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Influence of Sulphur doping on Optical and Structural Parameters of Thermally Evaporated Non-crystalline CdS_xSe_{1-x} Thin Films

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ABSTRACT

Cadmium sulfoselenide $CdS_xSe_{1-x}(x=2,4,6 \text{ and } 8\%)$ films were deposited by thermal evaporation technique under the vacuum of 3×10^{-6} Torr on the glass substrates. The structural, compositional, optical and thermal properties of the deposited CdS_xSe_{1-x} thin films were studied. X-ray diffraction studies confirmed that the deposited films are amorphous in nature. With the help of transmittance spectra, the refractive index (n), and excitation coefficient (k) are determined at room temperature in the wavelength range 800-1800 nm. Energy dispersive analysis by X-ray (EDAX) is used to investigate the compositional elements of thin films. The presence of Cd, S and Se of the CdS_xSe_{1-x} thin films and the composition of CdS_xSe_{1-x} thin films are estimated by EDAX analysis. The differential scanning calorimetry (DSC) was used to study the thermal properties of the deposited films.

Graphical Abstract



Highlights

- Cadmium sulfoselenide CdS_xSe_{1-x}(x=2, 4, 6 and 8%) films were deposited by thermal evaporation technique.
- X-ray diffraction studies confirmed that the deposited films are amorphous in nature.
- The presence of Cd, S and Se of the CdS_xSe_{1-x} thin films and the composition CdS_xSe_{1-x} thin films are estimated by EDAX analysis.
- DSC graph shows that on increasing the S concentration, the melting point of the glasses also increasing.

Keywords: Amorphous materials, Chalcogenides glass transitions, Optical Properties, Thin films.

INTRODUCTION

II-VI semiconducting materials have high potential in research field because of their interesting properties like wide band gap, high absorption coefficient, binding energy and high chemical stability. It can be applied as environment friendly device due to their unique interesting properties such as high photosensitivity, structural electrical and optical properties. The possibility of change their properties by exposing them to the external effects such as laser beams, gamma rays, light, x-rays and thermal waves are viable features of these materials [1-4]. Based on the type of the vacancy created during the deposition, and the deposition technique, the type of the semi conductivity and electrical behavior of the II-VI thin films can be determined. Amorphous chalcogenide compositions are widely used in optical DVDs and non- volatile memory devices [5-7].In the past few decades, the Cd-based chalcogenide alloys and compounds were subjected to extensive research [8-12] in optoelectronics, photovoltaics, photo-catalysis, gamma-ray detectors, sensors, quantum computing, thin film transistor and laser diodes.

The formation of amorphous materials requires the prevention of nucleation and growth processes which are responsible for the crystallization. This implies that the amorphous phase is thermodynamically less stable in comparison to the crystalline form as it possesses a greater free energy. Therefore, amorphous materials are prepared through non-equilibrium process. CdSe and CdS are II-VI semiconductors widely used as photoconductors in various electronic devices [13-19]. The thermal evaporation process is widely used because in this process we easily controlled the preparative conditions as lick evaporation rate, film thickness, surface morphology and the structural state [20, 21]. Many researcher deposited ternary CdS_xSe_{1-x} compositions by different techniques such as thermal evaporation technique [22-27], laser ablation technique [28], chemical spray pyrolysis [29, **30**], chemical bath deposition (CBD) technique [**31**, **33**] and mechanical milling technique [**34**]. The thin films of semiconducting compounds and alloys including CdSSe ternary systems were deposited by vacuum evaporation techniques. The amorphous chalcogenide semiconductors contain large number of localized states in the mobility gap [35-38]. In the present study, we report the influence of the S on the physical properties of the ternary chalcogenide non-crystalline CdS_xSe_{1-x} compositions. The cadmium sulfoselenide thin films were synthesized by using the physical evaporation technique. $CdS_xSe_{1-x}(x = 2,4,6 \text{ and } \%)$ thin films were characterized by XRD, EDAX, DSC and Optical properties and obtained results are discussed. A comparative study was carried out among the present thin films and similar samples of the previous literature.

