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HEAVY ION RADIATION DAMAGE ANNEALING IN GLASS DETECTORS

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The effect of radiation damage on track formation in solids is discussed. The annealing behaviour is studied for radiation damage caused by the passage of heavy ions, namely ^{139}La (14.6 MeV/n), ^{208}Pb (17.0 and 13.6 MeV/n) and ^{238}U (15.5 and 5.9 MeV/n), in sodalime glass detectors. The effect of fission fragments on phosphate glass detectors is also discussed. Three different stages of annealing in glass are proposed. A new formulation based on a single activated process for the annealing of heavy ion radiation damage is developed. This relation further supports the concept of a single activation energy of annealing.

1. Introduction

The passage of heavy ionizing charged particles in most insulating solids create continuous narrow trails of radiation damage and therefore establish the entity – a latent track [1]. Various techniques have been employed for the registration of latent tracks [2] and its development by the method of chemical etching [3]. However, the etching of a latent track is as complicated a chemical step in removing ordered and disordered layers in matter as was the original process of registration of the particle track. Chadderton [4] tried to resolve the development of tracks by optical microscopy rather than with the electron microscope during the study of fission track (FT) technique using etched fission fragment tracks.

Although the pace of development is rapid yet the phenomenon of thermal annealing of latent tracks is still not completely understood. There have been numerous investigations about the annealing behaviour of fission tracks in minerals and glasses, but there is no generally accepted description of the annealing kinetics. Here an attempt will be made to analyse the different stages that occur during the healing of solids.

2. Annealing stages

The mechanical and electrical properties have been found to change when insulating solids are bombarded with high energy particles. The displacement of atoms from their normal sites into interstitial positions is believed to be the main cause of these irradiation effects [5]. However, it has been observed that heating tends to restore the original properties of the solids. Therefore, the interstitial atoms tend to return to their normal sites, especially when the insulating solids are heated. We propose to define three different stages mainly responsible for healing of radiation damaged solids. These are

i) electrons recombine with those ions initially located within the damaged region surrounding the interstitial;
ii) recombination of electrons with ions initially located outside the damaged region around the original interstitial;
iii) finally, those electrons which were liberated from their original interstitials will recombine or be captured and start diffusing towards the damaged region.

The above three stages occur simultaneously during isothermal as well as isochronal treatments. Therefore, it has been found that only a single activated process can be isolated. The two parameters mainly responsible for understanding the annealing mechanism are track reduction rate and track etch rate. The track reduction rate is used to formulate a modified model for calculating the activation energy.

3. Activation energy of annealing

For a single activated process, the annealing kinetics are discussed within the framework of the two most commonly employed types of data, i.e., those obtained from isochronal and isothermal anneals. It has been assumed that the experimental technique measures the concentration of defects, N, after a fixed time at different temperatures (isochronal) or as a continuous function of time at a constant temperature (isothermal). The

most general chemical rate equation for a simple decay process is given by [6],

$$\frac{dN}{dt} = N^n K, \tag{1}$$

where N is the fractional concentration of the defect, K, a rate constant dependent on the temperature T in degrees kelvin and n, the order of the reaction. In general, the rate constant K is assumed to be given by the Arrhenius expression:

$$K = K_0 \, e^{-E/kT}, \tag{2}$$

where E is an activation energy, and k, Boltzmann's constant. If a discrete activation energy were involved and first order kinetics are assumed, the activation energy could be determined from eq. (2). Garlick et al. [7] also provide a method of analysis for determining both the activation energy and the order of the reaction n from isothermal and isochronal annealing data – assuming that the process is characterized by a unique activation energy.

In order to best fit the experimental data, a new empirical formula was proposed [8] relating the annealing rate V_a (defined as the rate of change of length for low cone angle etch pits and diameter for high cone angle etch pits) and activation energy E as follows:

$$V_a = A \, t^{-n} \, e^{-E/kT}, \tag{3}$$

where k is Boltzmann's consant, T, the annealing temperature, A, the proportionality constant and n, the exponent of annealing time, t.

Equation (3) can be rewritten as

$$\ln V_a = \ln A - n \ln t - E/kT. \tag{4}$$

The activation energy can be calculated from a plot of $\ln V_a$ against inverse annealing temperature.

4. Modified formula

Incorporating the errors in fitting the annealing data, it has been realised that the above formulation [eq. (3)] has a number of drawbacks. The chief shortcomings are the following:

i) the empirical relation uses only the approximate derivative i.e. $\Delta D/\Delta t$ and not the real one, dD/dt;
ii) this formula gives the change in track diameter $(D - D_0)$ and not the reduction in track diameter D/D_0 as supported by various authors [9–11].
iii) finally, differential equations always have larger errors as compared with integral equations.

