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Impact of electron irradiation on particle track etching response in polyallyl diglycol carbonate (PADC)

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Abstract. In the present work, attempts have been made to investigate the modification in particle track etching response of polyallyl diglycol carbonate (PADC) due to impact of 2 MeV electrons. PADC samples pre-irradiated to 1, 10, 20, 40, 60, 80 and 100 Mrad doses of 2 MeV electrons were further exposed to 140 MeV ^{28}Si beam and dose-dependent track registration properties of PADC have been studied. Etch-rate values of the PADC irradiated to 100 Mrad dose electron was found to increase by nearly 4 times that of pristine PADC. The electron irradiation has promoted chain scissioning in PADC, thereby converting the polymer into an easily etchable polymer. Moreover, the etching response and the detection efficiency were found to improve by electron irradiation. Scanning electron microscopy of etched samples further revealed the surface damage in these irradiated PADCs.

Keywords. 2 MeV electron; 140 MeV ^{28}Si; PADC; dose-dependent track registration properties; bulk etch-rate; etching response; critical angle of etching; detection efficiency; scanning electron microscopy.

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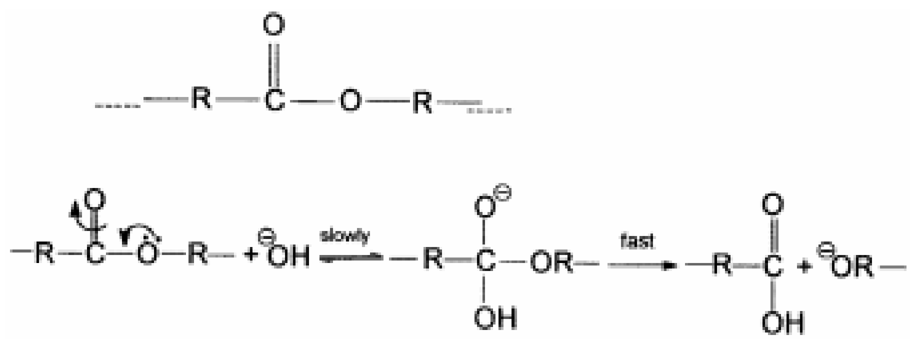
1. Introduction

The use of ion beams for polymer modification requires information on morphology of tracks created by the impinging ions. Tracks of energetic particles in polymers are complicated primarily because of greater multiplicity of chemically active defects that can occur – displaced atoms, broken molecular chains, free radicals etc. Since energy transfers needed to produce these defects are generally lower, polymers are a class of more sensitive track detectors. In polymers, the track core consists of a zone of drastically reduced molecular weight, corresponding to broken molecular bonds, surrounded by track halo where the chemically reactive species undergo secondary reactions. Chemical etching transforms the latent tracks into optically visible tracks by supplying the required amount of energy for enlargement process. Amount of chemical change depends both on total quantity of radiation energy available and on the rate at which energy is deposited. Linear energy transfer (LET) is a measure of rate of energy deposition and is defined as the linear rate of loss of energy by an ionizing particle traversing a material medium.

When the projectile consists of electrons, collision between two particles of equal mass can transfer whole of kinetic energy to the stationary particle. Large energy transfers are thus possible and path-length is much less well-defined. Further, a light particle is much more readily deviated by the nucleus and so scattering angles will be larger and the path-length much more crooked. It follows that range of an electron is not necessarily as great as its path-length. At higher energies (kinetic energy >1 MeV), the energy-loss of an electron is chiefly due to bremstrahlung and the probability of this process increases with energy. Electrons are low LET radiations, which mostly affect the physical and chemical properties of polymeric films. In the last few years, lot of work has been done to characterize and improve track detectors for better detection sensitivity [1] as well as charge and energy resolutions. The nuclear track technique is quite versatile [2], simple and accurate. The importance of latent tracks lies in the fact that they are the characteristic signatures of the charged particles producing them. The shape of the etched track is formed as a result of competition between bulk-etch rate and track-etch rate.

PADC belongs to the polyen of the type –CH_2–CHR– containing the diethyleneglycolcarbonate. All polymers containing the ester functional group in their molecule can be written as



Such a polymer is capable of alkali hydrolysing reaction by the above mentioned mechanism. PADC is essentially one macromolecule with less positive density and stronger space shielding effect. Therefore it is insoluble and not easily amenable to alkali hydrolyse. Due to this, the bulk etching rate of this polymer is less compared to that of other detectors.

PADC is a highly sensitive track recording plastic. It is a thermoset polymer that do not cross-link upon irradiation and are susceptible to interfacial degradation by a convenient etchant which constitutes an ideal polymer detector. Their amorphous nature and radiation sensitivity further enhance the track detection property [3]. It is about 100 times more sensitive than polycarbonate.

