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Structural, Morphological and Optical Properties of CDO: Al Thin Films Prepared by Chemical Spray Pyrolysis Methode

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Abstract: *Structural, Morphological and optical properties of CdO and Al doped CdO thin film were presented in this work. The Cadimium Oxide (CdO) semiconducting films are deposited on glass substrate by the chemical spray pyrolysis(CSP) method.*
The crystalline structure was studied by X-ray diffraction (XRD) having found the presence of the CdO cubic phase. The calculated values of absorption coefficient are in the order of 10^4 cm^{-1}. The value of direct band gap has been found to decrease from 2.5 to 2eV with increase of Al doping concentration.
***Keywords*:** *CdO: Al films, Spray pyrolysis, XRD, Morphological and Optical properties*

I. Introduction

Cadmium (Cd) is a metal that belongs to gether with zinc and mercury to group IIb in the Periodic Table. It has relatively low melting (320.9 °C) and boiling (765 °C) points . In the air cadmium is rapidly oxidized into cadmium oxide, cadmium oxide is used in batteries, electroplating baths, pigments, plastics [1]. There are many methods developed for the synthesis of CdO films such as spraypyrolysis [2, 3], sputtering, chemical bath deposition (CBD), pulsed laser deposition, MOCVD, sol-gel spin coating method and thermal evaporation methode [4]. The wide band gap properties of semiconductors, like CdO, are of interest particularly for applications such as solar cells and transparent electrodes TCO [5]. First reports of the use of pulses of laser radiation to remove, material from a solid (or liquid) target followed close on the heels of the first ruby lasers becoming available in the early 1960s. Given the obvious efficiency of the material ablation process, it was but a short step before pulsed laser ablation was first employed as a route to thin film deposition[6]. CdO one of these important semiconductors oxide which has high optical properties. According to these properties it has vast applications. Where it show high transparency in the visible region of solar spectrum and has high electrical properties which were represented low ohmic resistance. Although it is difficult to obtain simultaneously a high transmission coefficient, thin films have been carried out[7] .

In this work pure CdO and Al doped CdO films were deposited on glass by chemical spray pyrolysis methode. The structure, Optical and Morphological properties of CdO and Al doped CdO films.

II. Experimental

CdO and CdO: Al films were prepared by chemical spray pyrolysis method on glass substrates at 573 K. The effective area of the substrates was approximately 2.5 cm^2. The deposition parameters such as solution flow rate; and nozzle to substrate distance were kept constant at 5 ml/min, and (30±1) cm, respectively.
CdO was prepared using $CdCl_2.2H_2O$ and water. cadmium Chloride ($CdCl_2.2H_2O$) was dissolved in a water. Secondly, both solutions were mixed, so that the final concentration was 0.1 M.
which were then sprayed onto the heated substrates. The solution was stored in a volumetric reservoir at room temperature and connected to one side of the spray nozzle. The carrier gas, air was allowed to flow (8 l/min.) through the pressure-monitoring gauge, connected to the other side of the spray nozzle. The spray nozzle was moved in the x–y plane using the microprocessor controlled stepper motor system in order to a achieve uniform film coating. Moving the spray nozzle is just an option, so, it is possible to work in a stationary position too with the same setup. To prepared CdO:doped with Al,we added AlCl(1, 3,5wt%) to $CdCl_4.H_2O$ and $CS(NH_2)_2$ solution.

III. Result and Discussion

3.1X-ray Characterization

The X-ray diffraction (XRD) patetern of the CdO and CdO: Al thin films deposited on glass substrate is illustrated in Figure (1). the figur reveals a polycrystalline structure of the film. In this diffraction pattern, the peaks at 2θ (32.922, 37.942, 55.751) correspond to diffraction from (111) and (200) and (220) planes of the CdO cubic phase, respectively. It is apparent from this figure that all films are preferentially orientated along (111) crystallographic directions and the preferential orientation peak for Al doped film became more intense. This may be attributed to the crystallinity of the CdO film being improved with Al doping. This result is comparable with results obtained by [8, 9].

