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Etching and annealing kinetics of ^{238}U ion tracks in Makrofol-N plastic

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Abstract

Swift heavy ions of sufficient size and energy produce stable latent tracks in most of nuclear track recording materials. The etching and annealing kinetics of ^{238}U (17.17 and 16.34 MeV/u) ion tracks in a polycarbonate plastic (Makrofol-N) were investigated. The values of various etching parameters, viz., bulk etch rate (V_G), track etch rate (V_T), critical angle (θ_c), sensitivity ($S = V_T/V_G$) of the plastic for registration of ^{238}U ion, etching efficiency ($\eta = 1 - (V_G/V_T)$) and activation energies for bulk (E_b) and track etching (E_t) are reported. For study of annealing characteristics, the isochronal experiments were performed on Makrofol-N plastic irradiated with ^{238}U (17.17 and 16.34 MeV/u) ions from UNILAC accelerator at GSI. The annealing kinetics of heavy ion damaged Makrofol-N is explained by using modified version of Modgil and Virk (S.K. Modgil, H.S. Virk, Nucl. Instr. and Meth. B 72 (1985) 212) empirical formulation. Finally, the experimental range of ^{238}U ion in Makrofol-N has been compared with the theoretical values from different formulations.

1. Introduction

A knowledge of annealing mechanism of nuclear tracks in thin sheets of polycarbonate and crystalline minerals has become of increasing interest in recent years due to their wide applications in fission track geochronology [1–5]. The production of tracks by energetic ions in solid state nuclear track detectors (SSNTDs) and the subsequent revealing of these tracks by chemical etching is widely used to detect and identify light and heavy ions [6–8]. Makrofol-N ($C_{16}H_{14}O_3$, density ~ 1.23 gm/cm^3, thickness ~ 300 μm (manufactured by Bayer, Germany), has been found to be very useful in the detection of heavy ions [9].

Most of the previous studies on thermal recovery of the damage are restricted to the fission fragment tracks in minerals, glasses and plastics [11–14]. These studies do not reflect the exact picture of the etching and annealing mechanism because of the lower range, unidentified mass and energy and higher statistical errors inherent in fission fragment tracks, especially in case of plastics. So,

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for the present studies the behaviour of ion tracks in the Makrofol-N plastic produced by well-defined beam of ^{238}U (17.17 and 16.34 MeV/u) ions is investigated.

2. Annealing mechanism

Modgil and Virk [15] postulated a three step annealing model which explains the annealing behaviour of radiation damage in bulk materials. The authors favoured the concept of a single activation energy of track annealing and proposed an empirical formula

$$V_a = At_a^{-n}\exp(-E_a/kT_a), \quad (1)$$

where V_a (μm/h) is the annealing rate, dl/dt_a (μm/h) i.e. the rate of change of track length, l (μm), with respect to annealing time, t_a (h), A the proportionality constant; k (eV/K), the Boltzmann constant, T_a (K) the annealing temperature, and E_a (eV), the activation energy for the annealing of latent tracks.

In a paper on phosphate glasses, Price et al. [16] found that their annealing data could fit into the equation of Modgil and Virk [15] provided the thermal annealing rate is replaced by the fractional thermal fading rate

$$\frac{(S-1)_i-(S-1)_f}{(S-1)_i} = At_a^{1-n}\exp(-E_a/kT_a), \quad (2)$$

where $S = V_T/V_G$ and subscripts i and f refer to the initial and final values. The authors have further proposed that using this modified version of Eq. (1), the values of parameters A, n and E_a are reduced to certain constants for a given detector, irrespective of the ion beam used. We took the challenge of testing this formulation on our recent data on annealing of ^{238}U ion tracks in the polycarbonate plastic Makrofol-N.

