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Alternative Approach to Extract the Bulk Etching Rate of PADC Nuclear Detector

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Abstract

The paper aims to propose the maximum track length (L_{max}) measurement as an alternative approach to evaluate and extract the bulk etch rate (V_b) of the nuclear detector PADC CR-39, and compare it with the results obtained by the removal layer thickness measurement of the etched detector. The alternative L_{max}-method mainly relies on the measuring the length of the etched tracks, their maximum values and saturation times from the obtained track profile images. The detectors were irradiated with different energies of alpha particles emitted from the ^{241}Am source and then etched in a 6.5 N NaOH solution at 70±1°C for different successive time intervals. In order to calculate V_b, the maximum length of the tracks and their saturation times were determined corresponding to the energies of the alpha particles used. A direct proportion between the maximum length of the tracks and the energy of the alpha particle was observed. However, the value of V_b is found to be 1.344±0.0202 µm h^{-1} obtained using the L_{max}-method, and it was consistent with that computed by the method based on the measurement of the thickness difference of the etched detector which was 1.354±0.065 µm h^{-1}.

Keywords: PADC detector, SSNTDs, Bulk Etch rate, CR-39, Track Length.

طريقة بديلة لإيجاد معدل القشط العام للكاشف النووي PADC

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الخلاصة

يهدف البحث إلى اقتراح قياس القيمة القصوى لطول الأثر (L_{max}) كطريقة بديلة لتقييم وإيجاد معدل القشط العام (V_b) لكاشف الأثر النووي PADC CR-39، ومقارنتها مع النتائج المستحصلة من قياس سمك الطبقة المزالة للكاشف المقشوط. تعتمد الطريقة البديلة-L_{max} أساساً على القياس المباشر لأطوال الآثار المقشوطة، وقيمها القصوى، وزمن تشبعها من الصور الجانبية لمظاهر أو أشكال تلك الآثار. شُعِّع الكاشف بطاقات مختلفة بجسيمات ألفا المنبعثة من مصدر الأمريشيوم ^{241}Am، ثم قُشِطت لفتراتٍ زمنية متعاقبة بالمحلول الكيميائي NaOH بتركيز 6.5 N عند درجة حرارة 70±1°C . لإيجاد V_b، فقد تم تحديد القيم القصوى لأطوال الآثار وأزمان تشبعها تبعاً لطاقات جسيمات ألفا المستخدمة. لوحظ وجود تناسب طردي بين القيم القصوى لأطوال الآثار وطاقة جسيمات ألفا، إذ وُجِد أن قيمة V_b كانت تساوي 1.344±0.0202 µm h^{-1} باستخدام طريقة L_{max}، وإنها تتفق بشكل جيدٍ مع القيمة التي تم حسابها باستخدام الطريقة المألوفة التي تعتمد على قياس فرق السمك للكاشف المقشوط والتي كانت 1.353±0.065 µm h^{-1}

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1. Introduction

The solid-state nuclear track detectors (SSNTDs) are considered as reliable means to detect the charged particles. The poly allyl diglychol carbonate (PADC) polymer is regarded as one of the most favorite types of SSNTDs, especially the well commercially known detector CR-39 which is widely utilized in different fields of sciences. This preferred standpoint is due to its high sensitivity to the charged particles and protons recoil of neutrons [1, 2, 3]. However, a review of SSNTD's properties can be seen elsewhere [4].

It is well-known that the track creation and development in SSNTDs is strongly controlled by two essential parameters, the track etch rate (V_t) along the track depth in the detector, and the bulk etch rate (V_b) in undamaged areas of the detector, respectively. The shape development of the etched track passes through two phases or stages [5]. The first one is the acute-conical phase through which the track develops into a sharp conical etch pit when V_t>V_b. Whilst when the etching solution passes the end of the damaged path (i.e the end of the particle range) in the detector towards the undamaged region, the track enters the second phase of the development; the over-etched phase where the track develops into a spherical shape etch pit as V_t=V_b [6].

