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# Influence of deposition time in CdTe thin film properties grown by Close-Spaced Sublimation (CSS) for photovoltaic application

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#### ABSTRACT

Cadmium Telluride (CdTe) thin films were grown on borosilicate glass substrates by close-spaced sublimation (CSS) at a pressure of 1.5–2 Torr in Ar ambient. CdTe thin films were sublimed at a source temperature of 625 °C and substrate temperature of 595 °C. In this study, the impact of various deposition times on the structural, morphological, topographical, electrical and optical properties of CdTe thin films has been explored to achieve high quality thin film absorber layer for solar cells applications. The crystalline structure, surface morphology, surface topology, electrical and optical properties of the films were examined by using X-ray diffraction (XRD), Field Emission Scanning Electron Microscopy (FESEM), Atomic Force Microscopy (AFM), Hall Effect measurement and UV–Vis spectrophotometry, respectively. XRD investigation demonstrated that CdTe film shows polycrystalline nature pronounced with cubic zinc blende structure with a strong preferential (1 1 1) orientation. The FESEM images illustrated that the surface morphology and the average grain size of the film's were dependent on the deposition times of CdTe thin films. AFM analysis revealed noteworthy changes in the film's mere arm  $^{-3}$ . Band gap of CdTe thin film was found in the range 1.45–1.48 eV, which is suitable to be used in CdTe thin film solar cells.

Introduction

Improving the efficiency as well as reducing the cost has always been the spotlight of photovoltaic (PV) systems research [1]. Nowadays, energy harvesting by photovoltaics has turned out to be a key factor for assuring the world's ever increasing energy demand. Solar energy generation demand has been mostly compelled by quickly declining system costs. To maintain the high level of desirability for this energy source, there is a high necessity for further decline in production costs. This will guide to an assortment of manufacturing techniques, which merges high efficiency and cost-saving production [2]. The compound semiconductor thin films are one of the most promising materials for the fabrication of optoelectronic devices [18]. However, many of the compound semiconductors have been deposited by many processes. Though the mission of CdS/CdTe hetero-junction solar cell initiated in the early 1970s with an efficiency of about 6%, the current efficiency has reached 22.1% stated by First Solar Inc. [5]. Sustainable research efforts are being pursued to overcome the gap between the maximum attainable theoretical efficiency (~30%) and the highest recorded laboratory efficiency (~22.1%) of CdTe solar cells [17].

CdTe has been acknowledged as a foremost thin film material for its optimal bandgap of 1.45 eV. CdTe absorbs more than 90% of obtainable photons in 1  $\mu$ m thickness, therefore 1–3  $\mu$ m film are adequate for thin film solar cells [3–7,18]. CdTe solar cells such as homo-junction and

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hetero-junction have been inspected so far [8-10]. CdTe can be deposited by over 14 techniques which varies largely and can significantly influence the material characteristic and device quality [11-13]. Physical vapor deposition is the simplest method for the fabrication of thin film semiconductors. CdTe deposition methods include close-spaced sublimation (CSS), vapor transport deposition (VTD), thermal evaporation, sputtering etc. [14-18]. CSS and CdTe are directly interconnected due to widespread exploitation of CSS in the development of CdTe thin film [19,20]. The benefits of CSS method are high deposition rates and a tremendous exploitation of the raw material, leading to low manufacturing costs and viable module prices. CSS is a low-cost technique for the deposition of polycrystalline thin films owing to the reasonable operating pressure (0.01-10 Torr) and simple design [21,22]. CdS/CdTe solar cells are presently of technological significance because of their easiness of deposition method and comparatively high efficiency [23].

Nowadays, the obtainability of ample Cd and Te is a great worry for scientists and researchers for large scale manufacturing of CdS/CdTe solar panels. Apprehensions are more crucial for Te element because of its insufficiency compared to cadmium. This investigation has been executed by Zweibel [24] in detail. As a solution to this matter, the scientific research should be focused towards thin CdTe layers. Therefore, the key purpose of this study is to control the CdTe thickness by varying the deposition times. The effect of deposition times on the structural, morphological, topographical, electrical and optical properties of CSS grown CdTe thin films will also be discussed to comprehend its utilization in thin film solar cells.

#### **Experimental details**

CdTe thin films were grown on commercially available borosilicate glass substrates (3 cm  $\times$  3 cm) via CSS system. All substrates were cleaned by ultrasonic process and finally dried up by N<sub>2</sub> gas flow. Source material employed in this study was fine powdered CdTe (99.99%), which was first sintered at 700 °C for 30 min. Then, CdTe thin films were deposited via CSS process, where source and substrate temperature were ramped up at the same rate and finally differed for various duration as considered to be deposition time. A schematic of the in-house built CSS system is presented in Fig. 1.

