Journal of Materials and Environmental Science
ISSN : 2028-2508
e-ISSN : 2737-890X
CODEN : JMESCN

J. Mater. Environ. Sci., 2026, Volume 17, Issue 9, Page 1427-1437

http://www.jmaterenvironsci.com

Synthesis and Characterization of Green Dye-Modified Copper Oxide Thin Films for Dye-Sensitized Solar Cell Applications

Idu H. K[1]*, Sule. M. M[1,2], Silas. E[3], Saidu. Y[4]

[1,,3,4]*Department of Physics, Taraba State University, Jalingo, Taraba State, Nigeria*
[2]*Department of Pharmacy Technician, Taraba State Polytechnic, Suntai, Taraba State*
Department of Science Laboratory Technology, Federal Polytechnic, Bali, Taraba State

**Corresponding author: iduhyacinthkevin@gmail.com*

Received *08 Aug 2026,*
Revised *07 Sept 2026,*
Accepted *08 Sept 2026*

Keywords:
- ✓ *Copper oxide;*
- ✓ *Green dye;*
- ✓ *Spray pyrolysis;*
- ✓ *Dye-sensitized solar cells;*
- ✓ *Photoanode*

Citation*: Idu H. K., Sule M. M., Silas. E., Saidu. Y., O. (2026) Synthesis and Characterization of Green Dye-Modified Copper Oxide Thin Films for Dye-sensitized Solar Cell Application, J. Mater. Environ. Sci., 17(9), 1427-1437*

Abstract: This study reports the synthesis and characterization of green dye-modified copper oxide (CuO) thin films prepared by spray pyrolysis for possible application as photoanode materials in dye-sensitized solar cells (DSSCs). CuO-based thin films were deposited on cleaned glass substrates at 50°C, 100°C and 150°C, with and without green dye incorporation. The films were characterized using scanning electron microscopy/energy-dispersive X-ray spectroscopy (SEM/EDS), UV-visible optical analysis, refractive index estimation, extinction coefficient, absorption coefficient, dielectric constant, Tauc band-gap analysis and optical conductivity evaluation. The results showed that deposition temperature strongly affected film morphology and optical behaviour. Undyed CuO films became more optically ordered with increasing substrate temperature, while green dye incorporation enhanced visible-light response, particularly within the 400–600 nm region. The dye effect was strongest at lower to moderate temperature and decreased at 150°C, likely because of thermal degradation or volatilization of the dye molecules. The findings suggest that moderate-temperature spray pyrolysis can produce low-cost CuO/green-dye thin films with tunable optical properties for DSSC and related optoelectronic applications.

1. Introduction

The increasing demand for sustainable, affordable and environmentally benign photovoltaic technologies has intensified research into dye-sensitized solar cells (DSSCs). Recent reviews indicate that DSSCs remain important among third-generation photovoltaic technologies because their performance can be tuned through semiconductor morphology, sensitizer chemistry, electrolyte composition and electrode design (Agrawal *et al.*, 2022), (Kokkonen *et al.*, 2021), (Rahman *et al.*, 2023), (Korir *et al.*, 2024). DSSCs are attractive because they can be fabricated at comparatively low cost, can function under low-light conditions and can incorporate different sensitizers (O'Regan & Grätzel, 1991; Grätzel, 2003; Hagfeldt *et al.*, 2010; Kim *et al.*, 2022). However, their application is still limited by issues such as dye degradation, charge recombination, electrolyte leakage, material stability and modest efficiency, which remain major concerns in DSSC development (Hagfeldt *et al.*, 2010; Kokkonen *et al.*, 2021; Rahman *et al.*, 2023; Abouri *et al.*, 2023; Korir *et al.*, 2024).

Copper oxide (CuO) is a promising semiconductor material for thin-film photovoltaic and optoelectronic applications because copper is abundant, relatively inexpensive and capable of supporting useful optical and electronic behaviour. CuO has also been reported as a p-type oxide semiconductor with optical band-gap and conductivity characteristics that are relevant to absorber layers, photoelectrodes and related solar-energy devices (Prakash *et al.*, 2022; Kuryshchuk *et al.*, 2022; Prakash *et al.*, 2024). CuO thin films prepared by spray pyrolysis have been reported to show temperature-dependent morphological, structural, optical and dielectric properties relevant to solar-cell applications (Diachenko *et al.*, 2021), (Abiodun et al., 2024; Zare Asl & Rozati, 2024). In DSSCs, the photoanode is central to charge transport because it accepts electrons from excited dye molecules and transfers them through the external circuit. Improving the morphology and optical response of CuO thin films can therefore contribute to better light harvesting and charge movement (Hagfeldt *et al.*, 2010; Diachenko *et al.*, 2021; Agrawal *et al.*, 2022; Prakash *et al.*, 2022).

