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Study of charged particle tracks in barium phosphate (BP-1) glass

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Abstract

Samples of barium phosphate (BP-1) glass detector were irradiated by ^{238}U (11.4 MeV/u) ions from the UNILAC accelerator, at GSI, Darmstadt, Germany. Etching characteristics were studied using HF (48 vol%), NaOH (6.25N) and KOH (6.5N) at different temperatures. The sensitivity, $(S = V_t/V_b)$ of BP-1 glass is found to vary between 63 and 33 between temperature range 40–70°C. The experimental range of ^{238}U (11.4 MeV/u) ion in BP-1 has been determined and compared with the theoretical range values from different formulations. For study of annealing characteristics, both isothermal and isochronal experiments were performed on BP-1 glass irradiated with ^{238}U (11.4 MeV/u) and fission fragments from ^{252}Cf source. The annealing kinetics of heavy ion damaged BP-1 glass is explained by using different empirical formulations. The activation energy of annealing for heavy ion damaged BP-1 glass is found to be 0.405 ± 0.005 eV. © 1997 Elsevier Science B.V.

Keywords: BP-1 phosphate glass; ^{238}U heavy ion; Fission fragments; Annealing; Etching; Range

1. Introduction

Scientific and technological applications of solid state nuclear track detectors (SSNTDs) have stimulated efforts to develop better track recording solids [1]. The sensitivity of these track detectors can be represented by the ratio of track etch rate, V_t and bulk etch rate, V_b, i.e. V_t/V_b, or the minimum detectable value of charge to velocity ratio $(Z/\beta)_{min}$. In the past few years, a number of new latent track recording materials, viz., glasses, minerals, plastics, etc., have been reported in the literature. For heavy ion studies, glass track detectors play an important role due to some advantages over other detectors like discrimination of light charged particles, being free from mechanical ruggedness, being less prone to environmental changes and aging effects [2], etc. A large number of plastic track detectors have been developed showing high sensitivity to charged particles but their response is strongly dependent in a complicated way on the temperature, pressure and other environmental factors at the time of irradiation [3]. The outcome of the efforts made by different researchers working in this field led to the development of many new glass track detectors. Soda and quartz glasses have been used as track recorders successfully by different groups in the literature. Aschenbach et al. [4] identified ZnP glass with very steep response as a function

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of Z/β which they found to be superior to all other glasses tested by them. Other glasses have been studied from time to time by many groups [5–7], but none have shown promise of being useful in the study of relativistic heavy nuclei. In a search of new glass track detectors with both high resolution and high sensitivity, Price et al. [8,9] have found a very sensitive phosphate glass detector VG-13. However, VG-13 glass has a number of disadvantages due to its uranium content and it cannot be used in radiation environments where sizable concentration of neutrons is present, which may induce fission of the present uranium. It is mildly hazardous to handle and it corrodes quickly in air. Wang et al. [10] made a systematic search of the phosphate glasses of various compositions which were free from uranium. This work resulted in the development of barium phosphate (BP-1) glass, a glass with highest charge resolution and sensitivity among glasses. Moreover, like other glasses, the sensitivity of BP-1 is unaffected by the presence or absence of oxygen, so it can be used in space for identification of ultraheavy nuclei and isotopes of heavy nuclei in cosmic rays [11]. In the present investigation, a quantitative analysis of various etching parameters of BP-1 has been made by etching in different solutions at different temperatures. The experimental range value of ^{238}U ions in BP-1 is compared with the theoretical results.

2. Chemical etching of BP-1

The samples of BP-1 glass track detectors were irradiated from the UNILAC accelerator at GSI, Darmstadt, Germany by ^{238}U ions having energy 11.4 MeV/u. All these irradiations were carried out with ion fluences of 10^4 ions/cm^2 at angles of 45° with respect to the surface of the detector. These irradiated samples were etched in a series of constant temperatures of 40°C, 50°C, 60°C and 70°C in 48 vol% HF solution. After etching, the samples were washed in running water and then dried in the folds of a tissue paper. The etched and dried samples were scanned under a Carl Zeiss binocular microscope. The bulk etch rate, V_b is determined by using two different techniques: (1) the thickness measurement technique, and (2) the weight loss method.

