# Bulk Etch Rate and the Activation Energy of the CR-39 Detector using Thickness Difference Method

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## Abstract

The aim of this paper is to determine the bulk etch rate $V_b$ of the nuclear track detector CR-39 using the method based on measuring the thickness of the layer removed from the surface of the detector (the thickness difference) by the chemical etching, and then studying how to change it with the temperature of the etching solution to extract an empirical relationship between them. The detector samples were etched in a 6 N of NaOH solution at temperatures 80, 70, 60 and 50±1 $^o$C, and the thickness of the removed layer was determined by successive measuring the detector thickness for etching times of 1-9 h increasing with 1 h intervals. The results showed that the values of $V_b$ range between 2.351-0.605 μm/h for etching temperature 50-80 $^o$C respectively. An exponential relationship was obtained between the bulk etch rate and the etching temperature. It was noted that the results were consistent with that obtained by other studies using the track Length-diameter- ($L_e$-D) measurement method. The slight difference in $V_b$ magnitudes between the compared results is due to the slight difference in the concentration of the etching solution in certain cases as well as the difference of the detector origin and the method of measurement.

**Keywords**: SSNTDs, CR-39; Bulk etch rate; activation energy.

# معدل القشط العام وطاقة التنشيط لكاشف CR-39 بطريقة فرق السمك

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## الملخص

يهدف البحث إلى إيجاد معدل القشط العام لكاشف الأثر النووي CR−39 بطريقة قياس سمك الطبقة المزالة من سطح الكاشف (فرق السمك) بفعل القشط الكيميائي ودراسة كيفية تغيره مع درجة حرارة القشط لإيجاد علاقة تجريبية بينهما. قُشطت الكواشف بالمحلول الكيميائي NaOH بتركيز N 6 عند درجات حرارة قشط مختلفة $50\pm1^{o}C$ ,60 ,70 ,80 ، وتم ايجادها سمك الطبقة المزالة بقياس تتابعي لسمك الكاشف لازمان قشط h 9−1 بزيادة دورية 1 h، وقد أظهرت النتائج أن قيم $V_b$ كانت $0.605-2.351$ μm/h عند درجات حرارة قشط $50-80^{o}C$ على الترتيب. تم الحصول على معادلة أسية لتغير معدل القشط العام مع درجة حرارة القشط، ويلاحظ أن النتائج كانت متوافقة إلى حد كبير مع نتائج دراسات أخرى استخدموا طريقة قياس طول−قطر $(L_e-D)$ الأثر. إن الاختلاف البسيط في قيم $V_b$ المقارنة يعزى إلى الاختلاف القليل في تركيز المحلول القاشط في بعض الحالات فضلاً عن اختلاف منشأ الكاشف وطريقة القياس

**الكلمات الدالة :** SSNTDs؛ CR−39؛ معدل القشط العام؛ طاقة التنشيط.

## 1. Introduction

The bulk etch rate $V_b$ is one of the basic parameters for studying the track geometry in the SSSNTDs. Due to the significance of $V_b$ in controlling the development of the etched track as it is linked to the etch rate ratio $V(=V_t/V_b)$, its precise estimation is viewed as an important and decisive issue in studying the track shape and its profile development with the progress of the etching process.

The bulk etch rate is defined as the measure of the material removed from the undamaged areas of the detector surface in the horizontal direction during the etching process. The chemical reaction between the etching solution and the detector material leads to a degradation of the surface material by removing layers with specific thicknesses from the surface of the detector, which at last reduces the thickness and mass of the detector [1, 2]. The amount of the material that is removed by etching action from the detector surface depends on the type of the detector, the etching solution and its temperature and concentration, the etching time, the additives [2, 3, 4], as well as the environmental conditions that influence the detector [5, 6].

One of the critical elements that influence the accuracy of $V_b$ estimation is the swelling that outcomes from the absorption of water by the detector during the etching process, particularly in the case of etching for a long time. In this case, some steps are taken to get rid of the absorbed water and obtain the correction factor of the thickness and mass in order to minimize the measurement errors [7, 8]. The drying of the detector subsequent to etching process at a temperature not exceeding 50 $^o$C for 30 min is viewed as one of the regular methods to dispose of the absorbed water [4, 9, 10]. However, the value of $V_b$ is usually constant, under certain circumstantial and environmental conditions, over a little range of removal thickness of 7-8 μm from the detector surface (i.e. etching for short time), while its value may changes non-linearly when the detector is etched for quite a while [9].

