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GAMMA IRRADIATION EFFECT ON THE NONLINEAR REFRACTIVE INDEX AND OPTICAL LIMITING BEHAVIOR OF PYRONINE Y DYE SOLUTION

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ABSTRACT

In this work, the influence of gamma irradiation on the structural, third-order nonlinearities and optical limiting properties of Pyronine Y ($C_{17}H_{10}ClN_2O$) solution has been reported. The samples have been investigated via a typical absorbance spectra, and characterized in the wavelength range 300–700 nm. The nonlinear optical coefficient (nonlinear refractive index and nonlinear absorption) showed clear changes with different irradiation time. Also, the optical limiting property of the sample has been investigated using 532 nm continuous wave (cw) laser (SDL-532-100T). At the same time, the optical limiting behavior was found to increase with increasing dose. Gamma ray irradiation was performed by using ^{137}Cs source with an exposure rate of 0.56 Gy/min. This study also suggests that the gamma irradiation can be considered as a tool for the enhancement of the device properties amongst the other applications.

keywords: *gamma irradiation, pyronine y, nonlinear refractive index, optical limiting, optical device.*

1 INTRODUCTION

In recent years, owing to a number of practical applications in the field of micro-electronics and optoelectronics, a great deal of attention has been paid to the study of the properties of various materials. Recent years have witnessed a great demand for the organic materials for non-linear optical (NLO) applications because of their usage in devices such as second harmonic generators, electro-optic modulators, frequency conversion, etc [1,2]. The NLO phenomena take place when the optical properties of molecules change in the presence of strong external electric fields, i.e., high energy laser beams. They exhibit other additional advantages such as their broadband spectral responses, high thermal and chemical stability, along with their simple structures, low cost and ease of preparation in solution and thin polymer film [3,4]. Due to these advantages, the organic nonlinear materials have been extensively investigated for useful optical applications, such as all-optical signal processing, optical information storage, optical switching, and optical power limiting [5-8], which require large nonlinearities and fast response time. Dyes are a class of organic materials. They are very interesting materials for studying the optical nonlinear effects because they exhibit large optical nonlinearities, fast response time, and strong absorption in the visible spectral region [9]. In addition, they are characterized by their flexibility and thermal and chemical stability in dye-doped polymer solid films. These very important advantages make the dyes suitable candidate for nonlinear optical investigations. Optical power limiting behavior has received significant attention due to its functional and important applications. A wide range of materials with various nonlinear optical mechanisms contributing for optical limiting have been investigated [10-15]. The devices that based on this nonlinear behavior can be used for the protection of human eyes and sensitive optical equipments, such as sensors, from intense laser exposure by controlling the intensity of incident laser beam [16]. In this nonlinear optical process, the transmitted laser intensity (or power) of a material increases linearly with increased input laser intensity at low input intensities, but above certain input intensity(threshold intensity), the transmitted intensity becomes relatively constant. In this paper, the purpose of this paper is to report some of our results on the effect of different gamma irradiation dose on nonlinear optical coefficient of organic dye, we have chosen Pyronine Y dye for the study. We have reported the spectral characteristics of Pyronine Y dye in methanol and the third-order nonlinear optical properties of chosen dye in methanol measured by z-scan technique. Also the optical limiting behavior of the Pyronine Y dye have been studied for different gamma irradiation.

2 EXPERIMENTAL DETAILS

The chemical structure of Pyronine Y dye is ($C_{17}H_{10}ClN_2O$). Pyronine Y dye powder with concentration of 0.02 mM was irradiated at different Gamma-ray doses. Gamma ray irradiation was performed by using ^{137}Cs source with an exposure rate of 0.56 Gy/min. For each dose the film samples were placed simultaneously at the centre of the chamber surrounded for radiation equilibrium purposes. In the present method the sample was prepared as follows:

(0.01253 mol, 3g) was irradiated at different Gamma-rays, 21 KGy, 42 KGy and 62 KGy, respectively. After one month (21 KGy) 0.0121gm of an irradiation Pyronine Y dye powder was dissolved in 50 ml of concentrated methanol to get 0.08 mM as a bulk solution. The solution was stirred at room temperature for 50 min, and then the solution was filtered through a 0.2 mm syringe filter. 1.25 ml of bulk solution was dissolved in about 3.75ml of methanol solvent to get 0.02 mM. The other irradiated sample was also prepared in a similar manner. The codes of samples 0.02 mM unirradiation, 0.02 mM irradiated with 21 KGy, 0.02 mM irradiated with 42 KGy and 0.02 mM irradiated with 62 KGy separately is pristine, 21 KGy, 42 KGy, and 62 KGy respectively.

