Evaluation of CR-39 Track Parameters by Utilizing Chemical Organic Additives to NaOH Etchant

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Abstract

The nuclear track detector, CR-39, is widely used for detecting Alpha-particles which produce latent tracks on it and can be enlarged by using a suitable chemical etchant to be visible under an optical microscope. The commonly used etchant is an aqueous 6.25 N solution of NaOH at 70° C. The behavior of the track parameters were studied at different etching times using direct measurements of the track opening diameters. These parameters were re-evaluated after adding some organic solution to the Sodium Hydroxide etchant to improve the etching conditions and so the quality of the tracks. These parameters including the track density, track diameter, the bulk, and the track etch rates, as well as track length, and depth. It has been found that Methanol and Ethanol additives led to a decrease of the bulk and track etch rates but with about 30% increasing of the track etch rate ratio compared with their values in case of using NaOH without any additives. In the case of using Benzene and Acetone additives, there is no remarkable changes in these parameters. On the other hand, the etching efficiency was increasing more than the double in case of Methanol and Ethanol and decreasing near the half in case of benzene and acetone.

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1. Introduction

1.1 CR-39

Heavy charged particles can leave latent quasi-continuous trails of damage in dielectric media, which are relatively long-lived at normal temperature. Different dielectrics have different 'registration thresholds', such that only those particles which have linear rates of energy loss, above a critical value, will be registered in a given dielectric medium. These polymers have far lower registration thresholds than glasses and mineral crystals, and can therefore often record tracks of such low atomic numbers as alpha particles.

The net effect of the energy loss is the production of bond rupture to form a complex array of free radicals, ionized molecules and radical ions along the track of

heavy ion. These broken molecular chains reduce the molecular weight of the detector substance which increases the rate of a chemical attack on the detector. Subsequently, the area of the broken molecular chain (scission) is dissolved, and the resulting hole can become enlarged by factors of 10^2 to 10^3.

Solid-state nuclear track detectors (SSNTDs) have been successfully employed in different applications in science and technology [1], including in alpha auto-radiography [2], radiation dosimetry [3] and particle identification [4].

Among SSNTD, polycarbonate has the advantage that it can reveal tracks of low energies within the limit of the alpha energies. Among polycarbonate, Polyallyldiglycol carbonate with molecular formula $C_{12}H_{18}O_7$ is one of the most sensitive materials for recording alpha particles. These materials are commonly referred to as CR-39. Actually, CR is a registered trademark referring to Colombia Resin #39, products of Pittsburgh Plate Glass Company. The most important features of CR-39 are its sensitivity, stability and the uniformity also it has an important feature that it can alter its response within limits by suitable choice of etching condition. CR-39 is used extensively to give data in a different field such as Brachytherapy [5], Boron Neutron Capture Therapy BNCT [6-7], U-exploration Galleries [8-9], and measurements of Radon in building materials [10].

1.2 Track Geometry and Profile.

The damaged region resulting from the passage of a heavy ion through SSNTD will be chemically more reagent than the surrounding undamaged matter. This damaged region is attacked by a higher rate if it is treated with a properly chosen chemical reagent.

There are two kinds of etching rates that are the bulk etching rate for the undamaged material V_B and the track etching rate V_T for the damaged one, where the etching solution penetrates along the latent track. V_B and V_T are sufficient to describe the etched track where the simultaneous action of them can create a cone that has the original track as its axis. A result of competition between the effect of V_B and V_T is the visible track depth L_{depth} and track diameter D as shown in Figure (1).
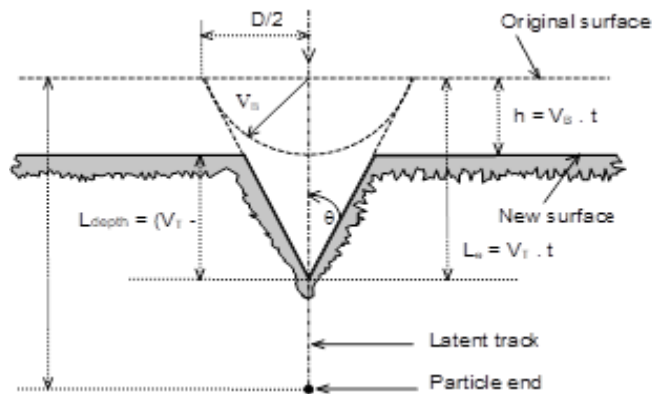
Fig.1. The etched track profile and its parameters at constant VT for a particle lying normal to the detector surface.

