ENERGY WINDOW OF MAKROFOL FOR ALPHA PARTICLE DETECTION

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Abstract

Determination of detection energy window of Makrofol detectors irradiated with alpha particles and chemically treated with PEW (potassium hydroxide, ethanol, water) solution is presented in this paper. Detectors were exposed to ^{241}Am source through the cylindrical collimators in order to control incident alpha particle energies and angles. Alpha particles were detected in the wide energy range from 0.4 MeV to above 5 MeV. The dependence of track diameter on particle incident energy was also examined. Changes of etchant concentration and bulk etch rate (V_b) during two-hour etching were investigated. A slight increase of etchant concentration was observed while V_b was nearly constant during the whole etching process. Alpha particle passage through collimators with various dimensions and the corresponding energy distribution were also simulated theoretically using Monte Carlo Method and computer program written in Fortran90.

Keywords: Makrofol; energy window; alpha particle; collimator; chemical etching

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1. Introduction

Solid-state nuclear track detectors (SSNTDs) are solid dielectric materials (photographic emulsions, crystals, glasses, or plastics) capable of detecting different types of radiation. Passing through SSNTD, charged particles break chemical bonds and decreases the average molecular weight in the central regions of their tracks. If the impinging particles have energy and incidence angles in specific intervals, the interaction results in formation of latent tracks which can be made visible by treating detectors with appropriate etching solution. Energy deposition and ionization along the particle path increase the etching rate inside the trajectory and its vicinity. According to theoretical model, track revelation process depends on the V-function obtained as the ratio between track etch rate (V_t) and bulk etch rate (V_b). The bulk etch rate depends on the structure of the polymer material and the etching conditions, such as etchant concentration and temperature. For a given detector etched under constant conditions, the track etch rate is a function of the particle energy deposited per unit path length. The necessary condition for a visible track formation is $V > 1$ [1, 2]. Since the V function depends on energy loss, tracks could be registered only when the critical value of energy loss $(dE/dx)_c$ is exceeded. dE/dx is a function of particle energy, therefore only particles with energies falling within energy window can be registered. Energy detection window depends on the type of detector and particles under consideration, as well as on etching conditions applied [3]. Besides, detection of particles by SSNTD can also be restricted by critical detection angle, which is dependent on particle incident energy and the thickness of the detector layer removed by etching.

Makrofol detector (Bisphenol-A polycarbonate, chemical formula $C_{16}H_{14}O_3$) is commonly used for registration of tracks produced by alpha particles, fast-neutron-induced charged particles and fission fragments. This detector is resistant to high vacuum and electro-magnetic fields and insensitive to electrons, X and γ rays. Makrofol is also resistant to environmental conditions such as light, moisture, and pressure [4]. It retains elasticity and toughness over a wide range of temperatures; it can be heated up to 130^0C without changing the sensitivity [5] and it starts softening at around 150^0C. Therefore, Makrofol is quite suitable for application in radiation dosimetry and particle detection. However, there are certain limitations with regard to particle incident energy [3, 6, 7].

The aim of this study was to investigate detection energy window for Makrofol detector irradiated with alpha particles and chemically etched in PEW (potassium hydroxide, ethanol, water) solution [8].

