Optimization of CR-39 as neutron dosimeter

Vijay Kumar\textsuperscript{a}, R G Sonkawade\textsuperscript{b}\textsuperscript{*} & A S Dhaliwal\textsuperscript{a}

\textsuperscript{a}Department of Physics, Sant Longowal Institute of Engineering and Technology Longowal Distt. Sangrur, Punjab 148 106, India
\textsuperscript{b}Inter University Accelerator Center, Aruna Asif Ali Marg, New Delhi 110 067, India

\textsuperscript{*}E-mail: rgs22@rediffmail.com

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The chemical etching parameters (etching time, temperature, normality of etchant etc.) for use of CR-39 as neutron dosimeter have been optimized. The CR-39 films placed in polyethylene radiator were exposed to $^{252}$Cf neutron source for different time intervals and the neutron fluences were varied from $4.68\times10^6$ to $2.7\times10^8$ n/cm$^2$. Etched tracks were analyzed and counted at 400X with microscope and neutron recoil track density was calculated after every hour of etching. It was found that the track density increases up to a certain etching time and then decreases for higher etching time, which is considered due to removal of shallow tracks and overlapping of the tracks due to their size enlargement. Optimized etching conditions found are, 7 N NaOH solution at 70$^\circ$C (±) for 9 hr with continuous stirring. However, we found a linear relationship between neutron fluence and track density. UV-visible spectra were also obtained and analyzed for the pristine and neutron irradiated CR-39 films in the wavelength range 200-700 nm at room temperature. As seen from absorption spectra, the absorption edge is shifted towards longer wavelength with increase of fluence and therefore, indicates a decrease in the band gap. The study may be important for the future high energy accelerators and Radioactive Ion Beam Facilities where CR-39 can be used as a neutron dosimeter.

Keywords: CR-39 polymer, Neutron irradiation, Etching, Track density, UV-vis spectroscopy

1 Introduction

The Solid State Nuclear Track Detectors (SSNTDs) are highly potential agents for novel applications in radiation research and radiological protection dosimetry in many domains. They are able to register the charged particles by the radiation induced damage caused along their interaction path. Since, the latent damage regions are very thin and even cannot be seen clearly from optical microscope. The damage regions produced by radiation on material are revealed and amplified for visualization in an optical microscope using a well reported technique known as chemical etching. Thus, chemical etching is one of the most important processes in track detection technique. It was observed that the zone of structure damage might be increased to $10^{-4}-10^{-2}$ cm by etching with suitable chemical reagent. The versatility of SSNTDs in neutron detection and dosimetry is well known through decades. SSNTDs have attractive characteristics compared to other detectors, such as non-fading of tracks, insensitivity to gamma, UV and X-rays etc. The most sensitive and well studied SSNTDs till now used for neutron dosimetry\textsuperscript{1-3} and track registration is poly [diethylene glycol bis-(allyl carbonate)], commonly named as CR-39 due to its high sensitivity to fast neutrons.

In early years, optical and etching characteristics of polymeric detectors were mainly studied by heavy ions, neutrons and gamma irradiation etc.\textsuperscript{4-13}, however, limited studies have been reported on neutron induced effects on etching and optical properties of detector\textsuperscript{4,14}. As neutron is a neutral particle, so it does not provoke any ionization in the detector directly and so, no tracks are produced because of neutrons directly. However, the interesting part lies in the secondary effects due to neutrons, in the form of recoiling atoms of the detector under neutron impact, leading to the production of charged particles that cause ionization, which produced etchable tracks. The tracks create molecular chain breaking, cross-linking and free radicals etc\textsuperscript{14-18}

The optimization for a specific purpose depends mainly on the standardization of chemical etching parameters such as etching time, temperature, normality of etchant etc. with respect to the rate of energy loss of the track by radiation. We have used optimized etching condition for the present study i.e. 7N NaOH solution at 70$^\circ$C (±) for 9 hr. In this experimental study, we have optimized the parameters. Hence, the effects of neutron radiation on track density and optical characteristics of CR-39 at optimized etching conditions have been investigated.
Our aim is to calculate track density and study optical characteristics at optimized etching conditions. Further, we have calibrated track density to neutron fluences. A linear relationship is observed between absorbance difference and neutron fluence. Such linear relations ensure pertinence of the study for the future high energy accelerator and Radioactive Ion Beam Facilities where CR-39 can be used as a neutron dosimeter.

2 Experimental Details
CR-39 films were commercially procured from Fukuvi Chemical Industry Co., Japan. Films with thickness 200 µm and density ~1.3 g/cm³ were used for this study. All samples of CR-39 polymeric detector were irradiated at room temperature in Inter University Accelerator Center (IUAC), New Delhi, India, with $^{252}$Cf neutron source. One side of these samples was exposed to fission fragments of $^{252}$Cf with yield of $2.16 \times 10^5$ n/s and average neutron energy of 2 MeV. The samples were placed in polyethylene radiator of thickness 1 mm. The use of a radiator in contact with CR-39 can enhance its response; because of the other proton recoils generated within the radiator. The irradiation facility was designed to decrease scattered neutron radiation from all sides. The samples were irradiated for various times to support the various fluences ($\text{yield \times time} \times r^{-2}/4\pi$, where $r$ is the distance of interest).