The substrate and source are detached by 2 mm and supported by proper holders. The system is sustained at the set temperatures by 2 kW halogen lamps. The thermocouples are employed to control the temperatures. Table 1 presents the deposition conditions of CdTe thin film growth. The chamber is reserved at 1.5–2 Torr (Ar gas) to generate the suitable deposition ambient. The argon gas is used as to maintain inert atmosphere inside the chamber. Fig. 2 depicts the CSS system along with CSS grown CdTe thin film.

The choice of deposition parameters particularly depends on the process of deposition. The significant process parameters in CSS are the temperatures of the source and the substrate, the nature of the atmosphere, the pressure in the reaction tube and the composition of the



Fig. 1. Schematic of close-spaced sublimation (CSS) system.

Table 1

Deposition	parameters	of	CSS	grown	CdTe	thin	films.
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Parameter	Condition		
Substrate Temperature	595 °C		
Source Temperature	625 °C		
Deposition Pressure	1.5–2.0 Torr (Ar gas ambient)		
Source-substrate Spacing	2 mm		
Deposition Time	1, 2, 3, 4, and 5 min		

source material. These parameters are interconnected. In this study, the deposition parameters such as source-substrate temperature, spacing, deposition time and pressure etc. have been selected based on the optimization from existing literature review, theoretical considerations, experimental data and in some cases by reasonable estimations. The source-substrate temperature, deposition pressure, and source-substrate spacing are adjusted to result in deposition rates of about  $0.5-1.0 \,\mu\text{m/min}$ . The key aim of this study is to control the film thickness below  $5 \,\mu\text{m}$  toward stable pinhole free CdTe thin films. By keeping other deposition parameters constant, deposition time is altered to achieve the desired film thickness.

Identification of phases and structural properties was inspected via X-ray Diffraction (XRD) using a BRUKER aXS-D8 Advance Cu K $\alpha$  diffractometer (Cu K $\alpha$  excitation wavelength of 1.541 Å) at room temperature. Surface morphology, grain size and cross-sectional images were viewed by using Carl Zeiss Merlin Field Emission Scanning Electron Microscopy (FESEM) operated at 3 kV. "NANOSURF EASYS-CAN 2 AFM" (Atomic Force Microscopy) SYSTEM" was utilized to investigate the surface topography and roughness. The electrical parameters were obtained by Hall Effect measurement system (HMS ECOPIA 3000). Optical characteristic analysis was done by using Perkin Elmer Lambda 950 UV/Vis/NIR spectrophotometer.

## **Results and discussion**

## Structural properties analysis

XRD patterns were documented in the  $2\theta$  range from  $20^{\circ}$  to  $60^{\circ}$  with a step size of  $0.02^{\circ}$ . The aim of XRD analysis was inspecting the impact of different deposition times on the structural and crystallographic properties of CSS grown CdTe thin films. The XRD patterns of CSS grown CdTe thin films are depicted in Fig. 3.

All the grown films illustrated polycrystalline property along the (111) plane and originated at  $2\theta = 23.8^{\circ}$  for all the deposition times confirming a cubic zinc blende structure. Another three low intensity peaks were found at  $2\theta = 39.30^{\circ}$  and  $2\theta = 46.51^{\circ}$  and  $2\theta = 56.91^{\circ}$ correspond to (220) and (311) and (400) plane, respectively. From the XRD peak, it is obvious that the pattern is dominated by the (111) peak as the intensity of other three CdTe peaks, (220), (311) and (400) are very weak. All these films are polycrystalline with cubic phase and are in agreement with the XRD results reported by Dharmadasa et al. and Spalatu et al. [30,31]. There are some factors affecting the intensity of XRD. Changing the atoms in the unit cell alters the diffraction intensity. Typically, differences in XRD peak intensities can be correlated to the variations in the scattering intensity of crystal components or their lattice arrangement and might be interpreted due to the crystallite size discrepancies that have resulted from many variables. It may also occur due to the discontinuity of the film texture, the large porosity of samples, great voids or the roughness of the substrate. Therefore, intensities of (111) diffraction peak differ for higher deposition times as the total area of crystal face is different conferring to crystal habit which are in agreement with the results found earlier [40]. Furthermore, the material volume reduces after each sublimation process in CSS deposition. Consequently, the surface has reached a steady state morphology demonstrating the difference in the deposition rate and final thickness for each run. The amount of deposited material



Fig. 2. (a) View of CSS system and (b) CSS deposited CdTe thin film.



Fig. 3. The XRD patterns of CSS deposited CdTe thin films.

should be the same to attain similar intensity. Peak intensity might vary if the thickness and the deposition time are changed. The sample deposited for 3 min displays the strongest preferred (1 1 1) orientation. The peak intensity showed declining tendency after 3 min deposition time which might also be ascribed to the growth of successive thin layers of material and decrease in surface mobility on glass surface [7]. All the diffracted peaks found from XRD were compatible with the JCPDS data file (15–0770); and were in good concurrence with the literature of CdTe cubic structure.

