High-Energy Fission of Bismuth. Proton Energy Dependence*

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Radiochemical studies of the fission of bismuth with protons of 75 Mev to 450 Mev have been performed. Radioactive products ranging from Cu^{61} to Cs^{131} were isolated and their cross sections were measured. Integration of the calculated cross section *vs* mass number curves for the various energies yielded values for the fission cross section which increase rapidly from a value of 0.016 b at 75 Mev to 0.12 b at 192 Mev, then more slowly to 0.20 b at 450 Mev. The most probable fission products decrease in mass and in neutron to proton ratio as the proton energy increases.

I. INTRODUCTION

DETAILED radiochemical studies of the fission of bismuth have been performed with 190-Mev deuterons[1] and 340 [2]–450 [3] Mev protons. These studies have shown that the cross section for fission at these energies is about one-tenth of the total cross section and that the fission products formed in high yield have a neutron to proton ratio substantially lower than that of the target nucleus. The excitation energy for the process is utilized in emission of particles, most of which are neutrons. Measurements have been made on the kinetic energy of the fission fragments from the bombardment of bismuth with 90-Mev neutrons[4] and the results are in agreement with the mechanism postulated by Goeckermann and Perlman[1] for the fission process, in which the target nucleus is de-excited by neutron emission, and the fission probability rises as the Z^2/A parameter increases. The fission cross section of bismuth was shown to rise steeply with energy[5] for neutrons of 25 to 84 Mev, and to be roughly constant[6] for protons in the energy range 225 to 340 Mev. In experiments on the photofission of bismuth[7] with x-rays of 48 and 85 Mev maximum energy, the steep energy dependence observed with neutrons in this same energy range was also observed. The fission products isolated from the lower energy photofission process were more neutron excessive than those observed with higher energy particles,[1] and the yield-mass curve appeared to be much narrower than at the higher energy.

The work reported here was undertaken in order to study the change in the fission process for a given target nucleus, Bi^{209}, as a function of the bombarding proton energy. Some of the radioactive products of the reaction, ranging from Cu^{61} to Cs^{131}, were isolated by radiochemical methods and their cross sections were measured. The data were then analyzed for information on the energy dependence of the mass degradation in the fission process, the cross section for the fission process, and the most probable charge *vs* mass number dependence of the fission products.

II. EXPERIMENTAL PROCEDURE

Irradiations of bismuth metal foils or powdered samples of bismuth oxychloride wrapped in aluminum were performed on the 2-inch target probe of the University of Chicago synchrocyclotron. The foil irradiation and beam monitoring procedure was essentially the same as that described elsewhere.[3] Irradiation times varied from 20 to 30 min and the nominal proton energy[8] from 75 Mev to 450 Mev. The energy variation was effected by setting the target at the radius corresponding to the desired energy. After the irradiation, the bismuth foil or powder was dissolved and aliquots were taken for radiochemical analysis of the products by standard procedures.[3,9,10]

Seventeen experiments were performed in all, and fourteen elements were isolated radiochemically, although not each of the elements was isolated in every experiment. The results represent an average of at least three samples from the same irradiation, and sometimes an average of two separate experiments. The elements separated were: copper, arsenic, bromine, rubidium, zirconium, niobium, ruthenium, palladium, silver, indium, tin, tellurium, iodine, and cesium. The separated samples were filtered onto a filter-paper disk, mounted on a cardboard card, covered with cellophane, and counted on a Tracerlab TGC1 end-window Geiger-Mueller counter of 2.3 mg/cm² window thickness. The background of the shielded counter was 21 to 25 counts per minute, and the dead-time correction was somewhat less than 1 percent per 1000 counts per minute.

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[1] R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).
[2] W. F. Biller, University of California Radiation Laboratory Report, UCRL 2067, December, 1952 (unpublished).
[3] P. Kruger and N. Sugarman, preceding paper [Phys. Rev. **99**, 1459 (1955)].
[4] J. Jungerman and S. C. Wright, Phys. Rev. **76**, 1112 (1949).
[5] E. L. Kelly and C. Wiegand, Phys. Rev. **73**, 1135 (1948).
[6] J. Jungerman, Phys. Rev. **79**, 632 (1950).
[7] N. Sugarman, Phys. Rev. **79**, 532 (1950); N. Sugarman and R. Peters, Phys. Rev. **81**, 951 (1951).
[8] The average proton energy is probably about 10 percent lower than the values quoted here because of the radial oscillations of the beam [R. C. Koch, Ph.D. thesis, University of Chicago, June, 1955 (unpublished)].
[9] Selected papers of Part VI, "*Radiochemical Studies: The Fission Products*" (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Div. IV, Vol. 9.
[10] W. W. Meinke, University of California Radiation Laboratory Report, UCRL 432, 1949.

