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Investigating the Impact of Substrate-to-Nozzle Distance on the Microstructure and Optical Properties of CuO Thin Films Deposited by Thermochemical Spray Technique

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ABSTRACT

The characteristics of copper oxide (CuO) thin films deposited by thermal spray method on moving substrates at various distances from the nozzle to the substrate were investigated using XRD, SEM, and UV-Vis methods. The most significant characteristic in all the deposited films, according X-ray diffraction (XRD) study, was the (111) peak. Stable crystalline structure of the films was demonstrated by varying d-spacing values ranging from 0.245 Å to 0.246 Å at varied distances. Moreover, the Scherrer equation was used to determine the crystallite size; it was found that the size dropped as the distance separating the nozzle from the substrate rose. Particle size and porosity varied between films deposited at 15 cm and 30 cm according to SEM investigation. Whereas the films produced at 30 cm revealed bigger particles (10 nm), those deposited at 15 cm showed reduced particle sizes (20nm). The UV-Vis data revealed that light transmittance rose while light absorption dropped as the distance separating the nozzle from the substrate increased. The energy band gap (Eg) was deducing using the Tauc equation A larger energy band gap (2.35 eV) was found in the films deposited at 30 cm than in those formed at 15 cm (2.04 eV). Furthermore, noted was a rise in the absorption coefficient and refractive index with increasing distance, suggesting improved film absorption and conversion capability of course.

Keywords: CuO thin films, moving substrate, structural properties, surfaces properties, optical properties.

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INTRODUCTION

Special properties of nano materials and metal oxide films led to substantial growth that enabled their essential adoption in advanced technological systems (Kumar, 2016). Modern technological innovation happens because of enhanced efficiency in electronic devices and optics through continuous improvements in materials engineering technology and thin-film properties (Shafi, 2023; Botas, 2024; Rajput *et al*., 2024). CuO is widely used in applications such as solar cells that convert light into electrical energy because it efficiently absorbs light (Wu *et al*., 2024), sensors used for gas (Nie *et al*., 2024) and light refers to electromagnetic radiation, which includes the visible spectrum as well as ultraviolet (UV) and infrared (IR) radiation. CuO (copper oxide) is used in sensors that can detect light-this could be in the form of measuring light intensity, wavelength, or changes in light conditions (Kwiatkowski *et al*., 2024) detection due to its high sensitivity to the surrounding environment, in addition to its use in photo catalysis for the degradation of organic compounds in environmental and purification applications (Rasheed *et al*., 2024; Sathish *et al*., 2025). The semiconductor characteristics of CuO operate as p-type together with high electrical conductivity properties and the band gap (Eg) of bulk copper oxide (CuO) is an important property for its semiconductor behavior. CuO is a p-type semiconductor, and its bulk band gap typically lies in the range of 1.2 to 1.5 eV. This band gap is ideal for applications in optoelectronic devices like solar cells, sensors, and other energy-related technologies, as it allows for efficient light absorption and charge carrier mobility. (Ren and Mindrov 2023; Lee *et al*., 2023). Among various methods for forming CuO films chemical spray pyrolysis (CSP) stands out because the technique provides cheap hardware, quick deposition speeds and straightforward setup procedures. The thermal spray technique outperforms thermal evaporation and chemical vapor deposition (CVD) because it has low costs while requiring no vacuum system and allowing big-area application (Toma *et al*., 2024).

The production of high-quality films occurs through two main deposition methods which include physical vapor deposition (PVD) and solution processing (Hossain and Mansour, 2023; Picavet *et al*., 2024). CdO film deposition occurs through three methods that include thin-film evaporation for achieving uniform structures and sol-gel deposition to modulate film characteristics (Toma *et al*., 2024) along with chemical vapor deposition (CVD) for creating high-quality homogeneous layers (Sivaramamoorthy *et al*., 2011).

