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Study of Optical, Electrical And Structural Properties of Spray Pyrolytically Deposited $CdZnSe_{2x}Te_{2(1-X)}$ Thin Films for X=0.25

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ABSTRACT

Spray pyrolysis is a simple, inexpensive and economical method to produce a thin film on large substrate area. Thin films of $CdZnSe_{2x}Te_{2(1-x)}$ for composition parameter x=0.25 at a substrate temperature of 300^0C are prepared by spray pyrolysis technique. From the optical transmission and reflection spectra, absorption coefficient (α) was calculated and was of the order of 10^4 cm^{-1}.Band gap energy were determined from absorbance measurement in visible range as 2.27 eV using Tauc theory. It shows that the main transition at the fundamental absorption edge is a direct allowed transition. The refractive index(n) and extinction coefficient(k) both decreases as wavelength increases which shows that the optical constants are most suitable for many scientific studies and technological applications such as heat mirrors, transparent electrodes and solar cells. The activation energy increases at higher temperature may be due to attributed to the increase of band gap. Hence the grain size of the films increases. This effect reduces the grain boundary effect. The XRD pattern shows number of peaks indicating that the films are poly crystalline in nature. The analysis of spectrum indicated that the films are having throughout cubic structure. The value of lattice parameter ' a' is 6.3702 A°

Keywords: $CdZnSe_{2x}Te_{2(1-x)}$ thin films, spray pyrolysis ,optical, electrical properties, activation energy, lattice parameter.

I. INTRODUCTION

In the recent years much more attention has been paid in semiconducting II-VI compounds because of their optoelectronic properties and their possible applications in switching and memory devices, photodiodes and solar cells. The ternary compounds including Cadmium zinc telluride and Cadmium zinc selenide have attracted much more attention in the field of solar cells due to their interesting properties of band gap. There are several binary and ternary semiconductors, such as GaAs, GaP, $CuInSe_2$, CdZnTe, CdZnSe, etc. which have band gaps in the required range(suitable for photovoltaic conversion). The evaluation of any material for application is complete and meaningful only when its structure and composition are precisely known. The reliability factor, which is the most important one for device application, can only be assured through a systematic and detailed study of the structural, electrical and optical properties. Cadmium zinc telluride and Cadmium zinc sellenide are among II-VI series of semiconducting compounds. The growth of quarternary compound is a opens up the possibility of their application for novel optoelectronic devices such as light emitting diodes, photo electrodes, blue green lasers etc.[1] the visible region of electromagnetic radiation [2]. The research of the optical and electrical properties of CdZnTe and CdZnSe system forms a basis of the active region of laser and LED.To the best of our knowledge, very less work has been reported on tellurium rich $CdZnSe_{2x}Te_{2(1-x)}$ polycrystalline material. Thus the present study is aimed at investing the optical and electrical properties of $CdZnSe_{0.5}Te_{1.5}$ in the form of thin films.

Several researchers studied properties in the II-VI semiconductor films using the variety of methods such as flash evaporation, chemical vapour deposition, r. f. sputtering, chemical bath deposition, electro deposition [3-5] and spray pyrolysis [6-7].

We have chosen spray pyrolysis due to simple, inexpensive and produce a thin film on large substrate area and it is suitable for scientific studies and for many technological and industrial applications. Very less work has been found on $CdZnSe_{2x}$ $Te_{2(1-X)}$ thin films. So our aim is to study optical, electrical and structural properties of $CdZnSe_{2x}Te_{2(1-x)}$ thin films with x=0.25 prepared by spray pyrolysis technique.

