RARE-EARTH FISSION PRODUCTS IN URANIUM FISSION INDUCED BY 660-Mev PROTONS

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Data on fission products in the rare-earth region for uranium bombarded by 660-Mev protons have been obtained by a radiochemical method. The total rare-earth yield was 0.7×10^{-24} cm^2, corresponding to approximately 20% of the total fragment yield. In the case of highly asymmetric fission the yields of stable nuclides and nuclides with neutron excess or deficiency are found to be approximately the same. The maximum yield in the light rare earths is for stable nuclides whereas the heavy rare-earth fission products are predominantly neutron-deficient. It is shown that the distribution of rare-earth elements found in nature cannot be explained by fission of heavy nuclei by high-energy particles.

IN an earlier work[1] we have indicated that the rare-earth region is a very convenient one for investigations designed to delineate the role of nuclear reactions in the creation of the elements. The importance of establishing basic trends in the production of rare-earth elements has been pointed out several times by A. P. Vinogradov in connection with some fundamental geochemical problems. For these reasons the study of uranium fission, which is a basic nuclear reaction in the heavy-element region, can not be complete without a knowledge of the yield of the rare-earth elements, which comprise a considerable fraction of the fission products. The yields of these nuclides can then be used to investigate the nature of highly asymmetric fission, a phenomenon which has not been studied to any great extent at the present time.

The rare-earth elements, furthermore lie in an interesting nuclear region in which shell-structure effects become important. It is well-known that there is a marked change in nuclear properties in the rare-earth region: for example, the excitation energy of the first collective level, the quadrupole moment, the isotopic shift of spectral lines, nuclear deformation, and so on. Some of the first members of this group have a shell containing 82 neutrons and gadolinium has a clearly defined sub-shell of 64 protons. All these factors provide motivation for examining the effects of nuclear structure on the fission process.

The determination of the yields of radioactive isotopes of the rare-earths formed in uranium fission induced by high-energy protons involves the solution of two basic problems in method: the effective separation of the elements of this group and a determination of the yields of nuclides which decay by K-capture. The problem of separating the elements has been discussed in Ref. 1; in the present paper main emphasis has been given to the development of a method for determining the yields of the nuclides indicated above.

METHOD OF INVESTIGATION

A target of spectrally pure metallic uranium, 0.5 to 1 gram in weight, was first irradiated in

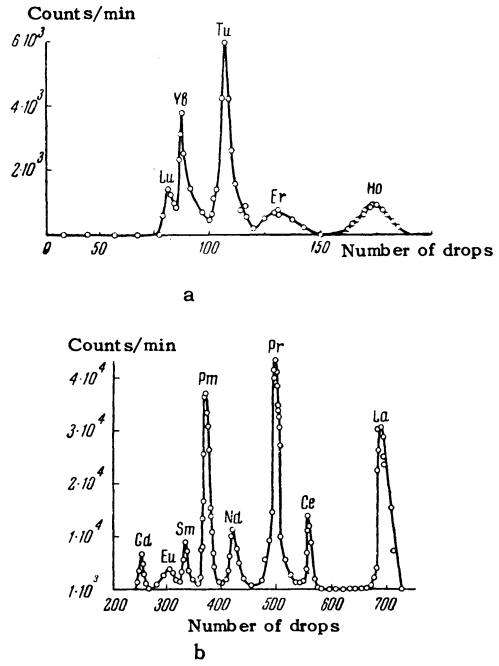

FIG. 1. Separation of rare-earth elements by a 3.6 percent solution of ammonium lactate (pH = 3.4 to 3.5): a – heavy rare earths, b – light rare earths.

