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Spontaneous Fission Half-Life of Am^{242m}†

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The spontaneous-fission half-life for the odd-odd nucleus Am^{242m} has been measured by a prompt-neutron fission-fragment coincidence method to be $(9.5\pm3.5)\times10^{11}$ years. A correlation of experimental spontaneous-fission half-life data for nuclei of all classes was found to be $\log_{10}\tau-(\beta-Z^2/A)\delta M=-9.268(Z^2/A)+374.0$, where β takes the values 42.22, 40.67, and 40.40 for even-even, odd-A, and odd-odd nuclides, respectively, τ is the spontaneous-fission half-life in years, and δM is the mass difference in milli-mass-units (mmu) between the values obtained from Green's smoothly varying mass formula and the experimentally measured values.

INTRODUCTION

SPONTANEOUS-FISSION systematics has been the subject of considerable study in recent years. It is of interest from the point of view of understanding fundamentals of the fission process, as well as from the aspect of heavy-element production. A large number of even-even nuclides are available, and their spontaneous-fission half-lives have been measured. Similarly, a number of even-odd nuclides have been measured. However, there are very few odd-odd nuclides available for investigation, although they are necessary for a thorough study of the systematics. Am^{242} is one of the few odd-odd fissionable nuclei available for such study. It is formed by neutron capture in the isomeric state Am^{242m}, which decays by the emission of a 48-keV gamma ray with a half-life of 152 years.[1] The ground state decays with a half-life of only 16 h, so that at equilibrium the ratio of Am^{242m} atoms to those in the ground state is approximately 10^5. The present report describes measurements on the spontaneous-fission half-life of Am^{242m}.

EXPERIMENTAL PROCEDURE

A sample of Am^{242m} was prepared by thermal neutron irradiation of Am^{241}. At the end of the irradiation it contained approximately 1% Am^{242m}. The sample was then passed through a mass separator to achieve a 20% Am^{242m} enrichment.

Cm^{242} contaminant builds up in the Am^{242m} sample through the following sequence:

$$Am^{242m}\xrightarrow[152\text{ years}]{\text{IT}} Am^{242} \xrightarrow[16\text{ h}]{(84\%)\beta^-} Cm^{242} \xrightarrow[163\text{ days}]{\alpha} Pu^{238} \xrightarrow[86.4\text{ years}]{\alpha} . \qquad (16\%)\downarrow\epsilon\ Pu^{242} \tag{1}$$

Since Cm^{242} has a spontaneous-fission half-life of 7.2×10^6 years, the fission rate from this daughter product gives rise to a large background of fission events which must be eliminated, insofar as possible, for an accurate spontaneous-fission half-life measurement. By repeated chemical separations it was found that the Cm/Am ratio could be reduced to less than 3×10^{-7}.

Two independent experiments were performed. The first yielded only an upper limit to the spontaneous-fission half-life of Am^{242m}, whereas the second established a definite value. For the first experiment, about 50 mg of the sample was electroplated on a foil to a thickness of about 500 μg/cm^2. A corona spark chamber[2] was chosen as the fission-fragment detector because of its property of excellent discrimination against α particles. The measurements which were started 24 days after the Cm separation were made intermittently over a period of 6 days. At the time t, the observed count rate R from the spark chamber containing the Am^{242m} is given by the expression

$$R=n(1-e^{-\lambda_{Cm}t})+k, \tag{2}$$

where the constant n is proportional to the spontaneous-fission decay constant of Cm^{242}, λ_{Cm} is the α-decay constant for Cm^{242}, and k is the rate due to spurious backgrounds in the detector and to spontaneous fission of Am^{242m}.

