounted on cardboard cards and covered with cellophane of 2.6-mg/cm^2 thickness. Counting was done with end-window Geiger or methane-flow proportional counters. The nuclides were identified by half-life determination and absorption measurements. Cross sections were calculated from the measured radioactivity by correction for chemical yield, dead-time losses of the counter, irradiation time, external absorption by the cellophane, air, and counter window, self-scattering, self-absorption, and backscattering,[9] and the branching ratio for beta decay. The fission-product samples and the Na^{24} monitors were counted under the same geometry conditions. The cross sections for the fission products were calculated from the previously measured cross section[10] of (10.8±0.5) mb at 450 Mev for the formation of Na^{24} from aluminum by the reaction $Al^{27}(p,3pn)Na^{24}$.

D. Nuclides Identified

The nuclides of each element were identified by comparison of the observed properties with literature values.[11] For a given element, it was noted that in general the lighter isotopes increased in yield relative to the heavier ones as the atomic number of the target decreased. Following is a list of the elements studied and the pertinent data on the observed radioactive nuclides. Not in all cases where identification of a nuclide was made was it possible to determine the cross section for formation of the nuclide.

Iron: All samples yielded the 46-day Fe^{59} isotope.

Nickel: 2.5-hr Ni^{65} and 56-hr Ni^{66} were identified in all samples. A small but increasing yield of 37-hr Ni^{57} appeared in targets of lower atomic number.

[7] *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), selected papers of Part VI, National Nuclear Energy Series, Plutonium Project Record, Div. IV, Vol. 9.

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[11] Way, Fano, Scott, and Thew, National Bureau of Standards Circular No. 499 and Supplements (U. S. Government Printing Office, Washington, D. C., 1950); Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

Copper: The isotopes 3.3-hr Cu^{61}, 12.8-hr Cu^{64}, and 61-hr Cu^{67} were found in copper samples of all targets.

Arsenic: In all arsenic samples, the 17.5-day As^{74}, 26.8-hr As^{76} (+25.8-hr As^{72}), and 40-hr As^{77} isotopes were present. In the higher atomic number targets, the 90-min As^{78} could be identified. To determine the separate yields of As^{72} and As^{76}, the arsenic samples were counted between the pole faces of a strong magnet around which an end-window counter could be rotated. At $+90°$ and $-90°$ from the vertical, β^+ radiation and β^- radiation, respectively, could be separately counted. Since the energies of the beta radiation of As^{72} and As^{76} are about the same, it was assumed that the counting efficiencies were the same, and that the same fraction of the energy spectrum entered the counter at $+90°$ and $-90°$. The ratio of the yield of 25.8-hr As^{72} to that of 26.8-hr As^{76} was found to be 0.8 for holmium, 0.79 for tantalum, 0.53 for rhenium, 0.25 for gold, 0.11 for bismuth, and 0.061 for thorium.

Bromine: In all samples, the 4.4-hr Br^{80m}, 35.5-hr Br^{82}, and 2.3-hr Br^{83} isotopes were found. A cross section for 16-hr Br^{76} was obtained in holmium.

Rubidium: All targets yielded the shielded isotope 19.5-day Rb^{86}.

Strontium: The 25.5-day Sr^{82} reported during this work[12] and characterized previously by Castner[13] and the 54-day Sr^{89} were present in all samples. The yield of ~30-yr[14] Sr^{90} was obtained through the isolation of its 65-hr Y^{90} daughter from old strontium samples.

Zirconium: The 17-hr Zr^{97} and 65-day Zr^{95} isotopes were identified in the higher atomic number targets. In the lower atomic number targets the increased yield of 85-day Zr^{88}, with its 104-day daughter, made accurate identification of 65-day Zr^{95} impossible.

Niobium: The 23-hr Nb^{96} and 35-day Nb^{95} isotopes were identified in the higher atomic number targets, but, again, long-lived isotopes or impurities made positive identification of 35-day Nb^{95} from the lower atomic number targets impossible.

Molybdenum: The 67-hr Mo^{99} was found in all samples.

Palladium: The 14-hr Pd^{109} was present in all samples. The higher atomic number targets yielded the 21-hr Pd^{112} and the lower atomic number targets gave the 4.0-day Pd^{100} and the 17-day Pd^{103} isotopes.

Silver: All samples contained 7.5-day Ag^{111}, 3.2-hr Ag^{112}, and 5.3-hr Ag^{113}. Samples of silver separated immediately after irradiation afforded a measure of the independent yield of Ag^{112}. Lower atomic number targets yielded the 8.5-day Ag^{108} isotope which decays by orbital electron capture.

Cadmium: All samples contained the isomers 43-day Cd^{115m} and 54-hr Cd^{115}. The yield of Cd^{115} was corrected for the In^{115m} daughter which decays 5 percent by β emission and 95 percent by isomeric transition,[15] 49 percent converted.[11] The isotopes 7-hr Cd^{107} and 470-day Cd^{109} were observed in lower atomic number targets.

Cesium: The shielded isotope 13-day Cs^{136} was obtainable only from thorium. Samples from the other targets yielded only the neutron deficient isotope 31-hr Cs^{129}.

III. RESULTS

The cross section for each nuclide from a given target was determined by measuring the radioactivity of the isolated nuclide, correcting for scattering, absorption, etc., and comparing the corrected saturation activity with that of Na^{24} in the monitor foils. The results of these measurements are given in Table I, where the values listed are averages of from one to eight independent determinations (the number of determinations is given in parentheses next to the measured cross sections). Where several determinations of the same cross section were made, the average deviation was about 15 percent. Also given in Table I is the classification of the yield as cumulative, C (including decay of precursors), or independent, I, for shielded nuclides.

An examination of the results given in Table I leads to several conclusions. (1) A general trend of decreasing cross section with decreasing atomic number of the target is noted. This is expected from the reported[4] decrease in fission cross section with decreasing atomic number. The cross sections of the neutron excessive fission products of mass number about 100, e.g., Sr^{89}, Mo^{99}, Ag^{111}, decrease by a factor of about 1000 in going from thorium to holmium, and even those of nuclides of mass numbers in the range 70 to 80, unless very neutron deficient, decrease by a factor of 10 to 100. (2) The cross sections for the assorted nuclides measured differ by a factor of about 300 in thorium, but by only a factor of about 10 in holmium. (3) For any given mass range, the decrease in cross section is greater for the more neutron excessive nuclides. In fact, the yields of nuclides closer to stability, e.g., As^{74}, Br^{80m}, Rb^{86}, actually increase somewhat in going from thorium to bismuth, then decrease more slowly with decreasing atomic number than the neutron excessive nuclides. (4) For the same degree of neutron excessiveness, the cross sections of the nuclides of low mass number (60 to 70) decrease much less than those of mass number about 100. (5) The cross sections of the neutron deficient nuclides of low mass number (60 to 70) show very little over-all change with atomic number of target.

