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Thermal Annealing of Heavy Ion Latent Tracks in Soda and BP-1 Phosphate Glasses

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Annealing behaviour of latent tracks in soda glass has been studied in the temperature range 100–300°C for different time intervals. These latent tracks are created by irradiating these samples with heavy ions viz., ^{238}U (11.4 MeV/u) and ^{132}Xe (11.4 and 5.9 MeV/u) from GSI Darmstadt and ^{56}Fe (4.0 MeV/u) and ^{48}Ti (4.0 MeV/u) from JINR, Dubna. A similar study has been made on Barium Phosphate glass (BP-1) irradiated with ^{238}U (11.4 MeV/u) and fission fragments of ^{252}Cf. The 'Single Activation Energy' model proposed by Modgil and Virk (1985) is used as an empirical approach for explaining the thermal fading of these latent tracks. This model always yields a unique value of activation energy of annealing and which is independent of the nature of the ion beam and the degree of annealing. In comparison to this model, Mark *et al.* (1973) and Green *et al.* (1985) approach on annealing phenomena has also been used to determine the activation energy. The value of activation energy for annealing of these heavy ion latent tracks is found to be 0.16 and 0.41 eV in soda and BP-1 phosphate glass, respectively. The results of 'Single Activation Energy' model are compared with those obtained by using models proposed by Mark *et al.* (1973) and Green *et al.* (1985). © 1997 Elsevier Science Ltd. All rights reserved

Introduction

The passage of heavy ion charged particles through most insulators like glasses, minerals and polymers leads to the formation (at the microstructural level) of the narrow region of radiation damage, known as latent track, where physical and chemical properties of the material are changed. These latent tracks undergo annealing under the heating environment. The phenomenon of thermal annealing of these latent tracks in nuclear track recording materials has been studied by many authors during the last three decades. However, due to the lack of a satisfactory theory of track formation in different materials, the phenomenon of thermal annealing of these latent tracks is not fully understood. The most successful latent track recording models apply to the minerals and the glasses and are related to the ion-explosion spike model. The track fading occurs as interstitially displaced atoms thermally penetrate the activation barrier to recover their initial lattice positions. Many of the authors consider a latent track as an aggregate of defects and kinetics of annealing is described by 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 (Fleischer *et al.*, 1975). It has been found that the activation energy associated with the repair of a damaged material is of the order of a few electron volts, which is typical energy involved in atomic diffusion. Hence it is assumed that the annealing of damaged materials occurs due to diffusion of defects in inorganic materials and movement of molecular fragments within a polymer. The interstitial atoms tend to return to their normal sites and the broken molecular chains may join and recombine with other active species when insulating solids are heated. During the heating, the electrons on receiving very small amount of energy, become free and recombine with the ions at the intersitial 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 neighbourhood atoms. This potential barrier is a measure of the activation energy, E_a, for annealing. The phenomenon of annealing of radiation damage in different dielectrics has been studied by several researchers (Fleischer and Price, 1964; Naeser and Faul, 1969; Khan *et al.*, 1984; Mark *et al.*, 1973; Dakowski *et al.*, 1974; Burchart *et al.*, 1979; Mark *et al.*, 1981; Gold *et al.*, 1981; Dartyge *et al.*, 1981; Modgil and Virk, 1984, 1985; Green *et al.*, 1985; Laslett *et al.*, 1986; Virk, 1991) and a number of empirical formulations have been proposed to explain the annealing mechanism as a function of both annealing time and temperature.

Mark *et al.* (1973) proposed that the annealing of fission tracks in apatite material can be explained by a summation series of exponential functions and suggested that the annealing behaviour of track

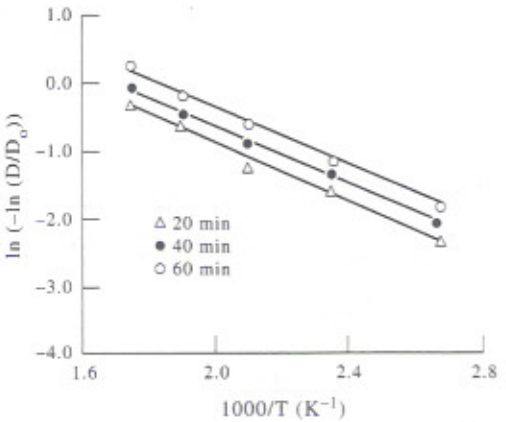

Fig. 1. The variation of ln (− ln (D/D_o)) vs $1/T$ (10^3 K^{-1}) for the annealing of tracks of ^{132}Xe (5.9 MeV/u) ion in soda glass.

