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Study of effect of substrate temperature on optical and electrical properties of $CdZnTe_2$ thin films deposited by spray pyrolysis technique.

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ABSTRACT:
Spray pyrolysis is a simple, inexpensive and economical method to produce a thin film on large substrate area. Semiconducting thin films of $CdZnTe_2$ have been deposited onto preheated glass substrate by varying substrate temperature from 250°C at an interval of 25°C to 325°C. The optimized deposition temperature is around 300°C.From optical transmission and reflection spectra, absorption coefficient(α) was calculated at various wavelengths ranging from 350 to 1100 nm and was of the order of 1.58×10^4 cm^{-1} to 7.635×10^4 cm^{-1}.Band gap energy were determined from absorbance measurement in visible range using Tauc theory. It shows that the main transition at the fundamental absorption edge is a direct allowed transition. At the temperature of 300°C, the optical band gap is found to be 2.04 eV. At the temperatures less than or greater than 300°C, the optical band gap goes on increasing. The band gap energy value 2.04 eV is most suitable for many scientific studies and technological applications such as heat mirrors, transparent electrodes and solar cells. SEM study provide the information regarding the morphology of the material which confirms the formation of nano sized, nanotubes. Electrical conductivity was measured by four probe method. Arrhenius plots shows the semiconducting nature of films.

Keywords: Spray pyrolysis, CdZnTe2, , Absorption coefficient, Band gap energy, Electrical conductivity.

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I. INTRODUCTION

The polycrystalline thin-film solar cells based on II-VI and I-III-VI compounds have paid more attention due to their potential applications such as solar cells, radiation detectors ,photo detectors and light emitting diodes. Among these semiconductors, cadmium zinc telluride(CdZnTe) is found one of the most promising candidate for two-junction tandem device applications due to its optimum tunable direct band gap1.4–2.26eV,good absorber and high atomic numbers of Cd and Te [1–3]. One of the most important applications of CdZnTe thin film is to use it as the top cell absorber layer in a tandem solar cell when tuned to its higher band gap. The nano-crystalline CdZnTe films may also be used in homo junction solar cells as window layer [4,5].Various methods have been used to prepare $CdZnTe_2$ thin films such as molecular beam epitaxy [6], liquid phase epitaxy [7],electrodeposition [8], close space vapour transport [9], laser ablation [10], thermal vapour evaporation [11], sputtering [12-13]and metal–organic chemical vapour deposition (MOCVD)[14-15] and spray pyrolysis[16]. We are using Spray pyrolysis technique as it is most simple and economical method. The advantage of the technique is that just by varying the concentration of precursors and substrate temperature, it is possible to control stoichiometry of the deposits. Hence the thin films of $CdZnTe_2$ have been prepared by spray pyrolysis technique. Most of the work has been done on $TeCd_{1-x}Zn_x$ system. So far no work has been reported on tellurium rich $CdZnTe_2$ polycrystalline material. The present study deals with the effect of substrate temperature on optical and electrical properties of spray pyrolitically deposited CdZnTe2, thin films.

II. EXPERIMENTAL

Aqueous solutions of cadmium chloride, zinc chloride and tellurium tetrachloride 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 1:1:3.2 by volume the film shows a selenium deficiency [16-17], if the ratio of proportion of solution was taken as 1:1:2 by volume. In order to find optimized condition for deposition of thin films, the deposition was carried out by varying one of the parameters as substrate temperature and keeping others at fixed value.

The sprayer was mechanically moved to and fro to avoid 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 air and the spray rate was 3.5 ml/min. with a maintaining pressure of 12 Kg/cm^2. The temperature of substrate was maintained at 250°C, 275°C, 300°C, 325°C and was measured by pre-calibrated copper constantan thermocouple. The thicknesses of the films were measured by weighing method on unipan microbalance and Michelson interferometer. The thicknesses of the films found by both the methods were found to be approximately same. The difference was of the order of 0.003 μm. The thickness of all the films was found to be maximum at substrate temperature 300°C. Optical transmittance and reflectance was taken on UV-1800-Shimandzu Spectrophotometer in the wavelength range 350 nm to 1100nm.Electrical conductivity was measured by four probe method .

