Uranium Photofission Yields*†

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(Received April 19, 1954)

The photofission yield-mass curve of natural uranium for 48-Mev x-rays was determined radiochemically. Isolation of selected peak and trough fission products at various maximum x-ray energies, 7 Mev, 10 Mev, 16 Mev, 21 Mev, 48 Mev, 100 Mev, and 300 Mev showed that the peak-to-trough ratio decreased from 300 to 4 as the maximum x-ray energy increased from 7 Mev to 300 Mev. The observed photofission yield curves may be quantitatively interpreted as the superposition of two components, a low-energy asymmetric (double-humped) curve and a high-energy single-humped curve. The average cross section for symmetric fission for photons in the energy range 16 Mev to 300 Mev is about 7 millibarns.

I. INTRODUCTION

MANY investigations of high-energy fission, induced by particles and x-rays, have been reported.[1–12] One striking feature of these studies is the decrease in the peak-to-trough yield ratio of the yield-mass curve as the energy of the bombarding particle increases, resulting in the one hump yield-mass curve as the energy of the particle enters the hundred-Mev range. A systematic investigation of the photofission of a heavy nuclide at various energies has not been reported.

The purpose of this work was essentially threefold; (1) to determine the yield-mass curve of natural uranium for 48-Mev maximum x-rays by the comparison method;[4,13,14] (2) to study the changes in the photoyield curve of uranium as a function of increasing maximum x-ray energy from 7 Mev to 300 Mev; and (3) to study the change in the neutron-to-proton ratio of primary fission fragments with increasing x-ray energy. On the assumption that the observed fission-yield curves are composites of two idealized curves,[14] one very asymmetric and the other symmetric, the excitation function and probability for symmetric fission as a function of energy were calculated.

II. EXPERIMENTAL PROCEDURE

A. Irradiation Setups

The University of Chicago 100-Mev betatron, described by Westendorp and Charlton,[15] was used primarily in this research. Ten-gram samples of analytical reagent grade uranyl nitrate (Mallinckrodt) in plastic vials were placed about 32 cm from the tungsten target (about 3 cm from the Pyrex glass doughnut) directly along the x-ray axis and within the half-angle of the x-ray beam. Average intensities of the x-ray beams at (21±1) Mev, (48±1) Mev, and (100±1) Mev, in roentgens/min at one meter from the target, were about 20, 250, and 1000, respectively. About 50 irradiations, most of them at 48 Mev, were performed.

Twenty gram samples in glass vials were irradiated at the University of Illinois 22-Mev betatron. A total of 10 irradiations were made at energies of 7 Mev (6.8±0.1), 10 Mev (9.7±0.1), 16 Mev (15.5±0.1), and 21 Mev (21.3±0.1). X-ray intensities at one meter for the 7, 10, 16, and 21 Mev x-ray beams were about 2.5, 9, 20, and 65 roentgens/min, respectively.

Six irradiations at 300 Mev were performed at the University of Illinois 340-Mev betatron. Cylindrical glass vials, containing 2.5 g of powdered uranyl nitrate, were placed about 55 cm from the platinum target. The intensity was about 1500 roentgens/min at one meter.

Since the photofission yields were determined by comparison with thermal-neutron yields,[4,13,14] it was necessary to obtain thermal-neutron irradiations of uranyl nitrate. These were performed at the Argonne Heavy-water Pile (CP-3) and at the University of Chicago 37-inch cyclotron. Six irradiations were performed at the pile, in each of which about 2 g of uranyl nitrate was bombarded in the thermal column for about 20 min at an average neutron flux of about 3×10^{11} neutrons/cm² sec. Ten irradiations were made at the

* This work was supported in part by a grant from the U. S. Atomic Energy Commission.
† Part of this document is based on work performed at the University of Illinois, Urbana, Illinois, supported by a grant from the U. S. Office of Naval Research.
‡ Presented in partial fulfillment for the Ph.D. degree in the Department of Chemistry, University of Chicago. Now at the University of Illinois, Urbana, Illinois.

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[12] H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).
[13] Finkle, Hoagland, Katcoff, and Sugarman in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 216 National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 1368.
[14] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951).
[15] W. F. Westendorp and E. E. Charlton, J. Appl. Phys. **16**, 581 (1945).

cyclotron. About 30 g embedded in 4 inches of paraffin was irradiated for about 20 min to 4 hr at a neutron flux of about 10^9 neutrons/cm^2 sec.

B. General Radiochemical Procedure

After irradiation, the uranyl nitrate was dissolved in $6N$ HNO_3 and was diluted to $3N$. Radiochemical analyses were performed on solutions containing about 20 mg of carrier and aliquots yielding an initial counting rate of about 1000 to 8000 counts per minute for the nuclide isolated. The radiochemical analyses, after slight modifications because of larger amounts of uranyl nitrate, were the same as those employed on the Plutonium Project.[16] Duplicate analyses for each nuclide from a given bombardment were carried out.

The final precipitates of about 25 mg were filtered onto 1.8-cm diameter filter paper disks, which were subsequently mounted on rectangular cardboards 6.3 cm $\times$7.6 cm and 60 mg/cm^2 thick. Cellophane of about 2.5 mg/cm^2 covered the precipitates. The radioactive samples, all beta emitters, were counted by a standard end-window ($\sim$2.5 mg/cm^2) Geiger-Mueller Tube (Tracerlab TGC-1) in a lead shield.

The nuclides isolated from betatron irradiations were also isolated from thermal-neutron bombardments. They were identified by tested radiochemical procedures,[16] by counting them for at least 3 to 4 half-lives, and by absorption curves when necessary. In some cases it was advantageous to follow the decay curves of a nuclide isolated from x-ray and neutron fission through a selected aluminum absorber.

