Structural and Optical Properties of $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ Thin Films Prepared by Pulsed Laser Deposition

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ABSTRACT. CdO thin films have been deposited at different concentration of SnO_2 ($x = (0.0, 0.05, 0.1, 0.15 \text{ and } 0.2)$) Wt. % onto glass substrates by pulsed laser deposition technique (PLD) using Nd-YAG laser with $\lambda = 1064 \text{ nm}$, energy = 600 mJ and number of shots = 500. X-ray diffraction (XRD) results reveal that the deposited $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films cubic structure and the grain size increase with increasing annealing temperature and increasing concentration of SnO_2 . The optical transition in the $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films are observed to be allowed direct transition. The value of the optical energy gap decreases with increasing of annealing temperatures and increase with increasing concentration of SnO_2 for all samples.

1. INTRODUCTION

Cadmium Oxide CdO The unique combination of cardio thin film properties which were represented by high electrical conductivity, high carrier concentrations and high transparency in the visible range of the electromagnetic spectrum, made it suitable for a wide range of applications in different fields [7,6]. The applications of CdO thin films can be summaries as follows:

- Its application in photovoltaic solar cells for front contacts window layer, or as heterostructure such as CdO/CdTe or CdO/Cu₂O solar cells [1, 8, 9].
- Photo electrochemical devices [3].
- Phototransistors [3, 5].
- Application in photodiodes [2, 4].
- Liquid crystal displays [5, 4].
- Antireflection coatings [3].
- IR detectors [5].
- Gas sensors [2, 1, 3, 4].
- Transparent electrodes, where was used as transparent anodes for organic light emitting (OLEDs) as a practical new display technology [1,3].
- It has been used as heat mirrors, due to its high reflectance in the infrared region together with transparency in the visible region [4].

Stannic Oxide SnO₂ In 1942 Masters [10] succeeded in preparing conductive transparent tin oxide, for the first time. A substance with white color has a molecular weight of (150. 69 g/mol). Its density (6.95 g/cm³), its melting point (1630°C) and its boiling point (1900°C) [11]. Stannic oxide is an n- type semiconducting material with a direct band gap of about 4.0 eV and an indirect band gap of about 2.6 eV [12]. The electron concentration in the conduction band arises primarily from the lack of stoichiometry produced by oxygen deficiency. The property of SnO_2 makes the material useful for many applications. There for increasing attention is begin paid to study this oxide especially on the method of operation, and its electrical and optical properties. SnO_2 thin films have been fabricated using different techniques including pulse laser deposition, electron beam evaporation [13], chemical vapor deposition [14], RF sputtering [15], evaporation and chemical spray pyrolysis [16]. SnO_2 as transparent conducting oxide is used extensively for a variety of applications such as transparent electrodes in solar cells, architectural windows and flat

panel displays [17]. Recently SnO_2 has been integrated into micro chemical silicon devices as a sensing element of micro sensor.

2. EXPERIMENTAL

2.1 Preparation Pellets

High purity powders (99.999%) of CdO and SnO_2 supplied from Fluka were used to form the target as a disk of 2.5cm diameter and 0.4 cm thickness by pressing it under 4 ton force. The pellets which containing the elements were heated to 873K for 3 hours then cooled to room temperature. The temperature of the furnace was raised at a rate of 10 °C/min. The amount of elements content of pellets was evaluated by using the following equation.

$$W_{(\text{CdO})_{1-x}(\text{SnO}_2)_x} = W_{\text{CdO}} \times (1-x) + W_{\text{SnO}_2} \times (x) \quad (1)$$

Where: $W_{\text{CdO}}=128.411$ (atomic weight for CdO), $W_{\text{SnO}_2}=150.69$ (atomic weight for SnO_2) and $x=0, 0.05, 0.1, 0.15$ and 0.2 (concentration of SnO_2).

2.2 PLD and Thin Film Preparation

The $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ films were deposited on glass slides substrates of (2.5×7.5 cm) were cleaned with dilated water using ultrasonic process for 15 minutes to deposit the films at room temperature by PLD technique using Nd:YAG with $\lambda=1064$ nm SHG Q-switching laser beam at 600 mJ, repetition frequency (6Hz) for 500 laser pulse is incident on the target surface making an angle of 45°. The under vacuum of (10^{-3} mbar) at room temperature and annealing temperatures 523 K were presented.

