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Structural, Topography and Optical Properties of Bismuth doped CuO films prepared by chemical spray pyrolysis

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Abstract.

CuO and CuO: Bi thin films is prepared by chemical spray pyrolysis (CSP). XRD offer that these films is polycrystalline monoclinic structure with a maximum peak at (200) resulting of the Bi, The crystallite size is set to increase from 15.28to 17.88 nm with Bi doping. whereas the dislocation density (□) parameter decreases from 4.28 to 3.12, while strain decreases from 2.26 to 1.93. AFM images reflect dependence of surface morphology and roughness onto doping. Transmittance offers good transparency, in the visible area between 67.5 and 62.5 , for undoped CuO and 4% Bi content. A small decrease in bandgap, its value was 2.1 eV and 2.0 eV for CuO and CuO:Bi respectively. The studied optical constants are increased via increasing Bismuth -doping.

Keywords: CuO, Bi, Topography, XRD, bandgap, refractive index

Introduction

CuO is a p-type semiconductor, own a bandgap through 1.21 to 1.51 eV [1-5]. CuO is non-toxic and abundance [6,7].There are many different techniques utilized for depositing CuO films such as diode applications [8], field transistors [9], solar cells [10],photovoltaic[11], secondary batteries [12],superconductors [13], gas sensors [14]. CuO can be deposited by various methods such as PLD [15], and ion-based deposition [16,54-74], CBD [17] and chemical spray pyrolysis [2, 3, 4, 18, 19]. CSP technique can employed to prepare metal oxide films due to high melting point of metal oxides [20]. n this research, the copper oxide has been prepared by CSP with various content of Bismuth particles, and study the influence of the additive on its physical properties.

Experimental

CuO and CuO:Bi films were grown onto a preheated glass substrate by CSP technique. After many trials, the preoperative parameters of the system were optimized to gain a regular, adherent thin films. The spraying solution was a mixing of concentration (0.1M) aqueous solution of (1.9965g) of $Cu(CH_3COO)_2$. H_2O resolved in distilled water D_w. 0.1 M of Bismuth tetrachloride dissolved in D_w was added to the matrix solution with 2% and 4% volumetric concentration as a dopant. The substrate temperature was controlled and it was measured using thermocouples. Thin films thickness were measured by weighing method and found to be in range of 330 ±10 nm. The preparation parameters were fixed after many trials as base temperature was 350 ^{O}C. N_2 was employed as a carrier gas. Deposition rate was 4 mL/min, spryer rate was 10 s hold by one min. to stop extravagant cooling. Distance between base and nozzle was 28 cm. The structural properties were obtained by XRD. AFM was employed to study surface topography. The transmittance is recorded via double beam spectrophotometer.

Results and Discussions

XRD styles of CuO and CuO: Bi is offered in Fig.1.. The XRD patterns exhibit peaks at 2θ ≈35. 49°, 38.10°,53.42° & 53.22° corresponding to (002), (200), (020) and (022) planes. These data are compared with ICDD file No.48-1548 data. Peak intensities increase with increasing doping in Bi. Films were polycrystalline and monoclinic, these results were fit with literatures [21-23].

The crystallite size (*D*) was obtained via Scherrer formula[24-27]:

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

Where, λ is the wavelength of X-ray, β is FWHM and θ is Bragg's angle.

Values of *D* were increased with increasing from 15.28to 17.88 nm with Bismuth doping (see Table 1). This increase may be due to the combined effect of the increase in Cu incorporation, an increase in the growth rate and reorientation effect.

The dislocation density δ was gained via the relation [28-31]:

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

the dislocation density (δ) parameter decreases from 4.28 to 3.12 with Bismuth doping

The strain (□) was calculated by the formula [32-34]:

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

We found that □ decreases from 2.26 to 1.93 with Bismuth doping. Table 1 illustrate the resultsof the studied parameters.Fig. (2) displays FWHM, D, δ and ε against Bismuth content.

