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UV effect on etching parameters and activation energy of CR-39 plastic detector

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Abstract. An investigation has been made relating the effect of UV radiations on etching parameters and activation energy of CR-39 (solid-state nuclear track detectors, (SSNTDs)). Corresponding changes in bulk etch rate (BER) (V_B), track etch rate (TER) (V_T), sensitivity (S), critical angle (θ_C) and efficiency (η) due to pre-irradiation and post-irradiation on CR-39 polycarbonate plastic detector have been reported and discussed in detail. Both bulk activation energy (BAE) (E_B) and track activation energy (TAE) (E_T) are reported to decrease with post-irradiation and are explained based on chain degradation, free radical formation, cross-linkage, chain scission and softening of the detector material. Further unusual decrease in BAE (E_B) and TAE (E_T) was observed for pre-irradiated CR-39 polycarbonate detectors, possibly due to the nature and density of the detector material.

Keywords. CR-39 detector; UV radiation; alpha-particles; bulk etch rate; track etch rate; activation energy.

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1. Introduction

Chemically etched solid-state nuclear track detectors (SSNTDs) have been widely developed and applied in fields such as nuclear physics, radiation dosimetry, 4π ion detection in plasma focus devices and in particle spectrometry. Gamma radiations, X-rays, UV, IR and protons impart significant changes in the track etch parameters [BER (V_B), TER (V_T), sensitivity (S), critical angle (θ_C) and efficiency (η)] of these SSNTDs due to their low linear energy transfer (LET) threshold [1–7]. CR-39 [polyallyl diglycol carbonate $(C_{12}H_{18}O_7)_n$] is one of the most widely used SSNTDs having excellent properties. CR-39 is sensitive to light and heavy ions but insensitive to background X-rays and γ-rays, making it an ideal choice [7, 8].

The energy carried by electromagnetic (lower LET) radiations and charged particles do not produce tracks in the CR-39 detector but lead to bond scission resulting in the degradation of the detector surface. This ultimately results in the modification of structural, optical and track etch parameters of CR-39. Change in the parameters is either due to the cross-linkage or chain scission process. Apart from this, the breaking of the long polymer chain leads to the formation of CO, CO_2 and H_2 [9]. Due to the passage of ionising radiations, initially, two alkyl radicals and a polycarbonate-ended radical are produced, which later results in 2,2-oxydiethanol diradical and CO_2 [10]. Alpha-irradiated CR-39 detectors prevent the recombination of free radical pairs in the presence of air (oxygen) leading to the formation of a permanently damaged track along the length [10]. Now it is very interesting to observe the influence of these LET radiations on the etching parameters of these detectors. The modification produced in the properties of the CR-39 detector due to these radiations results in the application of these detectors in science and technology.

In our earlier paper, [4], a detailed investigation of the UV effects at 160 nm on the Makrofol-E detector due to fission fragments has been reported. This paper reports a detailed investigation of the UV effect on the etching parameters of CR-39 at 120 nm and 160 nm. The prime objective of the present study is to experimentally verify the effects of etching conditions on track etch parameters of CR-39 and to observe the dependence of UV radiations on CR-39.

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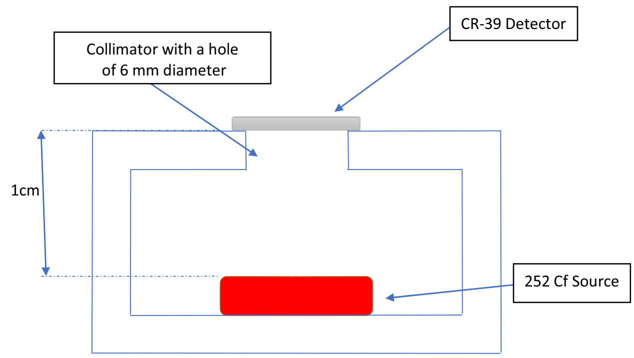

Figure 1. Schematic diagram of the normal irradiation of CR-39 in 2π geometry from Cf-252 source.