Green dye sensitization is also important from a sustainability perspective because natural or green dyes are generally less toxic, readily available and potentially biodegradable. Natural pigments such as anthocyanins, betalains, chlorophylls and related plant-derived dyes have been investigated as low-cost sensitizers for DSSCs because they absorb within the visible region and can support photoinduced charge transfer (Calogero *et al.*, 2012; Sawhney *et al.*, 2017; Kim *et al.*, 2022; Shukor *et al.*, 2023; Rahman *et al.*, 2023). Studies on natural-dye and mixed-dye sensitizers further show that pigment combinations can broaden visible-light absorption and improve photon harvesting when dye adsorption, pH, concentration and interfacial stability are controlled (Patni *et al.*, 2020; Mujtahid *et al.*, 2022; Hendi *et al.*, 2023). When incorporated into a metal oxide thin-film matrix, dye molecules may introduce additional visible-light absorption centres. This study therefore investigated how substrate temperature and green dye incorporation influence the morphology and optical characteristics of CuO thin films prepared by spray pyrolysis.

2. Materials and Methods

2.1 Materials

The materials used in the study included Copper(II) sulfate (CuSO4), sodium hydroxide (NaOH), ammonia solution, distilled water, green dye, glass slides, beakers, measuring cylinders, analytical weighing balance, magnetic stirrer, calibrated hot plate, compressor, spray pyrolysis machine, oven, filter paper, funnel, stopwatch and other standard laboratory items. All chemicals were used as received without further purification.

2.2 Substrate preparation

Glass substrates were cleaned rigorously to remove organic residues, dust and adsorbed contaminants that would otherwise impair nucleation, adhesion and optical uniformity. The slides were washed with detergent, rinsed thoroughly with distilled water, degreased in acetone and subjected to additional rinsing with distilled water or ethanol. The cleaned substrates were dried and stored under clean conditions until deposition. Adequate substrate preparation is essential because residual organic matter or surface hydroxyl variation can alter nucleation density, film roughness and optical reproducibility.

2.3 Precursor preparation and film deposition

A 0.1 M copper precursor solution was prepared by dissolving the stoichiometric quantity of Copper(II) sulfate in distilled water under continuous magnetic stirring until a clear solution was obtained. Sodium hydroxide solution was added dropwise while stirring to generate copper hydroxide intermediate species that subsequently convert to copper oxide during thermal decomposition. Ammonia solution, when employed, assisted complexation or pH adjustment.

For the dye-modified series, a measured quantity of green dye was introduced into the precursor solution and the mixture was stirred until macroscopic homogeneity was achieved. The resulting solutions (undyed and dye-containing) were used immediately for deposition.

2.4 Thin film deposition by spray pyrolysis

Copper oxide and green-dye-modified copper oxide thin films were deposited onto the preheated glass substrates by pneumatic spray pyrolysis, a simple, atmospheric-pressure, scalable technique well suited to oxide thin-film growth. Substrate temperatures were maintained at 50 °C, 100 °C and 150 °C by means of a calibrated hot-plate. The precursor solution was atomized with compressed air and sprayed onto the heated substrates under identical geometric and flow conditions for all samples. After deposition the films were allowed to cool naturally to room temperature. The experimental matrix is summarized in Table 1.

Table 1. Experimental sample grouping used in the study.

Sample group	Deposition temperature	Modification
CuO	50 °C, 100 °C, 150 °C	Without green dye
CuO + green dye	50 °C, 100 °C, 150 °C	Green dye incorporated into precursor

2.5 Characterization

Surface morphology and elemental composition of the deposited films were examined by scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM/EDS). Optical properties were investigated using UV–visible spectrophotometry. Absorbance, transmittance and reflectance spectra were recorded over the relevant spectral range.

Optical band gaps were extracted by constructing Tauc plots of $(\alpha h\nu)^{1/2}$ versus photon energy $h\nu$, appropriate for the indirect allowed transitions commonly observed in CuO, and extrapolating the linear portion of the absorption edge to the energy axis. All optical analyses followed established procedures for spray-pyrolysed oxide thin films.

3. Results and Discussion

3.1 SEM morphology

Scanning electron microscopy was used to examine the surface morphology of the deposited films (Figure 1a–f). A clear dependence on both substrate temperature and dye incorporation is evident. The undyed films (Figure 1a–c) show progressive improvement in surface continuity and compactness with increasing substrate temperature. At 50 °C the surface is irregular and contains discrete agglomerates, indicating incomplete precursor decomposition and limited surface diffusion. Deposition at 100 °C yields a more homogeneous and continuous film, while the film prepared at 150 °C is denser and more compact. These trends are consistent with earlier reports on spray-pyrolysed CuO, in which substrate temperature strongly influences grain development, packing density and residual porosity (Diachenko et al., 2021; Prakash et al., 2022; Zare Asl & Rozati, 2024). The dye-modified films (Figure 1d–f) exhibit additional surface features relative to their undyed counterparts. Altered particle aggregation and increased topographic contrast are observed, particularly at the lower deposition temperatures.

