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The etching of alpha-recoil tracks in phlogopite

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Abstract

This work uses three phlogopite samples to investigate the etching behavior of alpha-recoil tracks (α-recoil tracks), which is a key problem in dating procedures. At the initial stage of the etching process, the number of alpha-recoil tracks increased linearly with etching time, then the linearity was interrupted due to the overlapping of alpha-recoil tracks. The slope of the etching line could be similar and also could be different both in the same sample and in different samples. It is shown that the etching time could vary with different contents of U and Th and could not exceed 200 min in 4% HF acid at $25 \pm 1\,°C$ in order to get a satisfactory etching line and to calculate the exact density. We propose two valid methods that can accurately determine the position of the sample's surface and may reduce the alpha-recoil track dating errors.

Keywords: Alpha-recoil tracks; Etching; Track density; Phlogopite

1. Introduction

Uranium and thorium are trace elements in natural phlogopite. Their isotopes ^{238}U, ^{235}U and ^{232}Th as well as their daughter nuclei undergo radioactive decay by emitting a series of α particles (Wagner, 1998). The energy is released through successive α disintegration, which is predominantly the kinetic energy of the ejected α particles according to the principles of momentum and energy conservation. The heavy residual nuclei are recoiled and transfer energy to the crystal lattice of the mineral through nuclear collisions which form radiation damage zones that become observable as alpha-recoil tracks (ARTs) under an optical microscope with incident light and N-DIC (Nomarski-differential-interference-contrast) after chemical etching (Gögen, 1999; Huang et al., 1967; Katcoff, 1969). Visualization of etched ARTs by Nomarski-differential-interference-contrast microscopy (NDICM) and scanning force microscopy (SFM) enables the access to areal densities of ART etch pits beyond $10^4\,\mathrm{mm}^{-2}$ and thus the extension of the new ART-dating technique to an age range > 1 Ma. The successful application of SFM as a new tool in geochronology could open the way to a new field which could be characterized as nano-geochronology (Garrison et al., 1978; Glasmacher et al., 2003).

If the U and Th concentration and the ART volume density are known, a geological or archaeological age can be calculated. The following etching model (Gögen and Wagner, 2000) was established in order to obtain the volume density of ARTs (Jonckheere et al., 2005; Stübner and Jonckheere, 2006):

$$\rho_{\mathrm{ART}} = N_{\mathrm{ART}}[R_e - R_0(t_e) + v_v t_e] = N_{\mathrm{ART}}[R_c(t_e) + v_v t_e] \quad (1)$$

where N_{ART} is the volumetric track density, v_v is the vertical etching velocity, t_e is the etching time, R_e is the effective etchable range of the latent track and R_0 is the resolution of the optical microscope. R_0 cannot be assumed to be a constant, but for high microscopic magnification it is a function of etching time because both the R_0 and the effective etchable range R_e are unknown or cannot be determined easily. Their combined factor $R_c(t_e) = R_e - R_0(t_e)$ is introduced. $N_{\mathrm{ART}} R_e$ is the number of ARTs in a unit area for the unetched surface and $N_{\mathrm{ART}} v_v t_e$ is the number of tracks per unit surface added by etching. In principle, N_{ART} can be calculated from either the slope of the regression line fitted to a series of track counts at different etching times, but the intercept precision could be lower because of the steep slope of the regression line. Therefore the intercept of a regression line is not so good for a calculation of N_{ART} and

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it is better to calculate the N_{ART} value using the slope of the regression line. Eq. (1) is equivalent to the equations of Khan and Durrani (1972) for a fission track detector with $\theta_c = 0$.

The N_{ART} value could be obtained if the relationship between ρ_{ART} and t_e is known based on Eq. (1). Due to the possibility of uneven distribution of U and Th in phlogopite, the former results of slope of the etching line cannot give a convincing result of whether the number of tracks increases strictly linearly with etching time or not. It is possible that N_{ART} derived from a single etching curve plus the even value of U and Th concentrations of many randomly selected areas may result in a large error for the age determination of the mineral (Wolfman and Rolniak, 1978). For this reason, three samples were prepared, etched and observed under an optical microscope.