The track etching parameters, viz., the sensitivity, the etching efficiency and the critical angle of etching have been determined by using the following relations [12]:

$$\text{sensitivity } S = V_t/V_b, \tag{1}$$

$$\text{track etching efficiency } \eta = (1 - V_b/V_t), \tag{2}$$

$$\text{critical angle of etching } \theta_c = \sin^{-1}(V_b/V_t). \tag{3}$$

The track etch rate, V_t is determined by taking the slope from the linear portion of the plot between track length and etching time.

The variation of track length with etching time for ^{238}U (11.4 MeV/u) ion in BP-1 glass at different temperatures of the etching solution (48 vol% HF) is plotted in Fig. 1. The values of average track etch rates have been determined by taking the slope from the linear portion of these plots. The values of sensitivity (S), track etching efficiency (η) and critical angle of etching (θ_c) are summarized in Table 1. From the table, it is evident that sensitivity, S, decreases with the increase of temperature of the etching solution. In order to find the optimum etching conditions, BP-1 glass has also been etched in 6.25N NaOH and 6.5N KOH solutions. It is clear that the value of S is more in

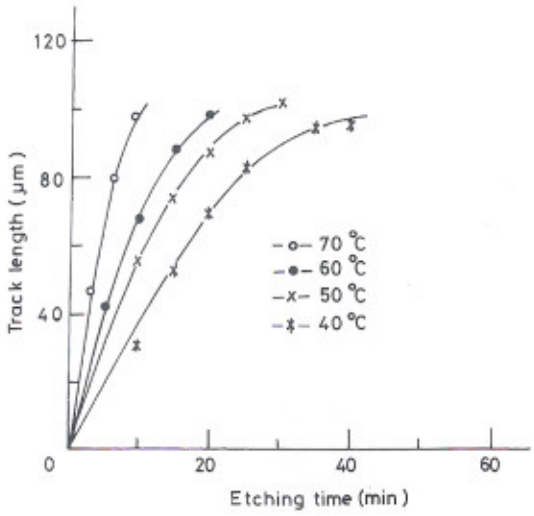

Fig. 1. Variation of track length with etching time (etched in 48 vol% HF) for ^{238}U (11.4 MeV/u) ion in BP-1 glass.

Table 1
Track-etch characteristics of BP-1 glass track detector etched in different etchants

Etching solution	Temperature of etching (°C)	Sensitivity $S = V_t/V_b$	Etching efficiency η (%)	Critical angle θ_c
48 vol% HF	40	63.46	98.42	0.90
	50	55.55	98.20	1.03
	60	49.03	97.96	1.16
	70	38.33	97.39	1.49
6.25N NaOH	40	48.73	97.94	1.18
	50	43.05	97.67	1.33
	60	33.56	97.02	1.70
	70	33.36	97.01	1.71
6.5N KOH	40	54.60	98.16	1.04
	50	44.35	97.74	1.29
	60	36.78	97.28	1.55
	70	35.48	97.18	1.62

case of 48 vol% HF than 6.25N NaOH and 6.5N KOH solutions. Hence, the 48 vol% HF is the most suitable etchant for the development of tracks in BP-1 glass. In comparison to 6.25N NaOH, 6.5N KOH solution gives somewhat better results.

The bulk etch rate, V_b and the track etch rate, V_t have exponential dependence on the temperature of etching solution as given by the relation [13]

$$V_b = A_b\, e^{-E_b/kT} \quad \text{and} \quad V_t = A_t\, e^{-E_t/kT}, \tag{4}$$

where A_b and A_t are the constants, E_b and E_t the activation energies for bulk and track etching, respectively and k the Boltzmann constant. The values of E_b and E_t are calculated by plotting ln V_b and ln V_t versus $(1000/T)$ for each detector. The determined values of activation energies for bulk and track etching are 0.76 and 0.35 eV, respectively (etchant 48 vol% HF).

3. Range measurement of ^{238}U ion in BP-1 glass

For this investigation the irradiated samples of BP-1 glass were etched in 48% HF solution at a constant temperature of 50 ± 1°C using a cryostat. 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 ×1000. After measuring the projected track length, the total etchable range was determined by applying the corrections due to bulk etching and over etching [14]. This total etchable range is taken as the range of ^{238}U ion in BP-1 glass. The experimental range value has been compared with the theoretical range values from the Benton and Henke [15], Mukherjee and Nayak [16] and Ziegler et al. [17] formulations (Table 2). TRIM-95 [18] based on the Ziegler et al. formulation gives the best agreement with the experimental range value in comparison to the results from the other formulations.

