ISSN: 0976-3031

Available Online at http://www.recentscientific.com

CODEN: IJRSFP (USA)

International Journal of Recent Scientific Research
Vol. 9, Issue, 4(H), pp. 26103-26106, April, 2018

International Journal of Recent Scientific Research

DOI: 10.24327/IJRSR

Research Article

EFFECT OF SOLUTION MOLAR CONCENTRATION ON CdO THIN FILMS PREPARED BY SIMPLIFIED SPRAY PYROLYSIS METHOD

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DOI: http://dx.doi.org/10.24327/ijrsr.2018.0904.1991

ARTICLE INFO

Article History:

Received 5th January, 2018
Received in revised form 20th February, 2018
Accepted 8th March, 2018
Published online 28th April, 2018

Key Words:

Thin films, X-ray diffraction, thickness, transmittance, Photoluminescence.

ABSTRACT

CdO thin films were deposited by perfume atomizer spray pyrolysis method on glass substrate at 200^0C by varying solution molar concentration such as 0.1 M and 0.2 M. X – ray diffraction (XRD) analysis show that the prepared cadmium oxide thin films belongs to cubic structure with preferential orientation along with (111) direction. The thickness of the films was determined by Stylus profiler. The average optical transmittance of the CdO films in the range of 300 – 800 nm, is about 60%. Estimated band gap energy (E_g) is 2.234 eV and 2.30 eV for 0.1 M and 0.2 M film respectively. Photoluminescence (PL) spectra showed a strong emission peak around 523 nm.

INTRODUCTION

Recent years, transparent conducting oxides such as cadmium oxide, zinc oxide, indium oxide, tin oxide find wide applications in optoelectronic devices, gas sensors and phototransistors [1–4]. In particular, cadmium oxide (CdO) is a promising material for solar cell applications due to its high electrical conductivity and high optical transmittance in the visible region of the solar spectrum [5]. CdO is a n-type semiconducting material with the band gap energy of 2.2 -2.28 eV [6]. Therefore, CdO is used as a window layer in heterojunction solar cell [7, 8]. Several techniques have been used to deposit CdO thin films such as chemical vapor deposition [9], RF magnetron sputtering [10], chemical bath deposition [11], sol–gel [12], SILAR [13, 14], thermal evaporation [15], layer by layer assembly [16] and spray pyrolysis [17–20]. Owing to simplicity and inexpensiveness, the perfume atomizer spray pyrolysis is a better chemical method for the preparation of thin films with larger area coating [21]. In this work, we report on the deposition of CdO films using perfume atomizer spray pyrolysis technique and studied the properties of the prepared CdO films with the role of precursor concentration on CdO thin films.

Experimental

CdO thin films were deposited on the glass substrates at 200^0C using perfume atomizer spray pyrolysis technique. Cadmium acetate dihydrate [$Cd(CH_3(COO)_2).\ 2H_2O$] was used as source material of Cd, double distilled water was used as solvent and a few drops of NaOH was added to stabilize the crystallite size. Spray solution of 50 ml was prepared with various precursor concentrations (0.1 and 0.2 M) of Cadmium acetate dihydrate. The optimized deposition parameters such as substrate- spray nozzle distance (25 cm), spray angle (about 45^0), spray time (3 s) and spray interval (30 s) were kept constant. The substrate temperature (Ts) was fixed at 200^0C. The prepared precursor solution was sprayed on substrate. We get the reddish brown CdO thin films.

After deposition, the coated substrates were allowed to cool down to room temperature. Thickness of the films was measured using a stylus profilometer (SJ-301, Mitutoyo). X-ray diffraction patterns were recorded using Philips X Pert PRO X-ray diffraction system (Cu Kα radiation; K = 1.54056 Å). Surface characteristics were carried out by AFM (A100 SGS). The optical transmittance spectrum in the wavelength region 190–900 nm was recorded using UV–Vis spectrometer (Shimadzu UV-1601). Room temperature luminescence

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spectrum was recorded using (SHIMADZU-5301) spectrofluorometer.

