

## **Color Characterizations of Pure ZnO and ZnO/ SeO<sub>2</sub> Thin Films Annealed at Different Temperature**

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### **Abstract**

Pure Zinc oxide and ZnO/SeO<sub>2</sub> oxide thin films were prepared successfully by sol gel method and annealed at different temperature under ambient condition. These films were characterized by means of XRD, AFM, and UV-visible. XRD patterns clearly showed the presence of crystalline ZnO/SeO<sub>2</sub> particles, the ZnO/SeO<sub>2</sub> film showed a good crystallinity like pure ZnO film, Optical transmittance spectra of films showed high transparency (>87%) in visible region. The color coordinate and tristimulus value of transmittance spectral showed that the best decolorization result was achieved at 7.5 at.% Se at 400°C, 600°C and 10 at.% Se at 500°C , for the best brightness result appeared at two point with 2.5 at.% Se at 600°C and 7.5 at.% Se at 400°C. AFM studies reveal that rms roughness of the thin films increased with the increasing of Se concentrations. Also, the surface roughness increased with the increasing of the annealed temperature.

**Keywords:** ZnO; SeO<sub>2</sub>; Sol-gel; decolorization; CIE; CIE LAB.

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## التشخيص اللوني للاغشية الرقيقة من اوكسيد ZnO النقي و ZnO/ SeO<sub>2</sub>

### و الملدنة عند درجات حرارة مختلفة

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#### الملخص

حضرت الاغشية الرقيقة لاوكسيد الخارصين النقي و ZnO/SeO<sub>2</sub> بطريقة السائل الهلامي و لدنت عند درجات حرارة مختلفة، ثم اجريت عليها فحوصات XRD و AFM و UV-visible للاغشية الملدنة. وقد اظهرت انماط حيود الاشعة السينية ان المادة متبلورة بصورة جيدة لكلا التركيبين النقي و المشوب. كما ان النفاذية كانت عالية (>87%) في المنطقة المرئية. بالنسبة للإحداثيات اللونية والقيم اللونية لطيف النفاذية اظهرت حصول افضل ازالة للون عند تركيز السلينيوم 7.5 at.% و عند درجة حرارة 400°C, 600°C وكذلك عند تركيز 10 at.% عند 500°C . و ظهرت افضل اضاءة عند تركيز السلينيوم 2.5 at و عند درجة حرارة 600°C و كذلك عند تركيز 7.5 at.% و عند درجة حرارة 400°C. بالنسبة لفحوصات AFM اظهرت ان معدل مربع خشونة السطح للاغشية الرقيقة يزداد مع زيادة تركيز عنصر السلينيوم وكذلك خشونة السطح تزداد مع زيادة درجة حرارة التلدين.

الكلمات الدالة: ZnO، SeO<sub>2</sub>؛ السائل الهلامي؛ ازالة اللون؛ CIE؛ CIE LAB.

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## 1. Introduction:

In the last decade ZnO has received good attention in fabrication of high quality thin films, by employing different methods such as magnetron sputtering [1], electrodeposition [2], pulsed laser deposition [3], chemical vapor deposition [4], spray pyrolysis [5], sol-gel process etc. [6]. Sol-gel preparation method represents the most attractive method, it provides a simple and an inexpensive of a large homogeneous area of a thin films with excellent compositional control, and uniform film thickness as well as lower crystallization temperature. The main factors which affecting on the microstructure and properties of the sol-gel films are; thickness, aging time and annealing treatment [7]. The ZnO thin films represent as a native n-type semiconductor ( $E_g \approx 3.3$  eV) and a large excitation binding energy of 60 MeV semiconductor, can gives high transparency and high electrical conductivity in the same time [8, 9].

For using a pure ZnO there are a few limitations for the integrated optical devices as they often require a much wider bandgap. Hence, the tuning of ZnO band gap becomes significant. ZnO band gap can be tailored by alloying with group II elements e.g. Ca, Be, Cd, Sr, and Mg [10]. The electrical properties of ZnO can be improved by using dopants, principally the group III elements (e.g., B, Al, Ga, and In) to substitute for Zinc atoms [11]. and alloying ZnO with group V elements e.g. Bi [12]. (ZnO:Al) thin-film has the equivalent electrical properties, with a lot of advantages, such as high electrical properties stability against hydrogen plasma, effective light trapping action, to improve the a-Si:H solar cell performance as a front electrode [13].

ZnO as a transparent conductive oxide (TCOs), has been widely studied because it is a non-toxic, thermally and chemically stable semiconductor with a low cost, therefore, ZnO thin films are a promising alternative to the commonly used ITO because of the high cost and scarcity of indium [14].

