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TRACK ANNEALING STUDIES IN GLASSES AND MINERALS

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ABSTRACT

An empirical formula relating annealing rate, V_a, and activation energy of annealing, E_a, is used to describe the annealing process in solid state nuclear track detectors and a three step annealing model is proposed. The activation energies for annealing are calculated and found that these vary from 0.16 eV to 1.0 eV for the detectors studied. It is observed that during thermal annealing of tracks, it is the annealing rate and not the activation energy which undergoes change with temperature.

KEYWORDS

Annealing rate; activation energy; interstitial diffusion; partial fading.

INTRODUCTION

Latent radiation damage tracks in various materials begin to fade when subjected to heating (Fleischer and co-workers, 1965). Annealing of latent tracks has considerable influence on the etch rates, etchable range, the critical angle for track registration in bulk materials and fission track ages (Storzer and Wagner, 1969; Khan and Durrani, 1973; Nagpal and co-workers, 1974; Singh and Virk, 1977). Studies on thermal annealing of damaged trails by Gentner and co-workers (1969), Storzer (1970) and Fleischer and Hart (1970) have shown that heating not only results in reduction of track densities but also of track diameters and lengths. Thus the study of annealing phenomenon is important for track analysis and understanding the mechanism of track formation in solid state nuclear track detectors (SSNTDs).

Annealing studies in various inorganic materials have been made by several authors using an Arrhenius type equation (Fleischer and Price, 1964; Neaser and Faul, 1969; Reimer and Wagner, 1972; Modgil and Virk, 1982). Various interpretations have been given for track density retention in SSNTDs with annealing time and temperature.

Neaser and Faul (1969) gave an explicit relation which expresses the track density retention as a linearly decreasing function of log t. It has been later confirmed by other authors (Haack and Potts, 1972; Burchart and co-workers, 1975; Haack, 1976). However, Mark and co-workers (1973) reported an experimentally established fact that logarithm of track retention is a linearly decreasing function of time. They also maintain that the retention lines on the Arrhenius graphs are parallel to each other. Mantovani (1974) proposed a linear relationship between retention and heating time for muscovite.

Gold and co-workers (1981) are of the view that Arrhenius equation should not be used to describe the decrease in track density induced by annealing and

have advanced a general reaction rate theory of annealing process in SSNTDs.

Recently, Mark and Mark (1981), Bertal and Mark (1981/2) proposed that the exact solution for isothermal fission track annealing can be well described as a diffusion process by an infinite series.

Though these models are frequently used yet their validity has not always been sufficiently proved. None of them is able to relate residual lengths and diameters as a function of time and temperature in case of partially annealed tracks. Moreover, the use of Arrhenius equation to describe the decrease in track density and to deduce the activation energy of annealing has been shown to be meaningless (Gold and co-workers, 1981).

FORMULATION OF TRACK ANNEAI ING MODEL

Considering the general discrepancies a three step annealing model based on diffusion process is proposed to describe the annealing phenomenon in glasses and minerals:

a) Electrons recombine with ions in the interstitial positions to form neutral atoms,
b) Readjustment of the electrostatic and mechanical stresses,
c) Diffusion of the atoms from the interstitial positions to their normal sites.

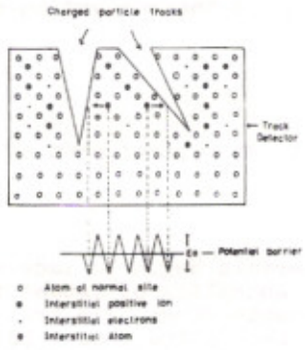

Fig. 1. Annealing of charged particle tracks through interstitial diffusion of atoms in inorganic solid detectors.

The process can be further elaborated as follows:

On receiving very small amount of thermal energy, the electrons recombine with the ions at the interstitial positions to form 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 the measure of the activations energy for annealing. Thus if the atom gets this much minimum energy it will be in a position to drift towards the core of the track.

EMPIRICAL FORMULA AND ITS APPLICATIONS

Based on the proposed model and in confirmity with the experimental observations an empirical formula relating anneal rate, V_a, and activation energy of annealing, E_a, is obtained.

$$V_a = A t^{-n} e^{-E_a/KT} \qquad (1)$$

where K Boltzman constant, T the annealing temperature, A proportionality constant and n the exponent of annealing time, t. The validity of the relation (1) has been tested for the annealing experiments carried out in our laboratory on sodalime glass and using the data on tektites, bronzite and muscovite published elsewhere (Durrani and Khan, 1970; Green and Durrani, 1977; Robert and co-workers, 1979).