These changes are attributed to the interaction of dye molecules with the growing oxide matrix during film formation.

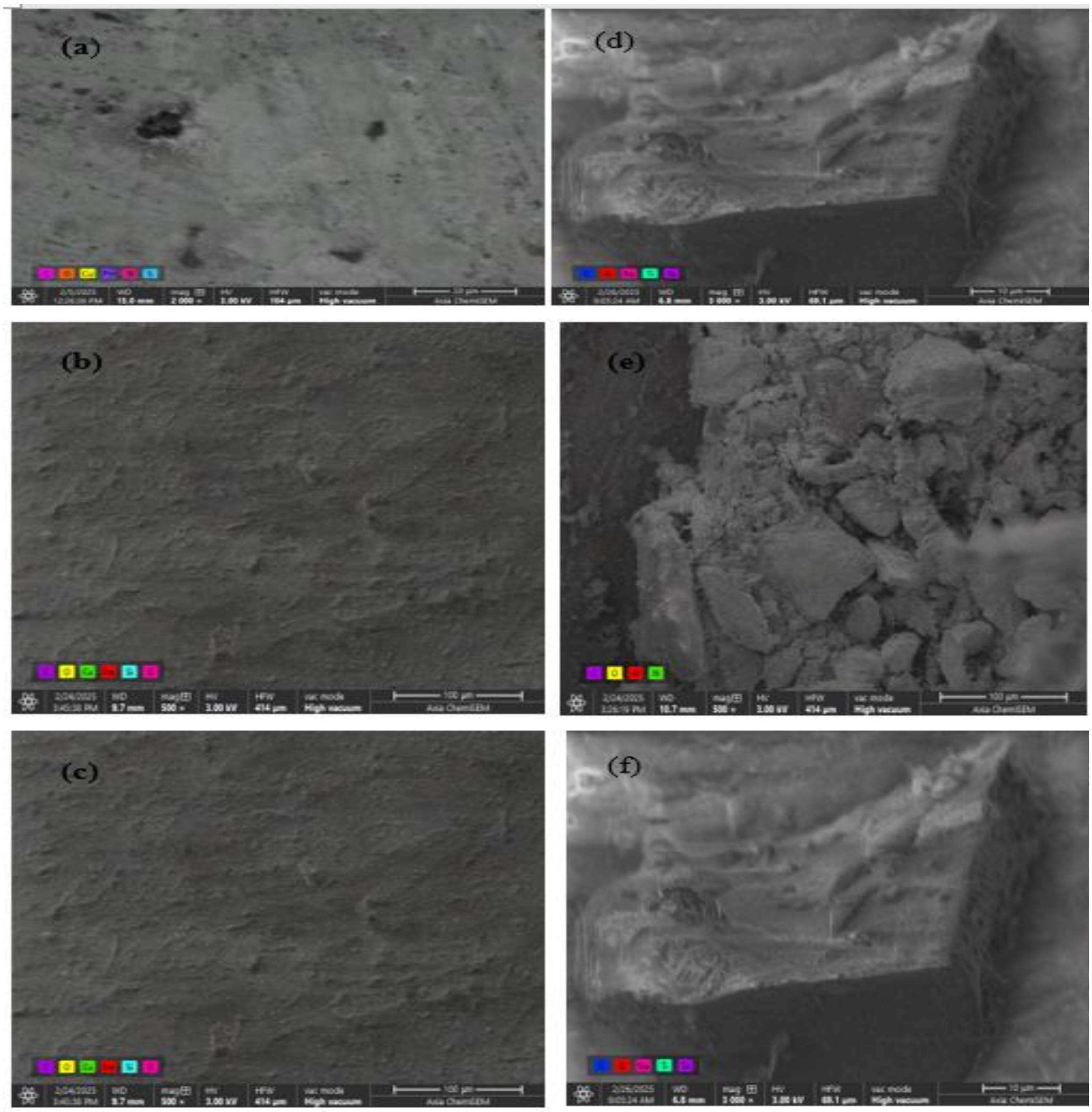

Figure 1. SEM images of CuO thin films deposited at (a) 50°C, (b)100°C and (c)150°C and green dye-modified CuO thin films deposited at (d) 50°C, (e)100°C and (f)150°C.

