# ZnCdS Thin Film Chalcogenide by Chemical Bath Deposition Method

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## Abstract

Chalcogenide ZnCdS thin films preparation by chemical bath deposition method is currently considerable interest because it is relatively inexpensive, simple and large area deposition technique. Different substrates are used since these are low temperature process which avoids oxidation and corrosion behavior of substrate. Photoelectrochemical solar cells, photoconductors, detectors, sensors, etc. are some of the applications of chalcogenide ZnCdS thin films. The effect of zinc content on structural, morphological and optical properties have been reviewed. Further, the changes occurred after annealing of the film at different temperatures and different time have also been incorporated and discussed in this article. In the present review, we have described in detail chemical bath deposition method for chalcogenide ZnCdS thin films. The method is capable of yielding good quality thin films. Their preparative parameters, structural, morphological and optical properties have been discussed in this review article.

**Keywords**: Metal chalcogenide, semiconductors, thin film, chemical bath deposition, solar cells

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## **INTRODUCTION**

During the recent decades, electronics is more central to our lives. Semiconducting materials are the building blocks of the entire electronics that affected many aspects of the technological society. Semiconductors are helpful for our everyday life and can be used by our society. John Koenigsberger classified solid materials into metals, insulator as well as conductor and J. Weiss introduced the word 'Semiconductor' in 1910 [1]. A substance which have crystalline structure and at room temperature it consists of few free electrons is referred to as semiconductors. In early generation, semiconductors are the heart of modern electronics including transistors, solar cells, light-emitting diodes, digital and analog integrated chips/microchip, circuits, computers, etc.

The growing importance of semiconductor devices is leading to challenging in the fabrication to produce cheaper and more reliable devices. Any imperfection can change the behavior of semiconducting materials. The development of cubic or hexagonal phase depends on many features including the deposition techniques [2]. John Barden expressed the behavior of semiconductor whose properties change dramatically based on the tiny amounts of impurities [3]. Semiconductor material is one whose electrical properties lie between an insulator and conductors. The resistivity of semiconductors is less than an insulator but greater than conductor and it have bandgap from 1–4 eV but it can go above 5 eV. The process adding impurities of to а semiconductor is known as doping. The effect of impurities on structural and optical properties of compound semiconductors are discussed [4-6]. Semiconductors are used in many devices for amplification, switching and energy conversion. Si and Ge are commonly used semiconductors but there are various semiconducting materials used for number of devices. From the past decades, the fabrication of nanocrystalline material is interested due to their unique physical and chemical properties. Among these, high surface-to-volume ratio is considerable and could lead to new atomic arrangements that affect the optical properties of nanomaterials [7]. The material which have one and two dimensions are in scale is referred to as 'quantum well' and 'quantum wire'.

The size reduction of semiconductor in which all the three dimension in low range of nanometer is called 'quantum dots'. Nanocrystalline materials can be classified into three categories:

- 1. Lamellar structures
- 2. Filamentary structure
- 3. Equiaxed structures

Nanocrystalline semiconductors exhibit the properties which lie in between bulk crystals and molecules [8]. Their electrical and optical properties are associated with quantum confinement of charge carriers leading to the blue shift of the bandgap with the shrinkage of their size [9]. When the dimension of the semiconductor quantum dots reduced close to the characteristic length referred to as exciton Bohr diameter, these characteristic lengths are usually in the range of few nanometers. There will be large change in their properties, this effect changes the surface-to-volume ratio and shifts electronic energy levels towards higher energy leading to an increase in the bandgap [10]. Nanocrystalline material properties are shape dependent, tenability in optical, electronic and catalytic properties [11–13].

Nowadays, fabrication of nanocrystalline thin film in basic research is considered as the backbone of solid state technology because of its applications in the field of photovoltaic cells, electronic components, photoconductors, sensors, optical filters, solar cells, etc. [14]. In modern era of technology, thin film have been interested for developing high efficiency solar cells and electric power with conventional source without emission of "greenhouse gases". Thin films are classified into two parts:

- 1. The layer and substrate where the films are deposited on it.
- 2. It can be composed of different layer such as thin film solar cells, electrochromic cells, etc.