ZnO has received good attention as a photocatalyst with respect to the degradations of various environmental pollutants [15]. In the present work, chromatic properties and chromatic coordinates of ZnO have been studied so that we can obtain the best optical window for different applications, especially in solar cells, as we know that one of the most important application of ZnO thin films were used as a window for the solar cell because of

its wide band gap and its high transparency of visible light, We need to manufacture transparent thin colorless films, also transparent conductive oxides (TCOs) characterized by its capable of transporting electrical charge and transmitting visible photon. We have attempted to study the color value of pure ZnO thin film and ZnO/SeO<sub>2</sub> thin films, in order to compare the optical effects of these films on the color values of transmittance spectral intensity to assuring if we have achieved a suitable TCO. five color value were used in this study, purity, dominant wave length, brightness, Chroma and hue angle in order to recognize the effect of SeO<sub>2</sub> additive and the annealing temperature on these color values.

For display window to complete the removal of the color from the sample, it is preferable to add additives whose ions have a absorption spectrum so that it gives a complementary color to the color of the sample so that we get a transparent colorless thin films, This process is called physical decolorization method.

## **2. Experimental details:**

### **2.1 Materials Used to Prepare the Sol-Gel:**

The main materials used in this work are:

- 1- Zinc acetate, Zn (CH<sub>3</sub>COO)<sub>2</sub>.H<sub>2</sub>O, 99+%, powder, Scharlau, Spain.
- 2- Diethanolamine (DEA), (C<sub>4</sub>H<sub>11</sub>NO<sub>2</sub>), 99.8+ %, liquid, Scharlau, Spain.
- 3- Selenium dioxide SeO<sub>2</sub>, 99+%, powder, Alpha chemika, India.
- 4- Ethanol, C<sub>2</sub>H<sub>6</sub>O, 99.9%, liquid, Scharlau, Spain.
- 5- Deionized water.

### **2.2 The Substrate:**

The substrate represents an important part of thin films system, but it is the source of most stress stands by the film and may be effects on many properties of the film. So that to minimize a thermal stress in the film, glass, quartzes and ceramic substrate are commonly used for polycrystalline films. In this work a microscopic glass slides with dimension (76.2×25.4×1) mm<sup>3</sup> are used as a substrate.

### 2.3 The Sol-Gel Preparation:

ZnO/SeO<sub>2</sub> solution was prepared using Zinc acetate dehydrate and selenium dioxide SeO<sub>2</sub> were dissolved in ethanol, The 0.75 molarity ZnO precursor was prepared by dissolving Zinc acetate dehydrate {Zn (CH<sub>3</sub>COO)<sub>2</sub>·2H<sub>2</sub>O} in ethanol, solution and stirring it at 60°C for 5 min after that add Diethanolamine; DEA (C<sub>4</sub>H<sub>11</sub>NO<sub>2</sub>) at a rate of one drop/sec, DEA was chosen as a stabilizer, the molar ratio of Zn:DEA was kept as 1.0, at that time 0.3M of selenium dioxide dissolved in ethanol was added as a doping source in the starting solution. the mixture solution stirred at 60°C for 1.0 h to obtain clear light solution. The weight of each component is found according to equation below. The doping level was varied by changing the atomic ratio [Se/Zn] in the solution from 0 to 10 at%. A clear solution was obtained during the process of stirring for 1h at 60°C. the gel solution was statically aged for 24h at room temperature, after that the gel solution was filtering to remove the sedimentation particle.

$$M = \frac{W}{M_W} \times \frac{1000}{V} \quad (1)$$

Where M is the molarities of the matter, M<sub>W</sub> is the molecular weight of the matter, V is the volume of the prepared solution.

For (ZnO)<sub>1-x</sub> (SeO<sub>2</sub>)<sub>x</sub> thin films, x = (0, 2.5, 5, 7.5, and 10 at%) of SeO<sub>2</sub> in ZnO solution.

### 2.4 Sample Preparation:

Thin films of oxides of ZnO and (ZnO)<sub>1-x</sub>/(SeO<sub>2</sub>)<sub>x</sub> with = (0, 2.5, 5, 7.5, and 10 at%) of SeO<sub>2</sub>, were deposited on glass substrates (76.2×25.4×1 mm) using sol-gel spin coating technique. The glass substrates were cleaned with acetone and then in ethanol for 20 min. The slides were removed from the ethanol and dried at room temperature.