An attempt has been made by Bhatia and Virk [17] to introduce a new parameter, instantaneous track etch velocity on the left-hand side of Eq. (1) to improve upon the previous formulation as follows:

$$\frac{d(V_T/V_G)}{dt_a} = At_a^{-n}\exp(-E_a/kT_a), \quad (3)$$

where V_T/V_G is the track etch to bulk etch rate ratio for a particular annealing time and temperature and this formulation gives a best fit for annealing of heavy ion tracks in Lexan polycarbonate. Green et al. [14] have also formulated the concept of single activation energy based on the results of annealing experiments on apatite. There are only a few reports on annealing of plastic track recording materials. It is true that the complex nature of plastics makes it difficult to present a satisfactory explanation of annealing mechanism.

In this investigation we present comparisons of various etching parameters, viz., bulk etch rate, V_G, track etch rate, V_T, sensitivity, S, critical angle of etching (θ_c, and etching efficiency, η, before and after annealing. The activation energies for bulk etching, track etching and annealing have also been determined.

3. Experimental details

Different sets of Makrofol-N palstic track detectors are prepared for irradiations. These sets were exposed to ^{238}U (17.17 and 16.34 MeV/u) ion beams from UNILAC accelerator at GSI, Darmstadt, Germany. The incidence angle of beams with respect to detector surface was 45°. Some samples were annealed in a temperature controlled oven with an accuracy of ±2°C in the range 50–150°C for a fixed interval of 30 min. The corresponding track lengths of both the unannealed and annealed samples were recorded after etching the different sets of samples in 6.0 N NaOH at different temperatures (40, 50, 60, and 70°C).

4. Measurement of track etching and annealing parameters

For measuring the track etch rate V_T before and after annealing, the exposed samples (unannealed) were etched at various temperatures, viz., 40, 50, 60 and 70°C and annealed at only 60°C in 6.0 N NaOH by using constant temperature shaker water bath. After each etching interval, the samples were scanned under an optical microscope to record the projected etched track length

at a magnification of 600×. The etching and microscopic observations were repeated until the maximum track lengths became invariant with further etching. The slopes of linear portion of the graphs (Figs. 1 and 2) give the track etch rates of different annealed and unannealed samples for ^{238}U ions of 17.17 and 16.34 MeV/u, respectively. The bulk etch rates were calculated by the relation [10]

$$V_G = \frac{\Delta x}{2\Delta t}, \tag{4}$$

where Δx is the thickness of plastic dissolved in etching time Δt.

The track registration sensitivity is determined by using the relation V_T/V_G for ^{238}U (17.17 and 16.34 MeV/u) ion tracks in Makrofol-N and the etching efficiency, η, is determined by the relation

$$\eta = 1 - \sin\,\theta_c = 1 - V_G/V_T, \tag{5}$$

where θ_c is the critical angle of incidence for track etching.

The activation energies for bulk etching is calculated using following relation [18]

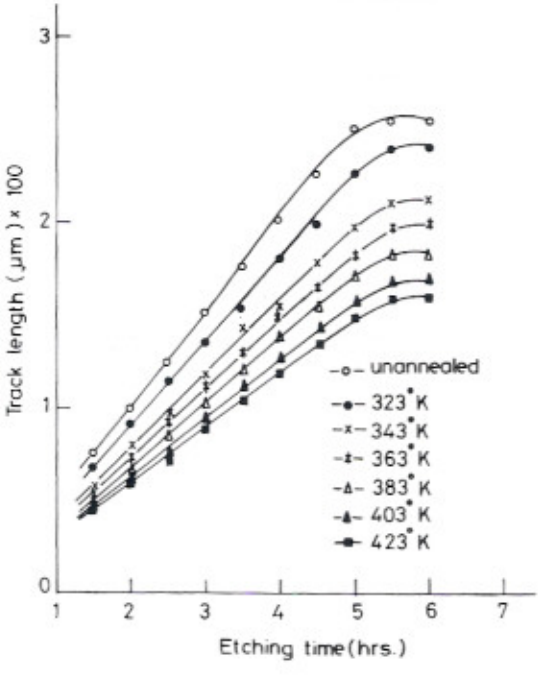

Fig. 1. Variation of track length with etching time (etched in 6.0 N NaOH at 60°C) for annealed and unannealed Makrofol-N irradiated with ^{238}U (17.17 MeV/u) ion.