MATERIALS AND METHODS

Synthesis and Preparation of Thin Film samples: Glassy alloys of CdS_xSe_{1-x} (x = 2, 4, 6, and 8 %) are prepared by melt quenching technique. High purity mixtures of starting elemental powders of (99.999%) from S, Se, Cd (Sigma-Aldrich) were weighed in required atomic percentages using a 5digit sensitive balance according in different stoichiometric quantities. Input powders of S, Se and Cd elements were agitated continuously in order to intermix the components as well as to ensure the homogeneity. In the present work, various compositions of CdS_xSe_{1-x} glasses in bulk form were prepared using melt quenching technique. The quenching rates extend from $\sim 10^{20}$ C s⁻¹ for chalcogenide glasses to $\sim 10^3 - 10^{50}$ C s⁻¹ for complex metallic glasses. The material was then sealed in evacuated for this purpose, cleaned quartz ampoules were used. The quartz ampoules (outer diameter ~ 10 mm, inner diameter ~ 8 mm and length ~ 10 cm), the weighed mixture of constituents is poured into the tubes and ampoules were formed by sealing the tubes under a vacuum of 10^{-3} Torr. In the beginning, temperature of the furnace is raised to 113°C at a rate of 2°C min⁻¹, in steps of 2h. Melting temperature, S=112.8°C,Se=217°C,Cd= 321°C.The ampoules were maintained at the temperature of 950°C and was held at that temperature for about 24h with continuously rocking for proper mixing and homogenization of the melt. The obtained melt was rapidly quenched in liquid nitrogen to avoid crystallization. The thin films CdS_xSe_{1-x} were deposited onto glass substrates and substrates were cleaned successively using isopropanol, ethanol, acetone, and finally ultrasonic cleaning. Vacuum

evaporation method by using Hind High Vacuum Coating Unit (12A4D) at a vacuum of the order 2×10^{-6} Torr. Molybdenum boat sources were used for the thin film growth process. To attain thermodynamic equilibrium the films were kept inside the deposition chamber for 24h as suggested by Abkowitz [**39**]. Film thickness and deposition rate were controlled during deposition by a quartz crystal oscillator. The deposition rate was kept constant at ~2.5 nm s⁻¹. The chalcogenide CdS_xSe_{1-x} thin films were controlled to get films have the same thickness. The accuracy of the film thicknesses was checked after the evaporation process using profilometer and obtained that the thin films have thicknesses about 200nm.

RESULTS AND DISCUSSION

Structural identification: The amorphous nature of the chalcogenide $CdS_xSe_{1-x}(x = 2,4,6 \text{ and } 8)$ thin films was checked by JEOL X-ray diffractometer (Model JSDX-60 PA). It was operated at 40kV and 35 mA. The used source was CuK_{α} -radiation which has a wavelength of 0.154 nm and energy of 8.04KeV. This power is sufficient to examine the thin films samples continuous scanning was applied with a slow scanning rate (1° min⁻¹) and a small-time constant (1S) to detect any probable diffraction line. X-ray diffractometers of all thin films samples were recorded at room temperature. The diffraction angle (2 θ) of XRD patterns was ranged from 20° to 70°. The X-ray diffraction pattern of different CdS_xSe_{1-x} thin films shown in figure 1 reveals that, no discrete or sharp diffraction lines were observed in all glassy samples, which confirm the amorphous nature of the prepared glass compositions. X-ray diffraction pattern of all samples were found to have almost similar trends.



Figure 1. X-ray diffraction patterns of CdS_xSe_{1-x}thin films with different compositions.

Optical parameters: Optical parameters of the chalcogenide thin films of CdS_xSe_{1-x} (x = 2,4,6 and 8%) matrix plays an important role in understanding the optoelectronic nature [40, 41]. These parameters can be interpreted in the interaction between the incident photons and the deposited films. Furthermore, optical properties of thin films can change or affect the characteristic incident light spectrum passing through these films. Optical investigation was measured using the transmittance

spectra in figure 2 within the range 800-1800 nm and with the help of UV-Vis-NIR Spectrophotometer (Shimadzu U-3600). Optical transmission (T) is a very complex function and is strongly dependent on the absorption coefficient (α) of the material. According to Swanepoel method [42, 43] the envelope of the interference maxima and minima, occurs in the spectrum can be utilized for obtaining optical parameters as refractive index and extinction coefficient (n and k).



Figure 2. Transmission Spectra of CdS_xSe_{1-x} thin films with different compositions.

