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REVIEW ARTICLE

Investigation of optical and electrical properties of copper oxide - polyvinyl alcohol nanocomposites for solar cell applications

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Abstract Copper oxide nanoparticles (CuO-NPs) were synthesized at room temperature via a green method that is easy, efficient, eco-friendly, and low-cost. The aqueous extract of *Bougainvillea* leaves was used as a reducing and capping agent. Pure polyvinyl alcohol (PVA) and CuO-PVA nano-composite thin films of 13*wt*%, 30*wt*%, 40*wt*%, and 51*wt*% were prepared by the solution casting method. The optical and electrical properties of the prepared CuO-NPs, pure PVA, and CuO-PVA thin films were analyzed using a UV–visible spectrophotometer and Keithley electrometer.

The band-gap energy (Eg) value for the synthesized CuO-NPs was determined to be 2.74 eV. This band separation lies in a range that is suitable for a variety of optical and optoelectronic applications such as solar cells for energy conversion. Further, increasing the dispersion of CuO-NPs in a PVA matrix extended the absorption spectrum of PVA distinctly into the visible region and decreased the optical band gap exponentially from 4.42 eV for pure PVA to 3.34 eV for 51 *wt*% CuO-PVA. In addition, an increase in the DC electrical conductivity with CuO-NPs dispersion was observed. The decrease in the band gap energy was usually accompanied by an increase in the DC electrical conductivity making the CuO-PVA nanocomposite more applicable in thin-film solar cell applications.

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1. Introduction

Copper oxide nanoparticles (CuO-NPs) are semiconducting materials with direct band gaps and high absorption properties. They have been applied in many areas including magnetic storage media (Kumar et al., 2000), photodetectors (Wang et al., 2011), field emissions (Zhu et al., 2004), solar cells (Santos-Cruz et al., 2005), nanofluid (Zhou et al., 2010). Additional applications involve biosensors (Rahman et al.,2010), gas sensors (Choi and Jang, 2010), super-capacitors (Zhang et al.,2011), photocatalysis (Liu et al., 2012), antibacterial activity (Al-Fa'ouri et al., 2021), and antiviral properties (Zhou et al., 2019). CuO-NPs can cause the virus's genome to degrade and its integrity to be compromised. It was reported that CuO-NPs can attack the COVID-19 virus's membrane and damage the virus's DNA structure (Fakhroueian et al., 2020). CuO-NPs can be impregnated into a N95 mask to create an antiviral respiratory safe facial mask, which in turn enhanced the masks antiviral function by five orders of magnitude (Zhou et al., 2020).

The most common methods for fabricating nanomaterials can be broadly classified into two groups: top-down methods (physical methods) and bottom-up methods (chemical and bio-assisted). However, these methods have various limitations that can be decreased by the green synthesis method which is becoming popular because of its simplicity and low cost.

Furthermore, polyvinyl alcohol (PVA) is a synthetic polymer that has recently gained significant attention due to its particular physical and chemical properties. It is both a nontoxic and environmentally friendly water-soluble polymer. It is also oil, grease, and solvent resistant, has high dielectric strength, is ductile but rigid, elastic, and acts as an excellent oxygen and odor barrier. In addition, its effective film-forming nature has been employed in developing inorganic–organic nanocomposites with better performance properties than typically filled polymer composites, which are made by loading inorganic nanoparticles as additives in a polymer matrix (Abd El-Kader and Orabi, 2002). Furthermore, PVA is also utilized in the production of surface coatings, artificial sponges, cosmetics, pharmaceutical aids, and ophthalmic lubricants as well as other products.

Controlling the band gap is essential when developing new organic solar cell materials. Researchers tried to develop and synthesize new materials with the greatest possible overlap between their absorption and solar emission spectra (Bredas, 2014). The addition of nano-inorganic particles such as CuO-NPs will regulate these two properties of organic materials. In addition, the balance between light absorption and charge carrier transfer is a major challenge for metal oxide semiconductors in solar energy harvesting. Both are based on band structure and are combined to assess the final solar conversion efficiency. Therefore, studying the optical and electrical properties of CuO-PVA nanocomposites is very important to decrease the band gap and enhance the harvesting of solar energy (Divya et al., 2014; Tessema et al., 2018).

PVA has a relatively wide band gap that limits visible-light absorption and reduces light harvesting. Consequently, this makes it inappropriate to be applied as an active layer in organic solar cells. The band gap and optical absorption of a semiconducting material are significant factors for its use as an active layer in solar cells. Combining the properties of organic PVA and inorganic CuO-NPs would assist in developing new nanomaterials with enhanced physical and chemical properties that can resolve the limitations of their counterparts (Sperling, 2005). It has been reported that inorganic–organic nanocomposites enhanced the band gap and the optical absorption and therefore found potential applications in gas sensors, super capacitors, solar cells, and flexible electronic devices (Srikanth et al., 2016; Al-Harbi et al., 2023).