From the Brag's law  $[d_{(hkl)} = (\lambda/2) \csc\theta]$  and Vegard's law  $[a_{cubic} = d_{hkl} (h^2 + k^2 + l^2)^{1/2}]$  [25], the lattice constant 'a' for cubic phase structure  $[h \ k \ l]$  was calculated. Crystallite size (D) of the films has been calculated by using the full-width at half maximum (FWHM)

of the (1 1 1) peak and Scherrer's formula  $[D_{hkl} = 0.9\lambda/(\beta cos \theta)]$  [26]. Full width at half maximum [FWHM] values have incredible correlation with the crystallinity. The higher values of FWHM point out the decline of the crystallinity, whereas the bigger crystallite sizes specify the enhanced crystallinity of the films [27]. The microstrain ( $\varepsilon$ ) has been measured from the relation [ $\varepsilon = \beta/4$ tan $\theta$ ] [28]. Monocrystalline property symbolizes lower microstrain value. In contrast, the high  $\varepsilon$ value denotes the polycrystalline film. By using the Williamson and Smallman's relation [ $\delta = n/D^2$ ] [29], the dislocation density has been calculated. The computed structural parameters of CSS deposited CdTe films are shown in Table 2.

The average crystallite sizes were in the range 40–55 nm as observed from the calculated values. The average crystallite sizes slightly decreased for higher deposition times because of the variation in the crystallinity, which may be attributed to recrystallization and reorientation of the films. Crystallite size was maximum (54.16 nm) for the films deposited for 2 min, which is a good indicator that the films can be used in solar cells as the absorber layer, since large grain size demonstrates good crystallinity, which reduces the parasitic resistances and consequently improves device performance [7]. The values of crystallite sizes found in this study are comparable with the study conducted by Mendoza et al. [32]. The lattice constant of 0.65 nm was found for all the CdTe diffracted peaks in all deposition times. The microstrain and dislocation density of CdTe films were strongly influenced by the deposition time. Fig. 4 shows the variations of the microstrain and dislocation density for different deposition times.

The highest microstrain of  $3.92 \times 10^{-3}$  was obtained for 5 min deposition time. Maximum value of dislocation density  $0.55 \times 10^{11}$  cm<sup>-2</sup> was also found for the same deposition time. Stress or stain in a material can cause visible fluctuations in the diffraction pattern of a material. The internal strain or stress and dislocation density illustrates different behavior for 2 min which may be due to the lattice disarrangement and imperfections originated in the crystal. Lower strain and imperfections implicate towards films with better crystalline quality which is desirable in the fabrication of high efficiency solar cell device. The microstrain and dislocation density are observed to rise with deposition time due to decline in corresponding average crystallite size

Table	2
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Structural parameters of CdTe thin films for various deposition times.

Deposition Time	hkl	d <sub>hkl</sub> (nm)	a (Å)	β (deg)	D (nm)	$\epsilon \ [\times 10^{-3}]$	$\delta \ [\times 10^{11}] \ (cm^{-2})$
1 min	(111)	0.3725	6.45	0.0031	45.133	3.714	0.491
2 min	(111)	0.3724	6.45	0.0026	54.160	3.094	0.341
3 min	(111)	0.3727	6.46	0.0030	47.787	3.510	0.438
4 min	(111)	0.3723	6.45	0.0031	45.134	3.712	0.491
5 min	(111)	0.3729	6.46	0.0033	42.756	3.924	0.547



Fig. 4. Microstrain and dislocation density variations for CdTe thin film.

which indicates the formation of high-quality thin films with enhanced crystallinity for lower deposition times. The results are in agreement with the reported work of Chander et al. [18].

## Surface morphology analysis

FESEM is a very proficient method to find the grain growth mechanism and surface morphology [29]. Fig. 5 shows the FESEM surface morphology and cross section images of CSS deposited CdTe thin films for different deposition times. The surface morphology and the average



Fig. 6. Variation of grain size and thickness of CdTe thin films for various deposition times.

grain size of the films are highly dependent on the deposition times as a function of thickness as perceptible from the FESEM images. The surface morphology studies imply that the films are uniform, smooth, homogeneous and nearly dense-packed as well as free from voids, cracks or pinholes. The obtained specific grains refer to the fastest, favoured growing direction in contrast to the round CSS grains, where no preferential growth direction can be found [33].

The material volume declines after each sublimation as the initial material on the graphite box decays after individual sublimation process. Consequently, thickness of the grown films fluctuates for different



Fig. 5. FESEM surface morphology and cross section images of CdTe thin films.

deposition times. The thickness for all the films was in the range  $1.50-2.40 \,\mu$ m. The grain sizes were almost constant for all the deposition times as evident from Fig. 6 and found in the range  $1.50-2.0 \,\mu$ m which are comparable with the film thickness and are in accordance with the measurement done by Hernández-Torres et al. [33]. Film grown at lower deposition time showed smaller grain sizes. The thickness and grain size of the film is maximum for 3 min deposition time and then reduces for higher deposition time. There is vital need to optimize CdTe deposition procedure to acquire larger grain. The different morphology and structure (crystalline) of the films might be due to the defects created by different thicknesses. This is because the rate of particle aggregation during the film growth is a key issue that directs the morphology and structure (crystalline) of the films [33]. The FESEM images and grain size support the XRD results and matches with the study by Spalatu et al. [31].