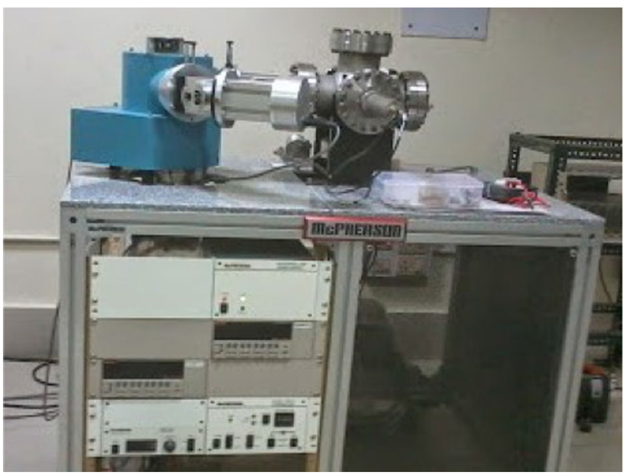

Figure 2. VUV Monochromator (224–324) McPherson used for UV irradiation.

2. Material and methods

2.1 *Irradiation source*

Cf-252 source was used for the irradiation of CR-39 with an activity 200 μCi having diameter 6 mm, in Nuclear Physics Laboratory, Physics Department, Banaras Hindu University (BHU), Varanasi, India. All the exposures were performed for 60 min at nearly normal incidence kept in the closed environment at a pressure of 10^{-2} torr. The detectors were kept at a distance of 1 cm from the source (figure 1) at an opening of 6 mm diameter.

2.2 *UV source*

A vacuum ultraviolet monochromator (VUV) (234/302) has been used to irradiate CR-39 (figure 2). It is an f/4.5, aberration-corrected monochromator with a focal length of 200 mm with 0.1 nm FWHM spectral resolution and 1200 g/mm grating (40 × 45 mm). The precision slits are micrometre adjustable from 0.01 to 3 mm. The wavelength varies from 115 to 370 nm with an accuracy of ±0.10 nm and a wavelength reproducibility of ± 0.05 nm. Deuteron lamp of 30 W was used at 30° angle of incidence at a standard pressure of 10^{-6} torr.

2.3 *Experimental details*

CR-39 plastic detector from Fukuvi Chemical Industry, Tokyo, Japan, with an average thickness of 250 μm and a density of 1.32 g/cm^3, was cut in an area of 1 cm × 1 cm. Twenty-five samples were equally divided into five sets (each having five detectors) as Set I, Set II, Set III, Set IV and Set V. Set I was first irradiated with α-particles and then exposed to UV radiations at 120 nm wavelength (referred to as Ist post-exposed set). Set II was also irradiated with α-particles and then exposed to UV radiations at 160 nm (referred to as 2nd post-exposed set). In Sets III and IV, the process was reversed, i.e. first exposed to UV radiations at 120 nm and at 160 nm, followed by irradiation of α-particles (referred to as 1st and 2nd pre-exposed sets, respectively). Set V was only irradiated with α-particles and used for reference purpose (unexposed set).

8.0 N NaOH solution was used for etching CR-39 plastic detectors after irradiation at five different temperatures, 60°C, 65°C, 70°C, 75°C and 80°C, for different time periods. Eight grams NaOH in 25 ml water and 25 ml ethanolamine was used as an etchant. Chavan *et al* [11] observed that 8 g NaOH in 25 ml water and 25 ml ethanolamine is very effective in enlarging alpha tracks in CR-39 SSNTDs. Error in the temperature was calculated to be ±1°C. The detectors were treated with ethyl alcohol and washed with distilled water to remove any contamination after etching. After that, the detectors were dried with tissue paper and kept in an insulated box. After etching, the Olympus microscope (BH-02) having a magnification of 100× (oil dipping) was employed to measure the track diameters of α-particles [4].

Different techniques such as the track diameter method [4], the gravimetric method [12] and the spectrometric method [13] are employed to calculate bulk etch rate (BER). Track diameter method, being the most widely used method, was used to calculate BER (V_B) at 60°C, 65°C, 70°C, 75°C and 80°C using the relation

$$V_{\mathrm{B}} = \frac{D}{2\Delta t}, \qquad (1)$$

where Δt is the time taken to develop diameter D of the α-particle in the CR-39 plastic detector.

Table 1. Various track etching parameters for post-exposed (alpha + UV) CR-39 detectors.

