



Structural and optical properties Investigation of $Zn_xCd_{1-x}S$ thin films

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Abstract – $CdZn_xS_{1-x}$ thin films with different composition have been deposited on glass substrate by the spray pyrolysis method at RT using $CdCl_2$ (0.1M), $ZnCl_2$ (0.1M) and H_2NCSNH_2 (0.1M) solution and a substrate temperature of $(400 \pm 20^\circ C)$. X-ray diffraction studies reveal that the films are polycrystalline in nature with hexagonal structure and preferential orientation along (002). The grain size of the films is found to increase from (37.397 to 46.902) nm with increasing Zinc concentration while the strain and the dislocation density of the films are found to decrease from $(7.15 \text{ to } 4.54) \times 10^{-4}$ rad and from $(3.82 \text{ to } 1.93) \times 10^{14}$ lines.m⁻² respectively. The transmittance spectrums of $CdZn_xS_{1-x}$ thin films reveal very pronounced interference effects for photon energies below the fundamental absorption edge by exhibiting interference pattern. The optical energy gap for $CdZn_xS_{1-x}$ thin films increases and shifts towards the UV region as the Zn concentration in the films increased.

Keywords: $CdZn_xS_{1-x}$ thin films, CdS thin films, ZnS thin films, spray pyrolysis technique

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Introduction

Recent investigations have evoked considerable interest in ZnS thin films due to their vast potential for use in thin film devices such as photo luminescent and electroluminescent devices and more recently as *n*-type window layer hetero junction solar cells [1]. Zinc sulfide has found wide use as a thin film coating in the optical and microelectronic industries. It has high refractive index (2×25 at 632 nm), high effective dielectric constant (9 at 1 MHz) and wide wavelength pass band (0.4–13 mm) [2]. It is commonly used as filter, reflector and planar waveguide.

Cadmium sulphide (CdS) are considered to be very important materials for a wide spectrum of optoelectronic applications as having good chemical, mechanical stability [3] and specific physical properties such as direct band gap widths, high absorption coefficients in the visible and infrared part of the solar spectrum, good electrical properties (e.g. carrier mobility and lifetime) and increased capability in obtaining adjustable *n*- or *p*-type conductivity by doping [4]

Thin film of $Zn_xCd_{1-x}S$ are known to have properties in between those of CdS and ZnS. Because The addition of Zinc to cadmium sulphide has resulted in very interesting properties related to photo electrochemistry and optoelectronics [5], because the ternary materials provide a possibility of tailoring their properties as per requirements and hence project themselves as important semiconducting materials for the applications in the field of device fabrication [6]. The band gap energy of CdS can be increased by the addition of ZnS ($E_g = 3.6$ eV). The ternary compound $Zn_xCd_{1-x}S$ offers a great range of tune ability both of its band gap (from 2.42eV for CdS to 3.6 eV for ZnS) and its lattice parameters[7]. Several techniques were employed for the growth of the ternary CdZnS films. In the present work, $Zn_xCd_{1-x}S$ thin films were deposited by Spray pyrolysis technique at different composition of (*x*). The effect of Zinc concentration on the structural and optical properties of these films was studied and discussed.

2.Experimental Details

$Zn_xCd_{1-x}S$ thin films were produced on a glass substrate by the spray pyrolysis technique. The $ZnCl_2$, $CdCl_2$ salts and H_2NCSNH_2 were dissolved in deionized water in separate beakers. Aqueous solutions of $ZnCl_2$, $CdCl_2$ salts and H_2NCSNH_2 were used as the sources of Zn, Cd, S, respectively. The $ZnCl_2$, $CdCl_2$ and H_2NCSNH_2 solutions were mixed for 30 min. With a magnetic stirrer. The compositions of the solutions used to fabricate the $Zn_xCd_{1-x}S$ thin films are shown in Table 1 in terms of the nominal concentrations in the deposition solution. The substrate temperature was regulated at ($400 \pm 20^\circ C$) during the deposition process using a resistive heater and a thermocouple. Glass substrates were prepared by cutting (2.5×2.5) cm^2 pieces and cleaning them by water and they were placed in the microwave until we used them. In order to spray the solution onto the substrate using an ultrasonic atomizer,

nitrogen (N_2) Was used as the carrier gas at a pressure of 3bar with a deposition rate of $3cm^3/min$ during the deposition process. At the end of the spraying process, nitrogen (N_2) was flowed onto the thin films formed on the glass substrate for 3min in order to dry them. They were then cooled down naturally to room temperature.