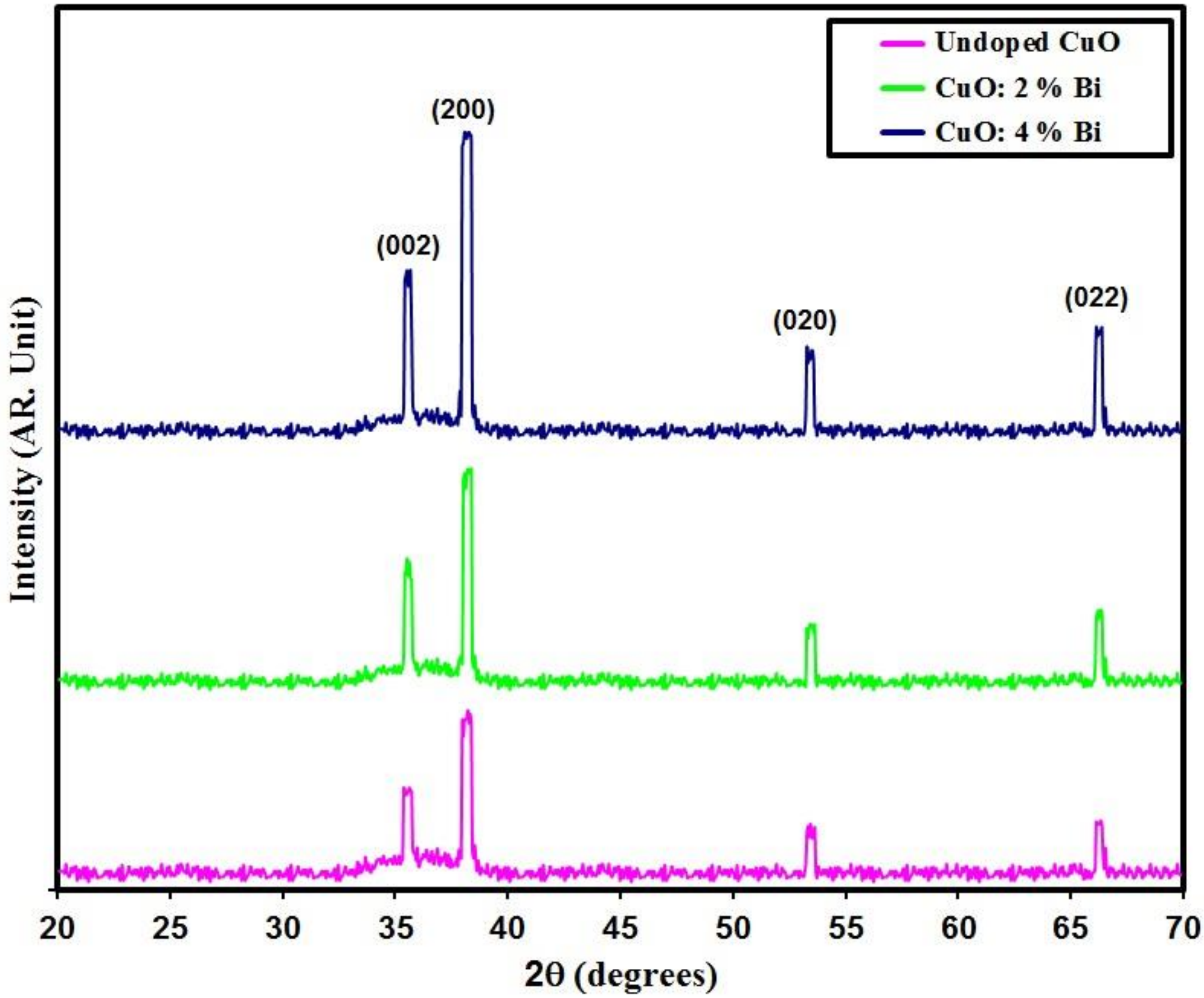

Fig.1. XRD styles of grown films.

Table 1. *D*, E_g, δ and ε of intended films.

Sample	(hkl) Plane	2 □ (º)	FWHM (º)	Optical bandgap (eV)	Grain size (nm)	Dislocations density ($\times 10^{15}$) (lines/m^2)	Strain ($\times 10^{-3}$)
Undoped CuO	200	38.10	0.55	2.10	15.28	4.28	2.26
CuO: 2% Bi	200	38.04	0.51	2.05	16.48	3.68	2.10
CuO: 4% Bi	200	38.00	0.47	2.00	17.88	3.12	1.93