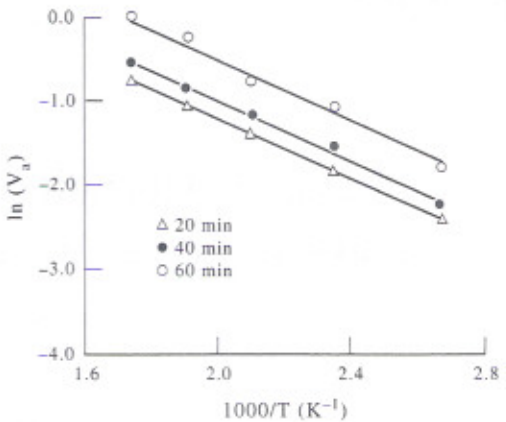

Fig. 3. The variation of ln (V_a) vs $1/T$ (10^3 K^{-1}) for the annealing of tracks of ^{132}Xe (5.9 MeV/u) ion in soda glass.

densities (ρ) in apatite could be explained by relationship

$$\rho(t) = \rho(\mathrm{o}) \exp(-\alpha(T)t) \qquad (1)$$

where $\rho(t)$ is the track density at annealing time t and $\rho(\mathrm{o})$ is the original track density. In this case the annealing coefficient $\alpha(T)$ is given by a sum of two exponentials

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

where α_{01} and α_{02} are annealing constants and E_{01} and E_{02} are the activation energies of two diffusion processes involved. However, at elevated temperature annealing, the data of Mark *et al.* (1981) could be approximated with a single exponential decay function. The two exponential equation (2) can be reduced to a single exponential approximation as

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

Using the above relation, equation (1) can be written as

$$\ln(-\ln r) = \ln \alpha_o - E_a/kT + \ln t \qquad (4)$$

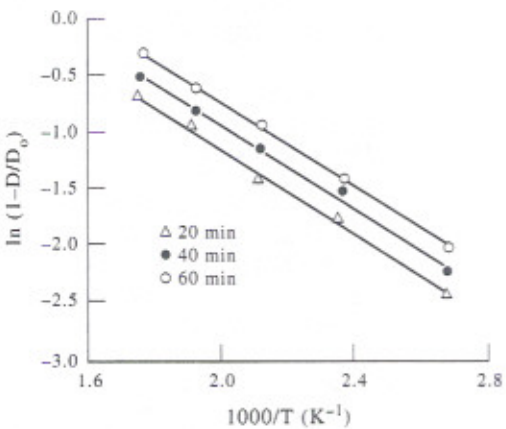

Fig. 2. The variation of ln ($1 - D/D_o$) vs $1/T$ (10^3 K^{-1}) for the annealing of tracks of ^{132}Xe (5.9 MeV/u) ion in soda glass.

where $r = \rho(t)/\rho(\mathrm{o})$ is the track density reduction ratio.

Mark *et al.* (1981) assumed the validity of $\rho/\rho_o \approx l/l_o \approx D/D_o$ (for apatite). Hence equation (4) to a first approximation for glass, reduces to

$$\ln(-\ln(D/D_o)) = \ln \alpha_o + \ln t - E_a/kT\,. \qquad (5)$$

A plot between ln (− ln (D/D_o)) vs ($1/T$) produces lines of slope E_a/K. This model has been used to explain the annealing of tracks in different materials. However, its shortcoming is the hypothesis of two activation energies for the annealing process.