The refractive Index (n) calculated with the help Transmission Spectra by using formula (Manifacier Envelope Method).

$$n = [N + (N^{2} + n_{0}^{2} \cdot n_{1}^{2})^{1/2}]^{1/2} \dots (1)$$

Where n_0 and n_1 are the refractive index of air and substrate respectively.

The number N is given by the following equation.

$$N = [(n_0^2 + n_1^2)/2] + 2n_0n_1[(T_{max} - T_{min}) / (T_{max} - T_{min})] \quad ...(2)$$

Where T_{max} and T_{min} are the upper extreme point and lower extreme point at a wavelength. The extinction coefficient (k) is given by -

$$K = (-\lambda/4\pi t). \ln P \qquad \dots (3)$$

Where't' is the thickness of the film and P is given by the following equation,

$$P = C_1/C_2 [1-(T_{max} / T_{min})] / [1+(T_{max} / T_{min})] \qquad \dots (4)$$

Where, $C_{1=}(n+n_0)$ $(n+n_1)$, n is the refractive index film.

 $C_{2=}(n-n_0)$ (n₁-n), n₁the refractive index of the substrates and n₀ is the refractive index of the air.

Figure 3 shows that at lower wavelength the refractive index (n) have a higher value. This is due to the frequency of incident photons becomes equal to the plasma frequency. When the wavelength is decreased the value of the refractive index becomes larger as well as the absorption of incident electromagnetic radiation is increased. The refractive index is sharply decreasing and increasing for all thin films irrespective of S percentage in CdS_xSe_{1-x} compositions. The value of the refractive index was found to be dependent upon the composition as well as on the stoichiometry of the CdSSe composition [44]. All thin films exhibit the same behavior of refractive index versus wavelength as a

normal dispersion. The extinction coefficient k can be calculated from the absorption coefficient, α by the following equation k=(- $\lambda/4\pi t$) lnP, where λ is the wavelength of the incident photons. Figure 4 represents the variation of k as a function of λ for the incident electromagnetic radiation. This figure reveals that, k is decreasing with increasing the wavelength of incident photons. Mottand Davis had mentioned a similar trend for various other amorphous semiconductors [45-48].



Energy Dispersive X-ray: The composition of the CdS_xSe_{1-x} film was investigated using energy dispersive analysis of x-ray (EDAX) is shown in figure 5, was used to study the quantitative elemental analysis as the study of the stoichiometry of CdS_xSe_{1-x} thin films. It is thought that some impurity elements with lest amount may probably results from glass used as substrate. The elemental analysis was carried out for Cd, S, and Se elements of the ternary CdSSe thin films. The sample was slightly selenium rich, which is in good agreement with the report of Tomkiewicz *et al* [49], Skyllas Kazacos



Figure 5. EDAX Spectra of CdS_xSe_{1-X} thin films with different compositions.

Differential Scanning Calorimetry: The Calorimetric measurements were carried out by using Differential Scanning Calorimetry (DSC) is shown in figure 6, at heating rate of 10 K min⁻¹ of different composition of CdS_xSe_{1-x} (x = 2, 4, 6 and 8) glasses. From the DSC graph, it is concluded that the melting points of prepared glasses CdS_xSe_{1-x} is increasing with increasing S concentration and the melting point achieved in the different composition is 474K, 489K, 491K and 501K.



Figure 6. DSC Spectra of CdS_xSe_{1-x} thin films with different compositions.

APPLICATION

The II-VI amorphous chalcogenide compositions thin films can be used in optical DVDs and non-volatile memory devices photovoltaics, photo-catalysis, gamma-ray detectors, sensors, thin film transistor and laser diodes.

CONCLUSION

The CdS_xSe_{1-x} (x = 2, 4, 6 and 8%) thin films deposited by thermal evaporation technique. The X- ray diffraction shows that the prepared material is amorphous in nature. It is concluded by this study that the transmission spectra of II-VI group CdS_xSe_{1-x} semiconductors thin films are sufficient to determine the optical constant such as refractive Index and extinction coefficient. The presence of elemental constituents was confirmed from EDAX analysis and image shows the presence of strong peaks for Se, Cd and S and no other impurity peaks confirm. DSC graph shows that on increasing the S concentration, the melting point of the glasses also increasing. These films can be used in optoelectronic devices.

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