According to International Energy Agency, electricity is dependent on coal, gas and oil for co-operation and development. Sun sends a mixture of heat and light in the form of energy on earth surface and both are essential for living things. Energy crisis can be minimized by using solar cells. Solar energy as one of the non-conventional source of energy and regenerated energy is abundant as well as pollution free. In 1839, E. Becquerel invented solar cell and defined that it is a device that captured sunlight and converted into electricity. Thin film solar cells can be developed because it decreases the cost of solar energy reaches to the grid parity level. Now a days, researches are progressed on thin film solar cells, CdTe solar cells and a-Si solar cells. For laboratory scale, CIGS based thin film indicates the highest efficiency among the various thin film technologies having record value of 22.6% and CdTe having 22.1%. Large number of elements and compounds have semiconducting properties including some pure elements such as silicon and germanium which are used because their outermost shell have 4 valence electrons gives the ability to gain and lose electron at same time. The synthesis of some II-VI compounds nanoparticles and their optical characterization have been studied [15–18].

Over the past 25 years, metal chalcogenides (sulphides, selenides and tellurides) has been studied in the form of thin film due to their applications in photoconductive cells, photovoltaic cells and in variety of optoelectronic devices. In recent years, the compound of chalcogenide family is more interested towards thin film because it has high efficiency and is used as an energy source for the production of solar cells. The compounds of chalcopyrite families had divides into: Binary (group II-VI, III-V and IV-IV), Ternary and Quaternary compounds. A detailed classification of various types of semiconducting materials is given below in Table 1.

**Table 1:** Classification of different types of semiconducting materials.

Compounds	Types of	Examples
	semiconductor	
Binary	II-VI compound	ZnS, ZnSe, ZnTe,
		CdS, CdSe, etc.
	III-V compound	AlAs, GaN, GaAs,
		InP, InAs, InSb etc.
	IV-IV compound	SiC, SiGe, etc.
Ternary		GaAsP, HgCdTe,
		AlGaAs, etc.
Quaternary		InGaAsP

Binary compounds of group IIB and VIA commonly known II-VI elements. as compound semiconductors exhibit larger degrees of ionic bonding than III-V compound semiconductor. Group II-VI semiconducting material of "Cadmium" based chalcogenide family have more attenuation by research community due to their unique properties and potential applications in the field of electrical, optoelectronic devices, light emitting, etc. Cadmium sulphide (CdS) is the member of II-VI compound. Semiconductor group applications materials with in several heterojunction photovoltaic systems includes cadmium telluride (CdTe), copper indium diselenide/sulphide and copper indium gallium diselenide/sulphide (CIGS) solar cells [19]. Researchers have great attenuation on CdS thin films due to its optoelectronic properties [20]. Processing of some III-V and II-VI binary compound semiconductors for technological applications have been studied [21-23]. The optical band-gap of CdS nanoparticles synthesized by mw assisted method is found to be increased as compared to bulk [24, 25].

Addition of impurities into binary semiconducting material causes dramatic change in their increasing properties. Ternary II-VI semiconducting materials have drawn more attention than the binary II-VI compound due to their physical properties which can be controlled by their constituent mole-fraction, particle size together with morphology. A lot of focus has been given to the doping of CdS nanostructures with the elements such as indium (In<sup>3+</sup>), aluminium (Al<sup>3+</sup>), gallium (Ga<sup>3+</sup>), boron (B<sup>2+</sup>), manganese (Mn<sup>2+</sup>), Iron (Fe<sup>2+</sup>), zinc (Zn<sup>2+</sup>), etc. that changes its optical, electrical and magnetic properties. Among these, zinc  $(Zn^{2+})$  is an important transition metal element having an ionic radius (0.074 nm) which is smaller than  $Cd^{2+}$  (0.097 nm). Therefore,  $(Zn^{2+})$  can easily be incorporated into CdS crystal lattice replaced Cd<sup>2+</sup> ions which improves its properties. Addition of Zn into CdS leads to an increase in the energy bandgap from 2.4 eV to 3.5 eV. The replacement of CdS with the higher bandgap ZnCdS led to a decrease in window absorption losses and result in an increase in the shortcircuit current density in solar cell [26-31]. The narrow bandgap partially blocks the transmission of high energy photon to the absorber layer below and decrease the power conversion efficiency. When CdS is replaced by  $Zn_xCd_{1-x}S$ , the wider bandgap leads to higher quantum efficiency in blue region of spectrum. Addition of Zn to CdS buffer layer material decreases the lattice constant with a lattice match to CIGS absorber to produce a more favorable conduction band alignment. It has been observed by adding Zn enhances both the open circuit voltage and short circuit current density of the device to yield a higher conversion efficiency of CIGS solar cells [32, 33]. ZnCdS is also useful for the fabrication of p-n junction without lattice mismatch in devices on quaternary materials like CuIn<sub>x</sub>Ga<sub>1</sub>- $_{x}$ Se<sub>2</sub> or CuIn(S<sub>x</sub>Se<sub>1-x</sub>)<sub>2</sub>. The growth parameters of the ZnCdS films can be tuned by varying the concentration of Zn. Different Zn contents regulates the growth rate of CdS and is the important factor for influence the properties of the film.

