

Measurement of the Average Neutron Yield per Fission for Am^{242m} †

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The average number of neutrons per thermal neutron fission, $\bar{\nu}$, was measured for Am^{242m} with the use of a spark-chamber fission detector and a paraffin-moderated neutron detector. The value obtained is 3.24 ± 0.12 neutrons per fission. To check the method, measurements also were made on U^{235} and U^{233} , which gave 2.43 ± 0.08 and 2.54 ± 0.04 neutrons per fission, respectively.

INTRODUCTION

A TRANSURANIC isotope is Am^{242} which is formed in the excited (isomeric) state, Am^{242m} . The isomeric state decays to the ground state, with a half-life of 152 years.¹ The isomeric state has a spin of 5, while the ground state has a spin of 1. The thermal-fission cross section for the isomeric state is 6400 b while that for the ground state is 3000 b. At equilibrium, approximately 10^5 Am^{242m} nuclei exist for each ground-state nucleus so that neutron fission is predominantly by the isomeric state. Am^{242} is one of very few fissionable odd-odd nuclei available for study. Its nuclear properties and their relation to nuclear systematics are therefore of considerable interest, and of vital importance to the understanding of nuclear processes of heavy elements. The present report describes the measurement of the number of neutrons emitted per thermal-neutron fission, $\bar{\nu}$, for Am^{242m} .

EXPERIMENTAL APPARATUS AND PROCEDURE

A paraffin-moderated neutron detector in which was inserted a small spark chamber was used for measuring $\bar{\nu}$. The neutron detector² had an ungated efficiency of

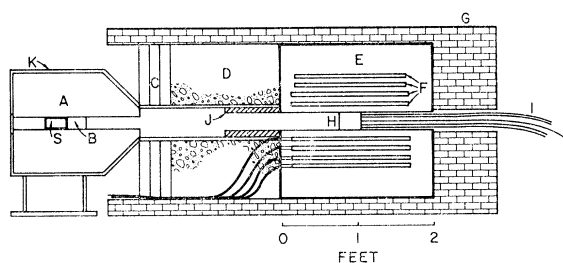


FIG. 1. Experimental arrangement for $\bar{\nu}$ measurements: *S* denotes the $10^7 n/\text{sec}$ source; *A*, the polyethylene moderator; *B* is a 3-in. plug of polyethylene; *C*, paraffin; *D*, pellets of borated polyethylene; *E*, the paraffin-moderated neutron detector; *F*, high-pressure BF_3 tubes; *G*, compressed boracic acid bricks; *H*, the spark chamber; *I*, electrical leads and air tubes to the spark chamber; *J*, a 3-in.-diam collimator of borated paraffin; *K*, 0.060-in. cadmium sheet.

† Work performed under the auspices of the U. S. Atomic Energy Commission.

¹ F. Asaro, I. Perlman, J. O. Rasmussen, and S. G. Thompson, *Phys. Rev.* **120**, 934 (1960).

² S. C. Fultz, R. L. Bramblett, J. T. Caldwell, and N. A. Kerr, *Phys. Rev.* **127**, 1273 (1962).

approximately 0.40, and consisted of a 2-ft paraffin cube containing 48 high-pressure B^{10}F_3 counters arranged in four concentric rings around a 3-in.-diam axial hole. A corona-type spark chamber³ 3 in. in diameter was placed at the center of the axial hole. It contained a single spark frame mounted in an aluminum container. Special precautions were taken in the construction of the spark chamber to give the lowest possible background from sparks arising from causes other than nuclear particles. Several materials were tried for the cathode, but most success was obtained from one constructed of aluminum and plated with gold. The samples, which were deposited on thin metal plates, were placed in close proximity to the spark frame. Fission fragments from the sample activated the spark chamber and started a gate on the scaling circuits. Fission neutrons from the sample were moderated in the paraffin and detected with the B^{10}F_3 counters. The decay time for the paraffin cube was approximately 100 μsec . The pulses from the detector were gated by 300- or 100- μsec gates as required by background considerations.

