

Significant Enhanced Optical Parameters of PVA-Y₂O₃ Polymer Nanocomposite Films

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Abstract

In our present work, $PVA-Y₂O₃$ nanocomposite films were prepared based on solution casting technique. The structures of the nanocomposite flms were examined by XRD and FTIR spectroscopy. From the XRD analysis, it is found that the flms are semi-crystalline and Y_2O_3 nanoparticles have cubic crystal structure. The average crystal size estimated via Scherer formula was 25 nm while W–H plot method gives 28 nm. FTIR spectra of all nanocomposite films exhibited good interaction between nano-Y₂O₃ and the PVA network. The morphological images obtained from SEM microscopy indicated good distribution of nano-Y₂O₃. Optical absorption spectra of the films enhanced with the incorporation of Y₂O₃. As the percentage of Y₂O₃ added increased, the direct optical energy gap, decreased from 5.42 to 5.31 eV. While the static refractive index, dispersion energy, third order optical susceptibility, nonlinear refractive index and optical conductivity increased.

Keywords PVA nanocomposites · Refractive index · Optical susceptibility · Optical conductivity

1 Introduction

Nowadays with the rapid advancement of nanoscience and technology, extensive research and development implemented on high performance polymeric nanomaterials for targeted applications in various fields $[1-3]$ $[1-3]$. The incorporation of nanoscale reinforcements into the polymer matrix is also an endeavor to develop polymer nanocomposites in

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diferent scientifc and industrial felds [\[4](#page-8-2)[–7\]](#page-8-3). Polyvinyl alcohol (PVA) features high elasticity, transparency and excellent relaxation properties therefore, selected for preparing PVA hybrid nanocomposites [\[8](#page-8-4)[–12](#page-8-5)]. Because of its high refractive index, large energy gap and low phonon energy, barium Y_2O_3 is an important ceramic to be applied as the inorganic filler in polymer nanocomposites $[13-15]$ $[13-15]$. By varying the loading concentration, polymer nanocomposites would also enable the fne tuning of refractive index and the realization of grade-index structure.

Optical properties of PVA/iron oxide nanoparticles have been investigated [\[16](#page-8-8)]. The band gaps decrease as the concentration of iron oxide increases and the refractive index enhanced as well. In the fuorescence spectra three peaks observed at 421, 451 and 469 nm. Chahal et al. [[17](#page-8-9)] have demonstrated the efect of irradiation on the optical properties of PVA-Ag nanocomposites. Accordingly, the optical band gap decreased from 4.57 to 3.05 eV. These nanocomposites showed high ability to block UV radiation. Morad et al. [[18\]](#page-8-10) have studied PVA as the matrix for dispersing different phases of $TiO₂$ nanoparticles. The optical band gap changed from 3.25 to 2.63 eV as the $TiO₂$ changed from anatase to rutile. It was found that the optical conductivity of PVA-TiO₂ (anatase) is higher than that for PVA-TiO₂ (rutile). M. Rashad [\[19\]](#page-8-11) considered PVA polymer matrix loaded with $Fe₂O₃$ and NiO nanoparticles. It is found that the addition of both $Fe₂O₃$ and NiO increased the optical energy gap as well as dispersion energy. At 1.0 wt% of nano-Fe₂O₃, the static refractive index, third order optical susceptibility and optical conductivity enhanced. PVA-CdS nanocomposites obtained by solution casting route [[20](#page-8-12)]. The UV absorption edge located at 502 nm and a sharp emission peak appeared at 502 nm. After irradiation of PVA-CdS composites, the defect emission quenched. Raju et al. [[21](#page-8-13)] studied the emission spectra of $PVA-Eu^{3+}$ polymer film. The optical absorption spectra indicated an absorption band at 275 nm. After UV irradiation the fuorescence spectra of PVA unchanged while the $PVA-Eu³⁺$ revealed a red emission. Abdullah et al. [[22](#page-8-14)] have developed PVA/PbS nanocomposites via solution casting and reduction technique. The PbS nanoparticles decreased the energy gap from 6.27 to 2.34 eV. It is found that the static refractive index values ranged from 1.09–1.20 as depicted from Cauchy model. Additionally, the refractive index and optical conductivity of PVA/PbS films enhanced with increasing the concentration of nano-PbS. Heiba et al. [[23\]](#page-8-15) prepared PVA-CdSe nanocomposites based on solution casting method. It is found that the optical band gap increased from 5.0 to 5.34 eV with the content of CdSe. Also, the refractive index enhanced with increasing CdSe ratios. As indicated multicolor emission peaks in the UV–Vis spectral region observed with blue shift. PVA-ZnO nanocomposite flms have been prepared and their optical properties investigated [\[24](#page-8-16)]. As the ZnO content increase the optical band gaps decreased from 4.76 eV to 2.38 eV. Interestingly, the PVA-ZnO nanocomposites showed enhanced UV luminescence at 10 mol% of ZnO.

