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Growth And Characterizations Of $Cd_{1-X}Zn_xSe$ Thin Film For Photovoltaic Applications By Solution Growth Technique

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Abstract
Cadmium Zinc selenide $Cd_{1-x}Zn_xSe$ (x = 0.6) thin film deposited on to glass substrate by solution growth technique. Deposition parameters were optimized. Structural, optical and surface morphology properties of as-deposited thin film were studied by XRD, UV-VIS, SEM and AFM, respectively. X-ray diffraction (XRD) analysis indicated polycrystalline cubic structure. SEM and AFM pattern revealed that, the film was uniformly deposited by crystalline grains over the entire glass substrate. UV-VIS absorption spectra were recorded within the range of 300-1100 nm. It shows high absorption with direct optical band gap energy equal to 2.39 eV. The electrical resistivity shows semiconducting behavior of the film.
Keyword: *Solution growth; semiconductor; photovoltaic; thin film*

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I. Introduction

Wide energy band gap II–VI semiconductor materials are recently more important for the optoelectronic devices as well as photovoltaic device fabrication process. For low cost and high-performance photovoltaic devices, it is necessary to use appropriate materials, which are technologically excellent. II-VI group semiconductor materials possess wide range of electrical and optical properties due to increased recombination efficiency, greatest absorption coefficient and strong photosensitive properties. These properties of materials are important to fabricate optoelectronic and photovoltaic devices like solar cells. CdZnSe is a ternary compound of II-VI group semiconductor materials. Band gap energy of such material is tuned by varying Cd and Zn concentrations. This can be done by varying value of x=0 to 1 in $Cd_{1-x}Zn_xSe$ composite thin films. The most important applications of $Cd_{1-x}Zn_xSe$ thin films reported are photo detectors [1, 2] solar cells [3, 4], PEC cells [5], LEDs [6,7], Photodiodes [8]. Syntheses of CdZnSe thin films have been carried out by many researchers by different techniques such as, thermal evaporation [9,10], electrodeposition [3,11], photoelectron deposition [12], epitaxial growth [13], pulse plating technique [14], Chemical bath deposition [7,15-18]. Among them chemical bath deposition (CBD) technique is the low cost, low bath temperature and no special instrumentation is required. In CBD, controlled chemical reaction plays a key role for the deposition of the thin film. The rate of deposition is controlled by adjusting the bath parameters like bath temperature, pH of solution, stirring rate and relative concentration of solutions in the bath [19]. In this technique substrates are immersed in an alkaline solution containing the chalcogenide sources, the metal ion, added base and complexing agent.

In the present investigation, I report the growth and characterization of Cadmium Zinc selenide $Cd_{1-x}Zn_xSe$ (at x=0.6) thin film deposited by Chemical Bath Deposition (CBD) technique and characterized for structural, optical, surface morphology and electrical study.

II.Experimental

Chemicals used

Chemicals used to deposit $Cd_{0.4}Zn_{0.6}Se$ thin film were Cadmium acetate [Cd $(CH_3COO)_2$ 2H2O], zinc acetate [$Zn(CH_3COO)_2.2H_2O$], sodium sulfite (Na_2SO_3), elemental selenium powder (99.9%), and triethanolamine (TEA). These chemicals were purchased from Merck, Mumbai, India and 30% ammonia solution was purchased from SD Fine-Chem Limited, Mumbai, India. All reagents used were of AR grade. Deionized water was used to prepare precursor solutions. Commercially available microscopic glass slides (BlueStar, Polar Industrial Corporation, Mumbai, India) with the dimension of 75mm x 25mm x 2mm were used as glass substrates.

Preparation of Cd $_{0.4}$Zn$_{0.6}$Se thin films

Aqueous solutions of cadmium acetate, zinc acetate and sodium selenosulfate were used as the sources of $Cd^{2+,}$ $Zn^{2+}$and Se^{2-} respectively. The electrolyte solutions were prepared by dissolving $Cd(CH_3COO)_2.2H2O$, $Zn(CH_3COO)_2.2H_2O$ in double distilled water. The substrates used for the deposition of $Cd_{0.4}Zn_{0.6}Se$ thin film were commercial microscope glass slides with the size of 75mm×25mm×2mm. Before deposition, the substrates were degreased in HNO_3 solution for 24h, cleaned by commercial detergent, washed in deionized water and dried in air. This process is done to ensure a clean surface, which is necessary for formation of nucleation centers, required for thin film deposition. The value of x=0.6 is adjusted by taking proportional volumes of cadmium and zinc precursor solutions of same molar concentration solutions. 8 mL of 1 M cadmium acetate and 12 mL of 1 M zinc acetate solutions were taken in a beaker of 50mL volume and magnetically stirred for few minutes. 5mL of TEA was added to this mixture with continuous stirring. On addition of TEA, it was found that the color of the mixture turned in to milky. Then, 30% ammonia solution was added slowly with constant stirring till the milky Solution turned in to clear solution. After this, 20mL of sodium selenosulfate (Na_2SeSO_3) solution was added in the beaker. Sodium selenosulphate was prepared by the method, reported earlier [20]. The whole reaction mixture was stirred for few minutes. The pre cleaned glass substrates were then immersed vertically in the beaker. The beaker was then kept at constant temperature of 80 Degree Celsius in a water bath for 5 hours. The pH of reaction bath was 11.

