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Etching and annealing studies of fission tracks in chlorite and their applications in dating

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Abstract

Etching characteristics of fission fragment tracks in chlorite from central Sierra Nevada Batholith, California, USA, are studied using HF as the track etchant. Systematic annealing experiments are carried out in order to obtain correction factor for the age of chlorite, which has suffered annealing during its geological history. Complete erasure of fission tracks in chlorite occurs for 4 hrs annealing at 600°C. Extrapolation of experimental annealing data suggests that a temperature of 155°C would remove all fission tracks in chlorite in 1 Ma. The annealing correction of 15% determined by the age-plateau method gives the corrected age as 161±30 Ma for this mineral. Uranium content in chlorite is quite low (~ 0.315 ppm). The mean value of activation energy is 1.6 eV.

Introduction

Micaceous minerals are being widely used in fission track geochronology because they are wide spread in nature. Besides, there is no need of grinding and polishing the specimens and their surfaces can be etched easily. Though a lot of work has been reported on muscovite, phlogopite and biotite (Lakatos and Miller, 1973; Kere, 1966; Nagpaul *et al.*, 1974; Singh and Virk, 1978), a very little data (Sharma *et al.*, 1977) are available with chlorite as the track detector. In the present investigations the etching and annealing studies are carried out in chlorite mineral from centrral Sierra Nevada Batholith, California, USA. The fission track (f.t.) age is determined for this mineral. A correction is applied for fading of tracks due to geothermal events during the geological history of sample. The correction obtained by age-plateau method (Storzer *et al.*, 1973) is rechecked by the method of calibration curve translating track length reduction into track density reduction (Storzer and Wagner 1969).

Experimental Procedure

1. Standardization of etching conditions in chlorite.

The etching conditions in chlorite were studied by using the samples irradiated with fission fragments from ^{252}Cf source. The samples were treated with 40% HF for

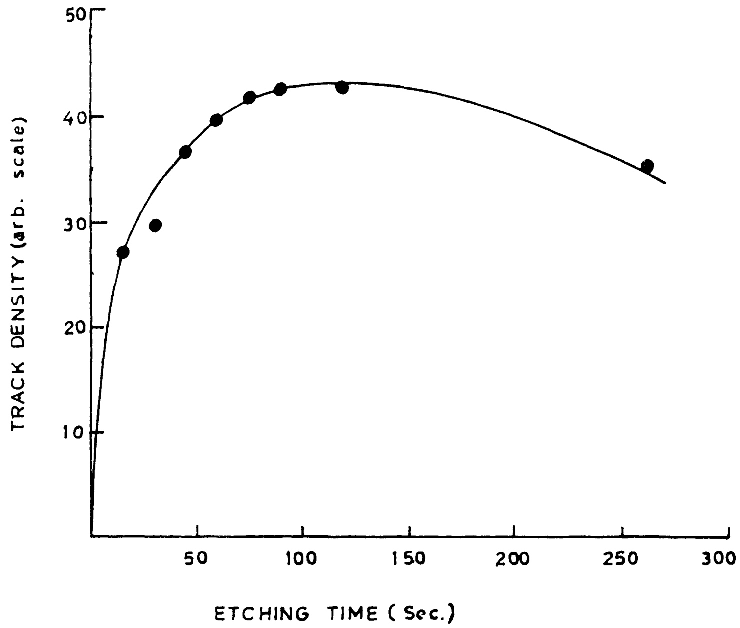

FIG. 1. The variation of number of tracks per graticule with etching time.