In the present study, a facile, non-toxic, and eco-friendly method was used for the preparation of CuO-NPs using *Bougainvillea* leaves aqueous extract. In addition, CuO-PVA nanocomposite thin films were prepared using the solution casting approach to investigate the effects of adding CuO-NPs at various concentrations to PVA on solar energy harvesting. Hence, studying the optical and electrical properties of the prepared CuO-NPs, pure PVA, and CuO-PVA thin films is the main objective of the present work.

In the literature, AC Conductivity and dielectric constant were studied without investigating any applications for this work (Manjunath *al.,* 2016). This is the first work done, according to our knowledge, to investigate the optical and electrical properties (DC-Conductivity) of copper oxide - polyvinyl alcohol nanocomposites for solar cell applications using the green route for the synthesis of copper oxide nanoparticles.

2. Experimental details

The experimental procedure to fabricate CuO-NPs from the *Bougainvillea* leaves and to prepare CuO-PVA nanocomposite films are presented in our previous study in which the green synthesized nanoparticles were subjected to characterization by XRD, FT-IR, and TEM, well crystalline, spherical shape and crystalline size in the range of 8–20 nm with an average size equal to 15 nm are presented (Al-Fa'ouri et al., 2021). The optical properties of the prepared CuO-NPs, pure PVA, and CuO-PVA thin films were studied and analyzed using a UV–visible spectrophotometer (Schimadzu 1601). The UV–vis absorption spectrum recorded for these samples is in the wavelength range of 200–800 nm. For the electrical properties, the two-probe technique with the aid of the Keithley electrometer (Model 6430 Sub-Femtoamp) was used to measure the temperature-dependent DC electrical conductivity for the pure PVA and the four CuO-PVA thin films over the temperature range of $T = 35 - 95^{\circ}C$. The thickness of each film has been measured using Swanepoel's envelope method and the results for pure PVA, 15 wt% CuO-PVA, 30 wt% CuO-PVA, 40 wt % CuO-PVA, 51 wt% CuO-PVA were 1730.84 nm, 1787.3 nm, 1996 nm, 2545.3 nm, and 2824.2 nm, respectively.

3. Results and discussion

The investigation of the optical absorption spectra in semiconducting materials provides essential information about the band structure and the associated energy gap. The optical band gap can be estimated from the absorption coefficient α of the films under investigation over a wide range of wavelengths (Tauc, 1970). The value of α specifies how far light of a specific wavelength can penetrate a material before being absorbed depending on the known Beer-Lambert's law $I = I_0 e^{-\alpha l}$, where I_0 is the incident intensity and I the transmitted one. It can be calculated from absorbance data using the relation

$$\alpha = \frac{2.303 \times A}{l}, \tag{1}$$

where A is the absorbance -which is the amount of light absorbed by the sample $[A = (I_0 - I)/I_0]$ - and l is the thin film thickness or the length of the light path through the sample. In a previous work (Al-Fa'ouri et al., 2021), the variation of α of the synthesized CuO-NPs with the wavelength has been found to have a high-intensity absorbance peak at 310 nm. Hence, a broad peak at 310 nm occurs in the UV–vis absorption spectrum. This peak indicates that CuO-NPs have maximum absorption in the ultraviolet range, which is an important optical characteristic for solar cell applications. In addition, α decreases exponentially in the visible region as the wavelength increases; it also decreases exponentially in the ultraviolet region as the wavelength decreases. Using the data that results from the optical absorption spectrum, Tauc plots have been used to determine the optical band gap E_g depending on the relation (Dahrul and Alatas, 2016)

$$\alpha h v = \beta (h v - E_g)^p, \tag{2}$$

where h is Planck's constant, v is the frequency of the incident photon, β is a parameter that depends on the transition probability, E_g is the optical band gap of the investigated film, while p is a parameter depending on both the type of transition (direct or indirect) and the profile of electron density in valence and conduction bands. In the above equation, $p = 1/2$ for an allowed direct, $p = 2$ for an allowed indirect, $p = 3/2$ for a forbidden direct, and $p = 3$ for a forbidden indirect transition. To determine the nature of the transition occurring, the value of p must be determined by plotting the relations $(\alpha h v)^{1/2}$, $(\alpha h v)^2$, $(\alpha h v)^{2/3}$, and $(\alpha h v)^{1/3}$ as functions of the incident photon energy hv (Lafi, 2016; Al-Harbi and Rajeh, 2022). The relation $(\alpha h v)^2$ vs hv for the synthesized CuO-NPs, presented in Fig. 1 is fitted quite well via Eq. (2) indicating that the absorption mechanism is identified as an allowed direct transition so $(\alpha h v)^2$ has been used; which fits well with the experimental result. The optical band gap (E_g) is obtained by extrapolating the linear portion of the plot to cross the point along the hv-axis at which $(\alpha h v)^2 = 0$ as shown in that figure. From this, the resulting value of the optical band gap of the synthesized CuO-NPs is determined from the intersection point to be $E_g = 2.74$ eV which is in good agreement with those obtained in the literature (Sarkar et al., 2020; Phang et al., 2021; Manjunath et al., 2016). This value of E_g makes CuO-NPs suitable for the solar conversion of a solar cell window.

