Fission of ^{241}Am with 14.8-MeV neutrons

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We have measured the mass-yield distribution from fission of ^{241}Am induced by 14.8-MeV neutrons. Yields were measured for 57 fission products from ^{86}Rb to ^{161}Tb. Data for 38 total chain yields were used to construct the mass-yield curve. Absolute fission yields were obtained with an accuracy of about 4% by the requirement of unit total yield in each half of the mass-yield distribution. Absolute yields were also obtained by means of the ^{238}U(n,f) monitor reaction, but the results from the two methods are not in agreement. The disagreement may be due to uncertainty in the ^{241}Am fission cross section; from our data we infer a cross section of 2.32 ± 0.09 b at 14.8 MeV. The peak-to-valley ratio in the mass-yield distribution is about 3.4. Several independent and cumulative fractional chain yields were measured, and Z_p values inferred from these measurements are in good agreement with those estimated from independent-yield systematics. Several partial isomer yields and isomer ratios were measured.

[NUCLEAR REACTIONS, FISSION ^{241}Am(n,f), $E=14.8$ MeV; measured fission yields, deduced fission mass distribution and $\sigma_{(n,f)}$. Measured independent and cumulative fractional chain yields and partial isomeric yields; deduced isomer ratios from fission.]

INTRODUCTION

We previously reported on the fission-yield distributions from the fission of ^{240}Pu with 14.8-MeV[1] and fission-spectrum neutrons[2] as part of our continuing studies of the fission process. In this paper we report results of measurements of products from the fission of ^{241}Am with 14.8-MeV neutrons. Previous studies of ^{241}Am fission have been limited to fission induced by thermal or reactor neutrons.[3-7]

We used two techniques to make the present measurements: one, conventional radiochemical separations, followed by radioactivity measurements on purified samples, and two, the recoil-catcher technique, followed by direct radioactivity measurements with Ge(Li) detectors on the catcher foil. The latter technique was generally limited to high-yield fission products with short half-lives because of the small amount of ^{241}Am target material used. We report results for a total of 57 nuclides extending from ^{86}Rb to ^{161}Tb. Absolute fission yields were obtained by requiring that the total yield in each half of the mass-yield distribution be unity. In two of the irradiations, ^{238}U monitor foils were used as an additional method of determining absolute fission yields.

EXPERIMENTAL PROCEDURES

Isotopically pure ^{241}Am was obtained from Oak Ridge National Laboratory. Because of the high level of alpha activity associated with this target material, all target preparations and much of the post-irradiation chemistry were performed in a gloved-box facility. This material was purified on an anion-exchange column to separate americium from its decay products ^{237}Np, ^{233}Pa, and ^{233}U.

We used both radiochemical and fission-catcher foil targets for the irradiations. Each radiochemical target consisted of about 2.5 mg of ^{241}Am as $Am(NO_3)_3$ cold-welded between two sheets of 0.025-mm aluminum foil. Each radiochemical target assembly consisted of an ^{241}Am target, either alone or sandwiched between ^{238}U monitor foils (99.8% ^{238}U, 0.2% ^{235}U) similarly encapsulated in aluminum. Fission-catcher foil targets contained about 100 μg/cm^2 of ^{241}Am within a diameter of 9.5 mm plated on 0.025-mm thick Pt disks. Each fission foil was separated from its catcher foil by a 1.6-mm-thick stainless steel spacer ring; a second catcher foil, placed behind the first, was used to verify that no fission fragments penetrated the first foil and that no fissionable nuclides were in the catcher-foil material. Catcher foils made of 0.025-mm-thick Be, Al, and Fe were tested; however, Al and Fe were not used in later irradiations because of the relatively high radioactivity induced by reactions with the foil material. Both types of target assembly were sealed in separate soldered Cu cans and finally placed inside a Cd target holder to minimize thermal-neutron activation.

The 14.8-MeV neutrons were produced at the Lawrence Livermore Laboratory (LLL) insulated-core-transformer (ICT) accelerator by reaction of 400-keV deuterons with a rotating titanium-tritide target. The maximum deuteron beam current was 20 mA, which produced a 14.8-MeV neutron source of up to 5×10^{12} n/s. The neutron flux density was monitored with a proton-recoil counter so that corrections could be made for the small changes in beam intensity. We performed nine irradiations, varying in duration from 1 to 14 h and producing up to 1.5×10^{11} fissions. The target holder was oriented at zero degrees to the deuteron beam and at distances of 10 to 100 mm from the neutron source. The longer distances were used on the monitored foil experiments to ensure that the front and back ^{238}U foils experienced similar irradiation conditions. The mean neutron energy in the target foils was 14.8 ± 0.3 MeV.

