

Radiation Effects and Defects in Solids: Incorporating Plasma Science and Plasma Technology

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/grad20>

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Published online: 19 Aug 2006.

To cite this article: H. S. Virk (1995) Single activation energy model of radiation damage in solid state nuclear track detectors, Radiation Effects and Defects in Solids: Incorporating Plasma Science and Plasma Technology, 133:1, 87-95, DOI: [10.1080/10420159508225760](https://doi.org/10.1080/10420159508225760)

To link to this article: <http://dx.doi.org/10.1080/10420159508225760>

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SINGLE ACTIVATION ENERGY MODEL OF RADIATION DAMAGE IN SOLID STATE NUCLEAR TRACK DETECTORS

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(Received May 24, 1994)

Passage of heavy ions produces radiation damage trails known as latent tracks in a variety of solid state nuclear track detectors (SSNTDs). The empirical formulation of the Single Activation Energy Model is based on track annealing data collected in our laboratory from both isothermal and isochronal experiments performed on different types of SSNTDs, viz. glasses, plastics and minerals. Some equivalent versions of our model have been proposed but the concept of single activation energy is vindicated in all empirical formulations. It is interesting to remark that this model always yields a unique value of activation energy independent of the nature of ion beam used and its degree of annealing. The anisotropy of mineral SSNTDs is revealed by variation of activation energy along different crystal planes and even at different orientations of ion beam. Some recent experiments are a pointer to the successful exploitation of this model for future cosmic ray studies using SSNTDs.

Key words: Solid state nuclear track detectors, defects, annealing, heavy ions, activation energy.

1 INTRODUCTION

Heavily ionising nuclear particles produce radiation damage tracks in a variety of dielectric materials known as Solid State Nuclear Track Detectors (SSNTDs). These damage tracks find applications in nuclear, cosmic ray and elementary particle physics, chemistry, radiobiology, earth sciences and nuclear engineering and host of other applications such as nuclear safeguards, virus counting, uranium exploration, archaeology and bird altimetry.¹

Nuclear tracks in solids are known since Young's investigations in 1958.² It is unfortunate that while trackologists have indulged in a plethora of fission track applications, there is hardly any break-through in the understanding of track formation both at the theoretical and experimental levels during the last 30 years. 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^{3,4} and direct observations using high resolution electron microscopy (HREM)^{5,6} and atomic force microscopy.⁷

Passage of a heavy ion in an SSNTD creates intense radiation damage which results in a series of point defects and extended defects along the latent track. It is observed that linear density of point defects closely follows projectile energy loss along the track while the density of extended defects is a steep function of it. Tombrello *et al.*⁸ suggest that extended defects are generated by atomic K-shell excitations in the heavier elements of the SSNTD. White *et al.*⁹ have emphasized that atomic or ionic defects can be created as a result of electronic excitation in track recording insulators. HREM⁵ reveals that latent tracks are constituted of extended defects, separated by gap zones loaded with point defects.

It has been an enigma for trackologists to get real knowledge of track formation in solids. It was evident to earlier workers that latent nuclear tracks disappear (anneal) when observed in the beam of electron microscope. Chemical etching renders the latent track visible under an optical microscope. Both chemical etching and annealing are controlled by the presence

of extended defects. By a judicious choice of annealing and etching experiments, we have developed a simple empirical approach to unravel the mystery of thermal fading of nuclear latent tracks in SSNTDs. Single Activation Energy Model was proposed for inorganic solids¹⁰ but to our utter surprise it holds its sway over all types of SSNTDs, viz. glasses, plastics and minerals.¹¹⁻²⁰

2 CONCEPT OF SINGLE ACTIVATION ENERGY

It is commonly assumed that track annealing is dominantly a diffusive process in which interstitially displaced atoms thermally penetrate an activation barrier to recover their initial lattice positions. Thus one is led to an Arrhenius type relation if latent track damage is solely a function of the interstitial atom density, i.e. concentration of defects in a solid.

If the annealing of a defect occurs by a single activated process with a constant activation energy, E_a , then the rate of change of the concentration of the defect is describable by the equation

$$\frac{dn}{dt} = -F(n)K = -F(n)K_0 \exp(-E_a/kT) \quad (1)$$

where n is the fractional concentration of the defect, $F(n)$ is any continuous function of n and K is the characteristic rate constant involving a Boltzmann factor, $\exp(-E_a/kT)$, for its dependence on annealing temperature, T . It is implicitly implied by equation (1) that activation energy, E_a , is independent of n .

