JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 28, Iss. 5 - 6, May - June 2026, p. 242 - 250

Structural, morphological, linear and third order nonlinear optical study of Al_2O_3 doped PVA composite thin films

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Aluminium Oxide (Al_2O_3) doped poly vinyl alcohol (PVA) composite thin films on silica glass plate were prepared via casting method and their structural, morphological, linear and 3rd order nonlinear optical (NLO) properties were reported. The polymer composite thin films were fabricated with varying weight percentages (1 wt %, 2 wt %, and 3 wt %) of Al_2O_3. The XRD technique revealed that the sample exhibited amorphous nature. The scanning electron microscopy (SEM) study showed the morphological features of the polymer composite thin films and the EDX spectrum reports the chemical configuration of the film. The 3rd order NLO study was carried out using a diode laser working at 650 nm wavelength. The composite films exhibited a negative nonlinear refractive index and nonlinear absorption coefficient due to self-defocusing and saturable absorption behaviors, respectively. The 3rd order NLO refractive index and absorption coefficient of the films were found to be the order of 10^{-7} cm^2/W and 10^{-3} cm/W. It was noted that, the NLO features of the thin film was increased with increase in Al_2O_3 concentration. The real and imaginary components of the 3rd order NLO susceptibility were measured to be the order of 10^{-6} esu. The results suggest that Al_2O_3 doped with PVA composite thin films may be an acceptable choice for future NLO applications.

(Received February 27, 2025; accepted June 2, 2026)

Keywords: Al_2O_3, PVA, NLO, Z-scan, Self-defocusing, Saturable absorption

1. Introduction

Research in nonlinear optics is an emerging area of material science owing to their extensive applications in optical switching, optical limiting, optical data storage, optical computing, two-photon microscopy, etc. [1-4]. Wide variety of materials are used in NLO study in the form of liquid and/or thin films [5-8]. Nanoparticles are a new type of materials that possesses significant NLO features including saturable absorption, reverse saturable absorption, multi photon absorption, self-focusing and self-defocusing, which are originates from nonlinear absorption and nonlinear refraction [9]. Polymer nanocomposites have received a significant attention in the past years as a relatively new class of materials because they combine the exceptional characteristics of nanoparticles and polymers [10]. Rather than being innate to the original parts, their special qualities result from the interaction of the two components. It has also been noted that, adding metal oxide nanoparticles to polymeric materials can change their optical and physical characteristics [10]. Polyvinyl alcohol (PVA) is a synthetic polymer that are biodegradable, nontoxic and soluble in water. Excellent film formation, optical transparency, easy processing, high dielectric strength, and good charge storage capacity are the main benefits of PVA. PVA contains hydroxyl groups along its carbon chain backbone, which can serve as a source of hydrogen bonding to facilitate the formation of polymer complexes. [10].

A large number of polymer nanocomposites used in the NLO study under continuous and pulsed laser regime [11-12]. El-naggar et al. [13] reported the structural, linear and NLO properties of PVA/CMC blended MnS/ZnS nanocomposites. NLO features of MgO nano-filler doped PVA nanocomposites was studied by Shamekh et al. [14]. Heiba et al. [15] reported that PVA/PVP blended polymer with ZnCo2O4/CdS nanocomposites for optoelectronics application. The 3rd order NLO characteristics of the samples are measured experimentally using different methods such as beam distortion, ellipse rotation, three and four-wave mixing, second harmonic generation, etc. [16–20]. Along with other existing methods, Z–scan method is a sensitive experimental technique that can be used to measure the 3rd order NLO features of a variety of materials

[21]. Z–scan is an accurate and user-friendly technique due to its simple experimental measurements. It can be used to determine the real and imaginary components of the susceptibility, as well as the sign and magnitude of the n_2 and β of the materials, which are directly measured from the transmittance curve. [21].

This paper presents the 3rd order NLO features of Al_2O_3 doped PVA composites with varying concentration of Al_2O_3 using low power diode laser. The sample is prepared by casting method and the morphology of the sample, crystalline property has been reviewed from SEM and XRD techniques, respectively.