3. RESULTS AND DISCUSSION

3.1 X-ray diffraction results

The main purpose of this section is to investigate the structural type of semiconductor material that is relevant to the work. Also, the effect of $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ ratio at room temperature and annealing temperature 523 K on the thin films structure have been studied. X-ray diffraction pattern of $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ at different concentration of SnO_2 ($x=0, 0.05, 0.1, 0.15$ and 0.2) showed that all these samples have a crystalline structure except ($x=0.2$ at R.T) also polycrystalline structure for it cubic phases (card No. 96-900-6688) with preferred orientation along (111) direction at 2θ around 32.9135° . As shown in Figure (1) to (2) and Table (1), which is in good agreement with the standard JCPDS (Joint Committee on Power Diffraction Standards). The grain size increase with increasing of concentration of SnO_2 , also In conducting the annealing process for films prepared were the results of X-ray diffraction showed that there is an increase in the height of the peaks and intensity decrease in (FWHM) any increase crystallized material membranes, this means that the thermal treatment caused the reduced crystalline defects caused due to the preparation and disadvantages of the interface by giving atoms material enough energy to re-arrange themselves in a crystalline lattice and disposal of the resulting stresses due to thermal lattice [18]. The grain size of thin film calculated using the Scherer's equation [19].

$$G = 0.94 \lambda / \beta \cos\theta \quad (2)$$

Figure (1): X-ray diffraction patterns for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films with different concentration of SnO_2 ($x = 0, 0.05, 0.1, 0.15$ and 0.2) at R.T.

Figure (2): X-ray diffraction patterns for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films with different concentration of SnO_2 ($x = 0, 0.05, 0.1, 0.15$ and 0.2) annealed at 523 K.

Table (1): Structural parameters inter-planar spacing, intensity, FWHM and crystalline size of (CdO)_{1-x}(SnO₂)_x thin films with different concentration of SnO₂ at RT and different annealing temperatures 523 K.

T _a (K)	x	2θ (Deg.)	FWHM (Deg.)	G.S (nm)	Int (a.u)	d _{hkl} Exp.(Å)	d _{hkl} Std.(Å)	hkl	phase	card No.
RT	0	33.6364	0.4141	20.0	20	2.6623	2.7108	(111)	CdO	96-900-6688
		38.5659	0.5432	15.5	17	2.3326	2.3477	(200)	CdO	96-900-6688
		55.2113	0.5326	16.8	10	1.6623	1.6600	(202)	CdO	96-900-6688
	0.05	32.9321	0.3998	20.7	18	2.7176	2.7108	(111)	CdO	96-900-6688
		37.9898	0.6543	12.8	14	2.3666	2.3477	(200)	CdO	96-900-6688
	0.1	33.0602	0.3633	22.8	15	2.7074	2.7108	(111)	CdO	96-900-6688
		38.2458	0.6543	12.9	11	2.3514	2.3477	(200)	CdO	96-900-6688
	0.15	33.3163	0.7689	10.8	15	2.6872	2.7108	(111)	CdO	96-900-6688
523	0	33.2409	0.3965	20.9	35	2.6931	2.7108	(111)	CdO	96-900-6688
		38.4111	0.6485	13.0	32	2.3416	2.3477	(200)	CdO	96-900-6688
		55.6873	0.3874	23.2	15	1.6493	1.6600	(202)	CdO	96-900-6688
		66.0908	0.4274	22.2	15	1.4126	1.4157	(311)	CdO	96-900-6688
	0.05	33.4300	0.3842	21.6	18	2.6783	2.7108	(111)	CdO	96-900-6688
		38.6003	0.5362	15.7	24	2.3306	2.3477	(200)	CdO	96-900-6688
		54.9937	0.3261	27.5	11	1.6684	1.6600	(202)	CdO	96-900-6688
	0.1	33.3039	0.3210	25.8	30	2.6881	2.7108	(111)	CdO	96-900-6688
		38.5372	0.5387	15.6	20	2.3343	2.3477	(200)	CdO	96-900-6688
		55.6242	0.5231	17.2	13	1.6510	1.6600	(202)	CdO	96-900-6688
	0.15	32.5473	0.2843	29.1	17	2.7489	2.7108	(111)	CdO	96-900-6688
		38.6633	0.7593	11.1	22	2.3269	2.3477	(200)	CdO	96-900-6688
	0.2	32.2951	0.2617	31.6	19	2.7697	2.7108	(111)	CdO	96-900-6688
		38.6003	0.5943	14.2	22	2.3306	2.3477	(200)	CdO	96-900-6688

3.2 The Optical Properties of $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin Films

The optical properties of deposited $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ films on glass substrates for different concentration of SnO_2 at room temperature and annealing temperatures 523 K have been determined by using UV-visible transmittance spectrum in the region of (360–1100) nm. Also the energy gap and optical constants have been determined.

