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A model for annealing of nuclear tracks in solids

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Abstract

A mathematical formula is derived for annealing of radiation induced damage in solids starting with the empirical equation having a form of the Arrhenius equation. This formula is transformed into a practical relationship in the usable form to fit the experimental track annealing data by making use of a relationship between fractional defect density in the latent track and etched track length. Calculated lengths of fission fragment tracks in annealed CR-39 based on the proposed model are compared with corresponding experimental measurements (our previously published results). A very good agreement is found between calculated results and experimental measurements. Physical meanings of parameters involved in the proposed model are given with appropriate theoretical explanation guided by analysis based on the model-fit on track annealing measurements. The results in this paper are useful for a wide spectrum of researchers including cosmic-ray physicists, geologists, nuclear track methodologists and semiconductor/accelerator physicists using ions implantation for doping semiconductors and materials modifications.

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Keywords: Ion; Insulating solid; Nuclear track; Annealing; Model

1. Introduction

Annealing of nuclear tracks has applications in correcting the data in cosmic-ray experiments (Westphal et al., 1998; Price et al., 1987) for comparison with laboratory data and in the exploration of uranium and geological history of earth (Green et al., 1985; Laslett et al., 1987). Annealing of nuclear tracks can also be used in understanding track formation as it can be thought of as the inverse process to track formation (Rana et al., 2000; Rana, 2006). Many authors have studied annealing of nuclear tracks and some of them have given mathematical formulae fitted to their data. A brief review of these fitted mathematical formulae on track annealing data, up to the year 2000, and discussion about them are given in our previous studies (Rana et al., 2000, 2001). Recently, Guedes et al. (2005, 2006) have proposed a model for annealing of nuclear tracks in minerals. Despite all this work, the annealing or fading of nuclear tracks is still poorly understood due to absence of comprehensive theories of track formation and track fading due to thermal effects.

Here, it is attempted to give an insight into the annealing of radiation damage, generally in track recording solids, using Arrhenius equation involving Boltzmann constant. This equation leads to a mathematical model for annealing of nuclear tracks after some justified assumptions, especially a relationship between fractional defect density in the latent track and etched track length. Calculations based on this model are compared with corresponding measurements of fission fragment tracks in the most widely used track detector CR-39. The plan of the paper is to outline the proposed model, discussion and comparison of calculated results with experimental results and finally conclusions. It should be noted that the word "defect" is used here in general meanings and it is an irregularity in a structure (crystal or amorphous) and it can be an interstitial, a vacancy and a lattice translation or rotation in a crystal and a broken chemical bond in an amorphous material. In other words, defect is a structure having an atomic spatial arrangement different from initial or bulk structure of the material.

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2. The model

2.1. Derivation

The Arrhenius type empirical equation can be used to relate annealing time and temperature as

$$t = t_0 \exp\left(\frac{E_a}{kT}\right), \tag{1}$$

where t and T are annealing time and temperature, respectively, E_a is the activation energy of annealing, k is the Boltzmann constant and t_0 is constant of proportionality and has same units as time. This equation gives the relationship between time and temperature for removal of damage inside a solid. Eq. (1) can be rewritten as

$$1 = \frac{t}{t_0} \exp\left(-\frac{E_a}{kT}\right), \tag{2}$$

where E_a is the minimum energy required to start the annealing process in which atoms start shifting from non-equilibrium positions in the latent track to equilibrium positions involving (on the average) a large number of steps. The energy needed to shift from one position in a solid to another is due to the energy barrier between any two possible positions of the atom. The highest energy point between two possible positions of an atom is called a saddle point. Left-hand side represents actually the complete removal of defects. In case of nuclear tracks, let $n(t, T)$ is the density of point defects in the form of atoms displaced from their equilibrium positions (lattice sites in crystals) after annealing time t at temperature T and n_0 is the initial un-annealed defect density. In case of complete removal of defects, the above equation can be expressed as

$$\frac{n_0}{n_0} = \frac{t}{t_0} \exp\left(-\frac{E_a}{kT}\right). \tag{3}$$

In case of partial annealing, the above equation can be written as

$$\frac{\Delta n(t, T)}{n_0} = \frac{t}{t_0} \exp\left(-\frac{E_a}{kT}\right), \tag{4}$$

where Δn is the decrease in defect density in time t at temperature T:

$$1 - \frac{n(t, T)}{n_0} = \frac{t}{t_0} \exp\left(-\frac{E_a}{kT}\right). \tag{5}$$

