[1] Rana Talib Saihood

[2]Nadir Fadhil Habubi

[3]Raghad Hamdan Mohsin

[4]Ibrahim Ramadhan Agool

[5]Sami Salman Chiad

[6]Abeer Ghalib Hadi

[7]Jameel M. Dhabab

Characterization of Indium Concentration Upon Properties of Nanostructured Copper Oxide Thin Films

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Abstract: - By applying the spray pyrolysis process at 350°C, CuO and In-doped CuO films with various In content of (0, 2 and 4%) were created on glass substrates. CuO films are confirmed to have a polycrystalline cubic structure by XRD analysis, with a recognizable peak at (200). As indium concentration rises, grain size grows from 13.74 nanometers to 15.82 nanometers, but the strain (%) parameter falls from 25.21 to 21.91. AFM measurements demonstrate that nanostructure exists. The average diameter size, surface roughness and rms values of the prepared films were (68.8, 49.7 and 31.2) nm, (9.77, 4,73 and 4.19) and (7.79, 6.60 and 2.23) nm for CuO, CuO: 2% In and CuO: 4% In respectively. Within the scope of this study, an investigation of the effect of indium dopant on the optical characteristics of CuO films was carried out. The transparency properties is decreased with indium doping increases, and the absorption coefficient (α) increases with indium content. The band gap values for the CuO and CuO: In films were found to be 1.98 ,1.93 and 1.87 eV with In content, respectively. The refractive index and the CuO thin film extinction values are changed with indium doping concentration change.

Keywords: Copper oxide, Indium doping, XRD, optical properties, Topography.

I. Introduction

The benefits of copper oxide, including its nontoxicity, affordability, and environmental appropriateness, make it a desirable material [1-3]. It receives a lot of attention because it is a well-known p-type semiconductor,and a band gaps of 1.3-2.1 eV and 2.0-2.6 eV [3,4]. Numerous applications, including catalysis, water splitting, gas sensors and solar cells [5-11], have shown the potential usage of cupric oxide thin films. Several deposition processes, like sputtering [12], plasma evaporation [13], spray pyrolysis [14-20], electro-deposition [21], and sol-

[1] *General Directorate of Education in Baghdad Governorate Rusafa second Ministry of Education* Baghdad, Iraq ranatalib353@gmail.com

[2]*Dept. Engineering of Refrigeration and Air Conditioning Technologies Alnukhba University College* Baghdad, Iraq

n.fadhil@alnukhba.edu.iq

[3]*General Directorate of Education in Baghdad Governorate Rusafa second Ministry of Education* Baghdad, Iraq rghdhmdan598@gmail.com

[4]*Bilad Alrafidain University College* Diyala, Iraq

dr.agool@bauc14.edu.iq

[5]*Dept. Physics College of Education, Mustansiriyah University* Baghdad, Iraq dr.sami@uomustansiriyah.edu.iq

[6]*General Directorate of Education in Baghdad Governorate Rusafa second Ministry of Education* Baghdad, Iraq abeerghalibhadia@gmail.com

[7]*Dept. Physics Alnukhba University College Baghdad, Iraq* j.mousa@alnukhba.edu.iq

gel dip-coating [5, 7], have been used to create copper oxide. Even though spray pyrolysis is a low-cost and straightforward processing technique, copper oxide thin films made using other soft chemistry processes, including this one, exhibit good qualities for most applications mentioned. It offers attractive benefits, such as being nonhazardous and excellent for depositing [11]. According to reports, under ambient deposition circumstances, it creates uniform film structures with broad areas, good crystallinity, a balance of thickness and stoichiometry, and phase purity [14, 15]. P type semiconductors make up the majority of copper oxide [22–25]. When different numbers of dips are analyzed and associated with morphological and structural features, the impact of the film thickness on the primary optoelectronic properties is examined. In this study, we investigated the effect of CuO films doped with indium on their structural, morphological, and optical characteristics.

