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Preparation and Tuning the Structural and Optical Properties of PVA/CuO-In₂O₃ Nanocomposites for Optoelectronic and Antibacterial Applications

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ABSTRACT

This research presents the fabrication and characterization of a multifunctional nanocomposite film by incorporating CuO and In₂O₃ nanostructures within a polyvinyl alcohol (PVA) matrix. Unlike conventional single-component composites, this work features a unique formulation that integrates the synergistic effect of CuO and In₂O₃ particles within the polymer, simultaneously enhancing the optical and antibacterial properties, targeting effective and low-cost applications in the medical and optoelectronic fields. The structure and surface are analyzed using optical microscopy and scanning electron microscopy. Optical studies, in the spectral range (260-860 nm), show a significant improvement in the absorbance and optical conductivity with increasing nanoparticle content. The incorporation of 3wt.% CuO-In₂O₃ increases the absorbance by about 55% and the optical conductivity by about 35%, with the optical energy gap, decreasing from 4.394 eV (for the pure polymer) to 1.824 electron volt (eV), and this indicates improved light absorption efficiency. The antibacterial activity also shows a proportional improvement with increasing nanoparticle concentration, highlighting the dual functionality of the composite. Compared with the PVA-based nanocomposite materials reported in the previous literature, the developed CuO-In₂O₃ system offers superior multifunctionality and provides a more efficient and scalable route for the design of antibacterial materials with superior optical performance in the future.

1. INTRODUCTION

In the twenty-first century, nanotechnologies are a rapidly developing focus. Nanoparticles have unique structural, optical, electrical, magnetized, and mechanical characteristics due to their nanoscale dimensions of 1 to 100 nm in contrast with their larger bulk counterparts [1, 2]. Metal oxide nanoparticles have increased in interest because of their wide application as catalysts in manufacturing processes, antimicrobial substances in medical applications, additives, chemical detectors, disinfectants, semiconductors, catalysts, and significant contributions to the advancement of cosmetics and microelectronics [3, 4]. Indium trioxide nanocrystals (In₂O₃), as individual n-type semiconductors, have garnered attention as superior semiconductor photocatalysts due to their outstanding electrical and physical properties [5, 6]. Their band gap of 3.71 eV and high exciton binding energy are what really set them apart [7, 8]. A wide range of applications are possible with these nanocrystals due to their strong oxidizing power and remarkable UV emission capabilities [9, 10]. These include photovoltaic, electronic devices, photo catalysts,

sensors, laser diodes, and ultraviolet photo detectors [11, 12]. The wide range of biological applications that In₂O₃ nanoparticles could serve is further enhanced by their biocompatibility and lack of toxicity [13]. Their luminosity at ambient temperature and extraordinary stability make them attractive for a variety of applications. In₂O₃ has a cubic crystal structure, more precisely a bixbyite structure [14, 15]. Eight indium atoms occupy the cube's corner positions, while six oxygen atoms are situated on its faces. Each indium atom is coordinated by six oxygen atoms in an octahedral arrangement. The lattice constant, corresponding to the cube's maximum edge length, is about 10.12 Å. Indium trioxide belongs to the Ia-3 space group, whose symmetry elements include a threefold rotation axis and inversion centers situated at the midpoint of each cube face [16, 17]. CuO is widely used a (p-type) semiconductor material with a narrow band gap of 1.2-1.9 eV, allowing electrons to quickly jump from the valence band to the conducting level [18, 19]. CuO nanopowder has many applications, e.g., wastewater treatment catalysts, sensitive gas sensors, super capacitors, and nanofluid applications [20, 21]. Polyvinyl alcohol is available

in the form of white to cream-colored brittle powder or beads. It can also dissolve in water to form a colorless solution that is widely used in the electronic device industry due to its exceptional insulation qualities and high dielectric permittivity value [22, 23]. Its molecular formula is (C_2H_4O) x, its density varies between 1.190 to 1.310 g/cm^3, and its melting point is 230 degrees Celsius [24, 25]. Recent studies have examined the integration of metal oxides such as CuO or In_2O_3 individually into polymeric matrices. However, the simultaneous integration of these two distinct species within a PVA matrix remains partially unexplored. This unique product offers a significant synergistic target between the strong antibacterial activity of CuO and the excellent optical and electronic properties of In_2O_3, resulting in a versatile material. The emerging PVA matrix is characterized by its stability, light compatibility, limited distribution, film formation, and solubility. Compared with previous work, using individual oxides or different host polymers, the $CuO-In_2O_3/PVA$ system exhibits a superior optical performance, high energy efficiency, and significantly improved antibacterial efficacy. That makes it highly promising for the next generation of optical and medical devices.