RESULT AND DISCUSSION

Thickness studies

Thickness of the prepared film was carried out using the stylus profiler. The obtained thickness was 0.38 µm for 0.1 M and 0.41 µm for 0.2 M cadmium acetate dihytrate solution concentration prepared CdO thin films respectively. When increasing the concentration of precursor solution, the thickness of the coated films also increases.

Structural studies

Figure 1 shows the XRD patterns of CdO thin films deposited at 200^0C using different precursor concentrations such as 0.1 M and 0.2 M. The peaks (111), (200), (220), (311) and (222) peaks are appeared the corresponding angles 33.23^0, 38.52^0, 55.47^0, 66.15^0 and 69.45^0. The CdO films exhibit predominant diffraction peak along the (111) plane with a cubic crystal structure and the peak positions are well agreed with JCPDS card no 75-0591(Lattice constant a = 4.694). From the XRD analysis, it is observed that all the prepared CdO thin films are polycrystalline in nature. The (111) plane intensity was found to be increased with increasing precursor concentrations [22]. It was observed that as the cadmium acetate concentration increases, the peak intensity increased correspondingly. This indicates the incorporation of more Cd^{2+} ions on the film during the spray pyrolysis process.

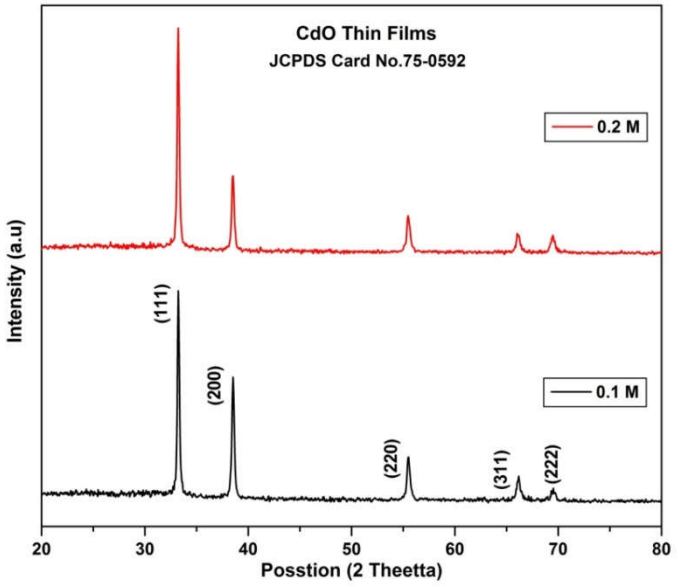

Figure 1 XRD pattern of CdO Thin films

The lattice constant 'a' is calculated from inter planer distance (d) and miller indices. The calculated values of lattice constant 'a' are listed in table 1, is found to be good agreement with the standard (JCPDS) values [23]

$$\frac{1}{d^2} = \left(\frac{h^2 + k^2 + l^2}{a^2}\right)$$

The structural parameter values of 0.1 M and 0.2 M solution concentration prepared CdO thin films are given in Table 1. The crystallite size is calculated using Debye - Scherer's formula [24].

$$D = \frac{0.9\lambda}{\beta Cos\theta}$$

Where, λ = 1.5406 A° for CuKa, 'β' is the full width at half maximum (FWHM) of the peak and 'θ' is the diffraction Bragg's angle. The Williamson-Hall equation according to UDM is given by [25]

$$\beta_{hkl} \cos\theta_{hkl} = \frac{K\lambda}{D} + 4\varepsilon \sin\theta_{hkl}$$

Dislocation density (δ) and strain (ε) for (111) plane is evaluated using the relations [26]

$$\delta = \frac{1}{D^2}$$

$$\varepsilon = \frac{\beta \cos\theta}{4}$$

The X-ray diffraction peak of films corresponding texture coefficient (T_C) is estimated using an expression [27]

$$T_c(h_i k_i l_i) = \frac{I(h_i k_i l_i)}{I_0(h_i k_i l_i)} \left[\frac{1}{n} \Sigma \frac{I(h_i k_i l_i)}{I_0(h_i k_i l_i)}\right]^{-1}$$