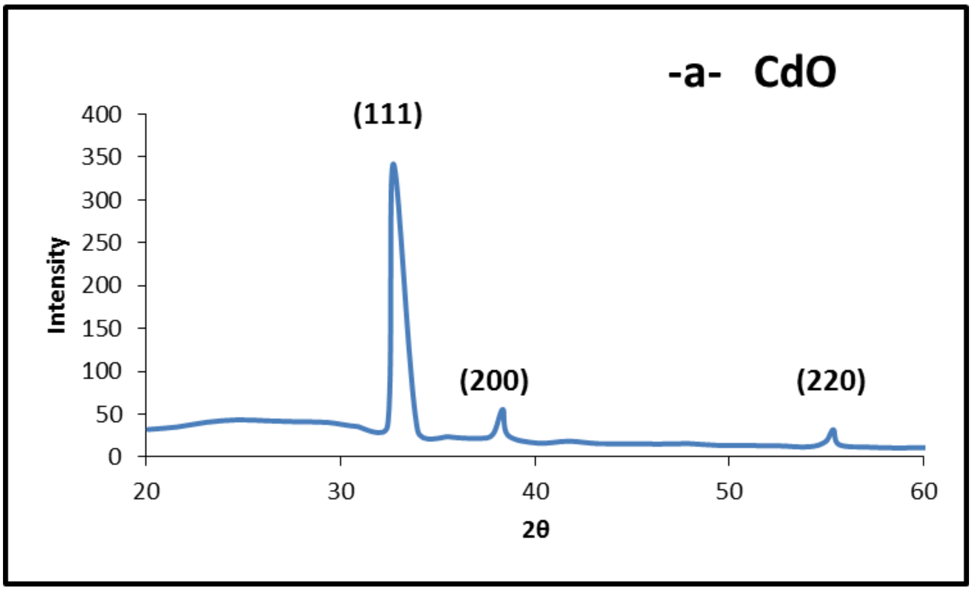


Figure (1-a) XRD pattern CdO thin film.

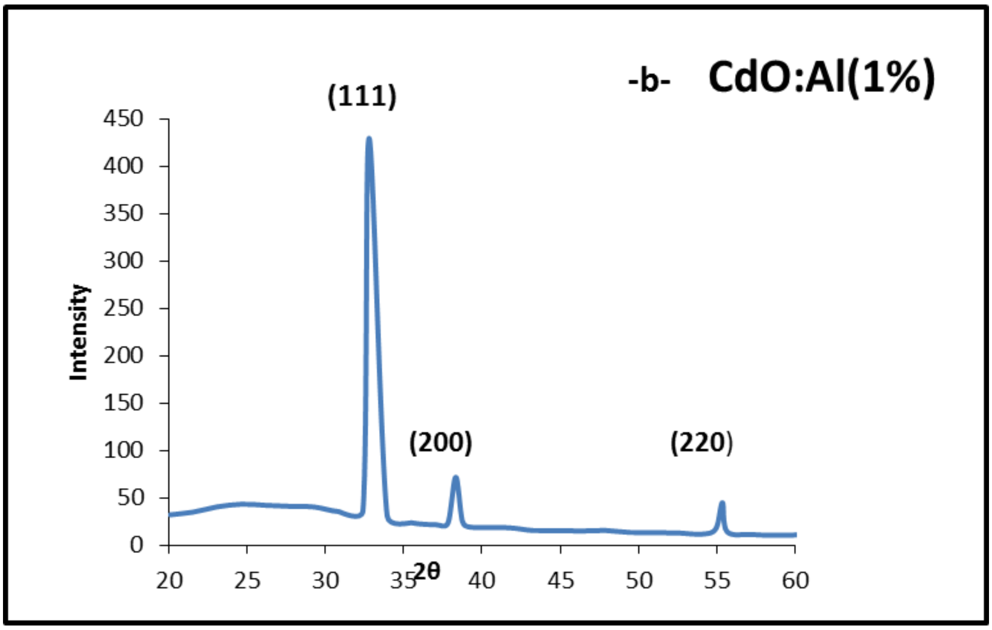


Figure (1-b) XRD pattern CdO:Al(1%) thin film.

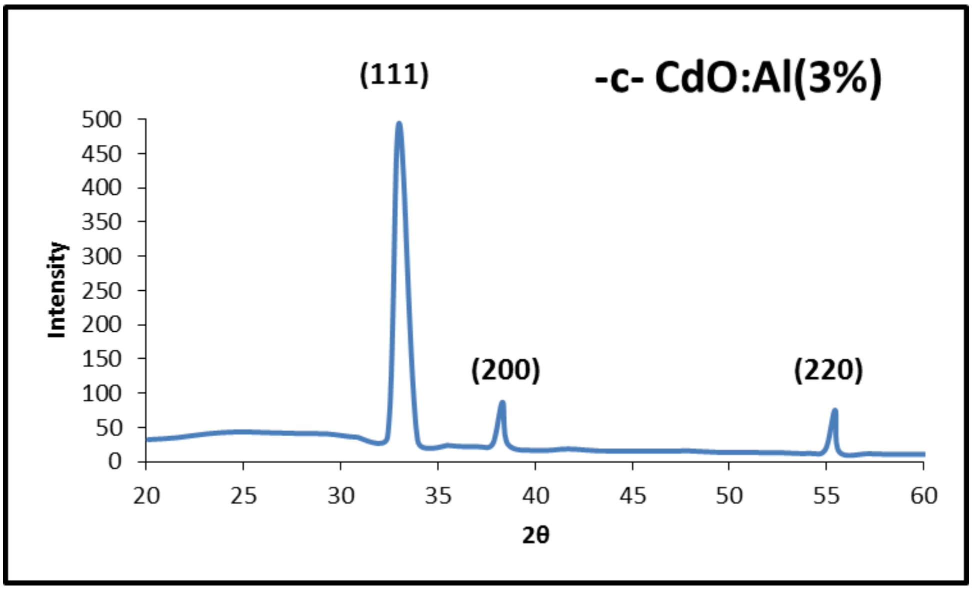


Figure (1-c) XRD pattern CdO:Al(3%) thin film.

