Mass distributions in monoenergetic-neutron-induced fission of ^{239}Pu

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Fission product yields for 24 masses were determined for the fission of ^{239}Pu with essentially monoenergetic neutrons of 0.17, 1.0, 2.0, 3.4, 4.5, 6.1, and 7.9 MeV. Fission product activities were measured by Ge(Li) γ-ray spectrometry of irradiated ^{239}Pu targets and by chemical separation of the fission product elements followed by β counting. Yields of near symmetric (valley) fission products increase nearly monotonically with incident neutron energy and do not exhibit a break in the slope of the yield versus neutron energy curve in the energy region where second-chance fission begins. This is in contrast to the curves for ^{232}Th, ^{235}U, and ^{238}U neutron-induced fission.

[NUCLEAR REACTIONS, FISSION ^{239}Pu(n,f), E_n=0.17, 1.0, 2.0, 3.4, 4.5, 6.1, and 7.9 MeV; measured mass yields.]

I. INTRODUCTION

In previous papers[1-3] we reported reasonably complete mass distributions (37 to 44 mass yields) for monoenergetic-neutron-induced fission of ^{232}Th, ^{235}U, and ^{238}U for neutrons with energies from 0.17 MeV (^{235}U) or near the fission threshold (^{232}Th and ^{238}U) to $\sim$8 MeV. In the present paper we report the yields of 24 masses in the fission of ^{239}Pu with neutrons of 0.17, 1.0, 2.0, 3.4, 4.5, 6.1, and 7.9 MeV energy. Heretofore very little fission yield data were available for this nuclide other than the yields compiled for thermal, fission spectrum ("fast"), and 14-MeV ("high energy") neutrons.[4,5] Cuninghame and Willis[6] reported the yields of six fission products, ^{99}Mo, ^{111}Ag, ^{140}Ba, ^{147}Nd, ^{153}Sm, and ^{156}Eu, for the neutron energy range of 0.13–1.7 MeV. To our knowledge there are no other radiochemical or mass spectrometric fission yield data for the ^{239}Pu(n,f) reaction.

II. EXPERIMENTAL

A. Neutron irradiations

Targets for the neutron irradiations were 12.7-mm diameter by 0.127-mm thick disks of plutonium metal with an average weight of 0.21 g. The disks were prepared in the Isotope Research Materials Laboratory, Solid State Division, Oak Ridge National Laboratory. They were of two types, I used for γ-ray measurements and II used for the radiochemical measurements. The isotopic compositions of these two types are given in Table I. In addition, type I disks contained 700 ppm aluminum and type II disks contained $\sim$1 wt. % aluminum. ^{241}Am was also present in the disks in amounts of $\sim 10^9$ dis/min ($<$0.01 wt. %).

The targets were sealed in a flat, thin-walled aluminum container and irradiated for periods of 8 to 16 h at the Argonne Fast Neutron Generator Facility[7] in the manner described by Smith and Meadows.[8] The aluminum container was attached to a low-mass fission chamber containing a thin, standardized deposit of ^{239}Pu to monitor the fission rate. The ^{239}Pu standard, obtained from the National Bureau of Standards, was deposited over a circular area 12.7 mm in diameter on a 0.13-mm thick platinum plate 19.1 mm in diameter. Its isotopic composition was 99.10 at. % ^{239}Pu and 0.883, 0.010, 0.006 at. % ^{240}Pu, ^{241}Pu, and ^{242}Pu, respectively. The fission chamber was positioned $\sim$3 cm from the neutron source. Neutrons with energies $<$5 MeV were produced by the ^{7}Li(p,n)^{7}Be reaction. Neutrons of higher energy were produced by the ^{2}H(d,n)^{3}He reaction.

Details of the monoenergetic neutron beam

TABLE I. Isotopic composition of plutonium targets.