A least-squares fit of Eq. (2) to the data is shown in Fig. 1. The error bars in the figure represent the counting statistical errors on the points which were used to weight the data for the least-squares fit. The constants of Eq. (1) were found to be $n=1\,112\,000$ $\pm125\,000$ counts/day and $b=3000\pm15\,400$ counts/day. These constants can then be combined to find the

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

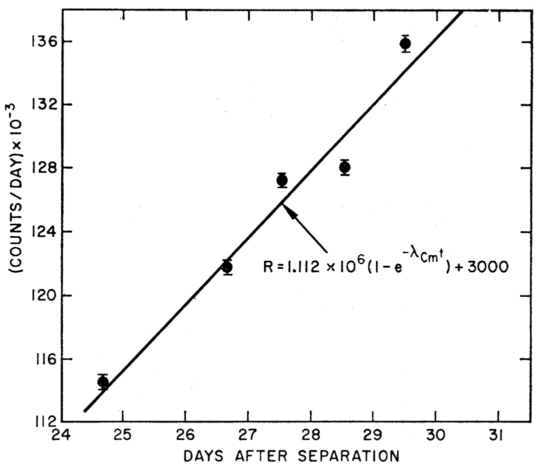

FIG. 1. Fission-count rate versus days after chemical separation of Cm^{242} from Am^{242m}. The straight line is given by the relation $F_s = 1.112\times10^6(1-e^{-\lambda_{Cm}t})+3000$.

spontaneous-fission half-life for Am^{242m}. Since the error on the constant b was so large, the error itself, 15 400, was used to determine a lower limit to the half-life of 2×10^{11} years.

The second measurement on the spontaneous-fission half-life of Am^{242m} was carried out with a method of prompt neutron-fission fragment coincidence. This experiment will be discussed in more detail than the previous one, since it yielded a specific value for the spontaneous-fission half-life rather than a lower limit. The apparatus used and the method of measurement have been described in detail in a report on the measurement of $\bar{\nu}$ for thermal-neutron–induced fission in this isotope.[3] Fission fragments were detected in a 3-in.-diam corona spark chamber. The α-particle activity of the Am^{242} source was about 10^8/cm^2/sec. Since the spontaneous-fission rate was about 10^{-11} of the total α-particle rate, only a small α-particle pileup spark rate could influence the results markedly. To minimize this effect, true spontaneous-fission events were distinguished from α-particle pileup pulses by placing the spark counter in the center of a 4π paraffin-moderated neutron detector and observing coincidences between fission fragments and the prompt neutrons moderated in the detector.

The fission-fragment–prompt-neutron coincidence rate for Am^{242m} is given by

$$R(\mathrm{Am}) = N_{\mathrm{Am}}\lambda_{FA}\epsilon_N\epsilon_F\bar{\nu}_{FA}\,, \quad (3)$$

where N_{Am} is the number of Am^{242m} atoms in the sample, λ_{FA} is the decay constant for spontaneous fission of Am^{242m}, ϵ_F is the spark-counter fission-detector efficiency, ϵ_N is the neutron-detector efficiency, and $\bar{\nu}_{FA}$ is the average number of neutrons emitted per fission for Am^{242m} spontaneous fission.

Similarly, for times short compared to the Cm^{242} α-particle half-life (163 days) the fission-fragment–prompt-neutron coincidence rate for the Cm^{242} which grows in is

$$R(\mathrm{Cm}) = N_{\mathrm{Am}}\lambda_{\mathrm{Am}}\lambda_{FC}\epsilon_F\epsilon_N\bar{\nu}_{FC}t\,, \quad (4)$$

where $\bar{\nu}_{FC}$ is the average number of neutrons emitted per fission from Cm^{242} spontaneous fission, t is the time measured from the zero time for the Cm^{242} which grows in, and λ_{Am} is the isomeric-transition (IT) decay constant for Am^{242m}, with an 84% branching ultimately through to Cm^{242}.