The neutrons used for the measurements were obtained from a Pu-Be neutron source which had a strength of $10^7 n/\text{sec}$. The arrangement of the apparatus is shown in Fig. 1. The neutron source was placed in the axial hole of a 14-in.-diam polyethylene moderator and aligned with the axis of the neutron detector. The thickness of the moderating plug immediately in front of the source was adjusted to give the smallest ratio of thermal-neutron counts (consistent with a reasonable count rate) when a cadmium sheet was placed in and out of a position in front of the polyethylene moderator, between the source and the detector. The accuracy of the method used for measuring $\bar{\nu}$ was checked by measuring $\bar{\nu}$ for U^{233} and U^{235} . Systematic errors appeared not to exceed 1.6%.

U^{233} AND U^{235}

The samples of U^{235} and U^{233} consisted of approximately 0.5 mg/cm^2 of material deposited within a 2-in.-diam circular area on aluminum plates 0.010 in. thick. The plate was fastened into place with the active de-

³ C. D. Bowman and R. W. Hill, *Nucl. Instr. Methods* **24**, 213 (1963).

posit of fissionable material 0.125 in. from the spark chamber frame. The characteristic curve of α counts (i.e., count rate vs voltage) was determined with a U^{235} α -source placed at the location of the sample. A similar bias curve for fission events was determined with a Cf^{252} source.

The threshold for detection of fission events was below that for α -particle detection, and the operating voltage of the spark chamber was chosen between these thresholds. The operating voltage was 2200 V for air at a pressure of 56 cm of mercury. The background rate for the spark chamber was obtained by counting the pulses with the neutron source removed. The source emitted many neutrons which were not thermalized in its polyethylene moderator, and these neutrons gave rise to a background which had to be subtracted from the total neutron count. This background was obtained with the neutron source present by measuring the neutron counts per gate when the gate was triggered by a pulse generator instead of the fission counter. It was measured with a statistical accuracy of 1% for each run, and had an average value of approximately 0.323 for U^{235} and 0.544 for U^{233} .

TABLE I. $\bar{\nu}$ Measurements for U^{233} and U^{235} .

Isotope	Fissions, F	True neutron counts, C_n	Av. No. of neutrons per fission, $\bar{\nu}$
U^{235}	4904	4269	2.43 ± 0.08
U^{233}	11 190	7884	2.54 ± 0.04

The efficiency of the neutron detector was measured with a Cf^{252} source. The fission particles from the Cf^{252} triggered the gate of the scaling circuits, and the true neutron counts C_n were observed. The efficiency ϵ of the neutron detector is given by the expression

$$C_n = \bar{\nu}_{252} F \epsilon, \quad (1)$$

where $\bar{\nu}_{252}$ denotes the value of $\bar{\nu}$ for Cf^{252} and F is the number of fissions. The value for $\bar{\nu}$ used for Cf^{252} was 3.78 (an average value between that of Asplund *et al.*⁴ and Hopkins and Diven⁵). The ungated efficiency of the detector is slowly dependent upon the average neutron energy. Thus, the value of ϵ obtained for Cf^{252} may not hold for neutrons from the fission of U^{235} or U^{233} . The efficiency was checked further by measuring the "ring ratio" of the neutron counts.⁶ This ratio is the ratio of the counts obtained from the fourth ring of BF_3 counters to the counts obtained from the first ring, in the paraffin-moderated detector. The efficiency and ring ratios had been measured as a function of neutron

⁴ I. Asplund-Nilsson, H. Condé, and N. Starfelt, Nucl. Sci. Eng. 16, 124 (1963).

⁵ J. C. Hopkins and B. C. Diven, Nucl. Phys. 48, 433 (1963).

⁶ J. T. Caldwell, R. L. Bramblett, B. L. Berman, R. R. Harvey, and S. C. Fultz, Phys. Rev. Letters 15, 976 (1965).

