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Bulk and track etch properties of CR-39 SSNTD etched in NaOH/ethanol

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Abstract

Recently, Matiullah et al. described the use of NaOH/ethanol as an etchant for the CR-39 detector, and have determined the corresponding bulk and track etch properties from the track diameter method. In the present work, the bulk and track etch properties of CR-39 in NaOH/ethanol were derived from direct measurements. The bulk etch rate has been found to increase with the molarity of NaOH/ethanol, reach a maximum at ~2.5 N and start to drop beyond 3 N. The bulk etch rate also increases with stirring. These phenomena can be explained by the insulation of the detector from the etchant by the etched products. Regarding the track etch, we have found a surprising result that the lengths of (pre-etched) tracks are actually shortened when the tracks are etched in NaOH/ethanol. Generally speaking, the remaining track depths obtained with stirring are longer than those for no stirring. The shortening of the tracks can be explained by the insulation of the pre-etched track wall from the etchant with the etched products.

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Keywords: Solid-state nuclear track detector; SSNTD; CR-39; Etchant; Ethanol

1. Introduction

With the discovery of a relative fast etchant of NaOH/ethanol solution for the CR-39 detector by Matiullah et al. [1], more etchants can be chosen for CR-39 in various applications. Matiullah et al. [1] have determined the corresponding bulk and track etch properties from the track diameter method. The bulk etch and track etch rates of Matiullah et al. varies from 15.83 to 60 m/h and 33.63 to 261.488 m/h at 55 °C, respectively, which depend on the molarities of the NaOH/ethanol solutions [1].

In the present investigation, the bulk and track etch properties of CR-39 in NaOH/ethanol were measured from direct measurements instead of using the track diameter method. The track diameter method employed in [1] was an indirect method, which might not be able to take into account the effects from precipitation of etch products onto the alpha-particle track walls. The latter has likely occurred in etching of CR-39 using NaOH/ethanol as found in the present work. The advantage of the direct methods used in the present paper is that the results will not be affected by such precipitation. Therefore, although measurements of etch properties for NaOH/ethanol through the indirect track diameter method were already reported in [1], further measurements through direct methods are still required.

The first objective is to measure the bulk etch rates of the CR-39 for different molarities of NaOH/ethanol through the masking method [2] and surface profilometry [3,4]. The effect of stirring on the bulk etch rate is also studied [5]. The second objective is to measure the depths of pre-etched tracks which are further etched in NaOH/ethanol. This part of work is performed because we have found a surprising but very interesting result that the lengths of the pre-etched tracks are actually shortened when they are etched in NaOH/ethanol. Alpha-particle tracks are first

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formed using normal etching (in NaOH/water). Both sharp-phase conical tracks and over-etched spherical tracks are studied with their lengths measured by the ''cross-section'' method [6,7] and the ''replica'' method [8], respectively, both of which are direct measurement methods. The effect of stirring on the track shortening is also studied.

2. Methodology

2.1. Bulk etch rates of the CR-39 in NaOH/ethanol

As mentioned above, the bulk etch rates of the CR-39 detector were measured by the masking method [2]. The bulk etch was determined by the height difference between the etched portion and the masked portion that masked by epoxy (Araldite® Rapid, England) which is resistant to etching through surface profilometry measurements (using Form Talysurf PGI (FTPGI) from Taylor Hobson, Leicester, England). The effect of stirring on the bulk etch rate of the CR-39 detector is also a concern [5] so the bulk etch rates were determined under the conditions with and without stirring during etching. Stirring was provided by a magnetic stirrer (Model no: SP72220-26, Barnstead/Thermolyne, IA, USA).

2.2. Track depths of pre-etched tracks etched in NaOH/ethanol

2.2.1. Irradiation of CR-39 detectors

The CR-39 detectors used in the present investigation were purchased from Page Mouldings (Pershore) Limited (Worcestershire, England). The original dimensions of the sheet of detector are $30 \times 47 \times 0.1$ cm (thickness). The detectors were cut into the size of 1×1 cm^2. Prior to etching, the detectors were irradiated with energies of 1, 2, 3, 4 and 5 MeV under normal incidence through a collimator. The alpha source employed in the present study was a planar ^{241}Am source (main alpha energy = 5.49 MeV under vacuum). The final alpha energies incident on the detector were controlled by the source to detector distances in normal air (from ~0.5 to ~3.5 cm). The relationship between the alpha energy and the air distance traveled by an alpha particle with initial energy of 5.49 MeV from ^{241}Am was obtained by measuring the energies for alpha particles passing different distances through normal air using alpha spectroscopy systems (ORTEC Model 5030) with passivated implanted planar silicon (PIPS) detectors of areas of 300 mm^2.