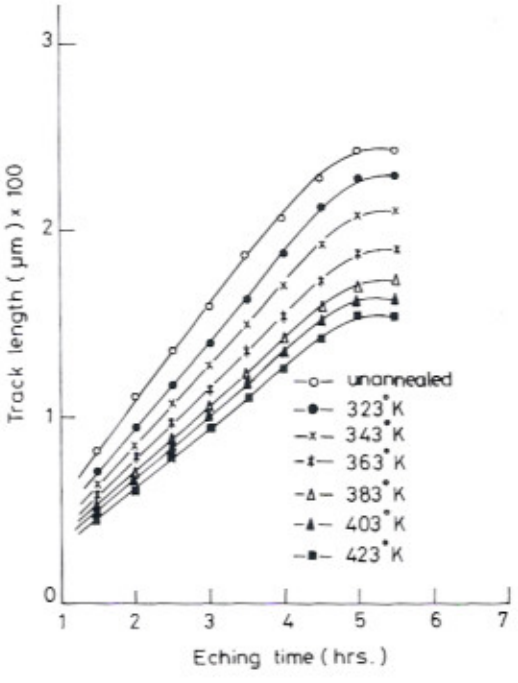

Fig. 2. Variation of track length with etching time (etched in 6.0 N NaOH at 60°C) for annealed and unannealed Makrofol-N irradiated with ^{238}U (16.34 MeV/u) ion.

$$V_G = A\,\exp(-E_G/kT), \tag{6}$$

where V_G is the bulk etch rate for unannealed sample, E_G is the activation energy for bulk etch rate,

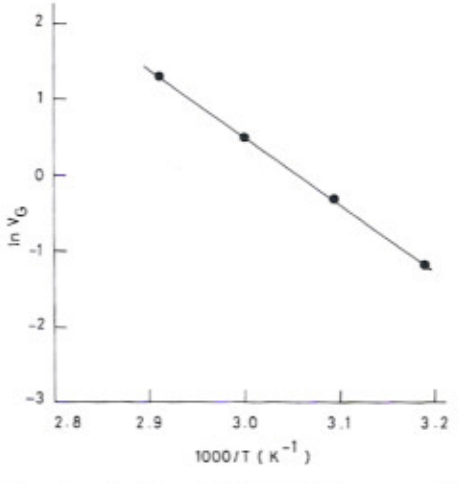

Fig. 3. The plot of ln V_G vs. $1000/T$ (K^{-1}) for unannealed Makrofol-N (etched in 6.0 N NaOH).

T is the etching temperature, k the Boltzmann constant and A is proportionality constant. E_G is determined from the slope of the graph (Fig. 3).

The activation energies for track etching is calculated using following relation [18]

$$V_T = B \exp(-E_T/kT), \quad (7)$$

where V_T is the track etch rate for unannealed sample, E_T is activation energy for track etching and B, k and T have usual meanings. E_T is determined from the slope of the graph (Fig. 4) for ^{238}U ion of 17.17 MeV/u energy.

The values of various etching parameters for unannealed samples, i.e. V_G, V_T, S, θ_c and η are given in Table 1. From this table it is concluded that the V_G, the bulk etch rate increases with increasing temperature of the etchant. The value of V_T also increases with the increase of etchant temperature. This value depends upon the incident particle energy. The values of the track registration sensitivity, S, at different temperatures have been determined and found to be almost same for a given energy of the ion. Hence, it can be concluded that for Makrofol-N the ratio V_T/V_G is generally independent of the etchant temperature. Table 2 gives activation energies for bulk and track etching. The activation energy for bulk etching, E_G, is found to be 0.77 eV. The value of E_G determined

