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Original Article

Deposition time dependent physical properties of semiconductor CuO sprayed thin films as solar absorber

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Abstract. This study aims to develop copper oxide (CuO) films on standard glass substrates using the spray pyrolysis technique and investigate the effect of different deposition times on their structural, morphological, wettability, optical, and electrical properties to enhance their optoelectronic characteristics. CuO thin films were fabricated at different deposition times (5 to 20 min) with a substrate temperature of 400 °C. X-ray diffraction (XRD) analysis confirmed the crystalline structure of all deposited CuO films, showing a monoclinic phase with preferential orientation along the (111) direction, indicating a well-ordered atomic arrangement. Atomic force microscopy (AFM) examination revealed the influence of deposition time on the surface morphology, with a low roughness value of 13.315 nm observed for the 10 min film compared to 19.432 nm for the 20 min film. Contact angle (CA) analysis showed a transition from hydrophilic to hydrophobic behavior as the deposition time increased, indicating significant changes in surface properties. This transition to a hydrophobic nature (CA = 105°) for the 20 min sample is important for protecting photovoltaic devices from humidity-related degradation, ensuring long-term reliable operation even in challenging conditions. The transmittance of the film in the visible region was low, indicating high absorbance of CuO. The optical gap decreased from 1.98 to 1.61 eV with increasing deposition time, making films suitable as absorber layers in solar cells. Electrical analysis showed improved conductivity with increasing deposition time, leading to a decrease in electrical resistivity (3.77 Ω.cm) and high charge density (1.269×10^{16} cm^{-3}) for the 20 min film. Therefore, the 20 min deposition film with a hydrophobic character exhibited good *p*-type electrical semiconductor properties and efficient absorption of solar light, making it promising for thin film solar cell applications.

Keywords: Thin films / CuO / deposition time / spray pyrolysis / AFM / hydrophobic

1 Introduction

Copper oxide (CuO) is a *p*-type semiconductor that exhibits several interesting properties. Its direct band gap, which is generally between 1.3 and 2.1 eV, gives it specific optical properties [1]. Moreover, CuO is non-toxic, chemically stable, easy to synthesize, abundant in nature and has good electrical properties. These characteristics make it an attractive material for a variety of applications, particularly in optoelectronic devices such as solar cells, light-emitting diodes [2], photodetectors, waveguides, optical filters, and antireflective coatings. Additionally, their ability to convert solar energy into electricity makes them potential candidates for energy storage and conversion applications. Different physical and chemical deposition methods were used to produce thin CuO films. In this research, we focused on the spray pyrolysis technique to deposit CuO thin films on ordinary glass substrates. This deposition method has many advantages, being a modest, flexible, inexpensive, void-free and easy to commercialize technique, which offers an expert solution in industrial applications involving surface engineering research [3,4]. Sputtering parameters, such as substrate temperature, precursor solution flow rate, carrier gas flow rate, precursor molarity, etc., influence the physical properties of CuO thin films. The majority of studies published in the literature, including these previous studies, have focused on the effect of these deposition parameters on the structural, morphological, optical and electrical properties of CuO films. So, along with other spray parameters, optimizing the deposition time (thickness) is a very important and crucial process parameter for the absorbent layer in photovoltaic applications. For example, Ben Saad et al. [5], prepared CuO thin films with various thicknesses between 180 and 2900 nm using spray pyrolysis, which demonstrates lowest resistivity of 8.51 Ω.cm and low band gap (Eg) of 1.52 eV. Kamli et al. [6] CuO thin films was synthesized on glass substrates at 300 °C via spray pyrolysis technique and reported the thickness increasing (deposition time from 10 to 40 min) have permitted the improvement of electrical characteristics (decrease of the resistivity and increase of

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the free carriers concentration). Surface wettability is an important aspect of surface properties and has significant implications for photovoltaic application [7]. Surfaces can be classified based on the contact angle (CA, θ) of liquids on the surface as hydrophilic ($\theta < 90°$) or hydrophobic ($90° < \theta$) [8,9]. Hydrophobic surfaces, in particular, have shown promise in numerous practical applications such as catalytic tissue engineering, self-cleaning mechanisms, oil recovery, anti-adhesion properties, photocatalysis, antibacterial properties, micro-droplet transport, anti-icing coatings, antifouling properties, and corrosion protection. Given the importance of hydrophobicity, our research focused on utilizing the spray pyrolysis technique to fabricate CuO films tailored for photovoltaic applications. This approach was driven by several factors: (i) hydrophobic films can protect photovoltaic components from moisture, thus ensuring their efficiency and durability. (ii) These films can reduce component corrosion, especially in humid or harsh weather conditions. (iii) By reducing water and contaminant accumulation, hydrophobic films can enhance solar cell performance by minimizing energy conversion losses. (iv) Hydrophobic surfaces are easier to clean, potentially reducing maintenance costs and extending the lifespan of photovoltaic components. For instance, Nezzari et al. [7] conducted a study on the synthesis and characterization of the wettability, as well as various physical properties, of cobalt oxide (Co_3O_4) doped with different nickel concentrations (0%, 2%, 4% and 6%) deposited by spray pyrolysis. They confirmed that the hydrophobicity improves the physical properties of the absorbent thin film, making it more suitable for use in photovoltaic applications. Moreover, Darenfed et al. [9] observed that the hydrophobic character ($\theta = 97°$) improves the optical and electrical properties of the CuO film doped with 6% Co deposited by spray pyrolysis. These intriguing results sparked our interest in the importance of this hydrophobic character for absorbent thin films in photovoltaic application.

