

## EFFECT OF NICKEL IONS AND PHYSICAL CHARACTERIZATION OF SELINIDE THIN FILMS BY CHEMICAL DEPOSITION METHOD

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### Abstract

Nickel doped zinc selenide (Ni:ZnSe) thin films by simple chemical bath deposition (CBD) method in an aqueous alkaline medium the bath temperature at 70°C. The doped Ni<sup>2+</sup> ions different concentrations varied from 1 to 7 mol % and also these deposited thin films were annealed in an open atmosphere with various temperatures at 400, 500 and 600°C respectively. XRD, UV-vis spectrum and PL spectroscopies. XRD results revealed that the Ni:ZnSe thin films are showed mixture of cubic and hexagonal systems along with some NiO and ZnO phase. The PL studies confirmed that the Ni<sup>2+</sup> ions doped ZnSe thin films were showed a prominent emission peak centre at 405 nm and a weak emission peak centre at 411 nm.

**Keywords:** ZnSe:Ni thin films, annealing; chemical bath deposition and optical properties.

### 1. Introduction

In the present investigation involved the first time preparation of different mole concentrations of Ni<sup>2+</sup> ions doped ZnSe thin films were deposited on non-conducting glass substrate by using the CBD method in an aqueous alkaline medium. Among these deposition methods, CBD is an excellent method for the integration in large scale fabrication process. The

attractive features of this method are its low cost, inexpensive, no requirement of sophisticated instrument, low processing temperature and non-polluting properties. The high purity of thin films deposited by using CBD, it can be used in optoelectronic devices. The as-deposited thin films were annealed at different temperature, such as 300, 400 and 500°C. Finally, the effect of rare earth metal ion concentrations and annealing temperatures on optical, structural and morphological properties of Ni<sup>2+</sup> ions doped ZnSe thin films were studied by XRD and UV-visible spectrum. ZnSe possesses great potential application such as red, blue and green light emitting diodes and tunable mid-IR laser source for remote sensing applications [1-10]. The luminescence of rare earth ions Ce, Tb, Eu, Pr, Tm, Gd implanted in the different host materials has been intensively studied because of the important applications of these materials in lasers and field emission display devices. Thin film of ZnSe has been prepared by using several methods such as molecular beam epitaxy, chemical vapor transport, pulsed laser deposition, vacuum beam deposition, thermal evaporation, electrochemical deposition and chemical bath deposition (CBD) [11-25]. The deposition rate of the ZnSe thin film is based on reaction between Zn<sup>2+</sup> and Se<sup>2-</sup> ions in an aqueous alkaline medium [26]. To the best of our knowledge, very limited reports are only available for Gd<sup>3+</sup> ions doped ZnSe thin films. In general, rare earth (RE) elements are effective luminescence centers in the rare earth elements doped semiconductor because of their sharp and temperature stable luminescence, attributing to the incompletely filled 4f shells that are well screened and slightly affected by the crystalline field [27-30]. Recently, ZnSe thin films are used for the deposition of high efficiency solar cell based on environmentally friendly, without Cd metal thin films like CuInSe<sub>2</sub>, CuInS and CuInGeSe<sub>2</sub> [31-35].

## **2. Experimental section**

The precursors of Zinc sulphate (ZnSO<sub>4</sub>), sodium metal with purity of 99.99%, gadolinium (III) acetate hydrate (Mn(CH<sub>3</sub>CO<sub>2</sub>)<sub>2</sub> · xH<sub>2</sub>O ) and ammonia solution (25%) were purchased from Sigma-Aldrich, USA. Hydrazine hydrate (N<sub>2</sub>H<sub>4</sub>.H<sub>2</sub>O) was purchased from Sd-fine, India. All the chemicals are used without any purification. The chemical bath deposition method was performed in an aqueous alkaline medium, where using commercially available non-conducting glass substrate. The substrate cleaning is significant role in the thin film deposition process. The glass substrate first cleaned with mild soap solution and successively washed by using acetone and boil in chromic acid for 1h. Then, substrate washed with double distilled

water, followed by dry in an oven 60 °C for 30 minutes. The sodium selenosulfate ( $\text{Na}_2\text{SeSO}_3$ ) solution was prepared by mixing of 0.625 g of selenium metal powder and 0.1875 g of sodium sulphite in 100 mL of double distilled water and the above solution was refluxed for about 7 h at 90 °C. This freshly prepared sodium selenosulphite solution was used for thin film deposition process because of it is relatively unstable.

