



EFFECT OF SUBSTRATE TEMPERATURE ON OPTICAL AND ELECTRICAL PROPERTIES OF CdZnSeTe THIN FILMS PREPARED BY SPRAY PYROLYSIS TECHNIQUE

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ABSTRACT

Polycrystalline semiconducting chalcogenide thin films of CdZnSeTe are prepared on large area of glass substrate by spray pyrolysis technique. Thin films of CdZnSeTe deposited at different substrate temperatures 250°C, 275°C, 300°C and 325°C were investigated. Absorption coefficients for the films were evaluated from optical transmission spectra in the wavelength range 350nm-1100nm. Composition was confirmed from EDAX while SEM pictures shows the presence of nanotubes. Graphs plotted between $(\alpha h\nu)^2$ and $(h\nu)$ yields straight lines indicating direct transition with optical band gap energies in the range 2.40eV- 2.44 eV. Optical properties such as extinction coefficient, refractive index and dielectric constant have been evaluated in wavelength range 350nm-1100nm. Electrical study was carried out by four probe method. Arrhenius behavior shows semiconducting nature of the films. Values obtained by this method are found to be suitable to use the thin films for many scientific and technical applications such as solar cells, heat mirrors detectors etc.

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INTRODUCTION

Cadmium and zinc based II–VI wide-bandgap compound semiconductors have potential applications for various opto-electronic devices as well as x-ray and γ -ray detectors (Sebastian, 1989; Matsumura, 2003; Noda et al., 2000; Feng et al., 1994). In addition to applications such as opto-electronic devices and x-ray/ γ -ray detectors, Cd-based binary compounds grown on Si, such as CdTe/Si, have been used as composite substrates for HgCdTe material, an infrared (IR) detecting semiconductor system (Chen et al., 1993; Wang, 1995; deLyon, 1994; Wijewarnasuriya et al., 1998; Brill et al., 2001; Dhar et al., 1996). Similarly, ternary alloys, such as CdZnTe/Si and CdSeTe/Si, as well as quaternary compounds, such as CdZnSeTe/Si, can also be used as composite substrates specifically engineered for exact lattice matching with HgCdTe material designed to detect in the long wavelength IR region. Various methods have been used to prepare CdZnSeTe

thin films such as molecular beam epitaxy (Dinan, 1985), liquid phase epitaxy (Pellicary, 1988), electrode position (Basol et al., 1989), spray pyrolysis (Tembhurkar, 2016, 2017), close space vapour transport (Gonzales-Hernandes et al., 1991), laser ablation (Aydinly, 1991), thermal vapour evaporation (Kim et al., 2004), sputtering (Becerril, 2004) and metal–organic chemical vapour deposition (MOCVD) (Chu, 1992).

Experimental details

Preparation of samples

Aqueous solutions of cadmium chloride, zinc chloride, selenium dioxide and tellurium tetrachloride were mixed in the ratio 1:1.3:2:3.2 by volume for spraying. The molarity of each solution was 0.02M and chemicals used were of AR grade. The films deposited have selenium and tellurium deficiencies if the ratio of solution is 1:1:2:2. The excess selenium and tellurium are used to remove these deficiencies (Tembhurkar, 1992).

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The temperature of substrate which are in the form of double washed and then heated biological glass plates. The temperature of the substrate was maintained at 300°C and was measured by a pre-calibrated copper constantan thermocouple. The solution was sprayed at a pressure of 12 Kg/cm². The glass sprayer was mechanically moved to and fro during spraying to avoid the formation of droplets on the hot substrate and ensure instant evaporation. The spray rate was maintained at 3.5 ml/min. The thickness of the films was measured by weighing method and Michelson interferometer. The only difference between these two methods is of the order of 0.003µm. Transmission was taken on UV-1800 Shimadzu spectrophotometer in the wavelength range 350nm-1100nm. Electrical conductivity was measured by four probe method. Chemical composition analysis of the film was tested by EDAX analysis. The surface morphologies of as deposited CdZnSeTe thin films were carried out by scanning electron microscope.

RESULTS AND DISCUSSION

Optical studies

Optical band gap: The Optical transmission spectra of as deposited CdZnSeTe thin films was taken with the help of UV-1800 Shimadzu spectrophotometer in the wavelength range 350nm-1100nm. Fig.1. shows the resultant optical transmission spectra of as deposited CdZnSeTe thin films at substrate temperatures 250°C, 275°C, 300°C and 325°C. For the calculation of absorption coefficient (α) at various wavelengths the method used by Rubin et.al.(1986) was adopted.

