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Optimization of Nanocrystalline Cadmium Sulphide (CdS) Thin Films for Solar Cells

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Abstract: Cadmium chalcogenides with appropriate band gap energy have been attracting a great deal of attention because of their potential applications in photovoltaic devices. CdS in the form of thin films is prepared at different substrate by a simple and inexpensive chemical bath deposition technique. The as deposited thin films have been characterized by XRD, SEM, and Optical techniques and FTIR. The XRD patterns shows that the films are polycrystalline with crystallite size 11 to 121 nm for the film deposited at optimized preparative parameters. SEM studies reveal that the grains are uniform with uneven spherically shaped, distributed over the entire surface of the substrates. FTIR spectrum shows that the percentage transmittance of the films has high transmittance in the range between 65 and 70% in the UV-VIS-NIR regions. Absorption spectrum shows a clear shift to the lower wavelength side over bulk crystallites. The optical band gap energy was found to be 2.57 eV with direct allowed band-to-band transition.

Keywords: Chalcogenides; Chemical Bath Technique; X-ray diffraction; Morphological & Optical properties.

I. INTRODUCTION

CdS is one of the most interesting II–VI semiconductors owing to its interesting optical, electrical and optoelectronic properties. Possessing a wide fundamental band gap, they have been used in a large variety of applications such as electronic and optoelectronic devices [1]. Thin films of CdS hold promise in photovoltaic applications as window coatings in many types of solar cells with absorber materials such as Cu(In,Ga)Se₂ [2], CdTe [3] or CuInSe₂ [4] and for thin film transistors [5]. Furthermore CdS nanocrystals are applied for lasers [6], as biological labels [7], and they are investigated as photo conducting cells in sensors for ultraviolet radiation [8]. In recent years, considerable interest has been shown in the synthesis and photoelectrochemical test of semiconductor thin films. CdS belongs to the II-VI group [9] and it is typically sulphur deficient, possessing the sulphur vacancies with a high electron affinity. This causes CdS to acquire electrons easily, resulting in CdS material n type in nature. Electron hole pairs generated in CdS are well separated with electrons being highly localized [10]. So it is the most studied nanocrystalline semiconductor as a photoanode in photoelectrochemical (PEC) solar cells because of its suitable bandgap, long lifetimes, important optical properties, excellent stability, easy fabrication and numerous device applications. CdS thin films have been prepared by diverse techniques: sputtering [11], vacuum evaporation [12], spray pyrolysis [13], electrode position [14] and chemical bath deposition (CBD) [15].

Among these various techniques, chemical bath deposition is a rather simple and inexpensive technique which enables the production of large area uniform and transparent films with good adherence and reproducibility at close to room temperatures. The technique of CBD involves the controlled precipitation from solution of a compound on a suitable substrate. Factors such as control of film thickness and deposition rate by varying the solution pH, temperature and reagent concentration are allied with the ability of CBD to coat large areas, in a reproducible and low cost process. Another advantage of CBD method with respect to other methods is that the films can be deposited on different kinds, shapes and sizes of substrates. [15]

The main objective of the present work is to developed, the CADMIUM based binary and ternary II–VI compounds n-type of semiconductors CdS thin films by using Chemical Bath deposition (CBD) Technique. Structural, Surface Morphology and optical properties of as deposited CdS films were investigated by XRD, SEM, FTIR and UV-VIS Spectrophotometer. We also studied the effect of Optimization parameters on the deposited films. The results obtained are discussed and compared whenever possible.