Where I_0 - represents the standard intensity, I - is the observed intensity of $(h_i k_i l_i)$ plane and n is the reflection number. In order to study the structural properties of the material, the Nelson – Relay function (NRF) method is employed [28].

$$NRF = 0.5\left(\frac{cos^2\theta}{sin\,\theta} + \frac{cos^2\theta}{\theta}\right)$$

Table 1 Structural parameter of CdO Thin films

Structural Parameters	Cadmium Acetate Dihytrate Concentration	
	0.1 M	0.2 M
Lattice constant (a)	4.669	4.670
Crystallite Size using Debye- Scherer's formula (nm)	40.813	40.779
Crystallite Size using Williamson-Hall equation (nm)	52.474	52.431
Dislocation (δ) X 10^{14} / m^2	6.004	6.013
Strain (ε) X 10^{-3}	2.517	2.614
Texture coefficient	1.399	1.993
Film thickness (µm)	0.38	0.41
NRF	5.075	5.078

*Standard Lattice constant a = 4.694

Optical studies

Transmittance spectra of CdO thin films with different precursor concentrations were recorded in the wavelength range 190–900 nm and are shown in Figure 3. The average optical transmittance is about 65% for 0.1 M concentration and the transmittance is reduced to 59%, for the films prepared with 0.2 M precursor concentration of cadmium acetate. The optical transmittance of CdO film increases with increasing the precursor concentration, which may be due to the change in the crystallite size as well as film thickness.

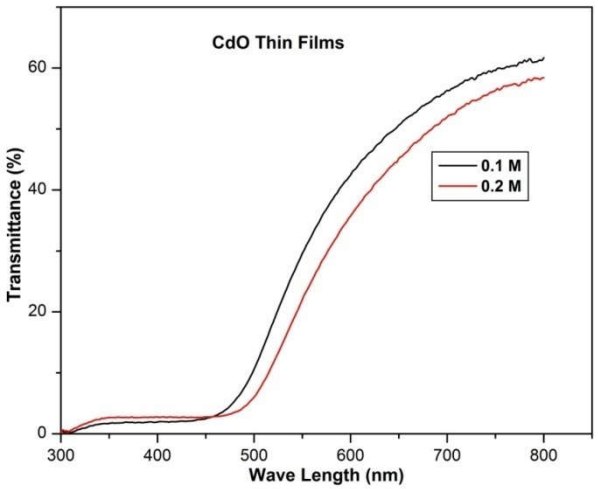

Figure 3 Transmittance of CdO Thin films

The optical band gap of various solution concentrations prepared CdO thin films was estimated by extrapolating the linear portion of $(\alpha h\nu)^2$ versus $h\nu$ plots. The absorption coefficient (α) is calculating the following equation.

$$\alpha = \frac{1}{d} In\left(\frac{1}{T}\right)$$

Where T is transmittance and d is film thickness. The absorption coefficient (α) and incident photon energy ($h\nu$) is related by the Touc's relation as follows.

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

Where $h\nu$, A and E_g are photon energy, constant and optical band gap respectively. Figure 4 shows the plot of $(\alpha h\nu)^2$ versus photon energy ($h\nu$) for various cadmium acetate dihytrate concentration CdO thin films. The optical band gap increased from 2.23 - 2.30 eV with the precursor concentration 0.1 M - 0.2 M. The optical band gap values increase from 2.14 – 2.35 eV for various precursor concentrations (0.025 M - 0.1M) of CdO thin films by spray method was previously reported [29].

