The Effect of Two Etching Solutions NaOH and KOH on Registration Properties and Etching of Polymer CR-39 Detectors

Submitted: 2023-04-03

Revised: 2023-08-11

Online: 2024-03-05

Accepted: 2023-08-21

Aqmur Sadoon Munshed^{1,a*}, Ahmed Abed Ibrahim^{2,b}

¹Department of Physics, College of Sciences, Kirkuk University, Kirkuk, Iraq ²Department of Physics, College of Sciences, Kirkuk University, Kirkuk, Iraq ^{a*}scphm012@uokirkuk.edu.iq, ^bDean.Ahmed2017@uokirkuk.edu.iq

Keywords: Solid-state Nuclear detectors, Track detectors, CR-39 detector, Etching solutions, Chemical Etching, Track etching parameters, Alpha particles.

Abstract. The solid-state nuclear track detector (SSNTD) CR-39 can be used to study the different factors affecting alpha particles. In this work, two different etching solutions (NaOH and KOH) on registration properties investigated the impact of diameter D, track Diameter growth velocity VD, number of tracks, and removal layer thickness of the CR-39 nuclear track detector.

The two types of NaOH and KOH were dissolved in water with varied normality (3, 5, 7, 9, 11)N at a temperature of 70 °C to create the etching solution. During the experiment, a source of particles with a 4 MeV energy, Alpha, was used to irradiate the pieces of detectors for 30 seconds. It was found that the values of the diameter, number of tracks, removal layer thickness, and track Diameter growth velocity V_D by using the etching solution (NaOH +water) were greater than the values in the KOH + water solution. The results showed excellent agreement with the outcomes of others who use different normalities.

Introduction

CR-39 detectors, made from polymers of C₁₂H₁₈O₇, are widely used to measure alpha, proton, and neutron radiation due to their sensitivity to high linear energy transfer (LET) radiation. When a CR-39 detector is irradiated by high LET radiation, defects are formed on its surface due to the breakdown of its molecular structure [1]. The defects are only about 10 nm in size and are therefore known as "latent prints". A method based on a chemical reaction ("chemical etching") is used to uncover alpha radiation signatures by increasing their dimension. Once the etching process is complete, the track dimensions are measured optically. Track formation is sensitive to the conditions of the etching process, such as temperature, etching time, and the concentration of the solution being used for the process. Therefore, it is necessary to determine the optimal concentration of the solution and the heating temperature. According to research, an etching process procedure that is based on the use of a solution of (KOH+water) and (NaOH+water) is recommended at a temperature of 70 °C and a concentration of (3, 5, 7, 9, 11) N.This study will investigate the effect of type etchant solutions on the proportion of CR-39 detector logs, which include track diameter, number of tracks, track diameter growth rate, VD, and removal layer thickness.

There are many factors that affect the response of nuclear track detectors, including ultrasonic waves, electric fields, light ions, temperature, high pressure, a high dose of electromagnetic rays, radiation time, the incidence angle of the radiation, the temperature of the etchant solution, the concentration of the etching solution [2], the type of detector, the energy of incident particles, and the exposure time of the detector to the radiation [3]. The sensitivity of nuclear detectors is influenced by two additional factors relating to the detector itself: the purity of the monomer and the molecular structure of the polymer [4].

It is generally known that different polyamide-etched-track nuclear detectors have different recording properties [5, 6]. The development of molecular arrays with sufficient sensitivity to find alpha and other charged particles in any application should be the goal of polymer latent detection research. CR-39 has been used as an etched nuclear sensor for more than 30 years [6]. The

polyalkyl chains in the CR-39 structure are connected by diethylene glycol-dicarbonate linkages [7]. The monomer has three bonded groups: (-CH2-O-CH2-), the carbonyl bonded group (C=O), and (-CH2-O-CH2-). On the other hand, these functional groups are in charge of the modifications in physicochemical properties that happen following exposure to each type of radiation [8]. The amount of damage depends on the specific ionization energy lost during charged particle deceleration [9]. It is crucial to have a thorough understanding of the material excitation characteristics and the track structure along a fast heavy ion's passage into a solid [10].

