Fission of ^{242}Pu with 15.1-MeV neutrons

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The mass-yield distribution from the fission of ^{242}Pu induced by 15.1-MeV neutrons was measured. Yields were measured for 65 fission products from ^{85}Kr to ^{151}Pm. From these data the total chain yields for 43 mass numbers were obtained and the mass-yield curve constructed. Fission product activities were measured by Ge(Li) γ-ray spectrometry of the irradiated ^{242}Pu target and by chemical separation of the fission product elements Pd, Ag, Cd, Sn, Sb, and Ce followed by β counting or γ-ray spectrometry. The peak-to-valley ratio in the mass-yield distribution is about 9.5. The average masses of the light and heavy groups are 101.5 and 136.5, respectively.

INTRODUCTION

Recently, the fission-yield distribution from the fission of ^{240}Pu with 14.8-MeV and fission-spectrum neutrons has been reported.[1,2] Studies of ^{242}Pu fission, however, have until now been limited to fission induced by fission spectrum neutrons.[3,4] No measurement of the mass-yield distribution from fission of ^{242}Pu with 14-MeV neutrons has been published.

In the present paper we report the mass-yield distribution from fission of ^{242}Pu induced by 15.1-MeV neutrons. The yields of 65 fission products were measured by high resolution γ-ray spectrometry of the irradiated ^{242}Pu target or, for some fission products in the valley of the mass distribution, by chemical separation followed by β counting or γ-spectrometric measurements.

EXPERIMENTAL PROCEDURE

The enriched ^{242}Pu used in this work was obtained as PuO_2 from Oak Ridge National Laboratory. It had the isotopic composition ^{238}Pu, 0.003%; ^{239}Pu, 0.018%; ^{240}Pu, 0.083%; ^{241}Pu, 0.096%; ^{242}Pu, 99.80%; and ^{244}Pu, 0.0005%. Since the fission cross sections of these Pu isotopes are all about the same at 15.1 MeV the contribution of the fission of the other Pu isotopes to that of ^{242}Pu fission was negligible. Measurements of the thermal neutron component of the neutron flux at the target by means of monitor foils showed that the contribution of thermal neutron fission of the isotopic impurities ^{239}Pu and ^{241}Pu could also be neglected.

Before each irradiation the ^{242}Pu was purified by a procedure described in Ref. 5 to remove the decay products of the Pu isotopes (mainly ^{241}Am and ^{237}U) and fission products. Because of the high level of α radioactivity of the ^{242}Pu sample, all target preparations and most of the postirradiation chemical procedures were done in a glove box. The $^{242}PuO_2$ sample containing 9.34 mg ^{242}Pu was dissolved in 1 ml 7 *M* HCl and a few drops of HBF_4 and of concentrated HNO_3 were added. After three days the PuO_2 was entirely dissolved. The solution was boiled down, dissolved in 10 *M* HCl plus some drops of concentrated HNO_3, and loaded onto a Dowex 1 anion exchange column. The absorbed Pu(IV) was washed with a mixture of 10 *M* HCl and concentrated HNO_3 (10:0.1). Finally the Pu(IV) on the column was reduced to Pu(III) and eluted with 10 *M* HCl containing NH_4I. The eluate was treated with aqua regia to remove I, and boiled down to near dryness. The residue was dissolved in 7 *M* HNO_3 and transferred in small portions onto a 0.05 mm thick polyethylene foil where the solution was evaporated to dryness. The residue was wrapped with the polyethylene foil and encapsulated in a small polyamide capsule of wall thickness 2 mm which in turn was sealed in a thin polyethylene foil. The sample was then wrapped in cadmium foil 1.0 mm thick to minimize the effect of any thermal neutrons produced in the bombardment.

In order to measure the low activities of fission products in the valley mass region it was necessary to employ radiochemical separations of the fission products. In this case the purified ^{242}Pu sample was enveloped in a 0.030-mm-thick aluminum foil and encapsulated in a small polyamide capsule which in turn was covered by a 1.0 mm cadmium foil.

