Nuclear Instruments and Methods in Physics Research B12 (1985) 212–218
North-Holland, Amsterdam

ANNEALING OF FISSION FRAGMENT TRACKS IN INORGANIC SOLIDS

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Received 30 August 1984 and in revised form 3 March 1985

A three step model is proposed to describe the annealing of fission fragment tracks in inorganic solids. An empirical formula relating the annealing rate, V_a and the activation energy of annealing, E_a is proposed. Activation energies for annealing are found to vary from 0.16 to 1.0 eV for the detectors studied. The observed activation energies are independent of the annealing temperature. They are assumed to be a property of the bulk material and hence must be constant. The concept that tracks may anneal as a whole is ruled out.

1. Introduction

Track annealing studies in different categories of solid state track detectors (SSTD) have been undertaken by various authors [1–9]. Most of the authors consider a latent track to be constituted of many defects aggregated to one single defect, and the kinetics of annealing is described by a Arrhenius relation, $\exp(-E_a/kT)$, where E_a, k and T are the activation energy of annealing, the Boltzmann constant and the annealing temperature, respectively. Previous studies of track annealing have shown that the thermally affected damage trails are healed both radially and axially [3,10,11] and that the extent of healing is a function of both temperature and duration of annealing.

Different interpretations have been given for track density retention in SSTDs with annealing time and temperature. Naeser and Faul [12] gave an explicit relation which expresses the track density retention as a linear decreasing function of logarithm of annealing time. Mark et al. [13] reported an experimentally established fact that logarithm of track retention is a linear decreasing function of time. Mantovani [14] proposed a linear relationship between track retention and heating time for muscovite. Gold et al. [15] have advanced a general reaction rate theory for the annealing process in SSTDs. Recently Mark and Mark [16,17] have proposed that the exact solution for isothermal fission track annealing can be well described as a diffusion process using an infinite series. Cantelaube [9] has tried to extend and extrapolate the Arrhenius law to the case when fading temperature varies over time. Dartyge et al. [18] have suggested different annealing rates for point defects and extended defects in minerals and also a critical temperature around which the annealing rate is much accelerated.

Though these models are frequently used yet their validity has not always been sufficiently proved. The various shortcomings of these track annealing models are listed below:

(i) The Arrhenius equation used for thermal fading of fission tracks is applicable under constant temperature conditions. It necessitates approximations as soon as the fading temperature varies with the time.

(ii) Most models are based on an a priori assumption that the track anneals as a whole. Hence it is not possible to apply these models to correlate the residual lengths or diameters of the partially annealed tracks with annealing temperature and time.

(iii) The activation energy of annealing changes with the degree of track loss which seems to be contradictory. Since there exists a transition line on the Arrhenius plots above which the tracks start fading, it seems justified to assume that activation energy is a constant parameter for a given type of SSTD. This fact is in consonance with the predictions of gap model proposed by Dartyge [19] for annealing of point defects in minerals.

2. Formulation of track annealing model

Considering the discrepancies of previous models, a three step annealing model based on diffusion process is proposed to describe the annealing phenomenon in inorganic SSTDs. The passage of highly ionising particles in SSTD produces interstitial ions due to the Coulomb repulsive force. These interstitial ions diffuse to their normal sites during the isothermal heating of the solid. The three steps of the model are:

(i) Electrons recombine with the ions in the interstitial positions to form neutral atoms.

(ii) Electrostatic and mechanical stresses due to Coulomb forces relax.