flowed onto the thin films formed on the glass substrate for 3min in order to dry them. Films thickness was determined by a multiple beam interferometry (Fizeau fringes in reflection). The films were characterized by X-ray diffraction technique using (Philips X-ray diffractometer) with $CuK\alpha$ radiation with wavelength (1.5406 \AA). A (UV-160A UV-visible recording) spectrophotometer supplied by Japanese company (Shimadzu) was used to record the optical absorbance and transmittance spectra of $Zn_xCd_{1-x}S$ thin films at wavelength range (480-1100) nm. Surface morphology of the films were studied by using (CSPM AA3000) Atomic Force Microscope (AFM) supply by Angstrom Company.

Table 1 Solution used for the production of $Zn_xCd_{1-x}S$ thin films

Nominal Composition of $Zn_xCd_{1-x}S$	$CdCl_2(0.1)$ (ml)	$ZnCl_2(0.1)$ (ml)	$H_2NCSNH_2(0.1)$ (ml)
0.0	25	-	25
0.25	18.75	6.25	25
0.5	12.5	12.5	25
0.75	6.25	18.75	25
1	-	25	25

3.Result and discussion:

3.1.Structural properties:

The X-ray diffraction patterns of $Zn_xCd_{1-x}S$ thin films where *x* equal (0, 0.25, 0.5, 0.75 & 1) deposited by spray pyrolysis method on glass substrate at R.T with thickness (500) nm are shown in Fig. (1). The XRD patterns for all cases reveal polycrystalline in nature for as-deposited films having the main diffraction peak corresponding to the reflection from (002) plane. The diffraction peaks for (002) are located at (27.07, 27.37, 27.76, 28.06, 28.55) for *x* equal (0, 0.25, 0.5, 0.75, 1) respectively, therefore it is clear that there is a shift toward higher value of 2θ when composition (*x*) change from 0 to 1.

As compared with ASTM cards, all films exhibit pure hexagonal structure and our results agree with [8]-[9]-[10]-[11]-[12]-[13]-[14]-[15]. The X-ray peak corresponding to (002) reflection is observed in all cases which represents the

preferential orientation in $Zn_xCd_{1-x}S$ films. The intense and sharp peaks in the XRD pattern reveal the good crystallinity of the thin films and confirm the stoichiometric nature of $Zn_xCd_{1-x}S$ thin films.

The preferential orientation (002) in $Zn_xCd_{1-x}S$ thin films where $x=0$ (CdS film) it was reported for thermal vacuum evaporated [16] - [17] - [18] - [19] while it was reported for chemical bathed[20]-[21] . Whereas for x equal to 0.5 and for $x=0.7$ has been also reported for chemical deposited [22] ,[23] Finally for $x=1$ (ZnS film) the preferential orientation (002) was reported for development technology solution [24] while it was reported for Chemical decomposition[25] . Table (2) illustrates bragg's angles , inter planers spacing , relative intensities , miller indices , and lattice constants of $Zn_xCd_{1-x}S$ thin films .

The diffraction peaks for (002) are located at 27.07° , 27.37° , 27.76° , 28.06° and 28.55° for x equal (0 , 0.25 , 0.5 , 0.75 & 1) respectively , therefore it is clear that there is a shift toward higher value of 2θ when composition (x) change from 0 to 1 .This shifting has been reported Rehana Zia [14]. This shifting in the peak position with increasing composition (x) suggests that the lattice constants increase with the increasing in Zn concentration as listed in table (2) and calculated from the following equation [26] :

$$\frac{1}{d^2} = \frac{4}{3} \left[\frac{h^2 + hk + k^2}{a^2} \right] + \frac{l^2}{c^2} \quad (1)$$

Where , d : is the interplaner distance , hkl : miller indices , a , c : lattice constants .