2. MATERIALS AND METHOD

PVA/CuO-In_2O_3 nanocomposite films are fabricated using a solution casting technique. Initially, 1 g of polyvinyl alcohol (PVA) is dissolved in distilled water using a magnetic stirrer for continuous stirring. CuO-In_2O_3 nanoparticles are then added to the polymer solution at concentrations of 1%, 2%, and 3% by weight. The mixture is stirred for 15 minutes at 70°C to ensure complete homogeneity. It is left to settle to eliminate air bubbles and to further stabilize the solution. The homogeneous solution is then poured into a 5 cm diameter Petri dish and left to dry at room temperature for 3 to 7 days. After complete drying, the formed films are carefully separated from the mould. Their thickness is measured using a micrometre. The structural, optical, and morphological properties of the composite nanofilms are analyzed using scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), and ultraviolet-visible (UV-Vis) spectroscopy. The optical properties are measured in the spectral range of 200-1100 nm using a Shimadzu UV-18000A dual-beam spectrometer.
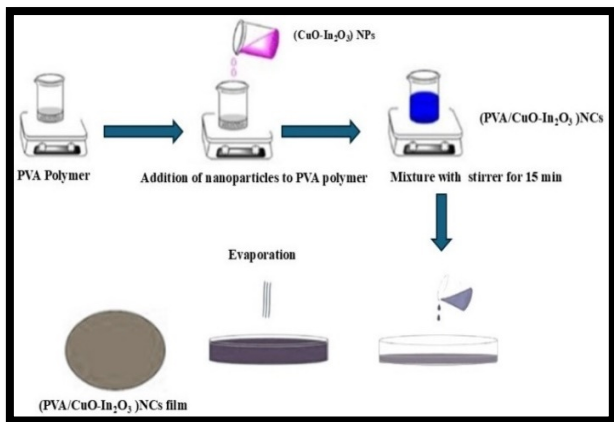

Figure 1. Schematic diagram of the process of preparation (PVA/CuO-In_2O_3) nanocomposites

The solution casting technique used in this study is an efficient low-cost method that is suitable for producing homogeneous nanofilms. Figure 1 illustrates the preparation steps, including the simultaneous distribution and incorporation of CuO-In_2O_3 particles within the PVA matrix during the preparation process. The solution casting method used to prepare PVA/CuO-In_2O_3 composites is simple and low-cost and can be easily scaled up for industrial production of large-area films. However, potential challenges on a large scale may include controlling nanoparticle aggregation. These challenges can affect homogeneity and optical performance, and can ensure a long-term stability of the composites under environmental conditions such as humidity, temperature changes, and UV exposure. These issues can be addressed by modifying the nanoparticle surface, adding stabilizers, and improving storage conditions.

The absorption coefficient (α) is defined as the following equation [25, 26].

$$\alpha = 2.3030 A / t \tag{1}$$

where, A represents absorbance, t represents the thickness of the sample.

Amorphous semiconductors utilize the indirect transition model [27, 28].

$$\alpha h\upsilon = B\left(h\upsilon - Egopt\right)r \tag{2}$$

B: constant;
$h\upsilon$: photon energy;
$E_{.g.}$, band gap energy;
r = 2 for the allowed indirect transition;
r = 3 for the forbidden indirect transition.

The coefficient of extinction (k) is calculated using the following equation [29, 30].

$$k = \frac{\alpha\lambda}{4\pi z} \tag{3}$$

The refractive index (n) is given by [31].

$$n = \sqrt{1+R} \, / \, \sqrt{1-R} \tag{4}$$

where, R is the reflectance.

3. RESULTS AND DISCUSSION

Figure 2 presents typical SEM images of (PVA/CuO-In_2O_3) nanocomposites without and with varying concentrations of (CuO-In_2O_3) nanoparticles. Figure 2(a) illustrates that the polymers are soft, homogeneous, and coherent. Introducing (CuO-In_2O_3) nanoparticles into the (PVA/CuO-In_2O_3) nanocomposites modifies the agglomerates of these systems, as shown in images A, B, C, and D. The surface morphology of the (PVA/CuO-In_2O_3) nanocomposites reveals numerous aggregates or chunks distributed randomly across the top surface [32]. Image (D) indicates an increase in the number of black dots on the surface as the concentration of (CuO-In_2O_3) nanoparticles rises. These findings are consistent with those of previous researchers [33, 34].
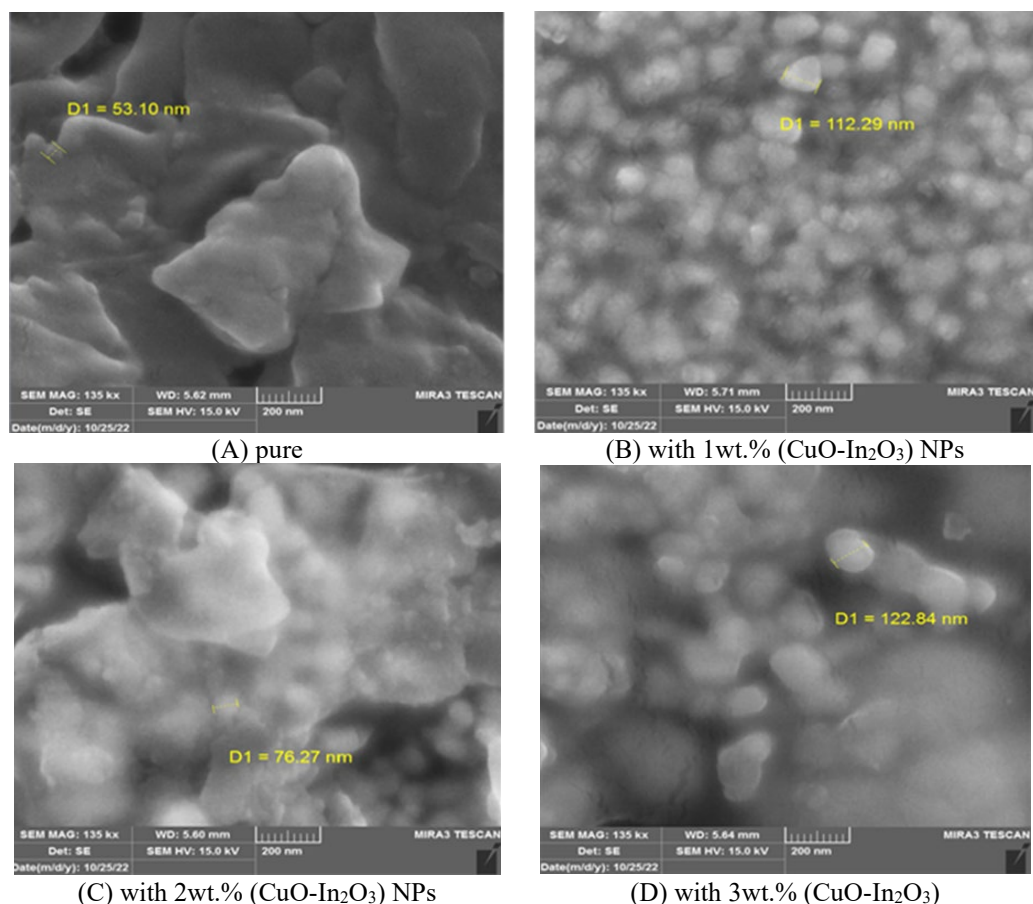
(A) pure