The cross sections of some selected nuclides, Cu^{64}, Cu^{67}, As^{74}, Rb^{86}, Sr^{89}, and Pd^{109}, are plotted in Fig. 1. The numbers in parentheses adjacent to the nuclides are the values of Z_A-Z, a measure of the neutron excessiveness or deficiency of the nuclides. The values of Z_A used are those of Coryell.[16] In comparing the extreme cases of Pd^{109} ($Z_A-Z=+1.3$) and Cu^{64} (Z_A-Z

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[16] C. D. Coryell, Ann. Rev. Nuc. Sci. **2**, 305 (1953).

TABLE I. Cross sections of fission-product nuclides (in millibarns).[a]

Nuclide	Yield type[b]	Th	Bi	Au	Re	Ta	Ho
Fe^{59}	C	0.30 (2)	0.23 (7)	0.16 (5)	0.082 (2)	0.062 (2)	0.032 (2)
Ni^{65}	C	0.59 (2)	0.60 (1)	0.32 (1)	0.14 (2)	0.088 (1)	0.021 (1)
Ni^{66}	C	0.63 (2)	0.46 (1)	0.21 (1)	0.081 (2)	0.048 (1)	0.012 (2)
Cu^{61}	C		0.091[c] (1)		0.013 (1)	0.0047 (1)	0.044 (2)
Cu^{64}	I	0.11 (1)	0.29 (2)	0.28 (2)	0.15 (4)	0.11 (4)	0.087 (2)
Cu^{67}	C	0.89 (1)	0.68 (7)	0.44 (6)	0.18 (4)	0.11 (4)	0.027 (2)
As^{72}	I	0.13[c] (2)	0.36 (2)	0.35 (1)	0.20 (3)	0.044 (1)	0.047 (1)
As^{74}	I	0.47 (2)	1.2 (2)	1.2 (1)	0.34 (3)	0.055 (1)	0.064 (1)
As^{76}	I	2.2 (2)	3.2 (2)	1.4 (1)	0.38 (3)	0.056 (1)	0.059 (1)
As^{77}	C	5.2 (2)	4.5 (2)	1.6 (1)	0.46 (3)	0.062 (1)	0.10 (1)
Br^{76}	C						0.0082 (1)
Br^{80m}	I	0.97 (1)	2.0 (1)	1.1 (1)	0.55 (1)		0.057 (1)
Br^{82}	I	1.4 (1)	1.7 (1)	0.78 (3)	0.21 (1)		0.019 (1)
Br^{83}	C	4.4 (1)	1.3 (1)	0.74 (1)	0.15 (1)		0.013 (1)
Rb^{86}	I	3.2 (1)	4.1 (1)	0.89 (1)	0.37 (1)	0.076 (1)	0.023 (1)
Sr^{82}	C		0.16 (2)	0.068 (1)	0.029 (3)		0.0034 (1)
Sr^{89}	C	15 (2)	5.4 (8)	1.2 (5)	0.22 (4)	0.087 (4)	0.029 (2)
Sr^{90}	C	7.2[c] (1)	3.8 (3)	0.73 (2)			
Zr^{95}	C	14 (1)					
Zr^{97}	C	19 (1)					
Nb^{95}	I	1.9[c] (1)					
Nb^{96}	I	3.7[c] (1)					
Mo^{99}	C	27 (2)	5.8 (6)	0.98 (5)	0.14 (2)	0.056 (3)	0.027 (2)
Pd^{109}	C	24 (2)	4.1 (1)	0.34 (1)	0.061 (4)	0.021 (3)	0.028 (2)
Ag^{111}	C	26 (2)	3.9 (2)	0.52 (1)	0.054 (3)	0.024 (3)	
Ag^{112}	I	5.1 (1)	1.3 (1)	0.21 (1)	0.022 (1)	0.0086 (1)	
Ag^{113}	C	16 (2)	1.8 (1)	0.091 (1)	0.011 (1)	0.0037 (1)	
Cd^{115m}	I	7.8 (2)	1.0 (2)	0.11 (3)	0.012 (3)	0.0044 (3)	0.0079 (2)
Cd^{115}	C	15 (2)	0.61 (2)	0.042 (3)	0.0041 (3)	0.0016 (3)	0.0145 (2)
Cs^{136}	I	1.3 (1)					

[a] Numbers in parentheses represent number of independent determinations.
[b] C ≡cumulative (including decay of precursors); I ≡independent, for shielded nuclides.
[c] Uncertainties larger than the 30 percent error quoted in text.

$=-0.3$), we note the exaggerated effects of the trends mentioned earlier.

The errors in the cross sections are rather difficult to determine because of the large number of individual correction factors applied to each measured activity. The activity of the longest-lived isotope of each element was obtained by counting to within 1 percent standard error. The activity of the shortest-lived isotope of an element, in the cases where three periods were present, such as Cu^{61}, As^{72} and As^{76}, Br^{80m} and Ag^{112} may have errors as large as 10 to 20 percent. In the case of the mixture As^{72} and As^{76}, where the percentage of each was determined by magnetic deflection, the yield of As^{76} is the more reliable one since its abundance ranged from 56 percent in holmium to 94 percent in thorium. The half-lives used for correcting the activities differed at most by a few percent from published values and may introduce a few percent error in the cross sections. The error introduced by the aluminum monitor foils may be as high as 10 percent in those cases in which the foils differed by 40 to 50 percent, aside from the error in the cross section for production of Na^{24}. The correction factors for absorption and scattering of nuclides decaying by beta emission of low energy, e.g., Fe^{59}, 0.26 Mev; Br^{82}, 0.47 Mev; Zr^{95}, 0.37 Mev; and Nb^{95}, 0.16 Mev, were large, from 1.2 to 1.9, and varied considerably depending on the sample weight. In these cases, the method of measuring cross sections by comparison to Na^{24} is distinctly less reliable. Where orbital decay occurred in part, e.g., Cu^{61}, Cu^{64}, and Br^{80}, it was assumed that the counting efficiency for the orbital capture was negligible. In all, it is expected that most of the reported cross sections may be in error by as much as 30 percent.

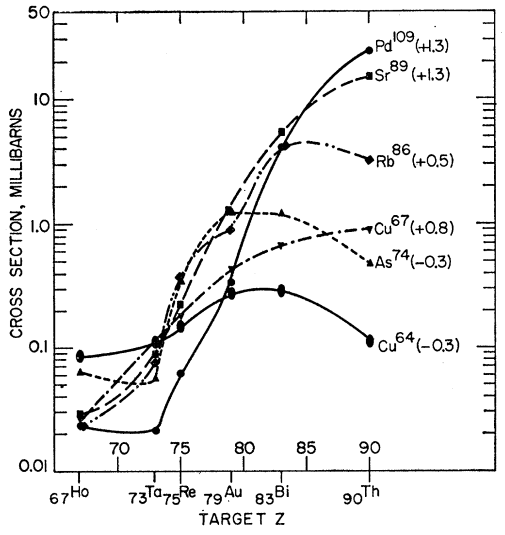

FIG. 1. Cross sections of selected nuclides *vs* atomic number of target. Numbers in parentheses are values of Z_A-Z. Circles—Pd^{109}; squares—Sr^{89}; diamonds—Rb^{86}; triangles ▲—As^{74}; triangles ▼—Cu^{67}; ovals—Cu^{64}.