Various parameters during deposition significantly affect the structure and properties of CuO, including substrate temperature, which plays a crucial role in controlling grain size and crystalline density (Lin *et al*., 2024), gas flow, which contributes to uniform distribution of the material and ensures consistency in the deposited layers (Moll *et al*., 2024), and deposition time, which impacts film thickness and the quality of optical and electrical properties (Darenfad *et al*., 2024).

Research proves that subjecting substrates to movement while deposition takes place enables during nucleation processes, substrate motion improved surface characteristics along with reduced film imperfections through uniform disposition. To evaluate structural and optical properties in CdO thin films properly one must assess spray pyrolysis deposition pressure variations. The quality improvement of CuO films continues to pose difficulties because researchers need to improve their crystallinity to maintain stability in actual applications (Xu *et al*., 2023; Ożga *et al*., 2024).

The method "thermal spray in the liquid phase" enables researchers to enhance efficiency alongside minimizing structural defects according to (Guignard, 2012). Additional investigation is essential to connect substrate selection to film structural characteristics because it will advance the creation of improved industrial materials (Balzano *et al*., 2021). Research on CuO thin film deposition through variable distance and moving substrate methods still lacks a comprehensive understanding of how these methods affect film structural and optical characteristics. Although deposition time significantly affects surface morphology and electrical properties, with longer times improving conductivity and lowering resistance (Darenfad *et al*., 2024), temperature and deposition conditions affect optical absorption, showing a systematic degradation with increasing wavelength, the effect of substrate motion during deposition on these electrical and optical properties has not been adequately explored, creating a research gap that requires deeper investigation. The research will address this

missing information through examination of variables that affect crystallization along with structural stability and optical of thin films. Our research focuses on improving CuO thin films in solar cell and sensor technologies through parameter control to maximize material performance.

MATERIALS AND METHODS

In this study, thin films were prepared on glass substrates with dimensions of 25.4×76.2 mm and thicknesses ranging from 1 to 1.2 mm. The substrates were precisely cut into dimensions of 12.7×76.2 mm using a specialized glass cutter. To ensure the quality of the deposition, the substrates underwent a multi-step cleaning process. This involved immersion in analytical-grade acetone for five minutes with stirring to remove organic contaminants, followed by immediate rinsing with distilled water twice. After drying the substrates with lint-free towels. Dissolve 1.7 g of $CuCl_2$ in 25 mL of distilled water to prepare a 0.01 M $CuCl_2$ solution and dissolve 0.56 g of KOH in 25 mL of distilled water to prepare a 0.02 M KOH solution, ensuring complete dissolution by slow stirring. Mix the two solutions slowly under magnetic stirring and heat the mixture at 80°C for 15 minutes to form copper oxide (CuO). The CuO thin films are then deposited onto a moving substrate at 250°C under a steady pressure of 1 bar with a moving speed of 6 cm/s to increase the homogeneity of the sample. The deposition occurs with a nozzle positioned at distances of 15 cm, 20 cm, 25 cm, and 30 cm from the substrate, with each spray lasting 10 seconds followed by a 2-minute evaporation phase to stabilize the substrate temperature. The temperature during deposition is maintained constant by a digital controller. The solvent used for the CuO deposition is distilled water, and after the deposition process, the films are placed in an evacuated desiccator for at least 2 hours to ensure proper drying and moisture removal. To confirm the formation of CuO, X-ray Diffraction (XRD) can be performed to verify the crystalline phase, UV-V is spectroscopy can analyze optical properties, and scanning electron microscopy (SEM) can be used to observe the morphology. The structural and optical characteristics of the films are analyzed after deposition by varying the nozzle distance, which affects the properties of the films (Ren and Mindrov, 2023; Lee *et al*., 2023). I designed the spray system specifically for deposition of films using CSP technology. The electrical components were carefully assembled and connected to ensure the deposition process was automated and efficient. CSP is considered one of the most efficient methods for deposition of oxides. It is characterized by its simplicity, low cost, and ability to deposit films of materials with high melting points while achieving wide-ranging homogeneity, making it ideal for preparing films that cannot be produced using other methods. The CSP system is composed as shown in (Fig. 1).