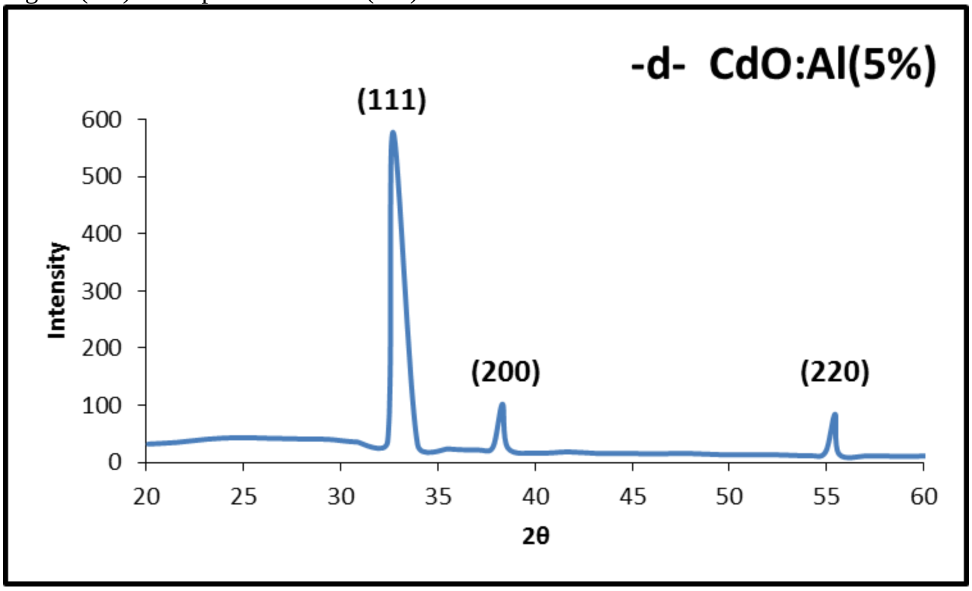


Figure (1-d) XRD pattern CdO:Al(5%) thin film.

Table (1): The Strucural Parameters of CdO:Al thin film

Film	$2\theta_{Exp.(deg)}$	$2\theta_{Stan.(deg)}$	$d_{Exp.}(A^o)$	$d_{Stan.}(A^o)$	(hkl)
CdO	32.922 37.942 55.751	33.001 38.285 55.258	2.792 2.349 1.6610	2.7120 2.3490 1.6610	111 200 220
CdS:Al(1%)	33.1649 37.701 55.4329	33.001 38.285 55.258	2.6994 2.3840 1.6561	2.7120 2.3490 1.6610	111 200 220
CdS:Al(3%)	32.9698 38.5658 55.1958	33.001 38.285 55.258	2.750 2.3327 1.6627	2.7120 2.3490 1.6610	111 200 220
CdS:Al(5%)	33.5310 38.1900 54.8570	33.001 38.285 55.258	2.6705 2.355 1.672	2.7120 2.3490 1.6610	111 200 220

as well as indicates that the lattice constants fluctuating with the increasing in Al concentration as listed in table (2) and calculated from the following equation [10] .

$$d = \frac{a}{\sqrt{h^2+k^2+l^2}} \quad ....(1)$$

Where ,
d : is the interplaner distance.
hkl : miller indices .
a : lattice constants .
The calculated values of lattice constants for CdO and CdO:Al thin films are in good agreement with ASTM data .
The grain size (D_{hkl}) of CdO and CdO:Al thin films where Al equal (1,3&5) deposited by chemical spray pyrolysis method on glass substrate at R.T are evaluated for the preferred planes [hkl] using the scherrer 's formula[11].

$$D = \frac{0.9\,\lambda}{\beta\,\cos\theta} \quad \dots\dots\dots\dots \qquad (2)$$

Where , β: is the full width at half maximum (FWHM) in radian and λ : is the X-ray wavelength (1.5406 A°).
The dislocation density (δ) of CdO and CdO:Al thin films which defined as the length of dislocation lines per unit volume of the crystal was calculated from this equation [12].

$$\delta = \frac{1}{D^2} \quad \dots\dots\dots\dots \qquad (3)$$

The values of the dislocation density of CdO and CdO:Al thin films are given in table (2) . It is evident from this table that the dislocation density of CdO and CdO:Al thin films Almost constant with increasing in Al concentration which can be also deduced from the Almost constant in the grain size where the dislocation density is proportion reversely with the square of the grain size according to eq.(4). The strain (ξ) developed in CdO and CdO:Al thin films can calculated from the relation [13] :

$$\xi = \frac{\beta\,\cos\theta}{4} \quad .... \ (4)$$

The values of the strain of CdO and CdO:Al thin films Almost constant with increasing in Al concentration .

