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Deposition and Characterization of Indium Selenide Thin Films for Opto-electronic Devices

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Amongst all the III-VI semiconductors, ones that crystallize with a layered structure have gained special interest due to its significant usage in photovoltaic devices. III-VI layered semiconductor such as InSe has low density of dangling bonds on its surface therefore it is considered as vital material for the fabrication of opto-electronic devices like photo sensor, solar cell etc. In present work, InSe thin films were fabricated through a simple and facile drop-casting method, where the thin films were drop-casted between two silver paste electrodes on a glass substrate. The structural, surface morphological, compositional, electrical and optical properties of the prepared films were obsrved by XRD, SEM, EDAX, high precision digital multi-meter and UV-visible spectroscopy, respectively. XRD analysis of the prepared film shows the existence of nano-crystalline nature with monoclinic crystal structure of InSe. SEM images show good continuity of InSe film. InSe thin films are n-type with bandgap of 1.8 eV and their electrical conductivity is in the order of 10^{-10} S/cm that makes them appropriate for using as an absorber layer in the solar cell.

Keywords: InSe, Thin film, Drop-casting, Absorber layer, Opto-electronic devices.

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1. INTRODUCTION

Alloy based chalcogenides are important from the application point of view because of their naturally thin body and exceptional semiconductor performances [1-2]. These chalcogenides have shown plenty of favorable applications in optoelectronics, photonics, super capacitors, flexible transparent electrodes and memory switching devices [3-4]. The metal chalcogenide semiconductor exhibit layered structure which can be exfoliated. Their bandgap depends on exfoliated layer thickness as well as 2D quantum potential well formed by their boundaries. Among them, III-VI binary semiconductors have prime application in optical, optoelectronic and thermal electrical devices [5-6].

Among several metal chalcogenides indium selenide is III-VI layered semiconductor having five crystalline phases designated as α, β, γ, δ and κ [7]. InSe has two crystalline surfaces with discreet physical properties [8]. The absorption coefficient of InSe is 10^5 cm^{-1} in the visible region of solar spectrum. InSe is thus a potential candidate for absorber layer of thin film solar cell since a major portion of the solar spectrum can be absorbed by it. InSe material can be both n-type and p-type depending on the doping and growth condition [9].

Various physical and chemical methods such as molecular beam epitaxy, thermal vapor deposition, e-beam deposition, spray pyrolysis, chemical bath deposition, chemical vapor transport, electrochemical atomic layers epitaxy have been employed for deposition of InSe thin films [10-21]. However numerous draw-backs are associated with these methods including high vacuum cycles, complexity, low yield and multistep hence resulting in raising expenses [22]. In this paper, a simple and facile technique i.e. drop-casting method for InSe thin films deposition on a glass substrate has been studied. The structural, surface morphological, compositional characterization, optical and electrical properties of InSe film has been studied. The above technique for the preparation of InSe thin films proposes several benefits including cost-effectiveness and convenience.

2. EXPERIMENTAL DETAILS

2.1 Thin Film Preparation

Both the constituent elements, Indium and Selenium were purchased from RESEARCH-LAB FINE CHEM INDUSTRIES, India, of 99.99 % purity. They were weighed in a stoichiometric ratio and processed further for sealing. Both the elements were taken into the quartz tube of diameter 1 cm and length 20 cm and was evacuated at 10^{-5} torr. The quartz tube was placed slightly slanted in the tube furnace at 900 °C, so that proper mixing takes place. Special care was taken to ensure that the temperature of 900 °C remained constant for 1 hour so that the entire charge gets into molten state. In order to inhibit frictional crystallization and precipitation after mixing, the molten form of the constituent elements was allowed to cool down very slowly for a day for the formation of the ingot. The resultant ingot so obtained was crushed into a fine powder which is suitable for InSe thin film fabrication.

2 g InSe (Indium Selenide) powder and 0.05 g PVP (Polyvinylpyrrolidone) were added in 60 ml of Isopropyl alcohol. The mixture was sonicated for 1 hour using ultrasonic probe sonicator. The solution slowly turned greyish black. The obtained solution was used to perform the drop-casting method at 180 °C on a glass substrate for the preparation of InSe thin film.