II. EXPERIMENTAL

The aqueous solutions of Cadmium Sulphate (CdSO₄) and thiourea ((NH₂)₂CS) were prepared by dissolving appropriate amounts of these salts (A.R. Grade) in double distilled water. The equimolar solutions were mixed together in appropriate volumes to obtain the

Cd: S ratio as 1:1 and then deposited on glass substrates. These substrates were washed with water, then cleaned in acetone and methanol ultrasonically, and finally, again washed with methanol ultrasonically before use. After cleaning the glass slides were kept vertically in a closed beaker with the help of a special holder attached with AC Motor having a constant speed of 60 r.p.m. Add liquid Ammonia in the bath for adjusting the pH of solution which is measured by pH meter and providing the temperature to the solution by heating coil was kept constant with the help of a temperature controller in the range 70°C to 72°C, by keeping all other parameters constant. After the deposition, the CdS films were washed with methanol ultrasonically to remove the loosely adhered CdS particles on the film and finally dried in air. The same procedure is repeated for different time durations [16, 17, 18].

The as deposited thin films of CdS were characterized for structural, morphological and optical properties. The CdS film thickness was measured using gravimetric weight difference method by assuming bulk density of corresponding materials. X-ray diffraction (XRD) patterns of the film were recorded on X-ray diffractometer (XPERT-PRO) by using Cu-K α lines ($\lambda = 1.54 \text{ \AA}$) for Cu K α radiation for the diffraction angle range 0–60°. The surface morphology and composition was studied by scanning electron microscopy. To study the optical characteristics of the film, absorbance spectra were recorded in the range 200–800 nm by means of UV-VIS-NIR spectrophotometer (Perkin Elmer: Lambda 35).

III. RESULTS AND DISCUSSION

A. Film Structure Studies

It has been reported that CdS may have either cubic or hexagonal crystal structure depending on the synthesis condition such as deposition temperature and precursor concentration. The as-grown CdS thin films were characterized by the XRD technique scanned in the 2θ range of 0–70°. The diffractograms were obtained for the films grown on the amorphous glass substrates (Figure 1).

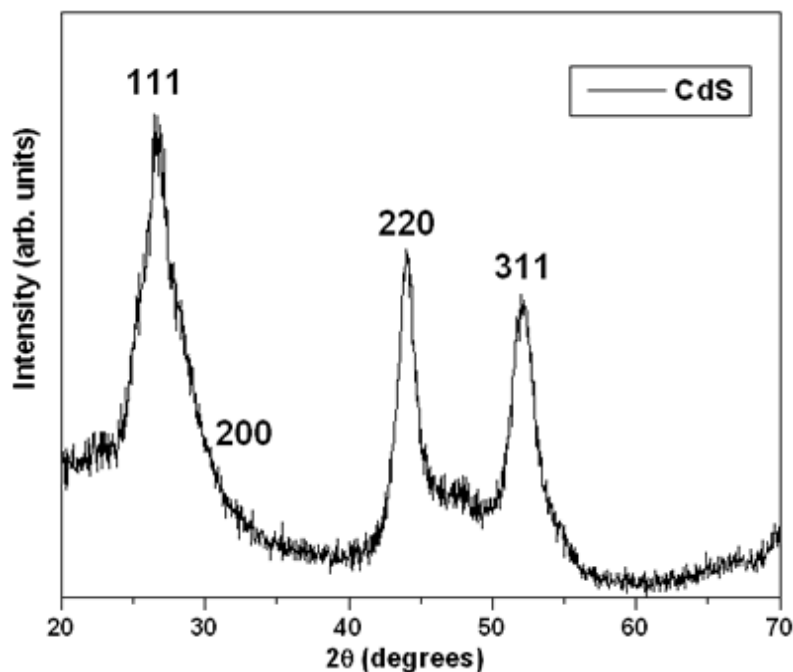


Figure 1: XRD of CdS thin films

The diffractograms depict that the deposits are polycrystalline in nature. It is seen that the plane (1 1 1) of CdS appears with higher peak intensity in all the diffractograms. A well matching of the observed and the standard d-values from JCPDS data card [19-20] confirms the formation of compound CdS with mixed cubic and Hexagonal crystal structure. The calculated values of lattice constant with cubic crystal structure are found to be $a = b = c = 5.8200 \text{ \AA}$ agreeing well with the standard values CdS [20]. The grain size was calculated for all cases for the reflections from the C (111) plane by using the well known Debye-Scherrer formula,

$$D = 0.9\lambda/\beta\cos\theta \text{----- (1)}$$

where D is the particle size, β is the full width broadening of the diffraction line measured at half of its maximum intensity in radians (FWHM), λ is the X-ray wavelength (1.5406 \AA) and θ is Bragg diffracting angle. From particle size analysis it is clear that the films are nanocrystalline in nature. The grain sizes were found to be within the range of 11 to 121nm. [21-24]