factors that affect the response of nuclear track detectors, including ultrasonic waves, electric fields, light ions, temperature, high pressure, Additionally, it is helpful for many other applications, including nanostructuring, charged particle detection, the manufacturing of tracking membranes, and cancer therapy [9,11]. The Bragg curve, with maximal projectile energy at around 3 to 4 MeV/u, is widely recognized as the peak-like credibility (the Bragg peak) of ion energy and the stopping power of ions in a solid, such as the CR-39 track detector [12]. To comprehend the etching process in nuclear tracks, it is vital to comprehend the link between the track etch rate and the bulk etch rate with reset energy [13].

Materials and Methods

The nuclear track detector in solid-state PADC CR-39 of thickness 400 µm from Track Analysis Systems Ltd. (TASL), England, is used to measure the diameters of alpha particles and calculate the track diameter growth rate V_D. The radioactive source ²⁴¹ Am was used to irradiate the detector; it is an alpha particle emitter with a maximum energy of 5.485 MeV and an average range in air of 4.16 cm. The required energy is obtained by adjusting the distance between the airborne radioactive source and the detector through an irradiation system. The detector was divided into two groups: the first group consisted of five samples exposed to alpha particles with an energy of 4 MeV, and the irradiation process was carried out using a 1 mm cross-sectional shooter to ensure a perpendicular fall on the detector. Samples were etched with 95% purity (KOH+water) and (NaOH+water) solutions at a temperature of 70 °C and with different concentrations (3, 5, 7, 9, 11, N) to etch the damaged areas to show the diameters of the tracks formed as a result of irradiation of the alpha particle detector, while the second group of five samples was also cut into pieces with dimensions of 1x1 cm² to do a preliminary analysis of the impact of varying the etching solution's concentration. The samples were etched under the same circumstances as described previously in terms of the etching solution type, temperature, and concentrations (3, 5, 7, 9, and 11) N at the bulk etch rate V_B without exposing the detector to alpha particles. Using Image Driving Software, which works with a specific program to transmit images of tracks, the lengths, and the diameters of the tracks directly to a PC where measurements are made, a digital camera (MDCE-5C) is connected to a light microscope (NOVEL) connected to the computer to take pictures of the etching tracks and measure the thickness of the removed layer.

Result & Discussion

The etchant solution type effect on the track diameter and etching time:

A CR-39 detector exposed to alpha particle radiation from a 241 Am source was etched in aqueous solutions of NaOH and KOH with different concentrations, and a temperature of 70 °C was used to achieve optimal processing conditions. After measuring the track diameter, the track diameter growth rate (V_D) was determined.

Compared to KOH aqueous solution, the diameter of the tracks rises for NaOH at shorter etching durations because the ionic size of NaOH is smaller than KOH, so the etchant solution's rising concentration causes an increase in the diameter of the alpha particle tracks that fall on the nuclear track detector CR-39.

Figure 1 shows the average alpha particle track diameter in relation to etching time for (3, 5, 7, 9, and 11)N in NaOH and KOH aqueous solutions at a temperature of 70 °C.

