

OPTICAL AND STRUCTURAL PROPERTIES OF $Zn_xCd_{1-x}S$ (x=0.2, 0.4, 0.6 AND 0.8) THIN FILMS PREPARED BY THERMAL EVAPORATION TECHNIQUE

^{a*}Humaira Latif, ^aRehana Zia, ^bMuneeb Irshad, ^bHuma Latif

^aDepartment of Physics, Lahore College for Women University, Lahore, Pakistan.

^bDepartment of Physics, University of Engineering and Technology Lahore, Pakistan

* Corresponding Author: humairalatif23@gmail.com

Abstract

Thin films of $Zn_xCd_{1-x}S$ (x=0.2, 0.4, 0.6 and 0.8) were deposited on cleaned soda lime glass substrates at room temperature by thermal evaporation technique, having source current 50-65 Ampere, chamber pressure 10^{-5} Torr and deposition rate 0.4 nm/sec. These conditions were same for all the thin films having different zinc concentrations. UV-VIS Spectrophotometry was used to study the optical properties of thin films of $Zn_xCd_{1-x}S$ in room temperature. XRD was used to study the structure of the thin films of $Zn_xCd_{1-x}S$ having various composition of „x“. UV-VIS studies revealed that as the concentration of zinc content increases, transmission spectra shift towards the shorter wavelength region from (575-526)nm, the percent transmittance was increased in the visible range with the increase of zinc content, absorption edges and absorption coefficient spectra also shift towards the shorter wavelength and hence the direct band gap energy varied non-linearly from 2.55eV to 2.84eV. It was also found that optical conductivity increases with photon energy and thin film of $Zn_{0.4}Cd_{0.6}S$ has high optical conductivity as compared to other value of „x“. The reflectance and optical constants such as the extinction coefficient and refractive index were also found to depend upon the zinc concentration in the films. XRD studies showed that all the thin films of $Zn_xCd_{1-x}S$ (x= 0.2, 0.4, 0.6 and 0.8) had a strong peak in between the diffraction angle 26.6° - 31.7° , which confirmed that all the thin films exhibited the wurtzite structure with a preferential orientation of (002) plane. It was also found that lattice constants, inter planer spacing, volume and grain size decreases except for $Zn_{0.4}Cd_{0.6}S$ thin film which had high crystallinity as compared to the other composition of the zinc content.

Keywords: Zinc Cadmium Sulphide ($Zn_xCd_{1-x}S$), Cadmium Sulphide (CdS), Zinc Sulphide (ZnS), Thermal evaporation, Thin film, Composition, Structural, Optical properties.

1. Introduction

The exceptional rise in thin film researches is, no doubt, because of their wide applications in the different fields of electronics, optics, space science and other industries [1]. Thin films have unique electrical, optical, mechanical and thermal properties due to the quantum confinement effect. This effect alters all the properties of the materials [2]. The binary and ternary II-VI compound semiconductors which are based on cadmium have received great attention because of their wide applications in optoelectronic devices. The band gap energy of ternary compound semiconductors is found to be tunable in the range of binary semiconductor band gap [1]. Cadmium sulphide CdS which is an n-type direct band gap (2.4 eV) semiconductor material, is extensively used as a window layer for ternary materials CuInSe₂, CdTe based solar cells [3]. By adding the zinc into cadmium sulphide material, Zn_xCd_{1-x}S n-type direct band gap compound semiconductor is developed, which has the properties in between the CdS and ZnS and its band gap lies in between 2.4 eV (CdS) and 3.6 eV (ZnS) at room temperature [4, 5]. Due to the tunable properties of cadmium zinc sulphide ternary material, it is widely used for the fabrication of photovoltaic solar cells and photoconductive devices. The band gap energy of Zn_xCd_{1-x}S is controlled by the change of zinc composition in order to suit the material properties with that of absorber material in solar cells. Thin films of Zn_xCd_{1-x}S are basically useful as a window material for the development of p-n junctions without lattice mismatch in devices based on quaternary materials such as CuIn_xGa_{1-x}Se₂ [6, 8] or CuIn(S₂Se_{1-z})₂ [9]. Thin films of Zn_xCd_{1-x}S have been developed by various techniques, including spray pyrolysis [10,14], ion beam deposition [15], molecular beam epitaxial growth [16,17], among of them, the thermal evaporation, is an attractive and effective method. Thin films developed by this technique exhibit the larger area uniformity.

In this work, optical and structural properties of n-type polycrystalline Zn_xCd_{1-x}S thin films developed by using thermal evaporation technique are investigated by varying the composition of zinc in the range (x=0.2, 0.4, 0.6 and 0.8).

2. Experimental Setup

2.1. Substrate Cleaning

Thin films of Zn_xCd_{1-x}S having different compositions (x=0.2, 0.4, 0.6 and 0.8) have been developed on soda lime glass substrates by using thermal evaporation technique. Soda lime glass slides having sizes 1*3*1/8 inch³ were used as substrates in present investigation and cleaning was done by chemical method. These soda lime glass slides were drastically washed with detergent in order to remove the grease or other layer of oil. Then the substrates were washed thoroughly in the de-ionized water. After that these slides were successively treated with isopropylalcohol (IPA) and were dipped in beaker which containing the acetone. The beaker was then subjected to the ultrasonic vibrations for at least 15 minutes. Then once again these slides were treated with IPA and then air dried.

2.2. Thin Films Development Procedure

Zn_xCd_{1-x}S (x=0.2, 0.4, 0.6 and 0.8) thin films were developed on thoroughly cleaned soda lime glass substrates through thermal evaporation technique. CdS and ZnS powder (Merck- 99.99% pure) were mixed and compound was developed. This prepared compound of various compositions was evaporated from tungsten boat kept in

vacuum evaporation plant through Edward Coating Unit (Model No: 306). The source current of about 50 to 65 Amperes, chamber pressures of about 10^{-5} Torr and deposition rate of about 0.4nm per second was maintained in all the evaporation processes. Soda lime glass substrates were kept at room temperature throughout the experiment.