Table (2): Variation of Grain Size , Lattice Constants**,** Dislocation Density and Strain of CdO and CdO:Al Thin Film

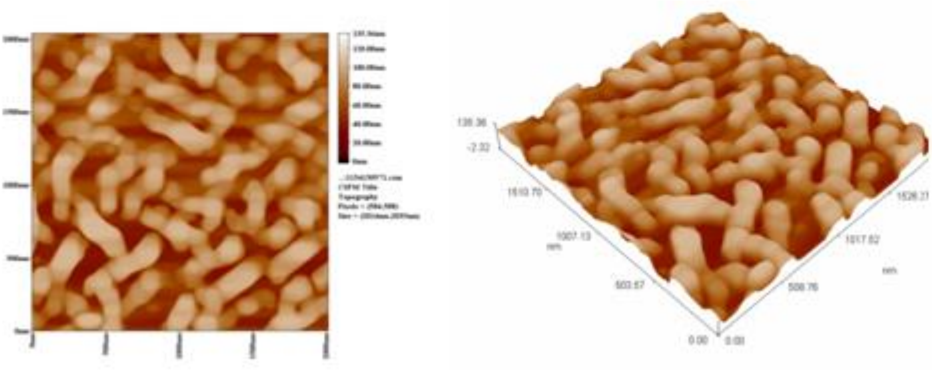
Film	**Grain size (nm)**	**Lattice Constants a(Å)**	**Dislocation density (lines.Å^{-2}) x 10^{-5}**	**Strain (rad) x 10^{-3}**
CdO	**271.27**	**4.6263**	**1.3588**	**1.2777**
CdS:Al(1%)	**271.420**	**4.6745**	**1.3574**	**1.277**
CdS:Al(3%)	**271.331**	**4.76313**	**1.358**	**1.2775**
CdS:Al(5%)	**307.449**	**4.6252**	**1.0578**	**1.274**

3.2. Morphological characterizations

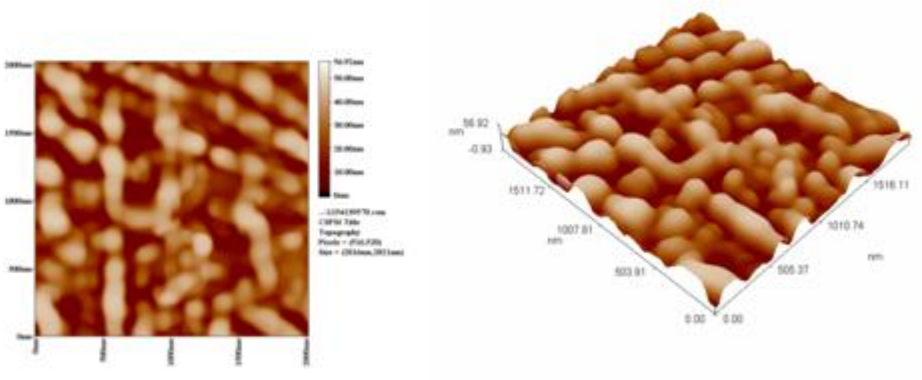
Atomic force microscopy (AFM) is a non-invasive and convenient technique to study the morphological characteristics and surface roughness of semiconductor thin films and to observe microstructure of thin films . It is well known that AFM is one of the most effective ways for the surface analysis due to its high resolution and powerful analysis software [14]. The two-dimensional AFM images at size equal to (2015.63x2027.34) nm of CdO and CdO:Al thin films . Fig. 2 shows two-dimentionnal (2D) and three-dimentionnal AFM scans of the crystallized CdO and CdO:Al thin films grown by spray pyrolysis on glass substrates

Table 3: Variation average diameter and surface roughness Rq values of the CdO and Al doped CdO thin films