In addition, UV–vis absorption spectra of pure PVA and four prepared CuO-PVA (13*wt*%, 30*wt*%, 40*wt*%, and 51*wt*%) nanocomposite films were recorded using a Schimadzu 1601 spectrophotometer in the range of 200–800 nm. These spectra are shown in Fig. 2. One can obtain from these figures that pure PVA films exhibit no apparent absorption in the visible light region (wavelength over 400 nm). However, films of PVA doped with different concentrations of CuO-NPs extend the absorption spectrum distinctly into the visible region with increasing concentration, as it appears from the absorption spectrum of 51*wt*% CuO-PVA. This result reveals that doping CuO-NPs into a PVA matrix enhances the harvesting of the visible part of solar radiation promising thereby a possible potential impact on solar-cell applications. The absorption spectra in Fig. 2 display that all films have relatively poor absorbance in the visible light region, and the absorbance rapidly increases with decreasing wavelength due to the direct transition of electrons.

Figs. 3-7 demonstrate the calculated band gaps of pure PVA and the four prepared CuO-PVA (13*wt*%, 30*wt*%, 40*wt*%, and 51*wt*%) nanocomposite films. CuO dispersion in the PVA matrix decreases the optical band gap from 4.42 eV for pure PVA to 3.34 eV for 51 wt% CuO- PVA. In other words, the optical band gap of the films decreases with the increasing concentration of the CuO nanofiller which is in good agreement with the literature ((Divya, et al.,2014; Radhakrishnan et al., 2014)). Apart from the predicted CuO-NPs aggregation, this could be due to the complex formation of the salt in the polymer matrix (hydroxyl groups of PVA). Besides, charge transfer transitions may cause a decrease in the optical band gap and an increase in the absorption spectrum (Divya et al., 2014a; Selvi, 2019). The shift in the absorption peaks in the UV region was employed to investigate the change in the optical energy band gap for pure PVA and

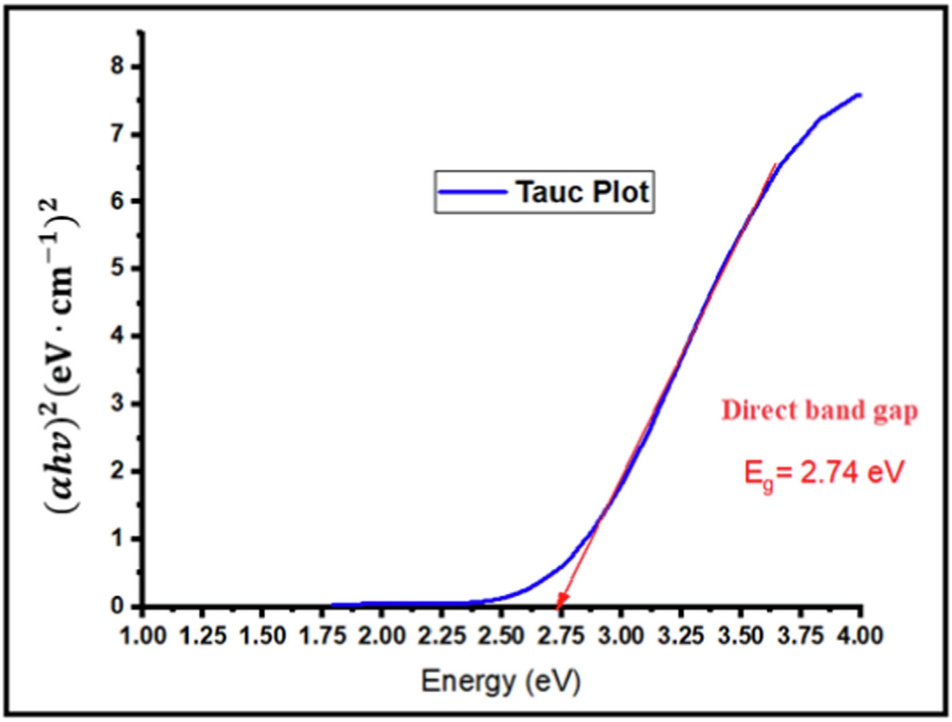

Fig. 1 Tauc plot for green synthesized CuO-NPs.