Pu isotope		at. %
Type I	238	$<$3 ppm
	239	99.955
	240	0.041
	241	0.002
	242	0.002
	244	$<$0.0003
Type II	239	98.8
	240	1.2

characteristics were given in a previous paper.[1] Spread in the principal neutron energy was 2—8 % for $E_n > 0.17$ MeV and 60% for $E_n = 0.17$ MeV. Fission rates in the target disks were typically 4×10^4 sec^{-1}. Other than the primary neutron beam, neutrons with lower energies also contributed to the fission rate. These neutrons were present from the ^{7}Li$(p,n)^7$Be* reaction, deuteron stripping reactions, and elastic and inelastic scattering from the room environment. Small corrections (1—15 %) were made for the effect of these secondary neutrons on the fission yields of masses that depend strongly on neutron energy ($A = 111$—127).

B. Fission yield determinations

Fission yields were determined by high-resolution γ-ray spectrometry of an irradiated plutonium target or by chemical separation of a fission product followed by β counting. These two methods are designated herein as the γ or RC-β method, respectively. Yields of ^{129}Sb were determined by both methods. The RC-β method was used to determine the yields of fission products with masses in the near symmetric (valley) region. The γ method was used for all other determinations.

For chemical separation of the fission products the irradiated plutonium metal targets were dissolved in concentrated hydrochloric acid containing $\sim 10\%$ concentrated nitric acid by volume. A hydrochloric acid solution of the carriers for the elements of interest was added followed by a drop of bromine to effect oxidation of antimony. Plutonium was extracted from this solution by contacting it twice with nearly equal volumes of 0.2 formal weight *bis*-2-ethylhexylphosphoric acid (HDEHP) in xylene. After extraction, sulfur dioxide was bubbled through the aqueous solution to reduce antimony. The fission products of palladium, silver, cadmium, tin, and antimony were then separated, purified, and mounted as samples for β counting following procedures described by Flynn.[9] The samples were counted in a calibrated low-background (0.5 counts/min) β proportional counter[9] equipped with an automatic sample changer. The radioactive purity of a sample was determined by following its decay over a period of several half-lives. Decay curves were analyzed with the least-squares computer program CLSQ.[10] The observed counting rate at the end of irradiation for each fission product was corrected for chemical yield, counting efficiency, decay, genetic relationships, and degree of saturation during irradiation to give the saturation activity A_∞.

For γ counting, the irradiated targets were mounted on aluminum plates and placed in a computer-controlled sample changer designed to give reproducible positioning of the samples. To reduce the intensity of the ^{241}Am 59.5-keV γ ray and thereby the dead time of the γ-ray spectrometer system, a 2.2 g/cm^2 steel absorber was placed between the sample and the detector. The latter was an 80-cm^3 lithium-drifted germanium detector with a resolution of 2.2 keV (FWHM) for the 1.33-MeV γ ray of ^{60}Co. Details of the γ-ray spectrometer and the γ counting method are given in Ref. 1. A large number (~ 40) of γ-ray spectra were recorded over a period of ~ 1 month to enhance statistical accuracy in the determination of the fission product activities. The spectra were analyzed with the computer program GAMANAL (Ref. 11) to obtain intensities of resolved photopeaks.

The measured fission product γ-ray activities as a function of time were analyzed with the decay program CLSQ (Ref. 10) to obtain activities at the end of irradiation. Further corrections were made for counting efficiency, cascade coincidence losses,[1] absolute γ-ray emission intensities,[2] genetic relationships, and degree of saturation to give the saturation activity A_∞. Values of A_∞ determined by either the RC-β or γ method are related to fission yields by the relationship

$$\text{fission yield} = A_\infty / \text{fission rate}.$$

In this work the fission rate was determined by counting the standardized sample of ^{239}Pu in the fission chamber as described previously. The uncertainty in this method is taken to be $\pm 5\%$.