TABLE I

Isotope	Decay mode	Observed half-life	Observed energy		Production cross section 10^{-27} cm²
			β-radiation, Mev	γ-radiation, kev	
$_{57}$La 140	β⁻,γ	38.5 hours	1.418	110; 165; 350; 480; 875; 1590	13.3
$_{58}$Ce 134	K	3.5 days		510	0.6
139	K,γ	145 „			
141	β⁻,γ	29 „	0.437	145	
143	β⁻,γ	35 hours	1.2	38; 62; 114; 305; 635; 755	17.3
145	β⁻,γ	2.5 „			
$_{59}$Pr 137	β+	1.4 „			
139	K,β+,γ	4.2 „			
142	β⁻,γ	20 „			6.3
143	β⁻	13.5 days	1.0		24.3
145	β⁻	4.5 hours			
$_{60}$Nd 139	K,β+,γ	5.75 „	2.3		1.3
140	K	3.4 days			3
147	β⁻,γ	10.5 „	0.9	39; 90; 190; 263; 310; 410; 520; 730	4.9
$_{61}$Pm 148	β⁻,γ	42.5 „			5.6
149	β⁻,γ	54 hours	0.92		14
150	β⁻,γ	4 „			
151	β⁻,γ	26.5 „	0.92	41; 65; 100; 135; 160; 175; 230; 290; 350—385	8.3
$_{62}$Sm 145	K,γ	113.3 days			
153	β⁻,γ	48 hours	0.6	40; 70; 100; 155	6
156	β⁻	10 „			2.6
$_{63}$Eu 146	K	36 „		35; 155; 237	0.36
148	K,γ	50.5 days			
152	K,β⁻,γ	9.5 hours	1.65		3
156	β⁻,γ	14.6 days	0.59		2
157	β⁻,γ	15.4 „			
$_{64}$Gd 149	α,K(?)	10.3 „			0.9
159	β⁻,γ	17 hours			0.075
$_{67}$Ho 161	K,γ	3.5 „			
162	K,β⁻,γ	65 days			
163	K,γ	7.5 „			
166	β⁻,γ	28 hours	1.62; 0.3		0.6
$_{68}$Er 160	K	28 „		47; 88; 136; 180—190; 1500	0.1
169	β⁻	9.5 days	0.35		0.4
171	β⁻,γ	7 hours			0.012
$_{69}$Tu 165	K,γ	33 „			0.3
166	K,γ	8 „			1.1
167	K,γ	9.3 days			1.6
170	β⁻,γ	140 „	0.66		0.09
$_{70}$Yb 166	K	36 hours		52—55; 84; 115; 185	0.5
169	K,γ	28.5 days			0.1
175	β⁻,γ	106 hours			0.02
$_{71}$Lu 170	K,γ	1.75 days			0.4
172	K,γ	6.75 „	1.2		0.13
174	K,β⁻,γ	175 „			
176 m	β⁻,γ	3 hours			0.01
177	β⁻,γ	6.75 days	0.5		0.02

the internal 660-Mev proton beam at the synchrocyclotron of the Laboratory for Nuclear Problems of the Joint Institute for Nuclear Research for 1 to 1.5 hours and then dissolved in several milliliters of concentrated hydrochloric acid containing 10 to 20 mg of cerium and hydrogen peroxide. The solution was passed through a column (0.8 × 10 cm) of Dowex — 1 — X8 in equilibrium with concentrated hydrochloric acid and the resin was washed by double the volume of this acid. The solution from the column was diluted to 0.5 to 3 N in HCl and again passed through the column with the anionoid in equilibrium with hydrochloric acid of corresponding concentration. The cerium hydroxide was then precipitated by ammonia and the precipitate dissolved in concentrated nitric acid; 1 to 2 mg of zirconium in the form of the oxychloride was added and the zirconium iodate was precipitated for sep-

aration of the thorium radioisotopes. The cerium hydroxide and zirconium iodate were precipitated several times. Double precipitation of the hydroxide and oxalate of cerium from the filtrate was carried out. The last precipitate of the hydroxide was dissolved in hydrochloric acid and from a 0.3 N hydrochloric acid solution the absorption of the rare-earth elements was carried out in Dowex -50 in NH_4 form. The cationoid was carried to the upper part of the column (25×0.2) cm, filled with the cationoid in equilibrium with the eluent. The elution was carried out with a 3.6 percent solution of ammonium lactate (pH = 3.4 to 3.5) at the rate of one drop per minute (approximately 0.03 ml/min) at 75 to 80°C. Each drop was deposited on a thin sheet of tracing cloth, dried under a lamp, and then measured for activity. In Fig. 1 are shown the elution curves for radioisotopes of the rare-earths formed in uranium fission induced by 660-Mev protons. The yttrium peak is not shown on the figure.