At a given time t, the total observed coincidence rate R_0 is

$$R_0 = R(\mathrm{Am}) + R(\mathrm{Cm}) \quad (5)$$

or

$$R_0 = N_{\mathrm{Am}}\epsilon_N\epsilon_F(\bar{\nu}_{FA}\lambda_{FA} + \bar{\nu}_{FC}\lambda_{\mathrm{Am}}\lambda_{FC}t)\,. \quad (6)$$

Thus, the total observed rate of spontaneous-fission neutron coincidences is linearly increasing with time, with a zero-time intercept b proportional to λ_{FA} and a slope m proportional to λ_{FC}. On taking the ratio m/b, we get

$$m/b = (\bar{\nu}_{FC}\lambda_{\mathrm{Am}}\lambda_{FC})/\bar{\nu}_{FA}\lambda_{FA}\,, \quad (7)$$

which is independent of detection efficiency. Furthermore, an examination of $\bar{\nu}$ systematics[4] indicates that $\bar{\nu}_{FA}\simeq\bar{\nu}_{FC}$ is valid to within 10%. Taking the IT half-life of Am^{242m} as 152 ± 7 years and expressing Eq. (7) in terms of half-lives τ, we get

$$m/b = 4.37\times10^{-7}(\tau_{FA}/\tau_{FC})\,, \quad (8)$$

where $\lambda_{\mathrm{Am}} = 5.20\times10^{-7}$ (h)$^{-1}\times0.84$ (branching to Cm^{242}). Taking[5]

$$\tau_{FC} = 7.2\times10^6 \text{ year},$$

we obtain

$$\tau_{FA} = 1.65\times10^{13}(m/b) \text{ year}. \quad (9)$$

On plotting the spontaneous-fission rate of the sample versus time in hours, starting immediately after the Am^{242m} sample was separated from the Cm^{242}, the growth of the spontaneous-fission rate of the Cm^{242} can be followed and is superposed upon a constant background of Am^{242m} spontaneous fission. Such data were obtained and are shown in Fig. 2.

As previously noted, two separations of the Cm were made, after which the ratio of Cm^{242}/Am^{242} was assured to be less than 3×10^{-7}. The zero time for completion of the chemical separation was found by following the

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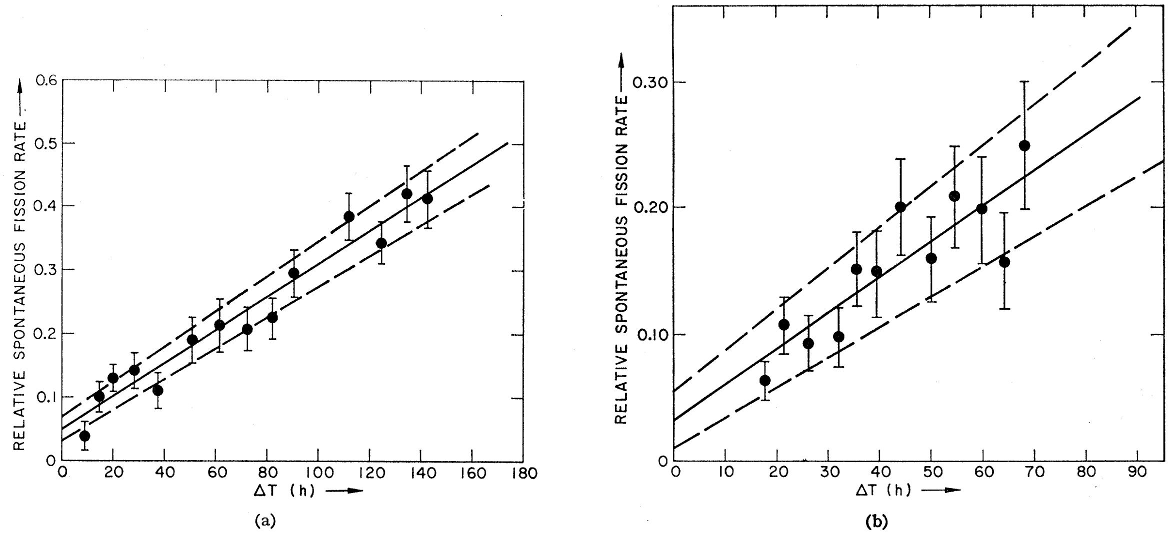