V_T must be exceed V_B to get a visible track, where the smaller the excess of V_T over V_B the larger the cone angle θ (=arc sin V_B / V_T).
The bulk etch rate V_B is calculated using the following equation:
$$V_B (h) = \Delta h / 2\Delta t \qquad (1)$$
Where Δh is the layer removed during the etching time Δt [(11].

The diameter etch rate V_D is calculated by measuring the diameter of alpha tracks at different etching times; then calculated by using the following equation:

$$V_D = \Delta D / \Delta t \tag{2}$$

The track etching rate V_T was calculated using the following equations [12]:

$$V_T = V_B \frac{4V_B^2 + V_D^2}{4V_B^2 - V_D^2} \tag{3}$$

There are many parameters used to describe the geometry of etched track, these are

- The full length of the latent track (L).
- The thickness of the surface removed by etching (h).
- The diameter of the etch pit (D).

The track length (Le) at etching time (t) is given by the following relation.

$$Le = V_T\, t \tag{4}$$

The surface is also begun removed at a rate V_B so that the track depth is given by the following relation [13]

$$L_{depth} = V_T\, t - V_B\, t \tag{5}$$

The etching efficiency (η) is defined as the ratio of the counted tracks and the number of particles incident on the detector surface, the efficiency depends on the track etched rate V_T and the bulk etched rate VB as follows:

$$\eta = 1 - \frac{V_B}{V_T} \tag{6}$$

The main objective of this paper is to improve the condition of track formation on CR-39 by using different additive to the etching solution and study different track etching parameters, including V_B, V_T, Le, L_{depth} and η.

2. Method Methodology

2.1. Radiation exposures of CR-39 detectors

CR-39 detector sheet of thickness 700 μm, manufactured by American Acrylics (US), was cut by laser beam into pieces of area 1×1 cm^2. CR-39 detectors were preserved in special plastic envelopes. All samples were exposed to ^{241}Am source (activity was 0.106 μCi and main alpha energy was 5.49 MeV). A collimator was used such that the incident alpha particles have energy of 3 MeV and 90° incident angle in air.

2.2. Chemical etching of CR-39 detectors

Immediately after irradiation, the detectors were etched in a 6.25 N aqueous solution of NaOH at different etching times equal to 2, 4, 5 to 8 h maintained at temperature of 70 ^{o}C in a water bath. The optimum etching time must be short enough to avoid over etched tracks [14]. The detectors were then taken out from the etchant, rinsed with distilled water, and dried. CR-39 detectors were then counted by an optical microscope to determine the track density in terms of tracks per cm^2. The background track density was determined by processing a virgin detector under the same etching conditions, and then the background was subtracted from the measured track density, to obtain realistic statistics of the tracks.

The other part of this work is the use of different solutions such as ethanol, methanol, benzene, and acetone mixed with sodium hydroxide solution and etched at the same optimum etching conditions.

3. Results and Discussion

3.1. The track parameters for NaOH etchant

Figure (2) shows the relation between the track densities at different etching time where the maximum track density is obtained around 6 hours etching times.
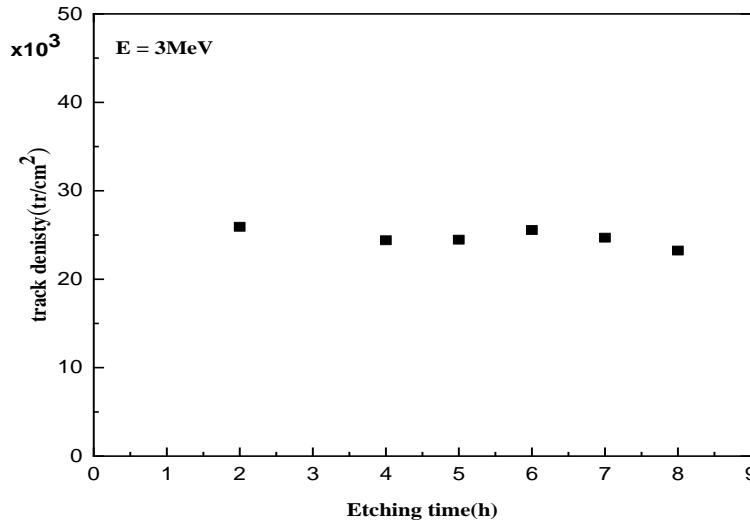

Fig. 2. The track density at different etching times at 6.25 N NaOH and 70° C.