The 100 mL of reaction solution obtained by mixing of 10 mL of 0.5 M zinc sulphate, 13 mL of 80 % hydrazine hydrate solution, 10 mL of (1, 3, 5 and 7 %) gadolinium (III) acetate hydrate solution, 5 mL of 25 % ammonia solution, 10 mL of 2 M sodium selenosulphate solution and sufficient amount of double distilled water. Then, the above reaction solution is stirred at ambient conditions for a few minutes. Then, the well cleaned glass substrates were inclined at vertically in the reaction solution containing beaker. The beaker was sealed to avoid solution evaporation. The gadolinium doped thin films were achieved by chemical bath deposition at 80°C for 1 h. The deposited substrates were subsequently removed from the reaction solution followed by washing with double distilled water and dried at room temperature for a few minutes. Then, these thin films have been annealed at different temperature at 300°C, 400°C and 500°C for 30 minutes in an open atmosphere.

### 3. Results and discussion

#### XRD analysis

The grain and structural characterization of the chemically deposited ZnSe thin films were studied by using XRD analysis. ZnSe is known to exist there are two types of crystalline phase such as metastable sphalarite cubic (zinc blend type) or stable hexagonal (wurtzite type) or some time mixture of both phase present [36-38]. The XRD patterns of as-deposited and different annealed (300, 400 and 500 °C)  $\text{Mn}^{2+}$  ions doped ZnSe thin films are shown in Fig. 1. The chemically as-deposited and annealed ZnSe thin films shows mixture of cubic zinc blend structure with preferred orientation of (111) and hexagonal wurtzite structure with preferred orientation of (100). This observed results matching with a previous report [39]. However, the phase transformation could not occur completely with increasing concentration of  $\text{Mn}^{2+}$  ions and annealed temperature because of the films is the poor cubic phase at various temperatures. When the annealed temperature increased between 400 to 500 °C, the diffraction peaks exhibits high intensity with mixture phase. Annealed films exhibit more dominant hexagonal and Se metal

peaks along with ZnO phase is formed by substitution of oxygen for selenium at the high temperature region. It is clearly indicated the Se contribution less for the film deposition processes at the increase impurity level. The lattice constant “a” determine for all prepare films from high intense diffraction peak of (hkl) plane using the following Bragg’s equation for cubic structure.

$$\frac{1}{d_{hkl}^2} = \left[ \frac{h^2 + k^2 + l^2}{a^2} \right] \quad (1)$$

and hexagonal structure,

$$\frac{1}{d_{hkl}^2} = \frac{4}{3} \left[ \frac{h^2 + hk + l^2}{a^2} \right] + \frac{l^2}{c^2} \quad (2)$$

where hkl is the miller indices and d is the interplane spacing of the atomic planes. The calculated lattice constant values are agreement with the standard values. The calculated average lattice constant values increase with increasing doping concentration and annealed temperature respectively. The lattice distortion appears in the structure of thin films by Mn<sup>2+</sup> ions in the ZnSe matrix. The particle size was calculated from the full width half maximum (FWHM) of high intense diffraction peak by using following Scherer formula.

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (3)$$

where D is the crystalline size of thin film, K is a constant which has a value of 0.94 for ZnSe [40],  $\beta$  is the FWHM of the diffraction line in radians,  $\lambda$  is the X-ray wavelength [41] and  $\theta$  is the Bragg diffraction angle.