B. Surface Morphology Studies:

The surface morphology of deposited CdS thin films were investigated by SEM at different magnifications as shown in Figure 2.

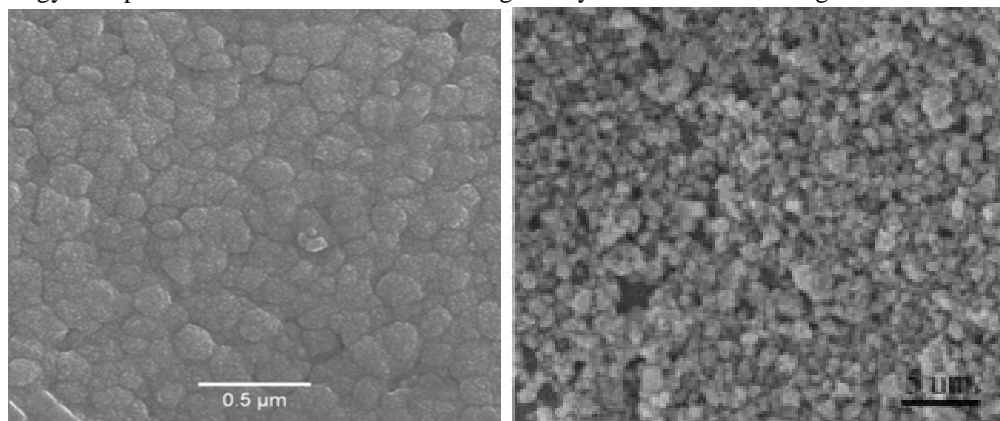


Figure 2. Scanning Electron Micrograph of as deposited CdS thin film at different magnification.

The deposits are compact, pinhole free with spherical grains from few nanometers up to clusters of 157 nm and the films are well covered on the substrate. From the figure, it is observe that the small nanosized grains engaged in a fibrous- like structure, which clearly indicates the nanocrystalline nature along with some amorphous phase of CdS thin films. The average grain size of the CdS nanoparticles is about 130 nm. This result is in consistent with results obtained from X ray diffraction studies [25-27].

C. Optical Absorption Studies

The optical absorption spectrum of the deposited CdS thin films on glass substrate was studied in wavelength range 200 to 800 nm. The nature of the transition involved (direct or indirect) during the absorption process was determined by studying the dependence of the absorption coefficient α , on photon energy $h\nu$ as [28]

$$\alpha h\nu = A (h\nu - E_g)^n \text{ -----(2)}$$

Where A is the constant, E_g is the bandgap energy, $h\nu$ is the photon energy, $n = \frac{1}{2}$ or 2 for direct or indirect transition. The value of absorption coefficient is found to be of the order of 10^4 cm^{-1} . The optical data was further analyzed to determine the nature of transition that takes place in CdS thin film. The plots of $(\alpha h\nu)^2$ versus $h\nu$ are shown in the Figure 3.