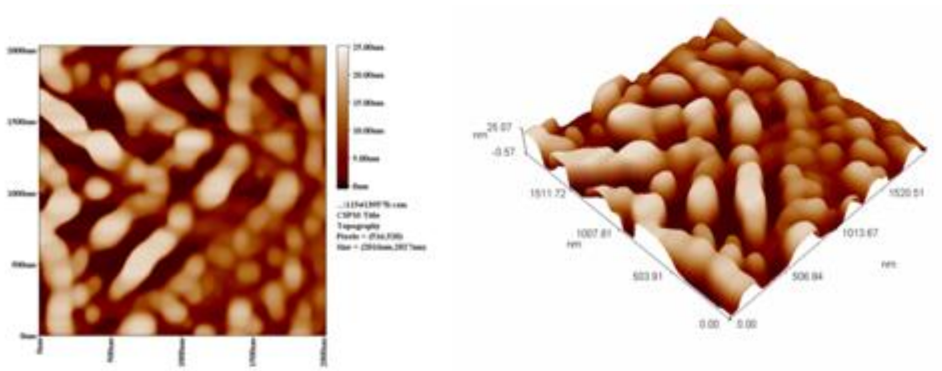
Film	Root mean square (nm)	Avg. Diameter (nm)
CdO	20.2	**86.50**
CdO:Al(1)%	11.1	**98.10**
CdO:Al(3)%	6.71	**100.44**
CdO:Al(5)%	1.39	**93.33**



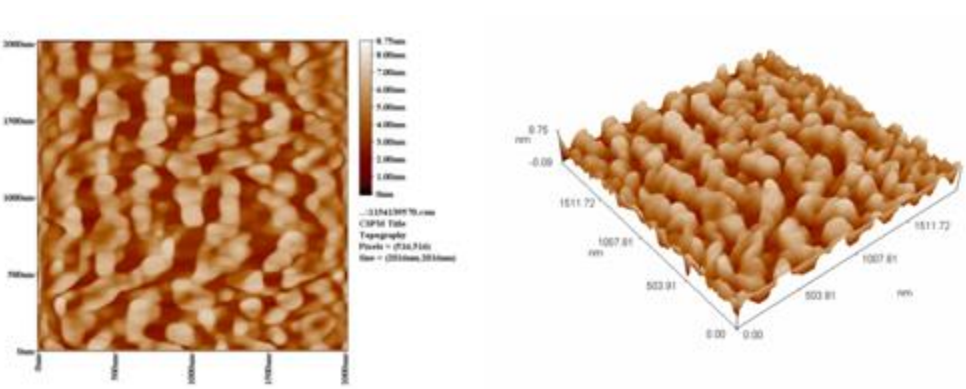
CdO



CdO:Al(1)%



CdO:Al(3)%



CdO:Al(5)%

Fig.(2) The three-dimensional and two – dimensional AFM images of CdO and CdO:Al thin films

3.2.Optical properties

The transmittance spectrum of CdO and CdO:Al thin films where Al equal (1 , 3 , 5)% are shown in Fig.(3). It is clear from this figure that the transmittance decreases with increasing in Al concentration [15,16].The absorbance spectrums of CdO and CdO: Al thin films where Al equal (1 , 3 , 5)% are shown in Fig. (4). It is clear that as the Al concentration increases the absorbance of CdO: Al thin films is increased .