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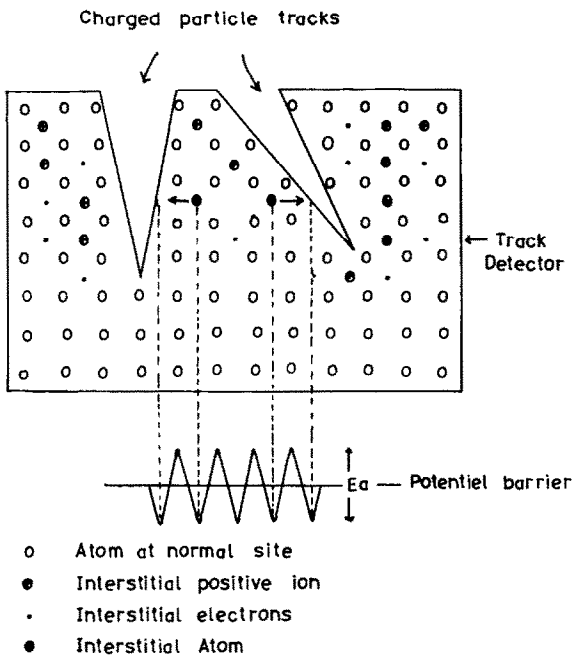


Fig. 1. Annealing of latent damage trails through interstitial diffusion of atoms in inorganic solid state track detectors.

(iii) The neutralized atoms diffuse from their interstitial positions to their normal sites.

During the isothermal heating, the electrons on receiving very small amount of energy, become free and recombine with the ions at the interstitial positions to form neutral atoms; with this the Coulomb stress is over and the only potential barrier for the interstitial atoms to migrate to their normal sites is the local stress presented by its neighbouring atoms (fig. 1). This potential barrier is a measure of the activation energy, E_a, for annealing. Thus if the atom gets this much minimum energy it will be in a position to drift towards the core of the latent track.

We may further infer that:

(i) the activation energy for annealing is that minimum energy which is required for an atom to jump from one interstitial position to another interstitial position. Hence it is a function of the bulk material and is independent of the energy, mass and charge of the traversing particle.

(ii) the use of Boltzmann equation, $t = A \exp(E/kT)$, gives a series of activation energies for different degrees of track loss. However, according to the proposed model, the activation energy is independent of temperature and time of annealing and may depend on a new parameter – track annealing rate.

3. Track annealing rate

To overcome the misconceptions in the use of the Arrhenius equation to explain the annealing results, a new parameter, track annealing rate, V_a, is proposed and introduced into the kinetics of annealing. It is the rate of change of length or diamter, as the case may be, and is given by:

$$V_a = \frac{dD}{dt} \quad \text{for high cone angle glasses} \qquad (1)$$

$$V_a = \frac{dl}{dt} \quad \text{for low cone angle minerals,} \qquad (2)$$

where D is the diameter and l is the length of the etched track. Hence it is proposed that during thermal annealing of tracks, it is the annealing rate and not the activation energy which undergoes change with temperature.

3.1. V_a dependence on temperature

Annealing experiments were carried out to study the effect of temperature on the track annealing rate. Samples were prepared from soda-lime microslide glass (Blue Star, PIC-2). These samples, after washing in alcohol and water, were irradiated with ^{252}Cf fission fragment source in 2π geometry.

Annealing of fission fragment tracks was carried out by heating the irradiated glass samples in Muffle furnace at temperatures 50, 100, 150, 200, 250, 300 and 350°C for an interval of 10 min at each temperature, successively. These samples were etched simultaneously with 1.25% HF for one hour at 17°C after the thermal annealing. Track diameters were measured with projection microscope (MP3, Polish make) using magnification 1000 ×.

Track diameter as a function of annealing temperature is plotted (fig. 2). Track annealing rate, V_a, is calculated using equation (1) assuming that V_a remains

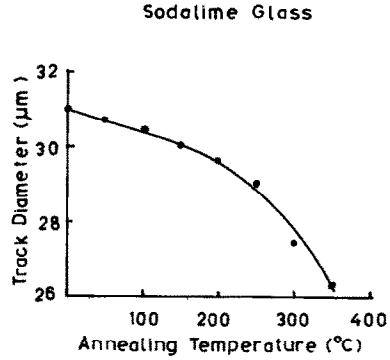


Fig. 2. The variation of mean etch pit diameter with annealing temperature in sodalime glass for heating times 10 min each.

