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A Comparison study of two Indium free alternative Cadmium based TCO thin films for optoelectronic applications

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Abstract - *The structural, compositional, optical and electrical properties of pure CdO thin films deposited on glass substrates by RF magnetron sputtering technique are compared with Cd_2SnO_4 thin films grown by the similar technique with the same deposition conditions. The films properties were characterized by XRD, AFM, EDS, UV-Visible spectroscopy and Hall Effect measurement. XRD analysis confirmed that Cd_2SnO_4 thin films crystallized into spinal cubic structure and CdO thin film has cubic structure with preferential orientation along (111) direction. The EDS spectra confirmed the presence of cadmium (Cd), oxygen (O) and Tin (Sn) in the appropriate films. The electrical and optical studies shows that Cd_2SnO_4 thin films have higher conductivity with similar transmittance in visible range compared to pure CdO thin films due to Sn doping effects.*

Keywords: *RF magnetron sputtering; TCO; thin films; Hall Effect; Cd_2SnO_4.*

I. INTRODUCTION

Transparent conducting oxide (TCO) materials possess unique combination of low resistivity and high optical transparency within the visible region of the electromagnetic spectrum and they are usually fabricated using thin film technologies, found many applications in display, photovoltaic, energy conservation, and lighting [1]. Current development of high-performance transparent conducting oxide films is limited with tradeoff between transparency and resistivity of the film since none of them can be improved without sacrificing the other. However, the functional properties of TCO can be tuned by optimizing the deposition conditions and to develop the super lattices by selective element doping or mixing. Pure tin oxide (SnO_2), indium oxide (In_2O_3), cadmium oxide (CdO), and zinc oxide (ZnO) are the well known binary compounds of TCO materials [2]. Doping these oxides resulted in improved electrical conductivity without degrading their optical transmission. Al doped ZnO (AZO), tin doped In_2O_3, (ITO) and antimony or fluorine doped SnO_2 (ATO and FTO), are among the most utilized ternary TCO thin films in modern technology [3]. In particular, ITO is used extensively in many applications. However, it is not clear that ITO will meet the stringent performance and cost requirements for next-generation opto-electronic technologies. Among the post-transition metal oxides, SnO_2 and CdO are the second and third member of the series followed by In_2O_3 and they are most significant representatives of n-type degenerate TCO material. In the present work, we focus on design, growth and characterization and comparing the properties of indium free cadmium and tin based transparent conducting oxide thin films.

II. EXPERIMENTAL PROCEDURE

CdO and Cd_2SnO_4 thin films were deposited on glass substrates by using RF magnetron sputtering technique (HINDHIVAC; Planar Magnetron RF/DC Sputtering Unit Model-12″ MSPT). Before deposition, the substrates were cleaned by using chromic acid followed by acetone. The Cd_2SnO_4 target was prepared by using commercially available pure CdO and SnO_2 powders (Merck) and the required amount of source material in the atomic weight ratio of 2:1 (CdO: SnO_2) was preheated at 800^0C for 8 hour in ambient atmosphere. Then, the powder was mixed with PVA and compacted in to the pellet of dimensions 5mm thickness and 5cm diameter respectively by using hydraulic pressure. The prepared pellet was sintered at 700^0C for 7 hours by using box furnace which could be used as the target material. The similar procedure was followed to prepare pure CdO target. The prepared target was loaded into the deposition chamber and the chamber was evacuated to the base pressure of 5×10^{-6} mbar using rotary and diffusion pumps. The deposition parameters such as substrate to target distance, working pressure, deposition temperature, deposition time and RF power for the deposition of CdO and Cd_2SnO_4 thin films were maintained at 6 cm, 3×10^{-3} mbar, RT, 30 min and 150 W respectively.

III. RESULTS AND DISCUSSION

The thicknesses of deposited thin films were measured by using stylus profilometer which are found to be in the range of 1.1µm. Figure 1 shows the X-ray diffraction (XRD) patterns of CdO and Cd_2SnO_4 thin films. This pattern revealed that the CdO thin film exhibited a preferential orientation along (111) direction at an angle of ~33˚ and three small intensity peaks were also appeared which corresponds to (200), (311), (222) planes of CdO. The observed peaks of CdO are well matched with standard joint committee powder diffraction system (JCPDS) card no.050640 which indicates that the deposited film is polycrystalline in nature with face centered cubic structure. On the other hand, Cd_2SnO_4 thin film is crystallized in spinal cubic structure, Fd3m (227) space group and the atoms are arranged in face centered lattice (FCC) arrangement with standard lattice parameter value a=b=c= 9.174Å which is confirmed by the standard JCPDS data, (No.80-1469) [4].