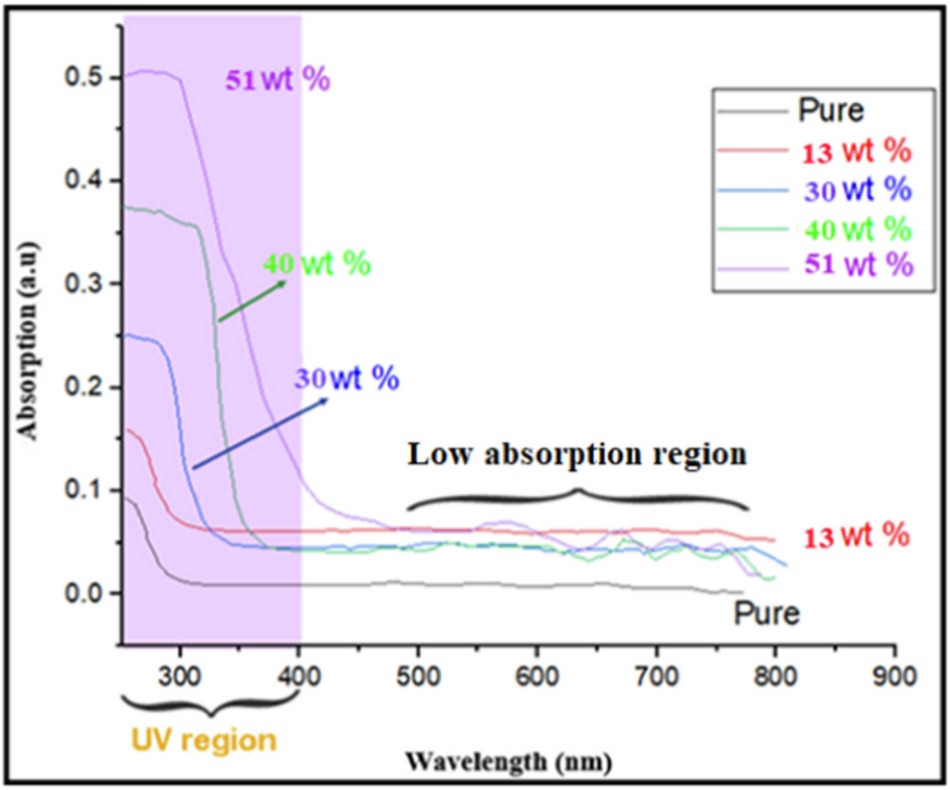

Fig. 2 UV–vis absorption spectra of pure PVA and four CuO-PVA (13*wt*%, 30*wt*%, 40*wt*%, and 51*wt*%) thin films.

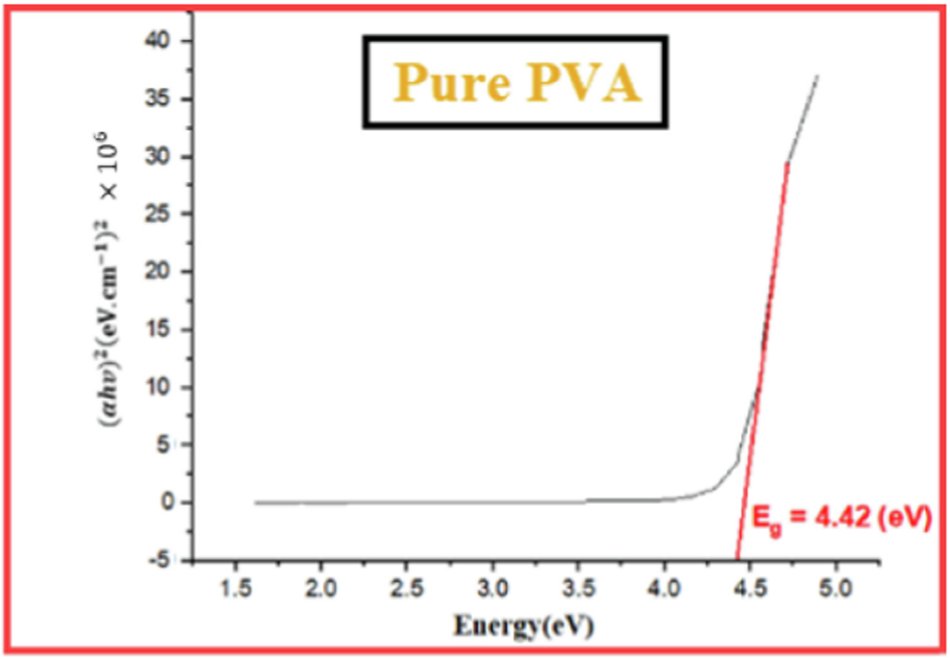

Fig. 3 Tauc plot for pure PVA film.

CuO-PVA nanocomposite thin films. A favorable band separation of CuO-PVA nanocomposite thin films of about 3.34 eV makes it suitable for solar cell applications. As a result, the optical band gap of CuO-PVA nanocomposite thin films can be tuned by changing the concentration of the CuO nanofiller in the PVA matrix, as well as changing the crystal size of CuO-NPs. This can be beneficial in the design of solar cell photoactive layers.