The 15.1-MeV neutron irradiations were performed at the linear accelerator of the Gesellschaft für Strahlen- und Umweltforschung (GSF) where neutrons are produced by the reaction of a beam of 900-keV deuterons on a tritium target. The maximum deuteron beam current was 160 μA, which produced a 15.1-MeV neutron source of up to 2.4×10^{12} n/s. The mean neutron energy in the ^{242}Pu target was 15.1 ± 0.7 MeV.[6] The target holder was placed at 0° to the deuteron beam at distances of 9 to 15 mm from the neutron source. The neutron flux intensity was monitored with a proton-recoil counter so corrections could be made for any changes in beam intensity. Each ^{242}Pu target assembly consisted of the ^{242}Pu target sandwiched between Cu and Au foils which were used as neutron fluence monitors.

The fast neutron fluence in the target was measured by ^{64}Cu and ^{196}Au produced by the (n,2n) reactions on the Cu and Au monitor foils, respectively. Cross sections of (975 ± 50) mb and (2250 ± 120) mb, respectively, were used for the (n,2n) reactions.[7] The thermal neutron fluence was measured by the reaction ^{197}Au(n,γ)^{198}Au. These measurements showed that the intensity of the thermal neu-

TABLE I. Nuclear properties of fission products detected in ^{242}Pu fission.

Nuclide	γ energy (keV)	Absolute γ intensity (γ/decay)	Half-life	Ref.
^{85}Kr^{m}	151.2	0.746	4.48 h	14
^{87}Kr	402.6	0.494	1.27 h	14
^{88}Kr	196.3	0.263	2.80 h	14
^{89}Rb	1031.9	0.636	15.15 min	14
	1248.1	0.467		
^{91}Sr	749.7	0.244	9.48 h	14
	1024.3	0.340		
^{91}Y^{m}	555.6	0.560	0.828 h	14
^{92}Sr	1383.9	0.900	2.71 h	14
^{92}Y	934.5	0.138	3.53 h	14
^{93}Y	266.9	0.0695	10.10 h	14
^{94}Y	919.2	0.49	18.70 min	15
^{95}Zr	724.3	0.444	63.98 d	14
	756.7	0.546		
^{97}Zr	743.4	0.946	17.00 h	14
^{97}Nb^{g}	657.9	0.983	1.20 h	16
^{99}Mo	181.9	0.0600	66.02 h	14
	739.5	0.123		
^{99}Tc^{m}	140.5	0.825	6.01 h	17
^{101}Tc	306.9	0.886	14.20 min	14
^{103}Ru	497.1	0.894	39.35 d	14
^{104}Tc	358.0	0.903	18.20 min	14
	535.1	0.134		
^{105}Ru	316.4	0.111	4.40 h	14
	469.4	0.175		
	676.4	0.157		
	724.2	0.473		
^{105}Rh	318.9	0.192	35.36 h	14
^{107}Rh	302.8	0.659	21.70 min	14
^{109}Pd	88.0	0.0389	13.46 h	14
^{111}Pd^{m}	172.0	0.324	5.50 h	14
^{111}Ag^{g}	342.1	0.0668	7.43 d	1
^{112}Pd	616.8[a]	0.433	20.12 h	14
^{113}Ag^{g}	298.0	0.0830	5.37 h	14
^{115}Cd^{g}	527.7	0.267	2.21 d	14
^{115}In^{m}	336.2	0.482	4.30 h	14
^{117}Cd^{m}	564.4	0.153	3.35 h	1
	1066.0	0.231		
^{117}Cd^{g}	273.3	0.284	2.56 h	1
	344.5	0.173		
	1303.4	0.176		
^{118}Cd	1229.5[a]	0.15	0.838 h	18
^{125}Sn^{g}	1066.6	0.886	9.64 d	14
^{125}Sb	427.9	0.297	2.77 yr	14
^{126}Sb^{g}	414.8	0.880	12.40 d	14
	666.3	1.00		
	695.0	1.00		
^{127}Sn^{g}	1095.6	0.152	2.10 h	14
	1114.3	0.298		
^{127}Sb	473.0	0.250	3.85 d	14
	685.7	0.357		
	783.5	0.147		
^{128}Sn	482.3	0.580	0.985 h	14
^{128}Sb^{m}	753.9	1.00	10.00 min	14
^{128}Sb^{g}	754.0	1.00	9.10 h	14
^{129}Sb	812.6	0.435	4.32 h	14
	914.7	0.188		
	1030.4	0.124		

TABLE I. (*Continued*).