It has been realized that the direct measurement of track lengths is difficult compared to the direct measurement of the track-opening diameters which is relatively simple and straightforward. Nevertheless, the formal description of the track evolution, the monitoring and imaging the profiles (longitudinal cross-sections) of the etched tracks, and measuring their lengths (or depths) have attracted the attention of many researchers to realize the real vision of the track growth and variation of its parameters [7-11].

Different methods have been proposed to measure the lengths of the etched tracks directly from their images. One of these methods included breaking and precisely polishing the detector side to reveal the lateral profiles or the longitudinal cross-sections of the etched tracks [12, 13]. Another method is based on using the confocal microscope through the 3D-imaging technique of the track shapes [7, 8, 14], or that based on the contact stylus profilometry through measuring the replica heights of the etched tracks [9, 15]. In our previous works [10, 16, 17], including this paper, the "lateral irradiation method" have been used to obtain the track images and depths based on irradiating the side edge of the detector. This method is straightforward and easier than the previously mentioned ones as there is no need for a complicated system.

Due to the importance of the bulk etch rate (V_b) in governing the track evolution as it is related to the etch rate ratio V(=V_t/V_b), the precise control and accurate measurement of V_b is crucial for track geometry studies. Various methods have been employed to determine V_b of the SSNTDs. One of the most widely used methods is the diameter measurement of the Fission fragments tracks method [4, 18]. Another approach is based on the measurement of the mass difference after etching the detector called the "gravimetric method" [19], or relies on the measurement of the detector thickness before and after etching and then the removed layer thickness called the "thickness difference method"[18] or the "standard method"[20]. The track length-diameter "L_e-D" method was also used to determine V_b of the CR-39 detector [17, 20, 21]. The method involves direct measurement of both the lengths and diameters of the etched tracks for successive etching times. Because of the constancy (saturation) of the track length in the over-etched phase of the track evolution, the measurements, in this case, should include only the acute-conical phase [17], i.e the sharp conical tracks, to obtain an accurate value for V_b.

Further methods have been employed to determine V_b for different track detectors. For example, the “peel-off method", “Masking method” in LR-115 and CR-39 detectors which they rely on the study of the surface profilometry of the detector and using atomic force microscope (AFM) [22, 23, 24]. The "non-destructive method" have also used to determine V_b through measuring the active-layer thickness of LR 115 using a color commercial document scanner [25], employing Fourier Transform Infrared (FTIR) spectroscopy [26], or measuring the removed layer using energy dispersive X-Ray fluorescence (EDXRF) [27].

In the present investigation, an alternative method is suggested to determine V_b named the "L_{max}-method". The method involves a direct measurement of the track lengths from the images of the etched tracks in both phases of evolution; the acute-conical and the over-etched phases. Accordingly, the maximum value of the track length (L_{max}) at the saturation point and also the corresponding saturation time (t_{sat}), which is the time required for track length to reach the maximum and constant value, to be determined in accordance with the energy of the alpha particle which in turn can be used to calculate V_b according to the Eq. (1) [10]:

$$L_{max} = R - V_b t_{sat} \quad (1)$$

where R is the range of the incident alpha particle in the detector material

2. Experimental approach

The solid-state nuclear track detector PADC CR-39 of thickness 200 μm from Page Mouldings (Worcestershire, England) was cut into the size of dimension 1.5x2 cm^2. The "lateral-irradiation" was used to irradiate the sides (the edges) of the detectors by alpha particles with energies of 5.2, 4.43, 3.61and 2.56 MeV under normal incidence. A 1 μCi ^{241}Am source of alpha particle emitter of main energy 5.485 MeV was used in the detector irradiation. The energies less than the main energy are obtained by changing the source to detector distance in air under atmospheric pressure. To reveal the track profiles, the irradiated detectors were chemically etched in a 6.5 N of NaOH solution at 70±1°C for different periods of time. The etched detector is positioned under the optical microscope so that the longitudinal cross-sections of the etched tracks can be visualized from the surface of the detector as shown in Figure-1.

