A comparative study of alternative methods to determine the response of poly-ethylene terephthalate nuclear track detector

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Abstract

Two widely used methods of determining the etch-rate ratio in poly-ethylene terephthalate (PET) nuclear track detector are compared. Their application in different regimes of ion's energy loss is investigated. A new calibration curve for PET is also presented.

Keywords: Nuclear Track Detector, Poly-ethylene Terephthalate, Restricted Energy Loss

1. Introduction

Nuclear track detectors (NTDs) find wide use in charged particle detection [1, 2]. They are particularly suited in the search for rare, highly ionizing particles against a large background from low ionizing particles [3, 4, 5, 6, 7, 8].

A charged particle losing energy while passing through an NTD, can create a permanent trail (“latent track”) of damaged bonds along its path [1]. On being treated with suitable reagents (chemical etching), if the material along the damage trail is etched out at a faster rate (the track etch-rate, \( V_T \)) than the etch-rate of the undamaged material (the bulk etch-rate, \( V_B \)), a conical

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etch-pit is formed. In nuclear track detectors the size and shape of etch-pits depend on the particle’s energy loss - more precisely on the restricted energy loss (REL) - along its path and on its angle of incidence [9, 10]. The minimum REL allowing the formation of an etch-pit, i.e. such that $V_T > V_B$, sets the detector threshold. For a particle impinging perpendicularly to the detector surface, if its energy loss is above threshold and constant, the etch-pit has the shape of a right circular cone. The process is similar to the Mach cone produced by an object moving in a medium at constant supersonic velocity. $V_T$ and $V_B$ play the role of the velocity of the object and that of the sound in the medium, respectively. We can identify the angle $\alpha$ (Fig. 1a) with the Mach angle and $\sin \alpha = V_B/V_T$. The shape of the etch-pit will not be a right-circular cone (or obliquely cut right-circular cone in case of angular incidence) if $dE/dx$ (as well as REL) varies along the latent track. The steepness of the etch-pit’s wall increases (Fig. 1b) until the energy loss reaches the Bragg peak [1]. Thus the etch-pit will look like a flared cone [11] as sketched in Fig. 1b.

The aim of this paper is to determine the etch-rate ratio of poly-ethylene terephthalate (PET) for ions with energy in the range 0.7 – 11.1 MeV/nucleon.

2. Methods to determine the etch-rate ratio

Different methods can be used to calculate the etch-rate ratio $V = V_T/V_B$ from the dimensions of the etch-pits. In this paper, two such methods are compared, which, in general, relate to different regimes of the ion’s energy loss. By one method [12], $V_T$ is calculated by measuring the length $L_h$ (Fig. 1a) of the etched out section of the latent track and the etching time ($t$). The bulk etch-rate $V_B$ is determined by the change in the thickness of the detector sheet over the etching time. With reference to Fig. 2h, for a particle impinging at an angle $i$ with respect to the normal to the detector surface, one has

$$\frac{V_T}{V_B} = \frac{d + V_B t}{V_B t \cos i} (i < \alpha) ; \quad \frac{V_T}{V_B} = \frac{\mu d' + V_B t}{V_B t \cos i} (i > \alpha)$$

where $d$ (or $d'$) is the vertical distance between the post-etching surface and the tip of the etch-pit cone, as measured by the microscope. The refractive index
Figure 1: Sketch of an etch-pit along the latent track of an ion incident normally to the NTD’s surface. The energy loss is (a) constant, (b) increasing along the ion’s trajectory. Here $\Delta t$ is the etching time at which the etchant reaches the level of the post-etch surface along the track.
Figure 2: Etched latent track when the ion impinges on the NTD’s surface at an angle (a) $i < \alpha$ and (b) $i > \alpha$. Of the detector material, $\mu$, has to be taken into account for $i > \alpha$ (Fig. 2b).

The refractive index of PET is $\mu_{\text{PET}} = 1.64 \pm 0.02$ in yellow light. It was determined following the same procedure as in [13] and it is identical to the value given in [14]. Henceforth this method of determining $\frac{V_T}{V_B}$ is referred to as “depth measurement method”.

By another method, the average $\frac{V_T}{V_B}$ can be determined from the size of the surface etch-pit opening [13, 16], using Eq. (2)

$$\frac{V_T}{V_B} = \sqrt{1 + \frac{4A^2}{(1 - B^2)^2}}$$

where $A = \frac{a}{v_{ni}}$ and $B = \frac{b}{v_{ni}}$; $a$ and $b$ are the semi-major and the semi-minor axis of the elliptical opening of the etch-pit, respectively. For a homogeneous and isotropic material, $B \leq 1$. For a particle impinging normally to the detector surface, the opening of the etch-pit is circular, and $a = b$. This method is
referred to in the following as “diameter measurement method”.

