Distribution of Mass and Charge in the Fission of ^{245}Cm†

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The distribution of mass and charge in thermal-neutron-induced fission of ^{245}Cm has been investigated by radiochemical determination of the fission yields for 34 mass chains and 2 shielded nuclides (independent yields). ^{245}Cm is the heaviest nuclide fissionable by thermal neutrons to be investigated in this manner. The mass distribution is asymmetric; the heavy-mass group is centered at about the same position as in all previously investigated cases of low-energy fission, while the light-mass group is shifted to a higher mass number. The fission-yield data for ^{245}Cm indicate a value of 4 ± 1 for $\bar{\nu}$, the average number of neutrons emitted per fission. The division of nuclear charge, as indicated by the independent yields of the shielded nuclides, adheres to a pattern characteristic of low-energy fission. A comparison of mass distributions in thermal-neutron-induced fission, reactor-neutron-induced fission, and spontaneous fission for several fissioning nuclei is given.

I. INTRODUCTION

THE published data on mass and charge distributions in thermal-neutron-induced fission cover a range of 12 mass units and 4 charge units in the fissioning nucleus (i.e., ^{229}Th to ^{241}Pu). The mass distributions are predominantly asymmetric. Variation in mass of the fissioning nucleus is manifested by a shift in the light peak of the mass-yield curve, whereas the average mass of the heavy fragments is relatively independent of the mass of the fissioning nucleus. The division of nuclear charge in low-energy fission is characterized by a deviation of about 0.5 charge unit in the most probable charge Z_p of the fission fragment from the original charge-to-mass ratio (charge density) of the fissioning nucleus.

With the recent availability of ^{245}Cm in quantities sufficient for radiochemical studies it was possible to extend the range of information to 16 mass units and 6 charge units for further correlations of fission characteristics with mass and charge of the fissioning nucleus. The large cross section of 1950 b (measured values: 1880 ± 150 b [1] and 2010 ± 150 b [2]) for thermal-neutron-induced fission of ^{245}Cm was advantageous in this investigation since only very small quantities of ^{245}Cm were available for fission-yield determinations.

II. EXPERIMENTAL

The fission yields of 37 nuclides (in 34 mass chains) and of 2 shielded nuclides in ^{245}Cm fission were determined radiometrically relative to the known yields for ^{235}U fission. Small aliquots of a curium solution with an isotopic composition[3] of 96.50% ^{244}Cm, 1.60% ^{245}Cm, 1.87% ^{246}Cm, 0.0220% ^{247}Cm, and $\leq$0.0044% ^{248}Cm were used in all the experiments except for two of the rare-earth determinations in which a fragment-recoil source of mass-separated ^{245}Cm (enriched to ~84%) was irradiated. The concentration of the ^{245}Cm in this solution was shown to be constant over the course of these experiments by absolute α-counting of small aliquots (assuming that essentially all α activity was due to ^{244}Cm with a half-life of 18.1 years[4]). The amount of ^{245}Cm in the recoil source was measured by α pulse-height analysis. Aliquots of an enriched ^{235}U solution (93.18%) were used as reference standards for all the experiments. The concentration of uranium in this solution was determined gravimetrically by weighing U_3O_8 ignited at 850°C.

Known amounts of ^{245}Cm (0.1 to 0.3 μg) and ^{235}U (1 to 10 μg) were irradiated simultaneously in the Argonne CP-5 heavy water reactor at a thermal neutron flux of approximately 10^{13} sec^{-1} cm^{-2}. One irradiation was performed at the Savannah River Reactor at a thermal neutron flux of $\sim4\times10^{15}$. With the exception of the rare earths the specific fission products of interest were isolated from both samples using standard radiochemical procedures, i.e., carriers were added, the samples were radiochemically purified, and thick samples (~10 mg/cm^2) were mounted for counting. In order to avoid working with highly α-active samples the curium was removed by several co-precipitations with LaF_3 or $La_2(C_2O_4)_3$ after the addition of the carriers.