In the present study, the primary aim of this research is to elaborate hydrophobic CuO thin films as absorber layers in photovoltaic applications. This approach entails a comprehensive investigation into the impact of varying deposition times (5, 10, 15 and 20 min) on the properties of copper oxide deposited on standard glass substrates using spray pyrolysis. So, the objective is to thoroughly understand the variations in their structural properties, wettability, as well as their optical and electrical characteristics. As far as we know, this is the first study to investigate the wettability of sputtered thin films of CuO for various deposition times, which highlights the novelty of our work.

2 Experimental

CuO films were elaborated on standard glass substrates using the Holmarc-type spray pyrolysis technique at different deposition times (5, 10, 15 and 20 min), resulting in varying film thicknesses (Fig. 1a). Spray pyrolysis is a chemical method for depositing thin films that relies on a precursor solution. Essentially, in spray pyrolysis, a thin film is formed by spraying a precursor solution onto a heated substrate, where the constituents react to form a chemical compound. First, we start by turning on the hood, then we place the well-cleaned substrates on the substrate holder. Before each deposition, standard glass substrates undergo a thorough cleaning process using acetone, ethanol, and distilled water, with each cleaning step lasting 15 min. After the cleaning process, the substrates are placed in boxes and allowed to air dry. The choice of precursor solution, substrate, and solvent is important. Initially, copper (II) chloride dihydrate ($CuCl_2.2H_2O$, purity 99.0%) is used as the copper source, with a mass of 1.8 g, dissolved in methanol (CH_3OH, purity 99.9%). The mixture is stirred for 1 h at a temperature of 30 °C to obtain a homogeneous solution. For the prepared samples, the distance between the atomizer and the substrate is set at 25 cm. The substrate holder is connected to an electrical resistance which serves as a temperature regulator. To avoid any thermal shock that could cause the substrates to break, the substrate holder is heated gradually from ambient temperature to the deposition temperature set at 400 °C. The carrier gas (compressed air) is maintained at a constant pressure of 2 bar and the solution is introduced into the spray nozzle at a pre-adjusted constant atomization pressure of 2 ml/min. Very fine droplets are sprayed onto the heated substrates. The effect of temperature activates the chemical reaction forming the CuO layer, while the other elements volatilize after the reaction as a result, as indicated by the following reaction:

$$CuCl_2.2H_2O + 2[CH_3OH] \rightarrow CuO\downarrow + 2[CH_3Cl]\uparrow + 3[H_2O]\uparrow.$$

At the conclusion of the deposition process, the heating is turned off, and the samples are allowed to cool slowly to room temperature to prevent thermal shock. Subsequently, the samples are recuperated (Fig. 1b).

To investigate the physical properties of the deposited CuO thin layers, specific and diverse techniques were utilized. The measurement of the thickness of our films is done using a MicroXam-100 type optical profilometer. The X-ray diffraction (XRD) analysis using a Philips X'Pert-PRO diffractometer with CuKα radiation source provides information about the structural properties, revealing the crystallographic structure of the material. The samples were characterized using the Nanosurf Flex-Axiom C3000 atomic force microscope (AFM) in static (contact) mode in open air and at room temperature. This mode of operation involves scanning the AFM tip point by point and line by line over the sample surface. The movement of the tip (in x, y and z) causes the movement of the laser beam which follows it, thus capturing the variations in relief of the layer (the sample). These variations are then transformed into a 3D image by an optical sensor. The specified scanning area of 25 μm^2 (5 $\mu m \times 5\ \mu m$) is used to obtain detailed information about the surface features. We conducted static contact angle measurements on the surfaces of the deposited films under ambient temperature and constant humidity (27% RH) conditions using an optical system. This system comprised a white light lamp for illumination and projecting the image of the droplet on the sample (LEYBOLD type light