2.2.2. Pre-etching in NaOH/water and further etching in NaOH/ethanol

The objective of this section is to first develop the alpha-particle tracks using normal etching (in NaOH/water) and then to investigate the change in the lengths of these pre-etched tracks for further etching in NaOH/ethanol.

After irradiation, one set of the detectors were pre-etched in 6.25 N NaOH/water maintained at 70 °C in a water bath for 1.5, 2 and 4 h (short pre-etching periods) for incident alpha energies of 1, 2 and 3–5 MeV, respectively, to reveal alpha tracks. All these tracks will be in the conical phase. Another set of the detectors were pre-etched in 6.25 N NaOH/water at 70 °C for 15 h (long pre-etching period) for all incident alpha energies as above. These tracks will be in the spherical over-etched phase.

After the pre-etching, we investigate the change in the lengths of the pre-etched tracks for different further etching periods in NaOH/ethanol. The etchants were prepared by dissolving NaOH directly in absolute ethanol (95%). The etching temperature was chosen to be 55 °C in the present investigation in order to have direct comparisons with the results given by Matiullah et al. [1] and to prevent too much evaporation of the ethanol. The tracks pre-etched for 1.5, 2, and 4 h were etched in different molarities of NaOH/ethanol solutions at 55 °C with an accuracy of ±1 °C. One set of the detectors was etched under no stirring while the other set of detectors were etched under stirring with a magnetic stirrer for the same time interval of 30 min. For the tracks pre-etched for 15 h, the detectors were also etched in different molarities of NaOH/ethanol solutions at 55 °C with an accuracy of ±1 °C for different time intervals of 30 min, 1 h and 2 h.

2.2.3. Measurement of track depths

Two different methods will be employed to measure the depths of conical and spherical tracks.

Yu et al. [8] proposed a method to derive the lengths of tracks in CR-39 detectors through measurements of their replicas (hereafter referred to as the replica method). The track length can be measured precisely through the heights of the replicas because measurements of heights of protruding objects will not be constrained by the stylus geometry of the surface profilometer. The replica method will be employed here to measure the track lengths for spherical tracks. Only the lengths of the tracks in the over-etched phase, but not those in the conical phase, can be measured using this method [7]. Whenever possible, the replica method is preferred as surface profilometry can provide a better spatial resolution(~40 nm) than that provided by general optical microscopes (~0.3 μm). The replicating fluid was prepared with Buehler Epo–Kwick resin (No. 20-8136-128) and Buehler Epo–Kwick hardener (No. 20-8138-032) with the mass ratio of 5:1. It was found that the replicating fluid could fill the tracks completely and the lengths of the tracks can be reflected by the heights of the replicas [7]. The tracks protruding from the resin surface were then measured by the FTPGI surface profilometer. The track depths were determined from the height difference between the top and the base of tracks in the lateral view of the image of replicas generated by FTPGI.

On the other hand, the tracks pre-etched for 1.5, 2 and 4 h for incident energies of 1, 2 and 3–5 MeV, respectively, in 6.25 N NaOH/water are conical. The cross-sections of these tracks will be revealed under the optical microscope operated in the transmission mode after polishing the edge

of the detectors. The depths of these tracks will then be measured directly from the cross-sections of the tracks [6,7] (hereafter referred to as the cross-section method).

Fig. 1 shows an example of the lateral view of a conical track from alpha particle with incident energy of 3 MeV and pre-etched in 6.25 N NaOH/water at 70 °C for 4 h. If this track is further etched in 1 N NaOH/ethanol solution for 15 min, it will become spherical, the lateral view of which is shown in Fig. 2. The same phenomenon also occurs for tracks from other incident alpha energies (1, 2, 4 and 5 MeV), i.e. the conical tracks from short pre-etching in 6.25 N NaOH/water at 70 °C will become spherical on further etching in 1 N NaOH/ethanol solution for 15 min. On the other hand, for long pre-etching in 6.25 N NaOH/water at 70 °C for 15 h (long pre-etching period), the tracks for all incident alpha energies will be already in the spherical phase. Therefore, regarding the track depth measurement techniques, only the depths of tracks from the short pre-etching periods are determined by the cross-section method, while the depths of all other tracks are measured by the replica method.

