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Etching studies of radiation damage in natural zircon

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The latent damage trails in natural zircon are etched using a new etchant reported by P Zaun and G A Wagner [*Naturwissenschaften (Germany)*, 72 (1985) 143]. The results for track etch rate (V_T), bulk etch rate (V_G) and etching efficiency (η) compare well with those for other etchants. It is observed that the new track etchant is more suitable for fission track age measurements than the other existing etchants.

1 Introduction

An understanding of the etching characteristics of zircon crystal is of considerable importance in view of the fact that it is a common and widely distributed accessory mineral in all types of rocks. Fission track (ft) dating has proved to be quite suitable for zircon due to its relatively high uranium content and low annealing rate of radiation damage compared to other minerals. However, the chemical etching of radiation damage in zircon is a tedious problem.

Historically, the search for a suitable etchant for zircon proved a long and difficult task. Fleischer *et al.*[1] originally described a boiling H_3PO_4 etchant at a temperature ranging from 375 to 500°C. Etching times under these conditions are only a few seconds or minutes which make precise control difficult. In addition, Naeser[2] developed an etching procedure using 100*N* NaOH solution at a temperature of 220°C. Under these conditions, etching time varies from 1 hr to many tens of hr. The important drawbacks to the use of this etchant are the formation of an obscuring precipitate on the zircon surface and the NaOH solution tending to dehydrate and solidify during the course of etching. Krishnaswami *et al.*[3] have developed an equivolume HF : H_2SO_4 etchant used at 150-180°C under pressure in a teflon-lined steel bomb. Gleadow *et al.*[4] have developed a binary eutectic etch melt of NaOH and KOH which overcomes the earlier difficulties.

More recently, Zaun and Wagner[5] have reported a new track etchant for zircon. This new etchant (NaOH : KOH : LiOH.H_2O = 6 : 14 : 1) reduces the etching time for track revelation compared to that with the eutectic etch melt. In our present study the latent damage trails in zircon were etched with three different etchants. The study was carried out using fission fragments from ^{252}Cf and induced tracks of ^{235}U.

2 Experimental Procedure

The zircon used in the present study is a large crystal of 2×1 cm^2 crystal surface. The crystal was procured from the Department of Geophysics, Osmania University, Hyderabad and extracted from the Khammam district (A.P.). Different samples from 100 plane were separated and prepared after grinding and polishing.

Track etching efficiency measurements—The fission fragments from ^{252}Cf source were allowed to pass through a collimator (circular slit) of 0.5 mm diameter in a vacuum chamber ($\sim 10^{-3}$ atm.) so that a parallel beam of fission fragments may be achieved. Then, polished samples from 100 plane of zircon were irradiated at two different angles, viz. $15 \pm 2°$ (for V_T measurements) and 90° (for V_G measurements). These irradiated samples were etched under three different etching conditions, described below in detail.

(i) In order to study the etching behaviour of fission tracks in zircon using acidic mixture[3], 5 ml of 48% HF was mixed with 5 ml of 98% H_2SO_4 in the teflon-lined steel capsule. The etching of the samples were carried out under pressure at 175°C. The samples were step etched and removed periodically from the capsule. The mean track length (corrected for dip) and diameter (major axis) were measured at each etching event using an optical microscope at a magnification of 1500× (+oil immersion).

(ii) Etching studies of fission tracks were also carried out using eutectic etch melt[4]. For this, 11.5 g of KOH was mixed with 8 g of NaOH in a crucible and brought up to a temperature of 200°C on a hot plate. The irradiated samples were placed in the molten etchant and the crucible was covered with inverted glass beaker for thermal insulation. The track length and diameter, were measured at various etching times.

(iii) After considerable experimentation, a slight modification was done in the new etchant reported earlier[5]. For this, 4 g of NaOH, 15 g of KOH and 6 g of LiOH H_2O were mixed with 2 ml H_2O in the teflon capsule. The irradiated samples were immersed in this improved etchant and the capsule was closed air-tight and placed in the muffle furnace pre-heated to 200°C. The track length and diameter were measured at various durations of etching.

Track density measurements—A few samples from 100 plane of zircon were annealed at a temperature of 800°C for 4 hr in order to remove fossil tracks. These samples were then irradiated with a thermal neutron fluence of 4.5×10^{14} n cm^{-2} from CIRUS reactor (IC-1 position) at the Bhabha Atomic Research Centre, Bombay, together with external muscovite detectors. Muscovite samples were pressed against the zircon surface during irradiation. The irradiated zircon samples (containing induced tracks of ^{235}U) were polished and etched using three different etchants. The density of etched tracks (Fig. 1) was determined at various durations of etching (Fig. 2). To reveal the induced tracks on external detector, the muscovite samples were etched for 50 min in 48% HF at 25°C.

