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Surface chemical etching behavior of LR-115 type II solid state nuclear track detector: effects of UV and ultrasonic beam

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Solid State Nuclear Track Detectors (SSNTDs) are well known for the detection of ionizing radiation through track formation of heavy ionizing particles. The LR-115 detector was commonly used SSNTD for the detection and measurement of Rn^{222} and its progeny. Several techniques of track revelation were known but, the chemical etching technique was the most frequently used. So the surface chemical etching behavior of LR-115 polymer is an important parameter in the study of ionizing radiation. The LR-115 type II SSNTD were etched by giving pre-UV and post-UV exposure to alpha irradiation on it, by the application of ultrasonic beam on the etchant after alpha irradiation and under normal conditions. The effects are discussed in terms of bulk etch rate V_b, track etch rate V_t, and track sensitivity. The result shows that the revelation of alpha particle tracks in the detector was enhanced by the application of ultrasonic beam on etchant. Both the bulk etch rate V_b and track etch rate V_t were found to be higher in case of ultrasonic etching than the normal etching conditions. Both the bulk etch rate V_b and track etch rate V_t were found to be higher in case of post-UV exposure after alpha irradiation to the detector than the normal alpha irradiated detector.

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1. Introduction

Solid State Nuclear Track Detectors (SSNTDs) are widely used in a variety of fields in science and technology, especially in the detection of ionizing radiation through track formation of heavy ionizing particles [1]. The composition, processing, and applications of solid-state nuclear track detectors (SSNTDs) are sufficiently discussed by many researchers [2-5]. SSNTDs, particularly LR-115, are used to measure the Rn^{222} and its progeny. The red LR-115 type II detector (from DOSIRAD) is a commercially available red cellulose nitrate detector with an active layer of 12 μm on a 100 μm clear polyester base (as declared by the manufacturer). Alpha particles emitted by Rn^{222} and its progeny hit the detector and leave the latent tracks in it. Many techniques of track revelation are known, e.g. grafting [5, 6] but, the chemical etching technique is the most frequently used in which tracks can be made visible under the optical microscope after chemical amplification via etching. Normally the radon monitoring is done by employing the detector in the natural environmental conditions that can change the detectors properties. These environmental effects are more prominent in countries like India, where temperature levels are high and solar radiations are very intense. As the track development in an SSNTD is based on two parameters V_t (track etch rate) and V_b (bulk etch rate) and it is known that V_b depends on many factors like structure, chemical composition and preparation of the detector, etching conditions (like chemical nature of etchant, temperature, concentration as well as the stirring conditions) aging, pre-irradiation treatment and storage conditions, environmental conditions during irradiation (ionizing as well as the non ionizing radiation) etc. [7-10], so the study of surface chemical etching behavior of this polymer by giving pre-UV and post-UV exposure to alpha irradiation on it is an important aspect. The measurement of radon values depends upon the exact counting of the tracks produced by alpha particles on these detectors. The counting of these tracks is normally done by an optical microscope of suitable magnification. So the quality of visualization of these tracks is also an important factor. Some of the workers have reported the methods of enhancing the revealed tracks for registration of alpha particles [11-12]. The ultrasonic beam is employed to the etchant to find its use as a tool for enhancing the revelation of the alpha particle tracks in the detector. The track sensitivity (Vt/V_b) describes the shape and size of the track and depends on both bulk etch rate and track etch rate. Various techniques were reported for determination of the bulk etch rate [13-16], however we used direct measurement technique discussed by Mehta et al. and Shikha et al. [17-18]. The track etch rates were also determined for finding the track sensitivity. This paper deals with two factors viz. conditions during track formation and the use of an enhancement tool for track revelation in LR-115 type II detectors. The results of both pre-UV and post-UV

exposure to alpha irradiation on the detector are compared with the results of normal etching conditions. The results of application of ultrasonic beam on the etchant after alpha irradiation are compared with the results of etching under normal conditions.

2. Experimental details

Pristine LR 115 type-II (cellulose nitrate) was procured from DOSIRAD, France in the form of thin films of active layer thickness of 12 µm on a clear 100 µm polyester base. Several samples of size 1.5' × 1.5' were cut from the films. In order to ascertain the effect of UV light and ultrasonic beam four sets of samples (set A, B, C and D) were made containing three samples in each set for this study so as to reduce the error in the results. The thickness of each sample was measured before etching on a specified area using a digital micrometer (Mitutoyo, No. 293-821) having least count of 1 µm.