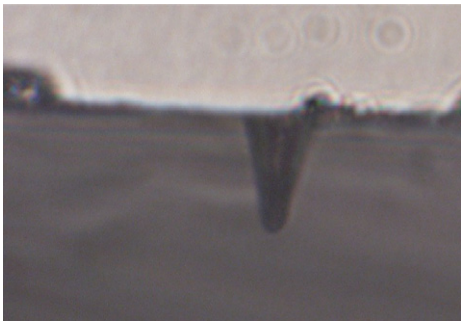

Fig. 1. The lateral view of a conical track (from an alpha particle with incident energy of 3 MeV and pre-etched in 6.25 N NaOH/water at 70 °C for 4 h) revealed under the optical microscope operated in the transmission mode after polishing the edge of the detectors.

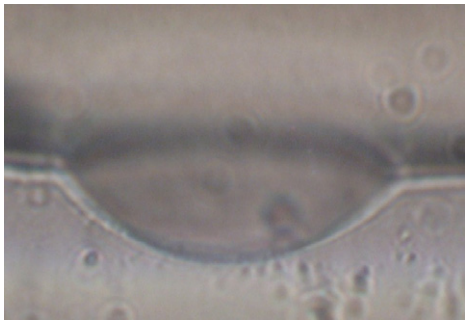

Fig. 2. The lateral view of a spherical track (from an alpha particle with incident energy of 3 MeV and pre-etched in 6.25 N NaOH/water at 70 °C for 4 h and then further etched in 1 N NaOH/ethanol for 15 min) revealed under the optical microscope operated in the transmission mode after polishing the edge of the detectors.

3. Results and discussion

3.1. Bulk etch rates of the CR-39 in NaOH/ethanol

After measurements of the amount of bulk etch using FTPGI, the bulk etch rates of the CR-39 detectors etched in different molarities of NaOH/ethanol solutions were determined, which are summarized in Table 1. From Table 1, we see that the bulk etch rate increases with the molarity of NaOH/ethanol, reaches a maximum at ~2.5 N and starts to drop beyond 3 N.

The differences between the bulk etch rates obtained in the present work and those reported in [1] are likely due to the direct and indirect measurement methods employed in the two works. As mentioned in the introduction, the direct measurement methods employed in the present work are not affected by the precipitation of etch products onto the alpha-particle track walls.

The fierce bulk etch rates for NaOH/ethanol etchants compared to that for the 6.25 N NaOH/H_2O etchant are due to the miscibility of ethanol with the organic etched products from etching the CR-39 (allyl diglycol carbonate) polymer [9]. However, sodium carbonate is also formed during etching of CR-39 [9]. In particular, the production rate of sodium carbonate from etching CR-39 in NaOH/ethanol is larger than that in NaOH/H_2O, and that a layer of precipitate consisting of sodium carbonate is accumulated on the surface of CR-39 in the former case due to the insolubility of sodium carbonate and its saturation in ethanol [9]. Therefore, if the molarity of NaOH/ethanol keeps increasing, the production rate of sodium carbonate will increase to a point such that the sodium carbonate can effectively insulate the detector from the etchant, and the bulk etch rate will decrease. Such insulation might be partially weakened through stirring during etching. With the help of stirring, the etched products can be removed more efficiently from the detector so that the bulk etch rate under stirring will be higher.

Table 2 shows the effect of stirring on the bulk etch rates of the CR-39 detectors in the NaOH/ethanol etchants. Three molarities have been studied and the bulk etch rate is indeed found to increase with stirring.