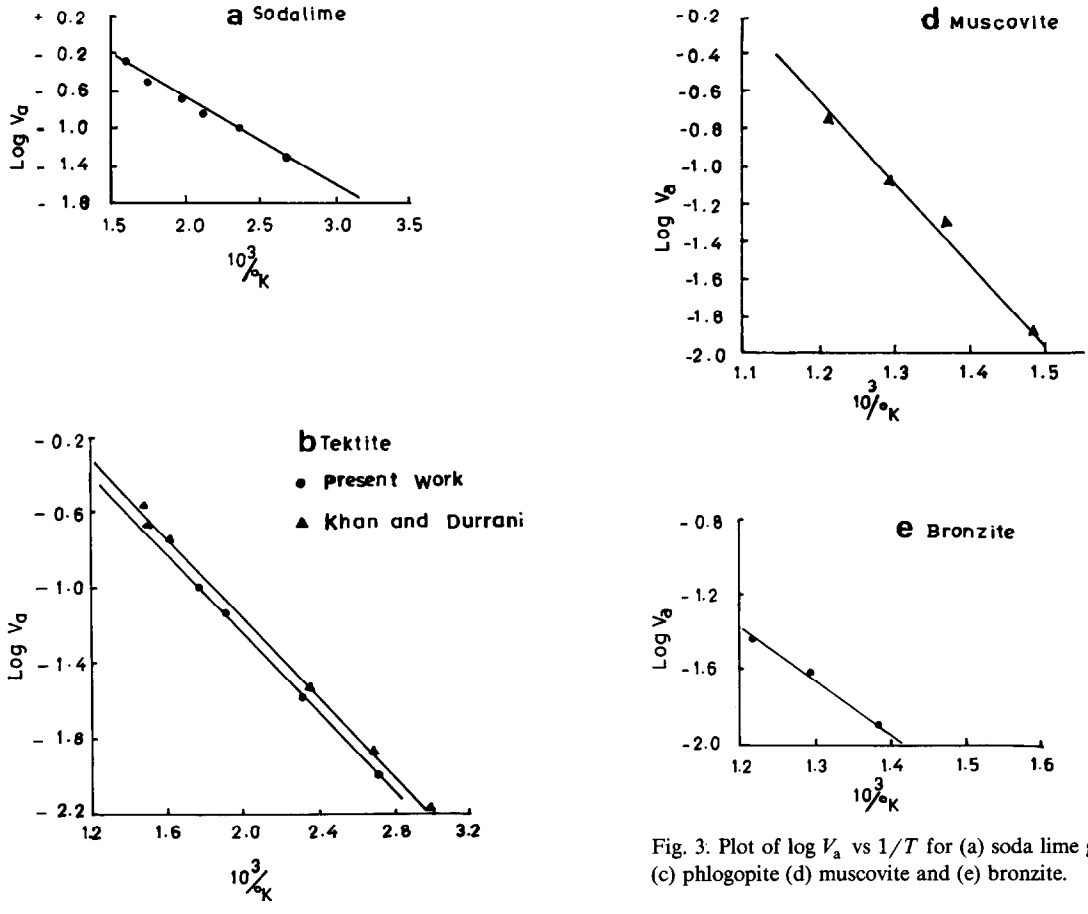


Fig. 3. Plot of log V_a vs $1/T$ for (a) soda lime glass (b) tektite (c) phlogopite (d) muscovite and (e) bronzite.

constant over the first 10 min of heating. Plot of log V_a vs $1/T$ (fig. 3a) shows an exponential dependence of V_a on the annealing temperature for sodaglass detector. The annealing experiments were repeated for tektite and phlogopite samples over the temperature range from 50 to 500°C and 450 to 700°C, respectively, using intervals of 50°C and heating time 10 min for each sample. These results also prove the exponential dependence of V_a on temperature (fig. 3b, c). Exponential dependence of V_a on annealing temperature is also confirmed from the data published by other authors [5,20,21] on tektites, bronzite and muscovite as shown in figs. (3b, d, e).