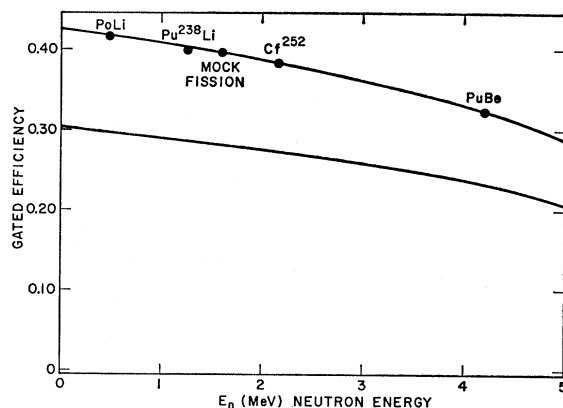


FIG. 2. Gated efficiency vs neutron energy for the neutron detector. The top curve includes data points taken with neutron sources which were calibrated with a MnSO_4 bath, and with a gate of 300- μsec duration. The lower curve applies to a 100- μsec gate.

energy by using a wide variety of neutron sources and a 14-MeV neutron generator. The relation between average neutron energy and neutron detector efficiency is shown in Fig. 2, for both 300- and 100- μsec gates.

With a knowledge of the number of fissions, the total number of neutrons counted, the α background, random-neutron background, and the efficiency of the neutron detector, the value for $\bar{\nu}$ can be calculated from Eq. (1) for which the neutron counts and fission counts are corrected for backgrounds. The results obtained for U^{235} are shown in Table I. For these measurements the gated efficiency of the neutron detector was 0.388, with a 300- μsec gate and having corrected for the difference between neutron spectra for Cf^{252} and U^{235} . The weighted average value of $\bar{\nu}$ for U^{235} was determined to be 2.43 ± 0.08 neutrons/fission at thermal energy. The results obtained for U^{233} also are shown in Table I. The gated efficiency for these data was 0.277, with a 100- μsec gate and again having corrected for the neutron spectrum. The average value of $\bar{\nu}$ for U^{233} was found to be 2.54 ± 0.04 . The reduced counting duty factor, obtained with the shorter gate, facilitated an increase in the strength of the neutron source and better counting statistics.

Am^{242}

Two samples of americium were prepared for the spark chamber, about 8 days apart. The material was separated in a calutron at Oak Ridge National Laboratory, to achieve an enrichment of approximately 20% Am^{242} from an original enrichment of 1% Am^{242} . The samples were prepared by electrodeposition of the americium on a 0.010-in.-thick nickel foil. The quantities of isotopes used for the deposits are shown in Table II.

The americium was uniformly deposited within a 2-in.-diam circle at the center of the nickel foil. Am^{242} decays by β decay to Cm^{242} with a half-life of 152

TABLE II. Quantities of americium isotopes used in the samples.

	Am ²⁴¹	Am ²⁴²	Am ²⁴³	Total Am
Isotopic abundance	0.7951	0.1980	0.0069	1.000
1st sample (mg/cm ²)	0.915	0.228	0.008	1.151
2nd sample (mg/cm ²)	0.952	0.237	0.008	1.197

years. The Cm²⁴² decays by spontaneous fission with a half-life of 7.2×10^6 years. Thus, to avoid the presence of an undesirably high fission background in the Am²⁴² samples, it was necessary to extract the curium isotopes just before preparing the samples. Hence, any fission background present at time zero would be due to spontaneous fissions from Am²⁴². The fission background from the Cm²⁴² would build up as the sample aged according to the relation

$$F_s = \lambda_{FC} \text{Cm}(t), \quad (2)$$

where F_s is the spontaneous fission rate, $\text{Cm}(t)$ is the number of atoms of Cm²⁴² at time t , and λ_{FC} is the decay constant for spontaneous fission of Cm²⁴². For the case considered, we have $\lambda_{Am} \gg \lambda_{FC}$, where λ_{Am} is the decay constant for Am^{242m}. Hence,

$$\text{Cm}(t) = \lambda_{Am} \text{Am}_0 t = (0.00456) \text{Am}_0 t, \quad (3)$$

where Am_0 is the number of atoms of Am²⁴² at time zero, and t is the time in years. Thus, for the spark detector used, which had an efficiency of approximately 0.01, we should expect to obtain an increase in the fission background of approximately 16 fission counts/h day. The growth of spontaneous fission was observed to be approximately this amount and corrections were made accordingly. The spontaneous-fission measurements will be discussed in detail in a subsequent article.⁷

