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

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(Received 23 August 1963)

Uranium impurities in minerals undergo spontaneous fission over geologic times, leaving radiation-damage trails whose number can be related to the mineral age, provided λ_F, the spontaneous-fission decay constant of U^{238}, is known. By requiring that the ages of a large number of minerals measured in this way agree with ages determined by decay of K^{40} and Rb^{87}, a value $\lambda_F=6.9\times10^{-17}$ yr^{-1} was deduced. By placing a sheet of natural uranium next to a sheet of mica for 6 months and then counting tracks entering the mica from the uranium, a value $\lambda_F=6.6\times10^{-17}$ yr^{-1}, in good agreement with the above value, was obtained. A weighted average of these two results gives the value $\lambda_F=(6.85\pm0.20)\times10^{-17}$ yr^{-1}.

THE necessity for making an accurate determination of the decay constant for spontaneous fission of U^{238} arose as a result of the recent discovery that various types of mica,[1,2] natural glasses,[3] and other minerals[4] contain trails of radiation-damaged material produced by fragments of uranium atoms which have spontaneously fissioned. By chemical etching, these "fossil tracks" can be enlarged to a convenient size for viewing in an optical microscope.[5,6] In mica the track density ρ_s per cm^2 of mineral surface is related to the time T, since solidification of the rock follows the relation

$$\rho_s=[\exp(\lambda_D T)-1]\lambda_F NRC_{238}/\lambda_D, \quad (1)$$

where λ_D and λ_F are the total decay constant and spontaneous fission decay constant for U^{238}, N is the number of atoms per cm^3, C_{238} is the fraction of these atoms that are U^{238}, and R is an effective etchable range of fission fragments in the mineral. Provided T is less than $\sim10^9$ yr, (1) reduces to

$$\rho_s\simeq\lambda_F TNRC_{238}. \quad (2)$$

The necessity for determining N, R, and C_{238} can be avoided by exposing the mineral to a dose n of thermal neutrons and then measuring the density ρ_I of new tracks resulting from thermal fission of U^{235},

$$\rho_I=n\sigma NRC_{235}, \quad (3)$$

where σ is the cross section for thermal fission of U^{235}. The mineral age is then simply

$$T=(\rho_s/\rho_I)n\sigma I/\lambda_F, \quad (4)$$

with $I=C_{235}/C_{238}$.

In recent studies of mineral ages by this technique,[3,7,8] all of the parameters except λ_F were either known or measured to within 10% accuracy. Previously measured values of λ_F[9–13], which appear to be most reliable, range from $\sim1.2\times10^{-16}$ to $\sim5.3\times10^{-17}$ yr^{-1}. In order to make mineral age determinations on an *absolute* basis by the fission track method, λ_F must also be known to at least 10% accuracy.

In Fig. 1, which uses data, some of which is from our recent paper,[3] we have compared our fission track ages of a group of micas[7] and natural glasses[3] (tektites) with age determinations based on the radioactive decay of K^{40} or Rb^{87}, whose decay constants are accurately known.[14] The fission track ages were calculated assuming $\lambda_F=8.27\times10^{-17}$ yr^{-1}. Taking the straight line drawn through the points as the best fit and requiring that the ages obtained by the two methods be concordant, we deduce a value

$$\lambda_F=(6.9\pm0.2)\times10^{-17}\text{ yr}^{-1}. \quad (5)$$

We now describe an independent experimental determination of λ_F which confirms the above value and permits us to use the age equation (4) on an absolute basis. The method is analogous to the fission-track method of dating minerals.

A sheet of synthetic mica, initially containing no tracks because of its young age, was placed next to a sheet of natural uranium and used to count spontaneous and induced fission fragments emerging from the uranium. The mica–uranium sandwich was cut in two. One-half was stored for six months to collect spontaneous fission events; the other half was irradiated in the

† This research was sponsored in part by Air Force Cambridge Research Laboratories, U. S. Office of Aerospace Research.
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[14] Two points at the top of the graph show too low a fission-track age. Such a discrepancy for old minerals has been shown[3] to result from track fading due to high temperatures, which are more likely to have occurred in very old samples.

