Spontaneous-Fission Decay Constant of ^{238}U†

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Precise measurement (approximately 1.6% relative error) of the spontaneous-fission decay constant of ^{238}U has been carried out with solid-state track recorders of pre-etched mica. The present experimental result, $\lambda_F=(7.03\pm0.11)\times10^{-17}$ yr^{-1}, is within the quoted experimental error of earlier measurements both by Fleischer and Price, and by Rao and Kuroda. It is in good agreement with the value inferred by Fleischer and Price from a comparison of different dating methods.

I. INTRODUCTION

THE first significant measurement of the spontaneous fission decay constant of ^{238}U was made by Perfilov[1] in 1947, who obtained the value $\lambda_F=(5.3\pm0.9)\times10^{-17}$ yr^{-1}. In 1952, Segrè[2] reported a value of $\lambda_F=(8.7\pm0.3)\times10^{-17}$ yr^{-1}. Using solid-state track recorders (SSTR) of synthetic mica, Fleischer and Price[3] obtained the value of $\lambda_F=(6.6\pm0.8)\times10^{-17}$ yr^{-1}. In addition, by requiring that the ages of a number of minerals measured by counting "fossil" fission fragments agree with ages determined by the decay of ^{40}K and ^{87}Rb, a value of $\lambda_F=6.9\times10^{-17}$ yr^{-1} was inferred. The weighted average of these two results was reported as $\lambda_F=(6.85\pm0.20)\times10^{-17}$ yr^{-1}. Rao and Kuroda[4] have employed a radiochemical method to obtain the value $\lambda_F=(7.8\pm0.9)\times10^{-17}$ yr^{-1}. These measurements are summarized in Table I.

In view of the general geophysical significance of the spontaneous fission of ^{238}U, precise measurement of this decay constant is highly desirable. The present experiment has been carried out to meet this goal. A preliminary account of the method employed here has already been reported.[5] This method is similar to that of Fleischer and Price[3] in that SSTR are utilized. Carefully selected samples of natural mica have been used in the present experiment. Absolute calibration of these detectors has been performed, independent of any knowledge of neutron flux or neutron cross section. A detailed description of this experimental method has been published.[6] A preliminary value of λ_F for ^{238}U so obtained has already been reported.[7]

TABLE I. Measurements of the spontaneous fission decay constant of ^{238}U.

Investigators	Year	Method	λ_F[a]
Perfilov[b]	1947	Fission chamber	5.3 ±0.9
Segrè[c]	1952	Fission chamber	8.7 ±0.3
Fleischer and Price[d]	1964	SSTR	6.6 ±0.8
		^{40}K and ^{87}Rb dating	6.9
		Weighted average:	6.85±0.20
Rao and Kuroda[e]	1966	Radiochemical	7.8 ±0.9

[a] Units of 10^{-17} yr^{-1}.
[b] Reference 1.
[c] Reference 2.
[d] Reference 3.
[e] Reference 4.

II. EXPERIMENTAL MEASUREMENT

Samples of clean muscovite mica were obtained (from Brazil) and cleaved to approximately 0.1 mm. The freshly cleaved mica was then etched in 49% HF at room temperature for about 7 h. The resulting "fossil" fission-fragment tracks, which arise due to uranium impurities in the mica, can be easily identified under a microscope. An example of a pre-etched track, as observed under suitable microscope magnification, is displayed in Fig. 1. In the mica samples selected, the fraction of surface area affected was negligible since the occurrence of pre-etched tracks was rare.

The choice of natural muscovite mica as the SSTR was not arbitrary, since a number of distinct advantages accrue from the use of mica in the present application. Tracks produced by fission fragments that lose most of their energy in the source are more clearly distinguished from surface imperfections in mica than SSTR of the polycarbonate resin variety. The small "diamond" shaped tracks in high quality mica are usually quite different from imperfections. The combination of asymptotically thick sources and polycarbonate resin SSTR can also present difficulties for absolute measurements since the etching process can remove surface from the polycarbonate resin. With mica, however, this danger does not exist since there is evidence that no etching occurs normal to the cleavage planes of the mica.[8] Finally, because of the lower