As shown in Table 2, the undyed CuO films contain copper and oxygen together with silicon, the latter arising from the glass substrate because the electron-beam interaction volume extends through the thin film and carbon remains low (<1 %). In contrast, the dye-modified films show a marked increase in carbon content (17.4–20.8 %), confirming incorporation of the organic dye. The highest carbon percentage occurs at 100 °C, coinciding with the strongest visible-light absorption. Oxygen is also elevated in the dye-modified series. Sodium, aluminium and nitrogen appear selectively and may originate from the substrate or residual dye components; occasional titanium and tantalum signals are most likely peak-assignment artefacts. Overall, the EDS data support the morphological observations and the optical evidence of successful dye incorporation at moderate temperature.

Table 2. Elemental composition (EDS) of undyed CuO and green-dye-modified CuO thin films deposited by spray pyrolysis at 50 °C, 100 °C and 150 °C.

Element	CuO 50 °C	CuO 100 °C	CuO 150 °C	CuO/dye 50 °C	CuO/dye 100 °C	CuO/dye 150 °C
C	0.9	0.5	0.5	17.4	20.8	17.4
O	7.2	7.6	7.6	43.0	49.9	43.0
Si	2.2	1.3	1.3	–	–	–
S	0.0	0.3	0.3	–	–	–
Cu	2.7	3.8	3.8	–	–	–
Pm*	86.5	86.5	86.5	–	–	–
Na	–	–	–	4.6	–	4.6
Ti	–	–	–	26.0	–	26.0
Ta	–	–	–	9.0	–	9.0
N	–	–	–	–	2.5	–

Note. *Values are expressed as weight percent (Wt. %). Dashes (–) indicate that the element was not detected or was below the quantification limit. *The high Pm signal is a typical and most likely a software peak-assignment artefact.*

3.2 Absorbance behaviour

The absorbance spectra of the undyed CuO films (Figure 2a) are characterised by strong absorption in the ultraviolet region, with a pronounced increase between approximately 300 nm and 400 nm. This feature arises from electronic transitions within the CuO lattice and is typical of copper-oxide thin films prepared by chemical spray pyrolysis (Diachenko *et al.*, 2021; Prakash *et al.*, 2022; Kuryshchuk *et al.*, 2022; Zare Asl & Rozati, 2024). Absorbance intensity rises systematically with substrate temperature; the film deposited at 150 °C exhibits the highest values across the measured spectral range. The enhancement is ascribed to improved structural order, increased packing density and a concomitant reduction in defect-related optical losses. Incorporation of green dye produces a substantial increase in absorbance, particularly in the visible region between 400 nm and 600 nm (Figure 2b).

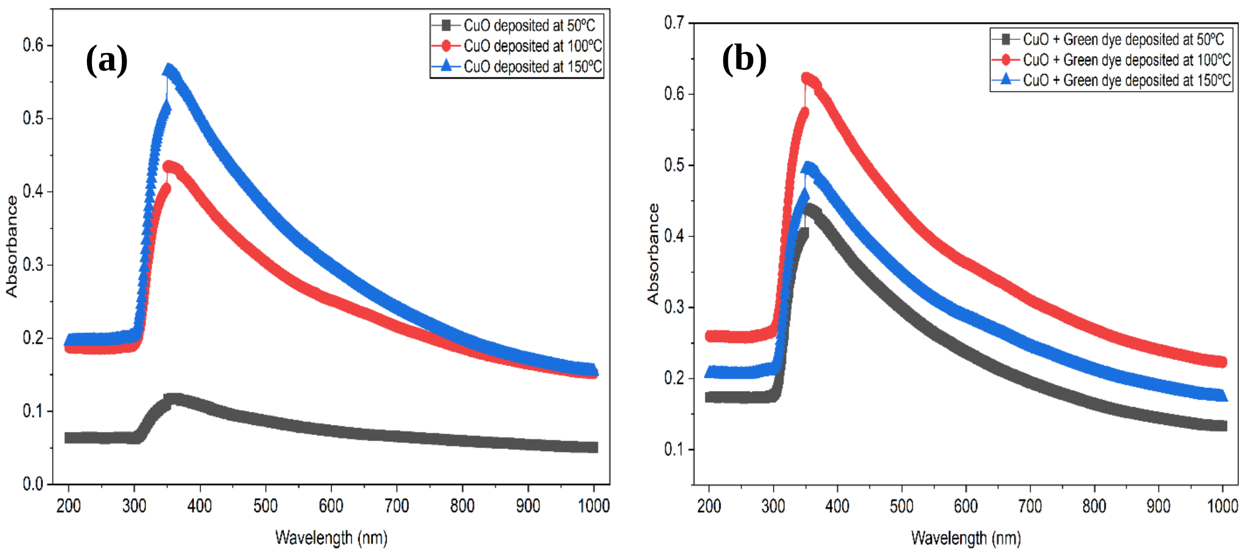

Figure 2. Absorbance versus wavelength of CuO (a) and (b) green dye-modified CuO thin films deposited at 50°C, 100°C and 150°C.

