

The results of four runs on the $F=3$ to $F=2$ transition are shown in Table III. The final value for the frequency of the $F=3$ to $F=2$ transition is 20.1 ± 0.4 Mc/sec, where the uncertainty quoted is the extreme deviation from the average. The value of b is then 2.8 ± 0.4 Mc/sec but, because of the difficulty of fitting

curves to the incompletely resolved resonances, we increase the uncertainty to ± 0.8 Mc/sec.

From Eq. (4), Q is found to be $(0.07 \pm 0.02) \times 10^{-24}$ cm².

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Thermal-Neutron Fission Cross Sections for Isotopes of Plutonium, Americium, and Curium*

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The following thermal-neutron fission cross sections have been measured in the thermal column of the Materials Testing Reactor at Idaho Falls, Idaho: Pu²³⁸, 18.4 ± 0.9 barns; Am²⁴¹, 3.13 ± 0.15 barns; Am²⁴², 6390 ± 500 barns; Am²⁴³, < 0.072 barn; Cm²⁴³, 690 ± 50 barns; Cm²⁴⁵, 1880 ± 150 barns. In addition, a pile neutron capture cross section of 520 ± 40 barns has been measured for Pu²³⁸.

I. INTRODUCTION

THE increasing interest in the fission process and the value of knowing the destruction rate of certain nuclides during long neutron irradiations have made it desirable to determine accurately a number of slow-neutron fission cross sections for isotopes of the transuranic elements. It has become possible to perform some of these measurements because of the increased quantities of isotopes available, and because of the enrichments toward the heavier isotopes of certain elements gained in long neutron irradiations of Pu²³⁹ and Am²⁴¹. Fission cross-section measurements were made on Am²⁴², Am²⁴³, and Cm²⁴³ for which there were known previously either upper limits, approximate values or no data. In addition, the fission cross sections of Pu²³⁸, Am²⁴¹, Cm²⁴⁵, and the capture cross section of Pu²³⁸ were remeasured for purposes of comparison.

II. EXPERIMENTAL METHODS

Most of the samples were prepared from the transuranic elements separated from two long neutron irradiations in the Materials Testing Reactor, one of Pu²³⁹ (NR-3)¹ for a total integrated flux of 1.85×10^{22} neutrons/cm², and the other of Am²⁴¹ (99B)¹ for a flux of 4.08×10^{21} . After gross separation of the transuranium fraction from fission products, each element was highly purified by methods in common use at this laboratory which have been reported elsewhere.² In

each case, the final step of the chemical procedure yielded the pure element in acid solution from which nearly all foreign inorganic salts and organic impurities had been removed. The sample of Pu²³⁸ was separated as the alpha-decay product of 99B curium when its Cm²⁴² content was greater than 80%. The plates to be fission-counted were prepared by vacuum-vaporizing aliquots of the samples from hot tungsten filaments onto 1 in. \times 0.002 in. platinum disks. These plates were attached to a graphite shuttle which was then seated into an ionization type fission counter constructed of graphite and Lucite.³

The measurements of the fission counting rate were made in the thermal column of the Materials Testing Reactor in a neutron flux of approximately 5×10^{10} neutrons/cm² sec. The fission counter was the double chamber type with one chamber containing a Pu²³⁹ standard acting as a flux monitor. In the second chamber alternate counts were taken on the sample, blank background plates and Pu²³⁹ standards. Of the six Pu²³⁹ standards used, three were nearly isotopically pure Pu²³⁹, while the others contained a known amount of Pu²⁴⁰. The cross sections reported here are based on an 806-barn fission cross section for Pu²³⁹ for neutrons with a Maxwellian energy distribution in the thermal region.⁴ The counting rates at different discriminator voltages over the counter plateau were measured for each sample and an extrapolation to zero discrimination was made to normalize differences in plateau slope due to varying

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¹ Arbitrary designations for samples obtained from particular irradiations.

² Thompson, Harvey, Choppin, and Seaborg, *J. Am. Chem. Soc.* **76**, 6229 (1954).

³ A. Ghiorso and W. C. Bentley, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.29, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV.

⁴ *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040 (Technical Information Division, Department of Commerce, Washington, D. C., 1955), second edition.