2. Experimental details

PVA (Mw=130000 g/mol) was obtained from Sigma Aldrich and used the sample as such. Al_2O_3 nanoparticle was purchased from Nanoshel India. The Al_2O_3 doped PVA composites were prepared by dissolving 1 g of commercially available PVA in 20 ml of distilled water and stirred for 2 h at 60 ^{0}C. The different weighing percentage (1 wt %, 2 wt % and 3 wt %) of Al_2O_3 was added separately into PVA solution and again stirred for 1 h at room temperature. The polymer composite with different weighing percentage of Al_2O_3 were prepared on thin silica glass plate by casting method and dried at room temperature for 24 h. The thickness of the polymer composite films was found to be 60 μm.

The 3rd order NLO features of the thin films were measured by Z–scan technique working at 650 nm wavelength. A 5 cm focal length of convex lens was used to focuses the beam. The composite thin films were placed on the micrometer translational stage and move the sample from –Z to +Z positions. A detector fed to the digital power meter calculates the transmittance from the sample. The experimental setup used in the present work was similar to that of our reported work [4]. The thin film's nonlinear refractive index and nonlinear absorption coefficient were measured using two distinct techniques: closed aperture (CA) and open aperture (OA). The beam transmittance was collected using the convex lens in the OA scenario and the aperture in the CA scheme.

3. Results and discussions

3.1. UV-Visible absorption study

The UV-Visible absorption spectrum of Al_2O_3 doped PVA nanocomposites with different weight percentage (1 wt %, 2 wt % and 3 wt %) of Al_2O_3 is shown in Fig. 1. From Fig.1, the spectrum covers 200 nm to 800 nm and the maximum absorbance is observed at higher weight percentage of Al_2O_3. A strong absorption peak observed in the UV region is attributed to the energy band gap of the samples [22]. Moreover, the absorption peaks of Al_2O_3-doped PVA nanocomposites with varying weight percentages decreases owing to reduction in particle size.

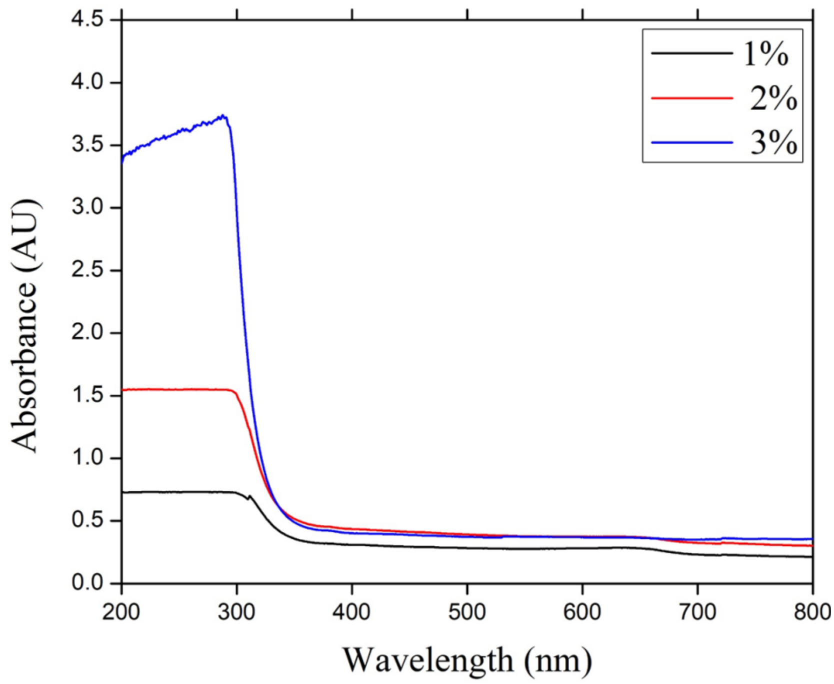


Fig. 1. UV-Visible absorption spectrum of Al_2O_3 doped PVA (colour online)

3.2. XRD study

The X-ray diffraction (XRD) image of Al_2O_3 doped PVA nanocomposites with varying weighing percentages of Al_2O_3 is shown in Fig. 2 (a-c). The XRD image of the nanocomposites are appeared as a polycrystalline property of the prepared thin films. When the concentration of Al_2O_3

of thin film increases, the polycrystalline nature of the film decreases.