The additional absorption confirms that the dye introduces chromophores capable of harvesting visible photons, thereby extending the spectral response of the composite film. Among the dye-modified samples, the film deposited at 100 °C displays the highest absorbance, whereas the film prepared at 150 °C shows a relative decrease. The diminution at the highest temperature is consistent with partial thermal degradation or volatilisation of the organic dye, a limitation frequently reported for natural-dye sensitizers (Patni *et al.*, 2020; Mujtahid *et al.*, 2022; Hendi *et al.*, 2023). The observed dye-induced extension of visible-light absorption is in accordance with the established behaviour of plant-derived pigments in DSSC photoanodes (Calogero *et al.*, 2012; Sawhney *et al.*, 2017; Kim *et al.*, 2022; Shukor *et al.*, 2023; Rahman *et al.*, 2023).

3.3 Transmittance and reflectance behaviour

Transmittance spectra (Figure 3) are complementary to the absorbance data. All films exhibit low transmittance in the ultraviolet region as a direct consequence of the strong CuO absorption edge. In the visible range, the transmittance of the undyed films (Figure 3a) increases with deposition temperature, reflecting the progressive improvement in structural quality and the associated reduction in scattering centres. The dye-modified films (Figure 3b) display lower visible transmittance than the corresponding undyed layers because a larger fraction of incident photons is absorbed by the incorporated dye. The reduction is most pronounced for the specimen deposited at 100 °C, reinforcing the conclusion that this intermediate temperature optimally preserves the optical contribution of the sensitizer.

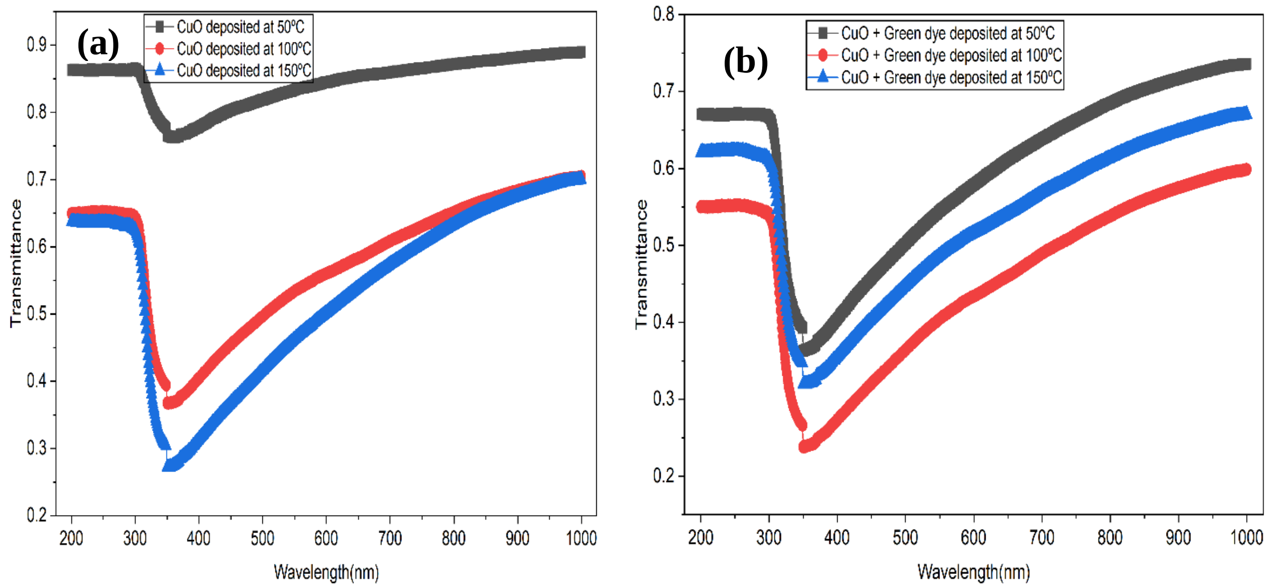

Figure 3. Transmittance versus wavelength of (a) CuO and (b) green dye-modified CuO thin films deposited at 50°C, 100°C and 150°C.

