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Influence of Air Annealing on the Optical Properties of CuO Thin Films Prepared by Chemical Bath Deposition (CBD)

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Abstract: The effect of air annealing on the optical characteristics of CuO thin films produced on glass substrates by Chemical Bath Deposition (CBD) process was studied by annealing the films in ambient air at different temperatures. The films as deposited were air annealed at temperatures of 350°C and 450°C for one hour. Optical properties of the films were studied by UV-Visible spectroscopy in the wavelength range of 200-1200 nm.

The results showed that the air annealing has a substantial effect on the optical behaviour of CuO thin films. As the annealing temperature increased, significant changes in the absorbance and transmittance spectra were observed, indicating an enhancement in the film quality and crystallinity. The absorption coefficient was observed to increase with annealing temperature due to increased grain development and decreased flaw density. The optical band gap was calculated using the Tauc's relation. The band gap energy was gradually changed with increasing annealing temperature. These modifications can be explained by structural reorganization, decrease of lattice defects and enhanced stoichiometry of CuO films during the annealing.

This work shows that air annealing is a promising post-deposition procedure for tuning the optical characteristics of CBD produced CuO thin films for their application in photovoltaic devices, photodetector and optoelectronic systems. The results provide useful information on how to optimize the annealing conditions for obtaining better optical performance in CuO-based thin-film devices.

Keywords: *CuO Thin Films, Chemical Bath Deposition, Air Annealing, Optical Properties, Optical Band Gap.*

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I. INTRODUCTION

Copper oxide (CuO) is a p-type semiconductor with a narrow band gap (1.2–2.1 eV) and a high optical absorption coefficient. It is non-toxic and is produced in plenty at low cost, and has therefore received much interest. These features make CuO a potential material for numerous applications such as solar cells, photodetectors, gas sensors, photocatalysts, lithium-ion batteries and other opto-electronic devices [1-4].

The optical characteristics of CuO thin films such as absorbance, transmittance, absorption coefficient and optical band gap plays an important role in establishing the suitability of thin films for optoelectronic applications [4]. These attributes are significantly dependent on the deposition procedure and post-deposition treatments. Hence, regulation of optical properties of CuO thin films is important for improving the device performance.

Among numerous thin film manufacturing techniques, Chemical Bath Deposition (CBD) is a simple, cost effective and scalable approach, which has the capability to produce homogeneous films over wide substrate regions [5]. The approach has the benefits of low process temperature, simple operation and effective control of the film thickness and composition. However, as-deposited films are typically suffered from poor crystallinity, structural flaws and residual stress, which could severely affect their optical and electrical properties.

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Post-deposition annealing is frequently used to improve the quality of thin films by increasing crystallinity, stimulating grain development, decreasing defects, and improving stoichiometry [6]. In particular, air annealing promotes oxidation and structural rearrangement, resulting to considerable changes in the optical properties of CuO films. The annealing temperature substantially affects the micro-structure and electrical structure of the material, which also affects the optical performance.

Although several investigations on annealing effects on CuO thin films created by diverse processes have been reported, only a few reports on the annealing of CuO films grown by CBD in air have appeared. Therefore, the present work is focused on investigating the effect of air annealing on the optical characteristics of CuO thin films generated by chemical bath deposition method. The films were annealed at different temperatures and evaluated by UV-Visible spectroscopy to measure the change in absorbance, transmittance, absorption coefficient and optical band gap. The results give useful information for optimization of the annealing conditions and enhancement of the performance of CuO-based optoelectronic and photovoltaic systems [7].

II. EXPERIMENTAL METHODS

CuO thin films were deposited on thoroughly cleaned glass substrates using the Chemical Bath Deposition (CBD) technique. A 0.1 M copper chloride precursor solution was prepared by dissolving 1.705 g of copper (II) chloride dihydrate ($CuCl_2 \cdot 2H_2O$) in 100 mL of double-distilled water under continuous stirring. The solution was heated from room temperature to 60°C to ensure complete dissolution of the precursor. Subsequently, 5 mL of aqueous ammonia (NH_3) was added dropwise as a complexing agent while maintaining constant stirring. The bath temperature was then increased to 70°C. Clean glass substrates were immersed vertically in the reaction bath, and the deposition process was carried out for 2 h. After deposition, the substrates were left in the growth solution for an additional 20 h to facilitate complete film formation and adherence. The deposited films were then removed, rinsed thoroughly with double-distilled water to eliminate loosely bound particles and residual impurities, and dried at room temperature before further characterization and annealing studies.