To explore the transport of charge carriers in the prepared samples, the DC electrical conductivity was measured at differ-

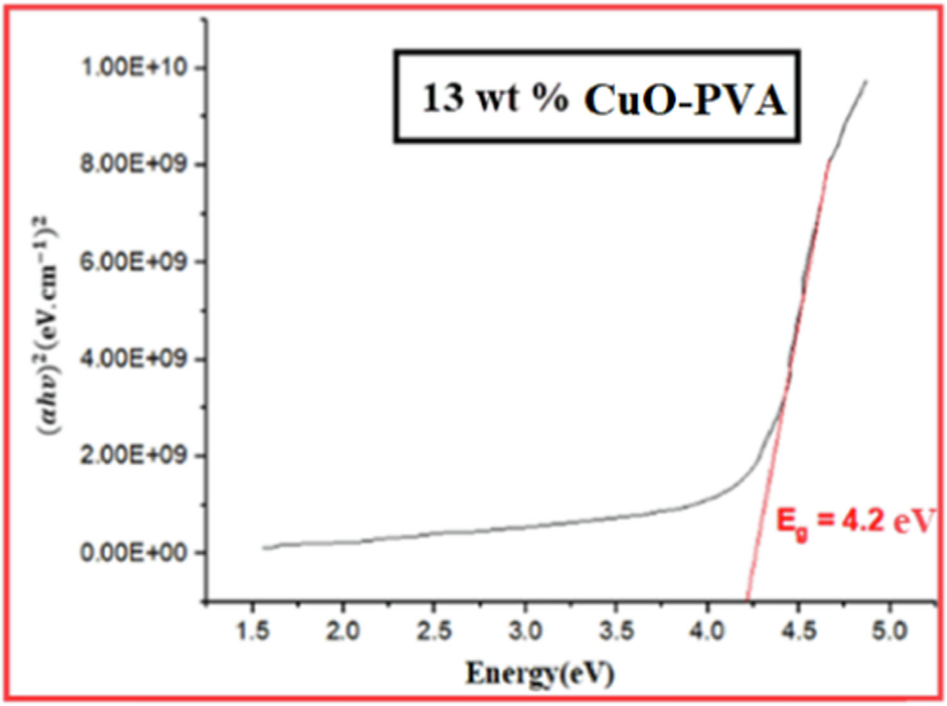

Fig. 4 Tauc plot for 13 wt% CuO-PVA film.

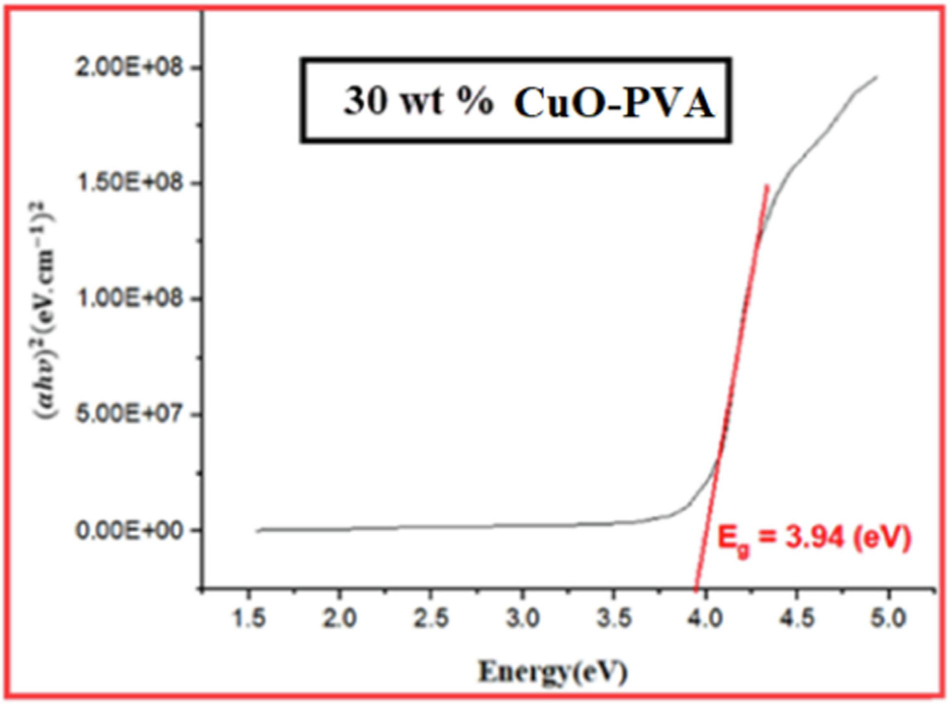

Fig. 5 Tauc plot for 30 wt% CuO-PVA film.

ent *temperatures* T in the case of the planner. The typical current–voltage I-V characteristics of the studied samples were recorded at different T, ranging from 35 °C to 95 °C, and in the voltage range (0–200 V). The I-V characteristics of the studied samples exhibit non-linear behavior, particularly at large electric fields. The DC electrical conductivity σ can be calculated using the simple relation

$$\sigma = \frac{IL}{VA} = \frac{IL}{V[W \times d]},$$

where I is the direct current through the thin film, V is the voltage drop across the film, L is the length of the film ($5cm$), A is the cross-sectional area $[W \times d]$, W being the width of the film ($2cm$) and d its thickness. Here, the values of I/V are obtained

