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FISSION TRACK ANNEALING MODELS AND THE CONCEPT OF A SINGLE ACTIVATION ENERGY

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The annealing behaviour of fission fragment tracks in sodalime glass detector is studied on the basis of three different models. It is observed that all three models yield identical values for the activation energy of annealing even when their mathematical formulations are different. The concept of a single activation energy, proposed earlier (Modgil and Virk, Nucl. Instr. and Meth. B12 (1985) 212) as an intrinsic property of the detector, seems to be fully justified.

1. Introduction

Fission track annealing behaviour has been studied by various authors [1–8] to determine the nature of radiation damage and its healing in a detector, in particular, and for estimation of age corrections to fission track ages of minerals, in general. A number of models [9–12] have been proposed to explain the kinetics of annealing in solid state track detectors (SSTDs) as a function of both heating time and temperature. According to their mathematical formulation, these models are classified into two categories: logarithmic and exponential. The limitations of these models are described elsewhere [13].

Annealing is a diffusion process and the relation between annealing time and temperature is generally expressed by the Boltzmann equation

$$t = A \exp(E_a/kT), \tag{1}$$

where E_a is an activation energy, k the Boltzmann constant and A is a material constant. Most workers have interpreted their annealing experiments on the basis of this equation and obtained a series of fanning lines or Arrhenius plots yielding a spectrum of activation energies for different degrees of annealing. The concept of a single activation energy was first proposed by Modgil and Virk [11] without taking recourse to Arrhenius plots. Green et al. [12] have also rejected the traditional fanning of Arrhenius plots and come out in support of the hypothesis of a single activation energy of annealing. Thus the conceptual difficulty of assigning a set of activation energies for the same SSTD, which is a mere experimental artefact based on Arrhenius approach, is finally removed.

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2. Comparison of annealing models

The empirical formula based on our model [11] relating annealing rate, V_a and activation energy, E_a is proposed as follows

$$V_a = At^{-n} \exp(-E_a/kT), \tag{2}$$

where A is a proportionality constant, n the exponent of annealing time, k the Boltzmann constant, and T the annealing temperature. The annealing rate is defined as the rate of change of length or diameter i.e., dl/dt or dD/dt and can be easily measured (table 1). To determine the activation energy, E_a, eq. (2) can be written in the form:

$$\ln V_a = \ln A - n \ln t - E_a/kT. \tag{3}$$

The special features of this model are that both time and temperature find explicit representation in its formulation, as a consequence, it takes care of variation of annealing rate for all times and temperatures. It also explains the partial fading of tracks as track length is used as a parameter in place of track density. The slope of a plot of $\ln V_a$ versus $1/T$ will give us a unique value for the activation energy.

According to Märk et al. [9], the track density reduction corresponds to track length or diameter reduction to a fair degree of accuracy and the annealing behaviour is given by

$$p(t) = p(0) \exp[-\alpha(T)t], \tag{4}$$

where $\alpha(T)$ is a decay constant (annealing coefficient) given by a sum of two exponentials [14]

$$\alpha(T) = \alpha_{01} \exp(-E_{a1}/kT) + \alpha_{02} \exp(-E_{a2}/kT), \tag{5}$$

where α_{01}, α_{02} are annealing constants and E_{a1} and

E_{a2} are two different activation energies of two different diffusion processes involved. For a high temperature annealing event, eq. (5) can be written in the single exponential approximation

$$\alpha(T) = \alpha_0 \exp(-E_a/kT). \quad (6)$$

Combining eqs. (4) and (6), we obtain

$$\ln(-\ln\, p/p_0) = \ln\, \alpha_0 + \ln\, t - E_a/kT. \quad (7)$$

To a first approximation, Märk et al. [14] assume that

$$p(t)/p(0) \equiv l(t)/l(0) = D(t)/D(0).$$

Therefore, we may write eq. (7) in the form:

$$\ln(-\ln\, D/D_0) = \ln\, \alpha_0 + \ln\, t - E_a/kT. \quad (8)$$

This model finds favour with many experimental groups as it explains the annealing behaviour over a wide spectrum of time and temperature ranges. However, its only shortcoming is that it assumes two sets of activation energies, one at low temperatures and the other for high temperatures.