Wavelength	Temp. (°C)	V_B (μm/h)	V_T (μm/h)	$S = V_T/V_B$	$\theta_C = \sin^{-1}[V_T/V_B]$	$\eta = 1 - (V_B/V_T)$
UV = 120 nm	60	0.742	2.517	3.392	17.14	0.70
	65	0.978	3.237	3.310	17.58	0.70
	70	1.355	4.258	3.142	18.55	0.68
	75	1.831	5.515	3.012	19.39	0.67
	80	1.987	5.758	2.898	20.18	0.65
UV = 160 nm	60	0.632	2.099	3.321	17.52	0.70
	65	0.897	2.920	3.255	17.89	0.69
	70	1.231	3.932	3.194	18.24	0.69
	75	1.721	5.170	3.004	19.44	0.67
	80	1.924	5.608	2.915	20.06	0.66

In case of fission fragments (ff), then formula used will be

$$V_B = \frac{D_{ff}}{2\Delta t},$$

where D_{ff} is the diameter of the over-etched fission fragments.

Both the methods are equally valid and Hermsdorf *et al* [14] discussed in detail the pros and cons of both the methods.

For the calculation of track etch rate (TER), the following relation is used:

$$V_T = \frac{\Delta L}{\Delta t}, \quad (2)$$

where the length of the track increases by ΔL in time Δt.

The optimum condition for the formation of the track is $V_T > V_B$. The sensitivity of the detector is the ratio of these etching rates

$$S = \frac{V_T}{V_B}. \quad (3)$$

The critical angle decides the appearance of the etched track and the relation is

$$\theta_C = \sin^{-1}\left[\frac{V_T}{V_B}\right]. \quad (4)$$

The last etching parameter is the etching efficiency of the detector, which can be calculated using the following equation:

$$\eta = 1 - \frac{V_B}{V_T}. \quad (5)$$

For the calculation of activation energies of bulk etch rate E_B and track etch rate E_T at a particular temperature, the following Arrhenius relations are used:

$$V_B = A\exp\left(\frac{-E_B}{KT}\right), \quad (6)$$

$$V_T = B\exp\left(\frac{-E_T}{KT}\right), \quad (7)$$

where E_B is the bulk activation energy, E_T is the track activation energy, k is the Boltzmann constant, T is the absolute temperature and A, B are constants.

3. Result and discussions

Pre- and post-effects of UV radiations on 252-Cf-irradiated (60 min duration) CR-39 have been reported. Table 1 shows the values of V_B, V_T, S, θ_C and η for Sets I and II (1st and 2nd post-exposed). The experimental values of V_B, V_T and θ_C display a linear increasing trend while S and η display a decreasing trend with the etching temperature.

Table 2 shows the values of V_B, V_T, S, θ_C and η for Sets III and IV (Ist and 2nd pre-exposed). The overall values of V_B, V_T and θ_C in table 2 show a decrease in their respective values while S and η display a significant increase in their values. Zaki *et al* [15] examined the result of He–Ne laser radiation on CR-39 and CN-85 detectors and predicted a downward trend in V_B up to a dose of 100 J/cm^2. Saffarini *et al* [9] found similar results, i.e. a rise in V_B and V_T with temperature and a fall with exposure to IR radiation on the CR-39 detector. Herrera *et al* [16] observed the effect of α-particles (1 MeV to 5.5 MeV) on CR-39 and found a decrease in mean track diameter for a lighter dose of X-rays while no change for higher doses.

Table 2. Various track etching parameters for pre-exposed (UV + alpha) CR-39 detectors.

Wavelength	Temp. (°C)	V_B (μm/h)	V_T (μm/h)	$S = V_T/V_B$	$\theta_C = \sin^{-1}[V_T/V_B]$	$\eta = 1 - (V_B/V_T)$
UV = 120 nm	60	0.514	2.101	4.087	14.16	0.75
	65	0.724	2.877	3.974	14.57	0.75
	70	0.995	3.837	3.856	15.02	0.74
	75	1.355	5.146	3.798	15.27	0.74
	80	1.678	5.561	3.344	17.56	0.70
UV = 160 nm	60	0.589	2.056	3.491	16.65	0.71
	65	0.765	2.641	3.452	16.83	0.71
	70	1.067	3.469	3.251	17.91	0.69
	75	1.568	4.723	3.012	19.39	0.67
	80	1.874	5.534	2.953	19.79	0.66

Table 3. Various track etching parameters for un-exposed (only alpha) CR-39 detectors.