The absorption spectra of the unirradiated and irradiated Pyronine Y dye dosimeters were measured using UV-Visible spectrometer (Cecil Reflectascan CE 3055) in the wavelength range 300-700 nm. Due to complex molecular structures of the dye, the super position of electronic and vibrational transitions produce typical overlapping continuous UV-Visible absorption bands. The absorption spectra produced two absorption bands in the visible region. These visible bands correspond to the excitation of outer electrons, which provide information on the electronic transitions of the molecules in the samples. They are attributed to the π-π^* transitions and to the presence of ions in the Pyronine Y dye [17,18]. UV absorption is mainly due to electron (or anion) transitions from the top of the valence band to the bottom of the conduction band [19]. The absorbance increases exponently with dose for 508 nm band and decreases exponently with dose for 550 nm band. As the dose increases, more chlorine ions break from carbon of Pyronine Y and radiation-induced H and OH free radicals are generated from the hydrolysis of solvent molecules. The effects of radiation on optical properties of unirradiated and irradiated Pyronine Y dye were measured using UV-visible spectrometer. The absorption spectra are shown in Fig. 1.

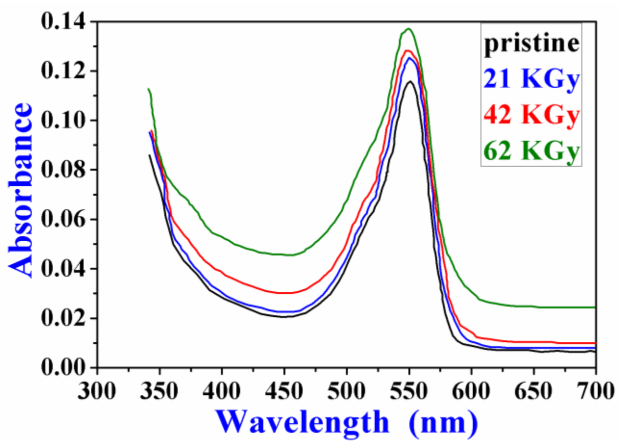

Figure1: Spectra of absorbance of Pyronine Y dye solution for different gamma irradiated.

2.1 Z-scan measurements

In Fig. 2, the schematic depicts the Z-scan experimental setup which is used to measure the non-linear optical properties. The experiment was performed by 532 nm diode pumped cw Laser (SDL-532-100T). A 5 cm focal length lens was used to focus the laser beam on the sample. The output transmittance was measured on a photodetector using an aperture of 2.5 mm. The samples were translated along the z

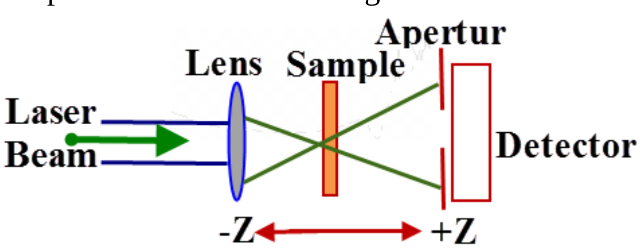

Figure 2. Schematic diagram of experimental arrangement for the Z-scan measurement.

direction from where the transmitted and initial intensity was measured. The Z-scan experiment involves two different setups, where, the closed aperture method is used to determine the non-linear refractive index (n_2) which is directly proportional to the real part of third order susceptibility Re[χ^3] and the open aperture method which is used to determine the non-linear absorption coefficient and is directly proportional to the imaginary part of third order susceptibility Im[χ^3].