Green *et al.* (1985) postulated a model describing the thermal annealing of tracks in inorganic solids, by using a different approach for presenting their experimental data through a best fit

$$\ln t - C_1 \ln(1 - l/l_o) = C_2 + C_3/T \qquad (6)$$

where C_1, C_2 and C_3 are constants which may be determined from the experimental data. This model predicts parallelism of lines of equal track length reduction on the Arrhenius plot and therefore a single activation energy can be determined. This equation is modified as

$$\ln(1 - D/D_o) = -C_2/C_1 + (1/C_1)\ln t - (C_3/C_1)/T \qquad (7)$$

$$\ln(1 - D/D_o) = \ln A_1 + n_1 \ln t - E_a/kT \qquad (8)$$

where $\ln A_1 = -C_2/C_1$, $n_1 = 1/C_1$ and $E_a/k = C_3/C_1$.

Modgil and Virk (1984, 1985) proposed a three step annealing model based on diffusion processes. The empirical formulation relating the annealing rate, V_a (rate of change length, l, or diameter, D with annealing time, $\mathrm{d}l/\mathrm{d}t$ or $\mathrm{d}D/\mathrm{d}t$) with the activation energy, E_a is given as

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

where A is a proportionality constant, n is the exponent of annealing time t and other symbols have their usual meanings. In order to determine the activation energy the equation can be rewritten as

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

Table 1. Activation energy values calculated from three different formulations for annealing of tracks of different heavy ions in soda glass material

Ion	Energy(MeV/u)	Activation energy of annealing (eV)		
		Mark *et al.* (1973)	Green *et al.* (1985)	Modgil and Virk (1985)
^{238}U	11.4	0.18 ± 0.002	0.16 ± 0.060	0.16 ± 0.003
^{132}Xe	11.4	0.18 ± 0.007	0.16 ± 0.004	0.16 ± 0.006
^{132}Xe	5.9	0.18 ± 0.007	0.16 ± 0.004	0.16 ± 0.006
^{56}Fe	4.0	0.20 ± 0.062	0.16 ± 0.005	0.16 ± 0.005
^{48}Ti	4.0	0.19 ± 0.050	0.16 ± 0.002	0.16 ± 0.004

This model favoured the concept of a single activation energy of annealing for all heavy ion and fission fragment damaged materials. In the present study, equations (5), (8) and (10) have been used for the determination of activation energy for annealing of heavy ion latent tracks in soda and BP-1 phosphate glasses.

Experimental Procedure

The samples of sodalime glass were irradiated by different heavy ion beams, viz., ^{238}U (11.4 MeV/u) and ^{132}Xe (11.4 and 5.9 MeV/u) from the UNILAC accelerator at GSI, Darmstadt, Germany and ^{56}Fe (4.0 MeV/u) and ^{48}Ti (4.0 MeV/u) from JINR, Dubna, Russia having a fluence of 10^4 ions/cm^2. All these irradiations were made at an angle of 90° with respect to the sample surface. The irradiated samples were cut into small pieces and annealed in a Solo furnace at different temperatures of 100, 150, 200, 250 and 300°C for time intervals of 20, 40 and 60 min, respectively. These annealed samples, for each ion and energy set, were etched along with the unannealed sample in an etchant, 48 vol.% HF + 96 vol.% H_2SO_4 + H_2O in the ratio 6:1:18, at a constant temperature of 40°C for 5 min. These etched samples were washed in running water and dried in folds of a tissue paper. The etched and dried samples were scanned using a Carl Zeiss binocular optical microscope at a magnification of 1000X. The tracks are circular in shape because the angle of incidence of the heavy ions is 90° with respect to the sample surface. The mean value of the track diameter is determined by measuring the diameter of a number of tracks. Similarly the samples of Barium Phosphate glass (BP-1) (Wang *et al.*, 1988) were irradiated by ^{238}U (11.4 MeV/u) heavy ion from the GSI at an angle of 45° with respect to the surface of the detector and by fission fragments of ^{252}Cf source at 90°. These irradiated samples were cut into small pieces and annealed at temperatures of 300, 350, 400, 450°C for 5 and 10 h in case of ^{238}U (11.4 MeV/u) and for 1 h in case of fission fragments. These annealed samples were etched in 48 vol.% HF solution at 50°C temperature for 30 min in case of ^{238}U and for 4 h in case of fission fragments. The value of mean track length in case of ^{238}U (11.4 MeV/u) and track diameter for fission fragment tracks in BP-1 glass is measured in the similar way by using the Carl Zeiss binocular microscope.