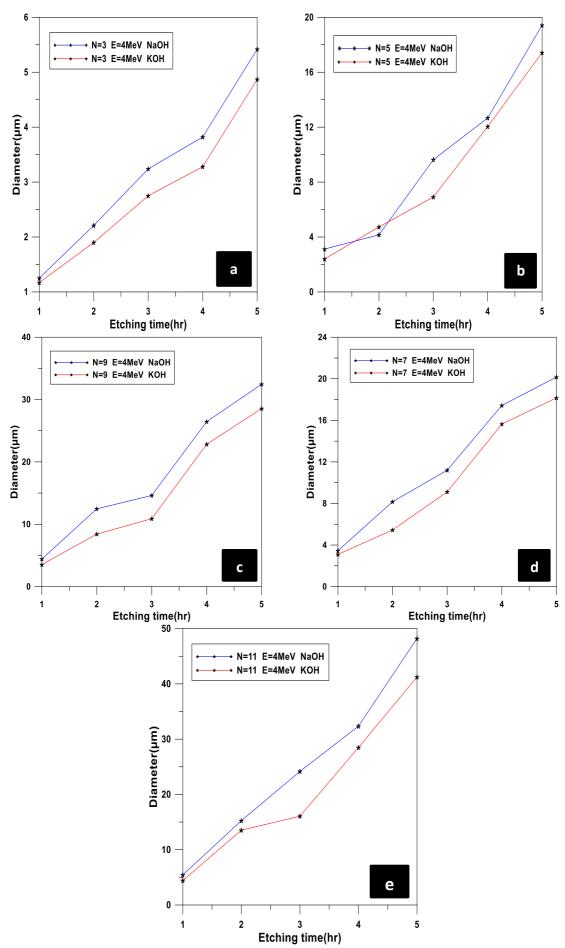


Fig. 1. (a-e) Average alpha-particle track diameter in CR-39 detectors etched in NaOH and KOH solutions at 70°C with varied concentrations(3,5,7,9,11) N as a function of etching time.

The change in the diameters in the nuclear track detector CR-39 of the traces left behind by the alpha particle's fall with the time of etching and the concentrations (3, 5, 7, 9, 11, N) Note from Figure 2 that the diameters of the tracks increase linearly with the increase in the concentrations of the etchant solution with the constant temperature of the solution, and the underlying cause of increasing the solution's concentration is increasing the number of reactant ions (OH⁻) in the detector, which in turn leads to an increase in the number of dissolved polymer molecules and then an increase in the amount of material removed from the surface of the detector per unit time.

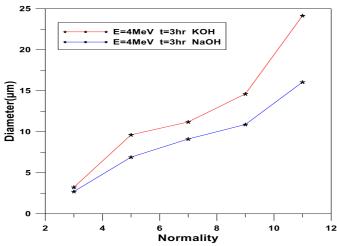


Fig. 2. Change in tracks diameter with the concentration of NaOH and KOH solutions.

The energy and the type of etchant solution effect on the number of tracks:

Table 1. The relationship between alpha particle energy and the number of tracks of NaOH and KOH solutions at 3hr etching time.

	a	queous NaC solution	ЭH					aqueous KOH solution			
E_{α} (MeV)	No. of track at N=3	No. of track at N=5	No. of track at N=7	No. of track at N=9	No. of track at N=11	No. of track at N=3	No. of track at N=5	No. of track at N=7	No. of track at N=9	No. of track at N=11	
2	133	127	126	121	112	125	135	142	138	129	
3	162	139	153	133	160	188	168	164	177	155	
4	166	171	163	146	170	192	200	198	194	170	
5	215	187	220	180	190	215	187	220	204	195	

From Table 1, It was found that the number of tracks begins to increase as the energy of the alpha particles increases, meaning that the relationship between the number of tracks and the energy is a direct relationship, and then the number of tracks begins to decrease as the energy increases; that is, the relationship between the number of tracks and the energy in the range 2-5 MeV is an inverse relationship, knowing that the etching time for each energy to obtain the largest number of tracks ranges between 1 and 5 hr for KOH and NaOH aqueous solutions.

_	aqı	ieous NaO solution	Н					aqueous KOH solution		
Etching time (hr)	No. of track at N=3	No. of track at N=5	No. of track at N=7	No. of track at N=9	No. of track at N=11	No. of track at N=3	No. of track at N=5	No. of track at N=7	No. of track at N=9	No. of track at N=11
1	150	159	169	189	163	176	196	174	181	160
2	167	176	176	167	165	183	200	192	184	163
3	166	172	169	146	170	192	205	198	194	170
4	163	162	163	140	165	180	188	194	192	167
5	156	159	154	136	140	170	178	187	185	162

Table 2. The relationship between the etching time and the number of tracks of NaOH and KOH solutions of 4MeV energy.

Note from Table 2 that the change in the number of traces begins to increase until it reaches its maximum value and then begins to gradually decrease with the increase in the time of etching as a result of the end of the trace and the merging of the traces with each other that occurs between the tracks with the progress of the etching process for KOH and NaOH aqueous solutions.

