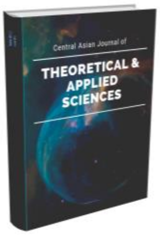

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Article

Analysing the Color Properties of Sol-gel Prepared Copper Oxide Thin Films

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Abstract: This research presented a study of the effect of annealing time at temperatures400 and 500°C on the Color properties of copper oxide films prepared using the sol-gel technique, and tests were conducted on them XRD, UV-visible and hall effect. .X-ray diffraction patterns showed the CuO, and the material was well crystalline. Visual tests showed that the transmittance was 85% at 500° C in the visible region. Color values were observed using the CIELAB colorimetric method, and two important color characteristics were measured: the color angle, which ranged between (25.54°-27.29°) and Chrome which was between (72.09 -74.15). The results of electrical measurements confirmed that the conductivity of the thin films increases with increasing annealing temperatures and that all thin films are of the p-type.

Keywords: Sol Gel, XRD, CIELAB, Then Films

1. Introduction

The current era is characterized by the dominance of modern science and technology over all aspects of life, which determine the fate of nations and the progress of peoples. It was necessary to find components that meet the purpose and keep pace with industrial and technological developments and progress. As a result, we notice the emergence of the science of microelectronics, through which we obtained materials with good electronic properties [1]. Oxide semiconductors have attracted great interest from the scientific community due to their interesting photoelectric properties as well as low-temperature deposition capabilities and low cost [2]. Metal oxides such as zinc oxide, titanium oxide, copper oxide, etc. are potential and candidate semiconductors and are widely used in photovoltaic devices such as photo anode, transparent electrodes, electron transport layer and absorption layer [3]. The most prominent of these oxides is copper oxide (CuO), which is considered an important semiconductor because it is one of the most prominent copper chemical compounds that is insoluble in water or bases, but rather dissolves in acids [4]. It is also characterized by its monoclinic crystalline structure, and its energy band gap ranges between (1.2-2.1) eV and shows p-type electrical conductivity. It is classified as a good material for many applications in devices because of its chemical and physical properties such as Lithium cells and gas sensors [5]. Thin-film technology has played an important and serious role in the industry, and the development of thin-film technology has begun to increase when the world has an urgent need to manufacture integrated circuits [6]. There are different methods that are used in the production of thin films with varying degrees of quality, speed, and cost. These methods are divided into two parts: physical methods and

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chemical methods. These methods include: chemical solution deposition [7], steam spray pyrolysis [8], DC magnetron spraying [9], spin coating [10], pulsed laser deposition, and electrodeposition [11]. Among the most important methods for preparing thin films is the spin coating method, which is characterized by ease of preparation, low cost, and the possibility of obtaining a homogeneous film with a large area. These methods also have the potential to control the final properties of the prepared thin film, depending on the concentration and type of material and annealing temperature [12]. In this study, we produced copper oxide thin films at various annealing temperatures, studied their color values, and compared the optical effects of these films on those values using the CIE LAB system, which contains three values (a*, b*, and l*). In addition, the value of color angle hab* and the amount of color Cab* and knowing the effect of annealing temperatures on these values. The researcher [13]discussed the effect of both temperature and molar concentration of CuO films prepared by spin coating method. The best crystallization and highest absorption were found for the annealed films at 550°C. XRD analysis also indicated that all films were polycrystalline in nature, with a monoclinic structure. The increase in molar concentration led to an increase in film thickness and a decrease in the energy gap from (2.44-3.68) eV at concentrations of (0.1-0.5) M, while increasing the annealing temperature from (350-550)°C led to a decrease in the energy gap from (3.5-2.44) and an increase in the thickness of the films from (136-252) nm.[14], studied the effect of annealing on the electro physical properties of copper oxide films, prepared by spin coating method, at annealing temperatures (400-500) °C. The results showed that the resistance of the films is more stable at 500°C than 400°C. The thickness of the films was 440nm, and the resistance ranged from 350 Ω.cm in un annealed samples to 20 Ω.cm in annealed samples. The films also showed a permeability estimated at about 45%. The researcher.[15] studied the effect of rotation speed on the physical properties of CuO films, prepared by the spin coating method and at rotation speeds of (2000-4000) rpm. The X-ray diffraction results showed the appearance of the CuO phase, and that the highest peak was 35.4° with the prevailing direction of the (002) plane, and that the crystal size increased at a rotation speed of (3000 rpm), and that the highest absorbance that could be obtained was at (3000 rpm). The researcher.[16]studied the color characteristics of pure ZnO films, ZnO/SeO2, annealed at different temperatures, and prepared by spin coating method, and the results showed that the optical transmittance was 85% in the visible region, where the color values and color coordinates were calculated according to the CIE1931, CIELAB system for the transmittance spectrum of the prepared samples with increasing annealing temperature, and it was noted that the best color removal result was achieved at 7.5% Se at 400°C, 600°C, and 10% Se at 500°C, and the best brightness result appeared at 2.5% Se at 600°C, and 7.5% Se at 400°C. In the research of [17], sol gel was employed to generate thin tin oxide films, which were subsequently spin-coated onto glass substrates. The structural, optical, and color properties of tin oxide films produced at different annealing temperatures were investigated. The XRD findings indicated that the films had a tetrahedral structure, with direction (110), (101), (211), and (301) being the prominent one. As the annealing temperature rises, the surface roughness, grain size, and brightness of SnO2 films increase, whereas transmittance, dominant wavelength, and purity decrease. The greatest transmittance was reached during annealing at a temperature of 200°C.