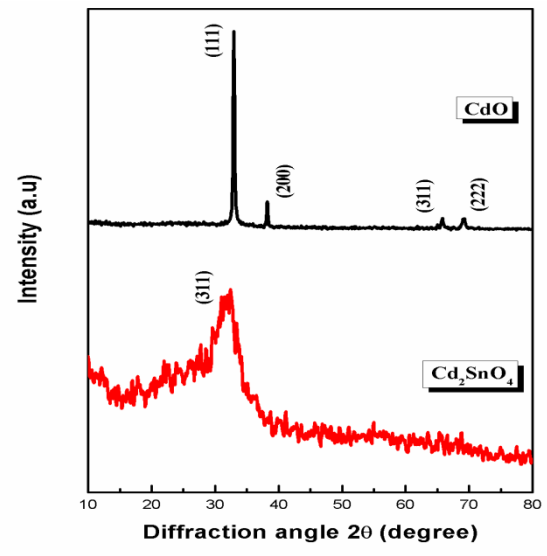

Figure 1. XRD patterns of CdO and Cd_2SnO_4 thin films

The compositional analysis of the prepared thin films is examined by energy dispersive X-ray spectroscopy (EDS). Figure 2a & 2b shows the EDS spectrum of CdO and Cd_2SnO_4 thin films respectively. The spectrum confirms the presence of cadmium (Cd) and oxygen (O) in the CdO thin films. In addition, Tin (Sn) also presents in the Cd_2SnO_4 thin films. Apart from the major constituent elements in the elemental mapping, another element silicon is also present which may be due to the glass used as substrate. Additionally, no other elemental peaks are appeared in EDS spectrum, which indicates the high purity of prepared thin films.

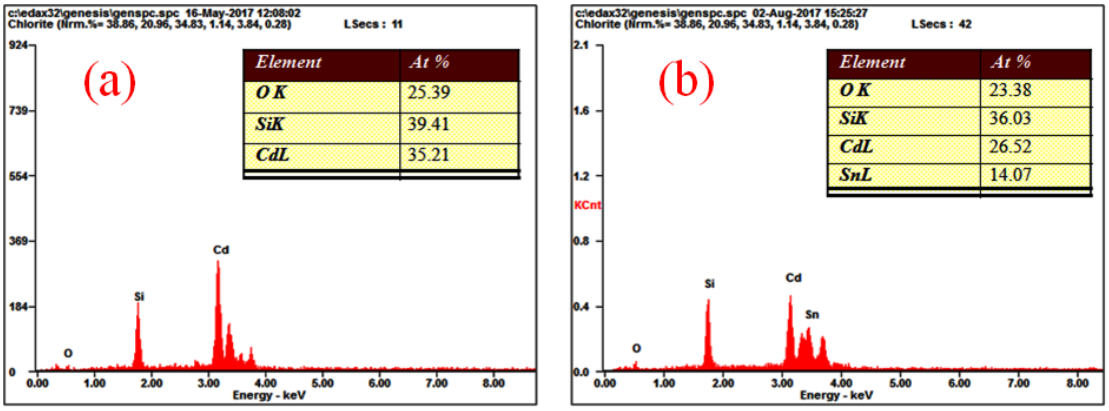

Figure 2. (a) & (b) EDS spectrum of deposited CdO and Cd_2SnO_4 thin films

The transmission spectra of pure CdO and Cd_2SnO_4 thin films are shown in figure 3a. It is seen from transmission spectra that the average transmittance in the range of 400-1100 nm is 85% for Cd_2SnO_4 thin films and 65% for pure CdO thin films which makes them more suitable for TCO and IR window applications. It is interesting to note that the absorption edge of Cd_2SnO_4 thin film is blue shifted of about 60-80 nm, which means that the optical transparent region is obviously widened. The blue shift of the absorption edge of Cd_2SnO_4 thin films can be attributed to the increase in carrier concentration, a phenomenon known as the Burstein - Moss (BM) effect [5]. In addition, transmittance of the films having an interference pattern due to generation of the wave fronts between the air and substrate interface which depends on the temperature during deposition and thickness of films. Further, the variation of refractive index of films and substrate may also be the reason for generation of the wave fronts. The band gap of the deposited thin films is evaluated by using Tauc's plot method shown in figure 3b. It is seen from the figure that the evaluated band gap values increased from 2.59 eV (CdO) to 3.01 eV (Cd_2SnO_4). This increase in energy band gap (blue shift) is attributed to the replacement of Sn ions in to CdO lattice which can also explained by BM effect.

