PHYSICAL REVIEW VOLUME 89, NUMBER 3 FEBRUARY 1, 1953

Radiochemical Studies on the Fission of Th^{232} with Li+D Neutrons*,†

A. TURKEVICH, J. B. NIDAY,‡ AND ALTHEA TOMPKINS§
Institute for Nuclear Studies, University of Chicago, Chicago, Illinois

(Received October 14, 1952)

The fission of Th^{232} with neutrons from lithium bombarded with 7.6-Mev deuterons has been investigated radiochemically. The relative yields of fourteen mass chains, mostly in the light group, have been determined. The yields of symmetrical products are about tenfold higher than in the fission of Th^{232} with pile neutrons. Very light mass chains (very asymmetric fission) show a much smaller increase in yield.

I. INTRODUCTION

THE radiochemical investigation of the fission of Th^{232} with pile neutrons (~2.7 Mev) has recently been reported.[1] There have also been accumulated similar data over a period of years on the fission of this nuclide with fast neutrons from the Li+D reaction at the University of Chicago 37-inch cyclotron. These are the subject of this communication.

Increasing the energy of excitation of a nucleus undergoing fission is known to increase the relative yield of products of symmetrical division.[2–7] For several reasons, Th^{232} represents a good nucleus to study this effect. First, the separation of the mass peaks from the fission of Th^{232} with relatively low energy neutrons is the largest[1] of all investigated nuclei; i.e., Th^{232} fission is the most asymmetric of all. Secondly, Th^{232} has one of the highest neutron to proton ratios among fissionable nuclei. This means that even if some neutron evaporation should occur prior to the fission process with high energy neutrons, the yields of the radioactive species near stability will still represent the total mass yields.

II. GENERAL EXPERIMENTAL PROCEDURE

The thorium samples were in the chemical form $Th(NO_3)_4 \cdot 4H_2O$ (Baker's Analyzed) and weighed about seventy grams. They were packaged in two Pyrex test tubes, and wrapped in cadmium. The samples were placed directly in back of the 2 in.×3 in. water-cooled lithium target of the cyclotron.

The results reported are from fourteen bombardments varying in duration from two to eleven hours and in integrated intensities from 15 to 400 microampere hours. The variation of intensity with time during bombardment was monitored via the beam integrator of the cyclotron. Since, however, the lithium targets tended to deteriorate during a prolonged bombardment, this may be a source of error in the determination of short lived species.

The fission rate in a sample of 70 grams of $Th(NO_3)_4 \cdot 4H_2O$ under our conditions was about 2.3×10^5 per microcoulomb of deuterons on the target. It was thus about one hundred times lower in a typical bombardment than in the pile work already reported. This made it impractical to determine the yields of nuclides having fission yields less than 0.05 percent.

The samples were wrapped in cadmium in order to reduce the thermal neutron activation of Th^{232} and of uranium and other possible impurities. The thermal neutron flux inside the samples was about $5 \times 10^5 n/cm^2$ microcoulomb of deuterons on the target. It was thus less troublesome than in the pile fission of thorium.

After irradiation, the samples of thorium nitrate were dissolved in hot water containing varying amounts of nitric acid and diluted to 100 ml in a volumetric flask. Aliquots of this master solution were pipetted out for analyses for the various fission products. The determinations were usually made in duplicate and only infrequently was the same aliquot used for more than one fission product.

The general radiochemical procedure, as well as some details of analysis for fission products from thorium, has already been given.[1] The results reported here were obtained concurrently with the ones reported for the pile fission of Th^{232}. The radioactivity of the isolated samples was also measured in the same way, using cylindrical glass-walled Eck-and-Kreb type Geiger counters, and then corrected for efficiency of detection of the radiation. As before, only relative yields were determined, 53-day Sr^{89} again serving as a standard and being isolated in each bombardment. The most serious general error in this type of relative yield determination occurs in the correction for absorption

---

* A preliminary report of this work was presented at the December, 1947, meeting of the American Association for the Advancement of Science, Chicago, Illinois.

† The operation of the University of Chicago 37-inch cyclotron was supported during this period by the ONR.

‡ Present address: California Research and Development Company, Livermore, California.

§ Present address: Institute for the Study of Metals, University of Chicago, Chicago, Illinois.

[1] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951).