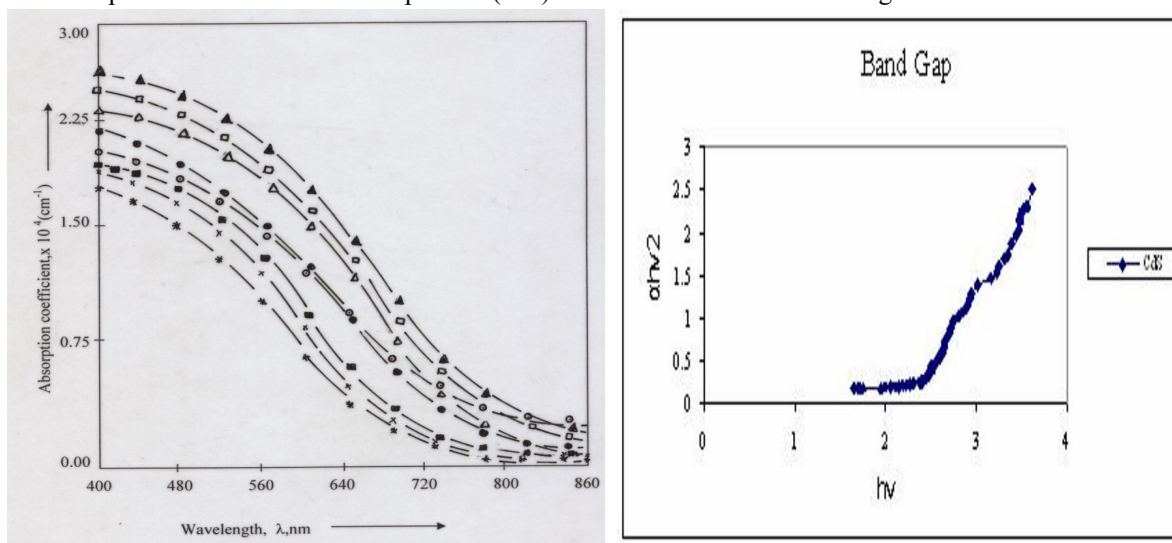


Figure 3. Plots of $(\alpha h\nu)^2$ versus $h\nu$ of as deposited CdS thin films on glass substrates.

The straight line nature of the graphs supports the direct bandgap nature of the semiconductor. The straight-line portion was extrapolated to the energy axis at $\alpha = 0$, to obtain the band gap of CdS thin films. The direct optical band gap of the CdS film estimated to be 2.57 eV agrees well with the reported value for CdS material [28].

D. IR Studies

IR spectra of CdS nanoparticles, is presented in figure 4.

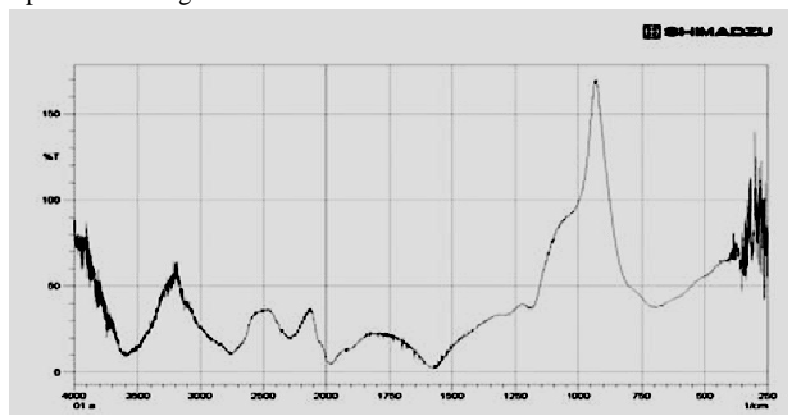


Figure 4. IR spectra of CdS thin film deposited on glass substrate.

The IR frequencies along with the vibrational assignments for CdS nanoparticles are given in table 1. The band at 3584.82 cm^{-1} is due to O-H stretching vibrations of water molecules. The band at 2746.73 cm^{-1} are due to C-H stretching vibrations. The bending vibrations of C=N appeared at 1574.93 cm^{-1} . CdS particles showed two stretching bands of C-O at 1182.4 and 1282.71 cm^{-1} . Trace amount of SO_4^{2-} as impurity is seen as there are small absorptions around 1018.4 cm^{-1} . At 668.36 cm^{-1} and 703.8 cm^{-1} , there are medium to strong bands which have been assigned to Cd-S stretching. The vibration absorption peak of the Cd-S band is at 262.33 cm^{-1} .