(B) with 1wt.% (CuO-In$_2$O$_3$) NPs

(C) with 2wt.% (CuO-In$_2$O$_3$) NPs

(D) with 3wt.% (CuO-In$_2$O$_3$)

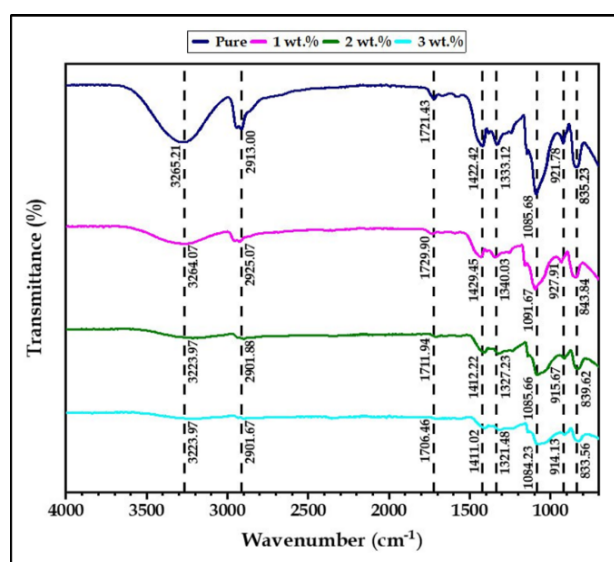**Figure2.** SEM images of (PVA/CuO-In$_2$O$_3$) nanocomposite

Figure 3. FTIR spectra for (PVA/CuO-In$_2$O$_3$) nanocomposites: (A) for (PVA) Polymer (B) for 1 wt.% (CuO-In$_2$O$_3$) NPs, (C) for 2 wt.% (CuO-In$_2$O$_3$) NPs, (D) for 3 wt.% (CuO-In$_2$O$_3$) NPs

The FTIR analysis is performed to detect the functional groups in the nanocomposite samples. FTIR is commonly used to verify the correct co-polymer blend ratio or quantify the amounts of a mixture of release agents, UV stabilizers, and other additives. It is a valuable tool for proper, qualitative and quantitative analyses of polymer constituents [35, 36]. Its primary application is quickly and definitively identifying materials such as compounded plastics, blends, fillers,

rubbers, coatings, and adhesives. Figure 3 depicts PVA and the prepared (PVA/CuO-In$_2$O$_3$) nanocomposites with varying weight percentages of the (CuO-In$_2$O$_3$) nanoparticle, and Table 1 summarizes the results. A comparison of observed wavenumbers for PVA, CuO, and In$_2$O$_3$ has also been performed [37, 38]. Table 1 shows the functional groups present in (PVA/CuO-In$_2$O$_3$) nanocomposites.

The change in absorbance (A) of (PVA/CuO-In$_2$O$_3$) nanocomposites with respect to the incident light wavelength (λ) is shown in Figure 4. The figures demonstrate that the absorbance of all the nanocomposites that have been made improves when the concentration of In$_2$O$_3$-CuO nanoparticles increases. This is because a higher concentration of nanoparticles provides better charge carriers [39, 40]. The absorbance of the nanocomposites is increased by almost 57% at $\lambda = 660$ nm when the concentration of nanoparticles is 3%. Optical, photo catalytic, and solar cell technologies all benefit from these behaviors. In general, the absorbance of all prepared nanocomposites decreases with increasing wavelength. This means that the incident photon is unable to excite the electron and move it from a lower energy level to a higher energy level, because its energy is less than the energy gap value of the nanocomposites [41, 42]. At these energies, donor-level electrons are excited to the conduction band, giving all nanocomposites a high UV absorbance. Photons have enough energy to interact with atoms, producing high ultraviolet absorbance. The UV absorption spectrum of PVA polymer is limited, but it is enhanced by adding In$_2$O$_3$-CuO nanoparticles due to the blend's high energy gap. The graphs of (PVA/CuO-In$_2$O$_3$) nanocomposites with high levels of nanoparticles show a clear peak in the UV region and less absorption in the visible range. The peak maximum absorption