According to the literature, crystallite size is a measure of the size of coherently diffracting regions of a material. The crystallite size is commonly determined from the XRD pattern using the Scherrer equation. In the Scherrer formula, a lot of assumptions are being made which could be different for the real samples. It assumes that all crystallites have the identical shape and size though the shape of crystallites is usually irregular. On the other hand, grains are volumes inside crystalline materials with a specific orientation. Grain is either a single crystalline or polycrystalline material and is present either in bulk or thin film form. Grain size usually refers the average diameter of the individual crystal orientations found in polycrystalline materials. During the processing, smaller crystallites come closer and grow to become larger due to kinetics. Therefore, in the most likely scenario, the grain is larger than a crystallite. Crystallite size is equal to grain size if the grain is perfectly single crystallite. Grain size and morphology are commonly determined by SEM (but not XRD). The grain size measured from an SEM could be the coalescence of small crystallites into one large grain made of several crystallites and it is sometimes difficult to resolve individual crystallite from an SEM [29]. The grain size measured from SEM is an average value whereas the Scherrer formula calculates the crystallite size using the diffraction information from a single plane at a specific 20 and FWHM value [29]. However, grains of sintered samples contain several dislocations and defects, which interrupt the periodicity of the crystalline nature. Hence, an individual grain may contain several crystallites. XRD technique provides the information of this crystallite size present in the grains whereas the microscopic investigation using SEM provides the grain size of the material. Since grain comprises many crystallites, the crystallite size and grain size are not same.

#### Surface topography analysis

AFM provides information about the surface topology of a thin film and allows topographies of the film surface to be plotted [37]. CSS grown CdTe film often suffers from poor coverage and is rough. Therefore, AFM analysis was carried out to inspect the correlation between the roughness and deposition times of CdTe thin films. AFM is utilized to achieve the surface topography, average roughness (R<sub>a</sub>) and RMS roughness (R<sub>q</sub>). Fig. 7 illustrates the topography (from 3D image) of CdTe film in a scanning area of 10  $\mu$ m  $\times$  10  $\mu$ m for deposition times 1 min, 2 min, 3 min, 4 min and 5 min, respectively.

Fig. 8 shows the variation of average roughness and RMS roughness with the change in deposition times of CSS deposited CdTe thin film. The grown films are rough, but the peak-to-valley detachment is less than the total thickness confirming even exposure. The average roughness values are in the range 59-120 nm which are in accordance with the study executed by Seth et al. [34] and Major et al. [35]. AFM results are also in agreement with the findings of the FESEM analysis indicating the improved grain size for higher deposition times. The roughness is an indication of the level of smoothness of the surface. Both  $R_a$  and  $R_g$  gives roughness of thin films but their approach of

calculating the roughness is different. Higher roughness would cause more light scattering that may lead to higher reflectance or even higher attenuation. Higher roughness is required to achieve greater optical length. For 1 min deposition time, the  $R_a$  and  $R_q$  were about 59.61 nm and 67.92 nm, respectively. When deposition time was 4 min, the Ra and R<sub>a</sub> boosted to 120.95 nm and 150.22 nm, respectively. The surface roughness increased up to 4 min and thereafter declined, which might be attributed to a partial phase transition as well as oxidization, which started at higher deposition time. The changes in surface roughness with time might also be accredited to surface defects, alteration in the degree of disorder and the porosity of the films as reported by Chandramohan et al. [41]. From the topography images, it is obvious that the surface roughness is affected by deposition times. Therefore, a smart trade-off between deposition time and roughness is required for better CdTe thin film quality and performance. The roughness is maximum for films deposited for 4 min, which is needed for a high-efficiency solar cell device as the optical length of the incident photon is greater owing to the enhanced probability of photon absorption because of total internal reflection at high roughness [36,37]. This property suggests that CSS grown CdTe films could be exploited as an absorber layer in singlejunction device as well as tandem device.

## Electrical properties analysis

The electrical parameters such as carrier concentration, mobility and resistivity were measured with an applied magnetic field of 0.55 T and probe current of 40nA. The measured electrical parameters for various deposition times are presented in Table 3.