II. EXPERIMENTAL

O.1 M of $CuCl_2$ was ressolved in a mixture of 1:1 deionized (W_D)and ethanol to create the CuO thin films. Indium trichloride ($InCl_3$), supplied by PubChem India, was used as the doping agent and was diluted in W_D. For the purpose of making the solution transparent, a few droplets of HCl were provided. Indium-doped CuO film was prepared via chemical spray pyrolysis and applied to a glass slide substrate. These are the preparatory requirements: 350 ^oC was the substrate temperature, 30 cm separated the nozzle from the substrate, 10 seconds of spraying time were followed by 90 seconds to prevent cooling, 4 ml/min was the spray rate, and N_2 was utilized as the carrier gas. To ascertain film thickness, the gravimetric method was employed., which was 335 ± 20 nm. By using XRD, structural characteristics were assessed. AFM was used to examine the films' surfaces. UV-Visible spectrophotometer was used to capture the 300-900 nm absorbance spectra.

III. RESULTS AND DISCUSSIONS

XRD patterns of the intended films are offered in Fig. 1. The obtained results show that the films were polycrystalline. These results matched wurtzite, CuO, and peaks at 37.26°, 43.24°, and 62.89° that correspond to the (111), (200), and (220) reflections (JCPDS card No 05-0661). Strong peak was seen approaching (111), which was in line with previous researchers' findings [26].

By applying the formula in Eq. 1, the grain size, denoted by "D," was calculated. [27-29]:

$$D = \frac{0.9\lambda}{\beta cos\theta} \qquad (1)$$

Where λ is wavelength of the X-rays, β and θ are (FWHM) and Bragg's angle, respectively. The gathered information is shown in Table 1. As the indium concentration rises, it has been demonstrated that grain size increases from 13.74 to 15.82nm. Indium concentration is hence appropriate for determining the grain size of a material [30, 31].

Evaluation is also done on other structural metrics, including dislocation density (δ). Table 1 reveals the structural characteristics obtained, where δ can be gained by [32-34]

$$\delta = \frac{1}{D^2} \qquad (2)$$

The strain (ε) is calculated by employing Eq. 3 [35-37]:

$$\varepsilon = \frac{\beta cos\theta}{4} \qquad (3)$$

Strain decreases as indium doping levels rise [38, 39]. Table 1 presents the determined structural parameters Pst.via Indium dopant as seen in Figure 2.

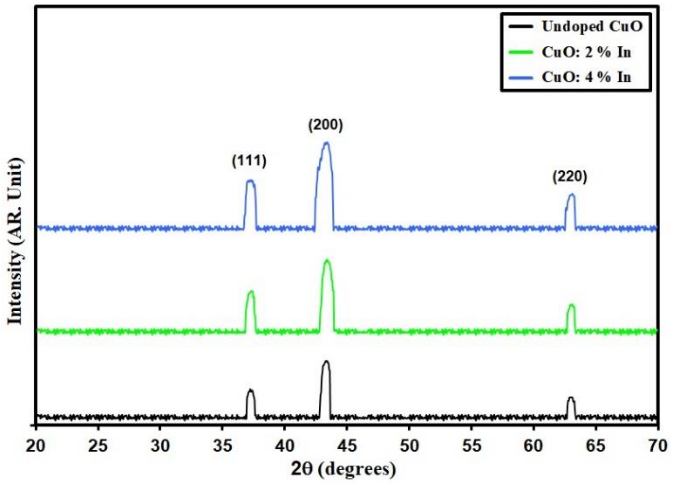

Fig.1. XRD-patterns.

The AFM micrograph for CuO and CuO is shown in Fig. 3: thin films Average Particle Size (P_{av}) values for Undoped CuO, CuO: 2% In, and CuO: 4% In were (68.8, 49.7, and 31.2) nm, respectively. By raising the CuO to CuO: 3% In, the surface roughness (Ra) decreases from 9.77 nm to 4.19 nm. The rms values of the formed films were 7.79, 6.60, and 2.23 nm for CuO, 2% In, and 4% In, respectively. Table 2 provides the AFM parameters Ap. These findings suggest that indium doping can significantly affect R_a, rms, and P_{av}, which could have implications for their electronic and optical properties [40, 41].