II. METHODS AND MATERIAL

The aqueous solutions of Cadmium chloride($CdCl_2$), Zinc chloride ($ZnCl_2$), Selenium dioxide (SeO_2) and Tellurium tetrachloride($TeCl_4$) each of 0.02 M were prepared in double distilled water. Chemicals used were of AR grade. The solutions are mixed in one in the proportion 1:1:1:3 by volume. The film shows a tellurium and selenium deficiency [8-9] if the ratio of proportion of solution was taken as 1:1:0.5:1.5 by volume. Sprayer was mechanically moved to and fro to avoid the formation of droplets on the substrate and insure the instant evaporation from the substrate. The distance between the sprayer nozzle and substrate was kept at 30 cm. The spraying was done in the atmosphere at the spray rate 3.5 ml/min. with a maintaining pressure of 12 Kg/cm^2. The temperature of substrate was maintained at 300^0C and was measured by pre-calibrated copper constantan thermocouple. The thicknesses of the films were measured by weighing method on unipan microbalance and was of the order of 0.1695 µm at substrate temperature 300°C. At this temperature deposition occurs at optimum rate resulting in terminal thickness. 0.1695 µm. It was found that the thin films had grayish color owing to the presence of more amount of tellurium. Optical transmittance and reflectance was taken on UV-1800-Shimandzu Spectrophotometer in the wavelength range 350 nm to 1100 nm. Electrical conductivity was measured by using four probe method [10]. Analytical method of indexing the X-ray diffraction pattern was used.

III. RESULTS AND DISCUSSION

3.1 OPTICAL STUDY

3.1.1 Transmission spectra

The optical transmittance (T) of the film was recorded at room temperature using UV-1800-Shimandzu spectrophotometer. Variation of transmittance with wavelength of the incident beam was recorded for the range of wavelength 350 nm to 1100 nm. And then the graph is plotted between % transmittance and wavelengths. **Fig.1**.Shows the variation of transmission versus wavelength for as deposited $CdZnSe_{0.5}Te_{1.5}$ thin films.

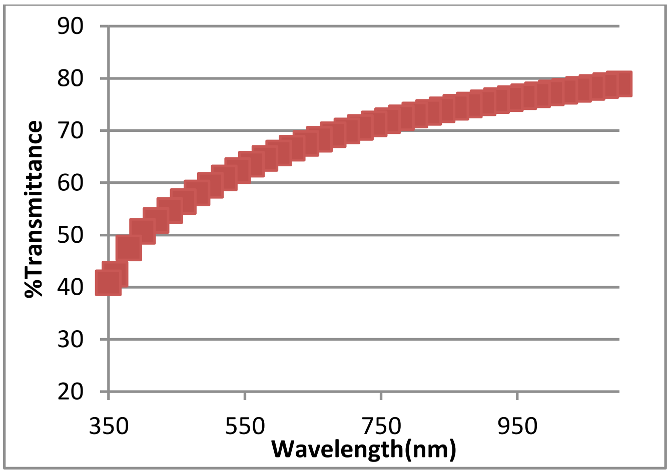

Fig.1Transmission spectra of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin films .

It was observed that the transmittance started decreasing after a particular wavelength, depending upon composition parameter (x) and remains constant for higher wavelengths.

It was also observed that onset of decrease of transmission gives the optical absorption edge. The optical coefficients were calculated for each wavelength given by relation,

$\alpha = (1/t)^{*} \ln(I_0/I)$ …………..(1)

Where, "t" thickness of the film," I_0" and " I" the intensities of incident and transmitted radiations respectively.

3.1.2 Reflectance spectra

Reflectance can be calculated using above values of % transmittance and graph is plotted between reflectance (R) and wavelength in nm. **Fig. 2** represents the reflectance spectra of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin films.

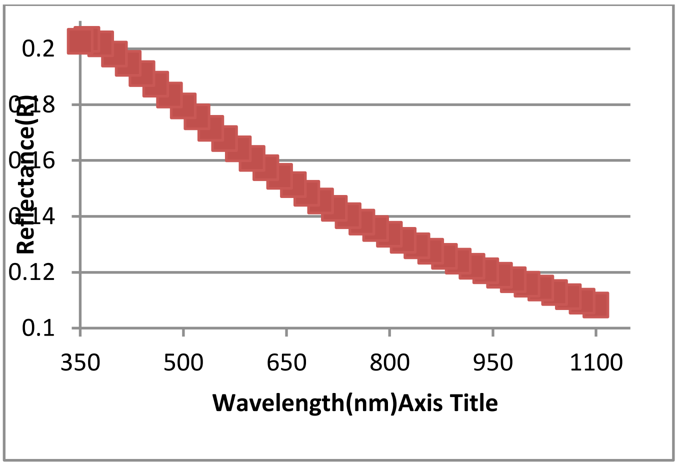

Fig. 2 Reflectance spectra of as deposited $CdZnSe_{0.5}Te_{1.5}$thin films.