Table 1
The bulk etch rates of the CR-39 detector obtained by the masking method for different molarities of NaOH/ethanol

Molarities of NaOH/ethanol (N)	With no stirring ($\mu m\ h^{-1}$)	Reference value [1] ($\mu m\ h^{-1}$)
0.5	22.364 ± 0.738	15.83
1.0	47.522 ± 0.248	28.42
1.5	61.328 ± 3.723	60.00
2.0	65.606 ± 1.736	38.85
2.15	75.615 ± 4.384	31.25
2.52	75.290 ± 2.637	36.38
3.0	61.584 ± 0.304	N/A

Table 2
The bulk etch rates of the CR-39 detector obtained by the masking method for different molarities of NaOH/ethanol, for the cases with and without stirring during etching

Molarities of NaOH/ethanol	With no stirring ($\mu m\ h^{-1}$)	With stirring ($\mu m\ h^{-1}$)	Reference value [1] ($\mu m\ h^{-1}$)
0.5	22.364 ± 0.738	29.820 ± 0.294	15.83
1.0	47.522 ± 0.248	62.248 ± 0.742	28.42
2.0	65.606 ± 1.736	69.192 ± 1.633	38.85

3.2. Track depths of pre-etched tracks etched in NaOH/ethanol

3.2.1. Short pre-etching periods

As described in Section 2, the depths of the tracks in the conical phase were measured using the cross-section method [6,7]. On the other hand, the depths of the tracks in spherical phase resulted from further etching in NaOH/ethanol were measured using the replica method. The track depths obtained with and without stirring during etching for 0.5, 1 and 2 N NaOH/ethanol etchants are shown in Tables 3–5, respectively. Generally speaking, the track depths are reduced after etching in NaOH/ethanol. Moreover, the track depths with stirring are longer than those for no stirring.

Table 3
Depths of pre-etched tracks in 6.25 N NaOH/water at 70 °C (pre-etch periods are 1.5, 2 and 4 h for incident alpha energies of 1, 2 and 3–5 MeV, respectively) for various incident alpha energies, and the depths of these tracks on further etching in 0.5 N NaOH/ethanol for 30 min under the conditions with and without stirring during etching

Incident alpha energies (MeV)	Depth of pre-etched track (μm)	Track depth with no stirring (μm)	Track depth with stirring (μm)
1	2.340 ± 0.051	0.373 ± 0.063	0.369 ± 0.007
2	7.667 ± 0.290	1.503 ± 0.072	2.074 ± 0.410
3	10.629 ± 0.260	1.518 ± 0.040	2.904 ± 0.112
4	5.763 ± 0.051	1.460 ± 0.043	1.698 ± 0.031
5	3.970 ± 0.051	0.867 ± 0.039	0.982 ± 0.340

The depths of pre-etched tracks are measured by the cross-section method, while those for further etching in NaOH/ethanol are measured by the replica method.

Table 4
Depths of pre-etched tracks in 6.25 N NaOH/water at 70 °C (pre-etch periods are 1.5, 2 and 4 h for incident alpha energies of 1, 2 and 3–5 MeV, respectively) for various incident alpha energies, and the depths of these tracks on further etching in 1 N NaOH/ethanol for 30 min under the conditions with and without stirring during etching

Incident alpha energies (MeV)	Depth of pre-etched track (μm)	Track depth with no stirring (μm)	Track depth with stirring (μm)
1	2.340 ± 0.051	0.470 ± 0.037	1.729 ± 0.029
2	7.667 ± 0.290	0.430 ± 0.008	1.460 ± 0.028
3	10.629 ± 0.260	2.867 ± 0.074	3.740 ± 0.060
4	5.763 ± 0.051	0.710 ± 0.112	1.097 ± 0.018
5	3.970 ± 0.051	1.224 ± 0.082	2.526 ± 0.069

The depths of pre-etched tracks are measured by the cross-section method, while those for further etching in NaOH/ethanol are measured by the replica method.

Table 5
Depths of pre-etched tracks in 6.25 N NaOH/water at 70 °C (pre-etch periods are 1.5, 2 and 4 h for incident alpha energies of 1, 2 and 3–5 MeV, respectively) for various incident alpha energies, and the depths of these tracks on further etching in 2 N NaOH/ethanol for 30 min under the conditions with and without stirring during etching

Incident alpha energies (MeV)	Depth of pre-etched track (μm)	Track depth with no stirring (μm)	Track depth with stirring (μm)
1	2.340 ± 0.051	1.169 ± 0.021	1.170 ± 0.023
2	7.667 ± 0.290	1.791 ± 0.052	1.974 ± 0.043
3	10.629 ± 0.260	2.580 ± 0.106	4.665 ± 0.075
4	5.763 ± 0.051	2.642 ± 0.075	3.083 ± 0.063
5	3.970 ± 0.051	1.897 ± 1.278	1.505 ± 0.670

The depths of pre-etched tracks are measured by the cross-section method, while those for further etching in NaOH/ethanol are measured by the replica method.

