(RESEARCH ARTICLE)

Effect of temperature in formation of nanocrystal on chemically deposited (Cd-Zn) S: CdCl$_2$; La, Eu films

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Abstract

Results of Photoconductivity (PC) and Photovoltaic (PV) studies for (Cd-Zn)S: CdCl$_2$;La,Eu films presented in different temperature of depositions along with Thiophenol (TP) and Poly Vinyl Chloride (PVC) using as a capping agent. Films have been characterized using Photoconductivity excitation spectra, optical absorption, Photoluminescence (PL), X-ray diffraction (XRD) and Scanning Electron Microscope (SEM). The band gap of such films is evaluated by Photoconductivity excitation spectra and optical absorption. The direct optical band gap of such films was found in the range 2.74-3.04 eV for different deposition temperature. These changing of band gap suggesting the quantum confinement effect in the developed nanoscale size of the particles. The PL spectra of both films nanoparticles showed emission peaks at 436nm, 461nm, 480nm, 504nm and 536nm respectively. SEM micrograph shows the morphological structure and better growth conduction in presence of capping agent at low temperature deposition. Results of XRD studies are associated to different prominent lines of CdS and ZnS. Both studies represent average particle sizes of the order 5.49 – 9.125Å.

Keywords: Chemical Deposition; Nanocrystalline Photoconductivity; Photovoltaic Effect; Photoluminiscence

1. Introduction

The II-VI group of semiconductor nanoparticles are widely utilised in thin-film transistor electronics, photovoltaic cells, photoconducting cells, photosensors, optical detectors, bulk phosphors, and photovoltaic cells. Numerous workers have tried Chemical Bath Deposition (CBD), which has been found to be a very practical, easy, and affordable process for creating such materials [1–9]. In their research on the photoconducting response of (Cd-Pb)S, (Cd-Zn)S and CdS films [10–12], Bhushan and group observed that under visible excitation with an incandescent bulb, the ratio of photocurrent to dark current was of the order of 10^7. Due to the quantum confinement phenomenon, the excitation and absorption of such semiconductor nanocrystals can be tuned by changing their size. A fascinating and expanding field of study is the use of quantized nanocrystals as absorbing materials in opto-electronic and photovoltaic systems [13–15]. (Cd-Zn)S films were found to have a photovoltaic response efficiency of 3.84%[16–17], and the outcomes of stability investigations of such films over a two-year period are also available[18]. Bhushan and their group also investigated photoluminescence in these films when UV (3650 Å) stimulation was used. The theme of the current work is films made by chemical bath deposition of (Cd-Zn)S: CdCl2;La,Eu at various temperatures and in the presence of capping reagents. The effects of particle size on photoconductivity (PC), photovoltaic (PV), photoluminescence, optical absorption, and excitation spectra of (Cd-Zn)S: CdCl2;La,Eu films have been discussed. This article also includes the findings of the XRD and SEM studies, in addition to these studies.

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2. Materials and Method

The films were applied to the surface of 24 mm x 74 mm conducting glass slides or microscopic glass slides of commercial quality. PV studies were conducted using the substrates of conducting glass slides. The spray pyrolysis process was used to create the conducting glass slides, with the resistivity of the plate being 20Ω cm^{-2} and $SnCl_2:2H_2O$ as the original chemical. Following acetone cleaning, these glass slide substrates were rinsed with double-distilled water. Once the substrate had been cleaned, the films were created by vertically dipping it in a solution of 1 M cadmium acetate/zinc acetate (totaling 7 ml), 2 ml. triethanolamine (TEA), 25 ml. 30% aqueous ammonia, and 7 ml. 1M thiourea (all A.R. grade). The combination had a pH of approximately 11. The depositions were carried out in a water bath with a constant temperature (Tempo) of 30 to 80^{0}C. The preparation of all solutions used double distilled water. The substrates were held vertically up against the side of a 50- or 100-ml beaker that contained the combination of the aforementioned liquids. Calculated ratios of CdCl2, Lanthanum, and Europium nitrates (all A.R. grade, 99.9%) were added to the initial mixture to prepare the doped films, and then depositions were carried out. Several ratios of the capping agent Poly Vinyl Chloride (PVC) and Thiophenol were used to create the films (TP). However, the best outcomes were found in 1.5 and.5 ml of (PVC) (TP).

The solution was first mixed with the substrates and then allowed to rest against the beaker walls before being dipped into the mixture. Throughout the deposition, no more stirring was done. A 100 W incandescent lamp was utilised as the excitation source for investigations on photoconductivity and photovoltaics (net illumination power = 2mW). Silver co-planer electrodes measuring 1.5 mm broad by 24 mm long with a 2 mm spacing were placed on the surface of the films for PC investigations. Silver electrodes with a surface area of 1 cm2 were placed on the back of the films to measure the PV. The second electrode was a conducting glass plate. 12 inch Hind-HiVac (12A4D) coating unit was used to apply the silver coating to the film.

A nanometer (DNM-121) was used to measure the associated photocurrent, and a digital multimeter was used to measure the voltage (Scientific HM-5011-3). By passing the light from a 1kW incandescent bulb through a prism monochromator and noting the photocurrent corresponding to various exciting wavelengths so generated. Several wavelengths were selected for photoconductivity excitation spectrum study. Optical absorption spectral investigations were conducted using a Varian (UV-VIS) DMS-100 spectrometer. XRD and SEM experiments conducted at the University of Delhi by using computerised Shimadzu differactometer (model- 98XRD with generator pw 1830 and pw 3710).