The absorption coefficient "α" is related to the optical transmission "T" and reflectance "R" by the relation [11],

$T = (1-R)^2\exp(-\alpha t)/1-R^2\exp(-2\alpha t)$ ………………(2)

Equation (2) is valid in the vicinity of fundamental absorption edge when $R^2 \exp(-2\alpha t) \ll 1$ and it is used to calculate the absorption coefficient "α".

From **fig. 2** it was observed that as the wavelength increases there is sharp decrease in the reflectance. The onset of decrease of reflectance gives the approximate value of band gap [12].

Knowing the approximate region of band gap from reflectance curve, α is calculated by using equation (2), from the knowledge of T, R and t.

An analysis of the spectrum showed that the absorption at the fundamental absorption edge can be described by the Taue relation [13],

$\alpha = (A/h\nu) * (h\nu-Eg)^n$ ………………(3)

Where hv –photon energy, A-constant which is different for different transitions, n = 1/2 for direct band gap transition and n = 2 for indirect band gap transition. To calculate the exact value of band gap, we plotted the graph between $(\alpha h\nu)^2$ versus hv of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin film of as shown in **fig.3.**

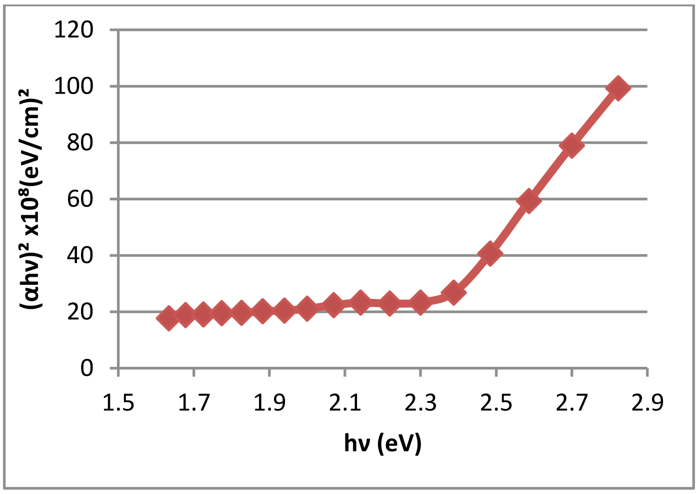

Fig.3 Graph between $(\alpha h\nu)^2$ versus hv of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin film .

The linearity of the graph showed the direct allowed transition, indicating the semiconducting nature of the films. The linear portion of the plot was extrapolated to meet on hv axis yield, the value of band gap energy was found to be 2.27 eV.

These results are found to be in good agreement with that obtained by Umeshkumar *et al.* and Murali *et al.*[14-15]

The optical transmission spectrum of the films under study shows that the transmission spectra mechanism is due to the direct allowed transition. [16]

The linear plot of $(\alpha h\nu)^2$ versus hv over wide range of photon energies shows thin film $CdZnSe_{0.5}Te_{1.5}$ has a direct allowed transition.