2. Materials and methods

2.1 Experimental determination of energy window for alpha particles

Makrofol (Iupilon®, Mitsubishi Gas Chemical Company, Inc., Japan) polycarbonate detectors with a thickness of 300 µm and density of 1.2 g·cm^{-3} were used for the experiments. Detectors were irradiated with almost monoenergetic alpha particles (≈ 5.5 MeV) emitted from ^{241}Am source (Eckert & Ziegler, Germany) which was electroplated onto disk made by stainless steel. The diameter of the source was 7 mm and the activity 500 Bq. Different alpha particle incident energies from 0.2 to 5.4 MeV were obtained using Plexiglas collimators with a variable thickness (1 – 40 mm) and with a cylindrical hole (diameter of 2 mm). Alpha particles slowed down by air column struck the detector almost at the right angle. A Fortran90 computer program was developed to estimate alpha particle incident energy, based on distances traveled in air. The data obtained from SRIM 2013 [9] were combined with Monte Carlo simulations for this purpose. After seven days of irradiation, the detectors were etched for 2 h in PEW solution (15 g KOH + 45 g H_2O + 40 g ethyl alcohol). The etching was performed in a laboratory glass beaker covered with a heavy watch glass. The beaker was kept in a water bath equipped with a vibrating stirrer in order to ensure constant temperature conditions of $(70\pm1)^0$C, as well as to produce a sort of convection in the etching solution, as proposed by Rana [10,11]. Washing with distilled water after termination of etching was used in order to stop further etching of detectors. The tracks were counted manually using optical microscope.

Additionally, unexposed detectors etched under the same conditions were used for determination of background track density. Bulk etch rates were determined by gravimetric method based on measuring masses of Makrofol detectors before and after the etching, using an analytical balance with the precision of 0.1 mg.

2.2 Monte Carlo simulation of alpha particles penetration through the collimator

A simple computer program in Fortran90 was written in-house in order to simulate alpha particle passage through collimators of variable thickness (D) and hole radius (R). Alpha particles slowed down by air column strike the detector at small angles (with respect to the normal to detector surface). Monte Carlo simulation and SRIM 2013 were applied. The points of alpha particle emission were randomly chosen at the circular source surface, as well as the direction of alpha particle emission. Fig 1 shows probability that alpha particles emitted from the source hit detector surface on the opposite side of the hole, with certain incident energy. Energy range is shifted to smaller values as D increases. The width of each bar in histograms correspond to ~1 keV energy interval. Percentage of particles emitted by the source, that enter the detector and the energy range for given thicknesses (D) and hole radii (R) are also presented in Table 1. Evidently, energy ranges are less than 0.05 MeV for hole radius used in the real experiment ($R = 1$ mm), therefore the incident alpha particles can be considered almost monoenergetic. The results were obtained by simulating ^{241}Am source described in previous section.