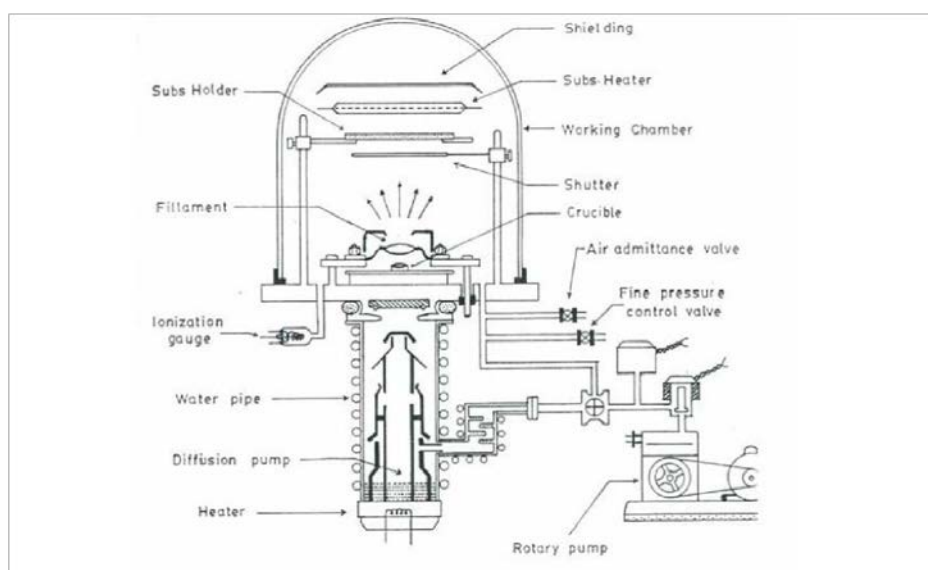


Figure 1: Schematic diagram of vacuum evaporation coating unit

The as grown thin films were uniform and stocked properly to the substrates at usual atmospheric circumstances. Thin films of CdS has orange yellow color and by increasing the zinc contents into cadmium sulphide yellowness also increased whereas the thin films of ZnS are colorless.

The optical characteristics of as grown $Zn_xCd_{1-x}S$ ($x=0.2, 0.4, 0.6$ and 0.8) thin films have been investigated through Hitachi UV-2800 double beam spectrophotometer and the structural properties of $Zn_xCd_{1-x}S$ thin films have been studied by using D_8 Diffractometer.

3. Results and Discussions

3.1. Optical Properties

Optical characteristics of zinc doped cadmium sulphide thin films had been investigated through Hitachi UV-2800 double beam spectrophotometer. This technique records transmission, as a function of wavelength. The transmission of zinc doped cadmium sulphide thin films having various compositions against wavelength are shown in Figure 2. The optical transmittance spectra have been studied by using "UV-VIS Spectrophotometer" having wavelength series from 300nm to 900nm. The transmission spectra shows that as the concentration of zinc contents increases, then spectra shift towards the shorter wavelengths from (575-480)nm and remaining thin films containing absorption edges in between (575-480)nm. It is clear from the spectra that all the thin films show high transmittance of 81-100% in the visible region (500-900) nm. The rapid increase in transmission and shifted of spectra towards the shorter wavelengths is due to the wide direct band gap of zinc sulphide. So actually by increasing the concentration of zinc sulphide, band gap also increases and that's why transmission increases in the visible region [18, 19, 20, 21, and 22]. The fluctuations of the transmittance are due to interference of radiations which passes from thin film and soda lime glass. The fluctuations represent the smooth thickness of the surfaces of films.

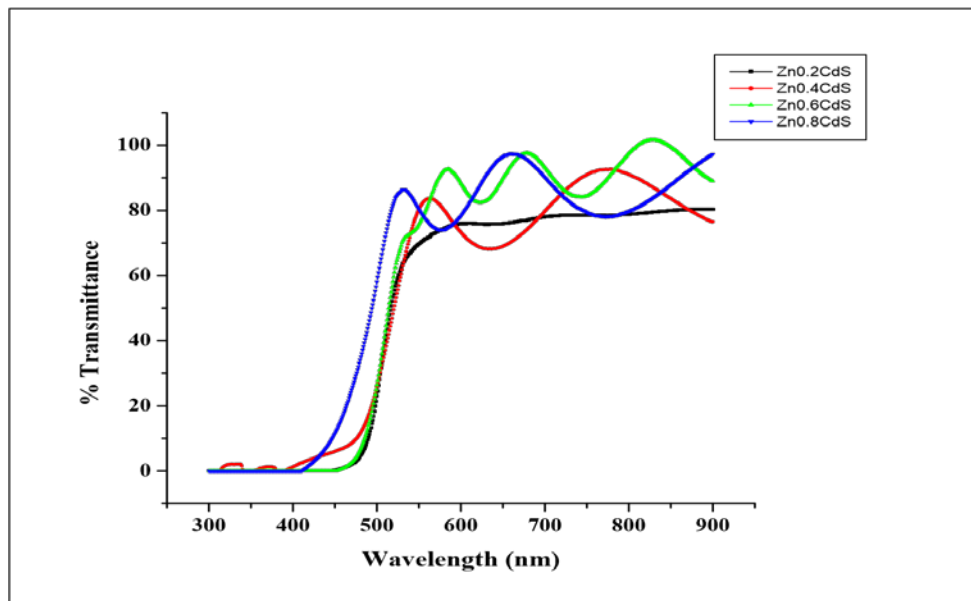


Figure 2: Variation in transmittance of $Zn_xCd_{1-x}S$ thin films With wavelength

A number of techniques can be used to find out the thickness of thin films. Out of this one theoretical technique is to find thickness through the transmission data taken over a range.

In this study Manificier et.al [23, 24] theoretical calculation was used to evaluate thickness of the developed thin films. This whole calculation was valid only if $k^2 \ll n^2$. This process is valid only when “Tmax” and “Tmin” functions build up from the figure2.

$$n = [N + (N^2 - n_0^2 n_s^2)^{1/2}]^{1/2} \tag{1}$$

$$N = \frac{(n_0 + n_s)^2}{2} + \frac{2n_0 n_s (T_{max} - T_{min})}{T_{max} T_{min}} \tag{2}$$

The thickness “t” is determined from two maxima or two minima, as follows:

$$t = \frac{M \lambda_1 \lambda_2}{2 [n(\lambda_1) \lambda_2 - n(\lambda_2) \lambda_1]} \tag{3}$$

Where, “M” is the number of oscillations between the two maximas at the wavelengths “ λ_1 ” and “ λ_2 ”, the value of “n” and “t” were found using the region of high transmission. The thickness of all the as grown thin films was 1.7 μm .

The percentage absorbance of zinc doped cadmium sulphide thin films having various compositions with wavelength are shown in Figure 3. The optical absorbance spectra have been studied from the wavelength of 380nm to 900 nm. The absorption spectra have divided into two zones; one is the larger absorption zone which is approximately from 388 to 488 nm and other one is the low absorption zone which is almost from 488 to onward nm. Absorption spectra confirm that at longer wavelength region transmission of radiations is maximum due to the minimum absorption in this region. Sharp edges appear in case of semiconductors due to the very little absorption take place when the energy of incoming photons is less band gap energy. It is clear from the spectra maximum absorbance for all the thin films of various composition is 10 percent [25]. The absorption spectra shows that as the concentration of zinc contents increases, absorption edges shift towards the shorter wavelengths. It is also notice that “Zn_xCd_{1-x}S” thin film at x=.4 covers larger area of shorter wavelength as compared to other composition of zinc contents, this might be due to the structural changes in the thin films that is increase in grain size.

