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CHARACTERIZATION OF VACUUM FLASH EVAPORATED CdTe THIN FILMS FOR SOLAR CELLS APPLICATION

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CdTe thin films were deposited by vacuum flash evaporation technique on commercial glass substrates at 100^0C, 175^0C and 250^0C temperatures. XRD results have shown that all samples exhibit a predominant sharp peak at 2θ around 23.8^0 which is assigned to the (111) plane of zinc blend cubic structure. It was found that the intensity of this peak (111) increases with decreasing the process temperature. Increase the process temperature leads to the appearance of two new diffraction peaks at 2θ around 39.3^0 and 46.3^0 positions corresponding to reflection from planes (220) and (311) which indicates the polycrystalline structure of these films. Average grain size calculated from XRD results and found to be 53.9 nm, 42.1nm and 57.4 nm for the films deposited at 100^0C, 175^0C and 250^0C substrate temperature, accordingly. AFM images have shown the surfaces are very smooth with the roughness less than 25 nm. Substrate temperature increase leads to increase the grain size, particularly the grains of maximum size 120 nm increased up to 160 nm (more than 30%).The average grain size calculated from AFM grain distribution diagram fits with the XRD average grain size. CdTe films composition measured by EDXRF analyzer. Revealed that the composition of films very close to the stoichiometric with a small excess of tellurium. The transmission and reflection spectrum of CdTe thin films measured in the range 400–1000nm and used to evaluate the band gap E_g. All the films have E_g (1.49 eV - 1.50 eV) correspond to the band gap of a bulk crystal. The absorption coefficient spectra used to evaluate Urbach tail. It was established that width of tail depends on the process temperature. The smaller values of tail width 31 meV and 64 meV which characterize the high perfection of crystalline structure have films deposited at 100^0C and 250^0C, accordingly.

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1. Introduction

In last decade cadmium telluride (CdTe) is of great interest as one of the most promising material for low cost, chemically stabile, large scale and effective solar cells application [1]. Except these advantages CdTe based solar cells have high radiation-firmness that makes them perspective for space use as well.

CdTe has been considered as one of the most attractive semiconductor absorber layer in thin film solar cells due to its optimal direct band gap 1.5 eV at room temperature and high (more than $5 \cdot 10^4$ cm^{-1}) adsorption coefficient. A theoretical limit of one-junction CdTe solar cells is more than 30% [1] for non concentrated sunlight of AM1.5.

Characteristics of CdTe thin films play an important role in the photovoltaic conversion and strongly depend on the applied deposition technique. Various techniques have been reported for deposition of CdTe thin films such as physical vacuum deposition [2-4], electro deposition [5], molecular beam epitaxy [6], metal-organic chemical vapor deposition [7], closed-space sublimation (CSS) [8], and screen-printing [9] techniques. According to First Solar, Inc. reports

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they broke record and achieved research cell efficiency of 21.5% and total area module efficiency of 18.2% [10], but the highest published in scientific journals efficiency of 16.5% is obtained by using CSS technology [11]. In spite of high value of efficiency the main disadvantage inherent to CSS technology related with the high (650-700^{0}C) temperature of deposition process. It requires high power consumption of process and expensive special heat-resistant glass substrates [12].That is why the further development and study of low-temperature methods of high-quality CdTe layers deposition are actual not only from the standpoint of reducing the cost of solar cells due to reducing the power consumption, but also from the standpoint of use of these techniques for production of solar cells on flexible polymer substrates. As such substrates can be used polyimide films allowing heating up to 400-450^{0}C, for example polymer films of the company DuPont [13].

Flexible solar cells permit to significantly (more than 95%) reduce the weight of the solar module compared with the same one on the glass substrate. This drastically increase the power produced by unit weight of solar module which makes them in demand for space application, especially due to the radiation firmness of CdTe. They are of great interest also for different terrestrial applications such as military application and power supply of low-power portable devices.

Among the promising low temperature techniques for manufacturing of CdTe solar cells are more suitable the magnetron spattering [14,15] and physical vacuum evaporation techniques [16,17].The best results were obtained by the method of vacuum thermal evaporation. This method allows performing a precise control of the deposition process and provides the ability to fully automate in-line manufacturing of large area solar cells.