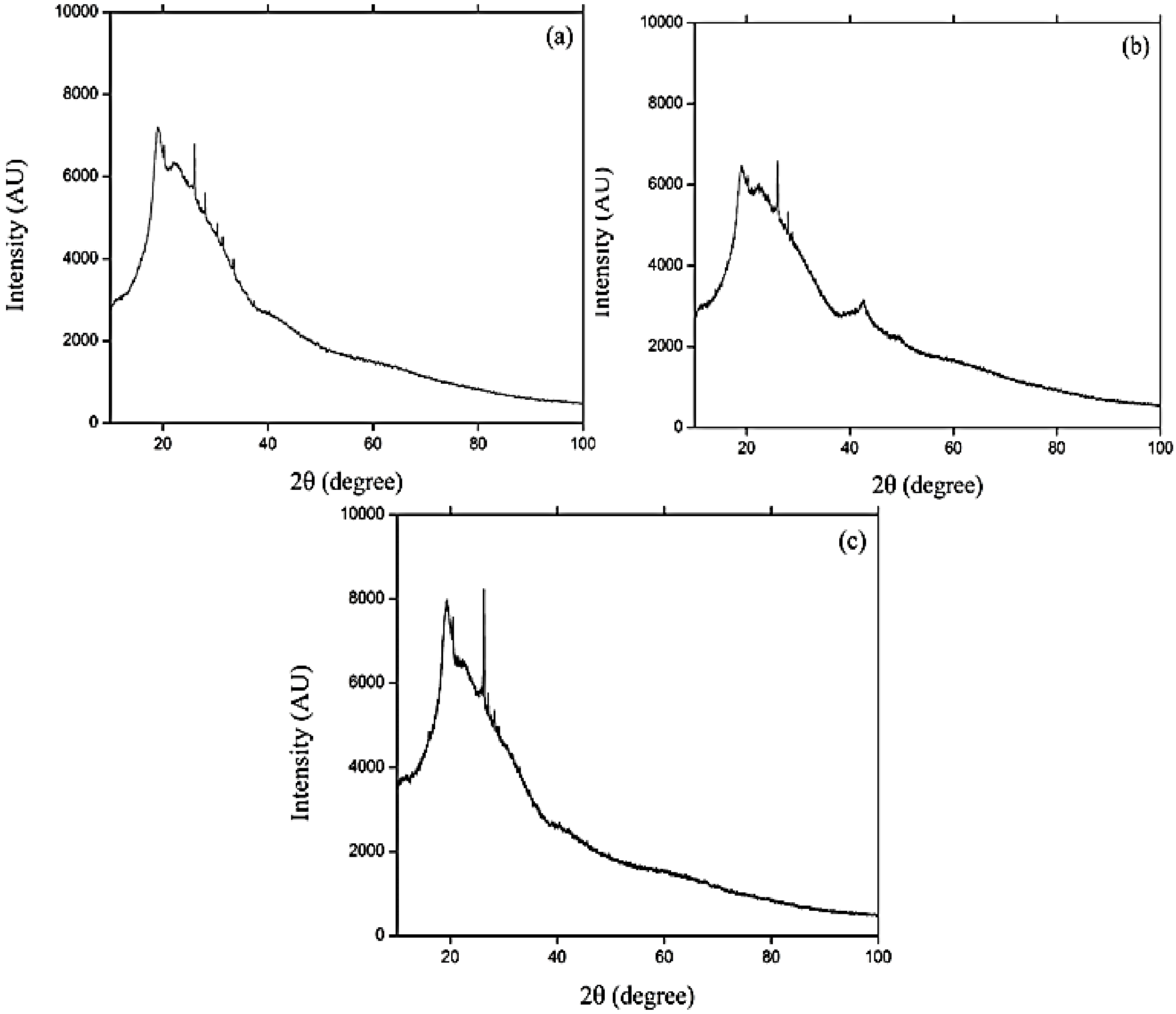


Fig. 2. X-ray diffraction image of Al_2O_3 doped PVA (a) 1 wt% (b) 2 wt% (c) 3 wt%

3.3 Scanning Electron Microscope (SEM) with EDX study

The scanning electron microscope image of Al_2O_3 doped PVA nanocomposites with varying weighing percentages of Al_2O_3 is shown in Fig. 3 (a-c). It is obvious from the images that the Al_2O_3 concentration increases, the size of the nanoparticles becomes decreases and agglomerated at a higher concentration of Al_2O_3. Furthermore, the particles are observed spherical in shape at higher concentration (Fig. 3 c). The surface is considered as smooth and little white spots is observed at lower concentration of Al_2O_3. The particle size is also decreased by increasing the concentration of Al_2O_3. Fig. 4 (a-c) shows the energy dispersive X-ray spectrum of Al_2O_3 doped PVA nanocomposites with varying weighing percentages of Al_2O_3. It is clear from Fig. 4 (a-c) that the aluminium percentages of the prepared sample increased from 1 wt% to 3 wt%.