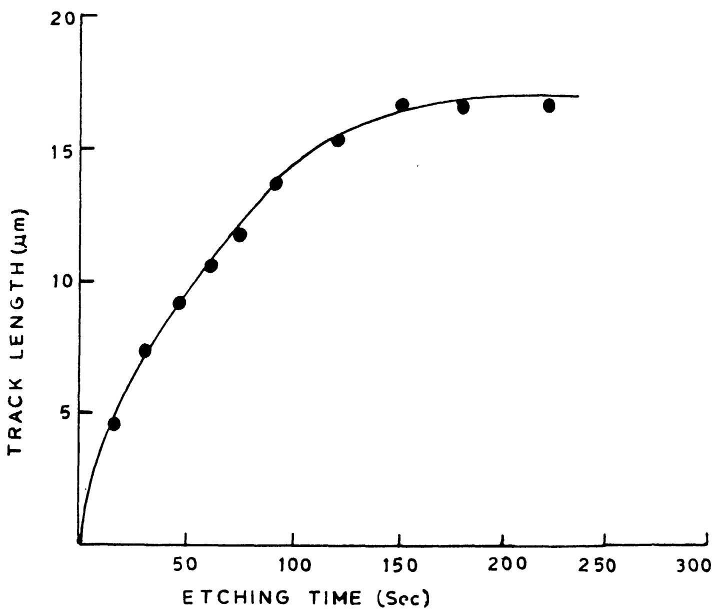

FIG. 2. The variation of track length with etching time.

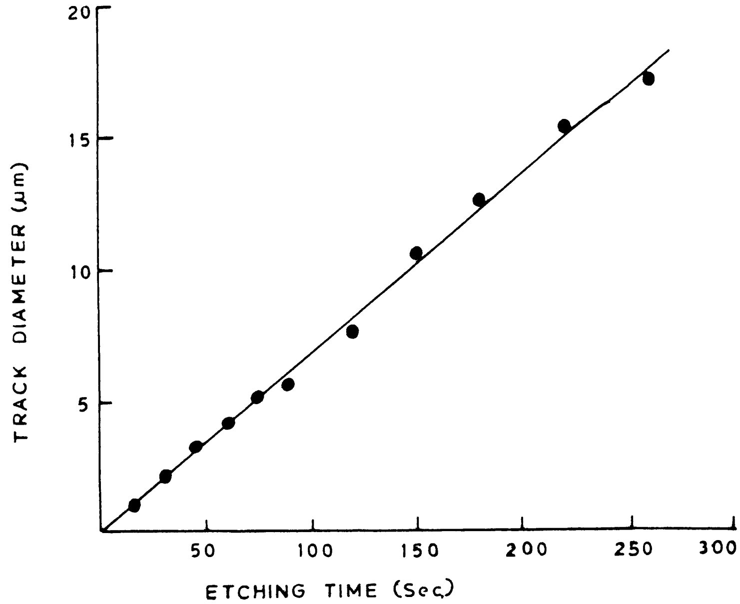

FIG. 3. The variation of track diameter with etching time.

different durations ranging from 15 to 360 s at 40°C. The track density, projected track length and track diameter were measured for different time intervals. The variation of track density, track length and track diameter with etching time is shown in Figs. 1, 2 and 3, respectively. The track etch rate, V_T, was calculated from the slope of the linear part of the curve, etching time versus track length (Fig. 2). The bulk etch rate, V_G, was calculated from the one half of the slope of the curve, etching time versus track diameter (Fig. 3).

The etching efficiency (η) was calculated from the relation (Fleischer *et al.*, 1975, Singh *et al.*, 1984):

$$\eta = 1 - \frac{V_G}{V_T} \tag{1}$$

From Fig. (1), it is observed that the optimum etch time required to reveal the maximum tracks for 40% HF, is 100 s at 40°C.

2. Age and Uranium content

The chlorite samples were prepared and etched in 40% HF for 100 s at 40°C for revelaton of fossil fission tracks. The fossil track density was recorded by counting the tracks under Olympus binocular microscope at a magnification 600 X using population method (Gleadow, 1981). A few samples were annealed at a temperature of 600°C for 4 h in order to remove fossil tracks. These samples were then irradiated with a thermal neutron fluence of 10^{16} n cm^{-2} from CIRUS Reactor at Bhabha Atomic Research Centre, Trombay, Bombay. The irradiated samples were etched and counted under as identical conditions as for fossil tracks and the induced track density was recorded. Fission track age was determined by using the age equation (Price and Walker, 1963; Fleischer and Price, 1964; Singh and Virk, 1978):

$$T = \frac{1}{\lambda_D} \ln \left(1 + \frac{\rho_s}{\rho_i} \frac{\lambda_D}{\lambda_F} \phi \sigma I\right) \qquad (2)$$

where,

λ_D =total decay constant of uranium (1.55×10^{-10} a^{-1})
λ_F =spontaneous decay constant of uranium
(7.03×10^{-17} a^{-1}) (Roberts *et al.*, 1968)
ρ_s =fossil track density
ρ_i =induced track density
ϕ =thermal neutron fluence (10^{16} –10^{17} n cm^{-2})
σ =thermal neutron cross-section for fission of ^{235}U
(580×10^{-24} cm^2)
I =Isotopic ratio of ^{235}U to ^{238}U (7.25×10^{-3})