The electrical resistivity was in the order of  $10^4 \Omega$  cm demonstrating insignificant changes with the growth conditions. The electrical resistivity was increased for 5 min and consequently the conductivity decreased which might be attributed to CSS chamber contamination during CdTe thin film growth. Therefore, the electrical conductivity of CdTe thin films is strongly influenced by the deposition time, which may be ascribed to the tunneling of the charge carriers through grain boundary barriers and recrystallization of grains during the growth process [43]. Resistivity values are in accordance with the data reported by Lianghuan Feng et al. [38]. The highest mobility  $(7.67 \text{ cm}^2/$ Vs) and the lowest resistivity (4.82  $\times$  10<sup>4</sup>  $\Omega$  cm) was obtained for CdTe film with 3 min deposition time. Mobility increased until 3 min and then declined with the increase of deposition time. This observation can be linked to the relatively higher bulk carrier concentration. Carriercarrier scattering consequence is much apparent for films with relatively higher bulk carrier concentration due to the increased probability of carrier collisions, which in return directs to lower mobility [32]. It was also found that the carrier concentration of the films was in the order of  $10^{13}$  cm<sup>-3</sup>. Understanding and controlling the carrier concentration in CdTe polycrystalline thin films has been enormously challenging and limiting. Ionic bonding between constituent atoms frequently leads to persistent intrinsic compensating defect chemistries and strong self-compensation that are difficult to control. The historical CdTe doping limit is an indication of defect compensation when relying on a mix of native point defects and poorly controlled impurities and doping mechanisms, which is a common issue with other polycrystalline compound semiconducting thin film materials. CdTe has the drawback as it is difficult to achieve high doping concentration due to self-compensation from intrinsic defects form e.g: vacancies (V<sub>Cd</sub>, V<sub>Te</sub>), interstitial defects (Cd<sub>i</sub>, Te<sub>i</sub>) and grain boundaries [45]. Therefore, CdTe carrier concentration is found low which is one of the key challenges to improve CdTe solar cells energy yield performance. It is therefore suggested that the variation of deposition time as a function of film thickness has notable impact on film properties. With the increase of deposition time, crystallinity has improved as evident from the XRD peaks. Aligning to literature, it is found that better crystallinity results in lower electrical resistivity, which is consistent from this study as well. This is mainly due to reduced carrier scattering and recombination



across the grain boundaries.

## Optical properties analysis

Optical characteristics were analysed by measuring the absorbance of deposited CdTe films. Absorbance of the reference glass is subtracted to equalize the influence of underlying substrate. Optical property analysis was carried out to examine the energy band gap, Urbach energy, refractive index and extinction coefficient of deposited CdTe films. The measured absorption coefficient values for all the samples are within the range of  $10^4$ – $10^5$  cm<sup>-1</sup> in the visible region which confirms the absorbance of more than 90% as desired for an effective absorber layer. To calculate the band gap, the optical absorbance data was employed. Direct band gap values can be estimated by plotting the straight-line portion of the graph to zero absorption coefficients. Basically, the energy band gap is found by the intercept on the energy axis [39]. The energy band gap,  $E_g$  and the nature of transition are analysed by the equation  $[\alpha h\nu = A (h\nu - E_g)^{1/2}]$  [26,29]. The graphs of  $(ah\nu)^2$  vs  $h\nu$  are plotted in Fig. 9 to determine the energy band gaps of the films.

The linear nature of the Tauc plot specifies that the CdTe is a direct band gap compound semiconductor. The optical energy band gap of CdTe thin films are found in the range 1.45-1.48 eV as tabulated in Table 4. The obtained band gap values are quite similar with the study conducted by Patel et el. [17] and Lianghuan Feng et al. [38]. The change in optical band gap may be due to the variation in plasma

6



Fig. 8. Variation of average and RMS roughness of CdTe thin film for various deposition times.

frequency which might be attributed to the change in film carrier density and mobility. The band gap of semiconductor might also be affected by the dislocation density, disorder at the grain boundaries, stoichiometric deviations, quantum size effect and change in preferred orientation.

Urbach energy (E<sub>u</sub>) signifies the degree of structural disorder in thin films. This quantity depicts the local defect or localized states near the optical energy gap. The dependency of the optical absorption coefficient of thin films with photon energy may arise from electronic transitions between localized states. The  $E_{\mathrm{u}}$  values are deduced from the slope of linear fit to the logarithmic plot of the absorption coefficient and photon energy using the equation,  $E_u = 1/[\Delta(\ln \alpha)/\Delta(h\nu)]$  [27]. The estimated values of  $E_u$  for all samples are given in Table 4. Generally, the Eu values are inversely proportional with the optical band gap of thin films. It is observed that the Eu value decreases with increasing optical energy gap for higher deposition times. The obtained values of Eu for 1 min, 2 min and 3 min CdTe films are 62 meV, 74 meV and 61 meV, respectively and then declines for 4 min and 5 min. This may be attributed to the appearance of defects, which cause redistribution of the localized states from band to tail. It also facilitates more possible transitions from band to tail and tail to tail, which leads to the simultaneous shrinkage and extending of optical energy gap and Urbach tail, respectively [42]. The higher values of Eu in the deposited CdTe films specify the higher phonon state disorder and/or higher doping concentrations in the films [27]. It is assumed that the nature of the structural disorder in as-deposited films is dominated by intrinsic defects such as dislocation and/or vacancies [27].