2. Materials and Methods

A. Materials used to prepare the Sol- Gel

1. Copper acetate (CH3.COO) CuH2O, which is a chemical compound and is in the form of green crystals with a molecular weight of (199.65 g/mol) and is produced by the Indian company (Thomas Baker).

2. Isopropanol alcohol as a solvent (C3H8O): It is a transparent, colorless liquid with a molecular weight of (90.10g/mol) and a purity of (99.5%).

3. Diethanolamine [CH2(OH)CH2]2NH as a stabilizer: it is a colorless, transparent, viscous liquid with a molecular weight of (105.14g/mol) produced by (Thomas Baker) Indian company.

B. Preparation of CuO thin films solution

A solution of thin copper oxide films was prepared using the materials (copper acetate, isopropanol alcohol, and diethanolamine). The weight of the aforementioned materials, which were dissolved in (50ml) of isopropanol, was calculated using the following relationship:

$$M = \frac{W_t}{M_{wt}} \times \frac{1000}{v} \qquad (1)$$

Wheres:

M = molar concentration (mol/l). Mwt=molecular weight of the substance (g/mol).

Wt = weight to be melted (g). V=Volume of the solution in which it was dissolved (ml).

0.2 g of copper acetate was dissolved in 50 ml of isopropanol. The mixture was stirred in a magnetic mixer for 20 minutes. The temperature of the solution was gradually increased to 60 °C and maintained at that temperature. Under continuous stirring, 1 ml of diethanolamine was added dropwise to the solution until the solution was uniform and showed a transparent dark blue color. The solution was removed from the magnetic mixer and stored in a sealed glass bottle for one day. After one day, the solution was filtered with filter paper and deposited on a glass substrate. The glass base was washed with water and detergent, and then with distilled water. The glass base was placed in acetone and ethanol for ten minutes. The base was dried in an oven at 250°C for ten minutes. A glass substrate was placed in the centre of the spin coater and the solution was dripped onto the substrate. The device was spun at 3000rpm for 30 minutes and the deposited film was then dried at 250°C. This process was repeated ten times. Finally, annealing was performed at a temperature between (400,500) °C. The morphology and optical characteristics were examined using AFM, FESEM and UV spectrophotometer images (300-1100 nm).