Knowing the total etchable range, it is possible to determine the residual range of the different heavy ion

$$\text{Residual range} = \text{Total etchable range} - 1/2 \text{ track length},$$
$$\text{RR} = R - L/2. \tag{5}$$

Table 2
Comparison between experimental and theoretical range values for ^{238}U (11.4 MeV/u) ion in BP-1 glass

R_{exp} (μm)	$R_{theoretical}$ (μm)		
	[15]	[16]	[18]
101.09 ± 1.53	82.58 (−18) [a]	93.87 (−7)	100.03 (−1)

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

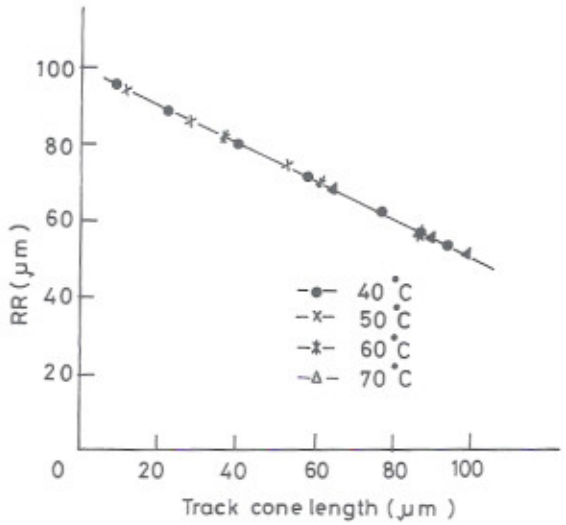

Fig. 2. Variation of residual range (RR) with track cone length (etched in 48 vol% HF) for ^{238}U (11.4 MeV/u) ion in BP-1 glass.

The track cone length, L and residual range, RR, gives a (L, RR) pair which forms the basis of single sheet particle identification [19] technique. Due to the high sensitivity of the BP-1 glass, it can be used for mass and charge resolution. Fig. 2 gives the variation of the track cone length versus residual range (etched in 48 vol% HF) for ^{238}U (11.4 MeV/u) in BP-1 glass. The falling of all points on a single line indicates the suitability of the BP-1 glass for single sheet particle identification technique. An interesting point also noted here is that the variation of track cone length with residual range is independent of the temperature of the etchant.

4. Annealing of heavy ion damaged BP-1 glass

The latent tracks in any material undergo annealing in the heating environment. The phenomenon of thermal annealing of these latent tracks in nuclear track recording materials has been studied by many authors during the last three decades. But due to the lack of a satisfactory theory of track formation in different materials, the phenomenon of thermal annealing of these latent tracks is not fully understood. The phenomenon of annealing of radiation damage in different dielectrics has been studied by several researchers [20–26] and a number of empirical formulations have been proposed to explain the annealing mechanism as a function of both annealing time and temperature. Many of the authors consider a latent track as an aggregate of defects and kinetics of annealing is described by Arrhenius relation, $\exp(-E_a/kT)$ where E_a, k and T are the activation energy of annealing, the Boltzmann constant and the annealing temperature, respectively [1]. The track fading occurs as interstitially displaced atoms thermally penetrate the activation barrier to recover their initial lattice positions. It has been found that the activation energy associated with the repair of a damaged material is of the order of a few electron volts, which is typical energy involved in atomic diffusion. Hence it is assumed that the annealing of damage materials occurs due to diffusion of defects in inorganic materials and movement of molecular fragments within a polymer. The interstitial atoms tend to return to their normal sites and the broken molecular chains may join and recombine with other active species when insulating solids are heated.

Mark et al. [21] proposed that the annealing of fission tracks in apatite material can be explained by a summation series of exponential functions and suggested that the annealing behaviour of track densities (ρ) in apatite could be explained by the relationship

$$\rho(t) = \rho(o)\ \exp\ (-\alpha(T)t), \tag{6}$$

where $\rho(t)$ is the track density after annealing time t and $\rho(o)$ the original track density. In this case the annealing coefficient $\alpha(T)$ is given by a sum of two exponentials

$$\alpha(T) = \alpha_{01}\ \exp(-E_{01}/kT) + \alpha_{02}\ \exp(-E_{02}/kT), \tag{7}$$

where α_{01} and α_{02} are the annealing constants and E_{01} and E_{02} the activation energies of two diffusion processes involved. However, at elevated temperature annealing, the data of Mark et al. [24] approximated with a single exponential decay function. The two exponential equation (7) can be reduced to a single exponential approximation as

$$\alpha(T) = \alpha_o \exp(-E_a/kT). \tag{8}$$

Using the above relation, Eq. (6) can be written as

$$\ln(-\ln r) = \ln \alpha_o - E_a/kT + \ln t, \quad (9)$$

where $r = \rho(t)/\rho(o)$ is the track density reduction ratio.