After preparing the sol-gels we deposit it on the substrate using a spin coater with 3000 rps for 30 sec. After deposition, the films were dried at 300°C for 10 min to evaporate the solvent and remove organic residuals. Then, the films were finally post annealing at three fixed temperatures; 400, 500, and 600°C.

## 2.5 Characterization:

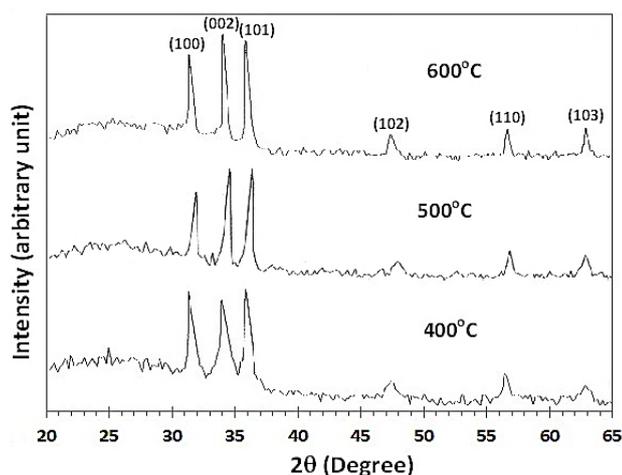
X-Ray Diffractometre Type Shimadzu XRD-6000(Cu-K $\alpha$ ), were used for x-ray diffraction records, through the courtesy of Iraqi Ministry of science and technology Lab. The atomic force microscopy (AFM) measurements were done using model (AA3000 SPM) scanning probe microscope through the courtesy of college of science, university of Baghdad. The spectrophotometric data were recorded using double-beam Shimadzu spectrophotometer. The thickness of the films was measured by using optical measurements.

## 3. Results and Discussion:

### 3.1 Structure and Morphology:

#### 3.1.1 X-ray Diffraction Test:

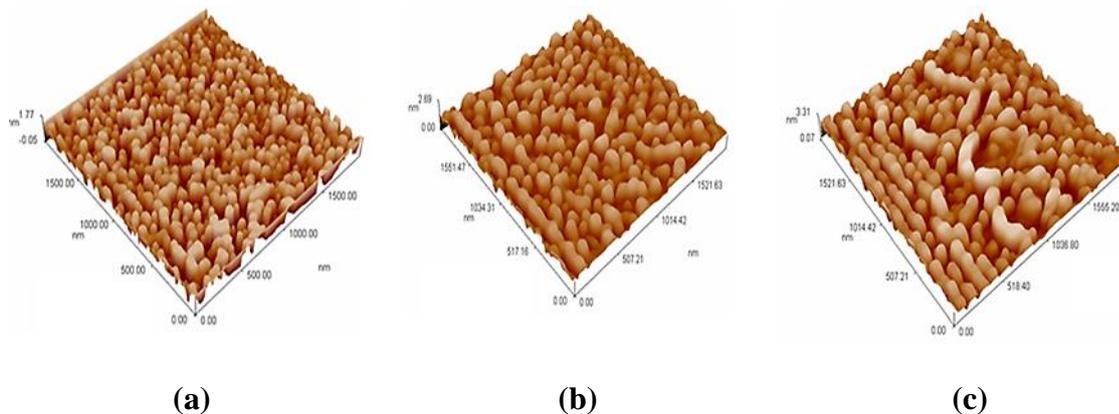
Fig. 1 shows The XRD patterns of undoped ZnO thin films annealed at three different temperatures (400, 500 and 600°C). From comparison with the standard XRD pattern of ZnO crystal (JCPDS 036-1451), reveal that these polycrystalline ZnO thin films have a hexagonal wurtzite structure [10], [16]. Films show good crystallinity with (100), (002), (101), (102), (110) and (103) peaks, the preferred orientation is observed with (002) plane at 34.26°. The growth of ZnO thin films with (002) orientation, reflecting that the highest density of Zn atoms is found along the (002) plane. The diffraction peak intensities revealing the crystallinity of ZnO thin films, were continuously improved with increasing annealing temperature, in the used temperature range.



**Fig.1:** XRD diffraction patterns of undoped ZnO thin films prepared by sol gel spin coating method at different annealing temperature.

### 3.1.2 AFM Test:

3D AFM images as shown in Fig. 2 reveal the influence of post-growth annealing on the ZnO thin films surface morphology. It was found that all samples are free from cracks and homogeneous. When careful examination of the AFM images, it observed that the grain sizes become larger with increasing annealing temperature. At high temperatures, the atoms will possess enough diffusion activation energy to propagate and occupy a favorable location in the crystal lattice and finally grain with lower surface energy has become the largest. In Table 1 values of surface roughness (RMS) are listed at annealing temperature of 400, 500 and 600°C, which are showed the increase of the roughness of thin films surfaces with increasing annealing temperature, as a result of grain growth. when a large grain growth the surface roughness increased rapidly. Also AFM analysis showed that the mean square root roughness of ZnO thin films increases with increasing Se concentration in the annealing temperature range used.



