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Gamma Irradiation Effect on Nonlinear characterization of Congo film for application in optical limiting

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***Abstract*— The influence of gamma irradiation on the nonlinear optical properties of Congo dye films prepared by casting method has been reported. The single beam Z-scan technique was used to determine the nonlinear optical properties of the Congo dye doped polymer film. The experiments were performed using cw SDL laser with a wavelength of 532 nm. This material exhibits a negative optical nonlinearity. Optical limiting characteristics of the polymer film were studied. The result reveals that Congo dye doped polymer film can be a promising material for optical limiting applications.**

***Index Terms*— gamma irradiation, Nonlinear optics, Laser, Z-scan technique, dye, optical limiting,**

I. INTRODUCTION

Demand for polymers having improved surface and bulk properties is continuously on the rise due to their use for various scientific and technological applications [1–2]. Polymer films and Irradiation of polymers has established itself as one of the most acceptable approach to alter polymer properties significantly [3–16]. As advanced technology keeps on developing every day, the demand for polymers having improved properties is continuously on the rise due to their use of various, scientific and technological applications [17]. Irradiation plays a prominent role in modification of properties of polymers significantly. Ionizing radiation passing through matter, deposit energy in the material and cause irreversible changes in the macromolecular structure of the target material. The most prominent effects of radiation involve a change of phase in the absorbing material due to the scissions or cross linking formation. At the same time, it is also likely possible to observe the creation of chemical bonds between different molecules (intermolecular cross linking) in the main chain.
The mechanism of gamma rays interaction with nonlinear materials fundamentally happens with a method for electronic ionization, electronic excitation and principally atomic separation of the orbital electrons [18]. So, this study report the results on the effect of gamma irradiation on optical nonlinearity of doped PMMA films with Congo dye. The experiments performed using the Z-scan technique to measure the magnitude and sign of the optical nonlinearity for Congo dye doped polymethylmethacrylate (PMMA) film. The optical limiting behavior of an irradiation and unirradiation polymer films has been studied.

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II. EXPERIMENT

The chemical structure of Congo dye are in Fig.1. The solution sample of the Congo dye was dissolved in chloroform. The concentration of the dye solution is 0.5 mM. To prepare the solid films, PMMA was selected as the host material because it is hard, rigid and has a glass transition temperature of 125 ^{o}C. It also exhibits good linear optical transmittance, optical stability, thermal stability and moreover better compatibility with organics [19,20]. A known quantity of PMMA and Congo dye were dissolved in chloroform separately, the concentration of the for un-irradiated and irradiated films in 42 GKy gamma dose Congo dye in chloroform is 0.5 mM, later both solutions were mixed and stirred for 2 hr using a magnetic stirrer. The ratio of PMMA solution and dye solution is 1:1. The film was prepared on a clean glass slide by the casting method and dried at 38 ^{o}C for 24 hrs. The film sample has a good purity and uniform thickness. The thickness of the pristine and irradiated film was measured using digital micrometer and is found to be 7μm.

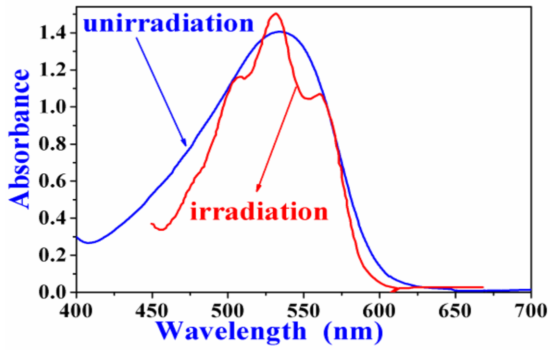

Fig. 2. UV-VIS absorption spectrum of an irradiation and unirradiation polymer films.

The UV-VIS (Ultraviolet-Visible) absorption spectra for un-irradiated and irradiated Congo dye films are recorded using Cecil Reflected-Scan CE 3055 reflectance spectrometer. The optical absorption for gamma irradiated Congo dye doped PMMA thin film with 0.5 mM concentration shows an absorption peaks at 507, 531 and 560 nm, respectively, as can seen in Fig.2. The spectrum of the optical absorption was computed from the absorbance data. The absorption coefficient (α) has been obtained directly from the absorbance against wavelength curves using the relation [21,22]

$$\alpha = 2.303A/d \quad (1)$$

where d is the sample thickness, A is the absorbance. The values of absorption coefficient (α) for 0.5 mM concentration of an irradiation and unirradiation Congo polymer film are listed in table 1.

The Z-scan technique was applied for the measurements of nonlinear optical characteristics of investigated samples. We used a SDL laser beam with 532 nm wavelength and power of 15 mw , which was focused by (+5 cm) focal length lens. The laser beam waist ω_0 at the focus measures 27.04µm and the Rayleigh length measured Z_R= 4.31mm. A 7µm thickness of an irradiation and unirradiation optical polymer films are translated across the region along the axial direction of the laser beam propagation. The transmission of the beam through an aperture placed in the far field was measured using a photo detector fed to the power meter. For the open aperture Z-scan, a lens was used to collect the entire laser beam transmitted through the sample replaced the aperture.