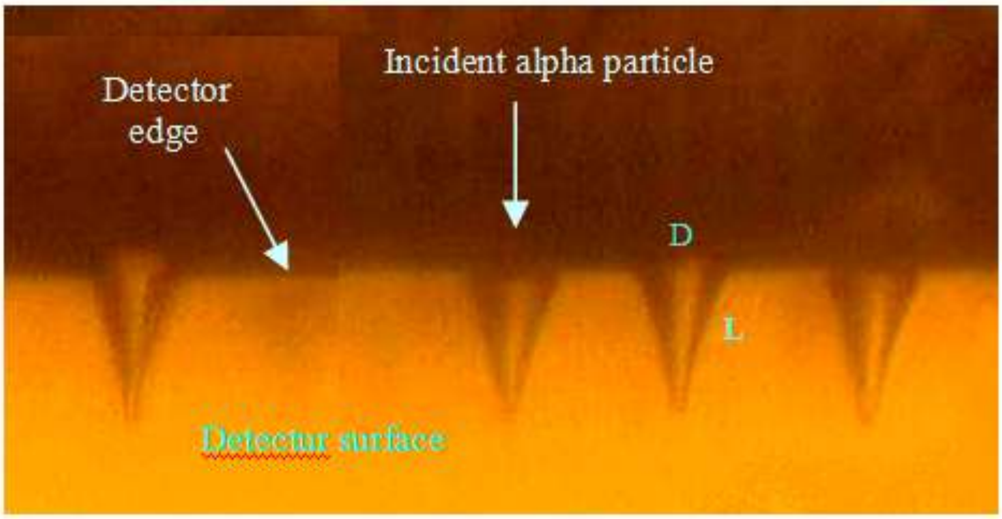

Figure 1- Lateral images of a 3.61 MeV alpha particle tracks in the CR-39 detector after etching for 4.25 h using a 6.5 N of NaOH solution at 70±1°C. The images of the etched tracks refer to that when alpha particles enter the edge (the side) of the detector under normal incidence and observed from the surface of the detector using a high-resolution digital camera attached to an optical microscope

To determine the bulk etch rate (V_b), the track lengths were directly measured from the images of the etched tracks, hence the maximum track length (L_{max}) and the saturation time (t_{sat}) can be determined according to the alpha particle energies. By using the L_{max}-method which is based on the Eq. (1), the V_b was calculated.

In order to confirm the accuracy of the results obtained using the L_{max}-method, the thickness difference method, which is one of the frequently used methods based on measurement of the removed layer thickness from the surface of the detector, was used to calculate V_b again. Unexposed CR-39 detector to alpha particles was cut into the size of dimension 1.5x1.5 cm^2 and etched under the same etching conditions, as before, for 1-5 h with successive intervals of 0.5 h. The thickness of the detector was measured before and after the etching for each interval from which the V_b was calculated based on the Eq. (2) [18, 20].

$$V_b = \Delta h / 2\Delta t \quad (2)$$

where Δh is the thickness difference of the detector after a Δt etching time.

However, imaging the track profiles and the detector edge, diameter measurements of the etched tracks together with the thickness of the detector have been done using the optical "XSZ-H Series

Biological Microscope", which attached to a PC through a high-resolution digital camera of type "MDCE-5A".

3. Results and Discussion

3.1. Bulk Etch Rate by "L_{max}-Method"

Figure-2 shows the relationship between the length of the etched tracks and the etching time for alpha energies 5.2, 4.43, 3.61 and 2.56 MeV of alpha particles. It can be seen from the Fig. that the track shape goes through two phases of growth and development [5]. The first one is the acute-cone phase where $V_t > V_b$, where the track length increases non-linearly with the etching time and reaches a saturation point at which it maximizes (L_{max}) and gets a constant value. The saturation point is reached when the etching solution reaches the end of the damaged path in the detector, i.e. the end of the alpha particle range, where V_t extremely reduces and approaches V_b. In this case, the damaged region created by the alpha particle is entirely etched and the formed track, which is called the "etched-out" track, see Figure-3, has a conical shape with a sharp tip at the end of the particle range in the detector [28].