appears at around 300 nm, owing to nanoparticle absorption caused by increased charge carriers. All nanocomposite samples show a wavelength-dependent decrease in absorbance in the visible and near-infrared spectrum. Because incident photons do not have enough energy to interact with atoms,

photon transmission occurs [43, 44]. The (PVA/CuO-In$_2$O$_3$) nanocomposites with 3% concentration of CuO-In$_2$O$_3$ NPs have the most outstanding absorbance because of the high diffusivity of (CuO-In$_2$O$_3$) nanoparticles in the polymer [45, 46].

Table 1. Details of functional groups present in (PVA/CuO-In$_2$O$_3$) nanocomposite

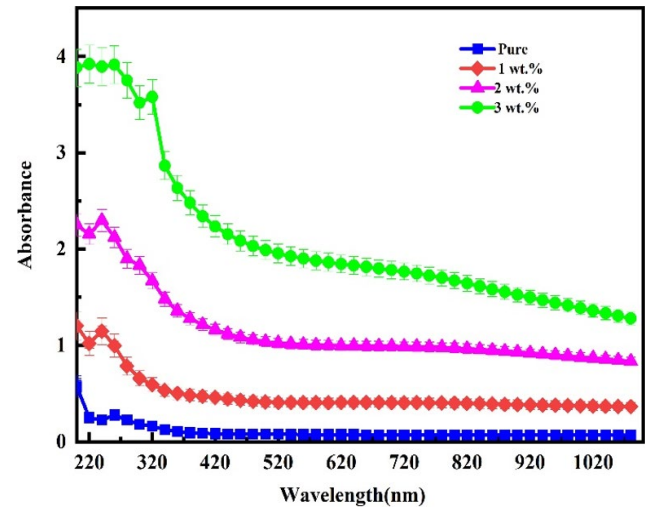| Observed Wavenumber for PVA	Observed Wavenumber for CuO	Observed Wavenumber for In$_2$O$_3$	Observed Wavenumber for (PVA/CuO-In$_2$O$_3$) Nanocomposites
3223-due to O-H stretching Table size	-	-	3265
2913-due to CH asymmetric stretching	-	-	2091
1721-due to C-O carbonyl stretch	-	-	1706
1422-due to C-H bending	-	-	1411
1085-due to C-H deformation vibration	-	-	1084
835-due to C-O stretching	530-due to Cu-O stretching bands	689-due to In-O stretching bands	833

Figure 4. (PVA/CuO-In$_2$O$_3$) nanocomposites absorbance spectrum as a function of wavelength

Figure 5 shows how (PVA/CuO-In$_2$O$_3$) nanocomposites' transmittance (T) varies with incident light wavelength (λ). This Figure shows that excellent excitation leads to a sharp electron that moves from the valence to the conduction band. Interestingly, adding (CuO-In$_2$O$_3$) NPs to polyvinyl alcohol, a polymer causes a gradual and nonlinear reduction in transmission in the visible region [47, 48]. Our results show that increasing the concentration of (CuO-In$_2$O$_3$) NPs wt.% reduces the optical transmission of (PVA/CuO-In$_2$O$_3$) nanocomposites to 65% at 660 nm. The lack of transparency can be attributed to dispersion by NPs within the matrix of polymers. This occurs when the size of the particle is smaller than the wavelength. Or, it could be caused by electron shifts between the polymer and (CuO-In$_2$O$_3$) nanoparticles. The transmittance of the polymer and nanocomposites reduces more in the UV region than in the visible region due to the addition of nanoparticles, which significantly reduces transmittance [49, 50].

The absorption coefficient α for (PVA/CuO-In$_2$O$_3$) nanocomposites is shown in Figure 6 as a function of wavelength. Because the energy of the incident photon is too low to transfer an electron from the valence band to the

conduction band, the absorption coefficient is the lowest at long wavelengths and low energy, ruling out the possibility of electron transfer. An increase in energy makes absorption better, which means there are more electron transitions. Consequently, an electron can move from the valence band to the conduction band by means of the incident photon's energy [51, 52]. At lower energies, when the absorbent coefficient is low (104 cm^{-1}), electrons are predicted to transition indirectly with the phonon maintaining their electronic momentum [53, 54].
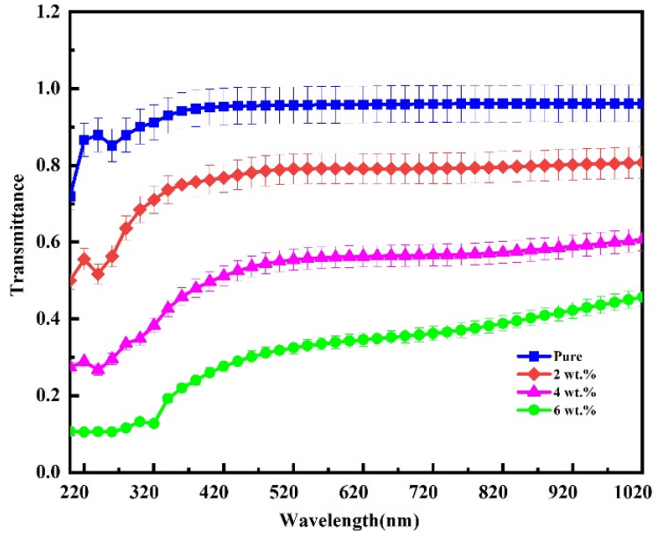