The refractive index gives information about vacancies existing in the deposited film and defined as a measure of density [43]. Usually, the refractive index increases with decreasing order of optical energy band gap and both parameters can be related as per Harve-Vandamme model [18]. The refractive index is calculated using the relation  $n^2 = 1 + [(A/E_g + B)]^2$  where, A and B are constants having values

13.6 and 3.4 eV, respectively [18]. The refractive index is found in the range 2.96–2.98 and are tabulated in Table 4. The refractive index is found corresponding to optical energy band gap and may be attributed to the variation in dislocation density and crystallinity [18]. The results are consistent with the earlier study conducted by Shaaban et al. [44] who observed a similar behavior of refractive index for CdTe thin films.

The extinction coefficient (k) provides information about the absorption of light in the materials medium owing to elastic scattering [43]. The extinction coefficient is estimated from the values of  $\alpha$  and  $\lambda$  using the known formula  $k = \alpha \lambda / 4\pi$  [37,44]. Fig. 10 illustrates the dependence of k on wavelength for CdTe of thin films. The extinction coefficient is found to increase with higher deposition times which may be attributed to the dominance in density temperature dependence of the extinction coefficient [43,18].

## Optimization and device structure

The experimental results revealed that the deposition times played crucial role to optimize the structural and optoelectronic properties of CdTe films. Considering all the characterization results, the source temperature of 625 °C, substrate temperature of 595 °C, deposition pressure of 1.5 Torr, source-substrate spacing of 2 mm are found optimum in this study. Deposition time of 1–4 min are suitable depending on the required thickness as higher deposition times might deteriorate the quality of the films. Based on the obtained characterization results, it can be stated that the CSS grown CdTe films exhibit desirable properties to be utilized as an absorber layer for the suggested Cd-based solar cells configuration as presented in Fig. 11.

However, all in all, it can be concluded that thorough optimization of process such as post-deposition annealing as well as subsequent layer deposition are necessary to achieve higher conversion efficiency of this kind of device configuration.

#### Conclusion

The impact of deposition time on the structural, morphological, topographical, optical and electrical properties of CdTe thin films has been inspected comprehensively with the optimization of properties to the solar cell applications. XRD analysis revealed that all the films illustrated polycrystalline characteristic along the (111) cubic plane as preferred orientation. The discrepancy in the crystallinity was acquired for different deposition times and the peak intensity showed declining tendency with lower deposition times. The surface morphology and the average grain size of the films were highly dependent on the deposition times and correlated the other properties. The average grain size of all the films was in the range 1.50-2.0 µm. Surface roughness was altered by the deposition times and depicted relatively less roughness value for lower deposition times. The highest mobility and the lowest resistivity were obtained for the CdTe film grown at 3 min deposition time. The optical transition was found to be allowed direct and the energy band gap was found in the range 1.45-1.48 eV. The refractive index was found in the range 2.96-2.98. It is therefore stated that the variation of deposition time in terms of CdTe thickness in CSS growth has a notable influence on CdTe film properties. All the investigated properties of CdTe thin films are well-matched in the range of standard reported values confirming their potential to be exploited as the absorber layers

Deposition time	Carrier concentration [ $\times 10^{13}$ ] (/cm <sup>3</sup> )	Mobility (cm <sup>2</sup> /Vs)	Resistivity [ $\times 10^4$ ] ( $\Omega$ cm)	Semiconductor type
1 min 2 min 3 min 4 min 5 min	2.50 2.31 1.54 2.16 2.38	5.30 7.30 7.67 5.88 3.62	5.90 4.93 4.82 5.82 7.25	p-type p-type p-type p-type p-type



Fig. 9. Plot of  $(ah\nu)^2$  versus photon energy  $(h\nu)$  for the evaluation of energy band gap of CdTe thin films.

Table 4			
Optical parameters	s of CdTe	thin	films.

Deposition time	Band gap energy, E <sub>g</sub> (eV)	Urbach energy, E <sub>u</sub> (meV)	Refractive index (n)
1 min	1.47	62	2.97
2 min	1.45	74	2.98
3 min	1.47	61	2.97
4 min	1.47	32	2.97
5 min	1.48	27	2.96

for photovoltaic application.

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Fig 10. Plotting of extinction coefficient of CdTe films as a function of wavelength.



Fig. 11. Schematic structure of CdS/CdTe thin film solar cells.

