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Structural and optical studies of hot wall vacuum evaporated CdTeSn thin films

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Abstract: Bulk compounds of CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ have been prepared by direct reaction of their high purity (99.9999%) elemental constituents employing rotating furnace. The hot wall system is optimized for the deposition of prepared alloys by using molecular flow studies with Monte Carlo simulation technique. Thin films have been deposited on well cleaned glass substrates using the prepared alloys by the optimized hot wall vacuum evaporation system. The compositions of the prepared bulk and thin films have been identified using energy dispersive X-ray analysis. The compositions are found to be same for both the bulk and thin films as the prepared alloys. The structural properties of the deposited films have been studied using X-ray diffraction technique. The results show that all the films are crystalline in nature and the peaks in the XRD graph of CdTe correspond to cubic zinc blende structure and that of $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ compounds to rock salt structure. The lattice parameters and grain sizes of all the films have been evaluated. The surface morphology of the thin films is studied using Scanning Electron Microscope (SEM). The SEM analysis shows that surface of the films are smooth and crystalline in nature. The optical transmittance spectra of thin films were recorded using spectrophotometer in the range of wavelength from 190 nm to 2500 nm. All the films exhibit direct optical band gap and their values are 1.45eV (CdTe), 0.9eV ($Cd_{0.25}Sn_{0.75}Te$) and 1.1eV ($Cd_{0.25}Te_{0.75}Sn$). Thicknesses of the thin films have been determined by multiple beam interferometric technique.

1. Introduction

CdTe is one of the suitable materials for the photovoltaic application because of its optimum band gap of 1.5 eV for solar spectrum and its direct band gap yielding high optical absorption coefficient. The energy conversion efficiency over 16% has been already realized in the research and development level [1-4]. Recently, binary and ternary compound semiconductors of II-IV-VI group elements have attracted extensive experimental interest because of their different energy related applications such as solar cells, infra red sensors and many more devices [5]. In the mixed system of $Cd_xSn_{1-x}Te$, the transition from a ten electron (x = 0) to a eight electron system (x = 1) changes in the crystal structure from rock salt (SnTe) to cubic zinc blende (CdTe) by the theoretical method and the variation of lattice parameter follows Vegard's rule. $Cd_xSn_{1-x}Te$ alloys have the unique feature of having the narrow range of lattice constants and direct band gaps, tunable from the visible to the far IR by adjusting the x composition of the alloy [6, 7]. As far as the authors' knowledge goes not much studies has been carried out on the CdSnTe compound. This is a novel material which is the major requirement for photovoltaic applications.

This paper presents experimental studies aimed at understanding the structural and optical properties of hot wall vacuum deposited CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films.

2. Experimental details

Bulk compounds of CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ have been prepared by direct reaction of their high purity elemental Cadmium, Tin and Tellurium (Sigma Aldrich 99.9999%). Stoichiometric amounts of the constituent elements according to the required composition are taken in three different quartz ampoules in which a pressure of 10^{-4} Torr is achieved and then sealed. The charged quartz ampoules are placed one by one in a furnace and heated at a rate of 100 K per hour in steps up to 1173 K and maintained at this temperature for about five hours. It is then allowed to cool slowly to room temperature. During the process of heating and cooling the ampoule has been rotated at 12 rpm continuously to ensure the homogeneity in the molten mixture. In the hot wall deposition system [8-10] two resistive windings serve to heat the source and wall of the tube independently. The whole apparatus is placed in vacuum as shown in the schematic diagram (Fig. 1). The main feature of the system is to heat linear quartz tube of radius 'r' and length 'L' which serves to enclose and direct the vapor from the source to the substrate, so that the molecular flow inside the tube is close to the thermodynamic equilibrium. The computer simulation for the deposition of CdSnTe thin films in the hot wall epitaxial system is carried out using a computer with the help of the MATLAB software [11]. The total number of intermolecular collisions, wall collisions and total number of molecules passing out through the exit plane directly from the entrance plane, the number density distribution along each volume section, energy of the molecule and flux density distributions at the exit plane are calculated with different source temperatures (673 – 873 K), wall temperatures (623 – 823 K) and design parameters (L/r*)* from 2 to 24 [10, 12]. The source and wall temperatures are optimized as 780 K and 760 K (for CdTe), 720 K and 700 K (for $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$) respectively and L/r as 16. The alloys are placed at the bottom of the quartz tube and well cleaned substrates are placed with the help of substrate holder cum heater which is mounted just above the open end of the tube. The source and wall temperatures are maintained at 780 K and 760 K (CdTe), 720 K and 700 K ($Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$) respectively.

