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The use of CH₃OH additive to NaOH for etching alpha particle tracks in a CR-39 plastic nuclear track detector

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H I G H L I G H T S

- The value of bulk etching rate is equal to 2.73 μm/h.
- Fast detection of alpha particles in CR-39 detectors.
- Samples of CR-39 have been irradiated with fission fragments.
- Etching efficiency was determined.

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Fast detection of alpha particles in CR-39 detectors was investigated using a new chemical etchant. ²⁵²Cf and ²⁴¹Am sources were used for irradiating samples of CR-39 SSNTDs with fission fragments and alpha particles in air at normal temperature and pressure. A series of experimental chemical etching are carried out using new etching solution (8 ml of 10N NaOH + 1 ml CH₃OH) at 60 °C to detect alpha particle in short time in CR-39 detectors. Suitable analyzing software has been used to analyze experimental data. From fission and alpha track diameters, the value of bulk etching rate is equal to 2.73 μm/h. Both the sensitivity and etching efficiency were found to vary with the amount of methanol in the etching solution. Pure NaOH was used as a control to compare with the result from etching in NaOH with different concentrations of CH₃OH. The etching efficiency is determined and compared with conventional aqueous solution of 6.25N NaOH at 70 °C for etching time equals 5 h. In this study, the obtained etching efficiency shows a considerable agreement with the previous work.

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1. Introduction

Solid state nuclear track detectors (SSNTDs) have been successfully employed in different applications in science and technology (Price, 2005, 2008). One of the most commonly used detectors is CR-39, because of its good sensitivity, stability against various environmental factors, and high degree of optical clarity (O'Sullivan and Thompson, 1980; Kodaira et al., 2009).

Chemical etching is an essential method being used in various studies of surfaces and technical applications (Fleischer et al., 1975). It is found that the fierce bulk etch rates for NaOH/ethanol etchants

compared to those for the 6.25N NaOH/H₂O etchant are due to the miscibility of ethanol with the organic etched products from etching the CR-39 detector (Tse et al., 2007). Recently, several new etchants are used to detect the registration of fission fragments and alpha particles tracks in the CR-39 detector, e.g., Matiullah et al. (2005a). They prepared etching solutions by including NaOH dissolved in methanol and NaOH dissolved in methanol/water (SMW) at 55 °C. In addition, they reported that these two etching solutions have higher etching efficiencies and much smaller etching time compared to the conventionally used solution 6 M aqueous NaOH at 70 °C. Matiullah et al. (2005b) introduced etching solutions as well as conventionally used 6 M NaOH at 70 °C for etching the CR-39 detector. These etching solutions included NaOH/ethanol and NaOH/1-propanol. They concluded that the etching efficiency of these two solutions is higher than that of aqueous NaOH. Ogura et al. (2001) showed that the etchant PEW solution (potassium hydroxide+ethanol+water) has

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high efficient etchant for CR-39 detectors to study the desensitization of proton tracks in the NCAR image taken by thermal neutron beams. Kodaira et al. (2009) reported that PEW solution works effectively to desensitize the registration sensitivity in the CR-39 detector.

The main objective of this work is introducing a new etchant solution for the CR-39 detector to investigate the etching characteristics of CR-39 for alpha particles registration in a small etching time.

2. Materials and methodology

CR-39 (TASTRACK) is a $C_{12}H_{18}O_7$ polymer with a density of 1.3 g/cm^3 and a thickness of $250 \text{ }\mu\text{m}$. CR-39 was cut into pieces with an area of $1.0 \times 1.0 \text{ cm}^2$. A group of CR-39 detectors were exposed to a thin open ^{252}Cf disk source as an alpha, fission fragment and fast neutrons source of activity $9.6 \times 10^{-3} \text{ }\mu\text{Ci}$ and surface area of 19.64 mm^2 . Another group was exposed to the thin ^{241}Am disk source that emits alpha with energy of 5.5 MeV and activity $0.924 \text{ }\mu\text{Ci}$. Using a variable length of air column, energies from 1 to 5 MeV were used.