2.1 Optimum etching conditions
The samples were etched with 7 N NaOH solution contained in a beaker at a temperature of 70°C (±) for 9 hr with constant stirring of magnet inside it, to keep up thermal homogeneity of solution. Etching was terminated when tracks formed were overlapped. After every etching, the detector were washed with deionized water and dried in dry air. The tracks were analyzed and counted using the optical microscope at a magnification of 400X. In this paper, we have calculated the track density after every one hours of etching of the samples. The average track densities as a function of etching time are shown in Fig. 1. It was seen that tracks started appearing from 2nd hour and become viewable at microscope from 1st hour onwards to 6th hour for lower fluence and 4th hour for higher fluence, beyond, which a fall in track density was observed. Further, it was seen from graph that track density increases for higher fluence at lower etching time, while at higher etching time it starts decreasing which is due to removal of shallow tracks and overlap of the tracks due to its track size enlargement. Whereas for lower fluences, it decreases initially, due to the lower linear energy transfer in the detector, indicating smaller damage region. Hence, these films need etching for higher time to visualize the tracks but the trend was the same with respect to etching time.

Apart from this, we recorded the calibration curve to check the sensitivity of detector for neutron detection. The curve between neutron fluence and track density after 9 hr of etching are shown in Fig. 2. The track density increases with the neutron fluence, a linear relationship is observed for the same by drawing the linear fit, which indicates a better sensitivity of detector for neutron detection (dosimetry) as shown in Fig. 2. This fact plays an important role in determining the equivalent dose in the field of neutron dosimetry. Hence, it is certained that such types of studies are very much important for the future high energy accelerators and Radioactive Ion Beam Facilities. To support these results, UV-visible spectroscopy was performed at room temperature for the pristine and neutron irradiated samples. Fig. 3 shows the UV-vis spectrum of CR-39 samples irradiated at difference fluences of neutron as

3 Results and Discussion
3.1 Measurement of track density
In order to obtained track density, the tracks were analyzed and counted under an optical microscope at a magnification of 400X. In this paper, we have calculated the track density after every one hours of etching of the samples. The average track densities as a function of etching time are shown in Fig. 1. It was seen that tracks started appearing from 2nd hour and become viewable at microscope from 1st hour onwards to 6th hour for lower fluence and 4th hour for higher fluence, beyond, which a fall in track density was observed. Further, it was seen from graph that track density increases for higher fluence at lower etching time, while at higher etching time it starts decreasing which is due to removal of shallow tracks and overlap of the tracks due to its track size enlargement. Whereas for lower fluences, it decreases initially, due to the lower linear energy transfer in the detector, indicating smaller damage region. Hence, these films need etching for higher time to visualize the tracks but the trend was the same with respect to etching time.

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well as pristine. Absorption peak shifts towards the visible region i.e., towards higher wavelength for irradiated samples, which show a decrease in band gap after irradiation\textsuperscript{6,8,20}.

The optical band gap energies ($E_g$) of the unirradiated sample was 5.4 eV while for neutron irradiated samples, calculated from the shift in optical absorption edge, are found to be 5.29, 5.26, 5.28 and 5.32 eV, respectively. Although there is a small shift in optical absorption edge at intermediated fluence but still it was seen that optical band gap energy decreases at lower neutron fluence and then increases at still higher fluences\textsuperscript{4}. Thus, it can be inferred that at lower fluence ion entrapment is predominant producing local charge regions leading to subsequent decrease in band gap. On the other hand, increase in band gap at higher neutron fluence may be due to chain scissioning of CR-39 after irradiation.

Figure 4 shows the variation of absorbance difference with neutron fluence at a characteristic wavelength of 234 nm. Absorbance difference for the neutron-irradiated samples with respect to unirradiated sample at 234 nm is the greatest. Therefore, we have chosen this wavelength as the characteristics wavelength. A linear curve obtained between the neutron fluence and the absorbance in the higher fluence i.e. at $7.31 \times 10^6$ n/cm$^2$, $1.77 \times 10^8$ n/cm$^2$ and $2.7 \times 10^8$ n/cm$^2$. It can be seen that there is an almost good agreement between the experimental points and a linear fit in the studied interval. The correlation coefficient was found to be 0.97. These results indicate the possibility of using CR-39 for the estimation of neutron dose or fluence using change in absorbance with respect to neutron fluence.

4 Conclusions

Our preliminary study shows that there is a possibility of the correlation between traditional method of neutron dosimetry using the total numbers of tracks per unit area with dose and the neutron fluence. Further, our experimental results with varying neutron fluence will give an exact picture of neutron dosimetry, optimization with respect to UV spectrophotometry. Calibration characteristics of track density to neutron fluences show a linear relation. The UV-vis spectral study also shows a linear relation between absorbance difference and neutron fluence. The experimental observation suggests the potentiality of CR-39 as neutron dosimeters in future high energy accelerators and Radioactive Ion Beam Facilities.
References