2.2 Characterization

XRD plots of the InSe thin films were recorded with a Rigaku Miniflex-II Desktop XRD diffractometer oper-

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ated from 10º to 90º with Ni-filtered Cu Kα radiation. The surface morphology of the prepared films was studied using Scanning Electron Microscopy (Hitachi S-3400N) at 15 kV operating voltage. The sample composition was confirmed by EDAX using JEOL JSM-5610LV. The variation in the optical properties was examined by transmission and reflection spectra in the range of wavelength from 400-1100 nm using UV-Vis spectrophotometer (Varian Cary 300). Electrical properties were observed using high precision digital multimeter (ET3240).

3. RESULTS AND DISCUSSION

3.1 X-ray Diffraction

InSe thin film was investigated with the help of XRD. InSe thin films generally crystallize in monoclinic structure. The XRD pattern of InSe thin film prepared on glass substrate is shown in Fig. 1. XRD peaks are observed at 2θ angles of 22.28º, 33.16º, 44.36º and 68.28º that corresponds to the reflections from (100), (004), (200) and (008) planes. Most of the peaks in the diffraction pattern are best matched with JCPDS card number 44-1007 for InSe. The strongest XRD peak in the diffraction pattern at 2θ, 22.28° is because of diffraction from the (100) planes that reports monoclinic crystalline structure of InSe thin film.

The crystalline grain diameter in the film evaluated using Scherrer's formula is 68 nm.

$$d = \frac{0.9\lambda}{\beta \cos\theta} \qquad (3.1)$$

where, d is diameter of nanocrystal, k is a constant ($k = 0.9$), λ is the wavelength of the X-ray ($\lambda = 1.54056$ Å), β is the FWHM (full width half maxima), and θ is the Bragg's angle.

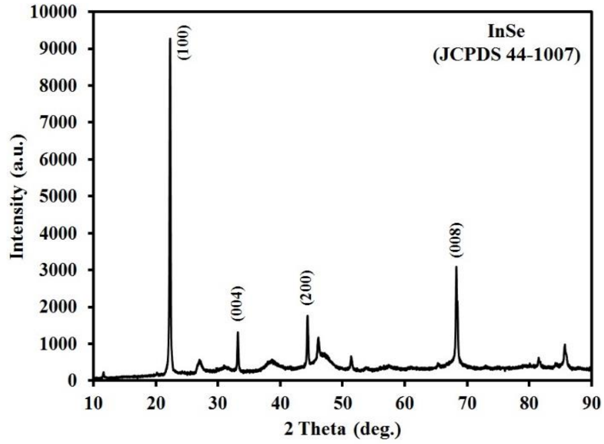
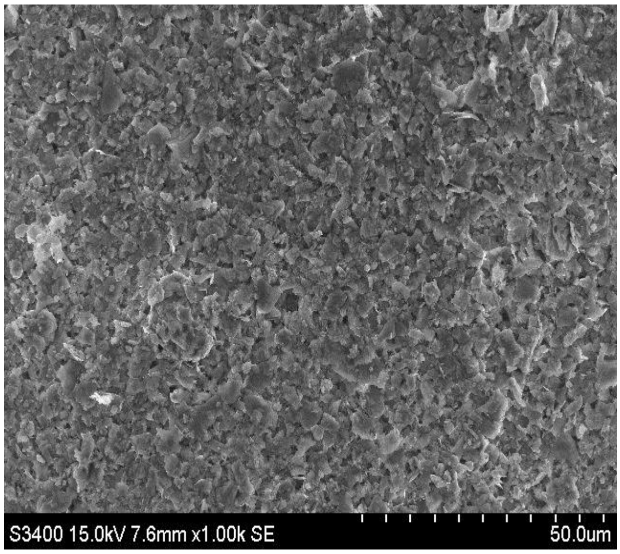


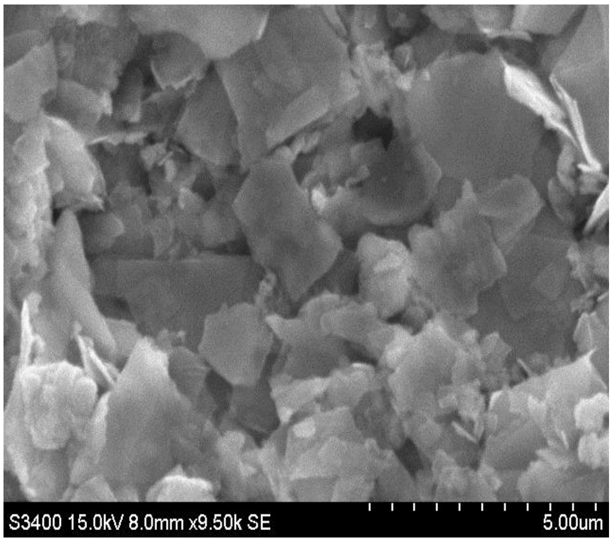
Fig. 1 – X-ray diffractogram of as-prepared InSe thin film