The as-deposited CuO thin films prepared by the Chemical Bath Deposition (CBD) technique were subjected to post-deposition air annealing to investigate the effect of annealing temperature on their optical properties. Annealing was carried out in a muffle furnace under ambient atmospheric conditions at two different temperatures, namely 350°C and 450°C. The samples were heated at a controlled rate to the desired temperature and maintained at that temperature for 1 h to ensure uniform heat treatment throughout the film. During the annealing process, the films underwent structural reorganization, grain growth, and reduction of defects, which are expected to improve the crystallinity and stoichiometry of the CuO films. After the annealing period, the samples were allowed to cool naturally to room temperature inside the furnace to minimize thermal stress and prevent film cracking. The annealed films were subsequently characterized and compared with the as-deposited films to evaluate the influence of air annealing on their optical properties, including absorbance, transmittance, absorption coefficient, and optical band gap energy.

III. RESULTS AND DISCUSSIONS

Figure 1 illustrates the optical transmittance spectra and the corresponding Tauc plots of CBD deposited CuO thin films. The films include the as-deposited sample (C1) prepared at 70 °C and the air-annealed samples C2, and C3 annealed at 350 °C and 450 °C, respectively.

The transmittance spectra recorded in the wavelength range 200–1200 nm show that the optical transmittance of the films increases progressively with annealing temperature. The as-deposited film (C-C1) exhibits the lowest transmittance, while the film annealed at 450 °C (C-C4) shows the highest transmittance. This increase in transparency after annealing can be attributed to the improvement in crystallinity, reduction of structural defects, and removal of residual impurities present in the as-deposited films [6]. Annealing also promotes grain growth and film densification, which reduces light scattering at grain boundaries and enhances optical transmission.

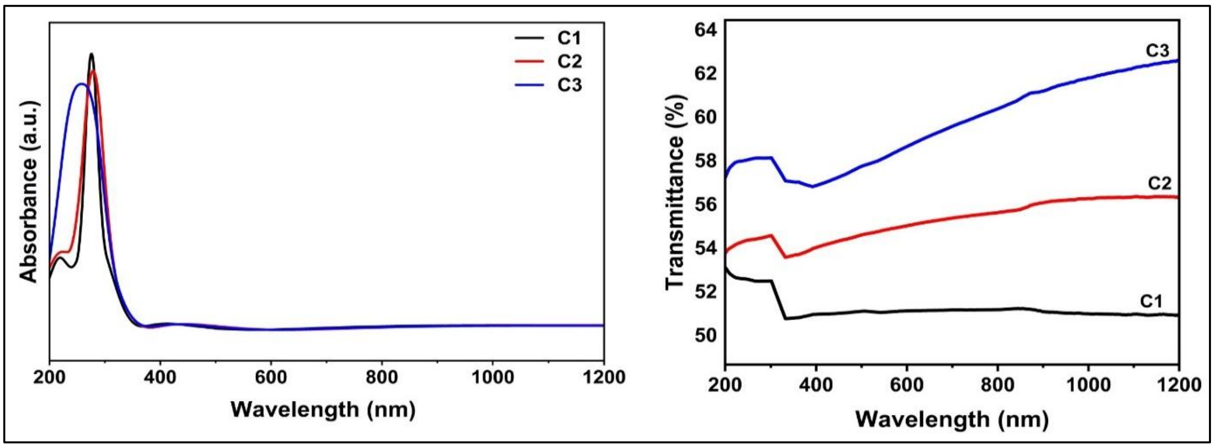

Fig 1: Absorbance and Transmittance Spectra of as Deposited(C1) and Post-Air-Annealed (C2, C3) Chemical Bath Deposited CuO Films