#### References

- Mungan ES, Wang Y, Dongaonkar S, Ely DR, Garcia RE, Alam MA. From process to modules: end-to-end modeling of CSS-deposited CdTe solar cells. IEEE J Photovolt 2014;4:954–61.
- [2] Enam FMT, Rahman KS, Kamaruzzaman MI, Sobayel K, Chelvanathan P, Bais B, et al. Design prospects of cadmium telluride/silicon (CdTe/Si) tandem solar cells from numerical simulation. Optik-Int J Light Electron Opt 2017;139:397–406.
- [3] Hossain MS, Rahman KS, Karim MR, Aijaz MO, Dar MA, Shar MA, et al. Impact of CdTe thin film thickness in Zn<sub>x</sub>Cd<sub>1-x</sub>S/CdTe solar cell by RF sputtering. Sol Energy 2019;180:559–66.
- [4] Ali AM, Rahman KS, Ali LM, Akhtaruzzaman M, Sopian K, Radiman S, et al. A computational study on the energy bandgap engineering in performance enhancement of CdTe thin film solar cells. Res Phys 2017;7:1066–72.
- [5] Chander Subhash, Dhaka MS. CdCl2 treatment concentration evolution of physical properties correlation with surface morphology of CdTe thin films for solar cells. Mater Res Bull 2018;97:128–35.
- [6] Chander Subhash, Dhaka MS. Time evolution to CdCl2 treatment on Cd-based solar cell devices fabricated by vapor evaporation. Sol Energy 2017;150:577–83.
- [7] Chander Subhash, Dhaka MS. Enhancement in microstructural and optoelectrical properties of thermally evaporated CdTe films for solar cells. Res Phys 2018;8:1131–5.
- [8] Hossain MS, Rahman KS, Islam MA, Akhtaruzzaman M, Misran H, Alghoul MA, et al. Growth optimization of Zn<sub>x</sub>Cd<sub>1.x</sub>S films on ITO and FTO coated glass for alternative buffer application in CdTe thin film solar cells. Opt Mater 2018;86:270–7.
- [9] Abbas A, West G, Bowers JW, Isherwood P, Kaminski PM, Maniscalco B, et al. The effect of cadmium chloride treatment on close-spaced sublimated cadmium telluride thin-film solar cells. IEEE J Photovolt 2013;3:1361–6.
- [10] Major J, Treharne R, Phillips L, Durose K. A low-cost non-toxic post-growth activation step for CdTe solar cells. Nature 2014;511:334–7.
- [11] Enam FMT, Rahman KS, Kamaruzzaman MI, Sobayel K, Aktharuzzaman M, Amin N. An investigation on structural and electrical properties of close-spaced sublimation grown CdTe thin films in different growth conditions. Chalcogenide Lett 2017;14(4):125–31.
- [12] Durose K, Boyle D, Abken A, Ottley C, Nollet P, Degrave S, et al. Key aspects of CdTe/CdS solar cells. physica status solidi (b) 2002;229:1055–64.
- [13] Razykov TM, Ferekides CS, Morel D, Stefanakos E, Ullal HS, Upadhyaya HM. Solar photovoltaic electricity: Current status and future prospects. Sol Energy 2011;85:1580–608.
- [14] Major J, Proskuryakov Y, Durose K, Green S. Nucleation of CdTe thin films deposited by close-space sublimation under a nitrogen ambient. Thin Solid Films 2007;515:5828–32.
- [15] Alamri S. The growth of CdTe thin film by close space sublimation system. physica status solid (a) 2003;200:352–60.
- [16] Cousins M, Durose K. Grain structure of CdTe in CSS-deposited CdTe/CdS solar cells. Thin Solid Films 2000;361:253–7.
- [17] Patel SL, Chander S, Kannan MD, Dhaka MS. Impact of chloride treatment on the physical properties of polycrystalline thin CdTe films for solar cell applications. Phys Lett A 2019.
- [18] Chander Subhash, Dhaka MS. Thermal evolution of physical properties of vacuum evaporated polycrystalline CdTe thin films for solar cells. J Mater Sci: Mater Electron 2016;27:11961–73.
- [19] Ferekides CS, Marinskiy D, Viswanathan V, Tetali B, Palekis V, Selvaraj P, et al.

High efficiency CSS CdTe solar cells. Thin Solid Films 2000;361:520-6.

