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ORIGINAL RESEARCH ARTICLE

Characterization of the Structural and Optical Properties of Copper Oxide for Use in Solar Cells Using Screen Printing Method

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ABSTRACT
Structural and optical properties of copper oxide for solar cells applications was studied using screen printing techniques. Annealing procedure was employed in order to test the temperature effect. Samples were annealed at 150 °C and 200 °C for 30 minutes, the last sample was allowed to dry at room temperature. Scanning Electron Microscopy (SEM), UV-visible spectroscopy, and Fourier transforms infrared spectroscopy were used to describe the materials (FTIR). The results demonstrate that the annealing temperature had a substantial impact on the sample's structural properties. SEM analysis shows that the 150 °C annealed CuO sample have good morphology, no brittleness or cracking, and no gaps between crystal grains. The UV-visible spectrometer machine type Cary 50bio (0906m12) was used to analyze the absorption and percentage transmittance of the samples at room temperature, dry CuO sample and sample annealed at 150 °C have the strongest absorption in the visible band. The entire samples have a good transmittance for solar applications according to FTIR results. CuO sample annealed at 150 °C has a band gap 1.29eV which makes it good material for solar applications.

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Copper oxide, temperature, Absorption, annealing, Screen printing, Morphology

INTRODUCTION
Meeting the world's growing energy demand is one of the century's most pressing challenges. At the moment, combustible fossil fuels meet the majority of global energy demand, despite the fact that they are inexhaustible and have an irreversible negative impact on climate due to global warming and ecological degradation. As a result, the need for an alternative source of energy that is clean, inexpensive, and, most importantly, renewable is critical at this time (Halder *et al.,* 2019; Bhamu *et al.,* 2018). Energy scarcity and pollution have raised a red flag for humanity, urging us to abandon traditional energy sources in favor of green energy sources such as solar energy, wind energy, geothermal energy, and tidal energy, these energies are typically collected as electrical energy and delivered to users (He *et al.,* 2019; Jayan *et al.,* 2019). Because of its essentially clean and inexhaustible nature, solar energy has emerged as an appealing alternative source of energy.

Because of the development of novel applications such as energy devices, there is a growing emphasis on the preparedness and depiction of semiconductor oxide materials (Daoudi *et al.,* 2018), Copper oxides as semiconductors have been studied for more than three decades due to the abundance of constituent elements and their unique properties, which lead to a wide range of applications. Copper oxides are earth-abundant elements with a low cost and nontoxic nature. Because of their high absorbing properties, band gaps, excellent PEC characteristics, and high performance energy conversion, they are widely used in many fields (Balka *et al.,* 2018). The Cu-O system produces two well-known oxides: cupric oxide (CuO), which is black, and cuprous oxide (Cu_2O), which is red or slightly yellow. CuO and Cu_2O are both thought to be p-type semiconductors with visible and near-infrared band gap energies of 1.21 to 1.51 eV and 2.10 to 2.60 eV, respectively (Khodair *et al.,* 2020; Balka *et al.,* 2018; Fasasi *et al.,* 2018; Daoudi *et al.,* 2018). Copper oxides are used in many different applications, including gas sensors, glucose sensors, lithium batteries, and solar cell excetra. Based on their absorptance and emittance properties, copper oxide thin films could also be used as thermal control coatings (TCC)

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in the interiors of spacecrafts (Prasanth *et al.,* 2019; Khodair *et al.,* 2020; Vinothkumar *et al.,* 2019), Spin coating, thermal evaporation, spray pyrolysis, and chemical vapor deposition are some of the techniques for depositing thin films (Khodair *et al.,* 2020; Balka *et al.,* 2018; Vinothkumar *et al.,* 2019; Prasanth *et al.,* 2019).

Using the method of screen printing techniques, impact of annealing on the structural and optical properties of CuO for solar cells applications was investigated in this study. Sultana *et al.,* (2018) grew CuO thin films using the CBD method and annealed them in an argon (Ar) environment for 10 minutes at 250, 550, and 850 °C, respectively. The annealing temperature has been used to study the growth kinetics, structure, and photo-response of CuO thin films. They discovered that the annealing temperature has a significant impact on the morphology of the CuO film, the amount of CuO phase formed, as well as the crystallite quality and size of the nanostructures. This is due to the fact that it provides enough energy to allow atoms to move to the correct location in the crystal lattice. There was a change in refractive index and dielectric constant in the ranges of 2.65-2.93 (w) and 7.2-9.7(w), as well as a change in absorption coefficient and band gap from 1.33 x 10^5 to 6.06 x 10^5 cm^{-1} and 1.5 to 2.16 eV respectively. Alzaid *et al.,* (2020) was successfully prepared CuO nanoparticles with an average size of XRD 13.81 to 18.75 nm using the Co-Precipitation technique, with copper chloride dehydrate and sodium hydroxide as the precipitate agents. X-ray diffraction, UV-Visible absorption spectroscopy (UV), scanning electron microscopy (SEM), and infrared spectrum (FTIR) are used to characterize the CuO nanoparticles. CuO nanoparticles have irregular flake-like shapes, a non-uniform distribution, and a high purity.

According to the optical measurements of the CuO nanoparticles, the band gap was estimated to be 2.03, 2.05, and 2.15 eV. In the current work, annealing procedure was employed in order to test the temperature effect on CuO samples. Samples were annealed at 150 °C and 200 °C for 30 minutes, the last sample was allowed to dry at room temperature. The samples were characterised using Scanning Electron Microscopy (SEM), UV-visible spectroscopy, and Fourier transforms infrared spectroscopy (FTIR) i.e. to study the structural and optical properties of copper oxide samples for solar cells applications using screen printing techniques. This work is organized as follows: introduction in section 1, Experimental details in section 2, results and conclusions in sections 3 and 4 respectively, followed by acknowledgment and references at the end.