In addition to the items mentioned above, there

TABLE II. Calculated mass yields for thorium, bismuth, gold, rhenium, tantalum, and holmium. Z_p is the most probable charge.

		Thorium			Bismuth			Gold			Rhenium			Tantalum			Holmium		
A	Z	Z_p	Fraction of yield	Mass yield, mb	Z_p	Fraction of yield	Mass yield, mb	Z_p	Fraction of yield	Mass yield, mb	Z_p	Fraction of yield	Mass yield, mb	Z_p	Fraction of yield	Mass yield, mb	Z_p	Fraction of yield	Mass yield, mb
59	26	25.0	0.950	0.31	26.1	0.659	0.35	26.0	0.700	0.23	26.0	0.700	0.12	26.0	0.700	0.089	27.0	0.300	0.11
65	28	27.4	0.880	0.67	28.5	0.493	1.2	28.5	0.493	0.66	28.5	0.493	0.28	28.5	0.493	0.18	29.3	0.198	0.10
66	28	27.8	0.760	0.83	29.0	0.300	1.5	29.0	0.300	0.70	29.0	0.300	0.27	29.0	0.300	0.16	29.7	0.095	0.13
61	29				27.0	0.050	1.8[a]				26.9	0.038	0.33	26.9	0.038	0.12	27.9	0.263	0.17
64	29	27.1	0.062	1.8	28.2	0.292	0.98	28.2	0.292	0.96	28.1	0.270	0.57	28.3	0.315	0.35	29.0	0.400	0.22
67	29	28.3	0.900	0.99	29.3	0.577	1.2	29.4	0.536	0.82	29.3	0.577	0.31	29.4	0.536	0.20	30.2	0.228	0.12
72	33	30.5	0.010	13[a]	31.5	0.134	2.7	31.5	0.134	2.6	31.5	0.134	1.5	31.5	0.134	0.33	32.0	0.250	0.19
74	33	31.3	0.095	4.9	32.3	0.315	3.8	32.4	0.344	3.5	32.3	0.315	1.1	32.2	0.292	0.19	32.7	0.379	0.17
76	33	32.1	0.270	8.2	33.0	0.400	8.1	33.1	0.396	3.6	33.1	0.396	0.97	33.2	0.389	0.14	33.4	0.366	0.16
77	33	32.5	0.840	6.2	33.4	0.536	8.4	33.5	0.493	3.3	33.5	0.493	0.94	33.6	0.458	0.14	33.8	0.370	0.28
76	35																33.4	0.114	0.073
80[m]	35	33.6	0.156	6.2	34.7	0.379	5.4	34.7	0.379	3.0	34.8	0.389	1.4				35.0	0.400	0.14
82	35	34.4	0.344	4.2	35.6	0.344	4.4	35.7	0.315	2.5	35.6	0.344	0.62				35.8	0.292	0.064
83	35	34.9	0.730	6.1	36.0	0.300	4.2	36.1	0.263	2.8	36.0	0.300	0.49				36.1	0.263	0.048
86	37	36.1	0.272	12	37.1	0.396	10	37.4	0.366	2.4	37.3	0.379	1.0	37.4	0.366	0.21	37.2	0.389	0.060
82	38				35.6	0.014	11	35.7	0.020	3.4	35.6	0.014	2.0				35.8	0.028	0.12
89	38	37.5	0.840	18	38.3	0.577	9.4	38.6	0.458	2.6	38.5	0.493	0.45	38.6	0.458	0.19	38.3	0.577	0.050
90	38	37.9	0.730	9.9[a]	38.8	0.370	10	39.1	0.263	2.8									
95	40	39.8	0.760	18															
97	40	40.7	0.410	46															
95	41	39.8	0.200	9.5[a]															
96	41	40.3	0.315	12[a]															
99	42	41.6	0.820	32	42.4	0.536	11	42.7	0.410	2.4	42.7	0.410	0.39	42.7	0.410	0.14	42.2	0.617	0.044
109	46	45.7	0.780	30	46.3	0.577	7.1	46.9	0.332	1.0	46.8	0.370	0.17	46.8	0.370	0.056	45.8	0.760	0.037
111	47	46.6	0.820	32	47.1	0.659	6.0	47.6	0.458	1.1	47.7	0.410	0.13	47.7	0.410	0.059			
112	47	47.1	0.396	13	47.6	0.344	4.0	48.1	0.225	0.92	48.1	0.225	0.097	48.2	0.200	0.043			
113	47	47.4	0.536	31	47.9	0.332	5.4	48.5	0.143	0.64	48.5	0.143	0.073	48.5	0.143	0.026			
115[m]	48	48.2	0.389	20	48.7	0.315	3.2	49.3	0.170	0.65	49.4	0.156	0.078	49.3	0.170	0.026	48.1	0.396	0.020
115	48	48.2	0.617	24	48.7	0.410	1.5	49.3	0.198	0.21	49.4	0.170	0.024	49.3	0.198	0.0081	48.1	0.659	0.021
136	55	56.7	0.095	14															

[a] Uncertainties larger than the 30 percent error quoted in text.

were other sources of error in some individual cases. The independent yield of 18-min Br^{80} was not measured because of the time elapsed between the end of the irradiation and the isolation of bromine. The yield of Br^{80} reported here is, then, that of the 4.5-hr Br^{80m} isomer counted via the 18-min Br^{80} in equilibrium with it. The yield of Sr^{82} is measured by attributing the observed radioactivity to the 76-sec Rb^{82} daughter. The yield of Sr^{90} was obtained by isolation and counting of the 65-hr Y^{90} daughter.

IV. DISCUSSION

A. Analysis of Results

The results listed in Table I were analyzed in order to determine the general characteristics of high-energy fission, such as the shape of the fission yield-mass number curve for each target element, the fission cross section as a function of atomic number of the target, and the excitation energy for the fission process.

The data were treated by a contour-mapping method[6,17,18] in which contour lines were drawn connecting points of equal cross section on a proton-neutron plot of the measured nuclides. The curve connecting the vertices of the contour diagrams determines the nuclide having the maximum yield for each mass number and, thus, the most probable neutron to proton ratio for each mass number. The center of the contour diagram represents the most probable fission product, from which the loss in mass and charge of the target nucleus undergoing fission can be determined. Since no distinction was made in this part of the analysis between nuclides whose yields are cumulative and those formed independently, the most probable neutron to proton ratio so obtained does not quite represent primary fission fragments, but lies closer to stability, because of the higher yield of a cumulative product than that corresponding to direct formation. The over-all result of this misrepresentation in these experiments, for the mass range 59 to 115, is an underestimate of the probability of forming neutron excessive fragments, so that the estimate of the mass lost (mostly neutrons) in fission is probably too high. The largest error from this source is made in the case of thorium and will be discussed later.