Temp. (°C)	V_B (μm/h)	V_T (μm/h)	$S = V_T/V_B$	$\theta_C = \sin^{-1}[V_T/V_B]$	$\eta = 1 - (V_B/V_T)$
60	0.453	1.630	3.598	16.13	0.72
65	0.675	2.272	3.366	17.28	0.70
70	0.989	3.139	3.174	18.36	0.68
75	1.374	4.074	2.965	19.71	0.66
80	1.652	4.432	2.683	21.88	0.63

Sensitivity and efficiency of CR-39 show a uniform decrease in their respective values, but the overall value of sensitivity and efficiency is slightly higher in pre-exposed CR-39 detectors than in post-exposed CR-39 detectors.

Table 3 shows V_B, V_T, S, θ_C and η for Set V (unexposed). These data are used to compare the overall change in track etch parameters during the pre- and post-exposure to UV radiations.

All CR-39 detectors show enhancement in V_B and V_T. However, post-exposed detectors have higher values of V_B and V_T than the pre-exposed detectors. Tse *et al* [10] also reported such patterns on CR-39 detectors on UV exposure. Farooq *et al* [17] reported a sudden surge by 9% in track diameters (bulk etch rate) when CR-39 detectors were post-irradiated (alpha + UV) with UV radiations. Caresana *et al* [18] reported the effects of fading and ageing of CR-39 plastic detectors leading to change in efficiency of CR-39 detector and around 45% reduction is observed when CR-39 detector is exposed in radon chamber and etched after six months.

V_B and V_T are strongly dependent on etching conditions, mechanical properties, chemical structure and hardness of the detector, and the ratio $S = V_T/V_B$ compensates for these influences. Internal and external conditions of detector sample treatment also play a vital role in the sensitivity of CR-39. Under low temperature and pressure, the sensitivity of CR-39 shows drastic variations from those at room temperature and normal pressure [19].

From tables 1–3, S of CR-39 shows a decreasing trend with an increase in temperature. The numerical values of S for pre-exposed samples are reported to increase while post-exposed samples report a decrease when compared to the unexposed samples. Hence, the peak sensitivity of CR-39 is recorded for the pre-exposed detector at 60°C. This temperature may be recorded as the optimum temperature for CR-39.

Figures 3 and 4 display the linear dependence of V_B and V_T on temperature inverse for post-exposed (120 nm and 160 nm), pre-exposed (120 nm and 160 nm) and unexposed CR-39. E_B and E_T are calculated with the help of the slopes of these plots. These values are reported in table 4.

Table 4 shows that the bulk activation energy of the unexposed CR-39 is 0.67 ± 0.04 eV. Nikezic and Yu [20]

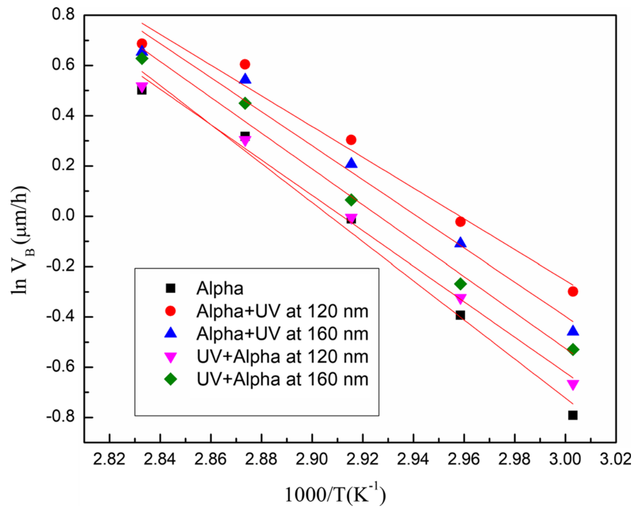

Figure 3. Variation of ln V_B (μm/h) with $1/T$ (1000/K) for pre-, post- and unexposed CR-39 detector.