Nuclide	γ energy (keV)	Absolute γ intensity (γ/decay)	Half-life	Ref.
$^{129}Te^{g}$	459.6	0.0714	1.16 h	14
$^{130}Sb^{g}$	330.9	0.778	0.667 h	14
	793.4	0.998		
	839.4	0.998		
^{131}Sb	943.6	0.440	23.00 min	14
$^{131}Te^{m}$	793.8	0.138	1.25 d	14
	852.2	0.206		
	1125.5	0.114		
	1206.6	0.0973		
^{131}I	364.5	0.824	8.04 d	14
^{132}Te	228.2	0.880	3.24 d	14
$^{132}I^{g}$	522.7	0.161	2.30 h	14
	630.2	0.137		
	667.7	0.987		
	772.6	0.762		
	954.6	0.181		
$^{133}Te^{m}$	647.7	0.293	0.923 h	14
	864.6	0.195		
	913.0	0.870		
^{133}I	529.9	0.873	20.80 h	14
^{133}Xe	81.0	0.371	5.25 d	14
$^{134}I^{g}$	595.4	0.114	0.877 h	14
	847.0	0.954		
	884.1	0.653		
	1072.6	0.153		
^{135}I	1131.6	0.230	6.70 h	14
	1260.5	0.303		
^{135}Xe	249.7	0.906	9.15 h	14
$^{138}Cs^{g}$	462.8	0.308	32.20 min	14
	1009.8	0.298		
	1435.9	0.763		
^{139}Ba	165.9	0.188	1.38 h	14
^{140}Ba	537.3	0.240	12.79 d	14
^{140}La	486.0	0.453	40.20 h	14
	1596.2	0.953		
^{141}Ce	145.4	0.481	32.45 d	14
^{142}La	641.2	0.490	1.55 h	14
^{143}Ce	293.3	0.434	33.0 h	16
^{144}Ce	133.5	0.111	284.6 d	14
^{146}Pd	453.6	0.55	24.20 min	19
^{149}Nd	211.3	0.273	1.73 h	14
	423.6	0.0945		
^{151}Pm	340.1	0.224	28.00 h	1

[a] γ ray emitted by the daughter in equilibrium.

tron fluence was only 0.01% of the intensity of the fast neutron flux. Ten separate irradiations were made varying in length from 1 to 28 h. After irradiation, the polyamide capsule containing the ^{242}Pu target was either sealed in a new polyethylene bag for γ-ray spectrometric analysis or transferred to a glove box for radiochemical separations. In this case the ^{242}Pu sample was dissolved together with its aluminum envelope in concentrated HCl in the presence of carriers of Pd, Ag, Cd, Sn, Sb, and Ce. The clear solution was diluted with a few ml of H_2O to precipitate AgCl which was separated by centrifugation. Ce and Pu were removed from the supernate by precipitating the fluorides. Pd, Cd, Sb, and Sn, left in the solution, were separated according to the procedures outlined by Flynn[8] which were slightly modified. The separated elements were finally radiochemically purified and samples prepared for counting, again according to the procedures compiled by Flynn.[8] Before the radiochemical purification of the Ce fraction, Pu had to be separated. The fluoride precipitate of Ce and Pu was dissolved in 10 *M* HCl and some drops of a saturated H_3BO_3 solution and Pu was separated by the procedure described above.[5]

The chemically purified samples of the elements were counted in a calibrated low-background (0.8 counts/min)

TABLE II. Yields of products from fission of ^{242}Pu with 15.1-MeV neutrons.

