ANNEALING CHARACTERISTICS OF HEAVY ION RADIATION DAMAGE IN SSNTDs AND CONCEPT OF SINGLE ACTIVATION ENERGY

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The concept of single activation energy of annealing introduced by Modgil and Virk [Nucl. Instr. and Meth. B12 (1985) 212] explains the annealing behaviour of radiation damage due to heavy ion beams in both organic and inorganic SSNTDs. The values of activation energy, E_a, for soda-lime glass are found to be identical using three different annealing models. Furthermore, the value of E_a is independent of the angle of incidence, the energy and the nature of the ion beam used for irradiation and is solely a detector property. It is interesting to note that track annealing based on length and diameter reductions yields similar values of E_a for soda-lime glass. The activation energy for CR-39 is found to be 0.19 eV using different ion beam irradiations.

1. Introduction

It is remarkable that no single theory has been able to explain satisfactorily the thermal annealing behaviour of latent tracks in both organic and inorganic solid state nuclear track detectors (SSNTDs).

To understand the annealing mechanism a number of models [1–5] were proposed. Mark et al. [1] put forward a model which explains the annealing kinetics by a summation series of exponential decay functions, mathematically represented as

$$p_t = p_0 \exp[-\alpha(T)t], \qquad (1)$$

where p_0 and p_t are density of states at time $t = 0$ and t, respectively; $\alpha(T)$ is a decay constant given by a sum of two exponentials

$$\alpha(T) = \alpha_{01} \exp[-E_{a_1}/kT] + \alpha_{02} \exp[-E_{a_2}/kT], \qquad (2)$$

where α_{01} and α_{02} are annealing constants while E_{a_1} and E_{a_2} are activation energies of two different diffusion processes. According to Mark et al. [1] the second term in eq. (2) may be neglected as its contribution is negligible at high temperature ranges; thus the equation may be written as

$$\alpha(T) = \alpha(0) \exp(-E_a/kT). \qquad (3)$$

Replacing track density ratios, p_t/p_0 by track length ratios, l_t/l_0, or track diameter ratios, D_t/D_0, which is a valid assumption for most of the SSNTDs and substituting the value of $\alpha(T)$, eq. (1) becomes

$$(D_t, l_t) = (D_0, l_0) \exp(-\alpha_0[\exp(-E_a/kT)]t) \quad \text{or} \qquad (4)$$

$$\ln\left[-\ln\frac{(D_t, l_t)}{(D_0, l_0)}\right] = \ln \alpha_0 + \ln t - E_a/kT. \qquad (5)$$

Green et al. [4] tried to understand track behaviour in inorganic solids, particularly in apatite mineral, by using a different approach. Their track length measurements of confined tracks and experimental data are described as a best fit by the expression

$$\ln t - C_1 \ln(1 - l_t/l_0) = C_2 + C_3/T, \qquad (6)$$

where $C_1 = 4.47$, $C_2 = -17.37$ and $C_3 = 19,000$

This model predicts parallelism of lines of equal track length reduction on Arrhenius plot and hence, as a consequence, a single activation energy of annealing. The authors are doubtful of its application in case of glasses. However, in our present work, eq. (6) is found to be suitable in its modified form

$$\ln\left(1 - \frac{(l_t, D_t)}{(l_0, D_0)}\right) = -\frac{C_2}{C_1} + \frac{1}{C_1} \ln t - \frac{C_3}{C_1 T} \quad \text{or} \qquad (7)$$

$$\ln\left(\frac{\mathrm{d}l_t, \mathrm{d}D_t}{l_0, D_0}\right) = \ln A + n \ln t - E_a/kT, \qquad (8)$$

Recently, Modgil and Virk proposed a model [5] relating annealing rate V_a ($\mathrm{d}l/\mathrm{d}t$ or $\mathrm{d}D/\mathrm{d}t$) with the activation energy, E_a as

$$V_a = At^{-n} \exp(-E_a/kT), \qquad (9)$$

where A is a proportionality constant, n, the exponent of annealing time t and k and T have their usual meaning.