III. RESULTS AND DISCUSSION

3.1Optical studies-

a)Optical band gap-

The optical transmission spectra of CdZnTe2, thin films deposited at different substrate temperature was taken in the wavelength range 350 - 1100 nm. **Fig.1**Shows the transmission versus wavelength of as deposited CdZnTe2 thin films at different substrate temperatures.It was observed that onset of decrease of transmission gives the optical absorption edge. The optical coefficients were calculated for each wavelength given by relation,

$$\alpha = (1/t) \ln(1/T) \text{------------------- (1)}$$

Where, t- thickness of the films, T- transmittance of the film. An analysis of the spectrum showed that the absorption at the fundamental absorption edge can be described by the Taue relation [18]

,

$$\alpha = (A/hv)\ (hv\text{-}Eg)^n \text{----------------(2)}$$

Where hv –photon energy, A-constant which is different for different transitions, n = 1/2 for direct allowed transition. To calculate the exact value of band gap, a graph is plotted between $(\alpha hv)^2$ versus hv of as deposited $CdZnTe_2$ thin film at different substrate temperatures as shown in **fig.2** The linearity of each 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 to evaluate the value of band gap energies. From graph it was found that optical band gap energy decreases from 2.09eV to 2.04 eV as substrate temperature increases from 250°C to 300°C beyond which band gap energy again increases with increase in substrate temperature. The results are in good agreement with earlier reported work of Dammak et al., Isshiki and Sivaraman[19-21].

b)Extinction coefficient(k) and refractive index(n)-

Extinction coefficient(k) is related to absorption coefficient(α) by the relation,

$$K = \alpha \lambda / 4 \pi \text{---------------------- (3)}$$

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

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

where α is the absorption coefficient , λ the wavelength and R the reflectance.

Fig.(5) shows the variation of extinction coefficient(k) of as deposited $CdZnTe_2$ thin film deposited at substrate temperatures 250°C,275°C, 300°C and 325°C as a function of wavelength in the wavelength range 350nm-1100nm. From graph it is clear that k goes on decreasing with increasing wavelength and for higher wavelengths remains approximately constant at each substrate temperature. It is also observed from graph that extinction coefficient is minimum for substrate temperature 300°C, above and below this substrate temperature ,it goes on increasing.

Fig.(3 &4) shows the reflectance spectra and variation of refractive index of as deposited $CdZnTe_2$ thin film deposited at substrate temperatures 250°C,275°C, 300°C and 325°C as a function of wavelength in the wavelength range 350nm-1100nm.**Fig.4** shows that refractive index goes on decreasing with wavelength and at the substrate temperature 300°C,it attains maximum value than its corresponding values at other temperatures. These results are in good agreement with the values by Adachi at.el.[22]

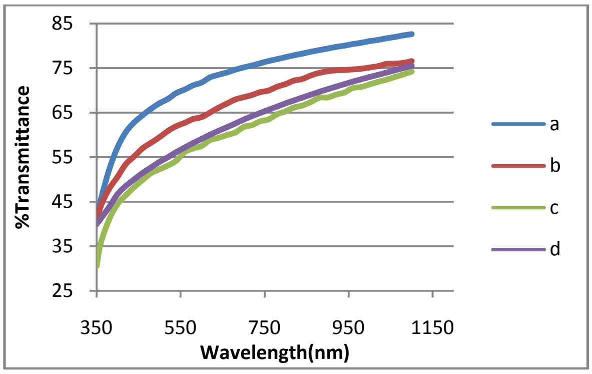

Fig.1. Transmission spectra of $CdZnTe_2$ thin films at Substrate temperatures a.250°C, b.275°C,c.300°C d.325°C.