It has to be noted that Eq. (2) provides the average of \( V_T/V_B \) over the length \( L_d = V_B t V/(V + 1) \) from the pre-etching surface (Fig. 1a), whereas with Eq. (1) the average is over the length \( L_h = (d + V_B t)/\cos i \) or \( L_h = (\mu d' + V_B t)/\cos i \). Therefore for a slowing down electrically charged particle, the etch-rate ratio computed using Eq. (1) yields a value larger than the one computed using Eq. (2), since \( V_T/V_B \) is an increasing function of \( REL \) \[1, 13, 17\].

3. Determination of \( REL \)

At the energies of ions used in this paper, the restricted energy loss for NTDs can be computed using Eq. (3) \[1\]

\[
\frac{dE}{dx} \bigg|_{E < E_{cut}} = C_1 \left( \frac{z^*}{\beta} \right)^2 \left[ \ln \left( \frac{W_{max} E_{cut}}{I^2} \right) - \beta^2 - \delta \right]
\]

where \( C_1 = 2\pi n_e e^4/mc^2; n_e \) is the number density of electrons in the detector; \( m_e \) is the electron mass; \( z^* \) is the effective charge of the incoming particle \[1\]

\[
z^* = z \left[ 1 - e^{-(-130\beta/\beta'^2)} \right]
\]

\( z \) and \( \beta \) being the particle’s electric charge and velocity in units of electron charge \( e \) and speed of light \( c \), respectively; \( W_{max} = 2m_e c^2 \beta^2 \gamma^2 \) in the “low-energy” approximation \[18\] is the maximum energy that can be transferred to an electron in a single collision; \( \gamma \) is the Lorentz factor; \( \delta \) is the density-effect correction term due to the polarization of the medium, relevant at relativistic energies; \( I \) is the material mean ionization potential \( (I = 73.2 \text{ eV for PET} \[19\]); \( E_{cut} \) is the maximum energy of delta-rays contributing to the formation of the latent track. Hereinafter, \( E_{cut} = 350 \text{ eV} \) is assumed to compute \( REL \) in polyethylene terephthalate. For calculating the average of \( REL \) along the ion’s trajectory, the Monte Carlo code SRIM \[20\] was used.

4. Experimental method

In order to compare the “depth measurement method” to the “diameter measurement method” when \( B \approx 1 \), we used 3.8 MeV/A \(^{35}\text{Cl}^{10+}\) beam from
the pelletron accelerator and the General Purpose Scattering Chamber at the Inter-University Accelerator Center (IUAC), New Delhi. Details of the beam are given in Table 1. Small pieces (5 cm × 5 cm) of 90 µm thick PET films (Desmat Co., India), were mounted on the aluminum holders and placed on the two arms inside the scattering chamber (Fig. 3). PET films were irradiated by \(^{35}\text{Cl}^{10+}\) ions backscattered from a gold foil target 250 µg cm\(^{-2}\) thick. Exposure duration was controlled such that the ion density on the detector never exceeded \(\sim 10^4/\text{cm}^2\) to prevent detector “burnout”. After the exposure, the detectors were etched in a tank (Julabo, Germany) equipped with a motorized stirrer, in 6.25 N NaOH aqueous solution at 55.0 ± 0.1°C for 3 hours. After etching, the detectors were observed under a Leica DM4000 B optical microscope with a 100x objective and 10x eyepieces. The image analysis software QWin was used for the measurement of etch-pits’ sizes.

5. Results

Data obtained from previous exposures of PET films and from the exposure to \(^{35}\text{Cl}^{10+}\) ions are summarized in Table 2 and Table 3. As shown in the last
Table 1: Properties of the $^{35}$Cl$^{10+}$ beam. Beam energy has an uncertainty $< 5\%$.

| Ion  | Energy per nucleus (MeV) | Beam current (nA) | Charge state |
|------|--------------------------|-------------------|--------------|
| $^{35}$Cl | 132                     | 15                | 10           |

two rows of Table 3 the values of $V_T/V_B$ for $^{35}$Cl$^{10+}$ ions of 70 MeV/nucleus and 77 MeV/nucleus, determined using Eq. (2) are significantly different, although the corresponding values of REL are very close. This happens because of the $(1 - B^2)$ term in the denominator of Eq. (2), when $B \sim 1$ (columns 6 of table 3 and Fig. 4).