The electrochemical reaction on the cathode takes place as follows:

$$Cd(CH_3COO)_2.2H_2O \longrightarrow Cd^{2+} + (CH_3COO)_2^{2-} + 2H_2O \quad (1)$$

$$Zn(CH_3COO)_2.2H_2O \longrightarrow Zn^{2+}+(CH_3COO)_2^{2-}+2H_2O \quad (2)$$

$$(1–x)Cd^{2+}_{(aq)} + 2e^- \longrightarrow (1–x)Cd_{(s)} \quad (3)$$

$$xZn^{2+}_{(aq)} + 2e^- \longrightarrow xZn_{(s)} \quad (4)$$

$$Na_2SeSO_3 + OH^- \longrightarrow HSe^- + Na_2SO_4 \quad (5)$$

$$HSe^- + OH^- \longrightarrow H_2O + Se^{2-} \quad (6)$$

$$(1–x)Cd_{(s)} + xZn_{(s)} + Se_{(s)} \longrightarrow Cd_{1-x}Zn_xSe \quad (7)$$

After 5 hours substrate coated with $Cd_{0.4}Zn_{0.6}Se$ was removed, rinsed with distilled water, and dried in open air at room temperature. Film obtained was uniform, well adherent and radish in color [17].

The glancing incidence X-ray diffraction (GI-XRD) patterns was recorded using Bruker AXS X-ray diffractometer (Model D8 Advanced, Germany), with CuKα1 radiation of λ = 1.5406A˚ and glancing angle θ = 0.5˚ in the detector scan mode (scan rate is 0.002) by keeping the samples fixed. SEM images are recorded by JOEL-JSM 5600. The optical absorption spectra were recorded on the Systronics spectrophotometer-17 within the wavelength range 300–1100 nm.

III. Results And Discussions

XRD Analysis

The structural determination and crystallite size of $Cd_{0.4}Zn_{0.6}Se$ thin film was carried out by X-ray diffraction (XRD) pattern. Fig. 1 shows X-ray diffraction pattern of as-deposited $Cd_{0.4}Zn_{0.6}Se$ thin film. One large and two small peaks are observed. More than one XRD peaks indicate that the as deposited film is polycrystalline in nature. The 2θ peaks at 26.43°, 41.16° and 52.046° are observed. These peaks are in between CdSe thin film (25.7°, 41.4° and 50.7°) [22] and ZnSe thin film (27.45°, 45.57° and 54.06°) [20]. These peaks are shifted towards ZnSe side as more concentration of Zn is large at x=0.6. This indicates that formation of $Cd_{0.4}Zn_{0.6}Se$ thin film with cubic structure. The crystallite size (D) was calculated by Scherrer's formula [9] from the full width at half maxima (β) of the peaks expressed in radians and found to 10.25 nm.

$$D = \frac{K\lambda}{\beta Cos\theta} \quad (8)$$

where 'K' is constant dependent on crystallite shape (0.89), 'λ' is wavelength of CuKα1 radiation, and 'θ' is angle between the incident and scattered X-rays. The average crystallite size (derived from Fig. 1) is found 10.25 nm.

