The optimum time of etching proved the Sensitivity of the CR-39 detector

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Abstract:
The ideal detector used in this study is the CR-39 polymer (Italy). A series of chemical etching experiments have been carried out on CR-39 detectors, irradiated with alpha particle. A alpha particle crossing a CR-39 sheet, leaves a trail of broken polymer links along its path (restricted to a microscopic cylindrical region). Experimental data has been analyzed to find out important track etch parameters. Both bulk etch rate and diameter of tracks are found to vary with time of etching and concentration of normality for NaOH, the average values for them are 1.190 µm/h and 1.172 µm/h, and 12.36 µm and 14.81µm, respectively. The critical angle of etching and track registration efficiency have also been determined using experimental data. All parameters depend on properties of incident ion and etching conditions. Results describing the dependence of track etch parameters on etching conditions have been presented. These results are useful in the interpretation of track data.
Keywords: Alpha tracks, CR-39NTDs, Ion tracks, Bulk etch rate, Critical angle, Etching efficiency.

INTRODUCTION

Track detectors were discovered by Young, Silk and Barner and they have been widely used in science and technology [1]. The detection technique based on using track detectors is presently called solid state nuclear track detection (SSNTD)[2]. One of the most commonly detector used is CR-39NTD, because of its good sensitivity, stability against various environmental factors, and high degree of optical clarity [3] . The bulk etch-rate (V_B) is one of the basic parameters required for the use of the SSNTDs. The value of V_B is required to be known many parameters such as track etch-rate, track efficiency and track diameters . Therefore, it is important to critically assess the methods of measuring V_B [4].

Fast heavy ions can produce latent tracks in many dielectric materials, after adequate treatment, such as chemical etching [5] .The operation of the solid state nuclear track detector (SSNTD) is based on the fact that a heavy charged particle will cause extensive ionization of the material when it passes through a medium. A recent review on SSNTDs has been given by Nikezic and Yu. If a piece of material containing the latent track is exposed to some chemically aggressive solution (such as NaOH or KOH solution), the chemical reaction would be more intensive in the latent track. Such a solution is called the etchant [1]. Through the etching, the latent track becomes visible as a particle ‘‘track” which may be seen under an optical microscope. Ion-track growth in SSNTDs has been

Surface after time t

suggested to base on two parameters, V_T and V_B as shown in Fig (1).

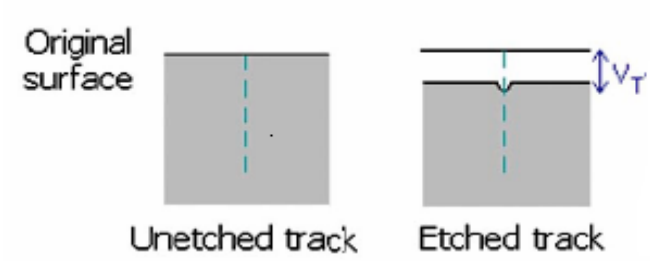

Fig.(1): Progressive effects of etching track detector[7]

V_T is the track etch rate (the rate of etching along the particle trajectory) and V_B is the bulk etch rate (the rate of etching of the undamaged detector surface) [8]. In other words, the bulk etch rate V_B is the rate of removing of the undamaged surface of the detector. Track development is governed by the ratio $V = V_T/V_B$ and track formation is not possible if V is smaller than or equal to 1 [1]. Research has been devoted to understanding the mechanisms of track growth in SSNTDs [9].

V_B is influenced by many factors, such as the intrinsic characteristics of the plastic material, preparation method and the etching conditions in terms of temperature, concentration and type of etchant, etching duration, and stirring. V_B has been measured by several experimental methods. Some have been based on the measurement of a parameter indirectly rated. In particular, direct methods for V_B determination based on 'thickness-measurement' before and after etching by measuring the thickness decreased of the detector during etching and based on 'mass-measurement' is recording the mass of the detector before and after etching [4]. The indirect method is 'diameter measurement' by measurement of the track diameter resulting after etching for specific time intervals [1].

a. Bulk etch rate (V_B)

Bulk etch rate is one of the crucial factors controlling the track development in SSNTDs[10]. Due t Original surface reaction between the etching solution (etchant) and the detector material, some molecules of the detectors are removed. The final effect is the removal of the material from the detector surface and the thickness of the detector becomes Latent ion track smaller and smaller [1]. There are many equation to determining the bulk etch rate (V_B).