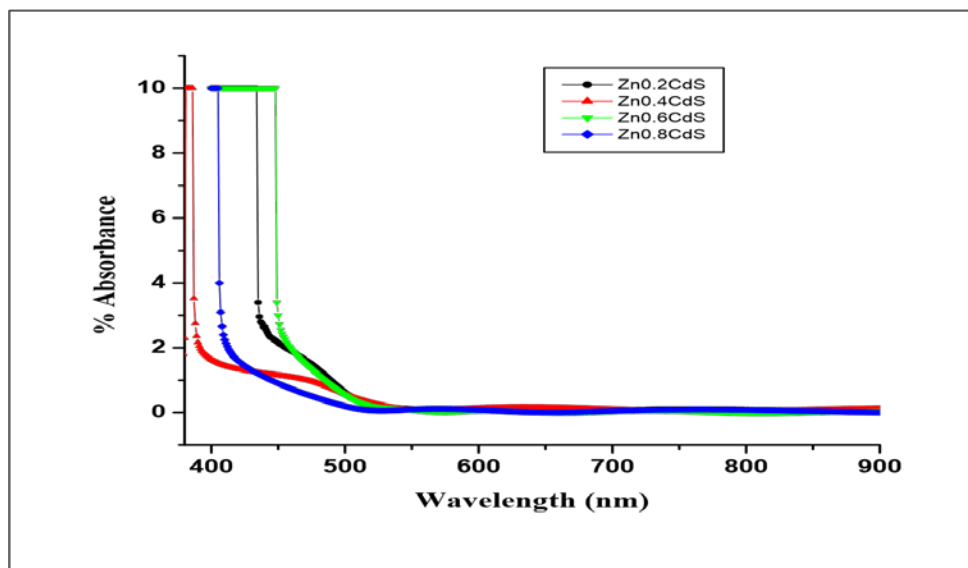


Figure3: Variation in absorbance of Zn_xCd_{1-x}S thin films with wavelength

Graph of absorption coefficient “ α ” of zinc doped cadmium sulphide thin films with wavelength is shown in fig 4. An absorption coefficient “ α ” has been determined from transmission measurements. It is expressed in the unit of reciprocal of length and given by the following relation [20, 26].

$$\alpha = 1/t \ln[1/T] \quad (4)$$

This relation shows that absorption coefficient and transmittance are inversely proportional to each other. Also in our case thickness is constant. That’s why in the wavelength region where the transmission is greater, at that region absorption coefficient is minimum and vice versa. It is clear from the Figure 4; absorption coefficient of all the films of different composition has same maximum value which is about 0.0038 cm^{-1} . “ α ” declines slowly with “ λ ” and reaches its minimum value near “absorption edge”, this indicates that absorption coefficient goes on decreasing with increasing wavelength. Figure 4 also shows that as the zinc content increases, curves shifts towards the shorter wavelength [21]. Also “Zn_xCd_{1-x}S” thin film at x = .4 covers greater portion of shorter wavelength because it has high absorption in that region as shown in Figure 4. .

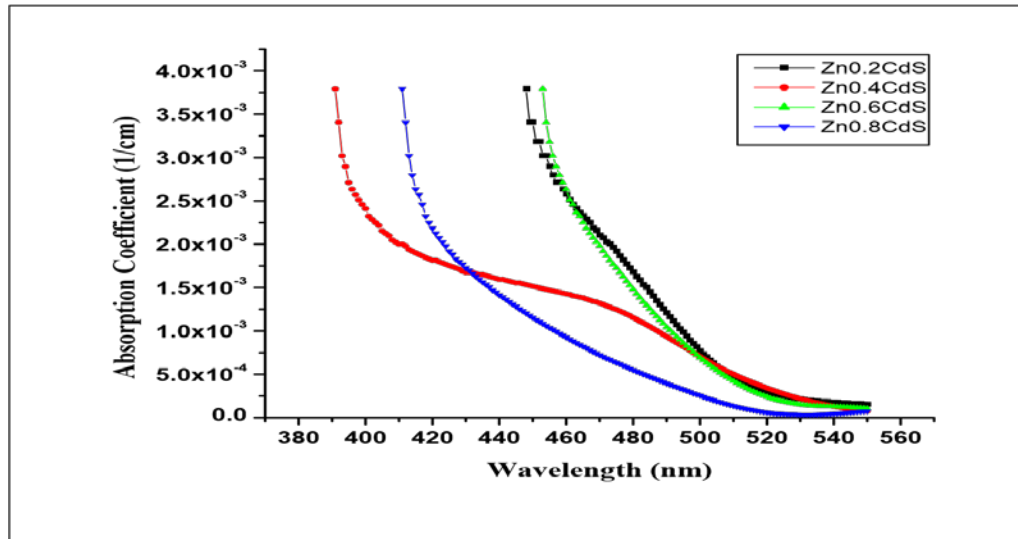


Figure 4.5: Variation in absorption coefficient of $Zn_xCd_{1-x}S$ thin films with wavelength

The band gap energies have been calculated from the plot of absorption coefficient against photon energy.

From “ α ”, the value of “ E_g ” was found out from the following relation [20, 27, and 28]:

$$\alpha = A (h\nu - E_g)^n / h\nu \tag{5}$$

Where,

n = related to the distribution of the states and its value is $1/2$ for direct band gap material and 2 for indirect band gap material, α = absorption coefficient $h\nu$ = photon energy and

A = constant

The values of α^2 as a function of $E (h\nu)$ were plotted by using the Tauc’s relation [29], which is given as;

$$\omega^2 E(\lambda) = (\hbar\omega - E_g) \tag{6}$$

Where,

$E(\lambda)$ = optical absorbance and

E_g = band gap energy

This relation shows linearity between $h\nu$ and “ $(\alpha h\nu)^2$ ”, therefore values of band gap energy was determined through the extrapolation of linear portion of curve to x-axis.

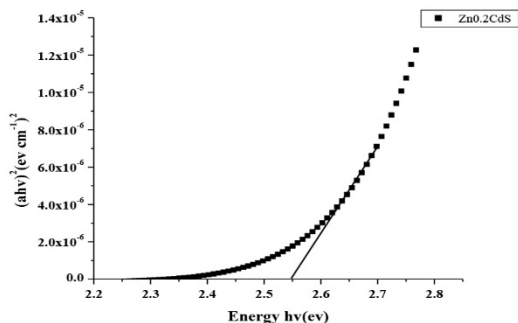


Figure 5 (a): Variation in $(\alpha hv)^2$ with Energy (eV) for $Zn_{0.2}Cd_{0.8}S$ Thin Film

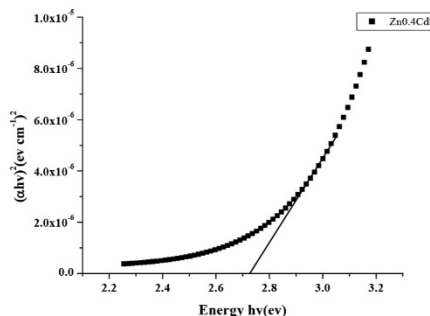


Figure 5(b): Variation in $(\alpha hv)^2$ with Energy (eV) for $Zn_{0.4}Cd_{0.6}S$ Thin Film