Each peak of this chromatogram was identified individually by half-life of each form of radiation (β^-, β^+, γ and x-ray) and by energy of the β- and γ-radiations. The measurements were carried out with both a magnetic analyzer with two end counters and with an ordinary apparatus with an end tube. The energy of the β-radiation was determined by absorption in aluminum while the energy of the γ-radiation was determined with a luminescent γ-spectrometer. The identification of the isotopes by γ-radiation consisted of taking the γ-spectra for each peak of the chromatogram and determining the half-life for the individual lines of the spectrum.[2]

DETERMINATION OF YIELDS OF RARE-EARTH NUCLIDES

The determination of the yields of β^-- and β^+-active nuclides was carried out by the method described earlier.[3] The determination of the yields of nuclides which decay by means of K-capture, however, was difficult because of the absence of any reliable method. Several methods are described in the literature. Murin and his colleagues[4,5] have used a method described by Wilkinson[6] for determining the absolute yields of K-capture radioisotopes of the heavy elements; this method is based on measurements of the different radiations using an argon counter and successive absorption in aluminum, beryllium, and lead foils. The counting was carried out using the x-ray K-radiation at low efficiency (0.5 to 1 percent).

In measuring the yields of the K-capture radioisotopes of elements of intermediate atomic weight, Mekhedov and Kurchatov[7] have used a magnetic analyzer with two end counters. The x-ray radiation and the γ-radiation were recorded by a krypton-filled end counter which had a high counting efficiency for the x-ray K-radiation for nuclei of intermediate atomic weight (50 to 70%). A shortcoming of both methods is the fact that it is necessary to take account of the decay scheme, the ratio of L/K captures, and soft γ-radiation; furthermore, the geometry is poor in the second method.

Recently, Malysheva[8] has proposed a method for measuring the K-capture decay isotopes of mercury, bismuth, and gold formed in the disintegration of bismuth by 660-Mev protons. This method makes use of the secondary x-ray L-radiation, using a standard argon counter. In this case the detection efficiency for the L-radiation of the heavy elements is 20% while the efficiency for the K-radiation is 0.5 to 1%. The latter can be neglected and account need be taken of K-capture only in the secondary L-radiation (with an error of less than 10%). The advantage of this method is the high counting efficiency for the L-radiation as compared with K-radiation and the fact that

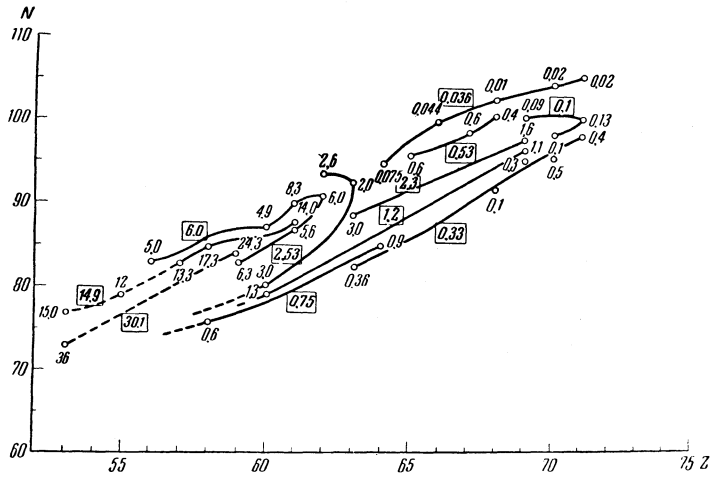

FIG. 2. Yields of radioisotopes of the rare earths.

there is no need to take account of soft γ-radiation nor of the exact ratio of L/K captures. The last factor is especially important since this quantity is either completely unknown or not known reliably in the majority of neutron-deficient nuclides.

The first method was found to be poorly suited to the present work so that the second was used. For this purpose we have determined the counting efficiency for x-ray K- and L-radiation for the rare-earth elements. The appropriate calculations show that the counting efficiency for K-radiation is 1% for cerium, less than 1% for lutetium and 95 and 55% respectively for L-radiation in these elements. In the remaining rare-earth elements the efficiency lies between these values. The magnitudes which have been obtained indicate a very high counting efficiency for L-radiation in the rare-earth elements and consequently indicate the convenience of the method described above for determining the yields of K-capture isotopes of these elements.

In the activity measurements made with the ordinary end counter corrections were introduced for the following factors.

1. Absorption of L-radiation in the counter window, in the air gap, and in the sample covering (16% for cerium and 22% for lutetium).

2. Absorption of the L-radiation in the ineffective counter volume which was (20% for cerium and 6% for lutetium).

3. L-fluorescent yield (using the data reported by Burhop[9]).

In addition, using the known decay schemes a correction was introduced for the fraction of x-ray K-radiation due to electronic conversion. The error in the determination of the yields was 40 to 50% for the known decay schemes and less than 100% for the unknown decay schemes.