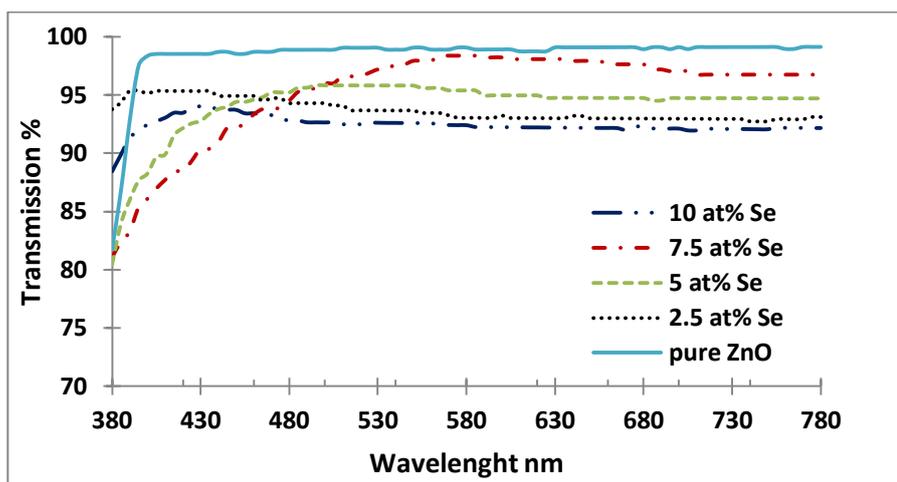
**Fig. 2:** 3D AFM image of, **a)** undoped ZnO deposited films annealed at 600°C,  
**b)** ZnO:Se 5 at% deposited films annealed at 600°C,  
**c)** ZnO:Se 10 at% deposited films annealed at 600°C.

**Table 1:** The root mean square (RMS) roughness of the ZnO thin film post annealed at different temperature.

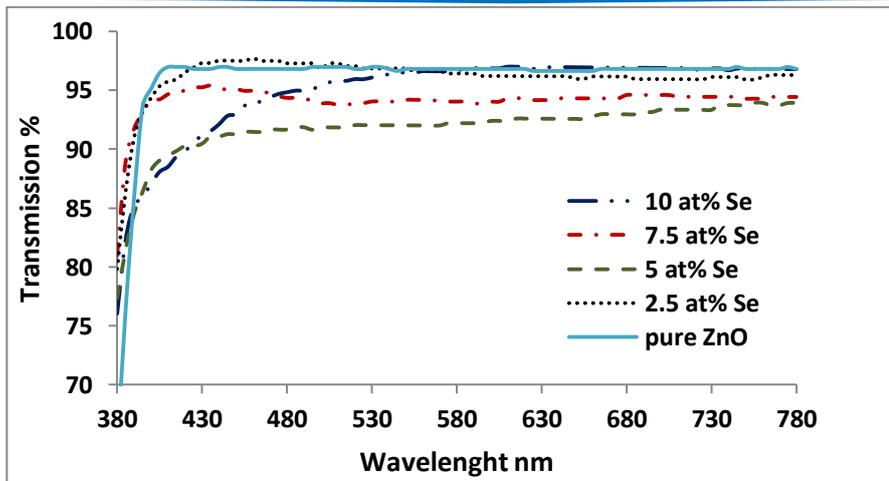
Samples	T = 400°C	T = 500°C	T = 600°C
	RMS (nm)	RMS (nm)	RMS (nm)
undoped ZnO	0.373	0.42	0.486
2.5 at %Se	0.41	0.433	0.511
5 at %Se	0.435	0.466	0.701
7.5 at %Se	0.554	0.612	0.833
10 at %Se	0.764	0.84	1.11