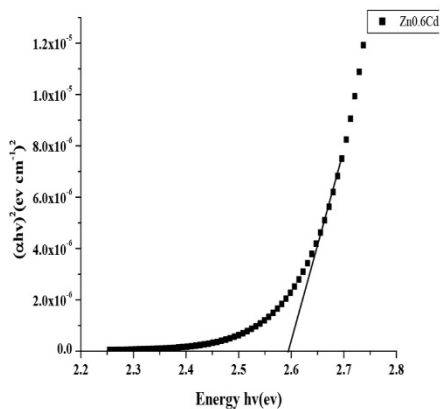


Figure 5 (c): Variation in $(\alpha hv)^2$ with Energy (eV) for $Zn_{0.6}Cd_{0.4}S$ Thin Film

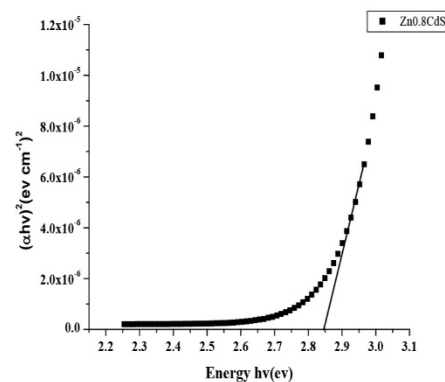


Figure 5(d): Variation in $(\alpha hv)^2$ with Energy (eV) for $Zn_{0.8}Cd_{0.2}S$ Thin Film

Table 1: Variation in bandgap energy of $Zn_xCd_{1-x}S$ Thin Films with Different Values of „x“

No. Of Samples	Zinc Contents (x)	Band Gap Energy (Eg)
1	0.2	2.55
2	0.4	2.73
3	0.6	2.59
4	0.8	2.84

It was noticed that as the concentration of zinc sulphide into cadmium sulphide increases band gaps also increases because zinc sulphide has wide band gap. Generally increase in band gap of zinc doped cadmium sulphide thin film is due to an increase in carrier concentrations which lead to “Burstein-moss effect”. The direct band gap energy varied non-linearly from “2.55 eV to

2.84eV” with continuously increasing value of x. Similar nonlinear behavior have been reported by [18,19,22].

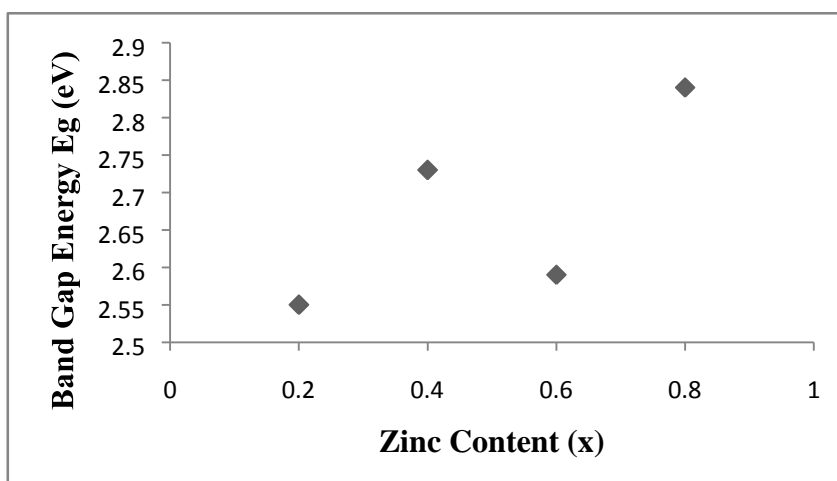


Figure 5(e): Bandgap energy Vs. Zinc contents

The variation in optical conductivity versus wavelength is shown in Figure 6. “ σ ” was found out through the relation [30];

$$\sigma = \alpha n c / 4\pi \quad (7)$$

The graph shows that as the photon energy increases, optical conductivity also increases. The increased in “ σ ” at larger “ $E (h\nu)$ ” is because of sufficient absorbance of incoming radiations at that region.

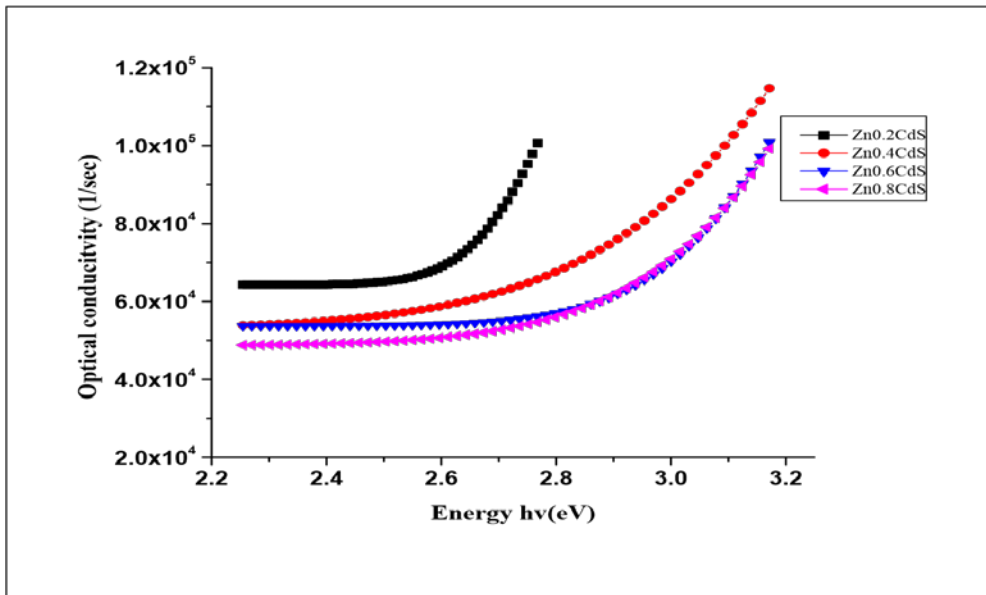


Figure 6: Variation in optical conductivity of $Zn_xCd_{1-x}S$ thin films with photon energy

Plot of percentage reflectance of zinc doped cadmium sulphide thin films having various compositions with wavelength is shown in the Figure 7. Reflectance was determined through the relation [31]:

$$R + T + A = 1 \tag{8}$$

Where, “R”, “A”, “T” is the reflectance, absorbance and transmittance respectively. It is clear from the figure all the thin films having various values of „x” shows maximum reflectance at the same value which is about 0.043. The onset behavior in reflectance of Zn doped CdS thin films occur in visible region near the absorption edge. It is noticed that “reflectance spectra” transfer toward shorter “wavelengths” owing to enhance concentration of zinc contents which conform that transmission is greater in the longer wavelength region [20].