[2] Nishina, Yasaki, Kimura, and Ikawa, Phys. Rev. **58**, 660 (1940).

[3] Engelkemeir, Freedman, Seiler, Steinberg, and Winsberg, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 204, National Nuclear Energy Series, Plutonium Project Record, Div. IV, Vol. 9.

[4] R. W. Spence, Brookhaven Conference Report BNL-C-9 (July, 1949), unpublished.

[5] Perlman, Goeckermann, Templeton, and Howland, Phys. Rev. **72**, 352 (1947); R. H. Goeckermann and I. Perlman, Phys. Rev. **73**, 1127 (1948).

[6] A. S. Newton, Phys. Rev. **75**, 17 (1949).

[7] Jones, Fowler, and Paehler, Phys. Rev. **87**, 174 (1952).

TABLE I. Fission yields in the fission of Th^{232} with Li+D neutrons.

(1) Mass No.	(2) Nuclide isolated	(3) No. of bombardments in which nuclide isolated	(4) $\frac{Y_x}{Y_{Sr^{89}}}$	(5) Y_x[a]	(6) Yield in pile fission	(7) $\frac{(Y_x/Y_{89})_{Li+D}}{(Y_x/Y_{89})_{Pile}}$
77	12-hr Ge—38-hr As	3	0.0034	0.022	0.009	2.7
	Total chain[b]		0.0078	0.052	0.020	
83	2.4-hr Br[c]	2	0.41	2.74	1.9	1.4
89	53-day Sr	14	(1.00)[a]	(6.7)	(6.7)	(1.00)
91	9.7-hr Sr	3	0.84	5.6	6.4	0.93
97	17.0-hr Zr—68-min Nb	2	0.74	4.95	5.4	1.0
99	67-hr Mo	3	0.46	3.1	2.9	1.0
103	42-day Ru	1	0.076[d]	0.51[d]	0.20	2.5
106	1.0-yr Ru—30-sec Rh	1	0.079	0.53	0.058	9.0
111	7.5-day Ag	3	0.094	0.63	0.052	13.1
115	2.25-day Cd—4.53-hr In	1	0.113	0.76	0.072	11.2
117	2.8-hr Cd—117-min In	1	0.055[d]	0.37[d]		
131	8.0-day I	2	0.35	2.3	1.2	2.0±1.0
132	77-hr Te—2.4-hr I	1	0.27[d]	1.8[d]	2.4	0.8±0.3
139	85-min Ba	2	1.34	9.0		
144	275-day Ce—17-min Pr	3	1.07	7.2	7.1	1.1

[a] 53-day Sr^{89} was the standard chosen in the relative yield determination. In both this work and in the pile neutron studies it was assigned a value of 6.7 percent to get absolute yields for other nuclides.

[b] The total yield of chain 77 was calculated assuming that 57 percent of the chain goes through the 59-sec Ge^{77} isomeric state (reference 1).

[c] Yield of 2.4-hr Br^{83} was calculated assuming 27/48 of the bromine came from 67-sec Se^{83} and the rest from 25-min Se^{83} (reference 1).

[d] These relative yields carry an estimated error of 50 percent.

of the beta-radiations in the counter walls. This limits the accuracy of these yields to 20 percent.

In many cases, however, the relative yields in the fast neutron fission of Th^{232} could be compared with the same quantities in the pile fission of Th^{232} with greater accuracy. This was possible because the samples were measured with the same (or very similar) Geiger tube. In this case, corrections for efficiency of detection of the radiations either cancel out completely or are much less serious than in the individual relative yield values.

The yields of fourteen mass chains, mostly in the light group of the fission products, have been determined. In all but five of these, the yields reported are averages of the results of at least two bombardments. The reproducibility from one bombardment to another was somewhat poorer than in the pile work. This was probably due to the lower intensities available—in several cases making it possible to check only crudely the purity of the isolated radioactivity by absorption methods. The lower intensities also increased the relative interference by thorium decay products. The lack of reproducibility may also be connected with slight differences in positioning of the samples during bombardment. This could give rise to variation in the yields of energy sensitive fission products.