$$V_B(h) = \Delta h/2\Delta t \text{ -----(1)}$$

where Δh is the difference layer removed during the etching time Δt.

$$V_B = \Delta m/2\Delta t\ A\ \rho \text{ ----(2)}$$

where Δm is mass difference, A the etched surface area, ρ the density of the detector and Δt is the etching time. However, this method is limited by the accuracy of mass measurements.

$$V_B\,(m, h) = (\Delta m)h_2/2\Delta t\ m_2 \text{ ---(3)}$$

$$V_B = D/2\Delta t \text{ ----(4)}$$

b. Track etch rate (V_T)

The track etch rate (V_T), is the rate of etching along the particle trajectory[11]. A track is formed when $V_T > V_B$. and calculated by the relation

$$V_T = V_B\,(4h^2+D^2)/\,(4h^2- D^2) \text{ ----(5)}$$

V_T depends on the etching temperature [12].conical holes (etch tracks), are produced on the surface at the position of particle impacts (two different etching rates (V_B and V_T)), as shown in Fig.(1)[13]

And all parameters determined by the following;

$V = V_T/V_B$—(6)

$D = 2h\sqrt{\frac{V-1}{V+1}}$ ---(7) $V \gg 1$, the previous equation becomes

$D=2h =2V_Bt$------(8)

Where D is track diameter, h_2 is the thickness of the detector, m_2 is mass of the detector after etching[1].

$L = V_T\, t - V_Bt$ ---(9)[14].

$V= h/t$----(10)

From Eqs. (9) and (10), V can be found as:

$V=1+L/h$------(11)

c. The critical angle (θ_c)

One of the geometrical limitations for the revelation of the etched tracks in SSNTDs is called critical angle (θ_c) [15], determined by

$\sin \theta_c = V_B/V_T$ or

$\sin \theta_c = 1/ (V_T/V_B)$,

$\theta_c = \sin^{-1}(V_B/V_T)$ ---- (12)

d. Detector Efficiency (f)

Registration efficiency is very important parameter for any dosimeter and should be obtained before its use. The efficiency of each of the etchants was determined by using the values of V_T and V_B in a standard formula [16]. The partial sensitivities depend on the V function ($V = V_T/V_B$) as well as the thickness of the removed active layer during etching and on a critical angle, can be obtained by the following equations

$f = 1 - (V_B/V_T)$

$f = 1- V_B/V_T = 1- 1/V$

$f = 1- \sin \theta_c$ -----(13)[9]

2-Materials and Methodology

a. CR-39NTD.

CR-39NTD is a $C_{12}H_{18}O_7$ polymer with a density of 1.31 g/cm^3 [17]. The range of 5 MeV alpha particles in it is 28.77 µm. Gaillard et al [18] [19], had a thickness of 700 µm cut into pieces with an area of (1.0 × 1.5) cm^2[6] [20], before use. The detectors used in the present study is the ideal detector, produced by Intercase Europe SRL (43100 Parma, Italy). The Intercast CR-39 has a low background for small etching process for its use in radon dosimetry [21]. The efficiency of CR-39NTDs changes with time of storage therefore, the calibration process is required before using it. CR-39NTD was calibrated and its efficiency was 79.5% . The main characteristics of the detectors are listed in Table (1). The sensibility of CR-39 is physically able to register low energy alphas [21].

b. Irradiation systems

Radium (^{226}Ra; activity 5µCi) was used as an alpha radiation source (for the irradiation of alpha particle). For controlling the final alpha energies incident on the detector distances in normal air the distance between the radium source and the detector surface has been determined at $E_\alpha(0)$ is 5.49 MeV, the energy of alpha source and the source detector distances in air at normal temperature and pressure (NTP). The alpha particle range in the air is 4.08 cm. The range of alpha particle in air was changed by varying the source detector distance. Irradiation to the sources for 6 minutes irradiation at 2 hours interval was carried out [2].