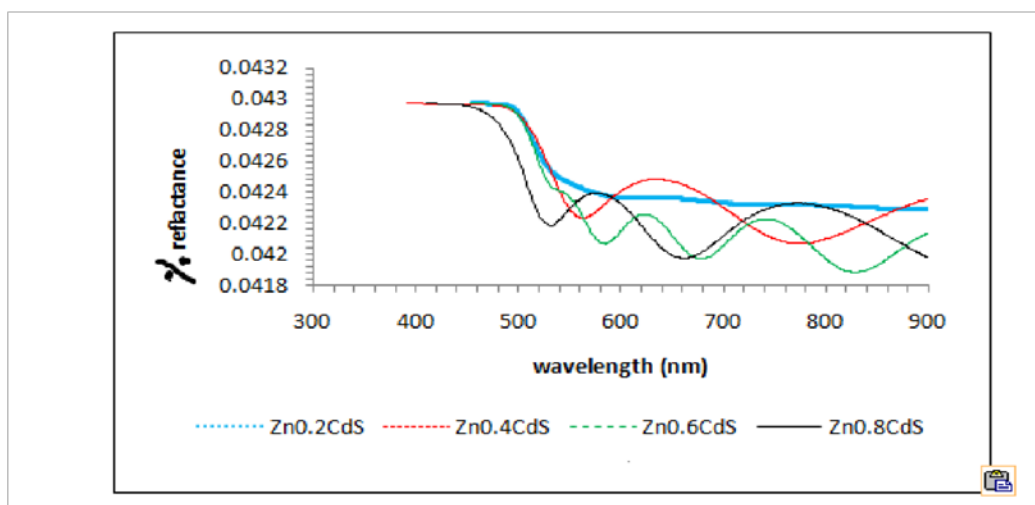


Figure 7: Variation in reflectance of $Zn_xCd_{1-x}S$ thin films with wavelength

The variation in refractive index with wavelength is shown in Figure 8. “n” was determined from the relation of reflectance [20, 32]:

$$R = (n - 1)^2 / (n + 1)^2 \tag{9}$$

Or,

$$n = (R)^{1/2} + 1 / (R)^{1/2} - 1 \tag{10}$$

This relation shows that there exists a direct relationship between the reflectivity and refractive index, therefore at low value of “R”, “n” will also be small. Initially values of “n” does not change at shorter wavelengths, all the thin films of various composition have the same value of

“n” which is about 1.523, but afterwards “n” decreases with increasing “λ” that is because of the “dispersion of light” as it moves deeper inside the material. Refractive index reduces as the absorption increases and reaches minimum value near the “absorption edge”. As the concentration of zinc contents increases the curve shifts towards the shorter wavelengths [20, 33].

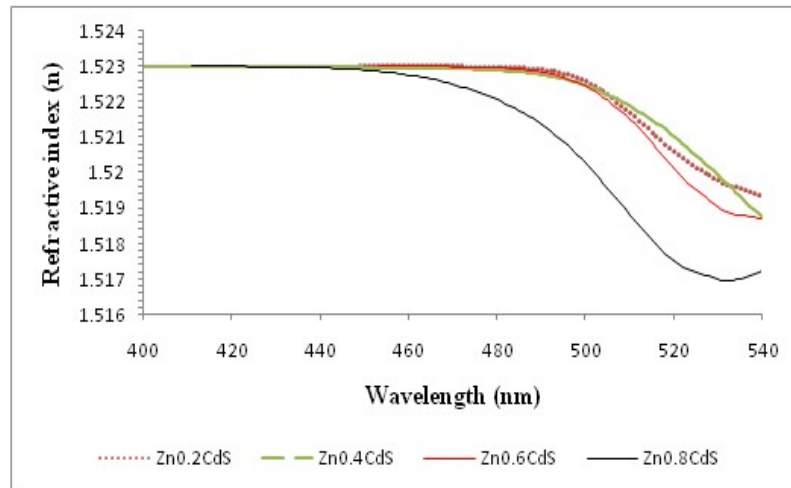


Figure 8: Variation in refractive index of Zn_xCd_{1-x}S thin films with wavelength

Plot of extinction coefficient “k” of zinc doped cadmium sulphide thin films with wavelength is shown in Figure 9. “k” determines how strongly a material absorbs or scatter light at a given “λ”.

Extinction coefficient was given by the relation [20]:

$$k = \alpha \lambda / 4\pi \tag{11}$$

The ups and downs in “k” is directly depend upon the absorption of radiation. It is clear from the figure that value of “k” decreases towards the longer wavelength. This decrease in extinction coefficient is due to decrease in absorption towards longer wavelength. By increasing the zinc contents, shifting of curves occur towards the shorter wavelengths. Similar behavior has been reported by [20, 33].

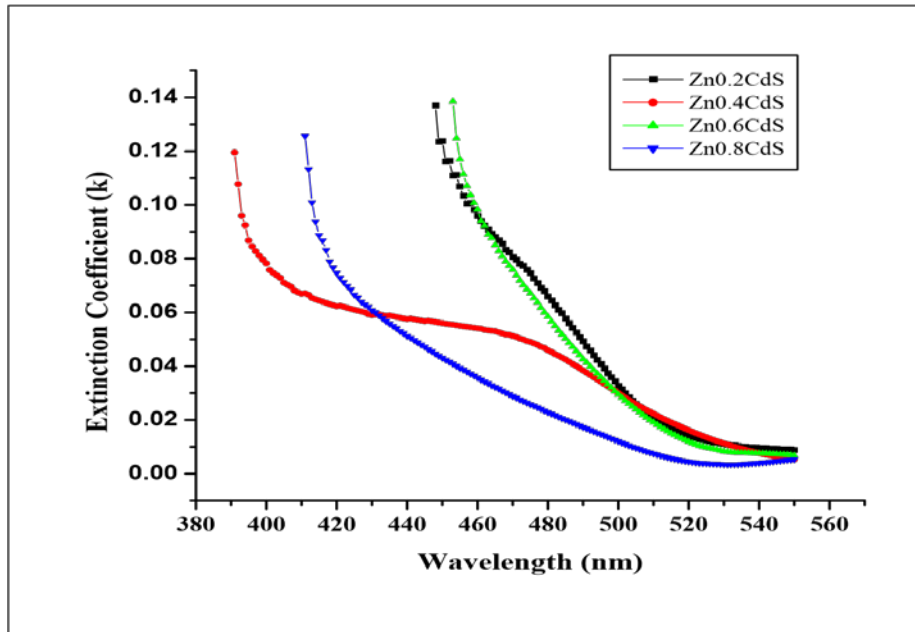


Figure 9: Variation in extinction coefficient of $Zn_xCd_{1-x}S$ thin films with wavelength

3.2. Structural Properties

Information about the structures of zinc doped cadmium sulphide thin films developed on soda lime glass substrates were obtained from “D8 Diffractometer”. Diffraction spectra were obtained through scanning of two theta in series of “10° to 85°”. Typical diffraction patterns of the prepared samples of zinc doped cadmium sulphide having various compositions are illustrated in diagrams 10(a), 10(b), 10(c) and 10(d) respectively.