When the etching solution crosses the end of the particle range (the saturation point) towards the undamaged region down the track tip, the second phase (the over-etched phase) of track development starts to evolve. In this phase, as seen in the Figure-2, the saturation process continues along with the length of the track at a constant and maximum value (L_{max}). Thus, the track length grow rate remains equal to zero (i.e dL/dt=0) with the progress of etching process. Therefore, the track as seen in Figure-3 starts to turn gradually into a rounded-tip track [28], and the track etch rate approaches the bulk etch rate ($V_t \approx V_b$) where shortly after the etching it continues at a constant rate V_b in all directions (scalar) and the track begins to change gradually to the spherical shape (Figure- 3).

Figure- 3 represents, for example, the Lateral images of a normally incident 2.56 MeV alpha particle tracks in the present study. The track images were observed from the surface of the detector under using an optical microscope after etching with 6.5 N NaOH solution at 70±1^{o}C for different time intervals from 0.5-7 h. The images show the track evolution phases, the variation of the track shapes and their lengths, the opening diameter of the track as well as the maximum length and the saturation time.

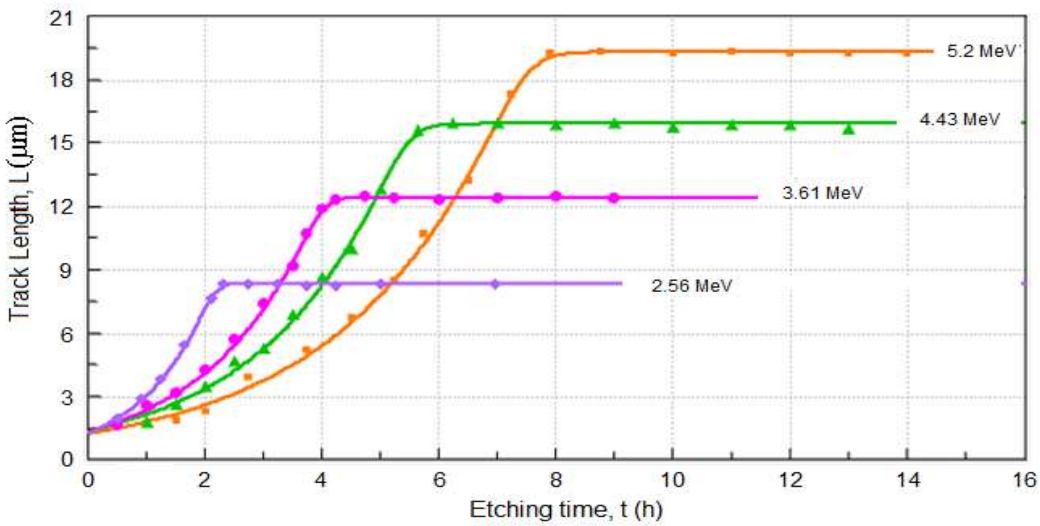

Figure 2- Relationship between the track length and the etching time for alpha particle energies 2.56-5.2 MeV in the solid state nuclear track detector CR-39, obtained from the direct measurements of the track lengths of the alpha particle track images using an optical microscope.

