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Effect of Doping on Thermoelectric Property of Spray Deposited Nanostructured CdO Thin Films

D. Mahesh Reddy, B.G. Jeyaprakash and R. John Bosco Balaguru
School of Electrical and Electronics Engineering, Centre for Nanotechnology and Advanced Biomaterials, SASTRA University, Thanjavur-613-041, Tamil Nadu, India

Abstract: Recently nanostructured based metal oxide thin films were preferred for thermoelectric applications due to its lower thermal conductivity than in bulk form. Cadmium oxide in bulk form has lower thermal conductivity and much work on thermoelectric studies in thin film is not reported. Hence in the present study, undoped, Zn and Mn doped CdO thin films were prepared on glass substrate using homemade spray pyrolysis technique and its structural, electrical, surface morphology and thermoelectric properties were analyzed and reported.

Key words: Thin film, thermoelectric, metal oxide, spray pyrolysis

INTRODUCTION

Power generation and cooling are the major applications of thermoelectric materials. Figure of merit (ZT) defines the performance of thermoelectric material and is given as $ZT = \alpha^2\sigma/\kappa$, where κ, α and σ are the thermal conductivity, thermoelectric power (Seebeck Coefficient) and electrical conductivity, respectively. Materials with large σ and low κ have high figure of merit. Bi_2Te_3 and Bi_2Se_3 are some of the compounds at room temperature with high ZT value in between 0.8 and 1.0. Many attempts have been made to improve the performance of thermoelectric materials (Venkatasubramanian *et al*., 2001). Lead telluride (PbTe) doped with different dopants has achieved ZT approximately 1.4-1.8 at 800 K (Heremans *et al*., 2008). However, telluride based compounds are toxic and less abundant in earth crust. Also at high temperature it gets oxidised. Hence, in recent years, due to good stability at high temperatures, low thermal conductivity, ease of deposition and low toxicity, metal oxide semiconducting thin film are recognized as promising candidates for power generation (Harman *et al*., 2002). However Figure of merit in oxides is still low (~0.34 at 1,000 K) (Venkatasubramanian *et al*., 2001), but their thermal stability as compared with bismuth compounds, gave a priority to use them as high temperature thermoelectric material (Meenakshisundaram and Rajagopalan, 2009). Also, nanostructured materials were preferred for thermoelectric application because of their low thermal conductivity (Li and Lin, 2004). CdO is an n-type semiconductor, which has a band gap of 2.20 eV and with low resistivity exhibit good thermoelectric property (Andrews, 1947). In this paper, the thermoelectric property of undoped, Mn and Zn doped CdO thin films prepared on glass substrate by spray pyrolysis method were studied. Also variation in structural, optical and surface morphology is also studied.

MATERIALS AND METHODS

Cadmium oxide films were deposited by a homemade spray pyrolysis system (Jeyaprakash *et al*., 2011) using aqueous cadmium acetate solutions. A 0.05 M of 50 mL aqueous cadmium acetate dihydrate solution was sprayed as fine mist on glass substrate kept at 250°C. Table 1 shows the optimized preparation condition to obtain uniform deposition. Dopant concentrations were fixed to 0.002 M. Before deposition, glass substrate of 7.5×3.5 0.13 cm was cleaned ultrasonically. Structural studies were carried out using PANalytical X-ray diffractometer (Model D/MAX ULTIMA III). Surface micrograph was obtained using HITACHI make (Model S-3000H) scanning electron microscope. Film thickness was measured using Stylus Profilometer (Mitutoyo Stylus Profilometer SJ-301). The optical studies were carried out using ELICO SL 159 UV-Vis spectrophotometer.

