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THERMAL ANNEALING OF PROTON TRACKS WITH ENERGIES OF 4 AND 6 MeV IN CR-39 POLYMER DETECTORS

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Abstract—In this paper, we study the thermal annealing of proton tracks of 4 and 6 MeV at temperatures ranging from 150 to 240°C in CR-39 polymer detectors. A special experimental set-up for irradiating the detectors was arranged to obtain adequate proton beams from the Cyclotron CV-28 at IPEN/SP, Brazil. We report experimental data on track densities, track diameters, and activation energies based on current annealing models for the annealing of proton tracks in the energy range investigated. A value of (0.20 ± 0.02) eV was determined as the mean activation energy of the annealing process in CR-39 detectors.

1. INTRODUCTION

Studies of the properties of track registration for charged particles in CR-39 solid state nuclear track detectors (SSNTD) are of current interest (Snowden-Ifft, 1993; Al-Jarallah *et al.*, 1993) especially in the case of thermal annealing of tracks (Virk, 1992; Karamdoust and Durrani, 1991). The reason for this interest is the wide use of the CR-39 polymer detectors in the studies such as nuclear reactions, neutron dosimetry and particle identification. However, it is still necessary to have a better understanding of the properties of track formation and of the thermal annealing process in CR-39 detectors. In the last case, the latent damage trails produced by charged particles can be totally or partially removed by the heating of these detectors for a sufficient length of time, so that the tracks cannot be registered by subsequent etching or their registration by etching is impaired (Durrani and Bull, 1987).

Studies of the annealing of proton tracks of energies between 0.1 and 20 MeV in CR-39 detectors are of considerable importance in neutron dosimetry and other applications with SSNTD where thermal effects are relevant. In the present paper, we study the thermal annealing of proton tracks at a temperature range from 150 to 240°C, with 4 and 6 MeV energies from the Cyclotron CV-28 at the Instituto de Pesquisas Energeticas e Nucleares/SP, Brazil, in CR-39 polymer detectors. A special experimental set-up to irradiate the detectors was arranged to obtain adequate proton beams.

Although there are numerous investigations of the thermal annealing process of latent tracks in minerals and glasses and present efforts in CR-39 detectors, this process is not yet fully understood. The purpose of this paper is to contribute some new experimental data about the annealing process of tracks in CR-39 and to discuss them based on current annealing track models (Märk *et al.*, 1973; Märk *et al.*, 1981; Green *et al.*, 1985; Modgil and Virk, 1984; Dartyge *et al.*, 1981). Thus, we report experimental data on track densities, track diameters, and activation energies for the annealing of proton tracks in CR-39 detectors in the energy range investigated.

2. ANNEALING MODELS

Annealing is a process by which the radiation damage produced in SSNTD at elevated temperatures is completely or partially removed. It has been found that the activation energies associated with the repair of damage trails are of the order of a few electron-volts (Fleischer *et al.*, 1975) which is typical of energies involved in atomic diffusion, and clear evidence of the atomic nature of the defects. Thus, the annealing models presume that the annealing of damage trails occurs due to diffusion of atomic defects through the crystal lattice or movement of molecular fragments within a polymer. The interstitial atoms tend to return to their normal sites and the broken molecular chains may rejoin and recombine with other active species when insulating solids are heated. Although some aspects of the annealing mechanism are well understood, the difficulty of calculating the changes in track structure as a function of annealing time and temperature has not yet been resolved (Durrani and Bull, 1987). A number of models (Märk *et al.*, 1973; Märk *et al.*, 1981; Green *et al.*, 1985; Modgil and Virk, 1984; Dartyge *et al.*, 1981) have been proposed to describe the kinetics of the annealing of radiation damage in various SSNTD as a function of annealing time and temperature. Märk *et al.* (1973) proposed a model where the track density reduction ρ/ρ_0 corresponds to track length reduction l/l_0 or diameter reduction D/D_0 and the annealing behaviour is given by

$$\rho(t) = \rho(0)\exp[-\alpha(T)t] \quad (1)$$

where $\rho(0)$ and $\rho(t)$ are the track densities at time $t = 0$ and t, respectively, and $\alpha(T)$ is an annealing coefficient which is given by the sum of two exponentials

$$\alpha(T) = \alpha_{01}\exp(-E_{a1}/kT) + \alpha_{02}\exp(-E_{a2}/kT) \quad (2)$$

where α_{01} and α_{02} are annealing constants, E_{a1} and E_{a2} are two different activation energies of two different diffusion processes involved, k is the Boltzmann constant and T, the absolute temperature. At elevated annealing temperatures, equation (2) can be written in the single exponential approximation