The track diameter is increasing with etching time as shown in Figure (3) where the relationship is close to the linear form. At etching time more than 6 hours, the increasing of the track diameter is responsible for the decreasing of the track density due to the tracks overlapping.
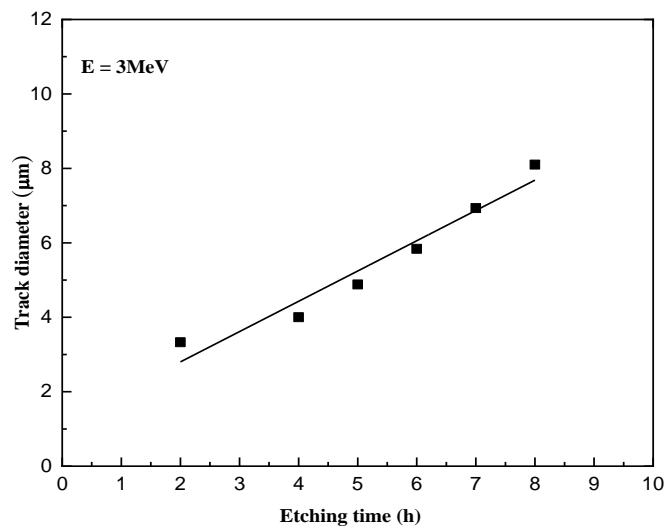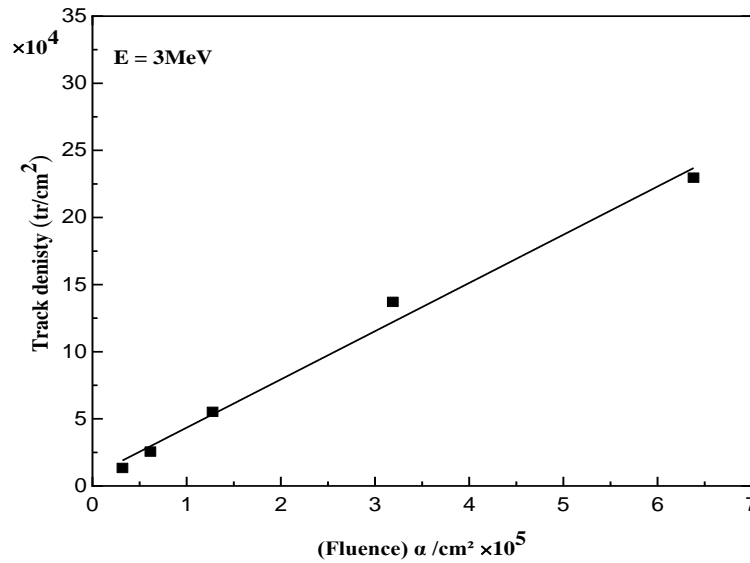

Fig. 3. Relation between the track diameter and the etching time at 6.25 N NaOH and 70° C.

The response of CR-39 is shown in Figure (4) as a relation between the fluence of the incident Alpha-particles and the registered track density. The relation is linear with a correlation coefficient equal to 0.99. The slope of the given line is equal to

0.36 ± 0.02 T/α which is the response of the CR-39 detector for 3 MeV normal incident Alpha particles.

Fig. 4. Relation between the fluence of alpha particles and the track density at the optimum etching condition of 6.25 N NaOH at 70°C for 6 hours.

After each etching time, the bulk etch rate V_B was calculated by measuring the removed layer Δh and use Equation (1), the mean value of V_B is equal to 1.89 µm/h which was shown as a horizontal line in Figure (5). The bulk etches rate V_B was measured to be 1.67µm/h under the circumstances of 6.25 N NaOH at 70°C for 6 h. This value as well as the average one is higher than that obtained by Ibrahim and Wathab, [15] who obtained a value of 1.29 µm/h using 6 N NaOH. Also, Najeba et al., [16] measured V_B as 1.174 µm/h, and Matiullah et al., [17] measured V_B as 1.25 µm/h.
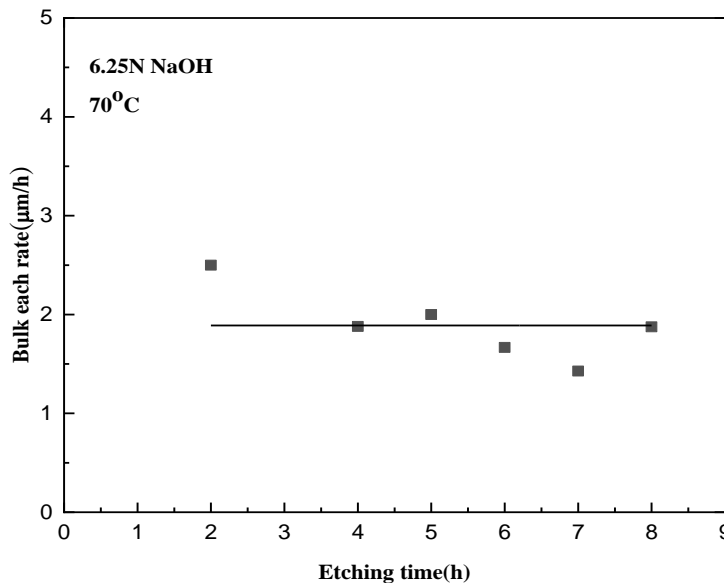

Fig. 5. The bulk each rate at different etching time.