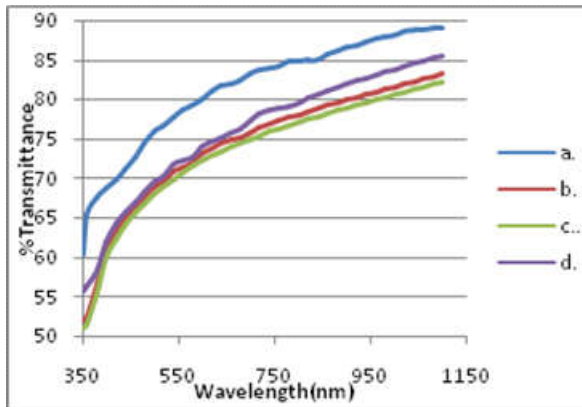


Fig.1. Transmission spectra of as deposited CdZnSeTe thin films at substrate temperatures a)250°C, b)275°C, c)300°C and d)325°C

For the sample of thickness *t*, α is given by the relation,

$$\alpha = (1/t) \ln(I_0 / I), \tag{1}$$

where *I*₀ and *I* are the intensities of incident and transmitted radiations respectively. Absorption coefficient is also related to % transmittance by the relation,

$$\alpha = (1/t) \ln(1/T) \tag{2}$$

Where *T* is the optical transmittance.

The optical study shows that the as deposited films are absorptive having high absorption coefficient. Optical band

gap can be determine from the graph between (α*hν*)² and (*hν*) and extrapolating the graph on (*hν*) axis as shown in fig.2.

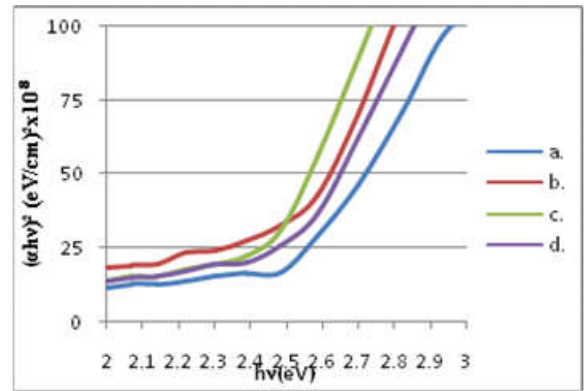


Fig.2. Variation of (α*hν*)² as a function of photon energy (*hν*) of as deposited CdZnSeTe thin films at substrate temperatures a) 250°C, b) 275°C, c) 300°C and d) 325°C

The values of optical band gap energies for as deposited Cd Zn Se Te thin films at different substrate temperatures are listed in table 1. The linearity of graphs in high energy region indicate direct type of transitions in the films. It is observed that optical band gaps of CdZnSeTe decreases from 2.44eV to 2.40 eV as substrate temperature increases from 250°C to 300°C which is due to increase in grain size with increase in substrate temperature and beyond 300°C, the optical band gap again increases with increase in substrate temperature as shown in fig.3 These results are found to be in good agreement with that obtained by Umeshkumar *et al.* and Murali *et al.* (2011-2009).

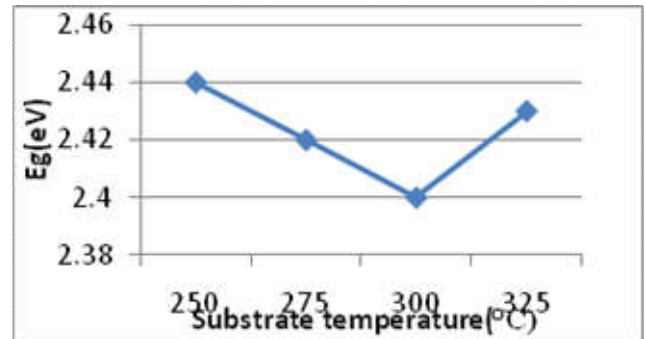


Fig.3 Variation of optical band gap with substrate temperature for as deposited CdZnSeTe thin films

Extinction coefficient and refractive index

The extinction coefficient *k* is related to absorption coefficient ‘α’ by the relation (Coutts et al., 2001,. Mahrov et al., 2004, Mahrov et al., 2004).

$$K = \alpha \lambda / 4 \pi \tag{3}$$

Refractive index *n* for the film is calculated using the relation,

$$n = (1 + \sqrt{R}) / (1 - \sqrt{R}) \tag{4}$$

where ‘α’ is the absorption coefficient, ‘λ’ the wavelength and ‘R’ the reflectance. The calculated values of extinction coefficient (*k*) and refractive index (*n*) at the wavelengths in the range 350 nm -1100nm are plotted as a function of wavelength as shown in figs.4 and 5 respectively. Figures shows that both *k* and *n* decreases with increasing wavelength but at higher wavelengths remains approximately constant.