Mixture of 10 N sodium hydroxide and pure methanol (CH_3OH) with different ratios was used as the etching solutions at 55, 60, 65 and $70 \text{ }^\circ\text{C}$. 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.

For an etching process, a water bath of stabilizing temperature with accuracy $\pm 0.5 \text{ }^\circ\text{C}$ was used. All etched samples were held at the same depth in the etchant solution. After etching the samples were immersed in running water for suitable time interval to remove all etchant products from the surfaces. Finally, the samples were carefully dried and then used for analysis. For estimation of track diameter an optical microscope fitted with a magnification of $400\times$ was used. The microscope was connected with a web digital camera to capture the sample image from the microscope and save it in P.C unit; software program (INFINITY ANALYZE software) was used to analyze the tracks after calibration.

The bulk etch rate V_B is the rate of removing of the undamaged surface of the detector due to the chemical reaction between the etching solution and the detector material, and it can be determined using the following equation:

$$V_B = \frac{D_{ff}}{2t_e} \quad (1)$$

where D_{ff} is the track diameter of fission fragment projectile that bombarded the detector and t_e is the etching duration. The traditional method for determining the track etching rate of a CR-39 solid state nuclear detector is based on the measurement of track diameter (D). The relation between V_T and D takes into account the removal of a detector layer $h = V_B t$ and bulk etching rate (Durrani and Bull, 1987). In this method CR-39 detectors were exposed to normal incident alpha particle of energy 2 MeV and then etched for etching time 45 min with different etching concentrations at different etching temperatures. From the track diameter and removed layer the track etching rate (V_T) was determined.

The sensitivity (V) of an etched track detector to alpha particle is a strong function of detector material properties, particle charge, energy and direction of the incident ions. The sensitivity (V) of the detector has been determined from the following equation (Durrani and Bull, 1987):

$$V = \frac{V_T}{V_B} = \frac{h^2 + r^2}{h^2 - r^2} \quad (2)$$

where V_T is the track etching rate, h is the removed layer and $2r$ is the track diameter. The critical angle (θ_c) is one of the geometrical

limitations for the revelation of the etched tracks in SSNTDs; it can be determined by this equation (Yu and Nikezic, 2009):

$$\theta_c = \sin^{-1} \frac{V_B}{V_T} \quad (3)$$

The etching efficiency of a detector is defined as the proportion of tracks etched out expressed as a fraction of particles actually incident on the detector surface. The etching efficiency of the detector (η) can be measured using the following equation (Yu et al., 2005):

$$\eta = 1 - \sin \theta_c \quad (4)$$

3. Result and discussion

For etching time reduction, great deal of efforts was done in order to achieve the goal of this work. Our basic movement was in the direction of finding out an optimum new etching condition to detect alpha particles registration in the CR-39 solid state nuclear track detector in short time. We test various stages of etching conditions until a best acceptable condition was reached. This new suggested condition was 8 ml of $10\text{N NaOH} + X \text{ ml CH}_3\text{OH}$ at $60 \text{ }^\circ\text{C}$. In this work, we study the effect of adding CH_3OH to the etching solution in the etching parameter (bulk etching rate, track etching rate, sensitivity, etching efficiency and critical angle).

Bulk etch rate of CR-39 was determined in temperature range from 55 to $70 \text{ }^\circ\text{C}$ in step of $5 \text{ }^\circ\text{C}$ using the new etching condition with different amounts of methanol, as shown in Fig. 1. Results show that the bulk etching rate increases with increasing the amount of CH_3OH and increases with increasing the temperature. The variation of V_B with etching time at different amounts of CH_3OH at $60 \text{ }^\circ\text{C}$ is illustrated in Fig. 2. One can notice that values of V_B at $3 \text{ ml CH}_3\text{OH}$ are higher than those of $1 \text{ ml CH}_3\text{OH}$.