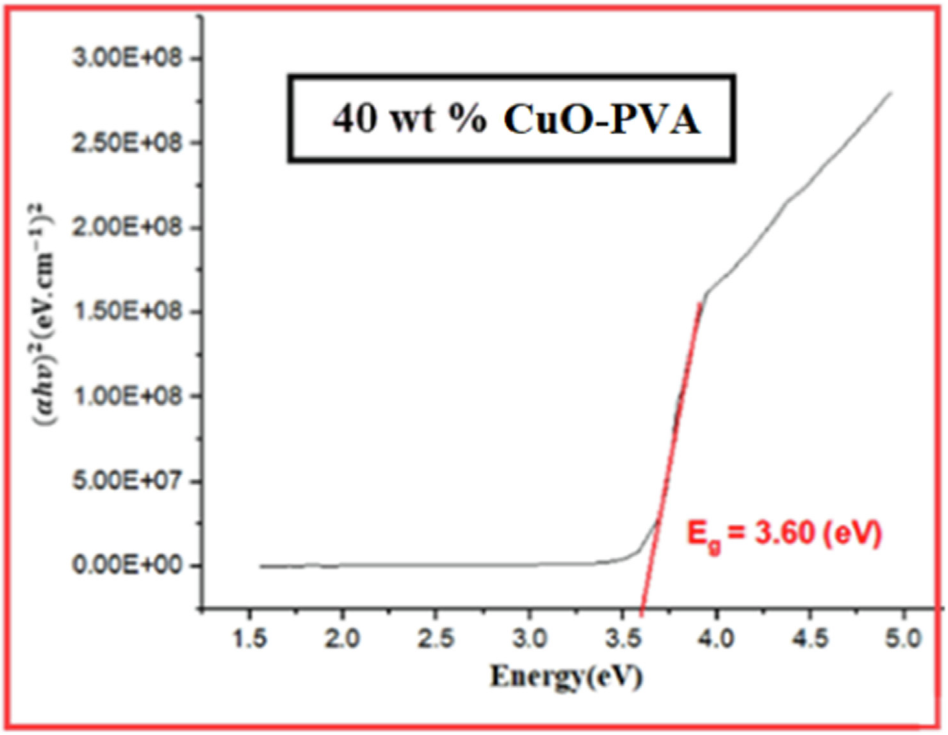

Fig. 6 Tauc plot for 40 wt% CuO-PVA film.

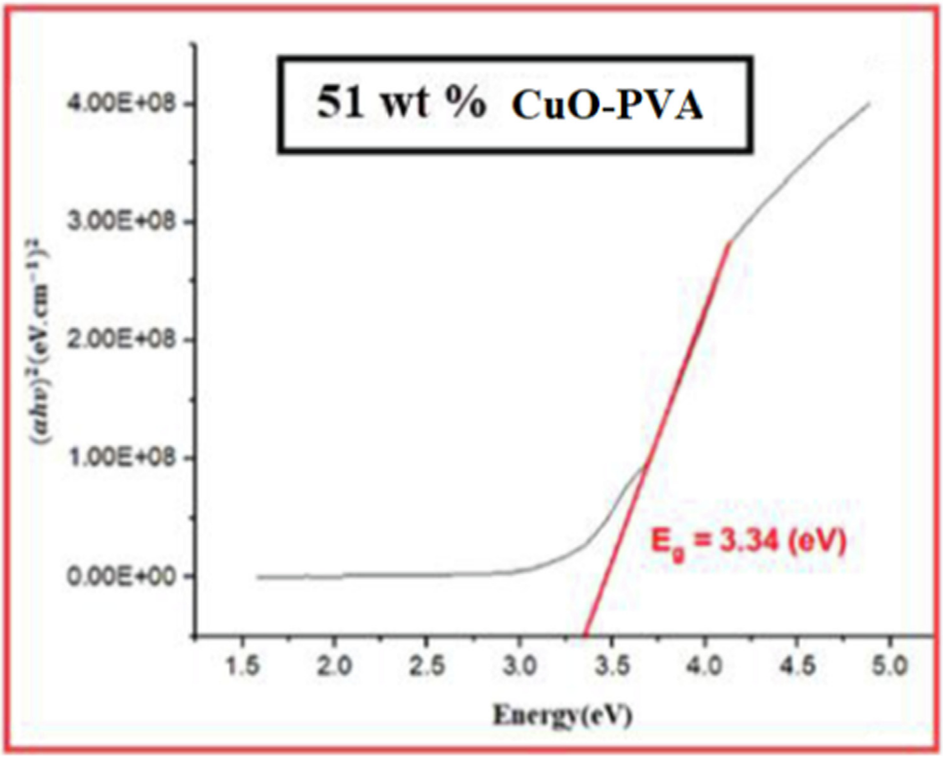

Fig. 7 Tauc plot for 51 wt% CuO-PVA film.

from Fig. 8. Note that σ versus T for pure PVA exhibits nonlinear behavior. Similarly, in the four CuO-PVA nanocomposites, I/V *curves* vary with T as shown in Fig. 8. It is clear from this figure that σ increases with T and with the concentration of the CuO nanofiller. The formation of charge-transfer complexes (CTCs) in the PVA polymer chain leads to the formation of new conducting routes inside the amorphous region of PVA, thus enhancing the conductivity (Mott & Davis.

