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[ISSN: 2412 -1150 ](Online)

**Optical Characterization of  $(\text{PbO})_{1-x}(\text{CdO})_x$  Deposited By Spray Pyrolysis Technique****Nahida B. Hasan<sup>1</sup>, Mohammed Ahmed Mohammed<sup>1</sup>**<sup>1</sup>Department of Physics, College of Science, University of Babylon, Iraq  
[iraq\\_moh\\_iraq@yahoo.com](mailto:iraq_moh_iraq@yahoo.com)**ABSTRACT**

Mixed  $(\text{PbO})_{1-x}(\text{CdO})_x$  thin films prepared by spray pyrolysis technique at a substrate temperature of 400°C. The films deposited were 160 nm thickness. The optical parameters of the prepared films as absorbance, absorption coefficient, optical energy gap, refractive index and extinction coefficient were found.

**Keywords:** Spray pyrolysis, Lead oxide, cadmium oxide, optical properties, Thin film.

**1. 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 improvise 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 optical 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.

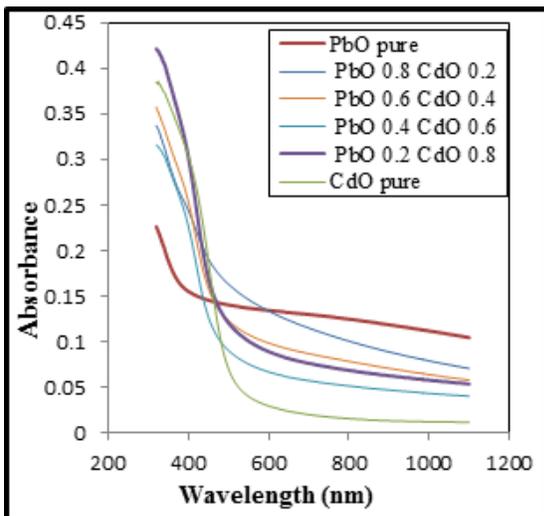
**2. EXPERIMENTAL DETAILS**

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  $6.5 \times 10^4 \text{ Nm}^{-2}$ . Optical transmittance spectra in the wavelength ranging 300–1100 nm were recorded using UV Visible spectrometer (UV-IR 1800 Spectrophotometer).

### 3. RESULTS AND DISCUSSION

#### A. Absorbance Spectrum

Optical transmission and absorption spectra depend on the chemical composition, crystal structure, energy of the incident photon, film thickness, and film surface morphology. The absorption spectra of the mixed  $(\text{PbO})_{1-x}(\text{CdO})_x$  films recorded in the wavelength from 300 to 1100 nm are compared as a function of CdO vol.% (x) in Figure (1). The average absorbance for the wavelength of visible region (500–850 nm) varies between 14–12 %. For x=0 (PbO pure) . For x=0.2 (PbO0.8 CdO0.2) the absorbance varies between 16-9 % , for x = 0.4 and 0.6 the absorbance which decreases from (12-7.4)% to ( 9-4)% respectively. This decrease was caused by the increasing crystallinity of PbO as has been confirmed in structural studies. While for x = 0.8 the absorbance which increases for 21-12% , this may probably be due to increasing CdO content. From Fig.(4.10) we note that the absorbance of (CdO pure) varies between 7-1%. From the above note that the increasing CdO vol.% (x) leads to increasing in absorbance in the visible region, this is consistent with researcher R.Kumaravel et.al [2]. The absorbance at NIR region (900-1100) nm has varied between 1-14 %.



**Figure 1:** Absorbance spectrum as a function of wavelength for  $(\text{PbO})_{1-x}(\text{CdO})_x$  films at different Vol.% of (x).