### 3.2 Optical Properties:

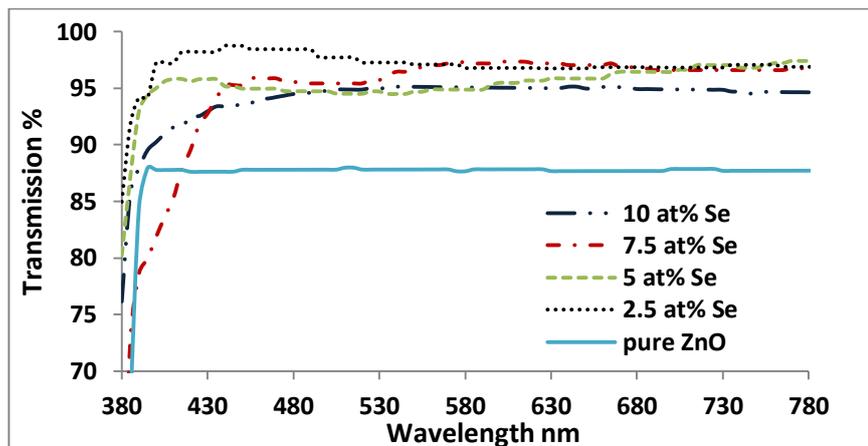
#### 3.2.1 Transmittance Test:



**Fig. 3:** The transmission spectra in visible wavelength range between 380 and 780 nm of ZnO/SeO<sub>2</sub> thin films with different at.% of Se annealed at 400°C



**Fig. 4:** The transmission spectra in visible wavelength range between 380 and 780 nm of ZnO/SeO<sub>2</sub> thin films with different at.% of Se annealed at 500°C.

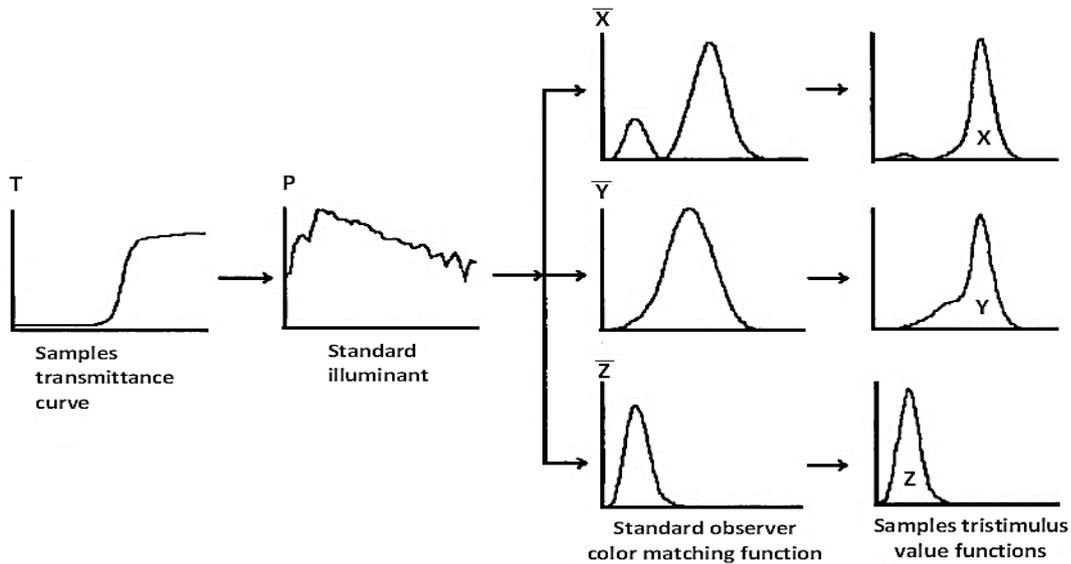


**Fig. 5:** The transmission spectra in visible wavelength range between 380 and 780 nm of ZnO/SeO<sub>2</sub> thin films with different at.% of Se annealed at 600°C.

The optical transmittance spectra in the Figures 3, 4 and 5 shows a good transmittance (between 87% and 95%) in the visible region. Figures shows that the transmittance decreased rapidly as the incident light wavelength shifted to short wavelength. This phenomenon is called the optical absorption edge ( $\lambda = 385\text{nm}$ ). From transmittance measurements the color values measurement could also be estimated by employing CIE and CIE LAB system for this purpose.

### 3.3 Color Values Measurements:

#### 3.3.1 CIE Color System Values:



**Fig. 6:** Calculation of the CIE tristimulus value by combining a samples standard illuminant with a transmittance curve and with functions of color matching of a standard observer. The enclosed area by the tristimulus value function curves would be the numerical value of tristimulus values. for present work 5 nm intervals are used [17].