The development of polymer nanocomposites with tunable optical properties is an active feld of research. The grand challenge for the generation of such materials is to develop an integrated synthesis strategy adapting particle generation, surface modifcation and integration inside the polymer. In the present research, we report the simple synthesis of PVA/Y_2O_3 nanocomposite films as well as their structural and optical investigations. The structures of the hybrid flms were examined by XRD and FTIR measurements. The morphologies of the cross-sectional areas of the flms were characterized by SEM. The linear and non-linear optical parameters like refractive index, dispersion energy and non-linear optical susceptibility were analyzed.

2 Experimental Procedure

The films of (PVA-Y₂O₃) nanocomposites in different Y₂O₃ wt% ratios (0.0, 1.0 and 3.0) were prepared by solution cast technique. 1.0 g of PVA dissolved in distilled water and stirred for 1.0 h to obtain a clear and homogeneous solution. A known amount of Y_2O_3 added and stirred continuously

until a viscous solution obtained. The solution was poured into the polystyrene petri dishes to get thin flms.

The crystal structure of nanocomposite flms was examined by a Rigaku XRD-6000 difractometer (Cu Kα radiation, $\lambda = 0.15418$ nm). Microscopic study of the PVA-Y₂O₃ surface conducted on a Quattro ESEM's environmental scanning electron microscope (Thermo Fisher Scientifc). The FTIR/ATR spectra were recorded for 400–4,000 wavenumber using PerkinElmer100 FTIR spectrometer (USA). Optical measurements were performed using High Precision UV–VIS Spectrophotometer PD-3000UV (Japan), having a dual light source capable of emitting ultraviolet as well as visible light.

3 Results and Discussion

Figure [1](#page-2-0) contains measurements of X-ray difraction for $PVA-Y₂O₃$ nanocomposite films at different ratios of nano- Y_2O_3 (0.0, 1.0 and 3.0 wt%). It is observed from this figure that the polyvinyl flm has a semi-crystalline structure of the monoclinic structure at $2\theta = 18.54^\circ$ which agree with the literature [\[9](#page-8-17)]. After doping the PVA flms the XRD spectrum showed diffraction lines of nano- Y_2O_3 as well. The spectrum of Y_2O_3 nanopowder match very well with the cubic crystal structure reported in ICDD PDF 41–1105 and agree very well with the data reported in the literature $[25, 26]$ $[25, 26]$ $[25, 26]$ $[25, 26]$. Numerous studies demonstrated that the Scherer equation applied to determine the average size of crystals (D) from the XRD patterns [\[27](#page-8-20)[–37](#page-9-0)]

$$
D = \frac{0.9\lambda}{\beta \cos \theta} \tag{1}
$$

where λ is the X-ray wavelength, θ is the diffraction angle and β is the corrected full width at half maximum.

After applying this equation, we obtained an average size for Y_2O_3 nanocrystals of about 25 nm. In view of the importance of applying the Williamson-Hall model to calculate the average size of crystals [\[38](#page-9-1), [39\]](#page-9-2), the linear ftting of data in Fig. [1b](#page-2-0) was used to calculate the size of Y_2O_3 crystals according to this model (see Fig. [1b](#page-2-0)). Thus, the average size of the crystals is approximately 28 nm. The microstrain (ε) value was 1.05×10^{-3} , which contributed to the broadening of the difraction peaks and thus the estimated crystal size is larger than that computed from Scherer formula.

To study the surface morphology of the prepared flms, scanning electron microscopy images collected and dis-played in Fig. [2.](#page-2-1) The SEM image of the Y_2O_3 nanopowder showed fne nanoparticles (see Fig. [2](#page-2-1)a).

