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ELECTRICAL PROPERTIES OF SPRAY DEPOSITED CDO THIN FILMS: EFFECT OF SUBSTRATE TEMPERATURE

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ABSTRACT:

The objective of this work is to study the influence of deposition temperature on electrical properties of the CdO thin films prepared by spray pyrolysis technique. These films were characterized for the structural and electrical properties by means of X-ray diffraction (XRD), and electrical resistivity. As deposited CdO films are polycrystalline with (111) preferential orientation. The information on crystallite size, microstrain and dislocation density is obtained from the full width-at half- maximum (FWHM) of the diffraction peaks. The electrical conductivity of the films was found to increase with increase in substrate temperature. The high conductivity of 6.17×10^3 $(\Omega\text{-cm})^{-1}$ and high transparency (87%) was obtained for the film grown at 400 ^{0}C.

Keywords: Thin films; X-ray diffraction; Cadmium oxide; spray pyrolysis; Electrical conductivity.

1. INTRODUCTION

Transparent conducting metal oxide semiconductor materials have attracted much attention owing to their potential applications in flat panel display, smart windows, light emitting diodes, heat reflectors, electronic, photovoltaic devices and solar cells [1–4]. Its high electrical conductivity and high optical transmittance in the visible region of the solar spectrum along with a moderate refractive index make it useful for various applications such as transparent electrodes, phototransistors, photodiodes, gas sensors, etc. [5-6]. CdO is an n-type semiconductor with a rock-salt crystal structure (FCC) and possesses a direct band gap of 2.2 eV [7]. Besides, the CdO will be attractive in the field of optoelectronic devices by making heterostructures with ZnO which has band gap energy of 3.3 eV. CdO thin films have been prepared by various techniques such as sol–gel, DC magnetron sputtering, radio-frequency sputtering, spray pyrolysis, pulsed laser deposition, chemical vapor deposition, and chemical bath deposition [8-14].

It is one step method operating at atmospheric pressure with very short production time [15]. This can be used to tune the band gap of materials. Due to these economical and flexible experimental conditions, spray pyrolysis has been employed to deposit CdO thin films and to study various properties of the films.

2. EXPERIMENTAL DETAILS

All the chemical reagents used in the experiments were obtained from commercial sources as guaranteed-grade reagents and used without further purification. The amorphous glass substrates supplied by Blue Star Mumbai, were used to deposit the CdO thin films. Before the deposition of CdO thin films, glass slides were cleaned with detergent and distilled water, then boiled in chromic acid (0.5 M) for 25 min, then slides washed with double distilled water and further ultrasonically cleaned for 15 min. Finally the substrates were degreased in AR grade acetone and used for deposition.

2.1. Thin film preparation

CdO films were prepared on preheated glass substrate using a spray pyrolysis technique. Spray pyrolysis is basically a chemical process, which consists of a solution that is sprayed onto a hot substrate held at high temperature, where the solution reacts to form the desired thin film. The spraying solution was prepared by mixing the appropriate volumes of 0.5 M cadmium sulphate ($CdSO_4$) and distilled water. The CdO films were deposited at different substrate temperatures of 250, 300, 350 400 and 450 ^{0}C. Samples deposited at various substrate temperatures are denoted by C250, C300, C350, C400 and C450, where numbers stand for substrate temperatures. The optimized values of important preparative parameters are shown in bracket viz. airflow rate which is used as carrier gas (1.2 kg/cm^2), spray rate (3 ml/min), distance between substrate to nozzle (30 cm), solution concentration (0.5 M) and quantity of the spraying solution (30 ml). After the deposition, the films were allowed to cool naturally at room temperature. All the films were transparent and well adherent to the substrate, were further used for structural, morphological, optical and electrical characterizations.

3. RESULTS AND DISCUSSION

3.1 Film thickness

Film thickness has a strong impact on the structural, optical and electrical properties of the device. Film layer thickness was measured by gravimetric weight difference method using the relation,

$$t = \frac{m}{\rho A} \qquad (1)$$

where, m is the mass of the film deposited onto the substrate, A is the area of the deposited film and ρ is the bulk density of CdO (8.15 g/cm^3). Thicknesses of as-deposited thin films were found to be in the range from 310 nm to 425 nm and listed in Table 1.

