Distribution of Mass in the Spontaneous Fission of ^{256}Fm†

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The distribution of mass in the spontaneous fission of ^{256}Fm has been investigated by the radiochemical determination of the fission yields for 28 mass chains. The mass distribution is asymmetric with a peak-to-valley ratio of ~12. The average masses of the light and heavy groups are 111.9 and 141.0, respectively. The fission-yield data indicate a value of 3 ± 1 for $\bar{\nu}$, the average number of neutrons emitted per fission. A comparison of the characteristics of the mass distribution for low-energy fission of nuclides ranging from ^{227}Th to ^{256}Fm is given.

I. INTRODUCTION

Recent physical studies of the fragment kinetic energy and mass distribution in spontaneous fission (sf)[1,2] and in thermal-neutron-induced fission (n,f)[2] of ^{257}Fm have indicated a marked departure toward mass symmetry from the asymmetric mass distribution typical of low-energy fission. It is therefore of interest to study other fissioning systems in this transition region. With the recent availability of ^{256}Fm in sufficient quantity (through ^{4}He ion irradiation of ^{253}Es), it was decided to determine the mass distribution for ^{256}Fm (sf) by the radiochemical method. This method eliminates the mass-resolution problem and the need for a mass-dependent correction for the number of neutrons emitted by the fission fragments. Both of these effects were important in the interpretation of the physical data from Refs. 1 and 2.

II. EXPERIMENTAL

The 2.62-h ^{256}Fm sources used for this study were prepared by 3-h irradiations of 10- to 100-μg ^{253}Es targets with 40-MeV ^{4}He ions at the Argonne National Laboratory cyclotron to produce the 1.25-h ^{256}Md parent. The ^{256}Md was then purified by a rigorous chemical-separation procedure[3] designed to provide the required decontamination (>10^{10}) from target materials and fission products produced in the cyclotron irradiation. The ^{256}Md was separated from Es, Fm, and most of the fission products by extraction chromatography using di(2-ethylhexyl)orthophosphoric acid (HDEHP) sorbed on Celite (diatomaceous earth). The procedure is essentially the same as that described in Ref. 3; however, a new slurry packing-pressure method[4] was used to prepare the columns. This technique enabled columns containing beds of 2.8 mm diam × 5 cm length to be operated at flow rates as fast as 8 ml/cm^2 min with high resolution. After loading the column, approximately 2 min were required to separate Md from Es and Fm. A single HDEHP column separation decontaminated the ^{256}Md from all fission products (except Dy, Ho, and Y) and from Es by factors of 10^3 to 10^4 and from Fm by 10^2. Three or four successive column separations were employed in order to give over-all decontamination factors for ^{253}Es of 10^9 to 10^{12}. In addition, the purified ^{256}Md (in $2N$ HCl solution) was passed through another extraction chromatography column containing tricaprylmethylammonium chloride sorbed on Celite, which provided additional decontamination from such fission products as Mo, Ag, Cd, Sn, Sb, Te, and I. The over-all decontamination from fission products (except Dy, Ho, and Y) was >10^{10}. Approximately 1 h was required for the chemical purification including the time required to dissolve the Es target.

After removal of small aliquots for fission counting, the purified sources of ^{256}Md were allowed to stand overnight, decaying as follows to the spontaneously fissioning ^{256}Fm:

$$^{256}Md \xrightarrow{92\%\ EC} {}^{256}Fm \xrightarrow{92\%\ SF} \text{fission products}.$$

Each source yielded about 10^7 total spontaneous-fission events. The specific fission products of interest were then isolated, and thick samples (10 mg/cm^2) were mounted for β counting. Fission yields were obtained for 28 fission products based on initial β activities ranging from 1 to 100 counts/min. The β counting was done in calibrated low-background (0.4-counts/min) counting equipment. The radioactive purity of each sample was verified by following its decay over several half-lives. The observed counting rate for each fission product was converted to the number of atoms after

correction for chemical yield, counter efficiency, and decay. Since the fission products are formed by the decay of ^{256}Fm which grows from ^{256}Md, the following three-membered Bateman equation was used to calculate the fission yields:

$$N^t_{FP} = FY \times N^0_1 \times \lambda_E \lambda_F \left[\frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_{FP} - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_{FP})(\lambda_{FP} - \lambda_2)} + \frac{e^{-\lambda_{FP} t}}{(\lambda_1 - \lambda_{FP})(\lambda_2 - \lambda_{FP})} \right],$$

where the subscript FP refers to the fission product, 1 refers to ^{256}Md, 2 refers to ^{256}Fm, E refers to the electron-capture branch of ^{256}Md, and F refers to the spontaneous-fission branch of ^{256}Fm. The time designation t in this equation is the interval between the time of separation of the fission product from the ^{256}Md-^{256}Fm fissioning source and the zero time for ^{256}Fm growth. N is the number of atoms of the particular nuclide, and FY is the fission yield of the particular fission product. Decay constants and branching ratios were taken from the work of Horwitz and Bloomquist,[3] and Hoff *et al.*[5] The number of atoms of ^{256}Md in each source at zero time, N^0_1, was determined by fission counting (Fig. 1) of an aliquot (1%) evaporated on a tantalum disk.

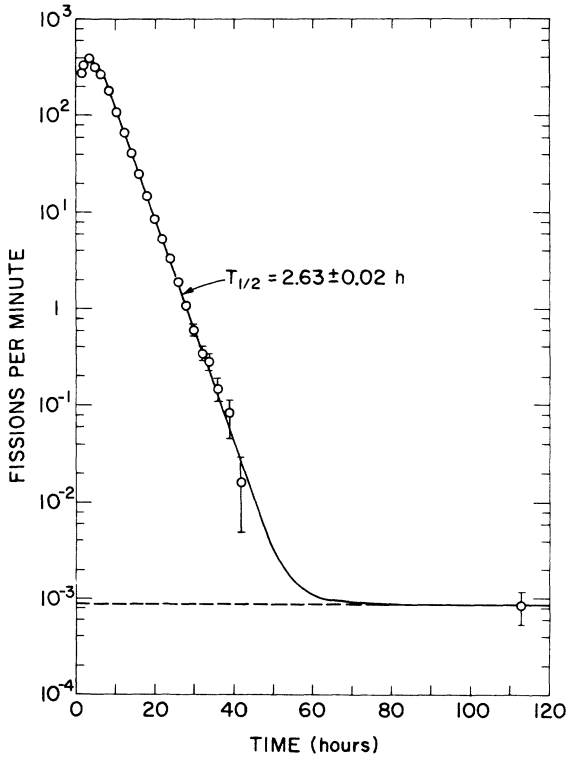

FIG. 1. Growth and decay of the spontaneous fission of ^{256}Fm from the parent ^{256}Md (1% aliquot).

III. RESULTS AND DISCUSSION

The purity of the ^{256}Md-^{256}Fm spontaneous fissioning source was confirmed by following the growth and decay of the fission rate. A typical curve (Fig. 1) shows no contaminating fission activity except for the expected low-intensity tail due to the daughter ^{252}Cf. A least-squares analysis of the data from two independent determinations gave a weighted average half-life for ^{256}Fm of 157.63 ± 1.27 min, in good agreement with the previously reported value[5] of 157 ± 2 min.

The fission yields for the 28 mass chains determined in this work are presented in Table I and plotted as a mass-yield curve in Fig. 2. The error for each fission-yield value was evaluated independently on the basis of the number of measurements, the statistical error in the count-rate de-

TABLE I. Fission yields for spontaneous fission of ^{256}Fm.