C. Characterization

The X-ray diffraction technique was, adopted to determine the nature of the crystalline structure of copper oxide thin films. A device of the type (XRD-6000) equipped by the Japanese company (Shimadzu) was used. A UV-Visible Spectro photometer with a wavelength ranging from (190-1100) nm was used, type (DU-8200) made in China and located in the research laboratory at the College of Education for Girls - University of Kirkuk to measure transmittance spectrum. The Hall effect of copper oxide films prepared at different temperatures was measured using an electromagnetic device model 6480 from PHYWE, Germany. The device connected to the computer provides us with information about the semiconductor type, resistance, conductivity, carrier mobility and concentration, and the Hall coefficient.

3. Results

A. Structure

a. Properties of X-ray Diffraction test result

X-ray diffraction patterns confirmed the formation of CuO films, which were detected by JCPDSstandardcardnumber00-045-0937. They have a polycrystalline structure, and Figure 1 shows the X-ray diffraction diagram for these films, where the peaks appeared (32.47,35.48,38.60,48.70,53.29,58.34,61.52,66.35,68.07,72.49,75.14) at levels($1\bar{1}0$,002 ,200, $\bar{2}02$,020,202, $1\bar{1}3$, $(3)\bar{1}1$, $2\bar{2}0$,311,004) and with the dominant direction of growth(002). these results are consistent with the results of researchers [13], [15], [18].

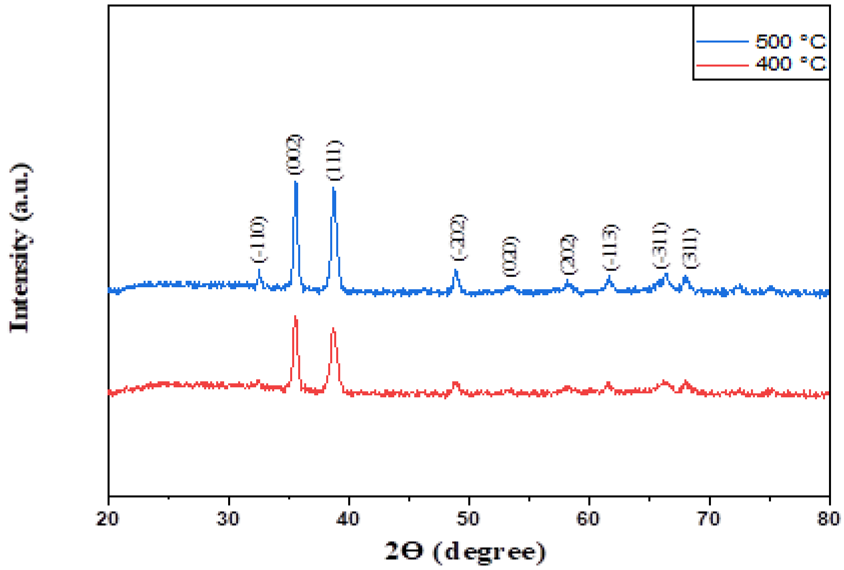

Figure 1. XRD diffraction patterns of CuO thin films prepared by sol gel spin coating method.

Calculating the average grain size (Dav) of the films made using the Scherer method [19] using equation (2), it was discovered that its value increases with increasing annealing temperature. This is because some grain boundaries are cancelled, causing the grains to coalesce into larger crystals. It also cancels out defects that remain after the growth process, causing the crystalline grains to rearrange themselves as they absorb sufficient energy for growth and arrangement within the network.

$$D = \frac{k\lambda}{\beta \cos\theta} \quad (2)$$

Where: k is a constant of 0.9, λ is the wavelength of X-ray radiation, θ is Bragg's angle, β is Full- Width Half Maximum (FWHM) in radian.

The density of dislocations was calculated from equation (3). The density of dislocations decreased with increasing annealing temperature, as shown in Table (1).

$$\delta = \frac{1}{D_{av}^2} \quad (3)$$

Table 1. Estimation some of the structural parameter values for CuO films.

(A-T)	Dav (nm)	δ×103	ε
400 (oC)	29.73	1.287	0.001225
500 (oC)	33.17	1.130	0.001022

B. Optical Properties

a. Transmittance Test

We notice that the transmittance increases with increasing wavelength and increasing annealing temperature. From transmittance measurements, color values can be found according to a CIE LAB system.