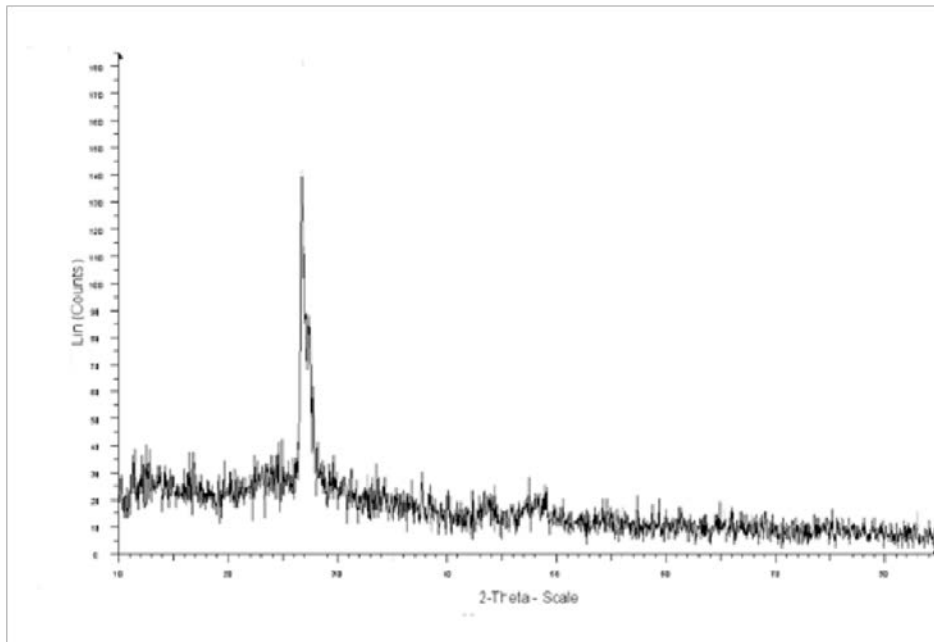


Figure 10 (a): XRD pattern of $Zn_xCd_{1-x}S$ thin film at $x= 0.2$

From diffraction pattern of “Zn_xCd_{1-x}S” thin film at x=.2, it can be seen that one get an intense peak, representing that film has polycrystalline structure. Diffraction peak appeared at $2\theta = 26.744^\circ$ having $d = 3.33075 \text{ \AA}$, which is calculated by using Bragg’s Law ($2d\sin\theta=n\lambda$). This indicates that the thin film has a wurtzite structure with the preferred orientation of the (002) plane, as suggested by JCPDS cards. A strong (002) diffraction line is present in the XRD pattern which indicates that maximum planes are oriented in that direction.

Lattice Constants and Volume of Unit Cell:

The lattice constant “a” and “c” for hexagonal planes of zinc doped cadmium sulphide thin film were found out from x-ray diffraction data by using the following relations [19]:

$$1/d^2 = 4/3(h^2 + hk + k^2 / a^2) + l^2 / c^2 \text{ (for hexagonal)} \quad (12)$$

And

$$c = 1.633a$$

(13)

Where,

$$a = b = 4.07 \text{ hexagonal \AA}$$

And

$$c = 6.66 \text{ hexagonal \AA}$$

Similarly, Volume of the unit cell is given by using the relation [19]:

$$V = (3)^{1/2} / 2 a^2 c \text{ (for Hexagonal)} \quad (14)$$

This is almost 95.54 hexagonal \AA .

Grain Size

The grain size of films from XRD patterns was determined by using Scherrer relation [22]:

$$\text{Grain Size} = k\lambda / \beta \cos\theta \quad (15)$$

Where,

“k” is 0.9, “ λ ” is wavelength of Xray of CuK α radiation and equals to 1.54nm, “ θ ” is the Bragg diffraction angle, and “ β ” is full-width half-maximum width of XRD peak appearing at the diffraction angle θ . The unit of β should be converted into radian. Therefore the above equation takes the form;

$$\text{Grain Size} = 50.97\lambda / \beta \cos\theta \quad (16)$$

The calculated grain size is 130 \AA .