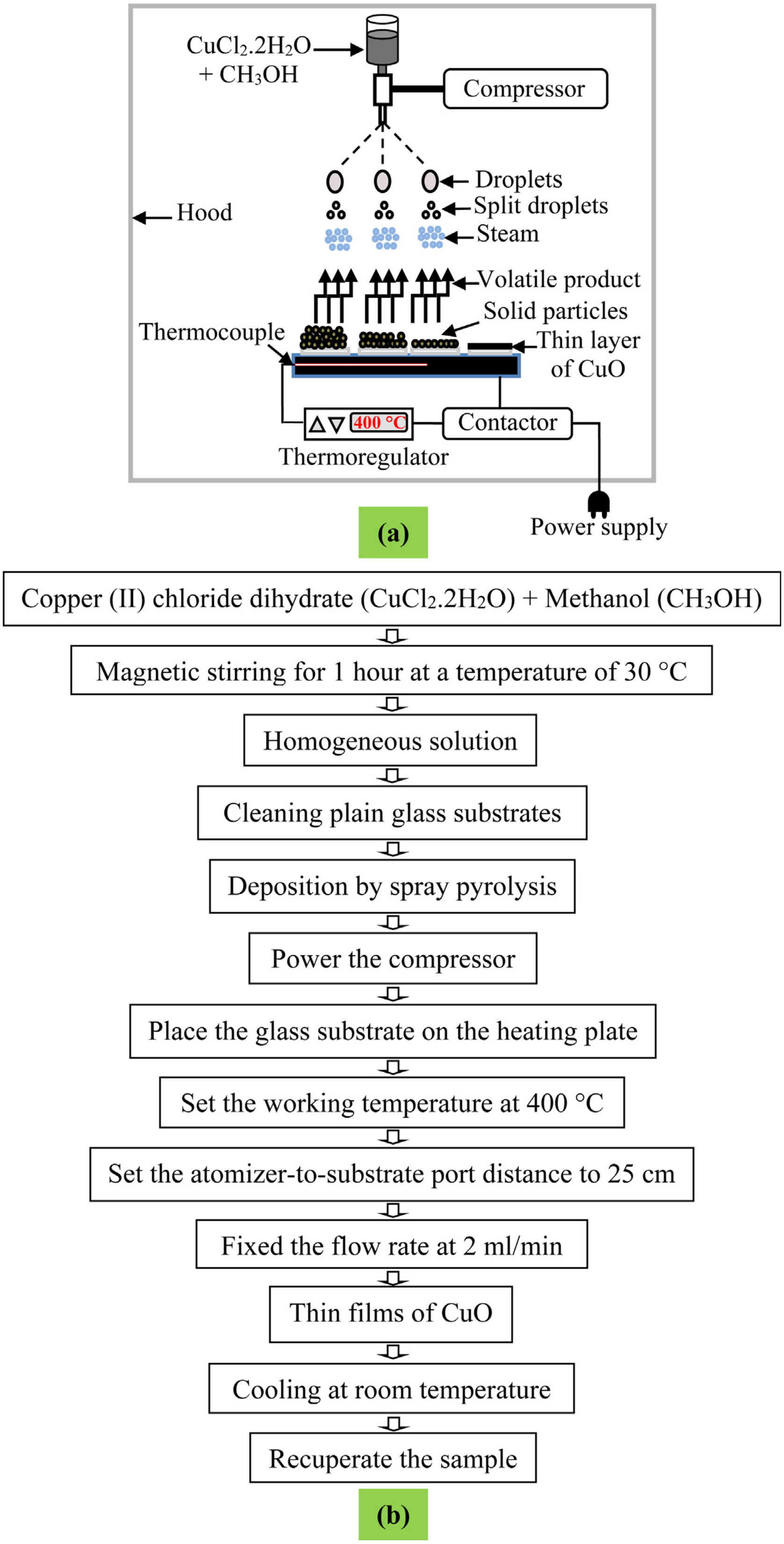

Fig. 1. a) The experimental device used for the production of our films is of the Holmarc type, b) processing steps used for CuO thin films deposition.

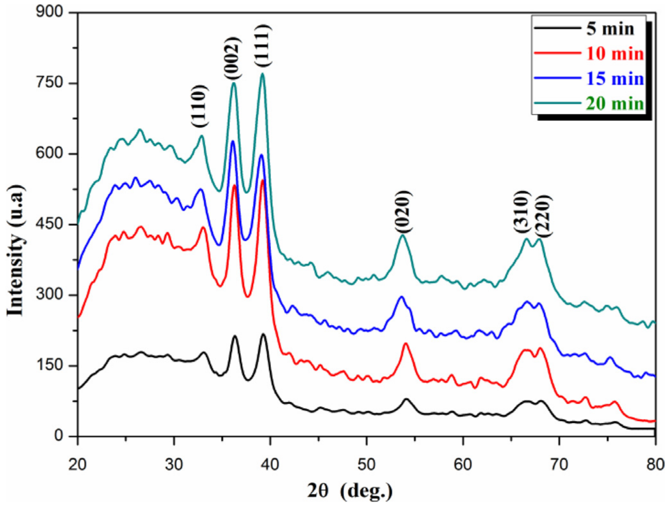

Fig. 2. XRD diffraction spectra of our films.

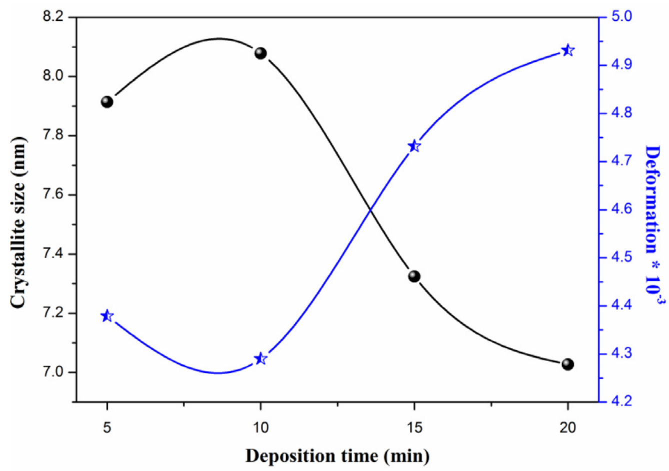

Fig. 3. Variation in crystallite size and deformation as a function of deposition time.

source (6 V, 30 W)), along with a projection lens to enlarge the image of the droplet projected on a translucent screen measuring 30×30 cm^2. To minimize evaporation of the distilled water droplet, all contact angle measurements were taken 5 s after depositing a 5 μl droplet using a micropipette. For each measurement, we calculated the average of 6 contact angle values taken at different positions on the surface. This method provides information on the wetting behavior of the material—specifically, how the water droplet interacts with the surface. The optical properties, including absorption and reflection characteristics, are investigated using a Shimadzu UV–Vis-NIR spectrophotometer in the wavelength range of 300–1000 nm. This helps in understanding how the material interacts with light across different wavelengths. For electrical properties, measurements such as electrical resistivity (ρ), carrier mobility (μ), and carrier concentration (n) are crucial. These are determined using the Hall effect measurement setup (HMS 3000). Overall, this comprehensive set of techniques covers structural, morphological, wettability, optical, and electrical aspects, providing a thorough understanding of the CuO films' properties.