A carrier-free procedure was used to determine the yields of the rare earths. A Dowex 1-X4 (100–200 mesh) column with an elutriant of 10% 6N HNO_3–90% methanol was used to separate the rare earths from aluminum (resulting from dissolution of the samples) and from other fission products. The rare earths were then chromatographically separated by di-(2-ethylhexyl) orthophosphoric acid (HDEHP) on a chlorodimethylsilane-treated diatomaceous silica column[5] with HCl elutriant in stepwise increasing concentra-

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[3] R. E. Cote, R. F. Barnes, and H. Diamond, Phys. Rev. **134**, B1281 (1964).
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[5] J. W. Winchester, J. Chromatog. **10**, 502 (1963).

tions. The radioactivity of the effluent was followed with a sensitive plastic scintillator to permit collection of each rare earth in a separate fraction. The rare earths were normalized relative to ^{141}Ce and ^{143}Ce which were determined with carrier using the hexone extraction procedure.[6]

In a chromatographic column separation of the rare earths there is always a possibility of fractionation which would lead to systematic errors in the heavy wing of the mass-yield curve. A tracer experiment with $^{141/144}$Ce and ^{152}Eu showed no evidence of fractionation during the column separations and also proved that the rare earths were recovered with yields of better than 99%.

The purified samples were counted with an end-window β proportional counter and/or γ-ray spectrometer using a 3 in.×3 in. NaI(Tl) crystal. The rare-earth fractions were measured in solution by γ-ray spectrometry. Here, and in all other cases where γ-ray spectrometry was applicable, the decay of one or more characteristic γ photopeaks was followed. Composite decay curves were resolved graphically.

If the targets of ^{245}Cm and ^{235}U are irradiated simultaneously, and if the counting of a ^{245}Cm fission product is done relative to the same fission product in ^{235}U fission, the fission yield for ^{245}Cm is given by

$$Y_x(^{245}\mathrm{Cm})=\frac{A_x(^{245}\mathrm{Cm})N(^{235}\mathrm{U})\sigma(^{235}\mathrm{U})Y_x(^{235}\mathrm{U})}{A_x(^{235}\mathrm{U})N(^{245}\mathrm{Cm})\sigma(^{245}\mathrm{Cm})},$$

where $Y_x(^{245}\mathrm{Cm})$ is the yield of fission product x in ^{245}Cm fission, $Y_x(^{235}\mathrm{U})$ is the yield of fission product x in ^{235}U fission, $A_x(^{245}\mathrm{Cm})$ is the activity of fission product x in the ^{245}Cm sample, $A_x(^{235}\mathrm{U})$ is the activity of fission product x in the ^{235}U sample, $N(^{245}\mathrm{Cm})$ is the number of ^{245}Cm atoms in the irradiated sample, $N(^{235}\mathrm{U})$ is the number of ^{235}U atoms in the irradiated sample, $\sigma(^{245}\mathrm{Cm})$ is the cross section for fission of ^{245}Cm by thermal neutrons, $\sigma(^{235}\mathrm{U})$ is the cross section for fission of ^{235}U by thermal neutrons.

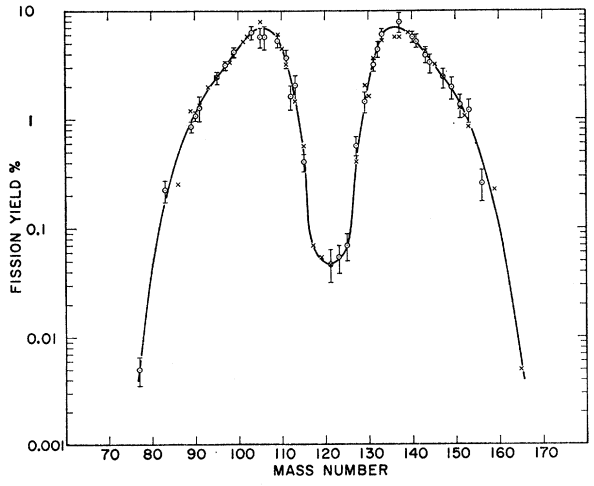

FIG. 1. Mass-yield curve for thermal-neutron-induced fission of ^{245}Cm. ○ measured points, × reflected points.

The activity of each sample was corrected only for chemical yield and relative amount of fissionable nuclide used in each irradiation since all other corrections essentially cancel in this comparison method.

III. RESULTS AND DISCUSSION

The fission yields for the 34 mass chains determined in this work are presented in Table I. Independent yields of the shielded nuclides ^{86}Rb and ^{136}Cs are considered separately (Table III). With a few exceptions the ^{235}U fission yields compiled by Katcoff[7] were used in calcula-

TABLE I. Fission yields for thermal-neutron-induced fission of ^{245}Cm.

