

## Influence of Bath Temperature on Structural, Optical and Electrical Properties of Cadmium Sulfide Thin Films Prepared by Chemical Bath Deposition

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**Abstract.** The effect of bath temperature (60–90 °C) on structural, optical and electrical properties of CdS thin films deposited by chemical bath deposition (CBD) at a constant precursor concentration and deposition time was studied. From the XRD analysis, it was found that the structure of CdS thin films varied with temperature. At lower temperature hexagonal structure was dominated while at high temperature, the cubic structure was prominent. The band gap of the as-prepared CdS thin films was calculated from the UV-Vis spectroscopic data, and it was found to be decreased with the increase of temperature. The resistivity of the CdS thin films also decreased with the increase in temperature.

**Keywords:** cadmium sulfide, CBD, temperature effect, structural change, growth mechanism, electrical properties.

### INTRODUCTION

CdS is an n-type semiconductor that belongs to the II-VI group of compounds with a direct band gap of 2.42 eV. CdS is a potential material for visible and near-infrared range applications. Particularly, it can be used in solar cells and photodetectors. For photovoltaic application CdS thin films must have appropriate optical absorption, high transparency, high photoconductivity and low film thickness to avoid absorption in the window layer [1]. Now a day's many of the researchers focused on Cadmium Sulphide (CdS) thin films because of excellent properties such as high carrier concentration ( $10^{16}$ – $10^{18}$  cm<sup>-3</sup>) and mobility (0.1–10 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup>), tunable band gap (2.1–2.45 eV), high electrochemical stability and high absorption coefficient ( $> 10^4$  cm<sup>-1</sup>) compared to bulk CdS counterparts [2]. Cadmium sulfide, in polycrystalline form, provides the best photovoltaic characteristics in CIGS- and CdTe-based thin film solar cells, respectively as buffer layer and window layer material. The maximum the-

oretical limit of photovoltaic conversion efficiency for CIGS and CdTe-based thin film solar cell with one hetero-junction is higher than 30 %, but the cell efficiency over 20 % was reached now. There is scope to increase the cell efficiency further 20 % by reducing the thickness of CdS window layer below 100 nm.

One of the most common methods for deposition of CdS layers is the Chemical Bath Deposition (CBD). This method is suitable for deposition of thin films over a larger area. It is also rather simple and cheap. At the same time, it is known that deposition temperature, availability of impurities, qualitative characteristics of substrates, etc. can affect the properties of CdS thin films. The thickness of CdS layer can be controlled by tuning the deposition parameters, particularly, the bath temperature showed greater influence on the film thickness due to changes in the reaction kinetics.

Therefore, the main purpose of this work is to study the influence of deposition temperature on the structural, optical, and electrical properties of CdS thin films deposited by CBD method at

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a constant precursor's concentration and deposition time.

## EXPERIMENTAL

### Deposition of CdS thin films

CdS thin films were prepared by using chemical bath deposition method. For the preparation of CdS thin films, the pre-cleaned and dried glass substrates were inserted vertically into the bath containing 0.0029 mol/l of  $\text{CdSO}_4$ , 0.15 of mol/l of  $\text{CS}(\text{NH}_2)_2$  and 1.72 mol/l of  $\text{NH}_3 \cdot \text{H}_2\text{O}$  and then deposition was carried out at various deposition temperatures in the range of 60–90 °C at a constant deposition time of  $t = 20$  min. Fig. 1 shows the arrangement of chemical bath deposition.

### Characterization

The structural properties were analysed using X-ray diffraction (XRD) (with  $\text{Cu-K}\alpha$  radiation, diffractometer ADP-2-01). In view of the low thickness of deposited films (50–200 nanometers), the XRD analysis is complicated. Therefore, the multiple depositions were carried out under the same conditions described above. The optical band gap of CdS thin films was calculated from the absorbance spectra, measured using Shimadzu UV-Vis 3101PC spectrophotometer in the wavelength range of 200–1700 nm. To determine the electrical resistivity of CdS thin films, indium contacts were deposited by thermal evaporation using an aluminum mask and then the potential sweep was applied to the contacts and the corresponding current was measured.