Alpha irradiation: All alpha irradiation were carried out using accessories (having a chamber to create vacuum so that collimated particles were incident normal to the surface of the sample) provided with PHOENIX (physics with home-made equipment & innovative experiments) interface developed by Inter University Accelerator Centre (IUAC) New Delhi India [19]. The Am^{241} source was used as a point source having alpha emission of 5.485 MeV and was placed at a distance of 1 cm from the LR-115 samples kept in a sample holder as shown in Fig. 1. All the samples were irradiated for same time period.

Fig. 1. (a) PHOENIX Interface (b) Vacuum Chamber with source and Sample.

UV irradiation: All the samples of LR-115 were exposed to UV light for two hours in a UV chamber using a black lamp (SAMSON made) of 125 W at λ = 320 nm as shown in Fig. 2(a). The UV chamber was having the facilities of a black lamp (λ = 320 nm), mercury lamp of 125 W (mixed light), UV tubes of 60 W (λ = 254 nm) and a simple white light tube of 30 W. The samples were fixed in a sample holder that was placed at a distance of 10 cm (so that spreaded UV light falls on the samples for the uniformity) directly below the UV lamp. The whole arrangement was suitable to study the photo-oxidation of the polymeric samples under the UV light. As pre- and post-UV exposure to alpha irradiation of the samples was a natural phenomena in the radon measurement experiments, so we tried to study both the phenomenon in this paper.

Fig. 2. (a) UV Chamber Fig. 2: (b) Ultrasonic Cleaner.

Ultrasonic Etching: All the samples of LR-115 used to study ultrasonic's effect were etched under the application of ultrasonic beam in a temperature controlled ultrasonic cleaner shown in Fig. 2(b) (Model No. GB-5000B) of 5.0 l capacity with a power output of 80 W. The samples were etched in a beaker (containing etchant) submerged in the water of the ultrasonic cleaner tank. The ultrasonic were passed through the water of the ultrasonic tank to the etchant of the beaker. The etching was done at a temperature of 60°C ± 1°C. Due to the passage of ultrasound through the etching medium, the temperature of etching medium rises. The rise in temperature depends on the acoustic power of the ultrasound. We have tried to isolate this effects produced by ultrasound by monitoring the temperature continuously and adding the cold water in the tank periodically. To avoid this effect we have also taken small time for the etching cycles of 20 min. each.

Normal Etching and different measurements: All batches of samples (except ultrasonic etched samples) were etched in 2.5 N NaOH in a constant temperature water bath at 60°C ± 0.5°C for different time intervals in the steps of 20 minutes each without stirring as described in our earlier paper [20]. After each etching interval the samples were washed thoroughly in distilled water and then dried in open air for 30 minutes for both normal and ultrasonic etching. The etchant was changed periodically so that concentration of etchant remained the same during the experiment. The thickness measurements for a particular sample were taken after etching on the specified area as discussed earlier. These samples were viewed through an optical microscope fitted with a camera as shown in Fig. 3 (Model No. 7001 IMS, Vaiseshika Ltd.). The photomicrographs of revealed alpha tracks in these detectors were taken using that microscope at 200X magnification.

Fig. 3. Optical microscope with a camera.

3. Results and discussion

The surface chemical etching behavior of LR 115 type-II was studied by measuring the cumulative thickness (total etched-out thickness of the sample, as measured by the digital micrometer before and after etching), removed. Table 1 shows the results obtained for the present study for etching in 2.5 N NaOH at 60°C ± 0.5°C for different sample etching conditions (Set A, B, C and D). It shows the values of bulk etch rate, track etch rate and the track sensitivity for all the sets of the samples. The bulk etch rate and track etch rate were calculated using the formulas given elsewhere for CR-39 detector [21] for the bulk etch rate V_b:

$$V_b = \Delta x/2t \quad (1)$$

where Δx is the thickness of the detector removed during the etching in time t.
for the bulk etch rate V_t:

$$V_t = V_b \ [1+Y^2]/[1-Y^2] \quad (2)$$

where $Y = d/2\Delta x$, d is the track diameter and Δx is the thickness of the detector removed during the etching in time t.