TABLE I. Summary of results.

Sample	Nuclides	Isotopic composition (percent)	Alpha pulse analysis ^a (percent)	σ_f (barns)	(by σ_f others)
Pu ²³⁸	Pu ²³⁸	99.71 ±0.01	100	Pu ²³⁸ 18.4±0.9	18±1 ^b 20 ^c
	Pu ²³⁹	0.092±0.003			
	Pu ²⁴⁰	0.194±0.007			
Am ²⁴¹	Am ²⁴¹	100	100	3.13±0.15	3.0±0.2 ^d 3 ^e
99B Am (Sample 1)	Am ²⁴¹	37.49 ±0.29	75.39	Am ²⁴² 6250±500	~6000 ^e ~2500 ^e
	Am ²⁴²	0.413±0.01			
	Am ²⁴³	62.10 ±0.28	6.98 Cm ²⁴² :17.63		
99B Am (Sample 1, purified)	Am ²⁴¹ Am ²⁴² Am ²⁴³	(same as Sample 1)	(same as Sample 1)	Am ²⁴² 6530±500	
NR-3 Am (Sample 1)	Am ²⁴¹	0.083±0.002	1.54	Am ²⁴³ <0.071	<25 ^e
	Am ²⁴²	0.00092 ^f			
	Am ²⁴³	99.91 ±0.004	96.58 Cm ²⁴² :0.25 Cm ²⁴⁴ :0.80 Pu ²³⁸ :0.82		
NR-3 Am (Sample 1, purified)	Am ²⁴¹ Am ²⁴² Am ²⁴³	(same as Sample 1)	1.56	Am ²⁴³ <0.073	
			97.42 Cm ²⁴² :0.25 Cm ²⁴⁴ :0.81		
NR-3 Cm	Cm ²⁴²	<0.03	1.08	Cm ²⁴⁵ 1880±150	2010±150 ^g
	Cm ²⁴³				
	Cm ²⁴⁴	95.50±0.07	98.92		
	Cm ²⁴⁵	1.62±0.1			
	Cm ²⁴⁶	2.88±0.2			
99B Cm (Sample 2)	Cm ²⁴²	23.16±0.35	94.05	Cm ²⁴³ 690±50	
	Cm ²⁴³	32.06±0.43			
	Cm ²⁴⁴	44.78±0.37	5.95		
	Cm ²⁴⁵	<0.17			

^a The following alpha-decay half-lives were used in calculating specific activities: Pu²³⁸ 89.6 yr, Am²⁴¹ 461.3 yr, Am²⁴³ 8200 yr, Cm²⁴⁴ 19 yr, and Cm²⁴² 0.4455 yr.

^b Reed, Manning, and Bentley, quoted in *The Actinide Elements* (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV, Chap. 20.

^c See reference 6.

^d B. B. Cunningham and A. Ghiorso, *Phys. Rev.* **82**, 558 (1951).

^e Street, Ghiorso, and Thompson, *Phys. Rev.* **85**, 135 (1952).

^f Calculated.

^g Fields, Studier, Diamond, Mech, Inghram, Pyle, Stevens, Fried, Manning, Ghiorso, Thompson, Higgins, and Seaborg, *Phys. Rev.* **102**, 180 (1956).

sample thickness, etc. The precision obtained by recounting a sample several days later was excellent, usually amounting to less than a one percent difference, but it was somewhat less for two separate samples of the same material.

To obtain the weights of the material on the fission counting plates, the alpha-particle disintegration rates of the samples were determined by counting each plate to a 0.5% standard deviation five to eight times in a standardized low-geometry alpha counter. Alpha-particle pulse analysis of volatilized samples was used to obtain the activity ratios whenever two or more isotopes in a sample each yielded observable alpha particles. In addition, the counting rates of the samples containing Cm²⁴² were followed for decay over a six-month period, and the decay rate was correlated with the Cm²⁴² content obtained by alpha-pulse analysis. For all samples containing Cm²⁴² or Cm²⁴⁴, the activity ratios were then corrected to the time of the mass

analysis. To minimize errors from these pulse analyses, the weight of material on a fission counting plate was calculated from the alpha-disintegration rate of the nuclide giving the largest percentage of alpha particles emitted.