The values of the most probable charge, Z_p, for the mass numbers studied in the various targets, as determined from the contour diagram analysis, are given in the third, sixth, etc., columns of Table II. The first and second columns give the mass number, A, and

[17] W. J. Worthington, University of California Radiation Laboratory Report, UCRL 1627, January 8, 1952 (unpublished).
[18] P. K. Kofstad, University of California Radiation Laboratory Report, UCRL 2265, June 30, 1953 (unpublished).

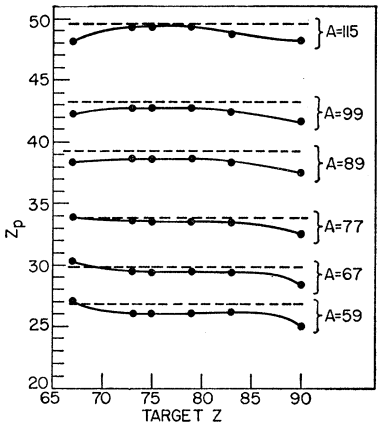

FIG. 2. Values of Z_p for selected mass numbers from $A=59$ to $A=115$ for the target nuclei holmium to thorium. The Z_A values at these mass numbers are also given (dashed lines).

charge, Z, of the nuclide studied. The neutron excess or deficiency of the most probable primary fragment of mass number A is determined by the difference between Z_A and Z_p. Figure 2 presents a plot of Z_p values for some selected masses, and the values[16] of Z_A corresponding to the chosen mass numbers. For thorium, the difference, Z_A-Z_p, is about $+1.5$ charge units, for bismuth, gold, rhenium, and tantalum, about $+0.5$ charge unit, and for holmium about -0.2 charge unit for light masses and $+1.5$ charge units for the heavier masses. Thus, the most probable primary products from thorium to tantalum are all neutron-excessive in the mass-number range 59 to 115, whereas those from holmium are slightly neutron-deficient for light masses, and neutron-excessive for heavy masses.

The total yield for a mass number from a given target is obtained by summing the individual cross sections of all isobaric nuclides, including estimates for stable or undetermined ones. Biller[6] determined cross sections for 63 nuclides from the fission of bismuth with 340-Mev protons and analyzed his data by plotting histograms of yield *vs* charge for isobaric nuclei including interpolated values for unmeasured species. In the work reported here, the spacing of the contour diagrams was not determined accurately enough for a detailed description of the charge distribution, since only some 30 nuclides were measured in all, and of these very few were isobaric. However, it was noted that most of the contour diagrams, as drawn, were consistent with a constant charge distribution, independent of mass number or target nucleus. This analysis of the problem is similar to that used by Glendenin, Coryell, and Edwards[19] for the thermal neutron fission of U^{235}. The charge distribution curve best representing the results of the experiments reported here is one in which the fractional yield for the most probable charge (Z_p) is 0.4, decreasing symmetrically for positive and negative values of $Z-Z_p$ to 0.36, 0.25, 0.134, 0.05, and 0.01 for $|Z-Z_p|$ equal to 0.5, 1.0, 1.5, 2.0, and 2.5, respectively. This charge distribution curve is somewhat broader than that[19] for the thermal neutron fission of U^{235}.

The cross section for a given mass number is calculated from the observed cross section given in Table I by correction for the fractional yield corresponding to the measured nuclide from the assumed yield *vs* charge distribution curve. The data and results are given in Table II where the fraction of yield is the fractional yield according to whether the nuclide studied represents a cumulative or independent product, and the mass yield is the full cross section for the mass number, obtained by dividing the cross section of a nuclide as given in Table I by its fractional yield. A plot of mass yield *vs* mass number for the target elements holmium to thorium is given in Fig. 3. The open symbols represent mass yields for independently formed nuclides and the filled-in symbols mass yield for cumulatively formed nuclides. Although the agreement between the calculated mass yields and the smooth curves drawn in Fig. 3 is by no means ideal, with disagreements in individual cases as high as a factor of 2 or 3, the general shape and

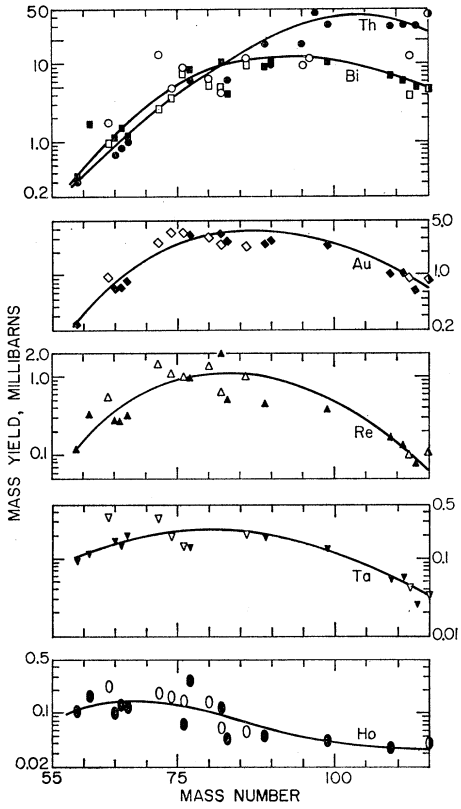

FIG. 3. Mass yield *vs* mass number curves for thorium, bismuth, gold, rhenium, tantalum, and holmium. Filled-in symbols represent yields calculated for cumulative products; open symbols represent yields calculated for independent products. Circles—Th; squares—Bi; diamonds—Au; triangles ▲△—Re; triangles ▼▽—Ta; ovals—Ho. Ordinate values of the yields appear alternately on the left and right.

[19] Glendenin, Coryell, and Edwards, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 52, National Nuclear Energy Series, Plutonium Project Record Div. IV, Vol. 9.

trend of the yield curves are reasonably well defined. The average discrepancies between the points and the smooth curves do not seem excessive in view of the sources of error contributing to the differences, namely, the experimental errors in the cross-section determinations, the errors in the Z_p values chosen from the contour diagrams, and the errors inherent in the choice of a universal charge distribution curve.

The yield curves of Fig. 3 were drawn with maxima at the mass for the most probable mass number as determined from the contour diagrams and given in Table III. If the nuclear process responsible for the formation of the elements in the mass range 60 to 115 is a fission process, then the integral under the symmetrical curve around the peak is equal to twice the total fission cross section. The results of this calculation, as well as summary results of the contour diagram analysis on the "fissioning nucleus," assuming mass degradation prior to fission,[5,6] are given in Table III. The fission cross section is seen to decrease from 0.67 b for thorium to ~0.002 b for holmium. The probable error estimated in the fission cross section is about 30 percent for the elements thorium to tantalum, and could be as high as a factor of 2 for holmium.