TABLE I. Cross sections of fission products of bismuth as a function of proton energy.

	Cross section (in mb) for given proton energy									
Nuclide	75 Mev	120 Mev	184 Mev	192 Mev	242 Mev	303 Mev	355 Mev	373 Mev	427 Mev	450 Mev
Cu^{61}		0.0030							0.029	
Cu^{64}		0.0059	0.044	0.025	0.038	0.067	0.090	0.11	0.14	0.091
Cu^{67}		0.038	0.15	0.14	0.23	0.37	0.39	0.41	0.63	0.38
As^{74}	0.0090	0.041	0.23	0.23	0.45	0.82	0.91	0.69	0.95	1.4
As^{76}		0.24	0.86	0.66	0.91	2.0	1.8	1.3	1.4	2.1
As^{77}	0.084	0.47	1.9	1.5	2.5	4.3	4.0	2.2	4.7	4.8
Br^{80m} [a]			0.76	0.28	1.2	1.3	1.0	1.8	3.0	1.0
Br^{82} [a]	0.0038	0.13	0.93	0.44	1.3	1.4	1.5	1.5	2.4	0.78
Br^{83} [a]	0.039	0.45	1.2	0.77	1.8	2.1	2.1	1.6	3.0	1.1
Br^{84} [a]	0.026	0.19								
Rb^{86}			0.064		0.052				0.20	0.92
Zr^{95}										1.8
Zr^{97}										0.50
Nb^{95m}	0.014	0.079	0.55		0.75	0.86	0.71	0.81		0.83
Nb^{95} [b]	0.019	0.14	1.1		1.2	1.1	2.0	2.7		1.4
Nb^{96}	0.030	0.70	2.8		3.7	5.1	3.3	4.1		4.6
Ru^{103}										3.9
Ru^{106}										1.6
Pd^{100} [c]				(0.020)						(0.0048)
Pd^{103} [c]				(0.012)						(0.0048)
Pd^{109}				0.58						1.0
Pd^{112}				0.14						0.32
Ag^{111}	0.40	1.7	2.9	3.0	3.1	3.1	3.3	3.1	3.0	3.0
Ag^{112}	0.13	0.69	0.99	1.5	1.5	2.7	1.4	1.9	1.3	1.4
Ag^{113}	0.32	1.5	2.1	1.5	2.1	0.91	1.9	1.1	1.4	1.3
In^{111}	0.14			1.2						4.7
In^{114m}	0.0050	0.13	0.32	0.67	2.4	2.4	5.3	4.9	4.5	4.1
In^{115m}	0.14			2.5						
Sn^{113} [c]										(2.7)
Sn^{117m}										3.2
Te^{118} [c]										(2.1)
Te^{121m} [c]										(5.8)
Te^{121}										1.9
I^{124} [d]	0.00034	0.0015	0.0079	0.012	0.020	0.032	0.031	0.031	0.028	0.027
I^{126} [d]	0.0014	0.0027	0.016	0.039	0.041	0.041	0.048	0.050	0.051	0.035
I^{128} [d]	0.0014	0.0025	0.0084	0.024	0.019	0.022	0.025	0.024	0.025	0.018
I^{130} [d]	0.00094	0.0017	0.0048	0.011	0.0083	0.012	0.011	0.014	0.013	0.0089
I^{132} [d]	0.00068	0.00082	0.0031	0.0085	0.0077	0.011	0.012	0.012	0.012	0.0084
Cs^{129} [c]	(0.00032)									(0.030)
Cs^{131} [c]	(0.0026)		(0.0033)			(0.012)			(0.0092)	(0.0037)

[a] The loss of bromine activity upon dissolution of the bismuth foil in HNO_3 was determined to be 67 percent of the total bromine activity in one experiment. This result is similar to that reported in reference 3. The bromine cross sections have been corrected for the loss of activity.
[b] Cross section of Nb^{95} includes formation from Nb^{95m}.
[c] ( ) Lower limit of the cross section because of unknown counting efficiency of the radiations.
[d] Lower limit of the cross section because of unknown loss of radioiodine upon dissolution of bismuth target in HNO_3 (see reference 9).