TABLE 1. *D, Eg and* P_{st} *of the grown films.*

Specimen	2 q (°)	(hkl) Plane	FWHM (°)	E_g (eV)	*D* (nm)	δ (× 10^{14}) (lines/m^2)	ε (× 10^{-4})
Undoped CuO	37.26	200	0.61	1.98	13.74	52.96	25.21
CuO: 2% In	37.23	200	0.56	1.93	14..97	44.62	23.14
CuO: 4% In	37.2	200	0.53	1.87	15.82	39.95	21.91

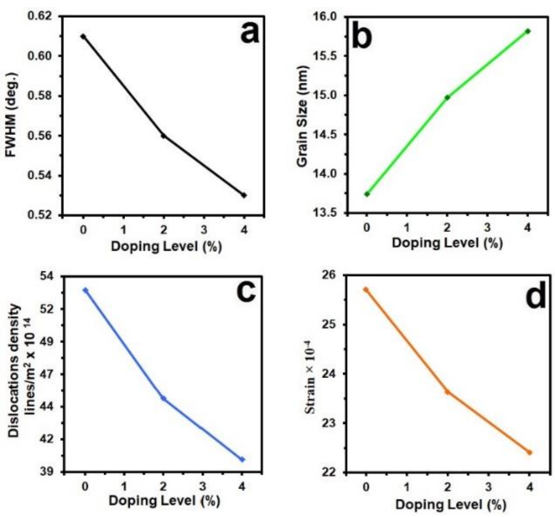

Fig.2. Structural parameters of the deposited films

TABLE 2. A_p of grown films.

Samples	P_{av} nm	R_a (nm)	rms (nm)
Undoped CuO	68.8	9.77	7.79
CuO: 2 % In	49.7	4.73	6.6
CuO: 4 % In	31.2	4.19	2.23

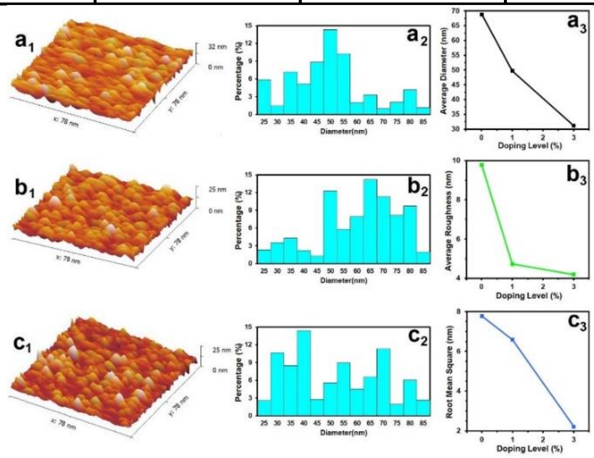

Fig. 3. A_P of the grown films.

Measurements of transmittance (T) in the UV-vis range were used to characterize the optically undoped and indium-doped CuO films. The transmittance curves of CuO and indium-doped thin films (2 and 4%) are shown in Fig. 3. When indium concentration was increased, the transmittance in the UV-vis region declined and reached over 66% at undoped CuO films. As indium doping concentration rises, the transmission edge shifts to the side with longer wavelengths. More indium being incorporated into the CuO lattice and a slight rise in the concentration of free charge carriers may be the causes of the loss in transparency of CuO: In films coated with 1 and 4% In concentration [42-44].

Fig. 4 shows a plot of absorption coefficient (α) versus wavelength. It can be calculated from equation (4) [45-47]:

$$\alpha = \frac{\ln(1/T)}{d} \qquad (4)$$

Where d is film thickness. Fig. 5 shows α rises indium dopant concentration rises. The carrier absorption with indium doping is the cause of the rise in absorption coefficient with an increase in indium doping concentration [48-51].

