

Electrical properties of $(\text{PbO})_{1-x}(\text{CdO})_x$ thin films fabricated by spray pyrolysis technique

Nahida B. Hasan and Mohammed Ahmed Mohammed

Department of Physics, College of Science, University of Babylon, Iraq

ABSTRACT

Nano particles of Mixed $(\text{PbO})_{1-x}(\text{CdO})_x$ thin films were prepared by spray pyrolysis technique at a substrate temperature of 400°C . The films deposited were 160 nm thickness. It has been making electrical measurements such as the Hall effect and D.C conductivity for all films. The results showed that (PbO) has conductivity about of $[209 \times 10^{-5} (\Omega \cdot \text{cm})^{-1}]$ at room temperature, and this conductivity increased with increasing of CdO Vol.%, as well as the results showed throughout the study that all films have low activation energy and this energy increase with increasing of Vol.% of (x) .

Keywords: Spray pyrolysis, Lead oxide, cadmium oxide, electrical properties, activation energy, Hall effect, D. C. conductivity, Thin films.

INTRODUCTION

PbO and CdO are an n-type semiconductor materials. Because of its good adsorptive properties and chemical stability, it can be deposited onto glass, ceramics, oxides, and substrate materials of other types. It has a high melting point and good transmission, and does not easily react with oxygen and water vapor in the air, so it has a high specific volume and good cycling performance. In addition, PbO and CdO thin films are also used for film resistors, electric conversion films, heat reflective mirrors, semiconductor–insulator–semiconductor (SIS) heterojunction structures, and surface protection layers of glass. At present, its most common application is as the anode material of solar cells [1,2]. Varieties of methods like dc reactive sputtering, chemical bath deposition [3], activated reactive evaporation [4], solution growth [5], thermal oxidation [6], sol–gel [7], and spray pyrolysis [8,9] have been reported in the preparation of CdO and PbO thin films. The electro optical properties of CdO make this material very convenient as a solar cell material [1]. In attempts to improve the properties of PbO, it is being tried out to mix with other oxides. Recently, Hosono *et al.* [10] reported amorphous semiconductor $2\text{CdO} \cdot \text{PbO}_x$ thin films with a novel information about the carrier generation through the formation of oxygen vacancies. In the present work, we bring out a detailed investigation electrical properties on these mixed films with $0 \leq x \leq 1$ prepared from spray pyrolytic decomposition of aqueous solutions of lead and cadmium acetates at 400°C .

MATERIALS AND METHODS

A simple homemade spray pyrolysis experimental setup was employed to prepare $(\text{PbO})_{1-x}(\text{CdO})_x$ mixed thin films on glass substrates ($35 \times 25 \times 1.35 \text{ mm}^3$) at a substrate temperature of 400°C . The difference in CdO vol.% (x) was achieved by mixing the aqueous solutions of 0.1 M of lead and cadmium acetates to pre-determined volume ratio. The value of (x) in the solution was varied from 0.00 to 1.00 ($x=0, 0.2, 0.4, 0.6, 0.8, 1$). The mixed solutions which were then diluted with water formed the final spray solution and a total volume of 25 ml was used in each

deposition. The deposition parameters such as spray nozzle-substrate distance (30 cm), spray time (4 s) and the spray interval (1 min) were kept constant. The carrier gas (filtered compressed air) flow rate was maintained at 6 l/min at a pressure of $6.5 \times 10^4 \text{ Nm}^{-2}$. The Hall effect was carried out according to the electrical circuit shown in Figure (1), which contains a D.C. power supply with (0 – 40) volt and two digital electrometers (HMS-3000) to measure the current and voltage. The electrical conductivity has been measured as a function of temperature for films in the range (R.T – 200) °C by using the electrical circuit. The measurements have been done using sensitive digital electrometer type Keithley (616) and electrical oven as show in Figure(2). The resistivity (ρ) of the films is calculated by using the following equation:

$$\rho = \frac{R \times A}{L} \dots\dots\dots (1)$$

Where R is the sample resistance, A is the cross section area of the films and L is the distance between the electrodes. The conductivity of the films was determined from the relation:

$$\sigma_{D.C} = \frac{1}{\rho} \dots\dots\dots (2)$$

The activation energies could be calculated from the plot of $\ln \sigma$ versus $1000/T$ according to equation (3).

$$\ln \sigma_{D.C} = \ln \sigma_0 - \frac{\Delta E_a}{kT} \dots\dots\dots (3)$$

Since : $\ln \sigma_0 - \frac{\Delta E_a}{k}$ is numerical constant .