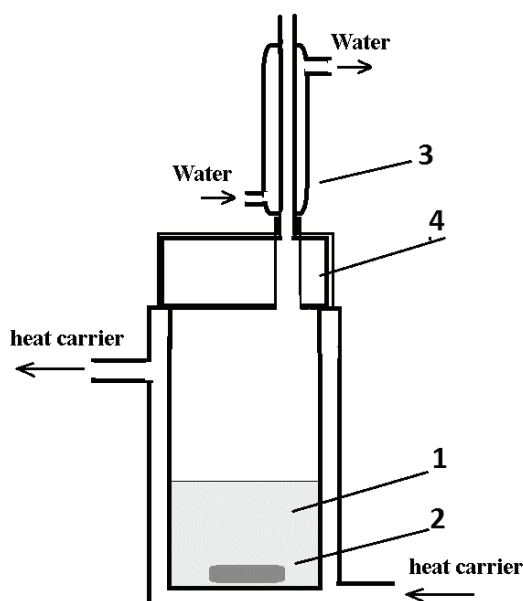


Fig. 1. Schematic of CBD thin film deposition for solar cell preparation

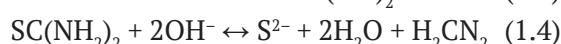
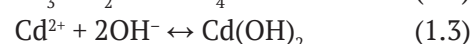
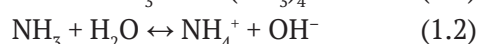
## RESULTS AND DISCUSSION

### Growth mechanism of CdS thin films

In the deposition of CdS thin films, ammonium hydroxide acts as the complex agent and it forms cadmium ammonia complex as shown below:



The formation of cadmium ammonia complex in solution provides the slow release of cadmium ions. Generally, the growth mechanism of CdS thin film using ammonia can be presented in the following reactions:



During deposition of CdS thin films, clusters were formed and their size differs depending on deposition temperature.

### Deposition temperature versus film thickness

Fig. 2 shows the dependence of film thickness on the deposition temperature. As shown in Fig. 2, the thickness of film was increased from 30 nm to 90 nm when the bath temperature was varied in the range, 60–80 °C and then it decreased at 90 °C. This is because of the increased growth rate with rise of deposition temperature until the film thickness reached an upper limit, thereafter it decreased. The decrease of film thickness after 80 °C was due to the dissolution of unstable nuclei adsorbed on the film surface, or after reaching a certain thickness film starts to peel off. A similar type of observation was also reported in literature by few investigators [2–4]. For example, Fangyang et al. [3] ob-

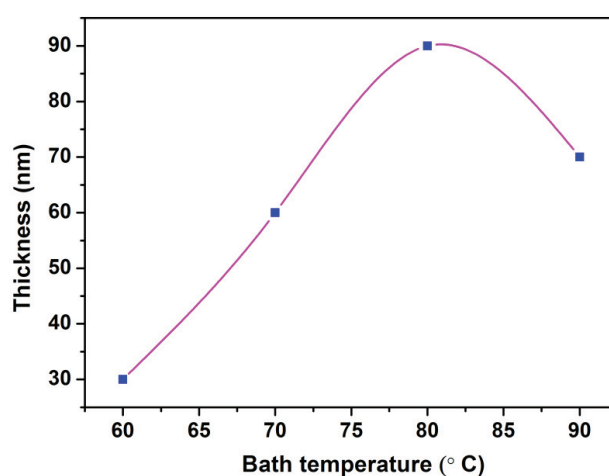


Fig. 2. Variation in the film thickness for different bath temperature

served that film thickness was increased for bath temperature in the range, 55–75 °C and then decreased in the range of 75–85 °C whereas Harietch et al. [4] reported that the thickness was increased for temperature that varied from 55 to 65 °C and then decreased when the temperature varied in the range, 65–75 °C.

### Structure analysis

Fig. 3 shows the XRD pattern of as-prepared CdS thin films grown at different bath temperatures. The lines in XRD spectra were found to be weak because of the low film thickness of CdS. From the XRD pattern (Fig. 3), it can be seen that CdS thin film prepared at 60 and 70 °C showed the diffraction peaks at 27.7°, 36.2°, 41.5°, 54.4°, 56.9°, and 69.1°, which respectively correspond to the (101), (102), (110), (004), (202), and (210) planes of hexagonal structure of CdS. However, for bath temperatures > 70 °C (i.e. for 80–90 °C), XRD pattern of CdS films showed the mixed structures of both hexagonal and cubic. The diffraction peaks observed at 30.1°, 54.6° and 64.6° correspond to the (200), (222) and (400) planes of cubic structure respectively. All the observed planes were well matched with the JCPDS card nos. 75–1545 and 75–0581 of hexagonal and cubic structure, respectively.

