Thermal-Neutron Fission of ^{242m}Am: Mass and Charge Distribution*

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35 chain yields from thermal-neutron-induced fission of ^{242m}Am were measured by a recoil-catcher method, using ^{235}U fission as a standard. Fractional cumulative yields of five krypton and of six xenon nuclides were measured by an emanation method. The results are in agreement with known trends.

INTRODUCTION

Studies of low-energy neutron-induced fission of ^{242m}Am are of interest because of the very high neutron fission cross section, 7600 ± 300 b,[1] the unusually high spin of the compound nucleus, $\frac{9}{2}-$ or $\frac{11}{2}-$, and because the compound nucleus is an odd-even nucleus. Americium enriched in ^{242m}Am was prepared by Hoff and co-workers[2] for cross-section measurements, and its availability made our mass-yield and cumulative-yield measurements possible.

The catcher-foil method was used to conserve the scarce ^{242m}Am and to avoid the handling of highly α-active solutions. Mass yields were usually measured by radiochemical assays of the catcher foils and in a few cases by γ counting of catcher foils. Known yields from thermal-neutron-induced fission of ^{235}U served as comparison standards. Fractional cumulative krypton- and xenon-yield measurements were made with the emanation technique.[3]

The results of our measurements agree with trends indicated in von Gunten's review article.[4]

EXPERIMENTAL

Preparation and Composition of Foils

Thin sources of americium and uranium were prepared by electroplating their oxides from oxalate solutions onto 2.2-cm-diam circles on 5-mil platinum plates. The americium was a portion of the enriched ^{242m}Am prepared by Hoff and co-workers.[2] The isotopic composition of the americium source was determined by a combination of α-pulse analysis and mass spectrometry on small portions of the material; final assay was accomplished by α-particle counting in a low-geometry zinc sulfide scintillation counter.[5] The isotopic composition of the uranium had been determined by mass spectrometry, and the material had a well-determined specific α activity.[5] Assay of the uranium foil was completed by measurement of its α activity in a 2π proportional counter. Composition of the foils is given in Table I. The estimated accuracy of the assay is 3% for the americium foil and 0.5% for the uranium foil.

Irradiations and Measurements

For the determination of mass yields the two foils were placed back to back in an aluminum container with each foil facing a 2-mil aluminum catcher foil. The container was irradiated in port 4W of the Los Alamos water boiler reactor in a thermal-neutron flux of $\approx 4 \times 10^{11}$ cm^{-2} sec^{-1} for periods varying from 5 min to 4 h. During the first irradiation after preparation of the foils, $\approx$0.4% of the americium was transferred to the catcher foil by recoil or mechanical means; after six irradiations, this decreased to $\approx$0.1%. Fission-product analyses were performed on the catcher foils; the fissile foils were reused. The catcher foils were usually dissolved separately in hydrochloric acid. Most fission products were determined by adaptations of standard radiochemical procedures.[6]

In the case of iodine and bromine assays[7] exchange was insured by having the carriers present during dissolution in hydrochloric acid.[8] The γ

TABLE I. Composition of foils.

Nuclide	Americium foil (μg)	σ_f (b)	Nuclide	Uranium foil (μg)	σ_f (b)
^{242}Cm	0.32	<5	^{234}U	0.15	<0.65
^{241}Am	148.9	3	^{235}U	232.8	569
^{242m}Am	37.05	7600	^{236}U	0.18	...
^{243}Am	1.31	<0.07	^{238}U	0.29	<0.5

activity of short-lived iodine products was measured on a sodium iodide scintillation counter with a single-channel analyzer gated between 760 and 1900 keV. The β activities of ^{133}I and ^{131}I were measured through a 35-mg cm^{-2} aluminum absorber.[8]

In several experiments ratios of the activities of various fission products were determined without chemical separation by measuring their γ rays directly from the catcher foils in a standard geometry with a 4-cm^2 × 7-mm planar Ge(Li) detector. The method of analysis was essentially the same as that of Gordon, Harvey, and Nakahara.[9] Comparison of some of these results with radiochemical results is given in the Appendix.

