Reports

Fission-Track Ages and Track-Annealing Behavior of Some Micas

Abstract. *The density of tracks from spontaneously fissioning uranium atoms in mica has been used to make quantitative measurements of geologic ages. In comparison with methods in which ages are derived from measurements of radioactive decay products, the fission track method is much simpler and is usually accurate for micas up to a few hundred million years old. Annealing experiments suggest that high ambient temperatures are responsible for track fading in the older samples.*

When heavy particles such as uranium fission fragments traverse certain minerals they leave trails of radiation-damaged material which can either be viewed directly in the electron microscope (*1*) or can be enlarged to optically visible size by preferential chemical etching (*2*). Reasoning that such tracks should have been produced in old minerals containing fissionable impurities, Price and Walker (*3*) obtained mica samples containing uranium inclusions and discovered "fossil" tracks emanating from the vicinity of the inclusions. Using a similar etching technique, Fleischer and Price (*4*) found fossil tracks in natural glasses, thus demonstrating that the phenomenon is a general one and not restricted to crystalline materials.

Price and Walker (*5*) observed fossil tracks in a wide variety of micas of different ages and examined in detail the possible ways in which tracks may be formed by natural radiation processes. They concluded that, with the exception of those samples which have been exposed to cosmic rays for a long time near the surface of the earth, almost all of the fossil tracks in terrestrial minerals are caused by the spontaneous fission of uranium impurities. They pointed out that the number of these "background tracks" is a measure of the age of the mineral, provided no annealing of tracks or diffusion of uranium has occurred. They described a simple experimental procedure by which the age can be measured and made preliminary measurements on two samples, which suggest that fossil tracks remain stored in mica for at least 10^8 years.

The possibility of making accurate measurements of mineral ages by the method outlined by Price and Walker has been examined and is treated here and in two companion reports. (i) In this report we give age measurements of a number of micas, some of which have been independently dated by radioactive decay techniques, and we describe track-annealing studies on muscovite and lepidolite. (ii) Maurette, Pellas, and Walker (*6*) have measured the fossil track ages of another group of micas and have studied the effect of high temperature annealing on tracks in phlogopite mica. (iii) Fleischer and Price (*7*) have measured the fossil track ages of tektites and other natural glasses and have done annealing experiments on tracks in glasses.

The experimental details of the dating method and a derivation of the age equation are given elsewhere (*5*). The number ρ_b of background or fossil tracks intersecting 1 cm^2 of surface is a function of the time T since cooling to a temperature at which tracks are retained, of the total decay constant λ_D of U^{238}, of the decay constant λ_F for spontaneous fission of U^{238}, and of the uranium concentration C_U. The amount of uranium in most mica crystals is very small (10^{-8} to 10^{-12} atom fraction) and is often distributed very nonuniformly on a microscopic scale (*5*). The concentration of uranium in the same region where background tracks were counted can be determined very accurately and easily by exposing the sample to a thermal neutron flux in a reactor and counting the number, ρ_n, of new tracks resulting from induced fission of U^{235} atoms in the region of interest.

The ratio of old to new tracks is a function of only one unknown quantity, the age, and any uncertainties pertaining to fission-fragment ranges or etching efficiency are thus eliminated. The exact equation is

$$\rho_b/\Delta\rho_n = [\exp(\lambda_D T) - 1]\,(\lambda_f/\lambda_D n\sigma I) \quad (1)$$

where I is the ratio of the abundances of U^{235} and U^{238} and n and σ are the thermal neutron dose and the cross-section for thermal fission of U^{235}.

To ensure that only U^{235} fissions are counted, a control sample is wrapped in heavy cadmium foil which absorbs thermal neutrons only. The difference in track densities in the cadmium-shielded and unwrapped samples is $\Delta\rho_n$. This extra step may be eliminated if the fast neutron flux is known to be a negligible fraction of the total flux.

For samples less than 10^9 years old, the equation simplifies to

$$T \simeq (n\sigma I/\lambda_F)\,(\rho_b/\rho_n). \quad (2)$$

The evaluation of the parameters in Eqs. 1 and 2 is discussed in detail elsewhere (*7*). We used the values $\lambda_D = 1.54 \times 10^{-10}$ yr^{-1}, $\lambda_F = 6.9 \times 10^{-17}$ yr^{-1}, and $I = 7.26 \times 10^{-3}$. The product $n\sigma$, which is the fraction of U^{235} that has undergone fission, cannot be monitored very accurately in a typical reactor irradiation, and it was therefore determined experimentally to within 10 percent by counting the radioactivity of a weighed uranium foil which is included with the mica samples. This can be performed inexpensively by commercial firms, so that the

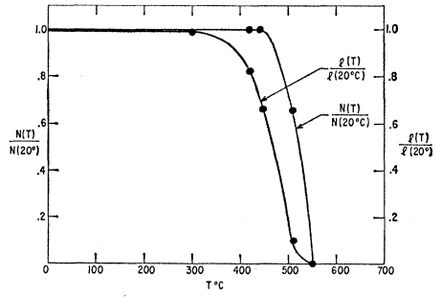

Fig. 1. Effect of annealing for 1 hour at a temperature T on the average length, l, and density, N, of etched tracks in muscovite from Renfrew County, Canada. In the graph, l and N are normalized to the average length and density measured with no heat treatment.