Figure (6) shows the track etch rate determined from Equation (3) for different etching times ranging from 4 to 8 hours. The Figure shows that the track etch rate increases with time, peaking at around 5 hours etching time before declining. After 5 hours of etching, the decayed values of V_T are expected to hit their minimum value, which is equal to V_B. For 6 h etching time, The track etch rate is equal to 1.98 μm/h which consistent with the work of Ansari et al., [18] who found that V_T is ranged from 1.88 to 2.0 μm/h, for different manufactured CR-39 sheets with the same etching condition of 6 N NaOH at 70 °C.
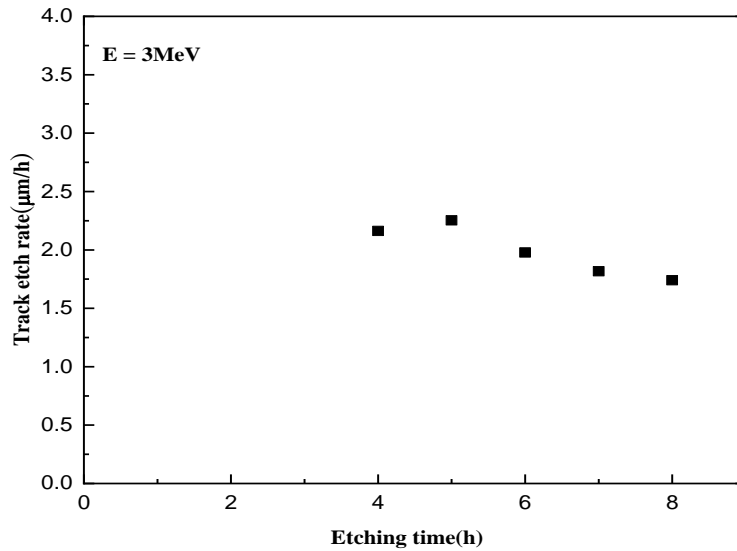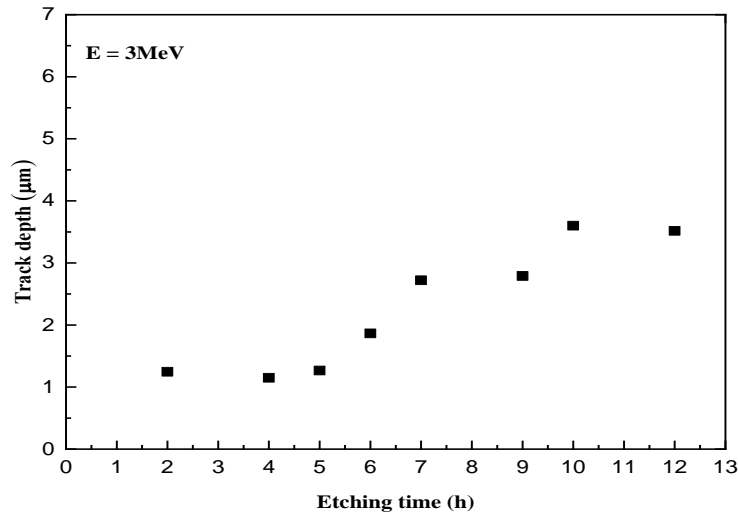

Fig. 6. Relation between the track etch rate and the etching time at 6.25 N NaOH and 70° C.