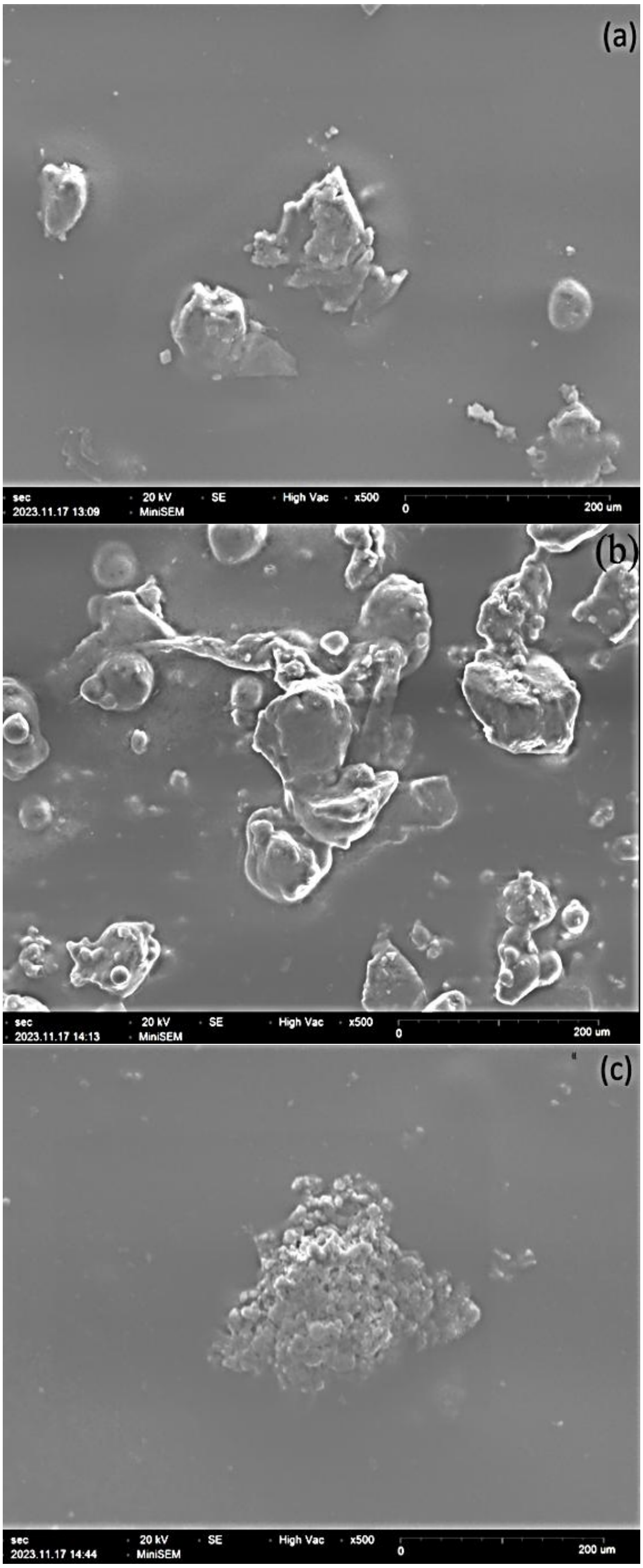


Fig. 3. SEM image of Al_2O_3 doped PVA nanoparticles (a) 1 wt% (b) 2 wt% (c) 3 wt%