Fission product	Number of determinations	Fission yield (%)
^{91}Sr	3	0.24 ± 0.03
^{97}Zr	1	0.75 ± 0.15
^{105}Ru	2	3.1 ± 0.4
^{109}Pd	1	3.6 ± 1.0
^{111}Ag	4	5.4 ± 0.4
^{112}Ag	4	4.7 ± 0.6
^{113}Ag (5.3 h)	4	4.0 ± 0.3 [a]
^{115g}Cd	2	5.6 ± 0.5 [a]
115 Total chain		6.0 ± 0.6 [b]
^{118}Cd	1	5.7 ± 1.0
^{121g}Sn	2	2.3 ± 0.2 [a]
121 Total chain		2.7 ± 0.3 [b]
^{125g}Sn	2	0.6 ± 0.2 [a]
125 Total chain		1.5 ± 0.5 [b]
^{127}Sb	1	0.44 ± 0.08
^{129}Sb	1	0.88 ± 0.15
^{131}I	1	2.6 ± 0.5
^{132}Te	3	3.9 ± 0.5
^{133}I	1	3.9 ± 0.8
^{135}I	1	5.4 ± 1.0
^{139}Ba	2	6.2 ± 0.8
^{140}Ba	2	6.0 ± 0.5
^{141}La	1	7.1 ± 1.4
^{143}Ce	3	6.1 ± 0.5
^{145}Pr	1	5.2 ± 1.0
^{149}Pm	2	2.7 ± 0.4
^{151}Pm	2	2.1 ± 0.3
^{153}Sm	1	1.4 ± 0.3
^{156}Sm	1	0.7 ± 0.2
^{157}Eu	2	0.53 ± 0.07
^{159}Gd	2	0.50 ± 0.07

[a] Isomer yield.

[b] Total chain yield calculated from isomer ratio for ^{235}U(n, f).

termination, and any uncertainties in the decay scheme of the particular nuclide measured.

Since the ratio of the fission yields of the two ^{115}Cd isomers has been well established from ^{235}U(n,f) studies,[6] and is expected to be about the same for ^{256}Fm(sf), the measured yield of the 53.5-h ^{115g}Cd was converted to the total chain yield using the known isomer ratio $[(m+g)/g = 1.072 \pm 0.02]$. Although the isomer ratios for ^{121}Sn and ^{125}Sn are not as well established, the available data from ^{235}U(n,f) were also used in these cases to convert the observed isomer yields to total chain yields. The isomer yield ratio for ^{121}Sn has been reported[7] as 1.156 ± 0.05, and the measured fission yields of the ^{125}Sn isomers[7,8] give an isomer ratio of 2.5 ± 0.2. Since the yield of the 1.2-min isomer of ^{113}Ag has not been measured, the total chain yield for mass 113 could not be estimated. The deviation of the yield measured for the 5.3-h ^{113}Ag isomer from the smooth mass-yield curve would indicate, however, that about 30% of the total chain yield decays by way of the 1.2-min ^{113}Ag isomer to ^{113}Cd.

Assuming that charge division and dispersion in ^{256}Fm(sf) are approximately the same as in other low-energy fissioning systems (i.e., $Z_p - Z_{UCD} \simeq 0.5$ and $\sigma = 0.56 \pm 0.06$), the cumulative fractional chain yields for the fission products in Table I are calculated to be >0.95 with the following exceptions: ^{118}Cd (~0.75), ^{125}Sn (~0.90), ^{129}Sb (~0.80), ^{132}Te (~0.90), ^{135}I (~0.80), and ^{156}Sm (~0.82). No charge-distribution correction was made for these cases in plotting the mass-yield curve (Fig. 2).

Summation of the mass-yield curve for ^{256}Fm gives a value of 212% (105% for the light group and 107% for the heavy group). Since the deviation of this value from 200% is within experimental error, no attempt was made to normalize the data to 200%. The observed mass distribution for ^{256}Fm(sf) is definitely asymmetric, with the mean masses (first moments) of the light and heavy groups located at 111.9 and 141.0, but with a peak-to-valley ratio of only about 12. This enhancement of the probability of fission in the region of mass