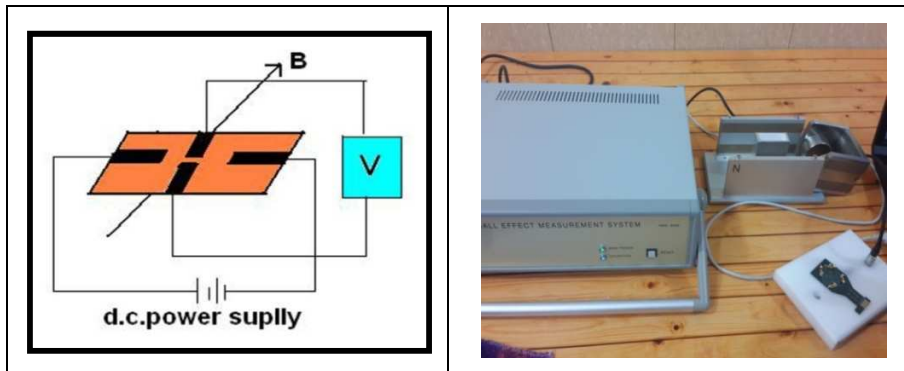


Figure 1: set-up and Photograph illustrates the Hall Effect

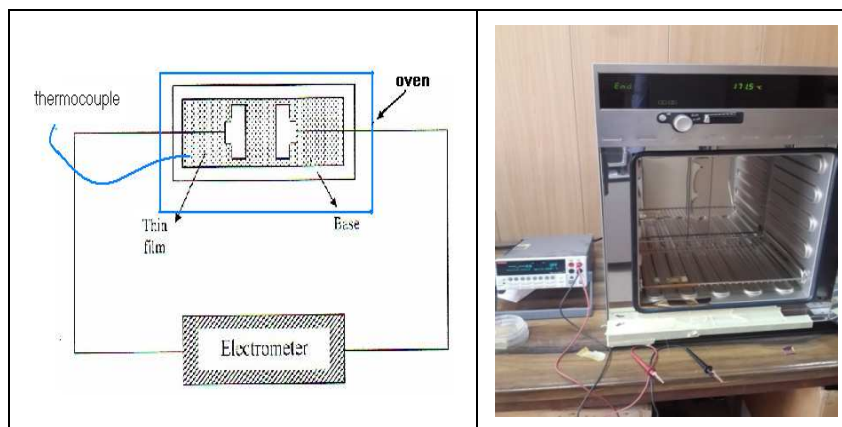


Figure 2: set-up and Photograph illustrates the D.C Electrical Conductivity

RESULTS AND DISCUSSION

The electrical properties of prepared mixed (PbO)_{1-x}(CdO)_x thin films were investigated as follows:

1. Hall Effect Measurements

Hall measurements of the deposited samples from x=0 to x=1 were shown n-type conductivity, which give negative values of R_H. These results followed the (PbO)_{1-x}(CdO)_x behavior as illustrated [11]. The type of charge carriers, concentration (n_H), Hall mobility (μ_H), conductivity (σ) and resistivity (ρ) have been estimated from Hall measurements in the table (1). From this table the conductivity (σ) of the solid solutions strongly depends on the composition.

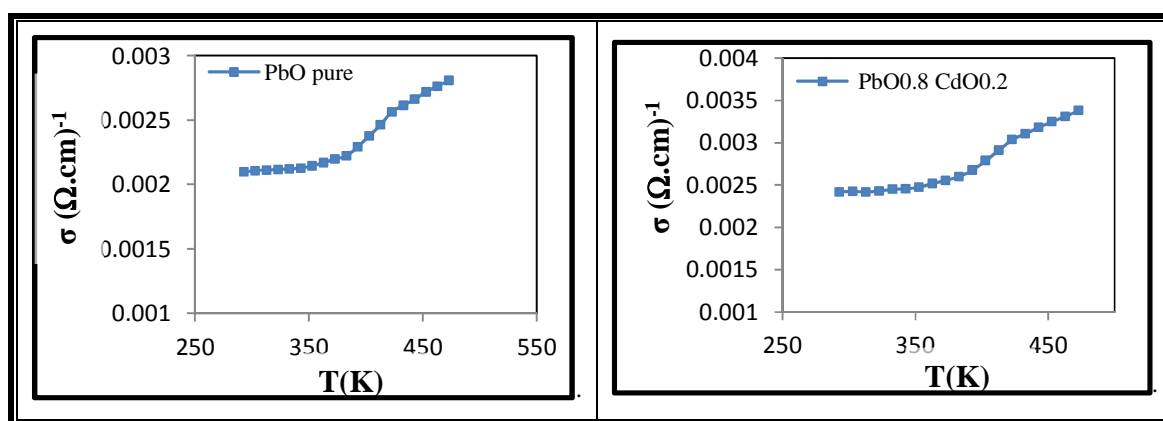
From table (1) the value of σ was increased with increasing CdO Vol.%. That the structure of thin film was changed to decrease the grain size and the decrease barrier potential of the internal grains, which make capture to the change carriers in the grain bounding and then increase the scattering. These results are in agreement which funded by [2]. Mobility decrease with increasing of CdO Vol.%. is due to the increasing of the carrier concentration.