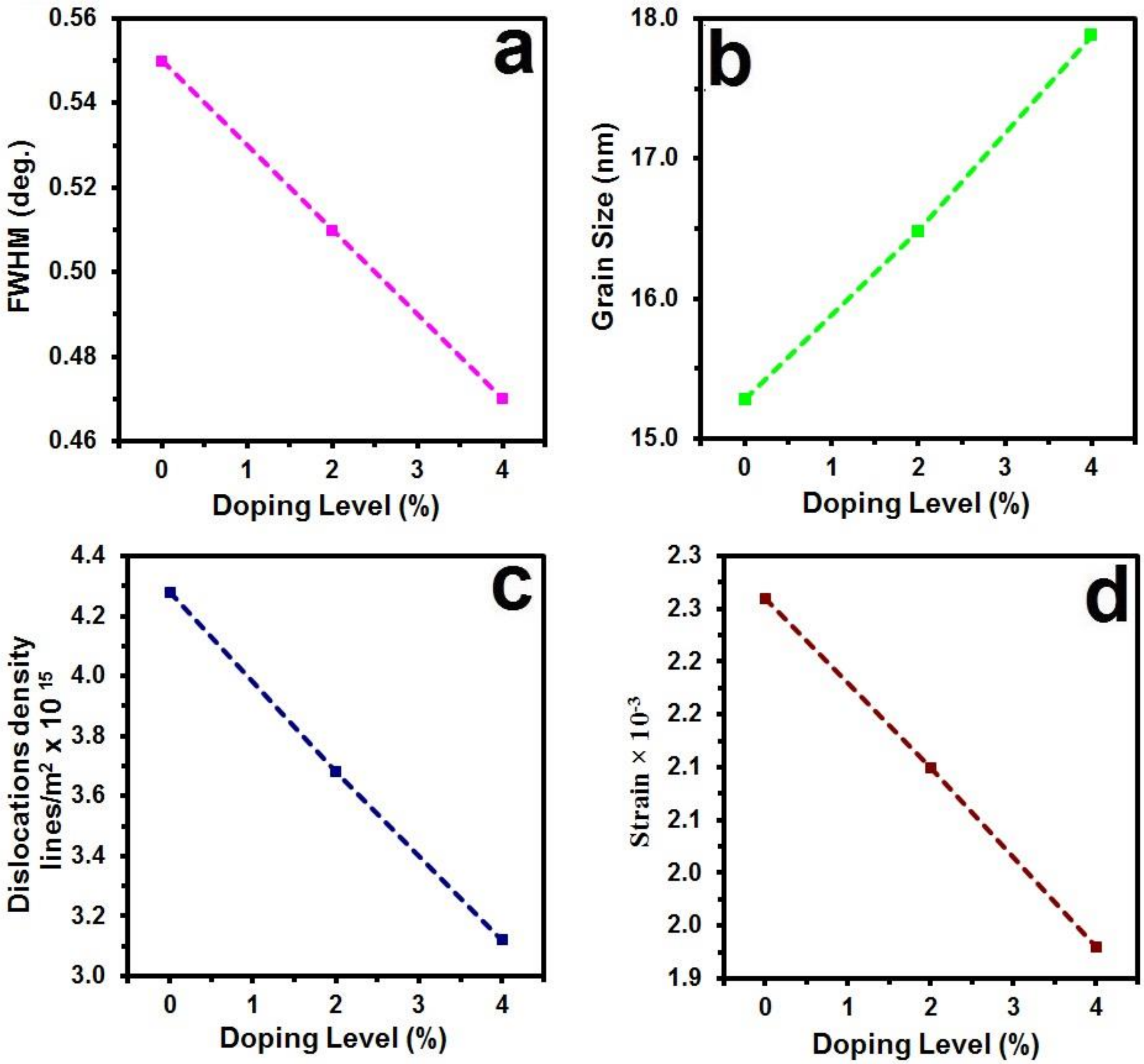

Fig.2. structural parameters of intended films.

Topography Films Undoped CuO and CuO: % Bi were obtained via AFM. Figure 3. Displays the granules ,which exist in films suffer scattered in some zones. surface morphology can be estimated via roughness (Ra) and root mean square roughness (rms). From Figure 3 (a_3, b_3 and c_3). Average Particle size and rms values of (65.50, 58.78 and 52.31) nm and (9.4, 8.71 and 5.46) nm, respectively, which strongly affected by Bismuth dopant.

Table 2 shows that the average roughness and average particle size P_{av} increases Bi content. surface area increment plays an important role in photocatalytic activity.