3.2.2. Long pre-etching period

As described in Section 2, the depths of the tracks in the spherical phase were measured using the replica method [8]. Examples of the lateral images of the replicas generated by FTPGI are shown in Figs. 3 and 4. Fig. 3 shows 2 MeV alpha-particle tracks pre-etched in NaOH/water at 70 °C for 15 h, and the average track depth is obtained as 7.617 ± 0.421 μm. Fig. 4 shows the tracks resulted from etching the above tracks (2 MeV alpha-particle tracks pre-etched in NaOH/water at 70 °C for 15 h) in 1 N NaOH/ethanol at 55 °C for 30 min, and the average track depth is obtained as 4.251 ± 0.159 μm. The depths of the tracks for the long pre-etching period (pre-etched in NaOH/water at 70 °C for 15 h) etched for different time at 55 °C in 0.5, 1 and 2 N NaOH/ethanol are shown in Figs. 5–7, respectively. It can be seen that the track depth also decreases with increasing etching time in general.

3.2.3. Shortening of tracks when etched in NaOH/ethanol

From the above results, we can conclude that the pre-etched tracks will be shortened on further etching in NaOH/ethanol. In fact, from our experiments, we have found that the pre-etched tracks will finally disappear under the optical microscope.

The shortening of the tracks in NaOH/ethanol can be explained by the insulation of the pre-etched track wall from the etchant with the etched products. With the fierce bulk etch rate of NaOH/ethanol, the pre-etched tracks will disappear fairly quickly. With the introduction of stirring during the etching, the track shortening may be lessened. However, this is not always the case, since the final result will be the combined effects from the production rate of etched products (which depends largely on the molarity of the etchant) and the dispersion rate of etched products from the track (which depends on the production rate of

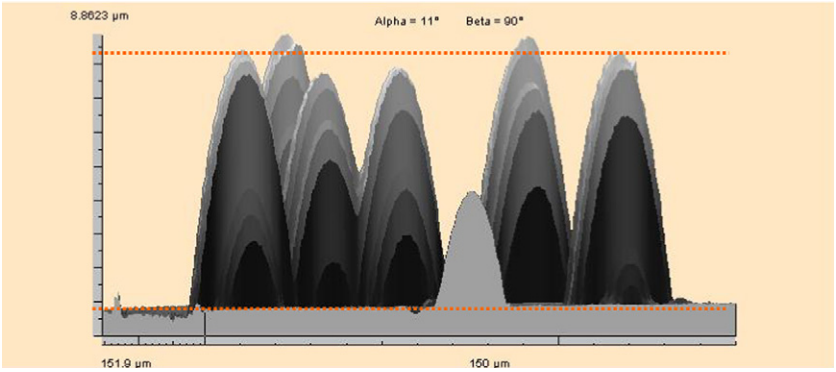

Fig. 3. The lateral view generated by the FTPGI Profilometer for replicas of alpha-particle tracks. The tracks are formed from 2 MeV alpha particles and by pre-etching in 6.25 N NaOH/water at 70 °C for 15 h. The average track depth is obtained by the difference between the two dotted lines as 7.617 ± 0.421 μm.

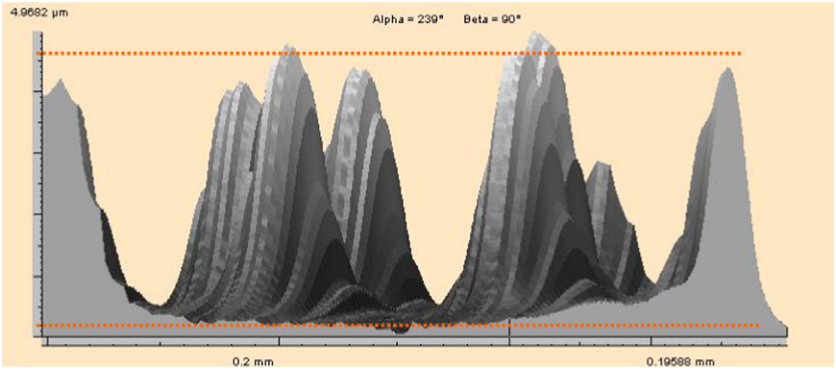

Fig. 4. The lateral view generated by the FTPGI Profilometer for replicas of alpha-particle tracks. The tracks are formed by etching the tracks in Fig. 3 (formed from 2 MeV alpha particles and by pre-etching in 6.25 N NaOH/water at 70 °C for 15 h) in 1 N NaOH/ethanol at 55 °C for 30 min. The average track depth is obtained by the difference between the two dotted lines as 4.251 ± 0.159 μm.