DISCUSSION OF THE RESULTS

The yields obtained for the radioisotopes of the rare-earths are shown in Table I. Using a method of interpolation and extrapolation which has been described in detail in Refs. 10 to 12, the yields of a large number of stable and unidentified radioactive isotopes were plotted on an isotope chart in N — Z coordinates (Fig. 2). Using the experimental and interpolated data, distribution curves were plotted for the yield of the various elements as a function of mass number; these are shown in Fig. 3. These curves, which are more or less dome-shaped, make it possible to extrapolate the yields of the other isotopes of the rare-earth elements. The yields of the isotopes of dysprosium and terbium, which could not be separated in a pure radioactive state, were determined in this way. This situation arises because of the fact that these two elements are eluted after yttrium, which has a very large yield, and the yttrium peak on the chromatogram tends to mask the dysoprosium and holmium peaks. The superposition of peaks, in separating various amounts of rare-earth elements, has been noted earlier.[13,14]

From the complete pattern of experimental and interpolated data it is possible to obtain a complete picture of the rare-earth fission products for uranium fissioned by 660-Mev protons and to estimate the fraction of stable nuclides and nuclides with neutron deficiency and neutron excess.

It follows from Table II that the yields for all three types of nuclides are approximately the same. Thus stable nuclides comprise 38.6%, the neutron-excess nuclides 36.6%, and the neutron-deficient nuclides 24.8% of the total rare-earth yield. However there is a marked dependence of the yield of these various types of nuclides on atomic number. For example, yields of stable nuclides and neutron-excess nuclides are reduced as Z increases whereas the yield of neutron-deficient isotopes increases as Z is increased. The dependence on atomic number is also observed in the position of maximum-yield nuclides with respect to the line of nuclear stability drawn through nuclides of greatest abundance in nature (solid line in Fig. 4). It is apparent from this figure that a line drawn through nuclides of maximum yield (dashed line) tends to move in the direction of neutron-deficient nuclei as the atomic number of the rare-earth element increases.

The data in Table II indicate a significant reduction in the total yield of rare-earth nuclides with increasing Z. The nature of this dependence is shown in the curve in Fig. 5, from which it is apparent that the yield of lutetium isotopes (Z = 71) is 150 times smaller than the yield of cerium isotopes (Z = 58). The smooth behavior of this curve is disturbed beyond gadolinium (Z = 64); this effect may be interpreted as an indication of the clearly defined sub-shell of 64 protons.

The total yield of rare-earth nuclides is 0.7×10^{-24} cm^2, about 20% of the yields of all fission products of uranium if it is assumed that the total fission cross section for uranium is 1.65×10^{-24} cm^2 (Ref. 11) and that this cross section does not change greatly as the energy is increased from 480 Mev to 660 Mev. The ratio of these quantities can be used to estimate the fraction of asymmetric fission which leads to the formation of elements with Z = 30 — 37 and Z = 57 — 71 (40% of the

