Distribution of Mass and Charge in the Spontaneous Fission of ^{244}Cm†

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The distribution of mass and charge in the spontaneous fission of ^{244}Cm has been investigated by radiochemical and mass spectrometric determination of the fission yields for 40 mass chains and 2 shielded nuclides (independent yields). The mass distribution is highly asymmetric with a relative probability of asymmetric-to-symmetric fission (peak-to-valley ratio) of greater than 5700. The average masses of the light and heavy groups are 103.1 and 138.7, respectively. The characteristics of the mass distribution, as well as the division of nuclear charge indicated by the independent yields of the two shielded nuclides, are consistent with the systematics established from other studies of low-energy fission.

I. INTRODUCTION

The distribution of mass in spontaneous fission (sf) has been studied for ^{238}U,[1] ^{240}Pu,[2] ^{242}Cm,[3] and ^{252}Cf.[4] Of these measurements only the work on ^{252}Cf provides sufficient data for a complete mass-yield curve. Since the characteristics of the ^{252}Cf mass distribution deviate somewhat from the systematic behavior of other low-energy fissioning systems,[5] the characteristics of other spontaneously fissioning systems are of some interest. The nuclide ^{244}Cm was available in sufficient quantities for such a study, and the fission yield of Xe isotopes for systems in the region of mass 244 are of cosmological interest.[6]

II. EXPERIMENTAL

A sample containing about 8 mg of curium (characteristics given in Table I) was molecular-plated[7] onto a thick aluminum plate over an area of 20 cm^2. This plate, containing 1.5×10^{12} α particles/min and 2×10^{6} sf/min, served as a master source from which the recoiling fission fragments were collected on 2-mil aluminum catcher foils. The catcher-foil method was used in order to avoid handling the highly-α-active material. Of the total sf events in the source 97.5% came from ^{244}Cm (Table I). The remaining 2.5% of the fissions came from ^{246}Cm and for the purpose of these measurements were considered to have negligible effect on the results.

With the exception of the rare earths and the noble gases, the specific fission products of interest were isolated by standard radiochemical techniques; i.e., the catcher foils were dissolved in the presence of carriers, each element was radiochemically purified, and thick samples (~10 mg/cm^2) were mounted for β and/or γ counting.

A carrier-free procedure was used to determine the relative yields of the rare earths. This procedure consisted of extraction chromatography using di- (2-ethylhexyl) orthophosphoric acid (HDEHP) sorbed on Celite (diatomaceous earth).[8,9] The rare-earth yields thus obtained were normalized by way of the cerium isotopes (^{141}Ce and ^{143}Ce) which were also determined with carriers using the hexone extraction procedure.[10]

The relative yields of the isotopes of krypton and xenon were determined by mass spectrometric analysis. The aluminum catcher foils were placed in double-walled Vycor tubes and attached to a vacuum system. The pressure of the system was reduced to 10^{-7} mm of Hg and the foils were melted. After cleaning the gases on titanium foil at 850°C the krypton and xenon were released into the mass spectrometer as separate fractions by selective retention of xenon on charcoal at −40°C. Standard air samples were analyzed by the same procedure before and after each foil analysis in order to determine the sensitivity and the mass discrimination of the spectrometer. The instrument used for these determinations has been described by Reynolds.[11] The detailed procedure

TABLE I. Isotopic composition and decay characteristics of curium source.

Nuclide	% mass	α half-life	Spontaneous-fission half-life	Spontaneous-fission (%)
242	0.000	163 day	7.2×10^{6} yr	...
243	0.055	32 yr	...	...
244	94.48	18.1 yr	1.25×10^{7} yr	97.5
245	1.95	8265 yr	...	...
246	3.45	4711 yr	1.80×10^{7} yr	2.5
247	0.057	1.6×10^{7} yr	...	...
248	0.014	3.84×10^{5} yr	4.22×10^{6} yr	...