3 Results and discussion

3.1 X-ray diffraction analysis

In Figure 2 we have presented the XRD diffraction spectra of different films as a function of deposition time. The influence of processing time on the films is clearly visible and suggests that the structural properties of these films are very sensitive to this parameter. The XRD spectra of the samples show a crystal structure with clear peaks characteristic of the monoclinic CuO phase, as found by comparison with the standard JCPDS file (45-0937) [10]. The diffractograms show prominent (002) and (111) planes located at 2θ values of 35.5° and 39.2° for all films prepared at various deposition times. Similar results were found by other authors [5,10]. However, small other planes linked to (110), (020), (311) and (220) are also observed indicating the polycrystalline nature of the films. No other secondary phases were detected for all samples.

The crystallite size (D) according to the orientation of the most intense peak (111) of the CuO films as a function of the deposition time was calculated according to the following Scherrer formula [11,12]:

$$D = \frac{0.9\lambda}{\beta \cos \theta} \tag{1}$$

where, λ: the wavelength of the X-rays. θ: the Bragg diffraction angle. β: is the width at half maximum (FWHM) in radians.

The strain values (ε) of our films for the (111) plane were calculated using the following equation [13,14]:

$$\varepsilon = \frac{\beta \cos \theta}{4}. \tag{2}$$

The calculated values of crystallite size and strain are shown in Figure 3. From this Figure, it is observed that crystallite size increases, while strain decreases for the sample prepared at 10 min. The reduction in strain density can be attributed to the improvement of film crystallinity and the elimination of defects [10,15]. Therefore, the best crystallinity was observed for the film deposited at 10 min. For the other films, the crystallinity begins to break down at deposition times greater than 10 min. A similar result was observed for the work of Akaltun et al. [15] and Welegergs et al. [16]. This behavior is due to stress formation (strain) because strain density is inversely proportional to crystallite size. All these results are in agreement with the literature [5,16].

3.2 Surface morphology and wettability analysis

Atomic force microscopy (AFM) was used to analyze the surface topography and roughness of our films. Figure 4 shows 3D atomic force microscopy images of nanostructured CuO over a 25 μm^2 image area synthesized at 400 °C in free air for varying elaboration times. The average surface roughness of the coatings decreased from 14.242 to 13.315 nm and then increased from 16.39 to 19.432 nm, as the deposition