Fig. (1): Chemical spraying system (CSP).

Below shows the shape of the samples after the sedimentation process, which included 4 samples that were deposited at different distances between the descent and the substrate, with dimensions of (15, 20, 25, 30 cm), As shown in the (Fig. 2).

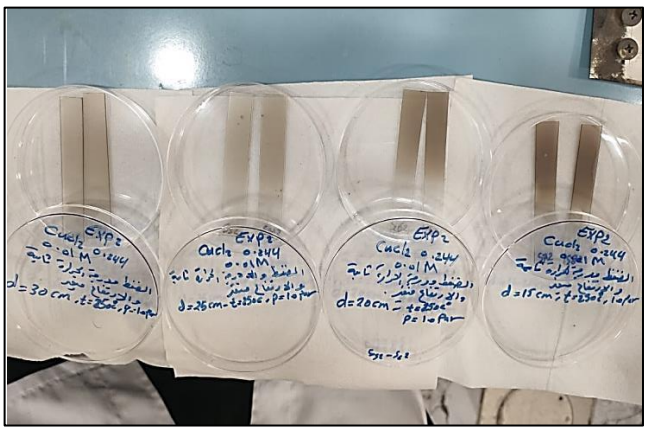

Fig. (2): Sample shape after sedimentation process.

The 721-2000 spectrophotometer, as shown in (Fig. 3), covers a wavelength range of approximately 320-1000 nanometers. The instrument was carefully refurbished by measuring all the ohms to obtain accurate results. The instrument used ohm transmittance frames within this range, which helped in calculating the optical properties.

Fig. (3): A photograph of an optical spectrometer.

RESULTS AND DISCUSSION

XRD Results

X-ray diffraction XRD (Shimadzu Company -XRD 6000) was employed to analyze the crystallographic structure of the thin films. The results revealed that the (111) peak, which is the main peak of copper oxide (CuO), was the dominant feature in all deposited samples, with a 2θ angle of approximately 35.79°, as shown in (Fig. 4). Other peaks corresponding to different crystallographic planes were also observed, but the (111) peak was the most prominent as shown in Table (1).

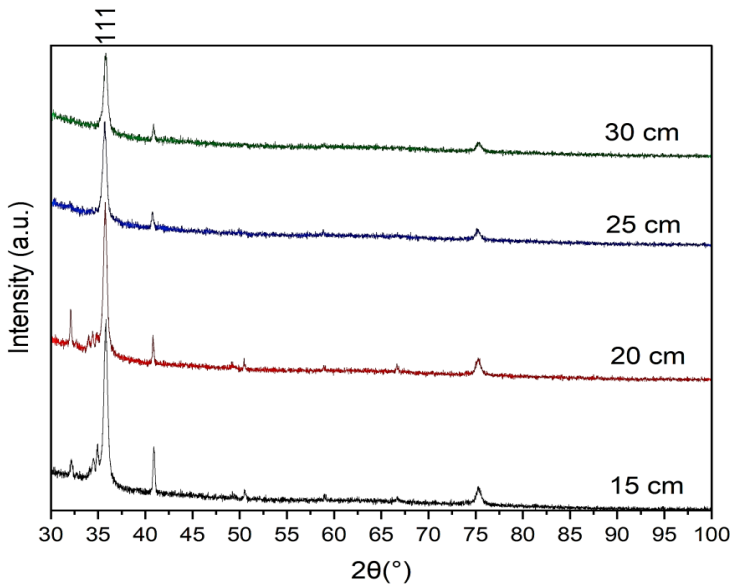

Fig. (4): Show X-ray diffraction (XRD) for samples at (15, 20, 25, 30 cm).

The indicating stability in the crystalline structure (cubic crystal). Additionally, the crystallite size was calculated using the Scherrer equation (Jaboyedoff *et al*., 1999):

$$D = \frac{K\lambda}{\beta cos\theta} \qquad \dots\dots\dots (1)$$

And it was observed that the crystallite size decreased as the distance between the nozzle and the substrate increased (ICDD: 00-048-1548).