1974). This implies that dispersing CuO nanofiller acts as a conductive filler in the PVA host matrix which considerably increases electronic conduction.

It is known that for semiconductors the conductivity follows the well-known Arrhenius equation (Mott and Davis, 1979)

$$\sigma = \sigma_0 exp^{(-E_a/k_B T)}, \tag{4}$$

where σ_0 is the electrical conductivity at T → ∞ including the charge carrier mobility and the density of states. E_a is the activation energy of conduction, and k_B is Boltzmann's constant. E_a is given by the slope of the plot of log(σ) vs $\frac{10^3}{T}$ as displayed in Fig. 9. The values of E_a are 0.0948, 0.0904, 0.0871, 0.0792,

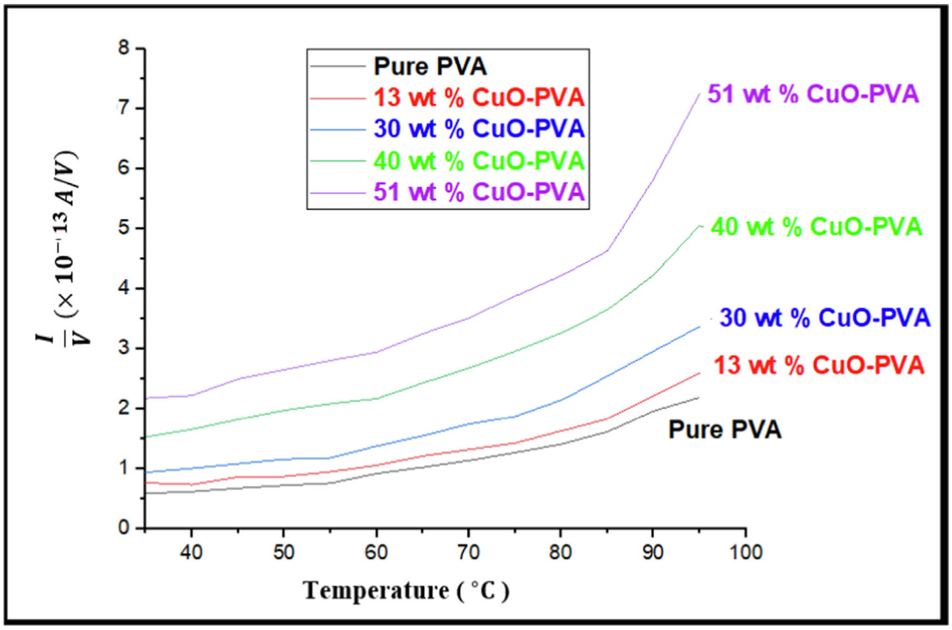

Fig. 8 Current / voltage I/V variation with temperature T for pure PVA and the four CuO-PVA nanocomposites films.