Product nuclide	Measured fission yield (atoms/fission)	Error (%)	Estimated fraction of total chain yield	Total chain yield[a] (atoms/fission)
^{85}Kr^{m}	0.00672	7.1	1	0.00672±0.00048
^{87}Kr	0.00993	7.6	0.998±0.001	0.00995±0.00076
^{88}Kr	0.0116	8.1	0.981±0.007	0.0118 ±0.0010
^{89}Rb	0.0133	11	0.999±0.001	0.0133 ±0.0015
^{91}Sr	0.0211	7.1	1	0.0211 ±0.0015
^{91}Y^{m}	0.0203	7.9	1	0.0203 ±0.0016[b]
Mass 91				0.0207 ±0.0011
^{92}Sr	0.0226	13	0.998±0.001	0.0227 ±0.0030
^{92}Y	0.0249	12	1	0.0249 ±0.0030
Mass 92				0.0238 ±0.0021
^{93}Y	0.0260	13	1	0.0260 ±0.0034
^{94}Y	0.0279	10	0.999±0.001	0.0280 ±0.0028
^{95}Zr	0.0337	8.9	1	0.0337 ±0.0030
^{97}Zr	0.0423	7.5	0.998±0.001	0.0424 ±0.0032
^{97}Nb	0.0422	7.1	1	0.0422 ±0.0030
Mass 97				0.0423 ±0.0022
^{99}Mo	0.0497	7.2	1	0.0497 ±0.0036
^{99}Tc^{m}	0.0470	7.4	1	0.0470 ±0.0035[b]
Mass 99				0.0484 ±0.0025
^{101}Tc	0.0504	9.9	1	0.0504 ±0.0050
^{103}Ru	0.0564	7.9	1	0.0564 ±0.0045
^{104}Tc	0.0545	12	1	0.0545 ±0.0066
^{105}Ru	0.0558	7.5	1	0.0558 ±0.0042
^{105}Rh	0.0542	7.9	1	0.0542 ±0.0043
Mass 105				0.0550 ±0.0030
^{107}Rh	0.0380	10	1	0.0380 ±0.0038
^{109}Pd	0.0253	8.6	1	0.0253 ±0.0022
^{111}Pd^{m}	9.4×10^{-5}	64	1	$(9.4\pm6.0)\times10^{-5}$[c]
^{111}Ag	0.0220	11	1	0.0220 ±0.0024
^{112}Pd	0.0192	7.2	0.998±0.002	0.0192 ±0.0014
^{113}Ag^{g}	0.0160	10	1	0.0160 ±0.0016
Mass 113				0.0178 ±0.0025[d]
^{115}Cd^{m}	≤0.0011	35	1	≤0.0011 ±0.0004
^{115}Cd^{g}	0.0112	13	1	0.0112 ±0.0015
Mass 115				≤0.0123 ±0.0016
^{117}Cd^{m}	0.00151	20	0.999±0.001	0.0015 ±0.0003
^{117}Cd^{g}	0.00503	20	0.999±0.001	0.0050 ±0.0010
Mass 117				0.0065 ±0.0011
^{118}Cd	0.0042	16	0.996±0.006	0.0042 ±0.0007
^{121}Sn^{g}	0.00658	20	1	0.0066 ±0.0013
Mass 121				0.0076 ±0.0017[e]
^{125}Sn^{g}	0.00684	16	0.990±0.009	0.0069 ±0.0011[c]
^{125}Sb	0.0135	18	1	0.0135 ±0.0024
^{126}Sb^{g}	0.0012	60	f	0.0012 ±0.0007
^{127}Sn^{g}	0.0166	11	0.886±0.050	0.0188 ±0.0023[c]
^{127}Sb	0.0187	12	0.999±0.002	0.0187 ±0.0022
^{128}Sn	0.0130	17	0.759±0.050	0.0171 ±0.0031[g]
^{128}Sn	0.0129	20	0.759±0.050	0.0170 ±0.0036[h]

TABLE II. (*Continued*).

Product nuclide	Measured fission yield (atoms/fission)	Error (%)	Estimated fraction of total chain yield	Total chain yield[a] (atoms/fission)
Mass 128				0.0171±0.0024
$^{128}Sb^{g}$	0.00678	11	0.994±0.003	0.0068±0.0008[c]
^{129}Sb	0.0200	10	0.967±0.011	0.0207±0.0021
^{129}Te	0.0216	10	1	0.0216±0.0022[b]
Mass 129				0.0211±0.0016
$^{130}Sb^{g}$	0.0142	12	0.881±0.029	0.0161±0.0020[c]
^{131}Sb	0.0240	13	0.676±0.051	0.0355±0.0053
$^{131}Te^{m}$	0.0159	10	0.987±0.005	0.0161±0.0016[c]
^{131}I	0.0391	6.1	1	0.0391±0.0024
^{132}Te	0.0456	7.4	0.935±0.018	0.0488±0.0037
^{132}I	0.0527	5.3	1	0.0527±0.0028
Mass 132				0.0513±0.0023
$^{133}Te^{m}$	0.0143	13	0.779±0.038	0.0183±0.0026[c]
^{133}I	0.0552	7.2	0.995±0.003	0.0554±0.0040
^{133}Xe	0.0566	7.6	1	0.0566±0.0043
Mass 133				0.0560±0.0030
^{134}I	0.0559	12	0.960±0.012	0.0583±0.0070
^{135}I	0.0510	8.7	0.848±0.032	0.0601±0.0057
^{135}Xe	0.0552	8.9	0.998±0.001	0.0553±0.0049
Mass 135				0.0575±0.0038
^{138}Cs	0.0500	9.1	0.996±0.016	0.0502±0.0046
^{139}Ba	0.0493	7.7	1	0.0493±0.0038
^{140}Ba	0.0484	9.0	0.999±0.001	0.0485±0.0044
^{140}La	0.0495	7.8	1	0.0495±0.0039
Mass 140				0.0491±0.0030
^{141}Ce	0.0486	8.2	1	0.0486±0.0040
^{142}La	0.0434	7.6	1	0.0434±0.0033
^{143}Ce	0.0366	7.1	1	0.0366±0.0026
^{144}Ce	0.0319	8.4	1	0.0319±0.0027
^{146}Pr	0.0218	8.7	1	0.0218±0.0019
^{149}Nd	0.0161	9.9	1	0.0161±0.0016
^{151}Pm	0.00915	9.7	1	0.0092±0.0009