MATERIALS AND METHODS

Preparation of copper oxide powder paste
CuO powder paste was made by combining 8.17 g of 99% pure CuO powder with 5 ml of Polyethylene glycol (PEG) solution, CuO and PEG in a 1:1 by weight ratio, and stirring for 30 minutes to generate a CuO solution with a concentration of 0.41 m.

Deposition procedure.
Before deposition, the substrates were cleaned with Decon 90, then acetone, and finally ethanol. They were then blasted dry before being etched with a plasma cleaner. A microscope slide (glass slide) measuring 75 by 26 mm (3 by 1 inch) and 1mm thick was utilized in this experiment. Masking tape was utilized to cover any unwanted regions of the substrate, and the copper oxide powder paste was screen printed in 50 by 20mm squares on a glass slide substrate. Three samples of CuO were prepared using the same procedure in this experiment.

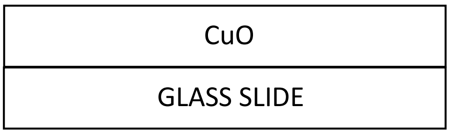

Figure 1: Substrate (Glass slide)

Annealing.
Annealing is the process of applying heat to the CuO layers of a thin film on a substrate (glass slide). The annealing procedure was utilized to accomplish the following goals: mending deposition errors, lowering layer resistivity, and enhancing crystal grain size (Yahaya *et al.,* 2022; AL-Hasan, 2010). CuO samples deposited on a substrate were heated at 150 °C and 200 °C for 30 minutes each in a heating furnance (Vecstar furnace, model spch12). Last sample was not annealed and allowed to dry at room temperature making it three sample's of CuO for studying in this work. (Yahaya *et al.,* 2022; AL-Hasan, 2010).

Sample characterization.
The sample was characterized by measuring absorption with a UV-visible spectrometer, the thickness of each layer with a mass-loss formula, transmittance with a UV-visible spectrometer, and the shape of the CuO material generated using a scanning electron microscope (SEM) were all studied (Yahaya *et al.,* 2022).
The mass added to the slide method is used to determine the thickness of the sample. The slide was weighted both before and after the film was placed to establish the approximate thickness of the film using the mass-loss formula (Yahaya *et al.,* 2022).

$$t = \frac{m}{A\rho} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

where
m = difference in masses before and after deposition of the sample's

A= Substrate deposition Area of measured in cm^2

ϱ = Materials density in gm/cm^3.

This thickness measurement method cannot provide an exact thickness of a deposited sample layer. To acquire an accurate thickness of a deposited sample, SEM micrographs taken in cross section, a stylus profilometer, spectroscopic Ellipsometry, or an X-ray reflectivity technique for a plat surface can be utilized (Yahaya *et al.,* 2022; Butt 2022; Elam *et al.,* 2003).

Using a focused beam of high-energy electrons, the scanning electron microscope (SEM) generates a variety of signals at the surface of solid materials. Electron signals give information on the sample's external morphology (texture), chemical composition, crystalline structure, and material orientation. Most of the time, data is collected across a defined area of the sample's surface, and a 2-dimensional image is generated to demonstrate spatial variations in these attributes (Yahaya *et al.,* 2022; Mohammed *et al.,* 2018). In a scanning mode, traditional SEM techniques can photograph areas spanning in width from 1 cm to 5 microns. SEMs are also often used to determine phases by doing qualitative chemical studies and/or examining crystalline structure. The SEM can also precisely measure very fine features and objects as little as 50 nm in size. A scanning electron microscope (SEM) was used to examine the morphology of all three CuO samples in this study (Yahaya *et al.,* 2022; Labroo *et al.,* 2022). The CuO samples' absorption and percentage transmittance were studied using UV-Visible Spectroscopy Analysis. The spectrum is a plot of the wavelength of the entire region versus the absorption (A) of light at each wavelength acquired by measuring the absorption of radiation by a sample at different wavelengths and mapping it using a recorder (Yahaya *et al.,* 2022; Tom, 2022). And the band gap of the sample can be estimated by plotting ((hv)^2 vs h) and extrapolating it along the x-axis. All three samples of CuO thin film prepared in this work are examined using a UV-visible spectrophotometer machine type Cary 50bio (0906m12) to evaluate absorption and percentage transmittance of the components utilized in this thin film (Yahaya *et al.,* 2022; Tom, 2022).

FTIR analysis can be used to analyze a wide range of materials, including bulk and thin films, liquids, solids, pastes, powders, fibers, and other materials. When proper standards are used, FTIR analysis can be used for both quantitative (quantity) and qualitative (identification) assessment of materials (Yahaya *et al.,* 2022; Berthomieu *et al.,* 2009; Nandiyanto *et al.,* 2019). FTIR can measure in bulk or the top 1 micrometer layer of materials with diameters of up to 11 millimeters. The FTIR spectra of pure compounds are typically so unique that they resemble a chemical "fingerprint." An Angilent Technology Cary 630 Fourier transform infrared (FTIR) spectrometer is used to perform FTIR analysis on all three CuO samples prepared in this experiment to measure the sample's transmittance. When kept at ambient temperature, the samples were analyzed in solid powder form (Yahaya *et al.,* 2022; Berthomieu *et al.,* 2009; Nandiyanto *et al.,* 2019; Khan *et al.,* 2022).This thickness measurement method cannot provide an exact thickness of a deposited sample layer. To acquire an accurate thickness of a deposited sample, SEM micrographs taken in cross section, a stylus profilometer, spectroscopic Ellipsometry, or an X-ray reflectivity technique for a plat surface can be utilized (Yahaya *et al.,* 2022; Berthomieu *et al.,* 2009; Nandiyanto *et al.,* 2019).

