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Effect of Annealing Temperatures on the Structural and Optical Properties of CdO Thin Films Prepared by SILAR Method

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Abstract: In this study, cadmium oxide thin films have been prepared onto glass substrate using SILAR method. The films were prepared at 85°C then annealed at 300°, 350° and 400°C for 1 h. The results of XRD reveal that all films have a polycrystalline structure with cubic phase. The UV-visible spectrophotometer analysis revealed that the value of energy band gap decreased with increase in annealing temperature from 2.24, 2-1.9 eV at 300°, 350° and 400°C.

Key words: CdO, SILAR, thin films, XRD, optical properties, temperature

INTRODUCTION

The transparent conductive oxides like Zinc oxide, Tin oxide, Indium oxide and Cadmium Oxide (CdO) have large applications in the field of optoelectronic devices (Thirumoorthi and Prakash, 2016). CdO can be classified as n-type semiconductor and its crystal structure is (FCC) a rock salt with direct band gap about 2.2 eV (Bari and Patil, 2014; Faizullah *et al*., 2013). From many transparent conductive oxides, CdO has received extensive interesting for solar cell application because of it has high transparency and low electrical resistivity in the vis-region of solar spectrum (Yusuf *et al*., 2016). Cadmium oxide films have been effectively used for numerous optoelectronic applications as solar cells, gas sensor (Ibrahim and Kadu, 2017), phototransistors, transparent electrodes, IR detectors photodiodes (Yildirim and Altiokka, 2017). Antireflection coatings and liquid crystal displays (Ezekoye *et al*., 2013). CdO thin films was deposited using many different techniques like chemical vapor deposition, spray pyrolysis deposition (Perumal *et al*., 2012). Pulsed laser deposition, chemical bath deposition (Usharani *et al*., 2015). Radio frequency sputtering, DC magnetron sputtering, sol-gel and reaction SILAR methods (Ibrahim and Kadu, 2017; Sahin *et al*., 2014). In this research, an attempt was conducted to fabricate thin films of CdO using low cost Successive Ionic Layer Adsorption and Reaction (SILAR) technique due to its an alternative and effective technique for preparing CdO thin films with large area coating and without any vacuum system.

MATERIALS AND METHODS

In Successive-Ionic-Layer-Adsorption and Reaction method (SILAR) the glass substrates were cleaned with (H_2SO_4:H_2O, 1:5, v/v) dilute sulfuric acid solution, then, it was ultrasonically cleaned utilizing acetone followed by deionized water for 15 min. The growth bath can be described as the following: $Cd(CHCOO)_2$ dissolved in 60 mL of deionized water. At room temperature the resulting solution stirred for 45 min utilizing the magnetic stirrer in order to get a well-dissolved and transparent solution. The value of Ph of the solution was reached to 12 by adding aqueous ammonia (NH_3). The resulting solution was heated to about 85°C. The clean substrates were immersed into the solution and left for 20 sec. Then, they were immersed into a hot deionized water at 85°C for 20 sec, after that the substrates dried for 20°C with hot air and this step represent one cycle. This process was repeated for 20 times in order to get sensible thick layers of CdO film. Finley the samples annealed for 1 h at different temperatures 300°, 350° and 400°C. The process of post-annealing of CdO films Utilizing electrical furnace (Carbolite) Model (CWF 1200) by (KJ Group Company). Structural properties for films were analyzed utilizing ((XRD-6000 Shimadzu X-ray diffractometer)) with (CuKα radiation and wavelength = 1.541 A°). The angle of scanning found in the range 10-80° with current 20 mA, voltage 40 kV and speed 5°/min). The optical properties were investigated by (Double bram UV-spectrophotometer) Model (MEGA-2100- SCINCO) in the wave length range (400-800 nm).

RESULTS AND DISCUSSION

Structural properties The structural characteristics of CdO films coated on glass substrate by SILAR technique was investigated utilizing the technique of X-ray diffraction. Figure 1 illustrates the patterns of XRD of CdO and all peaks of the thin films have well agreement with data of crystallographic (#73-2245 JCPDS). The basic