$$\alpha(T) = \alpha_0\exp(-E_a/kT). \quad (3)$$

From equations (1) and (3), we have

$$\ln(-\ln\rho/\rho_0) = \ln\alpha_0 + \ln t - E_a/kT. \quad (4)$$

Märk *et al.* (1981) assumed the validity of

$$\rho/\rho_0 \sim l/l_0 = D/D_0. \quad (5)$$

Then, equation (4) can be rewritten as

$$\ln(-\ln D/D_0) = \ln\alpha_0 + \ln t - E_a/kT. \quad (6)$$

Thus a plot of $\ln(-\ln D/D_0)$ versus $1/T$ produces straight lines of slopes E_a/k. This model has been used to explain the annealing of tracks in different SSNTDs. However, its only shortcoming is the hypothesis of two activation energies for the annealing process.

The model proposed by Green *et al.* (1985) described the thermal annealing of tracks in inorganic solids, particularly in apatite mineral, by using a different approach for presenting their experimental data through a best fit, using the expression

$$\ln t - C_1\ln(1 - l/l_0) = C_2 + C_3/T \quad (7)$$

where C_1, C_2 and C_3 are constants which may be determined from the experimental data. Equation (7) can be used for different models of isothermal and isochronal annealing. This model predicts parallelism of lines of equal track length reduction on the Arrhenius plot and therefore a single activation energy can be determined. An adequate and modified form of equation (7) is given by

$$\ln(1 - D/D_0) = -C_2/C_1 + (1/C_1)\ln t - C_3/C_1T \quad (8)$$

or in the form

$$\ln(1 - D/D_0) = \ln A' + n'\ln t - E_a/kT. \quad (9)$$

where $\ln A' = -C_2/C_1$, $n' = 1/C_1$ and $E_a/k = C_3/C_1$.

Modgil and Virk (1984) postulated a three-step annealing model relating annealing rate V_a with activation energy E_a as

$$V_a = At^{-n}\exp(-E_a/kT) \quad (10)$$

where A is the proportionality constant and n the exponent of annealing time. The annealing rate is defined as the rate of change of track length $(\Delta l/\Delta t)$ or diameter $(\Delta D/\Delta t)$. In order to determine the activation energy, equation (10) can be written in the form

$$\ln V_a = \ln A - n\ln t - E_a/kT. \quad (11)$$

In this model, the authors favoured the concept of a single activation energy of track annealing and proposed an empirical formula, equation (10), where the annealing rate is a function of annealing time and temperature. The activation energy can be determined from the slope of the plot of $\ln V_a$ versus $1/T$.

Dartyge *et al.* (1981) proposed a "gap model" to explain the radiation damage in dielectric minerals (muscovite mica, labradorite, pyroxene and olivine) from X-ray scattering studies. In this model, the latent tracks are constituted of extended defects, separated by gap zones loaded with point defects, and in this case the extended defects play the dominating role in the etching of tracks because of their annealing. According to the authors, upon thermal annealing, the extended defects are much more stable than point defects. Therefore, there are two different mechanisms explaining the annealing of latent tracks. This model will not be useful to calculate a single activation energy.

In the present paper, the above mentioned annealing models are used and compared with our data for thermal annealing of proton tracks of energies 4 and 6 MeV in CR-39 detectors.

3. EXPERIMENTAL

The irradiations in the samples of CR-39, supplied by Track Analysis Systems Ltd., TASTRAK, Bristol U.K., thickness between 650 and 850 μm, dimensions of approximately 3.0 cm × 1.5 cm, were carried out by means of proton beams in the Cyclotron CV-28 at the IPEN/CNEN/SP. An experimental set-up was arranged to obtain adequate proton beams in the detectors of about 10^2–10^4 protons/cm^2, conditions for optimum track density. In Fig. 1, we show the experimental set-up. At the scattering chamber, the incident proton beam of 23 MeV was collimated to a 3 mm diameter which then hit upon a 10 μm Ta target positioned normal to the incident beam. The samples of CR-39 were placed at 30° with respect to the scattered centre at a distance of 30 cm. A Faraday