3.2 Morphological Characterization and Compositional Analysis

SEM images in Fig. 2a and 2b show that the InSe thin film consist of compact particulates. Upon observation, the surface of the substrate was found to be covered with nano flakes. They also show good continuity of the film. Fig. 2a and 2b show the typical microstructure of the film. The particle sizes are ranging from 1-1.5 μm. Here, the particle sizes are larger than the crystallite sizes calculated with the help of the Debye-Scherrer equation. These results indicate that the InSe particles as a matter of fact are compact agglomeration of small grains due to inter atomic interaction between the particles.



a



b

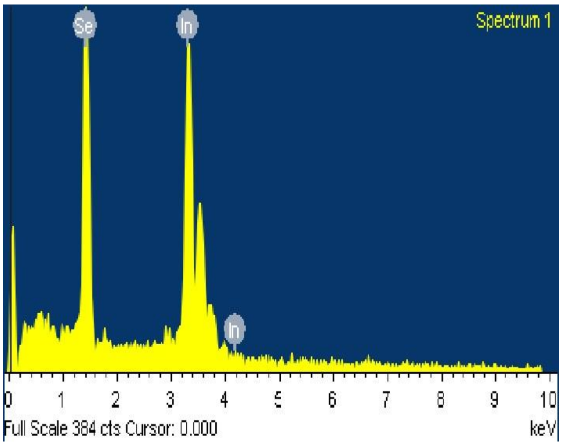
Fig. 2 – Scanning Electron Microscopic image of InSe thin film at ×1000 (a) and ×9500 (b)

Energy-dispersive X-ray spectroscopy (EDAX) of the InSe thin film is revealed in Fig. 3. The composition of the synthesized film is validated by EDAX. It indicates that 57.39 % Indium and 42.61 % Selenium is present in the sample. Some background peaks in Fig. 3 are due to the glass substrate.

3.3 Optical Properties

Fig. 4 shows the transmission plot of transmission versus wavelength for InSe thin film. The maximum transmission is observed in the range of wavelength

200 nm to 600 nm. After that the curve sharply decreases. The oscillating behaviour of the transmission curve for the InSe thin film shows that it has low surface roughness and good homogeneity.



Element	Weight %	Atomic %
Se L	42.61	51.92
In L	57.39	48.08
Total	100	100

Fig. 3 – Composition of InSe thin film by EDAX

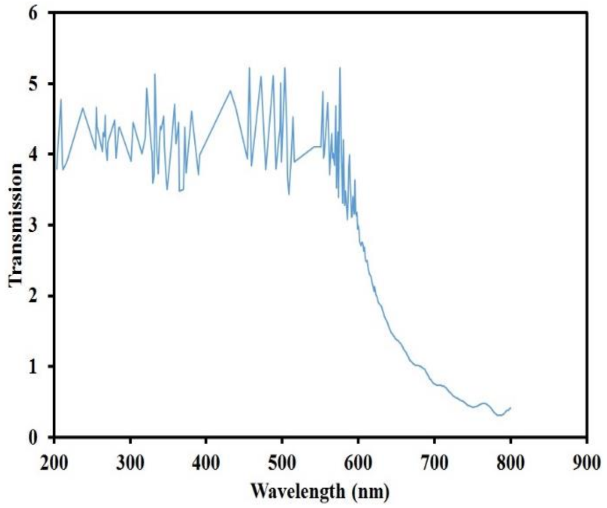


Fig. 4 – Transmission plot of InSe film

Fig. 5 shows the typical plot of $(\alpha h\upsilon)^2$ versus $h\upsilon$ for InSe thin film. The UV-Vis absorption spectra was carried out in the range of wavelength 400-1100 nm to prove the optical properties of synthesized InSe thin film. The film shows good absorption in the range from 400 nm-700 nm i.e. in visible region. Tauc relationship was applied to determine the optical bandgap of the film.

$$\alpha h\upsilon = A\left[h\upsilon - E_g\right]^n , \tag{3.2}$$

where, $h\upsilon$ is the photon energy, E_g is the bandgap energy, n is 1/2 for direct band gap.