III. SECONDARY NEUTRON FISSION

Since fast-neutron fission tends towards more symmetric cleavage of the nucleus,[4,8,17] it was important to show that the high photofission trough yields which are observed were not due to high-energy neutrons, resulting from (γ,n) reactions in the electron target, the doughnut, and the sample. The high ($\sim$15 Mev) potential barrier for protons and the small number of protons above 15 Mev excludes any appreciable proton-fission competition.

In order to determine the fast-neutron flux in the 10 g sample, and thereby, the fast-neutron fission contribution, a number of experiments were performed on the activation of phosphorus. The radioactivity from the reaction $P^{31}(n,p)S^{31}$ ($Q=-1.0$ Mev) was detected as 170-min Si^{31}. It was assumed that the average cross section[18,19] for the (n,p) reaction is 0.10×10^{-24} cm^2 from $\sim$2 Mev to 14 Mev.

Experiments at 48 Mev were performed to determine the fast-neutron flux resulting from (γ,n) reactions in the sample, and in the electron target and the Pyrex doughnut. About 1.1 g of red phosphorus was sandwiched between two plastic cylinders each of which contained 4.3 g of powdered uranyl nitrate. An identical setup, but with no uranyl nitrate, was irradiated for 10 min immediately after the first setup and at the same betatron intensity.

TABLE I. Fast-neutron measurements.

Sandwich	(nv), cm^{-2} min^{-1}	Fission rate, min^{-1}
(U—red P—U)	1.1×10^8	6.5×10^7
(no U—red P—no U)	5.3×10^7	

After irradiation, the phosphorus samples (Si^{31}) were counted. 85-min Ba^{139} was isolated from the combined uranyl nitrate samples of the first irradiation from which the total fission rate was calculated. The results are given in Table I, where (nv) is the fast-neutron flux from $\sim$2 Mev to 14 Mev, and the fission rate is that observed in the uranium samples. Assuming an (nv) of 1.1×10^8 cm^{-2} min^{-1} in the sample and a fission cross section[20] of 0.5×10^{-24} cm^2, we calculate that 1 percent of the total fissions at 48 Mev is due to fast-neutron fission. Similar results were obtained at 100 Mev. This low percentage of neutron fission is about the same as that found by Richter[21] using 16-Mev x-rays.

Experiments with 300-Mev x-rays yield a maximum value of 1 percent for the fast-neutron fission contribution. The thermal-neutron fission contribution, resulting either from external neutrons or internal neutrons was shown to be unimportant by cadmium shielding and indium foil activation experiments. If it is assumed that the peak-to-trough ratio for the fast-neutron fission is 6, as found with 14-Mev neutrons,[4] then the maximum error in the observed peak-to-trough ratios is 5 percent.

IV. EXPERIMENTAL RESULTS

The comparison method[4,13,14] was used in this research to determine the photofission yields in order to circumvent absolute beta and gamma counting. The ratio of two photofission yields is given by the equation:

$$\frac{Y'(N)}{Y'(S)}=\frac{Y(N)}{Y(S)}\cdot\frac{A^{\infty}(S)}{A^{\infty}(N)}\cdot\frac{A^{\infty\prime}(N)}{A^{\infty\prime}(S)}.$$

In this equation (N) and (S) designate, respectively, the nuclide isolated and the standard nuclide isolated (67-hr Mo^{99}); A^{∞} and $A^{\infty\prime}$, the saturation activities for

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[18] B. L. Cohen, Nucleonics **8**, No. 2, 29 (1951).
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[20] Ladenburg, Kanner, Barschall, and Van Voorhis, Phys. Rev. **56**, 168 (1938); National Research Council of Canada, Atomic Energy Project, Can, J. Phys. **29**, 203 (1951).
[21] H. G. Richter, Ph.D. thesis, Massachusetts Institute of Technology, August 16, 1952 (unpublished), showed by a modified technique that less than 0.8 percent of the fission of uranium with 16-Mev x-rays from the linear accelerator was caused by fast neutrons.

TABLE II. Natural uranium photofission yields.

Mass No.	Nuclide isolated and measured[a]	Thermal-neutron yield in U^{235}[b]	7 Mev	10 Mev	16 Mev	21 Mev	48 Mev	100 Mev	300 Mev
77	12-hr Ge→38-hr As	0.0067[c]					0.032±0.004[d]		
78	86-min Ge→91-min As[e]	0.020[b,c]					0.059±0.002		
83	2.4-hr Br	0.48					0.59±0.06	0.62±0.07	0.73±0.08
84	31-min Br	0.65					1.03±0.17	1.04±0.15	1.09±0.18
89	53-day Sr	4.3				2.6±0.1	2.8±0.1	2.8±0.1	3.0±0.1
91	9.7-hr Sr[f]	5.5					3.9±0.1		
97	17.0-hr Zr→74-min Nb[g]	6.0				5.7±0.2	5.8±0.2	5.8±0.2	
99	67-hr Mo[b]	6.8	6.6[i]	6.6[i]	6.6[i]	6.6[i]	6.6	6.6[i]	6.6[i]
103	38.5-day Ru	2.2				3.0±0.3	2.9±0.2	3.2±0.2	3.4±0.3
105	4.5-hr Ru	0.9					2.5±0.1		
106	1.0-yr Ru→30-sec Rh[g]	0.52				2.1±0.3	2.0±0.2	2.6±0.2	3.0±0.2
111	7.6-day Ag	0.018	0.046±0.004	0.065±0.007	0.30±0.01	0.43±0.01	0.77±0.02	1.02±0.02	1.88±0.06
112	3.2-hr Ag[j]	0.011	0.031±0.005	0.047±0.005	0.16±0.01	0.26±0.02	0.52±0.03	0.71±0.04	1.14±0.08
113	5.2-hr Ag	0.011[k]					0.60±0.03	0.77±0.04	1.21±0.06
115	54-hr Cd[l]	0.011		0.030±0.004	0.16±0.01	0.25±0.01	0.047±0.02	0.67±0.03	1.15±0.05
	43-day Cd	8×10^{-4}			0.013±0.001	0.018±0.001	0.041±0.002	0.048±0.002	0.20±0.02
117	2.83-hr Cd→1.95-hr In[e]	0.010		0.027±0.007			0.50±0.02	0.69±0.02	1.04±0.04
127	94-hr Sb	0.094[p]				0.70±0.02[p]	0.93±0.02[p]	1.07±0.03[p]	1.49±0.09[p]
131	8.0-day I	2.8				4.1±0.2	4.3±0.1	4.4±0.2	4.6±0.2
132	2.4-hr I[j]	4.2				5.0±0.1	4.9±0.1	4.6±0.1	4.3±0.1
133	22.4-hr I[m]	6.3					6.2±0.3		
137	33-yr Cs	6.1					4.7±0.3		
139	85-min Ba	6.3					4.6±0.1	5.1±0.1	
140	12.8-day Ba→40-hr La[g]	6.2	5.8±0.3	5.7±0.2	5.0±0.1	4.9±0.1	5.0±0.3	5.3±0.1	4.8±0.2
141	30-day Ce[n]	5.9					4.9±0.3		
143	33-hr Ce[o]	5.3				4.0±0.1	3.8±0.3	3.8±0.1	3.6±0.1
144	275-day Ce→17.5-min Pr[g]	4.6				3.8±0.2	3.4±0.2		