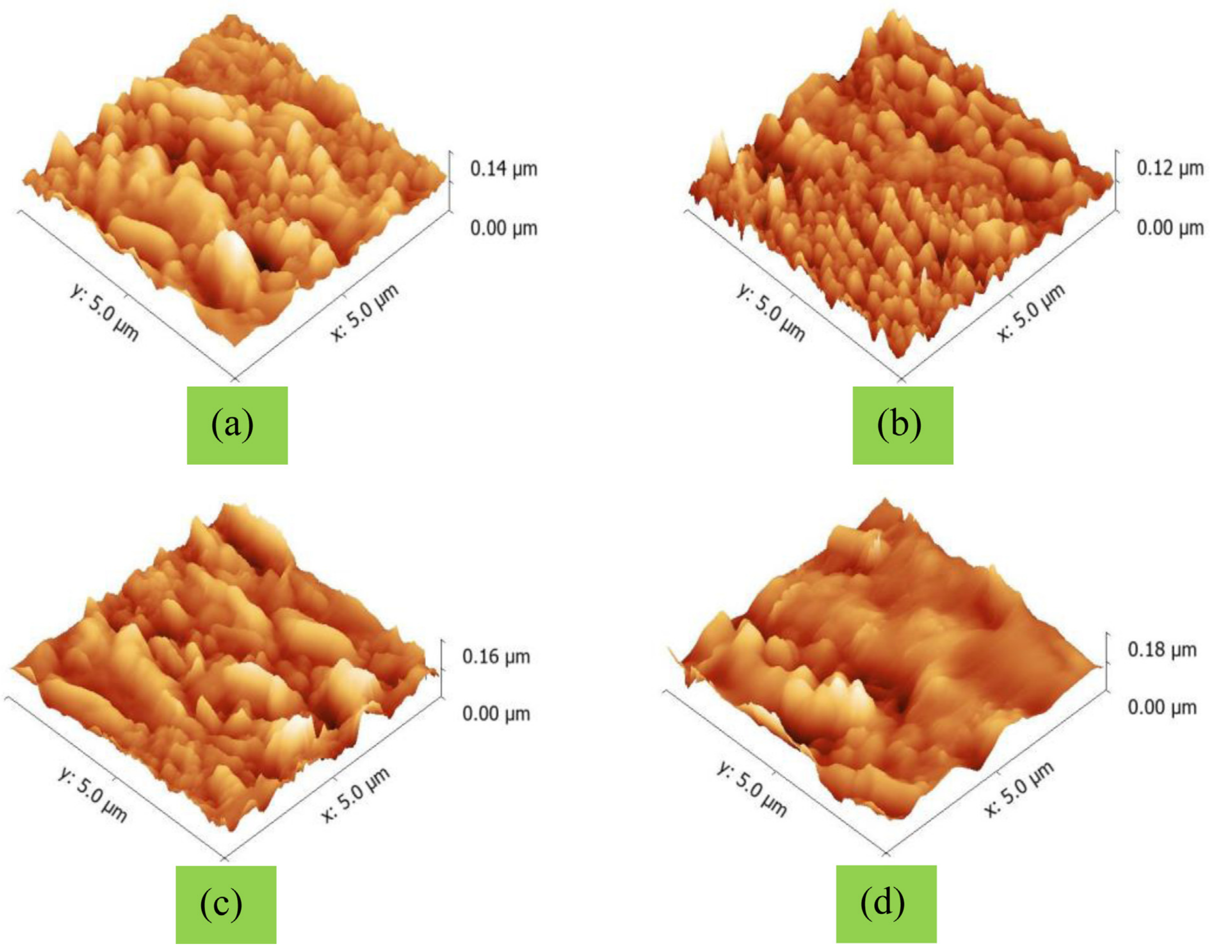

Fig. 4. AFM images of CuO thin films at a) 5 min, b) 10 min, c) 15 min and d) 20 min.

time increased to 5, 10 and 15, then to 20 min, respectively. These changes clearly indicate that deposition time plays a crucial role in the surface morphology of copper oxide coatings. Surface roughness is an important factor for absorber films, because incident light can be effectively trapped by increasing surface roughness through multi-reflections [16]. Scattering of light from irregularities in the absorber surface results in an increase in the light path length, which improves the absorption of CuO coatings [16]. Additionally, the distribution of peaks and valleys in the surface structure leads to effectively trapping more incident light in the internal structure of the coatings, thereby improving the intrinsic absorption of the coatings and thus contributing to better absorption properties [16]. Our AFM results agree with the XRD analysis.

Following Figure 5 we note that for the films deposited at 5, 10 and 15 min, the contact angle decreases, which suggests a hydrophilic character ($\theta < 90°$). This could be attributed to the presence of nanostructures such as nanowires, nanoparticles or nanotubes that promote water adhesion. Additionally, the ability of water droplets to penetrate hollows and cracks could lead to closer interaction between water and surface, promoting wetting. On the other hand, for the film deposited at 20 min, the contact angle is greater than 90° (CA = 105°), indicating a hydrophobic character. This could be due to a nanostructure that creates a surface texture that repels water and prevents the formation of a thin layer of water on the surface. The rougher surface of this CuO film deposited at 20 min compared to the other films could also contribute to this hydrophobicity. The results appear consistent with the AFM analysis shown in Figure 4, which strengthens the correlation between the surface nanostructure and the hydrophobic or hydrophilic properties of the films. Therefore, CuO films deposited by spray pyrolysis appear to have the ability to exhibit hydrophobic or hydrophilic properties depending on the deposition time, thus providing some flexibility in modulating the wettability characteristics of the surface. Based on these findings, the deposition time of 20 min can be considered as a critical value in the change of the surface state of CuO, which is related to the changes of the roughness during the crystallization process.

3.3 Optical analysis

In order to explore the optical properties, thin films of CuO were examined using a UV-Visible-NIR spectrophotometer. The transmittance T(λ) of CuO thin films with different values of deposition time (thickness) is shown in

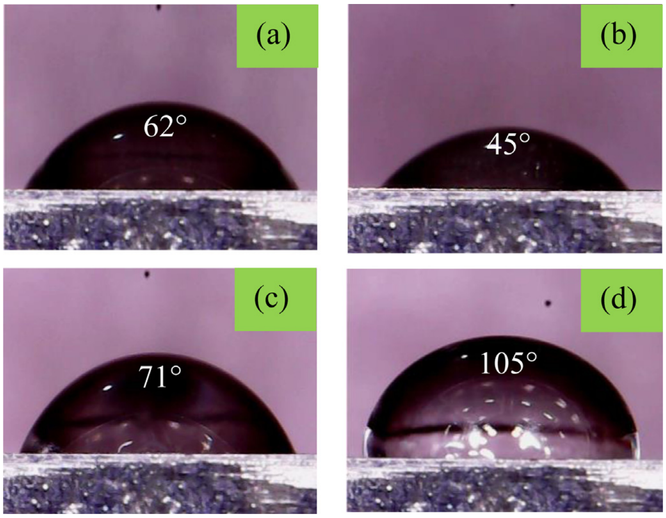

Fig. 5. Images of the contact angles with a drop of water of our CuO films.