c. Etching solution

The sodium hydroxide aqueous solution (NaOH) of 6.25 mol per liter was used as the etching solution. The etching was done in a bottle with a tight lid to prevent change in the concentration of the etching solution due to vaporization of water and absorption of moisture. The etching process is performed in a

thermostat bath which is electronically stabilized at 70 ºC. The chemical etching process involves 6.25M NaOH at 70±0.5 ºC, distilled water and a water bath-''gotech testing machines inc'' modelGT-7039-M,220V, 50 Hz. was used for heating the etching (6.25N NaOH) water solution at 70 ±0.5 Cº for 8h [2].

d. The bulk etch rates of the CR-39 in NaOH/ethanol

The standard determination of V_B is based on the measurement of the thickness of the detector after etching times and based on the determination of the mass of the detector after - before etching times. The thickness is measured with a digital micrometer of 1 ml accuracy in 10 positions on the detector foil and mass is weighted with an electric sensitive balance (Denver Instrument Company AA-200DS). The average bulk etch velocity was determined by using the above-mentioned values of V_B.

e. Irradiation and the etchings

The CR-39 detectors used in the present study are the ideal detectors (Intercast Europe srl (Italy) [21]. Each piece of CR-39 NTD for this study was irradiated at constant irradiation time [22] with alpha particles. Each detectors was been irradiated at 1 mSv/h with an ^{226}Ra source [23] (activity of 5µCi). After irradiation, these detectors were etched in an aqueous solution of NaOH by two methods. In the first method, many detectors were etched in different concentrations of N of NaOH maintained at 70 ºC for 8 hours of etching. In the second method, other detectors were etched in different time of etching with 6.25N of NaOH maintained at 70 ºC by a water bath ''gotech testing machines inc'' modelGT-7039-M, 220 V, 50 Hz". The etchant was continuously stirred in order to assure its homogeneity, which suggested the necessity of stirring to avoid the build up of etching products at the surface of the samples affecting etching rate. After etching, The sample detector was then taken out from the etchant, washed for 20 min in running water in order to definitively stop the etching process and removing the NaOH present on the detectors surface the sample was then rinsed with distilled water. The detectors were dried in air completely and then used for analysis [24].

f. Track Counting System / scanning process of CR-39NTDs:

After the chemical etching, equations (2) and (5) were employed to calculate the bulk etch-rate and track etch rate, for the thickness measurement, mass-measurement and diameter-measurement methods, respectively. The thickness and weight of each detector was measured before and after etching. The thickness removed layers were measured (micrometer was used to measure the alpha induced track thickness removed layer) and the mass of detector was weighted (using an electronic balance having an accuracy of 0.1 mg. For the scanning process, etched tracks were observed using an optical microscope fitted with a magnification of (40-100)X. The microscope image was viewed using a high quality monochrome charge coupled device (CCD) TV camera. The track densities ρ (number of tracks per unit area) are then found. Optimum values of time of etching and concentration of normality NaOH were determined. V_B, V_T and etching efficiency and the critical angle were calculated using equations in described in Ref.[1].

Result and discussion

The ideal detector used in this study is the CR-39 polymer(Italy). When an alpha particle crossing a cast CR-39 sheet, it leaves a trail of broken polymer links along its path (restricted to a microscopic cylindrical region). The CR-39 detectors irradiated

with alpha particle from ^{226}Ra source were etched in normality NaOH /1-propanol solutions. Various concentrations and time of etching were used to achieve the optimum processing conditions. For comparison purposes, CR-39 detectors were also etched in conventional aqueous solution of NaOH at different concentrations at 70 ºC. After weighting the mass of detector, V_B, V_T and f were determined using the Eqs.(2), (5) and (13) respectively as follows.

a. In the bulk etch-rate using thickness-measurement method, the values of diameter (D) have been calculated according to Eq.(8). The average value of diameter (D) is (12.36 ± 0.012) µm when the irradiation time is fixed (5min) and the etching time is changeable at 6.25N NaOH with (70±0.5) ºC. When the optimum etching time is 8hours, the value of diameter at this value of optimum time of etching is (15.70 ±0.025) µm and the value of thickness at this time is (7.85 ±0.035) µm, as shown in Table (1).

b. In the bulk etch-rate using mass-measurement method, the bulk etch-rate $V_B(M)$ is not a unique function of mass but depends up on either one or more the combination of other parameters namely, density of CR-39 is 1.31g/cm^3 [19], thickness is (700 µm) [21], area of CR-39 is 1.5cm x1cm as appear in Eq.(2). The mass of the CR-39NTD was measured before and after etching. The average value of diameter is (11.98 ± 0.028) µm when the time irradiation is fixed 6min and the etching time is 8hours and the concentration of normality N of NaOH is changeable with (70±0.5) ºC. when the optimum etching time is 8hours, the values of diameter at the optimum time of etching is (15,68±0.025) µm and the thickness is (7.84±0.035) µm, shown in Table (1).