Half-life determinations and absorption measurements of the radiations for energy determinations were made on all of the radioactivities and the results agreed well with accepted values.[11]

III. RESULTS

The cross sections of the isolated nuclides were calculated from the saturation activities corrected for self- and backscattering[12] of the radiations. The cross section used for the $Al^{27}(p,3pn)Na^{24}$ reaction in the aluminum monitor foils at all energies was 10.8 ± 0.5 mb.[13] The results of the calculated cross sections are

[11] Hollander, Perlman, and Seaborg, "Table of Isotopes," Revs. Modern Phys. **25**, 469 (1953).

[12] Engelkemeir, Seiler, Steinberg, and Winsberg, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 4, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV; Engelkemeir, Seiler, Steinberg, Winsberg, and Novey, NNES Paper No. 5.

[13] The cross section for the $Al^{27}(p,3pn)Na^{24}$ reaction is known to be relatively constant for protons of 90 Mev to 450 Mev energy. Below ~70 Mev the cross section decreases (reference 8).

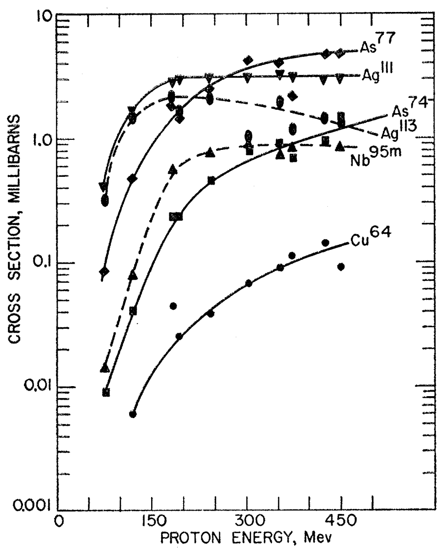

FIG. 1. Cross sections of selected nuclides from fission of bismuth *vs* proton energy. ●, Cu^{64}; ■, As^{74}; ♦, As^{77}; ▲, Nb^{95m}; ▼, Ag^{111}; ovals, Ag^{113}.

given in Table I. For those cases where the β-branching ratio or conversion coefficient is known accurately, it is expected that the cross sections are reliable to about 30 percent. In other cases, where the decay proceeds mainly by way of electron capture, the counting efficiency of the radiations is not well known and the cross section is undetermined. Comments on these specific cases are made in Table I. The uncertainty in the absolute cross sections for these nuclides does not, however, affect any comparison of the change in the ratios of cross sections as a function of the energy of the bombarding proton. The cross sections given for nuclides which are shielded from formation by β decay, e.g., Cu^{64}, As^{74}, As^{76}, etc., or have long-lived parents (Nb^{95}, Ag^{112}, In^{115m}, and I^{132}) are all *independent* cross sections for formation. The cross sections at 355 Mev of Table I, when compared to those of Biller[2] for the same nuclides measured with 340-Mev protons, agree to within a factor of 2 except for As^{77}, Nb^{95m}, Nb^{95}, and In^{114m}. In these cases, the cross sections differ by as much as a factor of 5. The I^{124} and I^{126} cross sections are about 10-fold lower than those of Biller, but as indicated in Table I, the undetermined loss of radioiodine in dissolving the bismuth targets could account for this disagreement. Comparison of the cross sections of the nuclides studied at 427 Mev and 450 Mev of Table I with those of Kruger and Sugarman[3] at 450 Mev again shows agreement to within a factor of 2 except for Cu^{61}, Cu^{64}, Rb^{86}, and Pd^{109}, where the cross sections of Table I for these nuclides are about 3- to 5-fold lower.