RESULTS

Prepared samples by method of screen printing.
After deposition of CuO powder paste on a substrate (glass slide) by method of screen printeing blackish film was produced.

Variance result based on annealing temperature and time.
150 ^{o}C annealed CuO sample, has a beautiful layer that has hardened without becoming brittle as a result of the proper annealing temperature. Because of the high temperature, 200 ^{o}C annealed CuO sample, has more brittleness on the substrate than the first sample, and the dry sample, has a more brittle condition and is not toughened due to the lack of annealing. Only the first of the three samples has a nice firm but not fragile coating.

Measurement of thickness result.
Mass difference equation (equation 1) was used in CuO samples thickness measurement of all the three samples.

1. Dry CuO sample.
m_1 = substrates mass prior to deposition = 5.408 g
m_2 = substrate mass after deposition = 6.482 g
Substrate mass difference before and after deposition
$m = m_2 - m_1 = 6.482$ g - 5.408 g = 1.074 g.
The density of the material used is, $\varrho = 6.315$ g/cm^3.
A= Film deposition area in cm^2 = 5cm x 2cm, = 10 cm^2.
Equation (1) was used to compute the thickness of a dry CuO sample.

$$t_1 = \frac{1.074}{10x6.315}$$

$$t_1 = 0.0170 \text{ cm}$$

2. 150 ^{o}C annealed CuO sample.
m_1 = substrates mass prior to deposition = 5.408 g
m_2 = substrate mass after deposition = 6.2968 g
Substrate mass difference before and after deposition
$m = m_2 - m_1 = 6.2968$ g - 5.408 g = 0.8888 g.
The density of the used material is, $\varrho = 6.315$ g/cm^3.
A= Film deposition area in cm^2= 5 cm x 2 cm = 10 cm^2.
Equation (1) was used to compute the thickness of 150 ^{o}C annealed CuO sample.

$$t_2 = \frac{0.8888}{10x6.315}$$
$$t_2 = 0.0140 \text{ cm}$$

3. 200 ^{o}C annealed CuO sample.
m_1 = substrates mass prior to deposition = 5.408 g

m_2 = substrate mass after deposition=6.1432 g
Substrate mass difference before and after deposition
$m = m_2 - m_1$ = 6.1432 g - 5.408 g = 0.7352 g.
The density of the used material is, ϱ = 6.315 g/cm^3.
A= Film deposition area in cm^2 = 5 cm x 2 cm, = 10 cm^2.
Equation (1) was used to compute the thickness of 200 ^{o}C annealed CuO sample.

$$t_3 = \frac{0.7352}{10x6.315}$$

$$t_3 = 0.0116cm$$

For all the CuO samples the thickness result is summarized in Table 1 below.

Table 1. Calculated thickness CuO samples.

Samples	Thickness (cm)
Dry CuO	0.0170
150 ^{o}C annealed CuO sample	0.0140
200 ^{o}C annealed CuO sample	0.0116

The results of thickness CuO samples calculated using the mass-loss formula are shown in the Table 1. the result shows that the higher the annealing temperature, the thinner the sample layer, which is an important aspect of solar cells because thinner layers absorb a higher percentage of solar energy, have higher efficiency, are more flexible, and are lighter in weight, all of which are desirable for thinner cells (Yahaya *et al.,* 2022).

Result of scanning electron microscope (sem).
The SEM morphology of the crystals in the sample is shown in the figures below. The Hitachi S – 4100 Scanning Electron Microscope was used to scan all three CuO samples, with an image magnification of x2000 and a power of 5 kv. To use the imaging machine, each sample was cut into 1 cm pieces.