Recent radiochemical investigations on the fission of uranium,[20] bismuth,[6] and tantalum[21] with 340-Mev protons have been reported and the results may be compared to those given in Table III. In general, the agreement is good if one makes allowances for the proton energy difference. For example, Biller[6] reports a loss of 2 protons and 18 neutrons in bismuth with 340-Mev protons, compared to 4 protons and 20 neutrons given here, and a cross section of 0.24 b compared to 0.21 b. Nervick and Seaborg,[21] in their proton bombardment of tantalum with 340-Mev protons, report 16 particles lost in fission including 2 or 3 protons, compared to 21 particles including 4 protons. The cross section for fission given by Nervick and Seaborg is 0.0041 b compared to 0.0050 b. The apparent increase in the neutron to proton ratio of the most probable products with increasing charge is perhaps the major difference between the results on bismuth and tantalum of Table III and those of Biller[6] and Nervick and Seaborg,[21] who observed an almost constant neutron to proton ratio. The sharp decline in fission cross section at 450 Mev with decreasing atomic number is similar to that reported by Kelly and Wiegand[3] and Jungerman[4] at lower energy, although the cross section for thorium[4] at 340 Mev is about 40 percent lower than that reported in Table III at 450 Mev, and those for gold and bismuth[4] at 340 Mev are about three-fold lower. The variation of the cross section with energy for gold[4] and bismuth[4,22] is too small to account for the difference in cross section reported at 340 Mev and 450 Mev.

[20] M. Lindner and R. N. Osborne, Phys. Rev. **94**, 1323 (1954).
[21] W. E. Nervick and G. T. Seaborg, Phys. Rev. **97**, 1092 (1955).
[22] L. G. Jodra and N. Sugarman, following paper [Phys. Rev. **99**, 1470 (1955)].

TABLE III. Summary of results of fission analysis.

	$_{90}Th^{232}$	$_{83}Bi^{209}$	$_{79}Au^{197}$	$_{75}Re^{185,7}$	$_{73}Ta^{181}$	$_{67}Ho^{165}$
Most probable fission product	$43.5^{103.5}$	40^{93}	$38^{87.5}$	36^{83}	$35^{80.5}$	32^{72}
Assumed "fissioning nucleus"	$_{87}Fr^{207}$	$_{80}Hg^{186}$	$_{76}Os^{175}$	$_{72}Hf^{166}$	$_{70}Yb^{161}$	$_{64}Gd^{144}$
Target reaction	$p,4p22n$	$p,4p20n$	$p,4p19n$	$p,4p17n$	$p,4p17n$	$p,4p18n$
n/p ratio of "fissioning nucleus"	1.38	1.32	1.30	1.30	1.30	1.25
Most probable n/p ratio at						
$Z=25$	1.36	1.25	1.26	1.26	1.26	1.14
$Z=35$	1.37	1.32	1.30	1.30	1.30	1.29
$Z=45$	1.38	1.35	1.32	1.32	1.32	1.37
Fission cross section, barns	0.67	0.21	0.061	0.019	0.0050	~0.002

B. Discussion of the Fission Process

The fission cross section for holmium represents about one-thousandth of the reaction cross section of holmium with 450-Mev protons, whereas for bismuth the ratio is about one-tenth. Inasmuch as the interaction of the primary proton with either nucleus by the cascade process,[23] followed by evaporation from the residual excited nucleus[24] is not expected to differ too much, the loss in fissionability for the lighter elements must represent poorer competition for the fission reaction compared to the other possible reactions. The fact that the fission reaction in bismuth and the elements of lower atomic number competes more favorably the higher the bombarding particle energy,[3,4,22] for particle energies in excess of 50 Mev, whereas for the fissionable elements uranium and thorium the fission cross section is roughly constant with energy, implies a considerably higher excitation energy to produce fission in the elements of lower atomic number. Goeckermann and Perlman[5] postulated that the fission of bismuth with 190-Mev deuterons occurred after the evaporation of 12 neutrons from the excited Po^{211} nucleus formed from the interaction of Bi^{209} and deuterons. The resultant nucleus, Po^{199}, being of lower neutron-to-proton ratio, is expected to be more fissionable than the original target nucleus. The radiochemical observations on the neutron-to-proton ratio of the fission products, and the need for particle energies far in excess of the calculated fission threshold for bismuth were nicely explained by this hypothesis. The results of energy measurements[25] of the high-energy fission fragments of uranium,

[23] R. Serber, Phys. Rev.**72**, 1114 (1947); M. L. Goldberger, Phys. Rev. **74**, 1269 (1948); Bernardini, Booth, and Lindenbaum, Phys. Rev. **85**, 826 (1952).
[24] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).
[25] J. Jungerman and S. C. Wright, Phys. Rev. **76**, 1112 (1949).

thorium, and bismuth, and of range measurements[26] of the high-energy fission fragments of uranium are in accord with the postulate of fission occurring after mass degradation, but as will be seen later, these experiments do not distinguish well between mass degradation before fission or after fission.

The selection of a fissioning nucleus for each target nucleus may be made from a determination of the most probable fission product found by the contour analysis method. The minimum energy required to produce the fissioning nucleus with 450-Mev protons can be calculated from evaporation theory[27] and calculated masses[28] and is found to be 330 Mev for thorium, 320 Mev for bismuth, 315 Mev for gold, 295 Mev for rhenium, 300 Mev for tantalum, and 340 Mev for holmium. For the lighter elements whose fission excitation functions appear to be steep, at least at energies up to 90 Mev,[3] the choice of one fissioning nucleus is probably not too bad an approximation. However, in the case of thorium, where the fission cross section is almost constant with energy, the contribution to the fission cross section per interaction from collisions involving low excitation energy will be as important as those involving high excitation energy. In this case, the most probable fissioning nucleus should be the one resulting from the most probable energy deposition[29] of about 100 Mev. The case of bismuth should be somewhat beween that of thorium and the lighter elements, in that the cross section from 200 to 450 Mev of bombarding proton energy increases only two-fold.[22] The distortion of the yield pattern of the primary fission fragments, as mentioned earlier in Sec. 4A, results from the contour analysis in which the cumulative and independent yields were plotted on the same diagram and lines of equal cross section were drawn. It is expected that conclusions drawn relative to the fissioning nucleus are most incorrect for thorium, weighted toward neutron deficiency which implies more energy deposited for the average fission, such that the calculated value is 330 Mev compared to the expected value[29] of about 100 Mev, and are probably not too bad an interpretation of the results for the lighter elements.