The results of Table I show a general trend of increasing cross section with increasing energy, for proton energies below 200 Mev, and except for the lighter nuclides, a much slower increase or even a decrease in cross section at higher energies. In Fig. 1 are plotted the cross sections of some selected nuclides, Cu^{64}, As^{74}, As^{77}, Nb^{95m}, Ag^{111}, and Ag^{113}, *vs* proton energy, in which the different energy dependences are shown. For nuclides whose mass numbers are not too different, such as the isotopes of the same element, the *independent* formation cross sections of the more neutron deficient isotopes generally increase faster with proton energy than the more neutron excessive ones. This effect is similar to that observed in high energy fission of uranium by particles[14] or photons[15] where the cross sections of the neutron excessive species are almost constant, and those of the neutron deficient species increase markedly with energy. In the case of the bromine isotopes, Br^{80m}, the most neutron deficient of the four isotopes studied, is not apparent in the decay curves for protons of 75 and 120 Mev, and appears in high yield at 184 Mev and higher, whereas Br^{84}, the most neutron excessive isotope, has a relatively high

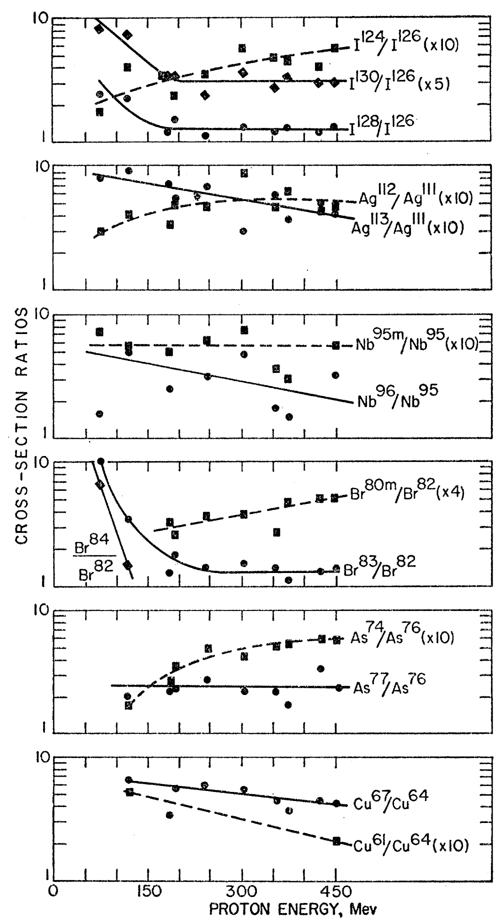

FIG. 2. Ratios of cross sections of isotopes of copper, arsenic, bromine, niobium, silver, and iodine. Isotope of each element chosen for comparison is one close to stability. The label $I^{124}/I^{126}(\times 10)$ means that the cross-section ratio $\sigma(I^{124})/\sigma(I^{126})$ is one-tenth the value read from the scale.

[14] M. Lindner and R. M. Osborne, Phys. Rev. **94**, 1323 (1954).
[15] R. A. Schmitt and N. Sugarman, Phys. Rev. **95**, 1260 (1954).

TABLE II. Summary of results of fission analysis.

	75 Mev	120 Mev	184 Mev	192 Mev	242 Mev	303 Mev	355 Mev	373 Mev	427 Mev	450 Mev
Most probable fission product	41.5^{99}	41.5^{98}	41^{97}	41^{97}	$41^{96.5}$	$40.5^{95.5}$	40.5^{95}	40^{94}	40^{93}	40^{93}
Assumed "fissioning nucleus"	$_{83}Bi^{198}$	$_{83}Bi^{196}$	$_{82}Pb^{194}$	$_{82}Pb^{194}$	$_{82}Pb^{193}$	$_{81}Tl^{191}$	$_{81}Tl^{190}$	$_{80}Hg^{188}$	$_{80}Hg^{186}$	$_{80}Hg^{186}$
Target reaction	$p,p11n$	$p,p13n$	$p,2p14n$	$p,2p14n$	$p,2p15n$	$p,3p16n$	$p,3p17n$	$p,4p18n$	$p,4p20n$	$p,4p20n$
n/p ratio of "fissioning nucleus"	1.385	1.365	1.365	1.365	1.355	1.360	1.345	1.350	1.325	1.325
Most probable n/p ratio at:										
$Z=25$	1.330	1.300	1.300	1.309	1.298	1.269	1.282	1.280	1.256	1.256
$Z=35$	1.360	1.343	1.342	1.341	1.340	1.330	1.335	1.333	1.314	1.315
$Z=45$	1.380	1.368	1.364	1.368	1.362	1.365	1.360	1.364	1.344	1.345
Fission cross section, barns	0.016	0.059	0.11	0.12	0.15	0.16	0.16	0.17	0.19	0.20