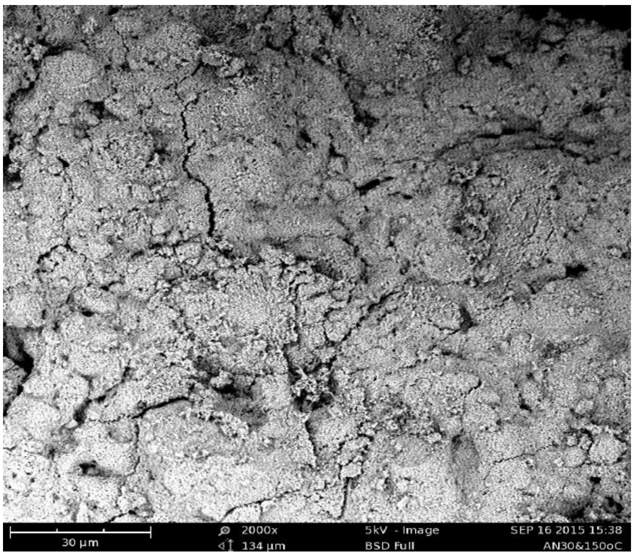

Figure 2: SEM micrograph of the dry CuO sample

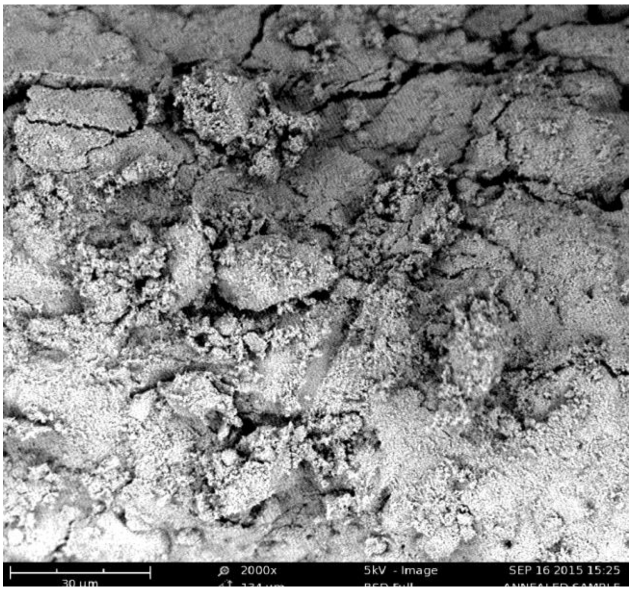

Figure 3: SEM micrograph of the 150 oC annealed CuO sample

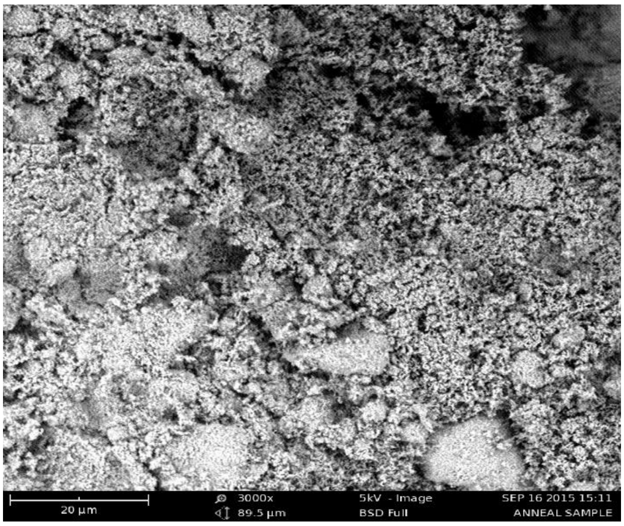

Figure 4: SEM micrograph of the 200^{o}C annealed CuO sample

Figure 2 shows the morphology of the dry sample, which reveals that the crystals are brittle and crystal compaction is low due to the lack of annealing completely. Figure 3 shows the morphology of a thin film solar cell that was annealed at 150 °C for 30 minutes. The morphology is good, with little brittleness or breaking, and there are few gaps between crystal grains. This indicates that the crystals are tightly packed together with little space between them. Figure 4 shows the morphology of a thin film solar cell annealed at 200 °C for 30 minutes; due to the use of an improper temperature and annealing time for the sample, the crystals in the samples generated are brittle and there is less compaction of the crystals. The temperature is higher, resulting in increased crystal brittleness and less crystal compaction. Gaps between the crystals are also likely to exist. The spaces between the crystals act as charge carrier traps, reducing the carriers' lifespan and hence increasing the resistivity of the materials deposited. The samples are annealed to lessen the effects of the crystal gaps. SEM analysis the samples reveals that the 150 °C annealed CuO sample has good morphology, less brittleness or breaking, and fewer gaps between crystal grains than the other two. The results of the SEM analysis in this study are comparable to those reported in the literature (Mohd *et al.,* 2013 and Roy *et al.,* 2015).

UV-visible spectroscopy analysis.
The investigation of the absorbed radiation and percentage transmittance of the annealed and dry CuO samples of the substrates were determined using UV – Visible spectroscopy.

1. Dry CuO sample.
2. 150 °C annealed CuO sample.
3. 200 °C annealed CuO sample

CuO samples produced were examined in a liquid form, and the powder was mixed with 3 milliliters of pure water. A recorder detects the sample's absorption radiation at different wavelengths (nm) and shows the spectrum, which is a plot of the wavelength (nm) of the entire region vs the absorption (A) of light at each wavelength (nm) (Yahaya *et al.,* 2022). The graphs below show the absorption and percentage transmittance findings obtained from UV - V spectrometer examination.