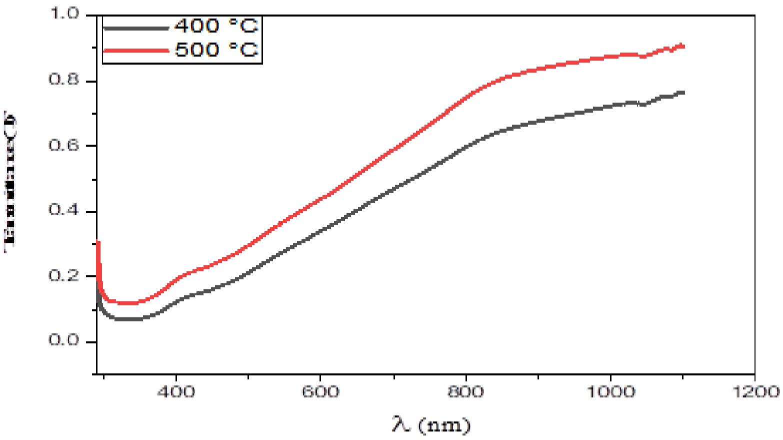

Figure 2. shows the transmittance as a function of wavelength for the prepared copper oxide films.

C. Measuring color values

a. CIE Color System Value

It is possible to perform some mathematical conversions on the transmittance or absorbance values after obtaining the transmittance spectrum curve for copper oxide using a UV visible spectrum device, through which we obtain a table of T_λ values for light with equal gradations of wavelengths for the range (380-770) nm. Tristimulus values (X_T, Y_T, Z_T) and color coordinates (x, y, z) of the transmitting light can be calculated by performing sequential steps on the transmittance values T_λ ,which were determined by CIE [20], [21].

$$X_T = \sum P_\lambda\ T_\lambda\ X_\lambda \tag{4}$$

$$Y_T = \sum P_\lambda\ T_\lambda\ Y_\lambda \tag{5}$$

$$Z_T = \sum P_\lambda\ T_\lambda\ Z_\lambda \tag{6}$$

whereas: $P_\lambda$is the power distribution curve for the light source used, (X_λ〗 _ ‹ Y_λ‹Z_λ) Values of distribution coefficients for light sources. The specimen's tristimulus value of X_T, Y_T, and Z_T is used to calculate the color coordinates values for the CIE system.

$$x = \frac{X_T}{X_T+Y_T+Z_T} \tag{7}$$

$$y = \frac{Y_T}{X_T+Y_T+Z_T} \tag{8}$$

$$z = \frac{Z_T}{X_T+Y_T+Z_T} \tag{9}$$

b. Color values according to CIE LAB system

The color values for this system were calculated through non-linear transformations of the CIE system, as it consists of three color values (a*, b*, and l*), and from these values we were able to obtain two other color values, which are the color angle $h_{ab}^*$and the color amount C_{ab}^*shown in Table 2 [21].

$$l^* = 116\left(\frac{Y}{Y_n}\right)^{\frac{1}{3}} - 16 \tag{10}$$

$$a^* = 500\left[\left(\frac{X}{X_n}\right)^{\frac{1}{3}} - \left(\frac{Y}{Y_n}\right)^{\frac{1}{3}}\right] \quad (11)$$

$$b^* = 200\left[\left(\frac{Y}{Y_n}\right)^{\frac{1}{3}} - \left(\frac{Z}{Z_n}\right)^{\frac{1}{3}}\right] \quad (12)$$

$$h_{ab} = tan^{-1}\left(\frac{b^*}{a^*}\right) \quad (13)$$

$$C^*_{ab} = (a^{*2} + b^{*2})^{\frac{1}{2}} \quad (12)$$

Table 2. Colorimetric values of the CIE LAB system for copper oxide films.

T(°C)	l^*	a^*	b^*	C^*_{ab}	h_{ab}
400	63.04	64.06	33.08	72.09	27.29
500	70.42	66.9	31.98	74.15	25.54

From Looking at Figure 4, it becomes clear to us that the color angle ranges between (25.54° - 27.29°), while the value of the color amount ranges between (72 – 74.15), The best color removal value was at 400°C, as the Chroma value decreased.