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The UV–Visible absorption spectra of the as-deposited and air-annealed CuO thin films are shown in the fig. 1. All the samples exhibit strong absorption in the ultraviolet region, followed by a sharp decrease in absorbance with increasing wavelength. The as-deposited film (C1) shows the highest absorbance, whereas the absorbance decreases progressively for the films annealed at 350 °C (C2) and 450 °C (C3). This reduction in absorbance with annealing temperature indicates an improvement in the optical quality of the films, which may be attributed to enhanced crystallinity, grain growth, and a decrease in defect states and scattering centres. In the visible and near-infrared regions, the absorbance remains very low, suggesting increased transparency of the annealed films. The observed decrease in absorbance is consistent with the corresponding increase in optical transmittance, confirming that air annealing improves the optical properties of CuO thin films. Among all the samples, the film annealed at 450 °C exhibits the lowest absorbance and highest transparency, indicating superior optical quality.

The optical band gap of the CuO thin films was estimated using the Tauc relation $(\alpha h\nu)^2 = A(h\nu - E_g)$assuming a direct allowed transition. The as-deposited film (C1) shows a band gap of about 2.16 eV**.** With increasing annealing temperature, the band gap gradually decreases to 2.14 eV (C2) and 1.92 eV (C4) [8]**.** This reduction in band gap may be attributed to improved crystallinity, grain growth, and reduced defect states after annealing.