III. RESULTS AND DISCUSSION

The results of fission product yield determinations are presented in Table II and shown graphically as mass-yield curves for several values of E_n in Fig. 1. Also shown for comparison in Fig. 1 is the mass distribution for 14-MeV neutrons based on data from Ref. 4. Uncertainties (1σ) in the fission yield values were obtained by consideration of all known sources of random and systematic error with the usual rules of error propagation. For peak fission yields ($> 1\%$) measured by the γ method, uncertainties fall typically in the range of 6—10 %. Larger uncertainties of about 15% are associated with the valley yields measured by the RC-β method, and of 20—50% with the independent yields of ^{134}I and ^{135}Xe. An assessment of possible error in determination of the mass yield due to direct formation in fission (independent yield) of chain members beyond the one measured was made from the energy-dependent charge distribution systematics of Nethaway.[12] The calculations show that the measured fission product yields over the E_n range of

TABLE II. Fission product yields of monoenergetic-neutron-induced fission of ^{239}Pu.

Fission product	Measurement technique	Incident neutron energy (MeV) 0.17	1.0	2.0	3.4	4.5	6.1	7.9
^{87}Kr	γ	0.76 ±0.10	0.83 ±0.09	0.93 ±0.09	1.04 ±0.11	1.38±0.11	0.81±0.15	0.93±0.16
^{88}Kr	γ	1.13 ±0.10	1.16 ±0.09	1.42 ±0.10	1.59 ±0.11	1.61±0.12	1.65±0.21	1.73±0.20
^{91}Sr	γ	2.72 ±0.16	2.71 ±0.15	2.94 ±0.17	3.18 ±0.18	3.24±0.18	3.07±0.22	3.03±0.19
^{92}Sr	γ	3.05 ±0.38	3.00 ±0.38	3.36 ±0.42	3.48 ±0.43	3.70±0.46	3.53±0.44	3.51±0.44
^{93}Y	γ	4.16 ±0.40	4.43 ±0.42	4.86 ±0.46	4.67 ±0.39	5.41±0.56	4.96±0.86	4.03±0.50
^{97}Zr	γ	5.67 ±0.32	5.37 ±0.33	5.83 ±0.33	5.84 ±0.39	5.93±0.35	5.99±0.65	5.13±0.52
^{99}Mo	γ	6.29 ±0.42	6.03 ±0.40	6.30 ±0.44	6.21 ±0.42	6.36±0.44		5.49±0.39
^{103}Ru	γ	7.57 ±0.50	5.61 ±0.54	6.97 ±0.47	6.68 ±0.48	5.83±0.95		5.66±0.39
^{105}Ru	γ	6.03 ±0.55	5.44 ±0.43	5.49 ±0.38	5.47 ±0.47	5.53±0.41	4.71±0.45	4.74±0.41
^{109}Pd	RC-β	1.14 ±0.17	0.94 ±0.14	1.10 ±0.17	1.12 ±0.17	1.41±0.21	1.21±0.18	1.16±0.17
^{111}Ag	RC-β	0.25 ±0.04	0.24 ±0.04	0.28 ±0.04	0.45 ±0.07	0.63±0.09	0.38±0.10	0.66±0.10
^{112}Pd	RC-β	0.14 ±0.02		0.16 ±0.02	0.26 ±0.04	0.36 ±0.05	0.47±0.07	0.59±0.09
^{115}Cd^{g}	RC-β	0.027±0.004	0.036±0.005	0.050±0.008	0.11 ±0.02		0.27±0.04	
^{121}Sn^{g}	RC-β	0.032±0.005	0.044±0.007	0.048±0.007	0.085±0.013	0.13±0.02	0.22±0.03	
^{125}Sn^{g}	RC-β	0.039±0.006	0.052±0.008	0.058±0.009	0.12 ±0.02	0.17±0.03	0.16±0.02	0.31±0.05
^{127}Sb	RC-β	0.34 ±0.05	0.37 ±0.06	0.35 ±0.05	0.76 ±0.11	0.77±0.12	0.82±0.12	1.08±0.16
^{129}Sb	γ, RC-β	1.18 ±0.24	1.20 ±0.24	1.27 ±0.25	1.77 ±0.35	1.65±0.33	1.58±0.32	1.67±0.33
^{132}Te	γ	5.52 ±0.38	5.11 ±0.35	5.33 ±0.51	4.99 ±0.28	5.37±0.56	5.07±0.52	4.92±0.39
^{133}I	γ	7.64 ±0.45	7.02 ±0.41	7.32 ±0.43	7.06 ±0.41	7.09±0.42	6.30±0.37	6.24±0.36
^{134}Te	γ	5.14 ±0.42	4.14 ±0.31	4.36 ±0.37	3.28 ±0.32	3.57±0.30	2.45±0.27	2.86±0.24
^{134}I[a]	γ	2.92 ±1.16	3.31 ±0.95	2.46 ±1.32	3.96 ±0.80	4.15±0.92	4.19±1.30	3.28±0.66
^{135}I	γ	6.89 ±0.37	6.29 ±0.34	6.67 ±0.38	6.43 ±0.37	6.39±0.39	5.40±0.31	5.21±0.29
^{135}Xe[a]	γ	0.85 ±0.37	0.92 ±0.34	0.73 ±0.61	1.15 ±0.31	1.58±0.54	2.43±0.84	1.83±0.48
^{140}Ba	γ	5.49 ±0.33	5.12 ±0.30	5.37 ±0.32	5.10 ±0.30	5.62±0.34	5.62±0.35	5.49±0.34
^{142}La	γ	4.88 ±0.41	4.49 ±0.37	4.92 ±0.44	4.50 ±0.36	5.00±0.40		
^{143}Ce	γ	4.57 ±0.36	4.40 ±0.35	4.44 ±0.34	4.22 ±0.33	4.15±0.33		