Also, the behavior of the track etches rate ratio V (which is the ratio between V_B and V_T) was found to be similar to that of V_T as shown in Figure (7). At 6 h etching time, V is equal to 1.19 and tends to reach 1 at the etching time higher than 8 hours.
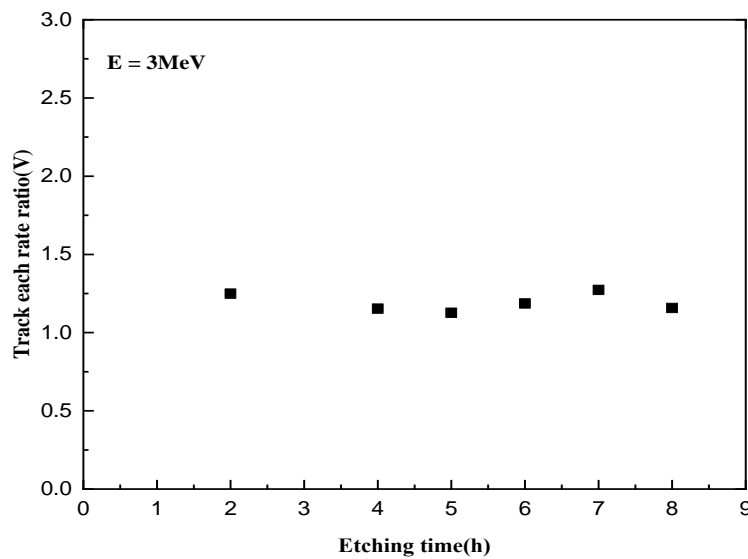
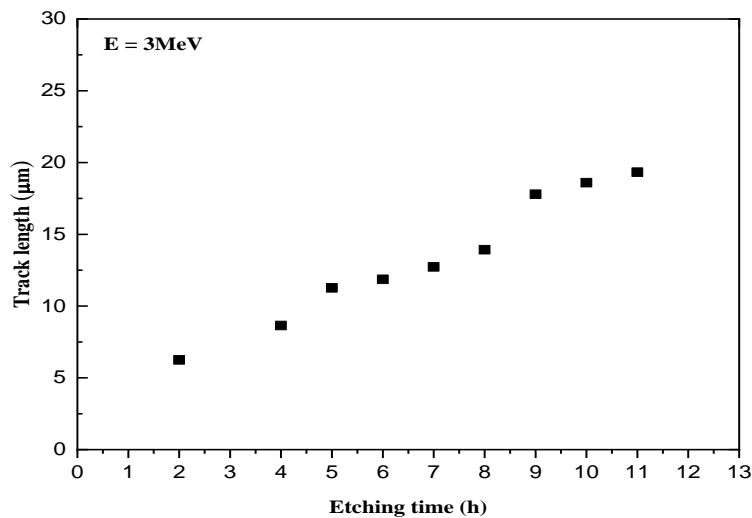**Fig. 7.** Relation between the track etch rate ratio and the etching time at 6.25 N NaOH and 70° C.

The track depth and the track length as a function of etching time are shown in Figures (8, 9). It can be observed that the track depth increases with the etching time up to a maximum constant value at the end of the track. Noting that the track length increases gradually with etching time leading to the maximum value at the end of the path of a particle in the detector, or the greatest absorption of energy, Figure (8).


Fig. 8. Relation between the track depth and the etching time at 6.25 N NaOH and 70° C.

The tracks lengths were plotted as a function of etching time in Figure (9). The track lengths are not linearly changing with etching time, they are growing gradually up to a maximum value and then access to constant values when the etching solution crossing the region under the end of the particle range in the detector. With the continued etching process, V_T decreases and drops sharply so that it equals the bulk etch rate (V_B), and then the etching ratio (V) ends to one (V=1) after the etchant completely reaches the end of the particle range. Applying the limited condition is that when the etching solution reaches the end of the incident particle range, further prolongation of the etching time does not result in the further growth of the track length because the etching now takes place with the bulk etch rate in all directions and this appears at etching times more than 8 hours.

Fig. 9. Relation between the track length and the etching time at 6.25 N NaOH and 70°C.

Figure (10) shows the efficiency as a function of etching time, which was calculated from the track parameters as given in Equation (6). At 6 hours of etching time, the efficiency is 0.16. The etching efficiency increases slightly with etching time, reaching a maximum of 0.28 after 8 hours, then dropping to a minimum of about 0.11 after 12 hours etching time.
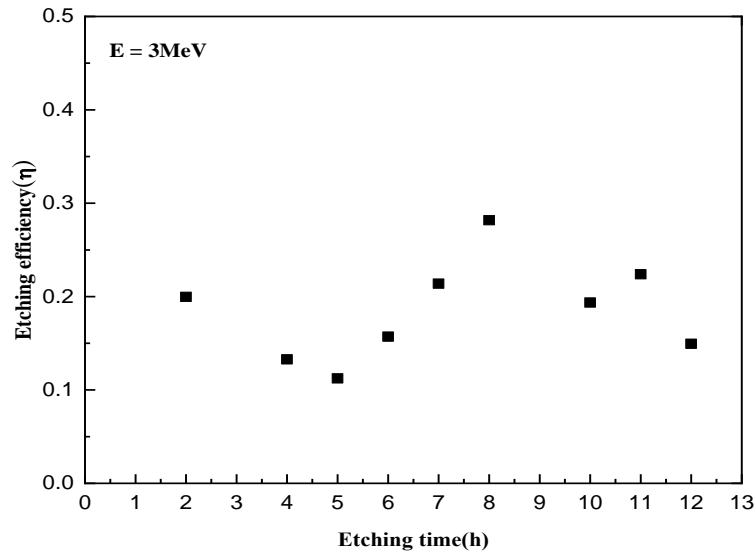
Fig. 10. The etching efficiency as a function of the etching time at 6.25 N NaOH and 70°C.