The difference between the peak and valley transmission (ΔT_{p-v}) in terms of the axis Phase shift at the focus is given by [20,22]:

$$\Delta T_{P-V} = 0.406(1-S)^{0.25}\Delta\phi_{\circ} \qquad (1)$$

where $S = 1-\exp(-r_a^2/\omega_a^2)$ is the aperture linear transmittance (=0.32) with r_a denoting the aperture radius and ω_a denoting the beam radius at the aperture in the linear regime. $\Delta\phi_{\circ}$, the on-axis phase shift is related to the third-order nonlinear refractive index by [23,24]:

$$\Delta\phi_{\circ} = kn_2L_{eff}I_{\circ} \qquad (2)$$

where L_{eff} is the effective thickness of the sample ($L_{eff} = (1-\exp(-\alpha L))/\alpha$),$\alpha$ the linear absorption coefficient, L is the thickness of the sample, the laser beam was focused by a lens of +50 mm focal length, and the solutions under investigation were contained in a 1mm thick quartz cell which was mounted on a step motor and translated along the beam direction. The transmission of the beam was measured using a photo detector fed to the digital power meter (Field Max II-To+OP-2 Vis Sensor).

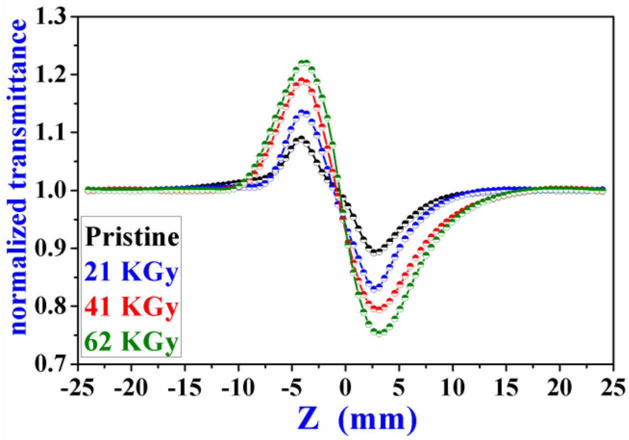

Figure 3: Pure Z-scan data and theoretical fitting curves of Pyronine Y day solution for different gamma irradiation.

Fig. 3 shows the pure Z-scan data for samples at incident intensity I_0 =1.045 kW/cm^2.The scans of all the samples have peak-valley configuration, corresponding to a negative nonlinear refraction index i.e. self-defocusing occur. In the previous work we shown that, in the case of solution and cw laser used in Z-scan technique, the defocusing effect is attributed to a thermal nonlinearity resulting from absorption of radiation. Fig. 4 shows the measured Z-scan data for open aperture setup of Pyronine Y dye sample for un-irradiated (pristine) and irradiated solutions in different dose.

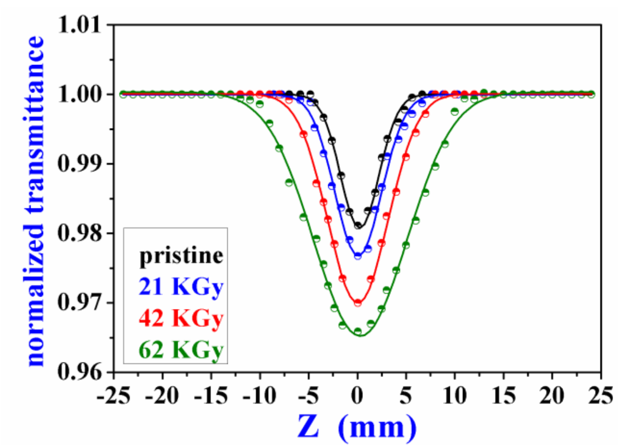

Figure 4: Open-aperture data and theoretical fitting curves for different gamma irradiation.

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 the absorption in the sample enhances the valley and decreases the peak in the closed aperture Z-scan curve and results in distortions in the symmetry of the Z-scan curve about Z=0.

The nonlinear absorption coefficient β can be estimated from the open aperture Z-scan data [25-27];

$$\beta = 2\sqrt{2}\Delta T / I_{\circ} L_{eff} \qquad (3)$$

where ΔT is one valley transmittances at the open aperture Z-scan curve, $I_o = 2P/\pi\omega_o^2$ is the intensity of the laser beam at focus and $\omega_\circ$ is the laser beam radius at the focus. The nonlinear absorption coefficient β (cm/W) and nonlinear refractive index n_2 (cm^2/W) for solution of Pyronine Y dye sample for unirradiated (pristine) and irradiated solutions in different dose are calculated from the open and closed aperture normalized transmittance and their values are listed in Table 1.

