Study of Neutron Induced Modification on Optical Band Gap of CR-39 Polymeric Detector

VIJAY KUMAR[A], R.G. SONKAWADE[B*], A.S. DHALIWAL[A] and ROHIT MEHRA[C]
[a]*Department of Physics, Sant Longowal Institute of Engineering and Technology Longowal Distt. Sangrur, Punjab-148106, India*
**E-mail:* rgs22@rediffmail.com, *vijays174@yahoo.com*

Neutron- irradiation effects on optical absorption of solid-state nuclear track detector, CR-39 was studied for the various fluences ranging from 2.38x106 to 2.7x108 n/cm^2 by ultraviolet-visible (UV-Vis) spectroscopy. The optical absorption spectra in the wavelength range of 200-700nm were recorded for the pristine and the neutron irradiated samples in the above fluence range. The UV-visible spectra shows a shift in the absorbance edge towards the higher wavelength, which can be correlated to the transition involved in the CR-39 polymer and variation in the band gaps (Eg) using Tauc's expression. Both the indirect and direct band gap in pristine and neutron irradiated CR-39 have been studied, it is interesting to note that the values of indirect band gap found to be lower then the corresponding values of the direct band gap. Indirect and direct bandgap of pristine and exposed CR-39 was found increasing with increasing fluences. However values of the indirect band gap found lower than that of direct band gap. A decrease in the optical energy gap with increasing fluence can be discussed on the basis of neutron-irradiation defects in CR-39 and the numbers of carbon atoms in a cluster were determined from UV-visible spectra of the neutron irradiated samples, using Tauc's expression.

Key Words: Neutron, UV-Visible, fluence, bandgap.

INTRODUCTION

Polymers have proven potential in all fields during the last few decades. Different studies of the effect of irradiation on polymers, reveals a variety of modifications of the physical and chemical properties such as optical, electrical, mechanical etc. of polymeric materials depending on the irradiation fleunce and the energy of the radiation used [1-5]. Interaction of radiation or highly energetic charged ion strikes a polymer target; it loses most of its energy in exciting or ionizing the atoms along its trajectory. Target ionization causes bond cleavage (scission), cross-linking, degradation, emission of atoms and molecules, creation

[b]*Inter University Accelerator Center, Aruna Asaf Ali Marg, New Delhi-110067*
[c]*Department of Physics Dr. B.R. Ambedkar NIT, Jalandhar (Punjab)*

of the free radicals and formation of saturated and unsaturated groups with stimulated evolution of gases [6]. These modifications have strong influences on the optical properties of polymers. Most of the previous work has been using electrons, protons, IR, X-rays and gamma rays irradiation etc. [7-10]. A very few studies have been carried out on neuron induced effects, to modify its property to be suitable for radiation dosimetry applications. The neutrons themselves do not cause ionization directly [11].

In the present work we have chosen important plastic CR-39 films, which is an amorphous polymer. The aim of present investigation is to study the effect of neutron irradiation on the UV-visible spectra of CR-39 polymeric detector before and after irradiation. From an idea of the direct and indirect optical band gaps, we have tried to analyze the relation between radiation doses against band gap. Optical properties of the polymers have been characterized by various authors [1, 4, 7-8] by correlating the change in optical properties with the mode of energy deposition. Also we investigate the Urbach's energy and the number of carbon atoms in a cluster using Tauc's expression [12].

EXPERIMENTAL PROCEDURE

The present study was performed by using a commercially purchased CR-39 sheets with approximate thickness 500µm having density 1.3 g/cm^3 of size 2cm x 2cm produced by Fukuvi Chemical Industry Co., (Japan).

All the samples of CR-39 were irradiated at room temperature in the Hot Lab, Inter University Accelerator Center, New Delhi using Am-Be neutron source of various fluences. The samples were placed in polyethylene radiator of thickness 1mm. The irradiation facility is designed so that the scatter neutron radiations from all sides are very minimal. The pristine and neutron irradiated CR-39 polymeric samples was subjected to spectral studies in the ultraviolet and visible region [7, 8] performed to observe the variation in optical band gaps and the number of carbon atoms in a cluster using Tauc's expression [3, 4] also investigated.

RESULTS AND DISCUSSION

The optical absorption spectra of pristine and neutron were recorded using a UV-visible spectrophotometer in the wavelength range 200-700 nm at room temperature are shown fig. 1. The region of optical absorption extends to higher wavelength for higher irradiation fluences [8]. It proves that neutron radiation damage effects are complex and non-linear in the most sensitive SSNTDs. The shift may be produced by the creation of free radicals, ions and hot molecules due to irradiation. One of the possibilities behind this behavior is interpreted as the formation of extended system of conjugated bonds or in other words the formation of carbon cluster.

The shift in the absorption edge is correlated with optical energy gap, E_g by Tauc's relation. The irregularities in the band gap level of the polymers is defined as Urbach's energy [4] and is calculated from the inverse of the slope of the linear part of the curve between logarithm of the absorption coefficient $\alpha(\upsilon)$ and energy $h\upsilon$ (eV), where α being the absorption coefficient. The calculated values are reported in table 1.

