# Spontaneous-Fission Decay Constant of $^{241}Am$†

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The $^{241}Am$ spontaneous-fission decay constant has been determined with solid-state track recorders of pre-etched mica. Measurements have been carried out with electroplated $^{241}Am$ sources. Consistent spontaneous-fission-rate measurements have been obtained for a large range of fission track densities over more than a 3-yr period. Systematic error due to source impurities has been established as insignificant relative to statistical error. The present experimental result for the $^{241}Am$ spontaneous-fission decay constant is $\lambda_f = (6.04 \pm 0.13) \times 10^{-15}$ yr$^{-1}$.

## I. INTRODUCTION

TECHNIQUES for precise fission-rate measurements with solid-state track recorders (SSTR) have already been established[1] and applied for the observation of very long spontaneous-fission half-lives.[2,3] This method has been employed in the present $^{241}Am$ experiments, which utilized SSTR of pre-etched mica. These experiments have been in progress for some time and a preliminary account of this work has been reported.[4]

Since the $\alpha$-to-fission rate for $^{241}Am$ is greater than $10^{11}$, this isotope provides a valuable test of the ability of mica SSTR to record fission-fragment tracks in extremely high $\alpha$-particle background. While surface damage to the mica was apparent in these experiments, fission-fragment tracks still stood out with distinct clarity. In one such experiment, mica was exposed to approximately $4 \times 10^{13}$ $\alpha$ particles/cm$^2$ and although considerable surface damage resulted, the fission-fragment tracks were still recorded with good clarity. This insensitivity to background effects demonstrates the suitability of properly selected pre-etched mica for this type of measurement.

## II. SOURCE PREPARATION AND CALIBRATION

In contrast with the measurement of the spontaneous-fission decay constant of $^{238}U$,[3] asymptotically thick sources of $^{241}Am$ are not readily available. Consequently, $^{241}Am$ sources were prepared by electrodeposition on platinum over an area of 1.96 cm$^2$. Such sources are known to be nonuniform and therefore require individual calibration.[1]

To examine the relative efficiency as a function of thickness $\mu$, $^{241}Am$ sources were prepared in the region $\mu < 500$ $\mu$g/cm$^2$. Four independent calibration exposures were performed with these sources on the uniform flux assembly. The results of these experiments are summarized in Table I.

In these irradiations, all sources are simultaneously exposed to a uniform flux of slow neutrons.[1] Fission induced by a uniform neutron exposure of these $^{241}Am$ sources provides track data for the *relative* efficiency calibration of these sources. The first exposure was bare, whereas the remaining exposures employed cadmium covers. Since source nonuniformity effects can be counteracted to a limited extent by introducing an annular mask between the SSTR and the electroplated source, annular aluminum masks 0.008 cm thick and 0.44 cm$^2$ in area were used for the thicker $^{241}Am$ sources $\mu > 100$ $\mu$g/cm$^2$. However, the existence of nonuniformity requires that all fission tracks be counted in each mica SSTR. In many cases, total track counts were obtained by two different observers and very good agreement was found.

To obtain an absolute scale for these relative efficiency measurements, at least one high-quality source must be obtained in the region $\mu < 100$ $\mu$g/cm$^2$. In this region, it has been established that the highest possible efficiency, i.e., the optical efficiency, is attained with high-quality fission sources. For mica, the optical efficiency is known to be 95%.[1] Since source 2 lies in this region and consistently yielded the highest track density per $\mu$g of $^{241}Am$ in all experiments, it can be assumed to possess the optimum 95% optical efficiency. All other efficiencies listed in Table I have been obtained in terms of this assignment for source 2.

In addition to the known 0.5% error in the optical efficiency,[1] the experimental error given in Table I contains independent contributions due to fission-track

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[2] R. Gold, R. J. Armani, J. H. Roberts, and A. Behkami, Bull. Am. Phys. Soc. **11**, 825 (1966).
[3] J. H. Roberts, R. Gold, and R. J. Armani, Phys. Rev. **174**, 1482 (1968).
[4] J. H. Roberts, R. Gold, and R. J. Armani, in Proceedings of the International Topical Conference on Nuclear Track Registration in Insulating Solids and Applications, 1969, Vol. 2, Chap. VIII, p. 15 (unpublished).