The calculated values of lattice constants for in $Zn_xCd_{1-x}S$ thin films are in good agreement with ASTM data . Similar results have been reported by Ghoneim [13].

It is clear from XRD patterns of in $Zn_xCd_{1-x}S$ thin films , that the full width at half maximum (FWHM) decreases with the increasing of Zn concentration in these films. This decreasing in (FWHM) indicates an increasing in the grain size of in $Zn_xCd_{1-x}S$ thin films as given in table (3) according to Scherrer's formula where the relation between the grain size (D) and (FWHM) is reversal as follows [27]:

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

Where , β : is the full width at half maximum (FWHM) in radian and λ : is the X-ray wavelength (1.5406 Å).

The increasing in the grain size with increasing in the Zn concentration had been also reported for chemical path deposition [28]-[21] .

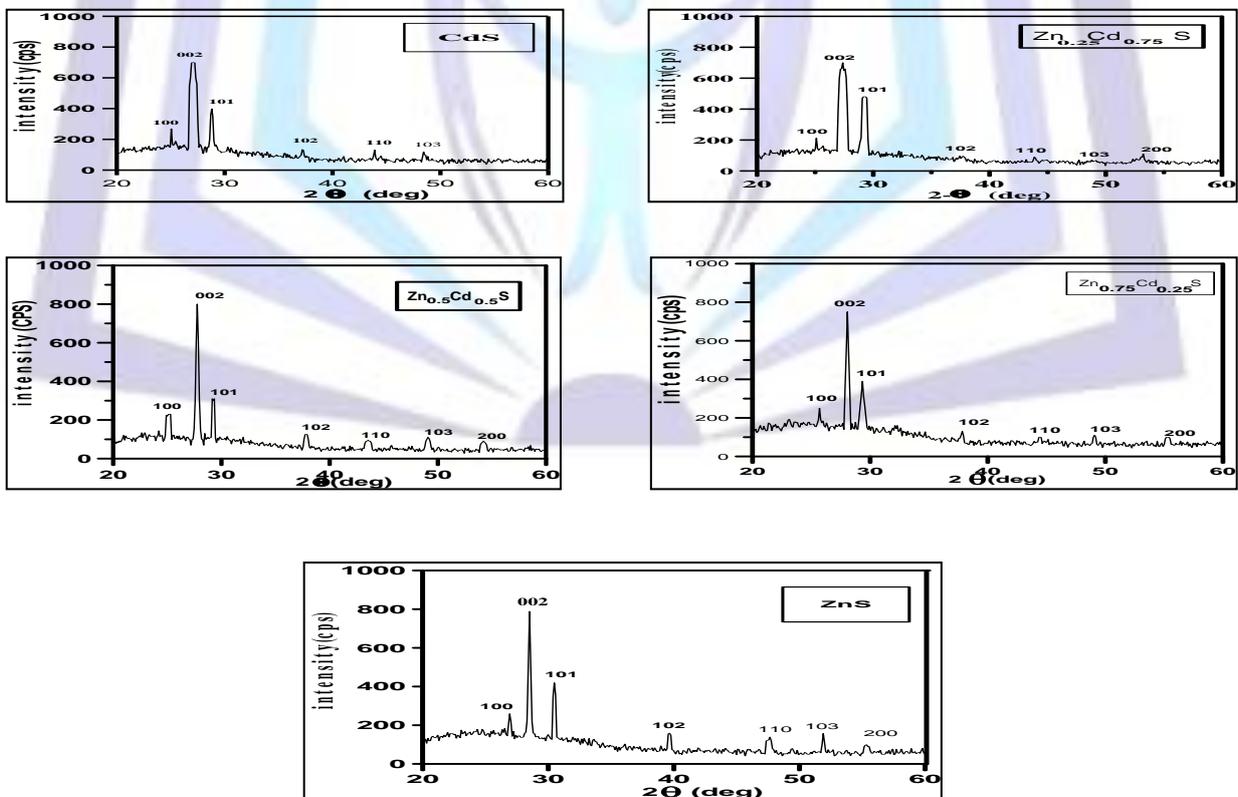


Fig1 X-ray diffractograms of thin film for different concentratio



Table (2) the structural parameters of $Zn_xCd_{1-x}S$ thin films

thin film	2Φ	d(exp)	d(STM)	Hkl	a(nm)
CdS	25	0.3565	0.358	100	0.4116
	27.0751	0.329	0.337	002	-
	28.753	0.310	0.316	101	0.405
	37.2412	0.241	0.245	102	0.408
	43.8541	0.206	0.2068	110	0.409
	48.3943	0.1879	0.1898	103	0.42
	$Zn_{0.25}Cd_{1-0.25}S$	25.1011	0.3543	0.3542	100
27.3712		0.325	0.325	002	-
29.1478		0.3056	0.3054	101	0.399
37.4386		0.239	0.235	102	0.407
43.8541		0.2062	0.2059	110	0.412
54.2306		0.17187	0.1718	200	0.3969
$Zn_{0.5}Cd_{1-0.5}S$		25.198	0.353		100