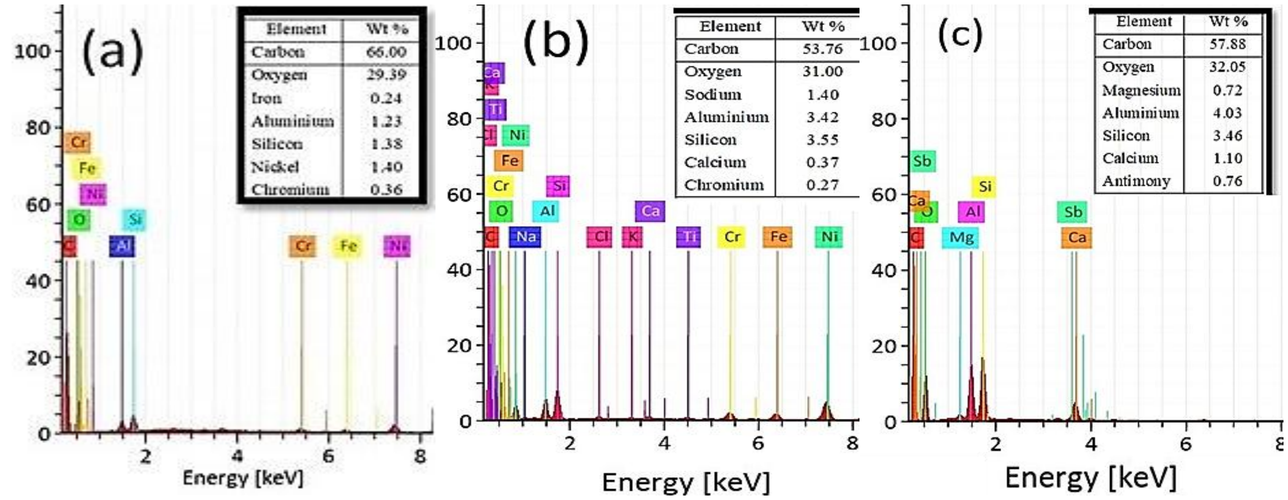


Fig. 4. EDX spectrum of Al_2O_3 doped PVA (a) 1 wt% (b) 2 wt% (c) 3 wt% (colour online)

3.4. 3rd order NLO study

3rd order NLO features such as n_2 and β of Al_2O_3 doped PVA composite thin films with different weight percentages of Al_2O_3 is studied from closed aperture (CA) and open aperture (OA) Z–scan techniques. Fig. 5 (a-c) shows the OA result of polymer nanocomposites with different weight percentages. In Fig.5 (a-c), the NLA image of the sample shows the negative nonlinear absorption owing to saturable absorption feature of the sample. Furthermore, the transmittance of the thin films increases with increase in weight percentage of Al_2O_3. The sample demonstrated a high normalized transmittance at 3 wt% and exhibited a significant nonlinear absorption coefficient. Saturable absorption is a type of nonlinear optical process that arises under intense illumination at the focal region, leading to a pronounced change in photon absorption prior to the system relaxing back to its ground state. The nonlinear absorption transmittance from the OA approach is provided by,

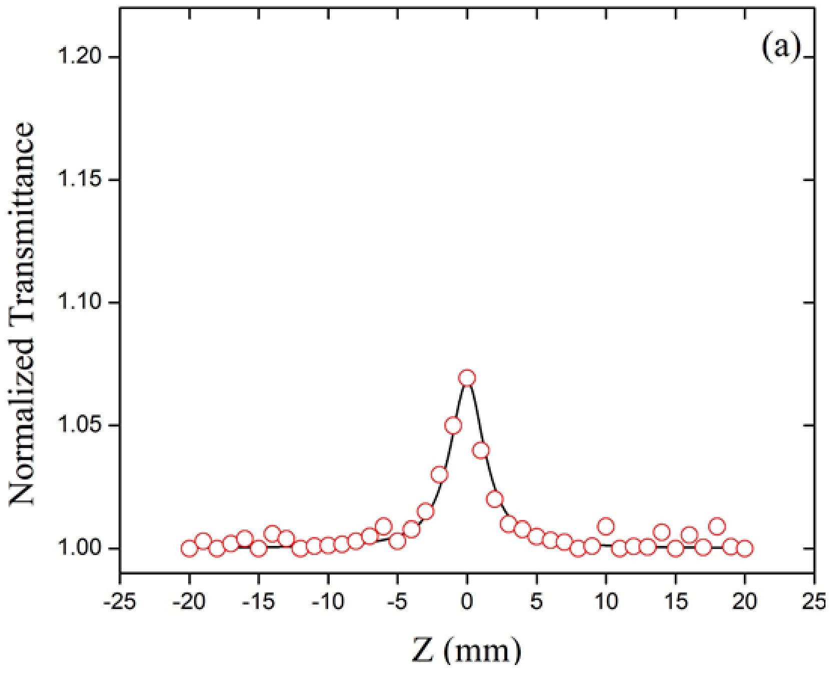
$$T\,(z, s=1) = \sum_{m=0}^{\infty} \frac{[-q_o(z)]^m}{[m+1]^{\frac{3}{2}}}, for\ |q_o(0)| < 1 \quad (1)$$

where

$$q_0 = \frac{\beta I_o L_{eff}}{\left(1+Z^2/Z_0^2\right)} \quad (2)$$

where L_{eff} and Z_o are effective and diffraction length of the thin films. The β of polymer nanocomposites thin films is given by,

$$\beta = \frac{2\sqrt{2}\Delta T}{I_0 L_{eff}} \left(\frac{cm}{W}\right) \quad (3)$$