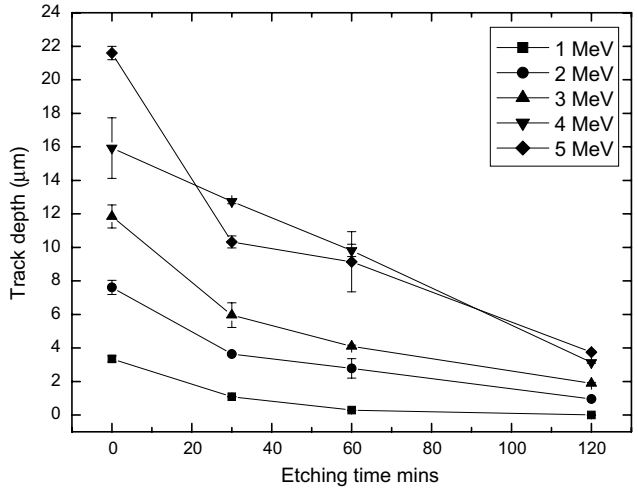

Fig. 5. Depths of the alpha-particle tracks (with incident energies from 1 to 5 MeV) from the long pre-etching period (pre-etched in NaOH/water at 70 °C for 15 h) etched at 55 °C in 0.5 N NaOH/ethanol for different time intervals.

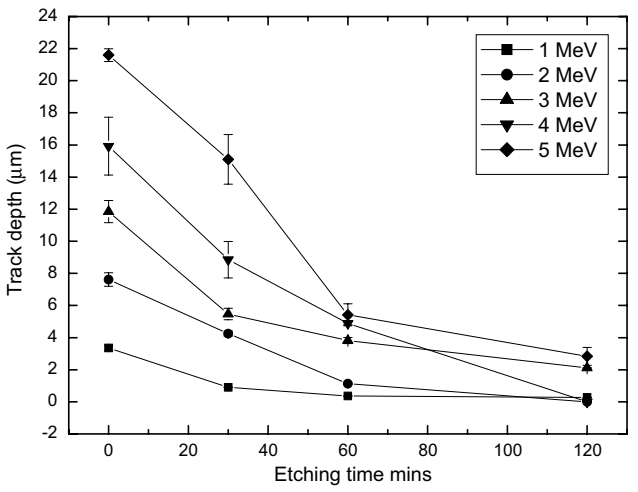

Fig. 6. Depths of the alpha-particle tracks (with incident energies from 1 to 5 MeV) from the long pre-etching period (pre-etched in NaOH/water at 70 °C for 15 h) etched at 55 °C in 1 N NaOH/ethanol for different time intervals.

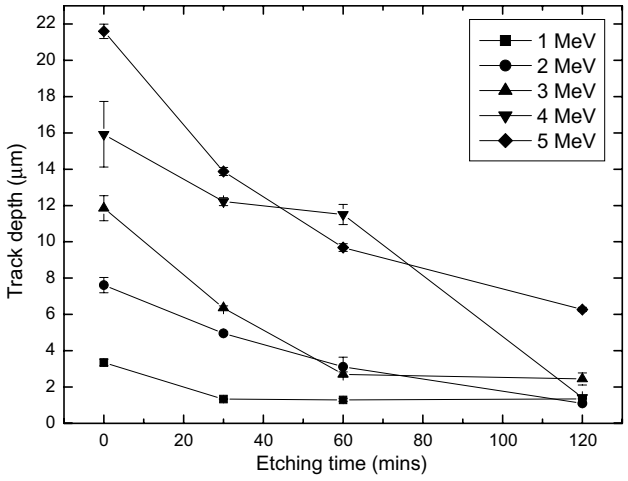

Fig. 7. Depths of the alpha-particle tracks (with incident energies from 1 to 5 MeV) from the long pre-etching period (pre-etched in NaOH/water at 70 °C for 15 h) etched at 55 °C in 2 N NaOH/ethanol for different time intervals.

etched products, viscosity of the etchant and etched products, size of the track opening and the track depth, etc).

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