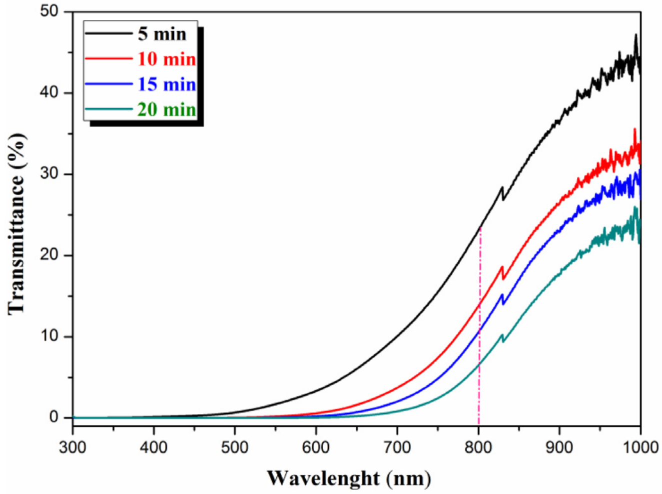

Fig. 6. Spectra of the transmittance of CuO films as a function of wavelength prepared at different deposition times.

Figure 6 in the spectral range 300–1000 nm. The transmission spectra show two regions, one of high transmission between 800 and 1000 nm and another in the visible with strong absorption. As can be seen, CuO films do not have good transparency in the visible spectral range. There is a decrease in the transparency (T%) of the deposited films from 23.12% to 6.48% with the increase in deposition time from 5 min to 20 min. According to Moumen et al. [17], this can be attributed to the high electronic transition between the valence band and the conduction band. Moreover, if the thickness increases, the scattering of incident light reaching the air-film interface becomes greater and this may this reduction in transmission may be associated with the growth of film thickness and/or the increase in film surface roughness. Indeed explain the decrease in T(λ). CuO thin films reveal high intrinsic absorption in the visible range and all transmission spectra exhibit a strong band edge transmittance drop when the incoming photon energy value is greater than or equal to the deviation from the semi-driver. In fact, this photon is absorbed by the material. Therefore, the transmission energy drops to eliminate this effect which precedes the onset of intrinsic interband absorption in CuO thin films in the visible spectral range (400–800 nm). This property allows CuO to have a favorable absorbing character for use in photo-detection of light in the visible range. Corresponding results have been obtained in other works [5,15]. On the other hand, the current results established by low-cost spray pyrolysis are comparable to those obtained using RF magnetron sputtering [17].

The optical gap value of CuO thin layers is important parameter to estimate the limit of the absorption band of thin layers. For this, we deduced the optical gap (Eg) of our films from the transmittance spectra according to the Tauc model [18,19]. The Eg value is determined by plotting $(\alpha h\nu)^2$ as a function of (hν) and extrapolating the linear region of the slope at zero absorption ($(\alpha h\nu)^2 = 0$) as shown in Figure 7.

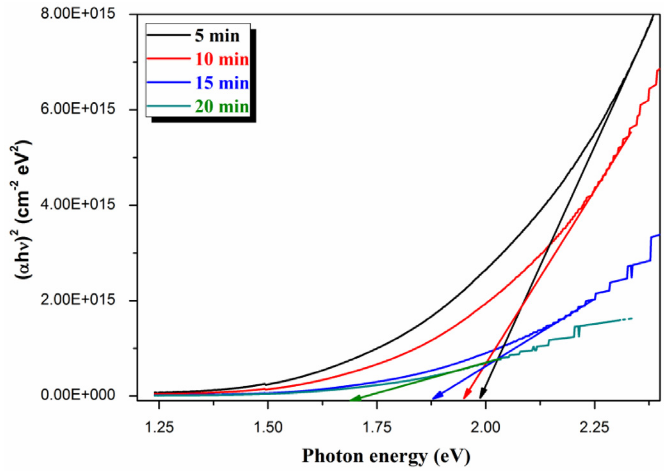

Fig. 7. Method for determining the optical gap.

Many experimental studies have pointed out that the band gap variation can be due to various factors such as: grain size, substrate temperature, thickness, doping concentration, lattice strain, structural parameters and the disorder. In Figure 8, we present the change in the optical band gap of CuO thin films as a function of deposition time. It is evident from the figure that as the deposition time increases, there is a decrease in the optical gap, indicating an increase in the thickness of the CuO film (Tab. 1). This decrease in the optical gap can be attributed to several factors. Firstly, a longer deposition time results in a thick film, leading to a higher density of electronic states near the edge of the optical gap. This increased density facilitates the excitation of electrons from the valence band to the conduction band. Secondly, the surface roughness of the film (as depicted in Fig. 4) can enhance the scattering of incident light, thereby increasing the probability of absorption and reducing reflection.

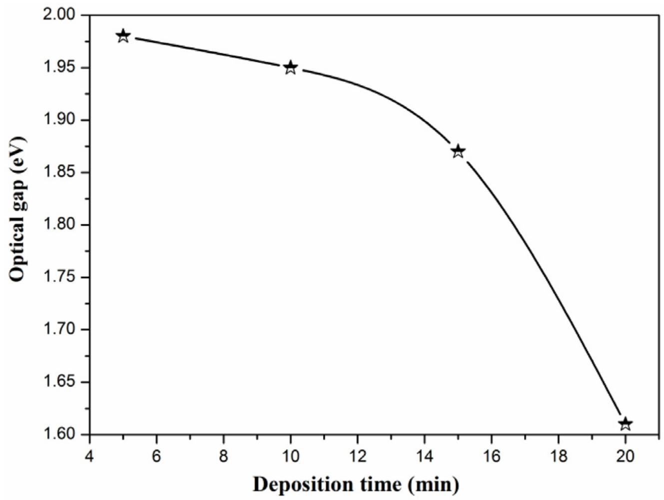

Fig. 8. Variation of the optical gap as a function of deposition time.