[a] The observed half-lives are given. For literature values, see reference 16 and "Table of Isotopes," Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).
[b] Yields above 2 percent and below 2 percent were taken from references 22 and 23, respectively.
[c] Yield of 38-hr As^{77} as reported by N. Sugarman, Phys. Rev. 89, 570 (1953).
[d] Yield of 38-hr As^{77}, calculated by assuming that the branching ratio of 12-hr Ge^{77} to 59-sec Ge^{77} is 0.52 as reported in reference c.
[e] Activity mixture counted. Half-lives of Ge and As are those of reference c.
[f] Counted through 199 mg/cm² Al to emphasize 9.7-hr Sr^{91}.
[g] Counted equilibrium mixture.
[h] Counted through 21.0 mg/cm² Al to absorb radiations of 6.0-hr Tc^{99m} daughter.
[i] Assumed yield; other yields normalized to it.
[j] The daughters 3.2-hr Ag^{112} and 2.4-hr I^{132} were separated from their parents during equilibrium. The yields of 112 and 132 are therefore the yields of their parents.
[k] Interpolated yield from smooth yield curve of reference 23.
[l] Counted through 91.0 mg/cm² Al to absorb radiations of 4.5-hr In^{115m} daughter.
[m] Counted through 56.9 mg/cm² Al to emphasize 22.4-hr I^{133}.
[n] Counted through 24.9 mg/cm² Al to absorb soft components of 30-day Ce^{141}.
[o] Counted through 67.4 mg/cm² Al to emphasize 33-hr Ce^{143}.
[p] The thermal-neutron yield of 0.15 percent reported by A. C. Pappas [Massachusetts Institute of Technology Technical Report No. 63, September 15, 1953 (unpublished)] raises the photoyields by a factor of 1.6.

thermal-neutron and x-ray irradiations, respectively; and Y and Y', the thermal-neutron and x-ray fission yields, respectively.

The thermal-yield values, $Y(N)$ and $Y(S)$, above 2 percent, obtained by Glendenin *et al.*,[22] and those below 2 percent, found by workers on the Plutonium Project,[23] were taken as absolute values (Table II). The absolute photo-yield of 67-hr Mo^{99}, $Y'(S)$, was found to be 6.6 percent for 48-Mev x-rays by normalizing the fission yield curve to 200 percent.

The most important advantage to be gained from the comparison method is the elimination of the need for absolute beta and gamma counting. The major disadvantages of the method are: (1) the photoyield curve is at best no more accurate than the assumed thermal-neutron yield curve, and (2) the yield determined in this manner, or by absolute counting, may not represent the full yield of the chain,[24,25] especially at higher energies where the most probable charge for a given mass is closer to stability.[3,26,27]

The photofission yield data of natural uranium at 7, 10, 16, 21, 48, 100, and 300 Mev are given in Table II, where, for the most part, the errors are the mean deviations of duplicate samples of an individual run. In some cases, such as Ru^{106}, Ag^{111}, Ag^{112}, Sb^{127}, and others, the yields at 48 Mev are the averages obtained from two or more experiments. In these cases, the errors are not appreciably different from those of a single run. Photofission values at 48 Mev are normalized to a total yield of 200 percent. The 7, 10, 16, 21, 100, and 300-Mev photoyields were calculated by arbitrarily assuming a yield for 67-hr Mo^{99} of 6.6 percent, the normalized 48-Mev value.

The experimental results of Table II are graphically represented in Figs. 1 and 2. Construction of the best 48-Mev photocurve (Fig. 1) was accomplished as follows. A binary fragment sum of 234, amounting to the emission of 4 neutrons per fission, was found by reflecting the experimental values of near-peak yields that are sensitive to the mass sum assumed, *viz.*, masses 89, 91, 140, 141, 143, and 144. A larger mass sum[28] of 237 is a better fit to the experimental yields of near-trough masses 111 and 127. Construction of the intermediate portion of the curve was accomplished by assuming that the mass sum ranged from 234 to 237. Similar

[22] Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. 84, 860 (1951).
[23] Reference 16, Appendix B.
[24] Glendenin, Coryell, and Edwards, reference 16, paper 52.
[25] L. E. Glendenin, Ph.D. thesis, Massachusetts Institute of Technology, Technical Report No. 35, July 29, 1949 (unpublished).
[26] P. Kruger, Ph.D. thesis, University of Chicago, January, 1954 (unpublished).
[27] L. Jodra, University of Chicago (unpublished).
[28] The effect of the apparent change in the number of neutrons emitted in other fission-yield curves was observed earlier by L. E. Glendenin and E. P. Steinberg of the Argonne National Laboratory, and C. D. Coryell of the Massachusetts Institute of Technology (private communication).

results were also found for the 21- and 100-Mev photoyield curves.