Table 1: XRD results average grain dimension (nm) of the thin films.

Distance (cm)	Theta (θ)	FWHM [°2Th.]	D (nm)	Average grain size
15 cm	28.7404 35.8464 40.9174 75.3254	0.33368 0.54019 0.31252 0.77649	24.57915 15.45761 27.13279 12.92233	20.02297
20 cm	29.0954 35.7694 40.8184 75.2704	0.57522 0.52325 0.31152 0.82189	14.37198 15.95458 27.21113 12.20401	21.70887
25 cm	28.6304 35.7364 40.7414 75.1164	0.70932 0.39645 0.62734 0.79028	11.46345 20.68246 13.30612 10.72366	14.10764
30 m	28.7404 35.8244 40.8844 75.2044	0.53737 0.63053 0.65147 0.91977	15.26243 13.24208 13.01461 10.90045	13.10489

SEM Results

Scanning electron microscopy (thermo scientific) was used to study the particle size and porosity of the thin films. The results showed a clear variation in both particle size and porosity across different distances between the nozzle and the substrate, as shown in (Fig. 5).

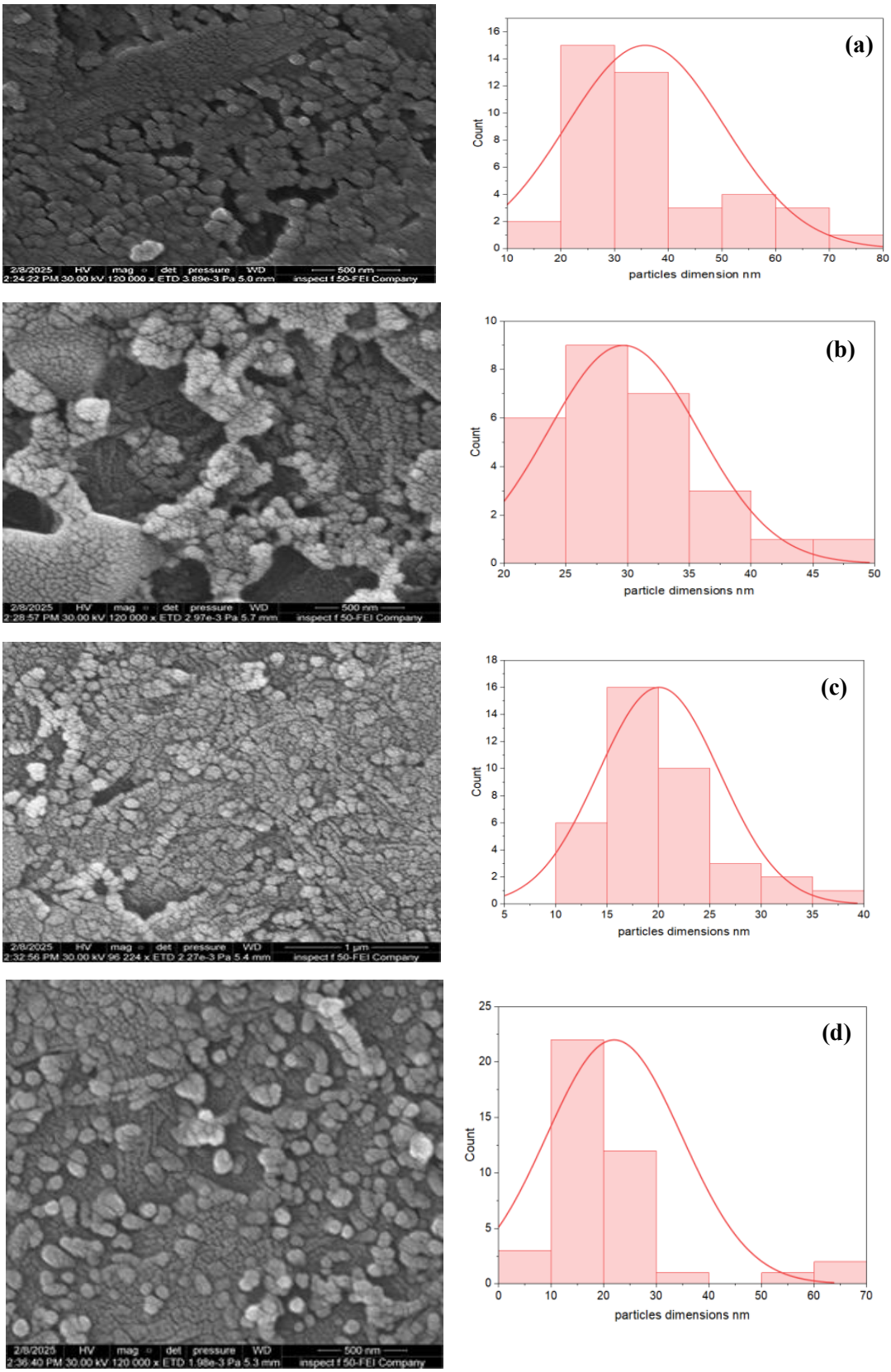

Fig. (5): SEM analysis of particle size and porosity across different distances between the nozzle and the substrate: (a) at distances d=15cm, (b) d=20cm, (c) d=25cm, (d) d=30 cm.

The films deposited at a distance of 15 cm exhibited the largest particle size, 20 nm, compared to the films deposited at a distance of 30 cm, which showed smaller particles 10 nm. It was also observed that porosity decreased as the distance between the nozzle and the substrate increased. These results indicate the impact of distance on the thin films' properties, particularly crystallization and porosity. Table (2) provides a summary of particle size and porosity across the different distances.

Table 2: SEM results-grain size and porosity of thin films.

porosity percentage (%)	Particle Size (nm)	Distance (cm)
91.415%	20	15
62.916%	25	20
68.997%	15	25
58.35%	10	30

UV Results