	27.766	0.321		002	-
	29.2465	0.305		101	0.4
	37.8334	0.2375		102	0.407
	43.5581	0.2069		110	0.413
	49.0852	0.1854		103	0.48
	54.2176	0.1689		200	0.39
$Zn_{0.75}Cd_{1-0.75}S$	25.6933	0.346		100	0.404
	28.0621	0.3176		002	-
	29.3452	0.304		101	0.4015
	37.8334	0.2375		102	0.413
	44.4463	0.2037		110	0.407
	49.0852	0.1854		103	0.443
	55.3033	0.166		200	0.383
ZnS	26.8777	0.3313	0.3309	100	0.382
	28.5556	0.3122	0.3128	002	-
	30.4309	0.2934	0.2925	101	0.383
	39.61	0.2272	0.2273	102	0.3825
	47.4073	0.1915	0.1911	110	0.383
	51.8488	0.1763	0.1764	103	0.3829
	55.3033	0.1659	0.1661	200	0.383

The dislocation density (δ) of $Zn_xCd_{1-x}S$ thin films which defined as the length of dislocation lines per unit volume of the crystal was calculated from this equation [29]:



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

The values of the dislocation density of in Zn_xCd_{1-x}S thin films are given in table (3) . It is evident from this table that the dislocation density of in Zn_xCd_{1-x}S thin films decreases with increasing in Zn concentration which can be also deduced from the increasing in the grain size where the dislocation density is proportion reversely with the square of the grain size according to eq.(3).This decreasing in the dislocation density indicates an improvement in the crystallinity of in Zn_xCd_{1-x}S thin films and their homogeneity as increasing in Zn concentration .

The strain (ξ) developed in in Zn_xCd_{1-x}S thin films can calculated from the relation [30] :