3.2. V_a dependence on annealing time

Another set of glass samples from Blue Star PIC-2 microslides was prepared to study the effect of annealing time on the track annealing rate. These specimen samples, after irradiation with ^{252}Cf source, were heated in a Muffle furnace at a temperature of 100°C for the intervals 10, 50, 100, 200, 500, 1000, 1500 min, successively. Annealed samples were etched with 1.25% HF for one hour at 17°C. Track diameters were measured

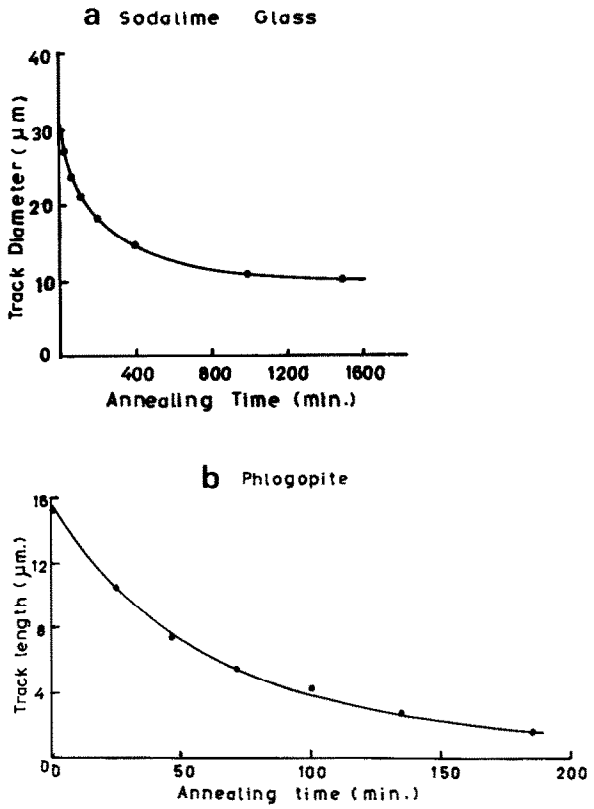


Fig. 4. Variation of mean track diameter with annealing time in (a) soda lime glass at 100°C (b) phlogopite at 500°C.

with projection microscope at total magnification of 1000×. The annealing experiment is repeated for phlogopite at temperature of 500°C for heating intervals of 25, 45, 70, 100, 135 and 180 min, respectively.

Mean track diamters as a function of annealing time are plotted (figs. 4a, b). Annealing rate, V_a, is calculated for different durations of annealing from the plots of track diameter vs annealing time. Plots of log V_a vs log t (fig. 5a, b) reveal that annealing rate follows a power law. This is very well established from the data published by other authors [5,20,21] as shown in fig. (5c, d, e).

4. Empirical formula

As a best fit to the experimental data, a new empirical formula relating annealing rate, V_a, and activation energy, E_a, is proposed as under:

$$V_a = At^{-n} e^{-E_a/kt}, \quad (3)$$

where k is Boltzmann constant, T, the annealing temperature, A, the proportionality constant and n, the exponent of the annealing time, t. The values of A, n and E_a are calculated for the detectors under reference from the respective plots and are summarised in table 1.

Track annealing rate, V_a, is calculated for sodalime glass, tektite, bediasite, bronzite, muscovite and phlogopite using eq. (3) at different temperatures and heating times (tables 2, 3). It is evident that the computed values of V_a for different detectors are in close agreement with the values obtained from the experimental data. Thus the validity of the proposed empirical formula is vindicated.

The problem of the use of rate constants of reaction rate theory to describe the annealing process by Arrhenius equation is solved with the proposed empirical formula. The constants in the empirical formula are independent of temperature and heating time.

5. Activation energy

The activation energies are deduced from the respective plots of log V_a vs $1/T$. The minimum activation energy of 0.16 eV is found in the case of soda lime glass whereas it is of the order of 0.55 eV for tektites and 1 eV for muscovite (table 1). The activation energies deduced from the empirical formula are low as compared to the values already reported by Storzer and Wagner [3], Reimer et al. [7] and Maurette et al. [22] but agree well with the energy of propagation for the interstitial diffusers in the bulk materials. The typical value of energy for interstitial diffusers is of the order of 1 eV and may be as low as 0.1 eV [23]. It has been shown that the energy of movement of an atom for silicon is 0.15 eV [24]. Thus the proposed diffusion model gives satisfactory explanation for the low activation energies obtained using the empirical formula.

