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Effect of Substrate Temperature on Mn doped CdO thin films Prepared by Perfume Atomizer Spray Pyrolysis Method

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Abstract—Mn doped CdO thin films were deposited by perfume atomizer spray pyrolysis method on glass substrate by varying substrate temperature (T=200^0 C, 225^0 C, 250^0 C, 275^0 C, 300^0 C). The 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 thicknesses of the films were determined by Stylus profiler. The average optical transmittance of the CdO films in the range of 300 – 800 nm, is about 58%. Estimated band gap energy (E_g) is 2.65 eV and 2.5 eV. Photoluminescence (PL) spectra showed a strong emission peak around 523 nm. A systematic study on the influence of the substrate temperatures on the optical, electrical and structural properties of Mn doped CdO thin films deposited by perfume atomizer spray have been reported.

Keywords—*Thin films, X-ray Diffraction,thickness, transmittance, photoluminescence.*

I. INTRODUCTION

Cadmium oxide (CdO) is one of the promising transparent conducting oxides (TCOS) from II to VI groups of semiconductors which has a great potential use for optoelectronic devices [1]. CdO has a high optical transmittance in the visible and NIR region [2]. CdO is a n-type semiconductor, has several attractive properties, high density, low resistivity, high melting point [3,4]. It was used in many industrial productions like solar cells, flat panel display optical communications, smart windows, photo transistors, Infrared detectors, ceramic glasses, storage batteries and other optoelectronic applications [5-11]. Doping suitable elements in metal oxide semiconductors show a critical role in tuning their optical and electrical properties of the potential application. The optical properties and bandgap can be modified by doping CdO with transition metals such as Aluminium, Fluorine, Tin, Copper, Zinc, Gadolinium, Boron and Indium [12-19]. These results infer that the optical and electrical properties of CdO can be enhanced by doping with suitable transition elements. In the present work, the effect substrate temperature on Mn doped CdO thin film on the structural, surface, optical and electrical properties of CdO thin films prepared by the perfume atomizer spray pyrolysis technique were reported.

II. MATERIALS AND METHODS

Mn doped CdO thin films were deposited on the glass substrates using the perfume atomizer spray pyrolysis technique. In the preparation of CdO films, 0.1M cadmium acetate [$Cd(CH_3(COO)_2)$] was used as a source material of Cd and double distilled water was used as the solvent. Mn doping was achieved by mixing aqueous solution of manganese acetate with the above precursor solution. Microscopic glass slides were used as substrates. The resultant precursor solution was sprayed on preheated substrates at different temperature such as 200, 225, 250, 275 and 300ºC. The substrate temperature was controlled through a thermocouple with the help of PID temperature controller. The optimized deposition parameters such as substrate-spray nozzle distance (25cm), spray angle (about45º), spray time (3s) and spray interval (30s) were kept constant. After deposition, the coated substrates were allowed to cool down to room temperature. The thickness of the films was measured using Stylus Profilometer (SJ-301, Mitutoyo). The X-ray diffraction (XRD) pattern of the prepared films were recorded using a Philips X Pert PRO X-ray diffraction system (Cu Kα radiation, K = 1.54056 Å). The Surface morphology of the prepared samples was analyzed using scanning electron microscope. Optical transmittance spectrum was recorded in the wavelength range of 190–900 nm using UV–vis spectrometer (Shimadzu UV-1601). Room

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temperature Photoluminescence spectrum was recorded using (SHIMADZU-5301) spectrofluorometer.

III. Results and Discussion

X-Ray Diffraction Studies

Thickness of the prepared Mn doped CdO thin films are measured by using the stylus profiler method. Film thickness increases with substrate temperature. The lattice constant of CdO thin films are calculated using the following formula,

$$\frac{1}{d^2} = \frac{h^2+k^2+l^2}{a^2} \quad \text{------------ (1)}$$

The calculated film thickness, lattice constant 'a' and d-spacing values are presented in table 1.

Figure 1 shows the X-ray diffraction pattern of Mn doped CdO thin films deposited by perfume atomizer spray pyrolysis method as a function of various substrate temperatures. The XRD pattern of reveals that all films are polycrystalline in nature and exhibits the cubic crystal structure as compared with JCPDS file no. 75-0591. The calculated lattice constant values agree with the standard JCPDS value. The Mn doping in the CdO film the intensity of the (200) plane is slightly decreased and the intensity of (111) peak is increased with temperature. The reason for the change in growth orientation is substitution of Mn ions into the Cd lattice sites. The results were well matched with F, Mo and Mn doped CdO thin films prepared by the spray pyrolysis method [20-23].