Fig. 3 depicts the relation between $\ln V_B$ and $1000/T (\text{K}^{-1})$ with different amounts of methanol. The dependence of $\ln V_B$ on $1000/T (\text{K}^{-1})$ reflects an exponential behavior of V_B with T ; it can be denoted by the relation:

$$\ln(V_B) = \ln(A) - \left(\frac{E_B}{kT} \right) \quad (5)$$

where A is a constant, E_B is the activation energy of etching, and k is the Boltzmann constant.

Activation energy is the energy required for activating the reaction between the detector material and the etching solution and it tells us about the kinetics of the reaction. Activation energy of etching is a measure of the thermal energy required to start bulk etching. Using a least square fit method the values of the bulk activation energy E_B for the CR-39 detector were calculated from

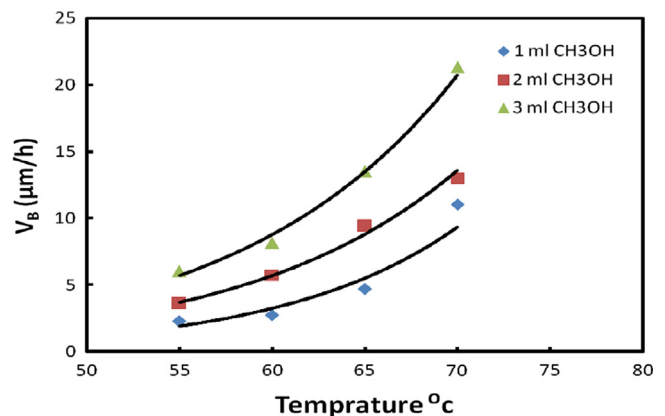


Fig. 1. Variation of V_B with temperature at different amounts of CH_3OH .

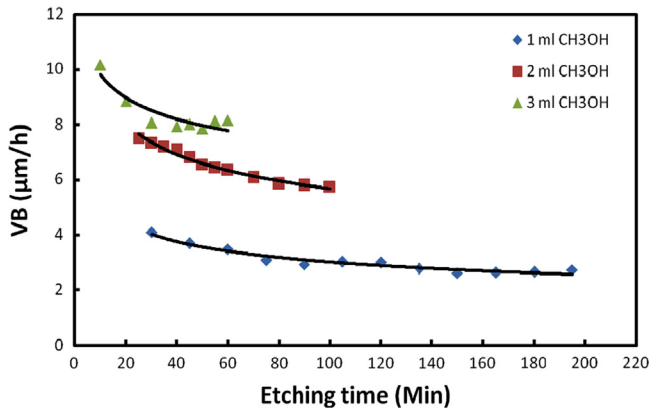


Fig. 2. Variation of V_B with etching time (min) at different amounts of CH_3OH at 60°C .

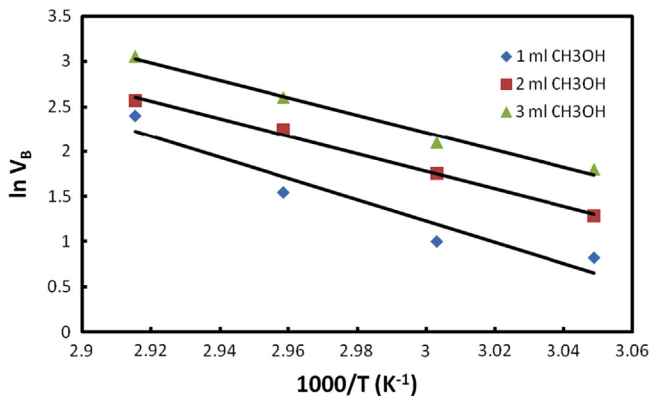


Fig. 3. Variation of $\ln V_B$ and $1000/T$ at different amounts of CH_3OH .