Table 1: Optimised preparation parameters

Parameter	Values
Volume of the solution (mL)	50
Pressure of carrier gas (kg cm^{-2})	2
Rate of deposition (mL min^{-1})	1
Distance between nozzle and heater (cm)	25
Substrate temperature (°C)	250

Corresponding Author: B.G. Jeyaprakash, School of Electrical and Electronics Engineering, Centre for Nanotechnology and Advanced Biomaterials, SASTRA University, Thanjavur-613 041, Tamil Nadu, India

RESULTS AND DISCUSSION

Structural characterization: X- ray diffraction studies for undoped and doped CdO were shown in Fig. 1. It indicates that the CdO thin films have polycrystalline nature and cubic structure. Peaks at different 2θ values are 38.52°, 55.5°, 66.2° and 69.39° which represent the orientations (111), (200), (220) planes of FCC CdO [JCPDS: 05-0640]. X-ray diffraction pattern shows the undoped, Zn and Mn doped CdO has a lattice constant of 4.688 Å, 4.697 Å and 4.693 Å, respectively.

Scanning electron microscopy: Figure 2 Shows scanning electron micrograph of undoped, Zn and Mn doped CdO thin films at two different resolutions (1 μm and 100 nm). It shows the films were deposited uniformly with grain size varying from 20-30 nm for all the films. Grains of Mn doped CdO are arranged closely to each other and this gives better conductivity and thermoelectric property.

Electrical properties: Figure 3 Illustrates the variation of resistivity verse temperature of undoped, Zn and Mn doped CdO thin film. It indicates that Mn doped CdO film

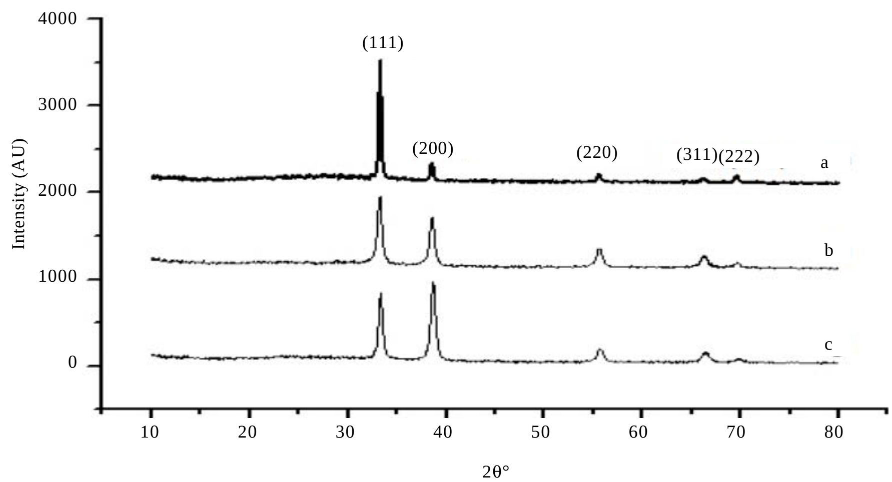

Fig. 1: XRD patterns of a: Undoped b: Mn doped CdO and c: Zn doped CdO thin films