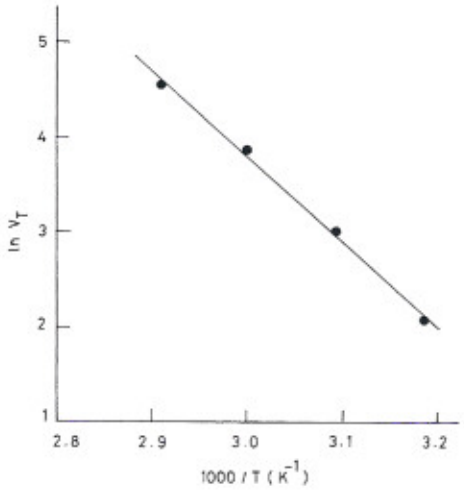

Fig. 4. The plot of ln V_T vs. 1000/T (K^{-1}) for unannealed Makrofol-N (etched in 6.0 N NaOH) for ^{238}U (17.17 MeV/u) ion.

Table 1
Values of etching parameters, V_G, V_T and S of ^{238}U (17.17 and 16.34 MeV/u) ion tracks for unannealed Makrofol-N (etchant 6.0 N NaOH).

Etchant temp. (°C)	V_G (μm/h)	V_T (μm/h)	S (V_T/V_G)
^{238}U ion of energy 17.17 MeV/u			
40	0.30	8.0	26.66
50	0.72	20.0	27.78
60	1.69	47.0	27.81
70	3.90	110.0	28.20
^{238}U ion of energy 16.34 MeV/u			
40	0.30	10.0	33.33
50	0.72	24.0	33.33
60	1.69	48.0	28.40
70	3.90	112.0	28.72

Table 2
Activation energy of bulk and track etching for ^{238}U ion irradiated Makrofol-N

Energy (MeV/u)	E_G (eV)	E_T (eV)
17.17	0.75±0.02	0.74±0.04
16.34	0.75±0.02	0.73±0.03

Table 3
Values of etching parameters, V_G, V_T and S of ^{238}U (17.17 and 16.34 MeV/u) ion tracks for annealed Makrofol-N (etchant 6.0 N NaOH at 60°C)

Annealing temp. (°C)	V_G (μm/h)	V_T (μm/h)	S (V_T/V_G)
^{238}U ion of energy 17.17 MeV/u			
50	1.69	45.0	26.63
70	1.67	43.0	25.75
90	1.63	41.0	25.15
110	1.60	38.0	23.75
130	1.55	34.0	21.93
150	1.54	30.0	19.48
^{238}U ion of energy 16.34 MeV/u			
50	1.69	47.0	27.81
70	1.67	45.0	26.94
90	1.63	42.0	25.76
110	1.60	40.0	25.00
130	1.55	36.0	23.22
150	1.54	34.0	22.08

by Enge [19] was found to be 0.65 eV. The activation energies for track etching, E_T, for ^{238}U ion of energy 17.17 and 16.34 MeV/u are found to be nearly equal, i.e. 0.74 and 0.73 eV, respectively.

The various etching parameters for annealed samples are given in Table 3. In case of annealed samples all etching parameters change after annealing at temperatures, viz, 50, 70 90, 110, 130 and 150°C. For annealed samples, bulk etch rate slightly decreases with increase of annealing temperature. By annealing studies, it is seen from Figs. 1 and 2, that the track etch rate, V_T, decreases with the healing of latent tracks. The track registration sensitivity, S, and etching efficiency, decrease with the annealing of latent tracks (Table 3). For the determination of activation energy for the annealing of tracks of ^{238}U ion in Makrofol-N Eqs. (2) and (3) have been used. The activation energy of annealing, E_a, has been determined from the slopes of the graphs (Figs. 5 and 6) for ^{238}U ion of energy 17.17 MeV/u by using the Eqs. (2) and (3), respectively. The determined values of activation energy for the annealing of ^{238}U ion tracks are 0.22 eV and 0.23 eV (Table 4) for 17.17 and 16.34 MeV/u energies, respectively.