Rewriting eq. (9), we have

$$\ln V_a = \ln A - n \ln t - E_a/kT \qquad (10)$$

Our present study is aimed at checking if the three mathematical formulations [eqs. (5), (8) and (10)] yield the same value of activation energy for a given SSNTD. Once having achieved this goal our next step will be to check the validity of the Modgil and Virk model [eq. (9)] in case of organic plastic, CR-39. The results ob-

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tained are encouraging as it appears that once and for all we have achieved an empirical relation which explains the experimental data on annealing in both organic (CR-39) and amorphous inorganic (soda-lime glass) solids.

2. Experimental procedure

Samples of soda-lime glass and CR-39 (Pershore Moulding) plastic were irradiated at the GSI UNILAC facility at Darmstadt using 17 MeV/n [208]Pb ion beam at an azimuthal angle of 45°. The former was also irradiated at angles of 60° and 90° whereas CR-39 was irradiated with other heavy ion beams, viz [139]La (14.6 MeV/n), [238]U (10 MeV/n) and [93]Nb (18 MeV/n) at an angle of 45°. Soda-lime glass was also exposed in our laboratory to fission fragments from [252]Cf source using 2π geometry.
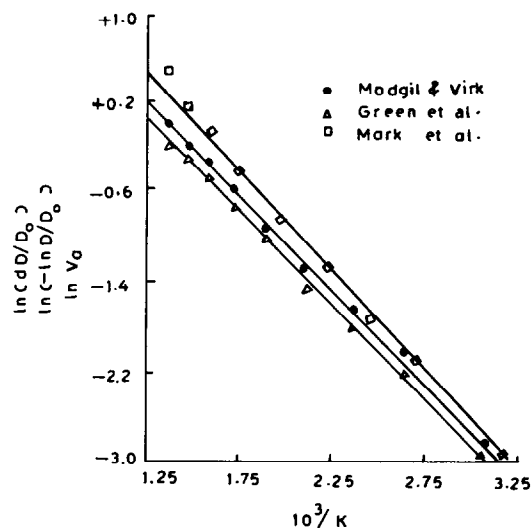

Fig. 1. Plots of $\ln V_a$, $\ln(dD/D_0)$ and $\ln(-\ln D/D_0)$ vs $1/T$ $(10^3/\text{K})$ for soda-lime glass detector using fission fragments.
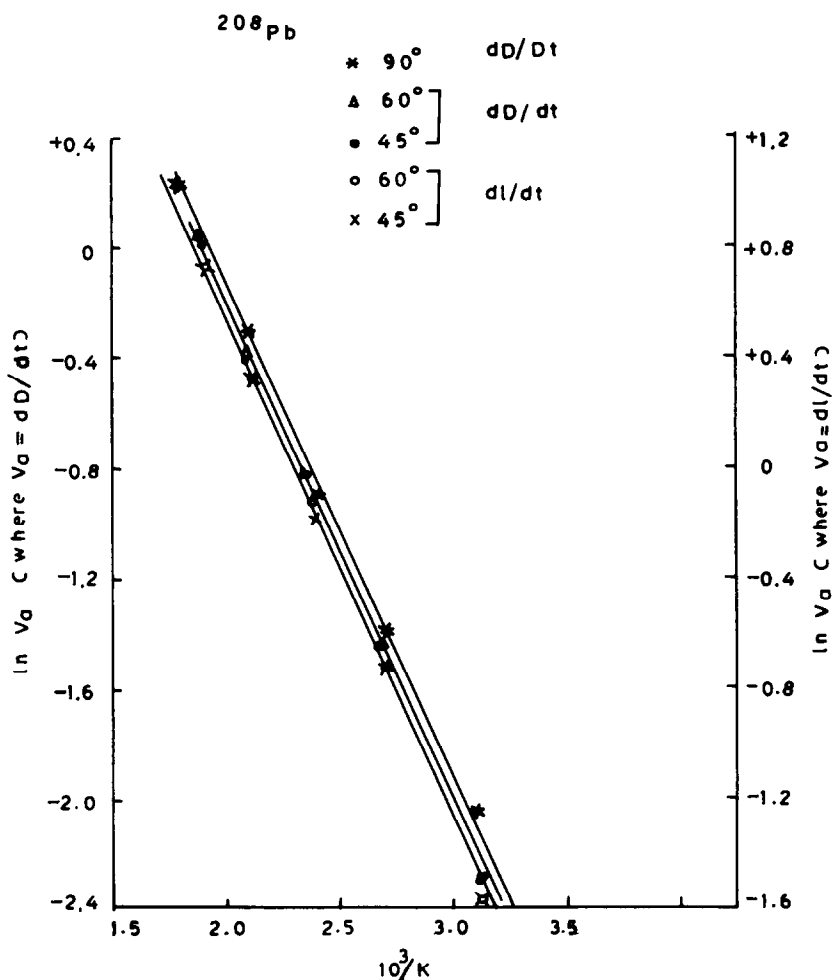