The probability for fission for a given fissioning nucleus may be related to the difference in energy between the fission threshold, E_f, and the neutron binding energy, E_n. This treatment is similar to that made by Seaborg[30] in the correlation of the relative probability of fission and γ-ray emission in the irradiation of heavy elements with slow neutrons. Fission threshold calculations made from the formula of Hill and Wheeler[31] and neutron binding energies calculated from the Metropolis and Reitwiesner[28] masses are given in Table IV, where it is seen that E_n of the fissioning nucleus does not change appreciably in comparison with the large increase in E_f with decreasing atomic number.

TABLE IV. Comparison of calculated fission thresholds with neutron binding energies.

Target nucleus	Fissioning nucleus	Fission cross-section, barns	Neutron binding energy, E_n, Mev	Fission threshold, E_f, Mev	E_n-E_f
$_{90}Th^{232}$	$_{87}Fr^{207}$	0.67	8.8	3.6	+5.2
$_{83}Bi^{209}$	$_{80}Hg^{186}$	0.21	9.4	6.1	+3.3
$_{79}Au^{197}$	$_{76}Os^{175}$	0.061	8.3	8.7	−0.4
$_{75}Re^{185,7}$	$_{72}Hf^{166}$	0.019	9.6	12.7	−3.1
$_{73}Ta^{181}$	$_{70}Yb^{161}$	0.0050	8.2	14.8	−6.6
$_{67}Ho^{165}$	$_{64}Gd^{144}$	~0.002	10.3	21.2	−10.9

Allowing for a spread in the charge of the fissioning nucleus by one unit, and the associated spread in E_n-E_f from this effect (most of this spread is in E_f), it is found that the fission cross section, σ_f, depends exponentially on E_n-E_f, and is given by the equation

$$\sigma_f=0.07e^{0.41(E_n-E_f)}.$$

The low fission cross section of the lighter elements may then be ascribed to the poor competition between the fission process and neutron evaporation when E_f is much higher than E_n, inasmuch as the energy required to achieve the observed fissioning nucleus is almost constant for all target nuclei, and the energy deposition spectrum is not expected to change much from holmium to bismuth. In the case of tantalum and 340-Mev protons,[21] for example, it was shown that less than 1 percent of the interactions involving energy transfer to the struck nucleus in excess of that necessary to achieve the fissioning nucleus results in fission.

A much steeper dependence of the fission cross section on E_n-E_f is seen in the photofission data of Huizenga, Gindler, and Duffield[32] on U^{238}, Th^{232}, and Np^{237} with 8-Mev photons where the two competing processes are fission and neutron emission. Also, the data on the dependence of the fission cross section of bismuth on proton energy,[22] when analyzed in the same way as the data of this paper, show a steeper dependence on E_n-E_f. This is borne out if one notes that the cross section of gold with 450 Mev protons of 0.061 b is about the same as that for bismuth with 120-Mev protons,[22] whereas the value of E_n-E_f in the two cases is −0.4 Mev and +2.1 Mev, respectively.

An alternate explanation of the high energy results is the one in which fission precedes de-excitation of the target nucleus. None of the conclusions reported here, or from the other radiochemical studies,[6,21] would be altered significantly by this interpretation, nor would the results on the energy[25] or range[26] of fission fragments

[26] E. M. Douthett and D. H. Templeton, Phys. Rev. **94**, 128 (1954).
[27] P. Morrison, *Experimental Nuclear Physics* (John Wiley and Sons, Inc., New York, 1953), Vol. II, p. 173.
[28] N. Metropolis and G. Reitwiesner, "Table of Atomic Masses," U. S. Atomic Energy Commission Report NP-1980, 1950 (unpublished).
[29] McManus, Sharp, and Gellman, Phys. Rev. **93**, 924 (1954).
[30] G. T. Seaborg, Phys. Rev. **88**, 1429 (1952).
[31] D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
[32] Huizenga, Gindler, and Duffield, Phys. Rev. **95**, 1009 (1954).

be less understandable. The term "fissioning nucleus" would now have little significance except as a measure of the overall change in mass and charge of the target nucleus. The energy required to effect the observed change by loss of particles from two fragments would be somewhat less. The competition of fission with de-excitation by other processes would be best at the highest excitation energy before the emission of particles —corresponding to the lowest Z^2/A—and would become progessively poorer as the nucleus lost energy, in contradistinction to the postulate discussed earlier where the fissionability increased as neutrons were lost and the Z^2/A parameter became larger. In either case, the fission products observed would be the sum of all interaction processes in which varying amounts of energy are deposited, and only in those cases of steep fission excitation functions would the fission products be representative of a fairly unique fission energy.

C. Fragmentation, Another Competing Process

The assignment of the radioactive nuclides formed in the mass range 59 to 115 to products of a fission process in which most of the mass of the target nucleus is present in two complementary fragments of the same yield seems secure in those cases where the yield-mass curve shows a pronounced hump at about half the mass of the target nucleus. In this case, the yield-mass curve decreases to a minimum value in the spallation region as the mass number decreases, then rises again sharply because of the onset of contribution from the fission process. The ratio of the peak yield of the hump to the yield at the minimum in the yield-mass curve is about 1000 for uranium and bismuth,[21] so that the differentiation between fission products and spallation products is clear-cut, except where the minimum in the yield-mass curve occurs. In the irradiation of tantalum with 340-Mev protons, Nervick and Seaborg[21] found the ratio of peak to minimum to be about 5, and concluded that this low ratio was more the result of an enhancement of highly asymmetric fission than the overlapping of the spallation and fission curves.

TABLE V. Cross sections of nickel, copper, and molybdenum nuclides from various targets.

	Cross section, millibarns				
Target	Ni^{65}	Ni^{66}	Cu^{64}	Cu^{67}	Mo^{99}
As^{75}	0.37	0.11	13	1.0	
Y^{89}	0.034	0.015	1.2	0.068	
Rh^{103}	0.00078		0.051	0.0028	0.038
$Ag^{107,9}$					0.085
In^{115}				0.00071	0.11
I^{127}	0.00033	0.00025			0.022
Pr^{141}	0.0011	0.00057	0.0041	0.0013	0.0059
Ho^{165}	0.021	0.013	0.087	0.027	0.027
Ta^{181}	0.088	0.048	0.11	0.11	0.056
$Re^{185,7}$	0.14	0.081	0.15	0.18	0.14
Au^{197}	0.32	0.21	0.28	0.44	0.98
Bi^{209}	0.60	0.46	0.29	0.68	5.8
Th^{232}	0.59	0.63	0.11	0.89	27

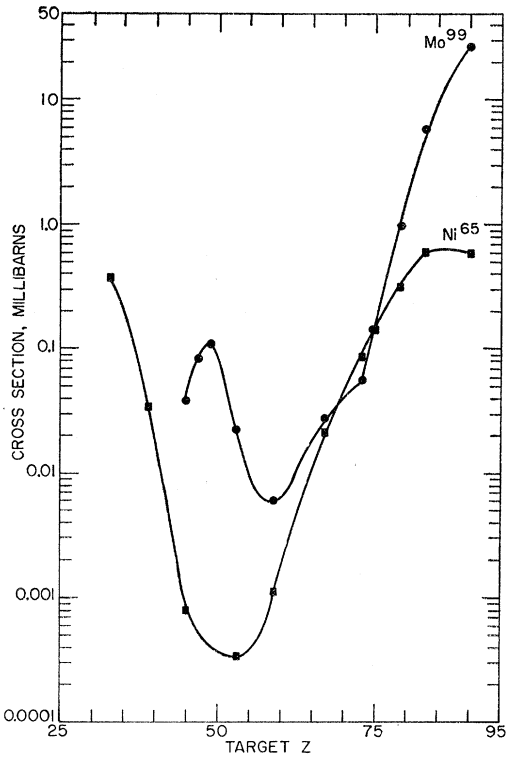

FIG. 4. Cross sections of Ni^{65} and Mo^{99} in targets of atomic number 33 (arsenic) to 90 (thorium). ●, Mo^{99}; ■, Ni^{65}.