Table 1. Comparison of ages of micas determined by the fission-track method and by radioactive decay method.

Location	Type	Age ($\times 10^6$ yr)		Uranium concentration [$\times 10^{-10}$ (atom/atom)]
		Fossil track method	Radioactive decay method	
Oregon	Muscovite	108 ± 27	95 to 100 (*10*)	5
Arizona	Muscovite	33 ± 7	32.7 ± 1.7 (*11*)	30
Baltimore, Md. Pegmatite, Clipper Mill Rd. and Ash Ave.	Muscovite	380 ± 80	400 to 450 (*12*)	~30*
Brown Derby, Pegmatite, Colo.	Lepidolite	1390 ± 250	1400 (*13*)	~2*
Pope's Claim, Southern Rhodesia; 17 km east of Salisbury on Chishawasha Farm	Lepidolite	670 ± 100	2600 to 2700 (*12*)	3
Salisbury, Southern Rhodesia (same as above)	Lepidolite	500 ± 100	2600 to 2700 (*12*)	7

* Uranium distribution nonuniform.

only parameters left to evaluate are ρ_b and ρ_n.

The results of measurements of ages and uranium concentrations of mica samples of various types, ranging in age from a few million to several billion years, are summarized in Tables 1 and 2. Only samples which have been dated independently by K^{40}-Ar^{40} or Rb^{87}-Sr^{87} decay methods, or both, are listed in Table 1. The principal errors are the accuracy of the n determination ($\pm$ 10 percent) and statistical errors due to a limited number of track counts. All but the Oregon sample were irradiated for the same time, so that errors in their relative ages are due only to counting statistics.

Our measured ages and uranium concentrations of micas which have not themselves been dated by radioactive decay methods are given in Table 2. The unreferenced samples were taken from museums and their locations are subject to some uncertainty. The ages calculated by the fission-track method and by radioactive decay methods appear to be in satisfactory agreement for the first four samples in Table 1, whereas the ages of the two African lepidolites are much too low according to the fission-track method. In Table 2, all the samples believed to be older than 500 million years gave fission-track ages which erred on the young side.

In the absence of information obtained by radioactive decay methods, it seems reasonable to suspect that the samples which gave low ages in Table 2 have been at sufficiently high temperatures for a long enough time to allow track fading to have taken place. Annealing experiments were therefore conducted to study the relation between temperature and track stability.

In previous electron microscope studies of artificially produced, unetched fission-fragment tracks in mica, it was found that the fine trails of radiation-damaged material were very resistant to high-temperature annealing and did not disappear completely until temperatures of 800° to 900°C were reached (*1*, *8*). At somewhat lower temperatures the trails appeared to be intermittent, suggesting that the damage was annealing out at different rates at various points along the tracks. Since the optical-detection technique employed in age determinations requires that the damaged trails be penetrated by an etchant, it is important to know what effect annealing at a high temperature will have on the etching of tracks.

We have performed annealing experiments on a muscovite from Renfrew County, Canada, and on a lepidolite from the Brown Derby Mine, Colorado. [Similar annealing experiments have been carried out on phlogopite by Maurette *et al.* (*6*).]

Large, thick, single-crystal samples of the Renfrew muscovite, in which the uranium concentration was known to be distributed rather uniformly, were irradiated in a reactor to produce a known density of "fresh" tracks. A large number of pieces were cut from each sample and annealed for 1 hour at temperatures ranging from 100°C up to 650°C. Each piece was then cleaved so as to expose a fresh surface, etched, and the number and distribution of track lengths intersecting the fresh surface were measured. The results are shown in Fig. 1.

Tracks appear to shorten before their number per unit area decreases. This result implies that short portions of a damaged fission-fragment trail "heal up" and prevent the acid from etching out the entire length. If these portions which heal up first represent a small fraction of the total track length, then the probability of cleaving through such repaired portions is small and the density of etched tracks will not be altered appreciably. The previous electron microscope observations of intermittent damaged regions in annealed specimens

Table 2. Ages of some previously undated micas determined by the fission-track method.

Location	Type	Age ($\times 10^6$ yr)		Uranium concentration [$\times 10^{-10}$ (atom/atom)]
		Fossil track method	Estimated age of region	
Gossan-Lead District, Va.	Muscovite	350 ± 60	350 to 450 (*14*)	50
New York City	Muscovite	400 ± 85	370 to 400	6
Bancroft, Ontario	Biotite	355 ± 40	1000 (*15*)	70
Renfrew County, Canada	Muscovite	360 ± 50	1000	8
Edenville, N.Y.	Phlogopite	300 ± 30	1300 (*16*)	~40*
Edenville, N.Y.	Biotite	240 ± 30	300 to 500	50
Natural Bridge, N.Y.	Biotite	720 ± 160	1150	~60*
Natural Bridge, N.Y.	Phlogopite	140 ± 15	1150	~80*
Natural Bridge, N.Y.	Muscovite	190 ± 20	1150	~10*
Somerville, N.Y.	Phlogopite	550 ± 80	1150	~14*
Chester County, Pa.	Muscovite	310 ± 28	?	16
Buckingham, Quebec	Muscovite	600 ± 85	?	3
Batchellorville, N.Y.	Muscovite	185 ± 17	?	20
Sacandaga, N.Y.	Muscovite	175 ± 80	1000	~25

* Uranium distribution nonuniform.

support this description. Incompletely etched tracks can also be seen optically in thin, annealed sheets of mica. Acid proceeds from both surfaces but stops when it meets a healed-up portion of track, leaving a constriction.

