Fast-Neutron-Induced Fission Cross Sections of Pu^{241} and Am^{243}†

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(Received July 10, 1961)

The cross sections for neutron-induced fission of Pu^{241} for neutrons of energies from 0.02 to 1.80 Mev and of Am^{243} for neutrons of energies from 0.3 to 1.7 Mev have been determined. The determination was made by measuring the ratio of each cross section to that of U^{235} using a back-to-back gas scintillation counter.

INTRODUCTION

THE neutron-induced fission cross sections of heavy elements in the plateau region above about 1.5 Mev have been found to have systematic dependences on the nuclear parameters of charge Z and mass A.[1–3] The thresholds for fission show analogous behavior.[4] In order to test the universality of the dependences, the cross sections of Pu^{241} and Am^{243} were measured. Pu^{241} was chosen because in addition to providing a test of the systematic behavior, its cross section was needed for use in the development of fast nuclear reactors. Am^{243} was chosen so that its threshold and plateau cross section could be compared to those of Am^{241}, the only isotope of americium studied prior to this work. The results obtained were found to be consistent with several of the systematic schemes, but not compatible, within experimental error, with some results obtained by other workers.[5,6]

The fission cross sections of Pu^{241} and Am^{243} were determined by measuring the ratio of the cross sections to that of U^{235}. Published values[1,5] for U^{235} were then used to obtain the absolute cross sections.

APPARATUS AND EXPERIMENTAL METHOD

The experimental technique was essentially that used in the previous measurement of the Pu^{242} fission cross section.[7] Therefore, only a general outline of the technique will be given here, together with details of some recent modifications. The same back-to-back gas scintillation counter was used as in the Pu^{242} measurements, the only change being the use of xenon instead of an argon-nitrogen mixture as the scintillating gas. This change was made in an effort to reduce the background of small scintillations which appeared at high neutron energies. It was assumed that some of these scintillations came from protons from $N^{14}(n,p)C^{14}$ reactions occurring near the photomultiplier where the light collection efficiency was high. On changing to xenon the background diminished. A small background remained, perhaps from an (n,p) or (n,α) reaction of very small cross section in the stainless steel sample supports. The xenon had an additional advantage in that it maintained high scintillation efficiency over a longer time, possibly because it was less influenced by contaminants in the counter.

A gas scintillation counter rather than a conventional ionization chamber was used for these measurements because of the shorter duration of the scintillator's output pulses. The shortness of the pulses was utilized to prevent pile-ups which would otherwise have resulted from the intense α activity of the Pu^{241} and Am^{243} samples. A simple back-biased diode was placed at the output of the photomultiplier with the bias set to eliminate most of the α pulses. In this way it was possible to obtain good fission pulse spectra with conventional electronic equipment. The spectrum obtained from the Pu^{241} sample, taken with the back-biased diode in operation, is shown in Fig. 1. The Am^{243} sample showed even greater separation between the α particles and fissions, indicating that the Pu^{241} sample was relatively thick to the fission fragments. The thickness sets a limit on the accuracy with which the fission rates of the Pu^{241} and U^{235} could be compared, the limit being about $\pm 5\%$. This is one of the larger sources of error in the Pu^{241} fission cross-section measurements. The greater separa-

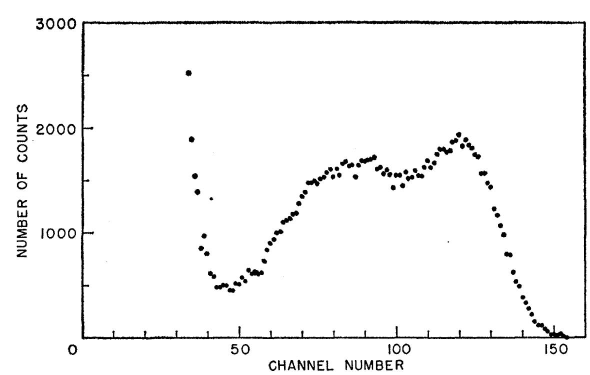

FIG. 1. Fission pulse spectrum from Pu^{241} counter.

† This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] D. W. Allen and R. L. Henkel, *Progress in NuclearEnergy. Series I—Physics and Mathematics* (Pergamon Press, New York, 1958), Vol. II, Chap. 1, p. 28.

[2] J. R. Huizenga, Phys. Rev. **109**, 484 (1958).

[3] Yu. S. Zamyatnin, Atomnaya Energia, Suppl. No. 1, p. 27 (1957), translated in *Physics of Fission*, Suppl. No. 1, Soviet J. Atomic Energy (Consultants Bureau, Inc., New York, 1957), Chap. 2, p. 21.