The slight increase of lattice constant is due to the increase in inter atomic spacing which results of substitution of smaller Mn^{2+} ions (0.83Å) into larger Cd^{2+} ions (0.95Å) [24, 25]. To further increase of substrate temperature Mn doping concentration, the diffraction angle slightly shifted towards the lower angle owing to mixed phases such as CdO and MnO structures. This new diffraction plane is corresponding to the MnO (matched with JCPDS75–1090), which may be due to incorporation of Mn^{2+} ions. The average crystallite size (D) of the CdO film was calculated from the following Scherrer's equation for the (111) plane [26].

$$D = \frac{k\lambda}{\beta Cos\theta} \quad \text{------------ (2)}$$

Where, k is a constant taken to be 0.94, λ is the X-ray wavelength, β is the full-width at half-maximum of the peak and θ is the reflection angle

Dislocation density (δ) and strain (ε) for (111) plane was evaluated using the relations [28, 29]

$$\delta = \frac{1}{D^2} \quad \text{------------ (3)}$$

$$\varepsilon = \frac{\beta Cos\theta}{4} \quad \text{------------ (4)}$$

The number of crystallites calculated using the formula

$$N = \frac{t}{D^3} \quad \text{------------ (5)}$$

The lattice distortion (L.D) developed in thin films can be evaluated from the relation [26]

$$L.D = \frac{\beta}{4\tan\theta} \quad \text{------------ (6)}$$

The calculated crystallite size, dislocation density, strain, Number of crystalline and lattice distortion values are presented in Table - 2.

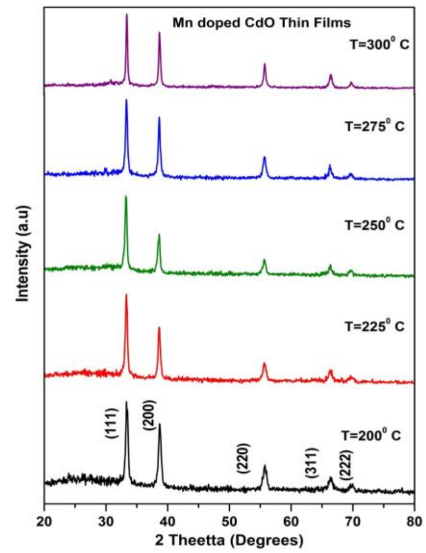

Figure 1: XRD pattern of Mn doped CdO thin films

Table 1: Micro-structural Parameter of Mn doped CdO thin films

Substrate Temperature (Ts) ^{0}C	Thickness (t) µm	d lattice spacing $\times10^{-10}$m	Lattice constant 'a' $\times10^{-10}$m
200	0.52	2.6704	4.629
225	0.63	2.6790	4.643
250	0.68	2.68607	4.657
275	0.77	2.68672	4.656
300	0.79	2.69012	4.663

*Standard Lattice constant a = 4.694

Table 2: Micro-structural parameters of Mn doped CdO thin films

Substrate Temperature (Ts) ^{0}C	Crystallite size (nm)	Dislocation density $\times10^{14}$	Strain $\times10^3$	No. crystalline $\times10^{-15}$	Lattice Distortion $\times10^3$
200	21.07	0.225	1.644	0.555	5.700
225	24.07	0.172	1.440	0.451	5.021
250	28.08	0.126	1.234	0.306	4.309
275	33.70	8.800	1.028	0.201	3.585
300	42.14	5.629	0.822	0.105	2.860

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Table 1 show that the d spacing and lattice constant slightly increases with substrate temperature. Table 2 shows the microstructural parameter of Mn doped CdO thin film at different temperature. As can be seen, the crystalline size of the Mn doped CdO films increases, but the strain and number of crystalline decreases with increasing temperature. The lattice constant and thickness slightly increases with temperature. However, it is observed that the crystalline size in the doped film does not vary in the XRD pattern, which is attributed to the lattice disorder produced in the films at Mn doping due to differences in the ionic radii of Cd^{2+} and Mn^{2+}ions.

Surface Morphological Studies

Figure 2. Shows the surface morphology of Mn doped CdO thin films at substrate temperature 200 and 275^{0}C. The grains formed on the surface are irregular in spherical shape and densely packed. The size of the grains is increased and uniformly distributed on the overall surface and the surface contains cauliflower like structures. The SEM images show that the surface is fully covered with grains of nearly uniform size and is free from pin holes. Thus it shows that the temperature of the substrate effectively modifies the surface morphology of the Mn doped CdO films. The composition of Mn doped CdO films was confirmed by energy dispersive X-ray spectroscopy (EDAX).The EDAX spectra of the Mn doped CdO thin film shown in Fig. 3. The elements Cd, Mn, and O are present in the film and the other element Si which is not expected in the film may be resulted from the glass substrate.