The etchant solution effect on the thickness of the removed layer:

The CR-39 detector was etched with aqueous NaOH and KOH solutions with concentrations of 3, 5, 7, 9, and 11 N at a temperature of 70 °C, and the thickness of the removed layer was measured sequentially. As shown in Figure 3, the relationship between them appears to be in a linear direction, so the etching time measured for each solution concentration and the general surface of the detector increase as the concentration of etching increases. The removed layer thickness of the solution increases with constant temperature.

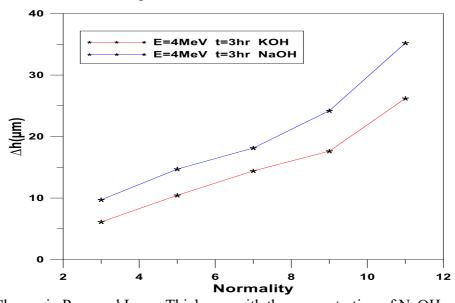


Fig. 3. Change in Removal Layer Thickness with the concentration of NaOH and KOH.

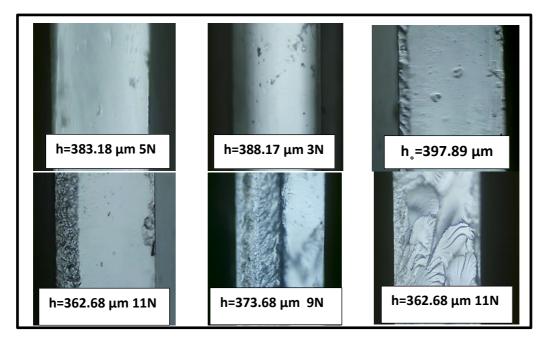


Fig. 4. Shows the thickness of the detector before etching h_{\circ} and after etching for three hours at (4MeV) energy, concentration (3,5,7,9,11) N and temperature (70±1 °C) for KOH solutions.

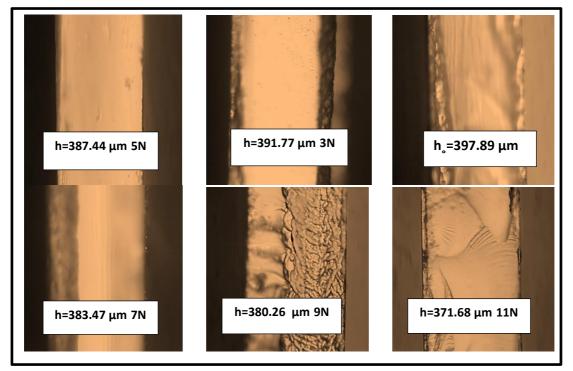


Fig. 5. Shows the thickness of the detector before etching h_{\circ} and after etching for three hours at (4MeV) energy, concentration (3,5,7,9,11) N and temperature (70±1 °C) for NaOH solutions.

Note from Figure 4 and Figure 5 that the thickness of the detector is a function of the concentration of the etching solution in the NaOH and KOH aqueous solutions.

The etchant solution effect on the track diameter growth rate V_D:

Figure 6 shows that the concentration effect of the track diameter growth rate V_D at a constant temperature of the etchant solution is increasing exponentially with increasing concentration. Also note that the increasing pattern of V_D takes the same shape at different concentrations (3, 5, 7, 9, and 11) of N in NaOH and KOH aqueous solutions, showing that the track diameter growth rate of VD increases exponentially. As the etchant concentration of NaOH aqueous solution and KOH aqueous solution increases.

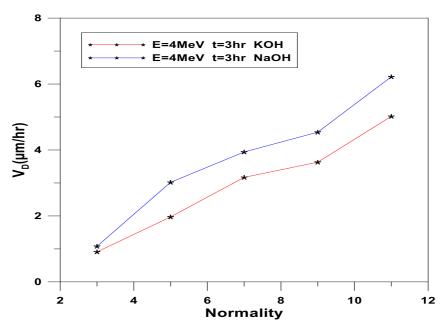


Fig. 6. Change in track diameter growth rate V_D with the concentration of NaOH and KOH.