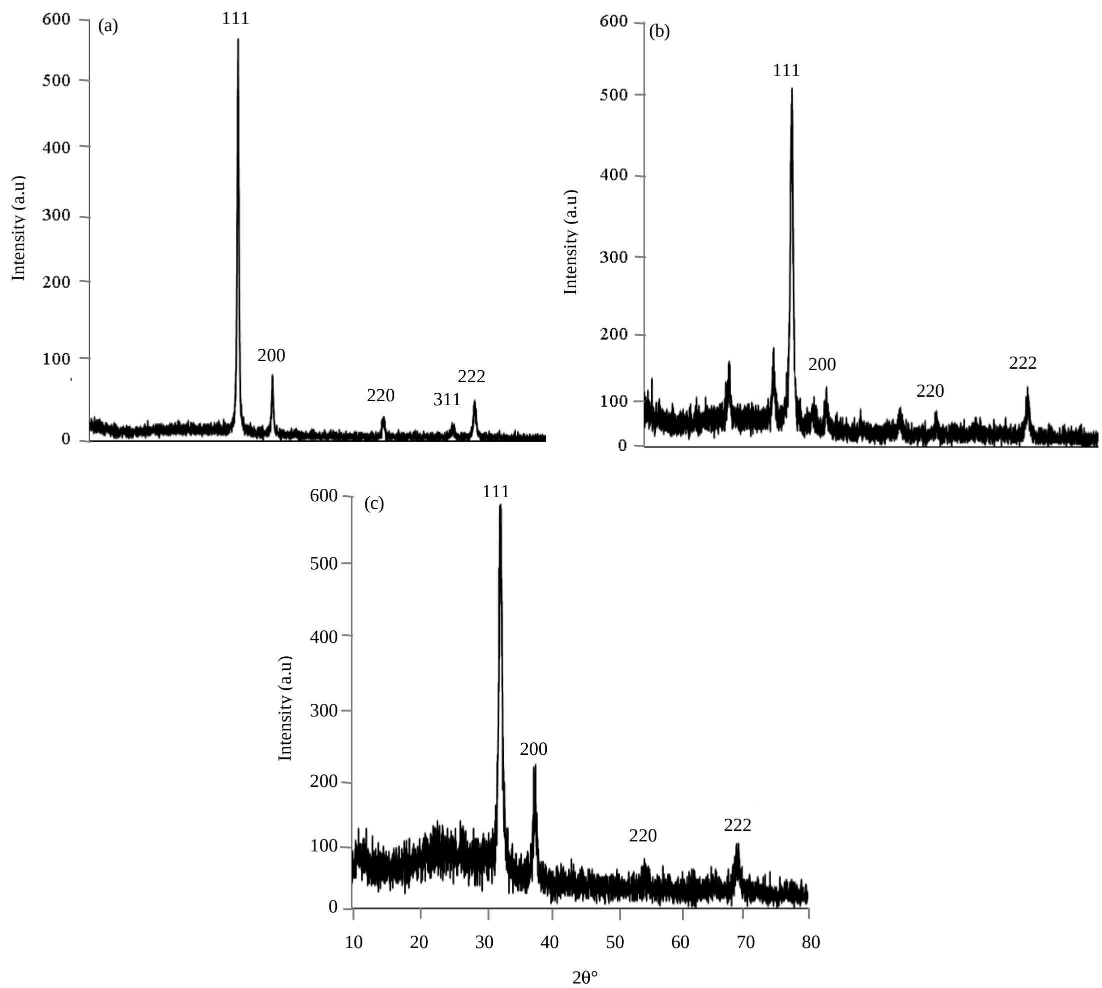

Fig. 1: XRD pattern of CdO/glass thin film deposited using SILAR method and annealed at: a) T = 300°C; b) T = 350°C and c) T = 400°C

Table 1: The results which obtained from the XRD for CdO thin films

Variables	2θ°	d(A°) observed	d(A°) standard	(hkl)	D crystallite size (nm)	$\delta\times10^{15}$ lines/m^2	a(A°)
CdO T = 300°C	32.783	2.7318	2.7130	111	52.637	0.3609	4.731
	38.067	2.3639	2.3495	200	85.447	0.1369	4.727
	55.023	1.6689	1.6614	220	45.537	0.4822	4.720
	65.704	1.4211	1.4168	311	30.046	1.1077	4.713
	69.122	1.3578	1.3565	222	28.725	1.2119	4.703
CdO T = 350°C	32.773	2.7326	2.7130	111	21.048	2.7225	4.733
	38.310	2.3495	2.3495	200	106.91	0.0874	4.699
	54.976	1.6702	1.6614	220	28.452	1.5323	4.724
	69.103	1.3582	1.3565	222	20.105	2.4739	4.704
CdO T = 350°C	32.839	2.7273	2.7130	111	23.394	1.8272	4.723
	38.132	2.3600	2.3495	200	30.525	1.0732	4.720
	55.680	1.6508	1.6614	220	45.674	4.7936	4.669
	69.088	1.3584	1.3565	222	25.129	1.5836	4.705

diffraction peaks of CdO thin film are 111, 200, 222 with one more additional peak at 311 plane when the films annealed at 300°C as demonstrated in Fig. 1a.

