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Exploration of the spray deposited Cadmium Telluride thin films for optoelectronic devices

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| ARTICLE INFO | A B S T R A C T | | | | |
|--|--|--|--|--|--|
| Keywords: Cadmium telluride (CdTe) Spray pyrolysis Structural Morphological Optical Electrical | Cadmium Telluride is one of the important partners used in heterojunction solar cells and various other opto- electronic devices. Spray pyrolysis method was employed to deposit CdTe thin films, on glass substrates, at 300 °C. The as-deposited films were black in color and having thickness of 497.28 nm. XRD analysis discovered that the as-prepared films were polycrystalline with a cubic crystal structure having preferred reflection (111), with a grain size of 12 nm. The abundance of Cd and Te elements in as-deposited films was ensured using the EDX technique. The SEM micrograph processing showed a compact morphology and an average grain size of 30 nm. The spectral peaks in Raman spectra were found at 139.30 cm^{-1} (1TO) and 165.33 cm^{-1} (1LO). The observed direct band gap was 1.52 eV . The calculated electrical resistivity at room temperature was noticed to be $1.2 \times 10^6 \Omega$ -cm. TEP study engrained the p-type conductivity in as-deposited films. | | | | |

1. Introduction

Cadmium Telluride is a semiconductor compound that has widespread applications in electronic and optoelectronic devices because of its special chemical, optical and transport characteristics [1]. It is one of the most studied and favorable compounds used in solar cells due to its special properties like a high average atomic number, the lowermost melting temperature, the large lattice parameter, the smallest negative formation enthalpy, and the high iconicity compared with other semiconductor compounds [2]. The p-type CdTe semiconductor material is widely utilized along with an n-type material in heterojunction solar cells [3]. It is also used in γ -ray detectors [4], photodiode and photo capacitor [5] infrared windows [6], light-emitting diodes and field effect transistors [7]. The direct optical band gap nearby 1.5 eV with maximal absorption coefficient (99%) make CdTe thin film useful for efficient solar energy conversion [8]. Depending on the preparation method and parameters used CdTe can be obtained with n-type or p-type conductivity which is very helpful in the construction of field effect transistors, diodes, and homojunction or heterojunction solar cells [9]. The electrical, optical and structural traits of solids are the important aspects of basic research as well as for their industrial applications. The thrust for the inexpensive mass production of polycrystalline solar cells for high conversion efficiency has increased the interest in structural, electrical and optical properties of semiconductor material like CdTe [10]. In recent years CdTe had been largely used in solar cell fabrication technology, so it is very crucial to investigate the microstructural, vibrational, electrical, optical, morphological and elemental assets of CdTe thin film at optimized preparative parameters.

The deposition methods and the corresponding preparative parameters used for the preparation of CdTe thin film are actually significant in the attainment of large solar cell efficiency [11]. The execution of CdTe solar cells strongly depends on the structural and morphological imperfections within the thin films so it is very imperative to deposit CdTe film with well-behaved composition and stoichiometry [10]. Various methods have been enrolled for the deposition of CdTe films like pulsed laser [12], electrodeposition [13], chemical bath deposition [14], molecular beam epitaxy [15], vacuum evaporation [3], physical vapor deposition [16], thermal evaporation [17], sputtering [18], spray pyrolysis [19], closed space sublimation [20], etc. Among all these thin film deposition techniques chemical spray pyrolysis is one of the modest, low cost and an easy method which includes spraying of a solution containing ions on the hot substrates with a jet of carrier gas [21]. This technique is very useful in the controlled deposition of a high-quality thin film on a large substrate area [21]. In spray pyrolysis, the film

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quality can be controlled by using various preparative parameters like substrate temperature, liquid flow rate, carrier gas pressure, solution quantity, the substrate to nozzle distance and molarity of the used precursors [21].

In this present article, we have employed a chemical spray pyrolysis technique aimed at the preparation of Cadmium Telluride thin films onto glass substrates. The electrical, structural, elemental, morphological, vibrational, and optical behavior of the as-deposited CdTe thin films has been studied and reported.

2. Experimental

Chemical spray pyrolysis method was used to develop Cadmium Telluride thin films on commercially gained amorphous glass substrates. Spray pyrolysis includes the formation of spray and atomization by adjusting the proper liquid flow rate and carrier gas pressure which is further followed by a chemical reaction on the heated glass substrate. The glass substrates were procured from Blue Star Company. The cleaning processes of substrate involves, cleaning with labolene cleaner followed by 30 min boiling of substrates at 300 $^{\circ}$ C in a bath containing 1 M chromic acid solution, these boiled substrates along with bath were allowed to cool for 12 h and then rinsed with double-distilled water and used.