From the curves in Fig. 1 we deduce that either or both the number and length of healed-up regions increases with increased annealing temperature and probably also with increased time.

In order to date samples accurately, it is necessary for the track densities not to have been lowered by past thermal treatments. These results show that densities are not affected by temperatures of 450°C or less for times of up to 1 hour. The stability of tracks for longer times is not yet known but we may estimate it by comparison with data on tracks in another material. It was found (*7*) that tracks in tektites were stable for 1 hour at temperatures below 500°C (which is comparable to our observation in muscovite of a decrease in number between 450°C and 510°C). If the tracks in mica anneal out under the same conditions, then the extrapolated tektite results would apply to muscovite also. We then calculate that tracks would be stable over the entire age of the earth at approximately 145°C, but would anneal out in only a million years at about 200°C.

The annealing data on lepidolite are less extensive than on muscovite because of the problem of long-lived radioactivity induced in a reactor irradiation. We have found that tracks begin to shorten after a 1-hour anneal at temperatures as low as 200°C. No data are available on track densities.

Let us now compare the relative effects of high temperatures on the reliability of dates derived from K-Ar measurements and those from fission track dating. In K-Ar dating, the measurements cease to be reliable if argon is able to escape. A reasonable value for the activation energy for argon diffusion appears to be $E_{Ar} \approx 1.1$ to 1.2 ev (*9*), and in typical samples the diffusion distance is of the order of millimeters. In contrast, the annealing out of fission tracks occurs at activation energies (E_t) of 2.0 to 2.5 ev in cases so far examined (*6*, *7*) but requires atomic motion of only a few atom spacings. These numbers imply that above some critical temperature, which is a function of $E_{Ar} - E_t$ and the ratio of the two diffusion distances involved, fission tracks will disappear more rapidly than will argon. This critical temperature is estimated to be 100°C when $E_t = 2.5$ ev.

In conclusion, fission-track dating provides a simple and usually accurate method of geochronology for micas less than a few hundred million years old. Although for older micas correct results are sometimes found, track fading appears to be common and may be due to a high ambient temperature over geological times or to a transient period at higher temperature. The extent of this fading may become a useful indicator of thermal history.

The most probable means of extending the technique of fission-track dating to older samples appears to require finding tracks in minerals with higher activation energies for track fading. Recent experiments show that fossil tracks in hornblende are more resistant to annealing than tracks in mica. Other minerals also have been found to contain fossil tracks and annealing and dating studies are in progress.

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Geomagnetic Polarity Epochs

In June of 1963 we published paleomagnetic results from six radiometrically dated volcanic flows from the Sierra Nevada of California (*1*) and 4 months later two additional studies appeared almost simultaneously, one by us on more material from the Sierra Nevada (*2*) and one by McDougall and Tarling (*3*) on volcanic rocks from the Hawaiian Islands. All of the data from these three studies are consistent with the geomagnetic field's having had normal polarity back to about 1 million years (eight radiometric dates from normal flows) and having had reversed polarity between 1 and 1.8 million years (19 radiometric dates from reversed flows). In rocks older than this several discrepancies appear between the results from California and those from Hawaii. In an attempt to account for these discrepancies, we have reexamined all of our data and have found no reason to distrust any of our paleomagnetic or radiometric results, but we have discovered errors in our geologic correlations which remove two of the discrepancies.

The radiometric dates of the rocks in our studies were all obtained in the geochronometry laboratory of Curtis and Evernden at the University of California. Curtis, Evernden, James, Lipson, and Savage (*4*) obtained the dates for our data points *S*1, *S*3, and *S*4, and the others were determined by Dalrymple (*5*). The dates were obtained for reasons other than their paleomagnetic interest, and collections of oriented samples for paleomagnetic study were made subsequently on the basis of published locality descriptions and unpublished field notes. The strength of this approach is that the petrographic criteria used to determine whether samples are suitable for dating were based solely on internal stratigraphic consistency and consistency with mammalian paleontology. These criteria were developed prior to and independently of the paleomagnetic studies. Because of this, the excellent consistency between the paleomagnetic and radiometric data back to 1.8 million years constitutes a strong, independent verification of the accuracy of the techniques developed at Berkeley for the dating of young volcanic rocks.

The weakness of working independently on previously dated material lies in the possibility of errors in geologic correlation. In reviewing our 16 data