As seen from Fig. 3, CdS thin film showed hexagonal structure for bath temperatures of 60 and 70 °C, whereas at bath temperatures of 80 and 90 °C, mixed structures of hexagonal and cubic were ob-

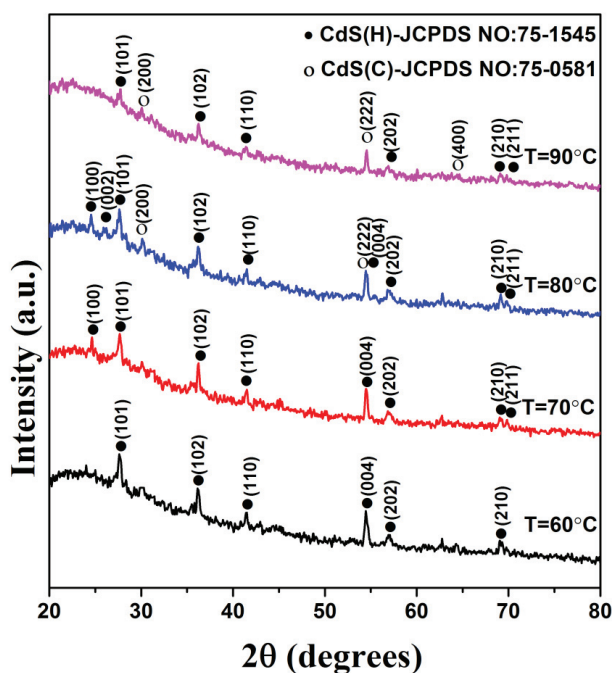


Fig. 3. X-ray diffraction pattern for as prepared CdS thin films at different bath temperatures

served. The observed structural changes were due to the following reasons. At lower temperatures, below 70 °C, ion-to-ion deposition process might take place during the growth of the film whereas the cluster-by-cluster process might occur for temperature > 70 °C. Therefore, due to cluster by cluster process at bath temperatures  $\geq 80$  °C, the mixed structures of hexagonal and cubic CdS was observed. A similar type of structural change during the growth process was also reported by other researchers in the literature [5, 6]. For example, George et al. [7] prepared CdS thin films by CBD using sequential mixing of 25 ml of 0.1M cadmium nitrate, 20 ml of 1 M sodium citrate, 7.5 ml of 4 M aqueous ammonia, 10 ml of 1 M thiourea and 37.5 ml of deionized water. The deposited CdS films showed hexagonal structure at a bath temperature of 60 °C. On the other hand, Oliva et al. [8] deposited CdS thin films by the chemical bath method using an aqueous solution of cadmium chloride ( $\text{CdCl}_2$ ), potassium hydroxide (KOH), ammonium nitrate  $\text{NH}_4\text{NO}_3$ , and thiourea ( $\text{CS}[\text{NH}_2]_2$ ). The deposited CdS films showed cubic structure at 75 °C. Lejmi et al. [9] deposited CdS thin films by CBD onto ITO-coated glass substrates using aqueous solutions of  $3.5 \cdot 10^{-2}$  M  $[\text{Cd}(\text{CH}_3\text{COO})_2]$ ,  $3.5 \cdot 10^{-1}$  M  $[(\text{NH}_2)_2\text{CS}]$ ,  $10^{-1}$  M  $(\text{NH}_4\text{Ac})$ , 0.3 M  $(\text{NH}_4\text{OH})$  containing  $10^{-5}$  M solutions of HPA or IPA. The deposition of CdS was carried out at 90 °C. XRD results of the deposited films indicated both cubic and hexagonal structures of CdS. The formation of cubic or hexagonal CdS phase depends on many factors including the deposition technique. In chemical bath deposition, the structure of the film depends upon bath temperature, composition and pH of the solution. In the present case, the rise in bath temperature led to a change in the deposition process, which changes the phase structure of CdS films.

### Optical properties

The absorbance of the material was defined as the logarithmic ratio of the amount of radiation falling on the material to the amount of radiation transmitted through it [10]. Fig. 4 (a) shows the absorbance spectra of CdS thin films prepared at various deposition temperatures. The absorbance of CdS thin films decreased with increase in the bath temperature due to an increase of film thickness as mentioned in section 3.2. (Fig. 2). The deposited CdS thin films showed a sharp absorption edge in the wavelength range of 500–550 nm with temperature. A shift in the absorption edge of CdS towards lower wavelength side was observed with increase of bath temperature due to the quantum size effect. The observed low optical absorption in the visible to near IR region for all films, (except the film pre-