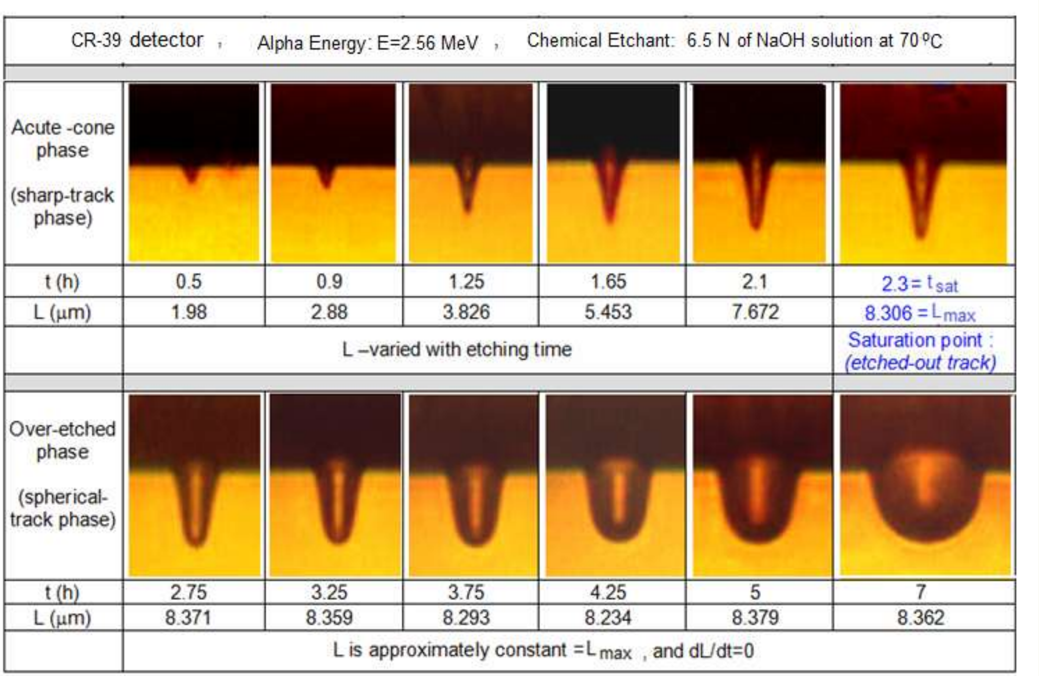

Figure 3- Lateral images and evolution phases of normally incident 2.56 MeV alpha particle tracks after etching for 0.5-7 h observed in the CR-39 detector using the optical XSZ-H Series Biological Microscope attached to a PC through a high-resolution digital camera of type MDCE-5A. The track shape goes through two phases of evolution: the acute-cone phase where $V_t > V_b$, and the over-etched phase where $V_t \approx V_b$

The maximum track length (L_{max}) and the saturation time (t_{sat}) mainly depend on the energy of the incident alpha particles. Accordingly, the L_{max} and t_{sat} values for tracks corresponding to the higher energies of alpha particles are greater than that of the lower energies as shown in Table-1. It can be noted from Figure- 4 that the L_{max} is Linearly proportional to the energy of the alpha particles while the t_{sat} changes exponentially with it as shown in the Figure- 5. It should be mentioned that the L_{max} depends on the energy of the incident particle while the t_{sat} depends not only on the energy of the incident particle but also on the etching rates, particularly the bulk etch rate V_b which in turn controlled by the etching conditions; the concentration [4], and the temperature of the etching solution [4, 17].

The increase in the concentration or in the temperature of the etching solution does not affect the values of the L_{max}, but it implies how fast the track reaches the maximum length and the saturation point (less t_{sat}) and vice versa. Hence, Figure- 4 could be employed as a L_{max}-E calibration curve by which one may directly identify the L_{max} of the alpha particle tracks at a particular energy in the CR-39 detector under the used etching circumstances.