3.1.3 Optical constants (n and k)

The refractive index was obtained from the relation [17],

$$n = [(1+R)/(1-R)] + [(4R/(1-R)^2)-k^2]^{1/2} \text{ ……….(4)}$$

Where k –is the extinction coefficient which is related to the absorption coefficient α and wavelength λ as, [18-20]

$$K = \alpha\lambda/4\pi \text{ …………………………..(5)}$$

The value of refractive index (n) and extinction coefficient (k) were calculated using relations (4) and (5). The calculated values of refractive index (n) and extinction coefficient (k) at the wavelengths in the range 350 nm -1100nm are plotted as a function of wavelength in nm as shown in **figs.4 and 5** respectively

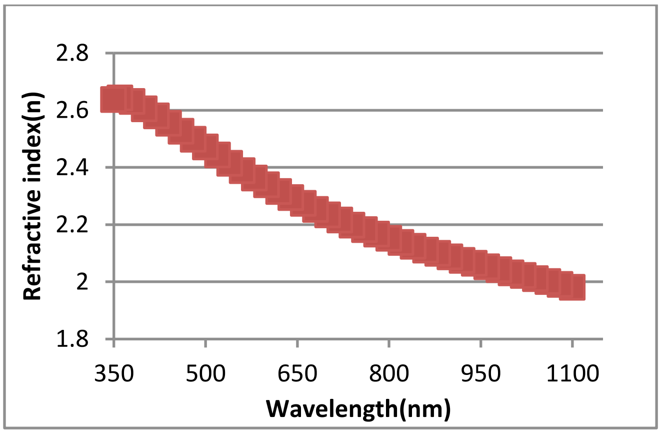

Fig. 4 Graph of refractive index (n) versus wavelength (λ) of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin film

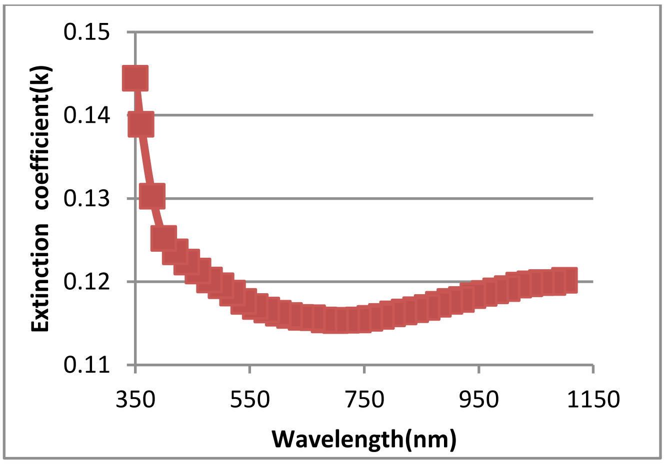

Fig. 5 Graph of extinction coefficient (k) versus wavelength (λ) of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin film

Figs. 4 and 5 shows that both k and n decreases with increasing wavelength but at higher wavelengths remains approximately constant. Our calculated values of n and k are in well agreement with the Saliha Ilican et.al [17] for spray pyrolitically deposited some ternary semiconducting $CdZn(S_{1-X}Se_x)$ thin films and other workers [21-22] also for ternary group of semiconducting materials. Quijada and Ross Henry[23] reported the refractive index for CdZnTe material to be 2.6.The This shows that optical constants are most suitable for many scientific studies and technological applications, such as sensors, heat mirrors, solar cells transparent electrodes and piezoelectric devices.

3.1.4 Real and imaginary parts of dielectric constant

The real (ε_1) and imaginary (ε_2) parts of dielectric constant are

given by the relations [24]

$$\varepsilon_1 = n^2 - k^2 \text{………………………….. (6)}$$

$$\varepsilon_2 = 2nk \text{ ………………………………(7)}$$

We also calculated real and imaginary parts (ε_1 and ε_2) of dielectric constants at several wavelengths ranging from 350nm-1100nm as it is directly related to the density of states within the energy gaps of the films. **Figs. 6 and 7** shows the variation of real and imaginary parts of dielectric constant for $CdZnSe_{0.5}Te_{1.5}$ thin films at substrate temperature 300°C.