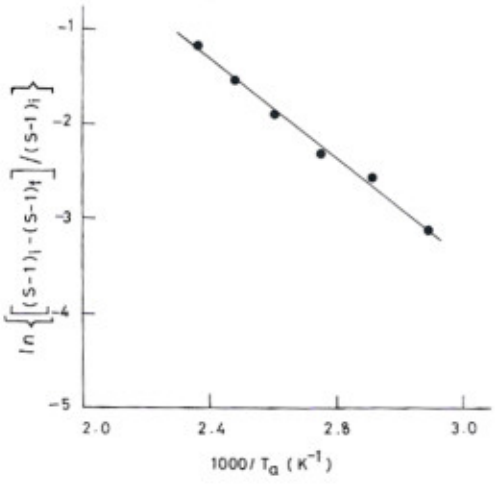

Fig. 5. Variation of ln $\{[(S-1)_i-(S-1)_f]/[S-1]_i\}$ vs. $1/T_a$ (10^3 K^{-1}) for annealing of tracks of ^{238}U (17.17 MeV/u) ion in Makrofol-N (etched in 6.0 N NaOH at 60°C).

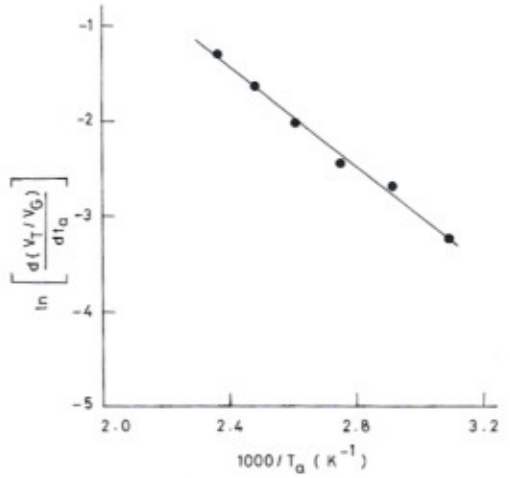

Fig. 6. Variation of ln $\left(\frac{d(V_T/V_G)}{dt_a}\right)$ vs. $1/T_a$ (10^3 K^{-1}) for annealing of tracks of ^{238}U (17.17 MeV/u) ion in Makrofol-N (etched in 6.0 N NaOH at 60°C).

Table 4
Activation energy values for annealing of tracks of ^{238}U ion in Makrofol-N.

Ion	Energy (MeV/u)	Activation energy of annealing (eV)	
		Formula of Price et al. [16]	Formula of Bhatia and Virk [17]
^{238}U	17.17	0.22 ± 0.04	0.22 ± 0.02
^{238}U	16.34	0.22 ± 0.02	0.23 ± 0.03

5. Range measurement of ^{238}U ion in Makrofol-N

For this investigation, the irradiated samples of Makrofol-N were etched in 6.0 N NaOH solution at a constant temperature of 60°C using a constant temperature bath. Etching of the tracks was performed for short intervals of time until the tips of the tracks became round. After successive etching, the samples were washed under running water for a few minutes and then dried in the folds of a tissue paper. The dried and etched samples were scanned at a magnification of 600×. After measuring the projected track length, the total etchable range was determined by applying the corrections due to bulk-etching and over-etching [20]. This total etchable range is taken as total range of ^{238}U

Table 5
Comparison between experimental and theoretical range values for ^{238}U ion in Makrofol-N

Energy (MeV/u)	R_{exp} (μm)	$R_{theoretical}$ (μm)		
		Mukherjee and Nayak [21]	TRIM-95 [23]	SRIM-97 [24]
17.17	254 ± 3	239 (−6)	288 (13)	283 (11)
16.34	242 ± 3	227 (−7)	274 (13)	270 (12)

Parentheses give percentage deviation of theoretical results from the experimental ones.

ion in Makrofol-N. The experimental range values have been compared with the theoretical range values from the Mukherjee and Nayak [21], and Ziegler et al. [22] formulations (Table 5). TRIM-95 [23] and SRIM-97 [24] based on the Ziegler et al. formulation are found to overestimate the experimental range values while, on the other hand, Mukherjee and Nayak formulation underestimates the experimental data.