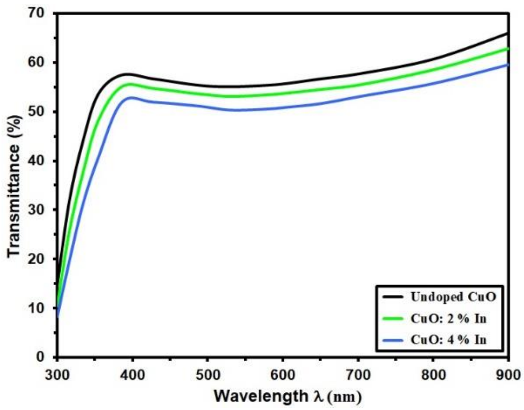

Fig. 4 T of the grown films

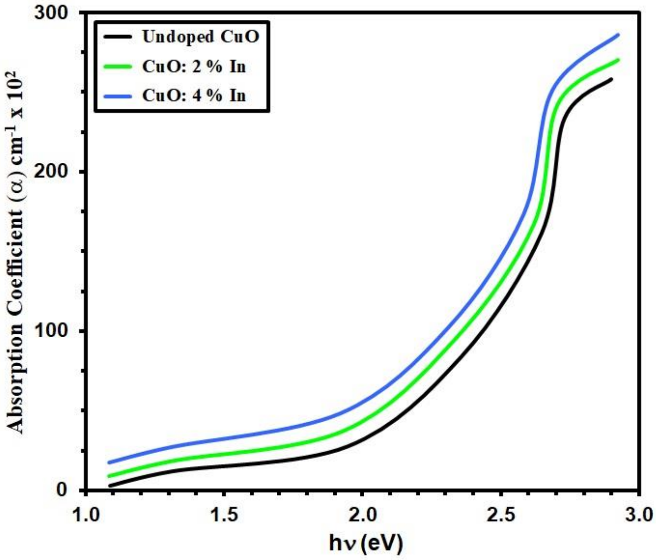

Fig. 5 αα for deposited films.

The optical band gap energy (E_g) is obtained via Tauc's formula[52-54]:

$$(\alpha h\nu) = A\left(h\nu - E_g\right)^{\frac{1}{2}} \qquad (5)$$

Where A is constant. The relationship was linearly dependent for n=1/2, E_g was demonstrated as offered in Fig. 6, the undoped CuO thin film recorded a greater band gap value than those doped with 0, 2, and 4% indium. When shown, as the proportion of indium doping increased, the band gap values of CuO film dropped. This drop is brought about by the dopant's propensity to widen the valence band of CuO film, which reduces the energy required for electrons to jump from the valence band to the conduction band. The visible light capture in solar cell applications can be enhanced by using this reduction in band gap [55, 56].

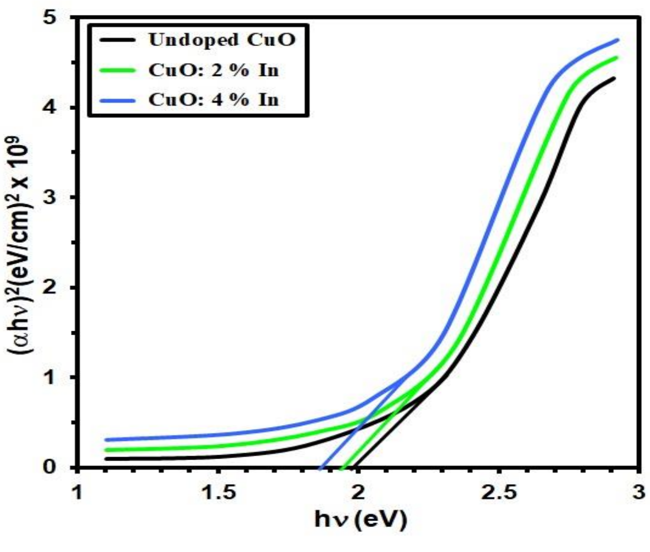

Fig. 6 E_g of the intended films.

The extinction coefficient (k) calculation by using the equation [57,58]:

$$k = \frac{\alpha\lambda}{4\pi} \qquad (6)$$

As the quantity of the dopant increased, the samples' extinction coefficient dropped, as seen in Fig. 7. The most significant value of the extinction coefficient, 0.75, was produced by the undepleted CuO films, followed by the 2% films with 0.72. The extinction coefficient of the 4% doped deposit was 0.67, and it was discovered that the extinction coefficient of the deposited CuO declined as the amount of indium dopant increased. Because α increased with the dopant content, k dropped [59, 60].