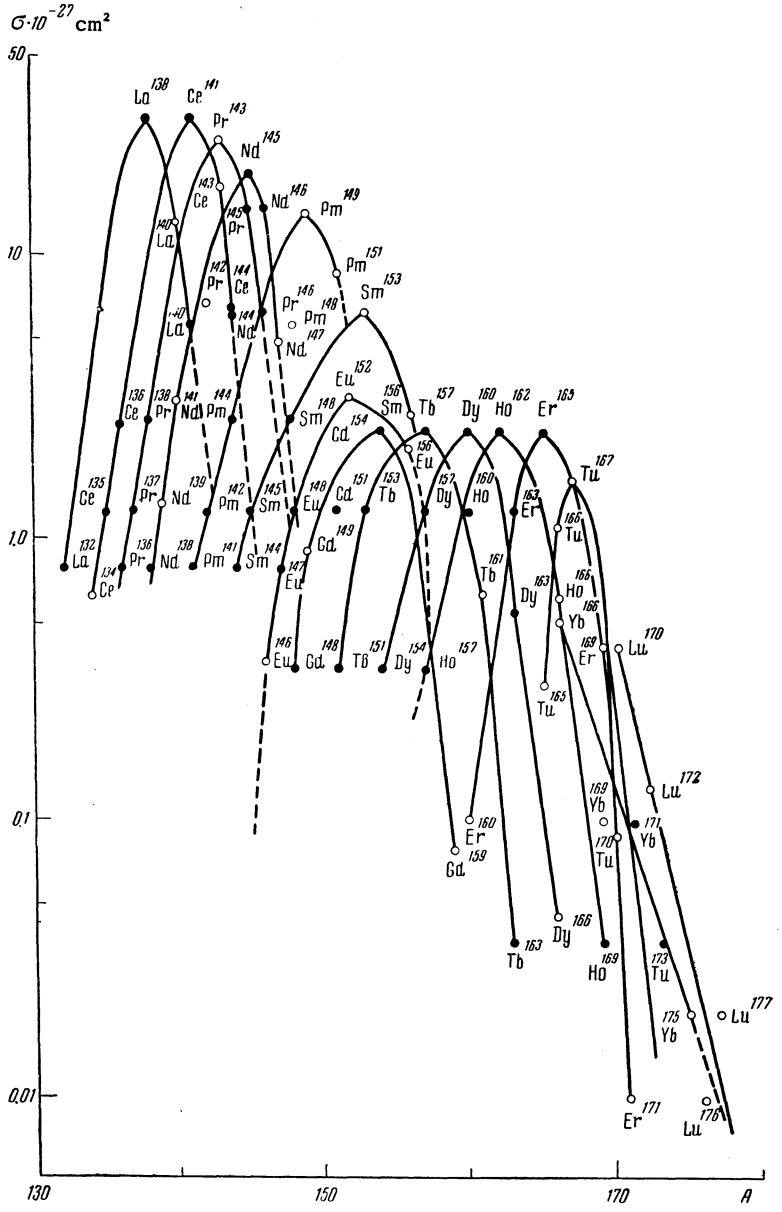

FIG. 3. Yields of rare-earth isotopes as a function of mass number. ● – interpolated data. ○ – experimental data.

TABLE II

Z	Element	Total isotopic yield, 10^{-27} cm²	Stable isotope yield		Yield of neutron-excess isotopes		Yield of neutron-deficient isotopes	
			10^{-27} cm²	%	10^{-27} cm²	%	10^{-27} cm²	%
57	La	131.1	53.1	40.5	21.2	16.2	56.8	43.3
58	Ce	137.5	59.5	43.3	55.9	40.6	22.1	16.0
59	Pr	107.0	15.5	14.5	71.5	66.7	20.0	18.8
60	Nd	88.0	72.2	82.0	5.5	6.3	10.2	11.7
61	Pm	65.3	—	—	54.9	84.0	10.4	16.0
62	Sm	44.0	23.7	53.8	17.6	40.0	2.7	6.2
63	Eu	23.2	5.7	24.5	13.3	57.5	4.2	18.0
64	Gd	14.3	8.4	58.9	0.08	0.5	5.8	40.6
65	Tb	15.3	1.5	9.8	1.8	11.8	12.0	78.4
66	Dy	13.0	8.7	67.0	0.2	1.4	4.1	31.6
67	Ho	13.0	1.1	8.5	2.6	20.0	9.3	71.5
68	Er	11.6	7.3	62.8	0.4	3.4	3.9	33.8
69	Tu	4.9	0.6	12.0	0.2	4.1	4.1	83.9
70	Yb	1.6	0.6	37.3	0.03	1.9	1.0	60.8
71	Lu	0.9	0.03	3.3	0.03	3.3	0.8	93.4
Total		670.7	257.9	38.5	245.3	36.5	167.6	25.0

total fission cross section for uranium).

The data on yields for all isotopes of the rare-earth elements can also be used to establish the distribution of isotopes over mass number (Fig. 3) and the distributions of isobars over atomic number (Fig. 6). These distributions are of the nature of identical dome-shaped curves for all elements, excluding the heavy elements, in which all nuclides are in the right-hand branch. In the distribution of isobars over Z there are noticeable deviations in the region Z = 64. This gives an indication of the role of shell structure in the fission process.