Table 1- Difference between range and maximum length (R-L_{max}), and the saturation time (t_{sat}) of the alpha particle tracks in the CR-39 detector for different energies using 6.25 N NaOH at 70 ^oC as the etchant

E (MeV)	L_{max} (μm)	t_{sat} (h)	R (μm)	R-L_{max} (μm)
5.2	19.29	8.35	30.54	11.25
4.43	15.88	6.15	24.23	8.35
3.61	12.40	4.5	18.28	5.88
2.56	8.33	2.5	11.67	3.34

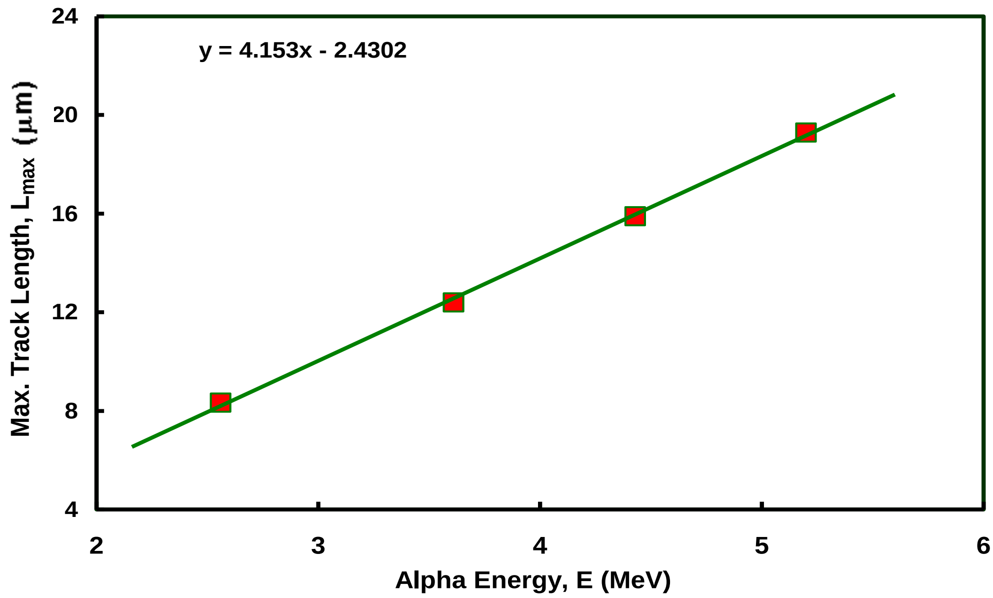

Figure 4-Variation of the maximum length of the alpha particle tracks in the CR-39 detector with alpha particle energies obtained from the direct measurement of the track image lengths using an optical microscope.