Mark et al. [24] assumed the validity of $\rho/\rho_o \approx l/l_o \approx D/D_o$ (for apatite); where l and D are the track length and track diameter after annealing time, t and l_o and D_o are the original track length and track diameter, respectively.

Hence Eq. (9) to a first approximation, reduces to

$$\ln(-\ln (l/l_o)) = \ln \alpha_o + \ln t - E_a/kT. \quad (10)$$

A plot between $\ln(-\ln (l/l_o))$ versus $(1/T)$ produces line of slope E_a/k. This model has been used to explain the annealing of tracks in different materials.

Modgil and Virk [25] proposed a three step annealing model based on the diffusion processes. This empirical formulation relating the annealing rate, V_a (rate of change of length, l, or diameter, D with annealing time, i.e., dl/dt or dD/dt) with the activation energy, E_a as follows:

$$V_a = At^{-n} \exp(-E_a/kT), \quad (11)$$

where A is a proportionality constant, n, the exponent of annealing time t and the other symbols have their usual meanings. In order to determine the activation energy the equation can be rewritten as

$$\ln V_a = \ln A - n \ln t - E_a/kT. \quad (12)$$

This model favoured the concept of a single activation energy of annealing for a given nuclear track recording material exposed to different heavy ions and fission fragments.

Green et al. [26] postulated a model describing the thermal annealing of tracks in inorganic solids, by using a different approach for presenting their experimental data through a best fit

$$\ln t - C_1 \ln (1 - l/l_o) = C_2 + C_3/T, \quad (13)$$

where C_1, C_2 and C_3 are constants which may be determined from the experimental data. This model predicts parallelism of lines of equal track length reduction on the Arrhenius plot and therefore a single activation energy can be determined. This equation is modified as:

$$\ln(1 - l/l_o) = -C_2/C_1 + (1/C_1)\ln t - (C_3/C_1)/T, \quad (14)$$

$$\ln(1 - l/l_o) = \ln A_1 + n_1 \ln t - E_a/kT, \quad (15)$$

where $\ln A_1 = -C_2/C_1, n_1 = 1/C_1$ and $E_a/k = C_3/C_1$.

In the present investigation, the annealing behaviour of latent tracks in BP-1 glass irradiated with ^{238}U (11.4 MeV/u) and fission fragments from ^{252}Cf radiative source has been studied. Eqs. (10), (12) and (15) have been used for the determination of activation energy from different models proposed by Mark et al. [21], Modgil and Virk [25] and Green et al. [26], respectively. The irradiated samples of BP-1 glass by ^{238}U (11.4 MeV/u) heavy ion at an angle of 45° with respect to the surface of the detector and by the fission fragments of ^{252}Cf at 90° were cut into small pieces and annealed at temperatures of 300°C, 350°C, 400°C, 450°C for 5 and 10 h. These annealed samples were etched in 48 vol% HF solution at 50°C temperature for 30 min. The values of mean track length in case of ^{238}U (11.4 MeV/u) and track diameter for fission fragment tracks in BP-1 glass are measured.

Figs. 3–5 show the variation of $\ln(-\ln (l/l_o))$, $\ln V_a$ and $\ln(1 - l/l_o)$ with the annealing temperature, $1000/T$ (K^{-1}) for the calculation of activation energy of annealing of tracks of ^{238}U (11.4 MeV/u) from formulations of Mark et al. [21], Modgil and Virk [25] and Green et al. [26],

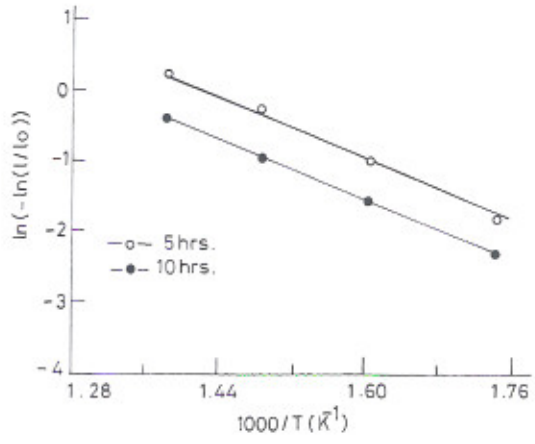

Fig. 3. Variation of $\ln(-\ln(l/l_o))$ versus $1/T$ (10^3 K^{-1}) for annealing of tracks of ^{238}U (11.4 MeV/u) ion in BP-1 glass.