Table 1 Nonlinear optical parameters of sample with different gamma irradiation

Sample	β x10^{-3} (cm/W)	Δn x10^{-4}	n_2 x10^{-7} (cm^2/W)
Pristine	5.5	0.69	0.66
21 KGy	6.9	10.9	10.4
42 KGy	9.06	14.3	13.7
62 KGy	10.3	14.9	14.2

The value of n_2 in a Pyronine Y dye sample for gamma irradiation in different dose are found to have large value than in the case of un-irradiated (pristine) solution. Irradiation of the Pyronine Y dye, $C_{17}H_{10}ClN_2O$, causing the loss of Cl and induce the detachment of ions and unsaturated groups (−C = C−). The values of n_2 and β are related to the un-irradiated and irradiated of the Pyronine Y dye solutions. The value of nonlinear refractive index increases when the gamma irradiation increases as shown in Fig. 5. This may attributed to the fact that the Irradiation in Pyronine Y dye 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 nonlinear optical, electrical, mechanical and chemical properties of the material [28]. The laser heating is produced while the laser beam passing through the sample medium. This process induces temperature and density gradients that change the refractive index of the dye medium and consequently thermal focusing effect arising. Also one can show from Fig. 5 the effects of each sample on nonlinear refractive index in percentage.

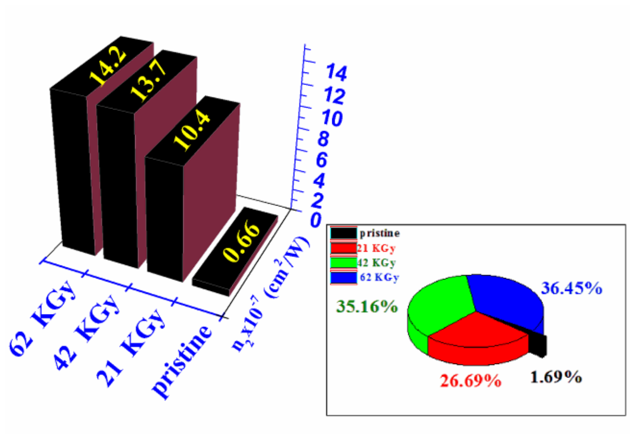

Figure 5. Histogram curve of the (n_2) as a function of gamma irradiation. Inset shows the effect of each sample in percentage.

2.2 Optical limiting

For optical power limiting measurements, based on the nonlinear refraction (self-defocusing), the sample was located at the position where the valley falls in the closed-aperture and the input power of the laser beam was varied. The input powers and the corresponding output powers of the laser beam were recorded by the power meters. Fig. 6 shows the optical limiting behavior of Pyronine Y dye. The laser output power was plotted as a function of the laser input power, varying over the range 0-45 mW. It is clearly seen that the output power initially varies linearly with increasing the input power (at low powers), but the transmitted output power starts to deviate from the linearity at high input powers. With still increasing the input power, the output power reaches a plateau region at certain value for the input power and the output power relatively remains constant (saturated) with further increase in the input power. The value of the input power at which the output power starts to saturate is known as the power limiting threshold which defined as the input power at which the sample transmittance falls to half of its linear transmittance [29]. The values of the power limiting threshold and limiting amplitude for the un-irradiated Pyronine Y dye solution and irradiated Pyronine Y dye solution were estimated, Table 2 summarizes these values. It is evident that the optical power limiting effect depends on the gamma irradiation dose of the Pyronine Y dye and increases with increasing gamma irradiation dose(time), while the power limiting threshold decreases with increasing gamma irradiation dose. At high Pyronine Y dye irradiation, the output power reached a plateau at low input power and the dye medium exhibits strong optical power limiting effect.

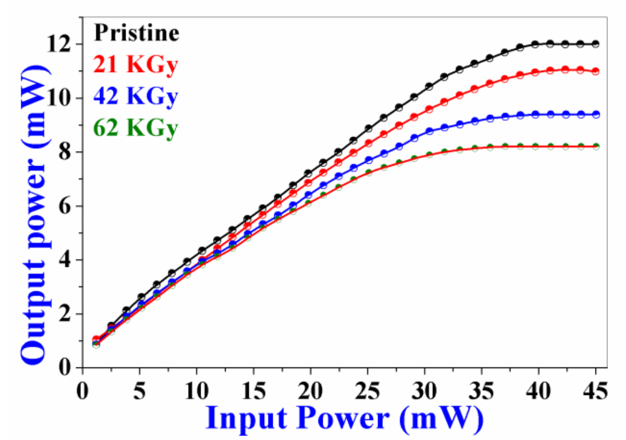

Figure 6: Optical limiting with different gamma irradiation of Pyronine Y dye solution.