3. Results and Discussion

3.1. PC studies

Table 1 Values of dark current (I_{dc}), photocurrent(I_{pc}) and the ratio (I_{pc} / I_{dc}) for different (Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu films (time of deposition = 1 hour, voltage = 15 volts.)

System	I_{dc} (nA)	I_{pc} (µA)	I_{pc} / I_{dc}
(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$;La,Eu(30^{0}C)	0.10	198	1.98×10^6
(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu (40^{0}C)	0.08	215	2.68×10^6
(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu (50^{0}C)	0.04	270	6.75×10^6
(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu (60^{0}C)	0.02	303	1.51×10^7
(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu (70^{0}C)	0.03	254	8.46×10^6

Figure 1 illustrates the findings of the rise and decay curves of photocurrent during the fabrication of the films of (Cd.90-Zn.10)S:CdCl2; La, Eu at various temperatures, and Table 1 lists the relevant values. Due to an increase in particle size, it has been found that when deposition temperature rises, dark current (Idc) drops and photocurrent (Ipc) increases. The size of the particles grew with the deposition temperature, which decreased the film resistivity. It has been found that rise and decay curves generally have comparable properties at various deposition temperatures. Also, it has been noted that in low temperature deposition circumstances, the rise consists of a rapid increase at first due to carrier creation, followed by saturation brought on by the balancing impact of generation and recombination phenomena. It is found that while photocurrent is higher in higher temperature films, rise and decay time duration is longer. Moreover, curves created for films of (Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu) employing capping agents of 0.5 ml of (PVC) and 1.5 ml of (TP) are shown in Figure 2 and the relevant values are listed in Table 2. It demonstrates that the rise and decay curves'

characteristics are the same whether a capping agent is used or not, but the latter case's photocurrent to dark current ratios are low. Also, it was found that the capping agent reduced the particle sizes.
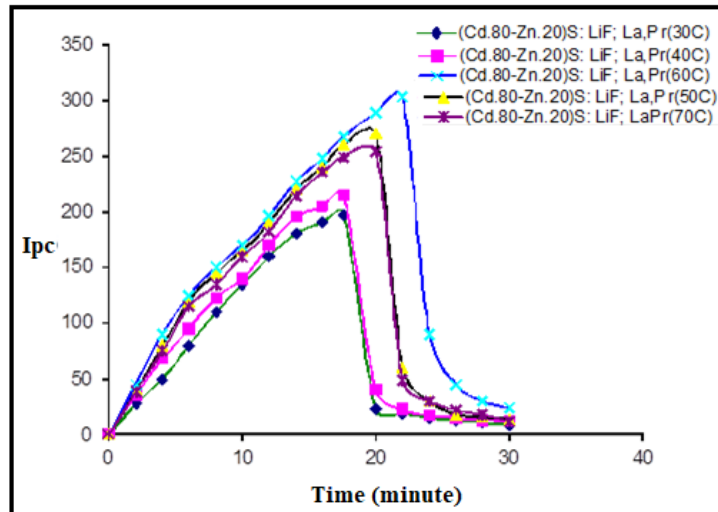

Figure 1 Rise and decay curves of different(Cd-Zn)S:LiF;La,Pr Films.

Table 2 Values of dark current (I_{dc}), photocurrent (I_{pc}) and the ratio (I_{pc} / I_{dc}) for different (Cd.$_{90}$-Zn.$_{10}$)S:CdCl$_2$; La, Eu films with capping agent{1ml of (PVA) and 2ml(TP)} [time of deposition = 1 hour, voltage = 10 volts.]

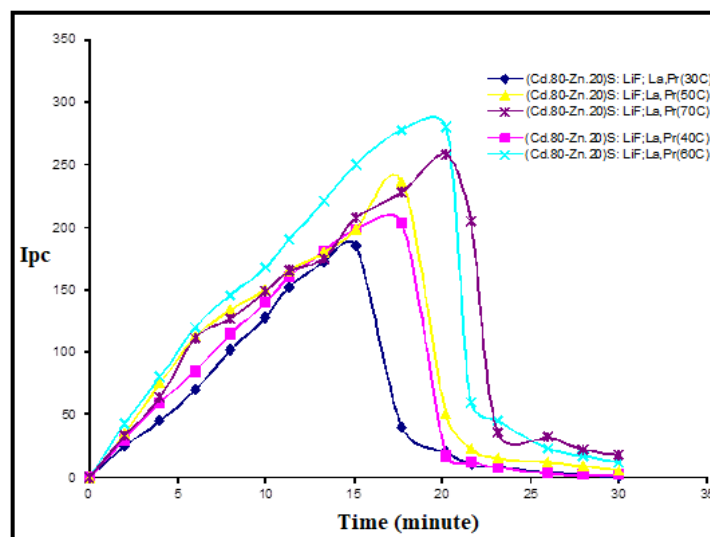| System	I_{dc} (nA)	I_{pc} (μA)	I_{pc} / I_{dc}
(Cd.$_{90}$-Zn.$_{10}$)S:CdCl$_2$;La,Eu (30^{0}C)	0.15	185	1.23×10^6
(Cd.$_{90}$-Zn.$_{10}$)S:CdCl$_2$; La, Eu (40^{0}C)	0.10	203	2.03×10^6
(Cd.$_{90}$-Zn.$_{10}$)S:CdCl$_2$; La, Eu (50^{0}C)	0.07	236	3.37×10^6
(Cd.$_{90}$-Zn.$_{10}$)S:CdCl$_2$; La, Eu (60^{0}C)	0.05	280	5.6×10^6
(Cd.$_{90}$-Zn.$_{10}$)S:CdCl$_2$; La, Eu (70^{0}C	0.05	258	5.16×10^6