Absorbance graphs

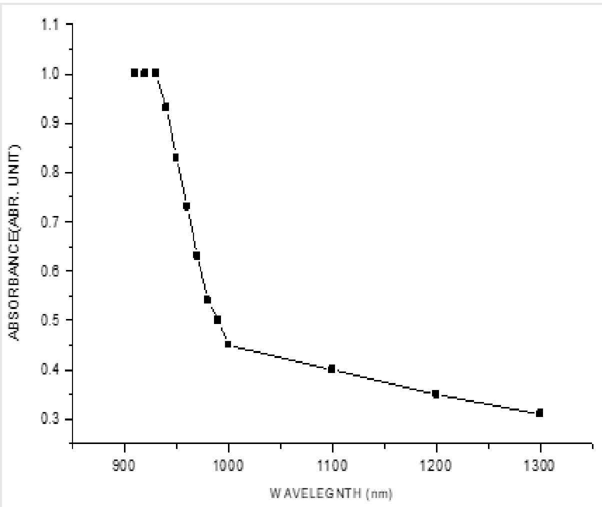

Figure 5: Dry CuO sample graph

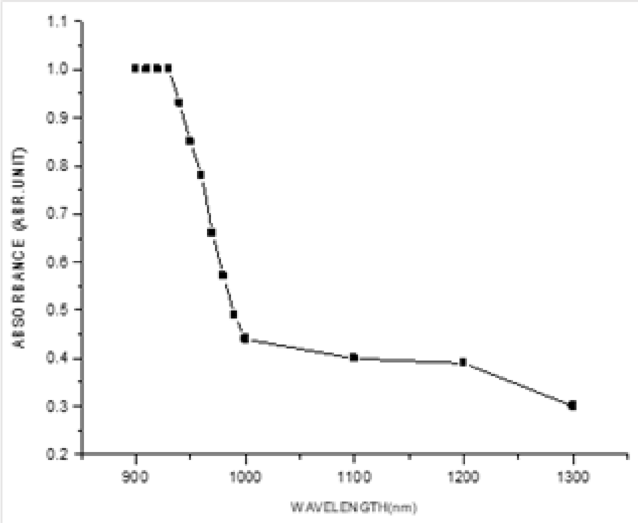

Figure 6: 150 °C annealed CuO sample graph

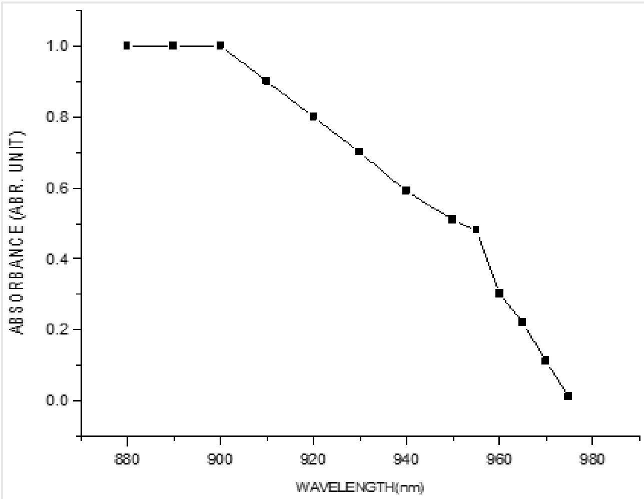

Figure 7: 200 °C annealed CuO sample graph

Transmittance graphs (%T)