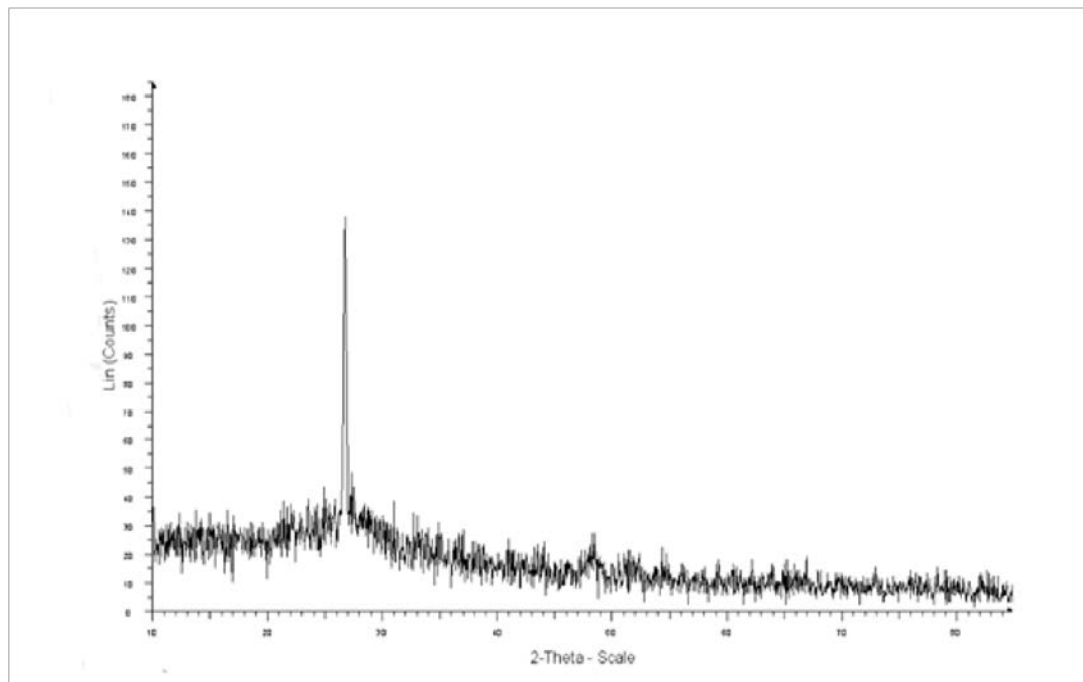


Figure10 (b): XRD pattern of $Zn_xCd_{1-x}S$ thin film at $x=0.4$

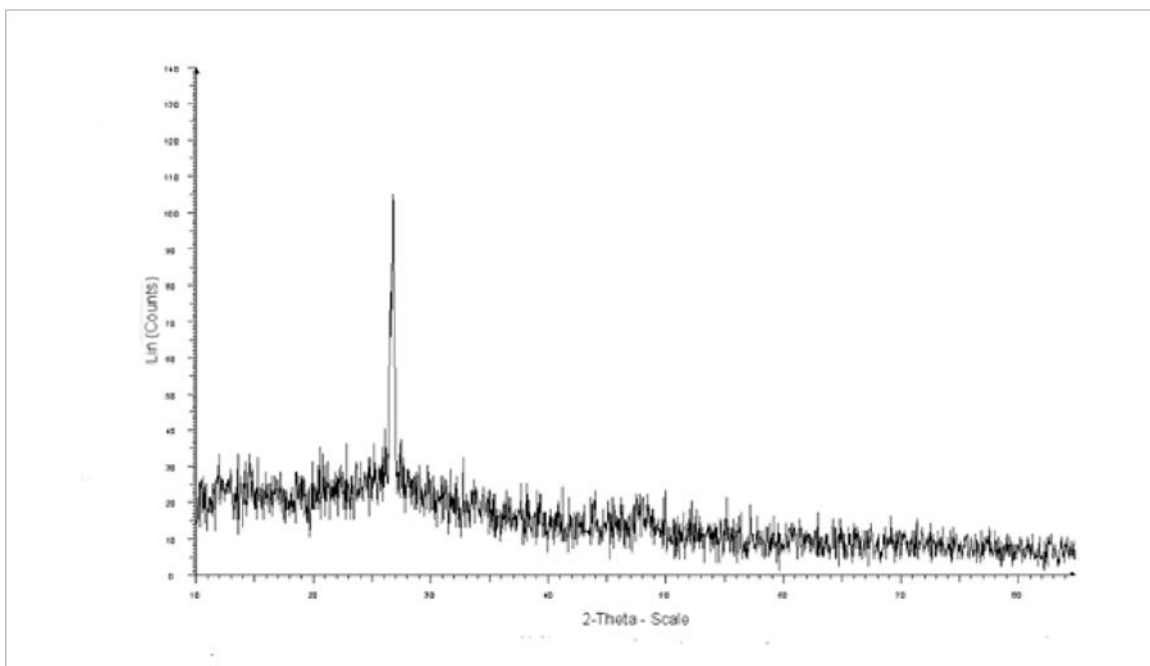


Figure 10 (c): XRD pattern of $Zn_xCd_{1-x}S$ thin film at $x=0.6$

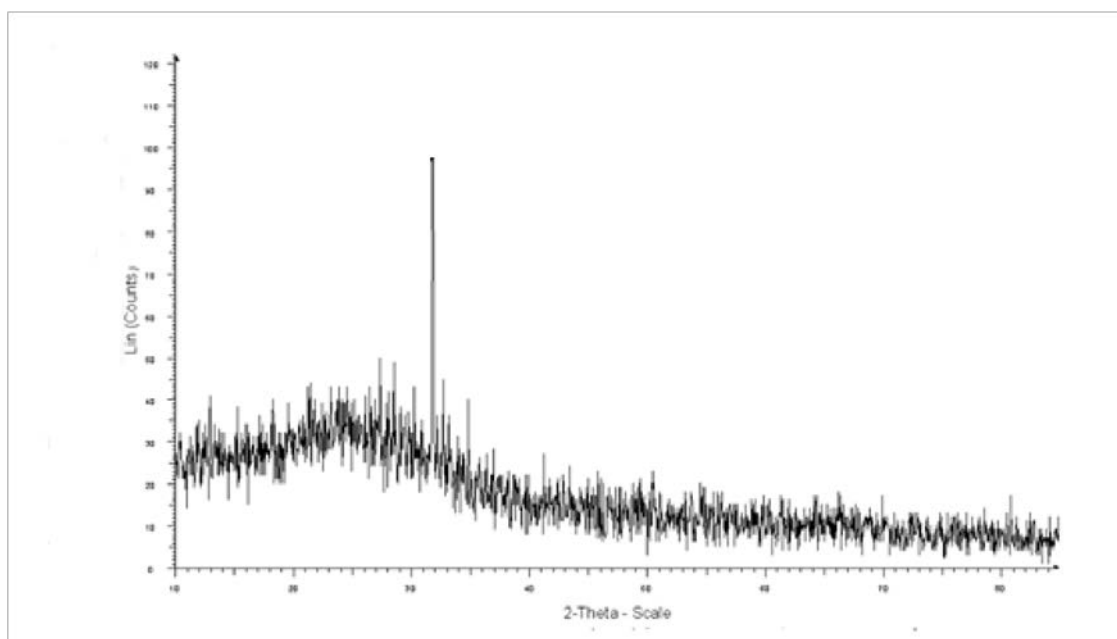


Figure 10 (d): XRD pattern of $Zn_xCd_{1-x}S$ Thin Film at $x=0.8$

Table 2 shows the values of the grain sizes, diffraction angles, Interplaner spacing, volume of the unit cells and lattice constants of $Zn_xCd_{1-x}S$ thin films calculated from equations (12), (13), (14) and (16).

Table 2: Parameters Extracted from XRD Micrographs [33]

Composition (x)	Diffraction angle (2θ) deg	Inter planer spacing (d) Å	Lattice constant „a“ Hexagonal (Å)	Lattice constant „c“ Hexagonal (Å)	Volume of Unit cell Hexagonal (Å ³)	Grain Size Å	Planes (hkl)
Zn _{0.2} Cd _{0.8} S	26.748	3.33075	4.07	6.66	95.54	130	(002)
Zn _{0.4} Cd _{0.6} S	26.683	3.33820	4.1	6.67	97.5	160	(002)
Zn _{0.6} Cd _{0.4} S	26.748	3.33018	4.04	6.6	93.29	110	(002)
Zn _{0.8} Cd _{0.2} S	31.720	2.8198	3.45	5.64	58.1	100	(002)