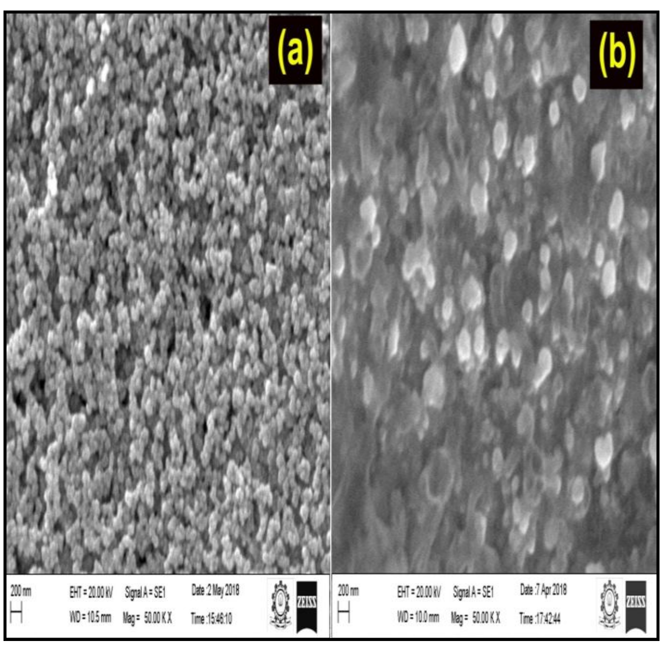

Figure 2: SEM image of Mn doped CdO thin films of substrate temperature (a) 200^0 C and (b) 275^0 C

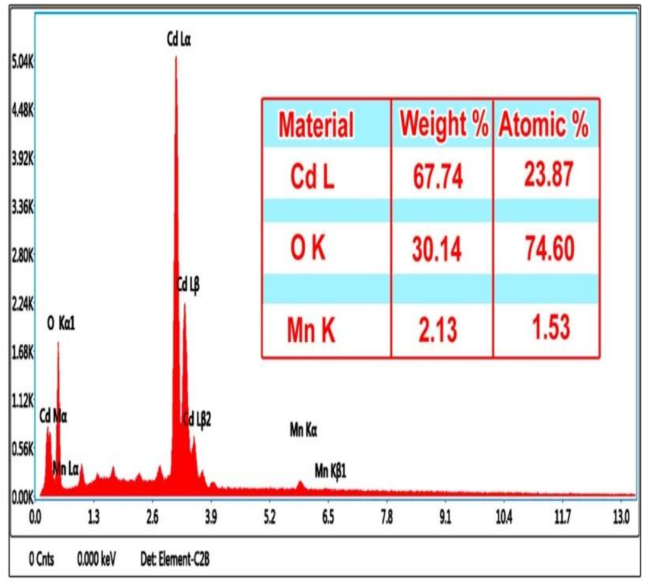

Figure 3: EDAX image of Mn doped CdO thin films

Optical Studies

The optical transmittance spectra of the Mn doped CdO films were recorded in the range of 400–800 nm at different temperature is shown in Fig. 4. The slight variation in transmittance was observed in the Mn doped films at different temperature. It may be due to the incorporation of more Mn in the lattice and interstitial positions and also a small increase in the free charge carrier concentration [30–32]. Moreover, obtained results from optical transmission patterns revealed that with the increasing substrate temperature, optical transmittance decreases.

The direct bandgap determination is based on the extrapolated linear regression of the curve resulting from a plot of photon energy hν vs $(\alpha h\nu)^2$. The absorption coefficient (α) can be calculated from the transmittance spectrum. The optical band gap (Eg) of the films was estimated from the transmittance data where the photon energy (hυ) and absorption coefficient (α) are related by the following equation [33].

$$\alpha h\nu = B(h\nu - Eg)^r \quad \text{--------(6)}$$

For allowed direct transition, the value of r = 1/2 and B is constant. The above equation can be rearranged as

$$In(\alpha h\nu) = InA + rIn(h\nu - Eg) \quad \text{--------(7)}$$

Figure 5 shows the Tauc's plot of Mn doped CdO film at different temperature. As clearly seen in Figure 5, the optical gap energy, decreased with increasing of substrate temperature from 2.65 to 2.5 eV, the band gap is narrowing due to the decrease in the transition tail width and shift effect [34, 35].