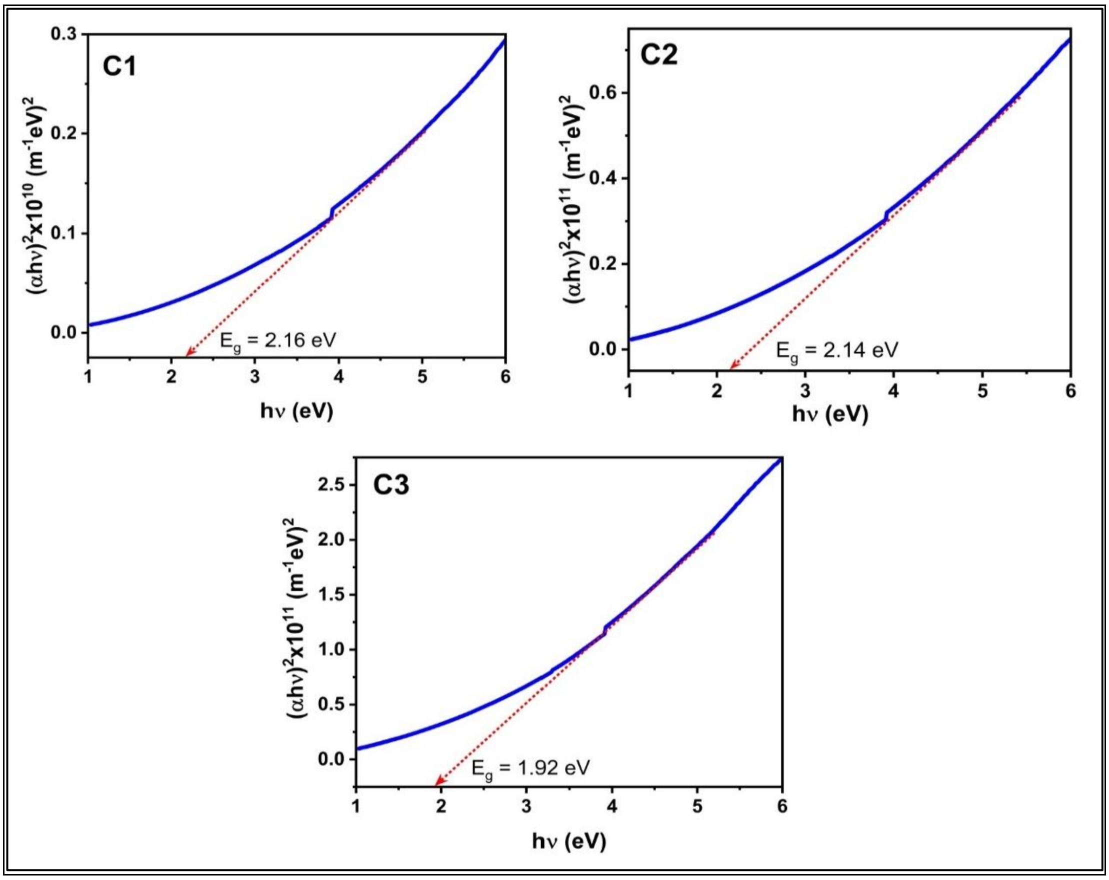

Fig 2: Tauc Plots of as-Deposited (C1) and Post-Air-Annealed (C2, C3) Chemical Bath Deposited CuO thin Films

Figure 2 presents the Tauc plots of CuO thin films deposited by the Chemical Bath Deposition (CBD) technique. The samples include the as-deposited film (C1) and films annealed in air at 350 °C (C2) and 450 °C (C3) for 1 h.

The transmittance spectra measured in the wavelength range of 200–1200 nm reveal a noticeable dependence of optical transmission on annealing temperature. The as-deposited film exhibits relatively low transmittance throughout the investigated spectral region, indicating the presence of a higher concentration of structural imperfections and scattering centres. Upon annealing, the transmittance of the films increases significantly, with the sample annealed at 450 °C showing the highest transparency. The enhancement in optical transmission can be associated with the improvement in film quality resulting from thermal treatment. Air annealing facilitates crystallite growth, reduces lattice defects and microstrain, and promotes a more homogeneous microstructure. These changes decrease the scattering and absorption losses within the film, thereby increasing the amount of transmitted light.

The optical band gap energy (Eg) was determined from the Tauc plots (Fig. 2) by extrapolating the linear portion of the $((\alpha h\nu)^2)$ versus $(h\nu)$ curves to the energy axis, assuming a direct allowed electronic transition. The estimated band gap values were found to be 2.16 eV for the as-deposited film (C1), 2.14 eV for the film annealed at 350 °C (C2), and 1.93 eV for the film annealed at 450 °C (C3) [9]. The observed decrease in band gap energy with increasing annealing temperature indicates a modification of the electronic structure of the CuO films. This behavior can be attributed to enhanced crystallinity, increased grain size, and a reduction in localized defect states within the band structure. The thermal treatment promotes better atomic ordering and reduces grain boundary density, resulting in the narrowing of the optical band gap. Such a reduction in band gap energy is advantageous for applications requiring enhanced visible-light absorption, including photovoltaic and optoelectronic devices.

Overall, the results demonstrate that air annealing is an effective post-deposition treatment for tailoring the optical properties of CBD-grown CuO thin films. Higher annealing temperatures lead to improved optical transparency and lower band gap energies, indicating enhanced material quality and suitability for optoelectronic applications [10].

Table 1: Film Thickness and Optical Bandgap of as-Deposited and Air-Annealed Chemical Bath Deposited CuO thin Films

Sample	**Film thickness (t) nm**	**Optical band gap (E_g) eV**
C1	79.68	2.16
C2	67.73	2.14
C3	46.36	1.92

IV. CONCLUSION

Overall, the results demonstrate that air annealing is an effective post-deposition treatment for tailoring the optical properties of CBD-grown CuO thin films. With increasing annealing temperature, the films exhibit enhanced optical transparency and reduced optical absorption in the visible region, indicating a significant improvement in film quality. The observed decrease in optical band gap energy may be attributed to improved crystallinity, grain growth, and the reduction of structural defects and localized states within the band structure. Annealing also promotes better atomic ordering and reduces defect-induced scattering, thereby facilitating more efficient light transmission through the films. These improvements in optical characteristics suggest that the annealed CuO thin films possess superior structural and electronic properties compared to the as-deposited films. Consequently, the films annealed at higher temperatures, particularly at 450 °C, are promising candidates for various optoelectronic and photovoltaic applications, including photodetectors, solar cells, transparent electronic devices, and optical sensing systems.

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