Figure 5. Transmission versus wavelength of (PVA/CuO-In$_2$O$_3$) nanocomposites at room temperature

Figure 7 illustrates the energy gaps for the allowed indirect transitions in (PVA/CuO-In$_2$O$_3$) nanocomposites, whereas Figure 8 depicts the energy gaps for the forbidden indirect transitions in the same nanocomposites. The obtained values are shown in Table 2. The values of both allowed and forbidden indirect energy gaps in all prepared nanocomposites decrease as the concentration of (CuO-In$_2$O$_3$) nanoparticles increases. At 3% concentration and λ = 660 nm, the energy gap of (PVA/CuO-In$_2$O$_3$) nanocomposites are decreased to approximately 57% for allowed indirect transitions and 77% for forbidden indirect transitions. This behavior makes

nanocomposites ideal for lightweight, low-cost electronics and optical devices [55, 56]. The progressive reduction in energy gap values as nanoparticle concentration increases can be explained by the creation of localized states within the energy gap. In this mechanism, electrons transition in two stages: first from the valence band to localized states within the energy gap, and from these states to the conduction band [57, 58].
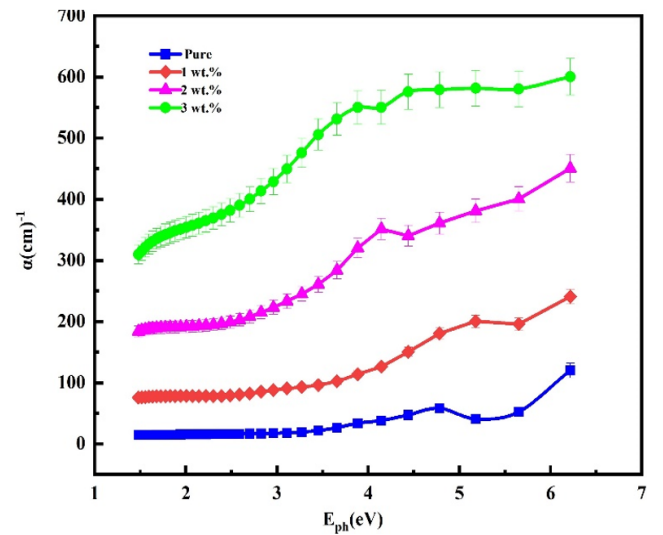

Figure 6. The absorption coefficient spectra as a function of wavelength for (PVA/CuO-In$_2$O$_3$) nanocomposites at room temperature
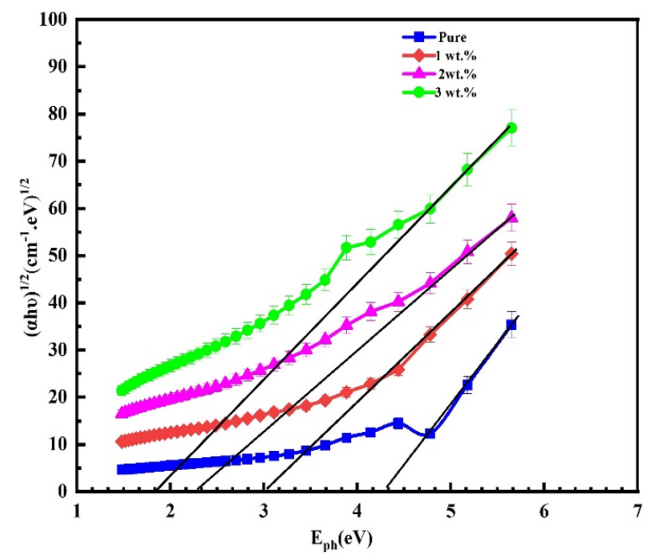

Figure 7. Photon energy affects $(\alpha h\upsilon)^{1/2}$ for (PVA/CuO-In$_2$O$_3$) nanocomposites

The optical energy gap of the (PVA/CuO-In$_2$O$_3$) nanocomposites decreases with the increase of the nanoparticle concentration. Additionally, charge transfer interactions between the PVA matrix and metal oxide nanoparticles may result in the formation of localized states at the band edges, leading to bandgap narrowing. These changes are consistent with the morphological observations from SEM images, which showed improved dispersion and interfacial interactions at higher nanoparticle content [59, 60]. Such structural modifications directly influence the electronic structure and consequently the optical response of the nanocomposites.
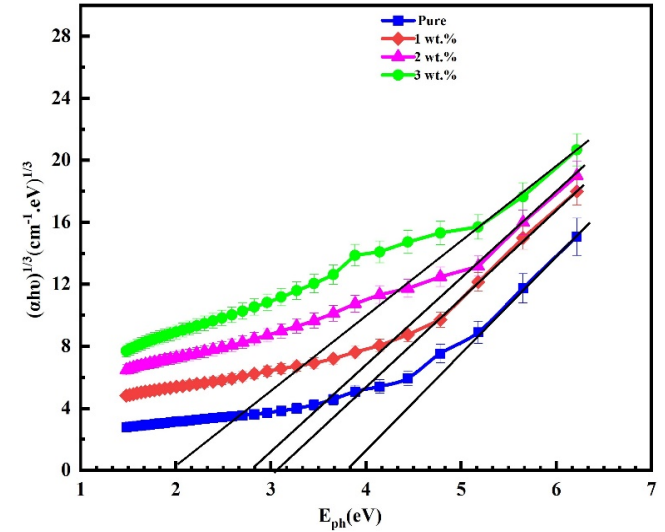

