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RECOVERY STAGES OF HEAVY ION PRODUCED DEFECTS IN QUARTZ CRYSTAL

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ABSTRACT

Heavy ions of sufficient energy, when traverse through the crystal lattice, cause the production of different types of defects along the environs of their trajectories. To understand the systematic annealing mechanism of such defects the various isothermal and isochronal annealing experiments were performed on ^{238}U(15.36 MeV/n) ion tracks in quartz crystal in the laboratory. From the analysis of such experiments, two recovery stages are found to be mainly effective to describe the annealing behaviour. The values of activation energies for the two annealing processes are reported in the present paper.

INTRODUCTION

The phenomenon of thermal annealing of radiation damage tracks in minerals has been studied now for over two decades by various authors [(Galazka & Burchart, 1977; Mark et al (1980); Price et al (1987)]. Most of the previous studies do not reveal the reaction kinetics to predict the annealing behaviour for arbitrary initial damage. The recent studies [Bertal & Mark (1983) and Singh et al.(1990)] have been attempted to elucidate the contribution of various reaction mechanisms. But still the existing theories of track fading are being questioned.

In the present paper an attempt has been made to understand the thermal response to ^{238}U(15.36 MeV/n) ion tracks in quartz crystal as a function of time & temperature, which as a consequence determines the microscopic spatial distribution of various defects produced along the environs of heavy ion trails.

ENERGY LOSS AND DEFECT PRODUCTION

Although the major part of the heavy ion energy loss per unit length of its range is accounted for by the ionization and excitation of the atoms in its way but still the other processes like localization of the excitons, formation of electron-hole pairs and self trapped excitons can not be ignored during the track formation (Singh, 1991). These defects are known as the point defects. The main type of the defects which plays a special role in the formation of etchable heavy ion trails, involve the Coulombic repulsive forces as well as the relaxation due to elastic and mechanical stresses and strains and are known as extended defects. So it is accepted that the heavy ion latent track consists of all the above types of defects, particularly of extended defects which are separated from the loaded zones of point defects.

RECOVERY STAGES OF DEFECTS

It is established that these defects diffuse to the defect sinks to heal the damage, particularly when the exposed samples are subjected to thermal treatment. To understand the systematic annealing mechanism of such defects produced with ^{238}U(15.36 MeV/n) ions in [10$\bar{1}$0] plane of natural quartz crystal, the various annealing & etching experiments are performed in the laboratory. From the analysis of the experimental data, two empirical relations are postulated which govern correspondingly two recovery stages. Basically we have two measureable physical parameters i.e. track etch rate V_T and track retention rate, r, as a function of time and temperature. These parameters can be related with activation energy for annealing. The first empirical formulation is based on the decrease in track etch rate V_T with temperature and is given by (Singh & Singh, 1989).

$$[\{V_T(o) - V_T(t)\}/t] = A\, t^{-n} \exp(-E_{eff}/KT) \qquad \ldots\ldots 1$$

where $V_T(o)$ and $V_T(t)$ are the etching rates before and after annealing, K the Boltzmann's constant, n, the exponent of annealing time and E_{eff} is the effective activation energy for monomolecular recombination of point defects. The etching rates used here are the instantaneous etching rates and are determined from the linear portion of the graph (Fig. 1). It is clear from the experimental data involving the etching rates of annealed and unannealed tracks that the above empirical relation fits best only upto a certain lower range of temperatures, however, at higher temperatures the