- [20] Luschitz J, Lakus-Wollny K, Klein A, Jaegermann W. Growth regimes of CdTe deposited by close-spaced sublimation for application in thin film solar cells. Thin Solid Films 2007;515:5814–8.
- [21] Terrazas J, Rodríguez A, Lopez C, Escobedo A, Kuhlmann FJ, McClure J, et al. Ordered polycrystalline thin films for high performance CdTe/CdS solar cells. Thin Solid Films 2005;490:146–53.
- [22] Luschitz J, Siepchen B, Schaffner J, Lakus-Wollny K, Haindl G, Klein A, et al. CdTe thin film solar cells: interrelation of nucleation, structure, and performance. Thin Solid Films 2009;517:2125–31.
- [23] Kim M, Sohn S, Lee S. Reaction kinetics study of CdTe thin films during CdCl2 heat treatment. Sol Energy Mater Sol Cells 2011;95:2295–301.
- [24] Zweibel K. The impact of tellurium supply on cadmium telluride photovoltaics. Science 2010;328:699–701.
- [25] Rahman K, Haque F, Khan N, Islam M, Alam M, Alothman Z, et al. Effect of CdCl<sub>2</sub> treatment on thermally evaporated CdTe thin films. Chalcogenide Lett 2014;11:129–39.
- [26] Rahman KS, Aris KA, Karim MR, Aijaz MO, Dar MA, Shar MA, et al. Impact of Cu incorporation to the CdTe thin film properties for photovoltaic application. Chalcogenide Lett 2018;15(5):293–306.
- [27] Islam MA, Rahman KS, Sobayel K, Enam FMT, Ali AM, Zaman M, et al. Fabrication of high efficiency sputtered CdS: O/CdTe thin film solar cells from window/absorber layer growth optimization in magnetron sputtering. Sol Energy Mater Sol Cells 2017;172:384–93.
- [28] Mariappan R, Ponnuswamy V, Ragavendar M, Krishnamoorthi D, Sankar C. The effect of annealing temperature on structural and optical properties of undoped and Cu doped CdS thin films. Optik-Int J Light Electron Opt 2012;123:1098–102.
- [29] Khan NA, Rahman KS, Aris KA, Ali AM, Misran H, Akhtaruzzaman M, et al. Effect of laser annealing on thermally evaporated CdTe thin films for photovoltaic absorber application. Sol Energy 2018;173:1051–7.
- [30] Dharmadasa I, Bingham P, Echendu O, Salim H, Druffel T, Dharmadasa R, et al. Fabrication of CdS/CdTe-based thin film solar cells using an electrochemical technique. Coatings 2014;4:380–415.
- [31] Spalatu N, Hiie J, Valdna V, Caraman M, Maticiuc N, Mikli V, et al. Properties of the CdCl2 air-annealed CSS CdTe thin films. Energy Procedia 2014;44:85–95.
- [32] Mendoza MF, Pérez RC, Delgado GT, Marín JM, Orea AC, Angel OZ. Structural, morphological, optical and electrical properties of CdTe films deposited by CSS under an argon–oxygen mixture and vacuum. Sol Energy Mater Sol Cells 2011;95:2023–7.
- [33] Hernandez-Torres M, Silva-Gonzalez R, Casarrubias-Segura G, Gracia-Jimenez J, SEM, EDS. PL and absorbance study of CdTe thin films grown by CSS method. Sol Energy Mater Sol Cells 2006;90:2241–7.
- [34] Seth A, Lush G, McClure J, Singh V, Flood D. Growth and characterization of CdTe by close spaced sublimation on metal substrates. Sol Energy Mater Sol Cells 1999;59:35–49.
- [35] Major J, Proskuryakov Y, Durose K, Zoppi G, Forbes I. Control of grain size in sublimation-grown CdTe, and the improvement in performance of devices with systematically increased grain size. Sol Energy Mater Sol Cells 2010;94:1107–12.
- [36] Patel SL, Purohit A, Chander S, Dhaka MS. Thermal annealing evolution to physical properties of ZnS thin films as buffer layer for solar cell applications. Physica E: Low-dimensional Syst Nanostruct 2018;101:174–7.
- [37] Patel SL, Chander S, Purohit A, Kannan MD, Dhaka MS. Influence of NH4Cl treatment on physical properties of CdTe thin films for absorber layer applications. J Phys Chem Solids 2018;123:216–22.
- [38] Feng L, Zhang J, Li B, Cai W, Cai Y, Wu L, et al. The electrical, optical properties of CdTe polycrystalline thin films deposited under Ar–O 2 mixture atmosphere by close-spaced sublimation. Thin Solid Films 2005;491:104–9.
- [39] Haque F, Rahman KS, Akhtaruzzaman M, Abdullah H, Kiong TS, Amin N. Properties of sputtered ZnS thin films for photovoltaic application. Mater Res Express 2018;5(9):096409.
- [40] Inoue M, Hirasawa I. The relationship between crystal morphology and XRD peak intensity on  $CaSO_4^{-}$  2H\_2O. J Cryst Growth 2013;380:169–75.
- [41] Chandramohan S, Sathyamoorthy R, Sudhagar P, Kanjilal D, Kabiraj D, Asokan K, et al. Influence of SHI irradiation on the structure and surface topography of CdTe thin films on flexible substrate. J Mater Sci: Mater Electron 2007;18:1093–8.
- [42] Studenyak I, Kranjčec M, Kurik M. Urbach rule in solid state physics. Int J Opt Appl 2014;4:96–104.
- [43] Chander S, Purohit A, Lal C, Dhaka MS. Enhancement of optical and structural properties of vacuum evaporated CdTe thin films. Mater Chem Phys 2017;185:202–9.
- [44] Shaaban ER, Yahia IS, Afify N, Salem GF, Dobrowolski W. Structural and the optical dispersion parameters of nano-CdTe thin film/flexible substrate. Mater Sci Semicond Process 2014;19:107–13.
- [45] Aris KA, Rahman KS, Ali AM, Bais B, Yahya IB, Akhtaruzzaman Md, et al. A comparative study on thermally and laser annealed copper and silver doped CdTe thin film solar cells. Sol Energy 2018;173:1–6.