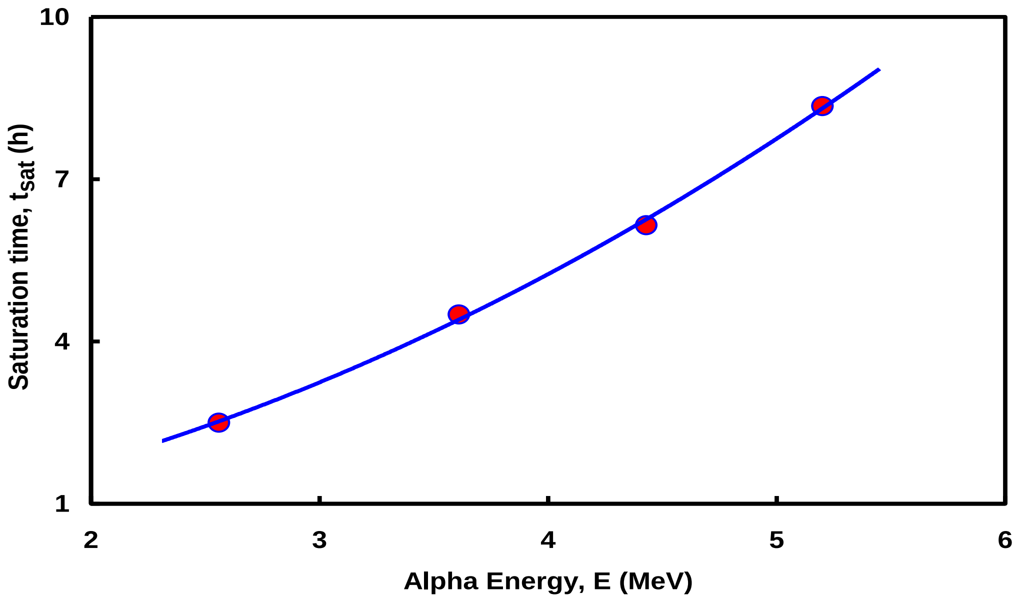

Figure 5-Variation of the saturation time of the alpha particle track lengths in the CR-39 detector with alpha particle energies, as they extracted from the curves shown in Figure-3.

In order to determine the bulk etch rate V_b using the suggested L_{max}-method, the results presented in Table-1 were used. The R-L_{max} differences were plotted against t_{sat} for the corresponding energies of the alpha particle as shown in Figure-6. Based on Eq. 1, the V_b was determined from the slope of the straight line obtained in Figure-6. The figure also shows that the V_b is equal to 1.344±0.0202 μm h^{-1} for the CR-39 detector under the etching conditions 6.5 N of NaOH solution at 70°C. It should be mentioned here that the range of the alpha particle (R) in the CR-39 detector for different energies was calculated using the SRIM -2008, Stopping Power and Range of Ions in Matter [29].

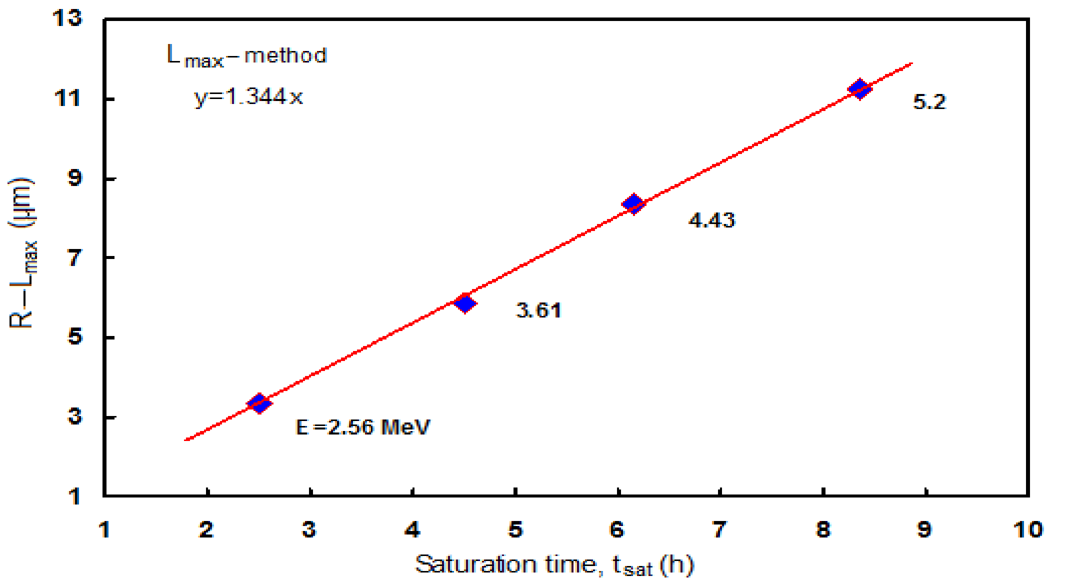

Figure 6-Relationship between the R-L_{max} differences and the saturation times of the alpha particle track lengths in the CR-39 detector for alpha particle energies 2.56-5.2 MeV, where the slope of the straight line equals to the bulk etch rate (V_b) of the detector using the L_{max}-method.