Table 1: Hall parameters for (PbO)_{1-x}(CdO)_x films at different Vol.% of (x).

Sample	R _H (cm ³ /C)	n _H (1/cm ³)	σ _{R,T} (Ω.cm) ⁻¹	ρ (Ω.cm)	μ _H (cm ² /V.s)
PbO _{pure}	-1.057×10 ⁷	-5.903×10 ¹¹	1.33×10 ⁻⁷	7.517×10 ⁶	1.407×10 ⁻²
PbO _{0.8} CdO _{0.2}	-8.56×10 ⁶	-7.293×10 ¹¹	5.176×10 ⁻⁶	1.932×10 ⁵	4.430×10 ¹
PbO _{0.6} CdO _{0.4}	-7.189×10 ⁶	-8.683×10 ¹¹	7.766×10 ⁻⁵	1.288×10 ⁶	5.583
PbO _{0.4} CdO _{0.6}	-3.219×10 ¹	-1.939×10 ¹⁷	3.443×10 ⁻¹	2.905	1.108×10 ⁻¹
PbO _{0.2} CdO _{0.8}	-1.015	-6.151×10 ¹⁸	1.279	7.817×10 ⁻¹	1.298×10 ⁻²
CdO _{pure}	-4.941×10 ⁻⁶	-1.263×10 ²⁴	6.973×10 ²	1.434×10 ⁻³	3.446×10 ⁻³

2.The D.C Conductivity and Activation Energy

Figures. (3) show the variation of D.C conductivity (σ) vs. temperature (T), this figure show that (σ) at x=0 increased with T, this is seems to be a normal behavior as one of semiconductor properties, due to the increasing carrier concentration with temperature. When CdO added to the solution (x=0.2,0.4,...,1), σ_{D,C} was increased with increasing T. The activation energy (E_a) obtained for these films is given in the table (2), which calculated from the slope of lnσ vs. 1000/T according to eq.(3), which shown in Figures.(4). From these figures the films having two activation energy depend on x value. This means there are two mechanisms for conductivity. The activation energy in the low temperature depends on the ionization impurity and at high temperature depends on the generation of electron-hole pairs. Table(2) shows that the value of Ea₁ is smaller than values of Ea₂. This indicates that the conductivity depends on the temperature where σ ∝ T^{3/2} [12].