Substituting these values the age equation reduces to

$$T = 6.45 \times 10^9 \ln \left(1 + 9.30 \times 10^{-18} \times \frac{\rho_s}{\rho_i} \times \phi\right) \qquad (3)$$

The uranium concentration of the mineral was calculated by using the relation (Price and Walker, 1963; Nagpaul, 1974; Virk and Koul, 1977):

$$C_W = \frac{2m\rho_i}{\sigma R \phi I N d} \qquad (4)$$

where

m = atomic weight of ^{238}U,
N = Avogadro's number (6.02×10^{23} mol^{-1}),
d = density of material (3.0 μg m^{-3})
R = mean range of induced fission tracks (14.5 μm).

The f.t. age and uranium concentration of chlorite samples calculated by equation (3) and (4) are summarized in Table 1. The mean observed fission track age and the uranium content in chlorite are found to be 138±17 Ma and 0.315 ppm, respectively.

TABLE 1. Fission track age and uranium concentration in chlorite. Thermal neutron fluence (ϕ) = 0.97 x 10^{16} n cm^{-2}

Laboratory symbol	Fossil track density ρ_s (x10^4)	Induced track density ρ_i (x10^4)	Fission track age T (Ma)	Uranium concentration C_W (ppm)
CCA-I	1.6	6.9	134±26*	0.307
CCA-II	1.7	7.1	137±33	0.316
CCA-III	1.8	7.3	143±29	0.323

* Estimated standard deviation, σ...

3. Correction for thermally affected fission tracks

The annealing correction to the f.t. age of chlorite was determined by following methods.

1) The calibration curve translating track length reduction into track density reduction.
2) The age-plateau method.

3.1 Method of calibration curve translating track length reduction into track density reduction:

In order to find the annealing correction by this method (Fleischer *et al.,* 1975; Singh and Virk, 1977), irradiated samples of known induced fission track density were heated at 400°C, for different time intevals in the range 10 – 70 m. Reductions in track length and track density were determined corresponding to each heating event (Table 2). A linear relation was observed between the track-length reduction and track density

TABLE 2. Percentage reduction in track density versus percentage reduction in track length in chlorite. Temperature = 400°C

Annealing time (minutes)	Percentage reduction in track density	Percentage reduction in track length
10	2.15	13.80
20	18.30	29.70
30	24.60	35.60
40	34.50	43.80
50	44.30	53.90
60	57.00	68.10
70	71.30	80.90

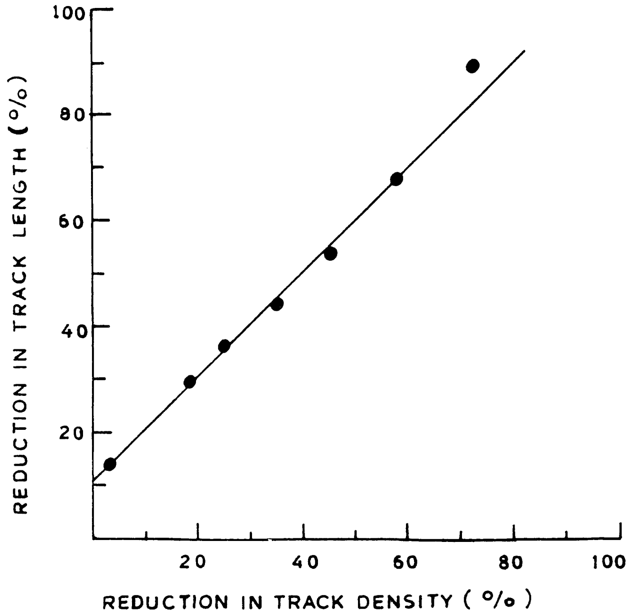

FIG. 4. Fission track length reduction versus fission track density reduction at the temperature of 400°C.

reduction (Fig. 4). The present study reveals that the track density reduction in chlorite lags behind the track-length reduction by 11%. The mean length of fossil tracks in chlorite has been observed to be 10.6 μm and that of induced fission tracks under the identical conditions is found to be 14.5 μm. The mean length of fossil fission tracks is thus less than the mean length of induced fission tracks by 27%. This much reduction corresponds to a 16% decrease in fossil track density. Hence the correction to fission track age of chlorite is 16%.

