
A.F.M. Faizullah¹ M.K.R.Khan, M.Mozibur Rahman
¹Department of Physics, University of Rajshahi, Bangladesh
E-mail: afmfaizullah07@gmail.com

Abstract

Thin films of undoped and (Al, N) doped CdO were prepared on glass substrate at substrate temperature 350 °C by spray pyrolysis method. The films are characterized by XRD, AFM, Optical and electrical properties. The films are highly poly crystalline and found to be cubic. AFM images show the surface properties of the film. It increases with increasing concentration of Al & N in the CdO samples. Optical transmittance is made at room temperature within the wavelength range 400 nm to 1100 nm. Direct band gap of undoped CdO has been measured and found to be 2.58 eV and it decreases with (Al, N) dual doping in CdO thin film up to 2.52 eV. The resistivity of undoped CdO shows the metallic behavior up to 370 K and then followed by semiconducting behavior. (Al, N) dual doping shows the carrier compensation during temperature range (355 - 375) K.

Keywords: spray pyrolysis, Dual doped, CdO, grain size.

1. Introduction

Transparent conductive oxides (TCO) are a type of nonstoichiometric semiconductor oxides of high conductivity arising from structural metal interstitials and oxygen vacancies. They have widespread use in many advanced technology applications. It is essential to investigate the means for improving the function of TCOs since they could have an enormous impact on the next generation flat panel displays and solar energy systems, and any progress in the field will require a dedicated, multidisciplinary effort [1]. It is well known that high carrier mobility is essential for TCOs with good quality electro-optical properties. From other side sometimes it is necessary to hybridize TCO in order to get some magnetic or other properties for various applications.

CdO is an n-type semiconductor with a rock-salt crystal structure (fcc) and possesses a direct band gap of 2.2 eV [2]. Its high electrical conductivity and high optical transmittance in the visible region of solar spectrum along with a moderate reflective index make it useful for various applications, photodiodes, gas sensors, etc [3, 4]. Undoped, mono-doped and dual doped CdO thin films have been prepared by various techniques such as spray pyrolysis [5], ion beam sputtering [6], sol-gel [7] etc.

In this study, undoped and (Al, N) dual doped CdO thin films have been deposited by spray pyrolysis method using cadmium acetate dehydrate, aluminium chloride and ammonium acetate as the solvent and the structural, morphological, optical and electrical properties of the films has been studied. We have chosen the spray pyrolysis method because of low cost, easy to control growth parameters, large area of fabrication.

2. Experimental Details

CdO thin films have been deposited on glass substrates at substrate temperature 350° C using spray pyrolysis technique. Cadmium acetate dehydrate [Cd(COOCH₃)₂.2H₂O] with constant concentration of 0.15 M, Aluminium chloride and Ammonium acetate were taken as the source of cadmium oxide, aluminium and nitrogen respectively. The distance between the spray nozzle and substrate was maintained at 20 cm to obtain maximum coverage throughout the film with substrate temperature 350 °C. Al and N were implanted at different ranges of 0, 1, 2, 3, and 4%. To study the structural property X-Ray Diffraction (XRD) patterns were obtained from Cu-Kα radiation (λ = 1.54056 Å) with scanning range of 20 from 30° to 65° to detect the possible peaks. Surface properties of the film were studied using AFM. Optical properties were taken by UV 1601PC double beam Spectrophotometer. Electrical properties were studied through the standard four probe technique. Fig. 1
shows the experimental setup for preparation of (Al, N) dual doped CdO thin film by using spray pyrolysis technique.

![Experimental setup diagram](image)

### 3. Results and Discussion

**Structural analysis:** XRD patterns were obtained for undoped, Al doped and (Al, N) dual doped CdO thin film and are given in Fig. 1. The diffracted peaks showed that it has polycrystalline nature with cubic crystal structure were confirmed from JCPDS file No. 05-0640. Both undoped and dual doped films show the preferential orientation along (200) plane. There are no additional peaks without CdO upon doping indicates the solubility of the dopant in the crystal structure. The grain size of the undoped film was found to be 27.15 nm which decreased to 26.873 nm for (Al, N) dual doping CdO. The values of peak angle, grain size and lattice parameter obtained from undoped and dual doped CdO films are given in Table 1.