| Ion  | Energy per nucleon (MeV/A) | Incidence angle | Time of etching (hours) | Depth measurement (µm) | Major-axis (µm) | Minor-axis (µm) |
|------|---------------------------|-----------------|-------------------------|------------------------|----------------|---------------|
| $^{30}$Si$^+$ | 2.1                       | 0°              | 1.8                     | 6.53 ± 0.30           | 2.87 ± 0.15   | 2.75 ± 0.14   |
|      | 3.4                       | 0°              | 2.0                     | 5.32 ± 0.29           | 2.91 ± 0.14   | 2.86 ± 0.14   |
| $^{39}$Cl$^+$ | 1.0                       | 0°              | 3.0                     | 5.11 ± 0.29           | 3.62 ± 0.15   | 3.34 ± 0.15   |
|      | 1.2                       | 0°              | 3.0                     | 4.05 ± 0.30           | 3.54 ± 0.16   | 3.21 ± 0.16   |
| $^{13}$C$^{4+}$ | 0.7                       | 25°             | 4.0                     | 5.12 ± 0.29           | 4.50 ± 0.14   | 4.09 ± 0.14   |
|      | 0.9                       | 25°             | 4.0                     | 4.03 ± 0.29           | 3.46 ± 0.14   | 2.76 ± 0.15   |
| $^{35}$Cl$^{10+}$ | 2.0                       | 0°              | 3.0                     | 12.25 ± 0.29          | 5.94 ± 0.15   | 5.87 ± 0.15   |
|      | 2.2                       | 0°              | 3.0                     | 10.81 ± 0.29          | 5.29 ± 0.14   | 5.26 ± 0.14   |

Table 2: Data used to compute $V_T/V_B$ using Eq. (1) and Eq. (2). Ions’ energies and angles of incidences have an uncertainty $< 10\%$ and $< 4\%$, respectively. Errors in column 5, 6 and 7 include statistical and systematic uncertainties.

It should be mentioned here that there exist innovative ways of increasing the sensitivity of $B$ as the so-called “two-step etching” process [22]. However this method is inapplicable here due to a comparatively shorter range of the incoming particles. As shown in Fig. 4 the normalized semi minor-axis $B$ levels out to $\approx 1$ at $REL > 17$ MeV/mg cm$^{-2}$ (similar to what observed in CR-39 at $REL \sim 6$ MeV/mg cm$^{-2}$ [23]). Such reduced sensitivity of the etch-pit diameter (or minor-axis) at large RELs [13] along with systematic uncertainties on $a$, $b$ and $V_B$ compels one to switch to the depth measurement method for $V_T/V_B$ determination.
Table 3: Data for the different ions incident on PET. Errors on REL are the standard deviation of values computed along the latent track. In column 6, the mean value of the semi-minor axis (b), normalized to the bulk etch length (V_B) [21] is given. In column 7 and 8 are the etch-rate ratios measured by the methods discussed in the text. Errors in column 6, 7 and 8 include statistical and systematic uncertainties.

### Calibration of PET nuclear track detector

In previous calibration campaigns, PET films had been irradiated with $^{238}$U, $^{129}$Xe, $^{78}$Kr, $^{49}$Ti beams at REX-ISOLDE CERN [17]; $^{56}$Fe, $^{32}$S, $^{16}$O beams at IUAC [24]; $^{12}$C beam at IOP [25]. In Fig. 5 the reduced etch-rate $S = V - 1$, is plotted against REL for data listed in Table 3 and data obtained from previous exposures. On the horizontal axis REL values are the average over the length $L_d$ or $L_h$ for $V_T/V_B$ determined from the depth measurement and the diameter measurement, respectively. Data are fit to a second-degree polynomial equation

$$S = p_0 + p_1 (REL) + p_2 (REL)^2,$$

where $p_0 = -(1.15 \pm 0.09)$, $p_1 = 0.331 \pm 0.014$ [MeV/mg cm$^{-2}$]$^{-1}$ and $p_2 = -(19.2 \pm 2.1) \times 10^{-4}$ [MeV/mg cm$^{-2}$]$^{-2}$. The fit adjusted $R^2$ is 0.987. The detection threshold is at $REL \approx 3.5$ MeV/mg cm$^{-2}$ determined by extrapolating the curve to $S = 0$. Therefore PET is able to record particles with $Z/\beta > 125$.

6. Conclusion and discussion

There is one clear advantage of determining $V_T/V_B$ from the diameter measurement method (Eq. (2)) over the depth measurement method (Eq. (1)). In the first case, it is relatively easier to focus the microscope on the opening of the
etch-pit only, thus reducing effectively the time for large area scanning, compared to depth measurement. However, as we have shown, the diameter measurement method becomes less sensitive for PET NTD detector when $REL \gtrsim 15$ MeV/mg cm$^{-2}$. Depth measurement provides higher resolution at larger $REL$ values. In conclusion in identifying a particle using nuclear track detectors, depth measurement method for determining $V_T/V_B$ should be adopted if normalized semi minor-axis $B \approx 1$.

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Figure 5: Reduced etch-rate data versus \( REL \) for PET using etch-pit depth measurements (filled symbols) and diameter measurements (empty symbols). The curve is the result of the fit to a second order polynomial. The adjusted \( R^2 \) value is 0.987.

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