Real and imaginary parts of dielectric constant

The real (ϵ_1) and imaginary(ϵ_2) parts of dielectric constant are given by the relations (Abd El-Wahabb, 2001).

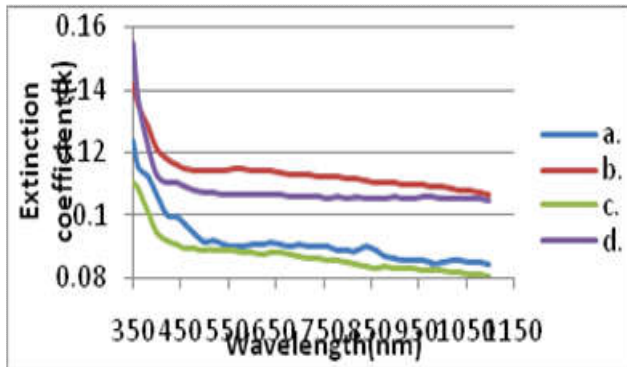


Fig.4. Variation of extinction coefficient with wavelength(nm)for as Deposited CdZnSeTe thin films at substrate temperatures a) 250°C, b)275°C, c)300°C and d)325°C

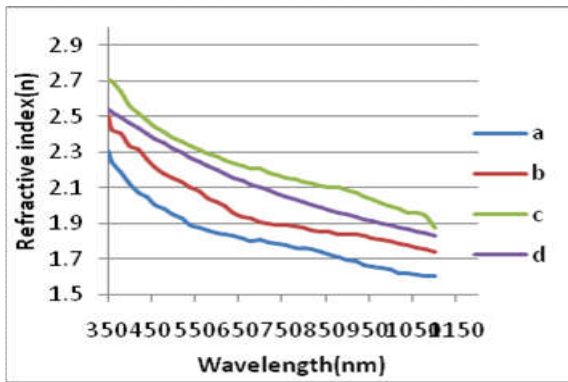


Fig. 5. Variation of refractive index as a function of wavelength for as deposited CdZnSeTe thin films at substrate temperatures a) 250°C b)275°C c)300°C d)325°C

$$\epsilon_1 = n^2 - k^2 \tag{5}$$

$$\epsilon_2 = 2nk \tag{6}$$

We also calculated real and imaginary parts (ϵ_1 and ϵ_2) of dielectric constants at several wavelengths ranging from 350 nm-1100nm as it is directly related to the density of states within the energy gaps of the films. Fig 6 and 7 shows the variation of real and imaginary parts of dielectric constant for CdZnSeTe thin films at substrate temperatures 250°C, 275°C,300°C and 325°C respectively.

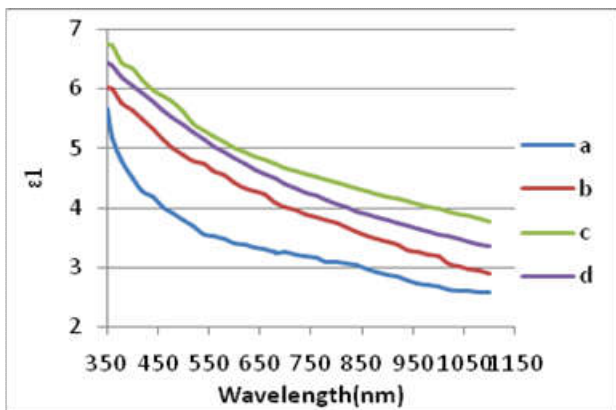


Fig. 6. Variation of real part of dielectric constant of CdZnSeTe films at substrate temperatures a)250°C b)275°C c) 300°C, d) 325°C

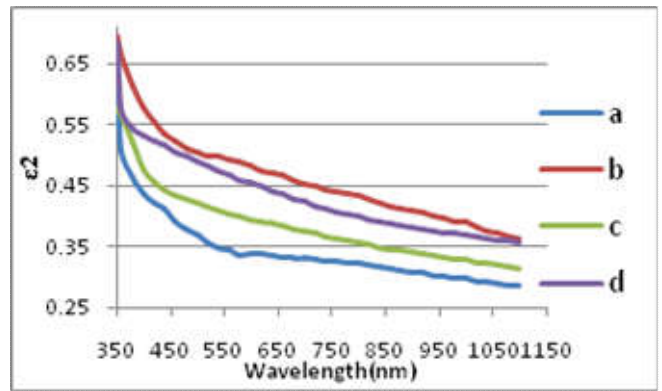


Fig. 7. Variation of imaginary part of dielectric constant of CdZnSeTe thin films at substrate temperatures a)250°C b)275°C, c) 300°C, d) 325°C

It is observed from the graph that both real and imaginary parts of dielectric constant decreases with increasing wavelength. The nature of curves for both ϵ_1 and ϵ_2 are found to be same, the only difference is that the values of real parts are higher than those of imaginary parts.