It is interesting to note that the activation energy calculated for no track loss (0% reduction curve) for soda lime glass using Arrhenius equation is 0.15 eV [25] and the activation energy obtained from the empirical formula is 0.16 eV. This shows that the minimum energy required for the tracks to start fading in both the cases is nearly the same. Thus the proposed formula seems to be the most appropriate to describe the annealing process in inorganic solids.

6. Partial fading

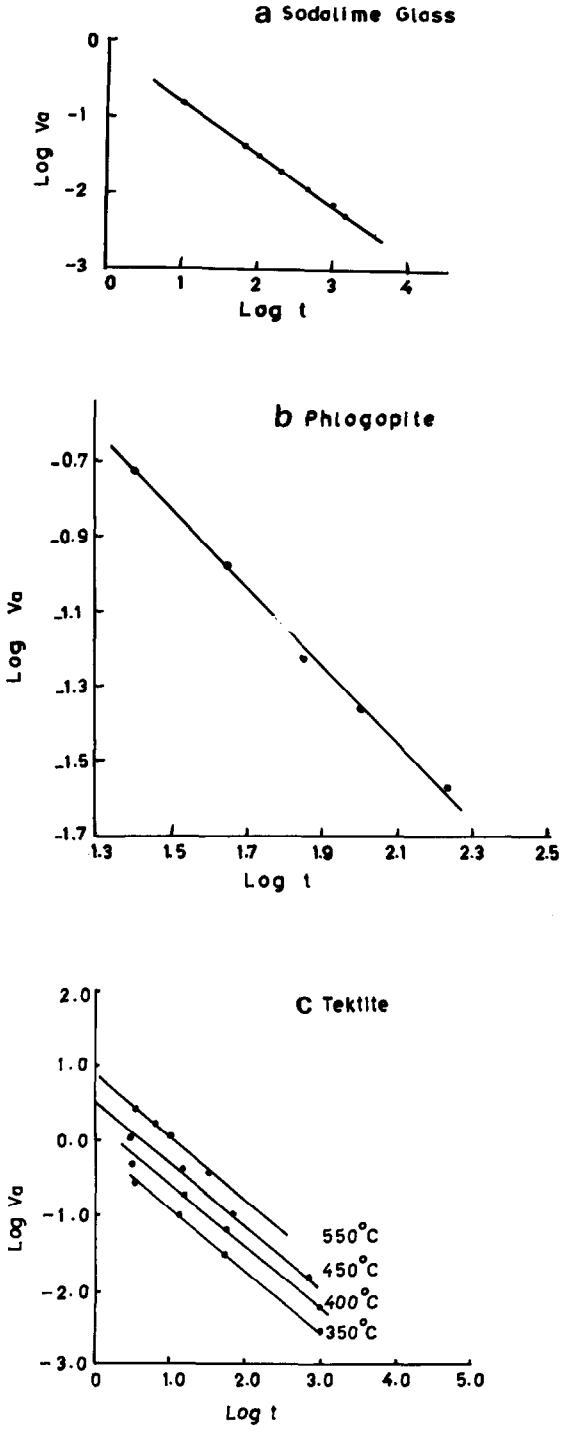
The partial fading of a single track i.e., residual length or diameter, can be well described with the help of the proposed model in the following way:

(i) The ions displaced at large distances, after recombination with electrons, will take more time to migrate to the core of the track as compared to those which lie near the damage trail.

(ii) In the beginning of the annealing process, the

number of interstitial ions is quite large and the probability of atoms to move back to their normal sites is quite high.

(iii) The number of interstitial ions decreases with the duration of annealing which lowers the probability of track healing.