yield at 75 and 120 Mev, and is not observed at proton energies greater than 184 Mev. Figure 2 shows the ratios of the cross sections of the isotopes of copper, arsenic, bromine, niobium, silver, and iodine to that of an isotope of each element close to stability. The trends in cross section just mentioned are evident for the isotopes of arsenic, bromine, and iodine, although the opposite trend appears in the ratios Cu^{61}/Cu^{64} and Ag^{112}/Ag^{111}. The Cu^{61} data are the least trustworthy because of the relatively short half-life of Cu^{61} (3.3 hr) and the time elapsed before counting was begun. The Ag^{112}/Ag^{111} ratio, although opposite in trend to the others mentioned, shows the expected behavior for the comparison of the *independent* formation cross section of a nuclide not too far removed from stability, with the *cumulative* formation cross section of a neighboring isotope, where in each case the most probable nuclide of the same mass is more neutron excessive than the observed nuclide. Hence, although the neutron to proton ratio of Ag^{112} is higher than that of Ag^{111}, an increase in the ratio Ag^{112}/Ag^{111} with proton energy is observed.

IV. DISCUSSION

The cross-section data of Table I were subjected to the same analysis as that used by Kruger and Sugarman,[3] as outlined below. The cross sections were plotted on a neutron-proton diagram and contour lines connecting equal cross sections were drawn. Inasmuch as many of the nuclides studied were shielded, the cross sections represent the independent formation of these nuclides, and there is less over-all tendency for the contour diagrams to be displaced toward stability as would occur if most of the nuclides were formed by decay of precursors. The contour diagrams for the 75-Mev and 120-Mev cases appeared rather irregular, probably the result of the poorer accuracy of the data because of the low cross sections at these energies, and, in part, the result of the mixing of cumulative and independent yields on the same contour diagram. The contour diagrams, as drawn, appeared to conform to the same charge distribution curve as that used in the analysis of the fission of the heavy elements holmium to thorium with 450-Mev protons, although the fit was poor for the 75-Mev and 125-Mev cases. The results of the contour analyses on the most probable fission product, the target reaction on the assumption that the particles are emitted before fission, and values of the most probable neutron to proton ratio for a given charge, are given in Table II.

Following are some observations on the analysis of the contour diagrams, the results of which are given in Table II. First, the maximum of the yield-mass curve for the fission products is at lower mass numbers for higher proton energies, consistent with the emission of a larger number of particles, mostly neutrons. Secondly, the most probable neutron to proton ratio at a given charge generally decreases as the proton energy increases, favoring the more neutron deficient isotopes of a given element. Thirdly, at a given proton energy, the most probable neutron to proton ratio increases as the charge increases, as was observed by Kruger and Sugarman.[3] The "fissioning nucleus" at each energy may be obtained by doubling the mass and charge of the most probable fission product. The fissioning nucleus found at 355 Mev is Tl^{190}, compared to Pb^{190} as the most probable fissioning nucleus reported by Biller[2] for 340-Mev protons, and that found at 450 Mev is Hg^{186}, identical with other experiments.[3]

The emission of the surprisingly large number of particles at 75 Mev and 120 Mev, as determined from the contour analysis, is inconsistent with the available energy, in that minimum energies in excess of that available are required. This erroneous observation is probably the result of the poor data at these energies and the shifting of the contour plots toward stability because of the presence of some cumulative cross sections, producing an apparently lower most probable neutron to proton ratio which is correlated with the emission of a larger number of neutrons.

The full cross section for a given mass number may be calculated from the measured cross section by estimating the fractional yield of the nuclide studied from the fractional yield *vs* charge dependence given elsewhere.[3] The results on nuclides of high-energy β radiation and of established decay schemes, such as Cu^{64}, Cu^{67}, As^{74}, etc., establish cross section *vs* mass number curves at the various proton energies, such that the integral under the curve is twice the fission cross section. In the cases, such as iodine, where it was expected that there would be a loss of radioactivity from the solution in dissolving the target because of