SEM studies

Surface morphology of thin films can be studied with the help of SEM analysis. Fig.9 shows the SEM picture of as deposited CdZnSeTe thin films at substrate temperature 300°C which shows the presence of nanotubes.

Electrical studies

The conductivity of CdZnSeTe thin films was measured by four probe method. The Arrhenius plots of conductivity for as deposited CdZnSeTe thin films deposited at substrate temperatures 250°C,275°C,300°C are shown in fig.8. Figures shows that conductivity increases with increase increasing substrate temperatures. The higher values of conductivity may be due to higher crystallinity (Garadkar *et al.*, 2010). Plots again shows that there are two distinct conductivity regions indicating more than one conduction mechanisms due to localized states responsible for this conduction process are the direct consequences of imperfections associated with thin films (More, 2003). The activation energies in two regions were calculated using the relation,

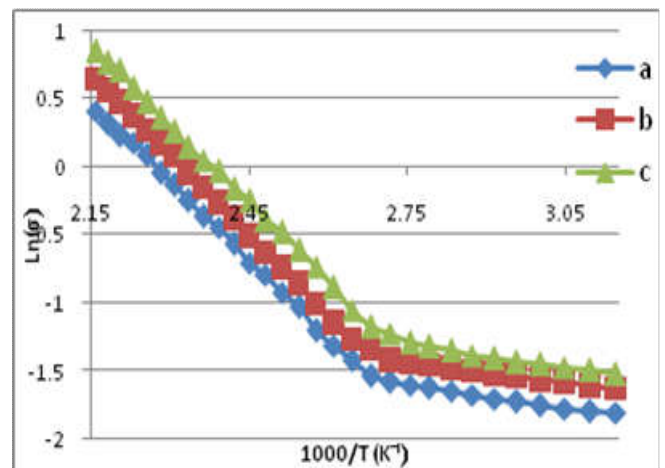


Fig. 8. Arrhenius plot of as deposited CdZnSeTe thin films at substrate temperatures a)250°C, b)275°C, c)300°C

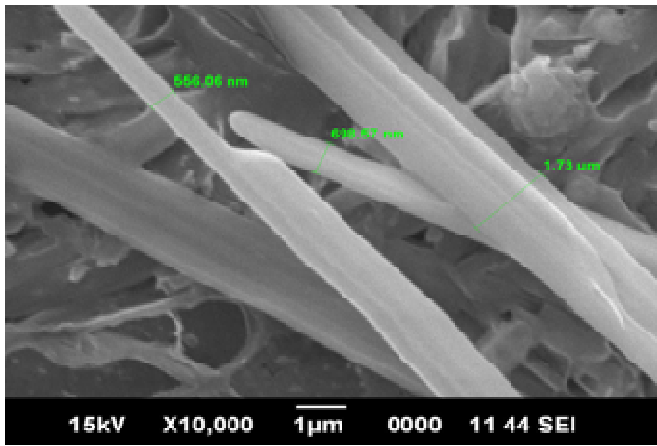


Fig.9 SEM image of as deposited CdZnSeTe thin film films at substrate temperatures 300°C.

$$\sigma = \sigma_0 \exp(-E_a/kT) \quad (7)$$

where 'k' is Boltzman constant, ' σ ' is conductivity of thin film at temperature 'T', ' σ_0 ' is a constant and ' E_a ' is the activation energy and 'T' absolute temperature. Activation energy represents the location of trap levels below the conduction band. It is found that activation energy in low temperature region is 0.084 eV and in high temperature region 0.65eV.

Conclusion

CdZnSeTe thin films at different substrate temperatures have been successfully synthesized by spray pyrolysis technique and effect of substrate temperature on optical and electrical properties have been studied. Optical studies shows that the films have high absorption coefficients. The optical band gap was found to be 2.40eV at substrate temperature 300°C, above and below which the optical band gap increases due to change in grain size with temperature.

Substrate temperature has an influence on all other optical parameters such as extinction coefficients, refractive indices and dielectric constants. Both real and imaginary parts of dielectric constant decreases with increasing wavelengths. The electrical studies shows that conductivity of CdZnSeTe thin films increases with substrate temperature as well as working temperature which shows that films are semiconducting in nature.

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