Refractive index (n) of the deposited Undoped and CuO: In was obtained from the formula [61,62]:

$$n = \left(\frac{1+R}{1-R}\right) + \sqrt{\frac{4R}{(1-R)^2} - k^2} \qquad (7)$$

Where R is the reflectivity. Figure 8 shows that at wavelength 520 nm, the Undoped CuO films had the most significant refractive index values (3.35) and the 4% Indium-Doped CuO films had the lowest values (3.14). The deposited materials' refractive indices showed an exponential drop with wavelength between 600 and 900 nm, indicating limited light absorption at long wavelengths [63]. Undoped and (CuO: In) films formed at indium doping concentration exhibit identical behavior of the extinction coefficient regarding wavelength [64].

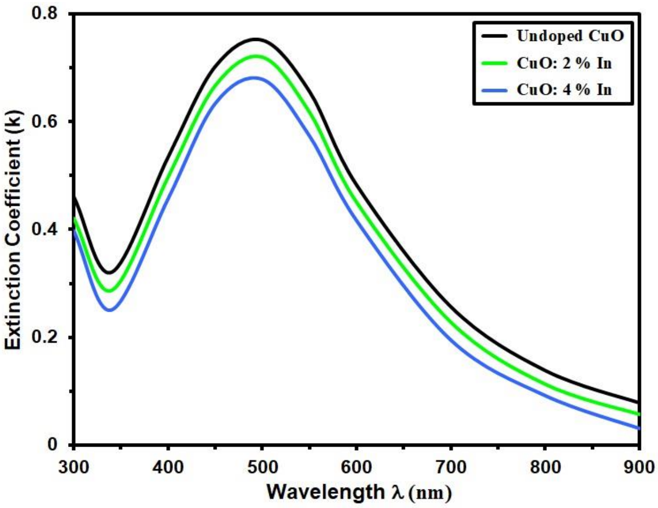

Fig. 7 k of the grown films.

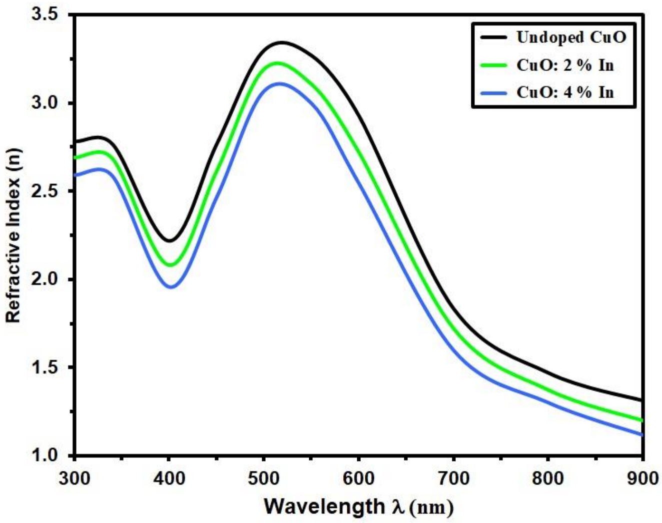

Fig. 8 n for grown films.

IV. CONCLUSION

On glass substrates, indium-doped CuO thin films have been created using (SPT) using different doping concentrations at 300 °C. According to XRD measurements, CuO films contain a dominating peak (200). The strain (%) parameter increased from 25.21 to 21.91, while the dislocation density reduced from 52.96 to 39.95. The grain size for undoped CuO particles is around 13.74–15.82) nm with CuO: 4% In. The average particle size grew from (68.8 to 31.2) nm, while the surface roughness decreases from 9.77 to 4.19 nm, according to the AFM picture. When indium concentration was increased, the transmittance in the UV-VIS region dropped and reached over 66% at undoped CuO films. With rising dopant concentration in the visible range, the absorption coefficient rises. The band gap values for the doped indium with (2 and 4%) and the undoped CuO thin film were 1.98 eV and 1.93 eV, respectively. With increasing doping concentration, the absorption coefficient decreases in a manner similar to how the n and k behave.

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