3.2. X-ray diffraction analysis

The XRD patterns of as-deposited CdO thin films as function of deposition temperatures are shown in Fig. 1. All the films are observed to be well grown with polycrystalline textures having cubic CdO phase. For all films, major crystalline texture is (1 1 1)-oriented one (JCPDS card No.: 75-0592) at 2θ~33.08^0. The intensity of the major reflex increased with increase in deposition temperature up to 400 ^{0}C and further intensity decreased. For the CdO films, the main characteristic peaks are assigned to the (1 1 1), (2 0 0), (2 2 0) (3 1 1) and (2 2 2) planes. A light preferential orientation in the (1 1 1) plane is observed for the CdO films and similar behavior has also been reported by other researchers [16].

The grain size, strain, lattice parameter, dislocation density and texture coefficient were estimated form the XRD pattern. The grain size was estimated using Scherrer's equation [17]

$$D = \frac{0.94\lambda}{\beta \cos\theta} \qquad (2)$$

where λ is the wavelength of incident X-ray, β is the full width at half maximum (FWHM) measured in radians and θ is the Bragg's angle of diffraction peak.

The strain and dislocation density values were calculated using the standard relations [18]:

$$\text{Strain } (\varepsilon) = \frac{\beta \text{Cos}\theta}{4} \qquad (3)$$

$$\text{Dislocation density } (\delta) = \frac{1}{D^2} \qquad (4)$$

where 'D' is the grain size. The strain values varied in the range of 1.17×10^{-3} to 0.67×10^{-3}. The dislocation density value was around 1.173×10^{15} lines/m^2. The calculated grain size, strain values and dislocation densities are tabulated in Table 1.

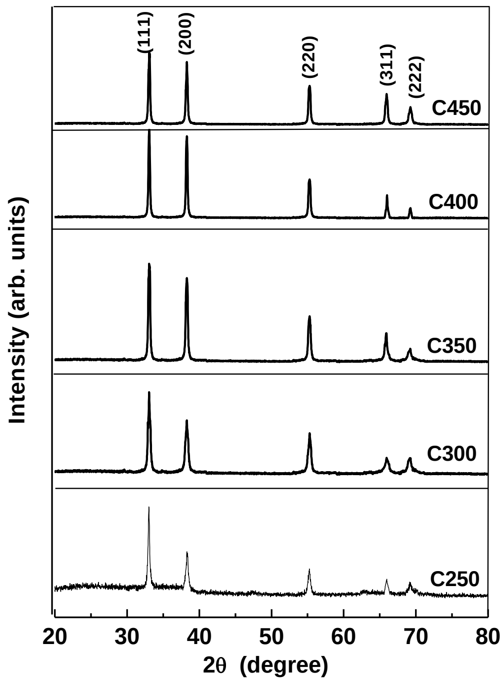

Fig.1. XRD patterns of CdO thin films grown at different substrate temperatures.

3.3 Electrical properties

The D.C. two point probe method of dark electrical resistivity measurement was used to study the variation of electrical resistivity (ρ) with temperature in the range 300-550 K. The variation of resistivity (ρ) with reciprocal temperature (K^{-1}) is depicted in Fig. 2. The deposition temperature variation affects the resistivity magnitude of CdO thin films because a more complete reaction occurs as the temperature is increased. The decrease in resistivity is also expected up to a certain temperature, (here 400 ^{0}C) considered as the optimum deposition temperature, above which the resistivity increases. A further increase in substrate temperature causes increase in the film resistivity. The Hall Effect measurements were carried out by using Van der Pauw's method. From the Hall Effect measurement the parameters like mobility, carrier concentration were determined and tabulated in Table 1. From the table it is seen that mobility of the CdO films increases with increase in substrate temperature attains the maximum value at 400 ^{0}C substrate temperature, for further increase in substrate temperature it decreases. The decrease of carrier concentration may be due to the decrease of native donors resulted from the enhancement of oxidation on the substrate surface [19]. Besides, the decrease of carrier concentration is also attributed to an increase in chemisorbed oxygen which acts as electron trap [20]. These results support the variation of resistivity with substrate temperature. The carrier concentration is found to increase with increase in substrate temperature. The increase of the film resistivity at 450 ^{0}C is ascribed to both the reduction in the carrier concentration and carrier mobility. The increase of electrical resistivity should be attributed to the increase of the grain boundary scattering. Variation of Carrier concentration and mobility with substrate temperature is depicted in Fig. 3.