Figure [2](#page-2-1) shows the SEM images containing a uniform distribution of Y_2O_3 nanoparticles without large agglomerations.

Fig. 1 a XRD spectra of PVA-Y₂O₃ nanocomposites and **b** βcos(θ) versus 4sin(θ) for Y₂O₃ nanopowder

Fig. 2 SEM scans of **a** Y₂O₃ nanopowder, **b** PVA pure film, **c** PVA + 1.0 wt% Y₂O₃ and **d** PVA + 3.0 wt% Y₂O₃ nanocomposite films

In Fig. [3](#page-3-0), the FTIR/ATR spectra of the polyvinyl alcohol films doped with nano-Y₂O₃ at ratios 0.0, 1.0, 3.0 wt%. Looking at this fgure, a strong absorption band at 3292 cm⁻¹ that is related to O–H stretching vibrations [[9,](#page-8-17) [40](#page-9-3)–[44](#page-9-4)]. The transmission bands at 2920 and 2850 cm^{-1} denote the asymmetrical and symmetrical stretching vibrations of C-H. Two absorption bands appeared at 1716 and 1088 cm⁻¹ comes due to C=O and C-O stretching vibrations of vinyl acetate group. It is evident here that the absorption peak at 1418 cm^{-1} related to –C-H- [[45\]](#page-9-5), shifted to

Fig. 3 FTIR/ATR spectra of PVA-Y₂O₃ nanocomposites

1416 cm⁻¹ when the ratio of Y₂O₃ was 3.0 wt%. The peak appeared at 1327 cm^{-1} is due to wagging vibrations of C-H groups. The bands at 944 and 833 cm^{-1} correspond to skeletal vibrations of PVA [\[46\]](#page-9-6). From the current data, it is apparent that there is a strong intercalation between the polymer vinyl alcohol network and Y_2O_3 nanoparticles.

Figure [4](#page-3-1) shows optical data such as absorbance, transmittance and reflectance for the present $PVA-Y_2O_3$ nanocomposite flms. It is clear that increasing the concentration of nano- Y_2O_3 improved the absorbance of the nanocomposite flms (Fig. [4](#page-3-1)a). Also, the optical transmittance decreased due to the increase in Y_2O_3 concentration (Fig. [4](#page-3-1)b).

Also in Fig. [4a](#page-3-1) an absorption peak at 280 nm noticed, which is associated with $n-\pi^*$ transition of $C=O$ [[47\]](#page-9-7). From the figure, it is observed that when the loading of Y_2O_3 increases, the intensity of this peak enhanced.

The direct and indirect optical energy gaps can be determined by applying the Tauc's equation to the optical absorption data [\[48](#page-9-8), [49](#page-9-9)];

$$
\alpha h v = B(hv - E_g)^n \tag{2}
$$

where α denotes the optical absorption, $h\nu$ is the energy of the photon beam, *B* is a constant and n equals 0.5 for direct allowed transitions but $n = 2.0$ in case of indirect allowed transitions. Figure [5](#page-4-0) shows a graph between $(\alpha h\nu)^n$ *vs.* $h\nu$ for the nanocomposite films. From this figure, the direct ($E_{\text{g(direct)}}$) and indirect ($E_{\text{g(idirect)}}$) energy gaps can be estimated via extrapolating the linear part to zero photon absorption ($(\alpha h \nu)^n = 0$). The estimated values of E_{g(direct)} are 5.42, 5.37 and 5.31 eV when the Y_2O_3 percentage is 0.0, 1.0 and 3.0 wt%, respectively. It is found that the samples have indirect energy gaps (4.90, 4.73 and 4.54 eV) for the concentrations 0.0, 1.0 and 3.0 wt% of nano- Y_2O_3 . The decrease of both energy gaps with addition of nano- Y_2O_3 may be due the formation of new levels [\[39](#page-9-2)].

It was demonstrated in the previous work that the linear refractive index (n) can be estimated from the following relation [[39,](#page-9-2) [50\]](#page-9-10):

$$
n = \left(\frac{1+R}{1-R}\right) + \sqrt{\frac{4R}{(1-R)^2} - k^2}, \ k = \frac{\alpha \lambda}{4\pi}
$$
 (3)

where R denotes reflectance and *k* is the extinction coefficient.