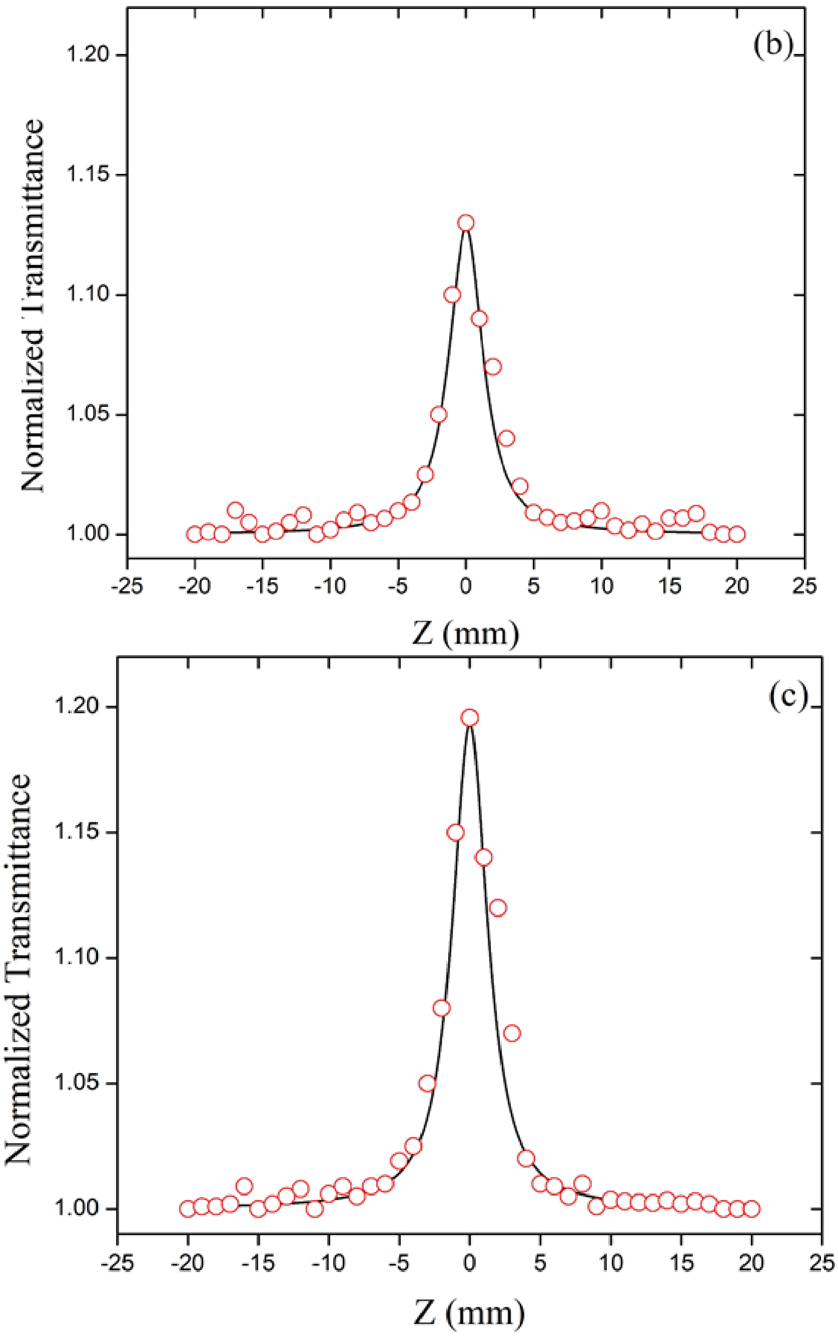


Fig. 5. Open aperture Z–scan result of Al_2O_3 doped PVA composites (a) 1 wt% (b) 2 wt% (c) 3 wt% (colour online)