Irradiated target containers were returned to the gloved-box facility for disassembly. The $Am(NO_3)_3$ targets were dissolved in the presence of small amounts of carriers for the products to be measured. Individual elements were sequentially separated from the ^{241}Am target material. Measurements with a Ge(Li) detector were made on the separated fractions to ensure that nearly complete removal of ^{241}Am was achieved before proceeding with final purification steps. After purification by standard radiochemical procedures, a combination of Ge(Li) and beta-counting techniques was used to determine the fission yields of the products, as described in Ref. 1.

Catcher foil measurements were made directly by Ge(Li) gamma-ray spectrometry. Because no chemical processing was performed on the catcher foils, measurements made soon after the end of irradiation permitted observation of short-lived fission products not observed in radiochemical samples. The following nuclides were measured on both catcher foils and separated samples: ^{95,97}Zr, ^{112}Pd, ^{115}Cd^{g}, ^{136}Cs, ^{140}Ba, ^{141,143}Ce, and ^{147}Nd. The results for these two sample types agreed to within the estimated accuracy of the measurements. From this agreement, we conclude that the ^{241}Am on the fission foils was thin enough to ensure that the mass distribution of fission products implanted in the catcher foil was not altered in the recoil process. This conclusion is also supported by the work reported in Ref. 8.

Nuclear constants (half-lives and photon abundances) used in the data analysis were reported earlier,[1] with the additions and exceptions noted in Table I. In addition, the beta-counting efficiency was redetermined for ^{153}Sm. The measured fission yields of ^{153}Sm reported previously[1,2] should be changed from 6.55×10^{-3} (Ref. 1) to 5.80×10^{-3} and from 5.22×10^{-3} (Ref. 2) to 5.48×10^{-3} atoms/fission. Likewise, the measured fission yield of ^{88}Kr reported in Ref. 1 should be increased to 0.0139 atoms/fission to reflect a change in photon abundances.

RESULTS AND DISCUSSION

The observed amount of each fission product was corrected for growth and decay occurring during and after the irradiation, based on the time history of the neutron flux and the half-lives of the product and its precursor. Results of fission-yield measurements are summarized in Table II, where yields are given for 57 products. Yields were first calculated relative to that of ^{99}Mo, which was measured in each ^{241}Am target, and the yield ratios for each product were averaged over all the experiments. The fission yields were then calculated by the requirement of unit total yield in each half of the mass distribution. This method gave a yield of 0.0436 atoms/fission for ^{99}Mo, with an uncertainty of about 4%.

We also calculated the number of ^{241}Am fissions in the two monitored irradiations from knowledge of the masses of ^{241}Am and ^{238}U, the 14.8-MeV-fission cross sections, and the average number of

TABLE I. Nuclear properties of fission products detected in ^{241}Am fission. (These data are additions or revisions to the summary given in Ref. 1.)

Nuclide	Gamma-ray energy (keV)	Photons/decay	Half-life	Reference
^{86}Rb	1076.6	0.0878	18.66 day	9
^{88}Kr	196.1	0.288	2.803 h	10,11
^{94}Y	918.24	0.56	19.1 min	12
^{104}Tc	357.99	0.903	18.2 min	13
^{107}Rh	302.8	0.66	21.7 min	14
^{133}Xe	81.00	0.364	5.227 day	11,15
^{156}Sm	203.8	0.24	9.4 h	16

TABLE II. Yields of products from fission of ^{241}Am with 14.8-MeV neutrons.