The only reported[8] value of the cross section for the neutron fission of Th^{232} is that of 10^{-25} cm^2 at 2.5 Mev. The neutrons from a thick target of lithium bombarded with 7.6-Mev deuterons have energies ranging up to 21 Mev. The neutron distribution from such a source, however, has been determined only at low deuteron energies.[9] Theoretical calculations on what the spectrum might be at a bombarding energy of 7.6 Mev indicated that the kinetic energy of most of the neutrons should be less than 14 Mev. This result was in agreement with $Cu^{63}(n,2n)Cu^{62}$ activation experiments, which showed that for comparable cross sections[8,10] about ten percent of the fissions in Th^{232} were induced by neutrons with energy greater than 12 Mev. We estimate that the majority of the fissions under our conditions are caused by neutrons with energy between 6 and 11 Mev.

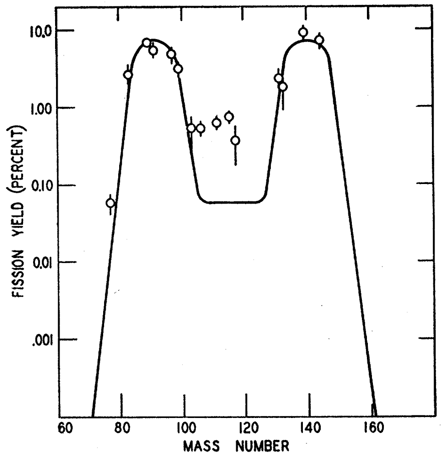

FIG. 1. Fission yields from Th^{232} with (Li+D) neutrons. The solid curve is the yield curve of Th^{232} with pile neutrons (see reference 1).

---

[8] Ladenburg, Kanner, Barschall, and Van Voorhis, Phys. Rev. **56**, 168 (1939).

[9] H. T. Richards, Phys. Rev. **59**, 796 (1941); L. L. Green and W. M. Gibson, Proc. Phys. Soc. (London) **62**, 407 (1949).

[10] J. L. Fowler and J. M. Slye, Phys. Rev. **77**, 787 (1950).

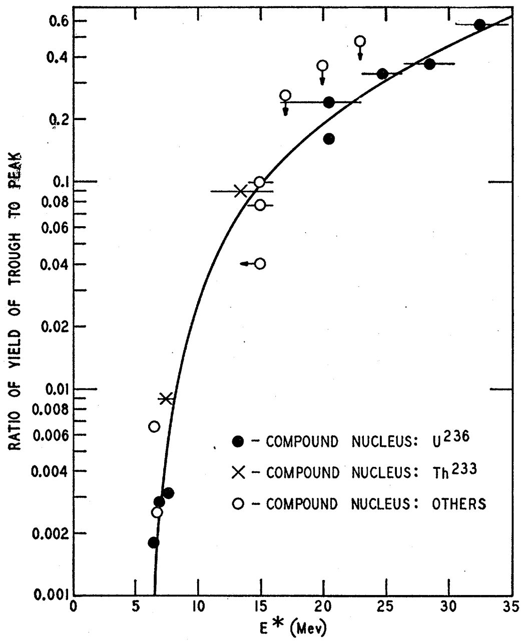

FIG. 2. Effect of energy of excitation of the compound nucleus (E^*) on the relative yield of symmetrical fission.

III. RESULTS

The results are presented in Table I. This lists for each mass number studied the long-lived radioactive members. The nuclide isolated radiochemically is underlined. The third column indicates the number of bombardments in which the nuclide studied was isolated. The fourth column gives the ratio of the yield of this nuclide to that of the standard, 53-day Sr^{89}. This is the experimental ratio of the activities corrected to an infinite bombardment and corrected for differences in detectability of the radiations.[1] The fifth column gives the yield of the chain in Th^{232} fission with Li+D neutrons, assuming Sr^{89} has a yield of 6.7 percent (the value obtained in the pile fission of Th^{232}). We assign a probable error of 20 percent to these relative yields, except for those indicated in the table, where special circumstances dictate a higher error.

The sixth column reproduces the yields in the pile fission of Th^{232} from the previous work.[1] Finally, the last column gives the ratio of the relative yield of the nuclide (referred to Sr^{89}) in fast neutron fission to the same quantity in pile fission. Unless indicated otherwise, these last ratios are believed to be good to 10 percent.