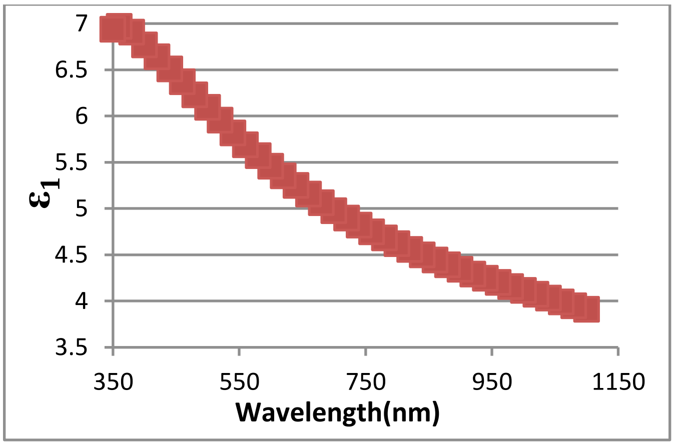

Fig 6 variation of real part of dielectric constant for CdZnSe0.5Te1.5 thin films at substrate temperatures 300°C.

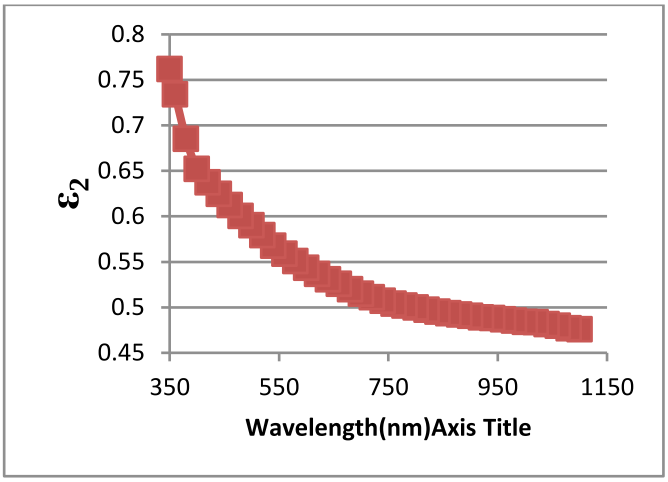

Fig 7 variation of imaginary part of dielectric constant for $CdZnSe_{0.5}Te_{1.5}$ thin film at substrate temperatures 300°C.

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.

3.2 ELECTRICAL PROPERTIES

The temperature dependence of conductivity was studied in the temperature range 300 K to 573 K. The resistivity were measured by four- probe method [25] given by the relation,

$\rho = [(V/I) * (2\pi S)] / [G_7(t/S)]$………(8) And

$G_7(t/S) = [2S/t] * \ln(2)$……………… (9)

Where S-the distance between the probes, t- the thickness of the film, I- the current generated from the constant current source between the inner probes, V- the voltage developed between the outer probes.

Fig.8 Shows the Arrhenius plot of conductivity versus inverse temperature of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin films The activation energy was calculated from Arrhenius plot using the relation [26-27],

$\sigma = \sigma_0 \exp\{ -E_a/kT\}$…….(10)

where 'σ0' is the pre-exponential conductivity,' E_a' is the activation energy and 'k' Boltzmann constant and 'T ' is the absolute temperature.

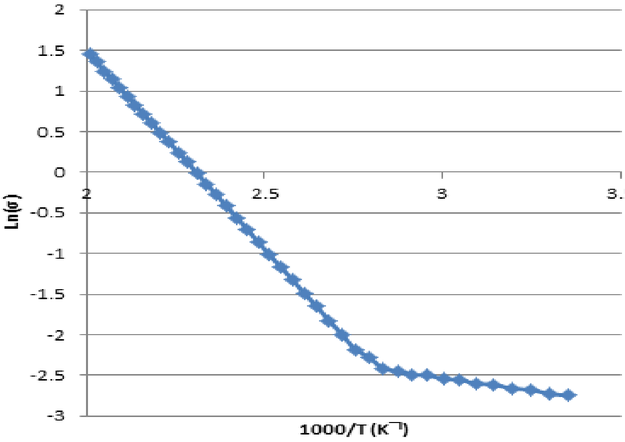

Fig. 8. Arrhenius plot of conductivity versus inverse temperature of as deposited $CdZnSe_{0.5}Te_{1.5}$ thin film