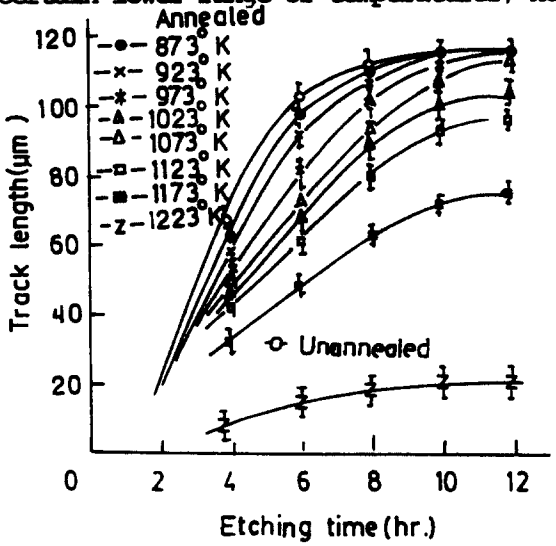

Fig. 1. Plot of etched track length versus etching time of ^{238}U(15.36 MeV/n) ion tracks incident at 30° in [10$\bar{1}$0] plane of quartz.

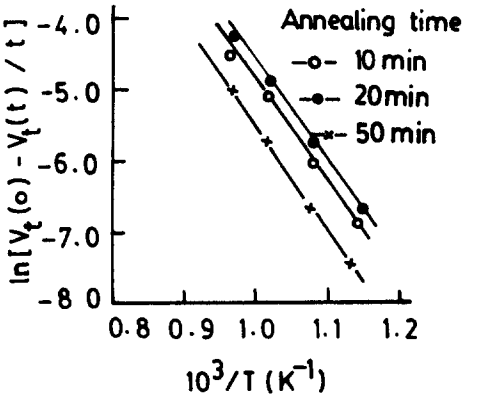

Fig. 2. Plot of $\ln[\{V_T(0) - V_T(t)\}/t]$ versus inverse of temperature

fitting diminishes gradually. But at the same time the etchable track length remains invariant during low temperature annealing. Thus this recovery stage may be termed as the stage of monomolecular recombination of the point defects or first recovery stage. For calculating the effective activation energy for this stage the eqn. 1 can be rewritten as-

$$\ln [\{V_T(o) - V_T(t)\}/t] = \ln A - n \ln t - E_{eff}/KT \qquad \ldots\ldots 2$$

The slopes of the graphs (Fig.2) between left hand side of this eqn. and T will give activation energy for monomolecular recombination at constant time of annealing. When the experiments are carried out at higher temperature range the experimental data relating etching rates at higher temperatures could not be described by eqn. 1. However at the same time the other

measureable physical parameter i.e. track retention rate plays a predominant role (Fig.3). It may be due to the healing of extended defects and consequently involves the second recovery stage. The activation energy for annealing of this stage can be related with temperature and time in the empirical relation, which describes the annealing behaviour at higher temperature range and is given by (Singh & Singh, 1989)-

$$1 - r = A.t^{1-n} \exp(-Q/KT) \qquad \ldots . 3$$

where $r = l(t)/l(o)$ is the ratio of annealed and unannealed track lengths and is known as track retention rate. A is the annealing constant depending upon the nature of ion and target material. To determine the activation energy for this stage the eqn. 3 can be rewritten as-