Results and Discussion

Figures 1 – 3 show how to calculate the activation energy of annealing by using least square fit method from different formulations, viz., Mark *et al.* (1973), Green *et al.* (1985), and Modgil and Virk (1985), respectively. The activation energies are calculated from the slopes of the plots of left hand sides of equations (5), (8), and 10 against the inverse temperatures in all the three cases. The values of activation energy of annealing calculated from

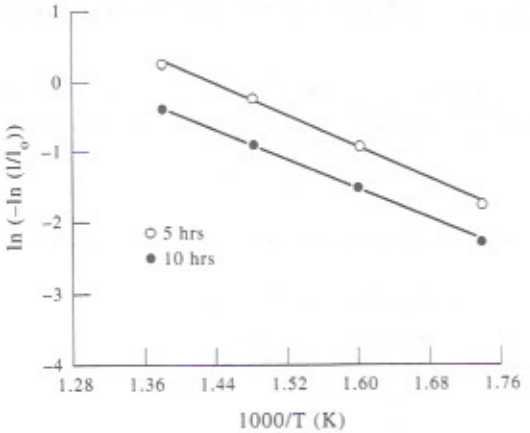

Fig. 4. The variation of ln (− ln (l/l_o)) vs $1/T$ (10^3 K^{-1}) for the annealing of tracks of ^{238}U (11.4 MeV/u) ion in BP-1 phosphate glass.

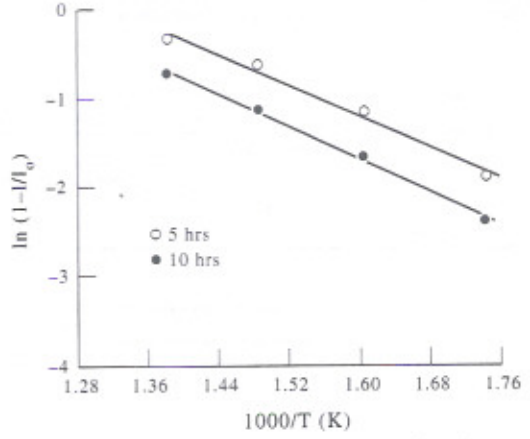

Fig. 5. The variation of ln (1 − l/l_o) vs $1/T$ (10^3 K^{-1}) for the annealing of tracks of ^{238}U (11.4 MeV/u) ion in BP-1 phosphate glass.

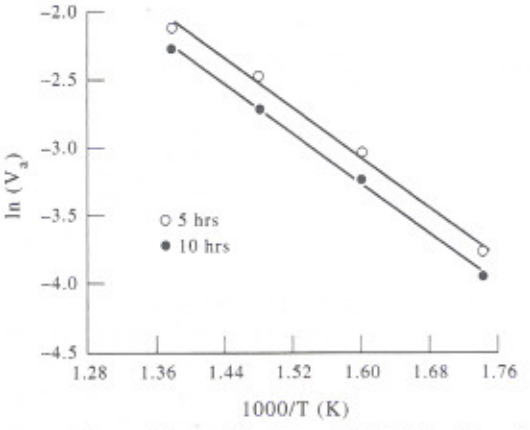

Fig. 6. The variation of ln (V_a) vs $1/T$ (10^3 K^{-1}) for the annealing of tracks of ^{238}U (11.4 MeV/u) ion in BP-1 phosphate glass.