FIG. 2. Relative spontaneous-fission–neutron-coincidence rate versus time after separation of the Cm^{242} from the Am^{242m} sample. Fig. 2(a) shows data obtained from the first sample, while Fig. 2(b) shows data obtained from the second sample. The solid lines are obtained from a least-squares fit to the data, with the dashed lines representing the rms deviations to these fits.

growth of the Cm^{242} α activity for a period of a few hours after the last chemical separation, then extrapolating linearly back to zero Cm^{242} α rate. Approximately 170 h after the last chemical separation, the α-particle rate gave 8.2×10^{-5} for the ratio Cm^{242}/Am^{242m}. The spontaneous-fission data correspondingly gave $(8\pm3)\times10^{-5}$ for this ratio, which is in good agreement.

RESULTS

The data shown in Fig. 2 are the fission-fragment–prompt-neutron coincidence-rate data plotted as a function of the time measured from the extrapolated time for zero concentration of Cm^{242}. The coincidence counting rate was corrected for cosmic-ray background (less than 3%) and for the small nonlinear effect due to the Cm^{242} α half-life decay of 163 days (a 2.5% correction after 140 h). As indicated in a previous report,[3] two independent runs were made with separate Am^{242} samples. The results from the first and second samples are shown in Figs. 2(a) and 2(b), respectively. The solid lines in each are least-squares fits to the data, while the dotted lines represent the rms deviations. Values of m/b were determined from each run, and when substituted into Eq. (9) yielded values for the spontaneous-fission half-life of Am^{242m}. For the first run, the spontaneous-fission half-life of Am^{242m}, τ_{FA}, is $(8.3\pm3.7)\times10^{11}$ years and from the second run it is $(1.38\pm0.98)\times10^{12}$ years. The weighted average of the two runs is

$$\tau_{FA}=(9.5\pm3.5)\times10^{11}\text{ years},$$

or the ratio of

$$\tau_{FA}/\tau_{FC}=(1.32\pm0.42)\times10^{-5}.$$

The errors indicated are based upon rms deviations of the least-squares fits to the data. Included are the effects of a 10% possible systematic error.

DISCUSSION

It is of interest to see how the measured half-life for spontaneous fission of Am^{242m} fits into possible systematics for other spontaneously fissionable nuclei. Swiatecki[6] derived an empirical correlation of spontaneous-fission data by using the parameters Z^2/A and δM, where Z^2/A is the usual fissionability parameter and δM is the mass excess as determined from the difference between a smoothly varying mass formula, such as that given by Green,[7] and the ground-state, experimentally measured mass. He found that an empirical relationship of the form below fitted observed spontaneous-fission half-lives quite well.

$$\log_{10}\tau_{FX}+K\delta M=K\theta+B+\text{higher-order terms in }\theta, \quad (10)$$

where τ_{FX} is the spontaneous-fission half-life in years of a nuclide denoted by X, and $\theta=(Z^2/A)-37.5$.

For values of Z^2/A less than 39, only the $K\theta+B$ terms are necessary for an adequate fit. Nuclides, when separated into even-even, even-odd, and odd-odd types, were found to be distinguished by a different value of B, while the coefficients of the powers of θ remained identical for good fits to the data. Other correlations based upon various semiempirical mass formulas

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[7] A. E. S. Green, Phys. Rev. **95**, 1006 (1954).