Since the 48-Mev curve is normalized to 200 percent and since the asymmetric yields at 16, 21, 100, and 300 Mev did not change appreciably when normalized to a yield of 6.6 percent for Mo^{99}, it is necessary to displace these photocurves vertically about +4, +3, −2, and −8 percent, respectively, to take account of the changing yields in the trough. These changes, however, do not affect the peak-to-trough yield ratios.

Table III presents data on independent yields of selected nuclides and their fractions of the chain yields for the thermal-neutron fission of U^{235} and for the x-ray work of this research. The species studied were 36.0-hr Br^{82}, 3.2-hr Ag^{112}, 52.5-min I^{134}, and 13.6-day Cs^{136}. It is noted that the independent yields of Br^{82} and Cs^{136} increase markedly at the higher energies. This effect is discussed in Sec. V. D.

V. DISCUSSION

A. General Observations of Photofission Results

Baldwin and Klaiber[29] determined the photofission excitation curve for uranium and reported a peak in the curve at about 16 Mev and a negligibly small cross section beyond 33 Mev. This peak at about 16 Mev was interpreted by Goldhaber and Teller[30] as a resonance in the photoabsorption cross section. Relative cross-section data of the photofission of U^{235} and U^{238} obtained by McElhinney and Ogle[31] indicate that in the 12-Mev to 22-Mev maximum x-ray interval the isotope U^{238} in natural uranium is responsible for about 99 percent of the fissions. Jungerman and Steiner[32] obtained similar results at 335 Mev.

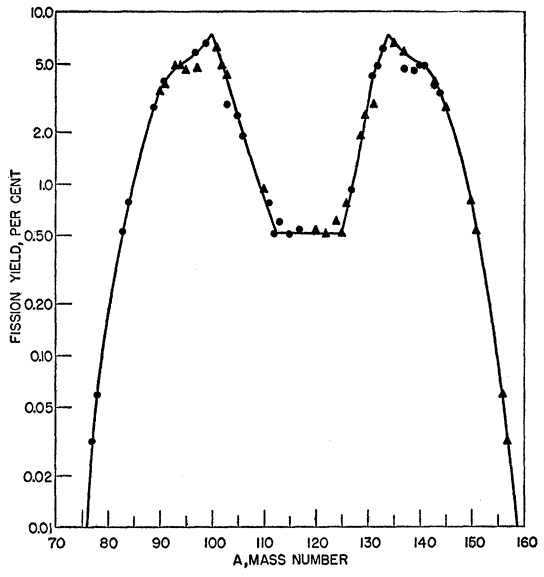

FIG. 1. Photofission yield curve for 48-Mev x-ray fission of natural uranium. ●, observed 48-Mev photoyields; ▲, reflected 48-Mev yields assuming that the fragment sum was 237 for masses 111 through 127, 234 for asymmetric masses, and mass sums in the 234 to 237 range for near-trough masses. Total fission yield is 200 percent.

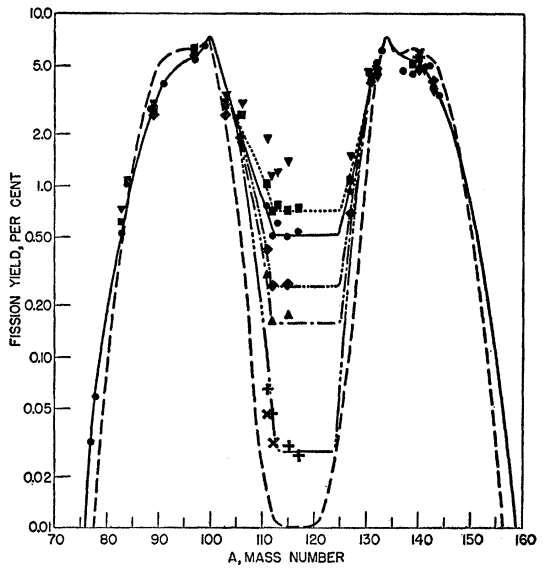

FIG. 2. Photofission yield curves for natural uranium as a function of x-ray energy. ×, 7 Mev; +, 10 Mev; ▲, 16 Mev; ◆, 21 Mev; ●, 48 Mev; ■, 100 Mev; and ▼, 300 Mev. The 7, 10, 16, 21, 100, and 300 Mev yields are normalized to 6.6 percent yield for mass 99 (48-Mev yield). ——, thermal-neutron U^{235} curve; — — — —, 10-Mev curve; — — — —, 16-Mev curve; — — — —, 21-Mev curve; ————, 48-Mev curve; and — — — — —, 100-Mev curve.

The fission yield curve in uranium was extensively investigated with 48-Mev x-rays so that excitation over the entire resonance region[33] could occur. Following are some comparisons of the 48-Mev x-ray fission curve of U^{238} with the thermal-neutron fission curve of U^{235} and other specified curves. The thermal-neutron fission curve of U^{235} is used for comparison since the fragment mass sum for the most probable mode of fission appears to be the same as that from 48-Mev x-rays and U^{238}.

1. The very-asymmetric yields of masses 77 through 84 are substantially higher than the corresponding thermal-neutron yields in U^{235} and the 10-Mev and 16-Mev x-ray yields[21] in U^{238}, where determined (see Table II and Fig. 3).