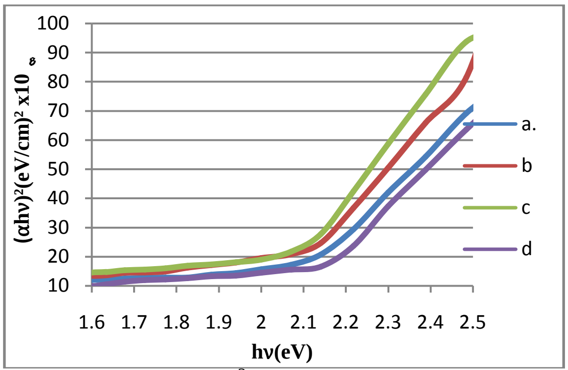

Fig.2 Variation of $(\alpha h\nu)^2$ as the function of photon energy for $CdZnTe_2$ thin films at Substrate temperatures a.250°C, b.275°C, c.300°C ,d.325°C

c)Real and imaginary parts of dielectric constant- Real and imaginary parts of dielectric constant(ε_1 and ε_2) are related to extinction coefficient(k) and refractive index(n) by the relation,

$\varepsilon_1 = n^2 - k^2$----------------------------(5)

$\varepsilon_2 = 2nk$----------------------------(6)

Figs.6&7 represents the variation of real(ε_1) and imaginary(ε_2) parts of dielectric constants as a function of wavelength for as deposited $CdZnTe_2$ thin films for substrate temperatures 250°C,275°C,300°C,and 325°C. From fig. it is very much clear that variation of both real and imaginary parts of dielectric constant follow the same nature of curves and as wavelength increases, both $\varepsilon_1$and ε_2 goes on decreasing. Also it is observed that values of real parts are higher than the values of imaginary parts of dielectric constants and values of both (ε_1)and (ε_2) are higher for films at substrate temperatures 300°C.

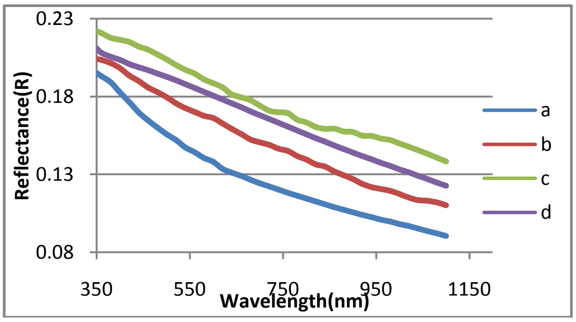

Fig.3.Reflectance spectra of $CdZnTe_2$ thin films at Substrate temperatures a.250°C,b.275°C,c.300°C,d.325°C

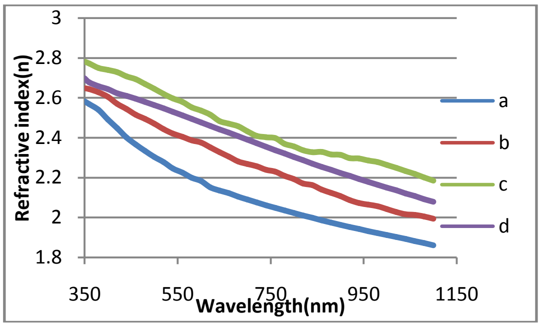

Fig.4 Variation of refractive index as a function of wavelength for $CdZnTe_2$ thin films at Substrate temperatures a.250°C,b.275°C,c.300°C,d.325°C

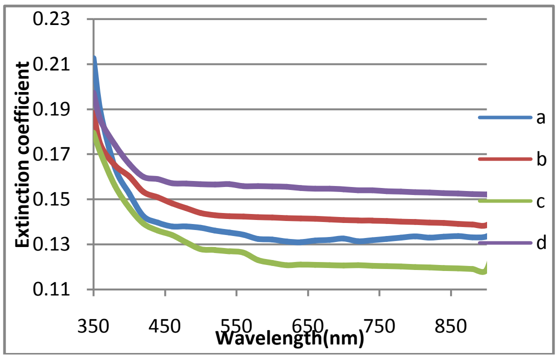

Fig.5 Variation of extinction coefficient as a function of wavelength for $CdZnTe_2$ thin film at substrate temperatures. a.250°C,b.275°C,c.300°C,d.325°C.