Figure 2 Rise and decay curves of different (Cd-Zn)S:LiF;La,Pr films with Capping agent

3.2. Photoconductivity Excitation Spectra

Figure 3 depicts the PC excitation spectra of films made of $(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu produced at various temperatures, whereas Figure 4 depicts films containing capping agents (0.5ml of PVC and 1.5ml of TP). Table 3 lists the band gaps that were thus acquired. As anticipated, it is revealed that the energy band gaps of the materials are slightly altered in both cases as the deposition temperatures decrease as a result of the peaks shifting towards the blue region. This shifting of peaks shows the change in particle size.
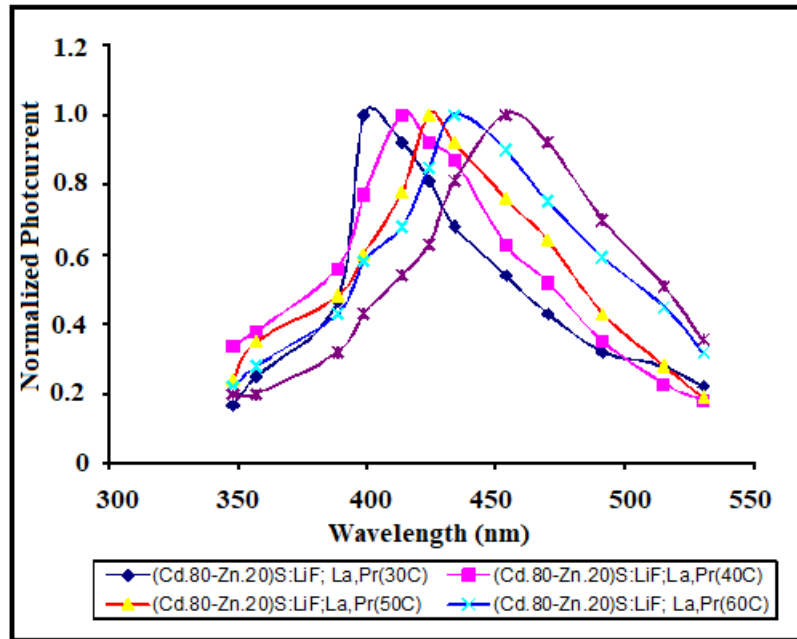

Figure 3 Photoconductivity excitation spectra of different (Cd-Zn)S: LiF; La,Pr films

Table 3 Values of band gap energies from Photoconductivity excitation spectra for different temperature of deposition $(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu with capping agent films

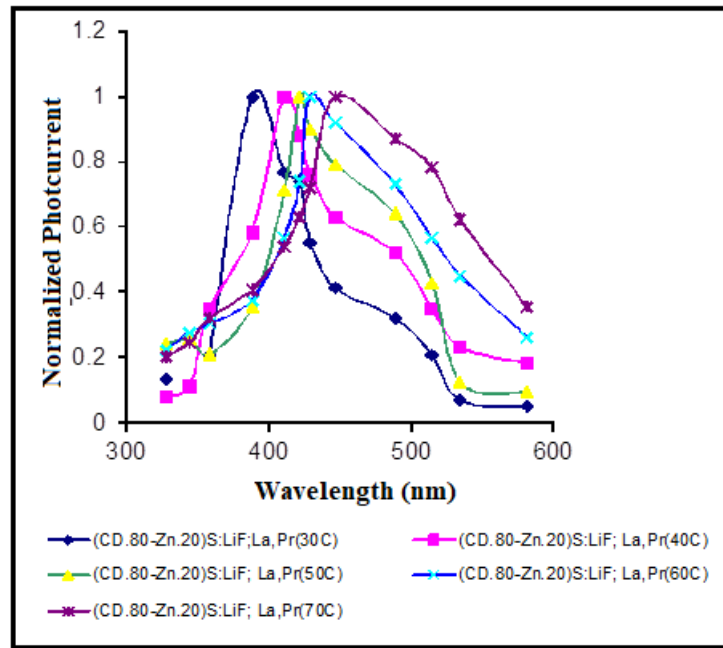| S.N.	System	E_g(eV)	E_g(eV) (with capping agent)
1	$(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu ($30^0C$)	3.1	3.19
2	$(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu ($40^0C$)	2.99	3.04
3	$(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu ($50^0C$)	2.91	2.93
4	$(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu ($60^0C$)	2.85	2.88
5	$(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu ($70^0C$	2.73	2.77

Figure 4 PC Excitation spectra of Different (Cd-Zn)S: LiF;La,Pr Films with capping agent

3.3. Optical absorption spectra

The optical absorption of $(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu films and that the using capping agent are shown in Figure 5. Plots of ($\alpha h\nu$) vs $h\nu$ were used to calculate band gap values. Table 4 provides an overview of the corresponding energy band gaps' values. The direct optical band gap of such films was lying in the range 2.72-3.02 eV for different deposition temperature. The values of direct band gap are higher than the bulk value (2.70 eV) and the increment of band gap also supports the nanometer size$(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu particle formation within the polymer matrix.