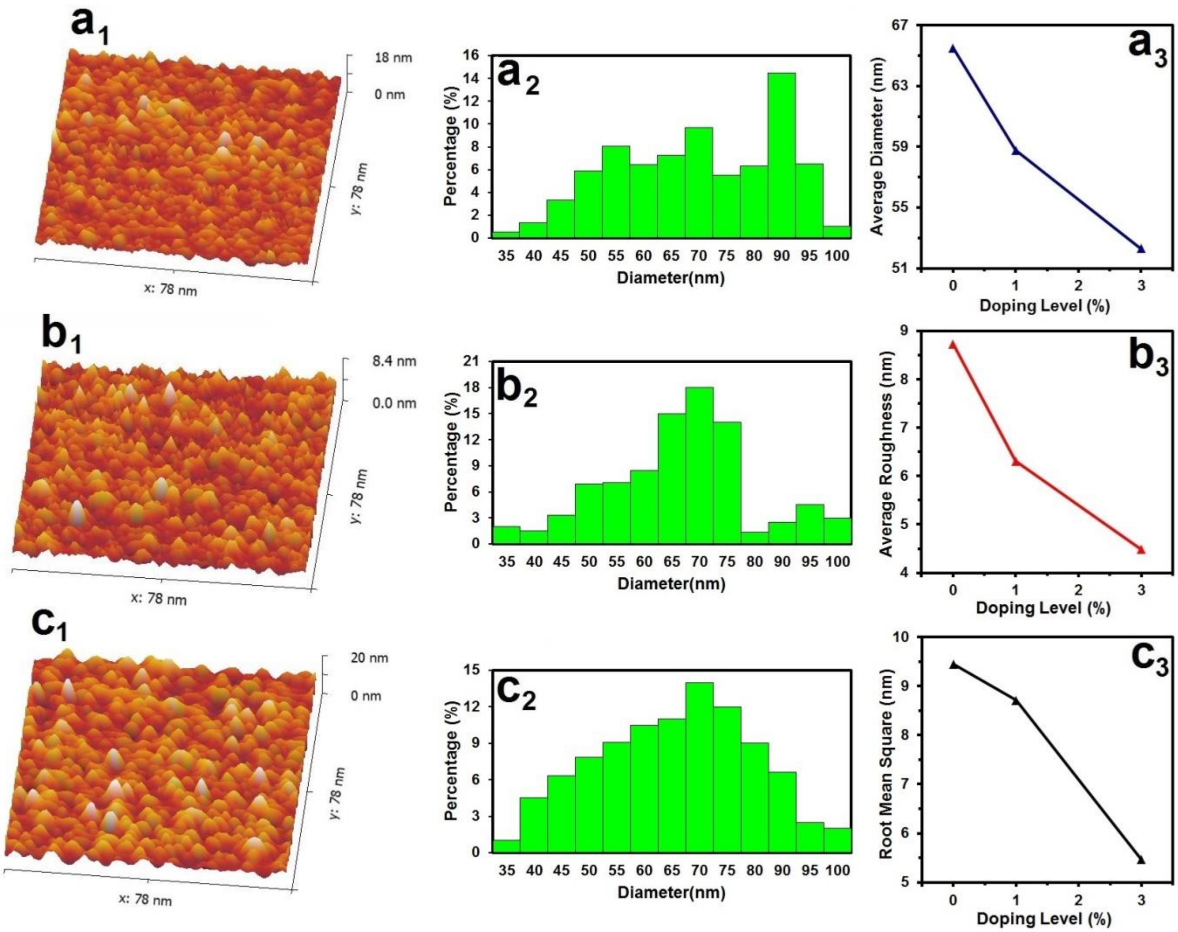

Fig.3. AFM information of intended films

Table 2. AFM parameters of intended films.

Specimen	P_{av} nm	Ra (nm)	rms (nm)
Undoped CuO	65.50	8.73	9.45
CuO: 2% Bi	58.78	6.31	8.71
CuO: 4% Bi	52.31	4.82	5.46

Transmittance (T) spectra are displayed in Fig.4. it can be noticed that T decrease with increase of Bi-doping and increasing with increases wavelength. T shows good transparency in the visible region between 67.5 and 62.5 %, for Undoped CuO and 4% Bi doping The optical absorption coefficient (α) in the fundamental absorption area was obtained using the following equation [35-38]:

$$\alpha = (2.303\times A)/t \quad (4)$$

Where (t) is film thickness and A is absorbance. A increased with increases Bismuth -doping as represented in Fig.5