Figure 8. Variation of $(\alpha h\upsilon)^{1/3}$ for (PVA/CuO-In$_2$O$_3$) nanocomposites with photon energy

Table 2. Energy gap values various with (CuO-In$_2$O3) concentration

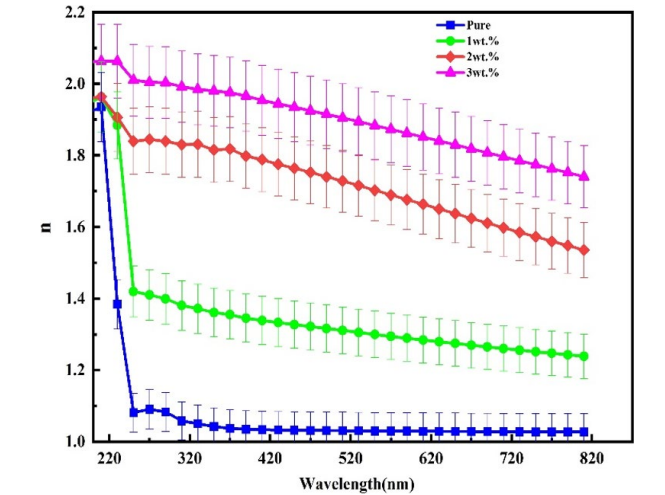| Sample	Concentrations of (CuO-In$_2$O$_3$) NPs wt.%	Eg (eV) (Allowed)	Eg (eV) (Forbidden)
PVA polymer (PVA/CuO-In$_2$O$_3$) nanocomposites	0	4.394	3.837
(PVA/CuO-In$_2$O$_3$) nanocomposites	1	2.958	3.05
(PVA/CuO-In$_2$O$_3$) nanocomposites	2	2.287	2.817
(PVA/CuO-In$_2$O$_3$) nanocomposites	3	1.824	1.997

Figure 9. Refractive index of (PVA/CuO-In$_2$O$_3$) nanocomposites variation with wavelength

Figure 9 presents the refraction index of (PVA/CuO-In$_2$O$_3$) nanocomposites as a function of wavelength. The findings show that as the weight proportion of the percentage of (CuO-In$_2$O$_3$) nanoparticles rises, so does the refractive index. The

noticed rise in refractive index is thought to result from increased nanocomposites' density. Nanocomposites are made denser by adding nanoparticles to a polymer matrix. With a higher density, there are more atoms and molecules per unit volume, which in turn raises the refractive index [61, 62]. The refractive index rises as incoming light interacts with substances that are highly UV-refractive. Finding a correlation between CuO-In_2O_3 nanoparticle concentration and refractive index has important implications for developing new nanocomposite materials [63, 64].

Figure 10 illustrates how the extinction coefficient of nanocomposites (PVA/CuO-In_2O_3) varies with the wavelength (λ). These figures demonstrate that as the wavelength of light that enters the UV region rises, so does the extinction coefficient of (PVA/CuO-In_2O_3) nanocomposites, which are highly absorbent in these regions. The practically constant absorption coefficient of the nanocomposites in the visible and near-infrared regions also explains why their extinction coefficient increases in these areas [65-67]. As a result, the extinction coefficient varies with the wavelength. The extinction coefficient of (PVA/CuO-In_2O_3) nanocomposites is directly proportional to the concentration of (CuO-In_2O_3) nanoparticles, which increases with higher concentrations. This is owing to the higher absorption coefficient of the nanocomposites [68-70]. The rise in extinction coefficient for (PVA/CuO-In_2O_3) nanocomposites is 53%.
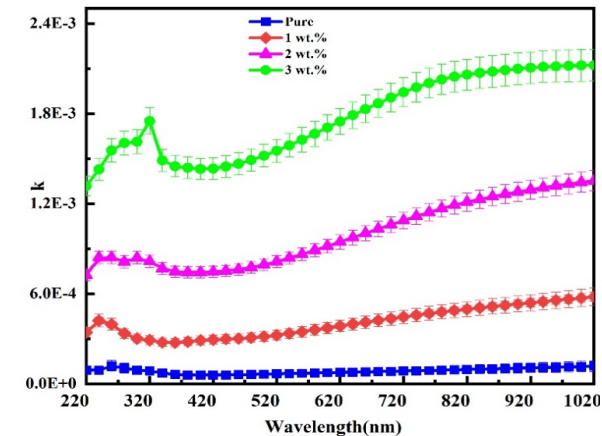