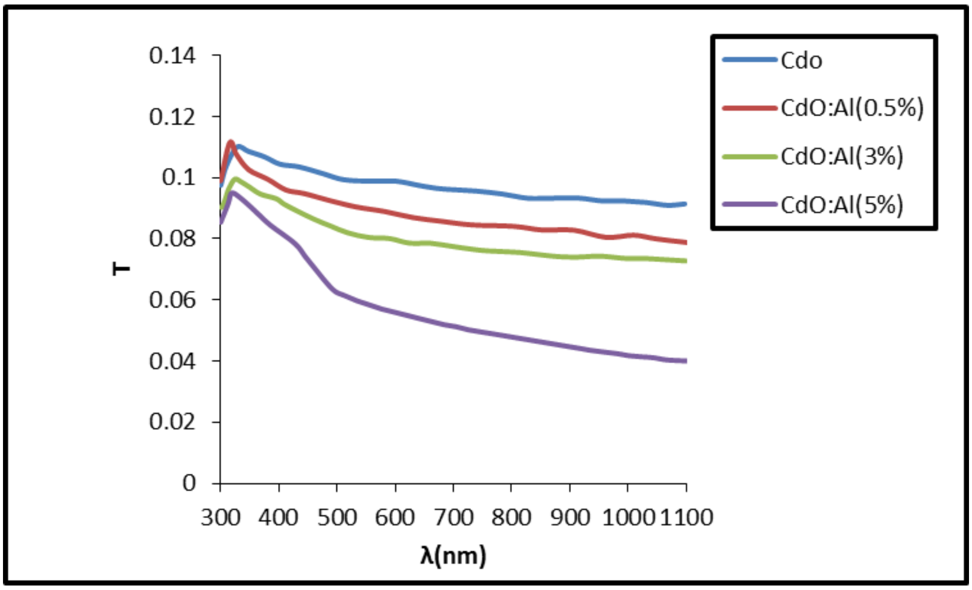


Fig.(3) Transmission spectrum of CdO and CdO:Al thin film

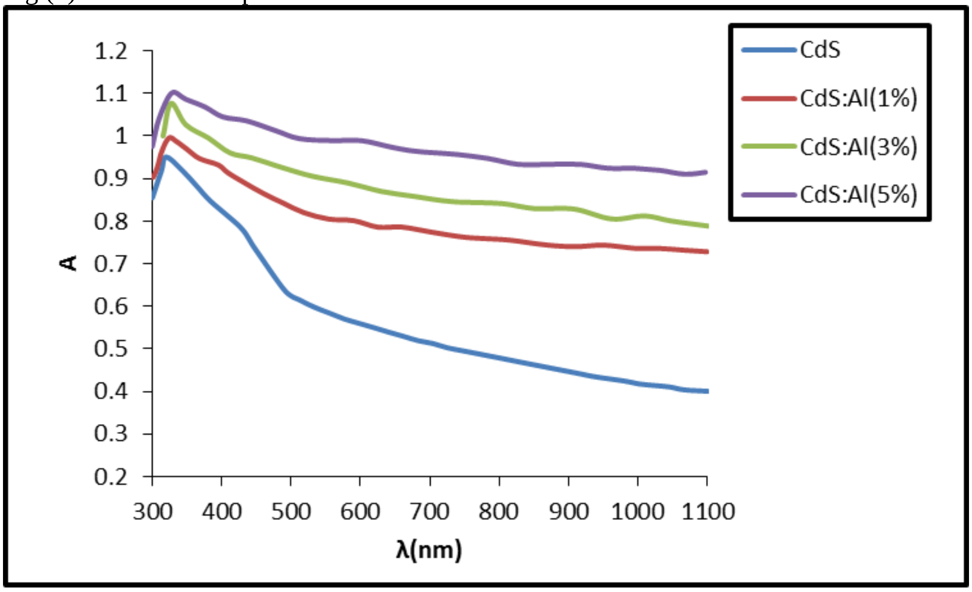


Fig.(4) Absorbance spectrums of CdO and CdO:Al thin film

The absorption coefficient (α) of CdO and CdO:Al thin films , was determined by using eq.(5) .

$$\alpha = 2.303 \frac{A}{t} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (5)$$

The variation of the absorption coefficient of CdO and CdO:Al thin films with the wavelength for Al constretion equal to (1 , 3 & 5)% is shown in figure (5) . It can be noticed that the value of the absorption coefficient of CdO and CdO:Al thin films is of the order of (10^4) cm^{-1} which supports the direct band gap nature of the semiconductor [17] . Our result agrees with [16]. It is also clear that the absorption coefficient of CdO and CdO:Al thin films increases with the increase in Al concentration.This is attributed to the increase in the absorbance of CdO and CdO:Al thin films with the increase in Al concentration causing an increment in their absorption coefficient where the relation between the absorbance and absorption coefficient is proportional at constant thickness according to eq.(5) .

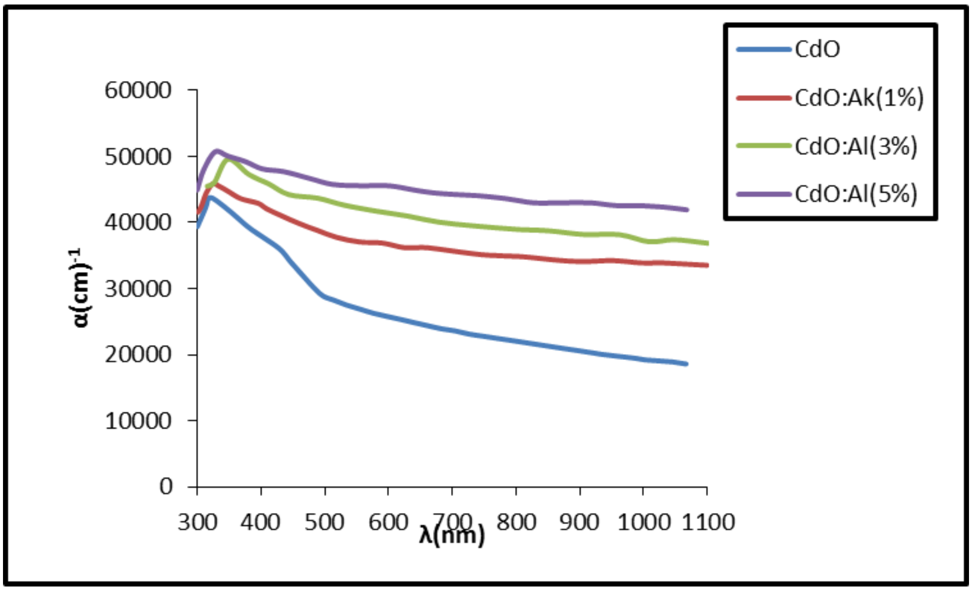


Fig.(5): absorption coefficient of CdO and CdO:Al thin film

The optical energy gap values (E_g) for CdO:Al thin films prepared by chemical spray pyrolysis methode have been determined from the region of the high absorption at the fundamental absorption edge of these films by using Tauc equation

$$\alpha h\upsilon = B_o \left(h\upsilon - E_g\right)^r \ldots\ldots\ldots\ldots \quad (6)$$