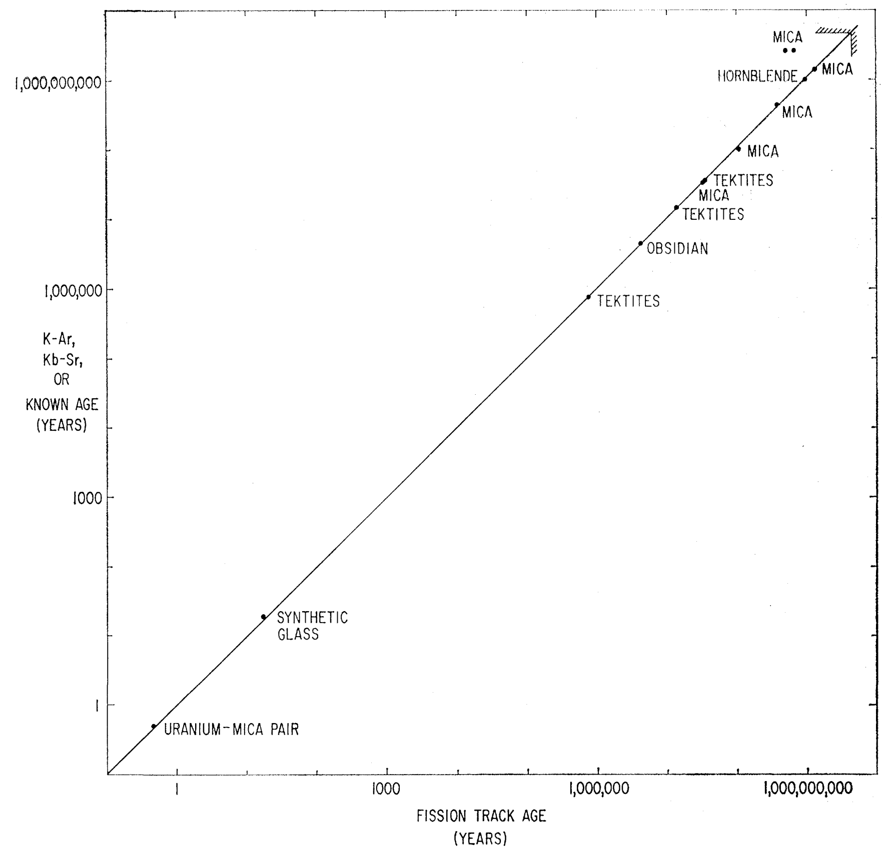

FIG. 1. The graph, which uses data from Ref. 3, unpublished data, and the new experiment reported here, shows that the ages of a large number of micas and glasses, as measured by counting spontaneous-fission tracks, bear a constant relationship to the ages determined by decay of K^{40} and Rb^{87} in the minerals. By drawing a straight line through the points on the log-log plot and equating the ages, a value $\lambda_F = 6.9 \times 10^{-17}$ yr^{-1} for the spontaneous-fission decay constant was deduced.

thermal column of the Brookhaven reactor to a dose of 1.04×10^{11} neutrons/cm^2 to collect induced fission events.

Equation (4) was used to calculate λ_F. The measured values of ρ_s and ρ_I were $(210 \pm 21)/\text{cm}^2$ and $(2.73 \pm 0.19) \times 10^6/\text{cm}^2$, respectively. The exposure time for the spontaneous-fission tracks was 0.51 yr and the best values of σ and I were taken to be $\sigma = (5.82 \pm 0.04) \times 10^{-22}$ cm^2 and $1/I = 137.8$.[15,16] Substitution into (4) gave

$$\lambda_F = (6.6 \pm 0.8) \times 10^{-17} \text{ yr}^{-1}, \qquad (6)$$

where the indicated standard deviation pertains to counting statistics. This value thus agrees with (5) within the counting errors.

Of the two values (5) and (6), the former, $\lambda_F = 6.9 \times 10^{-17}$ yr^{-1}, is probably more reliable since it is based on a comparison of a large number of experimental measurements of mineral ages ranging from 6×10^5 up to 1.4×10^9 yr (Fig. 1). The only significant sources of error are the decay constants for K^{40} and Rb^{87}, which are known to within 5%,[17] and the thermal neutron dose, which was measured to within ~5% by counting the Ba^{140} and Mo^{99} activity in a U foil. A weighted average of the two results gives $\lambda_F = (6.85 \pm 0.20) \times 10^{-17}$ yr^{-1}.

The main advantages of the fission-track method over previous methods involving electronic counting, emulsion counting, or radiochemical analysis are its simplicity and its ability to discriminate completely against a background of light particles such as cosmic rays and alpha particles.[18]

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