Reflectance spectra (Figure 4a-b) provide further insight into the optical response. Green-dye incorporation generally lowers reflectance across the visible spectrum at the lower and intermediate deposition temperatures, thereby increasing the proportion of light available for absorption. At 150 °C the reflectance of the dye-modified film rises again, consistent with a diminished contribution from the organic component. These optical trends underscore the necessity of balancing light-harvesting efficiency against film morphology and interfacial stability when the layers are considered for DSSC photoanode applications (Agrawal *et al*., 2022; Kokkonen *et al*., 2021; Korir *et al*., 2024).

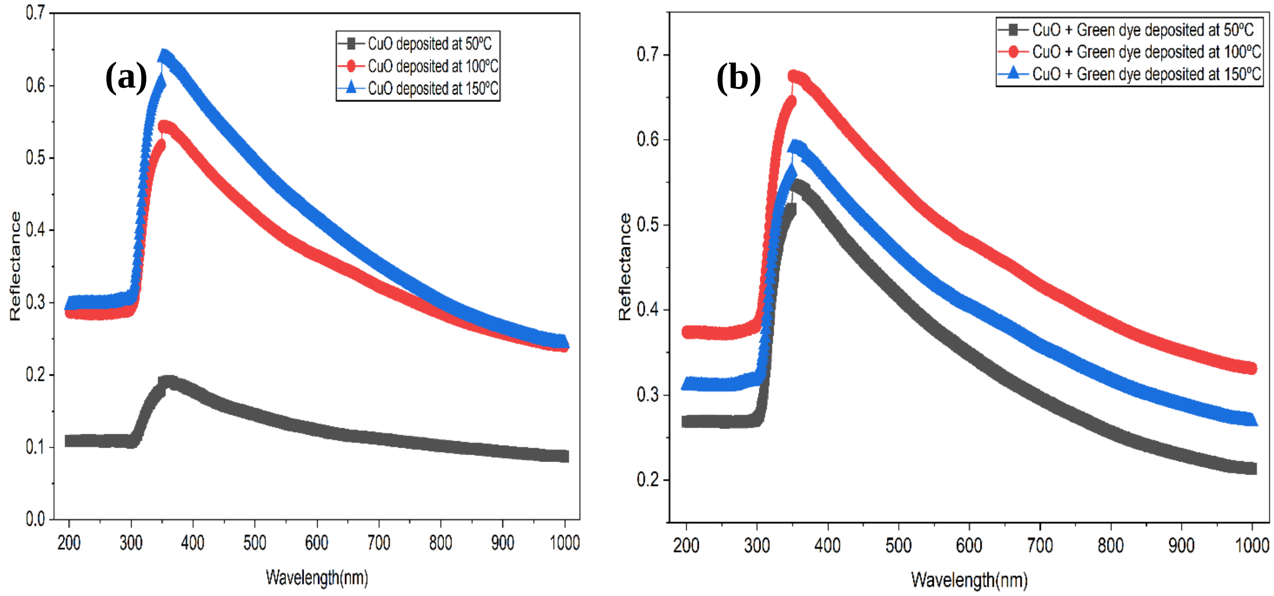

Figure 4. Reflectance versus wavelength of (a) CuO and (b) green dye-modified thin films deposited at 50°C, 100°C and 150°C.

3.4 Refractive index and extinction coefficient

The refractive-index spectra (Figure 5a-b) of the undyed films increase with substrate temperature, indicating progressive densification of the CuO matrix and an accompanying rise in optical density. Dye incorporation modulates the refractive-index response, the effect being most evident at 50 °C and 100 °C where the organic component remains largely intact. The extinction coefficient (Figure 6a-b) follows the absorption trends: undyed films exhibit high extinction in the ultraviolet, while dye-modified films show elevated extinction throughout the visible region.