Table 4 Values of band gap energies from optical absorption spectra for different temperature of deposition $(Cd_{.90}-Zn_{.10})S:CdCl_2$;La,Eu with capping agent films

S.N.	System	E_g(eV)	E_g(eV) (with capping agent)
1	$(Cd_{.90}-Zn_{.10})S:CdCl_2$;La,Eu (30^{0}C)	2.99	3.04
2	$(Cd_{.90}-Zn_{.10})S:CdCl_2$;La,Eu (40^{0}C)	2.89	2.94
3	$(Cd_{.90}-Zn_{.10})S:CdCl_2$;La,Eu (50^{0}C)	2.83	2.85
4	$(Cd_{.90}-Zn_{.10})S:CdCl_2$;La,Eu (60^{0}C)	2.77	2.80
5	$(Cd_{.90}-Zn_{.10})S:CdCl_2$;La,Eu (70^{0}C)	2.75	2.76
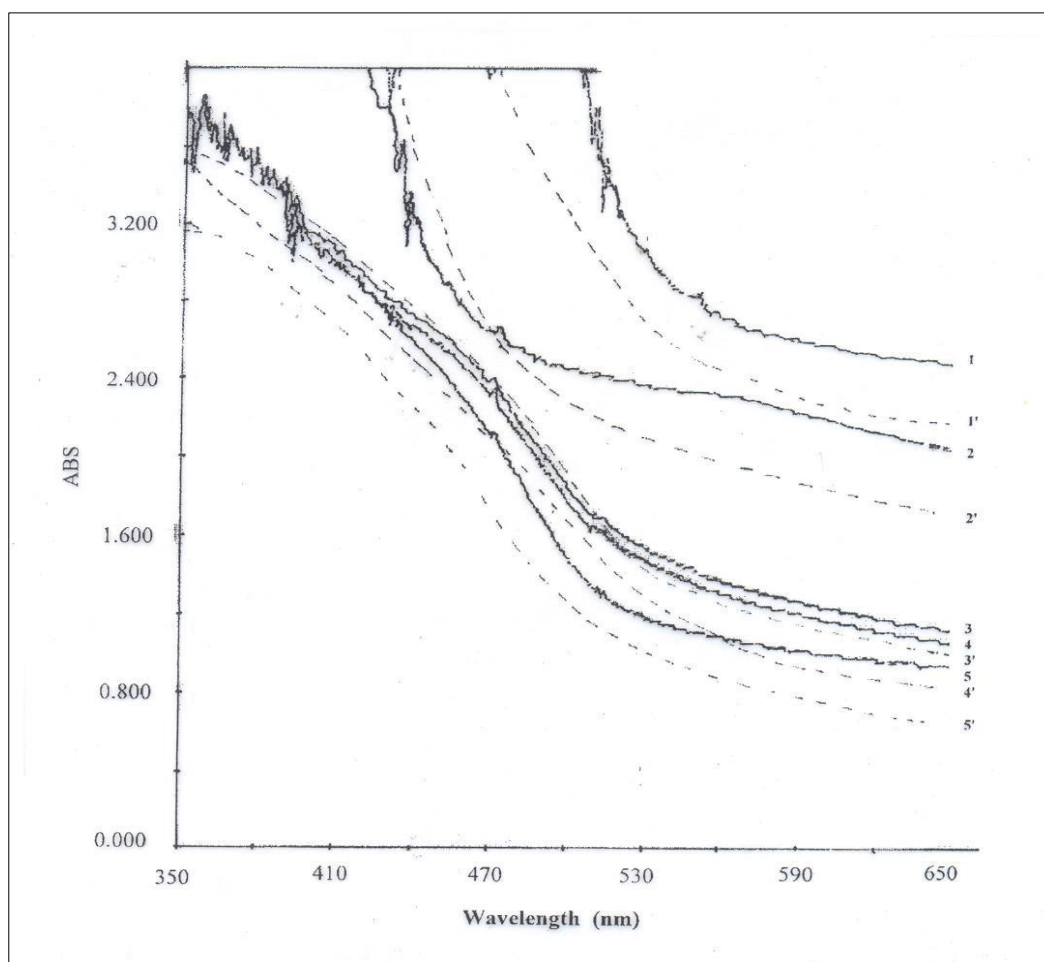
Figure 5 Optical absorption of different (Cd-Zn)S: LiF,Pr (--------) and with Capping agent (——)Films. 1. (Cd$_{.80}$-Zn$_{.20}$)S:LiF,Pr(30^{0}C) , 2. (Cd$_{.80}$-Zn$_{.20}$)S:LiF,Pr(40^{0}C), 3. (Cd$_{.80}$-Zn$_{.20}$)S:LiF,Pr(50^{0}C), 4. (Cd$_{.80}$-Zn$_{.20}$)S:LiF,Pr(60^{0}C), 5(Cd$_{.80}$-Zn$_{.20}$)S:LiF,Pr(70^{0}C).