Various, direct and indirect, methods have been employed for the determination of the bulk etch rate of SSNTD either by irradiating or non-irradiating the detector with the charged particles. One of the methods based on the mass difference of the detector before and after the etching process. This method is known as the gravimetric method [4, 11] and it is restricted to measurement accuracy of no less than the order of $10^{-5}$. Another method, which is used in this paper, relied on measuring the thickness of the detector before and after etching, and then the

thickness difference [12, 13]. The most commonly used method is measuring the track diameters of the fission fragments, particularly that from $^{252}$Cf source [2, 12]. Recently the $L_e$-D method is used to find $V_b$ for the detector CR-39 [13, 14, 15]. This method depended on the direct measurement of the track lengths and diameters from the images of the etched track. The precision of the results is limited by the measurements in the sharp conical phase of the track development [10, 13, 15].

Further methods have been used to determine $V_b$ in different SSNTDs. For example, the "masking" method in CR-39 using the atomic force microscope (AFM) [16, 17, 18], and the "peel-off" method to directly measure the bulk etch rate for the LR115 detector in the light of the investigation of the surface profilometry. A non-destructive method is also used to determine $V_b$ relied on measuring the thickness of the removed layer using energy dispersive X-Ray fluorescence (EDXRF) [19], or measuring the thickness of the active layer removed from the LR115 detector using a Fourier Transform Infrared (FTIR) spectroscopy [20].

The aim of the paper is to find the bulk etch rate ($V_b$) and the activation energy of the bulk etching of the nuclear track detector CR-39 for different etching temperatures and to find an empirical equation for the variation of the $V_b$ with the temperature of the etching solution.

## 2. Methodology

The TASTRAK plastic CR-39 detector used in the present study was purchased from Track Analysis Systems Ltd (TASL)(Pristol.UK). The original thickness of the detector sheet was 500 μm. The CR-39 detector was cut to a size of about $1.5 \times 1.5$ cm$^2$ and its thickness was measured before the etching process. The detector samples were etched separately in a 6 N aqueous solution of NaOH at different temperatures of 50, 60, 70, 80±1$^o$C for various intervals of etching time of 1- 9 h.

The thickness difference of the detector or the removed layer thickness was determined directly by successive measuring the detector thickness for etching times increasing with 1 h intervals. Based on the equation (1) [11], the $V_b$ were determined.

$$V_b = \frac{\Delta h}{2\,\Delta t} \tag{1}$$

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

It should be noted that the detector was etched from the two sides (i.e. the upper and lower surfaces), and the thickness of the detector before and after etching was measured directly from the images of the detector edges which transferred to the PC by a digital camera (MDCE-5C) installed on the optical microscope (Novel).

## 3. Results and Discussion

Fig. 1 shows the relationship between the thickness of the layer removed from the detector surface and the etching time at different etching temperatures. It is seen from the Figure that for used etching temperatures, the removed thickness increases linearly with the etching time, and this was in good agreement with that found by [15, 21]. Fig. 2 shows, for example, samples of the detector thickness images before and after etching for 1-6 h at etching temperature 80 $^{o}$C in the present study. The images appear a decreasing in the thickness of the detector and increasing the thickness of the layer removed from the detector surface with the progress of the etching process. Based on Eq. (1) the bulk etch rate $V_b$ has been calculated for CR-39 at different temperatures 50, 60, 70, 80$^{O}$C as shown in Table 1 .
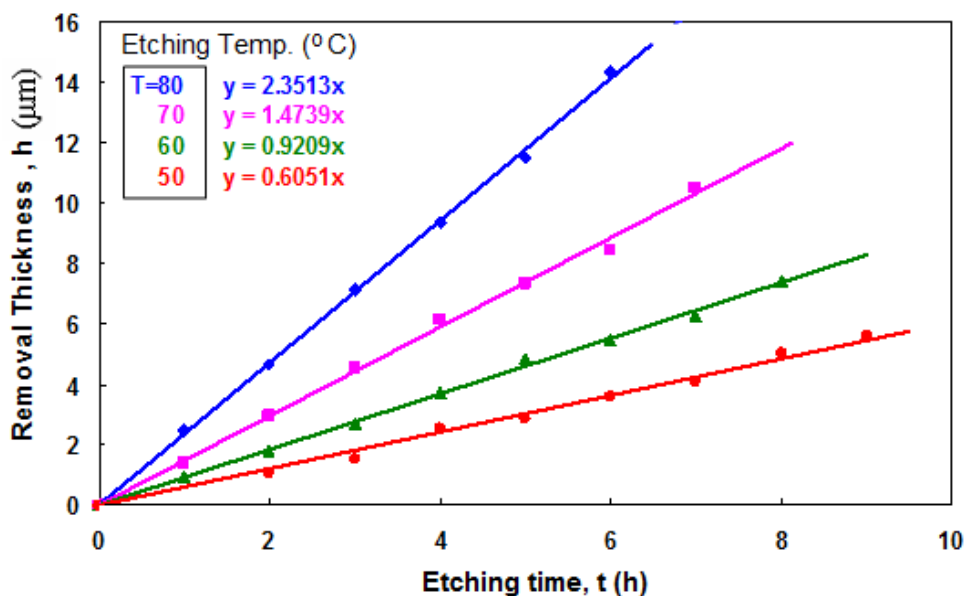