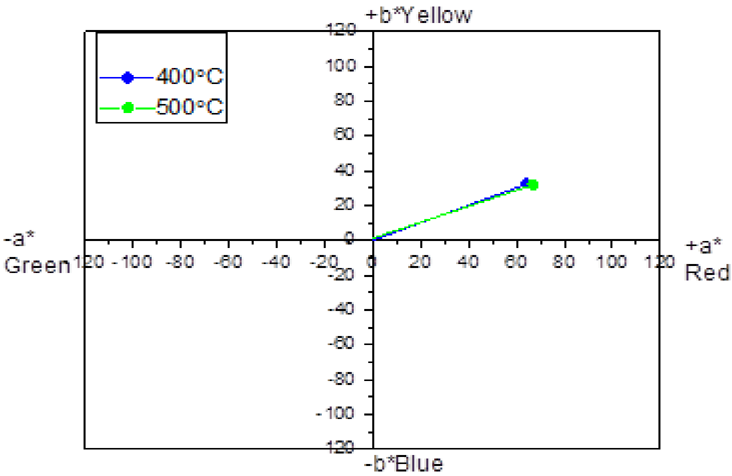

Figure 4. shows the color angle of the CIE LAB system for the prepared copper oxide films.

D. Hall effect

To determine the electrical properties, the Hall effect was measured at room temperature, and the conductivity, specific resistance, concentration, and majority carrier type were determined at different annealing temperatures, as shown in the table. Table (3) shows the results of the Hall measurements, and Figure (5) shows the variation of conductivity with temperature.

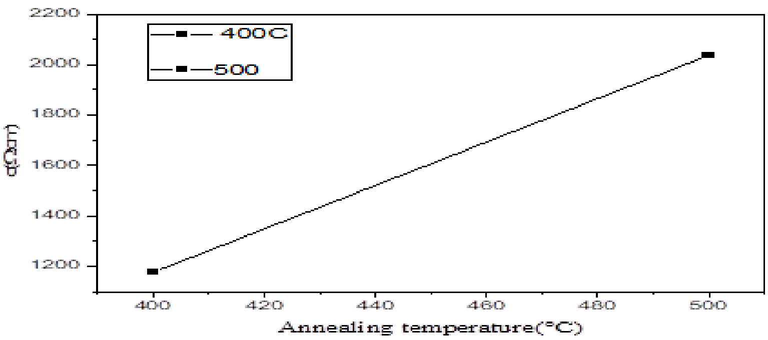

Figure 5. Hall effect measurement results.

Table 3. Shows the results of Hall effect measurements for CuO films.

Annealing Temperatures(°C)	RH (cm3 /C)	n (cm-3)	ϱ(Ω.cm)	σ(Ω.cm)-1
400	0.000187477	3.33375×10^{22}	0.000849712	1176.86986
500	0.00010827	5.77259×10^{22}	0.00049072	2037.82075

4. Discussion

The reason for the decrease in the density of dislocations is due to the increase in the average grain size, as the relationship between them is inverse. This indicates a decrease in crystal defects as a result of annealing. The hall effect was found that the sign of the Hall coefficient was positive and the majority carriers were holes, which means that the produced films were all p-type; this is consistent with the results of researchers. It was found that the conductivity increased with increasing annealing temperature; this was due to the crystallization process of the prepared films, which was confirmed by the XRD results.

5. Conclusion

Thin films of copper oxide were prepared on glass substrates using the sol-gel spin coating method, and the effect of annealing temperature on the textural, optical properties and color values was studied. XRD analysis CuO phases, and that increasing the annealing temperature led to increased crystallinity with increased crystallite size and decreased dislocation density. The highest transmittance of 85% was obtained at annealing temperature of 500 °C in the vision region. The color values showed that the best color removal and the lowest value of color amount were at 400°C. Electrical conductivity increases with increasing annealing temperature.

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