However by means of this technique it is difficult to deposit the films of II-VI compounds of strictly stoichiometric composition due to the partial pressures difference of the II and VI group components. From this point of view is more acceptable the method of the discrete vacuum thermal evaporation which is the version of the thermal vacuum evaporation known in literature also as the flash evaporation technique. In spite of the mentioned advantages of flash evaporation technique a very limited investigations dedicated to application this technique for deposition of chalcogenide thin films and investigation their characteristics [18-22].

As it is described in work [22] the stoichiometric composition of chalcogenide films is achieved because the deposition is performed by discrete evaporation of small portions of alloy to be deposited. Small particles of crushed chalcogenide alloy whose constituents have different vapor pressure fall down into the preheated flash exchanger. Since the particles have different sizes and fall down in a random way at each moment in the flash exchanger there are some amounts of particles which have different temperature and are at different evaporation stages. Simultaneous presence of such particles in the flash exchanger provides at the average the same fluxes of evaporated constituents and hence the stoichiometric composition of deposited film with pinpoints accuracy. In [22] this method has been successfully used particularly for deposition of CdS films. As it has been shown, these films are characterized by high perfection of the crystal structure and have the band gap coincides with the band gap of a bulk material.

In this work we applied vacuum flash evaporation technique to deposit CdTe thin films on glass substrate and investigated the substrate temperature effects on the stoichiometry, structural, morphological and optical characteristics of CdTe films which act as an absorbing and hetero-layer in chalcogenide solar cells.

2. Experimental details

Thin films of CdTe were fabricated by flash evaporation technique on commercial glass slides used as substrate with thickness 1 mm and diameter 20 mm. The substrates were thoroughly cleaned by soap-free detergent ultrasonically and then iteratively rinsing in distilled water to remove the traces of detergent. Thereafter the substrates were cleaned ultrasonically in ethanol and then dried by blowing air.

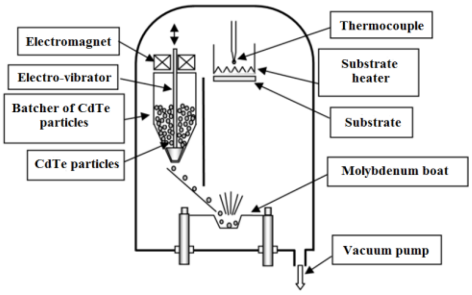


Fig.1. Schematic representation of the experimental setup

For deposition of CdTe films was used the same experimental flash evaporation setup as described in [22]. The schematic view of this setup is presented in Fig. 1.The source material was CdTe grains size of 2 - 6 mm and 99.999% of purity (purchased from Sigma-Aldrich Chemical Co., USA), which were crushed to a powder with particle dimensions of 50 μm –150 μm and then were placed inside a batcher. The vacuum chamber before evaporation was pumped to a base pressure of $1.5{\cdot}10^{-5}$ Torr.

Dispensing the CdTe particles from batcher onto the molybdenum flash exchanger was performed due to opening and closing of batcher outlet by the instrumentality of electro-vibrator (Fig.1).

The amount of CdTe particles falling into the flash exchanger was controlled by opening time of outlet. The outlet is opened when electro-vibrator attracts up by electromagnet. In our experiments the opening time was varied from 0.2 to 0.5 c. The outlet is opened again for evaporation of a next new portion of CdTe particles only after evaporation of all particles contained in the flash exchanger. The evaporation process monitored through the chamber's window and the pressure change in vacuum chamber caused by evaporation of CdTe.

Dispensing the CdTe particles was started after the flash exchanger is heated up to 700^0C - 800^0C temperature suitable to evaporate the CdTe powder. The distance between the flash exchanger and substrate was about 12 cm.

The film thickness and the duration of deposition were typically 1-2 μm and 20-40 min. The CdTe films were deposited at 100^0C, 175^0C and 250^0C substrate temperatures. The structural properties of samples were studied by X-ray diffractometer URD-6 in the θ-2θ mode using Cu - K_α (λ = 1.5405 Å) radiation. The surface morphology and roughness of CdTe films were investigated by atomic force microscopy (AFM) type NEXT supplied by NT-MDT Inc. Transmittance and reflectance, over the wavelength from 400 to 1000 nm, were measured using double beam Filmetrics F20 spectrophotometer.

The composition of deposited CdTe films was measured by ARLQUANT'X EDXRF analyzer.