However in our case of LR-115 type II detector, this detector has sensitive layer on only one surface and polyester layer on other surface i.e. resistant to etching in our case (the polyester layer was obtained by removing the active layer of LR-115 using a razor and then employed for etching in 2.5 N NaOH at 60°C ± 0.5°C, we found no removal of the layer for 100 min of etching time), so we have omitted the factor 2 for the calculation of these parameters [4, 22]. The passage of charged particles through the polymer leads to the chain scission and creates narrow paths of intense damage. Figure 4 shows the photomicrographs of the samples of different sets etched under different conditions revealing the alpha particles tracks in LR-115.

Table 1. Sample condition, etching time, bulk etch rate, track etch rate and the track sensitivity for the etching of different samples in 2.5 N NaOH at 60°C ± 0.5°C.

S. No.	Sample condition	Etching Time (min)	Bulk Etch Rate V_b (μm h^{-1})	Track Etch Rate V_t (μm h^{-1})	Track Sensitivity V_t / V_b
1	Only alpha irradiated (Set A)	100	4.2	8.51	2.03
2	Post irradiation (alpha irradiation + 2 h UV exposure) (Set B)	80	4.8	9.27	1.97
3	Pre irradiation (2 h UV exposure + alpha irradiation) (Set C)	80	4.7	8.44	1.76
4	Alpha irradiated + Ultrasonic etched (Set D)	80	5.33	12.68	2.38

Discussion: i) Effect of UV exposure: Table 1 clearly shows the effect of exposure of UV light for both pre-irradiated and post-irradiated samples. It can be seen from that with the exposure of UV light to samples the track sensitivity of the samples reduces than the track sensitivity of the normal condition samples irrespective of that the UV light exposure being done prior to alpha irradiation or after the alpha irradiation. The decrease of track sensitivity can be understood in terms of slight increase in the bulk etch rate of the samples and not much change in the track etch rate of the samples. The UV exposure of the polymer samples in the presence of oxygen causes deterioration of its properties. The possible method of this photo-oxidation is a free radical chain mechanism that goes with initiation, propagation, termination and chain branching. In case of LR-115 type II detector a die is being added to provide the detector a red color.

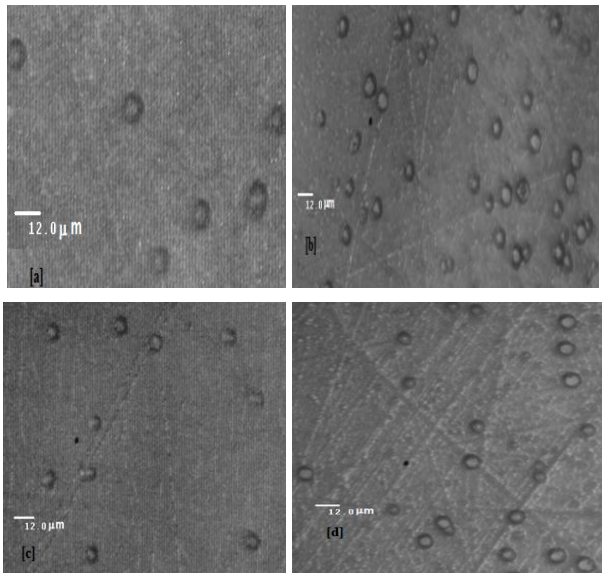

Fig. 4. Photomicrographs for the revelation of the alpha particle tracks in the LR-115 polymer for different sets of samples [a] normal alpha irradiated sample (set A), [b] alpha irradiated then 2h UV exposed (set B), [c] 2h UV exposed then alpha irradiated (set C) and [d] alpha irradiated then ultrasonically etched (set D).

