

Single-activation-energy model of radiation damage in solid-state nuclear-track detectors

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Passage of heavy ions produces radiation-damage trails known as latent tracks in a variety of solid-state nuclear-track detectors (SSNTDs). These tracks disappear in the beam of an electron microscope. The single-activation-energy model was proposed as an empirical approach for explaining the thermal fading of nuclear tracks. The empirical formulation of this model is based on track-annealing data collected from both isothermal and isochronal experiments performed on different types of SSNTDs. Some equivalent versions of the model have been proposed but the concept of single activation energy is vindicated in all empirical formulations. This model always yields a unique value of activation energy independent of the nature of the ion beam used and the degree of annealing. The anisotropy of mineral SSNTDs is revealed by variation in activation energy along different crystal planes and even with different orientations of the ion beam. Some recent experiments are a pointer to the successful exploitation of this model for future cosmic-ray studies using SSNTDs.

HEAVILY ionizing 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 in a host of other areas such as nuclear safeguards, virus counting, uranium exploration, archaeology and bird altimetry¹.

Nuclear tracks in solids are known since Young's investigations in 1958 (ref. 2). It is unfortunate that while 'trackologists' have indulged in a plethora of fission-track applications, there has hardly been any breakthrough in the understanding of track formation at both theoretical and experimental levels in the last 30 years. The opinions in favour of thermal spike and ion-explosion spike are as divided as ever^{3,4}. 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 direct observations using high-resolution electron microscopy (HREM)⁵⁻⁸.

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.*⁹ suggest that extended defects are generated by atomic K-shell excitations in the heavier elements of the SSNTD. HREM⁷ reveals that latent tracks are constituted of extended defects, separated by gap zones loaded with point defects.

Track formation in solids has been an enigma. It was evident to earlier workers that latent nuclear tracks disappear (anneal) when observed in the beam of an electron microscope. Chemical etching renders the latent track visible under an optical microscope but much of the useful information (physics) is destroyed. The same is true of annealing. Both chemical etching and annealing are controlled by the presence of extended defects. By a judicious choice of annealing and etching experiments, my laboratory has developed a simple empirical approach for unravelling the mystery of thermal fading of nuclear latent tracks in SSNTDs. A single-activation-energy model was proposed for inorganic solids¹⁰ but to our surprise it holds good for all types of SSNTDs, viz. glasses, plastics and minerals¹¹⁻²⁰.

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 methods²¹ for determination of activation energy from annealing-data curves: (i) method of cross-cut, (ii) ratio of slopes, (iii) constant rate of heating, and (iv) combination of isochronal and isothermal anneal. I discuss here only the method of cross-cut because of its simplicity and ease of performance compared with the other methods²¹.

Let n_0 be the initial concentration of defects in all samples that are annealed at 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:

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

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

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 the 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.

Comparison of annealing models

In many experiments a sample containing defects is gradually heated at a constant rate, and a physical