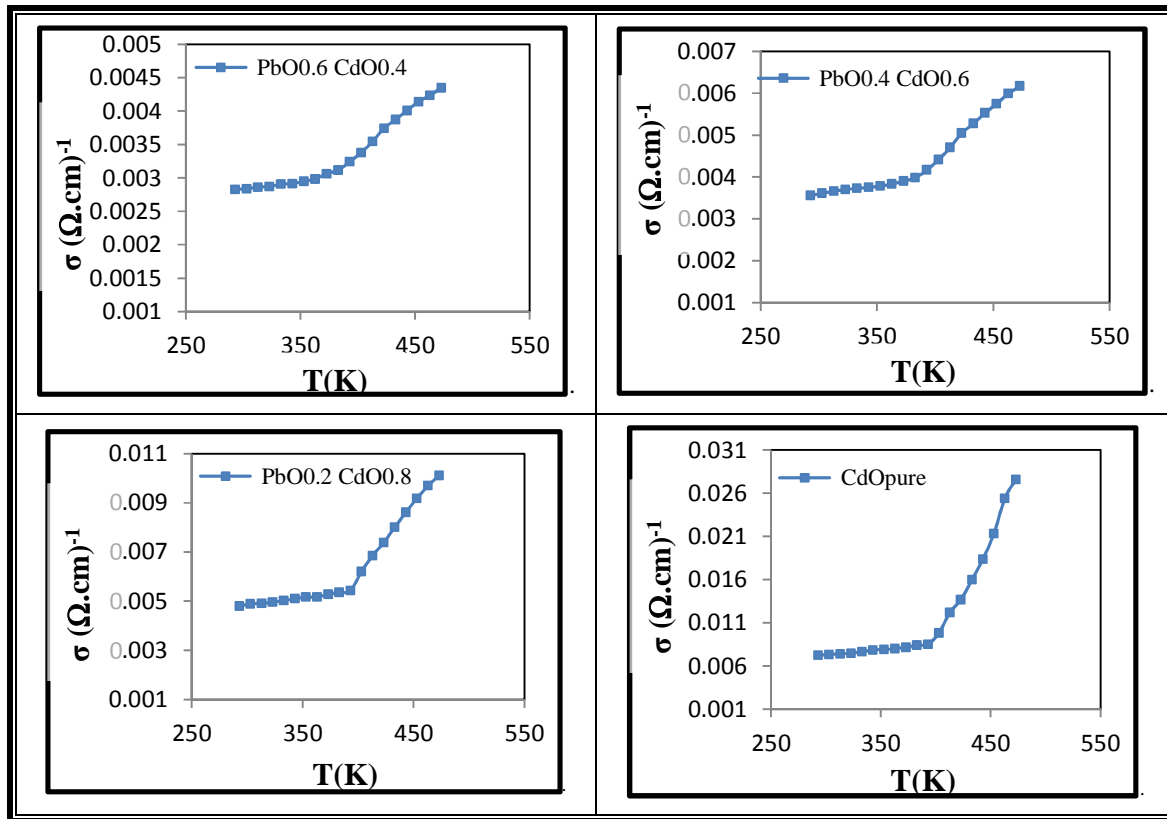
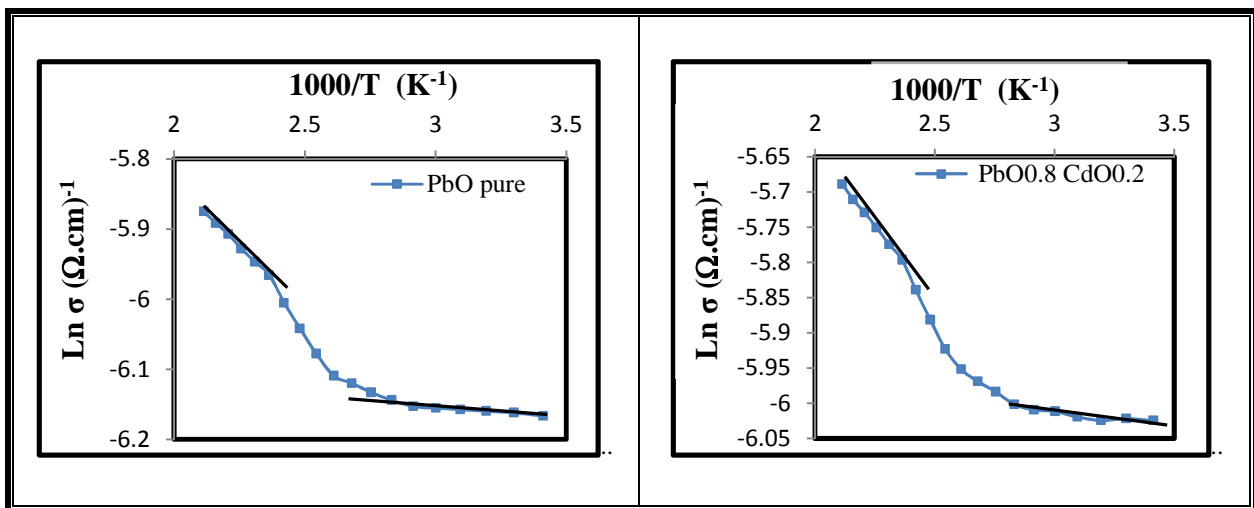


Figure 3 : Variation of σ_{dc} versus temperature for $(\text{PbO})_{1-x}(\text{CdO})_x$ films at different Vol.% of (x)