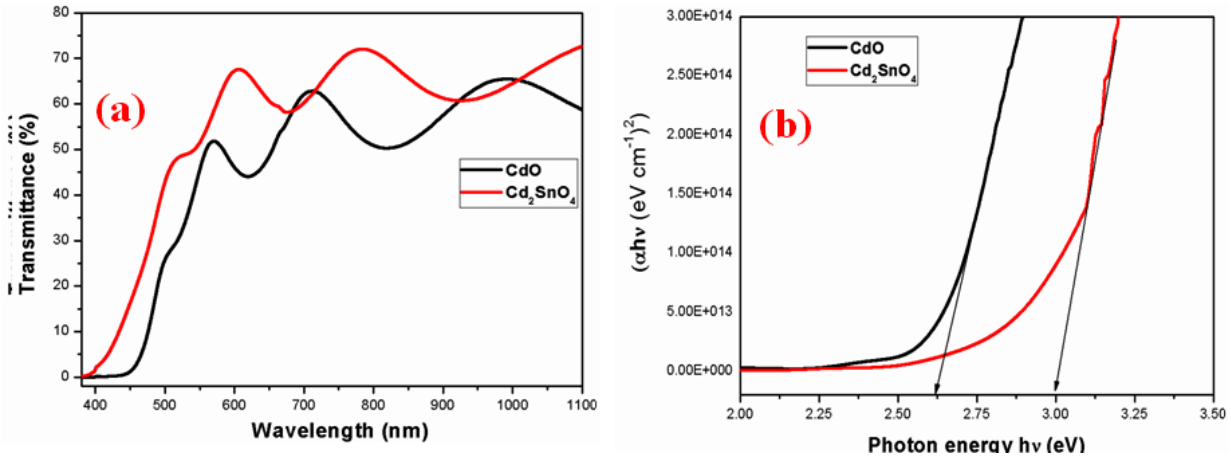

Figure 3. (a) Transmittance spectra of CdO and Cd_2SnO_4 thin films. (b) Tauc's plots of CdO and Cd_2SnO_4 thin films

The electrical properties of deposited CdO and Cd_2SnO_4 thin films were carried out by using Hall measurements. The negative sign of Hall coefficient indicates that the deposited thin films are n-type conducting nature. The comparison in electrical properties such as resistivity (ρ), conductivity (σ), carrier concentration (n_e) and carrier transport mobility (μ_e) of deposited CdO and Cd_2SnO_4 thin films are presented in table 1. The Cd_2SnO_4 thin films have very low resistivity value in the order of 7.448×10^{-4} Ω. cm than that of pure CdO thin films. It is noted that the variations of electrical resistivity and conductivity of deposited thin films are strongly related to the variations of carrier concentration and mobility of the thin films. The high value of carrier concentration in Cd_2SnO_4 thin films may be due to the creation of oxygen vacancies and metal interstitials that provide more donor states [6]. The mobility of the films shows decreasing trend with increasing carrier concentration which is in good agreement with the theory of electron scattering by ionized impurities for n type degenerate semiconductors.

Table 1. Electrical properties of CdO and Cd_2SnO_4 thin films

Electrical properties	*CdO thin films*	*Cd_2SnO_4 thin films*
Carrier concentration(cm^{-3})	2.343×10^{19}	4.493×10^{20}
Mobility (cm^2/Vs)	158.5	18.65
Resistivity (Ω. cm)	1.681×10^{-3}	7.448×10^{-4}
Conductivity (Ω^{-1}. cm^{-1})	595	1343

IV. CONCLUSION

In the present work, pure CdO and Cd_2SnO_4 thin films were deposited on glass substrates by RF sputtering technique at ambient temperature with same deposition conditions and the structural, compositional, optical and electrical properties of deposited films were investigated and compared to one another. XRD pattern revealed CdO thin films have cubic structure and Cd_2SnO_4 thin films were crystallized in spinal structure. The compositional analysis (EDS) confirmed the presence of Cd, Sn and O on the surface and proved the purity of deposited thin films. Cd_2SnO_4 thin films have high carrier concentration of 4.493×10^{20} cm^{-3} with lower resistivity of 7.448×10^{-4} Ω. cm compared to pure CdO thin films and this is due to the replacement of Sn ions in the CdO lattice. Cd_2SnO_4 thin films showed high optical transparency with shift of band gap to higher energies. Consequently, for their improved optical and electrical properties, Cd_2SnO_4 thin film could be an excellent candidate for future optoelectronic devices.

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