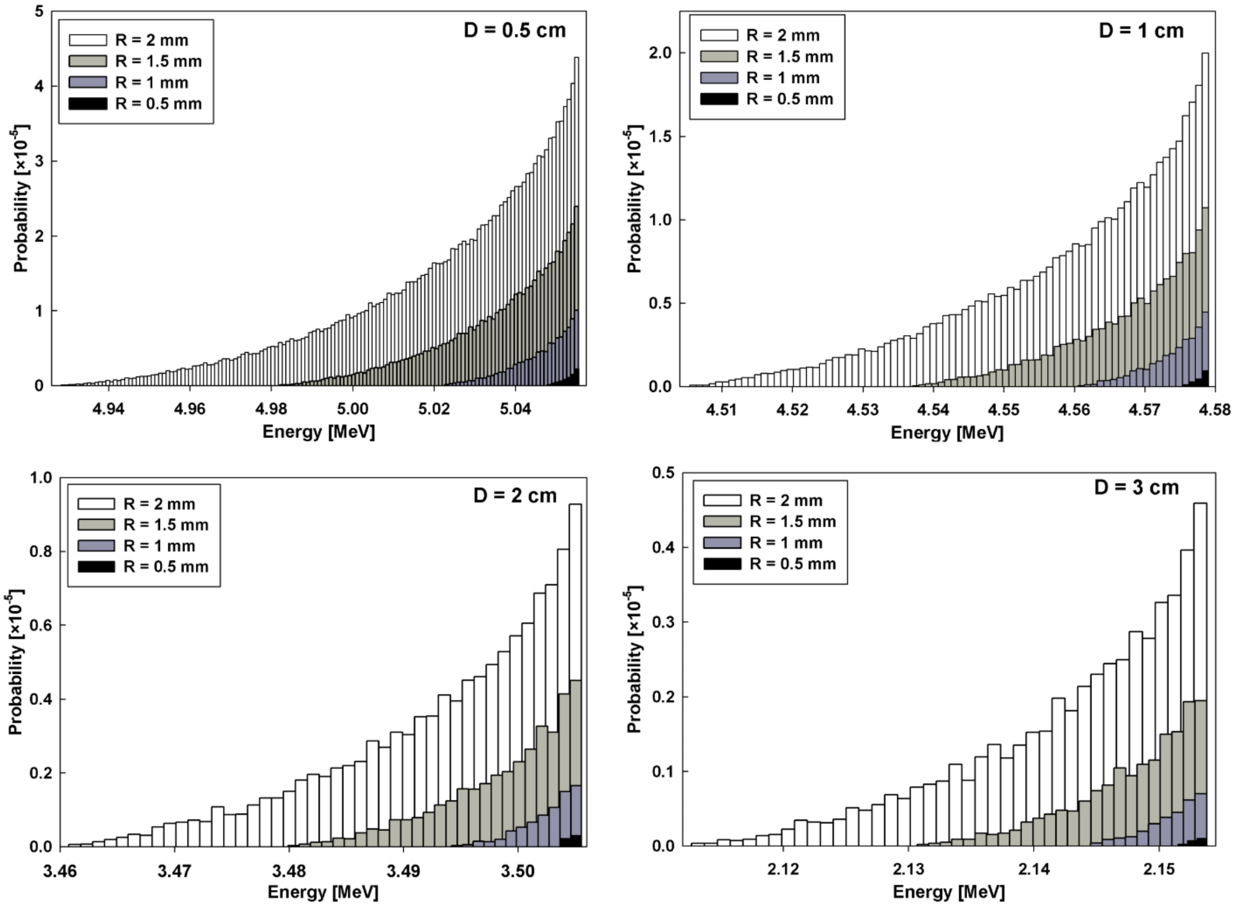

Figure 1. Probability that alpha particles emitted from the source hit detector surface with certain incident energy (please note that the energy range in abscise axis is very small)

Table 1. Results of Monte Carlo simulations: percentage of particles that enter the detector through the collimator and energy range

Collimator thickness	**R = 0.5 mm**	**R = 1 mm**	**R = 1.5 mm**	**R = 2 mm**
D = 0.5 cm	0.0050% (5.047–5.055) MeV	0.0769% (5.021–5.055) MeV	0.3656% (4.979–5.055) MeV	1.0679% (4.926–5.055) MeV
D = 1 cm	0.0013% (4.574–4.579) MeV	0.0200% (4.560–4.579) MeV	0.0999% (4.536–4.579) MeV	0.3084% (4.503–4.579) MeV
D = 2 cm	0.0003% (3.504–3.505) MeV	0.0051% (3.494–3.505) MeV	0.0255% (3.480–3.505) MeV	0.0805% (3.460–3.505) MeV
D = 3 cm	0.0001% (2.151–2.154) MeV	0.0023% (2.145–2.154) MeV	0.0115% (2.131–2.154) MeV	0.0361% (2.113–2.154) MeV