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

Fissioning nucleus	Mean mass (amu) Light group	Mean mass (amu) Heavy group	Peak width (amu) Half maximum	Peak width (amu) Tenth maximum	Symmetric yield (%)	Peak-to-valley ratio	Reference
			Thermal-neutron-induced fission				
^{228}Th	90.0	137.0	11.5	19	0.035	230	9
^{230}Th	87.4	140.6	12	18	0.017	500	10
^{234}U	93.3	138.2	14.7	22.5	0.015	440	11
^{236}U	94.9	138.6	15	21.8	0.0105	620	11
^{240}Pu	98.9	138.1	15.3	24.5	0.04	150	11
^{242}Pu	100.4	138.8	14.5	24	0.029	230	11
^{243}Am	100.9	139.1	16	25	0.02	350	12
^{246}Cm	102.8	139.2	13	26.5	0.045	155	13
^{250}Cf	106.1	138.9	16	28	<0.2	>30	14
			Reactor-neutron-induced fission				
^{232}Pa	91.1	139.0	13	22	0.07	100	15
^{233}Th	91.2	140.0	14	20	0.045	170	11
^{234}U	92.9	138.1	14	21	0.06	110	11
^{236}U	95.2	138.5	15	23	0.032	205	11
^{238}Np	97.1	138.5	14	22	0.04	175	16
^{239}U	97.2	139.1	16	24	0.04	160	11
^{240}Pu	99.1	138.1	15	24.5	0.06	115	11
^{242}Am	100.7	139.0	14	24	...	...	17
			Spontaneous fission				
^{238}U	94.9	140.6	13	20	<0.012	>600	18
^{240}Pu	100.0	137.8	13	21	<0.03	>250	19
^{242}Cm	101.6	137.5	13	22.5	<0.01	>800	20
^{244}Cm	103.1	139.2	12	23.5	<0.0014	>5700	21
^{252}Cf	105.9	142.1	15	27.5	≤0.009	≥650	22
^{256}Fm	111.9	141.0	16	29	~0.5	~12	This work

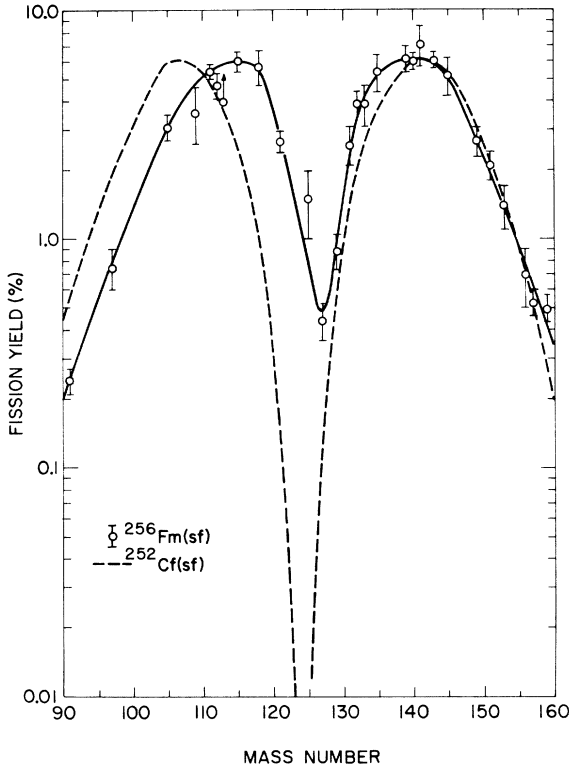

FIG. 2. Mass-yield curve for the spontaneous fission of ^{256}Fm compared with that for spontaneous fission of ^{252}Cf.

symmetry is clearly seen in Fig. 2 by comparison with the mass distribution for ^{252}Cf(sf). The full width of the peaks is 16 amu at half maximum (FWHM) and 29 amu at $\frac{1}{10}$ maximum (FWTM). The fission-yield data for the light and heavy groups are reflection symmetric around mass 126.5 ± 0.5 indicating a value of 3 ± 1 for the average number of neutrons emitted per fission.