[a]The experimental standard deviations given here do not include the systematic uncertainty in the yield determination. The additional uncertainty is calculated to be 2%.
[b]Branching ratio of the β decay of the precursor already taken into account.
[c]Partial isomeric yield only.
[d]Total chain yield was calculated using the branching ratio of 0.90 (Ref. 21) for the β decay of the precursor ^{113}Pd.
[e]Total chain yield calculated using a cumulative isomer ratio of $^{121}Sn^{m}/^{121}Sn^{g}=0.16\pm0.01$ (Ref. 22).
[f]Independent fission yield.
[g]Fission yield determined by measuring ^{128}Sn directly.
[h]Fission yield of ^{128}Sn determined by measurement of its daughter $^{128}Sb^{m}$.

proportional counter and/or in a γ-ray spectrometer. 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 corrected for chemical yield, counting efficiency, decay, genetic relationships, and relative degree of saturation.

The disintegration rates of individual radioactive fission products were determined by γ-ray spectrometry using three coaxial Ge(Li) detectors with relative efficiencies of 5.5%, 9.4%, and 15% and resolutions of 2.0, 2.1, and 2.1 keV full width at half maximum at 1332 keV, respectively. The absolute efficiency curve for each detector was determined with standardized sources from Amersham and the Physikalisch-Technische Bundesanstalt (PTB). The samples to be counted were mounted at positions at distances of 8.0, 9.0, or 12.0 mm, respectively, from the

Ge(Li) detectors.

Because of the relatively high geometric efficiency of the counting arrangements, corrections were required for losses from the photopeak caused by γ-cascade coincidence summing in the decay of nuclides in which two or more γ rays were emitted in coincidence. Correction factors for this effect were obtained for all measured fission products using the procedure given by Debertin.[9,10] The dead times of the γ spectrometers were determined by means of a pulse generator.[11,12]

A large number of γ-ray spectra were recorded over a sufficient period of time to encompass several half-lives for each of the fission products. These complex spectra were analyzed with the computer program SAMPO (Ref. 13) to obtain the intensities of the photopeaks. Whenever possible more than one γ line was used in the yield determinations. The fission product γ-decay data and half-lives used in the data analyses are presented in Table I.

Corrections were made for cascade coincidence losses, absolute γ-emission intensities, genetic relationships, and growth and decay occurring during and after irradiation based on the time history of the neutron flux and the half-lives of the fission product and its precursors. Corrections were also made for changes of neutron flux intensity as measured by the proton-recoil counter.