There are several methods for the determination of activation energy from annealing data. We discuss here only the method of cross-cut.²¹

Let n_0 be initial concentration of defects in all samples which are annealed at several different temperatures. The isothermal annealing curves are shown in Figure 1. If a line parallel to the time axis is drawn (dashed line) at a given value of n , say n_1 , then the integrand of equation (2) is equal to a constant

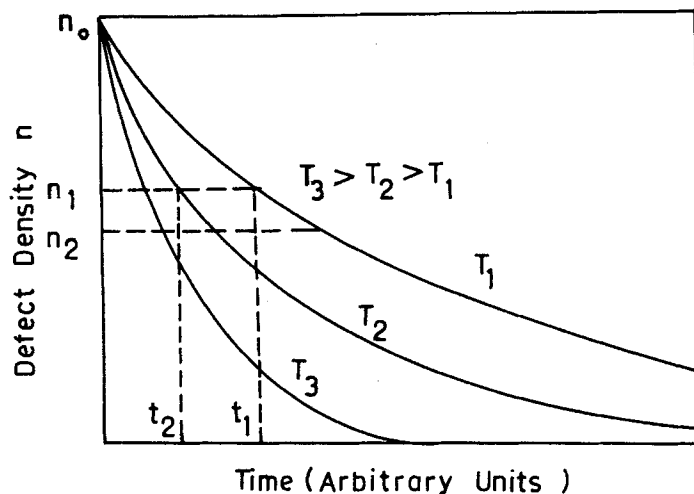


FIGURE 1 Determination of activation energy by the method of cross-cut.

$$\int_{n_0}^{n_1} \frac{dn}{F(n)} = K_0 t \exp(-E_a/kT) = C \quad (2)$$

$$\text{or,} \quad t \exp(-E_a/kT) = \text{Constant} = C^1 \quad (3)$$

Thus two times, t_1 and t_2 , required to reach a constant value of n at temperatures T_1 and T_2 , respectively, are related by

$$\ln t_1/t_2 = E_a/k(1/T_1 - 1/T_2) \quad (4)$$

When the cut crosses several curves, the times are related to the temperatures by

$$\ln(t_i) = \ln C^1 + E_a/kT_i \quad (5)$$

i.e., the logarithm of t_i is linear in $1/T_i$ with E_a/k as the slope of the curve. If a cut is taken at another point, n_2 , only the value of constant is altered in equation (5) and the lines described by this equation should be parallel. This characteristic is a check on the assumption that a single process with a constant activation energy is operative.

3 COMPARISON OF ANNEALING MODELS

In many experiments a sample containing defects is gradually heated at a constant rate and a physical property, p , related to the number of defects is observed during the annealing. Since n depends exponentially on the temperature, dp/dt is very small during the low temperature stages and then grows rather suddenly in the vicinity of a characteristic annealing temperature, T_a and then decreases to zero as all the defects responsible for a given annealing stage disappear.²¹

The physical property which is generally studied is either track density or track length/diameter reduction by performing two sets of experiments, viz. isothermal and isochronal. Track length reduction is considered to be a better parameter in recent investigations.²² Recently, other parameters based on length reduction, viz. annealing velocity, instantaneous track etch velocity and etch rate reduction have made significant contributions to the concept of single activation energy.^{10,15,30}

Track annealing models are broadly classified into three categories according to their mathematical formulations. These are logarithmic, linear and exponential models.

3.1 Logarithmic Model

Many of the earlier studies of annealing of fission tracks in minerals and glasses were based on this model using Arrhenius equation (3)

$$t \exp(-E_a/kT) = \text{Constant},$$

where E_a is effective activation energy, k is Boltzmann constant, t and T represent annealing time and temperature, respectively. Plots of $\ln t$ versus $1/T$ are called Arrhenius plots (Figure 2a) and must be parallel for different degrees of track fading if diffusion process is operative under single constant activation energy. However, it is observed¹ that the logarithmic model yields fanning Arrhenius plots (Figure 2b) with a spectrum of activation energies corresponding to different degrees of track fading. Hence, equation (3) is modified as

$$\ln(t) = C(r) + E(r)/kT \quad (6)$$

where $r = 1/l_0$ is a measure of length reduction and both $C(r)$ and $E(r)$ are functions of r only. This model has been exploited by many enthusiastic geochronologists^{23,24} to unravel the geothermal history of rocks, tectonic uplift and blocking temperatures. Its main limitation is that the Arrhenius equation used for thermal fading of tracks is applicable under constant temperature conditions only and does not allow the treatment of the problem for a given temperature history $T(t)$. It is also unable to explain fanning of Arrhenius plots and spectrum of activation energies.