Product nuclide	Measured fission yield (atoms/fission)	Error (%)	Est. fraction of total chain yield	Total chain yield [a] (atoms/fission)
^{86}Rb	$\leq 1.9 \times 10^{-5}$		b	
^{87}Kr	0.0105	4.1	0.967 ± 0.012	0.0109 ± 0.0005
^{88}Kr	0.0107	3.0	0.893 ± 0.029	0.0119 ± 0.0005
^{91}Sr	0.0201	3.1	0.994 ± 0.003	0.0203 ± 0.0006
^{91}Y	0.0208	3.0	1	0.0208 ± 0.0006
mass 91				0.0206 ± 0.0006
^{92}Sr	0.0215	3.0	0.964 ± 0.013	0.0223 ± 0.0007
^{94}Y	0.0255	3.0	0.985 ± 0.006	0.0258 ± 0.0008
^{95}Zr	0.0314	3.0	0.999 ± 0.001	0.0314 ± 0.0009
^{96}Nb	1.57×10^{-4}	3.0	b	
^{97}Zr	0.0339	3.0	0.967 ± 0.012	0.0350 ± 0.0011
^{99}Mo	0.0436	3.0	1	0.0436 ± 0.0013
^{103}Ru	0.0507	3.0	1	0.0507 ± 0.0015
^{104}Tc	0.0463	3.0	0.993 ± 0.005	0.0466 ± 0.0014
^{105}Rh	0.0476	3.0	1	0.0476 ± 0.0014
^{106}Ru	0.0410	3.0	0.997 ± 0.003	0.0411 ± 0.0012
^{107}Rh	0.0380	3.0	1	0.0380 ± 0.0011
^{109}Pd	0.0324	3.0	1	0.0324 ± 0.0010
$^{110}Ag^{m}$	$\leq 3.5 \times 10^{-5}$		b	
$^{111}Pd^{m}$	1.77×10^{-3}	8.4	c, d	
^{111}Ag	0.0231	3.0	1	0.0231 ± 0.0007
^{112}Pd	0.0242	3.0	0.951 ± 0.037	0.0254 ± 0.0012
^{113}Ag	0.0221	3.0	0.997 ± 0.001	0.0222 ± 0.0007
$^{115}Cd^{g}$	0.0160	3.0	1	0.0160 ± 0.0005
$^{115}Cd^{m}$	1.30×10^{-3}	3.0	1	$(1.30 \pm 0.04) \times 10^{-3}$
mass 115				0.0173 ± 0.0005
$^{126}Sb^{g}$	6.89×10^{-3}	12.4	b	
^{127}Sb	0.0223	4.6	0.960 ± 0.022	0.0233 ± 0.0012
^{128}Sn	4.93×10^{-3}	3.0	b	
$^{128}Sb^{g}$	0.0117	3.0	c	
$^{130}I^{g}$	6.67×10^{-3}	4.0	b	
$^{131}Te^{m}$	0.0140	7.1	c	
^{131}I	0.0445	3.0	0.996 ± 0.001	0.0447 ± 0.0013
^{132}Te	0.0216	3.0	b	
^{132}Cs	7.85×10^{-5}	13.4	b	
^{133}I	0.0430	3.0	0.883 ± 0.028	0.0487 ± 0.0021
^{133}Xe	0.0460	3.0	0.999 ± 0.001	0.0460 ± 0.0014
mass 133				0.0468 ± 0.0014
$^{134}Cs^{g}$	2.25×10^{-3}	4.9	b	
^{135}I	0.0181	4.1	b	
^{135}Xe	0.0449	3.0	0.927 ± 0.019	0.0484 ± 0.0018
^{136}Cs	0.0124	3.0	b	
^{137}Cs	0.0425	3.0	0.975 ± 0.008	0.0436 ± 0.0014
^{139}Ba	0.0402	3.0	0.993 ± 0.005	0.0405 ± 0.0012
^{140}Ba	0.0334	3.0	0.965 ± 0.010	0.0346 ± 0.0011
^{141}Ce	0.0323	3.0	1	0.0323 ± 0.0010
^{142}La	0.0296	3.3	0.988 ± 0.004	0.0299 ± 0.0010
^{143}Ce	0.0269	3.0	1	0.0269 ± 0.0008
^{144}Ce	0.0249	3.0	0.997 ± 0.001	0.0249 ± 0.0007
^{147}Nd	0.0181	3.0	1	0.0181 ± 0.0005
^{149}Pm	0.0130	3.0	1	0.0130 ± 0.0004
^{151}Pm	8.85×10^{-3}	3.0	0.998 ± 0.001	$(8.87 \pm 0.27) \times 10^{-3}$
^{153}Sm	6.68×10^{-3}	3.0	1	$(6.68 \pm 0.20) \times 10^{-3}$
^{155}Eu	3.39×10^{-3}	3.0	1	$(3.39 \pm 0.10) \times 10^{-3}$
^{156}Sm	2.20×10^{-3}	4.9	0.912 ± 0.039	$(2.41 \pm 0.16) \times 10^{-3}$
^{156}Eu	2.70×10^{-3}	3.0	0.999 ± 0.001	$(2.71 \pm 0.08) \times 10^{-3}$
mass 156				$(2.65 \pm 0.08) \times 10^{-3}$
^{157}Eu	1.69×10^{-3}	3.0	0.994 ± 0.004	$(1.70 \pm 0.05) \times 10^{-3}$

