JOURNAL OF SOLID STATE CHEMISTRY **18**, 313–316 (1976)

Kinetic Study of the Annealing of Fission Tracks in a Fe_2O_3–KPO_3 Glass Containing 100 ppm of Uranium

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Received January 16, 1976; in revised form March 29, 1976

The kinetics of the annealing of fission tracks in an artificial glass were studied using an optical microscope. The annealing experiments were performed in an electric furnace at 160–230°C for 15–160 min. The measurement of the fission tracks through an optical microscope was carried out after etching the annealed tracks with 10 *N* NaOH aqueous solution at 40°C. All reactions were found to be of first order within the annealing temperatures. An activation energy of 0.66 eV for the annealing reaction was obtained from the Arrhenius plot of the rate constants determined at 160, 190, 210, and 230°C.

Introduction

There are several papers concerning the annealing of fission tracks in minerals (*1–4*). Such studies are very interesting not only because they are useful for age estimation of a mineral at some ambient temperature, but because they can give us some information about the thermal stability of tracks in each mineral. It is also interesting to know whether the activation energy for the annealing is constant or not during the annealing process and within the annealing temperatures. The activation energy varies from one annealing process to another in (*3*), whereas the activation energy is almost constant in (*4*). Apparent rate constants were obtained by substituting the isochronal data into the ordinary equation of the rate constant in our previous paper (*5*). The order of annealing reaction and the value of activation energy were determined from the Arrhenius plot of the apparent rate constants on the basis of the assumption that the activation energy is constant during the reduction process.

This paper represents the results of the kinetic study of the annealing reaction of the fission tracks in Fe_2O_3–KPO_3 glass. The results were obtained from the ordinary kinetic study which is usually used in the reactions in solutions.

Experimental

The Fe_2O_3–KPO_3 glass containing 100 ppm of uranium was synthesized by quenching the melt of a mixture of Fe_2O_3–KPO_3–$UO_2(NO_3)_2 \cdot 6H_2O$ between two copper plates (*6*). Ferric oxide and uranium nitrate are of guaranteed reagent grade and potassium metaphosphate is of extra pure grade. Ferric oxide and potassium metaphosphate were weighed accurately so that the Fe/P ratio becomes 1:5 and they were mixed in a platinum crucible. An aliquot of aqueous solution of $UO_2(NO_3)_2 \cdot 6H_2O$ was added to the mixture and dried for a short time on an electric heater at about 110°C. The solid mixture was then heated in an electric furnace at 1200°C for 2 hr and then quenched. The glass synthesized in this way showed a transparent brown color. The glass was irradiated in the nuclear reactor of Rikkyo University ($\Phi = 5.5 \times 10^{11}$ N/cm^2·sec) for 30 min at room temperature. The glass was then cut up into smaller pieces and parts of them were mounted in Epoxylite resin on a slide glass. They were polished on a glass board with a little carborundum and then on felt.

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In order to observe the fission tracks in the glass by using an optical microscope, the chemical etching (i.e., enlargement of the tracks by chemical reagents) condition of the tracks was determined.

The annealing of a few pieces of glass in each experiment was performed in an electric furnace at 160, 190, 210, and 230°C for different times. The annealed glasses were then etched and the tracks were observed by using an optical microscope. The mean pit (etched track) density was measured by using an oil immersion objective (100X) and a glass section ruled into 5-mm squares at a 1000X magnification. The mean pit density was obtained by counting the etch pits in the square section, set over the Fe_2O_3–KPO_3 glass 100 times randomly (*5*). The error bars in the graphs correspond to the limits with a *t*-distribution reliability of 95%.

Results and Discussion

The etching curve is shown in Fig. 1. All pieces of glass were etched in 10 *N* NaOH aqueous solution at 40°C in a thermostat. The mean pit density (number of etch pits per square centimeter) for the individual etching time is plotted against the etching time. The etching condition for this glass was determined to be 1 min because the mean pit density became almost constant. The results of annealing experiments are shown in Table I. The order of the annealing reaction

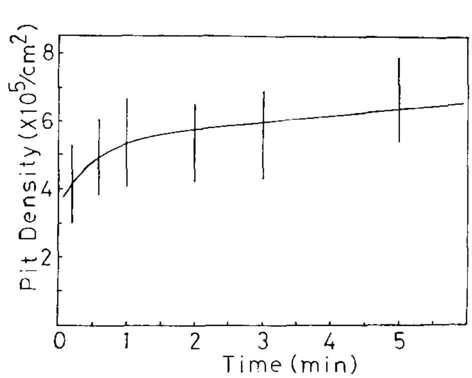

FIG. 1. Etching curve for the fission tracks in a Fe_2O_3–KPO_3 glass. The mean pit density is plotted against the etching time.