We have attempted to remove these drawbacks in the above formulation and to make a best fit. To make a formulation, we modify eq. (3) in the following way:

$$dD/dt = At^{-n} \, e^{-E/kT}.$$

Integrating both sides, we get

$$\int_{D_0}^{D} dD/dt \, dt = A \, e^{-E/kT} \int_0^t t^{-n} \cdot dt \quad \text{or}$$

$$D - D_0 = A \frac{t^{1-n}}{1-n} \, e^{-E/kT}, \tag{5}$$

where D_0 is the diameter of unannealed tracks.

On simplification, eqn. (5) becomes

$$1 - \frac{D}{D_0} = A' \left(t^{1-n}/1 - n \right) e^{-E/kT}, \tag{6}$$

where A' is another constant or

$$1 - r = A' \left(t^{1-n}/1 - n \right) e^{-E/kT}, \tag{7}$$

where $r = D/D_0$, the track diameter reduction.
We can rewrite eq. (7) as

$$\ln(1 - r) = \ln A' + \ln\left(t^{1-n}/1 - n \right) - E/kT. \tag{8}$$

The above relation is used to calculate the activation energy, E from a plot of $\ln(1 - r)$ against inverse temperature for a constant time interval.

The relation (7) is also true for low cone angle etch pits where $r = l/l_0$, the track length reduction and l_0 is the unannealed length.

Thus, a new formulation has been developed which removes the drawbacks of Modgil and Virk formulation [8] and shows similarity with the super model approach of Laslett et al. [12].

This formulation is tested for annealing experiments in sodalime and phosphate glass detectors to corroborate the hypothesis that the process is characterized by a single activation energy.

5. Experimental procedure

Glass samples of sodalime (microscopic slide) were irradiated with heavy ions, namely ^{139}La (14.6 MeV/n), ^{208}Pb (17.0 and 13.6 MeV/n) and ^{238}U (15.5 and 5.9 MeV/n) at the GSI heavy ion accelerator, UNILAC. The phosphate glass samples, commercially known as LG-700, and obtained from Schott Glass Technologies, USA, were irradiated with fission fragments from a ^{252}Cf in 2π geometry.

Irradiated samples of sodalime glasses were annealed in a muffle furnace at temperatures of 100, 150, 200 and 250 °C and for time intervals of 10, 20, 40 and 80 min. These samples, for each ion set, were etched along with the parent unannealed samples in 2.5% HF for 35 min at room temperature 30 °C. This etching condition is employed after determining the optimum etching conditions for track revelation in sodalime glass detectors. Here, the dilute etchant is preferred for glass detectors in comparison to concentrated etchants for obvious reasons [13]. All these samples were, then, washed in running water.

The irradiated samples of phosphate glass detectors were annealed isochronally from 400 to 650 °C with a 30 min hold for each 50 °C step. The etching of phosphate glass was carried out in 40% HF for 1 h at room temperature (30 °C) and then washed thoroughly in running water.

The mean observed track length (l') and diameter (D) were measured using a Carl Zeiss microscope with a resolution of 1 μm. The actual mean lengths were calculated using the relation $l = l'/\cos\theta$, where θ is the angle of incident beam with respect to the detector surface. Statistical errors were also applied to these measurements for each ion set and conform to a standard deviation of 1σ.

The mean lengths, l_0 and diameter, D_0 of unannealed tracks were also determined under identical conditions. The track diameters of only circular tracks were considered in phosphate glasses while dislocations and inhomogeneities were ignored.

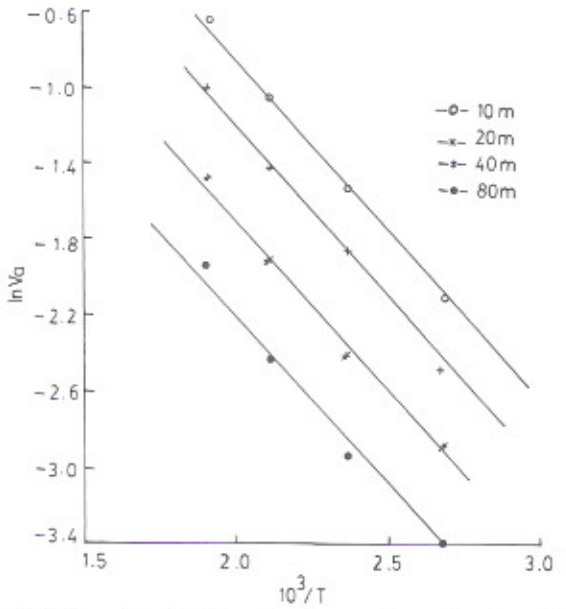

Fig. 1. The plot of ln V_a against inverse temperature in sodalime glass detector using ^{208}Pb (17.0 MeV/n) ion beam.

6. Results and discussion

The best fitting of any curve has been a subjective matter especially when the number of data points are few. Therefore, the method of least squares was used in plotting the best fit lines.

The annealing results for V_a (calculated as the rate of change of diameter) and the track diameter reduction, r are summarized in table 1. Fig. 1 shows the annealing results for only ^{208}Pb ion beam in sodalime glass using the Modgil and Virk formulation. The results for other ion beams are given in table 2, as they show correspondence with ^{208}Pb ion beam. The activation energy calculated from the slope of the plot of ln V_a against inverse temperature is found to be 0.16 eV.