Fractional cumulative yields of krypton and xenon nuclides from ^{242m}Am fission were determined by the emanation method[3] using barium or praseodymium stearates. The experimental details and treatment of the data are identical to those used in the determination of fractional yields of rare gases from thermal-neutron fission of ^{233}U and ^{239}Pu.[10]

Calculations

Calculation of chain yields from ^{242m}Am involved the comparison[11,12] of activities of late members of fission-product β-decay chains from the two catcher foils. The yield of ^{140}Ba from ^{242m}Am fission was established as a standard yield in the first two irradiations, while the assays of the foils were still reliable. In the following equations, unprimed terms refer to ^{235}U fission, and primed terms to ^{242m}Am fission. The subscript s refers to the standard fission product ^{140}Ba and the subscript x refers to any other fission product. The terms A, Y, N, and σ indicate the activity of a fission product, the fission yield of that product, the number of fissile atoms in the target foil, and the fission cross section of the target nuclide, respectively. The yield of ^{140}Ba from ^{242m}Am fission is given by

$$Y'_s = Y_s A'_s N\sigma/(A_s N'\sigma') . \quad (1)$$

In all subsequent irradiations, analyses for ^{140}Ba were performed along with those of other fission products. We define R as follows:

$$R = A'_x A_s/(A_x A'_s) . \quad (2)$$

Then the yield of fission product x is given by

$$Y'_x = Y_x Y'_s R/Y_s . \quad (3)$$

Somewhat shorter fission-product β-decay chains are expected from thermal-neutron fission of ^{242m}Am than from ^{235}U because ^{242m}Am has relatively more protons than ^{235}U. Except for $A = 135$ and 134 analyses were done for sufficiently late members of decay chains to obtain essentially the entire chain yield for both types of fission. ^{135}I has a fractional cumulative yield of 0.96 (see compilation[13]) for ^{235}U fission; approximately 0.92 is predicted for ^{242m}Am fission by the formulation discussed later. Appropriate corrections to the yield of this chain were included in the calculations. For the $A = 134$ chain a typical experiment involved separating 52.3-min ^{134}I from its 43-min ^{134}Te parent 45 min after a 30-min irradiation. The fractional independent yield of ^{134}I from ^{235}U fission is 0.11. This value is considered abnormal[13] relative to the predicted value of 0.24, and is attributed to the fact that $^{134}_{81}$I is one neutron removed from the 82-neutron shell. We would predict a "normal" ^{134}I independent yield of 0.33 for ^{242m}Am but revise this to 0.18 because of the shell effect. From the values 0.11 and 0.18 for the ^{134}I independent yields for ^{235}U fission and ^{242m}Am fission, respectively, we calculate and use a correction factor of 1.01 for our length of irradiation and separation times. The ^{134}I yield from ^{242m}Am is divided by this correction factor. A value of 0.33 for the independent yield of ^{134}I in ^{242m}Am fission would give a correction factor of 1.03. Thus the final results are quite insensitive to the assumed fractional independent yield of ^{134}I.

RESULTS AND DISCUSSION

Chain Yields

Chain yields for thermal-neutron fission of ^{242m}Am as well as the values of R from which they are derived are tabulated in Table II. The uncertainties in the values of R involve only the uncertainties from fission-product analyses; those in the chain yields also include uncertainties in the assay of the foils and the cross sections, and in the case of the 134 and 135 chains uncertainties in estimates of independent yields. The yields for ^{242m}Am fission, superimposed on the ^{235}U fission mass-yield curve, are shown in Fig. 1. With reasonably interpolated values for yields of unmeasured masses, we obtain values of 100.7 and 99.0% for the sum of the yields of the light and heavy peaks, respectively, for thermal-neutron fission of ^{242m}Am.

The immediately apparent differences between thermal-neutron fission of ^{242m}Am and ^{235}U are expected. The positions of the heavy peaks are about the same, while the light peak for ^{242m}Am fission is at higher mass numbers to accommodate the higher mass number of the fissioning nuclide. The individual peaks for fission of the heavier nuclide are also wider.

The average mass number of the light peak at half-maximum yield is 101.6; that of the heavy

peak is 138.5. The full width of the peaks at $\frac{1}{10}$-maximum yield is 25 mass units, and that of the valley at the same yield is 13 mass units. These values agree with the trends indicated in von Gunten's review article.[4] (The values of the parameters for ^{242m}Am listed in that article are based on our preliminary results.) The peak-to-valley ratio, ≈350, is greater than that for thermal-neutron fission of other nuclides except for ^{229}Th and ^{235}U, which have values of 530 and 600, respectively.

The mass-yield curve in the mass 131 to 135 region for ^{242m}Am fission is fairly smooth. Mass 134 does not have the abnormally high yield that it has for ^{235}U fission.

Fractional Yields of Rare Gases

Experimentally determined values of fractional yields of krypton and xenon nuclides are given in Table III. Values of Z_P, the most probable charge for an isobaric chain, are calculated from the

TABLE II. Yields from thermal-neutron fission of ^{242m}Am.