#### B. Absorption Coefficients

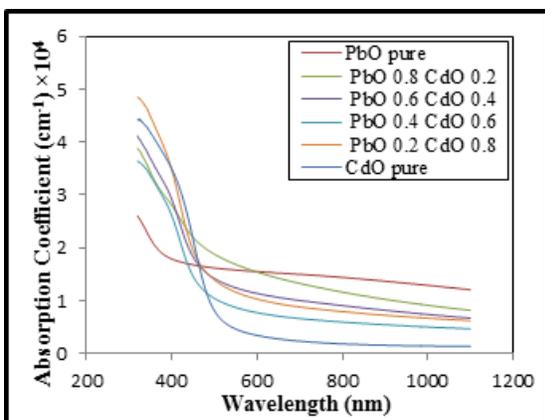
The absorption coefficient ( $\alpha$ ) for the prepared thin films was calculated from equation [2] :

$$\alpha = \frac{2.303 \times A}{t} \dots\dots\dots (1)$$

where:

t: is the thickness of thin film in (cm). , A: absorption.

The values of  $\alpha$  for all thin films are found to be greater than  $10^4 \text{ cm}^{-1}$  in the visible region , which means that the films have a direct optical energy gap [11]. The variation of the absorption coefficient ( $\alpha$ ) of  $(\text{PbO})_{1-x}(\text{CdO})_x$  films is shown in Figure (2) as a function of wavelength. The values of  $\alpha$  for all thin films are found in table (1) at different wavelength, we note that the absorption coefficient at 300 nm which increase with increasing CdO vol.%. The average value in the wavelength range from 500nm to 850 nm shows that the  $\alpha$  decreases with increasing CdO vol.% until x=0.6 but then increasing at x=0.8 . Also the sharp edge means the photon is absorbed directly from the electron , but the broadening of the absorption edge could be due to the grain boundary discontinuity effect and the lack of stoichiometry generally observed in crystalline material or may be due to another absorption mechanism because the films have multiple phase structures as mentioned in the XRD discussion. Also, it may be due to the localized tail states in the band gap. The sub -Band Gap (SBG) absorption in the thin films is believed to be related to the presence of a secondary phase at grain boundaries [12]. The lower value of absorption coefficient in the NIR region (900–1100 nm) attributes to the higher NIR transmittance of these films. The values of absorption coefficient are in agreement with those of  $2\text{CdOPbO}_x$  films by Hosono *et al.* [13].



**Figure 2:** Absorption Coefficients as a function of wavelength for  $(\text{PbO})_{1-x}(\text{CdO})_x$  films at different Vol.% of (x).

**Table 1:** The values of Absorption Coefficients for all thin films are found in at different wavelength.

sample	$\alpha \times 10^4$ at 300 nm	$\alpha \times 10^4$ at 500 nm	$\alpha \times 10^4$ at 850 nm	$\alpha \times 10^4$ at 1100 nm
PbO pure	2.61	1.62	1.4	1.2
PbO <sub>0.8</sub> CdO <sub>0.2</sub>	3.88	1.88	1.09	0.815
PbO <sub>0.6</sub> CdO <sub>0.4</sub>	4.11	1.41	0.58	0.66
PbO <sub>0.4</sub> CdO <sub>0.6</sub>	3.64	1.04	0.56	0.46
PbO <sub>0.2</sub> CdO <sub>0.8</sub>	9.85	2.46	1.42	1.25
CdO pure	4.43	0.8	0.164	0.135

### C. Optical Energy Gap

The optical energy gap of the most important constants in semiconductor physics, as it depends on the value of this constant use in the semiconductor optical and electronic applications [14]. A direct optical energy gap ( $E_g$ ) was calculated by using the relation (2) with  $r=1/2$ , and from the Figure (3), the values of  $E_g$  were

determined from the intersection point of the extrapolation of a linear curve with  $h\nu$  axis.

$$\alpha h\nu = B (h\nu - E_g^{\text{opt}})^r \quad \dots\dots\dots (2)$$

Where:

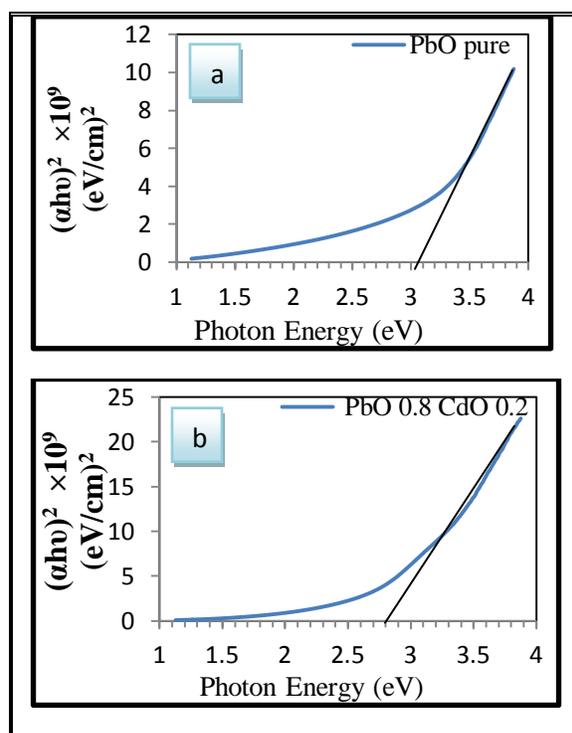
$E_g^{\text{opt}}$  : energy gap between direct transition.

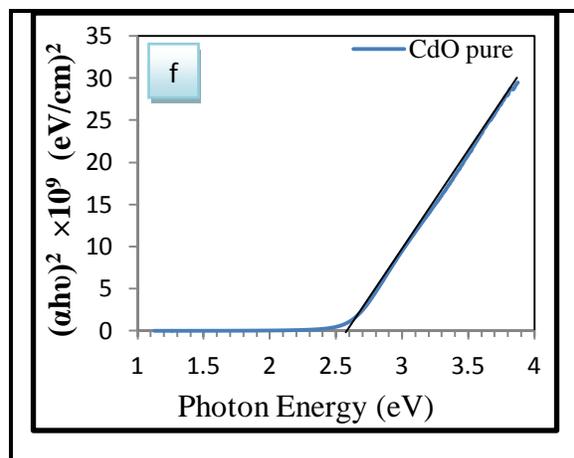
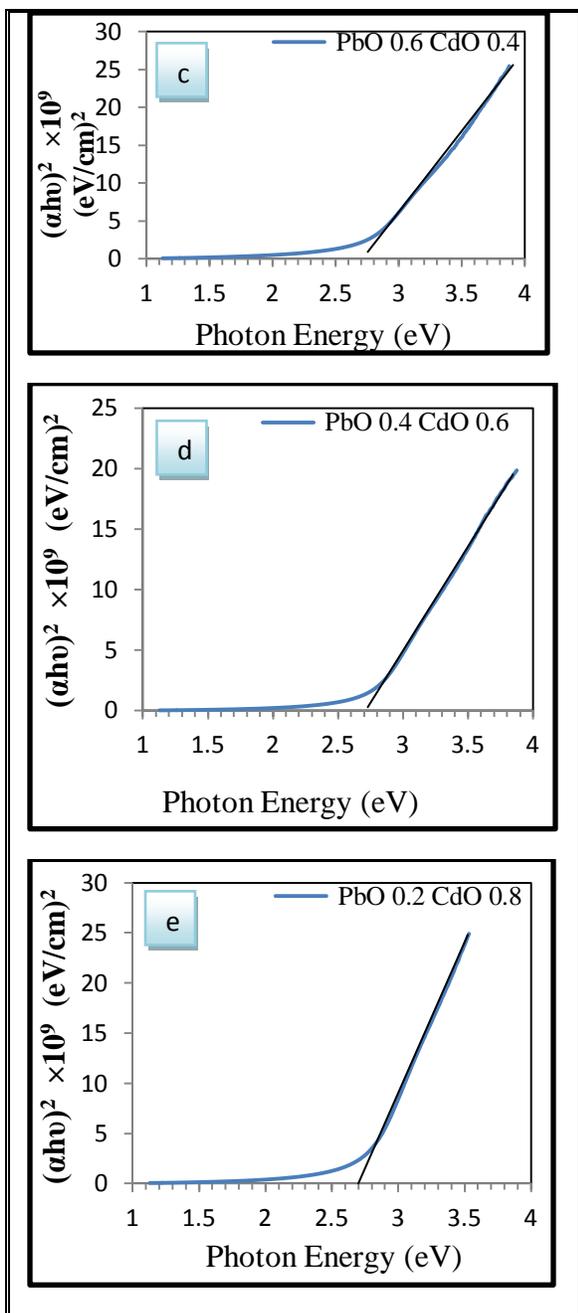
B: constant depended on type of material.