[a]Independent yield.

0–8 MeV represent essentially total (>90%) chain yields except for the independent yields of ^{134}I and ^{135}Xe. From the fission yields of the isomers ^{115}Cd^{g}, ^{121}Sn^{g}, and ^{125}Sn^{g} total chain yields may be estimated by using isomer ratios $(m+g)/g$ of 1.11±0.005 for ^{115}Cd^{g} (average value for several fissioning systems in Ref. 4), 1.16 for ^{121}Sn^{g},[13] and about 3.0 for ^{125}Sn^{g}.[4]

As with neutron-induced fission of ^{232}Th, ^{235}U, and ^{238}U,[1–3] the prominent features of the mass distributions shown in Fig. 1 are the strong dependence of fission yields in the valley mass region on E_n (increased probability of near-symmetric mass splits with increasing excitation energy) and the weak dependence of peak yields on E_n. These effects are illustrated in Fig. 2, where the yields of the valley fission products and a typical peak fission product (^{140}Ba) are plotted as a function of E_n. Also shown at the bottom of the figure is the cross section σ_F for neutron-induced fission of ^{239}Pu as a function of E_n. The arrow indicates the energy [6.4 MeV (Ref. 14)] where second-chance fission (n,nf) becomes possible. In contrast to the other fissioning systems, ^{232}Th(n,f), ^{235}U(n,f), and ^{238}U(n,f) studied at this laboratory, there is no apparent break in the slope $(d\ln Y/dE_n)$ of the yield vs E_n curves for ^{239}Pu(n,f) at the onset of second-chance fission.

The appearance of a break in the $\ln Y$ vs E_n curve depends on the relative cross sections of first- and second-chance fission in the energy region where second-chance fission becomes possible and on the magnitude of $d\ln Y/dE_n$ for the second-chance fissioning species, A_F-1, where A_F is the mass of the original or first-chance fissioning species. ($A_F=240$ in the present case.) The fission cross section σ_F for ^{239}Pu(n,f) increases about a factor of 1.3 between $E_n=5$ MeV and $E_n=8$ MeV. The fission cross section for ^{235}U(n,f) increases a factor of $\sim$1.5 between these two neutron energies. One may, therefore, assume that the relative increase in second- to first-chance fission for ^{239}Pu(n,f) is less than that for ^{235}U(n,f). Also the slope $d\ln Y/dE_n$ (or, more properly, $d\ln Y/dE_x$, where E_x is the excitation energy E_n+B_n, and B_n is the neutron binding energy) for the fission products shown in Fig. 2 is less in the energy region where only first-chance fission occurs than it is for the same fission products in ^{235}U(n,f). If one makes the assumption that $d\ln Y/dE_x$ for a