**Fig. 1:** the relationship between the removal layer from the CR-39 detector surface and the etching time at different etching time
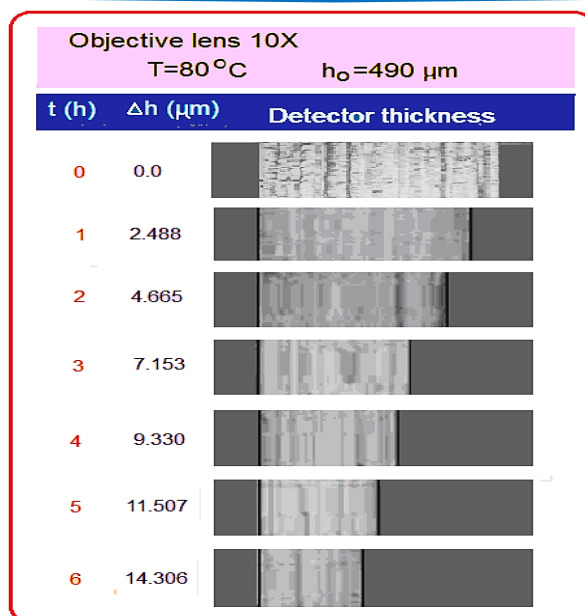
**Fig. 2:** Samples of the detector thickness images for CR-39 before and afteretching for 1-6 h at etching temperature 80$^o$C in the present study.

**Table 1:** The bulk etch rate for the CR-39 detector at different etching time

| Etching Temp. | | Bulk etch rate | |
|---|---|---|---|
| T (°C) | 1/T x10$^{-3}$ (K$^{-1}$) | V$_b$ (μm/h) | Ln V$_b$ |
| 80 | 2.833 | 2.351± 0.043 | 0.855 |
| 70 | 2.916 | 1.474± 0.0291 | 0.388 |
| 60 | 3.003 | 0.921± 0.049 | -0.0823 |
| 50 | 3.096 | 0.605± 0.0634 | -0.5025 |

   The more increase in the temperature of the etching solution, the more increase of the reaction energy and the reaction rate of the detector material with the chemical solution. This, in turn, causes more degradation in the surface layer molecules resulting in increases in the thickness of the layer removed from the detector surface and then the bulk etch rate. The bulk etch rate V$_b$ is a measure of the rate of the material that removed from the undamaged area of

the detector surface, and it is seen from Fig. 3 the $V_b$ increases exponentially with the temperature of the etching solution (T) according to the empirical formula given in Eq. (2) as,