3.2. The track parameters using different etchant additives

After adding 1 ml of Ethanol, Methanol, Benzene, or Acetone to 6.25 N NaOH solutions using an etching time of 6 h at 70°C, the track parameter were re-evaluated and compared to that obtained at the optimum etching condition using NaOH without any additives.
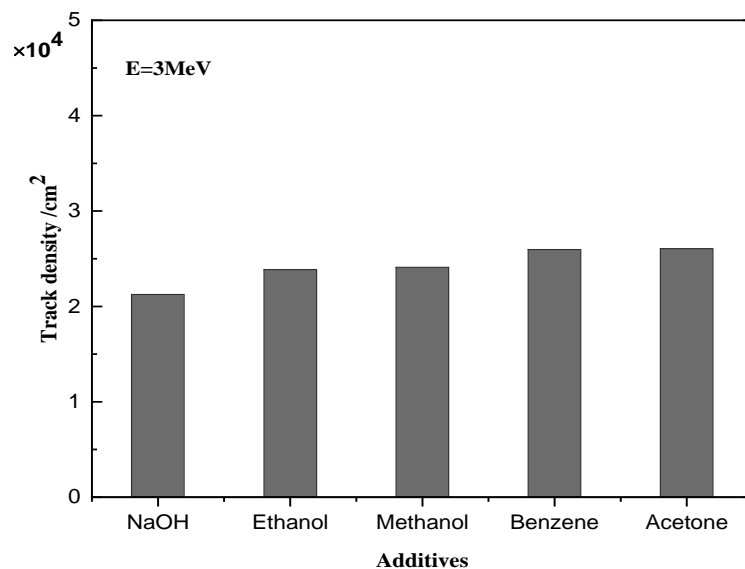
Fig. 11. The track density using 1 ml of different additives to 6.25 N NaOH solution at 70°C for 6 hours etching time.

Figure (11) shows the track density for CR-39 using these additives compared with NaOH without any additives. The Figure indicates a slight increase in the track density, especially for Benzene and Acetone. Otherwise, the track diameters are decreasing with using these additives as shown in Figure (12).
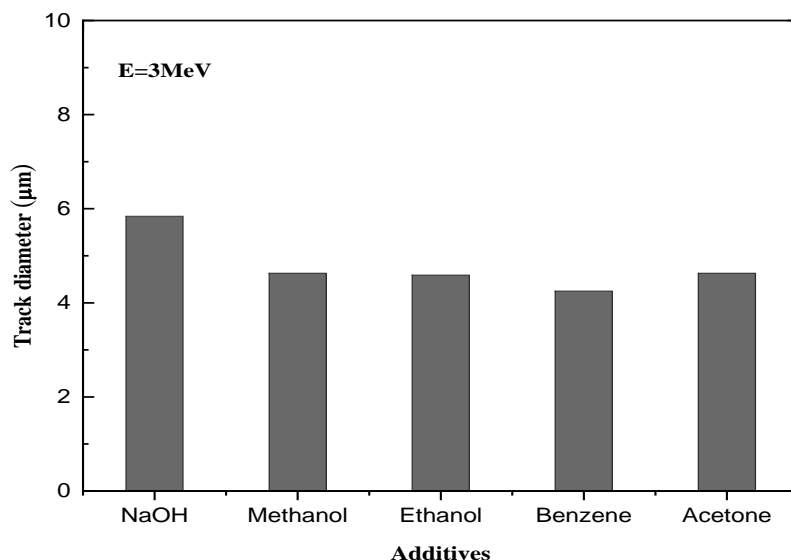
Fig. 12. The track diameters using 1 ml of different additives to 6.25 N NaOH solutions at 70°C for 6 hours etching time.