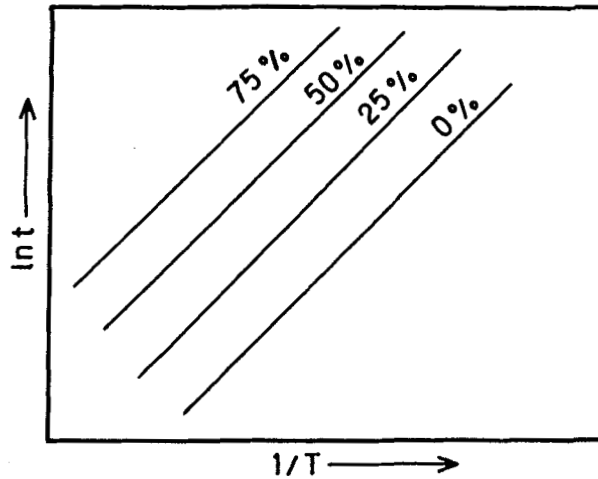


FIGURE 2(a) Parallelism of Arrhenius plots for different degrees of track fading under constant activation energy.

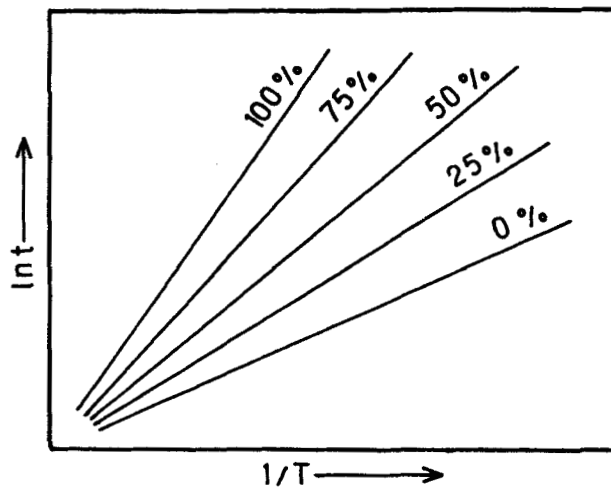


FIGURE 2(b) Fanning of Arrhenius plots corresponding to a variable activation energy.

3.2 Linear Model

Mantovani²⁵ proposed a linear relationship between track retention and heating time for muscovite,

$$r = A - Bt \quad (7)$$

where A and B are constants. Laslett *et al.*²⁶ modified this relation bringing in time and temperature explicitly and using $(1 - r)$ instead of r , that is

$$\ln(1 - r) = C_0 + \ln t + C/T \quad (8)$$

Dakowski *et al.*²⁷ also suggested a linear relationship between the track annealing parameter r and $\ln t$ for isothermal annealing of tracks in minerals. They proposed the following relationship for variation of E_a with r

$$E_a(r) = C(\alpha + \beta r + \gamma r^2) \quad (9)$$

where C , α , β , γ are constants to be determined from experimental data.

3.3 Exponential Model

The variation of the activation energy, E_a , with the degree of annealing, during isothermal annealing was, first of all, questioned by Mark *et al.*²⁸ They proposed a single activation energy of annealing but abandoned this concept in favour of two activation energies, one for low temperatures and the other one for the high temperature range. They proposed that the exact solution for isothermal annealing can be well described by using an infinite summation series with exponential decay functions

$$p(t) = \sum_{j=1}^n p_j \exp(-\alpha_j t) \quad (10)$$

where p_j and α_j are fitting parameters. To a good approximation, the track density may be expressed by a sum of two exponentials²⁹

$$p(t) = p(0) \exp[-\alpha(T)t] \quad (11)$$

where $\alpha(T)$ is a decay constant (annealing co-efficient) given by

$$\alpha(T) = \alpha_{01} \exp(-E_{a1}/kT) + \alpha_{02} \exp(-E_{a2}/kT) \quad (12)$$