This increased absorption within the optical gap range contributes to the observed reduction in the measured optical gap [5]. For deposition time equal to 20 min, the energy of the optical gap is approximately 1.61 eV. The band gap values obtained in our study align well with those reported in the literature [17,20]. For example, Akaltun et al. [15] investigated CuO films of varying thicknesses prepared using the SILAR method. They observed a decrease in the film's band gap from 2.03 eV to 1.79 eV with an increase in the CuO film thickness from 120 to 310 nm. Similarly, Ben Saad et al. [5] deposited CuO thin films on glass substrates using the spray pyrolysis method and observed a decrease in the band gap from 1.63 eV to 1.52 eV as the film thickness increased from 180 nm to 2900 nm. Furthermore, it is worth noting that smaller band gaps are favorable for achieving high absorptions, and the estimated band gap values tend to decrease slightly with increasing thicknesses [16].

Thin films characterized by small optical gap values exhibit potential as optimal candidates for integration as absorber layers within solar cells. This particular application necessitates materials possessing notable attributes such as high absorbance, low transmittance, and a low bandgap energy. Within the realm of thin film solar cell research, there exists a notable body of literature addressing the utilization of CuO. Notably, Ramya et al. [21] conducted a study focusing on CuO thin films fabricated through chemical bath deposition for solar cell applications, yielding commendable outcomes. However, their findings indicated that the transmission did not decrease by the anticipated 26% within the visible wavelength range. In contrast, our research expands upon this by revealing significantly diminished transmission levels (approximately 6.48% at 800 nm), coupled with heightened absorbance and a low bandgap energy of 1.61 eV. This remarkable performance can be attributed to the inherent hydrophobic characteristics of our film, further solidifying its viability as a prime material for solar cell fabrication. Furthermore, our results bear similarity to those attained using CIS, CIGS, CdTe, and Co_3O_4, esteemed materials widely recognized and employed as absorber layers in solar cell applications [22–24].

Table 1. Transmission values in the visible region of our films.

Deposition time, (min)	Max. T(%) for λ 800 nm	Thickness, (nm)
5	23.12	103.50
10	13.79	215.71
15	10.52	380.49
20	6.48	532.50

The refractive index (n) is very important in determining the optical properties of semiconductors, knowledge of the latter is essential in the design of hetero-structures of lasers and optoelectronic devices. From the transmission spectra we can determine the refractive index of our films, employing the Moss relationship [15].

$$E_g n^4 = k \tag{3}$$

where k is a constant of value 108 eV. A different relationship between refractive index and bandgap energy was presented by Herve and Vandamme as follows:

$$n = \sqrt{1 + \left(\frac{A}{Eg + B}\right)^2} \tag{4}$$

where A and B are numerical constants with values of 13.6 eV and 3.4 eV, respectively.

The variation of refractive index (n) with deposition time (film thickness) is shown in Figure 9 for these two models. As shown in Figure 9, the refractive index values increased from 2.72 to 2.89 with increasing deposition time. However, the rate of increase varied depending on the model employed. This observed behavior can be attributed to factors such as surface roughness (as depicted in Fig. 9), surface state, and the thickness of the produced films.

3.4 Electrical analysis

Hall effect measurements in Van Der Pauw mode were carried out on CuO films as a function of the deposition time varying from 5 to 20 min on ordinary glass substrates.

In Figure 10, the resistivity of CuO films is depicted as a function of deposition time. Following this figure, the variation of electrical resistivity with deposition time exhibits a bell-shaped curve: it increases with deposition time (t), reaching a maximum at $t = 5$ min, then decreases. This trend is directly related to the concentration of charge carriers. The decrease in resistivity can be attributed to less stable deposition conditions or weaker adhesion of CuO atoms to the substrate surface. This promotes faster but less homogeneous growth of crystallites, leading to larger crystallite sizes. Conversely, longer deposition times allow

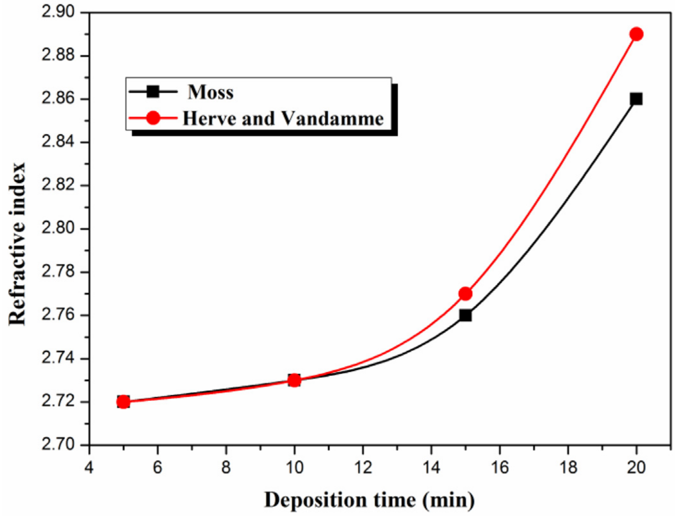

Fig. 9. Variation of the refractive index (n) of the film with deposition time for CuO thin films.