The yield curves of Fig. 3, although not covering as complete a mass range as that of tantalum with 340-Mev protons,[21] show the effect of "spilling-over" into the fission-product region on the high-mass side even at gold where the data indicate a not-quite symmetrical yield curve. The curve for holmium shows the onset of the minimum between the spallation and fission regions already at mass 110, whereas in tantalum this minimum occurs at about mass 130; in both cases the mass of the minimum is about 50 mass numbers lower than that of the target nucleus. In order to see if any sharp differentiation could be made between the production of a given nuclide by spallation or fission, several nuclides already studied, Ni^{65}, Ni^{66}, Cu^{64}, Cu^{67}, and Mo^{99}, were also isolated from irradiations of targets of lower atomic number, in which the major production process was expected to change from fission to spallation. Yields of most of the nuclides just mentioned were determined in praseodymium, iodine, indium, silver, rhodium, yttrium, and arsenic. The cross sections for production of these nuclides from all of the targets are given in Table V. A plot of the cross sections of Ni^{65} and Mo^{99}, both neutron-excessive with values of Z_A-Z about $+1$, *vs* the atomic number of the target nucleus is seen in Fig. 4.

It is tempting to identify the portion of the curve to the right of the minimum as resulting from fission, and that to the left from spallation. If this were true, most

TABLE VI. Cd^{115} isomer ratio data from high-energy bombardments.

	U	Th	Bi	Au	Re	Ta	Ho	Pr	I	Sb
$\sigma_{Cd^{115}}$	2[a]	2.0[e]	0.61[e]	0.39[e]	0.35[e]	0.36[e]	1.8[e]	0.48[e]	0.39[e]	0.33[h]
	14[b]	~14[d]	0.75[e]			0.59[g]				
$\sigma_{Cd^{115m}}$			~1.4[f]							

[a] 380-Mev alphas. [P. R. O'Connor and G. T. Seaborg, Phys. Rev. **74**, 1189 (1948)].
[b] Thermal neutrons. [Appendix B, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Div. IV, Vol. 9].
[c] 450-Mev protons. (This paper).
[d] 37.5-Mev alphas. [A. S. Newton, Phys. Rev. **75**, 17 (1949)]; 13- to 21-Mev protons [H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952)]; pile neutrons [A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951)].
[e] 340-Mev protons [W. F. Biller, University of California Radiation Laboratory Report UCRL 2067, December, 1952 (unpublished)].
[f] 190-Mev deuterons. [R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949)].
[g] 340-Mev protons. [W. E. Nervick and G. T. Seaborg, Phys. Rev. **97**, 1092 (1955)].
[h] 50- to 190-Mev deuterons, 380-Mev alphas. [M. Lindner and I. Perlman, Phys. Rev. **78**, 499 (1950)].

of the Mo^{99} found in bombarded holmium should be the result of fission, whereas the yield curve of holmium (see Fig. 3) suggests additional contribution to the yield by processes other than fission even at masses as low as 99. Also, the fact that the Z_p values for holmium are lower at the higher masses than those of tantalum, rhenium, and gold, whereas they are higher at the lower masses (see Fig. 2), again suggests the onset of another production process. The effect of the lowering of Z_p at mass 115 in holmium is great enough to cause the ratio of the Cd^{115} isomers to be almost the same as that found in thorium, indicating some "feed-in" by β decay. The possibility that the anomalous Z_p behavior of holmium might be due to impurity of thorium in the holmium was investigated and it was found that as much as 0.1 percent thorium impurity was necessary to account for the observation, an amount far in excess of the expected thorium impurity. The ratio of Cd^{115} to Cd^{115m} is almost constant at a value of about 0.37 for 450 Mev proton bombardment of gold, rhenium, tantalum, praseodymium, and iodine. This ratio is about 0.33 for high-energy deuteron and alpha-particle bombardment of antimony.[33] A summary of the Cd^{115} ratio data from the work reported in this paper and that of others is given in Table VI.

The observations just noted, which appear anomalous and in need of interpretation, are summarized. (1) High yields are found for the three products isolated in the mass range 99 to 115 from the 450-Mev proton bombardment of holmium. The effect is observed at higher mass numbers in tantalum.[21] (2) The production of these products appears to result from fission, if one interprets the cross section *vs* atomic number of target curve for Mo^{99} (Fig. 4) as composed of spallation and fission contributions. (3) The relation of the most probable charge, Z_p, for the products from holmium to the charge for stability, Z_A, changes from neutron deficiency at low masses to neutron excess at high masses. This effect is seen in Fig. 2 and in the ratio of the cross sections of the Cd^{115} isomers (Table VI). The presence of another process in the high-energy bombardment of heavy elements, important for the production of products between the fission and spallation regions, is indicated.

The proposed process in the heavy elements is the ejection in almost constant yield of fragments[34] in the mass range roughly 10 to 50, with charge values close to the Z_A values for the appropriate mass numbers. The difference between this process and that of "very asymmetric fission," proposed[21] as the explanation of the tantalum data at 340 Mev, is that the fission process produces nuclides in highest yield with an almost constant neutron to proton ratio, whereas the fragmentation process would lead to fragments of low mass close to stability, and products of much higher mass initially neutron excessive. This process corresponds to "intermediate mechanisms between fission and spallation mechanisms" suggested by Kofstad.[18]

Although the cross-section data on low-mass fragments from heavy elements are, at present, very meager, there is some evidence in support of this postulate. The cross sections for low-mass species are highest near stability, as observed in the spallation study of copper,[35] and even in silver,[18] antimony,[33] and tantalum,[21] where the spallation yields near the target are highest for neutron-deficient nuclides. The cross section for formation of Be^7 in gold[36] with 340-Mev protons is ~0.01 mb, those of Na^{24}, Mg^{28}, K^{42}, and K^{43} in tantalum[21] with 340-Mev protons are 0.006, 0.0035, 0.0066, and 0.017 mb, respectively. For lighter elements, such as silver,[18] this rough constancy in cross section no longer obtains. The cross section of Be^7 from silver is about 10 times higher than that in gold, the cross sections of C^{11}, F^{18}, and Na^{24} are all about 0.01 mb, and those of Mg^{28}, Cl^{38}, and Cl^{39} are about 0.001 mb, respectively. The excitation functions for these light fragments are very steep in copper[37] and lead[38] so that somewhat higher yields are expected at 450 Mev than at 340 Mev.