3. Results and discussion

3.1 Energy window

After determination of rough limits of detection energy window, collimator thickness was varied with a step of few tenths of mm in order to get a better precision. Totally 17 detectors were irradiated through the collimator with the hole radius R = 1 mm. After the etching, set of tracks formed a little white circle (corresponding to collimator hole) on detector surface, that could be observed even with a bare eye. Lower limit of detection energy window was estimated as 0.4 MeV corresponding to collimator thickness of 38.7 mm. The uncertainty of determining the energy limit was estimated to be less than 10%, considering the uncertainty of measuring collimator thickness and the energy spread out of alpha particle beam penetrating throughout the collimator hole. Further increase of collimator thickness i.e. reducing the incident alpha particles energy, did not produce visible tracks, although the software still predicted relatively high number of particles hitting the detector. On the other hand, the upper limit of the energy window could not be estimated in this experiment. Although some authors reported this limit to be ≈ 3 MeV [3, 7], a large number of visible tracks were clearly seen even for the incident energies of 5.4 MeV (collimator thickness of 1 mm). Exposing the detector directly to the source (without any collimator) gave the same result, indicating that the upper limit of detection energy window for Makrofol is probably higher than previously reported. However, a study of Cesar and Franco [6] also presented detection of alpha particles with incident energies above 5 MeV for the same etching conditions. Rammah et al. [4, 12] have reported detection of alpha particles with energies in a wide range from 1 to 5 MeV, although the etchant used in their experiment was rather different (10 N NaOH and methanol). On the other hand, Soares et al. reported the Makrofol energy widow of 0.7 MeV– 3.7 MeV for the same PEW solution, but for 30 min etching time and temperature of 65^0C [7]. The discrepancies in the results of different authors might be explained by different etching conditions applied in the experiments. A relatively high bulk etch rate and 2-hour etching time applied in our study resulted in a relatively large removed layer that has expanded the upper limit of the energy window.

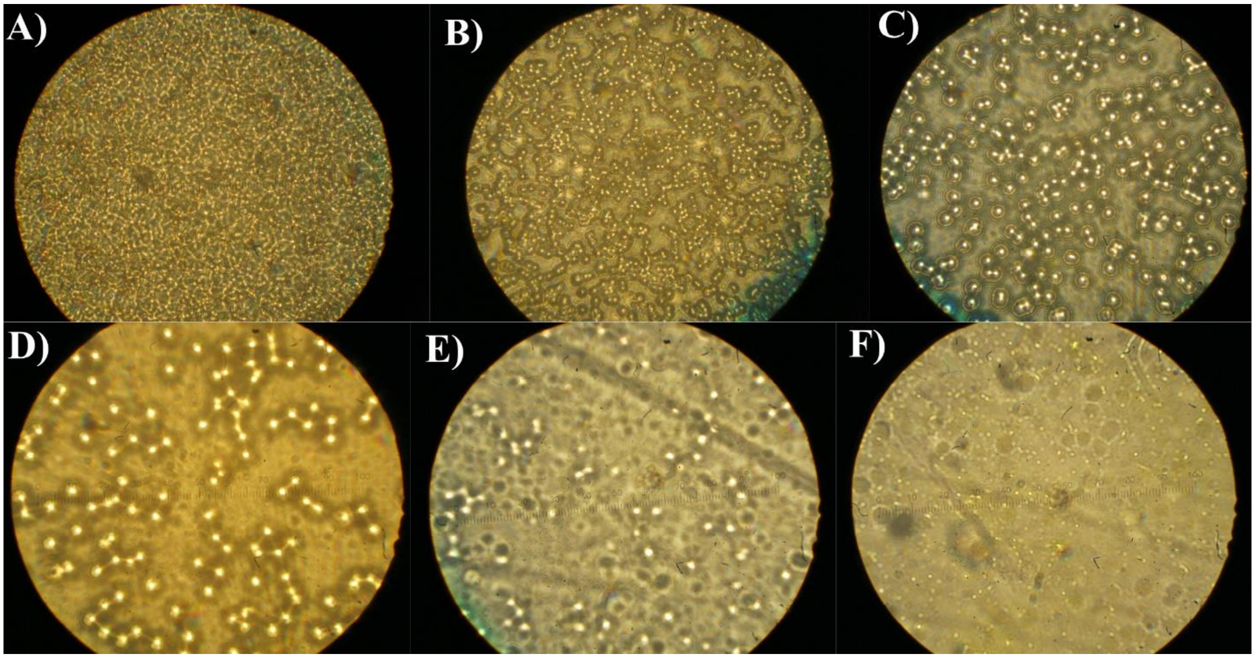

Figure 2. Microscope view images of alpha particle tracks in Makrofol for different energies A) 5.4 MeV; B) 4.6 MeV; C) 3.6 MeV; D) 2.3 MeV; E) 0.6 MeV; F) 0.4 MeV