Earlier studies indicate a modification in track registration properties of PADC on exposure to high gamma dose [4–11].

In the present study, the electron dose dependence on track registration properties of PADC, the most widely used polymer for charge detection, has been investigated. Moreover, the surface damage has been analysed by scanning electron microscope.

2. Experimental details

2.1 *Preparation of the targets*

PADC samples (composition: $C_{12}H_{18}O_7$, density: 1.32 g.cm^{-3}) of sizes 2×2 cm^2 were cut from commercially available sheets (thickness ≈ 1.5 mm), manufactured by Homalite Corporation, Wilmington, Del., USA. After removing the surface protective layers, these detector plates were washed thoroughly with soap solution and then with deionized water to remove surface contamination. The cleaned samples were then dried inside a vacuum desiccator.

2.2 *Irradiation and cooling*

Irradiation of the targets was done by 2 MeV electron beam from an electron generator at Hahn-Meitner Institute, Berlin. The electron beam was passed through a collimator and was allowed to fall on the target placed at a distance of 2 metres from the collimator. The dose of 2 MeV electron beam was varied, to carry out a dose dependent study. The doses used were 1, 10, 20, 40, 60, 80 and 100 Mrad. Irradiated samples were allowed to cool for about 24 h to allow induced radioactivity to fall below the safe limits of handling. The samples were then taken and preserved in plastic boxes.

2.3 *Exposure to 140 MeV ^{28}Si*

PADC detectors pre-irradiated to different doses of 2 MeV electron beam, were again exposed to well collimated beam of ^{28}Si ions with an initial energy of 140 MeV. The beam was allowed to fall on the gold foil scatterer and the optimum flux of ^{28}Si was measured to be 10^5 ions.cm^{-2} sec^{-1}. The current measured by the Faraday cup on the scatterer during irradiation was 0.8 pnA. The irradiation was done in 20° port of general purpose scattering chamber (GPSC) of 15 UD Pelletron Accelerator at the Nuclear Science Centre, New Delhi. The scattered beam was allowed to fall at an angle of 45° on the targets mounted on a specially designed zig-zag ladder. After irradiation, the samples were allowed to cool and then they were stored in plastic boxes.

2.4 *Chemical treatment*

The samples were washed thoroughly in lukewarm soap solution to avoid non-uniformity in etching due to surface contamination. Then the cleaned samples were etched in 6 N NaOH solution at an etching temperature of 55°C.

Successive etching was performed till the tracks were completely etched. Then the samples were washed in running water and dried under vacuum.

2.5 *Measurement of track parameters*

The track parameters were measured by Leitz optical microscope at a magnification of 625x. The diameters of 50 silicon ion tracks were measured for each detector to find out the most probable track-diameters at different etching times. The track lengths and the corresponding track-diameters were measured to an accuracy of $\pm 1.12\ \mu$m. The bulk-etch rate (V_G) were calculated from the slope of the plot between silicon ion track diameters vs. etching time. After every successive etching the track-lengths were measured at random all over the detector surface to average out the effects due to non-uniformity of the targets. The true maximum etchable track-lengths were then calculated from projected track lengths. The track-etch rate (V_T) were calculated from the slope of the plot between silicon ion track lengths vs. etching time. The errors associated with etch-rate measurements lies between 0.3 to 0.6 μm/h.

2.6 *Scanning electron microscopy of the etched samples*

The etched samples were washed thoroughly in running water and then dried. Gold plating of all the samples was done by ion sputter JFC-1100. The gold coating of thickness 150 nm was applied on the PADC samples under vacuum at 10 mA and 1 KV conditions. Gold plating was done to make the polymer surface conducting. After gold plating was done the system was allowed to cool and the samples were taken out from the chamber. Scanning electron microscopy of these coated samples were done by JSM-35 CF scanning electron microscope under complete vacuum conditions. The surface damage were studied by the photo-micrographs of the samples.

3. Theoretical aspects

The increase of track-diameter with etching time gives a measure of the bulk-etch rate (V_G) along the surface plane

$$V_G = (d/2t), \tag{1}$$

where, d is the diameter of the track after etching time t. The true track lengths (L) were then calculated from measured projected track lengths using the formulations [12] as follows:

$$L = (l/\cos\varphi) + (V_G t/\sin\varphi) - V_G(t - t_C), \tag{2}$$

where, l is the projected track length as observed by the microscope, φ is the angle of irradiation, $(V_G t/\sin\varphi)$ is the surface etching correction, t is the etching time, t_C is the complete etching time that corresponds to a fixed value of track diameter which is independent of temperature of a given etchant and dielectric [13]. $V_G(t - t_C)$ is the overetching correction.