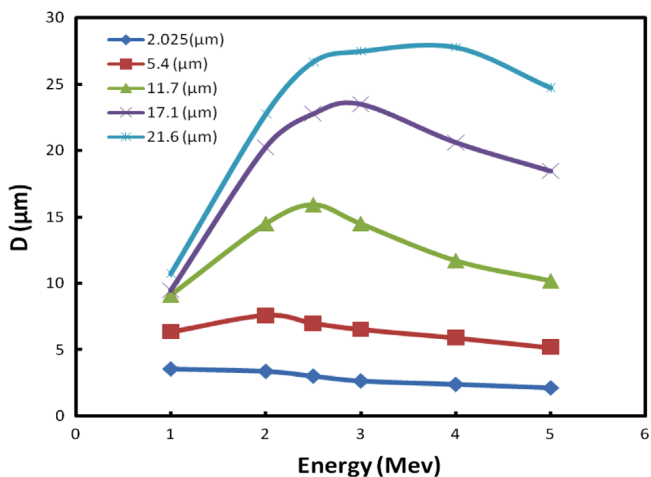


Fig. 4. Variation of alpha particle track diameter with energy at different removed layers (μm) etched in $1\text{ ml CH}_3\text{OH}$ at 60°C .

the slopes of the straight lines shown in Fig. 3. It shows that the average value of E_B was found to be 0.88 eV . In addition, the activation energy of track etching rate was calculated; its average value $E_T=0.718\text{ eV}$ was obtained. Thus track activation energy, E_T is found to be lower than the bulk activation energy E_B . These results are in agreement with previous work (Matiullah et al., 2005a).

Fig. 4 demonstrates the relationship between the energy of alpha particle and the etch pit diameter at different removed layers (μm). It shows that under the new etching conditions which

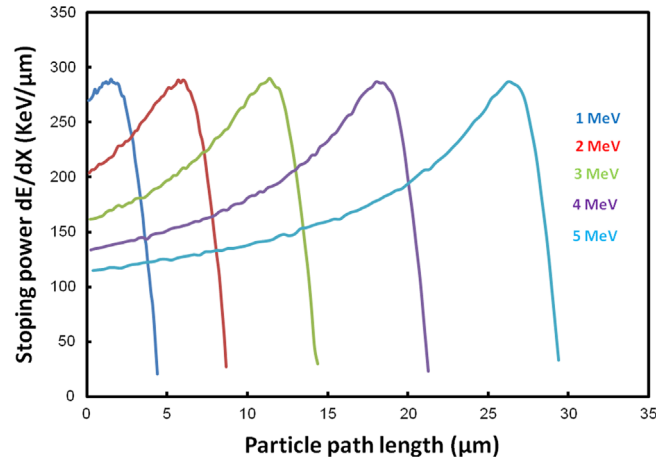


Fig. 5. Alpha particle energy loss as a function of particle path length in the CR-39 detector, data calculated using TRIM program (Ziegler et al., 2003).

used in this work, when the removed layer increase up to $2.0\ (\mu\text{m})$, the track diameter of 1 MeV alpha particles started to increase relative to 2 MeV alpha particles. This result may refer to increase in the path of 1 MeV alpha particles, after this point the value of damage inside the material started to decrease and the rate by which the track diameter was increasing started to decrease. Fig. 4 depicts the specific maxima, which are shifted to higher alpha particle energies for the higher removed layer. It was also found earlier that the location of the maximum is defined by the thickness of the external detector layer removed during the bulk etching process, and the range of the projectile in the detector material, as shown in Fig. 5. This means that tracks are etched out to the largest diameters only when the etching solution has unconstrained access to the end part of the particle trajectory, where the concentration of detector material defects is the highest. This takes place after removal of the detector external layer of a thickness equal to the projectile range (Szydłowski et al., 2013). This result supposes the availability of calibration curves of the track diameter versus particle energies for different removed layers (Sadowski et al., 1994).

The sensitivity was determined using Eq. (2); it was obtained by measuring the track diameter and the removed layer. The variation of the sensitivity of the detector with alpha particle energy when it etched with different amounts of CH_3OH at 60°C for etching time of 45 min shows that the sensitivity decreases with increasing alpha energy from 1 to 5 MeV and also decreases with increasing the amount of CH_3OH in the new etching solution.