Figure [6](#page-4-1) shows the linear refractive index profiles for PVA- Y_2O_3 films when the PVA matrix doped with 0.0, 1.0 and 3.0 wt% of Y_2O_3 . It is apparent that the refractive index enhanced

Fig. 4 Optical absorbance, transmittance and reflectance data for the PVA-Y₂O₃ nanocomposites

Fig. 5 a $(\alpha h \nu)^2$ and **b** $(\alpha h \nu)^{0.5}$ as a function of $h \nu$ for the PVA-Y₂O₃ nanocomposite films

when the ratios of nano-Y₂O₃ increased up to 3.0 wt%. This increase in the linear refractive index is due to the agglomeration of particles and therefore increasing the dispersion of the nanocomposite flms [[39\]](#page-9-2).

It is known that the refractive index dispersion is defned as [[51\]](#page-9-11);

$$
\left(n^2 - 1\right)^{-1} = \frac{E_0}{E_d} - \frac{1}{E_0 E_d} (hv)^2,\tag{4}
$$

where E_0 denotes the single oscillator energy and E_d measures the interband optical transition strength (disper-sion energy). Figure [7](#page-5-0) shows the graphs of $(n^2 - 1)^{-1}$ versus $(hv)^2$, which gives straight lines.

In Fig. [7](#page-5-0), the slope of the straight lines equals $\left(-\frac{1}{E_0 E_d}\right)$ λ and the intercept is (E_0/E_d) . Table [1,](#page-5-1) gives the estimated values of E_0 and E_d for the PVA-Y₂O₃ nanocomposite films. It is noted here that the single oscillator energy decreased from 4.15 to 2.91 eV while the dispersion energy increased

Fig. 7 The graphs of $(n^2 - 1)^{-1}$ versus $(hv)^2$ for the PVA-Y₂O₃ nanocomposite flms

Table 1 Estimated values of $E_{\rm o}$, $E_{\rm d}$ and static refractive index (n_0) for the PVA-Y₂O₃ nanocomposite flms

from 3.70 to 19.39 eV as nano- Y_2O_3 increased up to 3.0 wt%.

It was demonstrated in Wemple and DiDomenico model that the static refractive index at zero photon energy is given by [[39,](#page-9-2) [51\]](#page-9-11);

$$
n_0^2 = \left(1 + \frac{E_d}{E_0}\right) \tag{5}
$$

The values of n_0 tabulated in Table [1](#page-5-1) and showed increase with the Y_2O_3 content. In addition, the dipole strength defined as $f = E_0 E_d$ and the estimated values show increase with increasing Y_2O_3 content (see Table [1\)](#page-5-1).

It is important to estimate the linear optical susceptibility $\chi^{(1)}$ of PVA-Y₂O₃ films via the following relation [\[52\]](#page-9-12);

$$
\chi^{(1)} = \frac{E_d}{4\pi E_0} \tag{6}
$$

Table [1](#page-5-1) gives the values of $\chi^{(1)}$ that showed increase with the nano- Y_2O_3 content. Previous studies have shown

the possibility of calculating the third order optical susceptibility $x^{(3)}$ via Miller's rule like relation [\[53\]](#page-9-13);

$$
x^{(3)} = 6.82 \times 10^{-15} \left(E_d / E_0 \right)^4 \, (e.s.u)
$$
 (7)

In Table [1,](#page-5-1) the values of $x^{(3)}$ are given and shows increase as concentration of nano- Y_2O_3 increase up to 3.0 wt%. The nonlinear refractive index for the PVA-Y₂O₃ nanocomposite films described by the equation [[54](#page-9-14)];

$$
n_2 = \frac{12\pi x^{(3)}}{n_0} \tag{8}
$$

Table [1](#page-5-1) showed that the values of n_2 were enhanced when the concentration of nano- Y_2O_3 increased up to 3.0 wt%. By looking at previous research, we fnd that the values of nonlinear refractive index and third order optical susceptibility are greater than these found in the literature [[55](#page-9-15)]. Hence, $PVA-Y_2O_3$ nanocomposite films can be used in applications of nonlinear optical devices.