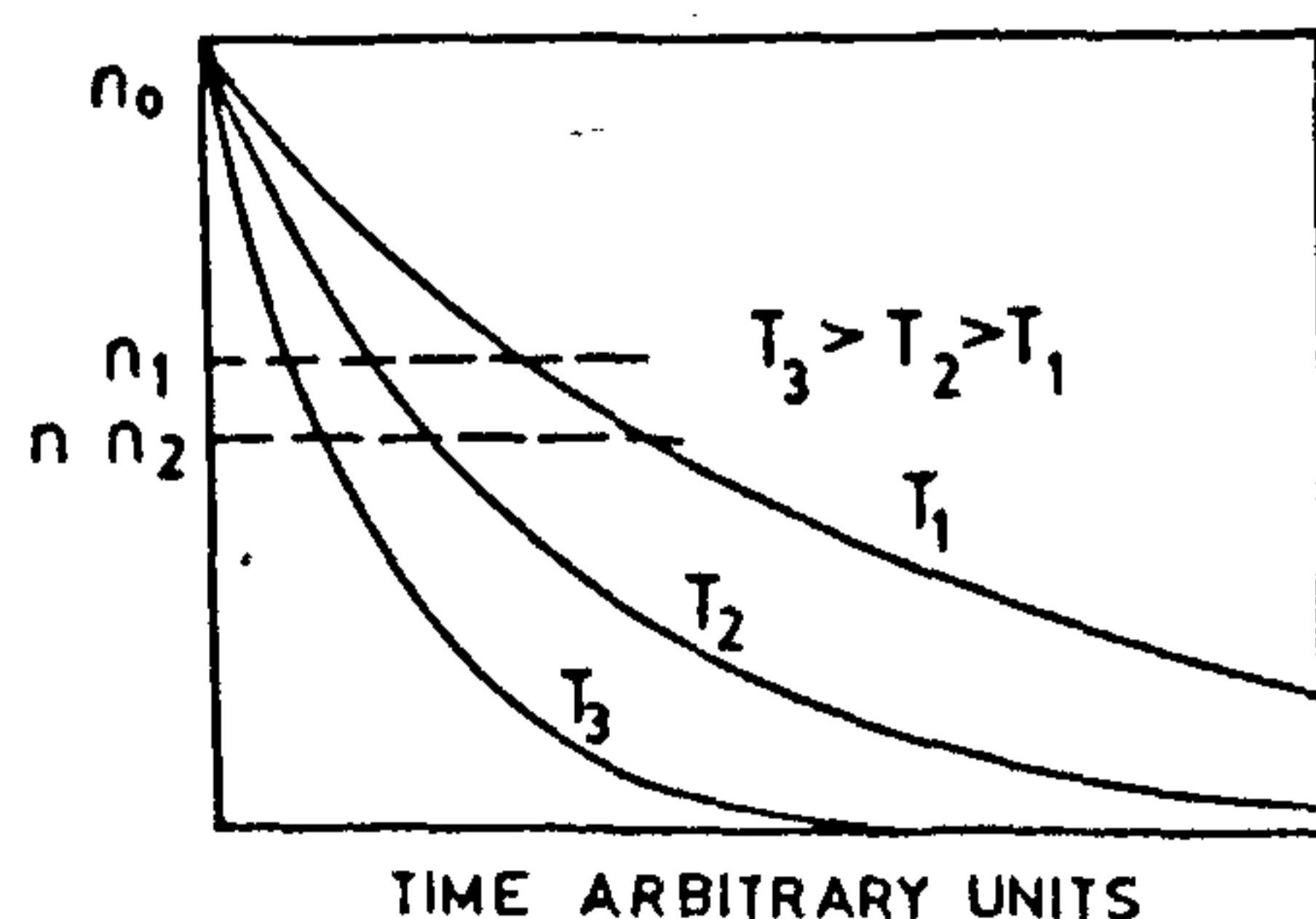


Figure 1. Determination of activation energy by cross-cut.

property, P , related to the number of defects is observed during the annealing. Since K depends exponentially on the temperature, dp/dt is very small during the low-temperature stages, then grows rather suddenly in the vicinity of a characteristic annealing temperature T_a , then decreases to zero as all the defects responsible for a given annealing stage disappear²¹.

The physical property generally studied is either track density or track-length/diameter reduction by performing two sets of experiments, viz. isothermal and isochronal. Track-length reduction has been considered to be a better parameter in recent investigations²². Of late, use of other parameters based on length reduction, viz. annealing velocity, instantaneous track-etch velocity and etch-rate reduction, has made significant contributions to the concept of single activation energy^{10,15,23}.

Track annealing models are broadly classified into three categories according to their mathematical formulation.

Logarithmic model

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

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

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

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

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 geochronologists^{24,25} 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 treatment of the problem for a given temperature history $T(t)$. It is also unable to explain the fanning of Arrhenius plots and spectrum of activation energies.