The CA method is used to determine the sign and magnitude of n_2. The effects of both NLR and NLA are considered when determining the NLR using the CA technique. [16]. While calculating the NLR using the CA technique, the influences of both NLR and NLA are taken into account. Fig. 6 (a-c) shows the pure nonlinear refraction curve of Al_2O_3 doped PVA nanocomposites thin films at different weight percentages of 1 wt %, 2 wt % and 3 wt %. The transmittance curve for all samples shows peak followed by a valley pattern indicates that the transmittance is the outcome of self-defocusing. Also observed from Fig. 6 (a-c) that, the peak-valley difference increases with increase in Al_2O_3 concentration. For a higher concentration, the photon largely interacts with molecules in the sample and the transmittance normally decreases. The normalized transmittance of the polymer nanocomposites thin films in CA technique is given by,

$$T(z) = 1 - \Delta\emptyset_o \frac{4X}{(X^2+1)(X^2+9)} \tag{4}$$

where $X=Z/Z_0$.

The n_2 of Al_2O_3 doped PVA nanocomposites thin films with different weight percentages is calculated by using the relation

$$n_2 \quad = \frac{\Delta\emptyset_0 \lambda}{2\pi I_0 L_{eff}} \left(\frac{m^2}{W}\right) \tag{5}$$

where $\Delta\emptyset_0$, λ and I_0 be the on-axis phase shift, light source wavelength and light Intensity at the focus.

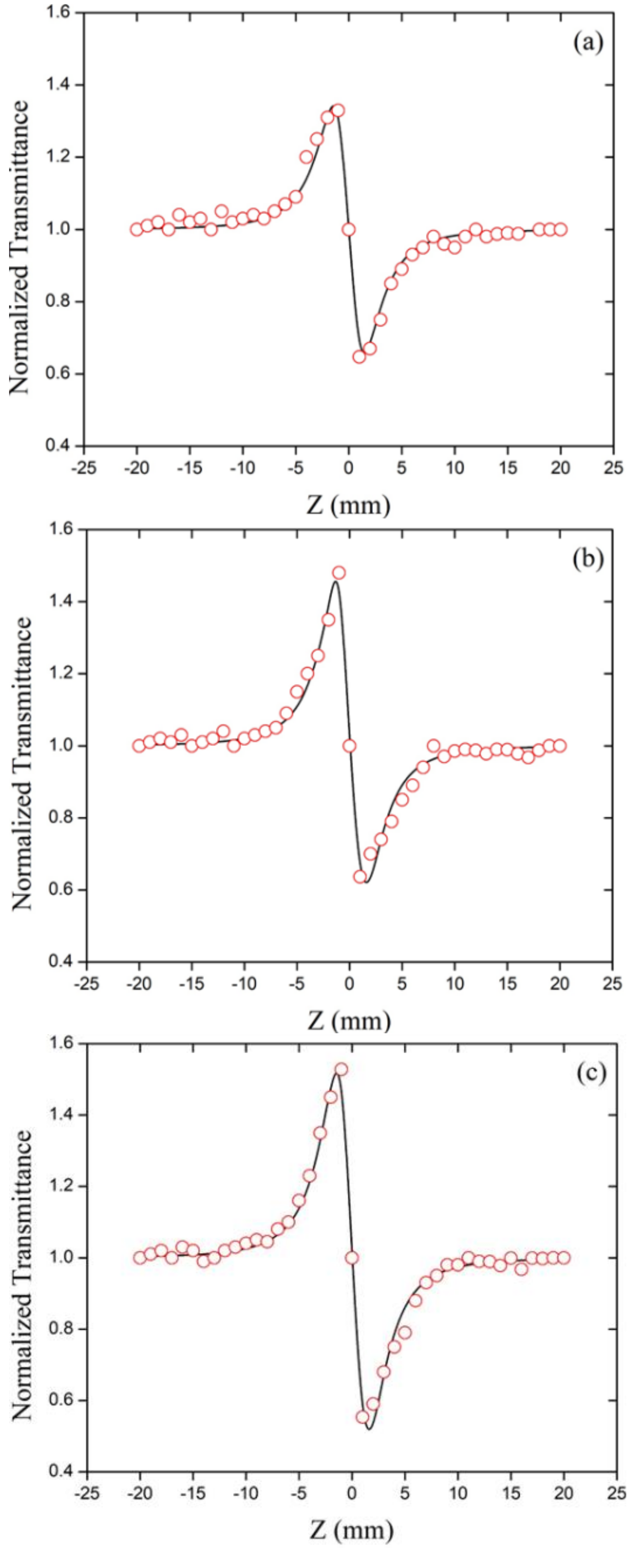


Fig. 6. Pure nonlinear refraction curve of Al_2O_3 doped PVA composites (a) 1 wt% (b) 2 wt% (c) 3 wt% (colour online)