TABLE II. *(Continued)*

Product nuclide	Measured fission yield (atoms/fission)	Error (%)	Est. fraction of total chain yield	Total chain yield [a] (atoms/fission)
^{159}Gd	1.31×10^{-3}	4.2	0.998 ± 0.001	$(1.31 \pm0.06)\times10^{-3}$
^{160}Tb	2.92×10^{-5}	5.1	b	
^{161}Tb	6.01×10^{-4}	3.0	1	$(6.01 \pm0.18)\times10^{-4}$

[a] The experimental standard deviations given do not include the systematic uncertainty in the yield determination. This additional uncertainty is estimated to be about 4%.
[b] Partial chain yield (see Table IV).
[c] Partial isomeric yield.
[d] In Ref. 1, the ^{111}Pd^m/^{99}Mo ratio was inadvertently listed instead of the ^{111}Pd^m fission yield. The correct yield is 4.80×10^{-4} atoms/fission. The calculated ^{111}Pd^m/^{111}Pd^g isomer ratio is 0.030 ± 0.003.

fissions in the pairs of ^{238}U monitor foils. The latter quantity was determined with a precision of about 1% from numerous prominent fission products, using yields given in Ref. 17. The ^{238}U-fission cross section is well known, with $\sigma_f(^{238}\text{U})/\sigma_f(^{235}\text{U}) = 0.571\pm0.010$,[18] leading to $\sigma_f(^{238}\text{U}) = 1.19 \pm 0.03$ b for $\sigma_f(^{235}\text{U}) = 2.08\pm0.05$ b.[19] The ^{241}Am-fission cross section at 14.8 MeV as measured recently is 2.68 ± 0.15 b $[\sigma_f(^{241}\text{Am})/\sigma_f(^{235}\text{U}) = 1.29 \pm 0.07]$.[20] We used this value, but it is not in agreement with older measurements which average around 2.4 b.[21]

The absolute yield of ^{99}Mo obtained by use of the ^{238}U(n,f) monitor reaction is 0.0378 atoms/fission, which is 13% lower than the value obtained by summing the mass-yield curve. We are unable to resolve this discrepancy, other than to suggest that the ^{241}Am-fission cross section is not known with sufficient accuracy. From our data, we infer a cross section of 2.32 ± 0.09 b for ^{241}Am fission, which is in good agreement with the older cross section data.[21] It is difficult to redraw the mass-yield curve in a reasonable manner and change the total yield in either half by more than 2%. For this reason we have rejected the absolute yield determination that is based on the use of monitor foils.

Using the value of 0.0436 atoms/fission for ^{99}Mo, we calculated the fission yields of other products from ^{241}Am fission from their relative yields to ^{99}Mo.

Table II also shows the estimated fraction of the total chain yield for each product nuclide. These estimates are based on tabulated values for the most probable charge Z_p[22] and on a Gaussian charge-dispersion curve with $\sigma = 0.56$. The Z_p values were derived from systematics inferred from an analysis of measured independent and cumulative fractional chain yields.[23] In general, if the estimated cumulative fractional chain yield was greater than 0.85, it was used to calculate the total chain yield for the particular mass number. Estimated fractional chain yields less than 0.85 are usually relatively inaccurate for this