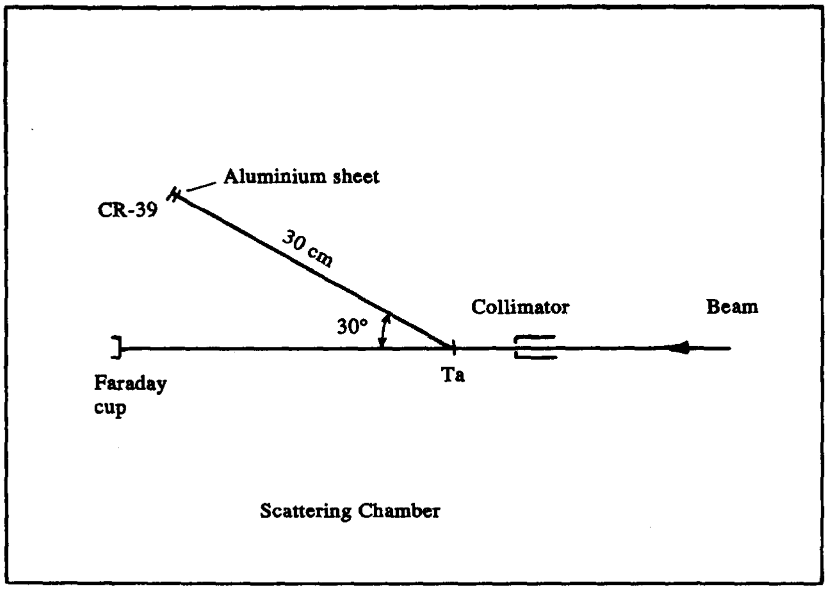

FIG. 1. Experimental set-up.

cup was used to control the number of incident protons in the samples during each irradiation. The energies of 4 and 6 MeV were obtained by superposition of 2500 and 2330 μm thick aluminium sheets over the CR-39 surface, respectively, in each irradiation with proton beams of 23 MeV. We have used the formalism of Andersen and Ziegler (1977) to calculate the energy loss of protons in aluminium. In a previous paper (Pinheiro Filho *et al.*, 1988) we have already described in detail the calculations of the energy lost per unit length of protons and heavy ions in nuclear emulsion with the use of this type of formalism.

The samples of CR-39 irradiated with protons were immersed in silicon oil and annealed in a controlled muffle furnace set for temperatures of 150, 180, 210 and 240°C for 30 min. These samples, for each energy set studied, were etched together with the unannealed samples in 6.25 N NaOH at 70°C for 16 h. This etching condition is employed after studies for determining the adequate etching conditions of proton track revelation in CR-39 detectors in the energy range investigated. The CR-39 detectors were analysed for measurements of track density and track diameter in optical microscopes Jena with 40 × and 20 × objectives and 10 × and 7 × oculars fitted with a moving filar micrometer attachment which was calibrated using a standard scale.

4. RESULTS AND DISCUSSION

The results obtained in measurements of the mean diameter of proton tracks of 4 and 6 MeV at temperatures ranging from 423 to 513 K for annealing times of 30 min in CR-39 detectors may be seen in Table 1. The mean diameters D_0 of unannealed samples were also measured taking into account the background tracks of CR-39 unirradiated samples of about (24 ± 7) tracks/cm^2 with diameters from 3 to 14 μm. The track diameters of circular tracks only were

Table 1. The annealing data for track diameter reduction (D/D_0) and annealing rate ($V_a = \Delta D/\Delta t$) of protons with energies of 4 and 6 MeV in CR-39 detectors for annealing times of 30 min

E (MeV)	T (K)	$(1/T) \times 10^3$ —	Mean diameter D (μm)	D/D_0	Annealing rate V_a (μm/min)
4	Unannealed	—	11.57 ± 0.14	—	—
	423	2.36	7.8 ± 0.1	0.674	0.126
	453	2.21	6.8 ± 0.3	0.588	0.159
	483	2.07	4.4 ± 0.3	0.380	0.239
	513	1.95	3.1 ± 0.1	0.268	0.282
6	Unannealed	—	10.28 ± 0.08	—	—
	423	2.36	7.5 ± 0.1	0.730	0.093
	453	2.21	6.3 ± 0.1	0.613	0.133
	483	2.07	4.3 ± 0.1	0.418	0.199
	513	1.95	3.0 ± 0.1	0.292	0.243

considered and the background tracks observed were not significant over the temperature range 423–513 K. The track diameter reduction D/D_0 and the annealing rate $V_a = \Delta D/\Delta t$ are summarized in Table 1. We may observe that the mean diameter of proton tracks of 4 and 6 MeV decrease with the increase of the temperature in the range from 423 to 513 K. The mean track diameter reductions are of 32.6 and 73.2% in 4 MeV and 27.0 and 70.8% in 6 MeV for the annealing temperatures of 423 and 513 K, respectively. Thus, the track diameter reduction D/D_0 is relevant with the increase of the temperature between 423 and 513 K. Otherwise, the annealing rate V_a rises with the increase of the temperature in the interval investigated (423–513 K) for proton tracks of 4 and 6 MeV.