The following equations are used to calculate the tristimulus value, with wave length range from 380 to 780 nm, [17],

$$X = k \sum P(\lambda) \bar{x}(\lambda) T(\lambda) \quad (2)$$

$$Y = k \sum P(\lambda) \bar{y}(\lambda) T(\lambda) \quad (3)$$

$$Z = k \sum P(\lambda) \bar{z}(\lambda) T(\lambda) \quad (4)$$

$$k = \frac{100}{\sum P(\lambda) \bar{y}(\lambda)} \quad (5)$$

Where  $P(\lambda)$  is the value of the spectral power distribution of the illuminant at the wavelength  $\lambda$ ,  $T(\lambda)$  is the transmittance factor of the sample at the wavelength  $\lambda$ , and  $\bar{x}(\lambda)$ ,  $\bar{y}(\lambda)$  and  $\bar{z}(\lambda)$  are the CIE color matching functions for the standard observer at the wavelength  $\lambda$ . The factor  $k$  normalizes the tristimulus value so that  $Y$  will have a value of 100 for the perfect white diffuser, a theoretical material that reflects or transmits 100 percent of the incident light. Fig. 6 shows the procedure of tristimulus values calculation for a transparent or reflecting specimen.

The calculation of the color coordinates values ( $x, y$  and  $z$ ) for CIE system of the specimen are found from its tristimulus value of  $X, Y$  and  $Z$ , [18]

$$x = \frac{X}{X+Y+Z} \quad (6)$$

$$y = \frac{Y}{X+Y+Z} \quad (7)$$

$$z = \frac{Z}{X+Y+Z} \quad (8)$$

The color coordinates values ( $x, y$  and  $z$ ) of CIE color system are used to found another important three color values, dominants wave length, color purity and the brightness. The values are presented in Tables 2, 3 and 4.

**Table 2:** Color coordinate and color values of CIE system of  $(ZnO)_x(SeO)_{1-x}$  thin films annealed at the temperature of 400°C

Brightness	purity	$\lambda_{\text{Dominants}}$	Color coordinates CIE			Samples Annealed At 400°C
			$z$	$y$	$x$	
98.937	11.533	608.8	0.3139	0.3313	0.3546	Undoped ZnO
93.567	12.21	613.5	0.3121	0.3293	0.3584	2.5 at.% Se
95.441	11.416	604.1	0.3142	0.3332	0.3525	5 at.% Se
97.252	8.86	602.4	0.3187	0.3376	0.3435	7.5 at.% Se
92.521	11.931	612.6	0.3127	0.3297	0.3574	10 at.% Se

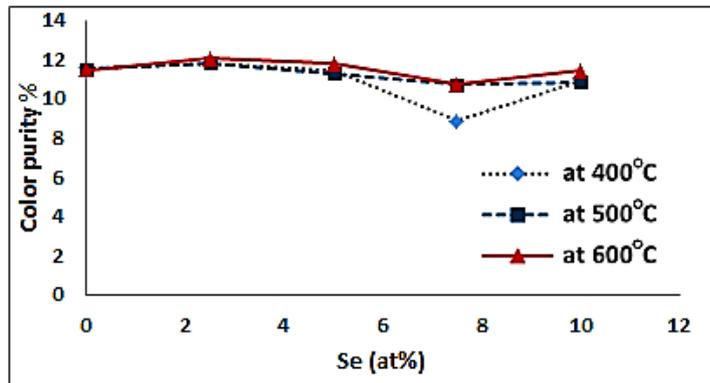
**Table 3:** Color coordinate and color values of CIE system of  $(\text{ZnO})_x(\text{SeO})_{1-x}$  thin films annealed at the temperature of  $500^\circ\text{C}$ .

Brightness	purity	$\lambda_{\text{Dominants}}$	Color coordinates CIE			Samples Annealed At $500^\circ\text{C}$
			z	y	x	
96.831	11.519	609.7	0.3137	0.331	0.3552	Undoped ZnO
96.735	11.826	611	0.3127	0.3304	0.3567	2.5 at.% Se
92.062	11.27	607	0.3148	0.3321	0.3530	5 at.% Se
94.116	11.69	614	0.3132	0.3298	0.3569	7.5 at.% Se
96.199	10.824	597	0.3170	0.3353	0.3475	10 at.% Se

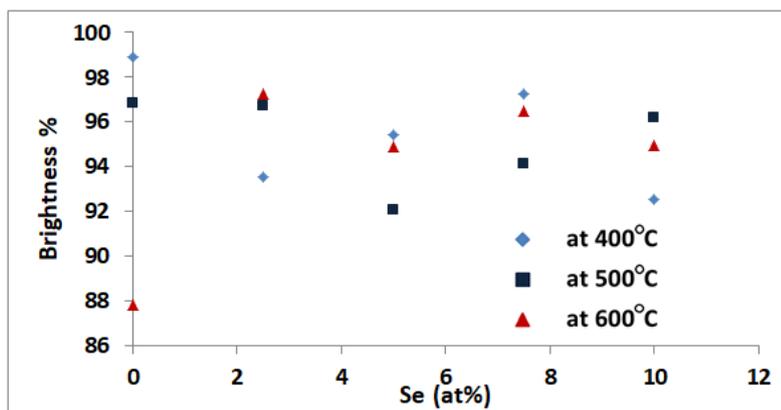
**Table 4:** Color coordinate and color values of CIE system of  $(\text{ZnO})_x(\text{SeO})_{1-x}$  thin films annealed at the temperature of  $600^\circ\text{C}$ .