For the deposition of CdTe thin films, a 36 ml solution was used which was the mixture of equal amount (1:1) of Cadmium Chloride monohydrate A.R. (CdCl₂.H₂O) and Tellurium Dioxide (TeO₂) solutions. Both these solutions were 0.025 M and the volume of solvent for each was comprising of DDW and ammonia in the ratio 4:1. Before mixing these two solutions, 0.3 ml Hydrazine Hydrate was dropped in Tellurium Dioxide solution and 0.5 ml Triethanolamine was dropped in the solution of cadmium chloride. The resultant precursor solution pH was maintained at ~11.5 using an ammonia solution. The substrate temperature was optimized and it was found to be 300 °C so for deposition temperature was kept 300 °C. Air was used as a carrier gas and was provided at pressure of 1 kg/cm³. A peristaltic pump is used to control the solution flow rate equal to 5 ml/min, straight-line distance between nozzle tip and substrate was 27 cm. These preparative parameters were optimized to obtain good quality polycrystalline thin film of precise thickness. Under these optimized preparative conditions deposition was done and substrates were let to cool to room temperature.

The weight difference method is used for the estimation of the thickness of the prepared films. Thermo Fisher Scientific DXR Spectrometer was utilized to record FT-Raman spectra. For the structural investigation, Shimadzu LabX XRD-6000 diffractometer is utilized. SEM and EDAX studies were carried out with JSM6360 LA, Japan fortified with EDXS hardware. The optical studies were done using JASCO V-570 UV–Vis–NIR spectrophotometer. The electrical conductivity measurements were carried out using two probe method in which for better ohmic contacts silver paste was applied.

3. Results and discussion

The pyrolytic decomposition of the resultant solution at 300 $^{\circ}$ C results in the growth of polycrystalline CdTe on hot glass substrates. After the cooling of substrates to room temperature, the films were observed to be black in color with uniform morphology. The films were strongly adhesive to glass substrates.

3.1. Thickness

The weight difference method is used for the estimation of the thickness of the as-prepared films. The relation used for thickness (t) calculation was $t = \frac{m}{A^*\rho}$, where 'm' is deposited thin film mass, 'A' is area acquired by a deposited thin film, and ρ is the density of deposited material in its bulk form (for CdTe ρ = 5.85 g/cm³). The as-prepared thin



Fig. 1. X-ray diffractogram for CdTe thin film prepared at 300 °C.

film was built with a thickness of about 497.28 nm. CdTe film with this much thickness is important for high-efficiency CdS/CdTe solar cells [22]. The calculated thickness of CdTe thin films was nearly the same as that of obtained earlier at nearly the same preparative parameters [13].

3.2. Structural analysis

The microstructural investigation of the as-prepared film was done using XRD. Fig. 1 demonstrate the X-ray diffractogram for thin film of CdTe, here 2θ is extending from 20^0 to 80^0 , from given diffractogram it is clear that the prominent diffraction peaks were found at 2θ values of 23.8283, 39.3800, 46.5800 which are associated with the planes (111), (220), (311) respectively, with cubic crystal structure and the equivalent evaluated values of 'd' are 3.71868, 2.27110, 1.93828 respectively, which were nearly identical with standard data [JCPDS Card No.: 15-0770]. An additional peak corresponding to Tellurium (Te) is found at 20 value 27.5700 [JCPDS Card No.: 04-0554] which is associated with (101) plane of Tellurium. The highest intensity reflection peak is along (111) plane, which confirms the growth of thin-film along this plane [23]. The reaction of Te ions with oxygen present in the ambience can result in the formation of oxides like TeO₂, CdTeO₃ [13], but with proper care and evacuation of air ambience inside the chamber or by using any other carrier gas like nitrogen this can be avoided.