3.4. Photovoltaic Studies

The photovoltaic studies were carried out for (Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu films emloying conducting glass plate as one electrode and that of with silver coating as the other electrode. As a result, it may act like an M-S contact. The measurements were made for both not using and with using capping agent. The corresponding I-V curves for such systems are shown in Figure 6 (a) and 6(b) respectively. The values of short circuit current (I_{sc}), open circuit voltage (V_{oc}), Fill Factor (FF) and Efficiency (η) are summarized in table 5a and 5b. It is evident that the values of efficiency are varying between 3.97 to 4.86 % and using capping agent 3.64 to 4.96% respectively. It is witnessed that with increase in the deposition temperature, the value of I_{sc} increases but V_{oc} decreases. It is clear that when the particle sizes are decreased then the films resistance increases.

Table 5a Parameters of Photovoltaic studies for different (Cd$_{.80}$-Zn$_{.20}$)S: LiF;La, Pr Films

S.N.	System	I_{sc} (mA)	V_{oc}(mV)	I_m(mA)	V_m(mV)	F.F.	η (%)
1.	(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$;La,Eu (30)	0.23	0.65	0.15	0.53	0.53	3.97
2.	(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$;La,Eu (40)	0.22	0.70	0.16	0.50	0.52	4.00
3.	(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$;La,Eu (50)	0.23	0.64	0.18	0.52	0.63	4.68
4.	(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$;La,Eu (60)	0.21	0.75	0.18	0.54	0.62	4.86
5.	(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$;La,Eu (70)	0.25	0.60	0.14	0.55	0.51	3.85

Table 5b Parameters of Photovoltaic studies for different (Cd.80-Zn.20)S: LiF, La,Pr Films with capping agent

S.N.	System	I_{sc} (mA)	V_{oc}(mV)	I_m(mA)	V_m(mV)	F.F.	η (%)
6.	(Cd.80-Zn.20)S: LiF;La, Pr (30)	0.24	0.65	0.14	0.52	0.46	3.64
7.	(Cd.80-Zn.20)S: LiF;La, Pr (40)	0.23	0. 70	0.15	0.50	0.47	3.75
8.	(Cd.80-Zn.20)S: LiF;La, Pr (50)	0.20	0.75	0.16	0.56	0.58	4.48
9.	(Cd.80-Zn.20)S: LiF,;La,,Pr (60)	0.21	0.77	0.18	0.55	0.61	4.95
10.	(Cd.80-Zn.20)S: LiF,;La,Pr (70	0.22	0.73	0.16	0.525	0.52	4.2
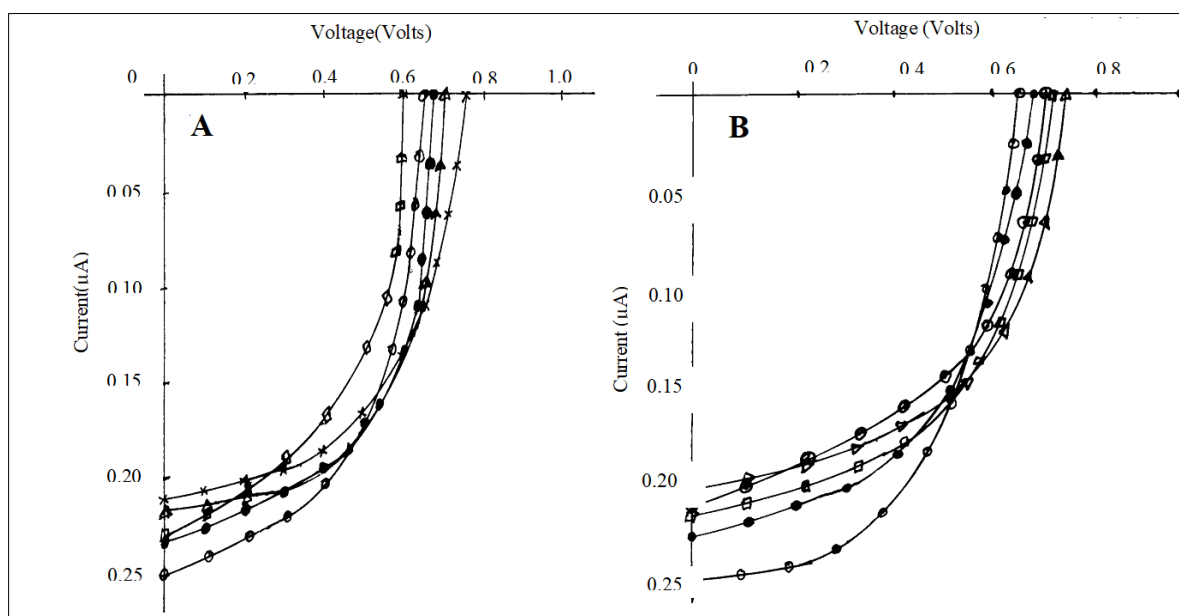
Figure 6 (a) I-V Characteristic of different (Cd.80-Zn.20)S:LiF,Pr films 1. (□) (Cd.80-Zn.20)S:LiF,Pr(30⁰C) , 2. (Δ)(Cd.80-Zn.20)S:LiF,Pr(40⁰C), 3.(•) (Cd.80-Zn.20)S:LiF,Pr(50⁰C), 4.(x) (Cd.80-Zn.20)S:LiF,Pr(60⁰C), 5 (O) (Cd.80-Zn.20)S:LiF,Pr(70⁰C). (b) I-V Characteristic of different (Cd.80-Zn.20)S:LiF,Pr films with capping agent 1. (•) (Cd.80-Zn.20)S:LiF,Pr(30⁰C) , 2. ((□) (Cd.80-Zn.20)S:LiF,Pr(40⁰C), 3. (Δ) (Cd.80-Zn.20)S:LiF,Pr(50⁰C), 4.(O) (Cd.80-Zn.20)S:LiF,Pr(60⁰C), 5 (●) (Cd.80-Zn.20)S:LiF,Pr(70⁰C).