There are a number of physical and chemical ways for preparing thin films and have been summarized below as shown in Table 2.

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Phys	ical deposition	Chemical deposition			
1. Ev	aporation Techniques	1.Chemical vapour			
a.	Vacuum thermal	deposition			
	evaporation	a. Low pressure (LPCVD)			
b.	Electron beam	b.Plasma enhanced			
	evaporation	(PECVD)			
с.	Laser beam	c. Atomic layer deposition			
	evaporation	(ALD)			
d.	Arc evaporation				
e.	Molecular beam				
	epitaxy				
f.	Ion plating				
	evaporation				
		2.Sol-gel technique			
		3. Chemical bath deposition			
		4. Spray pyrolysis technique			
		5.Plating			
		a. Electroplating technique			
		b.Electroless technique			

Table 2: Methods of thin film deposition.

Among these techniques, chemical bath deposition (CBD) is the oldest method to deposit the films on a substrate and is also known as solution growth technique or controlled precipitations. CBD method is simple,

low cost, versatile deposition over large area, does not require sophisticated instrument like vacuum system, other expensive equipment, required chemical are commonly available, working at lower temperature and capable of yielding good quality thin films. In this method, the precursor solution of metal ions must be complexed by ligands and complex solution is obtained with ammonia solution, citric acid, etc. When the complexation completed, the addition of anions come from the source of sulphur anions to deposit the chalcogenides.

#### EXPERIMENTAL DETAILS Chalcogenide ZnCdS Thin Films by CBD Method

In CBD method, substrates and solution are stationary and solution is stirred using magnetic stirrer. Aqueous or nonaqueous baths with constant stirring are used to heat the chemical bath to desired temperature under continuous stirring. Substrates are inserted vertical position inside the solution and left the desired film thickness is obtained. Depending upon deposition conditions such as temperature, stirring solution rate. concentration, etc. the film growth can take place by ion-by-ion condensation of materials on the substrate. In the present review article, a survey of chalcogenide ZnCdS thin films deposited by chemical bath deposition method is discussed (Figure 1).

The following section deals with chemical bath deposition of chalcogenide ZnCdS thin films. The detailed preparative conditions and properties are presented in Table 3.



Fig. 1: Experimental setup of chemical bath deposition.