6. Conclusions

1. The sensitivity of Makrofol-N to ^{238}U ion remains almost constant (within error of ±7%) with the increase of etchant temperature.

2. Price et al. [16] and Bhatia and Virk [17] formulations (both) can be used to explain the annealing kinetics of radiation damaged Makrofol-N plastic. The determined value of activation energy for annealing of heavy ion tracks is ~0.22 eV.

3. The theoretical range value computed from TRIM-95 and SRIM-97 based on Ziegler et al. formulation overestimates the experimental range values by more than 10% while Mukerjee and Nayak formulation underestimates the experimental results by 7%.

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References

[1] R.L. Fleischer, P.B. Price , R.M. Walker, Nuclear Tracks in Solids: Principles and Applications, University of California Press, Berkeley, Ca, 1975.
[2] M. Dakowski, J. Burchart, J. Galazaki, Bull. Acad. Sci. Ser. Sci. Terre 22 (1974) 11.
[3] G. Walder, T.D. Mark, Nucl. Instr. and Meth. B 32 (1980) 303.
[4] J.H. Adams, L.P. Beahm, Proceedings of the 11th International Conference on SSNTDs, Bristol, 1981, p. 163.
[5] A.S. Sandhu, J.A. Westgate, Earth Planet. Sci. Lett. 131 (1995) 289.
[6] S.M. Farid, Nucl. Instr. and Meth. 226 (1984) 501.
[7] G.S. Randhawa, H.S. Virk, Appl. Radiat. Isot. 46 (1995) 351.
[8] R.K. Jain, J. Rama Rao, S.K. Bose, K.K. Dwivedi, Nucl. Instr. and Meth. B 47 (1990) 48.
[9] A. Sexena, Measurement of heavy ion ranges in elemental and complex media using SSNTDs, Ph.D. Thesis, NEHU, Shillong, unpublished.
[10] S.A. Durrani, R.K. Bull, Solid State Nuclear Track Detection: Principles, Methods and Applications, Pergamon Press, Oxford, 1987.
[11] R. Gold, J.H. Roberts, F.H. Ruddy, Nucl. Tracks 5 (1981) 253.
[12] E. Dartyge, J.P. Durand, Y. Langevin, M. Maurette, Phys. Rev. B 23 (1981) 5213.
[13] T.D. Mark, R. Vartanian, F. Purtscheller, M. Pahl, Acta Phys. Austriaca 53 (1981) 45.
[14] P.F. Green, A.G. Ramli, S.A.R. Al-Najjar, P.P. Tingate, Nucl. Tracks Radiat. Meas. 10 (1985) 323.
[15] S.K. Modgil, H.S. Virk, Nucl. Instr. and Meth. B 12 (1985) 212.
[16] P.B. Price, G. Garbier, H.S. Park, M.H. Salamon, Nucl. Instr. and Meth. B 28 (1987) 53.
[17] R.K. Bhatia, H.S. Virk, Radiat. Eff. 107 (1989) 167.
[18] W. Enge, K. Grabisch, L. Dallmeyer, K.P. Bartholoma, R. Beaujean, Nucl. Instr. and Meth. 127 (1975) 125.
[19] W. Enge, Nucl. Tracks 4 (4) (1980) 283.
[20] K.K. Dwivedi, S. Mukherjee, Nucl. Instr. and Meth. 161 (1979) 317.
[21] S. Mukherjee, A.K. Nayak, Nucl. Instr. Meth. 159 (1979) 421.
[22] J.F. Ziegler, J.P. Biersack, U. Littmark, in: J.F. Ziegler (Ed.), The Stopping Power and Range of Ions in Solids, vol. 1, Pergamon Press, New York, 1985.
[23] J.F. Ziegler, TRIM-95: The Transport of Ions in Matter, IBM Research, 28-0, Yorktown, NY 10598, USA, 1995.
[24] J.P. Biersack, J.F. Ziegler, SRIM-97, Personal Communication.