TABLE I. Efficiency measurements for the $^{241}$Am sources.

| Exposure No. | Source[a] No. | $\mu$g $^{241}$Am | $\mu$[b] | Total tracks | Tracks/$\mu$g $^{241}$Am | Efficiency[c] |
|---|---|---|---|---|---|---|
| 1 (Bare) | 1 | 93.926 | 47.9 | 7 472 | 79.55 | 0.907±0.014 |
| | 2($p$) | 159.962 | 81.5 | 13 330 | 83.33 | 0.950±0.005 |
| | 3($m$, $p$) | 77.818 | 176 | 6 162 | 79.18 | 0.903±0.015 |
| | 4($m$) | 177.57 | 403 | 11 926 | 67.16 | 0.766±0.011 |
| | 5($m$) | 210.97 | 479 | 14 006 | 66.39 | 0.757±0.010 |
| 2 (Cd) | 1 | 93.926 | 47.9 | 9 327 | 99.30 | 0.950±0.014 |
| | 2($p$) | 159.84 | 81.5 | 15 878 | 99.30 | 0.950±0.005 |
| | 3($m$, $p$) | 77.818 | 176 | 7 315 | 94.00 | 0.899±0.014 |
| | 4($m$) | 177.57 | 403 | 13 340 | 75.12 | 0.719±0.009 |
| | 5($m$) | 210.97 | 479 | 16 398 | 77.73 | 0.744±0.009 |
| 3 (Cd) | 1 | 88.713 | 45.3 | 6 905 | 77.80 | 0.940±0.015 |
| | 2($p$) | 158.904 | 81.1 | 12 490 | 78.60 | 0.950±0.005 |
| | 3($m$, $p$) | 76.985 | 175 | 5 860 | 76.12 | 0.920±0.016 |
| | 4($m$) | 177.309 | 403 | 11 060 | 62.38 | 0.754±0.011 |
| | 5($m$) | 207.981 | 473 | 13 344 | 64.16 | 0.775±0.011 |
| 4 (Cd) | 2($p$) | 158.94 | 80.2 | 3 192 | 20.09 | 0.950±0.005 |
| | 3($p$) | 161.44 | 81.5 | 3 125 | 19.36 | 0.915±0.024 |
| | 4 | 950.42 | 480 | 16 243 | 17.09 | 0.808±0.017 |
| | 5 | 760.27 | 384 | 12 862 | 16.92 | 0.800±0.016 |

[a] $m$ denotes a mask has been used; $p$ denotes the source has been obtained from the second $^{241}$Am purification.
[b] Source thickness in units of $\mu$g/cm$^2$ of $^{241}$Am.
[c] Source No. 2($p$) has been assigned an optical efficiency 0.950±0.005, and all other efficiencies are obtained in terms of this assignment.

counting. The applicability of Poisson statistics in estimating random fission-track counting errors has already been established.[1]

## III. $^{241}$Am SOURCE PURITY

A principal source of systematic error in the measurement of long spontaneous-fission half-lives is the presence of contaminants of much shorter half-life. The $^{241}$Am used in these experiments was prepared by slow neutron capture in $^{240}$Pu and was obtained as pure $^{241}$Am.[5] Consequently, impurities of plutonium or curium are of great concern.

Mass spectroscopic analysis of this $^{241}$Am source revealed no evidence of either plutonium or curium isotopes within the detection sensitivity of the spectroscopy system. These detection limits are one part in $10^7$ and one part in $10^6$ for plutonium and curium isotopes, respectively. Since the expected $^{241}$Am spontaneous-

TABLE II. $^{241}$Am spontaneous-fission track data.

| Source No.[a] | $\mu$g $^{241}$Am | $\mu$[b] | Efficiency | Exposure time[c] | Observed tracks | $T_f$[d] |
|---|---|---|---|---|---|---|
| 2 (Cd, $p$) | 160 | 81.5 | 0.950±0.005 | 79 243 | 367 | 1.08±0.06 |
| 3 (Cd, $m$, $p$) | 77.8 | 176 | 0.907±0.009 | 79 273 | 179 | 1.03±0.08 |
| 4 (Bare) | 965 | 492 | 0.808±0.017 | 8 419 | 179 | 1.21±0.09 |
| 4 (Bare) | 962 | 491 | 0.808±0.017 | 51 506 | 1136 | 1.16±0.04 |
| 4 (Cd, $m$) | 177.6 | 403 | 0.745±0.006 | 50 381 | 213 | 1.03±0.07 |
| 5 (Cd) | 811 | 413 | 0.800±0.016 | 159 758 | 2909 | 1.17±0.03 |
| 5 (Cd) | 777 | 396 | 0.800±0.016 | 51 597 | 943 | 1.12±0.04 |
| 5 (Cd, $m$) | 211.0 | 479 | 0.758±0.006 | 50 370 | 274 | 0.97±0.06 |
| | | | | | Weighted average[e]: | 1.147±0.024 |

[a] $m$ denotes a mask has been used; $p$ denotes the source has been obtained from the second $^{241}$Am purification.
[b] $\mu$g/cm$^2$ of $^{241}$Am.
[c] Minutes.
[d] Units of $10^{14}$ yr.
[e] The source 5 (Cd, $m$) measurement has not been used in the formation of this statistically weighted average, since this result lies more than 2 standard deviations from the mean.

[5] Purchased from Isotope Sales Division, Oak Ridge National Laboratory, Oak Ridge, Tenn.