$$\ln (1-r) = \ln A + (1-n)\ln t - Q/KT \qquad \ldots . 4$$

where Q is the activation energy for second recovery stage and is

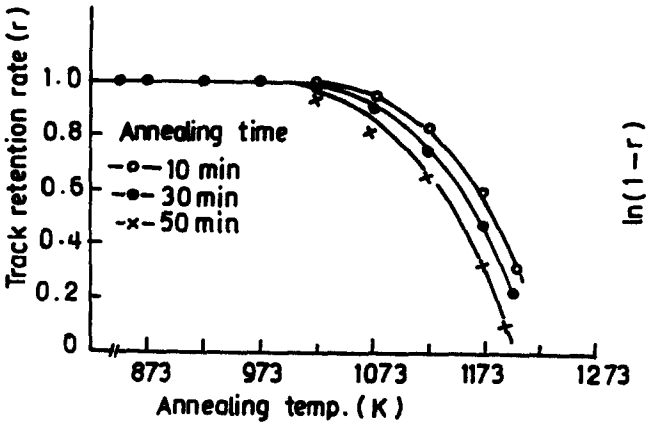

Fig. 3. Plot of track retention rate (r) versus inverse of annealing temperature

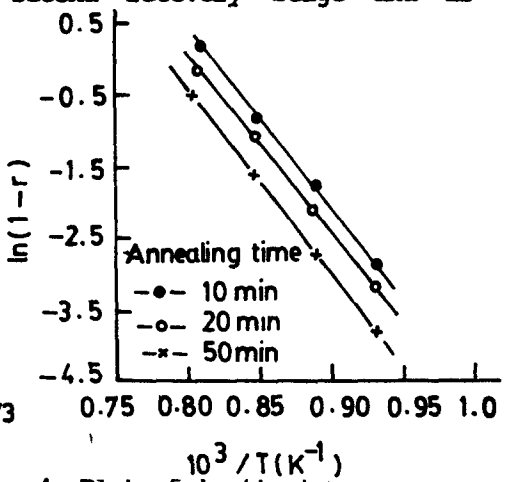

Fig. 4. Plot of ln (1-r) versus inverse of annealing temperature

determined from the slope of graph (Fig. 4) between left hand side of eqn.4 and T^{-1}.

EXPERIMENTAL PROCEDURE

The quartz samples are prepared by cutting, grinding and polishing the both sides of [10$\bar{1}$0] plane. The samples were then exposed with ^{238}U (15.36 MeV/n) ions at the incident angle of 30° using the facility of UNILAC heavy ion accelerator at GSI Darmstadt. The samples were then subjected to thermal treatment in a temperature controlled muffle furnace (± 3°C accuracy). Both the annealed and unannealed samples were ethed in 15N-KOH solution using oil bath and reflux condenser assembly. These samples were then scanned under optical microscope for measuring the observed track length.

RESULTS AND DISCUSSION

Fig. 1 shows the variation of mean track length versus etching time of unannealed and annealed tracks of ^{238}U(15.36 MeV/n) ions in [10$\bar{1}$0] plane of quartz crystal. The curves regarding the annealed tracks represent the data for 30 minutes annealing time at eight different temperatures (823, 923, 973, 1023, 1073, 1123, 1173 and 1223K). These temperatures are chosen to get preferential change in track etch rate and track retantion rate. It is found that track etch rate V_T goes on decreasing with annealing of tracks. This is due to the reason that free energy deposited along the

track decreases with annealing and it becomes more difficult for the etchant to dissolve the annealed latent tracks as compared to unannealed latent tracks. If we observe closely the upper part of the curves (Fig. 1), then it can be inferred that there is no change in maximum track length up to 973K for constant annealing time, however, a systematic variation of track etch rate V_T occurs at lower temperatures (873 – 1023K). The annealing at higher temperatures failed to yield the systematic results using V_T, but at the same time the track retention rate (r) plays a predominant role to study the systematic annealing behaviour. Thus two recovery stages are proposed to analyse the complete annealing behaviour.

The slope of the graphs (Fig. 2) between ln $[\{V_T(0) - V_T(t)\}/t]$ versus T will give the activation energy for first recovery stage while the slope of graphs (Fig. 4) between ln (1-r) versus T^{-1} will give the activation energy for second annealing stage. The results of the activation energy for each

Table 1. The values of activation energies for two recovery stages of ^{238}U(15.36 MeV/n) ion tracks incident at 30°C in the [10I0] plane of quartz crystal for different annealing times.

Ion (Energy)	Annealing time (min.)	Activation energy (eV) Ist. stage (E_{eff})	2nd stage (Q)
^{238}U(15.36 MeV/n)	10	1.44	2.10
	30	1.43	2.12
	50	1.43	2.13

annealing stage at annealing time of 10, 30 and 50 minutes are given in Table 1. It is interesting to note that activation energy for each anneal ing stage is almost independent of the annealing time.

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