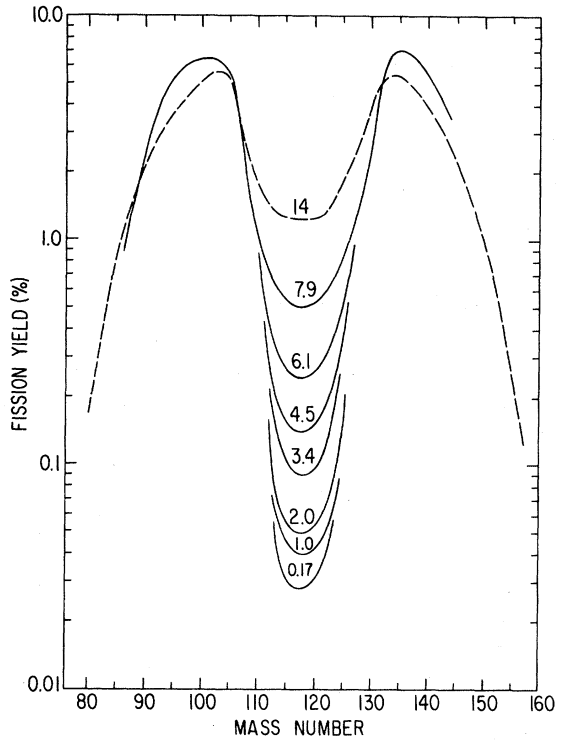

FIG. 1. ^{239}Pu(n_E,f) mass distributions. The solid curves represent the results of present measurements. The 14-MeV (dashed curve) data were taken from Ref. 4.

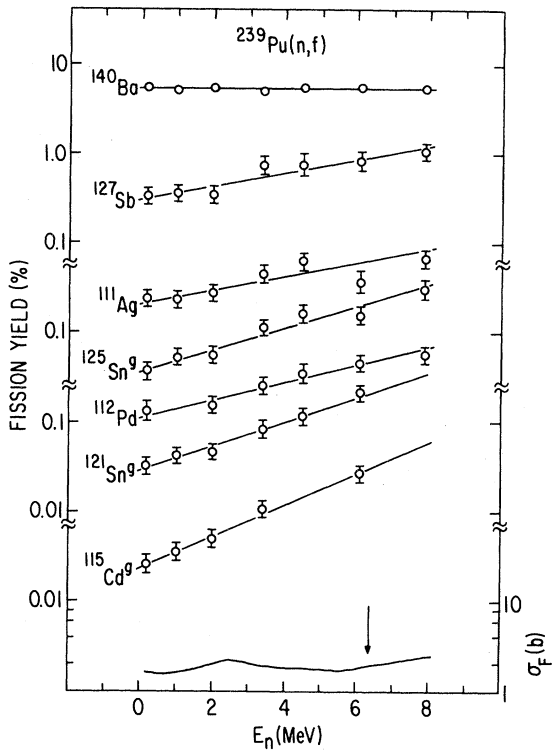

FIG. 2. Fission yields and cross section σ_F for fission of ^{239}Pu by monoenergetic neutrons as a function of neutron energy.

given fission product in the fission of ^{239}Pu or ^{235}U is the same, respectively, as $d\ln Y/E_x$ in the fission of ^{240}Pu or ^{236}U, then the emission of a neutron prior to fission will not cause as great a change in yield for the plutonium system as for the uranium system. Because of these two effects, a smaller second-to-first-chance fission cross section and smaller values of a $d\ln Y/dE_x$, it is less likely to observe any break in the $\ln Y$ vs E_n curves at the onset of second-chance fission for ^{239}Pu(n,f) than for ^{235}U(n,f). It should be pointed out, however, that there are fewer data points in this energy region for the present fissioning system than for others studied. Therefore, any small break in $d\ln Y/dE_n$ may have been overlooked.