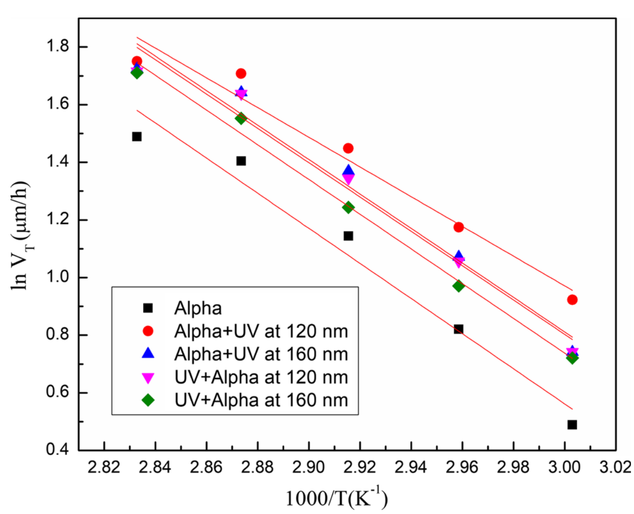

Figure 4. Variation of ln V_T (μm/h) with $1/T$ (1000/K) for pre-, post- and unexposed CR-39.

Table 4. Activation energy of post, pre- and unexposed CR-39 detector.

Irradiation type	E_B (eV)	E_T (eV)
Unexposed (Alpha only)	0.67 ± 0.04	0.52 ± 0.04
Aplha + UV at 120 nm (Ist post-exposed)	0.53 ± 0.04	0.45 ± 0.03
Aplha + UV at 160 nm (2nd post-exposed)	0.58 ± 0.04	0.51 ± 0.04
UV at 120 nm + alpha (Ist pre-exposed)	0.60 ± 0.04	0.52 ± 0.04
UV at 160 nm + alpha (2nd pre-exposed)	0.51 ± 0.04	0.52 ± 0.04

and Mhatre *et al* [21] reported the activation energy of CR-39 (different manufacturers) from 0.57 ± 0.04 eV to 0.92 ± not reported eV. The value reported in the present study lies in this range. Post-exposure to UV radiation at 120 nm reduces bulk activation energy to 0.53 ± 0.06 eV while post-exposure at 160 nm is calculated to be 0.58 ± 0.02 eV. This decrease in the value of bulk activation energy may be due to faster etching due to the exposure to UV radiations. Pre-exposure with UV radiations at 120 nm is calculated to be 0.60 ± 0.04 eV, while at 160 nm, the value reduces to 0.51 ± 0.03 eV. Track activation energy of the unexposed CR-39 is 0.52 ± 0.05 eV. When post-exposed with UV radiations at 120 nm, track activation energy reduces to 0.45 ± 0.04 eV while post-exposure to UV radiation at 160 nm reduces the value to 0.51 ± 0.06 eV. We can predict that post-exposure to UV radiation reduces the activation energy (E_B and E_T). This may be possible due to the energy carried by UV radiation. The energy of UV radiation softens the detector material and leads to the generation of free radicals. This process ultimately results in the chain scission of polycarbonate molecules by UV radiation.

When pre-exposed with UV radiations, CR-39 undergoes comparative softening, leading to smaller variation in activation energies. Unlike Jain *et al* [4, 5] and Jain and Kumar [6], the activation energy decreases slightly due to pre-exposure to UV radiation. A possible reason for this decrease in activation energy may be due to the nature and density of CR-39. It may further be pointed out from table 4 that only post-exposure to UV radiation at 120 nm, reduces the value of E_T (TAE) and no change in the value of E_T (TAE) was observed for the rest of the cases.

4. Conclusions

Based on the discussion, pre- and post-exposure to UV radiations at 12 nm and 160 nm on CR-39, the following conclusions may be drawn.

- An increase in V_B and V_T can be seen for pre- and post-irradiated CR-39. The values are comparatively higher for post-exposed CR-39.
- Sensitivity of CR-39 polycarbonate plastic detectors shows decrement in their numerical value due to post-exposure.
- An increase in sensitivity is reported for pre-exposed CR-39 and highest sensitivity is recorded for UV radiations at 120 nm at 60°C.
- Critical angle is found to decrease on pre-exposure while it increases on post-exposure.
- Efficiency decreases on increases on pre-exposure and decreases on post-exposure.

- A decrease in activation energy on post-exposure due to chain scission and free radical formation is recorded.
- An exceptional behaviour, a slight decrease in activation energy on post-exposure can be seen. This is possibly due to the nature and density of the detector material.

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