The thermal effects vary significantly between 150 and 240°C on the properties of track registration in CR-39 detectors. For instance, we have observed in unannealed samples of CR-39 a proton track density of about (725 ± 27) tracks/cm^2 and for annealed samples at 240°C about (75 ± 9) tracks/cm^2, i.e. a reduction of 89.7% which is greater than the mean reduction track diameter of 72.0% at 240°C obtained for the energies of 4 and 6 MeV.

For unannealed samples of CR-39 detectors, the bulk etch rate, $V_B = (1.31 \pm 0.13)\ \mu$m/h, was determined directly from the measurements of thickness of samples before and after etching. The track etch rates V_T for the two proton energies of 4 and 6 MeV were also determined from measurements of track diameter D and thickness h of the removed surface layer of the detector giving (1.52 ± 0.15) and $(1.48 \pm 0.15)\ \mu$m/h, respectively, by using the well-known formula

$$V_T = V_B(4h^2 + D^2)/(4h^2 - D^2). \quad (12)$$

The above mentioned data of the bulk and track etch rates of the unannealed samples allow us to determine the critical angle θ_c for proton track registration given by

$$\theta_c = \sin^{-1}(V_B/V_T) \quad (13)$$

which is dependent on the proton energy since the criterion for track formation is

$$V_T \sin\theta > V_B \quad (14)$$

where θ is the angle of incidence of the proton relative to the surface of the detector. We found in the energies of 4 and 6 MeV for the critical angle the values of 60° and 62°, respectively. These values are in good agreement with other recent determinations (Al-Jarallah *et al.*, 1993; Dörschel *et al.*, 1991).

In Table 2, we show the values of the activation energies determined from our experimental data of thermal annealing of proton tracks using different annealing models in this investigation. The plots for the activation energies determined by equations (6), (9) and (11) related to proton tracks of 4 and 6 MeV over the temperature interval of 423 to 513K for annealing times of 30 min in CR-39 detectors are shown in Figs 2, 3 and 4, respectively.

Table 2. The values of activation energies determined from our experimental data of thermal annealing of proton tracks using different annealing models in CR-39 detectors for annealing times of 30 min

	Activation Energy E_a (eV) Annealing Models		
Proton energy	Märk *et al.* (1981)	Green *et al.* (1985)	Modgil and Virk (1984)
4 MeV	0.26	0.18	0.18
6 MeV	0.29	0.21	0.21

Our results show that the annealing models of Green *et al.* (1985) and Modgil and Virk (1984) give the same values for the activation energies 0.18 and 0.21 eV, for proton tracks of 4 and 6 MeV, respectively. These values are in good agreement with existing determinations of activation energy for the annealing of heavy ion tracks of ^{238}U (10 and 16 MeV/nucleon) and ^{93}Nb (18 MeV/nucleon) in CR-39 by Virk *et al.* (1986) where the values 0.185 and 0.193 eV were found, respectively, and by Khan *et al.* (1984) who found the value of 0.194 eV for heavy ions of ^{238}U (16 MeV/nucleon). The values 0.26 and 0.29 eV for the activation energies determined from the model of Märk *et al.* (1981) are then greater than the values obtained with the annealing models

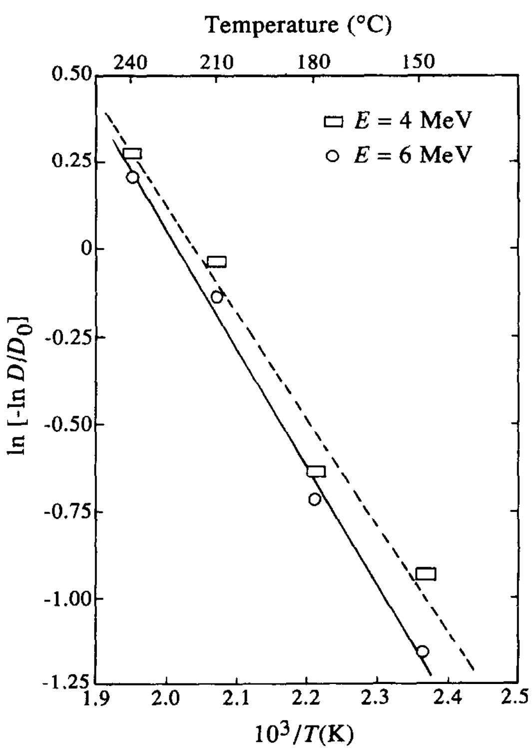

FIG. 2. The variation of $\ln(-\ln D/D_0)$ versus $1/T$ for annealing of proton tracks of 4 and 6 MeV in CR-39. The straight lines are least-squares fits of the experimental points (dashed line, $E = 4$ MeV and full line, $E = 6$ MeV).

