



Photoluminescence studies of $\text{Cd}_{0.8-x}\text{Pb}_x\text{Zn}_{0.2}\text{S}$ semiconductor compounds

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ARTICLE INFO

Keywords:

Co-precipitation
Photoluminescence
Blue shift
Mixed semiconductor compound
X-ray diffraction

ABSTRACT

The present study involves synthesis of bulk polycrystalline samples of $\text{Cd}_{0.8-x}\text{Pb}_x\text{Zn}_{0.2}\text{S}$ through low cost controlled Co-precipitation method. The synthesized compounds were characterized with respect to their crystal structure, particle size and photoluminescence properties using X-ray diffraction (XRD), and PL spectroscopy respectively. XRD results revealed that both cubic and Hexagonal phases are present in the compounds. PL studies indicated the photosensitive character of the compounds. Blue shift in the peak position in PL spectra with increase in concentration of Pb indicates the increase in the energy gap. PL spectra confirmed the formation of defect levels in the forbidden energy gap and the transition is thought to occur from donor levels to acceptor levels.

1. Introduction

Mixed compounds formed out of II-VI or III-V semiconductors are used for harvesting energy directly from Sunlight through Photovoltaic devices. Well before the synthesis of compound semiconductors, elemental semiconductor, Silicon was used for solar cell fabrication. In the search for low-cost alternatives to crystalline Silicon, thin film compound semiconductor materials are widely used as it is more efficient to create an electric field at hetero junctions using thin films. A typical polycrystalline thin film has a thin layer on top known as a “window” material. It allows most of the light through the interface to the absorbing layer. The absorbing layer has to have a high absorption, weak in generation of current and a suitable band gap to provide good voltage [1]. The CdZnS ternary compound is also potentially useful as a window material for the fabrication of p-n junction without lattice mismatch in the devices based on materials like CdTe [2].

Band gap energy is one of the most important parameters of a semiconductor compound which determines many of its electronic and optical properties. II – VI semiconductors with energy gap covering the visible spectral range are compatible for the fabrication of optoelectronic devices [3]. Also use of these materials in photovoltaic devices require better modeling of their electronic and optical properties as a function of alloy composition. Advanced growth technologies made it possible now to have better control process parameters and tuning the material properties of II – VI compounds [4].

As CdZnS and PbZnS are highly sensitive to light and in view of practical applications a study of their mixed compounds as

electrochemical converters is of technical importance [5]. Band gap engineering is also crucial prerequisite for the fabrication of quantum confined low dimensional hetero structures. This is based on the alloying process of different semiconductor materials by which the energy band gap of the material can be varied over a wide range.

A rapid increase in resistivity / energy gap of $\text{Cd}_{1-x}\text{Zn}_x\text{S}$ with increase in x, limits its usage in conductivity related applications like near infrared solar cells. Therefore, it is interesting to prepare samples of higher energy gap with reasonably good electrical conductivity. One way of achieving such property in compounds is by selecting suitable dopant or an element to be incorporated into the lattice. Hence, we have taken up the preparation of $\text{Cd}_{0.8-x}\text{Pb}_x\text{Zn}_{0.2}\text{S}$ system and their characterization by means of optical studies. Truly, there is very little information available on the ternary CdPbZnS system. Of course, there is an evidence in the literature showing an increase in energy gap with the incorporation of Pb in to the lattice [6]. Therefore, usage of these compounds help in the conversion of not only visible radiation but also UV radiation into electrical energy.

Room temperature PL mapping of band – edge emission and deeper defect-related emissions is proving to be useful technique to characterize the homogeneity of the crystalline quality of semiconductors [7]. An earlier examination of the usefulness of 300 K PL measurements in determining composition variations in II-VI semiconductors reported that the measured PL emission peak energy depends on the sample conductivity and concentration of dopants [8]. Brunett et al. [9] have used room temperature photoluminescence to correlate radiation detector performance with material quality.