The bulk etch-rate $V_B(M)$ was measured using the mass-measurement method in the first method, the average value of bulk etch-rate $V_B(M)$ is (1.190±0.091) µm/h when time of etching is changeable and the irradiation time is fixed at 6.25N NaOH with (70±0.5) ºC, but the value of bulk etch rate at the optimum time of etching is(1.208±0.090) µm/h. In the second method the average value of bulk etch-rate $V_B(M)$ is (1.172±0.092) µm/h when the etching time is fixed and the concentration of normality N of NaOH is changeable with (70±0.5) ºC, and the value of bulk etch rate at the optimum time of etching is (1.224 ±0.090) µm/h. Both methods generate good values of V_B. The value of V_B in the first is (1.208±0.090) µm/h, however, is in agreement with 1.2 µm/h[19] for an etchant concentration of 6.25N at 70 ºC for 8 hours in Refs[21], [25]

Two other, important parameters critical angle of etching , and track etching efficiency, have been evaluated using Eqs.(12) and (13). For the optimum time of etching at concentration 6 N at 70 ºC for 8hours. The critical angle of etching and track etching efficiency is 11.829 and %79.50, respectively. This is in agreement with Refs[21], [25]. The above suggested best etching conditions for CR-39 are useful for etching and for all samples.

Fig(1) shows the relation for the removed layer with the time of etching, when the time of etching is increased the removed layer also increased (linearly).

Fig(2) shows the relation of the diameter of track with the removed layer at 6.25N of NaOH concentration, when the removed layer increases, the diameter of track also increases (linearly).

Fig(3) shows the change in bulk etch rate V_B with normality of an aqueous solution of NaOH. When the concentration of normality is increased, the

V_B also increased but not linearly, therefore. Thus the first method of changing the etching time with fixed normality is much better and accurate. This method gave a good efficiency of %79.50 and approximates the real value of the detector efficiency of %80 used in this work and in agreement with the value in Refs.[25],[21].

Fig(4) shows the relationship for the efficiency of track with the time of etching at 6.25N of NaOH concentration and 70 ºC. When the time of etching increases, the efficiency of track increases until reaching 8hours then decreased, therefore the optimum time etching is 8hours. The relationship between track diameters and etching time is displayed in Fig. 3.

The linear fitting of experimental data is described in Tables (1) and (2).

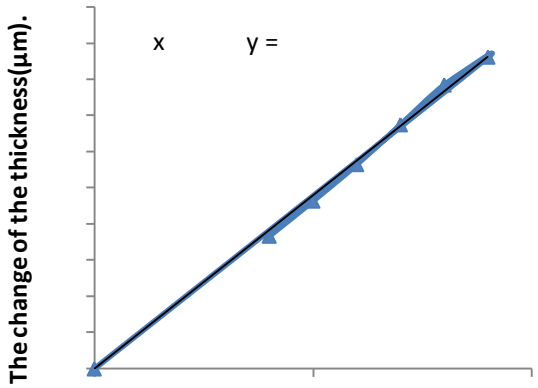

Fig. 2. Evolution of the detector thickness as a function of the etching time (h).

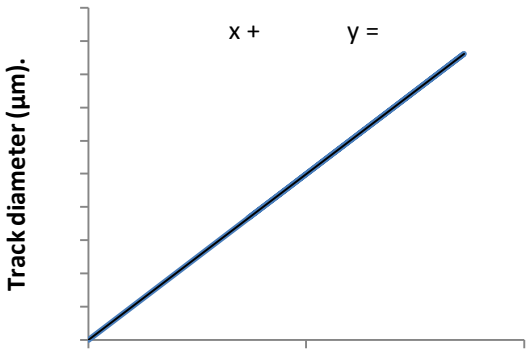

Fig.3.Track diameter versus removed layer for alpha particles at 6.25N NaOH concentration.