Fig. 2. Plot of $\ln V_a$ vs $1/T$ $(10^3/\text{K})$ for sodalime glass detector using [208]Pb ions.

Isochronal annealing of both the glass and CR-39 samples was carried out for 10 min each between selected ranges of temperatures; whereas for the CR-39 samples additional isothermal annealing for various time was also realised. After annealing, each ion set was etched along with the parent unannealed sample (to ascertain the range, l_0 and mean diameter, D_0) under the optimum etching conditions in 6M NaOH at 70°C for CR-39 and 2.5% HF at 30°C for soda-lime glass.

Measurements of track lengths and diameters (for soda-lime, 90° irradiation only) were recorded by optically scanning each sample with a Carl Zeiss microscope with a resolution of 1 μm. Statistical errors (standard deviation) were applied to these recorded diameters and lengths and their corresponding annealing rates obtained.

3. Results and discussions

The results on track annealing using three different models i.e. annealing rate, $V_a = dD/dt$ (Modgil and Virk, 1985), dD/D_0 (Green et al., 1985) and ratio of annealed to unannealed diameter, D/D_0 (Mark et al., 1973) were obtained for fission fragment irradiated glass. Using eqs. (5), (8) and (10); $\ln V_a$, $\ln dD/D_0$ and $\ln(-\ln D/D_0)$ were plotted against the reciprocal of the annealing temperature as shown in fig. 1. The values of activation energy obtained are recorded in table 1.

The hypothesis that activation energy should solely be a detector property was then checked for different angles using the set of ^{208}Pb ions in glass. From the plots between $\ln V_a$ vs $1000/T$ [K] (fig. 2) the activation energy is calculated. It is observed that the values of activation energy are the same irrespective of the track parameter (length or diameter) chosen. Our formulation yields low value of E_a for glass as compared to values determined by other authors [6–8] using the Arrhenius approach. However, our value agrees well with the energy of propagation for the interstitial diffusers in the bulk materials which may be as low as 0.1 eV [9,10]. We can conclude that although the mathe-
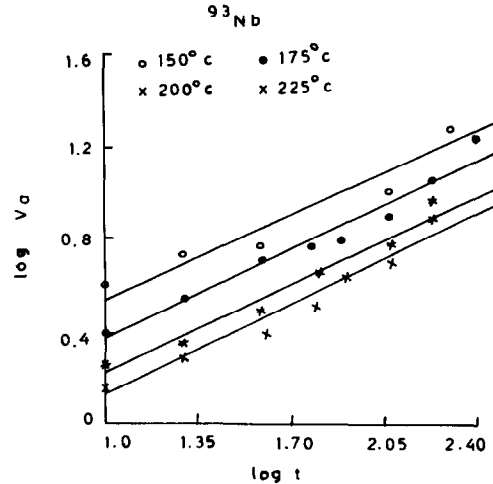