Critical angle was determined using Eq. (3). The values of the critical angle increase with increasing both the amount of CH_3OH at 60°C for etching time of 45 min , and the energy of incident alpha particles. This implies that the critical angle strongly depends on alpha particle energy. The relation between the etching efficiency as a function of alpha particle energy at different etching times, using our new etching condition (8 ml of $10\text{N NaOH}+1\text{ ml CH}_3\text{OH}$) at 60°C , is depicted in Fig. 6. It is observed that the etching efficiency increases with decreasing the etching time. From the obtained results, one can say that the optimum new etching condition is 8 ml of $10\text{N NaOH}+1\text{ ml CH}_3\text{OH}$ at 60°C for 45 min etching time.

The etching efficiency of CR-39 was determined from the critical angle measurements. Fig. 7 shows the variation of the etching efficiency of the detector with alpha particle energy at different amounts of CH_3OH at 60°C for etching time of 45 min . The results show that the etching efficiency decreases with increasing alpha energy from 1 to 5 MeV and also decreases with increasing the amount of CH_3OH . In addition, Fig. 7 includes a

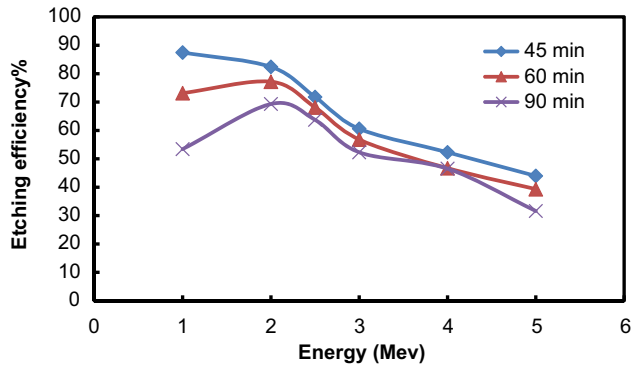


Fig. 6. Etching efficiency as a function of alpha particles energy at different etching times.

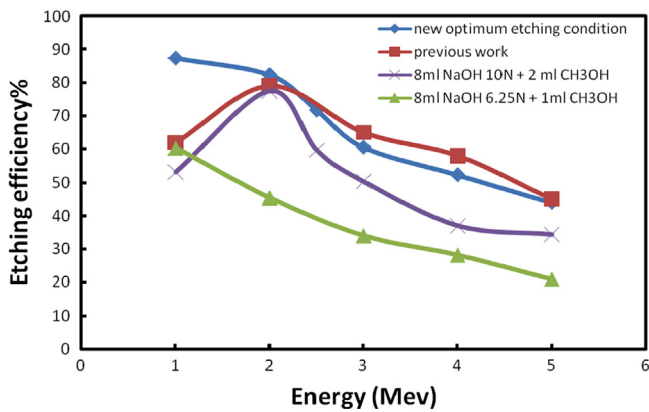


Fig. 7. Etching efficiency (η) as a function of alpha particles energy etched at different amounts of CH_3OH at 60°C and comparison between it in previous work and this study. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

comparison between the etching efficiency values for CR-39 detectors which are obtained in this work, as well as obtained by etchant in conventional aqueous solution of NaOH, and that obtained by El-Hawary et al. (1999). The green line shows the efficiency of the detector after etching time equals 1.5 h under etching condition (8 ml of 6.25N NaOH+1 ml CH_3OH at 60°C) with $V_B=1.91\ \mu\text{m/h}$; the blue line shows the efficiency of the detector after etching time equals 45 min under etching condition (8 ml of 10 N NaOH+1 ml CH_3OH at 60°C) with $V_B=2.73\ \mu\text{m/h}$, our new etching condition; the violet line shows the efficiency of the detector after etching time equals 45 min under etching condition (8 ml of 10 N NaOH+2 ml CH_3OH at 60°C), and the red line shows the efficiency of the detector after etching with conventional aqueous solution of NaOH (6.25N NaOH at 70°C for etching time equals 5 h). One can be conclude that our new etching condition has high etching efficiency for the CR-39 detector at low alpha energy compared to the other conditions. Thus, we claim that the new etching condition which introduced in this work becomes better than that used in previous works. Typical optical images of alpha particles track on CR-39 detectors with alpha energies between 1 and 5 MeV are shown in Fig. 8, since the CR-39 samples are chemically etched under the best etching conditions, i.e. 8 ml NaOH of 10N with 1 ml pure CH_3OH at 60°C for 45 min.