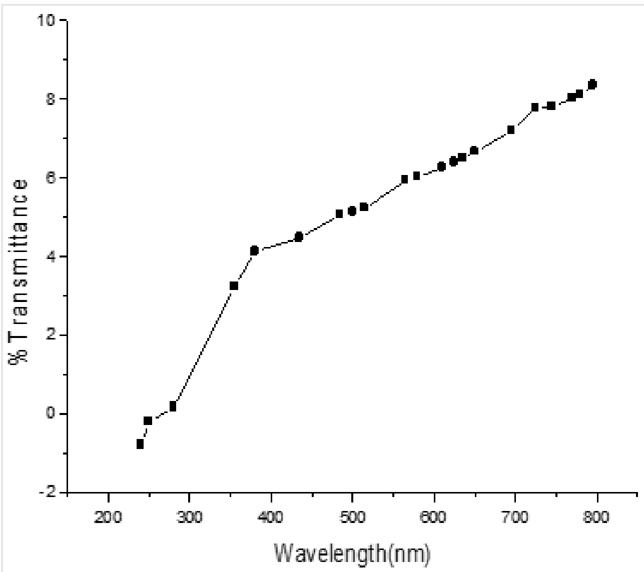

Figure 8: Graph of dry CuO sample

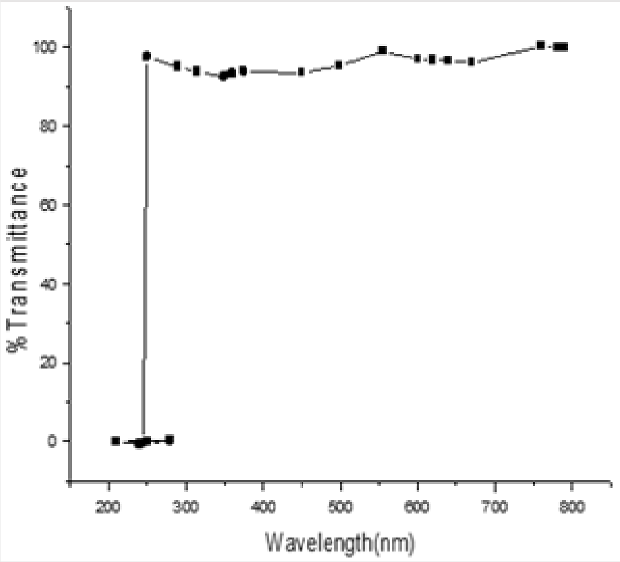

Figure 9: Graph 150 °C annealed CuO sample

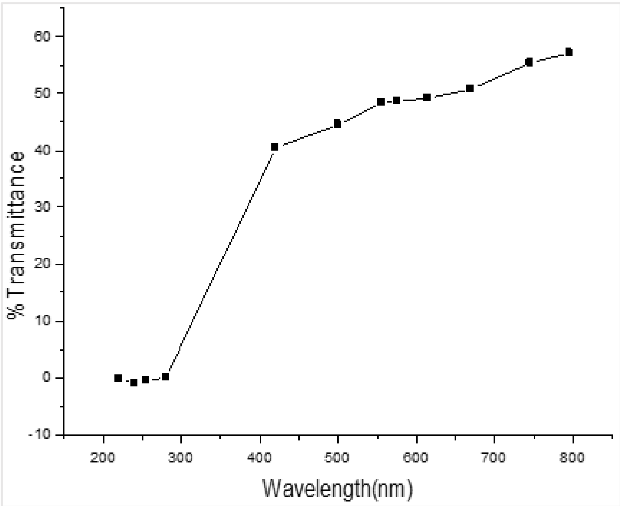

Figure 10: Figure 8: Graph 200 °C annealed CuO sample

At room temperature, the optical absorbance of the films was investigated in the wavelength range of 200–1300 nm. Using a spectrophotometer with UV, VIS, and NIR wavelengths Figures 5-7, show the fluctuation of absorbance (ABS) vs wavelength of each layer of the thin film sample (Yahaya *et al.,* 2022); we can observe from the figures that the absorbance values fell in general as the wavelength increased. Dry CuO, shows the highest absorption at wavelengths between 900 and 950 nm. CuO had the highest absorption at wavelengths ranging from 920 to 970 nm in the second sample, 150 °C annealed CuO sample. CuO shows the highest absorption at wavelengths spanning from 880 to 902 nm after being annealed at 200 °C for 30 minutes. 150 °C annealed CuO sample and 200 °C annealed CuO sample have substantial absorption at wavelength ranges of 920–970 nm and 880–902 nm respectively, making the samples appropriate for applications in solar cells. The optical absorbance in this investigation is comparable to those published in the literature by (Oluyamo, *et al.,* 2014; Bushra, *et al.,* 2014; Harish & Renu, 2013).

The optical% transmittance of the films was examined at room temperature in the wavelength range of 200-800 nm. Figures 8-10 demonstrate how the % transmittance of CuO thin films varies with wavelength as measured using a UV-VIS-NIR spectrophotometer. The percentage transmittance increases with wavelength and has a high transmittance in the NIR region, indicating that these films are appropriate for solar energy harvesting. CuO (Yahaya *et al.,* 2022), the dry CuO sample, has %T of 8.332 & at wavelength 795 nm. CuO exhibits a percent T of 99.836 % at wavelength 790 nm in the 150 °C annealed CuO sample. 200 °C annealed CuO sample, had a %T of 57.116 % at 795.0 nm wavelength. The CuO sample annealed at 150 °C for 30 minutes had the highest percentage transmittance at wavelength 790 nm, with 99.836 %. Furthermore, the percentage transmittance results demonstrate that some annealed materials have a greater % transmittance for solar cell application than dry CuO samples. The thin film's increased transmittance makes it ideal for solar energy collecting because it lowers solar radiation reflection and transmits it to the collector fluid when deposited on the collector's surface. The optical percentage transmittance result in this study is comparable to that published in the literature by (Samir & Riyam, 2013; Sandeep & Dhananjaya, 2012).

Calculated band gap energy

Figures 11–13 illustrate the band gap generated from calculated absorption coefficient values based on the absorbance result and sample thickness.

Dry CuO sample has 1.27eV band gap; 200 °C annealed CuO sample has 1.28 eV band gap; and 200 °C annealed CuO sample has 1.3 eV bandgap. The band gaps of the three CuO samples, are equivalent to those reported in the literature (Sultana *et al.,* 2018; Samir, *et al.,* 2013; Bushra, *et al.,* 2014).

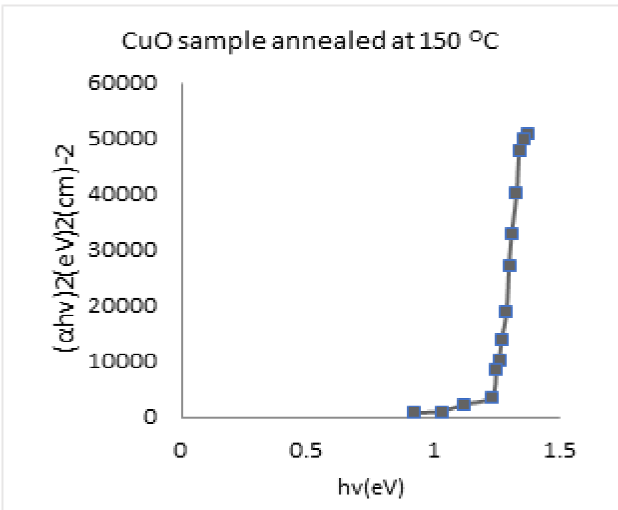

Figure 11: Dry CuO sample

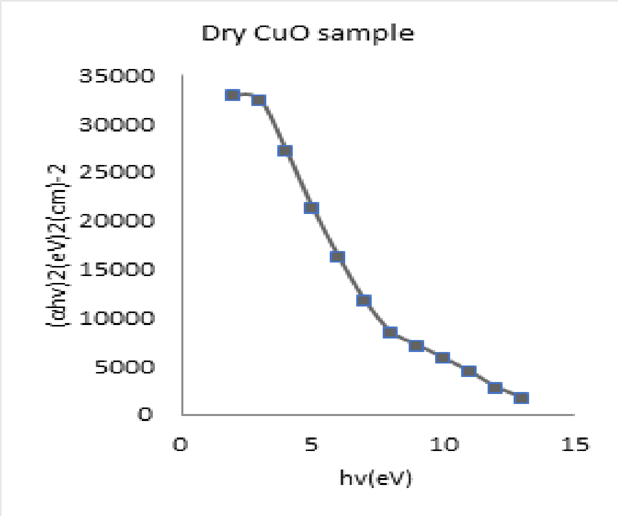

Figure 12: 150 °C annealed CuO sample

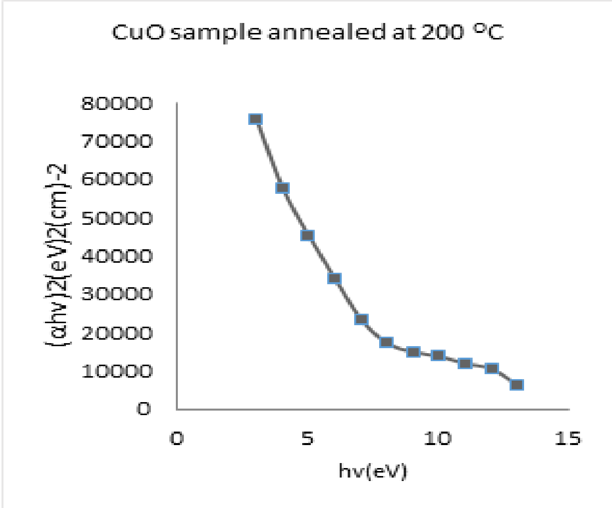

Figure 13: 200 °C annealed CuO sample

Analysis of Fourier Transform Infrared Spectroscopy (FTIR)