3 Results and Discussion

The track etch rate is calculated from the slope of linear part of the curve showing the variation of track length with etching time. The bulk etch rate is measured from the one half of the slope of the curve etching time vs track diameter. The etching efficiency (η) and critical angle (θ_c) were calculated from the following relations[6,7]:

$$\eta = 1 - \sin \theta_c \qquad \ldots (1)$$

and

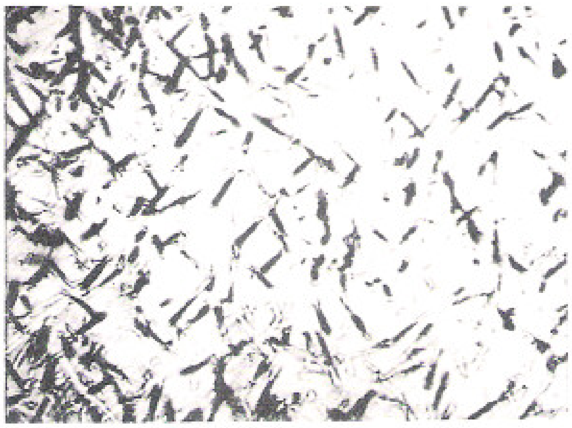

Fig. 1—Microphotograph of induced fission tracks in 100 plane of zircon using new improved etchant

$$\theta_c = \sin^{-1} \frac{V_G}{V_T} \qquad \ldots (2)$$

The value of V_T, V_G and η are found to be higher, using the new improved etchant (Table 1). This is due to the higher reactivity of LiOH etchant. The reactivity of the solutions increases in the order LiOH > KOH > NaOH. This has been qualitatively explained by considering the different radii of Li^+, Na^+ and K^+ ions in aqueous solutions. The small Li^+ ion with fewer water dipoles penetrates more easily and is, therefore, more reactive.

The induced tracks on zircon surface result from the passage of fission fragments across the internal surface from both above and below, producing a 4π geometry, while the induced tracks on external muscovite surface result from the one-way passage of fission fragments from the mineral into the external detector, giving a 2π geometry. The geometry factor (g) is calculated as:

$$g = \frac{\rho_{e.d.}}{\rho_{in}} \qquad \ldots (3)$$

where $\rho_{e.d.}$ and ρ_{in} are the induced track densities measured on the external surface of muscovite and internal surface of zircon, respectively. Fission track

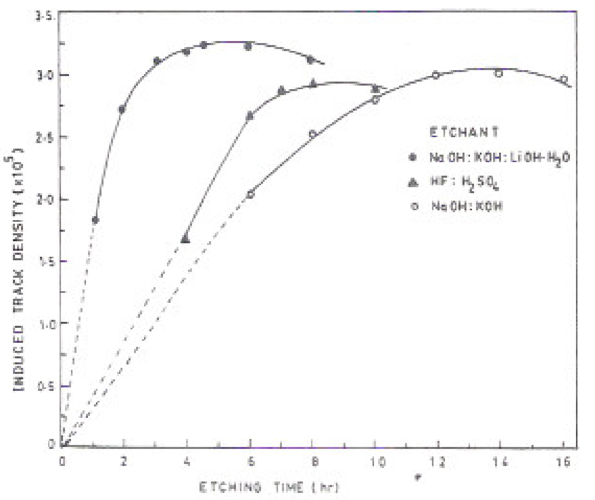

Fig. 2—Induced fission track density versus etching time for zircon, etched in different etchants

Table 1—Values of various etching parameters for zircon using three different etchants

Etchant	V_T (μm/hr)	V_G (μm/hr)	θ_c	η (%)
$HF : H_2SO_4$	1.37	0.108	4.5°	92.1
NaOH : KOH	0.45	0.030	3.8°	93.3
NaOH : KOH : $LiOH.H_2O$	6.20	0.150	1.4°	97.6

Table 2—Measured values of geometry factor (g) for 100 plane of zircon using different etchants

Etchant	$\rho_{e.d.}$ (muscovite) ($\times 10^4$)	ρ_{in} (zircon) ($\times 10^4$)	$g = \frac{\rho_{e.d.}}{\rho_{in}}$ ($= 2\pi/4\pi$)	Optimum etching time for zircon (hr)
$HF : H_2SO_4$	16.38	28.24	0.58	8
NaOH : KOH	16.57	29.58	0.56	12
NaOH : KOH : $LiOH.H_2O$	16.49	32.12	0.51	5

age of zircon is generally determined by using the external detector method[8,9], in which it is desirable that the etching efficiencies of both the zircon and the external detector (muscovite) be identical, so, that the appropriate geometry factor has an ideal value 0.5 ($=2\pi/4\pi$). With the new improved etchant, the value of g (Table 2) comes very close to the ideal value. Moreover, the optimum etching time required to reveal maximum track density (Fig. 2) is quite less using this new etchant.

4 Conclusion

The track etching efficiency of zircon is higher while using the new etchant. Thus, the new improved track etchant is most suitable for fission track dating work of zircon particularly when using the external detector method.

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