Figure 2 presents microscope view images of tracks in Makrofol detectors for several collimator thicknesses i.e. several alpha particle incident energies. As it was expected, increasing the collimator thickness decreases the track density. On the other hand, decreasing alpha particle energy leads to enlargement of track diameter to some point (at $E_\alpha \approx 1$ MeV). Further decrease of incident energy is followed by decrease in track diameter (Fig. 3). Variation of alpha particle diameter with energy following Bragg curve has also been observed by some other authors [4, 12, 13]. Similar observations have also been reported for LR115 and CR-39 detectors [14-17]. However, some authors investigating the penetration of swift heavy ions through solids have observed that the position of the maximum damage in a solid does not exactly coincide with the Bragg peak of the electronic energy loss. The radius of the track was found to depend not only on the ion energy loss, but also on its velocity i.e. its penetration depth [18-20].

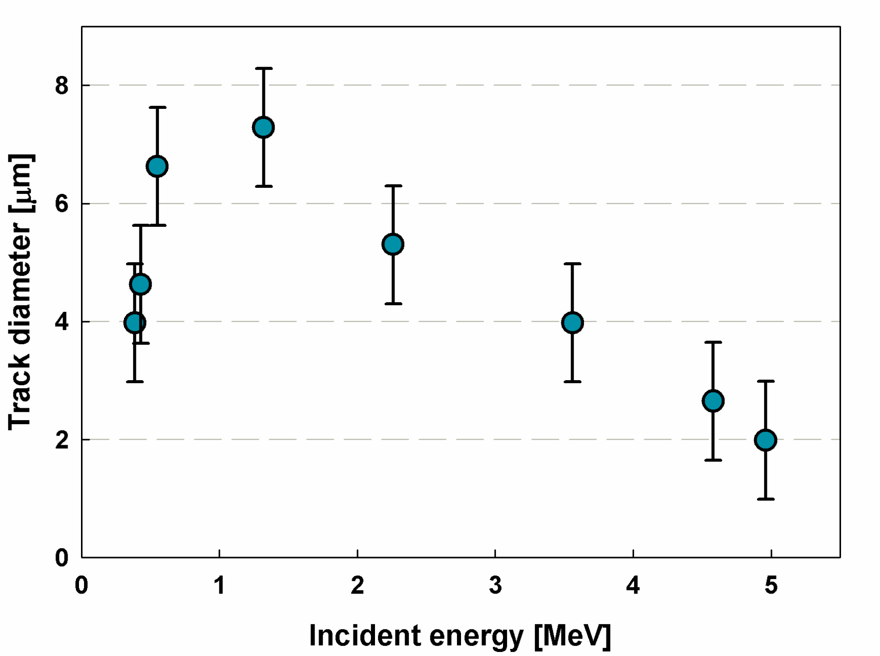

Figure 3. Variation of alpha particle track diameter in Makrofol with energy

3.2 The etchant concentration and bulk etch rate

Since ethanol is rather volatile even at the room temperature (the boiling point is 78.24^0C), it is reasonable to expect that certain amount of alcohol evaporates during the etching process. This might cause changes in the concentration of the etching solution, affecting the etching rates. In order to investigate this effect, an additional experiment was performed by etching a new set of non-irradiated detectors under the same etching conditions (2 h in PEW solution at 70^0C). The etchant was poured in a laboratory glass beaker covered with a watch glass (but not hermetically sealed) and heated in the water bath. After achieving a constant predetermined temperature of the solution, a certain amount of solution was sampled, and pH-measurements were performed by Metrohm 827 pH-meter with specified accuracy of ±0.003. Afterwards, four unexposed Makrofol detectors were successively immersed in the solution and etched for 30 minutes each, in order to detect possible changes in bulk etch rate. In addition, pH value of the etchant was measured in the middle of each 30-minute etching interval. Results are presented in Figure 4. A slight increase of pH value was observed indicating a small rise in the concentration of the etchant. However, changes of bulk etch rate were irregular and they were all within the range of measuring uncertainty (estimated to be about 10%). Therefore, small increase of etchant concentration can be neglected and V_b can be treated as nearly constant during the whole etching process ($V_b = (11.6 \pm 0.4)$ μm h^{-1} for the current conditions).