The crystallite size (D) was calculated from Scherrer formula and lattice strain (ε), dislocation density (δ) were evaluated from further relations as follows [24–27]:

$$D = \frac{k\lambda}{\beta \cos\theta}, \ \varepsilon = \frac{\beta \cos\theta}{4}, \ \delta = \frac{1}{D^2}$$

where λ indicates the X-ray wavelength, β indicates FWHM of the diffraction peak in radians, the Bragg angle is indicated by θ , Scherrer constant (k) was taken k = 0.9. The average crystallite size computed from the Scherrer formula was 12 nm, and it is nearly identical with the literature values [28]. The microstructural constants such as dislocation density (δ), lattice strain (ϵ), and texture coefficient obtained for (111) plane are 7.076×10^{-3} lines.m $^{-2}$, 2.916×10^{-3} , 1.165048544 respectively. tively and these values are well matching with previously reported terms [1]. Dislocation is a crystal imperfection produced in any part of crystal compared to whole crystal, if the distance between dislocations is more than interatomic distance and dislocation density is higher it may increase the band gap of nanomaterial [29] but for the as-prepared film the dislocation density value is very small which indicates no rise in optical band gap which was confirmed by optical band gap analysis. The obtained films possess a polycrystalline nature with the pure cubic crystal structure, which shows that ion by ion mechanism had been taken place during pyrolytic decomposition [30].

The lattice parameters for the cubic unit cell (a, b & c) were calcu-



Fig. 2. EDX spectra for the deposited CdTe thin film.

Table 1

The atomic percentage of Cadmium and Tellurium elements before and after deposition.

| As-ta atom perce in sp solut (%) | iken iic entage ray ion | As-obse mass percent the film analysis | rved age in by EDX s (%) | As-observed atomic percentage in the film by EDX analysis (%) | | Compositional Ratio (atomic) |
|---|-------------------------------------|--|-----------------------------------|---|-------|------------------------------|
| Cd | Те | Cd | Te | Cd | Te | Te/Cd |
| 50 | 50 | 47.07 | 52.93 | 50.24 | 49.76 | 0.99 |

lated using the relation [1]:

$$\frac{1}{d^2} = \frac{h^2 + k^2 + l^2}{a^2}$$

where 'd' denotes the interplanar spacing. The evaluated lattice parameters for the CdTe cubic crystal structure were found to be a = b = c = 6.47 A⁰. These values of lattice parameters were found

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matching with standard data [JCPDS Card No.: 15-0770].

3.3. Elemental and morphological inspection

Elemental analysis of the deposited CdTe thin film was carried out using the EDX spectra as shown in Fig. 2

Fig. 2 confirms the presence of Cadmium and Tellurium elements and their distribution over the film. The elemental analysis showed that the films possess good stoichiometry. Table 1 gives the elemental composition, in terms of Cadmium and Tellurium atomic percentage, within the precursor solution and within the prepared thin film, which is well matching with literature values [3,31].

Fig. 3 shows the SEM image of the as-deposited CdTe thin film.

Fig. 3, shows that the thin film is composed of needle-shaped grains that are spread along the entire substrate area indicating the well-defined and compact growth of polycrystalline CdTe thin film. It can be also seen that films are pinhole free, crack-free, uniform and densely packed. The SEM image processing gives the average grain size to be 30 nm and from XRD data analysis it was obtained to be 12 nm, this difference may have arrived due to the fusion of grains and cluster formation [32].



Fig. 4. Raman spectra of CdTe thin film.



Fig. 3. SEM micrograph of the CdTe thin film.





Fig. 5. a. Absorbance, b. Transmittance and c. Reflectance of CdTe thin film.

3.4. Vibrational analysis

Raman spectroscopy has been employed for the determination of the degree of crystallinity of the thin films. Investigation of lattice dynamics such as rotation or vibrations of molecules within thin films can be carried out using Raman spectroscopy [33]. Fig. 4 represents the FT-Raman spectra of the as-prepared CdTe film.

In Fig. 4 two intense peaks can be observed, out of which the peak at 139.30 cm^{-1} corresponds to fundamental transverse optic (1TO) and the peak at 165.33 cm^{-1} correspond to longitudinal optic (1LO) phonon modes of Cadmium telluride. These peaks also indicate the good crystalline quality of the as-prepared film. A very weak 2LO mode is observed at 341.79 cm^{-1} . A peak observed at 119.05 cm^{-1} is associated with Te–O bond which confirms the formation of oxides within the film. The analogous behavior was reported earlier also [13,31]. Raman intensity of Te peak is more as compared to the CdTe peak, this may be due to its good crystallinity which can also be confirmed by X-ray diffractogram in Fig. 1.

3.5. Optical analysis

3.5.1. Absorbance, reflectance, and transmittance study

Fig. 5a, 5b & 5c gives the absorbance, reflectance, and transmittance,

shown by the deposited CdTe film.