[4] J. D. Jackson, Atomic Energy of Canada Limited, Chalk River Project Report CRP-642-A, 1956 (unpublished), p. 125.

[5] *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

[6] M. E. Kazarinova, Yu. S. Zamyatnin, and V. M. Gorbachef, Atomnaya Energia **8**, 139 (1960).

[7] D. K. Butler, Phys. Rev. **117**, 1305 (1960).

tion of α particles and fissions in the Am^{243} counter resulted in a probable error in the Am^{243} measurements of $\pm 2.5\%$.

The neutrons for the measurements were produced by the $Li^7(p,n)Be^7$ reaction using protons from an electrostatic generator. The proton energy was determined using an electrostatic analyzer on the H_2^+ beam, and the neutron energy computed from the proton energy. The only change in the use of the neutron source from that described previously[7] was made for the low-energy measurements with the Pu^{241} sample. In order to avoid the second geometric group of neutrons below 120-kev neutron energy, the fission counter was placed at 115.5° with respect to the proton beam. At this angle, because of physical limitations, the counter had to be located twice as far from the neutron source as at 0° (18.0 cm at 115.5°, 8.9 cm at 0°). The increased distance together with the low yield from the target at 115.5° reduced the count rate almost a factor of ten.

At 18.0 cm from the target, the background count caused by low-energy neutrons scattered back by the surroundings became appreciable. In order to estimate this background a check of the dependence of count rate on distance from the neutron source was made with a $B^{10}F_3$ filled proportional counter. The departure of the dependence from $1/R^2$ indicated that 20% of the B^{10} count rate at 18.0 cm was due to scattered neutrons. 80% of the scattered neutrons had energies below the cadmium cutoff. Assuming that the background is actually moderated to thermal energies by the surroundings, and that the 0.26-v resonance[5] in Pu^{241} may overlap as much as a quarter of it, the effect on the fission ratio would be 5%. This must be considered as a possible systematic error as large as 5% in the Pu^{241} fission cross section below 150 kev. For all the data above 150 kev, which was taken at 0°, the error would be only 1%. The error in the Am^{243} measurements, where only the U^{235} was thermally fissionable, would be about $\frac{1}{2}\%$.

TABLE I. Composition of the fissionable samples.

Sample	Isotope	Half-life[a] (years)	Abundance[b] (atom %)	Mass (mg)
U^{235}	U^{234}	2.50×10^5	0.904 ± 0.009	0.046
	U^{235}	7.10×10^8	93.263 ± 0.05	4.725
	U^{236}	2.42×10^7	0.424 ± 0.004	0.022
	U^{238}	4.52×10^9	5.405 ± 0.05	0.277
Pu^{241}	Pu^{239}	2.44×10^4	1.16 ± 0.02	0.018
	Pu^{240}	6.59×10^3	2.32 ± 0.03	0.036
	Pu^{241}	13.22 ± 0.16	92.04 ± 0.03	1.434
	Pu^{242}	3.76×10^5	0.19	0.003
	Am^{241}	458 ± 2	4.31[b]±0.02	0.067
Am^{243}	Am^{243}	7.95×10^3	100 ± 0.5	0.237

[a] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 817–827 (1958).
[b] Abundances determined by mass spectrographic analysis except for Am^{241} which was determined by α counting.

The correction for absorption of neutrons in the foil backings was made by rotating the counter around the center of the foils, maintaining the cylindrical axis of the counter in the plane including the neutron source. The neutrons were thus successively incident on the counter at the four directions corresponding to 30° angles between the incident neutrons and the plane of the samples. This procedure also helped compensate for nonuniformity of the fissionable deposits. The Pu^{241} and U^{235} samples were uniform to within a few percent except for $\frac{1}{2}$-cm diameter circles at their centers. The Am^{243} sample was less uniform, but not sufficiently so to cause a measurable change in fission rate as the sample was rotated around its center.

The fissionable samples were all oxide deposits on platinum foil. The isotopic composition and masses of the samples are shown in Table I. The masses of the Pu^{241} and Am^{243} samples were determined by α counting with a low-geometry solid-state counter. The counter gave a resolution of 2% for 5-Mev α particles. The

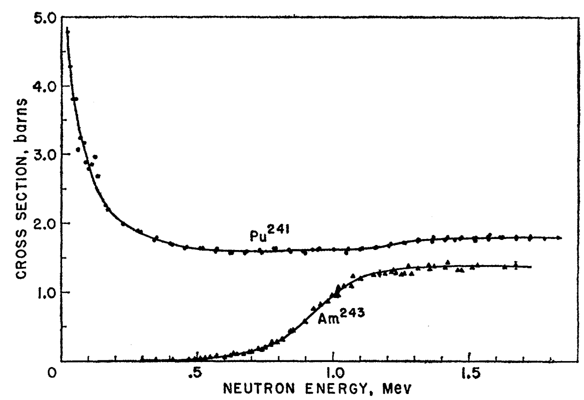

FIG. 2. Fission cross sections of Pu^{241} and Am^{243}.