Figure 10. Variation of the extinction coefficient of (PVA/CuO-In_2O_3) nanocomposite with wavelength
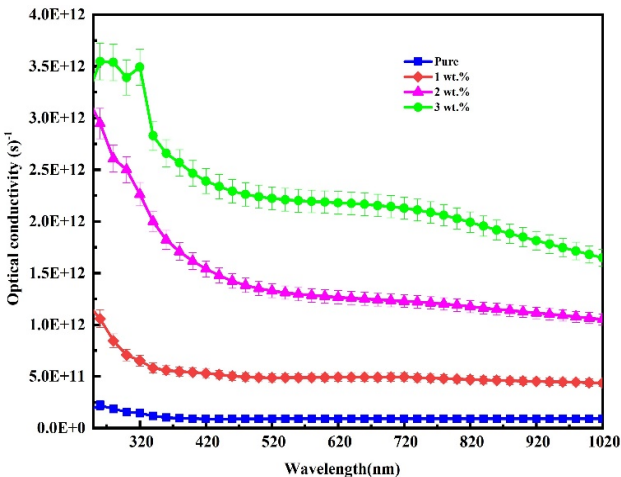

Figure 11. The optical conductivity of (PVA/CuO-In_2O_3) nanocomposites varies with wavelength

The optical conductivity of (PVA/CuO-In_2O_3) nanocomposites varies with wavelength, as shown in Figure 11. All of the prepared nanocomposites exhibit an increase in optical conductivity as the proportion of CuO-In_2O_3 nanoparticles grows. This is because the concentrations of nanoparticles increase, the average density of concentrated levels in the energy gap rises, which in turn raises the absorption coefficient and ultimately the optical conductivity of the nanocomposites (Table 3) [71, 72].

Table 3. Band gap energy and inhibition zone diameter for different PVA-based nanocomposites

Type of Nanocomposite	Band Gap (eV)	Maximum Diameter of Inhibition Zone (mm)
PVA/CuO-In_2O_3	1.887	24 current study
PVA/ZnO	-	24 [71]
PVA-CMC-SiO_2-Cr_2O_3	2.9	20 [27]

Figures 12-14 demonstrate that (PVA/CuO-In_2O_3) nanocomposites possess antibacterial properties against Streptococcus and Staphylococcus aureus. A higher concentration of (CuO-In_2O_3) nanoparticles results in a larger inhibitory zone. Their antibacterial action may be better understood if charge-negative bacteria are attracted to positively charged nanoparticles in nanocomposites by means of electromagnetic fields. Bacterial cells become oxidized and die instantly because of this process [73-75]. The release of electron-hole pairs from (CuO-In_2O_3) nanoparticles upon exposure to visible or ultraviolet light is another potential mechanism by which the nanocomposites exert their antibacterial activity [12, 76, 77]. The generation of reactive oxygen species (ROS) is a well-known and effective mechanism for the antibacterial effect of metal oxide-based nanomaterials. Although the mechanism proposed in this work is supported by previous results, direct experimental verification remains necessary to strengthen this hypothesis. Future studies suggest using specialized ROS detection assays, such as the DCFH-DA fluorescence assay, electron paramagnetic resonance (ESR), or chemical inhibitor tests, to directly confirm the generation of these reactive species.
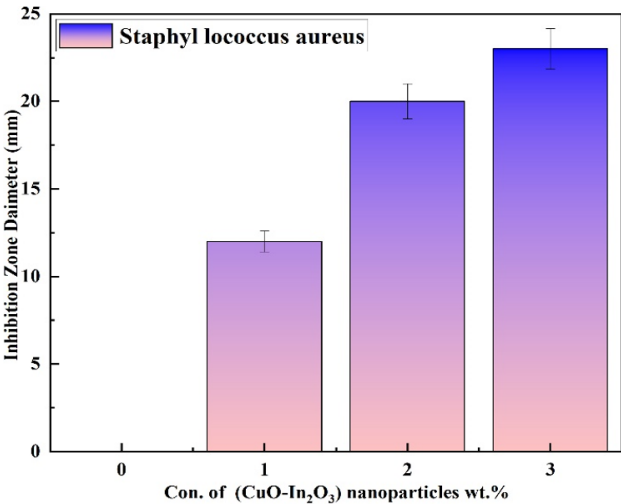