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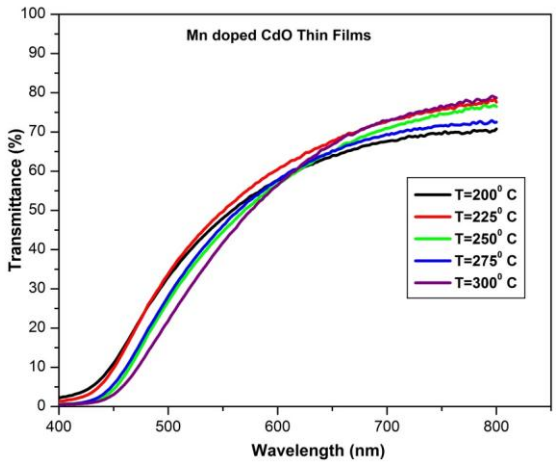

Figure 4: Transmittance spectra of Mn doped CdO thin films at different substrate Temperature

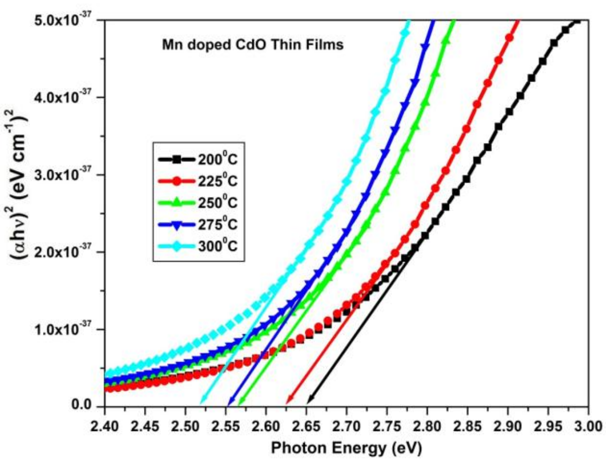

Figure 5: Tauc's plot of Mn doped CdO thin films at different substrate Temperature

Photoluminescence spectra

Photoluminescence (PL) is an important tool to investigate the quality of a thin film which depends on the size of the crystallites, morphology and chemical environment [36]. The PL spectra of Mn doped CdO at different substrate temperature is shown in figure 6. It was observed that the PL spectra consist of emission peaks centered at 494.76nm, 520.61nm and 594.28 nm for Mn doped CdO at all substrate temperatures. This confirms the perfect stoichiometries nature of the doped samples at different substrate temperature. The peak centered at 494.76 nm may be attributed to the combination of the electrons from the conduction band and holes from the valence band. The peak at 520 nm is attributed to the excitonic transition which is size-dependent. The peakat594.28 nm may be ascribed to the deep trap emission and surface state emission that is less size-dependent. This peak confirms the quantum confinement effect of the as deposited samples which is ascribed to the near-band-edge (NBE) emission of CdO. This NBE emission attributed to oxygen vacancies and cadmium interstitial [37, 38].

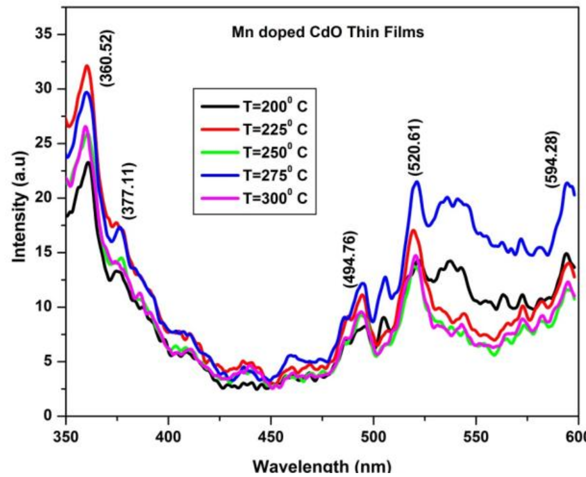

Figure 6: Photoluminescence spectra of Mn doped CdO thin films at different substrate Temperature

IV. CONCLUSION

The transparent and conducting Mn doped CdO thin films were grown on glass substrate at different substrate temperature using perfumer atomizer spray pyrolysis method. The effects substrate temperature on structural, morphological and optical properties of Mn doped CdO films were investigated. The prepared films show the cubic crystal structure of the CdO with a slight variation of lattice parameter due to the replacement of cadmium by manganese atoms. The substrate temperature can be effectively modified on the structure and surface morphology of Mn doped CdO films. The optical band gap is slightly influenced by the substrate temperature and decreases from 2.65 to 2.5 eV with temperature. SEM images show the evidence of morphological variation. EDAX spectrum shows the presence of Cd, O and Mn ions in the prepared films.

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