Linear model

Mantovani²⁶ proposed a linear relationship between

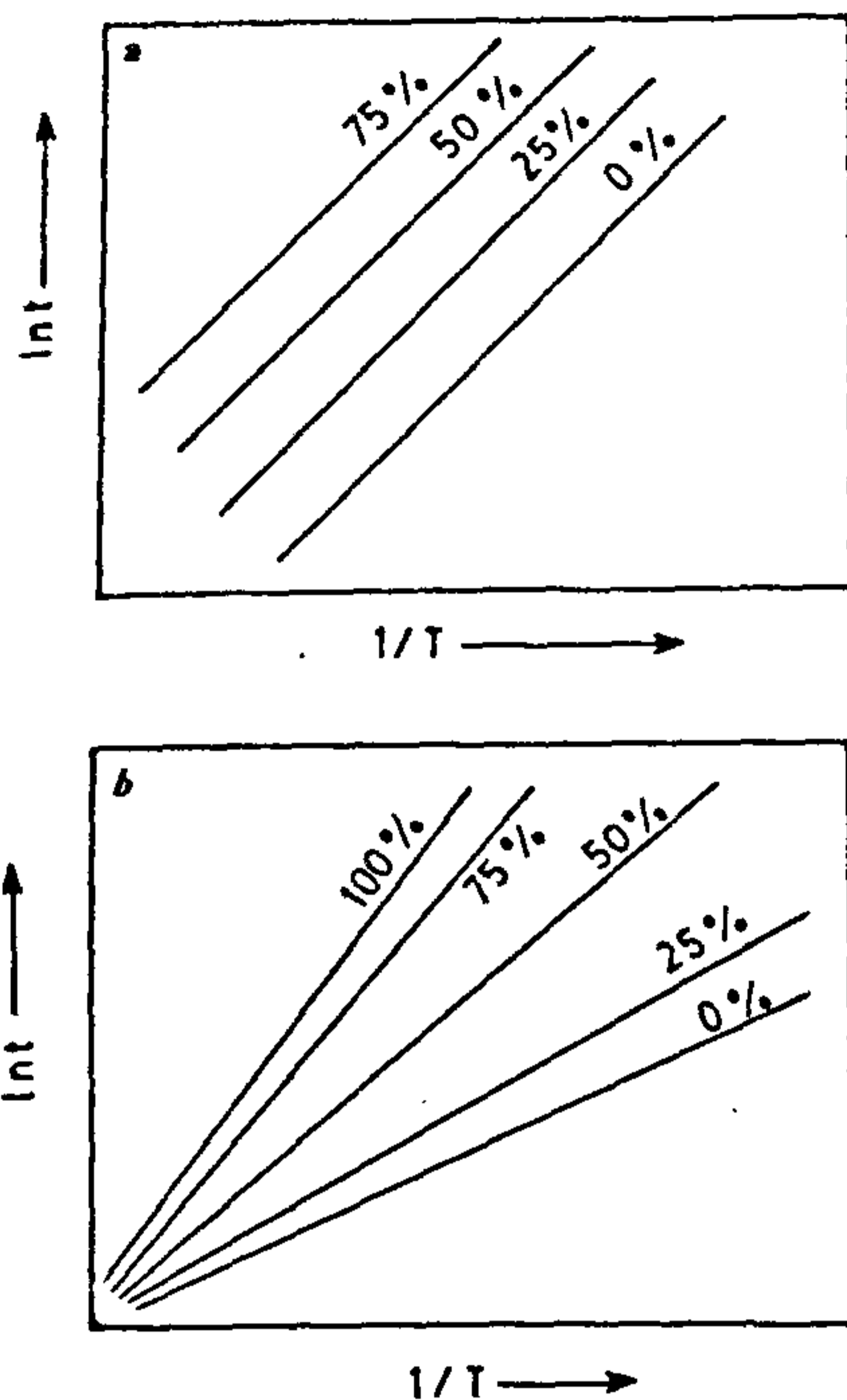


Figure 2. a. Parallelism of Arrhenius plots. b. Fanning of Arrhenius plots.

track retention and heating time for muscovite,

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

where A and B are constants. Laslett *et al.*²⁷ modified this relation, bringing in time and temperature explicitly and using $(1 - r)$ instead of r :

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

Dakowski *et al.*²⁸ 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.

Exponential model

The variation of activation energy E_a with degree of annealing during isothermal annealing was first questioned by Mark *et al.*²⁹ 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 for high-temperature range. They proposed that the exact solution for isothermal annealing can be well described using an

infinite summation series with exponential decay functions

$$p(t) = \sum_{j=0}^{\infty} p_j(0) \exp(-\alpha_j t), \quad (10)$$

where p_j and α_j are fitting parameters. To a good approximation, the track density may be expressed by a sum of two exponentials³⁰

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

where $\alpha(T)$ is a decay constant (annealing coefficient) given by

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

where 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, 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 follows¹¹

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

Single-activation-energy model

It is a well-established fact that 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 the 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 Arrhenius plots. However, this feature defies the very basis of the Arrhenius equation.

To resolve these contradictions of the Arrhenius approach, Modgil and Virk¹⁰ proposed a single-activation-energy model on the assumption that a latent track is created as an ion-explosion spike¹. The empirical formulation (equation 15) of this model

relates 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 model²³.