The information on yields for all rare-earth nuclides supplements the picture of uranium fission given earlier[11] and gives a detailed picture of the heavy fragments of highly asymmetric fission. Among these, neutron-deficient nuclides comprise a large fraction. This is in contrast with the light fission fragments, for which there is a clear predominance of neutron-excess nuclei. The quantity Z_p (the most probable charge for a given mass number) found from the isotope chart (Fig. 2), just as in Ref. 11, departs significantly from the line of nuclear stability and tends to be on the neutron-deficient side. This departure increases with increasing mass number (Fig. 7). On this figure the line of nuclear stability is characterized by a dependence of the quantity Z_A (the most probable charge of the stable nuclei) on mass number.[15] In light fission fragments[11] Z_p lies in the immediate vicinity of the line of nuclear stability. It should be noted that in uranium fission by slow neutrons the quantity $Z_p(n)$ lies in the region of high neutron excess.[15] The ratio n/p for heavy fragments is 1.3 to 1.51 whereas it is much lower for light fragments (1.14 to 1.4). The maximum is found for a nuclide with ratio n/p = 1.42 to 1.47. The considerable difference in the magnitude of n/p in the heavy fragments and light fragments indicates that uranium fission does not involve an emission mechanism since this mechanism is characterized by the same value of n/p for all fission fragments.

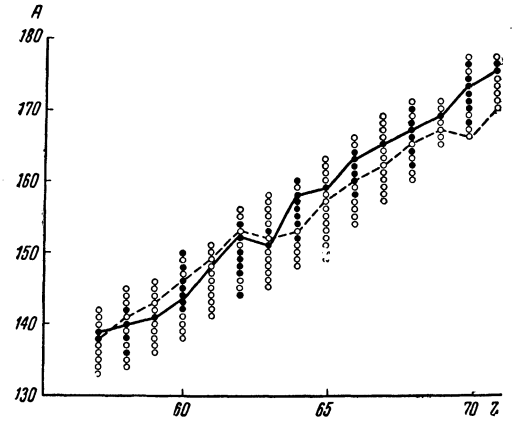

FIG. 4. Products of uranium fission in the rare-earth region. ● – stable isotopes, o – radioactive isotopes.

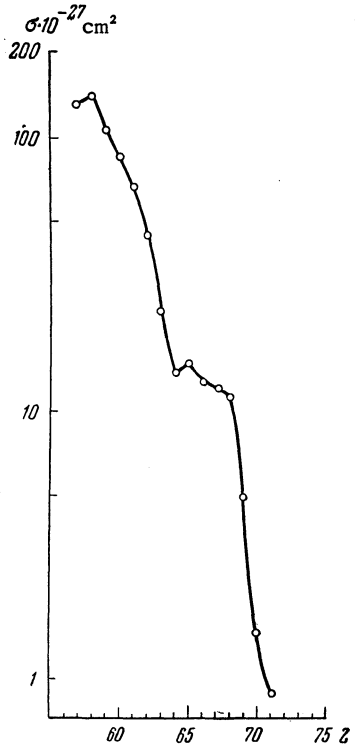

FIG. 5

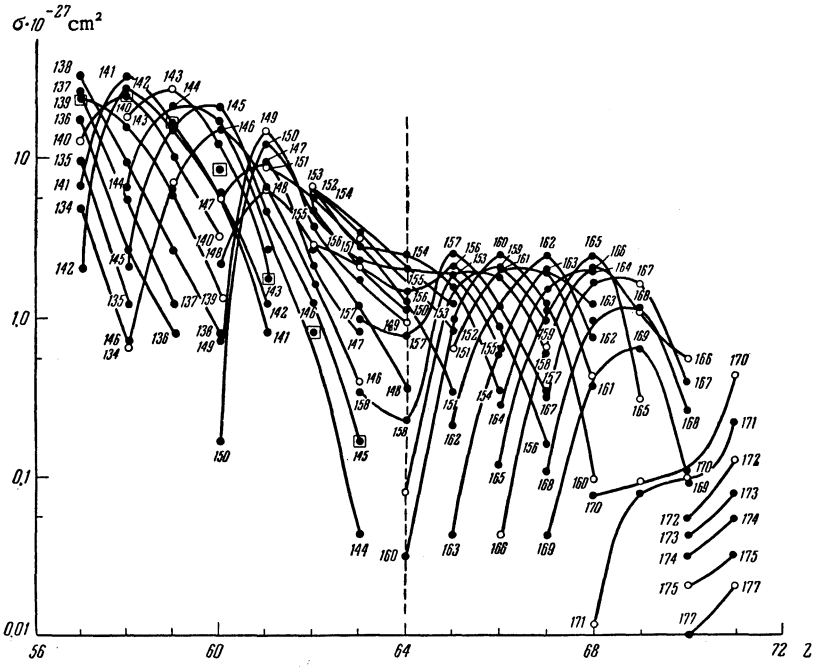

FIG. 6

FIG. 5. Total yields for isotopes of the rare earths as a function of atomic number.
FIG. 6. Distribution of isobars as a function of atomic number. ● – interpolated data, o – experimental data.