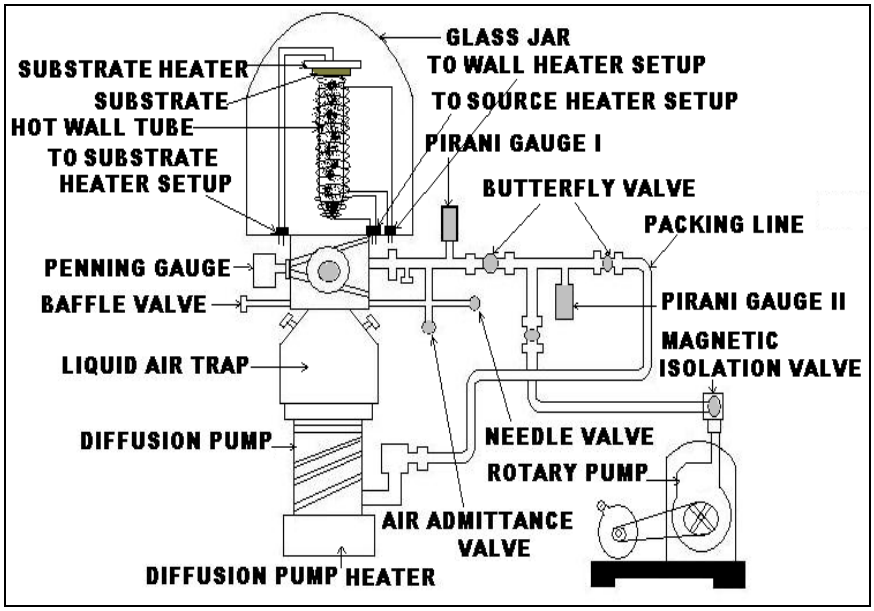

Figure 1. Schematic diagram of hot wall vacuum evaporation system

The deposition is carried out at a pressure of 4×10^{-5} Torr. CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ films are deposited with the substrate temperatures around 350 K, 400 K and 450 K. Composition of the bulk and films are determined by EDAX measurements (JOEL –Japan). The thicknesses of all the films are measured by using multiple beam interferometer technique. The structural analysis of the thin films is carried out using a computer controlled X-ray diffractometer system (model JDX 8030, Japan) fitted with Ni filter using Cu Kα radiation. The scanning is carried out using the θ –2θ scan coupling mode, the ratings being 40 kV, 20 mA. The optical studies are made on the films deposited on glass substrates in the wavelength range from 190 nm to 2500 nm at room temperature by using Spectrophotometer (JASCO-370V).

3. Results and discussion

All the films deposited on well cleaned glass substrate are smooth, uniform, adherent and dark brown in color. The darkness increases with increase in thickness of the thin films. Figure 2 shows XRD graphs and SEM images of CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films (deposited at substrate temperature 350 K) of thicknesses 220 Å, 334 Å and 375 Å respectively. The predominant peaks in XRD graph of CdTe thin film around 23°, 39°, 46°, 56°, 62°, 71° and 76° could be probably associated with (111), (220), (311), (400), (331), (422) and (511) reflections of the cubic zinc blende structure. Similar observations are found in XRD graphs of $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films around 28°, 40°, 50°, 60°, 67° and 75°could be probably associated with (100), (110), (111), (200), (210) and (211) reflections of the rock salt structure [5, 12-21]. The peak intensities of all the films increase with increase in substrate temperature, it is due to increase in the crystalline nature of the film. Full width at half maximum decreases with increase in substrate temperature attributes the increase in grain size and decrease in strain of the crystals in the film. The crystalline sizes (D) are calculated using the Scherrer's formula from the full width at half maximum (β) using the relation

$$D = \frac{0.94\,\lambda}{\beta \cos\theta} \qquad (1)$$

The strain (ε) is calculated from the slope of β cosθ versus sinθ plot using the relation