Figure 1b and c show the X-ray diffraction peaks of Cadimum Oxide thin films were appeared at 111, 200, 220 and 222 planes when the films annealed at (350° and 400°C). The values of the (2) and the values of d-spacing for CdO peaks are given in Table 1.

The X-ray diffraction peaks reveal to a preferred orientation of cadmium oxide which is along (111) plane

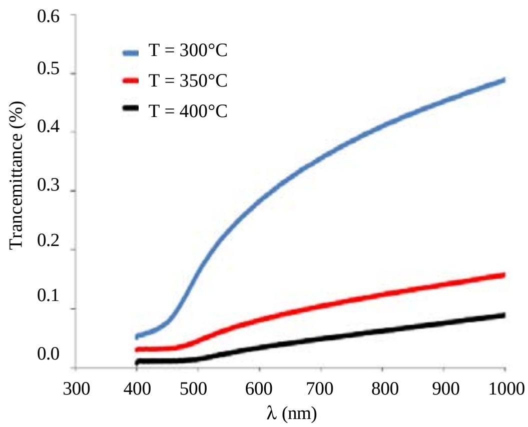

Fig. 2: Transmittance of CdO/glass thin films annealed at three various temperatures

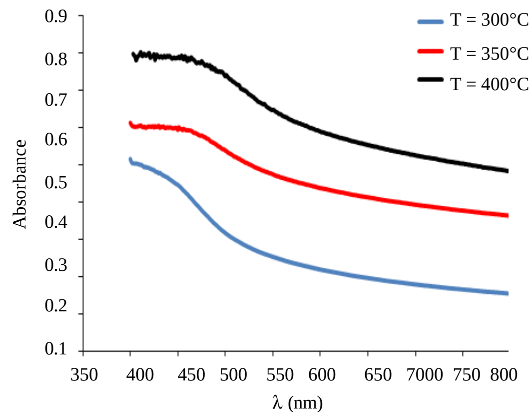

Fig. 3: Absorbance spectra of CdO/glass thin films annealed at three different temperatures

and its structure is a polycrystalline in nature with cubic phase. The intensity of preferential orientation reduced with increased annealing temperature which refer to reduced of crystallinity because of the annealing process effect on the crystalline quality of CdO thin film. Debye-Scherrer's formula was used to find the crystallite size (Cullity and Stock, 2001; Alkhayatt and Shymaa, 2015):

$$D = \frac{0.9\lambda}{\beta COS\theta} \quad (1)$$

Where:

D = The crystal size
λ = The X-ray's wavelength used
β = (FWHM) in radian
θ° = The diffraction angle for Bragg

The dislocation density (δ) was calculated using the following relations (Sahin, 2014):

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

In general, it can be observed that crystallite size was decreased with increasing annealing temperature this might because of increasing in grain boundaries and increasing in the amount of defect which lead to increase in dislocation density (δ) in the films (Table 1).

Optical properties: The transmittance spectrum recorded in the wavelength range of (400-800 nm) is demonstrated in Fig. 2. The average transmittance decreased rapidly with increasing annealing temperature to be 43, 13 and 7% at 300°, 350° and 400°C, respectively. There is a shift of the optical edge toward longer wavelength (red shift) when the annealing temperature is increased.

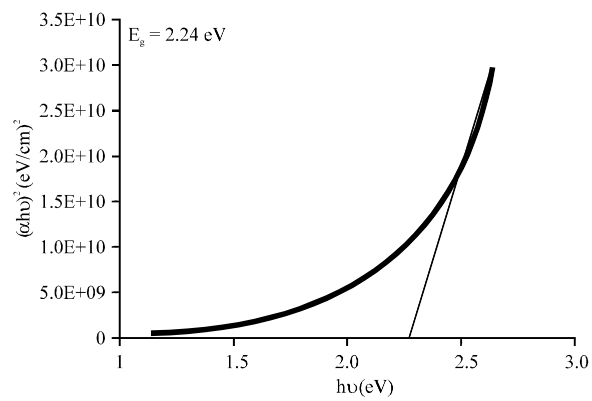

Fig. 4: Energy gap (E_g) of cadmium oxide thin film annealed at T = 300°C

Figure 3 shows that absorbance generally decreases with wavelength. From the figure can be observed the increases in absorbance value in the visible region after the annealing process. Tauce formula was used to determine the values of energy band gap as the following (Tauce, 1974):

$$\alpha h\nu = B(h\nu - E_g^{opt}opt)^r \quad (3)$$

Where:

B = Independent constant of an energy
E_g^{opt} = The optical band gap and r = 1/2 direct and r = 2 for indirect of transitions

The increase of the annealing temperature led to reduce in the energy gap to about 2.24, 2 and 1.9 at 300, 350 and 400°C, respectively as shown in Fig. 4-6. This variation might attributed to generating localized-states within the energy gap and rearrange the atoms in the crystalline lattice (Usharani *et al*., 2015). The reflectance R of the annealed films was determined using the relation (Mitchell, 2004):

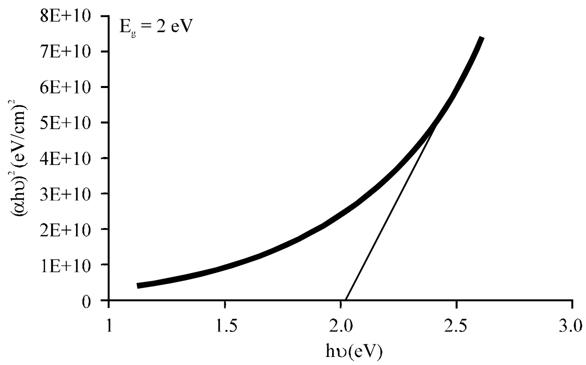

Fig. 5: Energy gap (E_g) of cadmium oxide thin film annealed at T = 350°C

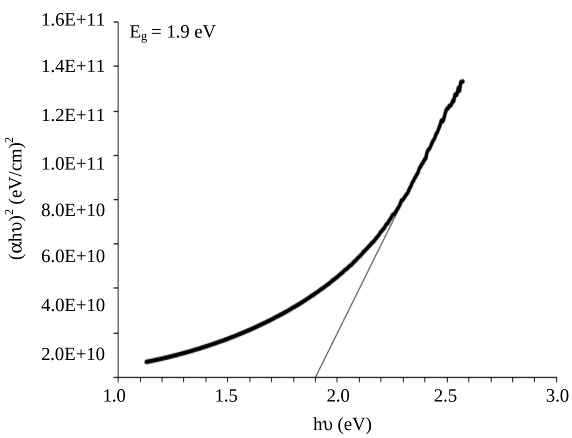

Fig. 6: Energy gap (E_g) of cadmium oxide thin film annealed at T = 400°C

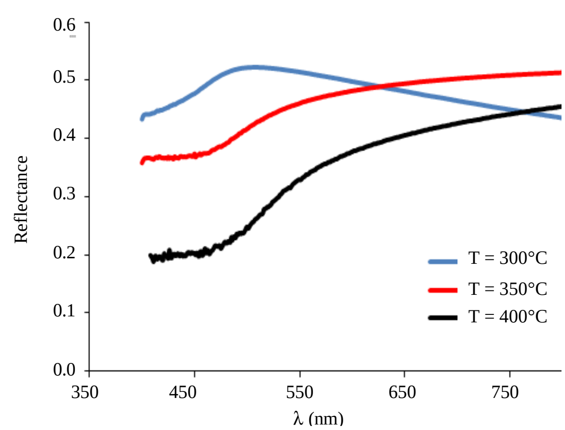

Fig. 7: Reflectance curves of CdO thin films

$$R+A+T = 1 \quad (4)$$

Figure 7 shows the cadmium oxide reflectance curves when annealed at three different temperatures.

CONCLUSION

The XRD studies have been indicated that the CdO thin films were prepared on glass substrates utilizing SILAR method have the (polycrystalline) structure with (cubic) phase. The crystallite size decrease as temperature of glass substrate increase by annealing process and the best crystallinity was found when the film annealed at 300°C. The energy band gap in CdO thin films was decreased from 2.24-2 eV as annealed temperature increases from 300-350 and 1.9 at 400°C. These consequences show that fabricating CdO thin films were attractive to many applications such as solar cells and optoelectronic device.

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