The yields in Th^{232} fission with Li+D neutrons are presented in Fig. 1. The vertical lines represent our estimate of the reliability of the yields relative to Sr^{89}. The figure shows also, for comparison, the smooth curve drawn through the yield data in the pile fission of Th^{232}. Neither of these studies on the fission of Th^{232} is accurate enough to look for the deviations from a smooth fission yield distribution indicated by recent mass spectrographic work.[11]

IV. DISCUSSION

The yield data illustrate the rise in relative yield of symmetrical fission when the energy of excitation of the nucleus undergoing fission is increased. There does not exist at present detailed information on how this probability increases for any one nucleus. The best one can do is to collect the fragmentary data at various energies for different compound nuclei and to compare them.[‖] This is done in Fig. 2. The ordinate (on a logarithmic plot) is the ratio of the yield for symmetrical division to that for the most probable (asymmetric) product. The abscissa gives the energy of excitation of the compound nucleus.

Table II gives the source of the data used in constructing Fig. 2.[1,4,6,7,12–16] The energy of excitation has been calculated from the kinetic energy of the bombarding particle and the binding energy of the projectile in the compound nucleus.[17] In the case of the photofission experiments,[4,12,13] the results have been plotted

TABLE II. Data for Fig. 2.

E^* Mev	Compound nucleus	$\frac{\text{Trough yield}}{\text{Peak yield}}$	Nuclear reaction	Reference
6.50	U^{236}	0.0018	thermal neutron fission of U^{235}	14
6.50	Pu^{240}	0.0065	thermal neutron fission of Pu^{239}	14
6.74	U^{234}	0.0025	thermal neutron fission of U^{233}	15
6.9	U^{236}	0.0028	U^{235}+~0.4-Mev neutrons	4
7.7	U^{236}	0.0031	U^{235}+1.2-Mev neutrons	4
7.5	Th^{233}	0.009	Th^{232}+pile neutrons	1
11–16	Th^{233}	0.09	Th^{232}+(Li+D) neutrons	This paper
~15	Th^{232}	0.10	Th^{232}+69 Mev (max)γ	12
~15	U^{238}	0.077	U^{238}+48 Mev (max)γ	13
~15	U^{235}	0.04	U^{235}+gamma-rays	4
20.5	U^{236}	0.16	U^{235}+14-Mev neutrons	4
17	Np^{239}	<0.26	U^{238}+cyclotron protons	7
20	Np^{239}	<0.36		
23	Np^{239}	<0.48		
16.5–23.0	U^{236}	0.24	$Th^{232}+\alpha$	6, 16
23.0–26.5	U^{236}	0.33		
26.5–30.5	U^{236}	0.37		
30.5–34.5	U^{236}	0.57		

---

[11] H. G. Thode and R. L. Graham, Can. J. Research **A25**, 1 (1947); MacNamara, Collins, and Thode, Phys. Rev. **78**, 129 (1950); Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. **84**, 860 (1951).

[‖] *Note added in proof:* A similar comparison has recently been made by Fowler, Jones, and Poehler, Phys. Rev. **88**, 71 (1952).

[12] D. M. Hitler and D. S. Martin, Abstract, American Chemical Society Meeting, Atlantic City, New Jersey (September, 1952).

[13] N. Sugarman and R. Schmitt, private communication.

[14] E. P. Steinberg, private communication. See also E. P. Steinberg and M. S. Freedman, Paper 219, of reference 3.

[15] Steinberg, Seiler, Goldstein, and Dudley, talk before the American Association for the Advancement of Science (December, 1947); USAEC declassified document, MDDC 1632 (January 6, 1948), unpublished.

[16] The data of Newton (see reference 6) on the yield of 43-day Cd^{115} relative to Ba^{140} as a function of depth of thorium target irradiated with 38-Mev alphas was normalized by assuming that this ratio has a value 0.00013 in the thermal neutron fission of U^{235} where the trough to peak ratio is 0.0018.

[17] G. T. Seaborg, Pi Lambda Upsilon Lecture, Ohio State University, Columbus, Ohio (March, 1952), unpublished.

at approximately 15 Mev, the reported[18] photofission resonance energy for U^{238}. The resonance is expected to be at approximately the same[19] energy in the case of Th^{232} and U^{235}. The ratios from the proton bombardment of normal uranium[7] are considered to be upper limits since the fission product selected in the work, Ag^{111}, is probably not at the bottom of the trough. These data are presented with downward arrows. With two exceptions, the remaining points lie on a reasonably smooth curve. The ratio for the thermal neutron fission[14] of Pu^{239} is definitely higher than the other data at comparable excitation. The other striking deviation is the lone experiment[4] reported on the photofission of U^{235}. It is possible that in this work the betatron energy was not sufficiently greater than 15 Mev to take advantage of the photofission resonance.[20] Within the accuracy of our knowledge of the energy of the Li+D neutrons causing fission in this study, the results reported here lie on the curve.