3.2.1 Transmittance

The transmittance of the $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films deposited with different SnO_2 concentration ($x=0, 0.05, 0.1, 0.15$ and 0.2) at room temperature and annealing 523K are shown in Figure (3). It is clear from this Figure that the transmittance spectrum of all deposited thin films increases with the increasing of wavelength (λ). On the other hand, the transmittance spectrum increases with the increasing concentration of SnO_2 and this is due to the increase of the surface roughness promoting the decrease of the surface scattering of the light, while the transmittance spectrum decreases with the increasing of annealing temperature. This decrease in the transmittance spectrum is attributed to decrease of the surface roughness promoting the increase of the surface scattering of the light.

Figure (3): The transmittance as a function of wavelength for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films with different concentration of SnO_2 at R.T and annealing temperature 523 K.

3.2.2 The Absorption Coefficient (α)

The absorption coefficient (α) of the $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films deposited with different concentration of SnO_2 ($x=0, 0.05, 0.1, 0.15$ and 0.2) at room temperature and annealing temperatures 523 K are shown in Figure (4). The absorption coefficient exhibits high values ($\alpha > 10^4$) which means that there is a large probability of the direct transition [20], and then (α) decreases with the increasing of wavelength. It is observed that the absorption coefficient (α) decrease with increasing the concentration of SnO_2 , and this is due to the increasing of energy gap with concentration of SnO_2 . Also, we can notice from this Figure (4) that (α) in general increases with the increasing of annealing temperatures and this is due to the decreasing of energy gap with annealing temperatures. The absorption coefficient (α) was calculated in the fundamental absorption region from the following Equation [21]:

$$\alpha = 2.303A/t \quad (3)$$

Figure (4): The absorption coefficient (α) as a function wavelength for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ films with different concentration of SnO_2 at R.T and annealing temperature 523 K.

3.2.3 Optical Energy Gap

The values of optical energy gap (E_g^{opt}) for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ films with a different SnO_2 concentration ($x=0, 0.05, 0.1, 0.15$ and 0.2) deposited at room temperature and annealing temperatures 523 K have been determined using Tauc equation E_g^{opt} is determined by the extrapolation of the portion at $(\alpha h\nu)^2$ from the relations between $(\alpha h\nu)^2$ versus the photon energy ($h\nu$), as shown in Figure (5) and Table (2). Thin films have been determined by using Tauc equation [22].

$$(\alpha h\nu) = A(h\nu - E_g)^{1/2} \quad (4)$$

In general, the values of direct optical energy gap increase with increasing concentration of SnO_2 (x) for all samples. The direct E_g^{opt} increases from $(2.64$ to $3.05)\text{ eV}$ and from $(2.4$ to $2.7)\text{ eV}$ for (R.T and 523) K respectively. This is due to the decrease of the density of state inside the optical gap, the increasing concentration of SnO_2 (x) leads to decreases from the secondary levels and structural defects, which lead to the contract tails region and this leads to expand in the optical energy gap, while the optical energy gap decrease with the increasing of annealing temperatures. Annealing causes a reduction in E_g , this may be due to the dilate of the lattice which causes a shift in the position edge of V.B and C.B because of the temperature dependence of the electron-lattice interaction that leads to change in the lattice constant by growth of grain size and the decrease in the defect states near the bands [23].

Figure (5): The variation of $(\alpha h\nu)^2$ as a function of photon energy ($h\nu$) for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ films with different concentration of SnO_2 at R.T and annealing temperature 523K.

Table (2): Show the values of E_g^{opt} at $\lambda=500$ nm for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films with different concentration of SnO_2 (x) at R.T and annealing temperature 523 K.

T_a (K)	x	E_g (eV)
R.T	0	2.64
	0.05	2.74
	0.1	2.82
	0.15	2.95
	0.2	3.05
523	0	2.4
	0.05	2.5
	0.1	2.55
	0.15	2.6
	0.2	2.7

4. CONCLUSIONS

Cubic structure is the CdO phase for $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ and orientated along (111). The optical transition in the $(\text{CdO})_{1-x}(\text{SnO}_2)_x$ thin films is observed to be allowed direct transition. The value of the optical energy gap decreases with increasing of annealing temperatures and increase with increasing concentration of SnO_2 for all samples.

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