3. Results and discussion

3.1. Structural analysis of CdTe thin films

Fig. 2 shows the X-ray diffraction patterns of CdTe thin films deposited by flash evaporation technique on a glass substrate. Diffraction patterns were recorded over 2θ range from 15^0 to 70^0 with the step 0.1^0. All CdTe films deposited at 100^0C, 175^0C and 250^0C temperatures exhibit a predominant sharp peak at 2θ around 23.8^0 which is assigned to the (111) plane of zinc blend cubic structure [ASTM 15-770]. These diffraction patterns shown that the intensity of predominant (111) orientation increases with decreasing the process temperature and is in good agreement with the same results observed in [23]. Increase the process temperature leads to the appearance of two new diffraction peaks at 2θ around 39.3^0 and 46.3^0 positions corresponding to reflection from planes (220) and (311) which indicates the polycrystalline structure of these films. The same diffraction peaks have been observed in many works for CdTe films prepared by thermal evaporation [24-26]. The interplanar spacing (d) and lattice parameter (a) for cubic crystalline structure were calculated using Bragg's diffraction law [27]:

$$2d \cdot \sin\theta = \lambda$$

and relation:

$$a = d \cdot \sqrt{h^2 + k^2 + l^2}$$

where λ is the wavelength of the X-ray used (λ=1.5405 Å), θ is the Bragg's angle and h,k,l are Miller's indices.

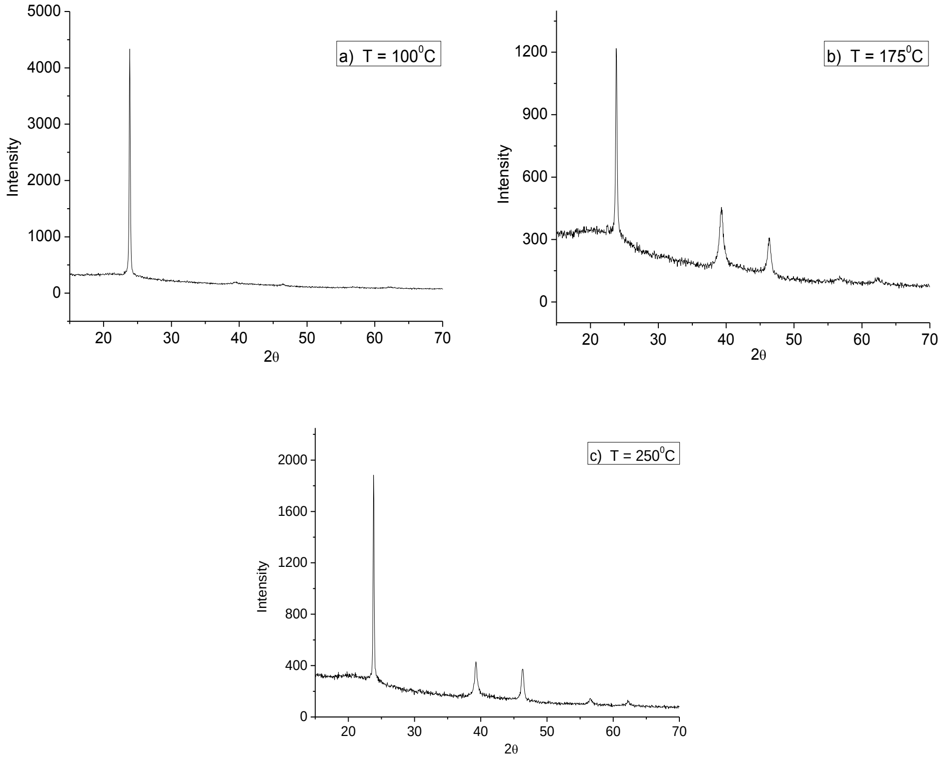


Fig. 2. XRD patterns of CdTe films deposited at substrate temperature: a) 100^{0}C, b) 175^{0}C, c) 250^{0}C

The summery of XRD data of CdTe thin films at different substrate temperatures are presented in Table 1. The results obtained within the accuracy of the measurements (0.1^0) are in good agreement with standard data of interplanar spacing (d) and lattice parameter (a). It is seen from Table 1 that with increasing temperature observed values of d and a approximate to the standard values.