Table 2 Optical limiting parameters at different levels of irradiation

Sample	limiting threshold (mW)	limiting amplitude (mW)
Pristine	16.57	9.6
21 KGy	13.23	8.0
42 KGy	12.06	6.7
62 KGy	11.41	5.8

The saturated output value at which limiting occurs for the samples solution are shown in Fig.7, for pristine and different gamma irradiation dose (time). It can be seen from Fig.7, that the saturated output value decreases with an increasing concentration.

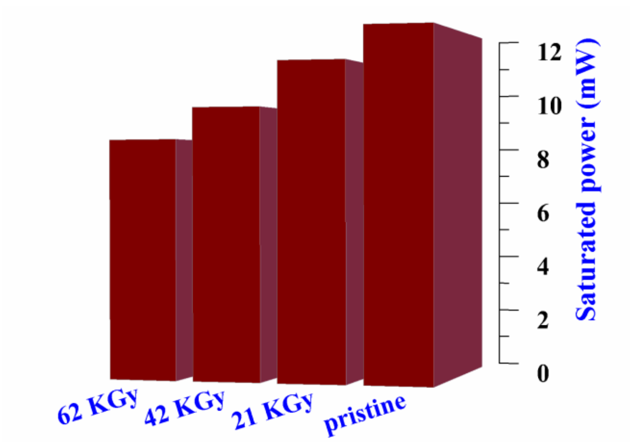

Figure7. Histogram curve of the gamma irradiation dose dependence of saturated output value of samples.

Conclusion

We carried out the measurements of third-order nonlinear optical properties in Pyronine Y dye sample for un-irradiated (pristine) and irradiated in different dose at the wavelength of 532 nm using Z-scan technique. The origin of optical nonlinearity observed in the CW regime is attributed to the thermal variation of local refractive index in the medium. The results indicate that the sample exhibits self-defocusing nonlinearities. It is found that the nonlinear refractive index and nonlinear absorption coefficient for irradiation samples are larger than that for pristine. The variation in the output power was studied as a function of input power for four different samples, one is a pristine sample and three samples at different levels of irradiation with gamma rays, and the influence of all these samples on the threshold limit and output clamping power was analyzed. The limiting threshold can be improved by a proper choice of design parameters such as the geometry of the configuration and the irradiation of the sample, based on the actual requirements of the sensor such as the dynamic range, sensitivity and the field of view. The mechanism of optical limiting in the low power regime is found to be predominantly of a thermal origin. All these experimental results show that this dye is a promising material for applications in various optical limiting devices.

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تأثير أشعة كاما على معامل الانكسار اللاخطي وسلوك الحد البصري لصبغة البيرونين y

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الخلاصة :

في هذا العمل تم الابلاغ عن تأثير التشعيع بأشعة كاما على التركيب والرتبه الثالثة اللاخطية وخواص المحدد البصري لمحلول صبغة البيرونين ($C_{17}H_{16}ClN_2O$) .ولقد تم فحص النماذج من خلال طيف الامتصاص النموذجي لمدى الطول الموجي 300−700 nm. أظهرت المعاملات اللاخطية (معامل الانكسار اللاخطي ومعامل الامتصاص اللاخطي) تغيرات واضحة بتغير زمن التشعيع. تم إجراء تشعيع بأشعة كاما باستخدام مصدر ^{137}Cs بمعدل تعرض قدره 0.56 Gy / min وكذلك تم فحص خاصية الحد البصري للعينه بأستخدام ليزر SDL−532−100T ذو الموجه المستمره عند 532nm . وفي الوقت نفسه وجد ان سلوك الحد البصري يزداد بزيادة الجرعة. كذلك هذه الدراسه تقترح ان أشعة كاما يمكن اعتبارها أداة لتحسين خصاص الجهاز لتطبيقات مختلفة .

الكلمات المفتاحية: التشعيع باشعة كاما، بارونين y، معامل الانكسار اللاخطي، المحدد البصري، الجهاز البصري.