4. Conclusion

Characteristics studies for fast detection of alpha particles in CR-39 detectors were investigated by using a new etching condition. The best chemical etching condition for fast detection of alpha particles is 8 ml of 10N NaOH+1 ml CH_3OH . The major advantage of the present method in this work is that CR-39 detectors are successfully used in the detection of alpha particle in short duration time (45 min) with $V_B=2.73\ \mu\text{m/h}$ with considerable efficiency. In addition, the effect of etching conditions upon the sensitivity, critical angle and etching efficiency has been

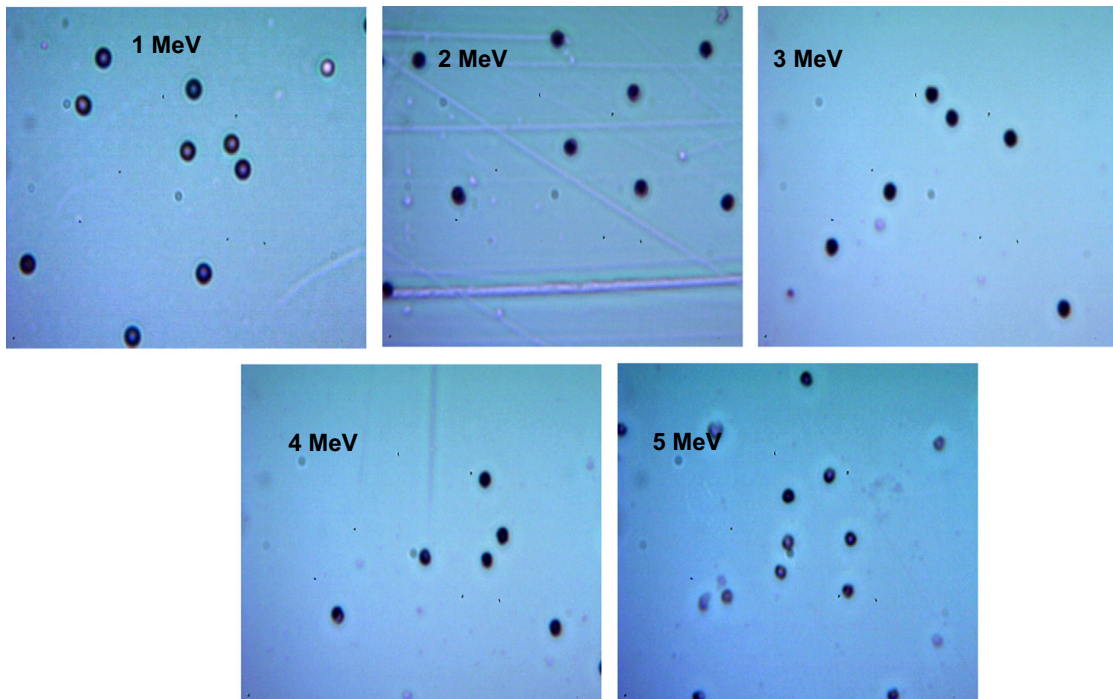


Fig. 8. Typical optical images of alpha particles track on the CR-39 detector with alpha energies between 1 and 5 MeV.

studied. The activation energies of this etchant have been calculated; moreover, track activation energy, E_T , has been found to be lower than the bulk activation energy, E_B , for CR-39 detectors.

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