The bulk etches rate V_B was re-evaluated after adding the different organic solutions. The additives of methanol and ethanol displayed a decrease in the value of V_B with about 50% (a value of 0.83 μm/h) while V_B remains unchanged for benzene and acetone (Table 1). For ethanol, Vivek et al., [19] varied the normality from 1.5 up to 3 with an increment of 0.5 and found that the values of V_B changed to 3.0, 3.99, 5.32, and 11.6 μm/h. When Ibrahim and Wathab, [15] changed the additive to be methanol with normality values of 1.5, 2, 2.5, and 3, the values of V_B were increased dramatically to be 3.009, 3.99, 5.32, and 7.38 μm/h. While Ashry et al., [20] measured V_B under different conditions (10 N NaOH + methanol, 50-70°C, and 45 min etching time) to be 2.73 μm/h. It can be deduced from the current and previous research that the bulk etch rate is highly affected by the additives and concentration of the etching solution.

It was observed that the track etch rate V_T decreased about 35% after adding ethanol solution (1.28 ± 0.39) μm/h and methanol solution (1.29 ± 0.39) μm/h. For benzene and acetone, V_T is slightly decreasing (about 8%) to give values equal to (1.82 ± 0.0071) μm/h and (1.86 ± 0.014) μm/h respectively (Table 1).

After adding organic solutions, the values of V were changed as shown in Table (1) from 1.19 (in the case of NaOH solution) to reach 1.09, 1.11, 1.53, and 1.55 for benzene, acetone, ethanol, and methanol respectively as listed in the Table (1). These results revealed that the tracks etch rate ratio (V) increases for methanol and ethanol by about 30% and slightly decreases in the case of benzene and acetone.

Table 1. The measured track parameters for different organic additives to 6.25 N NaOH.

Additives	V_B	V_T	V	θ_c	η	L_{depth}'	L_e
NaOH	1.67	1.98	1.19	57.44	0.16	1.86	11.86
Methanol	0.83	1.29	1.55	40.27	0.35	2.73	7.73
Ethanol	0.83	1.28	1.53	40.7	0.35	2.67	7.67
Benzene	1.67	1.82	1.09	65.98	0.09	0.95	10.95
Acetone	1.67	1.86	1.11	63.94	0.10	1.13	11.13

After adding the organic solutions, the results of track depth and length were reassessed; it was found that the track depth was increased in the case of methanol and ethanol, in contrast to what happened with the benzene and the acetone which started to decrease less than that of using NaOH (Figure 13). The etchant attacks the damaged region regardless of the used additives. But where methanol and ethanol show a reduced effect on the bulk etch rate then the depth of the track seems to be larger than the case of NaOH. Also, the bulk etch rate is larger in the case of benzene and acetone (equal to that of NaOH), then the depth, in this case, seems to be smaller like that of NaOH.
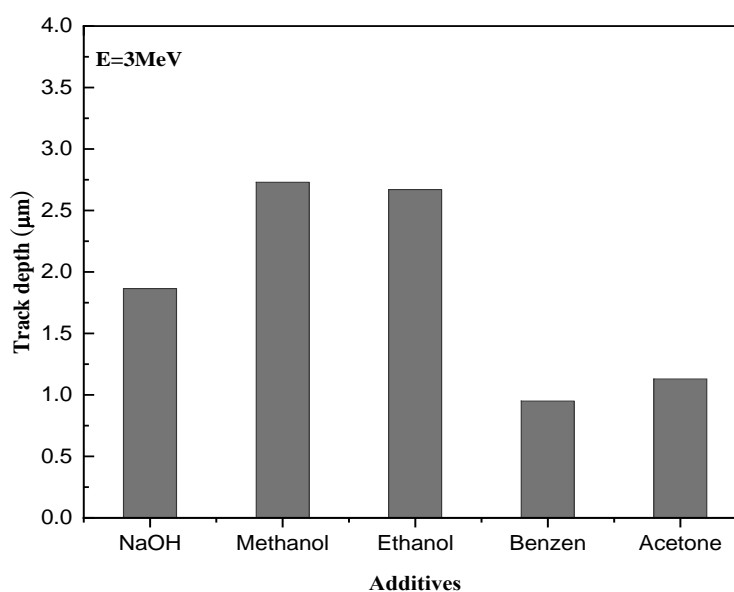
Fig. 13. The track depth using 1 ml of different additives to 6.25 N NaOH solutions at 70o C for 6 hours etching time.