Fission product	Method of counting	Number of determinations	Fission yield for ^{235}U (%)[a]	Fission yield for ^{245}Cm (%)
^{77}As	β,γ	3	0.0083	0.005±0.0012
^{83}Br	β	3	0.51	0.23 ±0.05
^{89}Sr	β	3	4.79	0.85 ±0.10
^{90}Sr	β	2	5.77	1.08 ±0.15
^{91}Y	β	1	6.11[b]	1.27 ±0.30
^{95}Zr	β,γ	3	6.2	2.40 ±0.30
^{97}Zr	β	2	5.9	3.10 ±0.35
^{99}Mo	β,γ	3	6.06	4.18 ±0.40
^{103}Ru	γ	9	3.0	6.27 ±0.90
^{105}Ru	β,γ	5	0.9	5.78 ±1.20
^{106}Ru	β	6	0.38	5.75 ±1.40
^{109}Pd	β	3	0.030	5.23 ±0.60
^{111}Ag	β,γ	6	0.019	3.63 ±0.70
^{112}Pd	β,γ	3	0.010	1.60 ±0.40
^{113}Ag	β	3	0.010[c]	2.02 ±0.50
^{115g}Cd	β,γ	4	0.0097[d]	0.41 ±0.07[e]
^{121g}Sn	β	5	0.015[d]	0.047±0.012[e]
^{123m}Sn	β	1	0.0013[d]	0.054±0.012[e]
^{125g}Sn	β	9	0.013[d]	0.060±0.015[e]
^{125}Sb	β,γ	4	0.021	0.071±0.015
^{127}Sb	β,γ	3	0.13	0.57 ±0.09
^{129}Sb	β,γ	3	0.8	1.42 ±0.30
^{129m}Te	β,γ	4	0.35[d]	1.48 ±0.20[e]
^{131}I	β,γ	3	3.1	3.18 ±0.40
^{132}Te	β,γ	4	4.7	4.41 ±0.80
^{133}I	β,γ	3	6.9	6.01 ±0.70
^{137}Cs	β,γ	7	6.15	7.89 ±1.60
^{140}Ba	β,γ	6	6.35	5.70 ±0.70
^{141}Ce	γ	6	6.4	5.20 ±0.70
^{143}Ce	γ	5	5.88[b]	3.85 ±0.60
^{144}Ce	γ	1	6.0	3.30 ±0.70
^{147}Nd	γ	3	2.21[b]	2.60 ±0.50
^{147}Pm	β	1	2.21[b]	2.03 ±0.50
^{149}Pm	γ	2	1.04[b]	1.97 ±0.40
^{151}Pm	γ	2	0.44	1.35 ±0.35
^{153}Sm	γ	3	0.159[b]	1.20 ±0.30
^{156}Eu	γ	3	0.0137[b]	0.25 ±0.06

[a] Yields are taken from the compilation in Ref. 7 unless another reference is given.
[b] From Ref. 8.
[c] Interpolated from smooth mass-yield curve (Ref. 7).
[d] Isomer yield.
[e] Total yield calculated by assuming the isomer ratio is the same in ^{245}Cm fissions as in ^{235}U fission.

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[7] S. Katcoff, Nucleonics 18, 201 (1960).

TABLE II. Comparison of mass distributions in low-energy fission.

Fissioning nucleus	Average mass number of fission product at half-maximum height: Light group	Heavy group	FWHM[a]	FWTM[b]	Peak-to-valley ratio	Reference
Thermal-neutron fission						
^{229}Th	86.5	141.5	12.5	19	500	c
^{233}U	93.5	138	14	22.5	390	d
^{235}U	95	138.5	14.5	22	650	d
^{239}Pu	99	138	14	24	150	d
^{241}Pu	~101	138	14	25	~30	e-g
^{245}Cm	104	138	14.5	27	130	this paper
Reactor-neutron fission						
^{232}Th	91	139	14	21	140	d
^{238}U	98	139	15	24	180	d
^{241}Am	102	139	14	24	150	h, i
Spontaneous fission						
^{238}U	~96	~140	13	20	...	j
^{240}Pu	101	137	13	21	>270	k
^{242}Cm	102	137	13	22.5	>700	l
^{252}Cf	106	142	15	27	750	m, n