Table 3. Registration parameters of CR-39 in NaOH and KOH solution at different concentrations and 70 °C.

		aqueous NaO solution	Н			aqueous KOH solution		
Concentration	D (µm)	V _D (μm.hr ⁻¹)	No. of track	∆h (μm)	D (µm)	V _D (μm.hr ⁻¹)	No. of track	∆h (μm)
3	3.24	1.08	166	9.72	2.75	0.91	192	6.12
5	9.63	3.02	171	14.71	6.91	1.97	200	10.45
7	11.2	3.94	163	18.14	9.12	3.17	198	14.42
9	14.62	4.54	146	24.21	10.89	3.63	194	17.63
11	24.16	6.22	170	35.21	16.06	5.02	170	26.21

Table 3 shows the h, D, V_D , and number of tracks values for the CR-39 detector. Etched at 70 °C in NaOH and KOH aqueous solutions at the (3, 5, 7, 9, and 11) N concentrations, it was noted that the diameter D and the growth of the trace diameter V_D are clear functions of the concentration of the KOH and NaOH aqueous solutions; that is, they are functions of the effect of the etching time on the detector irradiated with alpha particles.

It was observed that for each concentration used in this study, the diameter of the trace increases with the increase of the etching time until reaching a state of the end of the trace and the merging of the traces with each other; that is, It can be said that there is a direct relationship between the diameter of the effect and the concentration [14]. Also, NaOH aqueous solutions are more efficient than aqueous KOH solutions [15].

Conclusions

In this study, experimental verification was conducted of the following findings on the impact of various etchant solution types on nuclear track detectors (CR-39): The diameters of the tracks rise exponentially with the increase in concentration of the solution at the constant temperature of the solution, according to the influence of the etching solution type on the track diameter and on the track diameter growth rate V_D and etching time. This occurs as a result of an increase in the amount of (OH⁻) ions interacting with the detector material caused by a rise in solution concentration, which in turn causes an increase in the number of dissolved polymer particles. After then, there will be an increase in the volume of material taken from the detector's surface per unit of time. As a result of

this, there will be an increase in the diameters of the etched traces. Since the speed of the interaction of the etching solution with the detector material increases with etching solution concentration, the diameter of the etched traces will grow as a result. The rate of growth of the trace diameter was calculated from the graphical relationship between the diameter of the trace and the time of etching, which was also changing linearly with the time of etching, after determining the diameter of the alpha particle traces for the group exposed to alpha radiation under specific abrasive conditions. The number of tracks starts to rise as a result of the energy and type of etchant solution. The number of tracks is affected by the energy and the kind of etchant solution. The number of tracks starts to rise as the energy of the alpha particles rises, indicating a direct relationship between the two quantities. The number of tracks then starts to merge as the energy rises and then starts to gradually merge with the lengthening of the etching process due to the end of the trace and the merging of the traces with one another that happens between the tracks as the etching process advances. Effect of the etchant solution on the thickness of the layer that was removed with a constant etching time: increasing the concentration of the etching solution causes the interaction energy between the detector and the solution to increase, which works to degrade more detector material. This results in an increase in the number of dissolved particles on the surface of the detector. Additionally, increasing the etching time causes the detector to dissolve more completely, resulting in a reduction in detector thickness and an increase in removed thickness. For the CR-39 detector, NaOH and KOH solutions are both more effective than aqueous KOH because The ionic size of Na+ is smaller than that of K+. Hence, the attractive force between Na+ and OH is relatively strong, and so the ionization of NaOH is lower than that of KOH, which makes NaOH a weaker base than KOH. Upon dissolution, it turns into ions, and the ions vary in size (such as radii) from one element to another, which contributes to the so-called steric impedance of compounds. Therefore, the volume of Na⁺ in NaOH is sterically smaller than the volume of K⁺ in KOH, and often potassium is larger than sodium, so the alpha particles are affected by sodium greater than potassium. The increase in track diameter is an inherent function of etching time. That is, it is a function of the effect of etching time on the alpha particle irradiation detector. For any given energy applied in this study, It was found that the track diameter increased with increasing etching time until the end of the track was reached and the tracks fused together. In other words, it can be said that the relationship between track diameter and etching time is a direct one. The detector's maximum track occurs when maximum energy absorption occurs. This value is independent of the type of falling particles as well as the type of detector used. The registration properties and etching of polymer CR-39 detectors are affected by increasing the concentration of the etching solution for both NaOH and KOH aqueous solutions. The results are better with NaOH than with KOH aqueous solution and are also affected by the etching time and the energy used in the irradiation of the detector with alpha particles.