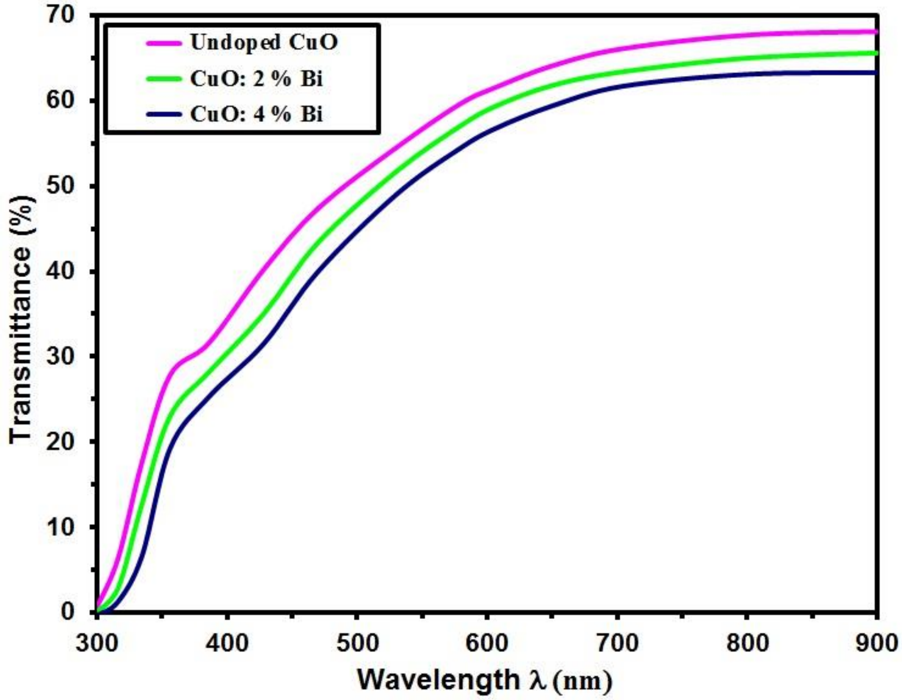

Fig. 4: Transmittance with wavelength of intended films.

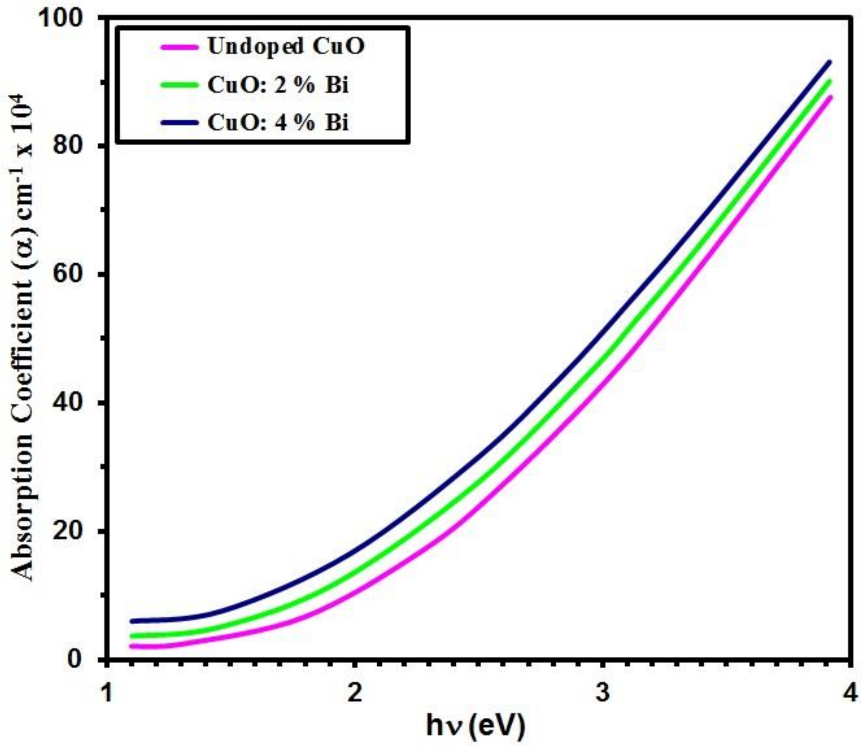

Fig. 5: α against wavelength of intended films.