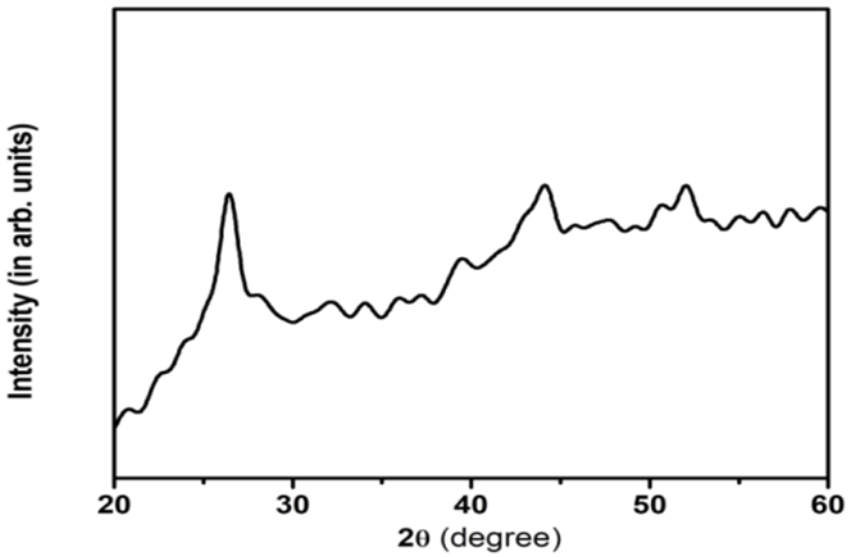

Fig. 1 XRD pattern of as deposited $Cd_{0.4}Zn_{0.6}Se$ thin film

Surface Morphology Analysis

Surface morphology of the films was studied by scanning electron microscope (SEM) and atomic force microscopy (AFM) images. Fig. 2 (a) shows SEM image of as-deposited ternary $Cd_{0.4}Zn_{0.6}Se$ thin films. It is observed that the spherical shaped nanoparticles with vacant spaces were covered on the entire substrate surface. The distribution of particles may become more ordered and the vacant spaces between them get occupied after the annealing of thin film [17]. The fine grains were well defined and spherical with different sizes, distributed over the substrate without any cracks and correspond to the crystalline phase of $Cd_{0.4}Zn_{0.6}Se$.

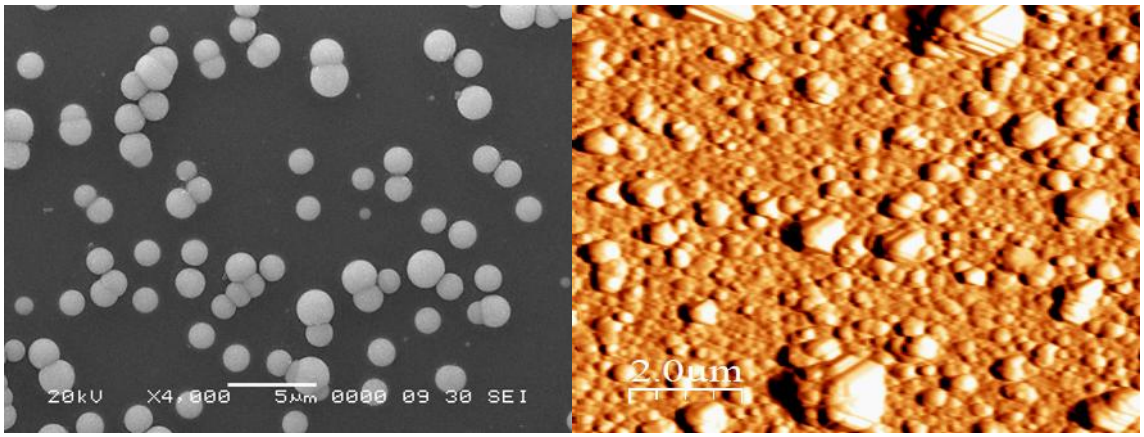

Fig. 2(a) SEM image of as deposited $Cd_{0.4}Zn_{0.6}Se$ thin film

Fig. 2(b) AFM image of as deposited $Cd_{0.4}Zn_{0.6}Se$ thin film

Fig. 2 (b) shows AFM image of as-deposited $Cd_{0.4}Zn_{0.6}Se$ thin film. From the image it is clear that the film is uniform and substrate surface is well covered by fine spherical or elliptical grains. The average cluster size was determined to be 130 nm and surface roughness was 0.3 nm. The cluster size and root mean square (rms) surface roughness were determined by using the software which was provided with the microscope. The surface roughness of the film is unavoidable in our case due to particles are spherical in shape.

Optical Analysis

Fig. 3(a) shows UV-VIS absorbance spectra of as-deposited $Cd_{0.4}Zn_{0.6}Se$ thin film. The absorbance spectra were used to study the optical transition in the films, which were studied at room temperature in the wavelength range of 300–1100 nm. The as-deposited $Cd_{0.4}Zn_{0.6}Se$ thin film showed an absorption edge at 524 nm. The optical absorption studies revealed that the film is highly absorptive and have direct type of transitions, which allowed the determination of optical band gap by the following Urbach relationship [19]

$$\alpha h\nu = A\left(h\nu - E_g\right)^n \quad (9)$$

where 'A' is the constant; depending upon the transition probability for direct transition, n = 1/2 for direct allowed transition and 'E_g' is the optical band gap of the material.