EFFECT OF NUCLEAR STRUCTURE

At the present time the effect of shell structure on the slow-neutron process in heavy nuclei is more or less established. The asymmetric nature of slow-neutron fission in these nuclei is considered by many authors[16,17] to be the result of the shell structure of the fissioning nucleus.

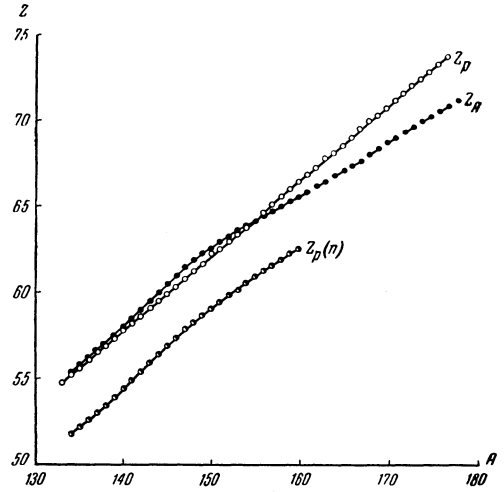

FIG. 7. The quantities Z_A and Z_p as functions of mass number in the rare-earth region for uranium fission induced by 660-Mev protons.

The fine structure in the yield curve for fission products of U^{233}, U^{235}, U^{238}, and Pu^{239} close to mass 133, 100, 84 and others,[18-25] which is observed by mass-spectroscope and radiochemical techniques, may be also be basically attributed to shell structure effects in nuclei with 50 and 82 neutrons. This fine structure is not observed at high bombarding-particle energies, however. This is an indication that the excitation energy of the fissioning nucleus is high. Thus the shell structure of the nucleus becomes apparent only in the last stage of the evaporation process, when the excitation of the nucleus has become small.

A consideration of the yield distribution curves for isobars as a function of atomic number, shown in Fig. 6, shows a marked departure in the behavior of these curves in the region of gadolinium ($Z = 64$). There is a marked reduction in yield for isobars containing 64 protons, corresponding to the filling of a sub-shell.[26] In isobars having a closed neutron shell at $N = 82$ there is also a very marked reduction in yield as the sub-shell of 64 protons is approached (in Fig. 6 isobars with $N = 82$ are enclosed in squares). In general these isotopes are not observed in gadolinium. In Fig. 5, as has already been indicated, there is a sharp break in the behavior of the distribution curve for the total yields of the rare-earth elements in the vicinity of gadolinium ($Z = 64$).

The reduction in yield which is observed for nuclei having a closed sub-shell can be explained from the point of view of the statistical model. It is well-known that nuclei with closed sub-shells have an abnormally low level density. In Ref. 27 it has been shown that in the evaporation process the yield of nuclei with low-level densities is considerably smaller than those with high-level densities.

The fact that uranium fission induced by 660-Mev protons exhibits nuclear shell structure effects, as in the neutron evaporation process, is evidence of the evaporation of neutrons from excited fragments. This finding tends to support the viewpoint that uranium fission is due to a barrier mechanism.

ORIGIN OF THE RARE-EARTH ELEMENTS

Any explanation of the origin of the elements must provide an understanding of the nuclear abundances found in nature. To delineate the role of the fission process in the formation of the rare-earth elements we have compared the natural abundances of stable isotopes of these elements