different formulations for different heavy ions in soda glass are presented in Table 1. Annealing models of Green *et al.* (1985) and Modgil and Virk give the identical values of activation energy of annealing (0.16 eV) for tracks of different heavy ions, ^{238}U (11.4 MeV/u), ^{132}Xe (11.4 and 5.9 MeV/u), ^{56}Fe (4.0 MeV/u) and ^{48}Ti (4.0 MeV/u) in soda glass. These values are in perfect agreement to the calculated values of Singh and Virk (1990) for annealing of heavy ion tracks ^{139}La (14.6 MeV/u) and ^{208}Pb (17.0 and 13.6 MeV/u) in soda-glass where an identical value 0.16 eV was determined. A similar result is predicted by Singh and Virk (1994) for sodalime glass irradiated by heavy ions, ^{208}Pb (17.0 and 13.6 MeV/u) and ^{139}La (14.6 MeV/u), where the activation energy is calculated by using the optical absorption spectroscopy (Ritter and Mark, 1983). Hence, a single value of activation energy for annealing of these latent tracks created by different heavy ions is observed for these two formulations. Mark *et al.* (1981)formulation gives value of activation energy (0.18–0.20 eV) higher than the value obtained by other two formulations.

In the similar way, Figs 4 – 6 show the variation of $\ln(-\ln(D/D_o))$, $\ln(1-D/D_o)$ and $\ln V_a$ with the annealing temperature, $1000/T$ (°K^{-1}) for the calculation of activation energy of annealing of tracks of ^{238}U (11.4 MeV/u) from formulations of Mark *et al.* (1973), Green *et al.* (1985) and Modgil and Virk (1985), respectively. The values of activation energy of annealing of latent tracks of ^{238}U and fission fragments in BP-1 phosphate glass are given in Table 2. Green *et al.* (1985) and Modgil and Virk formulations yield the identical values of E_a (0.405 ± 0.005 eV) for ^{238}U heavy ion and fission fragments. The values of activation energy, E_a are found to be 0.47 and 0.50 eV for ^{238}U (11.4 MeV/u) ion and fission fragments, respectively, using Mark *et al.* (1981) formulation.

The latent track is considered as an aggregate of defects produced by heavy ions in the given target and the whole physics of these defects depends upon the energy loss by heavy ions during swifting in the detector. The vaule of energy loss computed from TRIM95 (Ziegler *et al.*, 1985) for different heavy ions, viz. ^{238}U (11.4 MeV/u), ^{132}Xe (11.4 and 5.9 MeV/u), ^{56}Fe (4.0 MeV/u) and ^{48}Ti (4.0 MeV/u) in both the glass detectors is presented in Table 3. It is evident that the value of activation energy is independent of the energy loss suffered by heavy ions and hence, independent of the nature of the ion beam used for the creation of latent tracks. Annealing results of heavy ion latent tracks in both the glasses clearly establish the hypothesis of single activation energy as the intrinsic property of the material, independent of the nature and energy of the ion beam used. Some equivalent versions of the Modgil and Virk (1985) formulation have been proposed but the concept of single activation energy is vindicated in all these formulations (Salamon *et al.*, 1986 and Price *et al.*, 1987). Furthermore, Modgil and Virk (1985) model is helpful to understand the annealing phenomena in materials, track recording mechanism in different dielectrics, the estimation of annealing correction to the fission track ages (Sandhu and Westgate, 1995) and determination of isotope charge

Table 2. Activation energy values calculated from three different formulations for annealing of tracks of ^{238}U (11.4 MeV/u) heavy ions and ^{252}Cf fission fragments in BP-1 phosphate glass detector

Ion	Energy(MeV/u)	Activation energy of annealing (eV)		
		Mark *et al.* (1973)	Green *et al.* (1985)	Modgil and Virk (1985)
^{238}U	11.4	0.47	0.40	0.40
Fission fragments	—	0.50	0.41	0.41

Table 3. Energy loss values computed by TRIM-95 code for different heavy ions in soda and BP-1 phosphate glasses

Ion	Energy(MeV/u)	Target	Energy loss(MeV/mg/cm^2)
^{238}U	11.4	soda glass	108.18
^{132}Xe	11.4	—	55.18
^{132}Xe	5.9	—	63.86
^{56}Fe	4.0	—	28.96
^{48}Ti	4.0	—	22.02
^{238}U	11.4	BP-1 phosphate	27.66

and mass ratios of cosmic ray heavy ions (Price *et al.*, 1987).

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