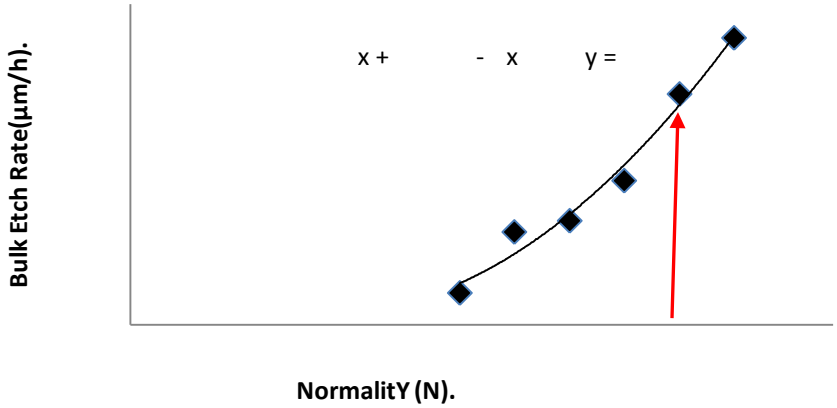

Fig. 4. The change of bulk etch rate with normality of an aqueous solution of NaOH.

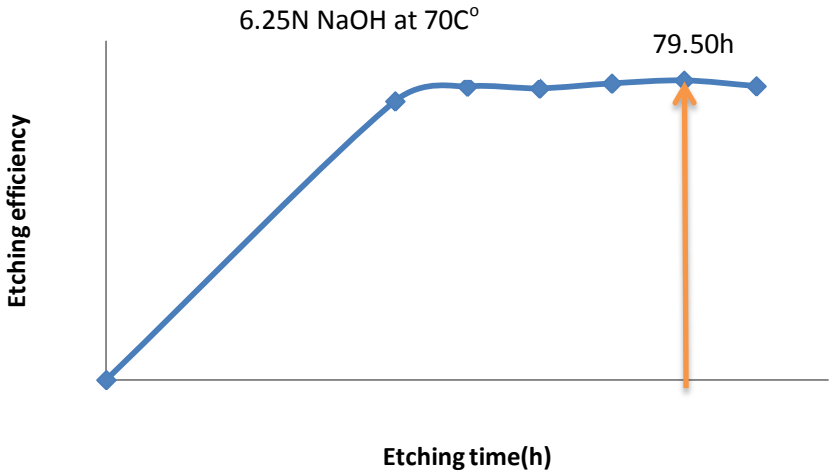

Fig. 5. Etching efficiency as a function of the etching time (h).

Table 1: Values of V_B,V, h, D, f and Θ_c for alpha particles of various etching in CR-39NTD.

Nu. Of detector	Thickness removed layer (µm)	Track diameter (µm)	Δm −Change of mass (m gm)	V_B (µm/h)	Track etch rate ratio V = V_T/V_B	Track length-L (µm)	Critical angle (Θc)	Efficiency f %
1	8.62	17.24	0.00425	1.199	4.526	38.043	12.764	77.93
2	7.85	15.70	0.00380	1.208	4.878	37.472	11.829	79.50
3	6.75	13.50	0.00330	1.197	4.697	30.975	12.364	78.71
4	5.65	11.31	0.00281	1.185	4.427	24.366	13.055	77.40
5	4.64	9.28	0.00232	1.178	4.271	19.265	13.540	76.59
6	3.67	7.35	0.00185	1.174	4.161	14.844	13.905	75.97

Table 2: Values of V_B,V, h, D, f and θ_c for alpha particles of various concentration in normality NaOH in CR-39NTD.

Nu. Of detector	Thickness removed layer (µm)	Track diameter (µm)	Δm −Change of mass (m gm)	V_B (µm/h)	Track etch rate ratio V = V_T/V_B	Track length-L (µm)	Critical angle (θc)	Efficiency f %
1	8.00	16.00	0.00425	1.259	4.411	34.352	13.000	77.37
2	7.84	15.68	0.00380	1.224	4.570	34.950	12.630	78.19
3	7.45	14.90	0.00330	1.170	4.456	32.340	12.960	77.56
4	7.24	14.48	0.00281	1.145	4.404	31.178	13.120	76.90
5	7.09	14,18	0.00232	1.138	4.087	28.104	14.160	75.50
6	6.83	13.66	0.00185	1.100	4.029	26.640	14.370	75.18

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