![XRD patterns](image)

**Surface analysis:** Fig. 2 shows the AFM (Atomic force microscopy) images obtained for undoped and (Al, N) dual doped CdO films. The roughness of the surface was estimated by calculating the Root Mean Square (RMS) value. The surface of (Al, N) dual doped CdO seems to be rougher than the undoped CdO. Doping of (Al, N) in CdO thin films increases the surface roughness with the increase of (Al, N) wt. % concentration.

**Optical properties:** Fig. 3 shows the optical transmittance spectra with wavelength from 400 nm to 1100 nm of the undoped, Al doped and (Al, N) dual doped CdO thin films. It is seen from Fig. 3 that the films are
transparent in the visible region. The optical transmittance increases with increasing (Al, N) doping concentration (Fig. 3).

The optical band gap of undoped and doped CdO thin films is determined from the data of transmittance and reflectance using the relations given below. The absorption coefficient ($\alpha$) is given by

$$\alpha = \frac{1}{t} \ln \left( \frac{(1-R)^2}{T} \right)$$  \hspace{1cm} (1)

where, ‘T’ is transmittance and ‘t’ is film thickness. The relation between the absorption coefficient and the incident photon energy ($h\nu$) is given by the equation,

$$\alpha = \frac{A(h\nu - E_g)}{h\nu}$$  \hspace{1cm} (2)

where ‘A’ is a constant and ‘$E_g$’ is optical band gap.

**Table 1.** XRD data obtained for undoped and (Al, N) dual doped CdO

<table>
<thead>
<tr>
<th>Types of samples</th>
<th>$2\theta$ (deg.)</th>
<th>Grain size (nm)</th>
<th>Observed d value (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Undoped CdO</td>
<td>33.002</td>
<td>27.150</td>
<td>2.7109</td>
</tr>
<tr>
<td>3% Al doped CdO</td>
<td>33.000</td>
<td>27.578</td>
<td>2.7111</td>
</tr>
<tr>
<td>(Al, N) doped CdO</td>
<td>33.014</td>
<td>26.873</td>
<td>2.7100</td>
</tr>
</tbody>
</table>

**Electrical properties:** The variation of resistivity with temperature is shown in Fig. 5. Resistivity measurements were done from room temperature to 470 K by Van der Pauw’s method. The resistivity of undoped and Al doped CdO thin film were slightly increases with temperature which shows the semi-metallic behavior (up to 378 K) and with the further increase of temperature the resistivity falls and shows the semiconducting behavior. But for (Al, N) dual doped CdO thin film the resistivity remains almost constant with temperature from 300 K to 360 K and after that the resistivity is sharply increases (at around 365 K) due to the effect of N doping which creates holes in the system. In this case compensation of charge is occurred and this compensation stops at nearly 380 K. And resistivity decreases with the further increase of temperature and shows the semiconducting nature. Finally it remains constant from 450 K to 470 K. This may be due to the spinning of Fermi energy.

![AFM images of (a) undoped and (b) 3% Al & 1% N dual doped CdO](image-url)
Fig. 3 Optical transmittance spectra of (Al, N) dual doped CdO thin films.

Fig. 4 The plot of $(\alpha h \nu)^2$ against $h \nu$ of CdO thin films.

Fig. 5 The plot of resistivity against temperature of CdO thin films.
4. Conclusions
(Al, N) dual doped CdO films are fabricated by spray pyrolysis technique. XRD patterns shows that it has polycrystalline nature with cubic crystal structure with (2 0 0) plane as preferential orientation. The grain size of undoped CdO film is 27.15 nm which increase for Al doped CdO thin film while it decreases with dual doping of Al and N. The optical band gap values were found to decrease from 2.58 eV to 2.52 eV for direct transitions with dual doping of (Al, N) in CdO. Electrical studies revealed that for dual doping compensation of charges (electron and hole) are occurs at certain temperature range (360-380) K.

5. Acknowledgement
We are gratefully acknowledged to Bangladesh Council of Scientific and Industrial Research (BCSIR), Science Laboratory, Dhaka, Bangladesh for providing XRD facilities.

6. References