The optical bandgap E_g of the samples was evaluated following relation [39-42]:

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

Where A is a constant, hυ is a photon energy. Fig. (6) shows the decreases of E_g with increasing Bismuth-doping in CuO film from 2.1 eV to 2.0 eV for 4% Bi-doping. This behavior is a result of make sub-levels in the bandgap that decrease E_g.

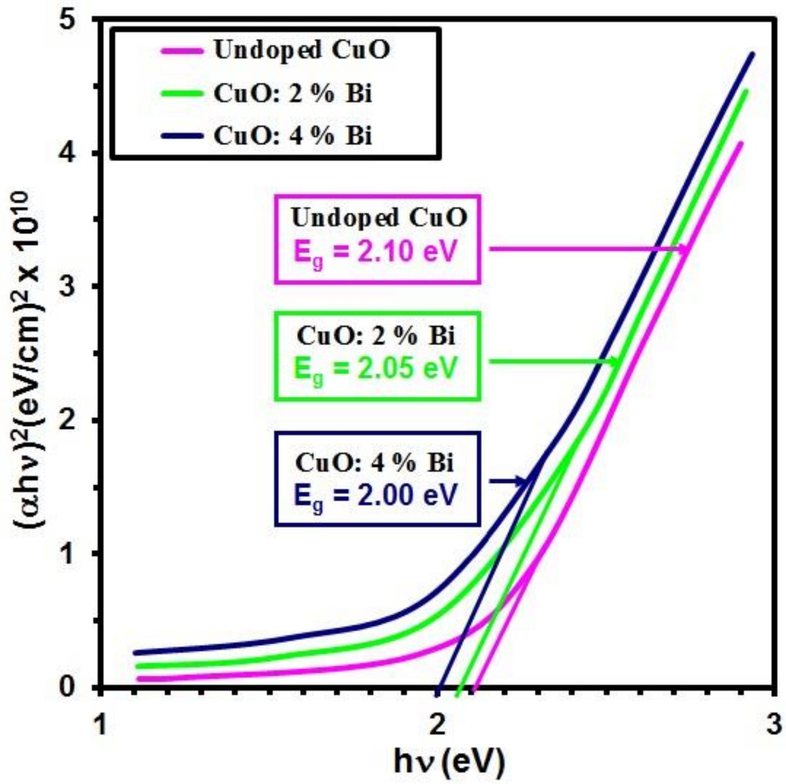

Fig.6: $(\alpha h\nu)^2$ versus $h\nu$ of intended films.

Extinction coefficient (k) has been calculated by the formula [43-46]:

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

Fig.7 shows k versus wavelength. There is an increment in k via increment of Bismuth.
The refractive index (n) was obtained by eq. 6 [47-51]:

$$n = \left(\frac{1+R}{1-R}\right) + \sqrt{\frac{4R}{(1-R)^2} - k^2} \qquad (6)$$

The difference of n versus wavelength is illustrated in figure (8) . n decreases with the increase in Bismuth i concentration.