Where , α : is the absorption coefficient , hυ : is the incident photon energy in eV , Bo : is a constant depends on the nature of the material
(properties of its valence and conduction band) and r : is a constant depends on the nature of the transition between the top of the valence band and bottom of the conduction band.

This equation is used to find the type of the optical transition by plotting the relations $(\alpha h\upsilon)^2$, $(\alpha h\upsilon)^{1/2}$, $(\alpha h\upsilon)^{2/3}$ and $(\alpha h\upsilon)^{1/3}$ versus photon energy (hυ) and select the optimum linear part[18]. It is found that the first relation yields linear dependence, which describes the allowed direct transition , then E_g was determined by the extrapolation of the portion at (α=0) as shown in Fig.(6). It is clear that the optical energy gap for CdO and CdO:Al thin films decreases as the Al concentration in the films increased. The optical energy gap values for CdO and CdO:Al thin films were (2.5 , 2.2 , 2.1 and 2) eV for Al concentration (0,1,3&5) respectively . Fig. (6) illustrates the variation of the optical energy gap for CdO and CdO:Al thin films with concentration (Al) [11,16,19].

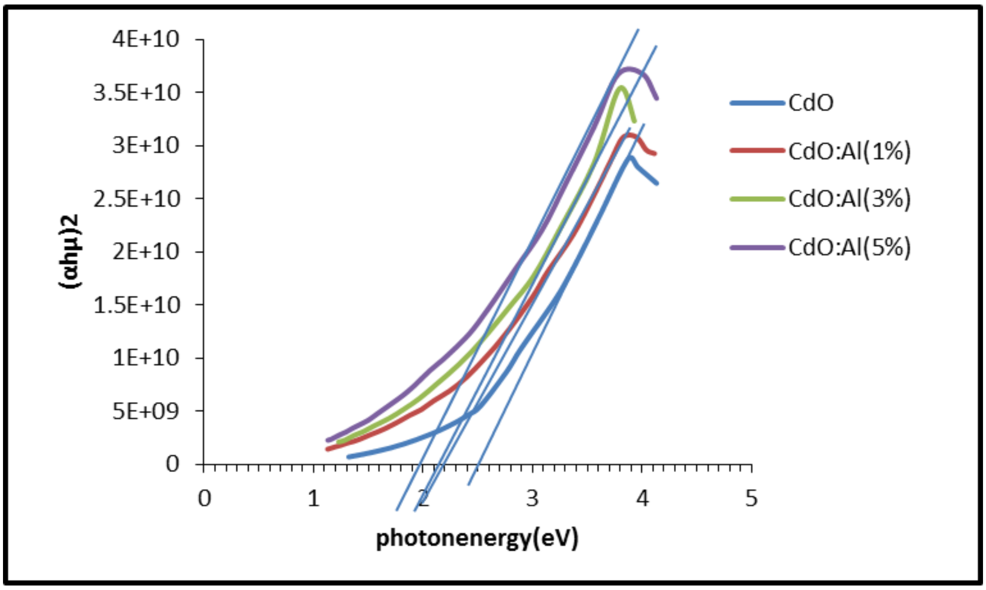


Fig.(6) $(\alpha h\upsilon)^2$ as a function of hυ for CdO and CdO:Al thin film

The refractive index (n°) of CdS and CdS:Sn thin films have been determined by using the following equation [20]:

$$n_\circ = \sqrt{\frac{4R}{(R-1)^2} - k_\circ^2} - \left(\frac{R+1}{R-1}\right) \quad (7)$$

Where , R : is the reflectance of the films and k_o : is the extinction coefficient.

The variation of the refractive index as a function of the wavelength for CdOand CdO:Al, thin films is illustrated in Fig. (7). It is clear from this figure that the refractive index decreases with the increasing in the wavelength of the incident photon . Also it can be observed , that the refractive index of CdOand CdO:Al thin films decreases with the increasing in the Al concentration[8].