Brightness	purity	$\lambda_{\text{Dominants}}$	Color coordinates CIE			Samples Annealed At $600^\circ\text{C}$
			z	y	x	
87.809	11.456	609.5	0.3138	0.3311	0.3550	Undoped ZnO
97.268	12.05	612.7	0.3123	0.3296	0.3579	2.5 at.% Se
94.890	11.778	608.6	0.3142	0.3303	0.3554	5 at.% Se
96.465	10.73	604.3	0.3162	0.3334	0.3503	7.5 at.% Se
94.945	11.426	602.2	0.3149	0.3328	0.3521	10 at.% Se

Fig. 7 shows the change of color purity with the concentration of the additive, if the sample has a color purity with less value, it indicates that the sample has low color, so that when purity becomes 0% means that the color is completely removed from the sample. Fig.8 shows the brightness of thin films, when the brightness value reached 100% that means a thin film was a perfect white diffuser.



**Fig. 7:** Color purity of ZnO/SeO<sub>2</sub> thin films annealed at (400,500 and 600°C) with different Se (at%).



**Fig. 8:** Brightness of ZnO/SeO<sub>2</sub> thin films annealed at (400,500 and 600°C) with different Se (at%).

### 3.3.2 CIE LAB Color System Values:

CIE LAB system with a uniform color spaces system is found by a nonlinear transformations to the CIE system, have three color value a\*, b\* and L\*, from these three value we can get two important color value its metric Chroma (C<sub>ab</sub>) and metric hue angle (h<sub>ab</sub>), were listed in the Tables 5, 6 and 7, [18].

$$L^* = 116 \cdot \left(\frac{Y}{Y_n}\right)^{\frac{1}{3}} - 16 \quad (9)$$

$$a^* = 500\left[\left(\frac{X}{X_n}\right)^{\frac{1}{3}} - \left(\frac{Y}{Y_n}\right)^{\frac{1}{3}}\right] \quad (10)$$

$$b^* = 200\left[\left(\frac{Y}{Y_n}\right)^{\frac{1}{3}} - \left(\frac{Z}{Z_n}\right)^{\frac{1}{3}}\right] \quad (11)$$

$$h_{ab} = \tan^{-1}\left(\frac{b^*}{a^*}\right) \quad (12)$$

$$C_{ab} = (a^{*2} + b^{*2})^{\frac{1}{2}} \quad (13)$$

**Table 5:** The color values of CIE LAB system of  $(ZnO)_x(SeO)_{1-x}$  thin films annealed at the temperature of 400°C.

C <sub>ab</sub> Chroma	h <sub>ab</sub> hue angle	Color values of CIE LAB system			Samples Annealed At 400°C
		b*	a*	L*	
3.2998	24.036	1.3440	3.0136	1.2975	Undoped ZnO
3.6948	21.249	1.3390	3.4436	1.2624	2.5 at.% Se
3.0560	27.061	1.3903	2.7215	1.3293	5 at.% Se
2.2189	38.654	1.386	1.7328	1.4056	7.5 at.% Se
3.5984	21.712	1.3312	3.3431	1.2689	10 at.% Se

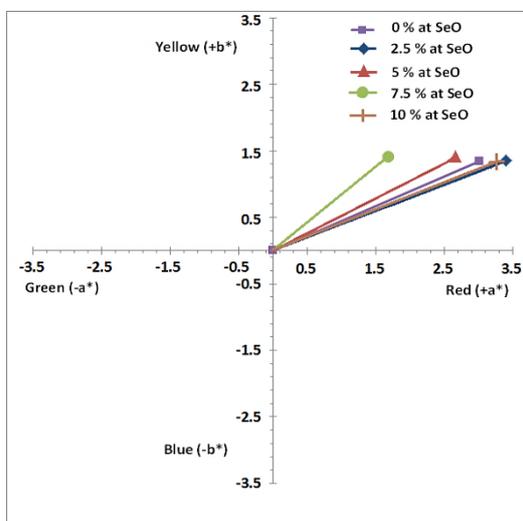
**Table 6:** The color values of CIE LAB system of  $(ZnO)_x(SeO)_{1-x}$  thin films annealed at the temperature of 500°C.