Table 1. XRD data of CdTe thin films at different substrate temperatures

Substrate temperature	2θ, (^{0})	d (Å) observed from XRD	d (Å) standard CdTe ASTM 15-770	[hkl] plane	a(Å) observed values	a(Å) standard CdTe ASTM 15-770
T=100^{0}C	23,85	3,726	3,742	111	6,454	6,481
T=175^{0}C	23,8	3,734	3,742	111	6,467	-
	39,3	2,289	2,290	220	6,474	-
	46,3	1,958	1,954	311	6,493	-
T=250^{0}C	23,8	3,734	3,742	111	6,467	-
	39,25	2,292	2,290	220	6,482	-
	46,3	1,958	1,954	311	6,493	-

The average grain sizes (D) of the film was calculated using the Scherrer's equation [28]:

$$D = \frac{0.9\lambda}{\beta\cos(\theta)}$$

where λ is the wavelength of the X-ray used (λ=1.5405 Å), β is the full-width at half-maximum (FWHM) of the (111) peak which has maximum intensity, and (θ) is the Bragg's angle.

The variation of the grain size with substrate temperature is presented in Table 2. It is seen that the grain size vary with increase the temperature non- monotonically. At first with increase the process temperature from 100^0C to 175^0C the grain size decreases from 53.9 nm to 42.1nm then from 175^0C to 250^0C increases from 42.1 nm to 57.4 nm. The same tendency, viz minimum value of (111) peak intensity is observed at substrate temperature 175^0C.

The other structural parameters of CdTe thin films such as dislocation density (δ), the number of crystallites per unit area (N) and micro-strain (ε) for comparison are presented in Table 2.

Table 2. Structural parameters of CdTe films deposited at substrate temperatures 100^0C, 175^0C and 250^0C

Substrate temperature	2θ (^{0})	FWHM (deg)	D (nm) from XRD results	[hkl] plane	δ 10^{10} lines/cm^2	ε x10^{-4}	N x10^{11} cm^{-2}
T=100^0C	23,85	0,184	53,9	111	3,44	6,43	3,19
T=175^0C	23,8	0,24	42,1	111	5,64	8,23	6,69
T=250^0C	23,8	0,176	57,4	111	3,03	6,03	2,64

The dislocation density δ defined as the length of dislocation lines per unit volume has been calculated using the equation [29]:

$$\delta = \frac{1}{D^2}$$

The number of crystallites per unit area (N) and the micro-strain (ε) of the films have been estimated with the use of the following equations [30]

$$N = \frac{1}{D^3}$$

$$\varepsilon = \frac{\beta\cos(\theta)}{4}$$

The smallest values of dislocation density, number of crystallites and micro-strain which characterize the structural performance of CdTe have thin films deposited at higher temperature 250^0C. Note that here also the relatively low crystalline perfection have CdTe films deposited at 175^0C.

CdTe films compositions were measured by EDXRF analyzer. It has been found, that the compositions of films very close to the stoichiometric with a small excess of tellurium. The results of measured elemental composition of CdTe films are presented in Table 3.

Table 3. Elemental composition of CdTe thin films deposited at substrate temperatures 100^0C, 175^0C and 250^0C by vacuum flash evaporation technique

Element	Substrate temperature								
	T=100^0C			T=175^0C			T=250^0C		
	Weight %	Atomic %	Std. Err	Weight %	Atomic %	Std. Err	Weight %	Atomic %	Std. Err
Te	51,8	48,74	0,67	54,57	51,4	0,69	54,58	51,34	0,69
Cd	44,26	47,28	0,21	45,22	48,35	0,21	45,1	48,21	0,21
Ba	2,31	2,01	0,99	-	-	-	-	-	-
Px	1,53	1,83	0,06	0,154	0,185	0,031	0,278	0,33	0,027
Se	0,056	0,09	0,005	0,031	0,049	0,005	0,036	0,05	0,005
Bi	0,047	0,02	0,009	0,02	0,012	0,008	-	-	-

Despite the fact that for thin film deposition as a source material we used high-purity (99,999%) CdTe, nevertheless results of measurements detect small amount of such elements as: Ba, Px, Se, Bi. The detected amount of these elements is considerably exceeding their concentration in CdTe source material and we assume the appearance of these elements can be related with their presence in glass substrate.