r: exponential constant, its value depended on type of transition,

$r=1/2$  for the allowed direct transition.

From Fig.(4.a), for  $x=0$  ( $\text{PbO}_{\text{pure}}$ ) the energy gap equal 3.11 eV, it decreases with increasing CdO Vol.% to (2.92, 2.8, 2.7 and 2.68) eV at  $x = 0.2, 0.4, 0.6$  and  $0.8$  respectively. As shown in Fig.(3.b to 3.e). Fig.(3.f) represents the optical energy gap of the film ( $\text{CdO}_{\text{pure}}$ ) which equal 2.6 eV. The observed optical band gap of PbO is compatible with the earlier reports [15]. To reduce the amount of optical energy gap here is important because it easily the transmission of electrons from the valence to the conduction band which contributes to the use of these films in various electronic applications [2].





**Figure 3:**  $(\alpha h\nu)^2$  as a function of  $h\nu$  for  $(\text{PbO})_{1-x}(\text{CdO})_x$  films at different Vol.% of (x).

**Table 2:** The values of optical energy gap for  $(\text{PbO})_{1-x}(\text{CdO})_x$  thin films.

Sample	$E_g$ (eV)
PbO pure	3.11
PbO <sub>0.8</sub> CdO <sub>0.2</sub>	2.92
PbO <sub>0.6</sub> CdO <sub>0.4</sub>	2.8
PbO <sub>0.4</sub> CdO <sub>0.6</sub>	2.7
PbO <sub>0.2</sub> CdO <sub>0.8</sub>	2.68
CdO pure	2.6

#### D. Extinction Coefficient

The values of extinction coefficient are calculated using the relation (3) [2]. The  $K_o$  value are plotted vs.  $\lambda$  for  $(\text{PbO})_{1-x}(\text{CdO})_x$  thin films for different values of (x) as shown in Figure (4).

$$K_o = \frac{\alpha \lambda}{4 \pi} \dots\dots\dots (3)$$

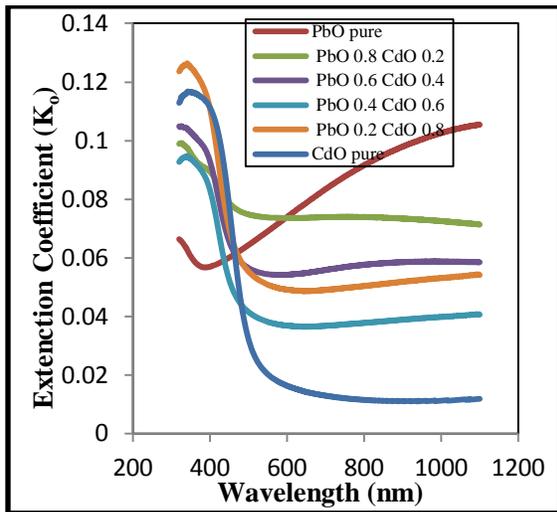
Where:

$\lambda$  : is the wavelength of incident photon rays.

$\alpha$  : absorption coefficient .