α_{01} , α_{02} are annealing constants and E_{a1} and E_{a2} are two different activation energies of two different diffusion processes involved. For high temperature annealing, the single exponential approximation is used and equation (12) reduces to

$$\alpha(T) = \alpha_0 \exp(-E_a/kT) \quad (13)$$

The exponential model can be expressed in terms of track density reduction or length reduction by using equations (11) and (13) as follows

$$\ln(-\ln r) = C_0 + \ln t - E_a/kT \quad (14)$$

4. SINGLE ACTIVATION ENERGY MODEL

It is a well established fact that the latent track annealing rate is a function of annealing time and temperature. Since most of the annealing experiments are performed under isothermal or isochronal conditions, the Arrhenius approach was adopted to explain the fading of tracks in minerals to correct their apparent ages. The various shortcomings of the track annealing models based on Arrhenius approach are listed below:

- (i) The Arrhenius equation is applicable under constant temperature conditions. It necessitates approximations as soon as the fading temperature varies with time.
- (ii) Most models are based on an 'a priori' assumption that the latent track anneals as a whole. Hence it is not justifiable to correlate the residual lengths or diameters of the partially annealed tracks with annealing temperature and time.
- (iii) The activation energy is a function of the degree of track loss in a given temperature-time plane which results in fanning of the Arrhenius plots. However, this feature defies the very basis of Arrhenius equation.

To resolve these contradictions of Arrhenius approach, Modgil and Virk¹⁰ proposed the Single Activation Energy Model on the assumption that the activation energy is a material dependent property. The empirical formulation (eq. 15) of this model relates the instantaneous annealing velocity ($V_a = dl/dt$ or dD/dt) explicitly with time and temperature, a crude justification for which has been provided by the assumption of a bimolecular reaction model.³⁰

$$V_a = A t_a^{-n} \exp(-E_a/kT) \quad (15)$$

where both A and n are ion-dependent constants and E_a is unique for a given SSNTD. To determine E_a , equation (15) is modified as follows

$$\ln V_a = \ln A - n \ln t_a - E_a/kT \quad (16)$$

The special features of this model are:

- (i) It predicts a single activation energy of annealing for all heavy ions and fission fragment tracks as required by the Arrhenius equation.
- (ii) It may be used for revealing the thermal history of track recording SSNTDs (minerals, meteorites and lunar rocks) as the annealing rate is given explicitly in terms of both time and temperature.
- (iii) It explains the partial fading of tracks due to environmental annealing as the track length is used as a parameter instead of track density.
- (iv) It has a universal application. Its validity is tested for all types of SSNTDs (both crystalline and amorphous) using a variety of heavy ion beams and fission fragments.¹¹⁻²⁰

5. DISCUSSION AND CONCLUSIONS

It seems that the time is ripe for exploitation of this simple but unique model. Some worthwhile attempts have been made to develop modified versions of our model. Salamon *et al.*³⁰ replaced the annealing velocity by the etch rate reduction of annealed latent tracks and found that the activation energy, E_a and other parameters, i.e. n and A , are also constants

for a given SSNTD. Price *et al.*³¹ have found an application of our model in their annealing experiments using phosphate glass detectors for recording of relativistic nuclei tracks. They report that significant annealing occurs in SSNTDs even at 0°C over a period of a few years. This has an obvious impact on experiments (e.g. Heavy Nucleus Collector and Space Shuttle 'Ions' Experiments) being carried out in space for recording cosmic rays using plastic and glass detectors. The charge and isotopic resolution is calculated after taking into account annealing correction using this model.

Recently, an equivalent version of the Single Activation Energy Model has been developed in our laboratory¹⁵ to overcome the shortcomings of our earlier formulation¹⁰ and that are proposed by Salamon *et al.*³⁰ The new formulation replaces the instantaneous annealing velocity, V_a , by the instantaneous track etch velocity, V_e , and gives a better fit of annealing data

$$d/dt_a(V_e) = A t_a^{-n} \exp(-E_a/kT) \quad (17)$$

The list of SSNTDs selected for our analysis is quite exhaustive and includes a variety of glasses, plastics and minerals. The results on activation energy data compiled in our laboratory are summarized in Table I. It is observed that all SSNTDs have a unique value of activation energy independent of ion beam and its energy as proposed in the empirical formulation of our model. Table II establishes the equivalence of the three different formulations of single activation energy model. While E_a is nearly the same, the value of n is different and shows a marked ion dependence.