The track-etch rate, V_T can be obtained by measuring the track length increase at different etching times. If over a small etching time period, Δt, the increase in track length is ΔL, then,

$$V_T \cong \Delta L/\Delta t. \quad (3)$$

If, V_T is a varying function of etching time, the critical angle of etching $\theta_{\rm C}$ is given by

$$\theta_{\rm C} = \sin^{-1}(V_G/V_T). \quad (4)$$

Critical angle of etching is the angle at or below which the damage trail of an impinging particle is not detectable. Critical angle in turn controls the efficiency of the detector. The detection efficiency of the detector is the ratio of the number of tracks created to the number of impinging particles, which can be calculated using the formula

$$\eta = 1 - \sin\theta_{\rm C}. \quad (5)$$

4. Results and discussion

The silicon track diameters were found to be an increasing function of dose. The bulk etch-rate, V_G was calculated from the slope of the plot of track diameters versus etching time. The V_G of PADC is shown in figure 1 as a function of electron dose. Though all PADCs were irradiated by ^{28}Si under similar conditions, they have different bulk-etch rates. The bulk-etch rate of PADC pre-irradiated to 100 Mrad dose of 2 MeV electron was found to be nearly 4 times that of pristine PADC which is not irradiated to electron as shown in table 1. Though the electrons cannot create etchable tracks of their own, they can affect etch rate values of the detectors and other heavy ion tracks depending on the absorbed dose [4]. With electron irradiation, the damage is dispersed in an atomic scale, so that it is not possible to see the individual etched features resulting from the defects responsible for accelerated etching. Electron irradiation has promoted chain scissioning, thereby increasing the V_G and converting them into rapidly etchable materials. As the particle slows down, its ionization rate increases. The track-etch rate (V_T) increases with ionization rate. Energy loss rate slightly increases along track trajectory. Therefore, increase in V_T is probable with increasing etched track length [14]. The track etch-rate was also found to be increasing with increase in electron dose as shown in figure 2. The etching response is the etch rate ratio V_T/V_G which was found to increase with increase in electron dose as shown in table 1. Variation of etching response in PADC with electron dose is shown in figure 3. The etching response is also known as the etching sensitivity of the detector. The larger its value, the better is the track developing efficiency of the detector. So, the electron irradiation has improved the sensitivity of the detector. In other words electron irradiation can be used to sensitize the detector. The critical angle of etching, $\theta_{\rm C}$ was found to decrease with electron dose as evident from table 1. The cone angles of etched tracks are bigger for detectors having higher values of critical angle. The detection efficiency of the detector was found to improve by increasing the electron dose on PADC, as shown in table 1.

The scanning electron micrographs of the samples after complete etching, shown in figure 4, revealed that the diameters of silicon tracks are maximum for the PADC pre-irradiated to 100 Mrad dose. The etched tracks are of larger diameters in the electron irradiated samples. In the pristine sample we observe that the surface other than the etched tracks is smooth. The sample surface other than the tracks shows maximum damage in case of electron irradiated samples. This extra damage is caused due to electron irradiation. It can also be observed from the photomicrographs that the portion of the sample other than the tracks has suffered maximum damage in the 100 Mrad dose electron irradiated sample.

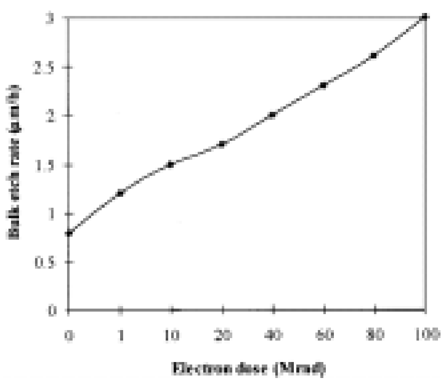


Figure 1. Variation of bulk etch-rate in PADC with electron dose.

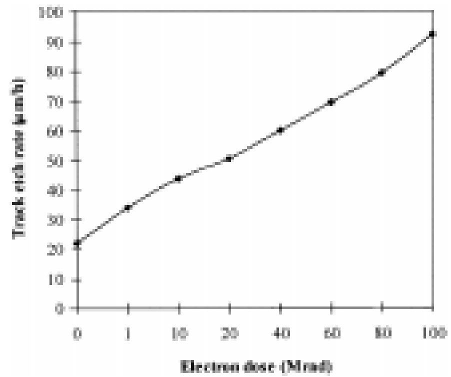


Figure 2. Variation of track etch-rate in PADC with electron dose.