A UV-Vis spectrophotometer-2000-721 (range from 320-1000 nm) was used to measure the light transmittance, absorbance, absorption coefficient, and spectrophotometer across different wavelengths of the thin films deposited using the thermal spray deposition technique. Light transmittance improved with greater nozzle-substrate distance, achieving best results at a separation of 30 cm. Light absorbance diminished as the distance between the nozzle and substrate rose, indicating enhanced film efficiency in light absorption. The films deposited at 30 cm had a larger energy band gap of 2.35 eV, whereas those deposited at 15 cm revealed a band gap of 2.04 eV. The altered optical properties in the thin films exhibit enhanced performance characteristics due to a greater distance between the nozzle and substrate. The absorption coefficient augmented with the band gap, as films applied at 30 cm exhibited superior light absorption relative to those at 15 cm. The absorption coefficient serves as a material indicator that demonstrates the efficiency with which a substance processes light energy prior to its conversion into other energy forms, such as heat or electricity. The refractive indices of films deposited at distances of 30 cm and 15 cm show a notable difference. The film deposited at 30 cm has a refractive index of 0.4, which indicates stronger light-path-altering characteristics compared to the film at 15 cm, which has a refractive index of 0.3. This change in refractive index is primarily due to differences in film thickness and density. Films deposited at greater distances tend to be denser or more uniform, resulting in a higher refractive index. In contrast, films deposited at shorter distances may be thinner or less dense, leading to a lower refractive index, as shown in Table (3) (All parameters are calculated automatically by the device).

Table 3: Summarizes efficiency (%), absorption (%) and energy gap (ev), across the different distances.