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

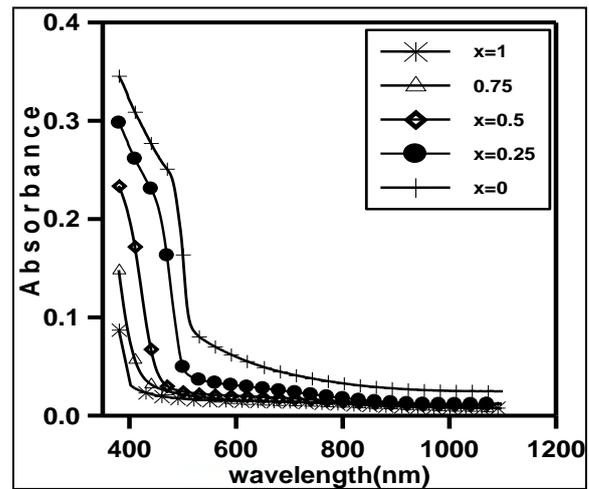
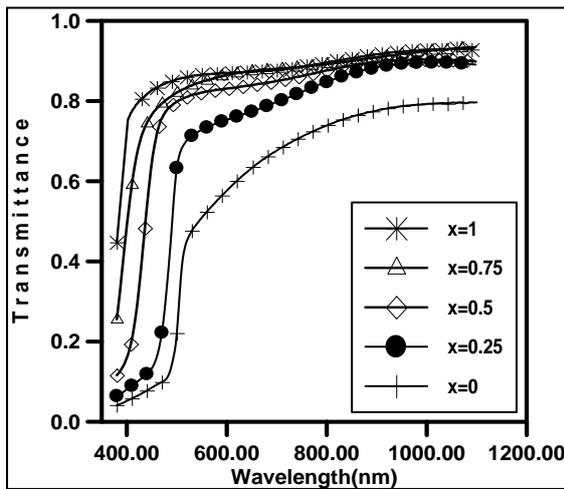
The origin of strain is related to lattice misfit which in turn depends upon the growing condition of the films [31] . The values of the strain of in Zn_xCd_{1-x}S thin films are given in table (3). It is clear from this table that the strain in in Zn_xCd_{1-x}S thin films decreases with increasing in Zn concentration which can be also deduced from the increasing in the grain size , where the decreasing in the strain and the dislocation density with the increasing in the grain size is a well-known phenomenon [32] . In polycrystalline films , the dislocated atoms occupy the regions near the grain boundary . Due to large number of grain boundaries and short distance between them , the intrinsic strains are always associated with such interface .The increasing in the grain size causing reduction in the number of grain boundaries and that leads a reduction in the intrinsic strains associated with the grain boundaries interface

Table 3: variation of the full width at half maximum, grain size, dislocation density and no.of crystals of Zn_xCd_{1-x}S thinfilms with composition(x) .

Thin film	Grain size (nm)	δ 10^{-4} (line/m ²)*1	N _o $\cdot 10^{-3}m^{-2}$
CdS	37.397	7.15	3.82
Zn _{0.25} Cd _{1-0.25} S	37.13	7.25	3.9
Zn _{0.5} Cd _{1-0.5} S	47.02	4.52	1.92
Zn _{0.75} Cd _{1-0.75} S	46.805	4.56	1.9
ZnS	46.902	4.54	1.93

Optical properties

The transmittance spectrum of Zn_xCd_{1-x}S thin films where x equal (0,0.25,0.5,0.75,1) are shown in Fig.(2). It is clear from the figure that the transmittance increases with increasing in Zn concentration which can be also deduced from the changing in the films' colure. This changing in the films' colure was also mentioned by Chaudhari *et al.* [33] . Also our results agree with Nadeem *et al.* [34] who found that with increasing in Zn concentration from 0 to 1 the transmittance of Zn_xCd_{1-x}S thin films increases. Also it is clear that the transmittance of Zn_xCd_{1-x}S thin films changes from ≈ 70% to ≈ 90 % and this is consider a wide range which can be useful in different application like optical filters and that agree with Kumar[20] . In addition the variation of the transmittance of Zn_xCd_{1-x}S thin films with the wavelength is very important because this variation will limit the transmitted wavelengths which play an important role in determination the category/type of the optical filters . The absorbance spectrums of Zn_xCd_{1-x}S thin films where x equal (0,0.25,0.5,0.75, & 1) are shown in Fig. (3). It is clear that as the Zn concentration increases the absorbance of Zn_xCd_{1-x}S thin films is decreased . This decreasing in the absorbance is attributed to the decreasing of Cd concentration which results in an decrease of the depth of donor levels and these levels will be available for the photons to be absorbed therefore the absorbance of Zn_xCd_{1-x}S thin films will decrease with increasing in Zn concentration. As well as from the same figure , it can be seen that the absorption edge shifts to the lower wavelengths as the Zn concentration increased and takes the values 506 nm , 496 nm , 458 nm,386nm and 364 nm for x equal 0 , 0.25,0.5 , 0.75 and 1 respectively . This shifting in the absorption edge was also mentioned by [15]-[23] . From this shifting in the absorption edge it can be deduced that the energy gap of Zn_xCd_{1-x}S thin films will increases with increasing Zn concentrations.