TABLE II. Relative isotopic abundances for krypton and xenon formed in spontaneous fission of ^{244}Cm.

Krypton		Xenon				
	Relative abundance		Relative abundance			
Isotope	foil No. 3	Isotope	Foil No. 1	Foil No. 2	Foil No. 3	Average
83	1.000	131	0.718 ± 0.015	0.687 ± 0.016	0.715 ± 0.012	0.707 ± 0.008
84	1.428 ± 0.428	132	1.000	1.000	1.000	1.000
85	2.648 ± 0.273	133	···	1.342 ± 0.030	1.364 ± 0.022	1.353 ± 0.019
86	2.485 ± 0.314	134	1.647 ± 0.025	1.595 ± 0.036	1.638 ± 0.028	1.627 ± 0.017
		135	···	1.756 ± 0.040	···	1.756 ± 0.040
		136	1.802 ± 0.029	1.726 ± 0.036	1.817 ± 0.032	1.782 ± 0.019

and method of data reduction have been reported by Alexander and Manuel.[12] The relative krypton and xenon yields were normalized to the radiochemically determined yields.

In several experiments ratios of the activities of various fission products were determined without chemical separation by measuring the γ-ray spectrum from the catcher foils with a 17-cm^3 coaxial Ge(Li) detector. These spectra were computer analyzed to determine the relative intensity of the various γ rays. This method of analysis has been described in detail by Gordon, Harvey, and Nakahara.[13]

The β and γ detectors used in these experiments were calibrated against standard sources which had been assayed by absolute counting techniques. Wherever possible a standard source of the specific isotope being determined was used. In the cases where this was not possible the efficiencies were determined from the known energy of the

TABLE III. Fission yields for spontaneous fission of ^{244}Cm.

Fission product	Method of determination	Number of determinations	Fission yield (%)	Fission product	Method of determination	Number of determinations	Fission yield (%)
^{77}As	β	4	0.0025 ± 0.0009	^{127}Sb	β	3	0.20 ± 0.02
^{83}Br	β	2	0.116 ± 0.012	^{129}Sb	β	3	0.74 ± 0.07
^{83}Kr	ms	1	0.116 ± 0.012 [a]	^{129m}Te	β	6	0.47 ± 0.05 [b]
^{84}Kr	ms	1	0.166 ± 0.034 [a]	^{131}I	β, γ	8	3.15 ± 0.32
^{85}Kr	ms	1	0.307 ± 0.031 [a]	^{131}Xe	ms	3	3.03 ± 0.15 [d]
^{86}Kr	ms	1	0.289 ± 0.045 [a]	^{132}Te	β, γ	18	4.27 ± 0.22
^{89}Sr	β	5	0.716 ± 0.036	^{132}Xe	ms	3	4.27 ± 0.22 [d]
^{90}Sr	β	4	0.808 ± 0.081	^{133}I	β, γ	7	7.45 ± 1.50
^{91}Sr	β	5	0.825 ± 0.083	^{133}Xe	ms	2	5.78 ± 0.29 [d]
^{95}Zr	β, γ	7	1.63 ± 0.16	^{134}Xe	ms	3	6.94 ± 0.35 [d]
^{97}Zr	β, γ	8	3.04 ± 0.30	^{135}I	β, γ	6	5.48 ± 1.10
^{99}Mo	β, γ	8	3.75 ± 0.20	^{135}Xe	ms	1	7.50 ± 0.38 [d]
^{103}Ru	γ	5	6.94 ± 0.70	^{136}Xe	ms	3	7.59 ± 0.38 [d]
^{105}Ru	β, γ	5	9.43 ± 0.94	^{137}Cs	β, γ	7	8.14 ± 0.81
^{105}Rh	γ	2	9.06 ± 0.91	^{139}Ba	γ	2	7.16 ± 0.72
^{106}Ru	β, γ	5	7.40 ± 0.74	^{140}Ba	β, γ	10	6.04 ± 0.32
^{109}Pd	β	4	5.02 ± 0.50	^{141}Ce	β, γ	21	5.79 ± 0.29
^{111}Ag	β	16	2.87 ± 0.14	^{143}Ce	β, γ	23	3.90 ± 0.20
^{112}Pd	β, γ	16	2.03 ± 0.20	^{144}Ce	β, γ	11	4.04 ± 0.40
^{113}Ag (5.3 h)	β	8	0.79 ± 0.08 [b]	^{147}Nd	γ	3	1.44 ± 0.29
^{115g}Cd	β	4	0.233 ± 0.012 [b]	^{149}Nd	γ	1	1.54 ± 0.15
^{115m}Cd	β	3	0.025 ± 0.002 [b]	^{149}Pm	β	1	1.66 ± 0.17
^{115} (total chain)			0.258 ± 0.013	^{151}Pm	β	1	0.70 ± 0.07
^{121g}Sn	β	1	<0.0012 [b]	^{153}Sm	β	1	0.52 ± 0.05
121 (total chain)			<0.0014 [c]	^{156}Sm	β	1	0.23 ± 0.03
^{125g}Sn	β	6	0.0067 ± 0.0013 [b]	^{156}Eu	β	1	0.19 ± 0.02
125 (total chain)			0.0168 ± 0.0029 [c]				