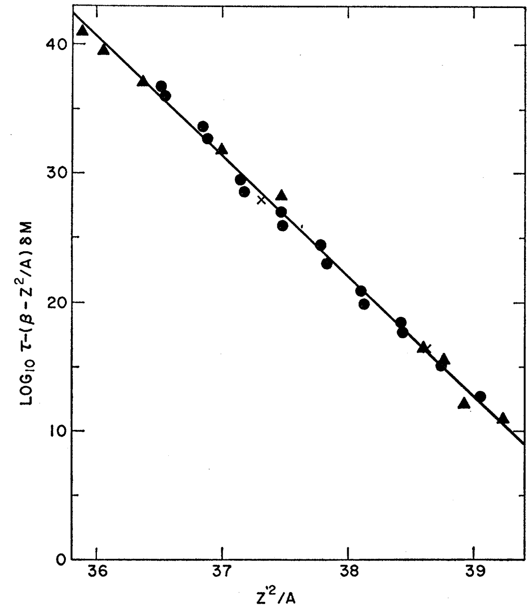

FIG. 3. Plot of $\log_{10}\tau-(\beta-Z^2/A)\delta M$ versus Z^2/A for even-even (filled circles), odd-A (filled triangles), and odd-odd (crosses) nuclei. The values for β are 42.22, 40.67, and 40.40, respectively, for these classes of nuclei.

have been advanced by Dorn,[8] Johannson,[9] and Viola and Wilkins[10] and have extended the correlation beyond measured mass values.

For the odd-odd case considered in these investigations, only the spontaneous-fission half-life of Es^{254} was available to determine the correlation constants. With the exception of the article by Johannson,[9] in which no attempt was made to account for odd-odd systematics, values for the spontaneous-fission half-life of Am^{242m} inferred from the correlations are high compared to that measured in the present experiment. Values for $\log_{10}\tau_{FA}$, as deduced from the results of Swiatecki,[6] Dorn,[8] and Viola and Wilkins,[10] are 14.7, 15.3, and 18.5, respectively. The value from the present experiment is 12.0.

In view of the large discrepancy between the measured value for τ_{FA} and the values deduced from the above results, an effort was made to improve the empirical correlation of spontaneous-fission half-lives. The emphasis in this correlation was to achieve a better understanding of the relative differences among even-even, odd-A, and odd-odd nuclei in spontaneous fission. For this purpose, a rather restrictive range (36–39) of Z^2/A was chosen for the correlation, since in this range of Z^2/A there are representative half-life data for all three classes of nuclei. In addition, previous empirical correlations of a similar type[6,8] indicated that only linear terms in Z^2/A would be required for a good fit to the data. Thus it is hoped the results of the present correlation will serve usefully as a guide to the general relationships among the three classes for all values of Z^2/A. The treatment in detail of particular half-life values for $Z^2/A>39$ should be approached more fundamentally.

For this correlation, the parameters Z^2/A and δM were used in a generalization of the Swiatecki[6] method. Here, δM is given as the difference in mmu between the mass value given by Green[7] and the experimental mass values given by Hyde *et al.*[11] The correlation is expressed in the relation

$$\log_{10}\tau-(\beta-Z^2/A)\delta M=\alpha Z^2/A+\gamma\,, \qquad (11)$$

where the parameters α, β, and γ are determined from the data. The nuclides used in this correlation, with

TABLE I. List of nuclides used in the least-squares correlation procedure. Z^2/A is the usual fission parameter, τ is the spontaneous-fission half-life in years, and δM is the mass difference in millimass units between the values obtained from Green's smoothly varying mass formula (Ref. 7) and the experimentally measured values. Values of τ used (other than for Am^{242m}) were obtained from the compilation by Hyde *et al.* (Ref. 11). The Am^{242m} value is from the present experiment.