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[3] R. L. Fleischer and P. B. Price, Phys. Rev. **133**, B63 (1964).
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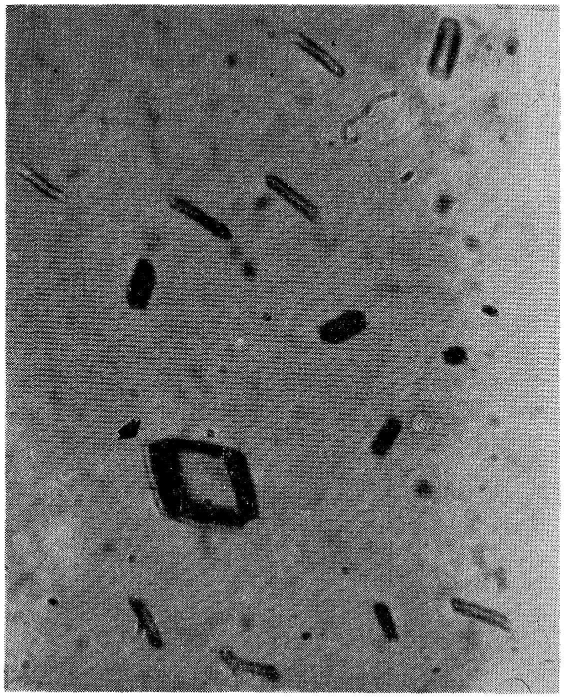

FIG. 1. Photograph of etched fission-fragment tracks in mica as observed under suitable microscope magnification. The pre-etched track (arrow) is clearly distinguished from all other tracks.

radiation damage threshold of the polycarbonate resin, a long-term exposure can produce more surface damage. In the present experiment, the mica surface was completely unaffected by a 4-month exposure.

Uranium foil, 0.127-mm thick and depleted to 0.2 wt.% in ^{235}U, was used. Depleted uranium was employed in order to reduce cosmic-ray neutron-induced fission in ^{235}U that might otherwise contribute an undesirable background. Two packages, one bare and one cadmium-covered, were prepared for exposure. As can be seen in Fig. 2, each package contained six sheets of pre-etched mica sandwiched between seven depleted uranium foils. Each of these packages was firmly pressed together to ensure complete surface area contact. Mica surface areas of 181.94 cm^2 and 182.29 cm^2 were exposed to the depleted uranium in the bare- and cadmium-covered packages, respectively. An additional 3×5-cm sheet of mica, unexposed to uranium, served as a background sample. It was placed in close proximity to these two packages during the exposure.

Both packages were exposed for 2806.18 h (approximately four months) in a low-level counting pit where the neutron background would be low. After exposure, the mica was etched at room temperature in 49% HF for 85 min. As has been demonstrated in Fig. 1, the "new" fission-fragment tracks are clearly distinguishable from the fossil tracks of the preetching process.[9]

[9] M. DeBeauvais, M. Maurette, J. Mory, and R. M. Walker, Intern. J. Appl. Radiation Isotopes **15**, 289 (1964).

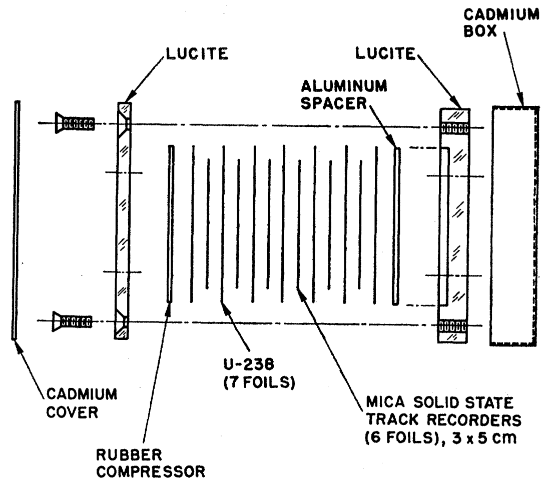

FIG. 2. Geometrical arrangement for ^{238}U spontaneous fission half-life experiment.