3.2. Optical analysis of CdTe thin films

The transmittance and reflectance of CdTe films deposited on glass substrate have been measured at room temperature in regard to the same clear glass substrate. Using the measured transmittance (T) and reflectance (R) spectra the absorption coefficient (α) has been calculated by the following equation:

$$\alpha = \frac{1}{t}\ln\frac{1-R}{T}$$

where *t* is the thickness of CdTe film.

The calculated relation between the absorption coefficient and the incident photon energy $\hbar\omega$ for CdTe thin films deposited at different substrate temperatures are shown in Fig.3.

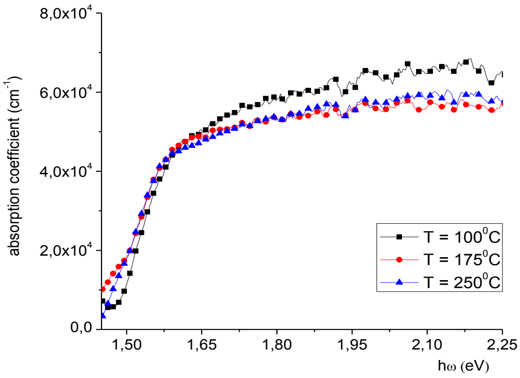


Fig.3. The plot of the absorption coefficient α v.s. photon energy (ħω) of CdTe thin films deposited at 100^oC, 175^oC and 250^oC substrate temperatures

Above the fundamental absorption edge the dependence of the absorption coefficient on the incident photon energy is given by Tauc's model [31]:

$$\alpha \cdot \hbar\omega = B\left(\hbar\omega - E_g\right)^n$$

where ħω is the photon energy, E_g is the optical bandgap, B is a constant and n is an exponent that depends on the type of optical transitions. For direct allowed transitions $n = ½$. The variation of $(\alpha \cdot \hbar\omega)^2$ of CdTe thin films deposited at different substrate temperatures as a function of photon energy are shown in Fig. 4.

Using the liner extrapolation method the value of the optical bandgap for CdTe thin films is determined to be 1.50eV, 1.49 eV and 1.49 eV for 100°C, 175°C and 250°C substrate temperature, accordingly. The thin film of CdTe deposited at 100°C has the same optical bandgap as a CdTe bulk crystal [32]. The same value 1.5 eV, for instance, has been obtained in [33] for nearly stoichiometric CdTe film deposited on a glass substrate.

These results shown, that the substrate temperature affects negligibly on the bandgap of deposited CdTe films and are within the accuracy of measurements ±0.01 eV.

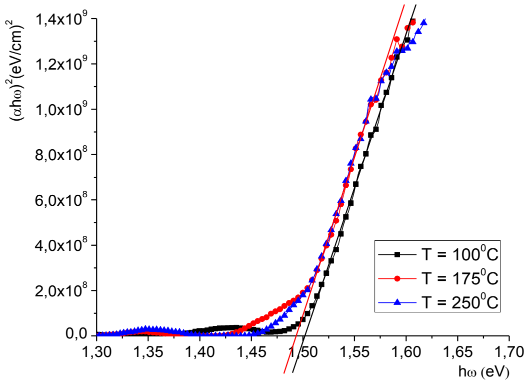


Fig.4. Plots of $(\alpha \cdot \hbar\omega)^2$ *v.s. photon energy (ħω) of CdTe thin films deposited at* 100^0C, 175^0C *and* 250^0C *substrate temperatures.* $E_g = 1.49eV$ *for* 175^0C *and* 250^0C *and 1.5eV for* 100^0C.

Increase of substrate temperature results in decrease of the bandgap. The same regularity was observed in [34,35] for vacuum evaporated CdTe thin films.

In the range of photon energy lower the bandgap the dependence of absorption coefficient from incident photon energy (Fig.3) can be presented by the Urbach rule given by [36-39]:

$$\alpha(\omega) = \alpha_0 \cdot e^{\frac{\hbar\omega}{E_e}}$$

where α_0 is a constant, E_e denotes an energy which is often interpreted as the width of the tail of localized states in the bandgap.

Urbach tails in optical absorption near of band edges related to disordered structure due to departure from the stoichiometry and imperfections of crystalline structure.