With the first sample of Am²⁴² the runs were made short so that the increase in the background due to spontaneous fission could be followed. These runs were

TABLE III. $\bar{\nu}$ Measurements on Am²⁴² for first sample.

Run	Fissions, F	True neutrons, C_n	$\bar{\nu}$
12	255	234	3.37
13	153	155	3.72
14	237	223	3.46
15	260	222	3.14
16	167	157	3.45
17	201	162	2.96
18	326	272	3.07
19	140	131	3.44
20	473	388	3.02
21	422	356	3.10
22	475	425	3.40
23	176	163	3.40
24	241	189	2.88
26	309	276	3.28
27	375	362	3.55
Weighted average			3.27 ± 0.18

⁷ J. T. Caldwell, S. C. Fultz, C. D. Bowman, and R. W. Hoff, University of California Report No. UCRL-70062, 1966 (unpublished).

about 2 hours long, and each was followed by a background run (neutron source removed) of 2 hours. (The average gated neutron background was approximately 0.560n/gate.) In this manner, the behavior of the spark chamber, as well as the growth of the spontaneous fission, was followed. Longer runs were made with the second sample to obtain better statistics. The data obtained for the two samples are given in Tables III and IV. The gated efficiency for the neutron detector was 0.272 for a 100- μ sec gate. The weighted average of $\bar{\nu}$ from all the data on Am²⁴² is 3.24 ± 0.12 neutrons/fission.

DISCUSSION OF $\bar{\nu}$ RESULTS

The measurements made of $\bar{\nu}$ for U²³³, U²³⁵, and Am²⁴² are summarized in Table V. In the third column are data which have been calculated or averaged from results obtained elsewhere. In the cases of U²³³ and U²³⁵ the values of $\bar{\nu}$ were obtained by least-squares averaging⁸ of a large number of experimental values. It is evident that the values measured in the present experiment agree within experimental error with the averaged values.

TABLE IV. $\bar{\nu}$ Measurements on Am²⁴² for second sample.

Run	Fissions, F	True neutrons, C_n	$\bar{\nu}$
33	595	555	3.43
34	685	628	3.37
35	714	582	3.00
36	611	568	3.42
37	546	450	3.03
38	490	443	3.32
39	508	417	3.02
Weighted average			3.21 ± 0.16

The neutron detector efficiencies used for the U²³³ and U²³⁵ were determined from ring ratios and the use of Fig. 2. The ring ratio for Am²⁴² was the closest of all to that for Cf²⁵², implying that the average energy of the neutron spectrum of Am²⁴² was the nearest to that for Cf²⁵². This would be expected, since its value for $\bar{\nu}$ was nearest to that for Cf²⁵². The relation between the average neutron energy, E_{av} , and $\bar{\nu}$ is given by Terrell⁹ as

$$E_{av} \cong 0.74 + 0.653(\bar{\nu} + 1)^{1/2}. \quad (4)$$

From this formula, the average energies of fission neutrons for the isotopes considered here were calculated, and are given in the fourth column of Table V.

It was not possible to obtain a comparison of the presently measured value of $\bar{\nu}$ with earlier data, as none has been published. A value of 3.36 ± 0.03 was obtained by Jaffee and Lerner¹⁰ at Argonne National

⁸ R. Sher and J. Felberbaum, Brookhaven National Laboratory, Rept. BNL 918, 1965 (unpublished).

⁹ J. Terrell, Phys. Rev. **113**, 527 (1959); **217**, 880 (1962).

¹⁰ A. H. Jaffee and J. Lerner (private communication; to be published).