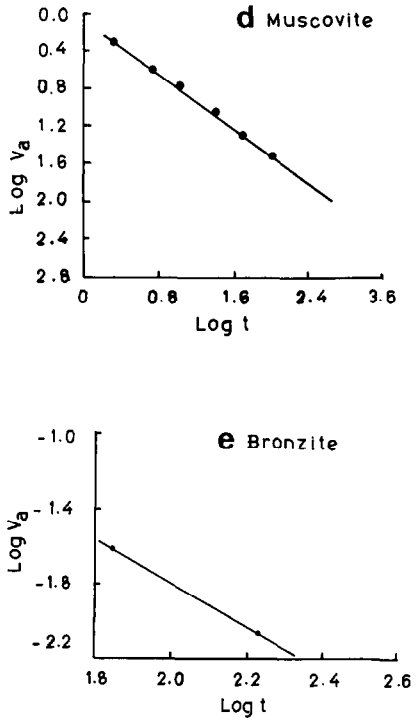


Fig. 5. Plot of log V_a vs log t for (a) soda lime glass (b) phlogopite (c) tektite (d) muscovite and (e) bronzite.

Therefore, the less damaged part i.e., the tail of the track will heal first causing a decrease in length whereas the highly damaged region i.e., the mouth of the track will take longer to heal. In other words, the lengthwise axial annealing rate will be higher than the radial rate along the diameter. Thus the new model successfully explains the partial annealing of tracks and is capable of measuring annealing rate at a given particular temperature and duration of annealing.

7. Ion-explosion model

Study of annealing phenomenon can give us insight into the track formation mechanism in solids. The proposed model and empirical formula lead us to conclude that annealing is a simple diffusion of interstitial ions, displaced during the process of track production by an ion-explosion mechanism, to their normal sites. Hence track annealing is just the reciprocal of track formation by an ion-explosion mechanism.

Table 1
Activation energy, E_a, n value and A value for different track detectors, (See equation (3).)

No.	Detector	Etchant	Activation energy E_a (eV)	n value	A value (μm/min)
1	Soda lime	HF	0.16	0.65	9.56×10^{1}
2	Tektite (Australite)	HF	0.53	0.80	1.93×10^{4}
3	Bediasite	HF	0.55	0.85	1.4×10^{4}
4	Bronzite	NaOH	0.53	1.20	1.1×10^{4}
5	Phlogopite	HF	0.63	1.00	5.85×10^{4}
6	Muscovite	HF	1.00	0.72	6.2×10^{6}

Table 2
Track annealing rate, V_a, as a funtion of temperature and time in soda lime, bronzite and muscovite detectors

Soda lime				Bronzite [a]				Muscovite [b]			
Temp. (°C)	Heating time (min)	Annealing rate V_a (μm/min) Exp.	Cal.	Temp. (°C)	Heating time (min)	Annealing rate V_a (μm/min) Exp.	Cal.	Temp. (°C)	Heating time (min)	Annealing rate V_a (μm/min) Exp.	Cal.
50	60	0.022	0.021	450	70	0.013	0.013	402	60	0.012	0.011
100	60	0.049	0.046	500	70	0.024	0.024	459	60	0.049	0.043
150	60	0.100	0.083	550	70	0.038	0.037	501	60	0.082	0.100
200	60	0.133	0.132	500	170	0.008	0.008	600	100	0.103	0.105
250	60	0.188	0.191								
300	60	0.245	0.261								
350	60	0.338	0.339								

[a] Green and Durrani (1977).
[b] Roberts et al. (1979).

Table 3
Track annealing rate, V_a, as a function of temperature and time in phlogopite and tektite (australite) detectors

Detector	Temperature (°C)	Heating time (min)	Annealing rate V_a (μm/min) Exp.	Cal.
Phlogopite	500	25	0.188	0.185
	500	45	0.104	0.102
	500	70	0.059	0.065
	500	100	0.043	0.045
	500	135	0.034	0.034
	500	180	0.027	0.026
Tektite [a]	300	10	0.071	0.067
	350	10	0.176	0.159
	400	10	0.296	0.330
	450	10	0.496	0.621
	500	10	1.071	1.077

[a] Khan and Durrani (1973).

The authors wish to acknowledge the coöperation of Mr. Amanjit Singh in carrying out the experimental data on phlogopite. The financial assistance rendered by the University Grants Commision, New Delhi and the technical assistance received from Mr. V.P. Gupta and Mr. Narinder Singh are thankfully acknowledged.

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