3.2. Bulk Etch Rate by "Thickness Difference Method"

To confirm the result obtained by the L_{max}-method, the thickness difference method was used to determine the bulk etch rate V_b once more. According to the relation in Eq. (2), the V_b was figured from the linear relationship between the removed layer thickness and the etching time in Figure- 7, which found to be 1.353±0.065 μm h^{-1} for CR-39 under the etching conditions 6.5 N of NaOH at 70°C. The obtained value is consistent with the result obtained using the L_{max}-method for CR-39 (V_b=1.344±0.0202 μm h^{-1}) under the same etching conditions.

Furthermore, the estimation of V_b that acquired by the L_{max}-method which was 1.344±0.0202 μm h^{-1} can be regarded as a reasonable value compared to the results obtained by Ho et al. [30] utilizing Form Talysurf PGI (Taylor Hobson, Leicester, England), and by Al-Nia'emi [17] using the track "L_e-D" (length-diameter) method to measure the bulk etch rate. The values of V_b for CR-39 detector were found to be 1.195 ± 0.028 and 1.424±0.0414 μm h^{-1} by Ho et al. and Al-Nia'emi respectively under the etching conditions of 6.25 N of NaOH solution at 70 °C.

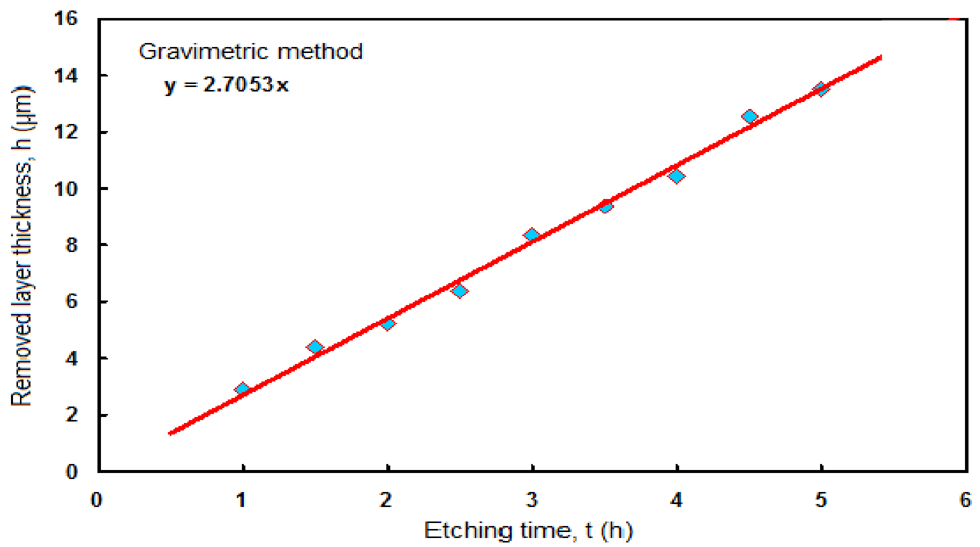

Figure 7-Relationship between the removed layer thickness and the saturation time of the alpha particle track lengths in the CR-39 detector for alpha particle energies 2.56-5.2 MeV. The slope of the

straight line divided by the factor 2 equals to the bulk etch rate (V_b) of the detector using the gravimetric method based on the mass difference measurements of the detector.

4. Conclusion

In this work, an alternative L_{max} method was used to determine the bulk etch rate (V_b) of the CR-39 detector. The results obtained by this method showed good agreement with that found using the thickness difference method. The results also indicated that the direct measurement of the lengths from the track images is rather more accurate than that of the thickness difference measurement. The advantage of the L_{max} method lies in its capability to determine the bulk etch rate implicitly from the data of the track lengths which can be achieved through fixing the maximum length of the track and its saturation time. This, in turn, helps those who are interested in the direct measurement of the track lengths instead of using different methods for measuring V_b. Thus, from the economic perspective, this procedure also saves the time and effort that might be spent on the utilization of other methods which may require additional means, detector strips, and chemical etching as well as additional measurements such as the measurement of the detector thickness or mass difference or any other parameters.

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