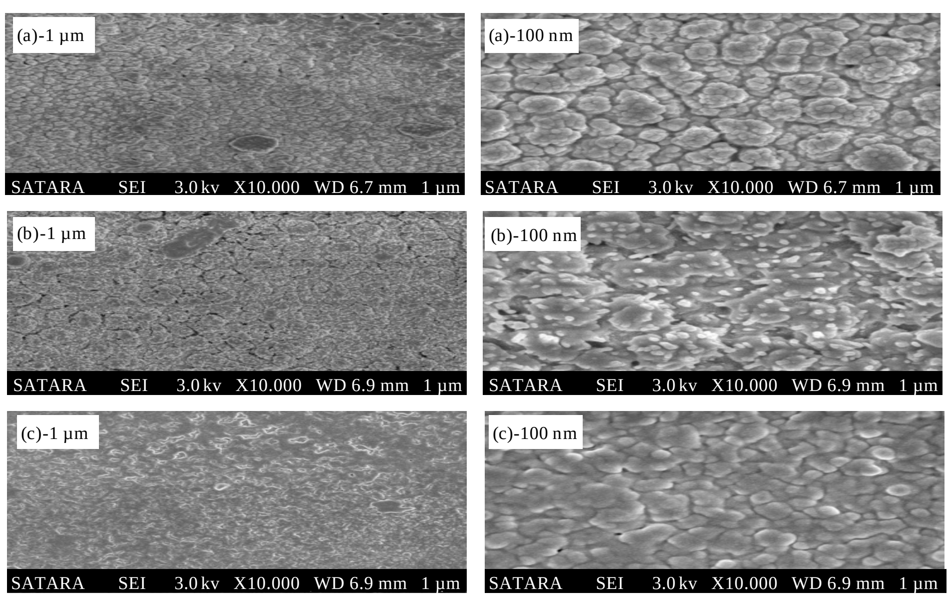

Fig. 2(a-c): (a) CdO thin film at 1 μm and 100 nm, (b) Zn doped CdO at 1 μm and 100 nm, (c) Mn doped CdO at 1 μm and 100 nm

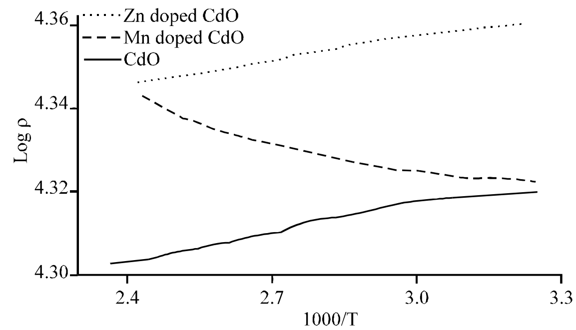

Fig. 3: Log ρ vs. 1000/T plot of undoped, Zn and Mn doped CdO thin film

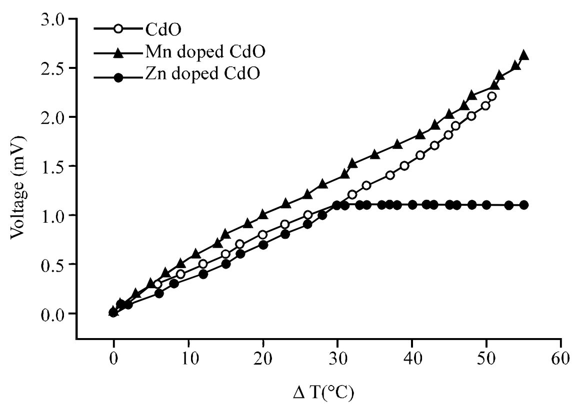

Fig. 4: Plot of thermo emf of undoped Mn and Zn doped CdO thin films

has metallic behavior with increase in temperature while undoped and Zn doped CdO shows semiconducting nature. The activation energy obtained from the plot was 0.0087, 0.0055 and 0.00988 for undoped, Zn and Mn doped CdO film, respectively.

Thermoelectric studies: Thermoelectric \studied were carried out in homemade glass test chamber. Initially the chamber was evacuated to 10^{-3} mbar. To measure thermo emf, one end of the film was heated locally using 100 W compact heater and the other end of film was maintained to room temperature by circulating cold water. The variation of thermo emf with different of temperature between two end of all the film is shown in Fig. 4. From the Fig. 4, it shows that the thermo emf is significantly high for Mn doped CdO thin film rather than undoped and Zn doped CdO film. This may be attributed to increases in the phonon scattering thereby thermal conductivity of the film decreases prominently. Also Mn doped CdO film shows a linear increase in thermo emf with respect to the temperature. Above 30°C, the thermo emf was saturated for Zn doped CdO film and indicates that Zn doping in CdO film increase the thermal conductivity. The Seebeck coefficient calculated from the plot were 36.58, 34.71 and 42.74 μV/K for undoped, Zn and Mn doped CdO thin film, respectively.

CONCLUSION

Undoped, Zn and Mn doped CdO thin films were prepared on glass substrate by spray pyrolysis technique. Structural studies revealed that all the films were polycrystalline in nature and grain size range from 20 to 30 nm. Visual inspection of Scanning electron micrograph reveals that the film is uniform and crystallite size varies with respect to change in dopants. Seebeck coefficient for Mn doped CdO film found to be high compared with undoped and Zn doped CdO thin film. Further research can be done to convert waste heat from the vehicle exhaust to thermo voltage and can detect harmful gases ejected from the vehicle. These sensors can also be used in medical, automobile industries and homemade appliances.

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