2. The light-peak yields of masses 89 through 106 and the yields of the complementary masses on the heavy peak are approximately equal to the corresponding yields of the fission-neutron (Fig. 4) or the 16-Mev photofission (Fig. 3) of U^{238}, and the 14-Mev neutron and ~20-Mev x-ray fission of U^{235} (Fig. 4).

3. Yields of the near-symmetric (111 and 127) and

[29] G. C. Baldwin and G. S. Klaiber, Phys. Rev. **71**, 3 (1947).
[30] M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).
[31] J. McElhinney and W. E. Ogle, Phys. Rev. **81**, 342 (1951).
[32] J. Jungerman and H. Steiner, Phys. Rev. **93**, 949(A) (1954).
[33] As discussed later in this paper, the cross section beyond the resonance region is small but measurable. J. Gindler, Ph.D. thesis, University of Illinois, February, 1954 (unpublished), using a different experimental approach from that reported here, has arrived at the same conclusion.

TABLE III. Independent fission yields.

Nuclide	Independent yield, percent: U^{235} thermal neutrons[a]	Independent yield, percent: Natural uranium, x-rays	Chain yield, percent: U^{235} thermal neutrons[a]	Chain yield, percent: Natural uranium, x-rays	Fraction of chain yield: U^{235} thermal neutrons[a]	Fraction of chain yield: Natural uranium, x-rays
36.0-hr Br^{82}	3.5×10^{-5}	1.8×10^{-4}[b] 2.7×10^{-2}[c]	0.20	0.38[d] 0.40[d]	1.75×10^{-4}	4.7×10^{-4}[b] 6.5×10^{-2}[c]
3.2-hr Ag^{112}		<0.01[b] <0.02[e] <0.07[c]		<0.53[b] <0.73[e] <1.21[c]		$<1.9\times10^{-2}$[b] $<2.8\times10^{-2}$[e] $<5.8\times10^{-2}$[c]
52.5-min I^{134}	1.0 0.9[f]		5.5 7.9[f]	7.4[d]	0.19[g] 0.11[f]	0.20[b,h]
13.6-day Cs^{136}	6.2×10^{-3}	0.044[b]	6.0	6.2[b,d]	1.0×10^{-3}	7.1×10^{-3}[b]

[a] Taken after L. E. Glendenin (see reference 25).
[b] 48 Mev.
[c] 300 Mev.
[d] These yields are read from smooth photoyield curves (Fig. 2).
[e] 100 Mev.
[f] A. C. Pappas and C. D. Coryell, Phys. Rev. 81, 329 (1951).
[g] The yield becomes 0.13 if 7.9 is assumed for the total chain yield.
[h] Obtained by repeated separations of I^{134}.

symmetric masses (112 through 126) are considerably greater than those of the thermal-neutron fission of U^{235} or the fast-neutron fission of U^{238}. The peak-to-trough ratio for 48-Mev maximum x-rays is 11.

4. Fine structure, which has been shown[22] to be present in the thermal-neutron fission of U^{235} at masses 100 and 134, may also be present in the x-ray fission of U^{238} at about the same masses. Tewes and James[12] have reported the existence of fine structure in the proton fission of thorium.

Analysis of the 7, 10, 16, 21, 100, and 300-Mev photofission results given in Tables II, III, IV (where the peak-to-trough ratios at various energies are reported) and Figs. 2 and 3 leads to the following observations:

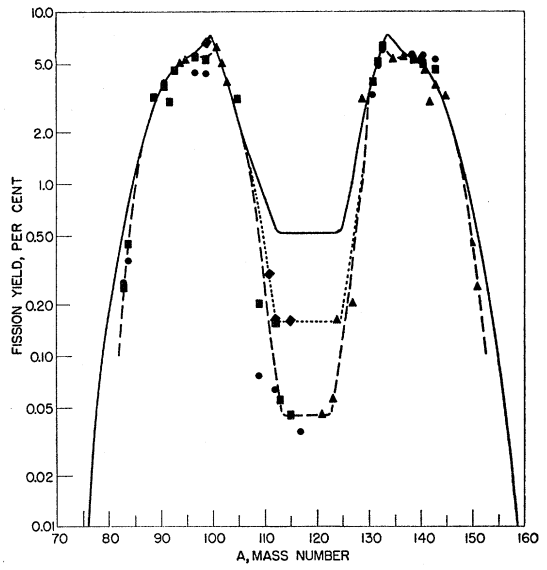

FIG. 3. Natural uranium photofission yield-mass curves. ●, 10-Mev and ■, 16-Mev photoyields by H. G. Richter (see reference 21). ▲, 16-Mev reflected photofission yields (arbitrarily assumed that the fragment mass-sum is 236 for masses 110 to 126 and 234 for all other masses). ◆, 16-Mev photoyields of this research. ———, 16-Mev curve of H. G. Richter, normalizing his curve to the 48-Mev photoyield of mass 139; – – – –, 16-Mev curve of this research, and ————, 48-Mev curve of this research (see Fig. 1).

1. The peak-to-trough ratios at 7, 10, 16, 21, 48, 100, and 300 Mev are: >300, 200, 38, 23, 11, 8, and 4, respectively. The lower limit to the 7-Mev ratio was obtained by assuming the same yield ratio of Pd^{112} to Cd^{115} at 7 Mev and 10 Mev.

2. Relative to the yield of Mo^{99} at 48 Mev (6.6 percent), the asymmetric peak yields of masses 89 through 103, and the yields of their complementary masses, for the x-ray energies 21 to 300 Mev hardly changed outside the experimental error.

3. The trough photoyields in U^{238} approach the thermal-neutron yields (0.01 percent) in U^{235} as the maximum x-ray energy decreases and approaches the threshold energy, 5.2 Mev.[34]

4. Irradiation of uranium by 100-Mev and 300-Mev x-rays caused no appreciable change in the "wings" of the photoyield curve, compared to the 48-Mev x-ray curve.