$$V_b = 0.059 \, e^{0.0461T} \tag{2}$$

If T is in Kelvin (K) rather than $^oC$, the $V_b$ will be,

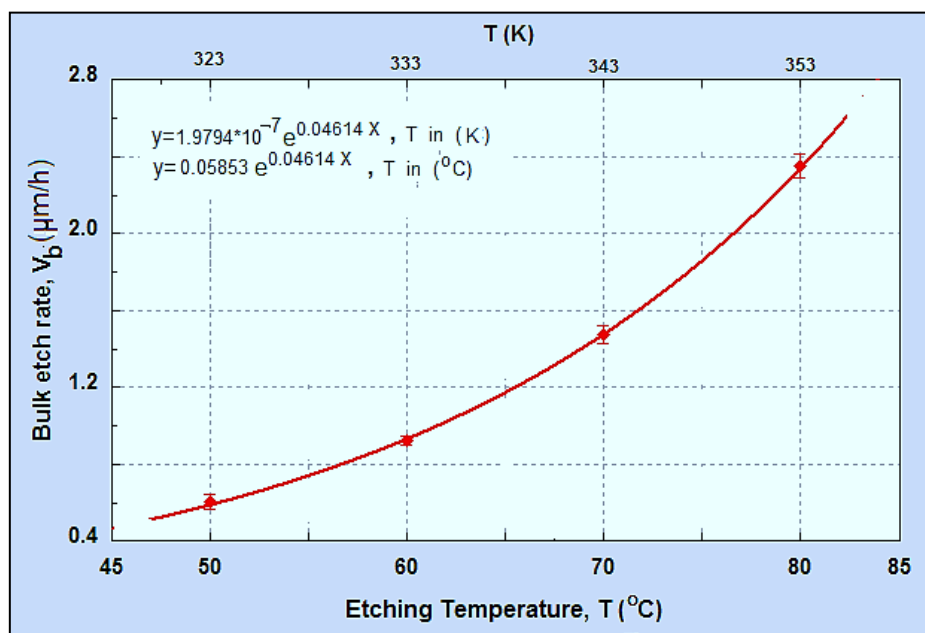$$V_b = 1.98 \times 10^{-7} \, e^{0.0461T} \tag{3}$$



**Fig. 3:** Variation of the bulk etch rate of CR-39 detector with the temperature of the etching solution

The mathematical formula of Eq. (2) appears similar to the exponential formula that is presented by [22], which have studied the bulk etch rate $V_b$ of the CR-39 detector using the chemical etchant NaOH with different concentrations and temperatures. The following mathematical formula was assumed [22]:

$$V_b = 1.27 \, e^{0.828C + 0.049T - 0.002CT - 17.624} \tag{4}$$

Where T and C are the temperature and concentration of the etching solution in Kelvin (K) and mol/L respectively.

Thus, for NaOH with a concentration of 6 N and temperature T, Eq. (4) becomes,

$$V_b = 4.05 \times 10^{-6} \, e^{0.037T} \tag{5}$$

Which is similar to the formula in Eq. (3) that investigated in this study.

Fig. 2 shows the variation of $LnV_b$ vrs 1000/T. According to the formula involving the activation energy as in Eq. 6 [23], the activation energy of the bulk etching have been determined from the slope of the line in Fig.2 on the basis of least square fit. The activation energy was found 0.446 eV for TASTRAK PADC CR-39 detector. However, this value is small in comparison with the values for such detectors which are between $0.75 < \varepsilon_o < 1$ [24].

$$V_b = V_o \, e^{-\varepsilon_o / kT} \tag{6}$$

Where k is the Boltzmann constant, $\varepsilon_o$ is the activation energy in eV, and $V_o$ is the constant of the proportion in μm/h.
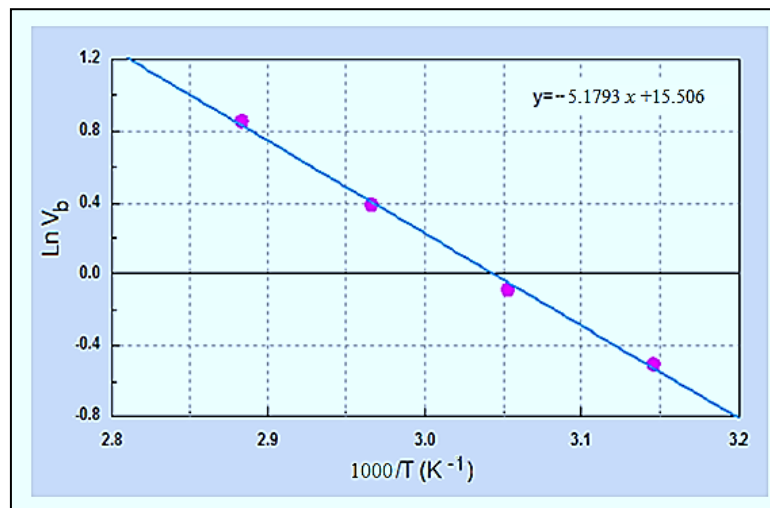


**Fig. 4:** The $LnV_b$ of the CR-39 detector against the reciprocal of the etching temperature 1/T

It is well known from various studies that the $V_b$ for different plastic nuclear track detectors is exponentially increasing with the etching temperature T, but what should be said here is that the detectors and even the same detectors of different origins may not always follow Eq. (6) to find the activation energy, but may follow other exponential equations such as Eq. (2) and (3) or other forms [15] in which the activation energy does not clearly appear. So, the activation energy of the detector cannot be calculated according to Eq. (6) in such a case, and if it is calculated, its value will not be accurate.