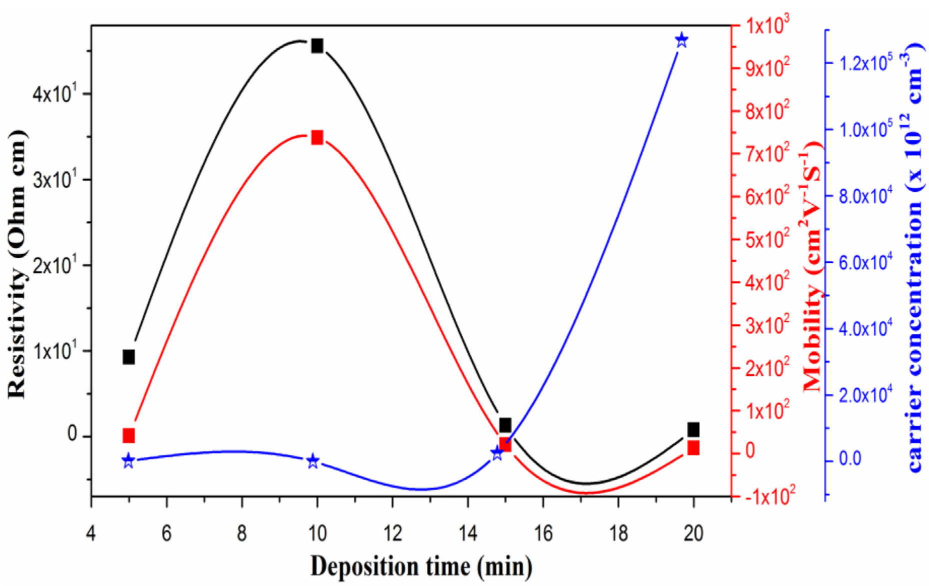

Fig. 10. Evolution of resistivity (ρ), charge carrier mobility (μ) and charge carrier concentration (n) of CuO films as a function of deposition time.

for slower but more controlled crystallite growth, resulting in better organization of CuO atoms and a narrower distribution of crystallite sizes. This can result in CuO films with improved structural homogeneity and smaller crystallite size, enhancing electrical conductivity by reducing obstacles to charge carrier diffusion. The surface morphology of the CuO layer may also change with deposition time, potentially affecting conductivity. A rougher surface can increase conductivity by enlarging the contact area between CuO grains, facilitating charge carrier transport. These results are consistent with the literature [5,6,16]. Ben Saad et al. [5] confirmed that the increase of CuO film thickness leads to an improvement of the conductivity. We can conclude that the lowest resistivity and the highest carrier concentration were obtained for deposition time equals to 20 min. We can also notice a correlation between the contact angle study and the carrier concentration of CuO thin films. According to Nezzari et al. [7], the hydrophobic nature of absorbent films contributes to the enhancement of electrical properties. This assertion is supported by a contact angle measurement of 105° obtained for a deposition time of 20 min. Moreover, the prolonged deposition time fosters an augmentation in the concentration of charge carriers, consequently leading to a reduction in resistivity for copper oxide (CuO) films produced through the spray pyrolysis method ($\rho = 3.77$ Ω.cm). This reduction contrasts with the resistivity of films produced via the electrodeposition method ($\rho = 342$ Ω.cm, [16]) over a 20 min deposition period.

4 Conclusion

CuO thin films were elaborated successfully on ordinary glass substrates by spray pyrolysis technique. The effect of deposition time on structural, morphological, wettability, optical and electrical properties of these films was investigated. XRD analysis indicates that all deposited film are polycrystalline in nature with monoclinic structure. Atomic force microscopy (AFM) highlights the effect of deposition time on the copper oxide surface, showing low roughness for the 10 min film, indicating a smooth surface compared to other films produced. The contact angles recorded for films deposited for less than 20 min are all below 90°, indicating their hydrophilic nature. Conversely, the 20 min deposition film exhibits a hydrophobic trait, as evidenced by the contact angle exceeding 90° (CA = 105°) who is important characteristic for solar cells. These findings align well with the AFM analysis. The optical characterization showed a low transmittance in the visible range of a value of 6.48% for a deposition time equal to 20 min. The optical band gap was found to be decreasing from 1.98 to 1.61 eV as the films' deposition time increases. The electrical study indicates that the electrical properties (resistivity and concentration of free carriers) of the samples are also improved by increasing the deposition time with a low value of resistivity for the film doped at 20 min ($\rho = 3.77\ \Omega$.cm). Furthermore, the hydrophobic nature of the film induced an improvement in the concentration of free carriers which gives better electrical properties. In conclusion, this study highlights the significance of increasing the deposition time as an important parameter for improving the physical properties of CuO films. The findings suggest that a hydrophobic CuO film deposited for 20 min shows potential as an absorbent layer in photovoltaic applications.

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Conflicts of 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.

Data availability statement

No data was used for the research described in the article.

Author contribution statement

The experimental work and the writing of the manuscript were done by Warda Darenfad. The experimental protocols, the data analysis and the interpretation of the results were performed by Warda Darenfad, Noubeil Guermat and Kamel Mirouh. Warda Darenfad and Noubeil Guermat contributed to the extensive revising of the manuscript. All authors discussed the results and commented the manuscript.

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