[a] Full width at half-maximum.
[b] Full width at 1/10 maximum.
[c] N. Ravindran, K. F. Flynn, and L. E. Glendenin, J. Inorg. Nucl. Chem. **28**, 921 (1966).
[d] Reference 7.
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[l] E. P. Steinberg and L. E. Glendenin, Phys. Rev. **95**, 431 (1954).
[m] W. E. Nervik, Phys. Rev. **119**, 1685 (1960).
[n] L. E. Glendenin and E. P. Steinberg, J. Inorg. Nucl. Chem. **1**, 45 (1955).

tion of the yields for ^{245}Cm. The yields for most of the rare earths were based on ^{235}U yields given by Bunney and Scadden.[8] Fission cross sections of 1950 and 577 b[9] for ^{245}Cm and ^{235}U, respectively, were used in the computations. Errors in the measured fission yields are estimated to be in the range of 10 to 25% and include experimental errors as well as uncertainties in the values

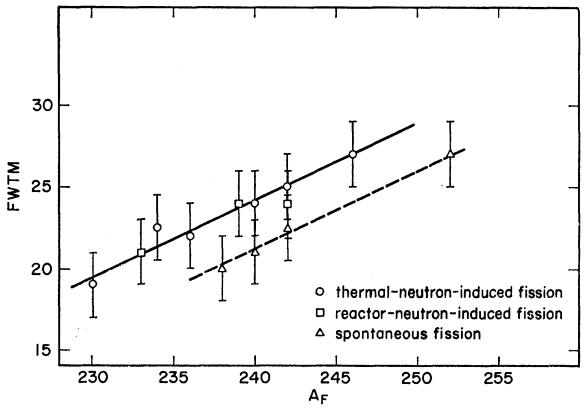

FIG. 2. Widths of mass distributions (FWTM = full width at one-tenth of maximum height) for various nuclides and different types of fission.

[8] L. R. Bunney and E. M. Scadden, J. Inorg. Nucl. Chem. **27**, 273 (1965).
[9] J. R. Stehn, M. D. Goldberg, R. Wiener-Chasman, S. F. Mughabghab, B. A. Magurno, and V. A. May, *Brookhaven National Laboratory Report No. 325* (U. S. Government Printing and Publishing Office, Washington, D. C., 1965), 2nd ed., Suppl. 2.

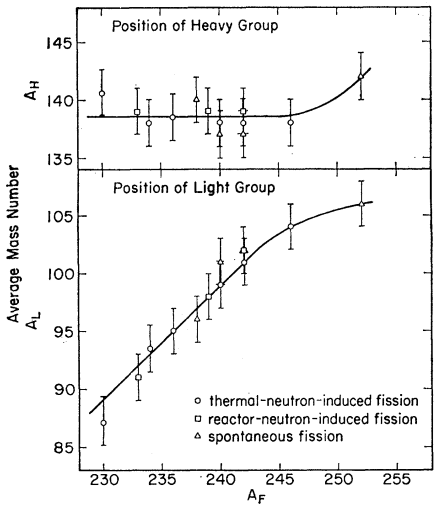

FIG. 3. Positions of light and heavy groups in the fission of various nuclides. The average mass number at half-maximum of the peaks is plotted.

of the fission yields for ^{235}U. Calculations showed that interference by neutron-induced fission products from other curium isotopes was negligible. Fission product separations from the unirradiated curium proved that the contribution from spontaneous fission was also negligible.

The observed fission yields from Table I are plotted as a function of fission product mass in Fig. 1. Within the estimated errors the observed data are consistent with the smooth mass-yield curve drawn through the experimental points. The data were put on an absolute basis by normalization of the curve to 200% total fission yield (2 fragments per fission). Summation of the mass-yield curve before normalization gave a value of 204% which is well within the experimental errors in the reported values of the fission cross sections. The measured points can be reflected through mass 121±0.5 (symmetric fission) indicating a value of 4±1 for $\bar{\nu}$, the average number of neutrons emitted per fission. To our knowledge no measurement of $\bar{\nu}$ for ^{245}Cm has been reported; however, its value can be estimated from fission energy systematics to be about 3.5.