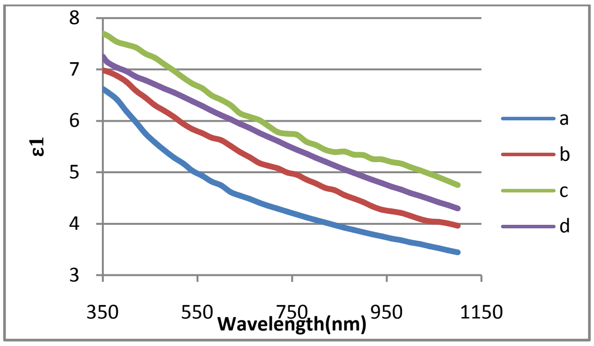

Fig.6 Variation of real part of dielectric constant as a Function of wavelength for $CdZnTe_2$ thin films at Substrate temperatures a.250°C,b.275°C,c.300°C,d.325°C

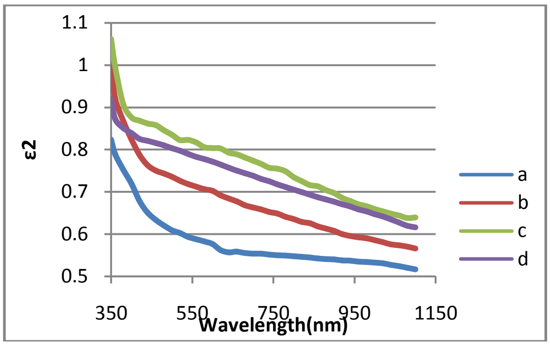

Fig.7 Variation of imaginary part of dielectric constant as a Function of wavelength for $CdZnTe_2$ thin films at substrate temperaturesa.250°C,b.275°C,c.300°C ,325°C

3.2Surface morphology studies-

SEM is one of the most useful techniquefor the invention of surface topography,microstructural features etc. because such properties of films influences their optical studies.**Fig.8** shows the surface morphology of as deposited $CdZnTe_2$ thin film deposited at substrate temperature 300°C.SEManalysis shows the presence of nanotubes.

Fig.8.SEM image of as deposited CdZnTe2 thin film at substrate temperature 300°C.

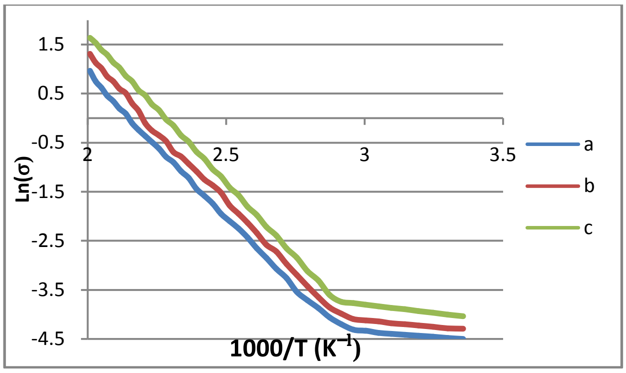

Fig.9 Arrhenius plots of as deposited $CdZnTe_2$ thin films at substrate temperatures a.250°C,b.275°C and c.300°C.

3.4 Electrical studies-

The conductivity of $CdZnTe_2$ thin films was measured by four probe method. Arrhenius plots of as deposited $CdZnTe_2$ thin films at substrate temperatures 250°C,275°C and 300°C are as shown in **fig.9.**It was observed that conductivity increases with increasing substrate temperature which may be due to higher crystallanity[23].Fig. shows two distinct conducting regions indicating more than one conduction mechanisms due to localized states responsible for this conduction process are the direct consequence of imperfections associated with the films[24]. The activation energies in two regions were calculated using the relation,

$\sigma=\sigma_0 \exp(-E_a/kT)$----------------------(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 0.11 eV and in high temperature region 1.04eV.

IV. CONCLUSION

$CdZnTe_2$ thin films were successfully deposited on glass substrate by spray pyrolysis technique and effect of substrate temperature on optical and electrical properties was studied successfully. Optical studies shows that films are highly absorptive in nature having high absorption coefficient. The optical band gap was found to be 2.04 eV at substrate temperature 300°C below and above which optical band gap increases.SEM analysis shows the presence of nanotubes. The electrical studies reveals that the conductivity of $CdZnTe_2$ thin films increases with substrate temperature as well as working temperature confirming semiconducting nature of the films.

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