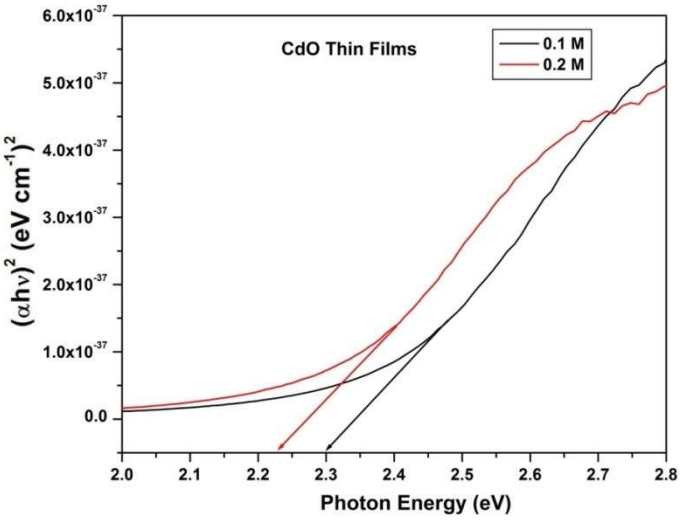

Figure 4 Touc plot of CdO Thin films

Photo Luminescence studies

Figure 5 shows the room temperature photoluminescence (PL) emission spectra were carried out at an excitation wavelength 320 nm for 0.1 M and 0.2 M solution concentration CdO thin films. There are four emission peak obtained all the samples. The weak peak appeared at 495 nm (2.50 eV) are attributed to transition between states at the bottom of the conduction band and the top of the valence band of the CdO thin film and band edge emission. The second peak appeared at 503 nm (2.46 eV). The strong emission peaks centered at 523 nm (2.37 eV) are assigned due to an exciton bound to a donor level. The other peaks at 508 and 535 nm are located in the visible spectrum at approximately 2.12 eV and are associated with emission due to impurity levels. The defect-related luminescence peak is obtained with broad bands due to radioactive transitions between oxygen vacancies. In other hand, it may have been raised Cd interstitials acting as shallow donors and Cd vacancies acting as deep acceptors. Which leads to the recombination mechanism and competes with excition-related luminescence depend on the stoichiometry and the preparation conditions (such as substrate temperature, concentration and pH) of the CdO thin films.

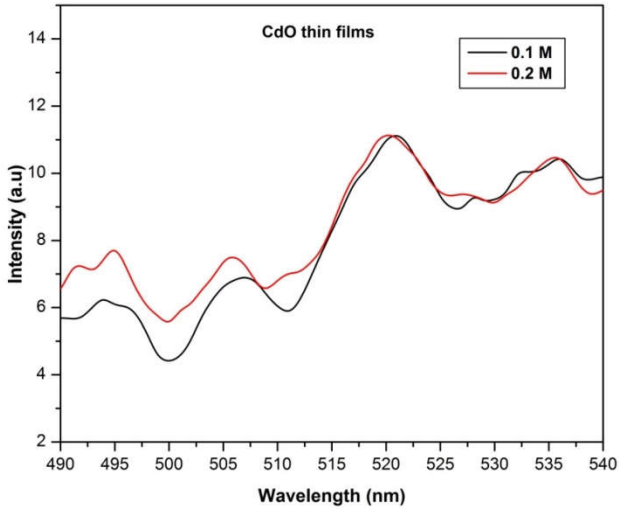

Figure 4 PL spectra of CdO Thin films

CONCLUSION

Cadmium oxide thin films were deposited by spray pyrolysis technique at a substrate temperature of 200^0C with 0.1 M and 0.2 M concentration of cadmium acetate dihytrate. Poly crystalline nature and cubic structure of CdO films were confirmed by XRD. The calculated average crystallite size was 40 nm. Average optical transmittance of 62% in the wavelength range 480–800 nm, is obtained for films with 0.1 M and the optical band gap energy (E_g) increases from 2.23 – 2.30 eV as the precursor molar concentration increases from 0.1 to 0.2. Intense PL emission peak is observed at 523 nm due to the presence of oxygen vacancies at high precursor concentrations. These films were found to good physical properties desirable for solar cell and other optoelectronic applications.

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How to cite this article:

Rajini M., Karunakaran M *et al*.2018, Effect of Solution Molar Concentration on Cdo Thin Films Prepared by Simplified Spray Pyrolysis Method. *Int J Recent Sci Res.* 9(4), pp. 26103-26106. DOI: http://dx.doi.org/10.24327/ijrsr.2018.0904.1991

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