3.5. Photoluminescence Studies

The Photoluminescence spectra of (Cd.90-Zn.10)S:CdCl₂; La, Eu films and that of using capping agent are shown in Figure 7. It is clear from these figures that the peaks are 436nm, 461nm, 480nm, 504nm and 536nm respectively. It is clearly observed that the peaks shift from green to blue emission, which shows nanocrystalline films.
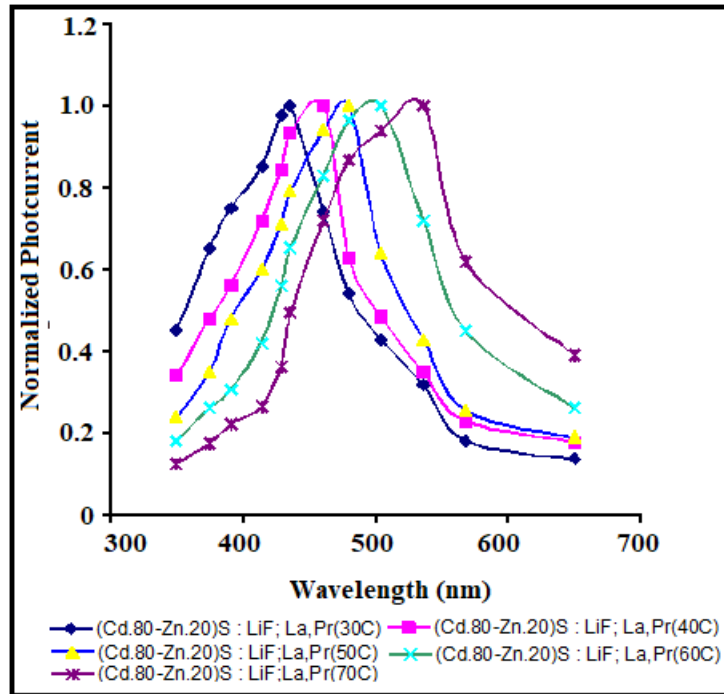
Figure 7 PL spectra of different (Cd-Zn)S: LiF; La, Pr Films

3.6. XRD Studies

The XRD diffractograms of(Cd.90-Zn.10)S:CdCl2; La, Eu and that of using capping agent films are shown in Figure 8 (a), (b) (c) and (d) respectively. We acquire the hexagonal and cubic phases. The intensity of peaks in X-ray diffractograms are shown to decrease as ZnS concentration increases, indicating that the addition of ZnS influences crystallinity. The $(111)_c$, $(220)_c$ lines of CdS and $(110)_h$ lines of α-ZnS were present in the (Cd-Zn)S: CdCl2; La,Eu XRD patterns. The evaluation of the evaluated lattice constant values and the reported values has been compared with the ASTM data to determine the peak assignments. The XRD patterns of using capping agent (Cd.90-Zn.10)S:CdCl2; La, Eu consisted of $(111)_c$, $(220)_c$ and $(311)_c$ lines of CdS, $(220)_h$ of ZnS. Several peaks are present, and there are differences in their intensities, which suggests that the presence of the capping agent is affecting the growth conditions. Using the formula [19], the crystals' particle sizes were determined.