The plot shows two segments i.e. two conduction regions. Region I(lower part of graph) between the temperature 298 K to 348 K and region II(upper part of graph) between the temperatures 349K to 498 K. These two segments correspond to two values of activation energy. The temperature dependence is weak at lower temperature which confirms the low activation energy. The slope increases at high temperature which reveals the possibility of conduction due to the extended state. This indicates semiconducting nature of the films. The polycrystalline $CdZnSe_{0.5}Te_{1.5}$ thin films of do have the electrical and optical properties similar to those of single crystal. At the same time, the deposition conditions have been found to have significant effect on the electrical properties [28-29]. The activation energy increases at higher temperature may be due to attributed to the increase of band gap. Hence the grain size of the film increases.This effect reduces the grain boundary effect .Thus it is evident that $CdZnSe_{0.5}Te_{1.5}$ thin films the possibility of shallow trapping state due to the interstitials of Cd/Zn or telluride vacancies are expected to dominate the extrinsic conductivity near the room temperature. Whereas at higher temperature deep traps states influence are probable appears. Similar results are also reported by the other workers [30-31] for same group of ternary compounds.

3.3 XRD STUDY

Phase analysis of deposited thin films is carried out by X-ray diffraction method using C_uK_α radiation(λ=1.5406A.U.) with 2θ=20° to 80°. XRD study of the sample was taken at room temperature. X-ray diffraction (XRD) spectra of as deposited $CdZnSe_{2x}Te_{2(1-x)}$ thin films deposited on glass substrate at the substrate temperature 300°C for the composition parameter x= 0.25, is shown in **Fig.8.**The XRD pattern shows number of peaks indicating that the films are polycrystalline in nature. The analysis of spectrum indicated that the ternary films are having throughout cubic structure It is observed that two main peaks corresponds to (111) and (220) planes. The experimental d-values for $CdZnSe_{0.5}Te_{1.5}$ thin films are calculated using Bragg's relation,

$2d_{hkl}\sin\theta = n\lambda$,………………..(11)

By taking θ values from the peaks of XRD pattern. These d-values are compared with the results of other workers [32-38}

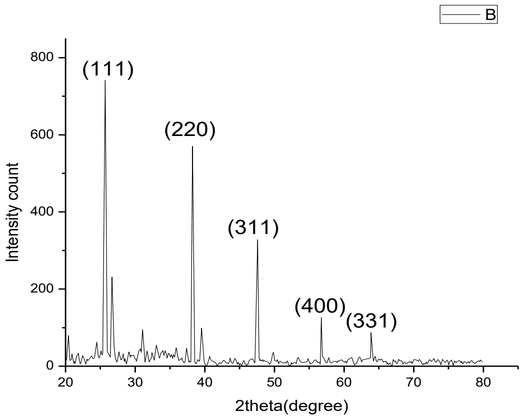

Fig.9 XRD of as deposited $CdZnSe_{2x}Te_{2(1-x)}$ thin film with x=0.25

The value of lattice parameter ' a' is found to be 6.3702 A° for for $CdZnSe_{2x}Te_{2(1-x)}$ thin films deposited at substrate temperature 300°C with composition parameter 'x=0.25'

4 CONCLUSION

Spray pyrolysis is a simple and inexpensive method to produce a thin film. Optical band gap of $CdZnSe_{0.5}Te_{1.5}$ thin film was found to be 2.27 eV which was calculated from $(\alpha h\nu)^2$ versus ($h\nu$) plot. The linearity of the plot shows the direct allowed transition. Arrhenius plot shows the two segments i.e. two conduction regions. Higher the conductivity value at low temperature is an evidence of the adsorption-distortion phenomenon. The XRD pattern shows number of peaks indicating that the films are polycrystalline in nature. The analysis of spectrum indicated that the films are having cubic structure(sphalerite). The value of lattice parameter ' a' is 6.3702 A° for $CdZnSe_{2x}Te_{2(1-x)}$ thin films deposited at substrate temperature 300°C with composition parameter 'x=0.25.'

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