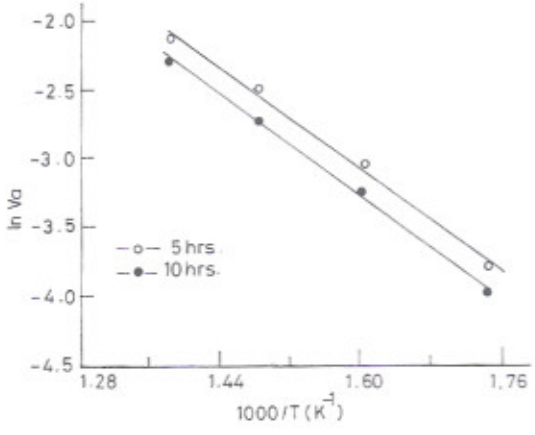

Fig. 4. Variation of ln (V_a) versus $1/T$ $(10^3\ K^{-1})$ for annealing of tracks of ^{238}U (11.4 MeV/u) ion in BP-1 glass.

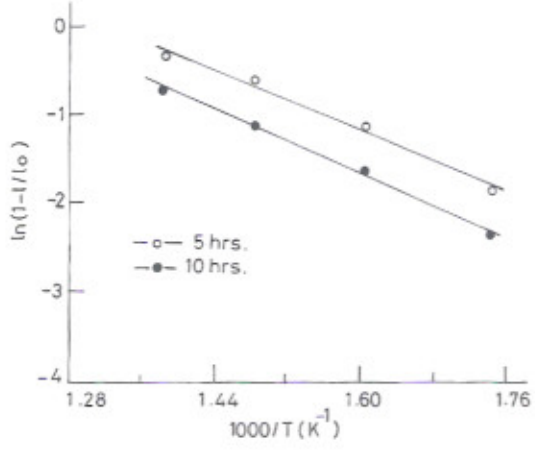

Fig. 5. Variation of ln $(1 - I/I_0)$ versus $1/T$ $(10^3\ K^{-1})$ for annealing of tracks of ^{238}U (11.4 MeV/u) ion in BP-1 glass.

respectively. The values of activation energy of annealing of latent tracks of ^{238}U and fission fragments in BP-1 glass are given in Table 3. The Green et al. and Modgil and Virk formulations yield the identical values of $E_a(0.405 \pm 0.005\ eV)$ for ^{238}U heavy ion and fission fragments. The values of activation energy, E_a are found to be 0.47 and 0.50 eV for ^{238}U (11.4 MeV/u) ion and fission fragments, respectively, using the Mark et al. formulation. Price et al. [27] studied the rate of thermal annealing of tracks of relativistic heavy ions, viz., U (930 and 350 MeV/u), Au (1000 MeV/u) and La (600 MeV/u) in VG-13 phosphate glass and a single value of activation energy 0.48 eV is determined. Singh and Virk [28] made a similar study on another type of phosphate glass exposed to Pb (17.0 and 13.6 MeV/u) and La (14.6 MeV/u) heavy ions. A single value of activation energy of annealing 0.56 eV is determined for this phosphate glass. Hence, a similar conclusion can be made for the BP-1 glass that the activation energy of annealing of latent tracks is 0.405 eV.

Table 3
Activation energy values calculated from three different formulations for annealing of tracks of ^{238}U (11.4 MeV/u) heavy ions and ^{252}Cf fission fragments in BP-1 glass detector

Ion	Energy (MeV/u)	Activation energy of annealing (eV)		
		[21]	[25]	[26]
^{238}U	11.4	0.47	0.40	0.40
Fission fragments		0.50	0.41	0.41

5. Conclusions

(a) The sensitivity of BP-1 glass decreases with the increase of etchant temperature and HF (48 vol%) is the most suitable etchant in comparison to NaOH (6.25N) and KOH (6.5N) solutions.
(b) The theoretical range value computed from TRIM-95 based on the Ziegler et al. formulation gives the best agreement with the experimental value.
(c) The Modgil and Virk [25] and Green et al. [26] formulations can be used to explain the annealing kinetics of radiation damaged BP-1 glass. The determined value of activation energy is 0.405 eV.

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