$D = K\lambda / (\beta_{1/2} \cos \theta)$ where K is a constant that depends on the grain form, $\beta_{1/2}$ is the full width at half-maximum (FWHM), λ is the X-ray wavelength, and θ is the Bragg's angle (in radians). Particle sizes were discovered to range from 3.14 to 7.125. Table 6 provides a summary of the matching XRD data
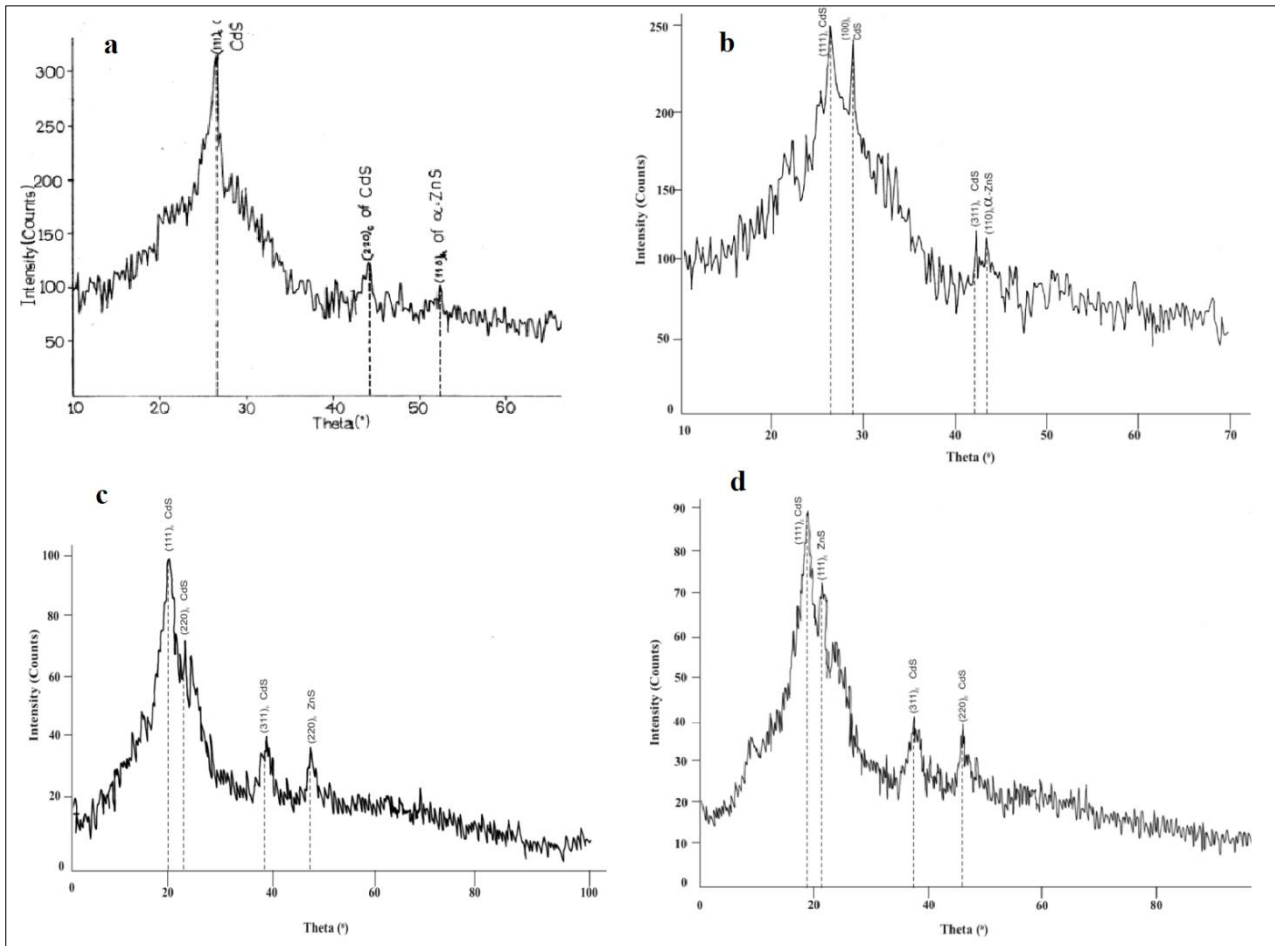
Figure 8(a) X-ray diffractogram of (Cd-Zn)S: LiF;La,Pr film at (50⁰C) (b) X-ray diffractogram of (Cd-Zn)S: LiF;La,Pr film (50⁰C)with capping agent (c) X-ray diffractogram of (Cd-Zn)S: LiF;La,Pr (60⁰C) film with capping agent (d) X-ray diffractogram of (Cd-Zn)S: LiF;La,Pr (60⁰C) film .

Table 6 XRD data of different (Zn-Cd)S:LiF;La,Pr films *(Preparation time = 60 min.; film thickness ~ 1-2 μm)*

d-values (A⁰)		Relative intensities		Hkl	Lattice constant (A⁰)		Average particle size (A⁰)	
Obs.	Rep.	Obs.	Rep.		Obs.	Rep.	XRD	SEM
(a)Zn-Cd)S: LiF;La,Pr								
3.3600	3.360	100	100	(111)c-CdS	5.818			
2.0518	2.058	35.5	80	(220)c-CdS	5.818		9.125	
1.7506	1.911	28	74	(110)h-α ZnS	3.50	3.82	3.00	
(b). *(Zn-Cd)S: LiF;La,Pr*								
3.5311	3.583	60	75	(100)h-CdS	4.077	4.135		
3.3685	3.360	100	100	(111)c -CdS	5.834	5.818		
1.9016	1.911	48	74	(110)h-α ZnS	3.803	3.820	3.254	
1.8267	1.753	50	60	(311)c-CdS	5.969	5.849	2.91	

(c) *(Zn-Cd)S: LiF;La,Pr*								
3.360		100		(111)c -CdS	5.8196	5.818		
2.0518	2.058	80		(220)c- CdS	5.8033	5.818	6.49	
1.7616	1.761	60		(311)c-CdS	5.5000	5.849	3.6	
1.8267	1.912	23	51	(220)c- ZnS	5.1600	5.406		
(d). *(Zn-Cd)S: LiF;La,Pr*								
3.3505	3.560	100	100	(111)c-CdS	5.807	4.135		
3.037	3.123	82	100	(111)c-ZnS	5.409	5.406	3.142	
1.9428	2.058	44.4	80	(220)c-CdS	5.882	5.818	2.074	
1.761	1.763	46	60	(311)c-CdS	5.86	5.848		