It can be seen from absorbance spectra (Fig. 5a) that the absorbance is decreasing with increasing wavelength. The high absorbance in visible range suggests that the more photons from this range could be absorbed which is well appreciated. Also, the good absorbing nature of the prepared film shows the uniformity in thickness [8] and uniform distribution which was also seen in the SEM micrograph. Fig. 5b showed that the as-deposited film possesses the maximum transmittance equal to 65% but in the visible region (λ < 700 nm) the transmittance is very less (<7%), which suggest that the prepared films can be employed in CdS/CdTe solar cells [24].

3.5.2. Optical band gap study

Optical band gap examination of a semiconductor thin film is very noteworthy if it has to be employed in optoelectronic devices [33,34], as the working efficiency of optoelectronic devices can be very much affected by their band gap. The optical band gap of the as-prepared CdTe thin film is obtained from the famous Tauc's equation [34] as $\alpha hv = A(hv - E_g)^s$ where α indicates absorption coefficient (α = measured absorbance/thickness), hv indicates incident photon energy, A indicates an empirical constant of energy, E_g indicates the band gap and 's' indicates an empirical constant having values $s = \frac{1}{2}$, s = 2 for direct and indirect allowed electronic transitions [35]. Fig. 6a illustrates



Fig. 6. a. Direct energy band gap and b. Indirect energy band gap plots for CdTe thin film.



Fig. 7. a. Absorption index plot and b. Refractive index plot, for CdTe thin film.

the graph between hv versus $(\alpha hv)^2$ and Fig. 6b illustrates the graph between hv versus $(\alpha hv)^{1/2}$. Extrapolating the linear part of these graphs up to the X-axis gives the optical band gap.

When the hole and electron possess a similar momentum in valence band as well as conduction band and an electron directly emit a photon then it is a direct band gap otherwise if an electron emits photon by transferring momentum with an intermediate state is an indirect band gap [36]. The obtained optical direct and indirect energy band gap values for the as-deposited thin film are 1.52 eV and 0.62 eV respectively, these values are found to be matching with literature values [28]. CdTe is a direct band gap material but sometimes there may be an occurrence of indirect transition because of the numerous allowed level transitions between valence band and conduction band.

3.5.3. Study of refractive index (n) and absorption index (k)

Fig. 7a presents the graphs of the absorption index (k) and Fig. 7b presents the graph of refractive index (n), both plotted against wavelength.

Absorption index (k) has been evaluated from the formula [37] $k = \frac{\alpha\lambda}{4\pi}$ and the refractive index (n) was evaluated using Fresnel's formula [38]

 $R=\frac{(n-1)^2+k^2}{(n+1)^2+k^2}$; where R indicates the measured reflectance.

The extinction coefficient or absorption index (k) is the ability of a material to absorb electromagnetic waves. Its value is less if the

electromagnetic wave gets absorbed easily and it is higher when the absorption of an electromagnetic wave by a material is less and wave gets extinct from it. The absorption index of the prepared film is maximum in visible region but with increasing wavelength, it is decreasing and above 1500 nm wavelength it becomes nearly constant. This decreasing absorption index indicates the loss of a portion of electromagnetic wave at higher wavelength, due to scattering. The refractive index can reveal the variation in the speed of electromagnetic wave after interaction with the material. From Fig. 7b it is clear that with an increase in wavelength the refractive index is also increasing.

3.5.4. Study of optical dielectric properties

The investigation of dielectric aspects like optical dielectric constant (ε_1), optical dielectric loss (ε_2), and loss tangent (tan δ) are crucial in understanding the absorption and loss of electromagnetic radiation because of dipole moment [36]. These dielectric quantities were calculated using the relations [39] $\varepsilon_1 = n^2 - k^2$ and $\varepsilon_2 = 2nk$ respectively. Fig. 8a, 8b & 8c shows the plots of λ versus ε_1 , ε_2 , and tan δ respectively.

It can be understood from Fig. 8a that at the higher wavelength the dielectric constant is maximum and it is lower in lower wavelength visible region, which indicates the prepared film has lower dielectric resistance in high wavelength region and higher dielectric resistance in the visible region. From Fig. 8b it is clear that the dielectric loss is higher





Fig. 8. a. Dielectric constant plot, b. Dielectric loss plot and c. Loss tangent variation plot, for CdTe thin film.

in the visible region and relatively lower and nearly the same in higher wavelength region. The loss tangent which gives the amount of loss of propagating signal and dielectric loss were observed to be very small for the as-deposited thin film which indicates the film has defect-free surface morphology.