Eq. (5) can be rewritten as

$$n(t, T) = n_0 \left[1 - \frac{t}{t_0} \exp\left(-\frac{E_a}{kT}\right)\right]. \tag{6}$$

Partial derivatives of Eq. (6) with respect to time and temperature are

$$\frac{\partial n(t, T)}{\partial t} = -\frac{n_0}{t_0} \exp\left(-\frac{E_a}{kT}\right), \tag{7a}$$

$$\frac{\partial n(t, T)}{\partial T} = -n_0 \frac{t}{t_0} \frac{E_a}{kT^2} \exp\left(-\frac{E_a}{kT}\right). \tag{7b}$$

Eqs. (7a) and (7b) give, respectively, time and temperature annealing rates of ion induced damage or latent tracks. They are important in understanding annealing behaviour of latent tracks or ion induced damage in solids. Using Eq. (7a) in Eq. (7b), we get an important relationship,

$$\frac{\partial n(t, T)}{\partial T} = \frac{E_a}{kT^2} t \frac{\partial n(t, T)}{\partial t}, \tag{8}$$

which is the first order partial differential equation and is called here annealing time–temperature equation. It shows that time and temperature annealing rates of ion induced damage or latent nuclear tracks are coupled. This is an important result due to its possible connection with the historical problem of geological thermal history reconstruction and analysis of time–temperature path (Green et al., 1985; Laslett et al., 1987; Guedes et al., 2005) in fission track dating. Eq. (8) shows that a small uncertainty in temperature will be reflected as a big uncertainty in annealing time.

After qualitative discussion about annealing of latent tracks or ion induced damage, we consider again Eq. (6) in attempt to achieve a relationship, which could be used to quantify annealing process of nuclear tracks. Eq. (6) represents the repair of defects or broken chemical bonds inside charged particle track during annealing. We cannot measure the density of point defects as a function of time in an annealing experiment. The most commonly used parameter is track length. The relationship between repair rate of individual defects (Nakata, 1999; Nordlund and Averback, 1997; Tombrello, 1993) in a latent track and decrease in etching rate of annealed latent track is understandable. Understanding the etchable track formation (Price and Walker, 1962; Chadderton, 2003; Enge, 1995; Ditlov, 2001; Fromm, 2005; Spohr, 2005) in solids is helpful in understanding the above mentioned relationship as annealing can be thought of as the inverse process to track formation. Here, an assumption is made by involving fractional quantities to minimize the free parameters to be used,

$$\frac{n(t, T)}{n_0} = \left(\frac{l(t, T)}{l_0}\right)^{1/\beta}, \tag{9}$$

where l_0 is the un-annealed track length, $l(t, T)$ is the length of track annealed at temperature T for time t, and β is a dimensionless free parameter included to incorporate the possibility of nonlinear nature of the relationship between number of defects and track lengths due to chemical etching process involved. The parameter β depends on the relationship between the removal of atomic defects in the latent track during annealing and reduction in the track length due to reduced etch rate. This relationship is difficult to understand due to complicated parameters involved like formation energy of defects and their diffusivity in the latent track, which is a phenomenon not completely understood. Further difficulty is added by the lack of thorough understanding of preferential chemical etching (Rana and Qureshi, 2002), which is normally thought as a threshold reaction depending upon deposited energy density in the latent track. The free parameter β should be fixed for, at least, specific experimental conditions. Experimental conditions mean