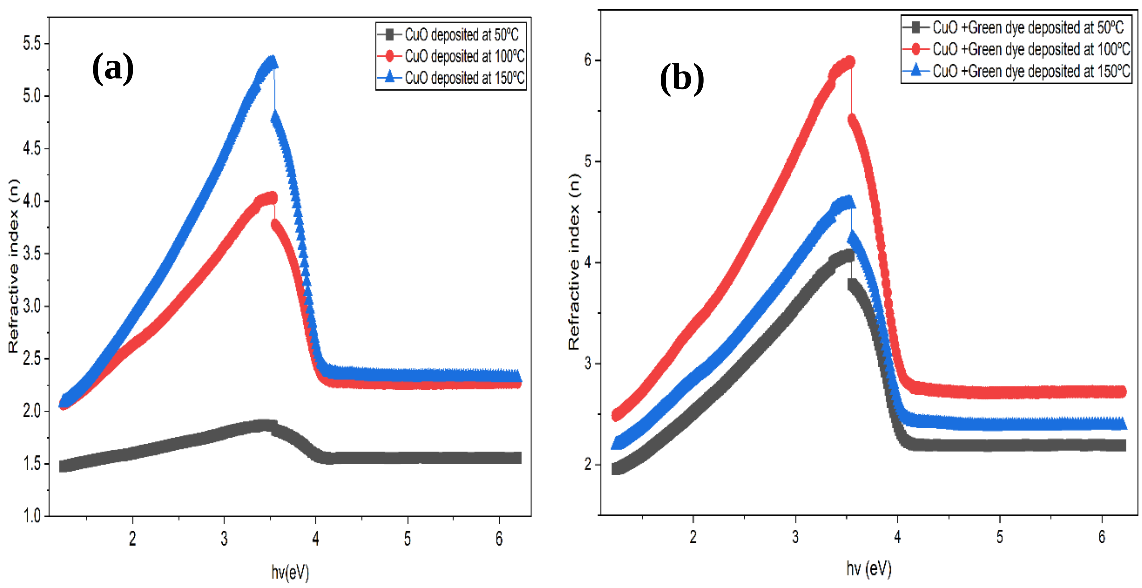

Figure 5. Refractive index versus photon energy of (a) CuO and (b) green dye-modified CuO thin films deposited at 50°C, 100°C and 150°C.

Conclusively, these optical constants demonstrate that substrate temperature governs a trade-off between the structural quality of the CuO host and the retention of dye-mediated visible-light interaction. Similar temperature-dependent optical tuning has been reported for spray-pyrolysed CuO thin films (Diachenko *et al.*, 2021; Prakash *et al.*, 2022; Kuryshchuk *et al.*, 2022; Zare Asl & Rozati, 2024). The dye-related changes are also consistent with DSSC studies that emphasise the joint importance of sensitizer absorption and semiconductor interface quality for efficient photon capture and subsequent charge separation (Mujtahid *et al.*, 2022; Rahman *et al.*, 2023).

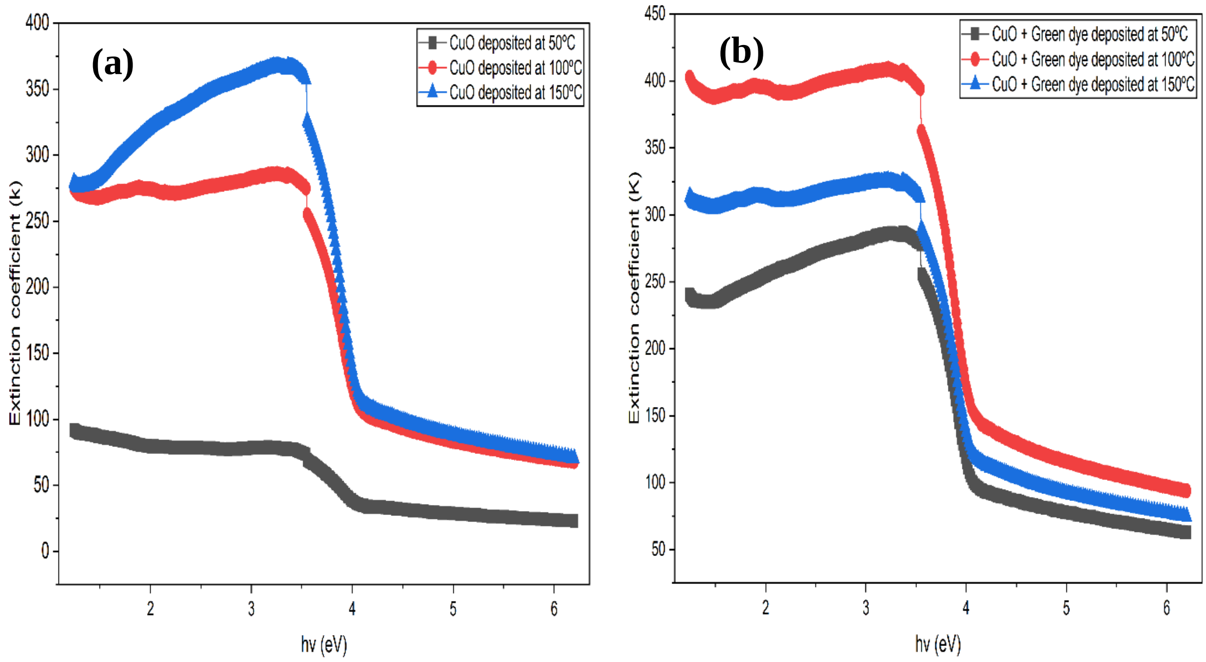

Figure 6. Extinction coefficient versus photon energy of (a) CuO and (b) green dye-modified thin films deposited at 50℃, 100℃ and 150℃.

