Nuclear Instruments and Methods in Physics Research B46 (1990) 149-151
North-Holland

ETCHING AND ANNEALING KINETICS OF HEAVY ION TRACKS IN QUARTZ CRYSTAL

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Swift heavy ions of sufficient size and energy produce stable latent tracks in most of the solid state nuclear track detectors. The etching and annealing kinetics of ^{208}Pb (13.8 MeV/n) ion tracks in natural quartz crystal were investigated. The values of various etching parameters, viz. track etch rate V_T, bulk etch rate V_B, etching efficiency η and activation energy for track etching E_T, are reported. The activation energy for annealing of latent tracks using a modified empirical formulation is determined.

1. Introduction

Recent developments in etching and annealing studies of heavy ion tracks in minerals and meteoritic crystals have an increasing interest due to their wide applications in geochronology and for the study of the charge spectrum of ancient cosmic rays [1-3]. Most of the previous studies on thermal recovery of the damage are restricted to the fission fragment tracks in minerals [4,5], glasses [6] and plastics [7]. These studies do not reflect the exact picture of the etching and annealing mechanism because of the lower range, undefined mass and energy and more statistical errors inherent in fission fragment tracks. So, for the present studies, ion tracks in the $10\bar{1}0$ plane of a quartz crystal, produced using a well defined beam of ^{208}Pb (13.8 MeV/n), is investigated. An attempt is also made to fit the laboratory annealing data in the modified empirical relation to describe the annealing kinetics of ^{208}Pb (13.8 MeV/n) ion tracks as a function of time and temperature which, as a consequence, determines the spatial distribution of various defects produced along the environs of heavy ion trails. The quartz crystal used in the present study was obtained from GSI, Calcutta, and originated from the pegmatite schists of Bihar State, India.

2. Experimental details

The quartz samples, after cutting along a $10\bar{1}0$ plane, were ground with SiC powder and finally polished with 8, 3, 1 μm diamond paste in successive steps. The polished surface was then exposed to a 30° incident collimated beam of ^{208}Pb (13.8 MeV/n) ions provided by the UNILAC heavy ion accelerator at Darmstadt, FRG. A few samples were also exposed with the incident beam normal to the surface in order to study the bulk etch rate V_B. The samples were cut into a number of small pieces for a wide study of etching and annealing phenomena. The samples were then annealed in a Muffle furnace over the temperature range 873-1223 ± 3 K for 10-50 min.

2.1. Measurement of etching rates (V_T and V_B)

For measuring the track etch rate V_T, the exposed samples (unannealed and annealed at various temperatures) were etched in boiling 20N KOH for different time intervals (1-12 h) using an oil etching bath and reflux condenser assembly. After each etching interval, the samples were washed in tap water, dried and scanned under an optical microscope to record the projected etched track length at a magnification of 400 ×. The etching and microscopic observations were repeated until the maximum projected track length became invariant with further etching. The slope of the linear portion of the graph (fig. 1) gives the track etch rate of different annealed and unannealed samples. The bulk etch rate, V_G, was calculated [8] from one half of the slope of the curve, etching time versus track diameter (fig. 2).

2.2. Measurement of η and E_T

The etching efficiency, η, of ^{208}Pb (13.8 MeV/n) ion tracks is determined using the following relations [9,10]

$$\eta = 1 - \sin\theta_c = 1 - V_B/V_T, \qquad (1)$$

where θ_c is the critical angle of incidence for track etching.

The activation energy for track etching, E_T, is calculated using a modified empirical ralation [11] taking into account the etchant concentration

$$V_T = \alpha c^n \exp(-E_T/kT'), \qquad (2)$$

where α is a proportionality constant; n, the exponent of concentration, c; T' the etchant temperature; k Boltzmann's constant and E_T, the activation energy for

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(North-Holland)

Table 1
Values of the etching parameters V_T, V_B, η, θ_c and E_T of ^{208}Pb (13.8 MeV/n) ion tracks (unannealed and annealed at different temperatures for a constant time of 30 min)

Track annealing temperature [K]	V_T [μm/min]	V_B [μm/min]	η [%]	θ_c [deg]	E_T [eV]
Unannealed (298)	0.40	0.0063	98.4	0.90	0.16
973	0.344	0.064	98.1	1.06	0.21
1073	0.120	0.064	94.6	3.05	0.30
1173	0.097	0.0063	93.5	3.70	0.37