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S.N.	Bath composition	Deposition Temp. (°C)	Substrates used	Deposition Time	Findings	Ref.
1	CdI <sub>2</sub> + ZnI <sub>2</sub> + thiourea + ammonia	800	Glass	20 min	The [Zn]/[Cd]+[Zn] ratio in (Cd, Zn)S thin films could be controlled by varying the mixture ratio of CdI <sub>2</sub> and ZnI <sub>2</sub> solutions. The resistivity of (Cd, Zn)S thin films are 4– $8\times10^7\Omega$ .cm which depends on the [Zn]/[Cd]+[Zn] in the solutions. The optical bandgap increased within the [Zn]/[Cd]+[Zn] ratio, yield bowing parameter of 0.724 eV.	31
2	CdSO <sub>4</sub> + NH <sub>4</sub> OH + EDTA + ZnSO <sub>4</sub> + thiourea	85°	Si (100) wafers	1–3h	From XRD spectra, the peak at 29.2° into three diffraction from (100), (002) and (101) planes of hexagonal ZnS and (111) diffraction from cubic ZnS overlap representing a single peak.	32
3	CdCl <sub>2</sub> + NH <sub>4</sub> Cl + thiourea + ZnCl <sub>2</sub> + NH <sub>3</sub>	80°-85°	Soda-lime glass (SLG)	20–90 min	From XRD studies, CdZnS films have hexagonal structures. Film surface consists of wider grains as the Zn-content increased in the solution and energy bandgap at different Zn content (Y= 0, 0.3, 0.5 was 2.40 eV, 2.55 eV, 2.70 eV). The thickness of the films is found to be 100–200 nm.	33
4	(0.1M) Zinc acetate + (0.1M) cadmium acetate + (0.1M) thiourea	70°	Glass	30 min	Thin layer with bright yellow color was formed on the film surface. From XRD studies, ZnCdS films have hexagonal structure with (002) crystal plane. As the Zn content increases intensity of the film increases and Zn-content varied from 45 nm to 72 nm. ZnCdS films showed a bandgap of 3.88 eV. SEM micrograph revealed the grains on the film surface were more compact and has dense structure.	34
5	(0.015M) cadmium acetate + (0.015M) Zinc acetate + (0.05M) thiourea + (0.3M) ammonium acetate + (0.8M) ammonia	75°	Glass	45 min	Specular yellow coloured films of CdZnS are obtained. Films are adhesive and sparingly reflecting. The optical bandgap was 2.8 eV-3.4 eV. SEM micrograph indicates smooth, dense, uniform grains are distributed with average grains values 25, 11 and 13 nm.	35
6	(0.1M) CdBr <sub>2</sub> .4H <sub>2</sub> O + (0.1M) [(CH <sub>3</sub> COO) <sub>2</sub> Zn.2H <sub>2</sub> O] + (7.4M) TEA + (1.0M) (NH <sub>2</sub> ) <sub>2</sub> SC	350K	Glass	24h	The optical energy bandgap is 2.85 eV which lies in between the values of CdS and ZnS thin films. The average optical conductivity is $4.03 \times 10^{13}$ S <sup>-1</sup> .	36
7	(0.06M) CdSO4 + (0.2M) ZnSO4 + (0.6M) thiourea + TEA + ammonia	72°	Glass	10–60 min	Bright yellow orange coloured films of ZnCdS are obtained. Yellowness increases with increasing Zn content (x=0, 0.2, 0.4, 0.6 and 0.8). As deposited films have both cubic and hexagonal structure and average grain size changes from 8.75 nm to 3.57 nm with increase in Zn content which is useful for gas sensing. SEM micrograph reveals that as Zn content increases, uniform films with fibers like structure appears. The bandgap is 2.48 eV and 3.62eV for Zn content between 0 and 0.8 respectively.	37

Table 3: Preparative conditions and properties of chalcogenide ZnCdS thin films by CBD methods.

8	$(0.05M) Cd(OA_c)_2$ + $(0.05M) Zn(OA_c)_2$	85°	Glass	0–30 min	SEM micrographs reveal that no 38 pinholes can be observed in the
	$+(12M) NH_4OH + (0.2M) Na_2S_2O_3$				300°C and 500°C film surface shows
					the grains are not completely covered the surface i.e. incomplete nucleation
					step with irregular grain growth rate.
9	$ZnCl_2 + CdCl_2 + NH_4Cl$	60°-90°	Glass	1.5h	From XRD studies reveals that as the 39
					bath temperature increases from $60^{\circ}$
					(002) becomes strong and then
					weakens. The as deposited ZnCdS
					films are annealed at 350°C in $N_2$ atmosphere and diffraction peak does
					not change. The average grain size
					changes from 17.5 nm to 32.8 nm.
					$Cd_{1-x}Zn_xS$ thin film are used as the window layer in CdTe thin film solar
					cells. The value of activation energies
					attributed to electron or hole
					valence band is about 1.67 eV.
10	(0.396M) Ammonium	85°	Glass	2h	The concentration of mixture ratio is 40
	nitrate + $(0.357M)$ KOH				varied from 0.5 to 0.9 using a relation $x = [7n^{2+1}/[Cd^{2+1} + [7n^{2+1}]]$ Changing
	$+(3.64\times10^{-3})$ thiourea				the concentration, $Zn^{2+}$ play a key
	(				role during the process of regulating
					the growth rate and forming ternary semiconductor CdZnS films. In CdS
					thin film, atom-by-atom and the
					hydroxide cluster mechanism play an
					important role during the growth process of thin film However in ZnS
					thin film, cluster-by-cluster is needed
11		1.000	N 1 1 1	15 '	for growth process.
11	(0.2M) CdCl <sub>2</sub> + $(0.12M)ZnCl2 + (0.4M) thiourea$	100*	Morybaenum	15 min	varied in the range from 0.2 to 0.5
	+40 ml ethylene glycol				with the help of formula taken x=
					[ZnCl <sub>2</sub> ]/ [CdCl <sub>2</sub> ]+ [ZnCl <sub>2</sub> ]. XRD
					deposited concentration change Zn
					doped ZnCdS films exhibited (002)
					plane at $2\theta = 26.76^{\circ}$ which shows the hexagonal structure As the Zn
					concentration increases grain size
					increases. FESEM micrograph
					and unevenly distributed over the
					film surface. EDS spectra indicates at
					0.2 concentration no traces of Zn is
					increases spectra of Zn is observed
1					and the ratio of atomic percentage of
					The energy bandgap increases from
					2.54 eV to 2.85 eV.
12	$(0.2M) CdCl_2 + (0.12M)$	160°	Molybdenum	15 min	From XRD spectra, the peaks at $42$
	+ 40 ml ethylene glycol				observed into three diffraction with
					(002), (201) and (202) planes of
1					hexagonal structure. SEM
1					film surface is smooth, uniform,
					pinholes, crack free, etc. The
1					bandgap was found to be 2.45 eV. PL emission spectra is estimated at 382
					nm and 496 nm.