5. The ratio of Mo^{99} to Zr^{97} is constant for 21, 48, and 100-Mev x-rays. This result may indicate that fine structure is present at all three energies.

6. Construction of smooth yield curves near the troughs for 16 to 300 Mev x-rays results in a near-symmetric fragment mass sum of 237.

The 10-Mev peak-to-trough ratio of 200 observed in this work and that of 150 found by Richter[21] (Table IV) agree within experimental error. However, the corresponding 16-Mev ratios, 38 and 120, are in serious disagreement. The former ratio 38, as reported in this paper, appears more reasonable in the light of the work of Fowler *et al.*[7] and Turkevich *et al.*[8] who showed that the trough yield is markedly enhanced near the threshold for energy increments of a few Mev. This effect is mostly dependent upon the extent of nuclear excitation and is fairly independent of the incident particle and the heavy fissionable nucleus involved. The ~20-Mev and 21-Mev photo peak-to-trough ratios in U^{235} and U^{238} of 20 and 23, respectively, and the 48-Mev and

[34] Koch, McElhinney, and Gasteiger, Phys. Rev. 77, 329 (1950); R. E. Anderson and R. B. Duffield, Phys. Rev. 85, 728 (1952).

69-Mev ratios in U^{238} and Th^{232}, of 11 and 10, respectively, support this conclusion.

B. Quantitative Interpretation of X-Ray Fission in U^{238}

The observed photofission yield curves (Fig. 2) may be interpreted as the superposition of two components, a low-energy asymmetric (double-humped) curve and a high-energy single-humped curve, produced by the absorption of high-energy photons. This postulate was suggested by Turkevich *et al.*[14] in order to interpret the observed neutron-fission yield curves of Th^{232} and U^{238}. The decrease in the peak-to-trough ratio with increasing energy (Table IV) will be assumed to be the result of an increasing contribution of high-energy fission. The cross section for high-energy fission may be obtained by calculating the fraction of high-energy fission and by assuming the measured cross section for photofission at the resonance energy.

The low-energy curve adopted for this analysis is essentially the photoyield curve observed at 7 to 10 Mev (Figs. 2 and 3) but with a peak-to-trough ratio of 600. This value was chosen to be roughly the same[16,35] as that found with thermal neutrons and U^{233} or U^{235}. In these cases the fissioning nucleus is excited by about 1 Mev above the fission threshold of 5.2 Mev.[34] With 7-Mev x-rays most of the excitation is at about 1–2 Mev (because of the resonance nature of the photoabsorption) where the contribution from symmetric fission would be expected to be higher.[7,36]

The single-humped high-energy photofission yield curve may be calculated from the differences in the trough yields of the 48- and 100-Mev curves. The value obtained, 2.4, is equal to the percentage of high-energy fission induced by the photons of 48 to 100 Mev with 100-Mev maximum x-rays. If it is assumed that only symmetric fission occurs in this interval, then 97.6 percent of the fissions for 100-Mev x-rays occur in the energy interval up to 48 Mev.

TABLE IV. Peak-to-trough ratios at various x-ray energies.

Energy of x-rays, Mev	Nuclide irradiated	Peak-to-trough ratio	Reference
2.6, fission neutrons	U^{238}	100	Engelkemeir, Seiler, Steinberg, and Winsberg, see reference 16, paper 218.
7	U^{238}	>300	This paper.
10	U^{238}	200	This paper.
10	U^{238}	150	See reference 21.
16	U^{238}	38	This paper.
16	U^{238}	120	See reference 21.
~20	U^{235}	20	See reference 4.
21	U^{238}	23	This paper.
48	U^{238}	11	This paper.
69	Th^{232}	10	See reference 9.
100	U^{238}	8	This paper.
300	U^{238}	4	This paper.

[35] W. E. Grummitt and G. Wilkinson, Nature **161**, 520 (1948).
[36] P. Fong, Phys. Rev. **89**, 332 (1953).

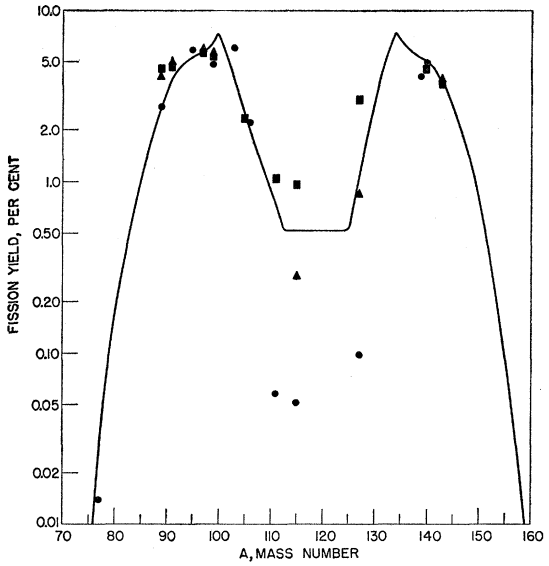

FIG. 4. X-ray and fast-neutron induced fission yields. ●, fission-neutrons and U^{238} (see reference 16); ■, 14-Mev neutrons and U^{235} (see reference 4); ▲, ~20-Mev x-rays and U^{235} (see reference 4). Solid curve is the normalized 48-Mev photofission curve of natural uranium of this research (see Fig. 1). The yields of reference 16 were normalized to the 48-Mev x-ray curve at mass 140 and those of reference 4 at mass 97.