Fission product	No. of determinations Radiochemical	No. of determinations γ-spectral	R	Chain yield ^{235}U (%) [a]	Chain yield ^{242m}Am (%)
^{83}Br	2		0.536 ± 0.012	0.52	0.237 ± 0.018
^{84}Br	2		0.445 ± 0.010	0.97	0.367 ± 0.027
^{89}Sr	2		0.291 ± 0.004	4.76	1.18 ± 0.08
^{90}Sr	2		0.283 ± 0.005	5.83	1.40 ± 0.10
^{91}Y	4		0.347 ± 0.017	5.90	1.74 ± 0.12
^{92}Sr	2		0.403 ± 0.011	5.98	2.05 ± 0.15
^{93}Y	2		0.473 ± 0.011	6.39	2.57 ± 0.19
^{95}Zr	2		0.589 ± 0.013	6.41	3.2 ± 0.2
^{97}Zr	2	2	0.84 ± 0.01	6.21	4.5 ± 0.3
^{99}Mo	4	2	1.02 ± 0.02	6.16	5.4 ± 0.4
^{103}Ru		2	2.71 ± 0.17	3.0	6.9 ± 0.5
^{109}Pd	2		130. ± 6.	0.030	3.3 ± 0.3
^{111}Ag	2		92. ± 2.	0.019	1.48 ± 0.11
^{112}Ag	2		57.8 ± 0.7	0.010	0.49 ± 0.04
^{115}Cd	2		8.0 ± 0.1		
^{115m}Cd	2		7.8 ± 0.3		
Total 115			8.0 ± 0.1	0.0104	0.071 ± 0.005
^{121}Sn	2		1.68 ± 0.04	0.014 [b]	0.0200 ± 0.0015
^{125}Sn	2		3.26 ± 0.09	0.027 [b]	0.075 ± 0.006
^{125}Sb	2		4.8 $^{+0.5}_{-1.9}$	0.027 [b]	0.11 $^{+0.01}_{-0.05}$
^{131}I	2	2	1.28 ± 0.04	2.91	3.2 ± 0.2
^{132}Te	2	2	1.12 ± 0.04	4.26	4.1 ± 0.3
^{133}I	2	2	1.02 ± 0.02	6.69	5.8 ± 0.4
^{134}I	2		0.90 ± 0.02	7.8	6.0 ± 0.4 [c]
^{135}I	2	3	1.18 ± 0.04	6.43	6.8 ± 0.5 [c]
^{136}Cs	2		2.4 ± 0.9	0.006 [d]	0.12 ± 0.05 [d]
^{137}Cs	2		1.11 ± 0.03	6.20	5.8 ± 0.7
^{139}Ba	2		1.02 ± 0.02	6.48	5.6 ± 0.4
^{140}Ba	2		"1"	6.34	5.39 ± 0.38
^{141}Ce	4	1	1.03 ± 0.03	6.1	5.3 ± 0.4
^{143}Ce	2		0.86 ± 0.02	5.91	4.3 ± 0.3
^{144}Ce	4		0.79 ± 0.02	5.40	3.6 ± 0.3
^{147}Nd	2		1.24 ± 0.04	2.19	2.31 ± 0.18
^{149}Pm	2		1.82 ± 0.04	1.04	1.61 ± 0.13
^{151}Pm	2		3.28 ± 0.07	0.43	1.20 ± 0.09
^{153}Sm	2		5.79 ± 0.14	0.158	0.78 ± 0.06
^{156}Eu	2		24.9 ± 0.8	0.0134	0.28 ± 0.02
^{157}Eu	2		28.3 ± 1.6	0.0066	0.159 ± 0.014
^{161}Tb	2		265. ± 11.	0.000082	0.0185 ± 0.0014

[a] Unless otherwise noted, ^{235}U fission yields are from Ref. 14.
[b] From Ref. 15.
[c] Values include correction for differences in fractional cumulative yields. See text.
[d] Independent yield.

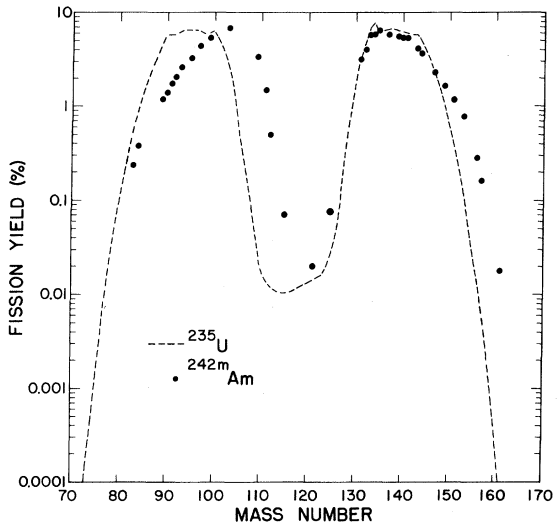

FIG. 1. Fission-product yields for thermal-neutron-induced fission of ^{242m}Am. A curve of fission-product yields for thermal-neutron-induced fission of ^{235}U is shown for comparison.