$$\beta = \frac{\lambda}{D\cos\theta} - \varepsilon\tan\theta \qquad (2)$$

The dislocation density (δ) is evaluated from the relation

$$\delta = \frac{1}{D^2} \qquad (3)$$

The width of the peaks decreases as the substrate temperature increases. This could be due to reduced strain within the film or an increase in grain size indicating a better crystalline perfection. The lattice parameter (a_0) of the crystal is determined by using the relation

$$\frac{\sin^2\theta}{(h^2+k^2+l^2)} = \frac{\lambda^2}{4a_0^2} \qquad (4)$$

where (hkl) is the miller indices of the peaks. The true lattice parameter is obtained for each XRD graph by plotting Nelson-Reliey factor versus lattice parameter [22, 23]. The lattice parameters, particle sizes, strain and dislocation densities of thin films deposited are given in table 1 [24, 25]. It is found that the size of the particle increases with the substrate temperature in all the films of all compositions. These results are in agreement with the SEM results. EDAX measurements of the bulk and films maintain the proposed composition and are percentage of each element is as shown in the table 2. The particle sizes in SEM micrograph of all the alloys are measured and the mean values of the particle sizes are found to be 410 Å (CdTe), 240 Å ($Cd_{0.25}Sn_{0.75}Te$) and 390 Å ($Cd_{0.25}Te_{0.75}Sn$) which are very close to the XRD results.

Table 1: XRD results of three representative CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films

Compound	Lattice parameter (Å)	Particle size (D) (Å)	Strain (ε) ($x10^{-4}$ lines^{-2} m^4)	Dislocation density (δ) ($x10^{14}$ lines^{-2} m^2)
CdTe	6.449	400	26.83	6.250
$Cd_{0.25}Sn_{0.75}Te$	7.886	220	18.24	20.66
$Cd_{0.25}Te_{0.75}Sn$	8.055	350	11.55	8.163

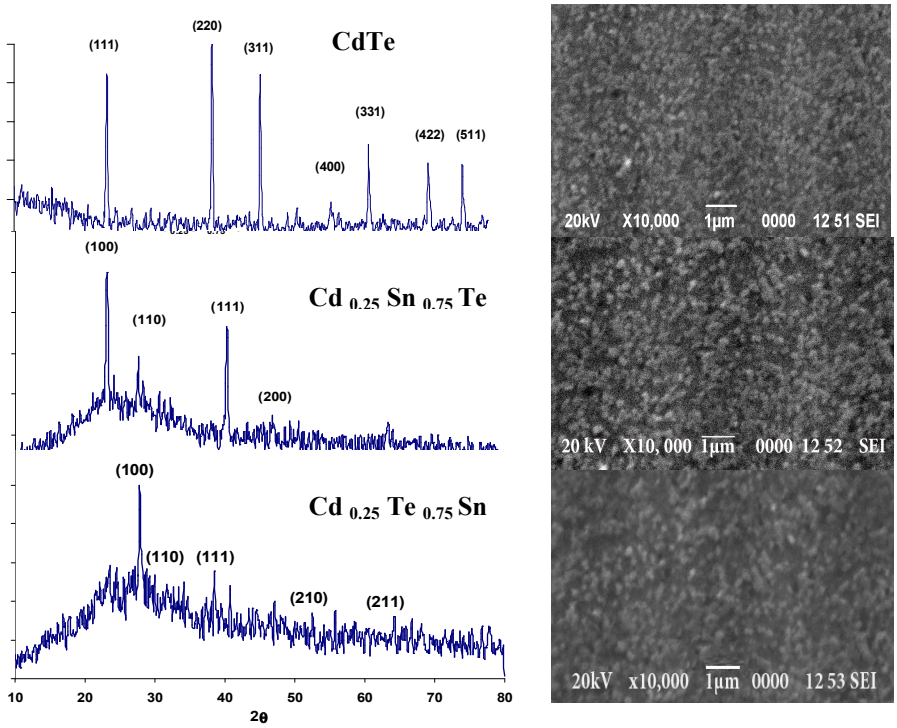

Figure 2. X-ray diffraction pattern and SEM images of CdTe, Cd $_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}$ Sn thin films