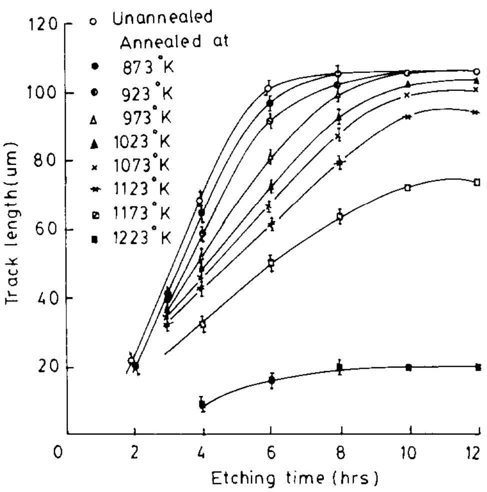

Fig. 1. The variation of track length with etching time of unannealed and annealed (30 min) ^{208}Pb (13.8 MeV/n) ion tracks in quartz.

track etching. E_T is determined from the slope of the graph (fig. 3) between track etch rate and inverse of etchant temperature. The values of V_T, V_B, η, θ_c and E_T of annealed and unannealed samples are given in table 1.

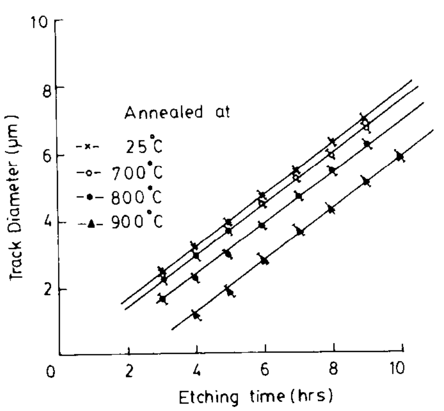

Fig. 2. The variation of track diameter with etching time of annealed and unannealed tracks.

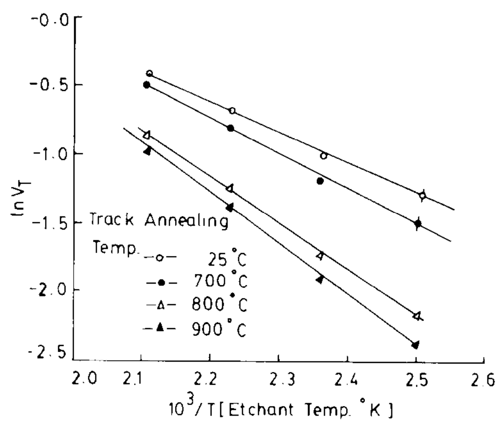

Fig. 3. The plot of ln V_T vs $1/T$ [K^{-1}] for unannealed and annealed samples.

3. Annealing kinetics and activation energy

Dartyge et al. [12] have predicted from the study of X-ray scattering of latent tracks in minerals that two types of defects, i.e. point defects and extended defects, are created during the passage of heavy ions. It is the latter which appears to dominate the observed track retention during annealing studies. It is experimentally found that the size and quantity of extended defects depends on annealing and on the size of the incident ion beam [13].

Fletcher and Brown [14] have proposed three annealing stages of the point defects caused by electron irradiation on the basis of electrical parameters including (1) monomolecular recombination of Frenkel defects (2) transient diffusion of point defects towards defect sinks, and (3) bimolecular recombination of the complementary defects. The annealing of heavy ion produced latent tracks has a parameter of track retention rate, which plays a major role towards understanding the annealing kinetics at elevated temperatures. On the basis of this parameter, we have proposed a modified form of the empirical relation of Modgil and Virk [15], which relates the track retention rate with the activation energy as a function of both time and temperature and which is given by

$$1 - r = At^{l-n} \exp(-E_a/kT), \tag{3}$$

where $r = l(t)/l(0)$ is the ratio of track length after and before annealing, A and n, annealing constants, and k is Boltzmann's constant. The slope of the plot of $\ln(1-r)$ versus the inverse of the temperature yields the activation energy for track annealing. It is observed that the laboratory annealing data at higher temperature (1073–1223 K) fit well according to eq. (2).