The yields of masses 111, 112, 115, 117, and 127 of the high-energy yield curve, calculated from the percentage of fissions[37] occurring in the energy interval 48 to 100 Mev and the observed fission yields of the trough species at 48 and 100 Mev, are 11.8, 9.1, 10.0, 9.6, and 7.3 percent, respectively. The proposed high-energy symmetric fission-yield curve is presented in Fig. 5. A curve of similar shape and roughly of the same yield values is found by like treatment of the 100- and 300-Mev trough data. The curve was assumed to approach zero at masses 106 and 130 because of the approximate constancy of the yield values at masses 103, 106, 131, and 132 (Table II).

The half-width (full width at half-height) of the proposed high-energy symmetric curve of U^{238} is ~21 mass-units. Sugarman,[5] investigating the photofission of Bi^{209} with 85-Mev x-rays, reported a half-width of ~20 mass-units for the symmetric yield curve. This contrasted sharply with a half-width of ~40 mass-units, found by Goeckermann and Perlman,[3] who determined the 190-Mev deuteron-induced yield curve in Bi^{209}. O'Connor and Seaborg,[1] bombarding uranium with 380-Mev alpha-particles, found a similarly shaped symmetric distribution of the fission products with a half-width of ~40 mass-units. Perhaps the 40 mass-unit widths result from a superposition of many narrow symmetric curves like that of Fig. 5.

[37] The percentage of symmetric fissions changes from 2.4 to 2.2 when correction is made for the change in bremsstrahlung spectrum for 48- and 100-Mev maximum energies.

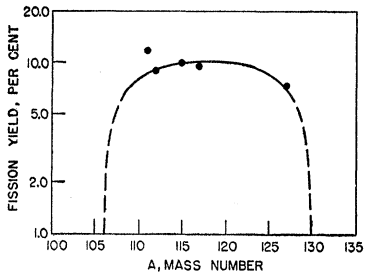

FIG. 5. Proposed high-energy symmetric fission-yield curve of U^{238}.

C. Average Cross Section for Symmetric Fission for Various X-Ray Intervals

The percentage of high-energy fission occurring in an energy interval may be obtained by subtraction of the trough values of the appropriate photoyield curves. This procedure was used earlier on the 48- and 100-Mev curves in order to obtain the high-energy symmetric fission-yield curve (Fig. 5). Corrections of the order of 10 percent are necessary for the change in the bremsstrahlung spectrum with energy. These data are given in column 2 of Table V where the energy intervals are given in column 1. The measured cross section[38] for the resonance absorption can be used to calculate the average cross section for high-energy fission for the various energy intervals. These values are given in column 3 of Table V, where it is seen that the cross section is roughly constant for photon energies above 16 Mev, with a value of about 7 millibarns. Thus, the assumption that was made on the analysis of the observed fission yield curve into low- and high-energy components leads to a total cross section for photofission that is constant at a value of about 8 millibarns from 48 to 300 Mev.

Gindler and Duffield[39] have made measurements on the photofissions of uranium in the energy ranges 10 to 22 and 125 to 300 Mev, by observation of the ionization from the fission fragments in scintillating crystals.

TABLE V. Symmetric fission data for various energy intervals.

Energy interval	Percentage of high-energy fission by subtraction of yield curves[a]	Average cross section for high-energy fission, barns	Average total fission cross section,[b] barns	Branching ratio for "symmetric fission"
5–10	0.2		0.03	
10–16	1.1		0.14	
16–21	1.0	0.006	0.06	0.1
21–48	2.4	0.006	0.02	0.3
48–100	2.2	0.007	0.02	0.3
100–300	5.2	0.009	0.05	0.2

[a] Subtracted values corrected for change in bremsstrahlung spectrum with energy.
[b] From upper curve of Fig. 6, taken from reference 39.

[38] R. B. Duffield and J. R. Huizenga, Phys. Rev. **89**, 1042 (1953).
[39] J. Gindler and R. B. Duffield, Phys. Rev. **94**, 759 (1954); J. Gindler, Ph.D. Thesis, University of Illinois, February, 1954 (unpublished).

Because of the absence of data from 22 Mev to 125 Mev two possible cross-section curves may be drawn depending upon the interpolated cross-section curve assumed for the unmeasured energy interval. These curves are given in Fig. 6. It is seen that the lower curve (dashed) agrees favorably with the cross-section value for the range 100 to 300 Mev given in column 3 of Table V, implying that high-energy fission is mostly symmetric. The upper curve, however, has cross-section values at high energy higher than those given in column 3 of Table V and are given in column 4. The values of the total cross section in the interval 21 to 300 Mev are about a factor of 3 greater than the average cross section as calculated on the assumption that only symmetric fission occurred in this interval. We may interpret the ratio of the cross section, found on the assumption that only symmetric fission occurred, to the total cross section as the branching ratio for "symmetric fission" (column 5).

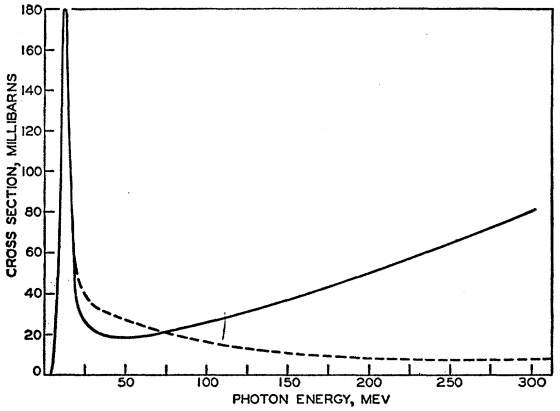

FIG. 6. Photofission cross section of U^{238}. Solid curve from 8 Mev to 19 Mev determined by Duffield and Huizenga (see reference 38); remainder of solid and dashed curves from 19 Mev to 300 Mev determined by J. Gindler (see reference 39).

Here, the term "symmetric fission" simply refers to the mass region 106 to 131.