Fig. 3. Plot of log V_a vs log t at different annealing temperatures for 18 MeV/n ^{93}Nb ions in CR-39 plastic.
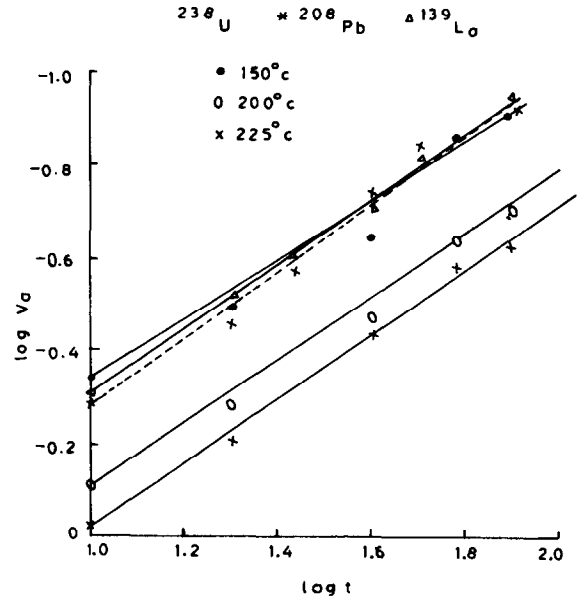

Fig. 4. Plot of log V_a vs log t for determination of n-value in CR-39 plastic using different ions.

Table 1
Identical values of activation energy of annealing for soda-lime glass using three different models and its independence of the angle of incidence

	Fission fragments			^{208}Pb (17 MeV/n)		
	Modgil and Virk	Green et al.	Mark et al.	90°	60°	45°
Activation energy, E_a (eV)	0.15	0.15	0.16	0.16	0.16	0.16

Table 2
The values of n, A and E_a for different ions in CR-39 plastic according to the Modgil and Virk model [5]

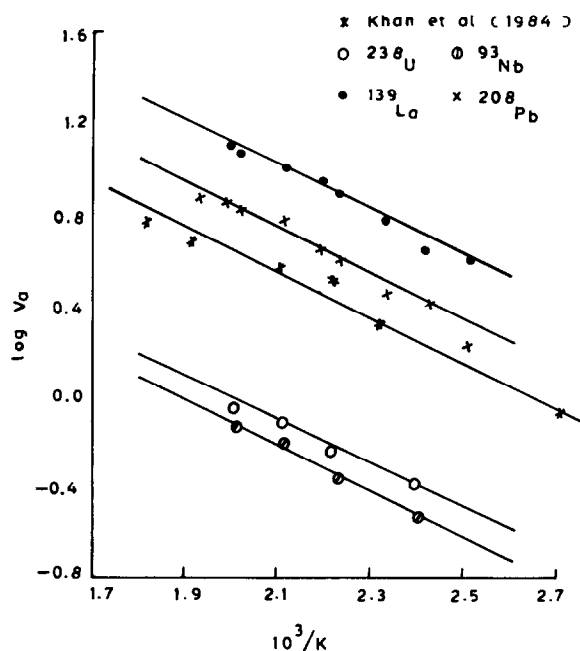| Ion beam	Energy (MeV/n)	n-value	Activation energy E_a (eV)	A-value $T = 200°C$ $t = 10$ min
^{238}U	10.0	0.65	0.185	396.15
^{238}U [a]	16.0	0.66	0.194	–
^{208}Pb	17.0	0.70	0.190	3242.75
^{139}La	14.6	0.68	0.192	50.50
^{93}Nb	18.0	0.61	0.193	245.50

[a] Khan et al. [3].

Fig. 5. Plot of log V_a vs $1/T$ (10^3/K) for different ions in CR-39 plastic for E_a determination.

matical formulation of the three models seems to be different, yet the identical values for activation energy imply that the physical nature of the process involved is the same. It is also evident from fig. 2 that E_a is independent of the angle of incidence.

The data obtained for organic track detector CR-39 is plotted as shown in figs. 3, 4 and 5. Annealing data of Khan et al. [3] for 16 MeV/n ^{238}U beam in CR-39 are also plotted in fig. 5. We observe that the exponent n is independent of the annealing temperature (fig. 3) but it

shows a weak dependence upon the ion beam used (fig. 4). It has also been observed that A [eq. (10)] is a constant dependent upon the stopping power of the ion beam used. The results regarding n, E_a and A values for CR-39 are summarized in table 2.

Our prediction that the activation energy of annealing is a detector property independent of the nature of the etchant, the annealing temperature, the energy and nature of ion beam used, is clearly indicated by the results obtained for both organic as well as inorganic solids. Hence, we may claim with confidence that our single activation energy model holds good for all types of SSNTDs.

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