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Prem Kumar and Sankaranarayanan [10] have prepared thin films of CdZnS by chemical bath deposition technique and studied room temperature PL at an excitation wavelength 390 nm. They observed that the film deposited under dark, white light and fluorescent white light illuminated chemical bath displays the broad blue emission centered at 494 nm and the weak green emission occurred at 532.9 nm. PL intensity was higher for the film deposited under fluorescent light. Navneethan et al. [11] synthesized N-Methyl aniline capped lead sulfide nano particles by wet chemical route. From PL spectrum, it is clear that emission wavelength is 388 nm. This peak shifted towards the blue when compared to bulk PbS. This emission is due to the recombination of electron and hole pair. The blue shift demonstrates the strong quantum confinement. Bhusan et al. [12] have reported the PL study on different CdS-Se films. It should be noted that PL spectrum of CdS film was found to show only one emission peak at 515 nm. This was related to the band edge transition in CdS. This peak shows a shift towards higher wavelength (575 nm) due to reduction in the band gap with increasing concentration of CdSe, resulting in a donor – acceptor transition occurring between donor levels formed by incorporation of excess Cd during acceptor levels formed by incorporation of Se (i.e., Cd vacancies). The higher PL intensity at higher concentrations of Se may be attributed to the increased density of surface states created by the potential fluctuations and compositional disorders in CdS – Se, providing channels for radiative recombination [13].

The Photoluminescence emission spectra of ZnS films of different thickness at room temperature have been studied by Jeyachitra et al. [14]. PL emission spectra for the ZnS samples excited with 450 nm (3.82 eV) exhibit low energy and high energy bands. The green emission bands corresponding to 3.28 eV are attributed to the transition of sulfur vacancy (Vs) to the valence band and the donor acceptor pair recombination. Since the energy separation between given band (3.28 eV) and the band to band transition (3.42 eV at room temperature) is approximately 2.72 eV, this probably excludes the possibility of the recombination of free electron with holes localized at Zn and S vacancies. The red emission band has been ascribed to surface states in particular Zn vacancies. Photoluminescence (PL) studies of CdS samples having different crystalline size were carried out at 300 K by Nanda et al. [15]. From the PL spectra of the samples, single peak is observed in case of bulk CdS (1450 nm) and three peaks are observed in case of sample of thickness (35 nm). As the PL peak energies are less than the band gap energy, these bands can be identified with transitions involving donors, acceptors, free electrons and holes. The formation of single peak in bulk CdS sample may be attributed to the crystalline size. As the crystalline size increases, the other two peaks, observed in the other sample come closer and merge, thereby increasing the intensity of single peak. This peak is ascribed to the donor – acceptor transition whose intensity increases with increasing crystalline size [16,17]. PL Study has been carried out on CdS bulk sample by Srinivasa Rao et al. [18]. One broad peak was observed in the PL Spectrum of CdS sample with excitation wavelength 328 nm at 410 nm and luminescence from the surface states (around 800 nm) has also been observed. The peak of eigen-transition is strong and steep, the emission peak of surface states is flat and weak. The position of peak shifts towards higher energy with the decrease of nano particle size.

Photoluminescence (PL) has been employed extensively to characterize defects and impurities in CdS, PbS and very few reports are available on CdZnS semiconductor compounds. Further, no reports are available on CdPbZnS semiconductors. Hence we are motivated to study PL of $Cd_{0.8-x}Pb_xZn_{0.2}S$ mixed semiconductor compounds with $x = 0, 0.1, 0.4, 0.6, 0.7, 0.8$. which led to qualitative model indicating that the recombination mechanism of excitons (electrons from the defect levels with the holes in valence band) involving defect levels. It also explained the relaxation of excitons in the stop over levels present in the energy gap, which resulted in the emission of phonons.