counter efficiency was determined using the U^{234} α's from the U^{235} sample. The intensity of these α's in turn was determined from the mass spectrum and the mass of the U^{235} sample. The Pu^{241} sample was prepared by electrodeposition.[8] The Am^{243} sample was prepared by evaporation of an ethylene glycol solution of the nitrate. Both a mass-spectrum analysis and an α-pulse analysis showed it to be essentially pure Am^{243}. The U^{235} sample was prepared by quantitative electrodeposition of a uranyl sulfate solution.

RESULTS

The results of the fission cross section measurements are given in Table II and shown in Fig. 2. The results have been corrected for the second group of neutrons from the $Li^7(p,n)Be^{7*}$ reaction above 600 kev using the data of Smith[9] and Bevington *et al.*[10] The total energy

[8] The sample was prepared at the Hanford Laboratories for B. R. Leonard, to whom the authors are indebted for its loan.
[9] A. B. Smith (private communication).
[10] P. R. Bevington, W. W. Rolland, and H. W. Lewis, Phys. Rev. **121**, 871 (1961).

spread of the neutrons due to geometry and target thickness was maintained less than 70 kev for the Am^{243} measurements, and less than 60 kev in the Pu^{241} measurements above 200-kev neutron energy. Below 200 kev it tapered down to 17 kev spread at 21-kev incident energy.

Statistical errors are shown in Fig. 2 for the Am^{243} cross section above 1.0 Mev. Below 1.0 Mev the statistical errors were too small in absolute size to be shown in the figure, but the percent error increased in proportion to $\sigma^{-\frac{1}{2}}$ reaching 30% at 0.3 Mev. For the Pu^{241} cross section above 150-kev neutron energy, the statistical errors are about the size of the data points, while below 150 kev, where the counting rate was low, the statistical errors were large enough to produce the scatter of points shown. In addition to the statistical errors it is necessary to consider systematic errors in the normalization of the curves. These arise primarily in the determination of the sample masses and in the correction for fissions causing pulses not large enough to be seen above the α pulses. No truly quantitative measure of these normalization errors is possible, but they can be estimated to be 6% for the Am^{243} measurements and 7% for the Pu^{241} measurements above 150 kev. For Pu^{241} below 150 kev, where the counter was farther from the neutron source, the low-energy air- and room-scattered neutrons may have caused an additional 5% systematic error.

DISCUSSION

The only published measurement of the Pu^{241} fission cross section is that of Kazarinova *et al.*[6] at 2.5 Mev giving 1.2 ± 0.2 b. This value is lower than a reasonable extrapolation of the present data. No published measurement of the Am^{243} cross section exists. However, three systematic empirical relationships between the fission cross sections in the region 2.5 to 3.0 Mev and other nuclear data have been observed. The plot[1] of the 3.0-Mev cross section against $Z^{\frac{4}{3}}/A$, and the plots suggested by Zamyatnin[3] and Huizenga,[2] both essentially of

TABLE II. Fission cross sections of Pu^{241} and Am^{243} in barns.

E_n (kev)	$\sigma_f(Pu^{241})$	$\sigma_f(Am^{243})$
20	4.8	
40	4.3	
60	3.5	
80	3.1	
100	2.9	
130	2.6	
160	2.27	
200	2.09	
250	1.94	
300	1.83	0.01
400	1.70	0.02
500	1.62	0.04
600	1.59	0.08
700	1.58	0.15
800	1.59	0.31
900	1.60	0.62
1000	1.61	0.96
1100	1.62	1.22
1200	1.67	1.31
1300	1.74	1.35
1400	1.77	1.38
1500	1.78	1.39
1600	1.78	1.39
1700	1.79	1.39
1800	1.79	

the cross sections against Z^2/A, predict that in this energy region the Am^{243} cross section should be about 25% smaller than that of Am^{241}. This would imply that extrapolation of the present data give better agreement with the Am^{241} cross section value obtained by Kazarinova *et al.*[6] than with that obtained at Los Alamos.[5] However, the scheme of Jackson[4] relating the 3-Mev cross section to the threshold energy, indicates that the present Am^{243} data are more consistent with the Los Alamos measurements. It is interesting to note also that neither americium cross section fits the plot against $Z^{\frac{4}{3}}/A$ well, but they both fit the theoretically preferable semiempirical plot against Z^2/A. Improved measurements of the Pu^{240} and Am^{241} cross sections as well as of those other heavy isotopes would help to determine which plot has the more general validity.