Here, the value of the absorption coefficient is observed in the order of $10^5\ \text{cm}^{-1}$. The extrapolation in the linear plot of $(\alpha h\upsilon)^2$ versus $h\upsilon$ at $(\alpha h\upsilon)^2 = 0$ results in the optical bandgap of 1.8 eV (Fig. 5) [21]. This is in agreement with reported value.

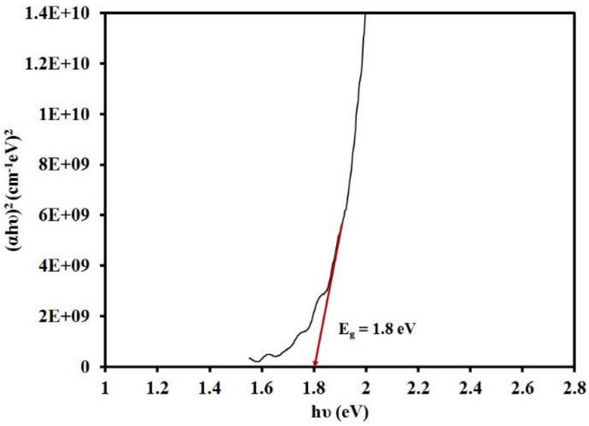


Fig. 5 – Tauc plot of $(\alpha h\upsilon)^2$ vs. $h\upsilon$ to determine band-gap of InSe film

3.4 Electrical Properties

Silver paint was used as contacts on InSe thin film as it gives ohmic contact [20]. For the assessment of electrical conductivity response, biasing of 20 V is applied across the electrode. The film shows n-type conduction as determined by hot probe method. *I-V* characteristics of typical InSe film in dark and under illumination is presented in Fig. 6.

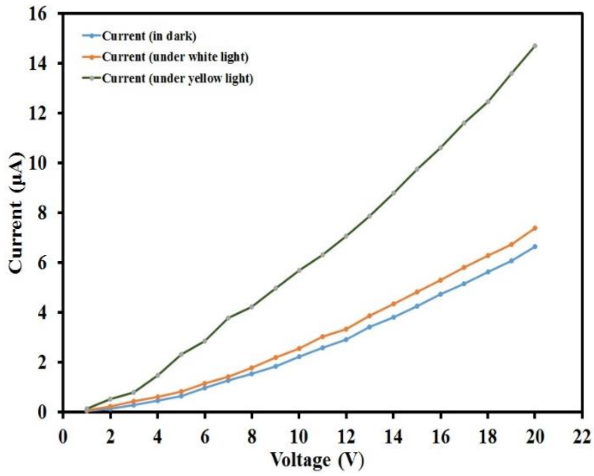


Fig. 6 – *I-V* characteristics of InSe film

The resistances of the film in darkness, under white light and under yellow light at 300°K were, $R_D = 2.82\ \text{M}\Omega$, $R_W = 2.55\ \text{M}\Omega$ and $R_Y = 1.31\ \text{M}\Omega$ respectively. The electrical conductivities of film in darkness, under white light and under yellow light at 300°K were, $\sigma_D = 7.10 \times 10^{-10}$ S/cm, $\sigma_w = 7.85 \times 10^{-10}$ S/cm and $\sigma_Y = 15.3 \times 10^{-10}$ S/cm respectively. The decrement in resistance and increment in electrical conductivity upon illumination clearly indicates the existence of photo conductivity in the sample.

4. CONCLUSIONS

InSe thin films were synthesized using a simple, facile and cost-effective drop-casting method. The XRD peaks show monoclinic crystalline structure of the synthesized thin film. The SEM images show good continuity of the film. The composition of InSe film is validated by EDAX. It indicated that 57.39 % Indium and 42.61 % Selenium exists in the sample. With the help of hot probe method, it is observed that the present film is *n*-type. The obtained value of electrical conductivity is in the order of 10^{-10} S/cm that is measured by *I-V* characteristics. The optical bandgap of InSe thin film is 1.8 eV with high optical absorption coefficient which makes it convenient for using it as an absorber layer in the solar photovoltaic cell.

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