The effect of the emission of fragments near stability in the mass range 10 to 50 with cross sections of ~0.01 mb would be less marked in the heavier, more fissionable elements, like uranium, thorium, and bismuth, than in holmium. In bismuth, for example, the peak cross section in the fission-product region is about 5 mb. Even if fragmentation had the same yield here as in tantalum, the effect would show up at masses of about 150 where the ratio of the cross section to the peak yield

[33] M. Lindner and I. Perlman, Phys. Rev.**78**, 499 (1950).

[34] This interpretation is an outgrowth of discussions held at Brookhaven National Laboratory in 1953–1954 on reactions in heavy elements at the Cosmotron. Participating in the discussions were R. B. Duffield, G. Friedlander, J. Hudis, N. Sugarman, A. Turkevich, and R. Wolfgang.
[35] Batzel, Miller, and Seaborg, Phys. Rev. **84**, 671 (1951).
[36] L. Marquez and I. Perlman, Phys. Rev. **81**, 953 (1951).
[37] Miskel, Perlman, Friedlander, and Miller, Phys. Rev. **98**, 1197(A) (1955).
[38] Baker, Caretto, Cumming, Friedlander, Hudis, and Wolfgang, Phys. Rev. **98**, 242(A) (1955).

in the fission-product region is about 1:1000. In holmium, however, two effects tend to make the fragmentation process more noticeable. First, the fission cross section is only about 0.002 b compared to 0.2 b for bismuth, so that the fragmentation process is now relatively more important. Secondly, the mass number corresponding to the peak fission-product yield from holmium is some 20 mass units closer to the target mass number than in the case of bismuth, the net result being that the heavy end of the fission-product region becomes "crowded" by the low-yield end of the spallation-fragmentation region. Since the fragments are assumed to be produced near stability the residual heavy-mass product is expected to be neutron excessive; the excitation energy left would be insufficient to overshoot stability. This effect explains the apparent anomalous behavior of the Z_p values from holmium, as evidenced by the relatively high ratio of Cd^{115} to Cd^{115m}. For still lighter target elements, such as silver[18] or copper,[39] the fission cross section is lower by a factor of 100 or more, so that a fission hump is no longer evident. This situation is in essence not much different from that which has been observed in the bombardment of bismuth with 2.2-Bev protons[40] where no discernable hump corresponding to the fission-product region is present, presumably because of the high cross section for fragmentation. Values in lead[38] in the Bev range for F^{18}, Na^{24}, Mg^{28}, and P^{32} of $\sim$1 mb have been determined. The residual heavy-mass fragment would be expected to be sufficiently excited to evaporate enough neutrons to lead to neutron deficiency in the observed high-yield products. For example, the neutron-deficient nuclides Ba^{128}, Ba^{129}, and Ba^{131} have yields in bismuth about 100-fold higher at 2.2 Bev[40] than at 340 Mev.[6]

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APPENDIX. SUMMARY OF CHEMICAL SEPARATION PROCEDURES

Iron.—Extractions into isopropyl ether from 9*N* HCl. Washings with 7.5*N* HCl. Scavenging precipitations of Sb_2S_3, CuS, $BaSO_4$, AgI, Nb_2O_5 and zirconium phenylarsonate. Volatilization of ruthenium from fuming $HClO_4$. Repeated $Fe(OH)_3$ precipitations. Final sample Fe_2O_3.

Nickel.—Precipitations of nickel dimethyl glyoxime. Scavenging precipitations of CuS, PdS, Sb_2S_3, $Fe(OH)_3$ and palladium dimethyl glyoxime. Final sample nickel dimethyl glyoxime.

Copper.—Precipitations of CuCNS and CuS. Scavenging precipitations of $Fe(OH)_3$ and $BaCO_3$ from NH_3 solution. Scavenging precipitations of CdS and AgCl. Final sample CuCNS.

Arsenic.—Precipitations of As_2S_3. Extractions of AsI_3 from 3*N* HCl-conc HI solution into chloroform.[41] Final sample As_2S_3.

Bromine.—Extractions of Br_2 into CCl_4. Purifications from iodine by oxidation of iodine with $NaNO_2$ and extraction into CCl_4. Final sample AgBr.

Rubidium.—Precipitations of $RbClO_4$. Scavenging precipitations of $Fe(OH)_3$ and $(Ba+Sr)CO_3$. Cesium separated as the silicowolframate. Final sample Rb_2PtCl_6.

Strontium.—Precipitations of $Sr(NO_3)_2$. Scavenging precipitations of $Fe(OH)_3$. Separation of barium as $BaCrO_4$. Final precipitate $SrC_2O_4 \cdot H_2O$. Sr^{90} determined by addition of yttrium carrier and repeated hydroxide and fluoride precipitations. Final sample $Y_2(C_2O_4)_3 \cdot 10H_2O$.

Zirconium.—Precipitations of $BaZrF_6$. Scavenging precipitations of LaF_3 and $BaSO_4$. Final sample ZrO_2 from ignition of cupferride.

Niobium.—Precipitations of Nb_2O_5. Extractions of cupferride into chloroform. Scavenging precipitation of $BaZrF_6$. Final sample Nb_2O_5.

Molybdenum.—Precipitations of molybdenum alpha-benzoinoxime. Scavenging precipitations of $Fe(OH)_3$ from NH_3 solution. Final sample Ag_2MoO_4.

Palladium.—Precipitations of palladium dimethyl glyoxime. Scavenging precipitations of $Fe(OH)_3$, AgI, and AgCl. Final sample palladium dimethyl glyoxime.

Silver.—Precipitations of AgCl and Ag_2S. Scavenging precipitations of $Fe(OH)_3$ from NH_3 solution. Final sample AgCl.

Cadmium.—Precipitations of CdS. Scavenging precipitations of $Fe(OH)_3$ and $In(OH)_3$ from NH_3 solution. Acid scavengings of PdS, and Sb_2S_3. Final sample $CdNH_4PO_4 \cdot H_2O$.

Cesium.—Precipitations of cesium perchlorate and silicowolframate. Scavenging precipitations of wolframic acid. Final sample $Cs_8SiW_{12}O_{42}$.

[39] R. E. Batzel and G. T. Seaborg, Phys. Rev. **79**, 528 (1950); **82**, 607 (1951).

[40] Sugarman, Duffield, Friedlander, and Miller, Phys. Rev. **95**, 1704 (1954).

[41] R. J. Prestwood, Los Alamos Report LA 1566, February 1, 1953 (unpublished).