Table 1: IR frequencies with vibrational assignments of CdS nanoparticles

Positions (cm^{-1})	Intensities	Assignments
3584.82	Strong	O-H stretching
2746.73	Strong	C-H Stretching
1574.93	Strong	C=N stretching
1182.4	Doublet	C-O stretching
1282.71	Medium	
1018.4	Weak (trace)	SO_4^{2-}
668.36	Doublet	Cd-S stretching
703.8	Medium	
262.33	Medium	Cd-S Stretching

E. Temperature Optimization

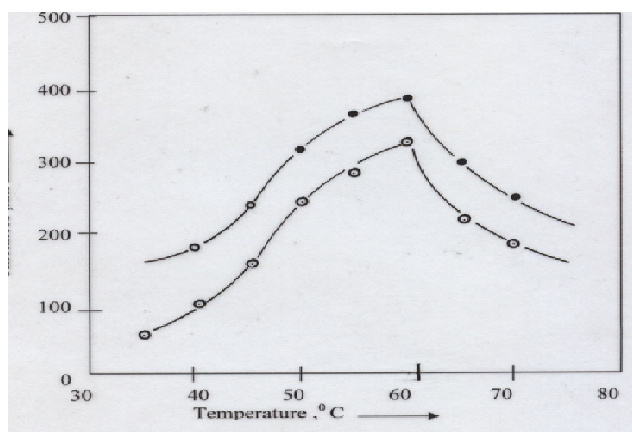


FIGURE 5 : Variation of film thickness as a function of deposition temperatures.

Figure 5 shows the variation of film thickness as a function of deposition temperatures. Various temperatures from 30°C to 70°C in steps of 10°C were used in the depositing CdS to obtain the optimal temperature. Figure 1 shows an increase in film thickness as the temperature increases from 313°K to 345°K. No film formation at room temp.; Ions in bound state, IP does not Exceed SP, At moderate temp, less thickness and at 60°C, max. thickness (312nm), ions get sufficient time to condense, above 60°C, thickness decreased, more and more ions released. The rise in the film thickness may be due to the dissociation of the Cadmium-complexes (with ammonia and with thiourea) and increase in the hydrolysis of $SC(NH_2)_2$ as the temperature increases. At higher temperatures, the decomposition of thiourea will be faster setting more S^{2-} ions free. Additionally, the Cadmium-complex dissociation is greater and gives higher concentrations of free Cd^{2+} in the solution, which in turn results in higher deposition rates or in other words for a constant buffer layer thickness, in shorter deposition times. Also the kinetic energy of the ions in solution is higher at higher temperature, which brings about increased interaction between them and subsequent deposition at volume nucleation centers of the substrate. It can be seen from figure, the average growth rate is high at 345°K (9.478nm/min) and it is slow at 313°K (7.622nm/min).

IV. CONCLUSIONS

The n-type semiconductor thin films of CdS have been successfully deposited by simple and inexpensive Chemical Bath Deposition Technique. Polycrystalline nature of as deposited thin films was predicted from X-ray diffraction studies. Scanning electron microscopy studies revealed uniform deposition with the average grain size of 130 nm. The UV absorption studies on films clearly show an increase in band gap with reduction in particle size as compared to bulk materials, which supports the formation of nanocrystallites in these films. The optical band gap was found to be 2.57eV with direct allowed transition. FTIR spectroscopy showed that the bonding peaks and the percentage transmittance of the films were found to have high transmittance in the range between 65 and 70% in the UV-VIS-NIR regions; hence they could be effective for window layers of solar cells. From the optimization study it is clear that the average growth rate is high at high temperature and low at lower one.

V. ACKNOWLEDGEMENT

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