The isotopic composition of each sample was determined by use of a 12-inch radius, 60° sector magnetic mass spectrograph. A double tungsten filament ionization source in conjunction with an electron multiplier detector made possible the analysis of very minute samples.⁵

III. EXPERIMENTAL RESULTS

The pertinent numerical data are summarized in Table I. The last column contains previously reported values for these cross sections with their references. The limits of error listed include standard deviations in

⁵ M. G. Inghram and W. A. Chupka, *Rev. Sci. Instr.* **24**, 518 (1953).

statistical events and the authors' estimates of indeterminate errors, but do not include those occurring in alpha-decay half-lives and the fission cross section of Pu^{239} .

The value of 8200 years for the half-life of Am^{243} was obtained from the mass and alpha-pulse analysis of 99B americium. A half-life for Am^{243} can also be calculated from the mass and alpha-pulse analysis data for the NR-3 americium, but one finds a less accurate value due to the unfavorable isotopic ratio of Am^{241} to Am^{243} . The fission cross section of Am^{243} was calculated after correcting for the fissions contributed by Am^{241} and Am^{242} . Although Am^{242} was undetectable in the mass spectrometer, it was necessarily in equilibrium with the Am^{241} present, and the amount of Am^{242} could be calculated on the basis of the equilibrium mixture found in 99B americium. After these corrections had been applied, which accounted for nearly half of the observed fission rate, the remainder were assigned to Am^{243} . An upper limit for the fission cross section is given since it is possible that the fissions could arise from an isomer of Am^{244} or an undetected impurity. The possibility of an isomer of Am^{244} with a half-life shorter than three years contributing the excess fissions was eliminated by fission counting one of the NR-3 americium samples one year after the first count, under the same experimental conditions. After correcting for the growth of Pu^{239} , no decrease in the fission counting rate was found. An impurity seems unlikely in view of the fact that after repurification (uranium, neptunium, and plutonium were separated from the americium in Sample 1, by the use of anion exchange resin) there was no appreciable change in the ratio of fission counts to sample weight. A similar repurification of the 99B americium likewise resulted in a nearly constant fission count to sample weight ratio.

A small decrease in the fission counting rate per unit mass was noted after the americium was again separated from a sample of 99B curium by elution through Dowex-50 resin with ammonium lactate at 87°C. The smaller cross section for Cm^{243} is considered the more reliable and is the only one listed in Table I. In reporting the thermal-neutron fission cross sections of Cm^{243} and

Cm^{246} , any fissions contributed by Cm^{242} or Cm^{244} in each sample were assumed to be negligible.⁶

IV. PILE-NEUTRON CAPTURE CROSS SECTION OF Pu^{238}

The pile-neutron capture cross section of Pu^{238} was measured by inserting three quartz capsules, each containing about a microgram of the same Pu^{238} used in the fission cross-section measurements, into the rabbit facility of the Materials Testing Reactor for approximately two, three, and five days' neutron irradiation, respectively. The pile-neutron flux was monitored by the neutron-capture activation of cobalt wire and the results were corrected for resonance neutron activation and self-protection. After separation and purification of the plutonium, plated samples were made, and the amount of Pu^{239} produced in each irradiation was determined by fission-counting. It was necessary to make a slight correction in the fission-counting rate for those fissions contributed by Pu^{238} . Allowing for the destruction of a small fraction of the Pu^{238} and of Pu^{239} during its formation, the resulting $\text{Pu}^{238}/\text{Pu}^{239}$ ratio and the known integrated neutron flux on each sample yield a capture cross section of 520 ± 40 barns. Similar measurements giving 455 ± 50 and 489 ± 3 barns for the capture cross section have been made at Argonne National Laboratory⁷ and Chalk River, respectively.⁸

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⁶ Hanna, Harvey, Moss, and Tunnicliffe, *Phys. Rev.* **81**, 893 (1951).

⁷ Bruhlman, Bentley, and Hyde, quoted in *The Actinide Elements* (McGraw-Hill Book Company, Inc., New York, 1954), National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV, Chap. 20.

⁸ Butler, Lounsbury, and Merritt, *Can. J. Phys.* **35**, 147 (1957).