Fig. 5 displays the dependence of $\ln\alpha = \ln\alpha_o + \hbar\omega/E_e$ against the photon energy ħω for the three substrate temperatures. In this plot are shown the fit of liner extrapolation carried out for linear region.

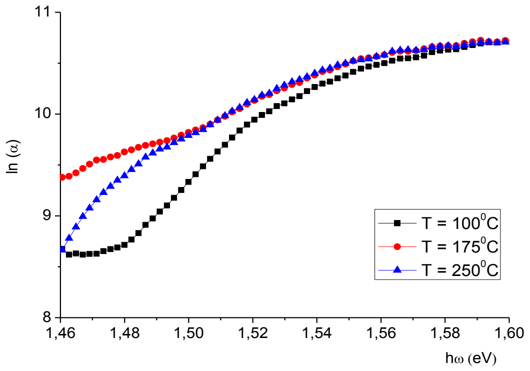


Fig. 5. The plot of ln(a) against the photon energy ħω for CdTe thin films

The calculated value of E_e varied in the range from 31 meV to 84 meV and is presented in Table 4. The higher value of E_e has CdTe films deposited at 175^0C and related to the imperfections of crystalline structure. These results correlate with the XRD results summarized in Table 2 that also confirm a low crystalline perfection of CdTe films deposited at 175^0C.

Table 4. The tail width E_e calculated from the liner fit of the ln(α) v.s. ħω

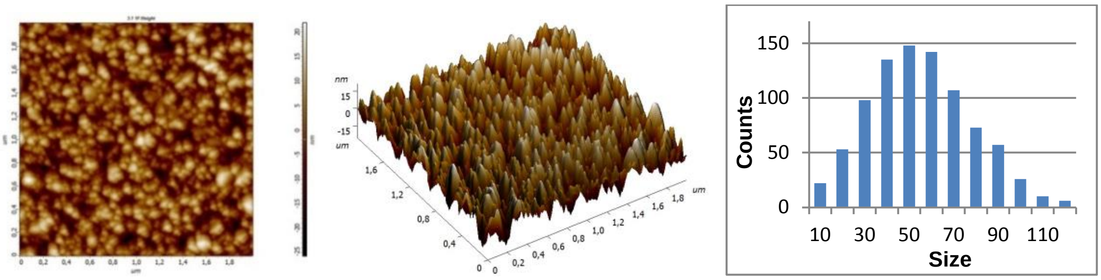
Substrate temperature	E_e (meV)
$T=100^0C$	31
$T=175^0C$	84
$T=250^0C$	64

The small values of E_e (31 to 84 meV) testify that CdTe films deposited by flash evaporation technique have high crystalline perfection and in order of magnitude agree with the E_e values (81–91meV) of vacuum evaporated CdTe thin films deposited at ambient temperature [24].

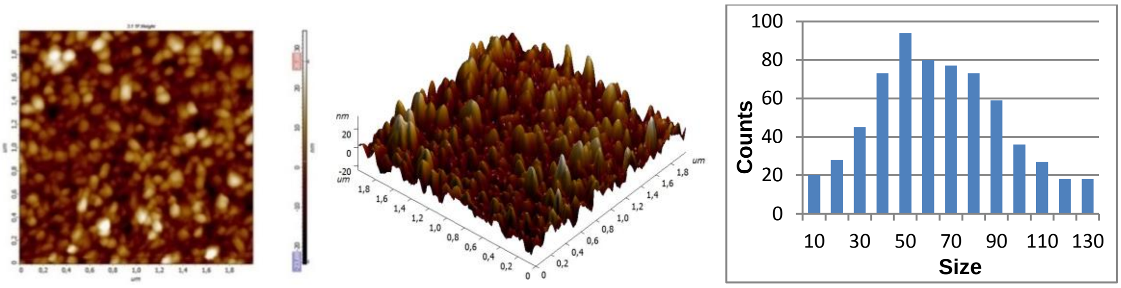
3.3. Surface morphology analysis of CdTe thin films

Fig. 6 shows typical 2μm x 2μm AFM images of CdTe films deposited on a glass substrate at different temperatures by flash evaporation technique. For statistics a few AFM images of different sites of the surface of each sample were investigated. The analysis of AFM images as well as the XRD results (Table 2) show that average grain size depends on the process temperature. From grain distribution diagram it is observed that the increase of substrate temperature leads to increase the number of grains having maximum size, consequently leads to increase the average grain size. With the increase of substrate temperature from 100^0C to 250^0C maximum grain size increases from 120 nm to 160 nm (more than 30%).