Laboratory. A comparison also can be made with a value calculated by Gordeeva and Smirenkin¹¹ by using an empirical expression based on early $\bar{\nu}$ measurements for other isotopes. They deduced the relation

$$\bar{\nu} = 0.1894Z + 0.007A - 16.60 + \delta_{\nu}, \quad (5)$$

where $\bar{\nu}$, Z , and A denote the average value of $\bar{\nu}$ at thermal neutron energy, the atomic number, and the mass number, respectively, and $\delta_{\nu} = 0.09\xi$, where $\xi = +1, -1, \text{ or } 0$ for odd-odd, even-even, and odd- A target nuclei, respectively.

From Eq. (5), the $\bar{\nu}$ value of 3.17 neutrons/fission was obtained for Am^{242m} . This agrees reasonably well with the present experimental value, 3.24 ± 0.12 . The

TABLE V. Values of $\bar{\nu}$ from this experiment and from other sources.

Isotope	$\bar{\nu}$ (this experiment)	$\bar{\nu}$ (calculated or averaged)	E_{av}^a (MeV)
U^{233}	2.54 ± 0.04	2.504 ± 0.007^b	1.96
U^{235}	2.43 ± 0.08	2.442 ± 0.006^b	1.95
Am^{242}	3.24 ± 0.12	3.17^c	2.09
Cf^{252}		3.78^d	2.16

^a Calculated from Eq. (4).

^b See Ref. 8.

^c See Ref. 11.

^d See Refs. 4 and 5.

Argonne value also agrees with the present value, within the limits of the experimental accuracy.

In the process of taking the data, the number of the neutrons per gate was measured. The statistics of these data, however, were not good enough to make possible accurate calculations of P_{ν} , the probability of obtaining ν neutrons per fission. However, by assuming a Gaussian form,⁹ the distribution can be calculated from the measured value of $\bar{\nu}$. This was done and the results are shown in Fig. 3.

¹¹ L. D. Gordeeva and G. N. Smirenkin, *Atomnaya Energ.* (USSR) **14**, (1963).

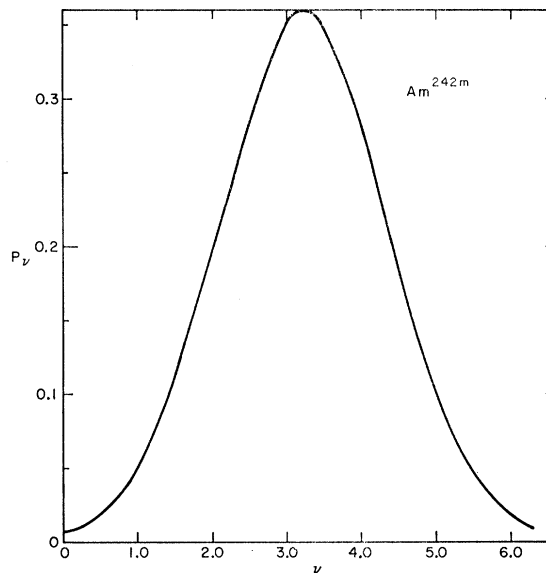


FIG. 3. Calculated values of P_{ν} (the probability of obtaining ν neutrons per fission) vs ν , based upon the measured value of $\bar{\nu}$. The method is that of Terrell (Ref. 9).

CONCLUSIONS

The number of neutrons emitted from Am^{242} per thermal neutron fission, $\bar{\nu}$, has been measured and found to be 3.24 ± 0.12 . The method was checked by using it on the measurement of $\bar{\nu}$ for U^{235} and U^{233} . For these measurements the efficiency of the neutron detector was corrected for differences in the neutron spectra of U^{235} and U^{233} from that of Cf^{252} . The maximum deviation of the U^{235} and U^{233} results from those obtained by averaging over many determinations⁸ is only 1.6%. The method therefore appears to be valid within the limits of the statistical accuracy.

ACKNOWLEDGMENTS

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