But when determining the track length after adding the organic solutions, it was found that ethanol and methanol showed track lengths smaller than that of NaOH, while benzene and acetone show larger track lengths but still less than NaOH (Figure 14). In this case, the track etch rate is the important parameter which is proportional to the track length and so has the same behaviour as shown in Table (1).
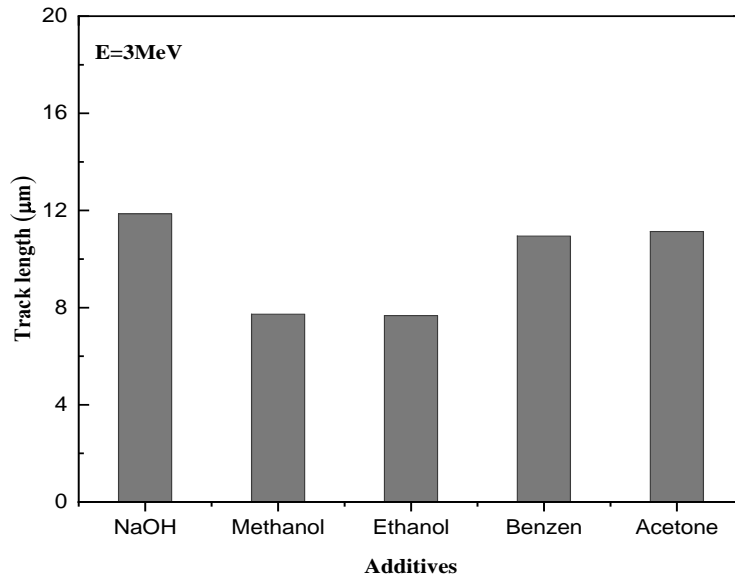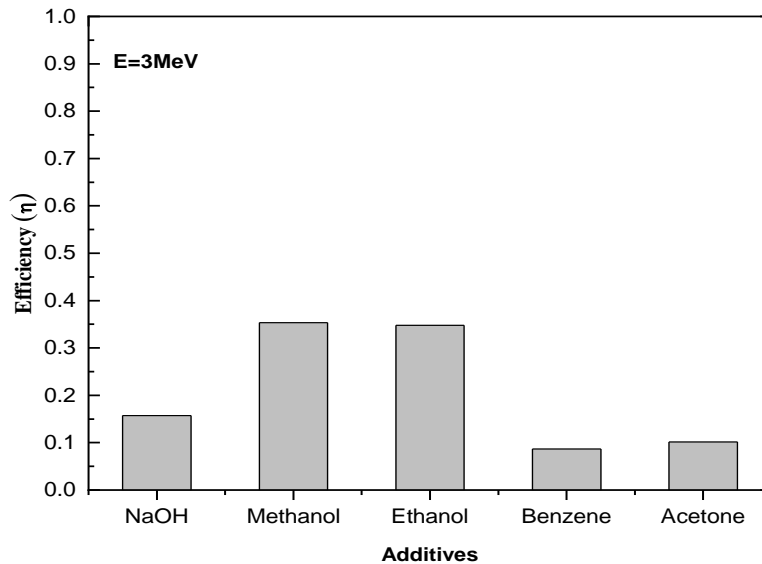
Fig. 14. The track length using 1 ml of different additives to 6.25 N NaOH solution at 70°C for 6 hours etching time.

The efficiency was re-evaluated after adding the used organic solutions. Comparing the results with that obtained for using NaOH without any additives, the efficiency was decreased 38% in the case of using benzene and acetone but increased more than twice (about 120 %) when using ethanol and methanol (Table 1) and Figure (15). This finding is a direct result of the combined effect of V_B and V_T as discussed before and this finally indicates some advantages of using ethanol and methanol as organic additives to the NaOH etchant.

Fig. 15. The detector efficiency using 1 ml of different additives to 6.25 N NaOH solution at 70°C for 6 hours etching time.

4. Conclusion

The track parameters of CR-39 were calculated for optimum condition of 6.25 N NaOH etchant at 70 ^{o}C at various etching times. The bulk etch rate V_B and the diameter etch rate V_D are critical parameters, and accurate measurements of these parameters allow for the proper estimation of residual parameters like the track etch rate V_T, the track etch rate ratio V, the detector etching efficiency η, and the track length and its depth.

The values of V_B and V_T obtained at the optimum etching time of 6 hours are compatible with those obtained by several authors. The estimated etching efficiencies obtained from measured track parameters at various etching times have a maximum value of about 0.28, which is lower than the value obtained from direct measurement of track densities at various Alpha-particle fluencies under the optimum etching time (about 0.35).

When the organic solvent is added to the NaOH etchant, the determined parameters are affected differently according to the used additives. The effects of methanol and ethanol additives on V_T, V_B, V, and η are significant. As compared to NaOH without additives, the etchant attacks the bulk of the detector twice as quickly. As a consequence, the length of the track is shortened while the depth is increased. As a result of these additives, the etching condition is improved, resulting in higher detector efficiency.

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