[a] Kr yields normalized at mass 83 to 0.116% (^{83}Br).
[b] Isomer yield.
[c] Total chain yield determined by using the isomer ratio from ^{235}U(n,f).
[d] Xe yields normalized at mass 132 to 4.27% (^{132}Te).

radiation by interpolation from calibration curves of efficiency versus energy.

III. RESULTS AND DISCUSSION

The relative yields of the krypton and xenon isotopes as measured by mass spectrometry (ms) are given in Table II. The isotopes of krypton occur in relatively low fission yield and are therefore rather difficult to measure. This accounts for the large errors associated with the values for the krypton isotopic abundances. The xenon isotopes occur in high fission yield and hence can be determined with much greater precision and correspondingly smaller errors. The xenon abundances are based on an average of three independent determinations.

The fission yields for the 40 mass chains determined in this work are presented in Table III. The independent yields of the shielded nuclides ^{86}Rb and ^{136}Cs are considered separately in Table IV. The mass spectrometrically determined yields of the krypton isotopes have been normalized at mass 83 to the radiochemically determined yield for ^{83}Br, and the xenon yields have been normalized at mass 132 to the yield for ^{132}Te. In the cases of ^{121g}Sn and ^{125g}Sn the measured isomer yields were converted to total chain yields by using the isomer ratios from ^{235}U(n,f) as determined experimentally by Erdal *et al.*[14,15] Since the yield of the 1.2-min isomer of ^{113}Ag is not known, no attempt was made to convert the measured yield for the 5.3-h isomer of ^{113}Ag to a total chain yield. At mass 129 the total chain yield is taken to be the yield of ^{129}Sb; the yield for ^{129m}Te is reported only as an isomer yield. The error for each fission yield was evaluated on the basis of the number of determinations, the precision of the measurements, and any uncertainties in the decay scheme and counting efficiency of the particular nuclide being measured. With the assumption that charge division and dispersion are given by the quantities $|Z_p - Z_{UCD}| \simeq 0.8$ and $\sigma = 0.56 \pm 0.06$ (see Table IV and discussion of independent yields of ^{86}Rb and ^{136}Cs), the cumulative fractional chain yields for the fission products in Table III are calculated to be >0.95 with the following exceptions: ^{112}Pd (~0.83), ^{135}I (~0.90), and ^{156}Sm (~0.87).