The real and imaginary parts of optical dielectric constant are given in terms of refractive index and extinction coefficient as follows $[16, 55, 56]$ $[16, 55, 56]$ $[16, 55, 56]$ $[16, 55, 56]$ $[16, 55, 56]$ $[16, 55, 56]$ $[16, 55, 56]$;

$$
\varepsilon_1 = n^2 - k^2 = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2},
$$

\n
$$
\varepsilon_2 = 2nk = \frac{4\pi^2 e^2}{m^2 \omega^2 V} \sum_{v,c,k} |\Psi_k^v| p_i^{-} |\Psi_k^c| \delta \left(E_{\Psi_k^c} - E_{\Psi_k^v} - hv \right)
$$
\n(9)

where ε_{∞} denotes the high frequency dielectric constant, $\omega_{\rm p}$ is the plasma frequency and Ψ_k^v defines the occupied but Ψ_k^c defnes the unoccupied electronic states. The real optical dielectric constant ε_1 versus ω^{-2} of three compositions of $PVA-Y_2O_3$ films are plotted in Fig. [8](#page-6-0).

As obtained from the figure above y-intercept equals ε_{∞} and the slopes give the values of plasma frequency ω_p (see Table [2\)](#page-6-1). It can be seen that the dielectric constants of the $PVA-Y_2O_3$ composites increase significantly with increasing Y_2O_3 content. For the PVA-Y₂O₃ nanocomposites, higher plasma frequencies achieved as the amount of Y_2O_3 in the composites increase.

From Eq. 9 when plotting ε_2 versus photon energy (*hv*) for the PVA-Y₂O₃ nanocomposite films (Fig. [9](#page-7-0)). The intercept of straight lines give the real energy gaps $E_{g(\varepsilon 2)}$ which listed in Table [2](#page-6-1). It was found that the values of $E_{g(\varepsilon 2)}$ were

Table 2 Estimated values of ε_{∞} , ω_{p} and the real energy gaps (E_{g(ε_{2}))} for the PVA-Y₂O₃ nanocomposite films

Y_2O_3 content $(wt\%)$	ε_{∞}	$\omega_{\rm p}$ × 10 ¹⁴ (Hz)	$E_{g(\varepsilon 2)}$ (eV)
$_{0.0}$	2.12	7.30	5.25
1.0	3.69	12.32	5.23
3.0	12.05	32.81	5.20

5.41, 5.34 and 5.31 eV. Looking at Table [1](#page-5-1) and comparing the energy gap values, it is found that the most probable transition is direct allowed.

As already mentioned, the optical conductivity (σ_{opt}) describes the response of medium to electromagnetic waves. To determine the optical conductivity we have to consider the relation between optical conductivity and refractive index given by [\[57](#page-9-17), [58](#page-9-18)];

$$
\sigma_{opt} = \frac{\alpha n C}{4\pi} \tag{10}
$$

where C defines the speed of light in empty space. Figure [10](#page-7-1) presents the optical conductivity data versus photon energy. The optical conductivity directly depends on refractive index and absorption coefficient of nanocomposites.

Fig. 10 Optical conductivity of the PVA-Y₂O₃ nanocomposite flms as a function of photon energy

When the percent of Y_2O_3 and photon energy increase, the optical conductivities are increased signifcantly. This is probably due to the formation of new levels inside the band gap, which facilitate the excitation of electrons from

the valence band to the nearest states [[55](#page-9-15), [58\]](#page-9-18). These outcomes open the door for employing the prepared PVA- Y_2O_3 nanocomposite films in the optical and optoelectronic applications.

4 Conclusions

The effect of nano- Y_2O_3 on conventional PVA films have been examined by XRD, SEM, FTIR/ATR and optical absorption spectroscopy. The crystal structure of Y_2O_3 nanoparticles is cubic with an average crystal size of 28 nm. The dispersal of Y_2O_3 nanoparticles to the PVA polymer surface showed high uniform distribution. The FTIR spectra showed possible interactions between Y_2O_3 nanoparticles and host PVA flms. Optical energy gaps at different concentrations of nano- Y_2O_3 decreased as the concentration reaches 3.0 wt%. The refractive index and optical dielectric constant of the nanocomposite flms increased with an increase of Y_2O_3 in the polymer system. The dispersion energy, third order optical susceptibility, nonlinear refractive index and optical conductivity increased as well. Due to improvement in linear and nonlinear optical parameters, $PVA-Y_2O_3$ nanocomposite films can be applied in the optical and optoelectronic devices.

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Declarations

Conflict of interest The authors declare that there is no confict of interest.

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