3.5.5. Study of optical conductivity and electrical conductivity

The UV–Visible spectroscopy can be employed to study the optical conductivity and electrical conductivity. The relations used for the

measurement of optical conductivity (σ_{opt}) and electrical conductivity (σ_e) of the as-deposited CdTe thin film were [40] $\sigma_{opt} = \frac{anc}{4\pi}$ and $\sigma_e = \frac{2\lambda\sigma_{opt}}{\alpha}$ respectively, where the symbols take their typical sense. Fig. 9a represents the graph of λ versus σ_{opt} and Fig. 9b denotes the graph of λ versus σ_e .

The optical conductivity happens to be when electrons are excited by electromagnetic radiation instead of the applied electric field. It is also associated with the dielectric properties of the film. It can be understood from Fig. 9a & b, that the optical conductivity is higher for the visible

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Fig. 9. a. Optical conductivity and b. Electrical conductivity plot for the CdTe thin film.



Fig. 10. plot for variation of logp versus 1000/T for CdTe thin film.

region and with an increase in wavelength it decreases, whereas electrical conductivity is less for the visible region and increases with an increase in wavelength.

3.6. Electrical conductivity analysis

The transport properties of the material can be well understood in contrast with electrical conductivity. D.C. two probe press technique is employed, to study the electrical resistivity of the prepared film, at a succession of temperatures from 308 to 573 K, under dark. The relation used for the evaluation of the dc electrical resistivity (ρ) was $\rho = \frac{RA}{t}$, where R indicates the resistance, A indicates the area of film which was $2.5 \times 1 \text{ cm}^2$ and t indicates film thickness. These obtained resistivity digits were fitted by using the relation [41] $\rho = \rho_0 \exp\left(\frac{E_a}{kT}\right)$), where T indicates the absolute temperature, ρ_0 denotes a constant, k denotes Boltzmann constant and E_a is the activation energy. The estimated room temperature electrical resistivity of the as-prepared thin film was $1.2 \times 10^6 \Omega$ -cm, and it is well matching with earlier reported values [42]. Fig. 10 represents the graph of 1000/T versus log ρ , which gives the association of the dark current with temperature ranging from 300 to 500 k.

Fig. 10 clearly indicates that resistivity of the CdTe film is temperature dependent and it is decreasing with increasing temperature which indicates that the conduction was taken place through the activated process and also it confirms the semiconducting behavior of the prepared thin film. The activation energy (E_a) values at low and hightemperature regions were found to be 0.31 eV and 0.93 eV respectively [42]. The semiconducting nature of the as-deposited CdTe film can also be established from the change in activation energy. According to carrier trapping theory the activation energy at room temperature should be lower than 50% of optical energy band gap and the results obtained for as-prepared film are well accordance with this theory, which suggest that when temperature is increased the activation energy barrier gets reduced and the electrons across grain boundaries are thermally emitted which results in higher conduction [9].

3.7. Thermoelectric power study

Seebeck coefficient or Thermoelectric power measurement was used to examine the type of conductivity possessed by the as-prepared thin films. It considers the polarity of voltage generated due to the temperature difference at the two ends of the film. If the voltage polarity towards the hot end of the film is positive, the film possesses n-type conductivity and if the voltage polarity towards the hot end of the film is negative, the film possesses p-type conductivity. The as-prepared film possesses the p-type conductivity.

4. Conclusions

CdTe thin films are prepared by employing a chemical spray pyrolysis technique at 300 °C, on glass substrates. The film thickness was found to be 497.28 nm. XRD data examination discovered that the fabricated film is polycrystalline and built with the cubic crystal structure and fancily grown-up along (111) plane. The average crystallite size calculated after XRD data analysis is found to be 12 nm. The elemental composition or the abundance of Cd and Te elements within as-prepared films was established with EDX investigation. The SEM micrograph processing showed that the film has a compact morphology without crystal defects and the grains were of needle shape with the average grain size 30 nm. The Raman spectra show that fundamental transverse optic (1TO) mode is at 139.30 cm^{-1} and longitudinal optic (1LO) phonon mode is at 165.33 cm⁻¹. The film possesses high absorbance and low transmission in visible region which suggests that they can be employed in solar cells as an absorber layer. The estimated optical direct band gap was 1.52 eV. The electrical resistivity at room temperature was $1.2\times 10^6\,\Omega\text{-cm}.$ TEP study confirmed the p-type conductivity of prepared films.

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Declaration of competing interest

The authors declare that there is no conflict of interest in current work.

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