III. RESULTS

The observed track data is summarized in Table II. Regions of exposed mica having clean surface area and relatively free of imperfections were scanned. Since the average track density for the bare package is not higher than that of the cadmium-covered package, background from cosmic-ray neutron-induced fission in ^{235}U, can be considered negligible. It has already been shown that the additional contribution to fission in uranium induced by other cosmic-ray components is negligible.[10] A total of 12 498 tracks were counted in an area of 50.249 cm^2, yielding a track density of 248.7±2.2 tracks/cm^2. The error quoted is that due to the random nature of track

TABLE II. Track data observed in ^{238}U-exposed mica.

Observer	Source	Side	Tracks counted	Area scanned (cm^2)	Tracks/cm^2
			Bare		
A	1	1	1120	4.471	250.5± 7.5
C	1	2	484	1.870	258.8±11.8
A	2	1	890	3.846	231.4± 7.8
A	3	1	992	4.134	240.0± 7.5
B	4	1	1420	5.649	251.4± 6.7
				Average:	245.7± 3.5
			Cadmium-Covered		
A	1	1	1035	4.086	253.3± 7.8
B	3	1	715	3.003	238.1± 8.8
B	3	2	1642	6.553	250.6± 6.2
A	4	1	2625	10.383	252.8± 4.9
A	5	1	1575	6.254	251.9± 6.4
				Average:	250.7± 2.9
			Background Sample		
A	...	...	26	15.0	1.7± 0.3
			Summary		
Total exposure time 0.3201 yr					
Average for all observed tracks, after background correction					247.0±2.2/cm^2

[10] P. B. Price and R. M. Walker, J. Geophys. Rev. **68**, 4847 (1963).

TABLE III. Asymptotic sensitivity measurements for mica.

Trial type	T_∞ tracks/cm²	$T(\mu)/\mu$ tracks/μg	$\mu[T_\infty/T(\mu)]$[a] μg/cm²	$s_\infty=\eta\mu\times[T_\infty/T(\mu)]$[b] μg ²³⁸U/cm²
Natural				
1	64 812	1853	34.97±0.87	4311±108
2	66 324	1870	35.47±0.89	4373±110
3	68 370	1955	34.97±0.87	4311±108
4	67 999	1886	36.05±0.90	4444±111
5	66 531	1818	36.60±0.92	4511±113
6	67 106	1894	35.43±0.89	4368±109
7	60 837	1766	34.45±0.76	4247± 94
8	63 040	1853	34.02±0.71	4194± 88
9	66 062	1890	34.95±0.73	4309± 91

[a] Quoted error is that of counting statistics only.
[b] An optical efficiency for mica of $\eta=0.948$ has been used.

counting and is obtained from the customary rules of Poisson statistics. The applicability of Poisson statistics has been verified for objective track-counting observations.[6]

A 15-cm² scan of the background sample revealed 26 events that appeared like fission-fragment tracks, corresponding to a background track density of 1.7 tracks/cm². When this background correction is made, the fission-track density in the exposed mica is 247.0±2.2 tracks/cm².

The absolute sensitivity for asymptotically thick depleted uranium foils, s_∞, has been measured with a relative error of 1.4%.[6] For SSTR of mica, $s_\infty=4.341\pm0.061$ mg/cm², corresponding to $(1.098\pm0.015)\times10^{19}$ atoms/cm². This asymptotic sensitivity has been obtained from epi-cadmium neutron-induced fission experiments in ^{235}U. A brief summary of these measurements can be found in the Appendix. According to the results reported by Rao and Kuroda,[4] the fission yield curve for spontaneous fission in ^{238}U is similar to that of slow neutron-induced fission in ^{235}U. In addition, mean fission-fragment ranges were calculated for various nuclides using the fission-product yield data of Katcoff[11] and the measured ranges of fission fragments in uranium obtained by Niday.[12] These calculated mean range differences were quite small, implying that the asymptotic-sensitivity calibration is unaffected within the present limits of experimental error.