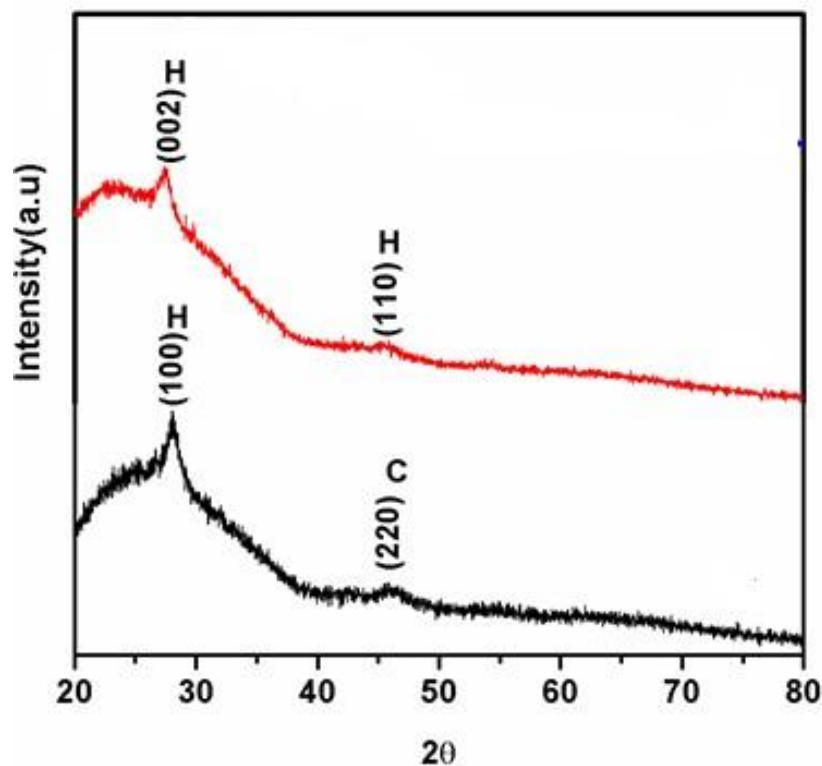


Fig. 1. XRD analysis of Ni:ZnSe thin films recorded at room temperature.

### 3.2 SEM analysis

The microstructure of Ni:ZnSe thin films were studied by using scanning electron microscopy is a very suitable method. The SEM images of the Ni:ZnSe thin films were deposited on glass substrate at different annealing temperatures are used, shown in Fig. 2 from this image as-deposited Ni:ZnSe thin film is homogenous, well covered to the glass substrate surface and without cracks or pinholes of film. The spherical particle of Ni:ZnSe thin films are consistently strewn over smooth back ground of film.

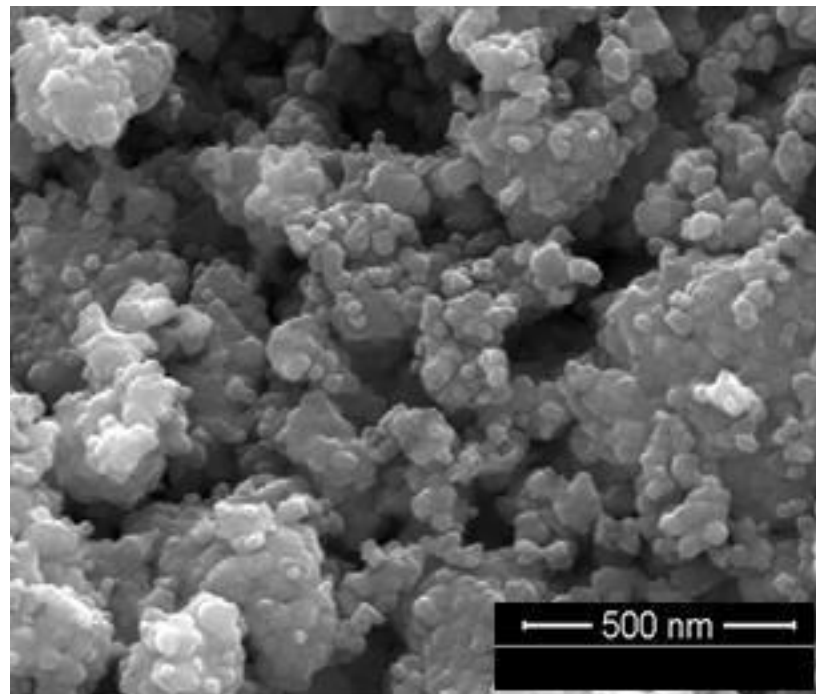


Fig. 2. SEM analysis of Ni:ZnSe thin films recorded at room temperature.