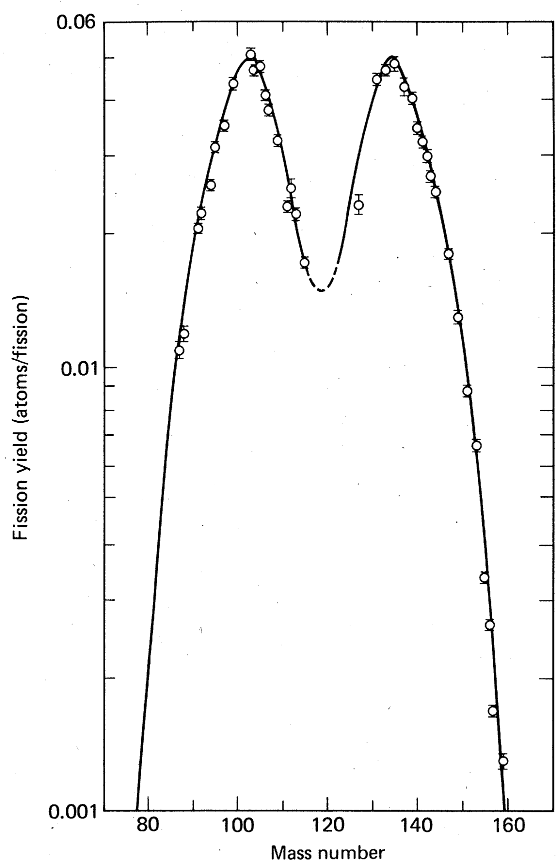

FIG. 1. Mass-yield curve for fission of ^{241}Am induced by 14.8-MeV neutrons. Measured yields are indicated by open circles with error bars. The curve was drawn with the aid of mirror points reflected about mass 118.5. The shape of the curve between masses 115 and 122 is only an estimate.

TABLE III. Recommended total chain yields for mass numbers 80 to 161. (These yields were taken from the smooth curve drawn through the data points in Fig. 1.)

Mass No.	Total chain yield (atoms/fission)	Mass No.	Total chain yield (atoms/fission)
80	0.0021	121	0.0158
81	0.0028	122	0.0173
82	0.0038	123	0.0188
83	0.0051	124	0.0209
84	0.0063	125	0.0233
85	0.0080	126	0.0262
86	0.0096	127	0.0292
87	0.0114	128	0.0324
88	0.0135	129	0.0357
89	0.0157	130	0.0392
90	0.0178	131	0.0427
91	0.0200	132	0.0460
92	0.0222	133	0.0490
93	0.0246	134	0.0502
94	0.0270	135	0.0497
95	0.0302	136	0.0486
96	0.0332	137	0.0463
97	0.0364	138	0.0436
98	0.0400	139	0.0400
99	0.0436	140	0.0364
100	0.0463	141	0.0332
101	0.0486	142	0.0302
102	0.0497	143	0.0270
103	0.0502	144	0.0246
104	0.0490	145	0.0222
105	0.0460	146	0.0200
106	0.0427	147	0.0178
107	0.0392	148	0.0157
108	0.0357	149	0.0135
109	0.0324	150	0.0114
110	0.0292	151	0.0096
111	0.0262	152	0.0080
112	0.0233	153	0.0063
113	0.0209	154	0.0051
114	0.0188	155	0.0038
115	0.0173	156	0.00282
116	0.0158	157	0.00206
117	0.0152	158	0.00151
118	0.0148	159	0.00113
119	0.0148	160	0.00083
120	0.0152	161	0.00060

purpose, so yields of such products were used solely as independent (or cumulative) fractional yield measurements.

From the 57 measurements given in Table II, we calculated total chain yields for 38 mass numbers. The remaining results are independent yields, partial isomer yields, or multiple measurements for the same mass number. The total chain yields greater than 10^{-3} are shown in Fig. 1 as a mass-yield curve. A smooth curve was drawn through the data, using yields reflected about mass 118.5 as an aid. We assumed the average mass of the fissioning nucleus was 237, which corresponds to an average emission of 5 neutrons per fission. There is no definite evidence of fine structure in the mass-yield curve. However, in the mass range from 111 to 113 there is evidence of a measurement problem, which repeated experiments failed to resolve.

The peak-to-valley ratio is about 3.4. The depth of the valley was estimated as shown in Fig. 1. A listing of recommended total chain yields for mass numbers from 80 to 161 is given in Table III. The yields are taken from the smooth curve shown in Fig. 1.

Partial chain yields were measured for several nuclides. These results are summarized in Table IV, together with the total chain yields (from Table III), the independent or cumulative fractional chain yields, and the values of Z_p inferred from the fractional chain yields. A Gaussian charge-dispersion curve with $\sigma = 0.56 \pm 0.06$ was used in calculating Z_p. The "measured" Z_p values are compared with those predicted from independent-yield systematics[22,23] and found to be in good agreement. The measured yields of ^{132}Cs and ^{134}Cs^g are higher than expected by factors of 18 and 3, respectively, and this may be caused by interference from $(n, 2n)$ and (n,γ) reactions on a trace of stable ^{133}Cs in the ^{241}Am target material.