Nuclide	Z^2/A	$\log_{10}\tau$	$-\delta M$	Class
Pu^{244}	36.21	10.4	4.67	even-even
U^{232}	36.48	13.9	4.00	even-even
Pu^{242}	36.51	10.9	4.42	even-even
Pu^{240}	36.82	11.1	4.19	even-even
Cm^{250}	36.86	4.3	5.32	even-even
Pu^{238}	37.13	10.7	3.70	even-even
Cm^{248}	37.16	6.7	4.33	even-even
Pu^{236}	37.44	9.5	3.69	even-even
Cm^{246}	37.46	7.2	3.96	even-even
Cm^{244}	37.77	7.1	3.91	even-even
Cf^{254}	37.81	−0.8	5.42	even-even
Cm^{242}	38.08	6.9	3.40	even-even
Cf^{252}	38.11	1.9	4.39	even-even
Cm^{240}	38.40	6.3	3.17	even-even
Cf^{250}	38.42	4.2	3.57	even-even
Cf^{248}	38.73	3.9	3.26	even-even
Cf^{246}	39.04	3.3	2.97	even-even
Pa^{231}	35.85	16.0	5.22	odd-A
U^{235}	36.02	17.3	4.81	odd-A
U^{233}	36.33	17.5	4.52	odd-A
Pu^{239}	36.97	15.7	4.35	odd-A
Am^{241}	37.45	14.4	4.33	odd-A
Cf^{249}	38.57	9.2	3.48	odd-A
Es^{253}	38.74	5.8	5.05	odd-A
Fm^{257}	38.91	2.0	5.74	odd-A
Fm^{255}	39.22	4.0	4.85	odd-A
Am^{242}	37.29	12.0	5.17	odd-odd
Es^{254}	38.59	5.2	6.25	odd-odd

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[9] S. A. E. Johannson, Nucl. Phys. **12**, 449 (1959).
[10] V. E. Viola, Jr. and B. D. Wilkins, Nucl. Phys. **82**, 65 (1966).
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the corresponding Z^2/A, $\log_{10}\tau$, and δM values, are presented in Table I.

Least-squares fits to Eq. (11) were performed for 17 even-even nuclides and 9 odd-A nuclides, each class of nuclide being treated independently. It was found that the parameters α and γ were identical within experimental error for both classes of nuclides, and that only a different value of β characterized the classes. As only two points were available for odd-odd nuclei, a three-parameter, least-squares fit could not be made. However, it was observed that a single value of β was obtained when either α or γ (or both) were assumed to be the same as for the other two classes. Thus, all classes of nuclei follow the empirical relation given by Eq. (11), with only the parameter β being different for different classes. By following an iterative least-squares procedure involving spontaneous-fission half-lives from all classes of nuclides, the constants of Eq. (11) were determined, and are given by

$$\log_{10}\tau-(\beta-Z^2/A)\delta M=-9.268(Z^2/A)+374.0, \quad (12)$$

where β takes the values of 42.22 ± 0.15 for even-even nuclides, 40.67 ± 0.14 for odd-A nuclides, and 40.40 ± 0.04 for odd-odd nuclides. The stated uncertainties are those computed by taking the rms sum of deviations between the experimental spontaneous-fission half-lives and those obtained from the correlation functions.

The three values of β thus obtained offer an interesting comparison of spontaneous fission for the three classes of nuclides. The even-even nuclei are set well apart from the other two classes. However, odd-odd and odd-A nuclei are quite close in their spontaneous-fission correlations. In fact, considering that values on only two odd-odd nuclei were available, the difference in β for these two classes is hardly significant. A plot of $\log_{10}\tau-(\beta-Z^2/A)\delta M$ versus Z^2/A is shown in Fig. 3. The agreement for all classes of nuclei is evident.

CONCLUSION

Generally speaking, spontaneous-fission half-lives obtained from the correlation functions of the present work (for the Z^2/A range 36–39) are in good agreement with those obtained for even-even and odd-A nuclei in previous correlations. However, the half-life values for odd-odd nuclei found in the present correlation generally result in lower values for odd-odd nuclei than had been predicted previously. The implication for heavy-element production is thus a negative one, in the sense that odd-odd nuclides cannot be expected to have a significantly greater stability against spontaneous fission than corresponding odd-A nuclei.

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