Efficiency (%)	Absorption (%)	Energy Gap (eV)	Distance (cm)
59.60	45.15	2.04	15
62.89	42.50	2.07	20
65.15	41.50	2.19	25
68.50	40.10	2.35	30

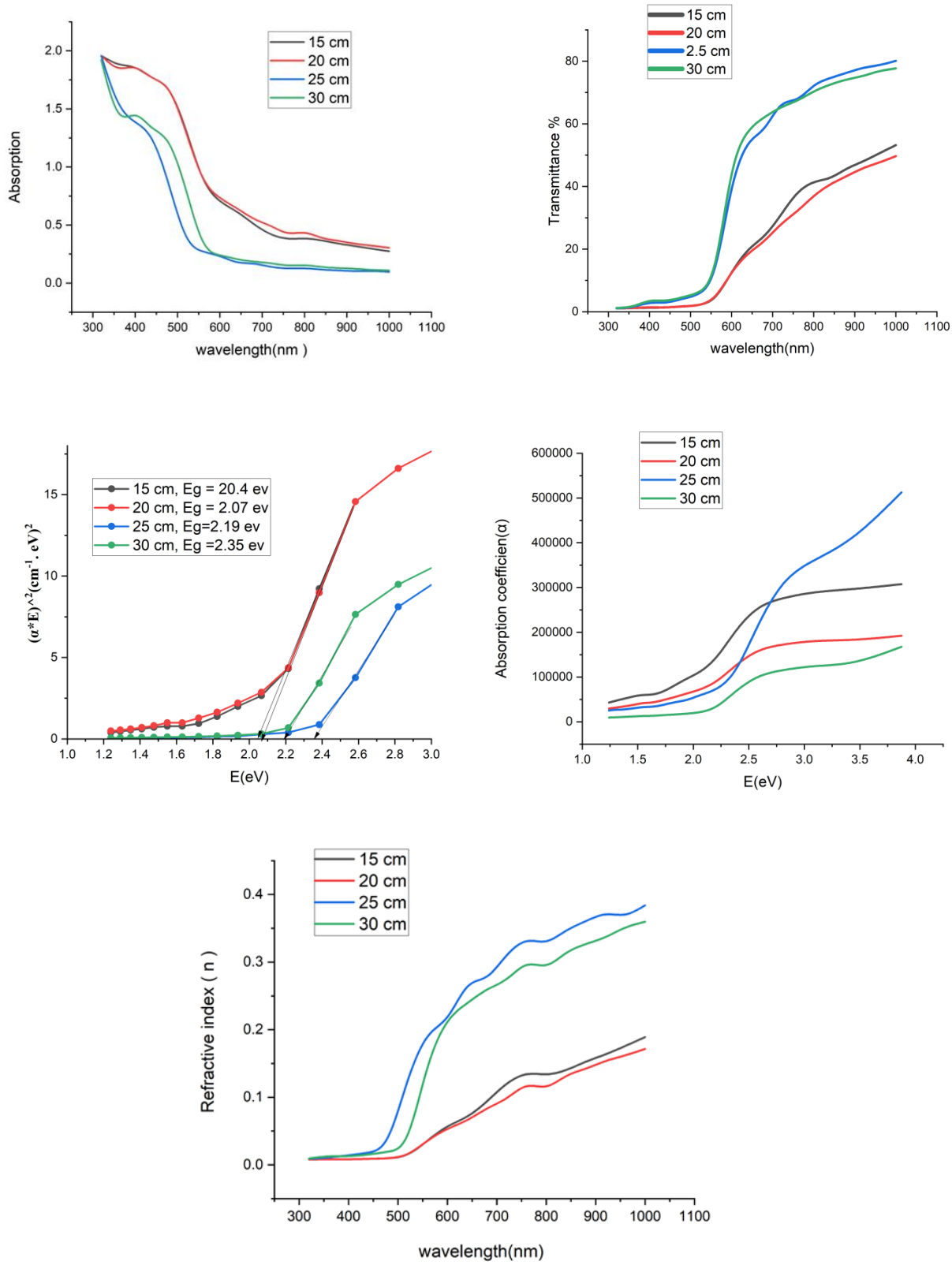

Fig. 6: The optical properties of CuO at distances (15, 20, 25, 30 cm).

