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Effects of high neutron doses and duration of the chemical etching on the optical properties of CR-39

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HIGHLIGHTS

- The variation of optical properties of CR-39 at very high neutron dose is analyzed. Etching process is found to play a crucial role for change in optical properties of neutron-irradiated CR-39.
- The optical absorbance varies linearly at lower dose, at very high dose absorbance saturation occurs. The dose at which saturation absorbance is observed shifts towards lower neutron dose with increase in etching time.
- The rate of decrease in optical band gap with respect to neutron dose is found to be more at higher etching durations.

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ABSTRACT

Effects of the duration of chemical etching on the transmittance, absorbance and optical band gap width of the CR-39 (Polyallyl diglycol carbonate) detectors irradiated to high neutron doses (12.7, 22.1, 36.0 and 43.5 Sv) were studied. The neutrons were produced by bombardment of a thick Be target with 12 MeV protons of different fluences. The unirradiated and neutron-irradiated CR-39 detectors were subjected to a stepwise chemical etching at 1 h intervals. After each step, the transmission spectra of the detectors were recorded in the range from 200 to 900 nm, and the absorbances and optical band gap widths were determined. The effect of the etching on the light transmittance of unirradiated detectors was insignificant, whereas it was very significant in the case of the irradiated detectors. The dependence of the optical absorbance on the neutron dose is linear at short etching periods, but exponential at longer ones. The optical band gap narrows with increasing etching time. It is more significant for the irradiated dosimeters than for the unirradiated ones. The rate of the narrowing of the optical band gap with increasing neutron dose increases with increasing duration of the etching.

1. Introduction

CR-39 is one of the solid polymeric track detectors (SPTDs) widely used at accelerator and reactor facilities for neutron spectrometry and dosimetry (Phillips et al., 2006; Castillo et al., 2013). The chemical composition of the detector, $C_{12}H_{18}O_7$, is close to that of human soft tissue (Zhou et al., 2007), which makes the material suitable for neutron dosimetry. The nuclear tracks produced in this polymer as a result of neutron-induced damage are almost permanent and widely used for radiation detection. The track formation in CR-39 is accompanied by changes in structural, optical, mechanical, electrical and chemical properties of the polymer (Calcagno et al., 1992; Tidjani and Watanabe, 1995; Steckenreiter et al., 1997; Fink et al., 1995; Mishra et al., 2000). These changes are caused by cross-linking, free radical formation, chain scission, irreversible bond cleavages, and other processes (Bouffard et al., 1997; Klaumünzer et al., 1996; Marletta, 1990; Lee, 1999). The changes in various properties of CR-39 induced by electrons, protons, alpha particles and swift heavy ions have been investigated by many authors (Lounis-Mokrani et al., 2003; Singh and Prasher, 2004; Sharma et al., 2007; Nouh and Abutalib, 2010; Kumar et al., 2012; Ghazaly and Hassan, 2014; Saad et al., 2014; Abdul-Kader et al., 2014; Butt et al., 2014). Characteristics of this polymer irradiated with neutrons have also been studied by some authors (Kumar et al., 2010, 2011; Kalsi and Agarwal, 2008).

It is well-known that the number of tracks increases with the neutron fluence and their quantification becomes difficult when

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Table 1
Neutron beam parameters and dosimetry data.

Beam current (nA)	Irradiation time (min)	Total charge on the target (μC)	Neutron dose (Sv)
200	1	122.1	0.82 ± 0.09
200	15	1891.5	12.7 ± 1.4
200	30	3281.7	22.1 ± 2.5
200	45	5339.7	36.0 ± 4.0
200	60	6460.8	43.5 ± 4.8

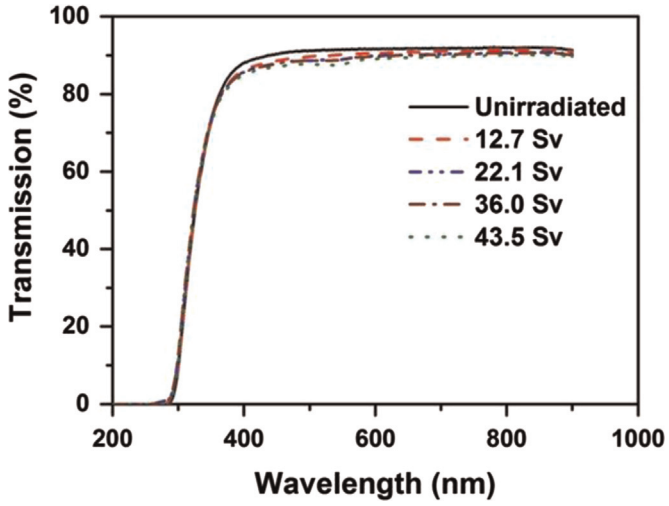

Fig. 1. Transmission spectra of an unirradiated and neutron-irradiated CR-39 samples before etching.