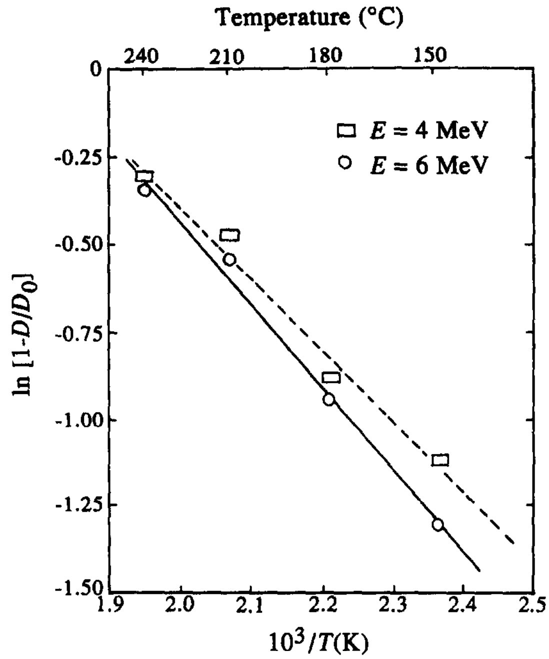

FIG. 3. The variation of $\ln(1 - D/D_0)$ versus $1/T$ for annealing of proton tracks of 4 and 6 MeV in CR-39. The straight lines are least-squares fits of the experimental points (dashed line, $E = 4$ MeV and full line, $E = 6$ MeV).

considered in this work (see Table 2), which may be comparable with other experimental data (Virk *et al.*, 1986; Khan *et al.*, 1984) in CR-39 detectors. Recently

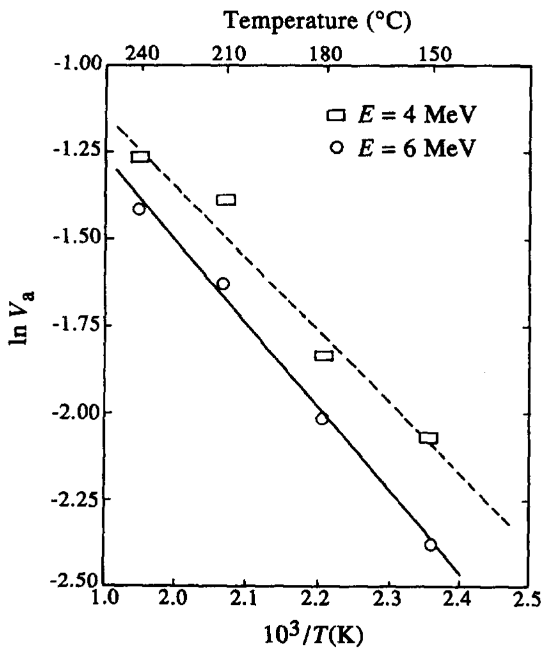

FIG. 4. The variation of ln V_a versus $1/T$ for annealing of proton tracks of 4 and 6 MeV in CR-39. The straight lines are least-squares fits of the experimental points (dashed line, $E = 4$ MeV and full line, $E = 6$ MeV).

Virk (1992) reported annealing data of activation energies from experiments carried out by his group for various types of SSNTD using the formalism of Modgil and Virk (1984). These activation energies for heavy ions of ^{93}Nb (18.0 MeV/n), ^{208}Pb (17.0 MeV/n) and ^{238}U (10.0 MeV/n) in CR-39 detectors presented 0.20 eV for each heavy ion analysed. These last results are in good agreement with our present experimental data (see Table 2) if we only consider the annealing models of Green *et al.* (1985) and Modgil and Virk (1984).

5. CONCLUSIONS

1) The effects of thermal annealing on the properties of density and diameter of proton tracks are significant in CR-39 detectors.

2) The annealing models of Green *et al.* (1985) and Modgil and Virk (1984) give the best fit for determining the activation energy for proton tracks in CR-39 detectors and as a consequence, a value of (0.20 ± 0.02) eV was determined as the mean activation energy of the annealing process.

3) The concept of a single annealing activation energy is fully justified in the case of CR-39 polymer detectors.

4) The experimental method used in this work is very adequate for studying the annealing process of proton tracks in CR-39.

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