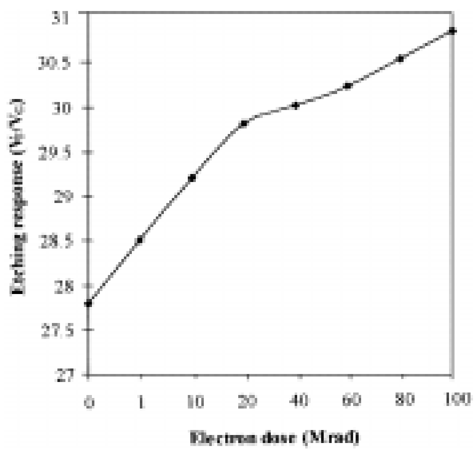


Figure 3. Variation of etching response (V_T/V_G) in PADC with electron dose.

Table 1. Variation of bulk-etch rate, etching response, critical angle of etching and detection efficiency with electron dose.

Electron dose (Mrad)	Track etch rate (μm/hr)	Bulk etch rate (μm/hr)	Etching response (V_T/V_G)	Critical angle of etching (θ_C in degrees)	Detection efficiency (η)
Pristine	22.2	0.8	27.8	2.1	0.96
1 Mrad	34.2	1.2	28.5	2.0	0.96
10 Mrad	43.8	1.5	29.2	1.96	0.97
20 Mrad	50.6	1.7	29.8	1.92	0.97
40 Mrad	60.0	2.0	30.0	1.91	0.97
60 Mrad	69.5	2.3	30.2	1.90	0.97
80 Mrad	79.4	2.6	30.5	1.88	0.97
100 Mrad	92.3	3.0	30.8	1.86	0.97

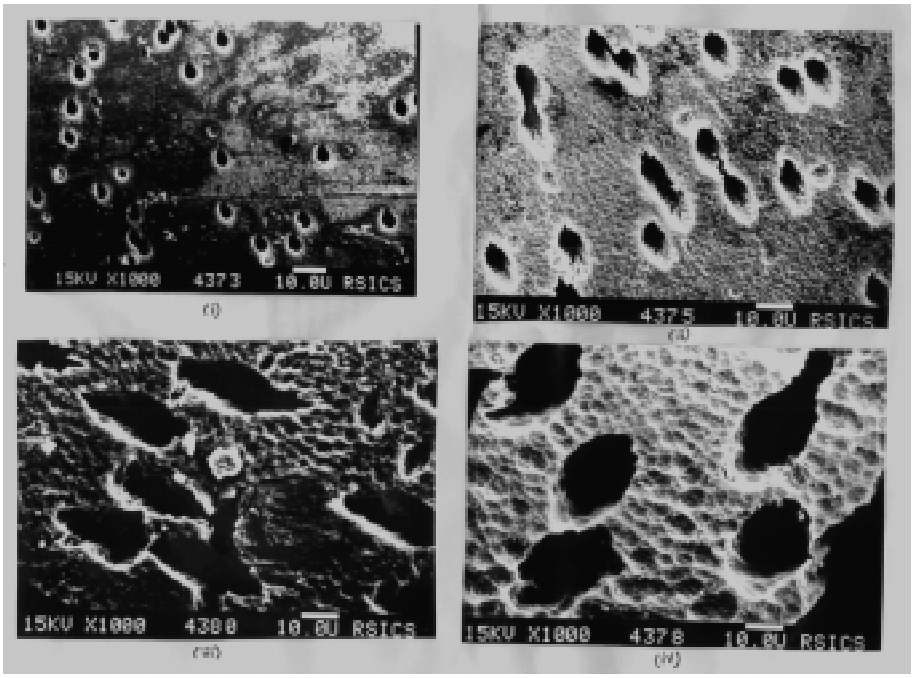


Figure 4. Photomicrographs of silicon tracks in PADCs clearly showing the increase of track diameter with increase in electron dose. (i) Pristine, (ii) 40 Mrad, (iii) 60 Mrad, (iv) 100 Mrad electron.

5. Conclusion

Dependence of track parameters on different doses of 2 MeV electrons have been studied. The electron irradiation has promoted chain-scissioning in PADC, which in turn has affected the etch-rates of PADC detectors. Bulk etch rate and track etch rate are directly related to electron dose. The bulk etch rate of PADC irradiated to 100 Mrad dose of electron was found to be 4 times higher than that of pristine PADC. The electron irradiation has made the PADC easily etchable. Moreover the PADC irradiated to 100 Mrad electron dose requires a comparatively less time for complete etching. Electron irradiation has improved the etching response and the detection efficiency of the detector. Thus, it can be concluded that the heavy ion track registration capacity of the PADC can be improved by preirradiating it with electrons. In other words, the electron irradiation can be used to sensitize the detector, PADC.

Acknowledgements

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