The Urbach's energy sharply decreasing with increasing neutron fluence. This sharp decrease indicates the irregularization of the band gap energy level; furthermore this is due to amorphous nature of CR-39 polymer. The decrease in band gap energy attributes to the decrease in the resistivity of CR-39. This means that there is change in the structural characteristics of CR-39 as a result of neutron irradiation. It may accounts for the scission of the polymer chain and formation of free radicals. It is observed that there

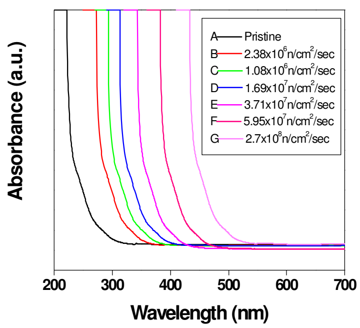

Fig 1: UV-Visible Spectra of the pristine and neutron irradiated CR-39.

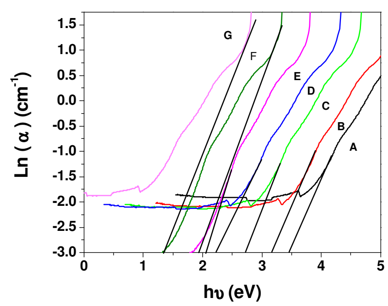

Fig 2: The dependence of the natural logarithm of α on photon energy for pristine and neutron irradiated CR-39 polymer.

Table 1: The variation of the band gap energy and Urbach's energy in the pristine and neutron irradiated CR-39, along with the number of carbon atoms. (N) per conjugated length.

Neutron Fluence n/cm^2	Band gap energy (eV)		Urbach's energy (eV)	No. of carbon atoms (N)	
	Indirect	Direct		Indirect	Direct
Pristine	3.42	4.90	0.29	5	4
$2.38x10^6$	3.22	4.62	0.74	6	4
$1.08x10^7$	2.7	4.02	0.51	7	5
$1.69x10^7$	2.6	3.67	0.48	7	5
$3.71x10^6$	2.10	3.17	0.45	9	6
$5.94x10^7$	1.60	2.68	0.36	11	7
$2.7x10^8$	1.35	2.30	0.31	13	8

is a decreasing trend of energy gap with increasing neutron fluence. This is due to the carbon enriched domains created in polymers during irradiation. In the studied range of wavelength the absorption bands are associated with π-π* electronic transition. The excitation of π electron require smaller energy and hence, transition of this type occurs at longer wavelength. In the high absorption region where absorption is associated with interband transition, the absorption coefficient α(ν) was given in the quadratic form by Tauc *et al.* and discussed more general form by Davis and Mott [8].

The values of the band gaps in any polymeric material is obtained by plotting $(\alpha h\nu)^{1/2}$ and $(\alpha h\nu)^2$ as a function of the photon energy (hν), respectively and extrapolating linear portion of the plot to hν axis, is used to define the so called optical

band gap in polymers i.e. direct and indirect have been determined for pristine and neutron irradiated CR-39 polymer and are shown in figures 3 and 4. The values of the indirect and direct band gap (E_g) for pristine and neutron irradiated samples are reported

in table 1.The simultaneously existence of indirect and direct band gaps in CR-39 polymer has rarely been reported [13], although such as coexistence of direct and indirect band gaps has been observed in some other materials [8]. M F Zaki (2008) also found similar pattern with gamma induced modification on optical band gap of CR-39. The number of carbon atoms (N) in the cluster per conjugation length [4] can be reported in table 1.

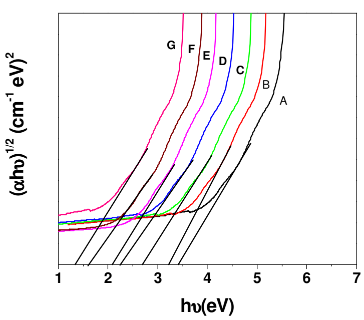

Fig 3: The dependence of $(\alpha h\nu)^{1/2}$ on the photon energy (hν) for unexposed and exposed with Am-Be neutron source of different fluences

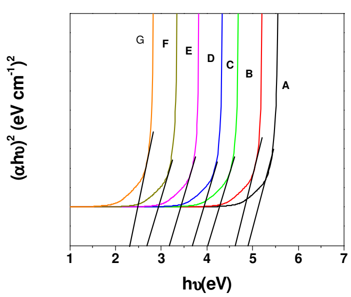

Fig 4: The dependence of $(\alpha h\nu)^2$ on the photon energy (hν) for unexposed and exposed with Am-Be neutron source of different fluences

Conclusions

Significant changes are observed in optical response of the CR-39 polymer after irradiation. From this experimental study of pristine and neutron irradiated CR-39, the values of the optical band gap (E_g) and Urbach's energy (E_u) were determined from the optical absorption spectra. It is concluded that the values of the indirect band gap are lower then the corresponding values of the direct band gap in pristine and neutron irradiated CR-39 polymers. Also the band gap (E_g) decreases with the increases neutron fluences, due to the degradation of CR-39 and the formation of defects and cluster in the material, while the cluster size increase with the increase in the fluence for both the cases.

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