References

- [1] A.P Fews, D. L. Henshaw, High resolution alpha particle spectroscopy using CR-39 plastic track detector, *Nuclear Instruments and Methods in Physics Research*. 197 (1982) 517–529.
- [2] D. Hermsdorf, M. Hunger, S. Starke, F. Weickert, Measurement of bulk etch rates for polyallyl-diglycol carbonate (PADC) and cellulose nitrate in a broad range of concentration and temperature of NaOH etching solution, *Radiation Measurements*. 42 (2007) 1–7.
- [3] D.S. Manar, A.R. Ali, F.K. Nada, A. El-Taher, *Journal of Radiation and Nuclear Applications*. 5 (2020) 119–125.
- [4] M. el Ghazaly, On alpha particle spectroscopy based on the over-etched track length in PADC (CR-39 detector), *Radiation Effects and Defects in Solids*. 167 (2012) 421–427.
- [5] A. F. Saad, M. Fromm, M. H. Ibraheim, A. A. El-Namrouty, A. M. Nwara, S. A. Kandil, M. S. Dawood, M. S. (2021). Loss of chemical bonds induced by high doses of γ-radiation in a PADC polymer film: The influence of dose and dose rate on radiation chemical yields. *Radiation Physics and Chemistry*. 187 (2021) 109579.
- [6] T. Yamauchi, S. Kaifu, Y. Mori, M. Kanasaki, K. Oda, S. Kodaira, T. Konishi, N. Yasuda, R. Barillon, Applicability of the polyimide films as an SSNTD material. *Radiation Measurements*. 50 (2013) 16–21.

- [7] K.C.C. Tse, F.M.F. Ng, K.N. Yu, Photo-degradation of PADC by UV radiation at various wavelengths, *Polymer Degradation and Stability*. *91* (2006) 2380–2388.
- [8] M. El Ghazaly, H. E. Hassan, Spectroscopic studies on alpha particle-irradiated PADC (CR-39 detector), Results in Physics. 4 (2014) 40-43.
- [9] A.V.E. Bagulya, L.L. Kashkarov, N.S. Konovalova, N.M. Okat'eva, N.Y.G.E. Polukhina, Starkov, N.I, Search for superheavy elements in galactic cosmic rays, JETP letters. 97(2013) 708-719.
- [10] F.F. Komarov, Nano-and microstructuring of solids by swift heavy ions, Physics-Uspekhi. 60.5 (2017) 435.
- [11] V.Alexeev, A.Bagulya, M.Chernyavsky, A. Gippius, L. Goncharova, S.Gorbunov, A.Volkov, Charge spectrum of heavy and superheavy components of galactic cosmic rays, Results of the olimpiya experiment, The Astrophysical Journal. 829.2 (2016) 120.
- [12] R. A. Rymzhanov, S. A. Gorbunov, N. Medvedev, A. E. Volkov, Damage along swift heavy ion trajectory, Nuclear Instruments and Methods in Physics Research section B. Beam Interactions with Materials and Atoms. 440 (2019)25-35.
- [13] B. Dörschel, D. Hermsdorf, K. Kadner, Studies of experimentally determined etch-rate ratios in CR-39 for ions of different kinds and energies, Radiation measurements. 35.3 (2002) 183-187.
- [14] D. Nikezic, K. N. Yu, Formation and growth of tracks in nuclear track materials, Materials Science and Engineering: R: Reports. 46.3-5 (2004)51-123.
- [15] S.A. Amin, D.L. Henshaw, Effect of various etching solutions on the response of CR-39 plastic track detector. Nuclear Instruments and Methods in Physics Research. 190.2 (1981) 415-421.