				1		
13	(0.2M) CdCl <sub>2</sub> + (0.12M) ZnCl <sub>2</sub> + (0.4M) thiourea + 40 ml ethylene glycol	160°	Molybdenum	15 min	From XRD, the crystallite size of the 43 as deposited and annealed ZnCdS thin films is calculated by using Scherrer's relation and is found to be 36 nm and 41 nm respectively. The increase in average grain size due to annealing improves the crystallinity of the films. The RMS roughness of both the films are estimated to be 127 nm and 39 nm. The optical bandgap is found to be 2.45 eV and 2.52 eV for both the as deposited and annealed films.	3
14	$\begin{array}{c} (0.05M) \ CdCl_2 + (0.5M) \\ C_6H_5O_7Na_3 + (0.5M) \\ KOH + (0.5M) \ CS(NH_2)_2 \\ + (1.5 \times 10^{-3}M) \\ Zn(CH_3COO)_2 \end{array}$	70°	Glass slide	15, 30,60 and 90 min	XRD studies reveals that an intense 44 peak centered at 26.7° corresponds to hexagonal phase of (002) plane. Three less intense peaks are appeared at 44°, 48° and 55° with (110), (103) and (104) planes. The growth mechanism of the film is affected by Zn concentration in the solution bath as Zn increases, the hydroxide growth mechanism is important and decreases the deposition rate as well as crystallite size of the films. The bandgap values increase up to 2.6 eV as Zn content in the film increased.	1
15	Cadmium sulfate + Zinc sulfate + thiourea + ammonia + sulfuric acid	70°	Glass slide	35 min	Zinc compositions changes from $x=0$ , 45 0.05, 0.08, 0.12 and 0.18. From XRD spectra reveals that diffraction peak intensity of (002) diffraction line decreases with increasing mixture ratio in the solution. SEM micrographs indicates that Cd <sub>1-x</sub> Zn <sub>x</sub> S changes with the increasing of Zn molar fraction. EDS measurements show that surfaces contain pure CdS and agglomerates formed by Cd, O, S and Zn for x= 0 to 0.18. The mobility increases from 2 to 4.8 cm <sup>-2</sup> /Vs with increasing x up to 0.12 while beyond this molar fraction, it shows decreasing tendency.	5

## CONCLUSIONS

In the present review article, we have described the synthesis of ZnCdS thin films by chemical bath deposition method. This method is simple, low cost, large area deposition and capable of yielding good quality of thin films. The data presented in the tabular form indicate that the film formation could be carried out on different substrates at different conditions. The review work deals with the synthesis of ZnCdS thin films and their structural, morphological and optical properties have also been discussed. The devices such as solar cells, photoconductors, detectors, sensors, etc. made from ZnCdS thin films show the promise of the method in modern thin film technology.

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## **Cite this Article**

S. Kumar, K.P. Tiwary. ZnCdS Thin Film Chalcogenide by Chemical Bath Deposition Method. *Nano Trends: A Journal of Nanotechnology and Its Applications*. 2020; 22(1): 19–27p.