2. Experimental procedure

In order to determine the relationship between ρ_{ART} and t_e, the three phlogopite samples were used from the 12th crater situated in the area of Hama pond, Jingbo Lake in the northeast part of China. The samples were gathered from the same locality of the crater. Each of the samples was fixed on a HF acid resistant Teflon holder by resinous glue, which is also HF acid resistant. A clean flat surface on each phlogopite sample was prepared. Then the samples were etched in 4% HF acid at $25 \pm 1\,^{\circ}\mathrm{C}$. The etching process was interrupted at 15 min intervals for track observation under an optical microscope with IL-NDIC (Lang et al., 2003). Different areas were randomly chosen for the samples. In order to accurately determine the position of every observed area after each etching interval, two valid methods, termed "manual" and "automated" methods, respectively, have been developed. In the automated method, a zero point needs to be determined on the PTFE (Polytetrafluoroethylene) holder and in the manual method orthogonal marks were made on the surface of the phologopite.

The etching process continued until ARTs overlapped to an extent where most tracks could not be distinguished under the microscope (Rufe and Hochello, 1999). Tracks were counted after each time interval and so the curve for etching time versus track numbers could be plotted. The observed area was $1.869 \times 10^{-4}\,\mathrm{cm}^2$ after calibration.

3. Results and discussion

During the etching process, some areas were gradually destroyed and became inappropriate for further counting. Indeed, single tracks can still be distinguished although there were overlaps of ARTs at the initial stage of the etching process. As the etching time increased (Jonckheere and Gögen, 2001; Ziegler et al., 1985), tracks overlap to such an extent that the number of tracks making up the clusters on the etching surface could no longer be confirmed. The linearity of the etching line was interrupted due to the non-proportional relationship between overlapping tracks and newly produced tracks and this finally reached a fluctuating "plateau" (Hashemi-Mezhad, 1998). Old layers of phlogopite were removed while new layers were exposed to the chemical etchant during the etching process. The track number may vary between layers due to the variation of U and Th contents or bulk composition in both vertical and horizontal directions. Further studies on the variation of the constitution of bulk composition are required in the future.

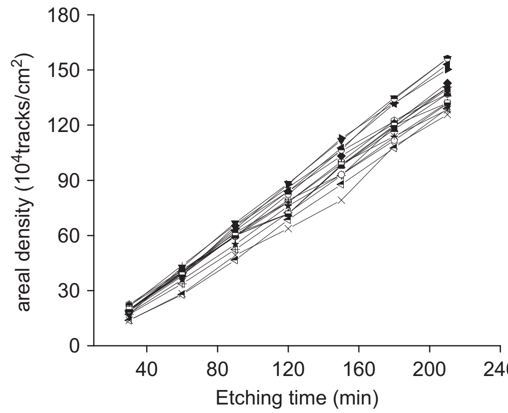

Fig. 1. Etching lines of all the observed areas in sample1.

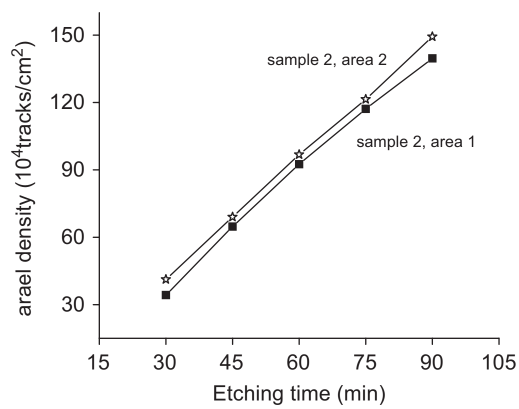

Fig. 2. Etching lines for sample 2.

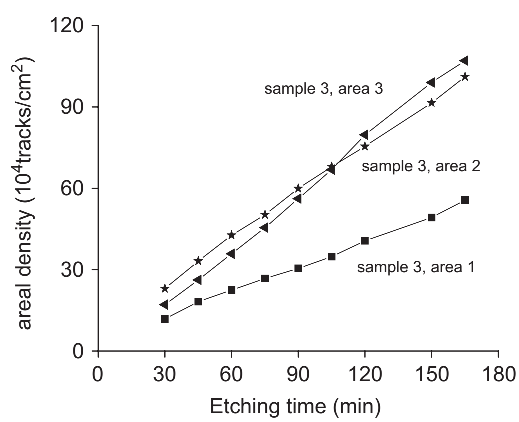

Fig. 3. Etching lines from sample 3.

The etching line of each selected counting area was drawn and isochronal data in different areas was finally summed up in order to get another etching line. Tracks increased linearly with etching time in every observed area (Figs. 1, 3, and 5).