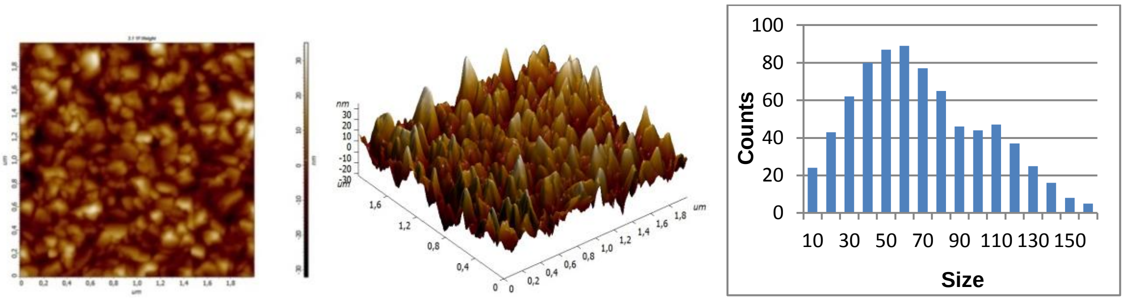
The average grain sizes calculated from grain distribution diagram (Fig.6) fit with the average grain sizes calculated from XRD results (Table 2).



a) Substrate temperature 100^{0}C



b) Substrate temperature 175^{0}C



c) Substrate temperature 250^{0}C

Fig.6. A typical AFM images and the distribution of grain size of CdTe thin films deposited on a glass substrate at (a) 100^{0}C, (b) 175^{0}C and (c) 250^{0}C temperatures.

4. Conclusions

The structural, optical and morphological properties of CdTe thin films deposited on glass substrate at different temperatures (100^{0}C, 175^{o}C and 250^{o}C) by vacuum flash evaporation technique have been investigated. XRD results have shown that all CdTe thin films have zinc blend cubic structure and exhibit a predominant sharp peak at 2θ around 23.8^0 which is assigned to the (111) plane. It is found that decreasing the process temperature leads to increases the intensity of diffraction peak (111). At the same time it is found that increase the process temperature leads to the appearance of two new diffraction peaks at 2θ around 39.3^0 and 46.3^0 positions corresponding to reflection from planes (220) and (311) which indicates the polycrystalline structure of these films. It is determined that the average grain size calculated from XRD results vary with increase the temperature non-monotonically. At first with increase the process temperature from 100^{0}C to 175^{0}C the grain size decreases from 53.9 nm to 42.1nm then with increase the temperature from 175^{0}C to 250^{0}C grain size increases from 42.1 nm to 57.4 nm.

Analysis of XRD results have shown that the smallest values of dislocation density, number of crystallites and micro-strain which characterize the structural performance of CdTe have thin films deposited at higher temperature 250^{0}C.

According to AFM images all CdTe films have very smooth surface with the roughness less than 25 nm. AFM images have shown that increase of substrate temperature promotes to increase the average grain size and the number of grains having maximum size. For instance, with the increase of substrate temperature from 100^0C to 250^0C maximum grain size increases from 120 nm to 160 nm (more than 30%). The average grain size calculated from AFM grain distribution diagram fits with the XRD average grain size.

The results of measurements performed by EDXRF analyzer confirmed that flash evaporation technique provide the composition of deposited CdTe films very close to the stoichiometric composition with a small excess of tellurium.

Optical analyses have shown that CdTe thin films deposited at 100^0C have 1.5eV bandgap and 1.49eV bandgap have films deposited at 175^0C and 250^0C. These values correspond to the bandgap of bulk crystal.

The results of optical measurements used to evaluate Urbach tails. These results have shown that width of tails depend on the process temperature and the smaller values of tail width 31 meV and 64 meV have films deposited at 100^0C and 250^0C, accordingly. The small values of tail testify a high crystalline perfection of CdTe films.

The results of XRD, compositional, optical and AFM measurements have shown that the flash evaporation technique can be successfully used to deposit good quality CdTe thin films for high-efficiency solar cells application.

Acknowledgments

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