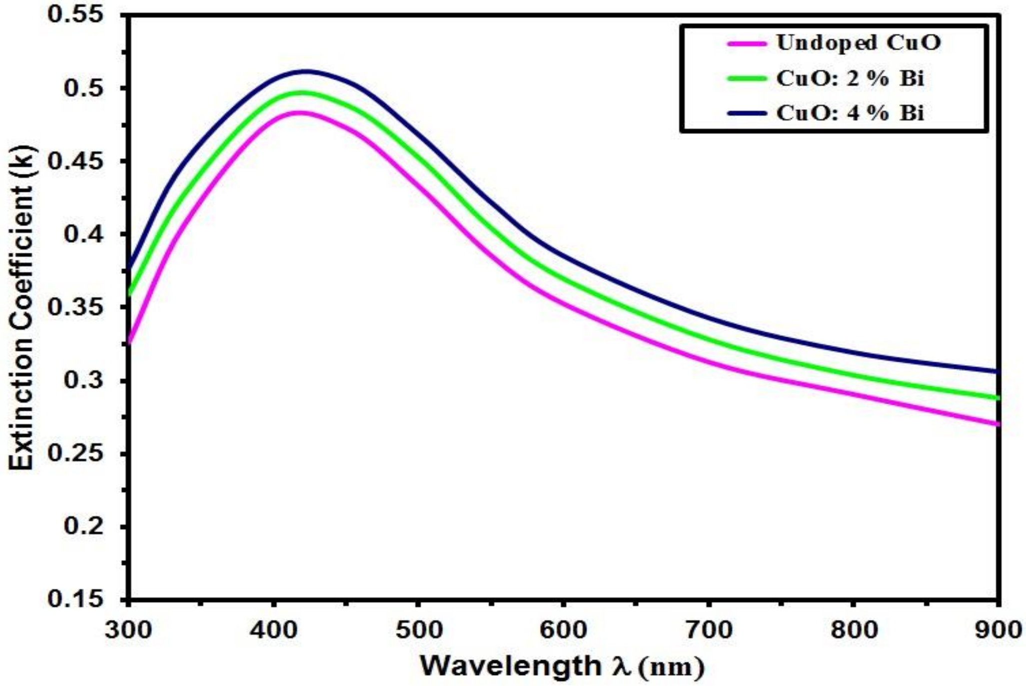

Fig.7: k of deposited films.

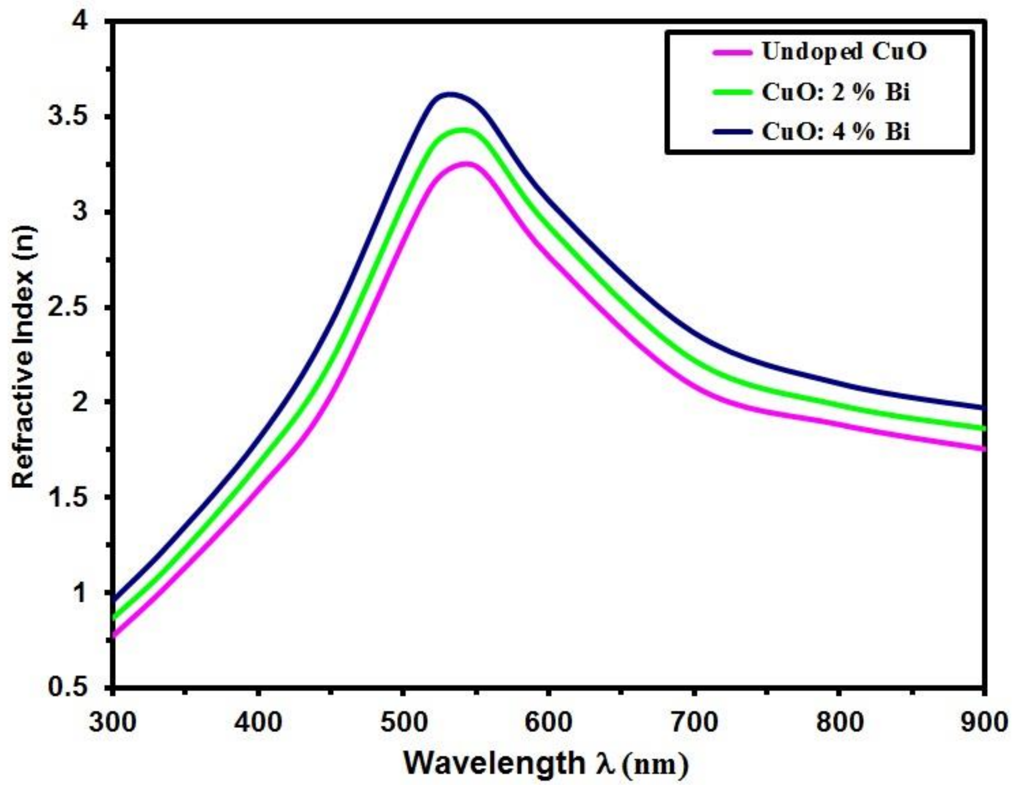

Fig. 8. n of deposited films

Conclusion

In this paper, Undoped CuO and CuO: % Bi thin films have thicknesses 330 ±10 nm were deposited CSP technique. XRD approves that these films were polycrystalline monoclinic structure with a maximum peak at (200) resulting of the Bi, The Grain size for Undoped

CuO particle is about (15.28-17.88) nm with CuO: 4% Bi, where strain increased from 2.26 to 1.98. AFM images display that average particle size were seen in the area of 65.50 nm to 52.31 nm with undoped CuO and CuO: Bi with 2% and 4% concentrations respectively, The transmittance is decreased with increasing Bi -doping. It is found that α , *k* and n are increased with increasing Bi-doping, E_g of undoped CuO film have the highest (2.1eV) and subjected to decrease visa doping.

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