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Section B

Heavy ion radiation damage annealing in track recording insulators and single activation energy model

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A variety of track recording insulators has been irradiated using heavy ion beams from ^{40}Ca to ^{238}U (energy range 10–18 MeV/n) at the GSI UNILAC facility. Annealing data have been collected using a series of isothermal and isochronal experiments. The single activation energy model is proposed to explain the annealing kinetics of heavy ion radiation damage in track recording insulators. An empirical relation, $V_a = At_a^{-n} \exp(-E_a/kT)$, expressing the annealing rate V_a in terms of annealing time t_a and temperature T, gives the best fit to data yielding a unique value of activation energy E_a. Surprisingly, the concept of single activation energy as a unique property of the track recording insulators, both crystalline and amorphous, is vindicated, irrespective of the nature and energy of ion beam used for irradiation. Some interesting applications of our formulation are also discussed.

1. Introduction

Heavily ionising nuclear particles produce radiation damage tracks in a variety of insulators. Recently, some attempts have been made to understand the anatomy of radiation damage latent tracks in solids using small angle X-ray and neutron scattering, and to make direct observations using high resolution electron microscopy [1–4].

The annealing kinetics for a single activated process are discussed within the framework of two most commonly employed types of data, i.e. those obtained from isochronal and isothermal anneals. Therefore, it has been presumed that the experimental technique measures the concentration of defects N after a fixed time t different temperatures (isochronal) or as a continuous function of time at a constant temperature (isothermal). Then, the most general chemical rate equation for a simple decay process is given by [5]

$$\mathrm{d}N/\mathrm{d}t = N^n K \tag{1}$$

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

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

where E_a is an activation energy and k the Boltzmann constant. If a discrete activation energy were to be involved and first order kinetics were assumed, the activation energy could be determined from eq. (2). Several methods employed to determine the activation energy E_a are described by Dienes and Damask [6].

It has been observed empirically by many workers [7–10] that an Arrhenius equation

$$t = A \exp(E_a/kT) \tag{3}$$

can be applied to the thermal annealing of heavy ion radiation damage tracks in minerals and glasses. However, it is observed that eq. (3) yields a spectrum of activation energy in contradiction of single activated process. The following is the modified version of eq. (3):

$$\ln(t) = C(r) + E_a(r)/kT, \tag{4}$$

where $r = l/l_0$ or D/D_0 is a measure of length or diameter reduction of annealed tracks and $E_a(r)$ is a function of r only. Hence the activation energy depends on the degree of track annealing and is not unique for a given insulator.

To resolve this problem, Modgil and Virk [11] proposed a new empirical formula expressing the annealing rate in terms of annealing time and temperature by the following equation:

$$V_a = At_a^{-n} \exp(-E_a/kT_a), \tag{5}$$

where $V_a = \mathrm{d}l/\mathrm{d}t$ is the annealing rate, t_a and T_a are the annealing time and temperature, respectively, n the exponent, and A the proportionality constant. The unique feature of this formulation is that it yields a single activation energy E_a for all track recording insulators. A crude justification for eq. (5) has been provided by the assumption of a bimolecular reaction model [12].

Table 1
The value of activation energy E_a for various types of insulators using Modgil and Virk formulation [11]

Detector	Ion (energy)	E_a [eV]	Detector	Ion (energy)	E_a [eV]
Apatite	^{93}Nb (18.0 MeV/n)	0.71	Sodalime	^{139}La (14.6 MeV/n)	0.16
	^{208}Pb (17.0 MeV/n)	0.74	Glass	^{208}Pb (17.0 MeV/n)	0.16
	^{238}U (10.0 MeV/n)	0.72		^{238}U (15.0 MeV/n)	0.16
Chlorite	^{40}Ca (15.0 MeV/n)	0.80	Phosphate	^{139}La (14.6 MeV/n)	0.56
	^{139}La (14.6 MeV/n)	0.78	Glass	^{208}Pb (17.0 MeV/n)	0.56
	^{238}U (16.5 MeV/n)	0.77		^{238}U (15.0 MeV/n)	0.56
Muscovite	^{40}Ca (15.0 MeV/n)	0.98	Lexan	^{238}U (16.0 MeV/n)	0.17
	^{139}La (14.6 MeV/n)	0.98		^{208}Pb (17.0 MeV/n)	0.17
	^{238}U (16.5 MeV/n)	0.96		^{139}La (14.6 MeV/n)	0.17
Zircon	^{93}Nb (18.0 MeV/n)	3.61	CR-39	^{93}Nb (18.0 MeV/n)	0.20
	^{208}Pb (17.0 MeV/n)	3.58		^{208}Pb (17.0 MeV/n)	0.20
	^{238}U (10.0 MeV/n)	3.57		^{238}U (10.0 MeV/n)	0.20

2. Experimental technique

Samples of various track recording insulators, both crystalline and amorphous, were prepared as per specifications given by GSI for irradiation at UNILAC facility. A variety of heavy ion beams from ^{40}Ca to ^{238}U, with energies ranging from 10–18 MeV/n, were made available for irradiation yielding a track density of 10^4–10^6 cm^{-2}. A set of annealing experiments were performed for each track recording insulator under both isothermal and isochronal conditions, respectively. A lot of annealing data were collected and analysed from these experiments [13–17].

3. Discussion of results

The experimental data on annealing of heavy ion tracks in apatite, chlorite, muscovite, zircon, sodalime glass, phosphate glass, Lexan polycarbonate and CR-39 plastic gives a best fit to the empirical relation (eq. (5)) proposed by Modgil and Virk [11]. The values of activation energy deduced by plotting ln V_a versus $1/T$ are summarized in table 1. It is interesting to note that all categories of heavy ions, despite their different beam energies, yield almost identical values of activation energy of annealing for a given insulator. This shows that the minimum energy required to start the annealing process is independent of the nature and energy of the track forming ion and is a characteristic property of the detector material.

It seems that the time is ripe for exploitation of this simple but unique formulation which establishes activation energy as a detector property independent of nature and energy of ion beam used as well as annealing history of the sample. Price et al. [18] have used a modified version of eq. (5) in their annealing experiments using phosphate glass detectors for determination of charge and isotopic resolution of relativistic heavy ion cosmic ray tracks.

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