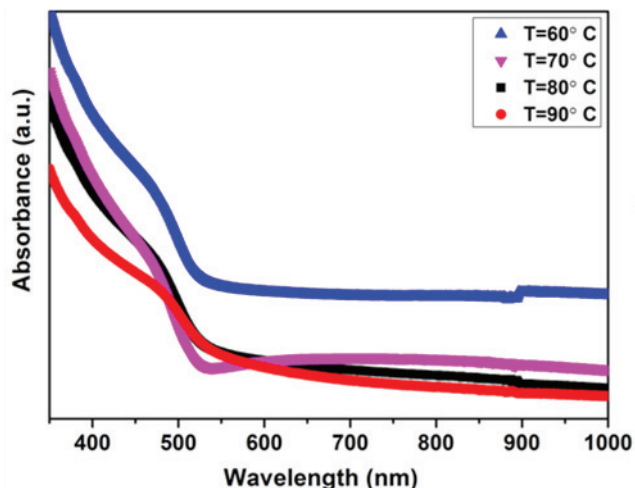


Fig. 4. Absorbance spectra of CdS thin film deposited at different deposition temperatures

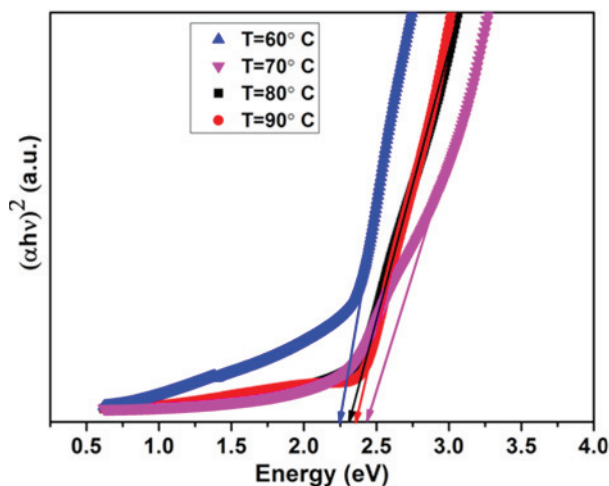


Fig. 5. Plot of  $(\alpha hv)^2$  versus  $hv$  of CdS thin film deposited at different deposition temperatures

pared at 60 °C), indicates high transparency in this region, which is the desired for CdS to prove as a good buffer layer in solar cells.

Tauc plots were used to estimate the energy band gap ( $E_g$ ) of CdS thin films that were drawn using the relation of  $(\alpha hv)^{1/n} = A(hv - E_g)$ , where A is a constant,  $\alpha$  is the absorption coefficient,  $hv$  is the photon energy, and  $n$  is equal to 1/2 for direct and 2 for indirect allowed transitions. Fig. 5 shows the relationship between  $(\alpha hv)^2$  and  $hv$  for CdS films prepared at different bath temperatures. The  $E_g$  value can be obtained by extrapolating the straight line onto the energy axis whose intercept onto the x-axis gives the optical band gap as shown in Fig. 5. The evaluated energy band gap of the films varied in range, 2.25–2.45 eV with the change of bath temperatures. The films deposited at 60, 70, 80 and 90 °C had the energy band gaps at 2.25, 2.44, 2.35, and 2.3 eV, respectively. These band gaps are in good agreement with the values reported in literature [11–14]. In general, the change in band gap is due to the influence of various factors like variations in structural parameters such as grain size, lattice strain, impurities, changes in the stoichiometric ratio Cd/S and variations in carrier concentration in the film [15–19]. In the present case, the band gap of CdS thin films was increased from 2.25 to 2.44 eV with the rise of bath temperature up to 70 °C and thereafter it decreased to 2.3 eV. This change in  $E_g$  was mainly attributed to the structure change from hexagonal structure to cubic in the film.

As seen from the XRD analysis, at low bath temperatures, ion- to-ion growth process took place that favored the hexagonal structure. At high temperatures, cluster-by-cluster growth process oc-

curred in the films, which supported the cubic structure. A similar type of trend in the band gap with the change of bath temperature was also reported by others. Kumar et al. [1] prepared CdS nanofilms formed by CBD method for different deposition temperatures that vary in the range 50–90 °C. They observed an increase in the band gap of CdS nanofilms in the bath temperature range between 50 and 70 °C and thereafter the band gap decreased for the bath temperatures, 80 and 90 °C. Similarly, Sachin et al. [2] prepared CdS thin films by CBD method for different deposition temperatures i.e. 65, 75, and 85 °C. They observed that the band gap of CdS films was increased with bath temperature between 65 to 75 °C and afterwards it decreased for bath temperatures >75 °C.