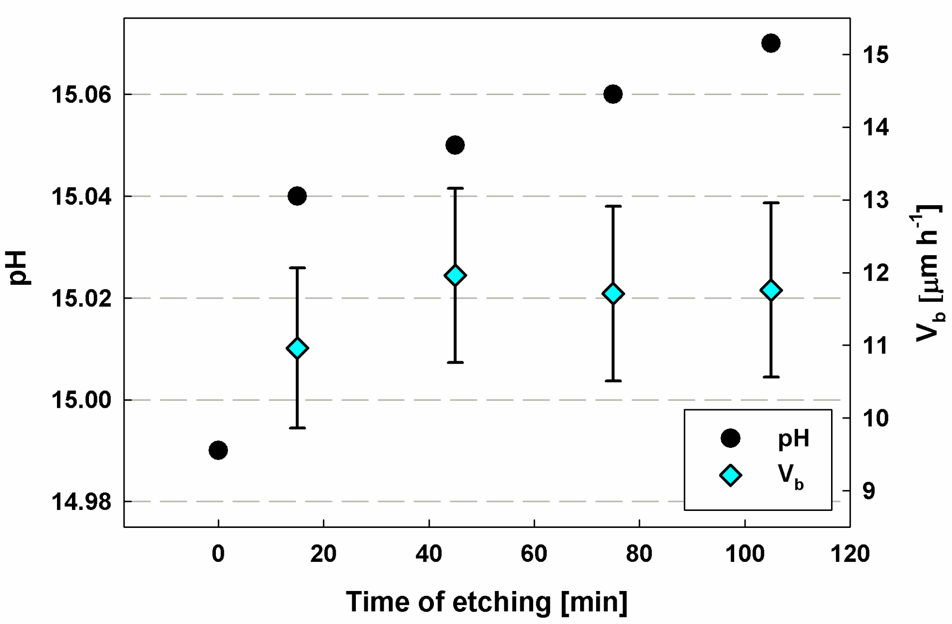

Figure 4. Changes in bulk etch rate and pH of the etching solution during the etching process

For the purpose of comparing bulk etch rates, several pieces of non-irradiated CR-39 detector were also etched for 2 h at 70^{0}C, using the same PEW solution. The bulk etch rate obtained by gravimetric method for CR-39 was (34.5 ± 0.5) μm h^{-1} (it was three times higher than that for Makrofol).

4. Conclusion

Makrofol detectors were irradiated with alpha particles from ^{241}Am source and chemically treated by PEW solution. Particles were detected in the wide range of energies from 0.4 MeV to almost 5.5 MeV. The upper limit of the energy window could not be estimated in this study. The dependence of track diameter on particle incident energy follows Bragg curve.

Bulk etch rate was determined by gravimetric method (V_b = (11.6±0.4) μm h^{-1}) and it can be treated as almost constant since the increase of etchant concentration during the two-hour etching process can be neglected. Probability that alpha particles emitted from the source hit detector surface as well as the corresponding energy distribution were also investigated theoretically using computer program written in Fortran90. It has been found that collimator acts in such a way that the energy of alpha particles can be defined with a very small uncertainty. Reducing the collimator diameter results in narrowing of the energy interval, but also in decreasing the number of penetrating particles.

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Declarations of interest: none

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