3.7. SEM Studies

The SEM images of $(Cd_{.90}-Zn_{.10})S:CdCl_2$; La, Eu films at 5000 magnification are depicted in Figure 9 (a), (b), (c) and (d) . Some samples show grains with spherical shapes. High temperature case obtained spherical shape, but deposition temperatures are decreased then the irregular shape and this growth increased further, which can be seen in third and fourth case. The particle sizes were calculated by Heyn's intercept method and were estimated to lie between 2.074 - 3 Å. Table 6 is a list of the values for particle sizes.
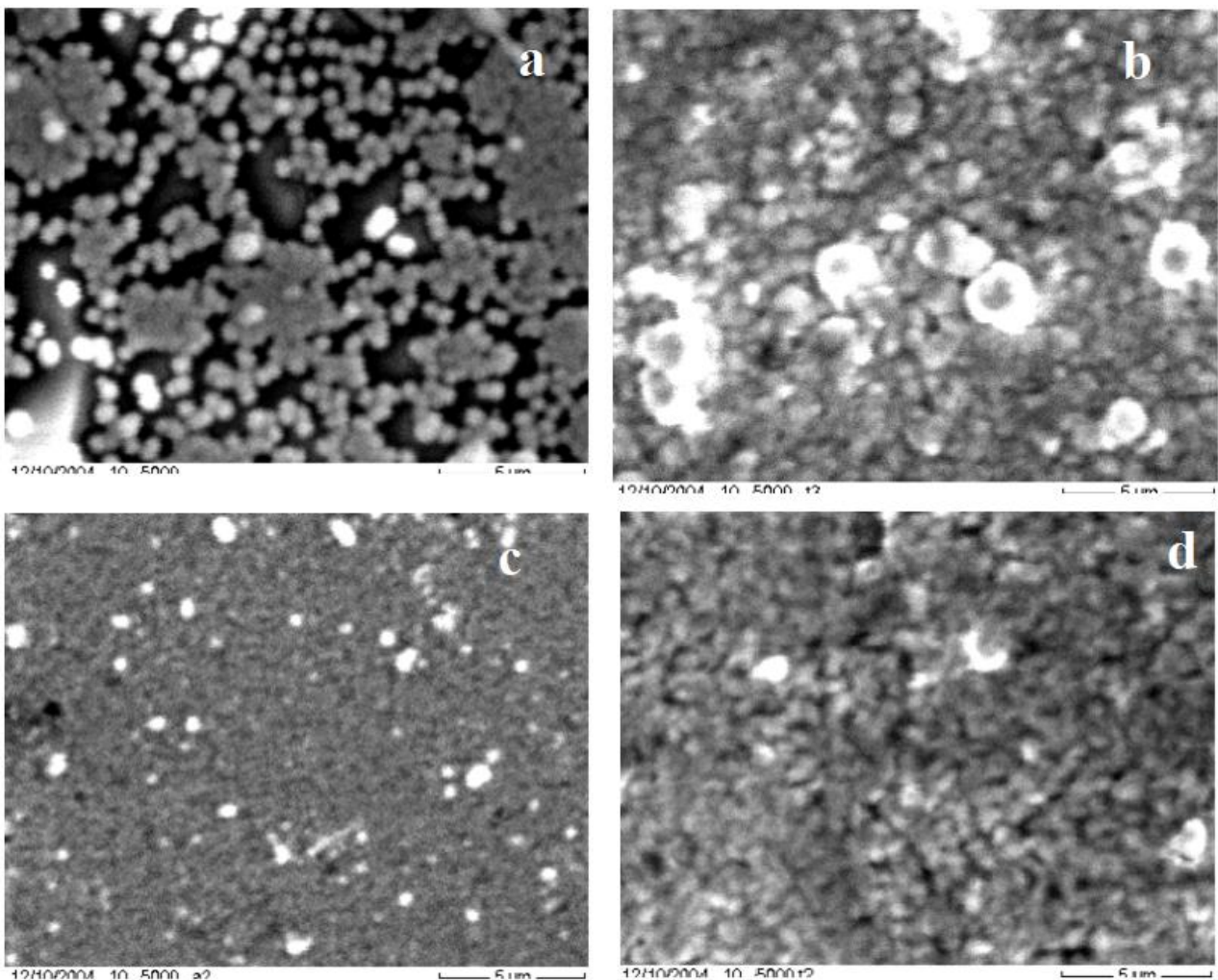

Figure 9 (a) SEM micrographs of Cd-Zn)S: LiF;La,Pr (60⁰C) film. (b) SEM micrographs of Cd-Zn)S: LiF;La,Pr (60⁰C) film with capping agent. (c) SEM micrographs of Cd-Zn)S: LiF;La,Pr (30⁰C) film with capping agent. (d) SEM micrographs of Cd-Zn)S: LiF;La,Pr (30⁰C) film. (Magnification = 5000)

4. Conclusions

The preparation of (Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu films by chemical deposition technique using PVC and TP as a capping agent two types of films have been studied as(Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu and with capping agent. The films have been characterized by photoconductivity, photovoltaic, photoconductivity excitation spectra, optical absorption spectra, photovoltaic effect, photoluminescence, XRD and SEM measurement. By altering of the ratios of photocurrent to dark current (I$_{pc}$/ I$_{dc}$) of (Cd$_{.90}$-Zn$_{.10}$)S:CdCl$_2$; La, Eu along with capping agent at different particle sizes were obtained. The band gap changes up to 2.72-3.02 eV were observed due to quantum size effect. The band gap variations are dependent on the capping agent and deposition temperature. The PL spectra of simple and capping agent shows the similar peaks. The variations of emissions are obtained yellow- green to blue Along with XRD and SEM measurement particle sizes are found nano- scale.

Compliance with ethical standards

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