All of the samples were measured using an Angilent Technology Cary 630 Fourier transform infrared (FTIR) spectrometer. The materials were examined in solid powder at room temperature. The transmittance of the films was measured at room temperature in wave numbers ranging from 600 to 4000 cm-1. The deposited film spectra shown in Figures 14-16 depict the FTIR analysis outcome of this experiment, which looked at the transmittance of all tested specimens.

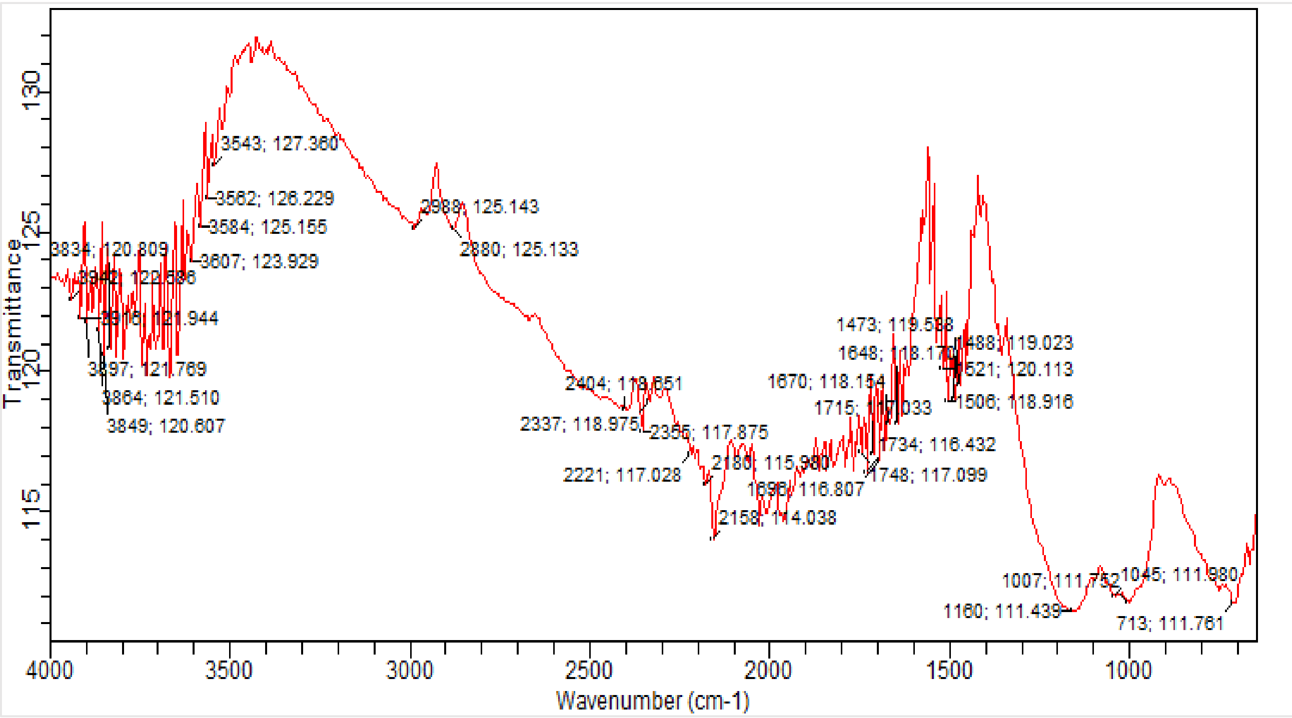

Figure 14: Dry CuO FTIR graph

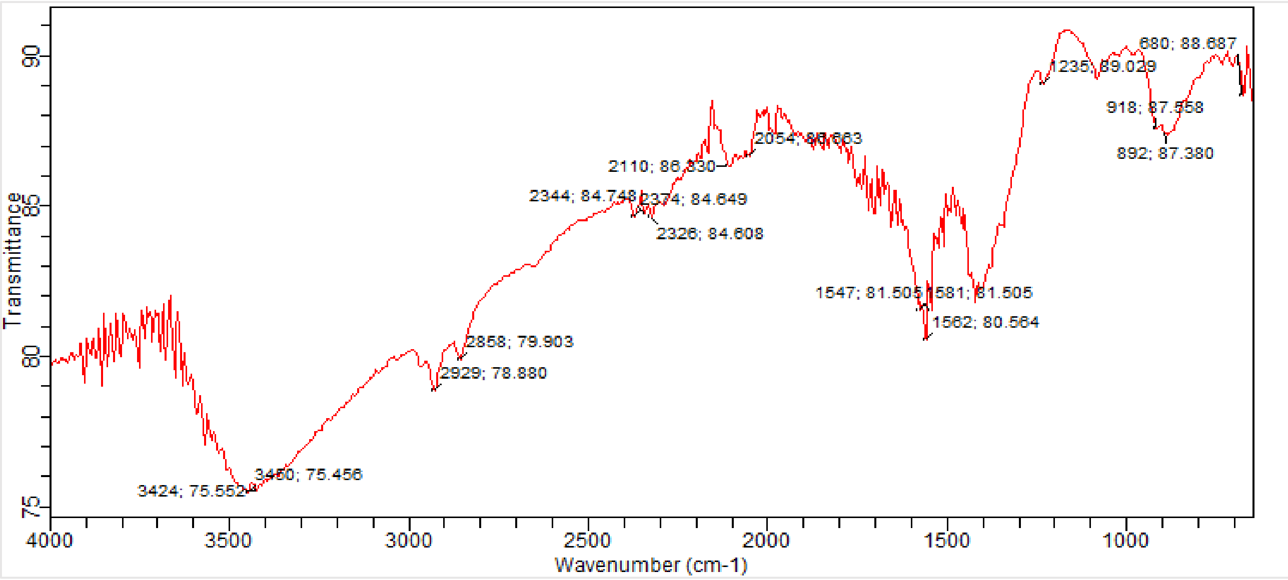

Figure 15: 150 °C annealed CuO sample FTIR graph

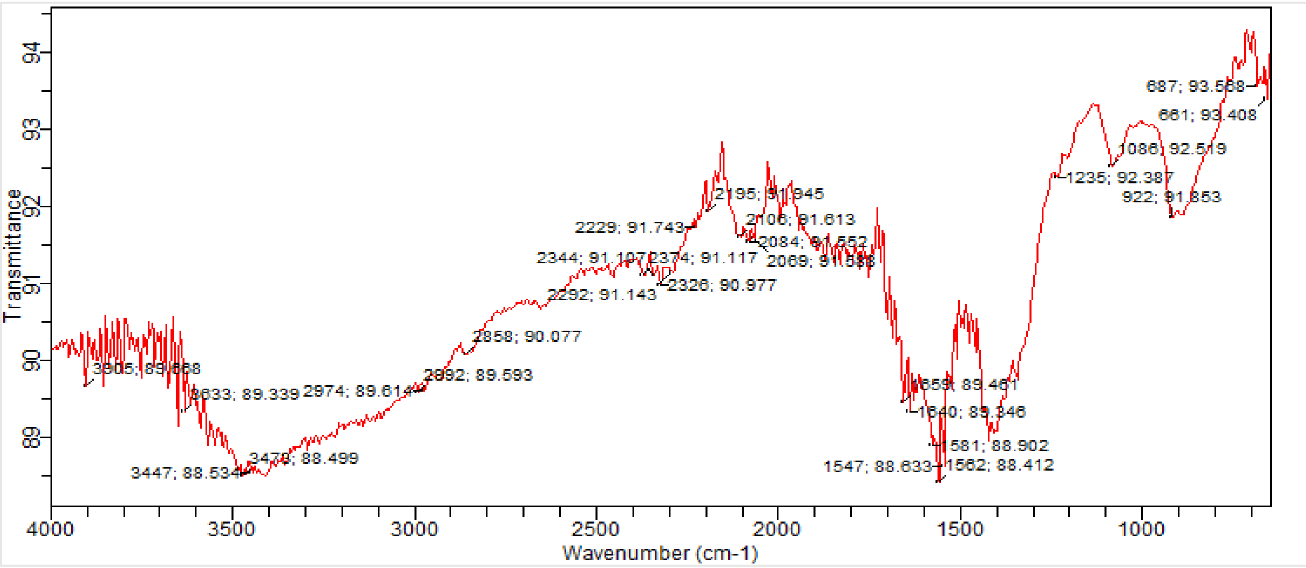

Figure 16: 200 °C annealed CuO sample FTIR graph

The FTIR analysis results are shown in the figures above. The transmittance of Dry CuO sample is 11.802 at 647 cm^{-1} wave number, while 150 °C annealed CuO sample has a 89.567 transmittance at wave number 640 cm^{-1}, and 200 °C annealed CuO sample has 93.408 transmittance at wave number 661 cm^{-1}. These results show the broad transmittance of copper oxide annealed at different temperatures per wave. The spectrum CuO has a prominent peak at 601 cm^{-1}, these results are equivalent to the literatures of (Amar, *et al.,* 2014; Amar, *et al.,* 2014; Asha, and Beena. 2014).

CONCLUSION

Screen printing techniques were utilized to investigate the optical and structural properties of CuO for usage in solar cells. CuO was successfully deposited on the substrate (glass slide), three samples were created, and the thickness of the samples was calculated using the equation of mass difference.
SEM was used to explore the morphology of the films, specifically the changes in changing film preparation circumstances, particularly the annealing temperature. On the sample annealed at 150 °C, the surface morphology of the deposited CuO thin film on glass substrate shows less brittleness and complete toughening.

Using UV–VIS Spectroscopy, the absorbance and % transmittance of thin film materials were determined, and the absorbance analysis result was used to determine the optical band gap of the thin film materials. According to FTIR spectroscopic investigation, the samples have good transmittance peaks for photovoltaic application.

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RECOMMENDATIONS

The result for this work indicates that more research should be carried out on CuO using the same techniques to gear the efforts toward the fabricating CuO solar cells, substrate thickness should also improve so that it can accommodate a fair amount of dye capable of capturing more light.

We recommend use of any alternative way of finding accurate thickness of film deposited.
The annealing process should be improved so that the band gap values match the expected values of CuO band gap.

The measurement of photo current, photo voltage, and power conversion efficiency characteristics should be carried out to ensure that the substrate can work properly towards achieving the desired goals.

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