fractional yields by assuming that the shape of the charge-distribution curve for low-energy fission processes is the same as that for thermal-neutron fission of ^{235}U.[10] We use the Gaussian curve with a value of σ, the width parameter, of 0.56 ± 0.06 recommended by Wahl and co-workers.[13] The deviation from unchanged charge distribution is $Z_P - A'(Z_F/A_F)$ where A' is the fission-fragment mass before prompt neutron emission, Z_F is 95, and A_F is 243. There is no information on ν_f, the number of neutrons emitted from individual fragments in fission of ^{242m}Am. For thermal-neutron fission of ^{233}U, ^{235}U, and ^{239}Pu, the values of ν_f in the mass ranges involved are almost the same.[16] Accordingly, we use the values derived by Wahl and co-workers[13] by the Terrell summation method[17] for ^{235}U fission. The values of $Z_P - A'(Z_F/A_F)$ in Table III are reasonably consistent with the value of 0.45 ± 0.10 for the light fragment (or -0.45 for the heavy fragment) used in the formulation for calculating "normal" yields from ^{235}U fission for products with masses less than 102 or greater

TABLE III. Fractional cumulative yields from thermal-neutron fission of ^{242m}Am.

Fission product	100 × fractional cumulative yield	Z_P	A'	$Z_P - A'(Z_F/A_F)$
^{89}Kr	87 ± 1	35.87 ± 0.10	90.32	0.56 ± 0.10
^{90}Kr	66 ± 1	36.27 ± 0.05	91.35	0.56 ± 0.05
^{91}Kr	33 ± 1	36.75 ± 0.05	92.38	0.63 ± 0.05
^{92}Kr	13 ± 1	37.13 ± 0.10	93.41	0.61 ± 0.10
^{93}Kr	$2.7^{+0.3}_{-0.1}$ [a]	37.58 ± 0.13	94.46	0.64 ± 0.13
^{139}Xe	50 ± 1	54.50 ± 0.02	140.17	-0.30 ± 0.02
^{140}Xe	28 ± 1	54.83 ± 0.05	141.19	-0.37 ± 0.05
^{141}Xe	$8.2^{+0.4}_{-0.2}$	55.28 ± 0.10	142.21	-0.32 ± 0.10
^{142}Xe	$2.4^{+0.3}_{-0.2}$	55.60 ± 0.15	143.23	-0.46 ± 0.15
^{143}Xe	$0.32^{+0.03}_{-0.01}$	56.03 ± 0.18	144.25	-0.36 ± 0.18
^{144}Xe	0.12 ± 0.01	56.20 ± 0.20	145.27	-0.59 ± 0.20

[a] Not corrected for delayed neutron emission. The corrected value of 2.8 was used for calculating Z_P. Corrections for delayed neutron emission for other yields are much smaller than experimental uncertainties.

TABLE IV. Comparison of radiochemical and γ-spectrum analyses.

Fission product	R Radiochemical	R γ-spectrum	Energy (keV)
^{97}Zr	0.84 ± 0.01	0.85 ± 0.01	658 (Nb)
^{99}Mo	1.03 ± 0.02	1.02 ± 0.02	140 (Tc)
^{131}I	1.24 ± 0.03	1.32 ± 0.03	364
^{132}Te	1.12 ± 0.02	1.13 ± 0.05	228
^{133}I	1.01 ± 0.02	1.02 ± 0.02	530
^{135}I	1.17 ± 0.05	1.21 ± 0.05	1140, 1265, 80 (Xe)
^{141}Ce	1.02 ± 0.03	1.05 ± 0.03	146

than 134.[13] When the measured fractional yields from thermal-neutron fission[10,18] of ^{233}U and ^{239}Pu in these mass regions are examined in the same manner, the deviations from unchanged charge distribution are also consistent with those of ^{235}U fission.

APPENDIX. COMPARISON OF RADIOCHEMICAL AND γ-SPECTRUM ANALYSES

Measurement of several fission products was performed by both the standard radiochemical analyses and the γ-spectrum analyses of unseparated fission products. The γ spectra of ^{235}U and ^{242m}Am fission products are somewhat different because of the differences in the mass-yield curves. Comparison of the data is given in Table IV.

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[1]This cross section is a revision of our earlier result [see K. Wolfsberg, G. P. Ford, and H. L. Smith, J. Nucl. Energy Pt. A, B 20, 588 (1966)]. The revision consists of using the currently accepted value of 432.7 yr [see R. E. Stone and E. K. Hulet, J. Inorg. Nucl. Chem. 30, 2003 (1968)] for the half-life of ^{241}Am instead of the previously accepted value of 457.7 yr in interpreting the assays of the foils used for fission counting.

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