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

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 to

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

The special features of this model are:

- (i) It predicts 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 thermal history of track-recording SSNTDs (minerals, meteorites and lunar rocks), as 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 track length is used as a parameter in place of track density.
- (iv) It has universal application. Its validity has been tested for all types of SSNTDs (both crystalline and amorphous) using a variety of heavy-ion beams and fission fragments¹¹⁻²⁰.

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.*²³ replaced annealing velocity with etch-rate reduction of annealed latent tracks and found that activation energy E_a and other parameters, i.e. n and A , are also constant for a given SSNTD. Price *et al.*³¹ have applied the model in their annealing experiments using phosphate-glass detectors for recording tracks of relativistic nuclei. They report that significant annealing occurs in SSNTDs even at 0°C over a period of a few years. This has an obvious influence 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 are 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 the earlier formulation¹⁰ and that proposed by Salamon *et al.*²³ The new formulation replaces instantaneous annealing velocity V_a by instantaneous track-etch velocity V_p , and gives a better fit of annealing data:

$$d/dt_a (V_p) = At_a^{-n} \exp(-E_a/kT). \quad (17)$$

Table 1. Values of activation energy E_a for various SSNTDs using Modgil and Virk formulation¹⁰.

Detector	Ion (energy per nucleon)	E_a (eV)	Detector	Ion (energy)	E_a (eV)
Apatite	⁹³ Nb (18.0 MeV)	0.71	Sodalime glass	¹³⁹ La (14.6 MeV)	0.16
	²⁰⁸ Pb (17.0 MeV)	0.74		²⁰⁸ Pb (17.0 MeV)	0.16
	²³⁸ U (10.0 MeV)	0.72		²³⁸ U (15.0 MeV)	0.16
Chlorite	⁴⁰ Ca (15.0 MeV)	0.80	Phosphate/glass	¹³⁹ La (14.6 MeV)	0.56
	¹³⁹ La (14.6 MeV)	0.78		²⁰⁸ Pb (17.0 MeV)	0.56
	²³⁸ U (16.5 MeV)	0.77		²³⁸ U (15.0 MeV)	0.56
Muscovite	⁴⁰ Ca (15.0 MeV)	0.98	Lexan	²³⁸ U (16.0 MeV)	0.17
	¹³⁹ La (14.6 MeV)	0.98		²⁰⁸ Pb (13.6 MeV)	0.17
	²³⁸ U (16.5 MeV)	0.96		¹³⁹ La (14.6 MeV)	0.17
Zircon	⁹³ Nb (18.0 MeV)	3.61	CR-39	⁹³ Nb (18.0 MeV)	0.20
	²⁰⁸ Pb (17.0 MeV)	3.58		²⁰⁸ Pb (17.0 MeV)	0.20
	²³⁸ U (10.0 MeV)	3.57		²³⁸ U (10.0 MeV)	0.20

The list of SSNTDs selected for our analysis is quite exhaustive and includes a variety of glasses, plastics and minerals. Activation-energy data compiled in our laboratory are summarized in Table 1. It may be 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 2 establishes the equivalence of three

Table 2. Values of activation energy E_a and exponent n for two plastic track detectors using different formulations.

Ion beam	Energy (MeV per nucleon)		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		
CR-39								
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	0.214	0.195	1.60	
²³⁸ U	10.0	0.185	0.66					
²³⁸ U*	16.0	0.194	0.65					
Lexan								
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	

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

Table 3. Activation energy for fission-fragment-track annealing in the case of mineral SSNTDs.

SSNTD	Crystal plane	E_a (eV)
Apatite	10 $\bar{1}$ 0	0.71
	1 $\bar{1}$ 01	0.62
	0001	0.57
Quartz	10 $\bar{1}$ 0	1.73
	10 $\bar{1}$ 1	1.44
	01 $\bar{1}$ 1	1.27
	0001	1.15
Zircon	100	3.60
	011	2.87
	001	2.15
Garnet	110	2.53
	011	2.52
	101	2.53

Table 4. The value of activation energy in muscovite using different ion beams.

Incident ion	Energy (MeV per nucleon)	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

different formulations of the single-activation-energy model. While E_a is nearly the same, the value of n is different and shows a marked ion dependence.

Tables 3 and 4 provide convincing proof of the anisotropy of mineral SSNTDs. The activation energy on 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 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, has been vindicated in all the experiments using different ion beams and both organic and inorganic SSNTDs.