RESULTS AND DISCUSSION

Relative yields were first calculated as the ratio of a particular yield to that of ^{143}Ce which was measured in each ^{242}Pu irradiation, and the ratios for each fission product were averaged over all the irradiations. The absolute fission yields were then calculated by normalization of the complete mass distribution to 200% total yield, the undetermined yields being interpolated or extrapolated from measured yields. This method gave a yield of 0.0366 atoms/fission for ^{143}Ce. Assuming an error of 20% of all interpolated or extrapolated undetermined yields, the uncertainty (1σ) in the fission rate obtained by the normalization procedure is only 2%. Using the value of 0.0366 atoms/fission for ^{143}Ce, we calculated the fission yields of other products from ^{242}Pu fission from their relative yields to ^{143}Ce. Results of fission-yield measurements are summarized in Table II. Uncertainties (1σ) in the fission yield values were obtained by consideration of all known sources of error with the usual rules of error propagation. They do not include the 2% uncertainty in the number of fissions, which is a constant uncertainty in all of the measurements. The possible error in the determination of the mass yield that may be caused by direct formation in fission (independent yield) of chain members beyond the measured nuclides was estimated according to Nethaway.[20] Table II shows the estimated fraction of the total chain yield for each fission product nuclide. From the measurements of 65 individual fission products given in Table II the total chain yields for 43 mass numbers have been calculated.

The number of fissions occurring in the ^{242}Pu targets was also calculated from the number of ^{242}Pu target atoms, the 15.1-MeV fission cross section of ^{242}Pu, and the neutron fluence. The integrated neutron fluence in the target was obtained by averaging the results from the Cu and Au monitor foils in front and back of the ^{242}Pu sample. The fission cross section used for ^{242}Pu at 15.1 MeV is (2.01 ± 0.08) b.[23,24] The estimated uncertainty in the number of fissions calculated by this method, however, is no less than 15%. This uncertainty mainly arises from the uncertainty in determining the neutron fluence in the ^{242}Pu sample by means of monitor foils. For this reason we did not use this method for calculating absolute yields; however, the absolute yield of ^{143}Ce estimated by this method is 0.032 ± 0.007 atoms/fission.

The total chain yields greater than 0.5% are shown in Fig. 1. Because no distinct structure could be found, a smooth curve has been drawn through the data points using values reflected about mass 119 as an aid. The average mass of the fissioning nucleus was assumed to be 238, which corresponds to an average emission of 5 neutrons per fission event. According to Madland and Young[25] the average number of prompt neutrons emitted per fission event is estimated to be 5.13.

The peak-to-valley ratio in the mass-yield distribution is found to be about 9.5. This result agrees with the trend found for 14.8-MeV fission of the isotopes of U (Ref. 26) and of ^{239}Pu (Ref. 27) and ^{240}Pu (Ref. 1), i.e., the peak-to-valley ratio increases with increasing target mass number from 3.2 (for ^{233}U) to 6.8 (for ^{238}U) and 4.1 (for ^{239}Pu), 5.5 (for ^{240}Pu) to 9.5 (for ^{242}Pu). The average masses of the light and heavy mass group are derived from the fission yield data as 101.5 and 136.5, respectively. This is in agreement with earlier observations that the average mass of the heavy mass group is nearly constant for 14.8-MeV

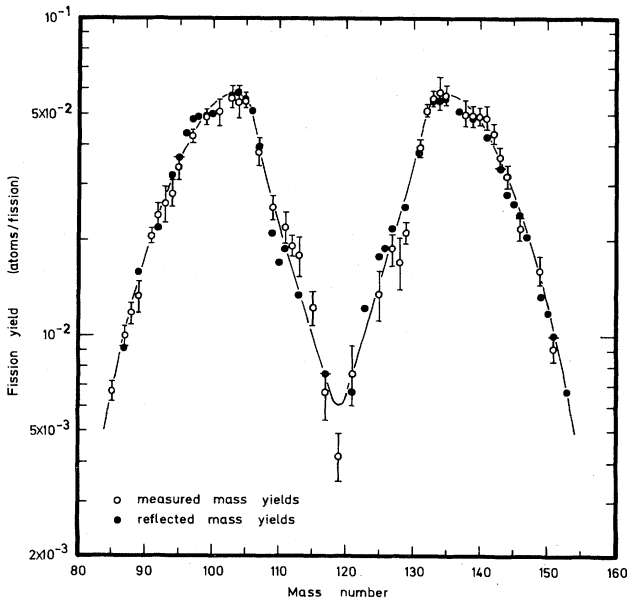

FIG. 1. Mass-yield curve for fission of ^{242}Pu with 15.1-MeV neutrons. Measured yields are indicated by open circles with error bars. The curve was drawn with the aid of mirror points reflected about mass 119.0.

fission of ^{232}Th,[28] U isotopes,[26,28] Pu isotopes,[1,27,28] and ^{241}Am,[29] whereas the average mass of the light mass group increases linearly with the mass of the fissioning system.

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