The fission yields from Table III are plotted as a function of fission-product mass in Fig. 1. A mass-yield curve for the sf of ^{252}Cf is also shown for comparison. The data were put on an absolute basis by normalizing the curve to 200% total fission yield. The observed mass distribution is asymmetric with the mean masses (first moments) of the light and heavy groups located at mass 103.1 and 138.7, respectively. The relative probability of asymmetric to symmetric fission (peak-to-valley ratio) is greater than 5700. The full width of the peaks is 12 amu at half maximum (FWHM) and 23.5 amu at 1/10 maximum (FWHM). The fission-yield data for the light and heavy groups are reflection symmetric around mass 121 ± 0.5, indicating a value of 2 ± 1 for the average number of neutrons emitted per fission. These characteristics of the mass distribution are consistent with the systematics of low-energy fission.[16]

The observed independent fission yields for the shielded nuclides ^{86}Rb and ^{136}Cs are given in Table

TABLE IV. Independent fission yields and empirical Z_p values.

Shielded nuclide	^{86}Rb	^{136}Cs
Independent fission yield (%)	0.0018 ± 0.0004	0.13 ± 0.02
Total chain yield (%)	0.291 ± 0.045	7.72 ± 0.40
Fraction of chain yield	0.0062 ± 0.0017	0.0168 ± 0.0027
Z_p (empirical)	35.08 ± 0.1	53.30 ± 0.1
Primary mass (A') [a]	86.9 ± 0.1	137.3 ± 0.1
$\Delta = Z_p - A'(Z_F/A_F)$	0.89 ± 0.2	-0.72 ± 0.2

[a] 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).]

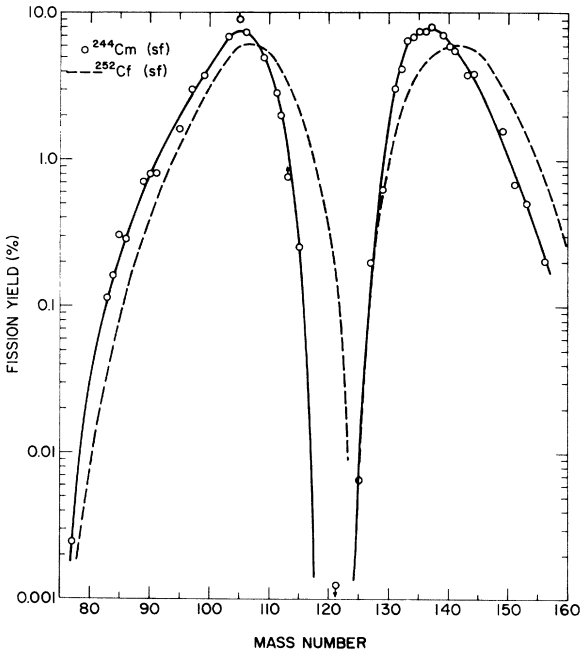

FIG. 1. Mass-yield curve for the spontaneous fission of ^{244}Cm compared with that for spontaneous fission of ^{252}Cf. Postneutron masses are shown.

IV. 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 of a Gaussian charge dispersion with $\sigma = 0.56 \pm 0.06$ as recommended by Wahl *et al.*[17,18] A convenient quantity in the consideration of charge division is the difference Δ of Z_p from the value for unchanged charge density (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 difference Δ is then $Z_p - A'(Z_F/A_F)$ and has a value of about 0.45 charge units (positive for light fragments and negative for heavy fragments) in most cases of spontaneous or low-energy neutron-induced fission ($E_n < 14$ MeV) which have been studied.[5,18-21] The Δ values determined in this work for ^{86}Rb and ^{136}Cs are 0.91 and −0.72 charge units, respectively. These values are significantly larger than 0.45. Recent measurements[21] of charge distribution in fission using high-resolution x-ray techniques and an analysis[21] of radiochemical data on cumulative yields of Kr and Xe isotopes also indicate a larger value of ~0.66 for Δ in thermal-neutron fission of ^{239}Pu.

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