The value of n_2 of the polymer nanocomposites is tabulated in Table 1. The real and imaginary components of the 3rd order NLO susceptibility is given by,

$$Re[\chi^{(3)}](esu) = \frac{\varepsilon_0 c^2 n_0^2}{10^4 \pi} n_2 \left(\frac{cm^2}{W}\right) \tag{6}$$

$$Im[\chi^{(3)}](esu) = \frac{\varepsilon_0 c^2 n_0^2 \lambda}{10^2 4\pi^2} \beta \left(\frac{cm}{W}\right) \tag{7}$$

where c and ε_0 are the speed of light in vacuum and permittivity of free space. The 3rd order NLO susceptibility of Al_2O_3 doped PVA nanocomposites thin films with different weight percentages is given by,

$$\chi^{(3)} = \sqrt{(Re(\chi^3)^2 + (Im(\chi^3)^2} \, (esu) \tag{8}$$

The calculated value of 3rd order NLO susceptibility $\chi^{(3)}$ of Al_2O_3 doped PVA nanocomposites with different weighing percentages such as 1 wt %, 2 wt % and 3 wt % of Al_2O_3 is presented in Table 1. From Table 1, the 3rd order NLO features of the polymer nanocomposites increases with increase in Al_2O_3 concentration. At 3 wt %, the nanocomposites film exhibits large optical nonlinearity. Furthermore, the higher concentrated sample (3 wt % of Al_2O_3) exhibits smaller particle size which in turn increases the optical nonlinearity. The result is very good agreement with reported work [24].

The obtained results are compared with some recently reported nanoparticles doped PVA. It is observed that the values of NLO parameters of Al_2O_3 doped PVA nanocomposites are higher than that of some reported materials. Y.S. Tamgadge et al. [25] reported the NLO properties of $Zn_{(1-x)}Sr_xO$ doped PVA composite thin films. The nonlinear absorption coefficient of the thin film with varying concentration was observed to be the order of 10^{-4} cm/W. M. Halajan et al. [26] reported the ZnSe doped PVA nanocomposites with varying the concentration of ZnSe. They reported that the nonlinear refractive index of the polymer nanocomposites was found to be the order of 10^{-10} cm^2/W. Xiushan Zhu et al. [27] reported the NLO properties of Co_3O_4 doped PVA thin films and observed that the nonlinear refractive index of the film is found to be the order of 10^{-10} cm^2/W. Moreover, earlier studies have reported that increasing the concentration of Al_2O_3 nanocomposites leads to significant changes in the structural and morphological properties of the sample [28,29].

Table 1. 3rd order NLO features of Al_2O_3 doped PVA nanocomposites

Wt %	n_2 X 10^{-7} (cm^2/W)	β X 10^{-3} (cm/W)	Re (χ^3) X 10^{-6} (esu)	Im (χ^3) X 10^{-6} (esu)	$\chi^{(3)}$ X 10^{-6} (esu)
1	– 1.91	– 1.89	– 0.76	– 0.48	0.90
2	– 2.59	– 3.89	– 1.03	– 1.00	1.44
3	– 3.70	– 7.25	– 1.47	– 1.87	2.38

4. Conclusions

The Al_2O_3 doped PVA nanocomposites with different weighing percentages such as 1 wt %, 2 wt % and 3 wt % of Al_2O_3 were prepared and doped on silica glass plate via casting method. The thin films show poly crystalline nature by increasing the Al_2O_3 concentration. The morphology and chemical composition of all the samples were identified by SEM and EDX analysis. The EDX analysis confirm the increasing weight percentages of aluminium in polymer nanocomposites thin films. 3rd order NLO features of polymer nanocomposites thin films were measure by Z–scan technique and observed self-defocusing and saturable absorption behavior of the polymer film. The 3rd order NLO features of the polymer nanocomposites thin film increases with increase in concentration of Al_2O_3. The 3rd order NLO susceptibility was found to be the order of 10^{-6} esu. Based on the experimental results, the Al_2O_3 doped PVA nanocomposites is a low-cost material for application in photonics and optoelectronics.

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