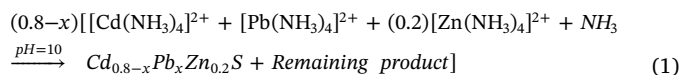
2. Experimental details

2.1. Synthesis of samples

Bulk polycrystalline samples of $Cd_{0.8-x}Pb_xZn_{0.2}S$ are prepared by the decomposition containing cadmium, lead and zinc acetate salts. The preparation process is based on the slow release of Cd^{2+} , Pb^{2+} , Zn^{2+} and S^{2-} ions in solution. The slow release of Cd^{2+} , Pb^{2+} and S^{2-} ions is achieved by the dissociation of a complex species of Cd, Pb and Zn such as tetra amine cadmium, tetra amine lead and tetra amine zinc complex ions $[Cd(NH_3)_4]^{2+}$, $[Pb(NH_3)_4]^{2+}$ and $[Zn(NH_3)_4]^{2+}$

S^{2-} ions are supplied by the decomposition of organic sulfur containing compounds, such as thiourea in alkaline medium reacts with Cd^{2+} , Pb^{2+} and Zn^{2+} ions to form $Cd_{0.8-x}Pb_xZn_{0.2}S$ precipitate. The reaction mechanism is as shown below.

The three tetra amine complexes $[Cd(NH_3)_4]^{2+}$, $[Pb(NH_3)_4]^{2+}$ and $[Zn(NH_3)_4]^{2+}$ combine in the reaction mixture leading to the formation of $Cd_{0.8-x}Pb_xZn_{0.2}S$ as per the chemical equation:



The precipitate was filtered and dried at room temperature for 24 h. The dried precipitate was heated for 2 h at 300 °C under nitrogen atmosphere and then slowly cooled to room temperature at 2 °C/min.

The precipitate was ground to fine powder and made into pellets under a pressure of 10 t per sq.cm. by using a punch dye of 1.0 cm diameter. The thickness of pellets thus prepared was seen to be about 1 mm by taking approximately same amount of powder every time.

The pellets were sintered at 800 °C for 2 h in nitrogen atmosphere. The furnace was cooled slowly to room temperature at the rate of 2 °C per min and these pellets are used for further studies of XRD and Photoluminescence [19].

2.2. Photoluminescence studies

Cary Eclipse fluorescence spectrophotometer with Xe source has been used to record photoluminescence emission spectra of $Cd_{0.8-x}Pb_xZn_{0.2}S$ semiconductor compounds. Powder of each sample is taken in a capillary tube, which in turn is placed in the cuvette of spectrophotometer and the emission spectrum of each sample is recorded.

3. Results and discussion

3.1. X-ray diffraction

Crystallographic studies of mixed ternary compounds were carried out using X – ray diffraction (XRD) technique. X-ray diffractograms of all $Cd_{0.8-x}Pb_xZn_{0.2}S$ samples ($x = 0-0.8$), published in our earlier article [19], indicated that the compounds were polycrystalline.

The values of inter planar distance (d) corresponding to the observed diffraction peaks were computed and compared with JCPDA data (PDF No. 400835 and PDF No. 050592) of $Cd_{0.8}Zn_{0.2}S$ (hexagonal) and PbS(cubic) phases and assigned the miller indices. The details of the parameters are given in Table 1.

It is found from the Table 1 that only hexagonal peaks are observed in the compound $Cd_{0.8-x}Pb_xZn_{0.2}S$ with $x = 0$, both hexagonal and cubic phases are present in the mixed crystals with $x = 0.1-0.6$ where as only peaks corresponding to cubic phase are seen in samples with $x = 0.7$ and 0.8. Overall, one can see that $Cd_{0.8-x}Pb_xZn_{0.2}S$ compounds undergo a crystalline transition from hexagonal to cubic i.e. the structure changes from lower symmetry to higher symmetry. These results are also in agreement with our earlier reports [22,24].

Hexagonal structure of $Cd_{0.8-x}Pb_xZn_{0.2}S$ compound is due to major peaks occurring from the reflections of (100), (002), (101) planes and

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