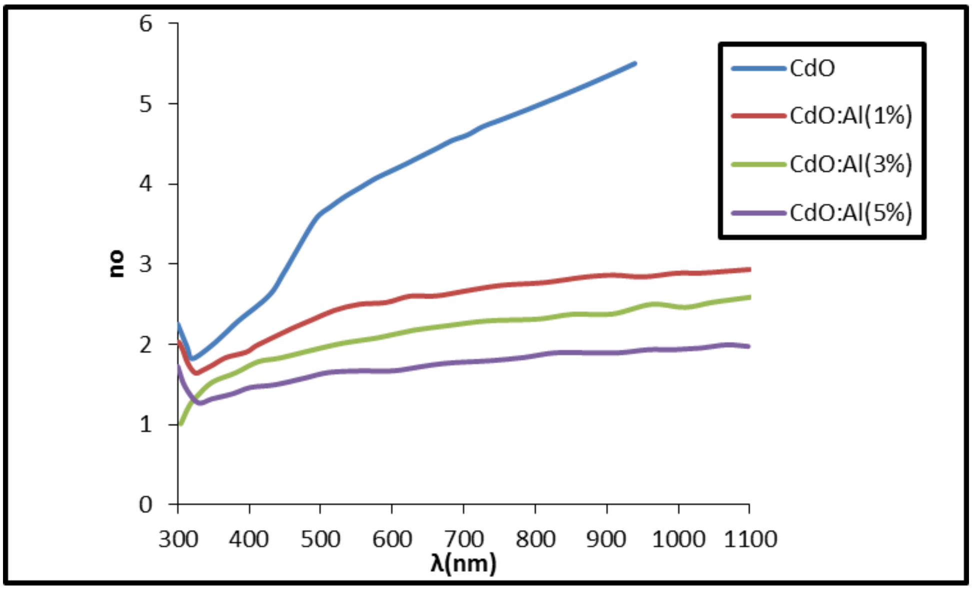


Fig.(7) The variation of the refractive index for CdO and CdO:Al thin films

The extinction coefficient (k°) have been determined by using the following equation [21]:

$$k_{\circ} = \frac{\alpha\lambda}{4\pi} \ldots\ldots\ldots\ldots\ldots \quad (8)$$

Where , α : is the absorption coefficient and λ: is the wavelength of the incident photon .

It is clear from this equation that k° depends on α and has a similar behavior to α . Fig. (8) illustrates the variation of the extinction coefficient of CdO and CdO:Al thin films with the wavelength , with the increasing in the Al concentration the extinction coefficient k° increases . Therefore k° will increase with the increasing in the Al concentration since it has a similar behavior to α and depends on it .

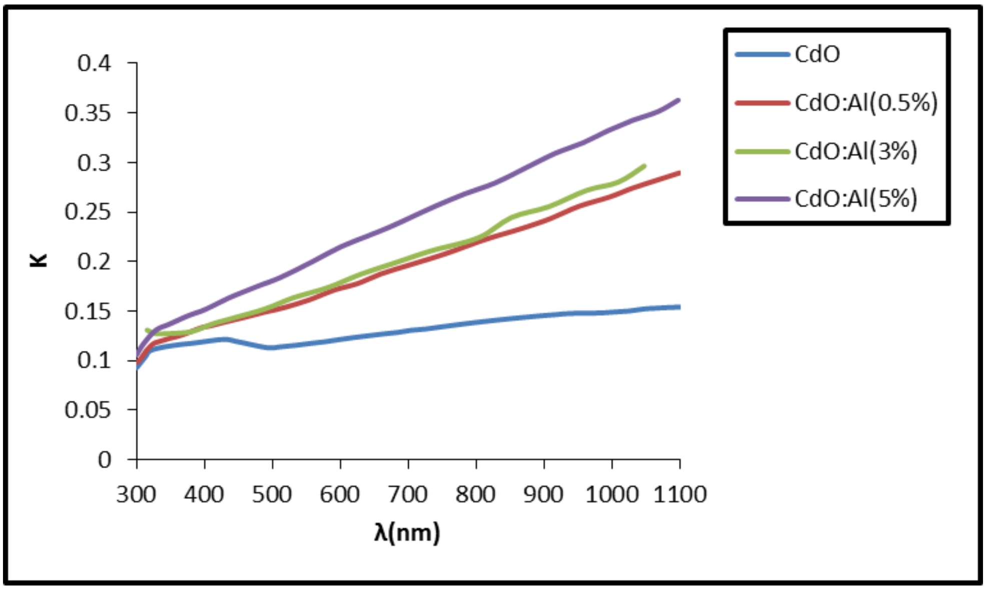


Fig.(8) Extinction coefficient of CdO and CdO:Al thin film s

IV. Conclusions

CdO and CdO:Al thin film were successfully deposited using spray pyrolysis method at substrate temperatures 573 K. The crystalline and cubic CdO and CdO:Al thin films with only (111) . Optical studies show that the transmittance and band gap energy decreases with increase in Al concentration. The variation of band gap energies from 2.5 to 2 eV.

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