The limiting effect of the samples was studied using the same laser in the z-scan technique. The experimental set-up for the demonstration of Z-scan and optical limiting is shown in Fig. 3. A Congo dye doped PMMA thin film of an irradiation and unirradiation are kept at the position where the transmitted intensity shows a valley in closed aperture Z-scan curve. A variable beam splitter (VBS) was used to vary the input power. The input laser intensity is varied systematically and the corresponding output intensity values are measured by the photo detector.

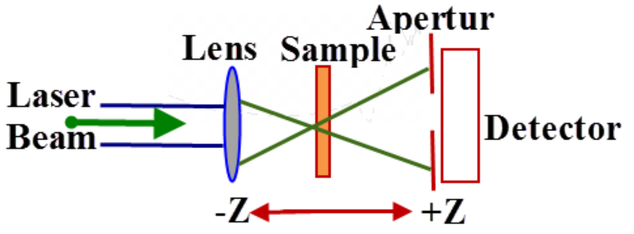

Fig. 3. Experimental set-up for Z-scan and limiting effect.

III. Results and Discussions

Fig. 4 shows the closed aperture Z-scan data for 0.5 mM concentration of an irradiation and unirradiation polymer film at incident intensity I_0=1.306 kW/cm^2 of the laser beam at focus z =0. The scan of both the samples have peak-valley configuration, corresponding to a negative nonlinear refraction index i.e. self-defocusing occur. The defocusing effect is attributed to a thermal nonlinearity resulting from the absorption of a tightly focused beam traversing through the absorbing dye medium produces spatial distribution of the temperature in the sample and, consequently, a spatial variation of the refractive index that acts as thermal lens resulting in phase distortion of the propagating beam.

Let $\Delta Ø_0$ be the on-axis phase shift at the focus which is related to the difference in the peak and valley transmission $\Delta T_{p\text{-}v}$ as [23]:

$$\Delta T_{p\text{-}v} = 0.406\,(1\text{-}S)^{0.25}\,\Delta Ø_0 \qquad (2)$$

Where $S=1\text{-}\exp(-2r_0^2/\omega_0^2)$ [24] is the aperture linear transmittance with r_0 denoting the aperture radius and ω_0 denoting the beam radius at the aperture in the linear regime. Then, the nonlinear refractive index, n_2, is given by [25]

$$n_2 = \Delta Ø_0\, \lambda\, /2\pi I_0 L_{eff} \qquad (3)$$

Where λ is the laser wavelength, L_{eff} =[1-exp(-αd)] /α [26] is the effective thickness of the samples.

Fig. 5 shows the measured Z-scan data for open aperture set-up for the irradiation and unirradiation polymer films. The typical Z-scan data with fully open aperture is insensitive to nonlinear refraction, therefore the data is expected to be symmetric with respect to the focus, but absorption in the sample enhances the valley and decreases the peak in the closed aperture Z-scan curve and results in distortions in the symmetric of the Z-scan curve about Z=0 [27-30].

The nonlinear absorption coefficient β can be estimated from the open aperture Z-scan data [31-33]