3.5 Optical band gap behaviour

Optical band gaps were estimated from Tauc plots of $(\alpha h\nu)^{1/2}$ versus photon energy (Figure 7), a representation appropriate for indirect allowed transitions that are commonly observed in CuO. The undyed films display absorption edges whose linearity improves with increasing deposition temperature, indicating a progressive reduction in structural disorder and defect density. The extracted band-gap values lie within the range reported for spray-pyrolysed copper-oxide thin films (Diachenko *et al.*, 2021; Prakash *et al.*, 2022; Kuryshchuk *et al.*, 2022; Zare Asl & Rozati, 2024; Prakash *et al.*, 2024). Incorporation of green dye produces a modest perturbation of the apparent band-gap behaviour. This perturbation is attributed to the introduction of dye-related electronic states in the vicinity of the CuO band edges. The results are consistent with the Tauc analysis framework (Tauc *et al.*, 1966) and illustrate that both process temperature and sensitizer chemistry can be employed to tune the optical response of CuO-based photoelectrodes.

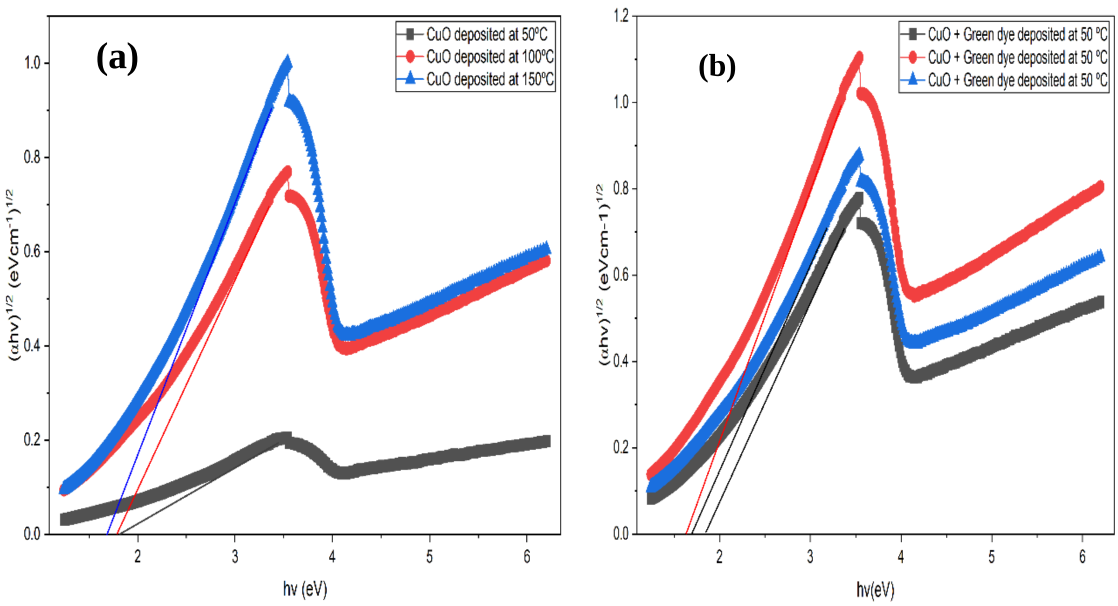

Figure 7. Tauc plot, (αhv)1/2 versus photon energy, of (a) CuO and (b) green dye-modified thin films deposited at 50°C, 100°C and 150°C.

4. Conclusion

Green dye-modified CuO thin films were successfully prepared by spray pyrolysis at substrate temperatures of 50 °C, 100 °C and 150 °C. Substrate temperature exerts a decisive influence on both film morphology and optical response. Undyed CuO layers become denser and optically more ordered with increasing temperature, whereas incorporation of the green dye markedly enhances visible-light absorption in the 400–600 nm range. The dye contribution is maximised at 100 °C, which affords an optimal compromise between morphological quality of the CuO host and retention of the organic sensitizer. At 150 °C the dye-related optical enhancement diminishes, consistent with partial thermal degradation of the organic component.

These findings establish moderate-temperature spray pyrolysis as a simple, low-cost and scalable route for the fabrication of CuO/green-dye composite thin films. The films prepared under the optimised conditions exhibit the morphological continuity and visible-light harvesting characteristics required of photoanode materials in dye-sensitized solar cells and related optoelectronic devices.

Acknowledgement, The technical inputs of Mr Lawrence Celestino Nnaemeka of the Nano Lab, Department of Physics, University of Nigeria, Nsukka, is acknowledged.

Disclosure statement: *Conflict of Interest:* The authors declare that there are no conflicts of interest.

Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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