When  $X=0$ , i.e. the film is PbO, the minimum value of K equal to  $65 \times 10^{-3}$  at wave length 400nm and increase with increasing  $\lambda$  to  $105 \times 10^{-3}$  at  $\lambda = 1100$  nm. This means that there are different absorption mechanisms, depending on the film structure. With  $x=0.2$  the maximum value of K is  $98 \times 10^{-3}$  at  $\lambda = 300$  nm then decreased as  $\lambda$  increased to a minimum value at

$\lambda = 1100 \text{ nm}$  with value  $71 \times 10^{-3}$ . For  $x > 0.2$ . From this fig. at  $x = (0.4 - 0.8)$ ,  $K$  was decreased with increased  $\lambda$  and showed two characteristic regions with different wavelengths, which may be attributed to the structure of the film translated into two phases, one PbO blend and the other CdO, where the maximum value of  $K$  are increasing from  $104 \times 10^{-3}$  to  $125 \times 10^{-3}$  at  $\lambda = 300 \text{ nm}$  for  $x = 0.4$  and  $0.8$  respectively. While at  $x = 1$ , i.e (CdO pure) also the maximum value at  $\lambda = 320 \text{ nm}$  but with value of  $112 \times 10^{-3}$ . In general the behavior of  $K$  similar the behavior of  $\alpha$



**Figure 4:** Extinction coefficient as a function of wavelength for  $(\text{PbO})_{1-x}(\text{CdO})_x$  films at different Vol.% of (x).

### E. Refractive Index

The refractive index is the ratio between the speed of light in vacuum to its speed in material which doesn't absorb this light. So the value of  $n$  begins when  $K$  is constant. The refractive index ( $n$ ) was calculated from the relation (4)[2].

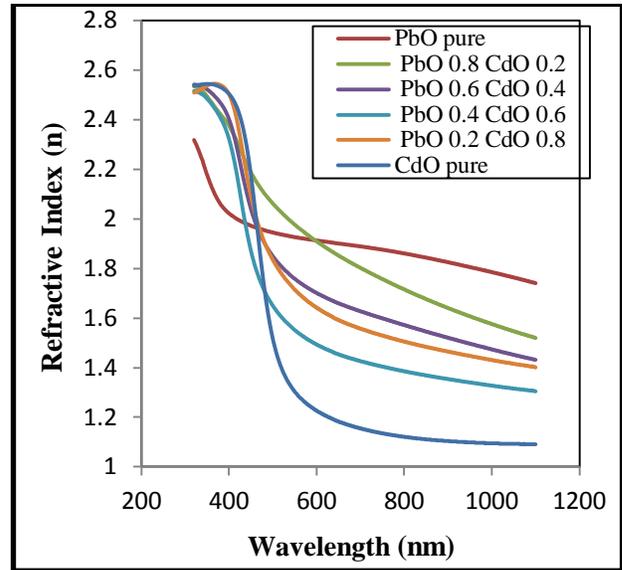
$$n = \left[ \left( \frac{1+R}{1-R} \right)^2 - (K_0^2 + 1) \right]^{\frac{1}{2}} + \frac{1+R}{1-R} \cdot \dots (4)$$

where :

R: is reflectance.

The values of  $n$  vs.  $\lambda$  were shown in Figure (5). From those Fig. the maximum value

of  $n$  for all thin films is approximately equal to a value of 2.5. From Figure (5), the graph  $n$  value decreasing with increasing  $\lambda$ , the explanation of this behavior may be related to the polarization of thin film because  $n$  depends on material polarization where with increasing polarization the velocity of light was decreased so  $n$  changed. The polarization depends on the crystalline and on the grain size of thin film so these depend on preparation conditions.



**Figure 5:** Refractive index as a function of wavelength for  $(\text{PbO})_{1-x}(\text{CdO})_x$  films at different Vol.% of (x).

### 4. CONCLUSION

$(\text{PbO})_{1-x}(\text{CdO})_x$  thin films have been successfully deposited using spray pyrolysis technique. All samples exhibit high transmittance more than 60% in the visible light spectrum. Refractive index was 2.5 and the absorption coefficients were in the range of  $0.8 - 1.62 \text{ cm}^{-1}$  at wave length 500 nm. Energy gap of these films were found to be decreasing with increasing in Vol.% of (x), while the photon transition was allowed direct transition.

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