The model postulated by Green et al. [12] is based on track length measurements of confined tracks and the experimental annealing data is described as a best fit by the equation:

$$\ln\, t - C_1\, \ln(1 - l/l_0) = C_2 + C_3/T, \quad (9)$$

where $C_1 = 4.47$, $C_2 = -17.37$ and $C_3 = 19000$.

Eq. (9) can be modified to explain different modes of annealing–isothermal, isochronal and Arrhenius plots. This model predicts parallelism of lines of equal track length reduction on the Arrhenius plot and hence, as a consequence, a single activation energy of annealing.

The authors of this model are doubtful of its validity in the case of glasses. However, in the present study, the relation [9] is found suitable for a sodalime glass detector in its modified form:

$$\ln(1 - D/D_0) = -C_2/C_1 + 1/C_1 \ln\, t - C_3/C_1 T, \quad (10)$$

$$\ln(dD/D_0) = \ln\, A' + n' \ln\, t - E_A/kT. \quad (11)$$

The plot of $\ln(dD/D_0)$ versus $1/T$ will give the activation energy in terms of the ratio C_3/C_1.

The gap model of Dartyge et al. [10] postulates different annealing rates for radiation damage produced point defects and extended defects in minerals, and a critical temperature around which the annealing rate is much accelerated. As this model assumes an Arrhenius type relation, it will not be useful to consider it here for development of a single activation energy concept.

To sum up, eqs. (3), (8) and (11) represent three different mathematical formulations but the same physical process. Hence it will not be susprising if all three models yield the same value of activation energy in support of the hypothesis of a single activation energy of annealing.

3. Experimental procedure

Annealing experiments were carried out to study the effect of temperature on the size of fission fragment tracks in glass. Samples were prepared from sodalime microslide glass (Blue Star, PIC-2) and irradiated with a ^{252}Cf fission fragment source in a 2π geometry for 2 min each.

Irradiated samples were annealed in a Muffle furnace at temperatures varying from 50 to 450°C, with 10 min hold for each 50°C. All these samples were etched in 2.5% HF at room temperature (31°C) for 35 min, after determination of optimum etching conditions for track revelation in glass. Track diameters (major axis in elliptical tracks) were measured using a Carl Zeiss binocular microscope at a magnification 1000×.

A reference standard was prepared and etched under identical conditions to determine the mean diameter D_0, of unannealed tracks. There is a smooth variation of mean track diameter with temperature as shown in fig. 1. The track annealing rate, V_a, is calculated at different temperatures from the observed mean diameters (table 1).

Table 1
Experimentally determined values of annealing parameters for fission tracks in sodalime glass detector.

Temp. (°C)	Temp. (K)	$\frac{10^3}{T(K)}$	Diameter $D(\mu m)$ [a]	Annealing rate [b]	$\ln V_a$	$\frac{D_0 - D}{D_0}$	$\ln(\frac{dD}{D_0})$	$\frac{D}{D_0}$	$\ln(-\ln\frac{D}{D_0})$
Unannealed	–	–	11.85	–	–	–	–	–	–
50	323	3.09	11.25	0.06	−2.81	0.05	−2.99	0.95	−2.97
100	373	2.68	10.52	0.13	−2.04	0.11	−2.21	0.89	−2.15
150	423	2.36	10.00	0.19	−1.66	0.16	−1.83	0.84	−1.75
200	473	2.11	9.15	0.27	−1.31	0.23	−1.47	0.77	−1.34
250	523	1.91	8.03	0.38	−0.97	0.32	−1.14	0.68	−0.95
300	573	1.74	6.37	0.55	−0.60	0.46	−0.78	0.54	−0.48
350	623	1.60	4.85	0.70	−0.36	0.60	−0.51	0.41	−0.11
400	673	1.48	3.75	0.81	−0.21	0.70	−0.36	0.32	+0.13
450	723	1.38	2.50	0.94	−0.06	0.79	−0.23	0.21	+0.44

[a] $D_0 = 11.85$. [b] $V_a = \mathrm{d}D/\mathrm{d}t$.