3.2 Age-plateau method

For estimating annealing correction by age plateau method (Storzer *et al.,* 1973 and Singh *et al.,* 1985), different pairs of chlorite samples were taken. In each pair, one sample contained only fresh fossil tracks which had been thermally affected in the past and the other contained only neutron-induced fission tracks. The pair of samples were heated in a muffle furance at 100, 200, 300, 400, 500 and 600°C for 1 h in each case and the track density reduction was calculated for each pair after etching and scanning of the samples. For the given pair of samples, f.t. age was calculated corresponding to each temperature by using equation (3). The results are given in Table 3. The apparent age increases at low temperature but reaches a plateau at the temper-

TABLE 3. Annealing correction for fission track age by age plateau method. Annealing time = 30 minutes

Temperature (°C)	Fossil track density ρ_s ($\times 10^4$)	Induced track density ρ_i ($\times 10^4$)	Fission track age T (Ma)
Unannealed sample	1.68	7.13	137±28
100	1.68	7.12	138±31
200	1.68	6.99	140±29
300	1.68	6.89	142±31
400	1.66	6.69	147±30
500	1.06	3.89	158±31
600	0.25	0.89	161±28
700	0.01	0.03	161±30

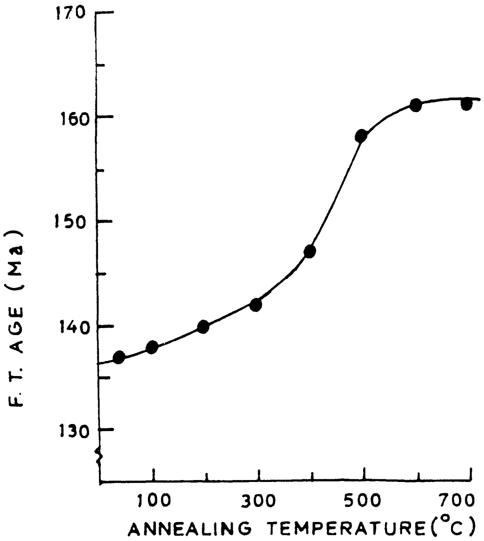

FIG. 5. Fission track age versus annealing temperature for chlorite.

ature above that corresponding to a previous heating event or at a temperature at which the tracks fade naturally. The curve obtained is shown in Fig. (5). The true age obtained by this method is 161±30 Ma and the age determined in the proceeding section is 137±28 Ma. Thus the annealing correction to f.t. age of chlorite is 14.9%, which is in good agreement with that determined by the method of calibration curve translating track length reduction into track density reduction.

4. Annealing characteristics of chlorite

The annealing procedure adopted in the present work is that of Naeser and Faul (1969). The irradiated chlorite samples were heated at different constant temperatures of 400, 425, 450 and 475°C in a muffle furance for times varying from a few minutes at higher temperatures to over 1,000 minutes at lower temperatures. The samples were then etched and the track density of residual tracks was measured. The percentage of tracks faded after each heating event versus log of heating time is plotted (Fig. 6). Fig. 7 depicts the annealing data of the mineral on Arrhenius diagrams. Individual points on this graph represent 0%, 25%, 50%, 75% and 100% track density reductions determined at various temperatures corresponding to the different times of Fig. 6. The data have been extrapolated to geologically meaningful times and temperatures. All the points on the Arrhenius plot that have the same reduction in track density lie on a straight line, the slope of which defines the activation energy. The activation energy for thermal fading of fission tracks was calculated from the following equation:

$$t = a\,e^{E/kT} \tag{5}$$

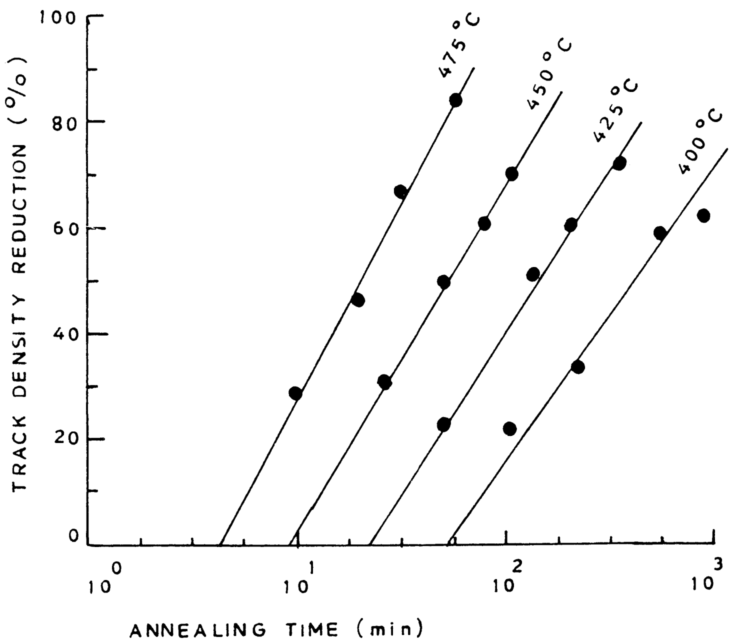

FIG. 6. Experimental results for track annealing in chlorite.

where

t = annealing time
a = constant, depending on the material
E= activation energy (eV)
k = Boltzmann's constant
(8.62×10^{-5} eV k^{-1})
T= absolute temperature (Kelvin).

Results and discussion

The values of V_T, V_G and η for chlorite are found to be 0.18 μm/s, 0.033 μm/s and 81.67%, respectively. The fission track age of 161±30 Ma for chlorite determined in the present investigations is comparable to the age of some accessory minerals obtained by Naeser and Dodge (1969), for the same region. The errors shown in the results are the standard deviations estimated from counting statistics. From the annealing data (Fig. 7) it is evident that a temperature of 155°C will erase all fission tracks in

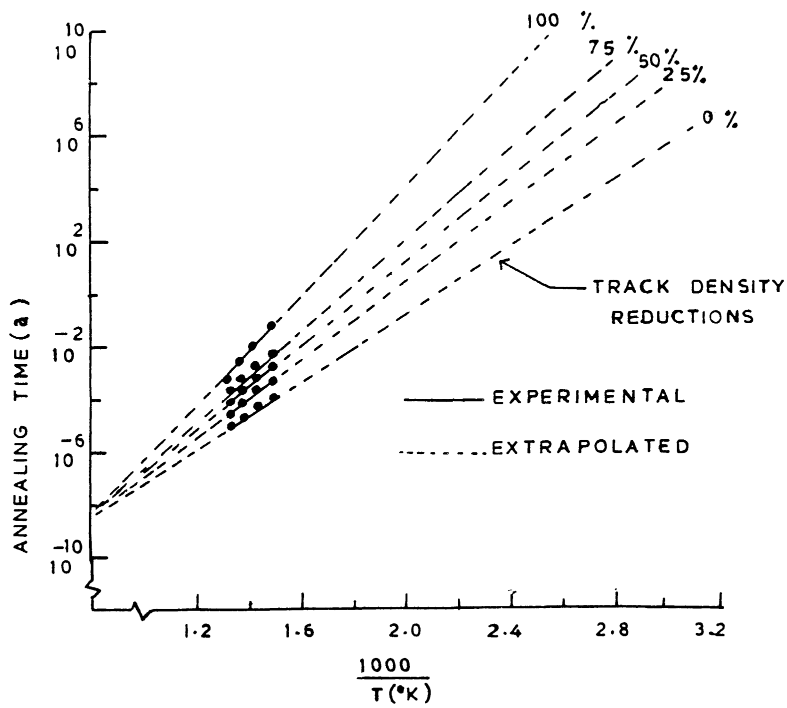

FIG. 7. Arrhenius diagrams for track density reduction at different temperatures.

chlorite in 1 Ma. The mean activation energy for thermal fading of fission tracks in chlorite as calculated from equation (5), is 1.6 eV.

Conclusions

Chlorite has high efficiency for track revelation and can retain tracks even at high temperatures. It is thus suitable for fission track dating.

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