The track annealing rates (rate of change of length or diameter) in the SSNTD

of interest as a function of annealing temperature and time are calculated using relation (1). The activation energies are deduced from the respective plots of log V_a vs 1/T (Fig. 2). The minimum activation energy 0.16 eV is found in the case of sodalime glass whereas it is of the order of 0.53, 0.55 and 1.0 eV for tektite and bronzite, badiasite and muscovite respectively (Table 1).

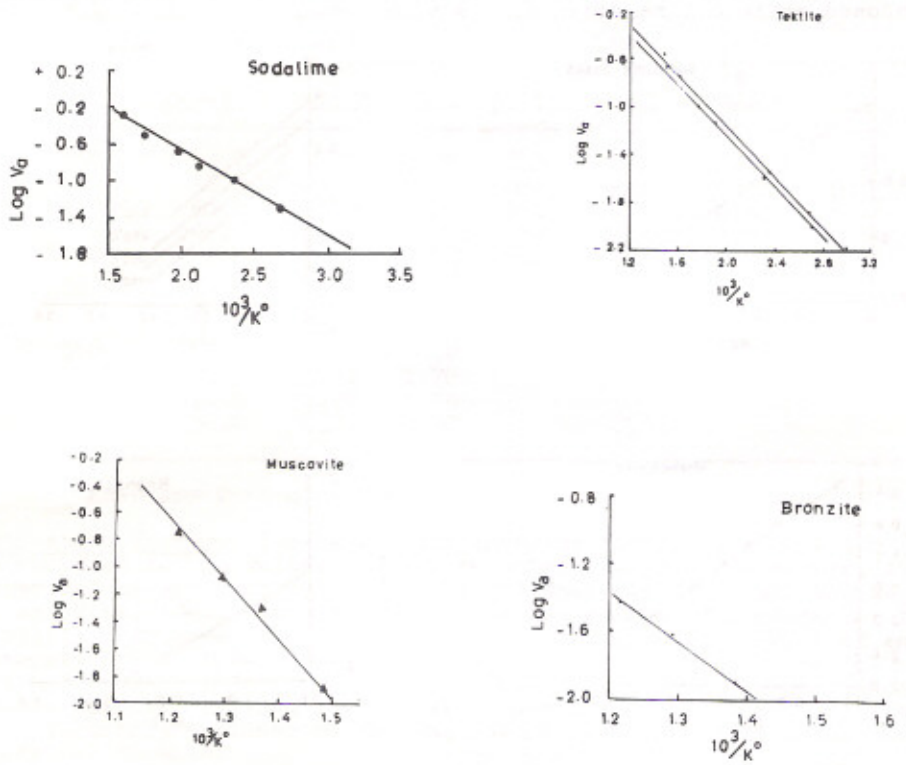

Fig. 2. Plots of log V_a vs 1/T for sodalime, tektite, muscovite and bronzite.

TABLE 1 Activation energy for annealing, n-value and A-value for different track detectors

Detector	Etchant	Activation energy of annealing (eV)	n-value	A-value (μm/min.)
Sodalime	HF	0.16	0.55	9.56×10^{1}
Tektite	HF	0.53	0.80	2.33×10^{2}
Bediasite	HF	0.55	0.85	2.28×10^{2}
Bronzite	NaOH	0.53	1.20	1.10×10^{4}
Muscovite	HF	1.00	0.72	6.20×10^{6}

DISCUSSION AND CONCLUSION

(a) Track Annealing Rate. Since the introduction of SSNTD for fission track dating by Fleischer and co-workers (1969), attempts have been made by several authors (Mark and co-workers, 1973; Burchart and co-workers, 1975; Mantovani,

1974) to explain the annealing experiment results of track retention with annealing time. However, in the present study we have introduced the concept of a single individual track annealing and have found a linearly decreasing relationship between annealing rate and heating time (Fig. 3). It is evident from Table (2) that the computed values of V_a for the SSNTDs are in close agreement with the values obtained from experimental data. Thus the validity of the proposed empirical relation (1) is vindicated.