### 3.3. Optical studies

The optical properties of as-deposited and annealed  $\text{Mn}^{2+}$  ions doped ZnSe thin films were studied by using UV-visible absorption spectrometer at room temperature in the wavelength range from 300 to 1200 nm. Figure. 3 shows the absorption coefficient of  $(\alpha h\nu)$  Vs photon energy  $(h\nu)$  curve of  $\text{Gd}^{3+}$  ions doped ZnSe thin films. The fundamental absorption is corresponding to the transition from the valence band to the conduction band. It can be used to investigate the band gap energy of the chemically deposited semiconducting material. The band gap energy ( $E_g$ ) was calculated by using the following relationship,

$$\alpha h\nu = A(E_g - h\nu)^n \quad (7)$$

where  $\alpha$  is the absorption coefficient,  $h\nu$  is the photon energy,  $A$  is the constant,  $E_g$  is the energy band gap [42-45]. The average band gap energy of the films was decreased from 3.88 eV to 3.16 eV with increased doping concentrations and annealed temperatures. The absorption coefficient edge was not sharp obtained from 1 mol %  $\text{Mn}^{2+}$  ions doped thin film for different annealed temperature. When increase doping concentration about 3 to 5 mol % the absorption coefficient edge was sharp. While using doping concentration about 7 mol % the absorption coefficient edge was not sharp. From this observation may be attributed to the fact that films obtain from 1

and 3 mol %  $\text{Mn}^{2+}$  ions doped and corresponding different annealed films having cubic and hexagonal phase.

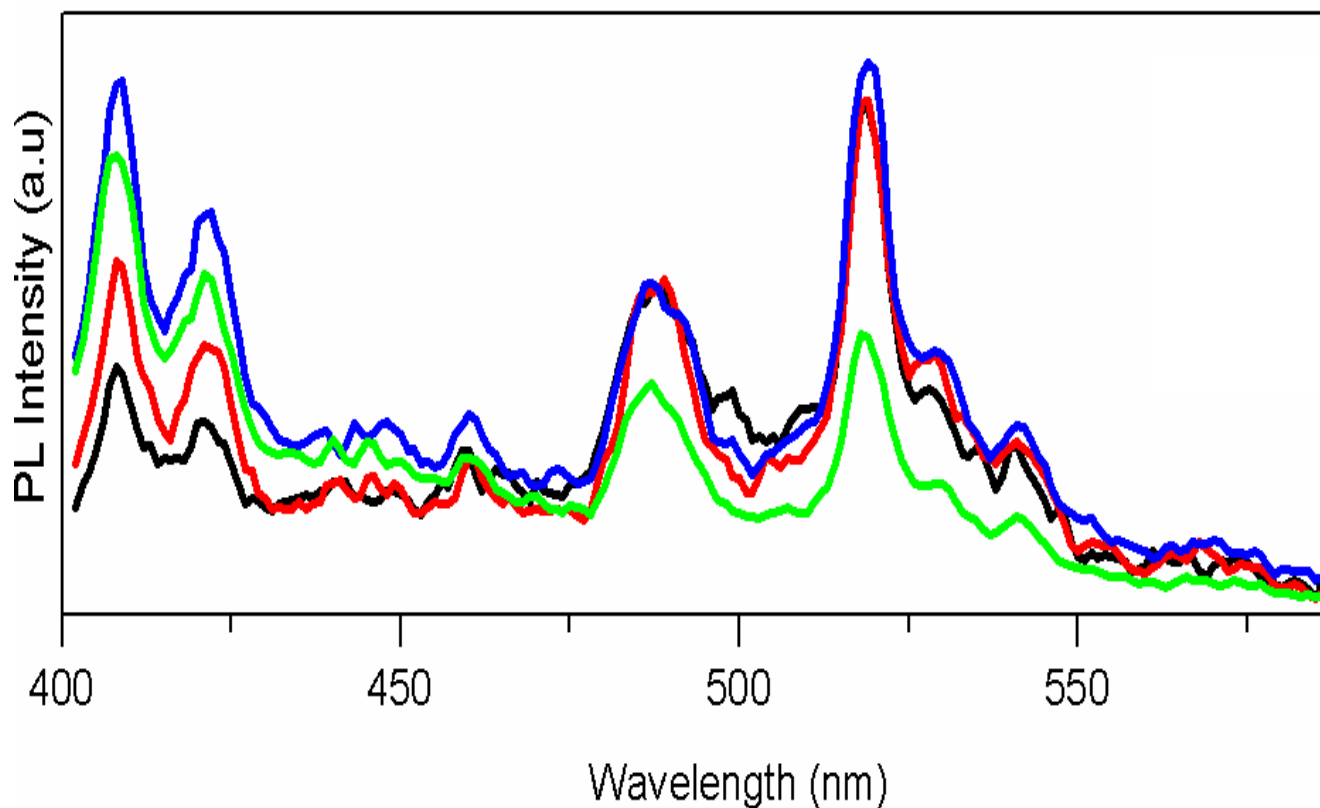
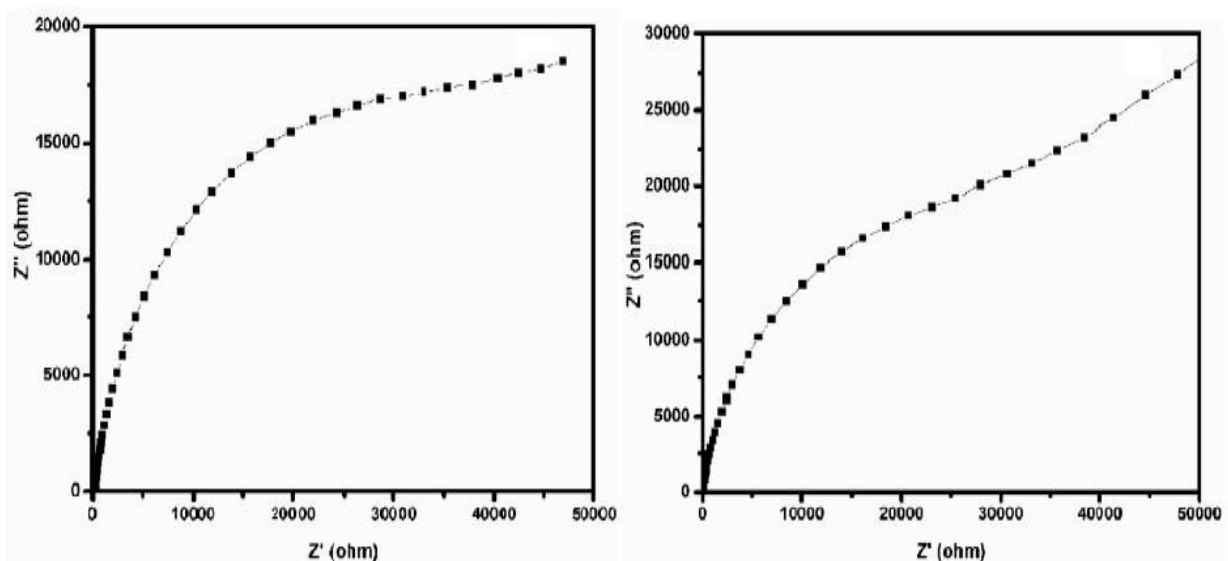