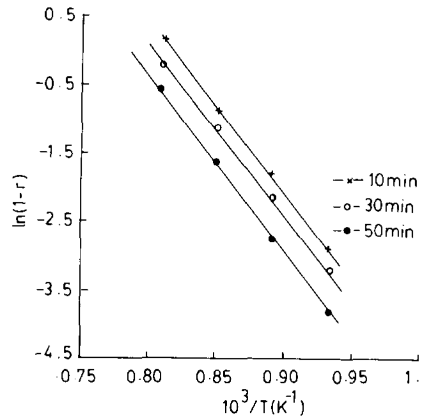

Fig. 5. The plot of $\ln(1-r)$ vs $1/T$ [K^{-1}] for different annealing times.

4. Discussion

By annealing studies, it is seen from fig. 1 that the track etch rate, V_T, decreases with healing of latent tracks. However, the bulk etch rate, V_B (fig. 2), remains almost invariant. The etching efficiency, η, decreases while the activation energy, E_T, for track etching increases with the annealing of latent tracks (table 1). This is due to the fact that free energy deposited along the damage goes on decreasing with thermal treatment and consequently it becomes difficult for the etchant to attack the annealed as compared to the unannealed tracks. Fig. 4 describes the variation of track length retention ratio, r (as in eq. (3)) versus temperature for different annealing time intervals (10–50 min). The single value of the activation energy at higher temperature (800–950 °C) is determined using eq. (3). The parallelism of the lines in fig. 5 shows that the single value of the activation energy 2.11 eV is isolated at different annealing time intervals (10–50 min).

We are thankful to Drs. R. Spohor and J. Vetter for providing heavy ion irradiation facilities at the UNILAC accelerator, Darmstadt. Two of the authors (L. Singh) and (A.S. Sandhu) gratefully acknowledge the financial support of the Council of Scientific and Industrial Research (CSIR), New Delhi.

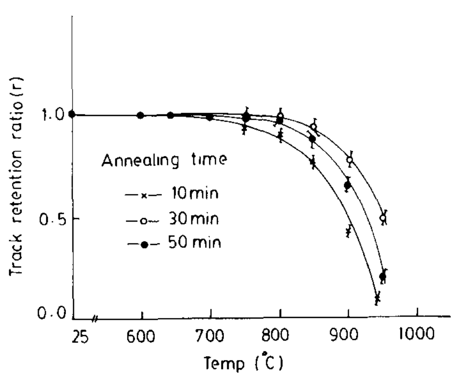

Fig. 4. The variation of track length reduction ratio with annealing temperature for different annealing times.

References

[1] R.L. Fleischer, P.B. Price, R.M. Walker and M. Maurette, J. Geophys. Res. 72 (1967) 333.
[2] C. Perron and M. Maury, Nucl. Tracks 11 (1986) 73.
[3] K. James and S.A. Durrani, Earth Planet. Sci. Lett. 87 (1988) 229.
[4] J.N. Gowswami, R. Jha and D. Lal, Earth Planet. Sci. Lett. 71 (1984) 120.
[5] W. Ritter and T.D. Mark, Nucl. Instr. and Meth. B14 (1986) 314.
[6] G. Walder and T.D. Mark, Nucl. Instr. and Meth. B32 (1988) 303.
[7] J.H. Adams and L.P. Beahm, Proc. 11th Int. Conf. on SSNTDs, Bristol (1981) p. 163.
[8] W. Enge, K. Grabisch, R. Beaujean and K.P. Bartholoma, Nucl. Instr. and Meth. 115 (1974) 263.
[9] R.L. Fleischer, G.M. Comstock and H.R. Hart, J. Geophys. Res. 77 (1972) 5050.
[10] R.L. Fleischer, P.B. Price and R.M. Walker, Nucl. Tracks in Solids (Univ. California Press, Berkeley, 1975).
[11] G. Somogyi and I. Hunyadi, Proc. 10th Int. Conf. on SSNTDs, Lyon (1979) p. 443.
[12] E. Dartyge, J.P. Duraud, Y. Langevin and M. Maurette, Phys. Rev. B23 (1981) 5213.
[13] L. Singh, A.S. Sandhu, S. Sing and H.S. Virk, Radiat. Eff. Defects Sol. (1989) in press.
[14] R.C. Fletcher and W.L. Brown, Phys. Rev. 92 (1953) 585.
[15] S.K. Modgil and H.S. Virk, Nucl. Instr. and Meth. B12 (1985) 21.