In light of this discussion, the exponential form of $V_b$ given by Eq. (6) which involves the activation energy may not be applied on the TASTRAK PADC plastic CR-39 detector used in the present study, and in contrast the exponential formula in Eq. (2) which have no activation energy term can be used for this detector.

The exponential change pattern of the bulk etch rate with the etching temperature is consistent with the results recorded by [4, 15]. The results obtained by Al-Nia'emi for the change of $V_b$ with the etching temperature Fig. 5, using the direct measurement of the track length-diameter ($L_e$-D) of a 2.5 MeV alpha particles in the CR-39 detector, ranged between 1.42-1.102 μm/h at etching temperatures 55-70$^o$C in a 6.25 N of aqueous solution of NaOH, while the values of $V_b$ obtained, in the present paper, for the CR-39 detector using the thickness difference method were 2.351-0.605 μm/h at etching temperatures 50-80$^o$C in a 6 N of aqueous solution of NaOH.

For the purpose of comparison, we present some values of $V_b$ for the CR-39 detector in other studies used the same or different method of measurement under similar or very similar etching conditions. The values of $V_b$ that have been found in [25, 26, 27] were 1.45, 1.23, 1.317μm/h respectively for the detector etched in a 6.25 N aqueous solution of NaOH at 70 °C, while the values that have found by Al-Hubayti and Younus [28, 29] were 1.268 and 1.421 μm/h respectively for the detector etched in a 6 N aqueous solution of NaOH at 70 °C. However, these results seem in good agreement with that found in the present study.
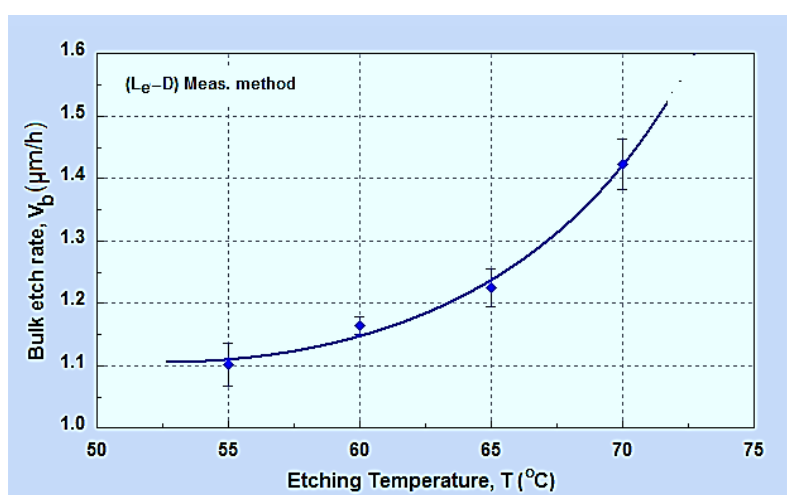


**Fig. 5:** Variation of The bulk etch rate with the etching solution temperature using the track length-diameter ($L_e$-D) measurement method [15]

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## 4. Conclusion

One of the positive aspects of the thickness difference method is that it provides high accuracy results, and it does not need to irradiate the detector with particles and to measure the track diameters and lengths. The effect of the etching temperature is significantly shown on the thickness of the detector and the Bulk etch rate. It was found that increasing the etching temperature by 10 $^o$C leads to an increase of 50-60% in $V_b$. Increasing the temperature of the etching solution leads to a linear increase of the thickness of the layer removed from the surface of the detector and it enhances the bulk etch rate of the detector, which increases exponentially with the temperature according to the exponential formula $V_B = a\,e^{bT}$. The TASTAK plastic CR-39 detector used in the present study was found to follow the exponential formula that does not involve the activation energy, where it may not be calculated because it gives an inaccurate value of up to 0.446 eV. This value is much lower than the known values for CR-39 which are between $0.75 < \varepsilon_o < 1$ [24].

## Acknowledgement

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