C <sub>ab</sub> Chroma	h <sub>ab</sub> hue angle	Color values of CIE LAB system			Samples Annealed At 500°C
		b*	a*	L*	
3.3685	23.456	1.3408	3.0901	1.2908	Undoped ZnO
3.5087	22.669	1.3523	3.2377	1.2813	2.5 at.% Se
3.1412	25.239	1.3394	2.8413	1.3102	5 at.% Se
3.5528	21.793	1.3190	3.2989	1.2701	7.5 at.% Se
2.5911	31.89	1.3688	2.2000	1.3660	10 at.% Se

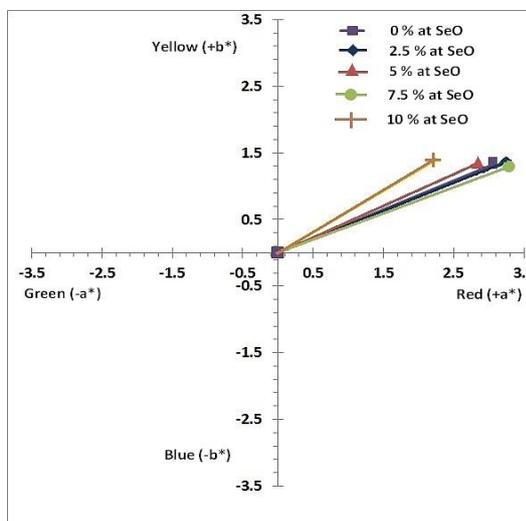
**Table 7:** The color values of CIE LAB system of  $(ZnO)_x(SeO)_{1-x}$  thin films annealed at the temperature of 600°C.

C <sub>ab</sub> Chroma	h <sub>ab</sub> hue angle	Color values of CIE LAB system			Samples Annealed At 600°C
		b*	a*	L*	
3.3439	23.641	1.3411	3.0631	1.2930	Undoped ZnO
3.6392	21.635	1.3417	3.3828	1.2678	2.5 at.% Se
3.4167	22.456	1.3051	3.1576	1.2787	5 at.% Se
2.8771	27.667	1.3359	2.5481	1.3326	7.5 at.% Se
3.0452	26.51	1.3592	2.7250	1.3231	10 at.% Se

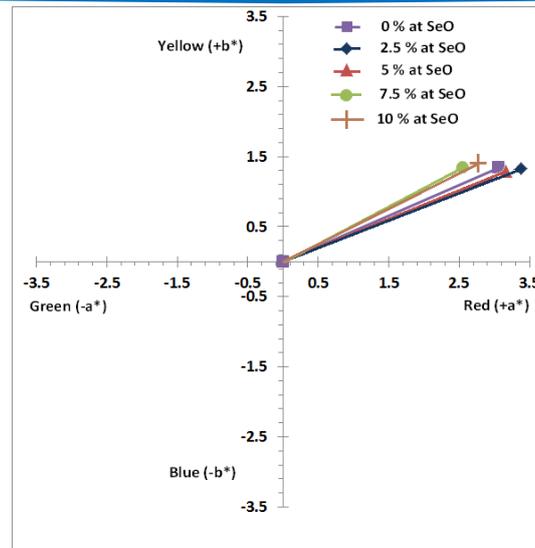
Fig. 9-a, b and c shows that the metric hue angle are very close to red color coordinate and their values are between (21° and 38°), the best decolorization values were obtained with additive of 7.5% SeO as seen the chroma value decreased to the lower value at this additive.



(a)



(b)



(c)

**Fig. 9:** Metric hue angle and metric Chroma for the color of thin films annealed at (a) 400°C, (b) 500°C and (c) 600°C.

#### 4. Conclusions:

The goal of the present study is to controlling on the color values of the ZnO thin films, the colors value is changed due to the additives and annealing temperature and consider as a function of color values. Using the color transformation of CIE and CIE LAB are useful to reveal the effect of additive on the colorization of ZnO Thin films. The XRD patterns of the samples showed that the thin films had a polycrystalline of a hexagonal wurtzite structure. the increase in the grain size improves c-axis orientation, as is indicated by increase in intensity of the (002) peak and decrease in the FWHM with an increase in the annealing temperature. All the films are high transparent. Optical transmittance between 85% and 95% within the visible region has been obtained.

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