the number becomes large because of the tracks overlapping. Although there are various methods to estimate neutron doses with CR-39, optical absorbance measurements (Sahoo et al., 2014a) are one of the suitable options, especially when the track overlapping is extremely high. Overlapping of a large number of tracks makes it difficult to determine track parameters by isolating each track, no matter what of the available image analysis programs, such as AxioVision, Image J (Abramoff et al., 2004), or autoTRAK (Paul et al., 2012), is used. Sahoo et al. (2014a) and El-Badry et al. (2008) have shown that the optical absorbance varies linearly with the neutron dose, which makes implementing the methodology in the dosimetry of high neutron doses, where the track overlapping is high, advantageous. As reported earlier (Sahoo et al., 2014b), differences between the absorbances of the CR-39 detectors irradiated with neutrons of different fluences become significant only after chemical etching. Therefore, CR-39 detectors need to be etched between irradiation and measurements of the transmission or absorption spectra. Etching procedure needs to be standardized because these optical properties depend on the concentration of the etchant and the duration of the etching. The effect of the etching time on optical properties of the CR-39 polymer irradiated with 2.17 and 3.95 MeV alpha particles has been studied by Eissa (2014). In this work, we attempted to investigate the effect of etching on the optical properties of CR-39 irradiated to high neutron doses. Earlier, Sahoo et al. (2014a) investigated optical and chemical properties of CR-39 irradiated with neutrons up to 5 Sv and found a linear relationship between the optical absorbance and the neutron dose. The prime objective of our work was to verify that the linear relationship holds at higher neutron doses and also to study the effect of the etching time on it. In addition, effects of the etching on the optical band gap width for both direct and indirect transitions were investigated.

2. Methodology

2.1. Irradiation

CR-39 detectors ($10 \times 10 \times 1.5$ mm^3; Intercast, Parma, Italy) were irradiated with neutrons emitted by a thick beryllium target bombarded with 12-MeV protons at a 6-m port of the BARC-TIFR Pelletron-Linac Accelerator Facility (India). The dosimeters were irradiated at a distance of 17.5 mm from the ^{9}Be target along the beam direction. The ^{9}Be target was 6 mm thick, which was sufficient to stop all protons. The total charge accumulated on the ^{9}Be target was measured with a current integrator attached to it, which counted the number of projectiles (protons) hitting the target. The total charge on the target deposited by the total number of projectiles provided the dose by means of a scaling factor, neutron dose per μC. Table 1 lists the irradiation parameters and the neutron doses to which the dosimeters were irradiated. Initially, one detector was irradiated for 1 min to get a significant number of tracks in the material that can be counted with an optical microscope. Then other detectors were irradiated for 15, 30, 45 and 60 min to absorb higher neutron doses.

2.2. Post irradiation procedure

After irradiation, the detector irradiated for 1 min was etched with 6.25 N NaOH at 70 °C for 6 h. Images of the developed tracks were captured with an optical microscope (Axioscope A1, Carl Zeiss, Germany) at a magnification of 200 × . As the number of the projectiles was low (corresponding to the total charge of 122.1 μC), the neutron-induced recoil tracks were well-separated and could be processed with the image analysis software autoTRAK_n developed by Paul et al. (2013). In this case, the measured neutron dose was 0.82 Sv, and it was found that 1 μC charge on this target corresponded to a neutron dose of 6.74 ± 0.75 mSv (Paul et al., 2013, 2014). Neutron doses to the detectors irradiated for 15, 30, 45 and 60 min were estimated using the dose scaling factor 6.74 mSv/μC (Table 1). These irradiated detectors, accompanied with one unirradiated detector, were subjected to stepwise chemical etching at a time interval of 1 h. After each etching step, the ultraviolet–visible transmission spectra of all the detectors were recorded in the range of 200–900 nm with a resolution of 1 nm (scanning spectrophotometer UV 2080 Plus, Analytical Technologies Ltd., India). The procedure was repeated until the transmission values of the irradiated detectors became negligible.

3. Results and discussion

3.1. Effect of the chemical etching duration on the transmission spectra

Fig. 1 shows the transmission spectra of unirradiated and neutron-irradiated CR-39 samples in the range from 200 to 900 nm before etching. There was no significant change in the transmission due to neutron irradiation because the detectors had not been etched. Sahoo et al. (2014a,b) have observed similar effects on CR-39 irradiated to neutrons of different fluences. Fig. 2 represents the transmission spectra of the unirradiated and neutron-irradiated CR-39 samples after 1, 2, 3 and 4 h of etching. A comparison of Fig. 2 with Fig. 1 shows that the transmission changes at each etching step.

The transmission in the visible region was found to decrease