The optical transmittance spectra of three representative thin films of all alloys are as shown in figure 3. The insets in each graph show the corresponding plot of $(\alpha h\gamma)^2$ versus hγ and the extrapolation of the linear portion to the abscissa are the band energy gap of the film. The absorption coefficient α is estimated from the optical transmittance spectra using the relation

$$\alpha = \frac{2.303\log(1/T)}{t} \qquad (5)$$

where T is the transmittance (in %) and t is the thickness of the film. All such graph satisfies the condition for a direct transition in the excitation process i.e. $\alpha \propto (E_v - E_i)^{\frac{1}{2}}$ for allowed direct transition, where E_v is the top of the valence band and E_i is energy of the initial state from which the transition is made. All the films exhibit direct band gap structure and their band gap values are found to be 1.45 eV (CdTe), 0.9ev ($Cd_{0.25}Sn_{0.75}Te$) and 1.1ev ($Cd_{0.25}Te_{0.75}Sn$) [5]. Transmittance of all the films deposited at different substrate temperatures is analyzed and it is found that there is decrease in the transmission percentage and the minimum value of transmittance shifted to the higher wavelength side and band edge decreased as the substrate temperature increased [26].

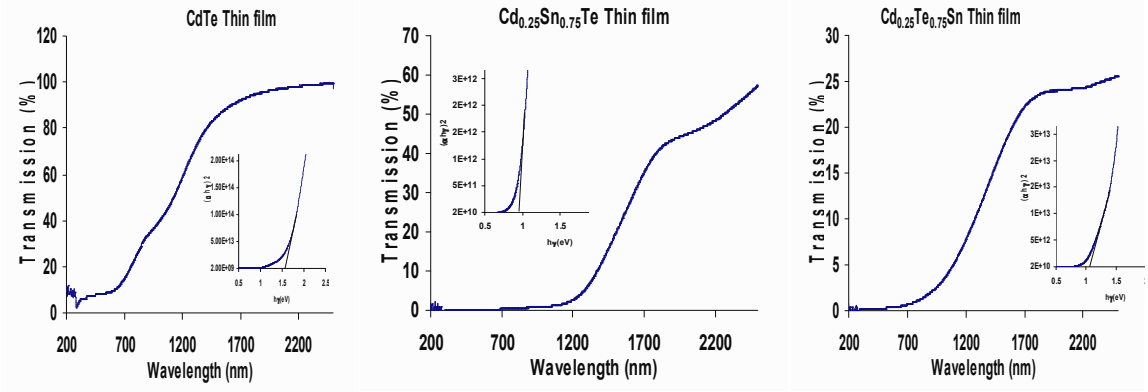

Figure 3. Transmittance spectra and dependence of $(\alpha h\gamma)^2$ on photon energy for CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}$ Sn thin films

Table 2: Results of EDAX studies of CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films

Compound	Cd (%)	Sn (%)	Te (%)
CdTe	50.01	--	49.99
$Cd_{0.25}Sn_{0.75}Te$	10.86	38.12	51.02
$Cd_{0.25}Te_{0.75}Sn$	10.52	50.15	39.33

4. Conclusion

Bulk compounds of CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ have been prepared by direct reaction using rotating furnace and thin films have been deposited on well-cleaned glass substrates using optimized hot wall vacuum evaporation method. The compositions are found to be the same for both bulk and thin films. The macrostructural parameters such as crystalline size, strain, dislocation density and true lattice parameter are calculated from the appropriate formula. The structures of the deposited films are cubic zinc blende structure for CdTe and that of $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ are rock salt structure. With the increase in substrate temperature, a marked increase in crystallinity of the film is observed. The scanning electron microscope images confirm crystalline nature of the film with many particles and the size of particles are increased with the increase in substrate temperature. The optical parameters are calculated from the transmission spectra of the CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films. The optical band gap decreases with increase in the substrate temperature and thickness of the films. The optical absorption in the films obey the direct allowed transition. It is observed that the absorption coefficient and the optical band gap of the thin films depend on the substrate temperature that subsequently modifies the density of states. In the process of thermal treatement some of the unsaturated bonds are annealed out to some extent, reducing the band energy gap and consequently decreasing the optical gap. The characterization of CdTe, $Cd_{0.25}Sn_{0.75}Te$ and $Cd_{0.25}Te_{0.75}Sn$ thin films confirm the formation of crystalline films and these are suitable for photovoltaic applications and infra red detectors.

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