The next plot (fig. 2) shows the annealing results using our modified formula for ^{208}Pb ion beam in sodalime glass. The value of activation energy calculated from the slopes of the plot of $\ln(1-r)$ against inverse temperature comes out to be 0.16 eV. The comparison of activation energies using both the formulations for different penetrating beams in sodalime glass is shown in table 2.

The results of fission fragment tracks in phosphate glasses are calculated for isochronal annealing. The

Table 2
The values of the activation energy for different ion beams using both the formulations

Ion beams used (MeV/n)	Activation energy (eV)	
	Modgil and Virk Formula	Modified Formula
^{208}Pb (17.0)	0.16 ± 0.005	0.16 ± 0.001
^{208}Pb (13.6)	0.15 ± 0.006	0.16 ± 0.002
^{139}La (14.6)	0.16 ± 0.005	0.16 ± 0.002
^{238}U (15.5)	0.16 ± 0.004	0.16 ± 0.001
^{238}U (5.9)	0.15 ± 0.006	0.16 ± 0.002

Table 1
The annealing data for V_a and r using both the formulations for ^{208}Pb (17.0 MeV/n) ion beam in sodalime glass detector. Annealing time = 10 min.

Sample no.	T (K)	$\frac{10^3}{K}$	Mean observed diameter D (μm)	Annealing rate, $V_a = \frac{dD}{dt}$	Track diameter reduction $r = D/D_0$	ln V_a	$\ln(1-r)$
1	Unannealed	–	18.00 ± 0.2 [a]	–	–	–	–
2	373	2.68	16.75 ± 0.15	0.12	0.93	−2.12	−2.66
3	423	2.36	15.75 ± 0.15	0.22	0.88	−1.51	−2.12
4	473	2.11	14.50 ± 0.12	0.35	0.81	−1.05	−1.66
5	523	1.91	12.80 ± 0.15	0.52	0.71	−0.65	−1.24

[a] D_0.

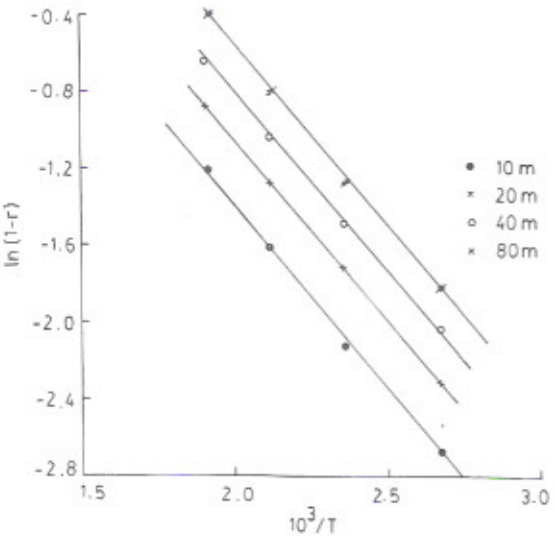

Fig. 2. The plot of $\ln(1-r)$ vs inverse temperature in sodalime glass detector using ^{208}Pb (17.0 MeV/n) ion beam.

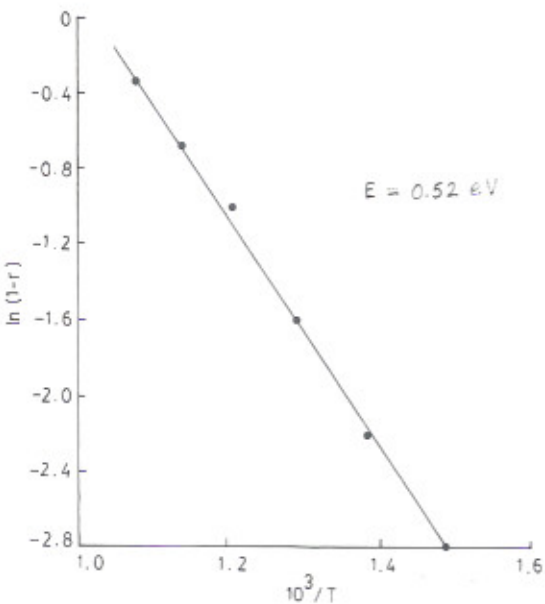

Fig. 3. The plot of $\ln(1-r)$ vs inverse temperature in phosphate glass detector irradiated with fission fragments.

activation energy is calculated using the modified relation (8) by plotting $\ln(1-r)$ against inverse temperature (fig. 3) and found to be 0.52 eV.

The results obtained from our modified formula show minimum deviation. Therefore, we conclude that while constructing empirical models to be used as a basis of prediction, simple models with fewer fitted parameters are generally preferable. This model further supports the concept of a single activation energy for annealing and removes the the ambiguities of the earlier approach adopted in our laboratory to understand annealing kinetics in SSNTDs.

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