A branching ratio of 0.3 for "symmetric fission" in the high-energy region implies that the fission-yield curve is still asymmetric, but with a small peak-to trough ratio. A nearly symmetric curve of 50 mass-unit width with a dip at symmetric fission corresponding to a peak-to-trough ratio of about 1.5 satisfies the experimental data.

D. Variation of Independent Fission Yields with Energy

Measurements of the independent fission yields of the nuclides, Br^{82}, Ag^{112}, I^{134}, and Cs^{136}, were made at various x-ray energies and are given in Table III. For comparison, the independent yields of these nuclides in the thermal-neutron fission of U^{235} are also presented.[25] The fraction of the chain yield measures the probability that fission leading to a given mass will result directly in the charge represented by the nuclide

studied. The fact that the values for Br^{82} and Cs^{136} from x-ray fission are higher than those from slow neutron fission of U^{235} means that the primary fission fragments from high-energy x-ray fission of U^{238} are formed closer to stability. These data, and the observed change in the ratio of the yields of the Cd^{115} isomers in the region 100 to 300 Mev, may be used to estimate the most probable neutron-to-proton ratio for a given mass. If it is assumed that the neutron-to-proton ratio does not change in high-energy fission,[3,40] the neutron-to-proton ratio of the fissioning nucleus for photons of 100 to 300 Mev is thus obtained. The assumption that the neutron-to-proton ratio of the primary fragments in high-energy fission is constant appears to be questionable in the light of recent experiments.[26]

The ratio of 54-hr Cd^{115} to 43-day Cd^{115m} for 300-Mev x-rays was found to be 5.7 as compared to 14 for x-rays below 100 Mev. A ratio of $\sim$14 is also observed[14,16] in the fission of U^{235} and Th^{232} with low-energy neutrons. If it is assumed that the ratio changed above 100 Mev because of the independent formation of Cd^{115} isomers, whereas below 100 Mev all of the Cd^{115} was formed from decay of Ag^{115}, then we may calculate the fraction of the chain formed at Cd^{115}. The ratio of 54-hr Cd^{115} to 43-day Cd^{115m} formed independently in high energy proton fission of heavy elements[26] (tantalum to gold) is 0.36. Using this ratio and an increase of 20 percent in the total fissions as the energy is increased from 100 to 300 Mev (upper curve of Fig. 6), then it is found that 7 percent of the chain of mass 115 is formed directly at cadmium. This fraction of the chain yield corresponds to a charge displacement of 1.7 from the most probable charge if the variation of fractional yield with charge displacement is the same as that found in thermal-neutron fission[24,25] of U^{235}. The neutron-to-proton ratio for the most probable charge of 46.3 for mass 115 is calculated to be 1.49. If the neutron-to-proton ratio of the fissioning uranium nucleus is taken to be 1.49, then it is found that there is associated with the fission of uranium with photons of 100 to 300 Mev, the emission of 9 neutrons. For an increase of 5 percent in the total fissions, such as is found if it is assumed that only symmetric fission occurs at high energy (Table V), calculations lead to 12 neutrons emitted.

The high fractional yield of the shielded nuclide Br^{82} with 300-Mev x-rays may be ascribed exclusively to the energy range 100 to 300 Mev. Below 48 Mev the yield is low, and only about 5 percent of the fissions with 300-Mev x-rays occur between 48 and 100 Mev where the yield may be expected to be intermediate between 48 and 300 Mev. The neutron-to-proton ratio found from the fractional yield of Br^{82} is 1.46, in good agreement with the value obtained from the Cd^{115} measurements.

The measurements at 48 Mev on I^{134} and Cs^{136} both lead to neutron-to-proton ratios of about 1.57. The fissioning nucleus of the same ratio would be U^{236}, which agrees well with the observed mass sum of 234 (Fig. 1) for the fission ascribed mainly to the 14-Mev resonance absorption. The higher independent yield of Cs^{136} for 48-Mev x-rays on U^{238} relative to thermal-neutron fission of U^{235}, where the fissioning nucleus in both cases is thought to be U^{236}, in this analysis is ascribed to a difference in the splitting of the nucleus. At higher energies, a constant neutron-to-proton ratio fission process results in a higher independent yield for Cs^{136}.

The upper limits to the independent yields of Ag^{112} at 48, 100, and 300 Mev are not inconsistent with values calculated from the neutron-to-proton ratios of 1.57 and 1.49 for the energy intervals 5 to 48 Mev, and 100 to 300 Mev, respectively. The normalized fission yield of Te^{132} decreases from 5.2 percent with 21-Mev x-rays to 4.0 percent with 300-Mev x-rays, whereas the yield of I^{131} for this energy interval is constant at 4.3 percent. This may be interpreted on the basis that I^{132} is being formed independently in increasing yield. The fractional yield of I^{132} found in the 100- to 300-Mev interval agrees well with that expected.

It is thus seen that there are increased yields of products closer to stability in the fission of U^{238} with x-rays of 48 Mev or greater. For 48-Mev x-rays, the increased yields can be explained by an unchanged neutron-to-proton ratio in the fission process. For 100–300 Mev x-rays, there is evidence for the emission of about 9 neutrons in the fission act.

ACKNOWLEDGMENTS

We wish to acknowledge the assistance of Mr. B. C. Cook and the Betatron Group, Mr. M. Fielding and the staff of the 37-inch cyclotron of the University of Chicago, and the personnel of the Argonne National Laboratory responsible for the thermal-neutron irradiations. The irradiations at the 22-Mev and the 340-Mev betatrons of the University of Illinois were accomplished through the generous cooperation of Professor D. W. Kerst, Mr. T. J. Keegan, Mr. T. B. Elfe, and the Betatron Groups.

[40] W. F. Biller, University of California Radiation Laboratory Report UCRL-2067, 1953 (unpublished).