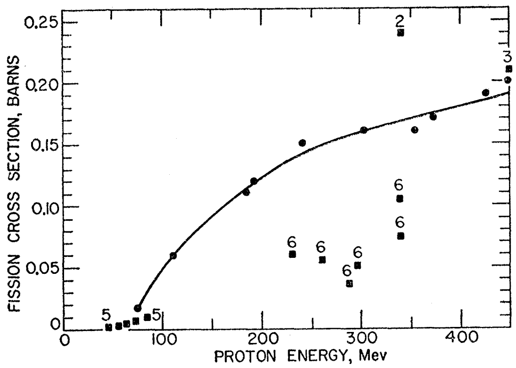

FIG. 3. Fission cross section of bismuth *vs* proton energy. ●, results of this work (Table II); ■, results of other workers. 2: Biller, reference 2; 3: Kruger and Sugarman, reference 3; 5: Kelly and Wiegand, reference 5; Relative data normalized to fission cross section of thorium of 0.5 b with 84-Mev neutrons); 6: Jungerman, reference 6.

the volatility of the element, the cross sections were uniformly low. In other cases, where the β-ray energy was low, e.g., Nb^{95}, or where the decay scheme is not too well established, e.g., In^{114m}, the cross sections staggered relative to the smooth curves.

The fission cross sections of bismuth for the various proton energies studied, as determined by integration of the cross section *vs* mass number curves, are given in Table II. Because of the many sources of error in these determinations, it is expected that the fission cross sections may be in error by as much as 30 percent. The values at 75 Mev and 120 Mev are somewhat more uncertain. The cross section increases by about a factor of 6 from 75 Mev to 200 Mev, then by about a factor of 2 to 450 Mev. The increase in fission cross section with proton energy is the major effect responsible for the shape of the curves of Fig. 1. In the case of Ag^{111}, the ratio of the cross section of Ag^{111} to the fission cross section of bismuth is almost constant at a value of 0.025 from 75 Mev to 192 Mev, then decreases slowly to 0.015 at 450 Mev. Since Ag^{111} is a cumulative product, the change with energy in the neutron to proton ratio of the most probable product at a given mass number does not affect the cumulative yield as long as the most probable products are neutron excessive, 2 or 3 charge units from stability. As the energy is increased, the decrease in the most probable mass and in the neutron to proton ratio both tend to make the yield of Ag^{111} in fission lower. The shapes of the other curves given in Fig. 1 are different from that of Ag^{111} inasmuch as the yields of the other nuclides are affected differently by the changes in mass and neutron to proton ratio with energy.

The fission cross sections *vs* proton energy are plotted in Fig. 3 (circles). The low-energy neutron fission cross sections of Kelly and Wiegand,[5] normalized to a fission cross section for thorium of 0.5 b with 84-Mev neutrons, the high-energy proton data of Jungerman,[6] and the cross sections found at 340 Mev by Biller[2] and at 450 Mev by Kruger and Sugarman[3] by methods similar to those described here, are also given in Fig. 3 (squares). The cross sections determined by ionization chamber techniques[5,6] are uniformly low relative to those determined by the radiochemical analytical method.[2,3] This discrepancy in the fission cross section measured by the two methods has already been pointed out for thorium, bismuth, and gold.[3]

The increase in the fissionability of bismuth with proton energy results from an increase in energy deposition in the target nucleus.[16] For proton energies in the range 75 Mev to 200 Mev, where nuclear transparency is not too important, the fission cross section rises steeply with proton energy, either because of better competition of the fission process with other energy dissipating processes, e.g., particle evaporation, or because of the formation of a more fissionable nucleus after particle evaporation. In the energy range 200 to 450 Mev, the most probable energy imparted to the target nucleus becomes a smaller fraction of the particle energy (about 100 Mev) and does not change appreciably with particle energy. In this energy range, the fission cross section rises more slowly with proton energy, a result of the slow enhancement of the high-energy component of the energy deposition spectrum. The most probable fission product, as determined from the contour analysis, defines the "fissioning nucleus," whose values at the various proton energies are given in Table II. Where the excitation function for fission is steep, e.g., in the energy range 75 to 200 Mev, the fissioning nucleus is fairly unique and is a good parameter for the fission process. However, in the energy range where the fission cross section varies only slightly with energy, the fissioning nucleus determined from the contour analysis is an average of all the fissioning nuclei whose individual contributions are difficult to determine.

V. ACKNOWLEDGMENTS

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