Fig. 3. Optical spectra of as- deposited Ni:ZnSe thin films recorded at room temperature.

### 3.4 Electrochemical impedance spectroscopy (EIS) studies

The electrochemical impedance spectroscopy (EIS) was performed for semiconducting Ni:ZnSe thin films to resolve the current response to application of an ac voltage as function of the frequency. The EIS measurements were carried out for the semiconducting ZnSe thin films to resolve the current response to application of an ac voltage as function of the frequency. . The Nyquist plot for Ni:ZnSe thin films are shown in Figure 4. The Nyquist plot for pure ZnSe and Ni:ZnSe thin films are shown in Figure 4. The measured  $R_{ct}$  is  $8 (\Omega\text{cm}^{-2})$  for Ni:ZnSe thin film. From the observed result, the  $R_{ct}$  of Ni:ZnSe thin film is vary less when compare to the others films, this results suggested that the addition of  $\text{Ni}^{2+}$  into ZnSe host lattice is favorable to the faster interfacial Ni-Ni charge transferee reaction.



**Figure. 4 Electrochemical impedance spectroscopy of (Nyquist plot) ZnSe thin film.**

#### 4. conclusions

The  $\text{Ni}^{2+}$  ions doped ZnSe thin films were deposited on non-conducting glass substrate by chemical bath deposition (CBD) method. The effect of  $\text{Ni}^{2+}$  ion concentrations and different annealed temperature of deposited thin film on optical, structural and morphological properties were studied. The XRD studies revealed that the crystalline nature of thin films has cubic and hexagonal structure. The X-ray diffraction results revealed that the pure ZnSe and  $\text{Ni}^{2+}$  ion doped ZnSe thin films are hexagonal (wurtzite) structure. along with small amount of Se metal peak was observed. The EIS study indicates that the  $\text{Ni}^{2+}$  ion doped ZnSe thin film shows less charge transfer efficiency with fine conductivity when compare to the other thin films.

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