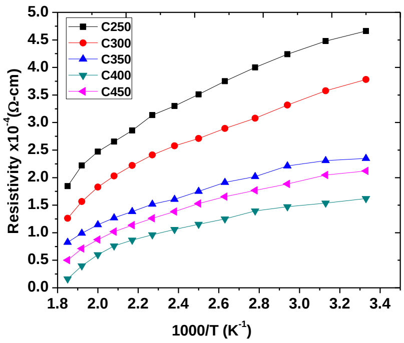

Fig.2. Variation of resistivity with substrate temperature for sprayed CdO thin films.

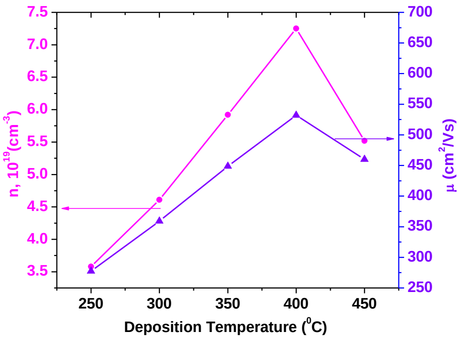

Fig.3. Variation of carrier concentration and mobility with substrate temperature for sprayed CdO thin films.

The activation energies were calculated using the relation

$$\rho = \rho_0 \exp\left(\frac{E_a}{kT}\right) \qquad (8)$$

where, ρ is the resistivity at temperature T, ρ_0 is constant, k is Boltzmann constant, T the absolute temperature and Ea is the activation energy. The activation energy represents the location of trap levels below the conduction band. From Fig. 2, one can observe two distinct activation energy regimes. This clearly indicates that different scattering mechanisms are operative in the two regimes. Fig. 3 shows the activation energies for low temperature and high temperature regions for various substrate temperatures.

4. CONCLUSIONS

In this study, the influence of the deposition temperature on the structural, surface morphology, optical and electrical properties of CdO thin films grown on amorphous glass substrates by spray pyrolysis was investigated. From the X-ray diffraction (XRD) pattern, it was observed that the CdO thin films were polycrystalline with cubic structure. The dc electrical resistivity of CdO thin films were decreased from 4.66 × 10^{-4} to 1.62 × 10^{-4} (Ω cm) with increase in deposition temperature. The electron carrier concentration (n) and mobility (μ) of CdO films deposited at 400 ^{0}C were estimated to be of the order of 7.25 × 10^{19}cm^{-3} and 532.1 cm^2 /Vs. Our results suggest the potential application of sprayed CdO thin film which can be used as a transparent conducting oxide layer for different optoelectronic and photovoltaic devices.

Table 1. Values of film thickness, lattice parameter, grain size, dislocation density, strain and energy band gap of the CdO films for different deposition temperatures.

Deposition Temperature (^{0}C)	Thick. (nm)	Lattice Para. (Å)	Grain Size (nm)	Strain (× 10^{-3})	Dislocation density (× 10^{15})	TC (111)	E_g (eV)
250	310	4.6957	21	1.178	2.225	0.348	2.46
300	360	4.6902	24	0.925	1.682	0.385	2.41
350	395	4.6874	27	0.724	1.424	0.508	2.38
400	**425**	**4.6872**	**29**	**0.672**	**1.173**	**0.612**	**2.34**
450	390	4.6905	25	1.093	1.626	0.421	2.37

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