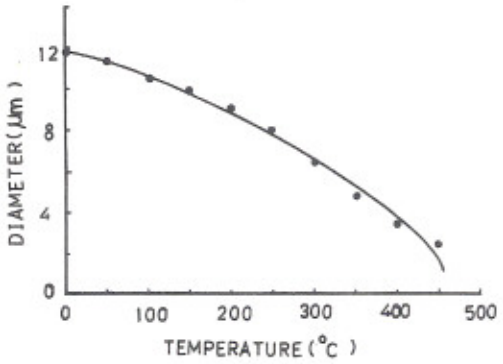

Fig. 1. The variation of mean etch diameter with annealing temperature ranging from 50 to 450°C in sodalime glass for heating times 10 min each.

4. Discussion of results

The results on track annealing in terms of annealing rate, V_a, rate of change of diameter in terms of unannealed diameter, dD/D_0, and ratio of annealed to unannealed diameter, D/D_0, are summarized in table 1. Using eqs. (3), (8) and (11), ln V_a, ln dD/D_0 and ln($-$ln D/D_0) are plotted against reciprocal of annealing temperature ($10^3/T$ K) as shown in figs. 2–4. In all three cases, experimental annealing data show an excellent fit to a straight line. The slope of the lines gives us a measure of the activation energy of annealing.

It is interesting to note that all three models give almost identical values for the activation energy i.e., 0.15 eV each on the basis of Modgil and Virk [11] and Green et al. [12] models, and 0.16 eV according to the Märk et al. [9,14] formulation. These values also correspond to the activation energy of 0.15 eV for no track loss curve (0% reduction) on the Arrhenius plot for

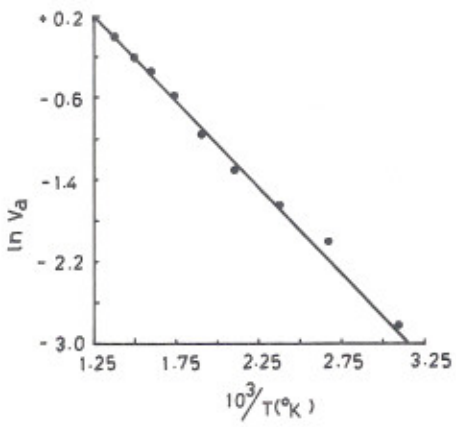

Fig. 2. Plot of annealing rate, ln V_a vs $1/T$ for sodalime glass (eq. (3)).

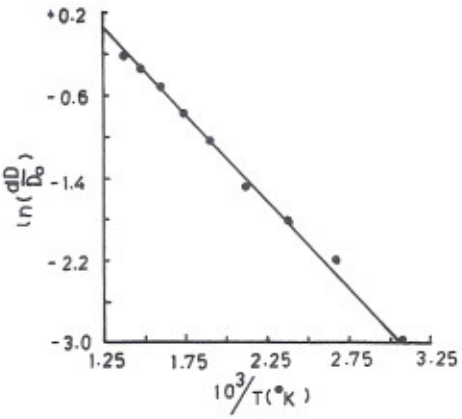

Fig. 3. Annealing results plotted as ln dD/D_0 vs $1/T$ for sodalime glass (eq. (11)).

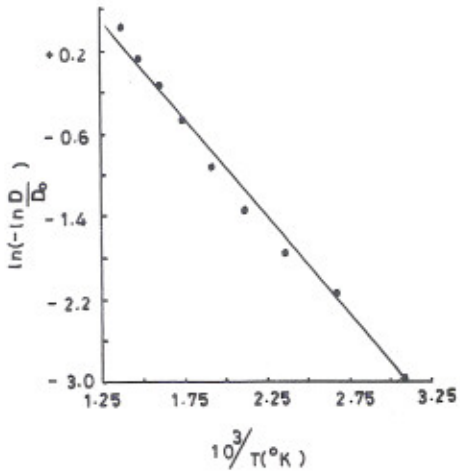

Fig. 4. Annealing results plotted as ln($-$ln D/D_0) vs $1/T$ for sodalime glass (ref. eq. (8)).

sodalime glass [15]. The concept of a single activation energy which is characteristic for a given inorganic SSTD, independent of the nature of the track forming particle, as proposed earlier in our model [11] seems to be finally vindicated. The model is in the process of being tested in case of organic SSTDs and the preliminary results are quite encouraging.

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