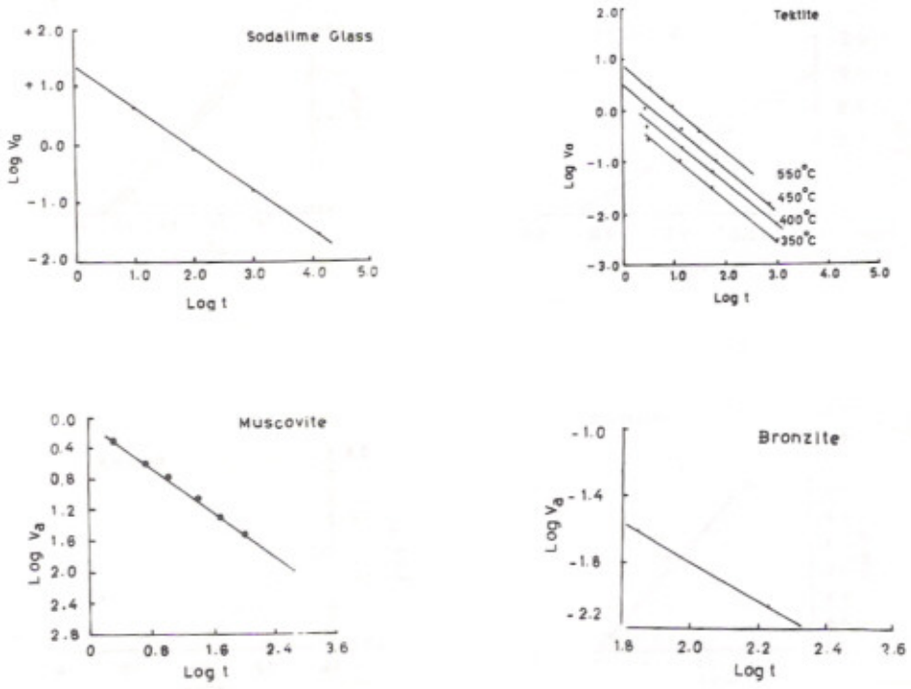

Fig. 3. Plots of log V_a vs log t for sodalime, tektite, muscovite and bronzite.

TABLE 2a Annealing rate as a function of temperature and time in various solid state track detectors

Sodalime				Bronzite				Muscovite			
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								

+ Green and Durrani (1977)
++ Robert and co-workers (1979)

TABLE 2b

Tektite				Badiasite			
Temp.	Heating time	Annealing rate V_a (μm/min.)		Temp.	Heatihg time	Annealing rate V_a (μm/min.)	
(C°)	(hrs)	Exp.+	Cal.	(C°)	(hrs)	Exp.+	Cal.
350	3.2	0.28	0.28	350	4.0	0.11	0.10
	16.0	0.11	0.10	400	4.0	0.21	0.23
	60.0	0.23	0.28	450	4.0	0.45	0.45
	940.0	0.005	0.003	500	4.0	0.61	0.64
450	2.9	0.47	0.53	550	4.0	1.24	1.25
	14. 5	0.21	0.17				
	60.0	0.059	0.066				
	720.0	0.008	0.006				
550	3.1	3.08	3.10				
	6.0	1.90	1.78				
	10.0	1.26	1.24				
	30.0	0.52	0.37				

+ Khan and Durrani (1970)

(b) Activation Energy. The activation energies found using relation (1) are low as compared tc the values already reported (Storzer and Wagner, 1969; Fleischer and co-workers, 1969; Durrani and Khan, 1970; Reimer and Wagner, 1972; Modgil and Virk, 1982) 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 (Elliot and Gibson, 1976). Seitz (1972) have shown that the energy of movement of an atom for silicon is 0.15 eV. Thus the proposed diffusion model gives satisfactory explanation for the low activation energies obtained using the empirical formula.

The parallel lines on the plot of log V_a vs 1/T for tektite at different annealing times show that the activation energy must not change. The proposed model also confirms this fact, which clearly states that the activation energy is a property of the bulk material and must be a constant.

(c) Partial Fading. The partial fading of a single track (residual lengths and diameters) can be well described with the help of this model in the following way.

i) The ions displaced at far distances (after recombination with electrons) will take more time tc migrate to the core of track as compared to those which lie near to the track.
ii) The number of interstitial atoms decreases with the duration of annealing which results in the decrease in the probability of track healing.

Thus the less damaged part of the track will heal first causing a decrease in length whereas the highly damaged region will take much time resulting in the decrease in diameter. Hence the proposed concept that the track anneals as a whole is ruled out. The proposed model and the empirical formula lead us to conclude that annealing is simply diffusion of interstitial ions displaced during the process of track production and the ion-explosion model is the most realistic track formation mechanism in inorganic solids.

The proposed relation holds good for partially annealed tracks and is capable of measuring annealing rate at a particular temperature and duration of annealing. It also clears the misconception of variation of activation energy

with temperature introduced by the use of Arrhenius equation to explain the results of annealing experiments.

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