Tables III and IV provide a convincing proof of the anisotropy of mineral SSNTDs. The activation energy along different crystal planes has different unique values, garnet being the only exception. The anisotropy is so marked in muscovite that the value of E_a varies even with the incidence angle of the beam on the same plane. However, the most remarkable feature of our model, the concept of single activation energy as a unique property of the detector, is vindicated in all the experiments using different ion beams and both the organic and inorganic SSNTDs.

TABLE I
The value of activation energy, E_a , for various types of SSNTDs using Modgil and Virk formulation.¹⁰

Detector	Ion(Energy)	E_a (eV)	Detector	Ion(Energy)	E_a (eV)
Apatite	⁹³ Nb(18.0 MeV/n)	0.71	Sodalime Glass	¹³⁹ La(14.6 MeV/n)	0.16
	²⁰⁸ Pb(17.0 MeV/n)	0.74		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 Glass	¹³⁹ La(14.6 MeV/n)	0.56
	¹³⁹ La(14.6 MeV/n)	0.78		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(13.6 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

TABLE II
The values of activation energy, E_a and exponent, n in plastic track detectors using different formulations.

Ion Beam	Energy (MeV/n)	Modgil and Virk ¹⁰		Price <i>et al.</i> ³¹		Bhatia and Virk ¹⁵	
		E _a (eV)	n	E _a (eV)	n	E _a (eV)	n
<i>CR-39</i>							
⁹³ Nb	18.0	0.197	0.63	0.201	0.88	0.198	0.44
¹³⁹ La	14.6	0.198	0.69	0.196	1.80	0.197	1.28
²⁰⁸ Pb	17.0	0.196	0.74	0.199	0.21	0.195	1.60
²³⁸ U	10.0	0.185	0.66				
²³⁸ U	16.0	0.194	0.65				
<i>Lexan</i>							
¹³⁹ La	14.6	0.173	1.54	0.172	0.913	0.170	2.14
²⁰⁸ Pb	13.6	0.173	2.40	0.170	0.935	0.171	3.00
²³⁸ U	16.0	0.171	2.84	0.169	0.895	0.170	2.63

**H. A. Khan, N. A. Khan, K. Jamil and R. Brandt, *Nucl. Tracks Radiat. Meas.*, 8, 377 (1984).

TABLE III
Activation energy for fission fragment track annealing on different crystal planes of mineral SSTNDs.

S. No.	SSNTD	Crystal plane	Activation energy E_a (eV)
1.	Apatite	10 $\bar{1}$ 0	0.71
		1 $\bar{1}$ 01	0.62
		0001	0.57
2.	Quartz	10 $\bar{1}$ 0	1.73
		10 $\bar{1}$ 1	1.44
		01 $\bar{1}$ 1	1.27
		0001	1.15
3.	Zircon	100	3.60
		011	2.87
		001	2.15
4.	Garnet	110	2.53
		011	2.52
		101	2.53

TABLE IV
The variation of activation energy with angle of incidence in muscovite using different ion beams.

Incident ion	Energy (MeV/n)	Activation energy E_a (eV)	
		15°	75°
⁹³ Nb	18.0	0.98	0.78
²⁰⁸ Pb	17.0	0.98	0.79
²³⁸ U	10.0	0.97	0.78
²⁵² Cf F.F		0.96	0.78

ACKNOWLEDGEMENTS

The author wishes to express his gratitude to all the workers of the SSNTD laboratory for their help in data collection and preparation of this manuscript. He is also grateful to Professors L. T. Chadderton (Canberra), P. B. Price (California), T. D. Mark (Innsbruck), R. Brandt (Marburg), M. Dakowski (Warsaw), S. A. Durrani (Birmingham) and S. Mukherjee (IIT Kanpur) for their helpful suggestions and critical evaluation of the model during its various stages of development.

I am really indebted to the Director of the GSI and Drs. Reimer Spohr and J. Vetter for beam time facility at UNILAC.

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