Figure 2 indicates the strong energy dependence of symmetrical fission and can be interpreted as an excitation curve for the formation of symmetrical products relative to asymmetrical ones in the fission of heavy elements. It would indicate, for example, that the yield of symmetrical products in spontaneous fission (if the main peaks are well separated) would be immeasurably low.

Table I and Fig. 1 also illustrate that the increase in neutron energy from pile neutrons to Li+D neutrons has a much less drastic effect on the low yields on the light side of the light peak. The yield of Ge^{77} has increased 2.7-fold; that of Br^{83} only by 40 percent. The increase in yield in this region in the case of Th^{232} can be explained semiquantitatively by the postulate that an appreciable fraction of the fissions with Li+D neutrons involve the emission (before or after fission) of one or two more neutrons than in the case of pile neutron fission. This insensitivity to energy in this region of the yield curve confirms the data of Newton.[6] In his work the same two mass chains had approximately the same yields from U^{236} excited by about 33 Mev as in U^{236} formed from thermal neutrons on U^{235}. Newton's work on U^{236} excited by 33 Mev also shows little increase in yields on the heavy side of the heavy group. Thus, both the work on Th^{232}+37.5-Mev alphas[6] and this work emphasize the difference between the restrictions preventing symmetric and very asymmetric fission.

---

[18] W. E. Ogle and J. McElhinney, Phys. Rev. **81**, 344 (1951).
[19] M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).
[20] R. W. Spence (private communication).

PHYSICAL REVIEW VOLUME 89, NUMBER 3 FEBRUARY 1, 1953

Ionization Probability Curves for Krypton and Xenon near Threshold

R. E. Fox, W. M. Hickam, and T. Kjeldaas, Jr.
Westinghouse Research Laboratories, East Pittsburgh, Pennsylvania

(Received October 10, 1952)

The shapes of the ionization probability curves of krypton and xenon have been determined near the ionization potential by measuring the ionization produced by nearly monoenergetic electrons in a mass spectrometer. Ionization processes associated with the $^2P_{\frac{1}{2}}$ and $^2P_{\frac{3}{2}}$ ground states of the ions are clearly resolved. Ions are observed which are attributed to auto-ionization of atoms excited to the higher states having the $^2P_{\frac{1}{2}}$ core configuration.

I. INTRODUCTION

LITTLE information is available concerning the true shapes of ionization probability curves near threshold. This is largely owing to the difficulty in obtaining an electron beam with a sufficiently narrow energy spread. Lawrence,[1] and later Nottingham,[2] in their study of the ionization probability of mercury used a magnetic analyzer to reduce the energy spread of the electron beam. In this type of experiment where total ionization is measured precautions are necessary to distinguish photoelectric effects and true ionization and to eliminate impurities in the gas sample. If, however, only the shape of the ionization probability curve is to be determined, one may use a mass spectrometer in which a positive analysis of the ions produced largely eliminates the need for these precautions.

On the other hand, the problems associated with the production of a sufficiently monoenergetic electron beam in a mass spectrometer were solved only recently. With the use of this new method it was demonstrated[3,4] that within the experimental accuracy then attainable, the ionization produced was directly proportional to the excess energy of the bombarding electrons in the region near the threshold.

In the earlier work,[3] it was reported that a slight break sometimes observed in the ionization probability curves for krypton might be attributed to the doublet ground state of the krypton ion. Since then, an increase in sensitivity, due in part to a new pulsing circuit,[5] has permitted a more detailed study of ionization prob-

---

[1] E. O. Lawrence, Phys. Rev. **28**, 947 (1926).
[2] W. B. Nottingham, Phys. Rev. **55**, 203 (1939).
[3] Fox, Hickam, Kjeldaas, and Grove, Proc. of the Symposium on Mass Spectroscopy in Physical Research, Natl. Bur. Standards Circular 522, January, 1953.
[4] Fox, Hickam, Kjeldaas, and Grove, Phys. Rev. **84**, 859 (1951).
[5] To be published.