$$\beta = 2\sqrt{2}\Delta T \big/ I_0\, L_{eff} \qquad (4)$$

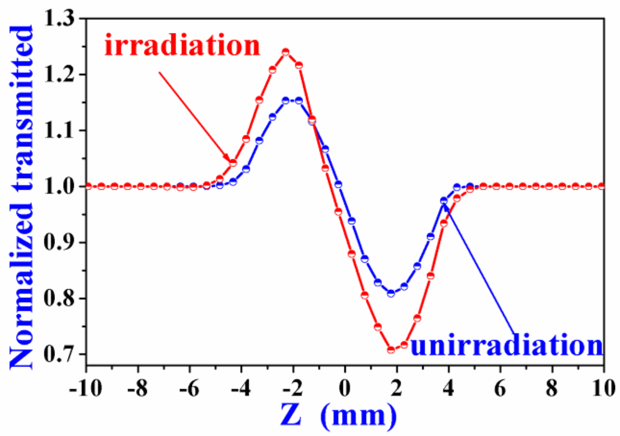

Fig.4. Open data for polymer film

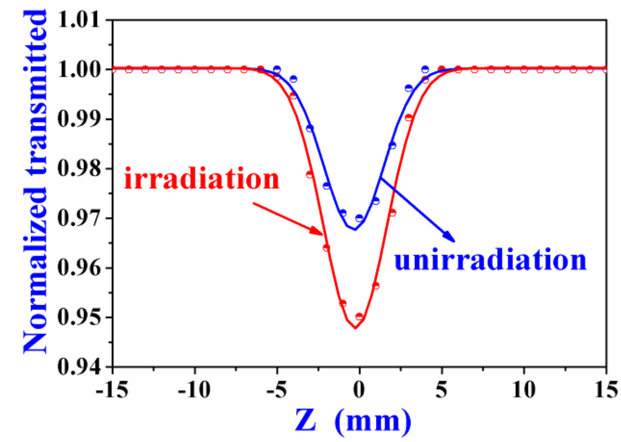

Fig. 5. Closed data for polymer film

In general, the measurements of the normalized transmittance versus the sample position for the cases of the closed and open aperture allow for the determination of the nonlinear refractive index, n_2, and the nonlinear absorption coefficient β, since the closed aperture transmittance is effected by the nonlinear refraction and absorption. The determination of n_2 is less straight forward from the closed aperture scans. It is necessary to separate the effect of the nonlinear refraction from that of the nonlinear absorption. A method [34] to obtain a purely effective n_2 is to divide the closed aperture transmittance by the corresponding open aperture scans. The data obtained in this way purely reflects the effects of the nonlinear refraction. The ratio of Figs 4 and 5 scans is shown in Fig. 6. The values of the nonlinear refractive index and nonlinear absorption for the irradiation and unirradiation polymer films at 0.5 mM concentration are given in Table 1. The value of n_2 in irradiation dye doped film is found to have large value than in the case of unirradiation, this may be attributed to two reasons, firstly absorption coefficient (α) for the irradiation dye doped film is large than in the case of unirradiation polymer film[35], secondly the Irradiation in polymers destroys the initial structure by way of cross linking, free radical formation, irreversible bond cleavages, etc., resulting in the fragmentation of molecules and formation of saturated and unsaturated groups. All these processes introduce so-called defects inside the material that are responsible for change in the optical, electrical, mechanical and chemical properties of the material [36,37].

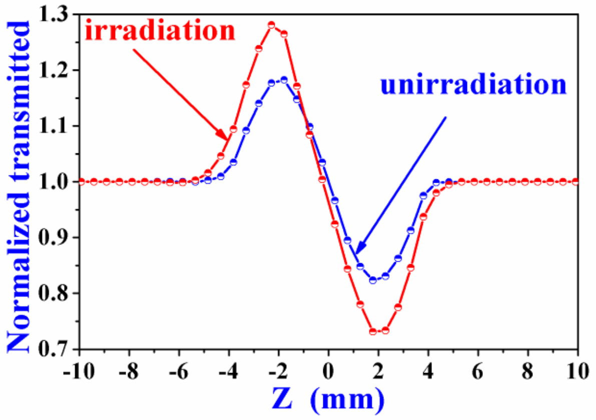

Fig. 6. Pure data for an irradiation and unirradiation polymer film.

Table 1: Linear and nonlinear optical parameters for an irradiation and unirradiation films .

Sample	$\alpha(cm^{-1})$	$n_2(cm^2/w)$ $\times10^{-6}$	$\beta(cm/w)$ $\times10^{-2}$
irradiation	4619.19	29.46	32.20
unirradiation	4951.71	48.19	55.69

The optical limiting curve for the irradiation and unirradiation polymer films at 0.5 mM concentration is shown in Fig.7. The output power rises initially with an increase in input power for both samples, but after a certain threshold value the samples start defocusing the beam, resulting in a greater part of the beam cross-section being cut off by the aperture [38-40]. Thus the transmittance recorded by the photo detector remained reasonably constant showing a plateau region. The limiting behavior observed in both samples is attributed mainly to nonlinear refraction. Since the samples were pumped with cw laser beam the arising nonlinearities are predominantly thermal in nature. The experiment was repeated for the pure solvent chloroform to account for its contribution, but no limiting was observed.

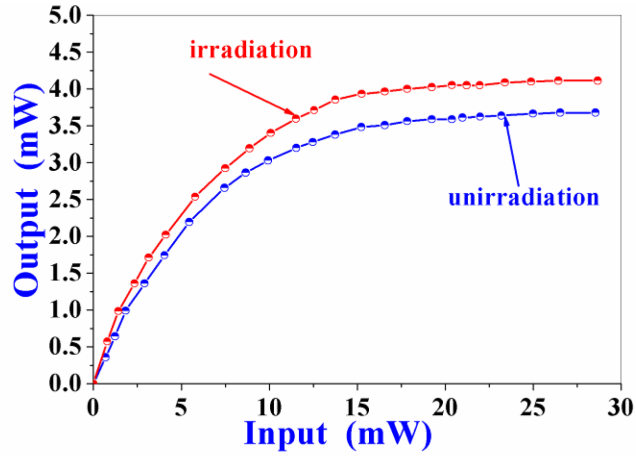

Fig. 7. Optical limiting effects of an irradiation and unirradiation polymer film.

IV. Conclusion

UV-VIS Spectrophotometric studies of non-irradiated and irradiated polymer films at 0.5 mM concentration reveals the large nonlinear optical parameters. We have measured the nonlinear refraction, n_2, and the nonlinear absorption coefficient, β, for irradiation and unirradiation polymer films using the Z-scan technique with 532 nm of SDL laser. The results of Z-scan indicate that the samples have a large optical nonlinearity compared with other optical materials: n_2 of the samples is the order of 10^{-6} cm^2/W at 532 nm. The sample show good optical limiting behavior at 532 nm. All these experimental results show that the sample is a promising material for applications in nonlinear optical devices.

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