Fig(2) Transmittance spectrums of $Zn_xCd_{1-x}S$ thin films

Fig(3) absorbance spectrums of $Zn_xCd_{1-x}S$ thin films

The optical energy gap values (E_g) for $CdSe_xS_{1-x}$ thin films prepared by thermal evaporation method have been determined from the region of the high absorption at the fundamental absorption edge of these films by using Tauc equation [35]:

$$\alpha h\nu = B_0 (h\nu - E_g)^r \quad (5)$$

Where, α : is the absorption coefficient, $h\nu$: is the incident photon energy in eV, B_0 : is a constant depends on the nature of the material (properties of its valence and conduction band) [36] 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 [37].

This equation is used to find the type of the optical transition by plotting the relations $(\alpha h\nu)^2$, $(\alpha h\nu)^{2/3}$, $(\alpha h\nu)^{1/2}$ and $(\alpha h\nu)^{1/3}$ versus photon energy ($h\nu$) and select the optimum linear part. 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 ($\alpha=0$) as shown in Fig.(4). It is clear that the optical energy gap for $Zn_xCd_{1-x}S$ thin films increases as the Zn concentration in the films increased. This is attributed to the decreasing of Cd concentration which results increase of the depth of donor levels associated which in turn causing an increase in the optical energy gap for $Zn_xCd_{1-x}S$

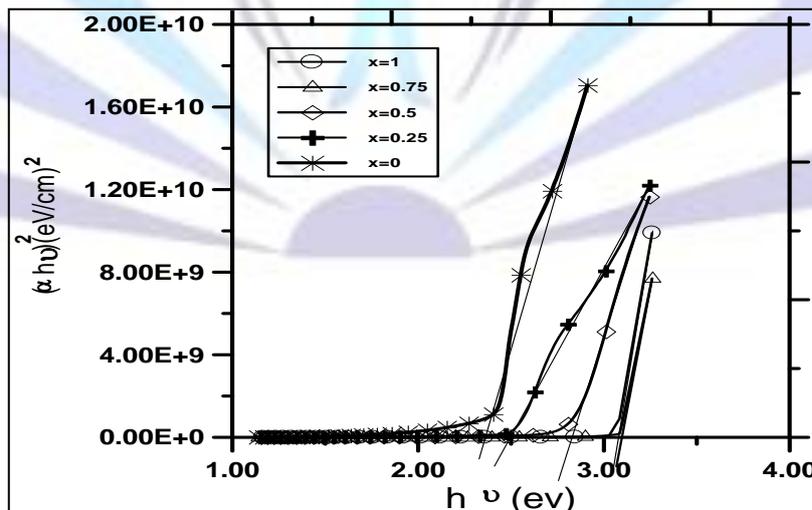


Fig (4) energy gap of $Zn_x Cd_{1-x}S$ thin film for different concentration .

thin films . This shifting and increasing in the optical energy gap have been also reported for Martin et.al. [23]. The optical energy gap values for $Zn_xCd_{1-x}S$ thin films were 2.45 eV , 2.5 eV , 2.7 eV,3.2 and 3.43 eV for x equal (0 , 0.25,0.5 , 0.75 & 1) respectively as shown in table 4 . The obtained values of the optical energy gap match well with the reported values of CdS [38]-[39][40] and ZnS [34]-[41], the other composition have in between values .

**Table 4. Comparison of band gap, energy wavelength of ZnxCd_{1-x}S film**

X=	E _g (ev)	λ(nm)
0	2.4	516
0.25	2.5	496
0.5	2.8	443
0.75	3.15	394
0.75		
1	3.1	400

II. Conclusion

Chemical spray pyrolysis technique can be successfully employed for the deposition of uniform morphologist and polycrystalline Zn_xCd_{1-x}S thin films with hexagonal phase at R.T . The increasing of Zn concentration in these films has improved the crystallinity of the films and their homogeneity because the decreasing in the strain and the dislocation density with the increasing in the grain size according to the increasing of Se concentration . Morphological studied indicates that surface roughness decreases with increasing in Zn concentration . Optical studied indicates that Zn_xCd_{1-x}S thin films exhibit direct band gap which is strongly depends on the Zn concentration almost cover the entire visible spectral that makes these films are suitable for optoelectronic devices especially for solar cell and optical filter

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