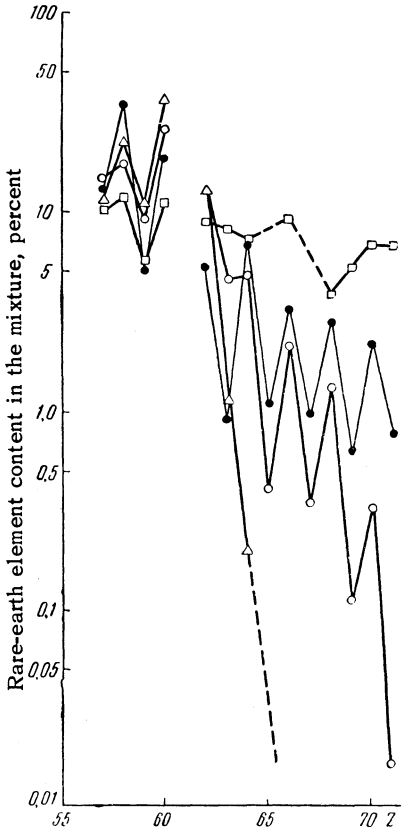

FIG. 8. Rare-earth content as a function of atomic number: ● – in the earth's crust, □ – in the sun and in certain stars, △ – in products of thermal-neutron fission of U^{235}, ○ – in products of fission of natural uranium by 660-Mev protons.

TABLE III

Element	Mass number	Content, percent: In the earth's crust[28]	Content, percent: In fission products
$_{57}$La	138	0.089	34.1
	139	99.911	65.9
$_{58}$Ce	136	0.193	2.5
	138	0.25	12.1
	140	88.48	55.9
	142	11.07	29.5
$_{59}$Pr	141	100.0	100.0
$_{60}$Nd	142	27.13	8.6
	143	12.2	30.7
	144	23.87	24.65
	145	8.3	22.9
	146	17.18	11.9
	148	5.72	1.16
	150	5.6	0.09
$_{62}$Sm	144	3.16	1.05
	147	15.07	22.2
	148	11.27	12.4
	149	13.84	27.35
	150	7.47	21.5
	152	26.63	8.4
	154	22.53	7.1
$_{63}$Eu	151	47.77	58.6
	153	52.23	41.4
$_{64}$Gd	152	0.2	12.9
	154	2.15	17.1
$_{64}$Gd	155	14.73	26.3
	156	20.47	21.2
	157	15.68	13.5
	158	24.87	6.4
	160	21.90	2.6
$_{65}$Tb	159	100.0	100.0
$_{66}$Dy	156	0.0524	1.05
	158	0.0902	22.2
	160	2.294	12.4
	161	18.88	27.35
	162	25.53	21.5
	163	24.97	8.4
	164	28.18	7.1
$_{67}$Ho	165	100.0	100.0
$_{68}$Er	162	0.136	4.9
	164	1.56	24.2
	166	33.41	30.2
	167	22.94	24.6
	168	27.07	15.6
	170	14.88	0.5
$_{69}$Tu	169	100.0	100.0
$_{70}$Yb	168	0.140	15.0
	170	3.03	35.9
	171	14.31	25.8
	172	21.82	10.7
	173	16.13	7.0
	174	31.84	4.8
	176	12.73	0.8
$_{71}$Lu	175	97.40	83.3
	176	2.60	16.7

with the fission yields. In Fig. 8 are shown the rare-earth abundances in the earth's crust,[28] in the sun, and in various types of stars,[29] together with yield curves for the stable isotopes of these elements in thermal-neutron U^{235} fission[30,31] and fission of natural uranium by 660-Mev protons. The yields and abundances are plotted as functions of atomic number (the percentage of each element in the rare-earth totals). In estimating the stable-isotope yields the contribution of radioactive chains has been taken into account.

An examination of these curves shows the same increase in the even elements both in nature and in the uranium fission products. The relative positions of these curves in Fig. 8 indicates that the rare-earth abundances observed in nature can not be explained by thermal-neutron fission of U^{235}, since elements heavier than terbium are not formed in this process. The fission-product curve for 660-Mev proton-induced fission in uranium differs from the natural abundance curve only in the region of heavy rare-earth elements. If it is assumed, however, that the heavy rare-earth yield increases with increasing proton energy at cosmic-ray energy, one would expect completely similar behavior for the two curves. For this reason there would seem to be little basis for the proposal that the abundances of the rare-earth elements observed in nature can be explained by fission of heavy heavy nuclei by high-energy protons.

A detailed examination of the data in Table III indicates a noticeable difference between the isotopic composition of the even rare-earth elements found in nature and those produced in the fission process. In fission due to fast protons there is a clear preponderance of light isotopes as compared with the natural distribution, especially for heavy elements. This preponderance of light isotopes becomes more pronounced as the proton energy is increased. Thus, fission of heavy elements by high-energy particles would not seem to be a likely mechanism for explaining the abundances of the rare-earth elements which are presently observed in nature.

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