The observed characteristics of the ^{256}Fm(sf) mass distribution are compared in Table II[9-22]

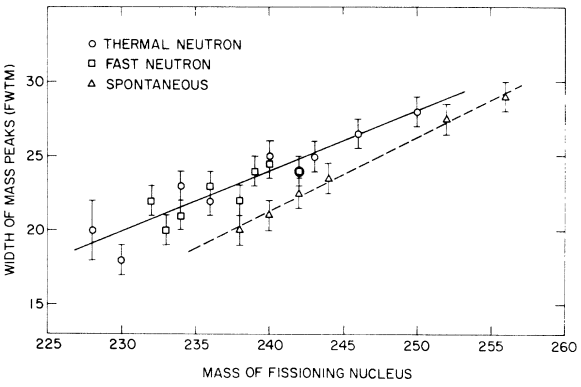

FIG. 3. Widths of mass distributions as a function of the mass of the fissioning nucleus.

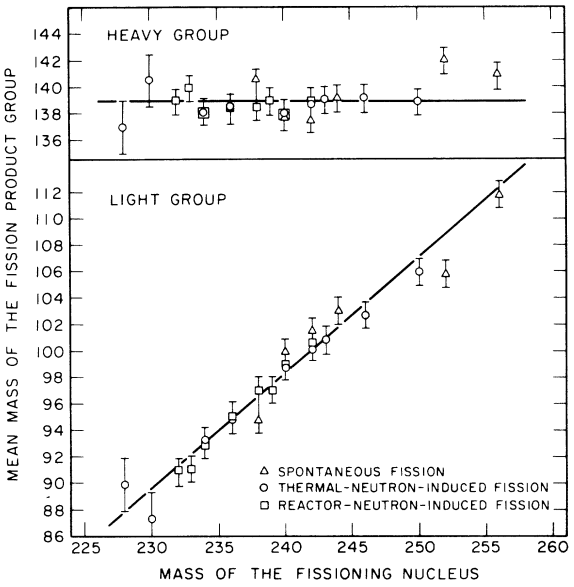

FIG. 4. Average masses of light and heavy groups as a function of the masses of the fissioning nucleus.

with those of other low-energy fissioning systems determined radiochemically. Peak width (FWTM) versus mass of the fissioning nucleus (A_F) is shown in Fig. 3 from which it is apparent that the previously reported[13] linear increase in peak width as a function of A_F continues through mass 256. The mean masses of the fission product groups as a function of A_F are shown in Fig. 4. The data for ^{256}Fm(sf) are seen to be consistent with the systematic pattern, wherein the average heavy-group mass remains approximately fixed at mass 139 ± 1, while the light-group mass increases linearly with A_F. [It is to be noted, however, that ^{252}Cf(sf) deviates from this systematic pattern; i.e., the mean light mass is significantly lower and the mean heavy mass is significantly higher than would be predicted.]

Although the observed width and positions of the light and heavy groups for ^{256}Fm(sf) are as expected from extrapolation of the trends observed in the lighter asymmetric fissioning systems, the marked decrease in peak-to-valley ratio clearly suggests the approach toward the predominantly symmetrical fission reported[2] for ^{257}Fm(n,f). An investigation is now underway to determine the mass distribution for ^{255}Fm(n,f) by the radiochemical method.

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(^{3}He, d) Stripping to Unbound Analog States in Tc Isotopes

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A systematic study of 14 unbound analog states in Tc isotopes was performed via the ^{92,94,96}Mo(^{3}He, d) reactions. Proton form factors were calculated using a simple single-particle resonance model. The proton spectroscopic factors deduced with the distorted-wave Born approximation agree well with the known neutron spectroscopic factors for the parent states obtained from (d, p) reactions. We conclude that this simple model is adequate for the calculation of spectroscopic factors to unbound analog states in these isotopes.

Recently there has been a growing interest in the problem of proton-stripping reactions to unbound analog states. The conclusions obtained by McGrath *et al.*[1] suffered from the lack of distorted-wave Born-approximation (DWBA) calculations for unbound states. Since then, a few approaches to the problem were suggested by several authors,[2-6] and some experimental results are available from both (^{3}He, d) and (d, n) studies.[1,2,7] It is the purpose of this letter to report the results of a systematic study of 14 unbound analog states in Tc isotopes, which were populated via the (^{3}He, d) reaction. A simple method for the analysis of the results, which takes into account the unbound nature of these states, is proposed and tested against all our experimental results with very good agreement.

A 30.2-MeV ^{3}He beam from the Saclay cyclotron