1. Fleischer, R. L., Price, P. B. and Walker, R. M., *Nuclear Tracks in Solids*, University of California Press, Berkeley, 1975.
2. Young, D. A., *Nature*, 1958, **182**, 375.
3. Chadderton, L. T., *Nucl. Tracks Radiat. Meas.*, 1988, **15**, 11.
4. Fleischer, R. L., Price, P. B. and Walker, R. M., *J. Appl. Phys.*, 1965, **36**, 3645.
5. Dartyge, E., Duraud, J. P., Langevin, Y. and Maurette, M., *Phys. Rev.*, 1981, **B23**, 5213.
6. Albrecht, D., Armbruster, P., Sphor, R., Roth, M., Schauptert, K. and Stuhrman, H., *Appl. Phys.*, 1985, **A37**, 37.
7. Houpert, C., Hervieu, M., Groult, D., Studer, F. and Toulemonde, M., *Nucl. Instrum. Methods Phys. Res.*, 1988, **B32**, 393.
8. Chadderton, L. T., Biersack, J. P. and Koul, S. L., *Nucl. Tracks Radiat. Meas.*, 1988, **15**, 31.
9. Tombrello, T. A., Wie, C. R., Itoh, N. and Nakayama, T., *Phys. Lett.*, 1984, **A100**, 42; Tombrello, T. A., *Nucl. Instrum. Methods*, 1984, **B1**, 23.
10. Modgil, S. K. and Virk, H. S., *Nucl. Instrum. Methods*, 1985, **B12**, 212.
11. Virk, H. S., Modgil, S. K., Singh, Gurinder and Bhatia, R. K., *Nucl. Instrum. Methods Phys. Res.*, 1988, **B32**, 401.

12. Virk, H. S., Modgil, S. K. and Bhatia, R. K., *Nucl. Tracks Radiat. Meas.*, 1986, **11**, 323.
13. Bhatia, R. K. and Virk, H. S., *Indian J. Pure and Appl. Phys.*, 1987, **25**, 282.
14. Singh, Ravi Chand, Bhatia, R. K. and Virk, H. S., *Indian J. Pure Appl. Phys.*, 1988, **26**, 673.
15. Bhatia, R. K. and Virk, H. S., *Radiat. Eff.*, 1989, **107**, 167.
16. Sandhu, A. S., Bhatia, R. K., Singh, Surinder and Virk, H. S., *GSI Sci. Rep.*, 1987, 242.
17. Sandhu, A. S., Singh, Surinder and Virk, H. S., *Indian J. Pure Appl. Phys.*, 1987, **25**, 97.
18. Sandhu, A. S., Singh, Surinder and Virk, H. S., *Mineral. J.*, 1987, **13**, 254.
19. Sandhu, A. S., Singh, Surinder and Virk, H. S., *Mineral. J.*, 1988, **14**, 1.
20. Singh, Lakhwant, Sandhu, A. S., Singh, Surinder and Virk, H. S., *Radiat. Eff. Def. Solids*, 1989, **108**, 257.
21. Dienes, G. J. and Damask, A. C., *Point Defects in Metals*, Gordon and Breach, New York, 1963.
22. Green, P. F., Duddy, I. R., Gleadow, A. J. W. and Tingate, P. R., *Nucl. Tracks*, 1985, **10**, 323.
23. Salamon, M. H., Price, P. B. and Drach, J., *Nucl. Instrum. Methods Phys. Res.*, 1986, **B17**, 173.
24. Sharma, K. K., Bal, K. D., Prashad, R., Lal, Nand and Nagpal, K. K., *Tectonophysics*, 1980, **70**, 135.
25. Carpena, J., Pognante, U. and Lombardo, B., *Tectonophysics*, 1986, **127**, 117.
26. Mantovani, M. S. M., *Earth Planet. Sci. Lett.*, 1974, **24**, 311.
27. Laslett, G. M., Green, P. F., Duddy, I. R. and Gleadow, A. J. W., *Chem. Geol.*, 1987, **65**, 1.
28. Dakowski, M., Burchart, J. and Galazka, J., *Bull. Acad. Pol. Sci., Ser. Sci. Terre*, 1974, **22**, 11.
29. Mark, E., Pahl, M., Purtscheller, F. and Mark, T. D., *Min. Petrogr. Mitt.*, 1973, **20**, 131.
30. Mark, T. D., Pahl, M. and Vartanian, R., *Nucl. Technol.*, 1981, **52**, 295.
31. Price, P. B., Gerbier, G., Park, H. S. and Salamon, M. H., *Nucl. Instrum. Methods Phys. Res.*, 1987, **B28**, 53.

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