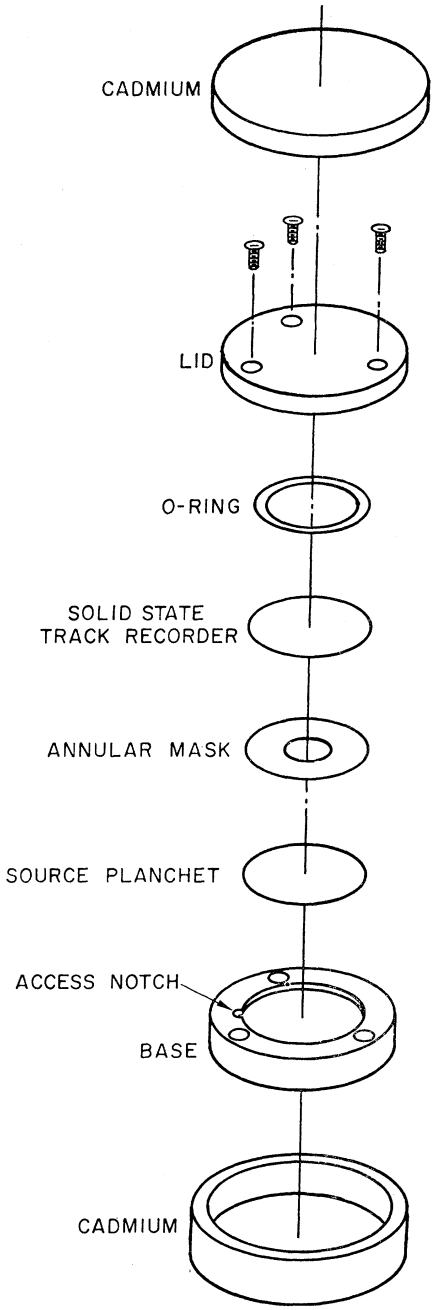


FIG. 1. The cadmium-covered source-holder assembly used for the $^{241}$Am spontaneous-fission half-life measurements. Source-holder assemblies, without cadmium, were also used for bare $^{241}$Am spontaneous-fission half-life exposures.

fission half-life is roughly $10^{14}$ yr, whereas the half-lives of possibly troublesome plutonium isotopes are about $10^{11}$ yr, then no significant systematic error can arise from plutonium impurities. This conclusion did not hold, however, for curium isotopes since the latter have spontaneous-fission half-lives around $10^6$ yr, and therefore curium impurities of as little as one part in $10^8$ could be serious.

An $\alpha$ pulse-height spectrum analysis also failed to reveal the presence of either $^{242}$Cm or $^{244}$Cm. Unfortunately, this method did not possess a sufficiently high sensitivity to rule out troublesome quantities of $^{242}$Cm and $^{244}$Cm. Hence, further precautions against curium contaminants were necessary. To this end, after the first set of $^{241}$Am spontaneous-fission half-life exposures, the $^{241}$Am was chemically repurified by a method due to Ryan and Pringle.[6] In this repurification process, curium and plutonium impurities are reduced by a factor of roughly $10^5$. To test for the effects of these contaminants, further spontaneous-fission half-life exposures were carried out with these repurified sources.

## IV. RESULTS

The $^{241}$Am sources were pressed against selected pre-etched mica SSTR as shown in Fig. 1. Exposures were carried out with and without annular masks as well as with repurified sources (No. 2 and No. 3). Bare and cadmium-covered packages were placed in the same shielded low-level neutron environment used for the measurement of the $^{238}$U spontaneous-fission decay constant.[3] The experimental track data obtained from these exposures are summarized in Table II.

The efficiencies used for sources 3 (Cd, *m*, *p*), 4 (Cd, *m*), and 5 (Cd, *m*) have been obtained from statistically weighted averages[7] of the calibration exposures 1 (bare), 2 (cadmium), and 3 (cadmium) displayed in Table I. The experimental error assigned to each half-life measurement in Table II consists of independent contributions due to efficiency error and track-counting statistics.

Comparison of bare and cadmium-covered exposures in Table II reveals that neutron-induced fission events can be neglected. Table II also discloses good agreement between measurements obtained from both the repurified and original $^{241}$Am. It can, therefore, be concluded that plutonium and curium contaminants were also negligible in the original $^{241}$Am.

The final statistically weighted average of the half-lives listed in Table II is $(1.147\pm0.024)\times10^{14}$ yr, or a corresponding spontaneous-fission decay constant of $(6.04\pm0.13)\times10^{-15}$ yr$^{-1}$.

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[6] V. A. Ryan and J. W. Pringle, Dow Chemical Company, Rocky Flats Plant, R.F.P. Report No. 130, 1960 (unpublished).

[7] In the formation of a statistically weighted average the weights are taken as proportional to the total number of observed tracks.