Figure 12. Inhibition zone diameter varying with (CuO-In_2O_3) nanoparticle concentrations against Staphylococcus for (PVA/CuO-In_2O_3) nanocomposites
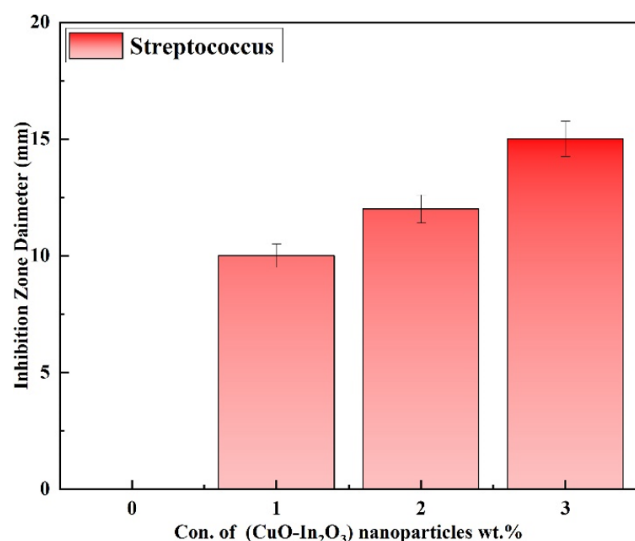
Figure 13. Inhibition zone diameter varying with concentrations of (CuO-In$_2$O$_3$) nanoparticles against Streptococcus for (PVA/CuO-In$_2$O$_3$) nanocomposites
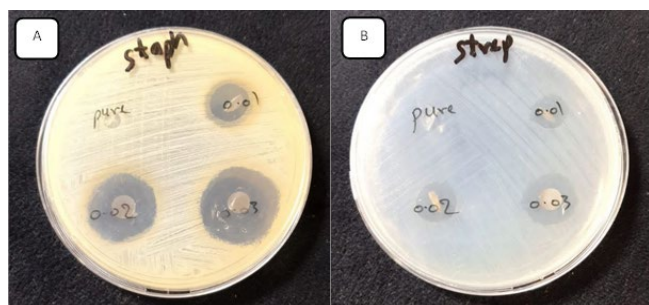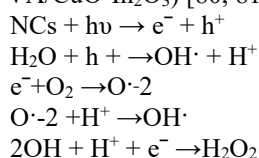

Figure 14. Antibacterial activity of (PVA/CuO-In$_2$O$_3$) nanocomposite. Zone of inhibition of (PVA/CuO-In$_2$O$_3$) nanocomposite against A: Staphylococcus, B: Streptococcus

Previous studies [9, 75] have demonstrated the effectiveness of such tests in confirming the ROS-based mechanism in similar nano systems. Therefore, the inclusion of these analytical techniques in future research will contribute to supporting the current results and clarifying the true role of reactive oxygen species in the antibacterial effect.

Interactions between these electron-hole pairs and water molecules can result in the formation of hydroxide (OH-) and hydrogen (+H) ions. Upon exposure to oxygen, nanocomposites generate peroxide anions (O$_2^-$), which combine with hydrogen ions (H$^+$) to form hydroperoxide (HO$_2$). This intermediate can further react with additional hydrogen ions (H$^+$) and electrons (e$^-$) to yield hydrogen peroxide (H$_2$O$_2$). The concentration of H$_2$O$_2$ molecules increases proportionally with the surface area of the nanoparticles [78, 79]. Bacterial cell walls may be permeable to these reactive oxygen species (ROS), which may cause damage and eventual death. The technique by which these nanocomposites are formed is called ROS generation by (PVA/CuO-In$_2$O$_3$) [80, 81]:

NCs + hυ → e$^-$ + h$^+$

H$_2$O + h + →OH· + H$^+$

e$^-$+O$_2$ →O·2

O·2 +H$^+$ →OH·

2OH + H$^+$ + e$^-$ →H$_2$O$_2$

Only hydrogen peroxide (H$_2$O$_2$) can cross bacterial membranes and destroy them. The fact that the membrane has negative charges makes it impossible for superoxide anions (O·2) and hydroxyl radicals (OH) to get through and stay on its surface. Nevertheless, oxidative stress is produced when light interacts with free radicals, which in turn inhibits the multiplication of bacteria.

4. CONCLUSIONS

This work includes synthesizing (PVA/CuO-In$_2$O$_3$) nanocomposites and investigating structural, optical and antibacterial activity. The scanning electron microscope reveals uniform and compact aggregates or clusters on the surface. FTIR analysis indicates that the surface structure of the pure polymer is uniform, without any signs of agglomeration or porosity. The observed reduction in peak intensity in the FTIR spectrum after the additive incorporation suggests an increase in the number of PVA polymer chains within the film structure. This implies that the polymers, after the addition, may have formed interconnected chain networks in the polymer nanocomposites. These structural changes are reflected in both the optical and electrical properties. Optical coefficients of PVA polymer (absorption and extinction coefficient) are increased with the increase of (CuO-In$_2$O$_3$) nanoparticle concentration. Absorbance, optical conductivity, and refraction of PVA polymer increase with the increase of (CuO-In$_2$O$_3$) nanoparticle concentration; the highest values are observed at 3% of (CuO-In$_2$O$_3$). At a concentration of 3%, the energy band gap of the nanocomposites reduces from 4.934 eV to 1.824 eV for (CuO-In$_2$O$_3$) nanoparticles. The synthesized (PVA/CuO-In$_2$O$_3$) nanocomposites demonstrated successful antibacterial activity against two distinct pathogenic bacterial strains, which could benefit the food, pharmaceutical, and beauty industries. The large optical bandgap decreases (from 4.934 to 1.824 eV) and high photoconductivity (~ 35%), making PVA/CuO-In$_2$O$_3$ nanocomposites promising applications in UV detectors and flexible optical sensors. Their antibacterial activity, with an inhibition zone diameter of up to 24 mm against Staphylococcus aureus and Streptococcus, qualifies them for use in medical coatings, dressings, and food packaging. The combination of optical and antibacterial properties gives them a competitive advantage over similar materials. This study gives to the presentation of the low-cost and easy synthesis process by PVA/CuO-In$_2$O$_3$ nanocomposites compare with other materials (such as PVA/ZnO).

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