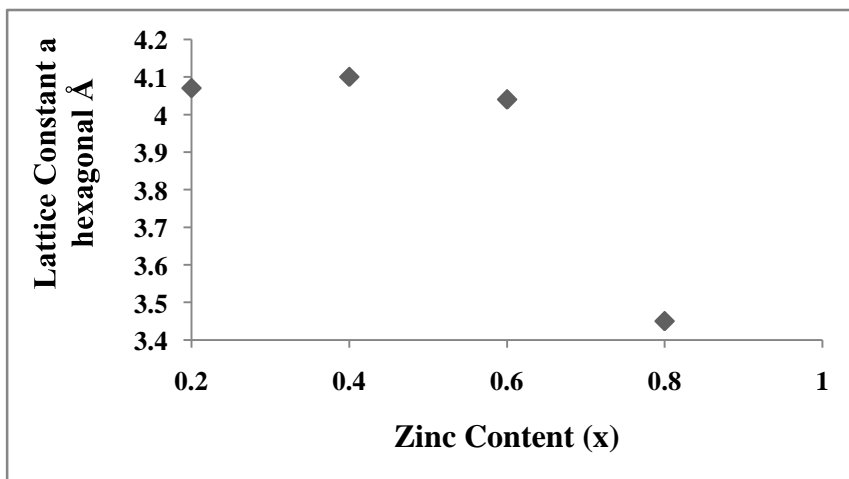


Figure 10 (e): Variation in lattice constant “a” with Zinc content “x”

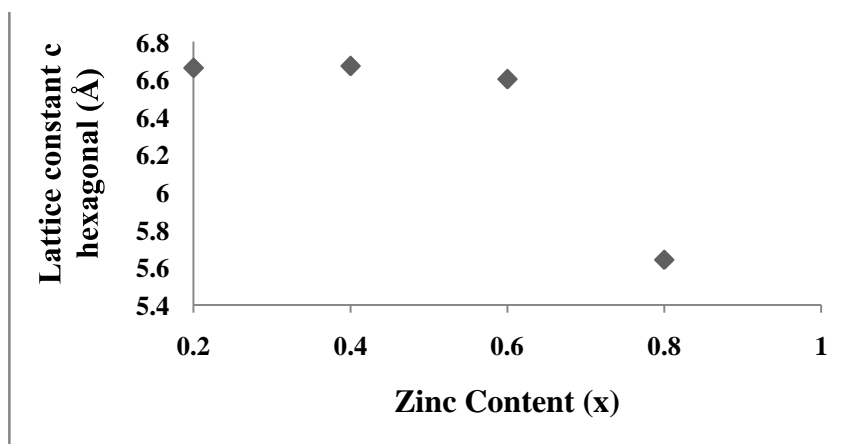


Figure 10 (f): Variation in Lattice Constant “c” with Zinc Content “x”

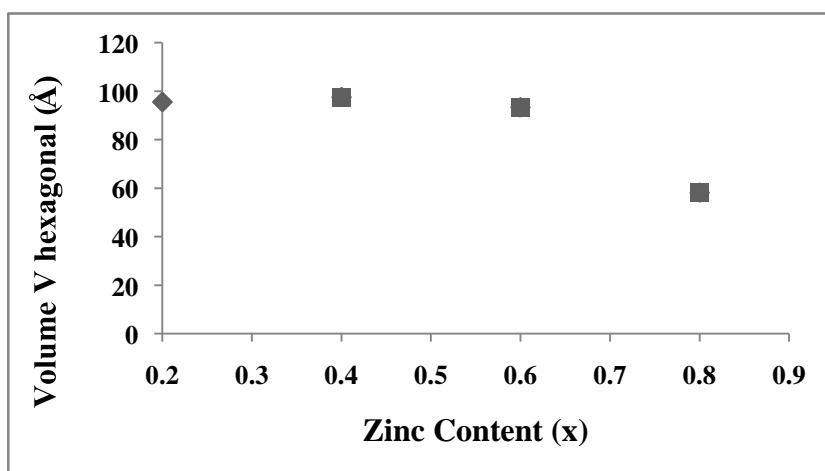


Figure 10 (g): Variation in Volume “V” with Zinc Content “x”

The main peaks for these films occurred between 26.6° – 31.7° and are identified as reflections from (002) planes. From the x-ray diffraction patterns, it is clear that diffraction angle approaches to some extent towards greater angle at the value of $x=0.2, 0.6$ and 0.8 (Increasing Zn contents) in the zinc doped cadmium sulphide thin films except at $x=0.4$. The increasing trends in the diffracting angle is because of the replacement of “ Zn^{2+} to Cd^{2+} ”, in which the ionic radius of cadmium ion (0.095nm) and zinc ion (0.074nm) approaches towards the reduction in “ a ” as well as “ c ”, as the size of lattice constant depends on the size of ion. A gradual decrease in “ a ” and “ c ” with the increasing contents of zinc follow the “Vegard’s Law” [20, 34, 35]. Thin film of “ $Zn_{0.4}Cd_{0.6}S$ ” contradicts the above results, Instead of increasing the diffraction angle with increasing the concentration of Zn contents, its angle decreases and its lattice parameters, grain size and also volume of unit cell increases. The larger grain size implies the good crystallization that indicates less stress in the film. This could enhance the band gap due to the lessening of the crystal defects within the material.

4. Conclusion

In this work thin films of $Zn_xCd_{1-x}S$ ($x=0.2, 0.4, 0.6$ and 0.8) have been successfully deposited on to soda lime glass substrates by using vacuum evaporation technique. UV-VIS studies show that as the concentration of zinc content increases transmission spectra shift towards the shorter wavelength region from (575-526) nm, percent transmittance increases from 81-100% in the visible region and absorption edges shift towards the shorter wavelength region. Also it is observed that thin film of $Zn_xCd_{1-x}S$ at $x=0.4$ covers the larger portion of shorter wavelength region as compared to other composition of zinc contents. The absorption coefficient spectra shift towards the shorter wavelength region by increasing the zinc contents and become high at the value of about 0.0038cm^{-1} . It is also noticed that optical conductivity increases with photon energy and thin film of $Zn_{0.4}Cd_{0.6}S$ has high optical conductivity as compared to other value of

“ x ”. Optical band gap of the zinc doped cadmium sulphide thin films varies non-linearly from 2.55eV ($x=0.2$) to 2.84eV ($x=0.8$) that is band gap increases with increasing the zinc contents of the compound. In the longer wavelength region reflectance is minimum but as the concentration of zinc contents increases reflectance spectra shift towards the longer wavelength region and become high at the value of 0.043. The optical constants such as the extinction coefficient and refractive index is also found to depend upon the zinc concentration in the films. X-ray diffraction studies show that all the thin films have wurtzite structure with a preferential orientation of (002) plane. The main intense peaks of the prepared thin films having various compositions occur between the diffraction angles 26.6° - 31.7° . It also confirms that by varying the composition of zinc content diffraction angle increases but inter planer spacing, lattice constants, volume and grain size decreases obey the Vegard’s Law except for $Zn_{0.4}Cd_{0.6}S$ thin film which has high crystallinity as compared to the other composition of the zinc content. As $Zn_{0.4}Cd_{0.6}S$ thin film has wide band gap, high optical conductivity and good crystallinity, it can be used as a window layer for solar cell, which can reduce the absorption losses of window layer. So this composition of zinc content is considered the best composition for the fabrication of window layer for solar cells.

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