Employing the experimental data of Table II, one has 771.6±6.9 fissions/yr cm² and therefore,

$$\lambda_F=(7.03\pm0.11)\times10^{-17}\ \mathrm{yr}^{-1} \tag{1}$$

for the spontaneous fission decay constant of ^{238}U, with a relative error of 1.6%. As can be seen from Table I, this value is in good agreement with that obtained by Fleischer and Price.[3]

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[12] J. B. Niday, Phys. Rev. 121, 1471 (1961).

APPENDIX: ASYMPTOTIC SENSITIVITY MEASUREMENTS

The effective mass per unit area from which all fission fragments are observed is denoted by s_∞, the asymptotic sensitivity. A detailed discussion of the measurement of s_∞ can be found elsewhere.[6] Because of the importance of this quantity in this experiment, these measurements are also summarized below.

Asymptotically thick ^{238}U foils (of the same thickness as used in the spontaneous fission-decay-constant exposure) are placed in good contact with pre-etched 0.1-mm thick mica. On the other side of the mica, a thin uranium source (either UF_4 or U_3O_8) is placed. The uranium used to prepare these thin sources is enriched to about 93% ^{235}U. The isotopic composition is precisely known from mass spectrograph measurements and the thickness μ of each source ($\mu<100$ μg/cm² U) is determined by α counting. Fortunately, the uranium that was employed also contained approximately 0.85 wt.% ^{234}U, which could be used as a tracer. Consequently, the total α activity of the thin uranium sources was increased by about a factor of 25, which permitted the use of a low-geometry counting chamber.

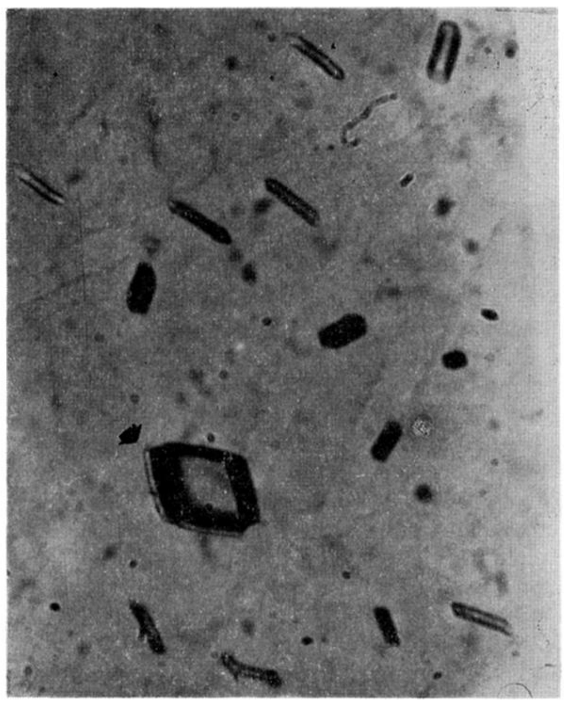
Nine of these mica "sandwiches" were assembled in cadmium covers and placed on the rim of a wheel that could be rotated in the graphite reflector of the Argonne Thermal Source Reactor (ATSR). This geometry ensured exposure to the same time-integrated epi-cadmium neutron flux. The results of this exposure are shown in Table III, where the asymptotic sensitivity is given in the form

$$s_\infty=\eta\mu[T_\infty/T(\mu)].$$

Column 1 contains T_∞, the observed tracks per cm² of the asymptotically thick source, which for this exposure was natural uranium containing 0.71150% ^{235}U. Column 2 contains $T(\mu)/\mu$, the number of tracks per μg of ^{235}U in the thin source, which for this experiment contained 93.185375% ^{235}U. Column 3 gives the ratio of column 1 to column 2.

In addition to these measurements, the optical efficiency η must be known in order to determine s_∞. Using thin sources of ^{244}Cm, the value $\eta=(94.8\pm0.53)\%$ was obtained for pre-etched mica. The fission rate of the thin ^{244}Cm sources was accurately measured by both α-particle counting and direct fission-fragment counting. For a more detailed description of these calibrations see Ref. 6.

The last column in Table III gives the asymptotic sensitivity obtained for each of the nine mica "sandwiches." The final result obtained for s_∞ is 4341±61 μg ^{238}U/cm² or $(1.098\pm0.015)\times10^{19}$ ^{238}U atoms/cm².



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