Four partial isomer yields (not independent yields) were measured for which we can calculate the isomer ratios. These data are summarized in Table V. The isomer ratios for ^{111}Pd and ^{115}Cd are both significantly larger than found for 14.8-MeV-neutron-induced fission of ^{240}Pu (Ref. 1 and footnote d to Table II), reflecting an increased fraction of the Pd and Cd formed directly in fission. The isomer ratios of ^{128}Sb and ^{131}Te formed independently in fission can be calculated from the measured fission yields and the calculated fractional chain yields. Assuming a charge-dispersion curve with $\sigma = 0.56$, the isomer ratios (m/g) from fission alone are 0.80 ± 0.12 and 1.09 ± 0.18 for ^{128}Sb and ^{131}Te, respectively. The isomer ratios from 14-MeV fission have been predicted to be 0.75 for ^{128}Sb and 4.59 for ^{131}Te, the former being in good agreement with measurement and the latter not in agreement.[24]

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TABLE IV. Measured fractional chain yields (FCY) and values of Z_p.

Nuclide	Fission yield [a]	Total chain yield [b]	Measured independent FCY	Z_p value Measured [c]	Z_p value Predicted
^{86}Rb	$\leq 2.0 \times 10^{-5}$	0.009 6	$\leq$0.002 1	$\leq$34.88 ± 0.17	35.06 ± 0.09
^{96}Nb	1.57×10^{-4}	0.033 2	0.004 74 ± 0.000 24	39.05 ± 0.16	39.07 ± 0.09
^{110}Ag^m	$\leq 3.5 \times 10^{-5}$	0.029 2	$\leq$0.001 2	$\leq$44.80 ± 0.18	44.87 ± 0.17
^{126}Sb^g	6.89×10^{-3}	0.026 2	0.26 ± 0.03	$\geq$50.16 ± 0.08 [d]	50.20 ± 0.16
^{128}Sn	4.93×10^{-3}	0.032 4	0.152 ± 0.008 [e]	51.08 ± 0.08	50.87 ± 0.12
^{130}I^g	6.67×10^{-3}	0.039 2	0.170 ± 0.010	$\geq$51.97 ± 0.08 [d]	51.60 ± 0.08
^{132}Te	2.16×10^{-2}	0.046 0	0.469 ± 0.023 [e]	52.54 ± 0.04	52.41 ± 0.08
^{132}Cs	7.85×10^{-5}	0.046 0	0.001 7 ± 0.000 2	52.86 ± 0.20	52.41 ± 0.08
^{134}Cs^g	2.25×10^{-3}	0.050 2	0.044 7 ± 0.002 8	$\geq$53.55 ± 0.12 [d]	53.28 ± 0.08
^{135}I	1.81×10^{-2}	0.049 7	0.364 ± 0.021 [e]	53.70 ± 0.06	53.69 ± 0.08
^{136}Cs	1.24×10^{-2}	0.048 6	0.256 ± 0.013	54.15 ± 0.05	54.06 ± 0.08
^{160}Tb	2.92×10^{-5}	0.000 83	0.035 2 ± 0.001 8	63.49 ± 0.12	63.25 ± 0.14

[a] Measured fission yield from Table II.
[b] Total chain yield from Table III.
[c] The "measured" Z_p is inferred from the measured fractional chain yield. The uncertainty given for the measured Z_p includes the uncertainty in the width of the Gaussian charge-dispersion curve, $\sigma = 0.56 \pm 0.06$, which is by far the major part of the uncertainty.
[d] The result is a lower limit to Z_p because only the partial isomeric yield was measured.
[e] Cumulative fractional chain yield.

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TABLE V. Isomer ratios from fission of ^{241}Am with 14.8-MeV neutrons.

Nuclide	Spin and parity	Fission yield (atoms/fission)	Isomer ratio (m/g)	Fractional production [a] Fission	Fractional production [a] β decay
^{111}Pd^m	$\frac{11}{2}^-$	0.001 77 [b]	0.073 ± 0.007	0.31	0.69
^{111}Pd^g	$\frac{5}{2}^+$	0.024 1 [c]			
^{115}Cd^m	$\frac{11}{2}^-$	0.001 30 [b]	0.081 ± 0.003	0.06	0.94
^{115}Cd^g	$\frac{1}{2}^+$	0.016 0 [b]			
^{128}Sb^m	5^+	0.016 5 [c]	1.41 ± 0.16	0.70	0.30
^{128}Sb^g	8^-	0.011 7 [b]			
^{131}Te^m	$\frac{11}{2}^-$	0.014 0 [b]	0.68 ± 0.09	0.77	0.23
^{131}Te^g	$\frac{3}{2}^+$	0.020 7 [c]			

[a] The fractional production was estimated from the cumulative fractional chain yields of the isomer pair and its β-decay precursor.
[b] Measured yield from Table II.
[c] Yield estimated from the total chain yield (Table III), the cumulative fractional chain yield, and the measured yield of the other isomer.

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