However the use of dies can minimize photo-oxidation in the polymer and thus reduce the polymer degradation. So the increase in bulk etch rate may be due to thermo-oxidation whose process is almost similar to photo-oxidation. Normally there is sharp increase in bulk etch rate with UV exposure as reported in [23] for CR-39 SSNTD case, but in our case the increase is not very sharp as explained above. The track sensitivity of set C (2 h UV exposure then alpha irradiated) samples is less than the track sensitivity of set B (alpha irradiated then 2 h UV exposed) samples and so the efficiency of Set C samples would be higher than that for set B samples. This can be explained due to more track etch rate in post-irradiation case. The reason behind that may be the creation of some preferential sites for the absorption of UV light quanta due to the damage created by alpha particles. Those preferential sites are created at the track sites that lead to the little increase in the track etch rate and hence the track sensitivity. This result is in agreement with the suggestions given in [24]. However the decrease in track sensitivity is not drastic for both pre-irradiated and post-irradiated samples of LR-115, which shows that the sensitive layer of this detector is not much affected by the exposure of UV beam for the 2 h of exposure time. This may be due to the protective behavior of its red dye against UV exposure.

ii) Effect of ultrasonic etching: Fig. 4 (a) and (d) shows the tracks of the alpha particles revealed by the normal etching method and ultrasonic etching method respectively. In both the cases the tracks were round but latter were more pronounced and easy to identify and have slightly more diameter than the former one. The ultrasonic etching process reveals the tracks in less time than the normal etching case.

The rate of dissolution of the LR-115 in NaOH depends on the concentration of the active layer of cellulose nitrate in the etchant. It becomes slower as the concentration of the cellulose nitrate in the etchant increases. Under normal conditions, if the bulk etching rate is slower than the rate of etching along the radiation damaged region, alpha tracks can be etched out. However, as the concentration of cellulose nitrate dissolved in the track core region becomes higher, the rate of dissolution becomes slower; so the bulk etching rate may be in comparison to the track etching rate. This explains the situation of the normal etching in which tracks are revealed but are not of good quality. Both the bulk etching rates and track etching rates were found to be more in ultrasonic etching condition than in the normal condition. However the track etching rate increases more than the bulk etching rate in ultrasonic etching condition that leads to the increase in the track sensitivity. This suggests that the track etching response is better in ultrasonic etching in comparison to normal etching. To understand these results we have to look into the mechanism of ultrasonic etching. Ultrasonic's travel in the form of compression and rarefaction in the etchant, they produce rapidly changing pressure in the liquid that causes the formation of cavities where the pressure is relatively low. These cavitation bubbles of the etchant are supported only by the negative pressure or rarefaction of the ultrasonic's surrounding them. This bubble, subjected to alternating compression and rarefaction, starts to oscillate in resonance with ultrasonic's and grow larger and finally results in a violent collapse or "implosion". During implosion, the inward rush of the bubble upon itself driven by the compression of the ultrasonic's results in form of energy of collapse concentrated on a very small area. The energy of this collapse is directed towards the sample. These implosions generate, momentarily, local pressures up to several GPa and temperatures at the centre of bubble of the order of 10^4 - 10^6 K. Ultimately the passage of ultrasonic's through etchant creates the cavitation-implosion process, and many tiny vacuum regions on the surface of the sample. The action of the vacuum on the track core region will suck out the solution saturated with the dissolution of active layer of cellulose nitrate and replace it by fresh solution. Hence the tracks etch rate increases more than the bulk etch rate in the case of ultrasonic etching.

Conclusion: Our study shows that there is an increase in bulk etch rate of UV exposed samples than the normal samples. However, the track etch rate of UV exposed samples does not show this behaviour. Although there is loss of track sensitivity of the UV exposed samples, but that change is not too drastic so as to decrease the efficiency of the samples drastically. The LR-115 detectors can be used effectively in the radon measurement study in natural environment where the exposure of the samples to sunlight is for a small time (1-2 h) during the whole day. However the response of LR-115 detector in the radon measurement study for the exposure time of nearly 10 h in a day to the Sunlight for long time (15-20 days) has to be checked out. Our study also indicate that the geometrical shapes of the etched tracks due to alpha

particles in LR-115 detector registered from Am^{241} source are more sharper and easily identifiable in ultrasound-induced etching than those obtained by conventional etching. This tends to suggest that the track etching response is better in ultrasonic etching as compared to normal etching. The result supports the well established idea that heat enhances the chemical etching of particle tracks and agitation is required for removing the etch products.

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