TABLE I

RESULTS OF THE ANNEALING EXPERIMENTS OF THE FISSION TRACKS IN A Fe_2O_3–KPO_3 GLASS

Temperature (°C)	Time (min)	Mean pit density (N_i) ($\times10^5/cm^2$)
160	40	3.98 ± 0.19
	80	3.02 ± 0.15
	120	2.58 ± 0.15
	160	1.56 ± 0.12
190	20	4.00 ± 0.18
	40	2.99 ± 0.16
	80	1.47 ± 0.21
	120	0.860 ± 0.109
210	20	2.54 ± 0.18
	40	1.69 ± 0.16
	60	0.768 ± 0.104
	80	0.568 ± 0.094
230	15	2.31 ± 0.19
	30	0.776 ± 0.105
	45	0.232 ± 0.067

was determined by substituting the annealing data into the N_0 and N_i containing terms in the ordinary formula of the rate constant and by plotting them on a graph. N_i and N_0 are the mean pit densities with and without annealing and the latter is $(5.38 \pm 0.25) \times 10^5/cm^2$ in the case of this glass.

All the annealing reactions were found to be of first order for this glass. The first-order reaction plot is shown in Fig. 2. Almost all plots are well matched on the straight lines and the rate constants can be obtained from the slope of the individual line. The second- and third-order reactions were lying far away from the straight lines, and therefore they are not shown in this paper. The rate constants for the first-order annealing reactions are shown in Table II. An activation energy of 0.66 eV was obtained from the Arrhenius plot of these rate constants (Fig. 3).

The thermal stability of fission tracks in glasses is generally known to be lower than those in the other minerals. The activation energy for the annealing reaction of fission tracks in this Fe_2O_3–KPO_3 glass is about 3–4 times lower than that in zircons (*5*).

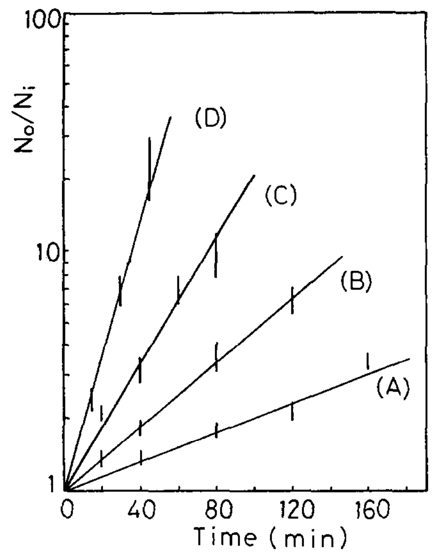

FIG. 2. First-order annealing reaction plot for the fission tracks in a Fe_2O_3–KPO_3 glass at (A) 160°C; (B) 190°C; (C) 210°C; (D) 230°C. N_0/N_t is plotted against the annealing time.

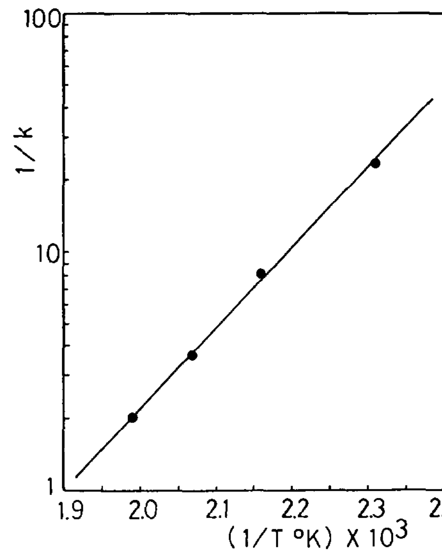

FIG. 3. Arrhenius plot for the annealing reaction of the fission tracks at 160, 190, 210, and 230°C. $1/k$ is plotted against the reciprocal of the annealing temperature.

TABLE II

RATE CONSTANTS FOR FIRST-ORDER ANNEALING REACTION OF FISSION TRACKS IN A Fe_2O_3–KPO_3 GLASS

Temperature (°C)	$1/T$ (°K) × 10^3	$k \times 10^3$ (min^{-1})	log k
160	2.31	44.6	−1.35
190	2.16	125	−0.90
210	2.07	276	−0.56
230	1.99	507	−0.29

In the case of zircon the activation energy ranged between 1.7 and 2.9 eV and the orders of the annealing reactions were almost third or fourth. These results showing that the order of annealing reaction of fission tracks is higher in the solids having higher thermal stability and lower (i.e., first-order reaction) in the solid having lower thermal stability can lead to the following useful idealization. The order of annealing reaction of fission tracks in solids seems to be closely related to the thermal stability of the solids. Thus the atoms around the fission track are less mobile and accordingly the track is less annealed in solids having the higher order of annealing reaction. If the annealing of the fission tracks in glasses occurs by bulk viscous flow, first-order reactions will be obtained. The rate of the annealing will be proportional to an appropriate strain rate which is inversely proportional to the viscosity. As shown in Fig. 3, the activation energy for the annealing of the tracks in this glass is constant within the annealing temperature (160–230°C). It is therefore concluded that the energy to anneal the tracks will be independent of the temperature. These results confirm that the kinetic study which is usually used in the reactions in solutions can be applicable in the case of the annealing of fission tracks in solids.

Acknowledgment

We are grateful to the staff of Rikkyo University for generous assistance in carrying out the neutron irradiation of the sample.

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