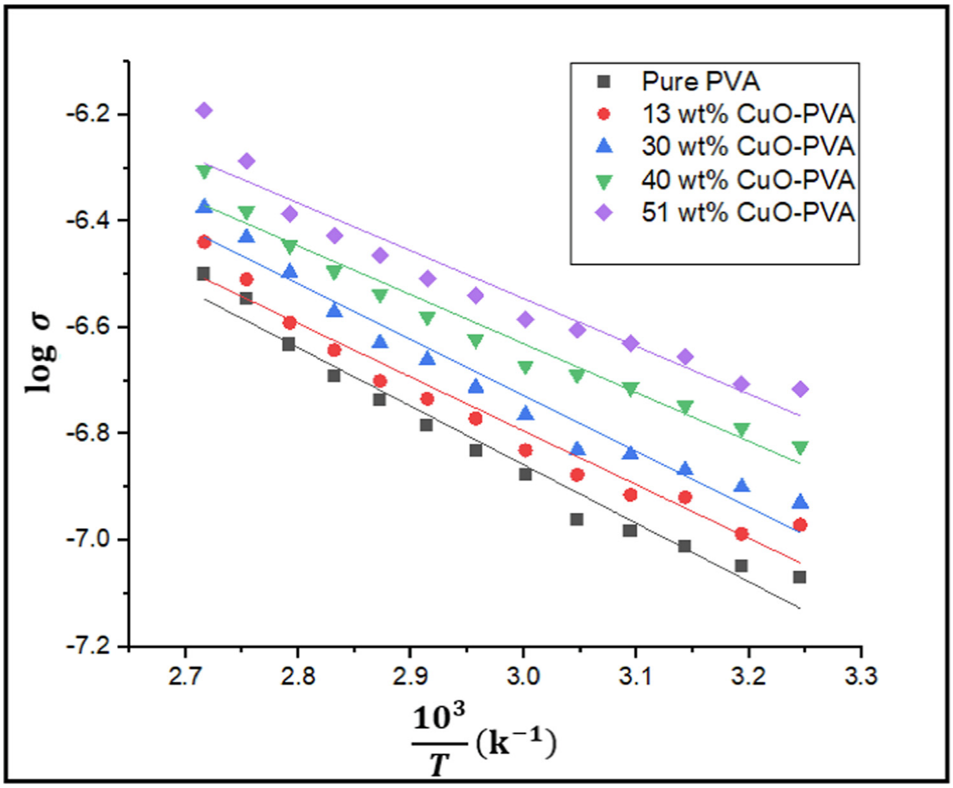

Fig. 9 log σ vs $\frac{10^3}{T}$ of the pure PVA and the four CuO-PVA nanocomposites.

and 0.0724 eV for 0,13*wt*%, 30*wt*%, 40*wt*%, and 51*wt*% samples, respectively. It is clear from these values that E_a decreases with the increasing concentration of CuO-NPs in the PVA host matrix. The decrease of E_a and the increase of electronic conduction result from dispersing the CuO nanofiller that acts as a conductive filler in the PVA host matrix as mentioned above. This agrees fairly well with the results obtained for the optical band gap. In the former case E_a represents the energy difference between the mobility edge and Fermi level or the difference between Fermi level and valence edge, that is $E_a = E_c - E_F$ or $E_a = E_F - E_v$. Usually, E_a is approximately half of the optical band gap E_g indicating that the Fermi level lies near the mid-gap ($E_g \approx 2E_a$) (Imran & Lafi, 2013). The E_a values are much smaller than the corresponding values of the half-width of the optical band gap (E_g = 4.42–3.34 eV). Such small values may be an indication that the conduction takes place because of the thermally-assisted charge carrier's movement in the band tails of the localized states (Imran and Lafi, 2013). As a result, the values of the optical and electrical parameters that have been experimentally obtained in this work are indicative of the fact that the optical band gap of CuO-PVA nanocomposite thin films and the electronic conductivity are enhanced by increasing the concentration of the CuO nanofiller in the PVA matrix. This in turn enhances its visible light absorption to develop a good candidate material for photoactive layers in solar cell applications, with more safety since NPs can be prepared using green synthesis.

DC conductivity electrical measurement showed that; as the temperature increases the DC conductivity increases. Also, DC conductivity increases gradually as the concentration of the CuO nanofiller increases. This implies that dispersing CuO nanofiller acts as conductive fillers in the PVA host matrix, and considerably increases electronic conduction. As a result, the produced CuO-PVA nanocomposites will be more useful in electronic and solar cell devices.

It would be interesting to study the effect of different sizes of CuO nanofiller on tuning and engineering the optical energy band gap of CuO-PVA nanocomposite. Incorporating a conductive component into nanocomposite and creating a hybrid composite material will improve the electrical conductivity of CuO-PVA nanocomposites, so it would be beneficial to study the effect of adding Cu or Au nanofiller on the absorption spectrum and band gap of CuO-PVA nanocomposites. It is recommended to study different coating techniques in future research to lower the manufacturing costs of photovoltaic systems.

4. Conclusion

Green synthesized CuO-NPs, pure PVA, and CuO-PVA nanocomposite thin films were prepared and characterized through spectrophotometry. The DC electrical conductivity σ at different temperatures T has been measured and analyzed using a Keithley electrometer. The key results and conclusions are as follows:

- The peak of the UV–vis absorption spectrum of synthesized CuO-NPs indicates that they have maximum absorption in the ultraviolet range and the estimated value of the optical band gap was determined to be E_g = 2.74 eV. This value of E_g makes CuO-NPs suitable for solar cell applications.
- The UV–vis absorption spectrum of PVA films showed that it extends distinctly into the visible region with increasing concentrations of CuO-NPs. This enhances the harvesting of the visible part of solar radiation that could promise a potential impact on solar cell applications.
- The values of the optical band gap for (0, 15, 35, 56, and 51) wt % CuO- PVA composite films were obtained to be (4.42, 4.20, 3.94, 3.60, and 3.34) eV, respectively. This means that the optical band gap of CuO-PVA nanocomposite films enhanced its visible light absorption.
- The DC electrical conductivity increases with the temperature and also gradually with the concentration of the CuO nanofiller. In addition, the variation of the estimated values of the activation energy gives the same trend as that of the optical band gap values.
- Results of this work showed that the prepared samples can be used to develop a good candidate material for photoactive layers in solar cell applications, with more safety since NPs could be prepared using green synthesis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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