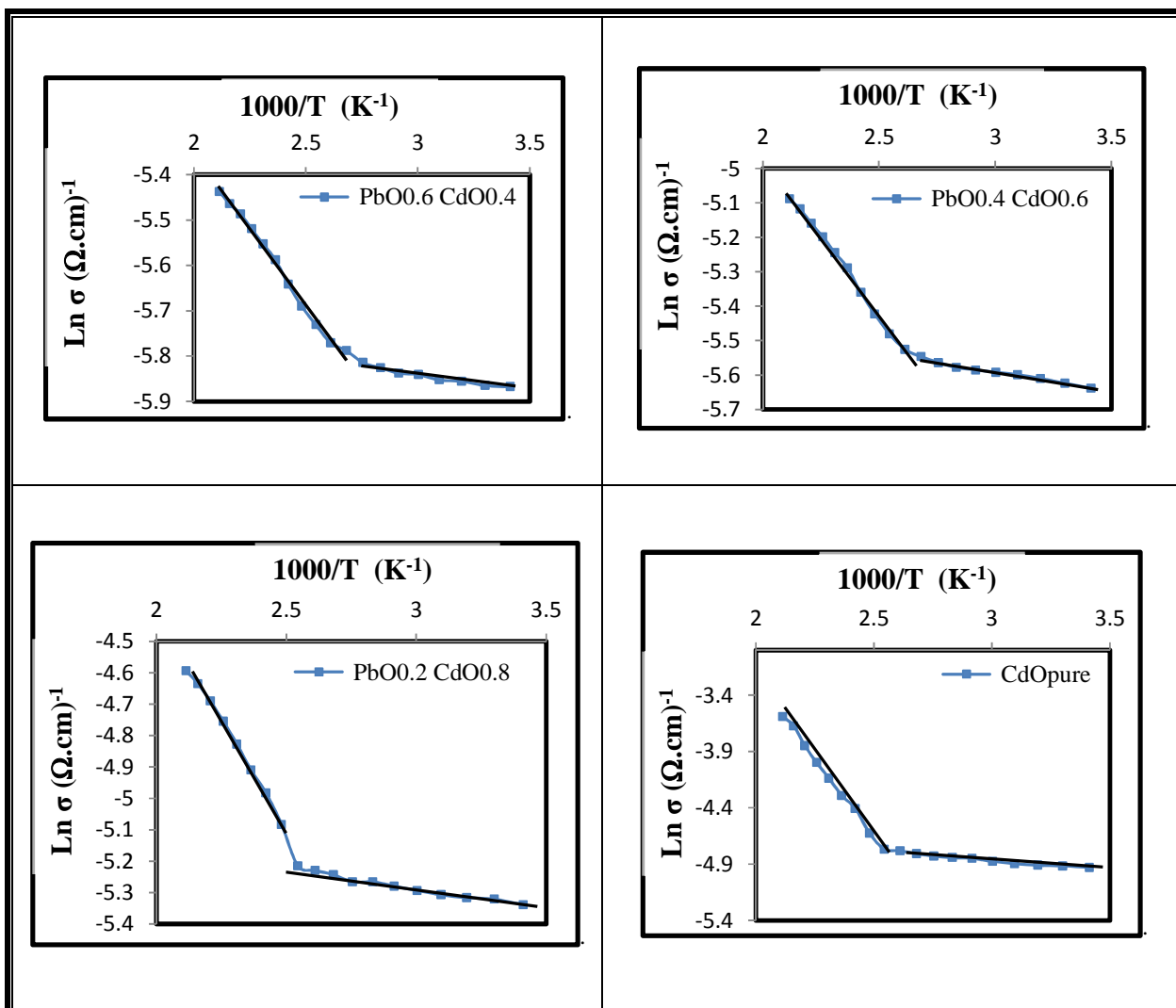


Figure 4 : Lnσ versus 1000/T for (PbO)_{1-x}(CdO)_x films at different Vol.% of (x)

Table 2: D.C. conductivity parameters for (PbO)_{1-x}(CdO)_x films at at different Vol.% of (x)

Sample	Ea ₁ (eV)	Temp.Range (K)	Ea ₂ (eV)	Temp.Range (K)	σ _{R.T} × 10 ⁻⁵ (Ω.cm) ⁻¹
PbO _{pure}	0.00223	(293-363)	0.03156	(423-473)	209
PbO _{0.8} CdO _{0.2}	0.00274	(293-353)	0.04035	(403-473)	241
PbO _{0.6} CdO _{0.4}	0.00685	(293-363)	0.05985	(403-473)	282
PbO _{0.4} CdO _{0.6}	0.00984	(293-373)	0.07918	(383-473)	355
PbO _{0.2} CdO _{0.8}	0.01654	(293-393)	0.11577	(403-473)	480
CdO _{pure}	0.01552	(293-393)	0.24001	(403-473)	724

CONCLUSION

(PbO)_{1-x}(CdO)_x mixed films were deposited on glass substrates at 400 °C and studied as a function of CdO vol.%, x(0 ≤ x ≤ 1). All thin films had ohmic behavior. Mixed (PbO)_{1-x}(CdO)_x thin films had n-type, low conductivity. Also, two activation energy. Mobility decrease with increasing of CdO Vol.%. σ_{D.C} was increased with increasing of CdO Vol.%.

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