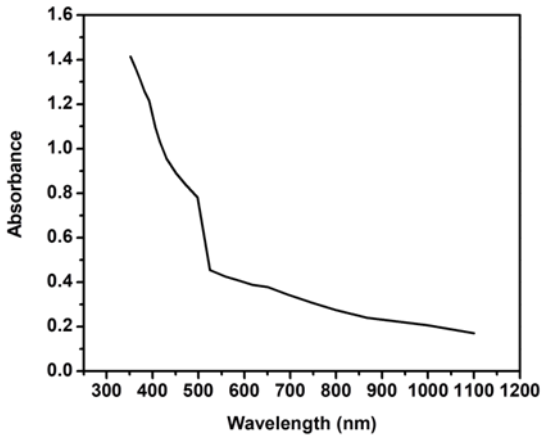

Fig. 3(a) Absorbance spectra of as deposited $Cd_{0.4}Zn_{0.6}Se$ thin film

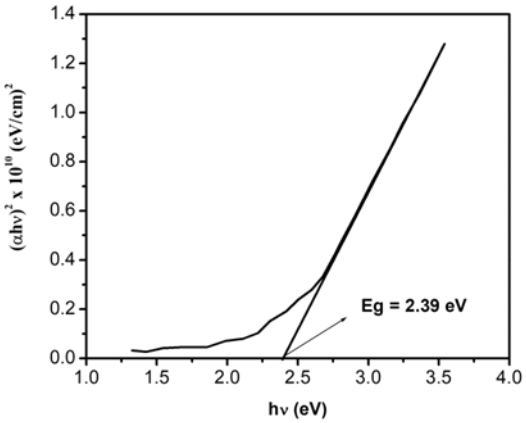

Fig. 3(b) plot of $(\alpha h\nu)^2$ vs hν of as deposited $Cd_{0.4}Zn_{0.6}Se$ thin film

Fig. 3(b) shows the variation of $(\alpha h\nu)^2$ versus hν. Extrapolating the straight-line portion of the plot of $(\alpha h\nu)^2$ versus hν for zero absorption coefficient value gives the band gap energy, which is found to 2.39 eV. The band gap energy value is shifted towards ZnSe side as band gap energy of CdSe is 1.88 eV [21] and of ZnSe is 2.7 eV [19]. It indicates that exact x=0.6 composition $Cd_{0.4}Zn_{0.6}Se$ thin film is formed.

Electrical Resistivity Analysis

The dark electrical resistivity of the thin film was measured using a DC two-probe method in the temperature range 300–500K. A plot of inverse absolute temperature versus logρ for cooling cycle is shown in Fig. 4. The dependence is almost linear indicating the presence of only one type of conduction mechanism in the film. Our experimental data fit in to the relation,

$$\rho = \rho_0 exp\left(\frac{E_a}{KT}\right) \quad (10)$$

where 'ρ' is the resistivity at temperature T, 'ρ_o' is a constant, 'k' is Boltzmann's constant, 'T' is the absolute temperature and 'E_a' is the activation energy.

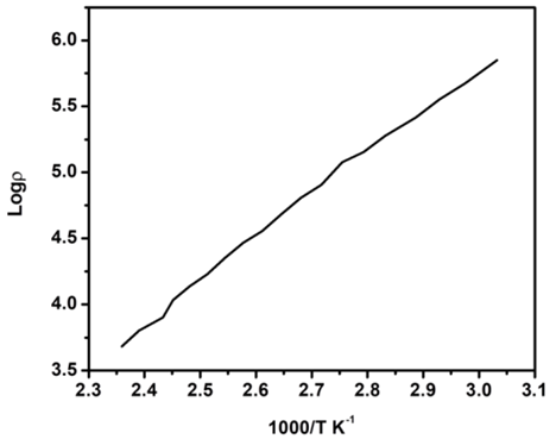

Fig. 4 Variation of logρ (Ω cm) vs. 1000/T (K^{-1}) of as-deposited $Cd_{0.4}Zn_{0.6}Se$ thin film

The high-temperature conductivity is a thermally activated excitation of charge carriers from grain boundaries to the region of the grains. The decrease in resistivity with increase in temperature confirms the semi-conducting behavior of the film. The room temperature electrical resistivity was found to be 0.99 X 10^6 Ω cm.

IV. Conclusion

The thin films of $Cd_{0.4}Zn_{0.6}Se$ were deposited on to glass substrates from aqueous alkaline reaction bath by CBD. The XRD studies showed that the as-deposited film has polycrystalline cubic structure. Electrical resistivity study showed semiconducting nature of the film. SEM and AFM iages showed uniform deposition of film on to the glass substrate. Optical study showed band gap energy equal to 2.39 eV. The optical band gap energy shifted toward ZnSe side, because Zn concentration is more than the Cd concentration for x=0.6. So in $Cd_{1-x}Zn_xSe$ thin film, by adjusting the value of x from 0 to 1, optical band gap energy varies from 1.88 eV to 2.7. Optical band gap energy is the more important physical property of semiconductor material that is useful for various photovoltaic and photoelectro-chemical cell applications, which is tunable in $Cd_{1-x}Zn_xSe$ film.

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