CONCLUSIONS

The XRD, SEM and UV-V is techniques generated results which show that the moving substrate plays an essential role to enhance structural along with optical characteristics of deposited

thin films. Thin films deposited with 30 cm distance between nozzle and substrate had a superior energy band gap at 2.35 eV than those deposited with 15 cm distance whose band gap was 2.04 eV. Improvements in energy band gap values indicate that the films become better at absorbing light which improves their performance in solar cells along with sensor applications. The XRD results show that film crystallite size along with particle size decreases as the nozzle-substrate distance extends while the crystallinity of the films improves correspondingly. A modified crystalline arrangement found in films obtained at 30 cm distance results in better optical performance according to UV-V is analysis because it improves their light-to-energy conversion capability. The system with the moving substrate produced an equalized distribution of material that enhanced both the deposition uniformity and final film structure. The use of moving substrate enhances crystallization and stabilizes structural film properties which leads to better optical characteristics with greater transmittance energy band gap and absorbance. The distance between the nozzle and substrate serves as a vital factor for enhancing both structural quality and performance of the film. Research has proven that thin film crystalline structure enhances effectively when using a moving substrate thus resulting in better light absorption performance. These findings open new avenues for optimizing performance in advanced applications such as solar cells and sensors, and emphasize the importance of precise control over deposition parameters to achieve the best performance of materials in future applications.

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دراسة تأثير المسافة بين الركيزة والفوهة على البنية الدقيقة والخصائص البصرية لأغشية أكسيد النحاس الرقيقة المترسبة بتقنية الرش الكيميائي الحراري

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الملخص

تمت دراسة خصائص اغشية أكسيد النحاس (CuO) الرقيقة المترسبة بواسطة طريقة الرش الحراري على الركائز المتحركة عند مسافات مختلفة بين الفوهة والركيزة باستخدام طرق XRD وSEM وUV-Vis. وكانت الخاصية الأبرز في جميع الاغشية المترسبة، وفقًا لدراسة حيود الأشعة السينية (XRD)، هي القمة (111). تم إثبات الهيكل البلوري المستقر للأغشية من خلال تغير قيم المسافة بين الطبقات (d-spacing) التي تراوحت بين 0.245 Å و0.246 Å عند المسافات المختلفة. علاوة على ذلك، تم استخدام معادلة شيرر لتحديد حجم البلورات؛ وقد تبين أن الحجم ينخفض كلما زادت المسافة بين الفوهة والركيزة. اختلف حجم الجسيمات والمسامية بين الاغشية المترسبة عند 15 سم و30 سم وفقًا لفحص الميكروسكوب الإلكتروني الماسح (SEM). بينما أظهرت الاغشية المنتجة عند 30 سم جسيمات أصغر (10 نانومتر)، أظهرت الاغشية المترسبة عند 15 سم أحجام جسيمات أكبر (20 نانومتر). أظهرت بيانات الأشعة فوق البنفسجية (UV-Vis) أن نفاذية الضوء زادت بينما انخفض امتصاص الضوء كلما زادت المسافة بين الفوهة والركيزة. وبذلك، تحسنت الاغشية في امتصاص الضوء. تم استنتاج فجوة الطاقة (Eg) باستخدام معادلة تاوك (Tauc equation)، وقد تبين أن فجوة الطاقة كانت أكبر (2.35 eV) في الاغشية المترسبة عند 30 سم مقارنة بتلك التي تم تشكيلها عند 15 سم (2.04 eV). علاوة على ذلك، لوحظ زيادة في معامل الامتصاص ومعامل الانكسار مع زيادة المسافة، مما يشير إلى تحسين قدرة الفيلم على الامتصاص والتحويل بالطبع.

الكلمات الدالة: CuO الأغشية الرقيقة، الركيزة المتحركة، الخصائص التركيبية، الخصائص البصرية، الخصائص السطحية.