### Electrical properties

Fig. 6 shows the variation in electrical resistivity of CdS thin films with bath temperature. It can be seen from Fig.6 that electrical resistivity of as-prepared CdS thin films was decreased with increase of bath temperature from 60 to 80 °C and thereafter it increased slightly. The decrease in electrical resistivity of CdS films was due to either interstitial Cd ions or sulfur vacancies existing in the film. These defects might acted as donors thereby resulting in an increase of the carrier concentration. Consequently, the electrical resistivity of CdS was decreased [20]. In addition, the variation in the structure of CdS films with bath temperature was also responsible for the observed variation in the resistivity.

A similar type of trend in the electrical resistivity of CdS films was also reported by Liu et al.

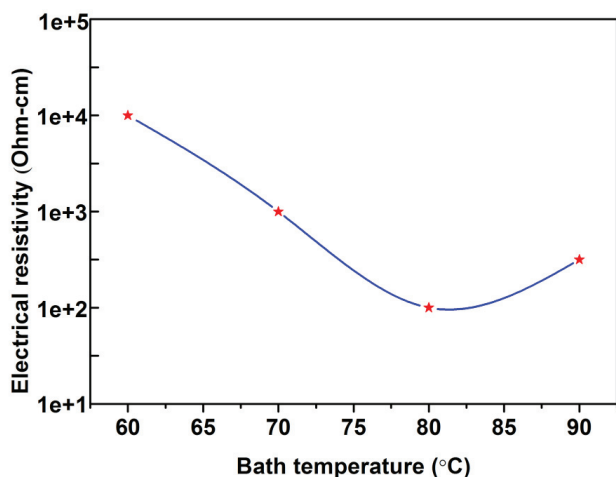


Fig. 6. Dependence of electrical resistivity on bath temperature

[21]. They deposited CdS thin films by CBD using a chemical bath containing 0.03 M cadmium sulfate ( $\text{CdSO}_4 \cdot 8\text{H}_2\text{O}$ ), 0.1 M thiourea ( $\text{H}_2\text{NCSNH}_2$ ) and 25–28% ammonium hydroxide ( $\text{NH}_4\text{OH}$ ). The deposition of CdS thin film was carried out at various temperatures in the range of 55–85 °C. The XRD pattern of CdS film deposited at 55 °C showed hexagonal structure whereas an increase of bath temperature to 65 °C promoted the cubic-hexagonal transformation in CdS. Consequently, the measured electrical resistivity of CdS films was decreased from  $5 \cdot 10^5 \Omega \text{ cm}$  to  $8.5 \cdot 10^3 \Omega \text{ cm}$  with the variation of bath temperature from 55 to 85 °C. Thus, in the present case, the decrease of electrical resistivity in CdS films can be attributed to the concentration of defects present in the films [3, 21–22] as well as the structural changes observed with bath temperature as described earlier in the XRD analysis. The increase of electrical resistivity of CdS films at 90 °C was due to the peeling of the film after reaching a limited thickness of 90 nm.

### CONCLUSIONS

The effect of bath temperature on the structural, optical, and electrical properties of CdS thin films formed by CBD process were investigated. It was found that the as-prepared CdS thin films showed hexagonal structure at lower temperature and mixed (hexagonal and cubic) structure at higher temperatures. The band gap energy of CdS films varied in the range of 2.25–2.45 eV with the change of bath temperature. The electrical resistivity considerably decreased with the increase of deposition temperature. The main reason for these changes in structural, optical, and electrical properties was attributed to change in the growth mechanism of CdS thin films with deposition temperature.

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### CONFLICT OF INTERESTS

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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## Влияние температуры химического осаждения на структурные, оптические и электрические свойства тонких пленок сульфида кадмия

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**Аннотация.** Изучено влияние температуры синтеза (60–90 °С) на структурные, оптические и электрические свойства тонких пленок CdS, осажденных методом химического осаждения (CBD) при постоянной концентрации прекурсора и времени осаждения. Из рентгеноструктурного анализа было установлено, что структура тонких пленок CdS изменяется в зависимости от температуры. При более низкой температуре преобладала шестигонная структура, а при высокой температуре – кубическая. По данным спектроскопии в области УФ и видимого диапазона была рассчитана ширина запрещенной зоны конечных тонких пленок CdS, и установлено, что она уменьшается с повышением температуры. Удельное сопротивление тонких пленок CdS также уменьшалось с увеличением температуры синтеза.

**Ключевые слова:** сульфид кадмия, метод химического осаждения (CBD), температурный эффект, структурные изменения, механизм роста, электрические свойства.

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