Some characteristics of the mass distributions derived from the fission yield data for monoenergetic-neutron-induced fission of ^{239}Pu are given in Table III. The relative change in mean mass for the light and heavy groups as a function of E_n indicates that the increase in neutron emission with increasing excitation energy is greater from the heavy fragments. Values of $\bar{\nu}$, the average number of neutrons emitted per fission, calculated from the mean masses are in reasonable agreement with experimental values based on direct measurement by fission-coincident neutron counting,[15] with the exception of the value at 7.9 MeV.

The dissipation energy, that is, the energy transformed from collective energy into nucleonic

TABLE III. ^{239}Pu(n_E,f) mass distribution characteristics.

E_n	Peak to valley ratio	Mean mass (u) Light group	Mean mass (u) Heavy group	$\bar{\nu}^a$	$\bar{\nu}^b$
0.17	230	99.0	138.3	2.7	2.90
1.0	160	98.5	138.2	3.2	3.01
2.0	130	98.7	138.3	3.1	3.18
3.4	70	98.7	137.9	3.4	3.35
4.5	50	98.6	137.9	3.5	3.60
6.1	25	98.6	137.7	3.7	3.81
7.9	13	98.8	137.7	3.6	4.05

[a]Calculated from conservation of mass.
[b]Evaluated from experimental measurements by fission-coincident neutron counting (Ref. 15).

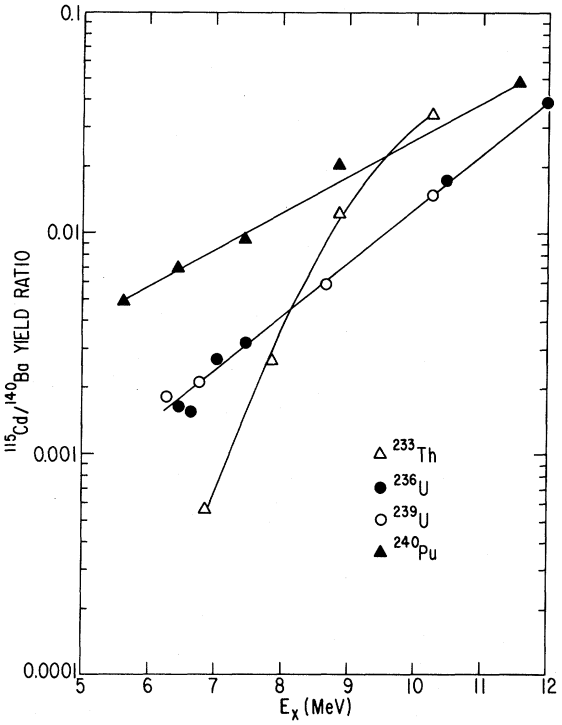

FIG. 3. The ratio of ^{115}Cd to ^{140}Ba yields as a function of excitation energy for the fission of ^{233}Th, ^{236}U, ^{239}U, and ^{240}Pu in the region where only first-chance fission occurs. The curves are merely to guide the eye.

excitation as the fissioning nucleus moves from its saddle to its scission configuration, has been deduced[16,17] from the ratio of ^{115}Cd to ^{140}Ba yields vs E_n or E_x for neutron-induced fission of ^{232}Th, ^{235}U, and ^{238}U. The magnitude of the dissipation energy has been found to be inversely correlated with the slope of the ^{115}Cd to ^{140}Ba yield ratio vs E_x in the energy region where only first-chance fission can occur. Because the slope of this yield ratio is smaller for $^{239}Pu(n,f)$ than for either of the other three fissioning systems (see Fig. 3), the dissipation energy for ^{240}Pu is expected to be greater. A detailed analysis for this energy has not yet been completed. However, a linear extrapolation of dissipation energies based on the fission parameter Z^2/A indicates it to be ~ 9 MeV for ^{240}Pu.

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