The observed mass distribution is asymmetric with the light- and heavy-mass groups centered at mass 104 and 138, respectively. The relative probability of asymmetric to symmetric fission (peak-to-valley ratio) is about 130. The full width at one-tenth maximum (FWTM) of either mass group is approximately 27 mass units.

The observed characteristics of the ^{245}Cm mass distribution are compared with those for thermal-neutron-induced fission of ^{229}Th, ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu in Table II. Also included are the same characteristics for reactor- (~2 MeV) neutron-induced fission of ^{232}Th, ^{238}U, and ^{241}Am and for spontaneous fission of ^{238}U, ^{240}Pu, ^{242}Cm, and ^{252}Cf. Two significant trends are ob-

TABLE III. Independent fission yields and empirical Z_p values.

Shielded nuclide	^{86}Rb	^{136}Cs
Independent fission yield for ^{235}U (%)[a]	2.9×10^{-5}	6.8×10^{-3}
Independent fission yield for ^{245}Cm (%)	$(6.2\pm1.5)\times10^{-5}$	0.16 ± 0.03
Chain yield for ^{245}Cm (%)	0.45	7.0
Fraction of chain yield	$(1.4\pm0.35)\times10^{-4}$	$(2.3\pm0.4)\times10^{-2}$
Z_p(empirical)	34.22 ± 0.2	53.25 ± 0.15
Primary mass (A')[b]	86.8 ± 0.2	137.4 ± 0.1
$Z_p-A'(Z_F/A_F)$	0.35 ± 0.25	-0.37 ± 0.2

[a] Yields from compilation in Ref. 7.
[b] Neutron emission from primary fragments assumed to be the same as in spontaneous fission of ^{252}Cf [H. R. Bowman, *et al.*, Phys. Rev. **129**, 2133 (1963)].

served in all three types of low-energy fission: (1) The width of the mass distribution increases with increasing mass of the fissioning nucleus; (2) The change in mass is accompanied by a shift in the position of the light group, with the position of the heavy group remaining relatively fixed. In Fig. 2, it is seen that the full width at one-tenth maximum (FWTM) is a linear function of the mass of the fissioning nucleus A_F and is 3 mass units wider for thermal- and reactor-neutron-induced fission than for spontaneous fission.[10] This effect can probably be attributed to the higher excitation energies in neutron-induced fission, as higher excitation energies generally tend to broaden the mass distribution.

From Fig. 3 it is apparent that the position of the heavy-mass peak remains fixed at mass 138.5 ± 2 over a range in A_F of about 15 mass units, whereas the position of the light-mass peak is linear with A_F (slope of 1) over the same range. It is to be noted, however, that the heaviest nuclide ^{252}Cf, and possibly also the lightest nuclide ^{229}Th, depart from this pattern.

It will therefore be interesting to investigate the mass distributions for thermal-neutron-induced fission of ^{227}Th, ^{249}Cf, ^{251}Cf, and ^{254}Es as these nuclides become available in sufficient quantities.

The observed independent fission yields for the shielded nuclides ^{86}Rb and ^{136}Cs are given in Table III. The chain yields are taken from the smooth mass-yield curve of Fig. 1. Empirical values for the most probable charge Z_p were calculated from the fractional yields under the assumption[11] of a Gaussian charge dispersion with $\sigma=0.62\pm0.06$. A convenient quantity in the consideration of charge division is the deviation of Z_p from the value for the unchanged charge distribution (UCD) which is given by $Z_{UCD}=A'(Z_F/A_F)$, where A' is the primary mass of the fission fragment (before neutron emission) and Z_F/A_F is the charge-to-mass ratio (charge density) of the fissioning nucleus. The deviation is then $Z_p-A'(Z_F/A_F)$, or Z_p-Z_{UCD}, and has a value of about 0.5 charge unit (positive for light fragments and negative for heavy fragments) in all cases of spontaneous or low-energy neutron-induced fission ($E_n<14$ MeV).[12] The Z_p-Z_{UCD} values for ^{86}Rb and ^{136}Cs (Table III) are 0.35 ± 0.25 and -0.37 ± 0.2, respectively, indicating a pattern of charge division consistent with other cases of low-energy fission.

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[10] It should be noted that this effect is hardly discernible if the comparison is made on the basis of the full width at half-maximum (FWHM) (Table III, column 4).

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