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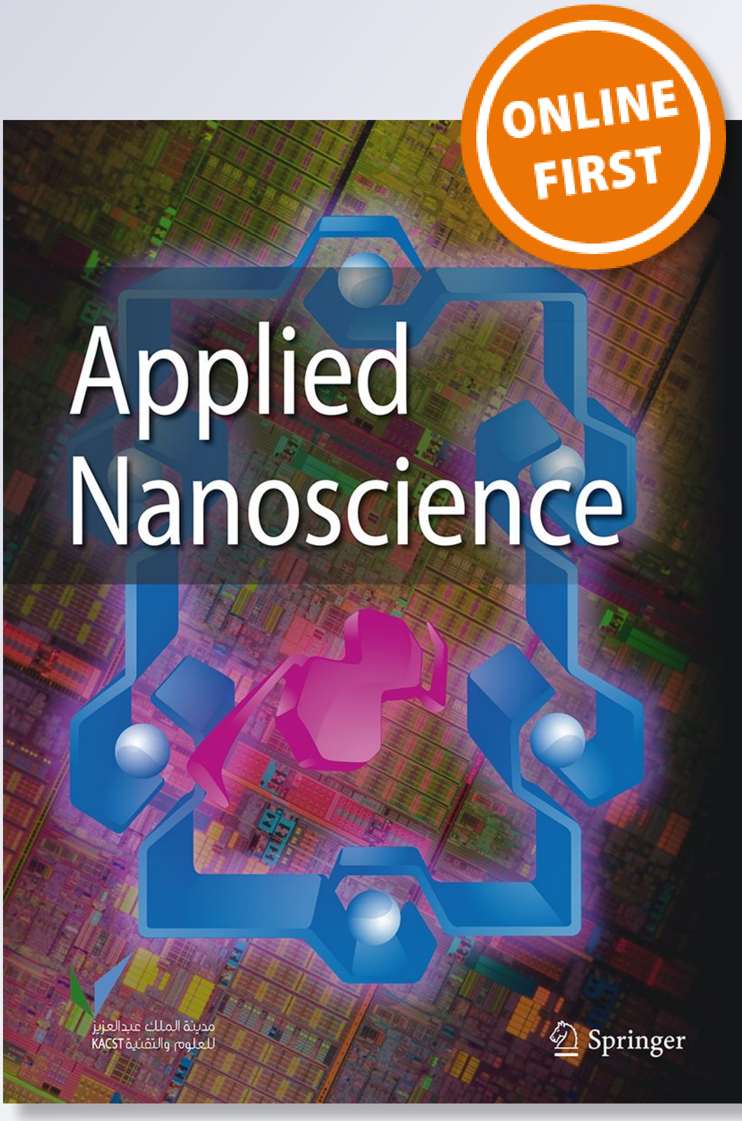

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ORIGINAL ARTICLE

Structural and optical properties of cadmium telluride obtained by physical vapor deposition technique

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Abstract

The structural and optical properties of cadmium telluride (CdTe) thin films prepared by thermal evaporation using physical vapor deposition technique and obtained under different process conditions (different deposition time) were investigated. CdTe thin films were obtained on (100) silicon and glass substrates. The analysis of atomic force microscopy images of surface revealed a certain periodicity of the objects' location on the surface. After detailed analysis by autocorrelation functions, it has been established that the films have a clearly expressed periodicity along the axis OX, which is due to the orientation of growth on (100) Si substrate. The optical properties indicate a smooth film with nanoparticle size (25–28) nm on surface for both thin and thick films of CdTe. The optical transparency is observed in the short infrared radiation. An analysis of the optical properties was performed using the Swanepoel method. Hence, the basic properties of the films such as refractive index, film thickness, absorption coefficient and optical conductivity are determined. The optical transparency is observed in the short infrared radiation.

Keywords Thin films · Cadmium telluride · Swanepoel method · Thermal evaporation

Abbreviations

CdTe	Cadmium telluride
CIS	Copper indium selenide
CIGS	Copper indium gallium di-selenide
CdS	Cadmium sulfide
PV	Photovoltaic
PVD	Physical vapor deposition
CSS	Close spaced sublimation
PLD	Pulsed laser deposition method
RF	Radio frequency
VTD	Vapor transport deposition
MOCVD	Metal-organic chemical vapor deposition
AFM	Atomic force microscope
SEM	Scanning electron microscope
EDS	Energy-dispersive X-ray spectroscopy

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Introduction

The problem of alternative energy sources is the highest priority area of modern research, due to fossil resources limits. Renewable energy technologies produce sustainable, clean energy from sources such as the sun, the wind, plants, and water (Ruvinskii et al. 2016; Dresselhaus and Thomas 2001; Ismail and Ahmed 2009; Barnhart et al. 2013; Prokopiv et al. 2017). Photovoltaic (PV) energy conversion plays an important role in advancing energy solutions (Shah et al 1999). There are three main generations: silicon, thin film and organic solar cells (Hamakawa 2013; Wisz et al. 2017). At present, the most commonly used solar cells are photovoltaic elements based on silicon technologies. An alternative to these converters is thin-film solar cells designed to reduce the cost of solar modules and increase production using straight-line semiconductors, deposited on a cheap substrate of large areas (glass, metal foil, plastic) (Aramoto et al. 1997; Dobson et al. 2000). The straight-line semiconductor is capable to absorb solar radiation at a thickness of the layer two orders of magnitude lower than the thickness of the silicon cells. Using materials with higher optical absorption coefficient, it is possible to reduce the thickness of photovoltaic elements (Alvarez et al. 1997). It decreases the costs of production. Moreover, it is possible to reduce the weight of photovoltaic devices depending on the used substrate. As a result, using the thin-film semiconductor materials, such as CIS, CIGS, CdTe and CdS, and suitable manufacturing technologies, enable to obtain PV devices at relatively low cost, good electrical performance, low weight and high flexibility (Znajdek et al. 2014).

Cadmium Telluride is one of the promising materials for producing low-cost thin-film photovoltaic second-generation devices (Mahabaduge et al. 2015). CdTe is II–VI compound semiconductor with a direct optical bandgap of ~1.5 eV that is optimally matched to the solar spectrum for PV energy conversion (Wu 2004). The use of cadmium telluride as thin-film material for solar cells has attracted wide attention due to its high availability, low cost, and simplicity in the large-scale manufacture. The theoretical value of efficiency is about 33% (Polman et al. 2016) and the practical efficiency is about 22% of the thin-film photovoltaic materials (Gloeckler 2016).

In recent years, the interest for this material has significantly grown. Worth to note that by 2010 such thin-film elements had an efficiency not exceeding 17%, and in the previous 7 years, the efficiency had risen to 22.5% (Gloeckler 2016). This is due to the fact that the big producers of solar cells are focused on reducing the cost of such solar panels. The cost was ~1$/Wp at 16.7% (2001), decreased to ~0.75$/Wp at 22.1% (2016) and First Solar Inc. predicts efficiency about ~25% achievable with current cell structure ~0.67$/Wp (Green 2016, Nykyruy et al. 2017).

The PV elements based on CdTe thin films can be deposited by various deposition techniques. It includes physical vapor deposition (PVD) such as close spaced sublimation (CSS) (Krishnakumar et al. 2017), pulsed laser deposition method (PLD) (Bylica et al. 2006), thermal evaporation (Brus et al. 2014; Saliy et al. 2017a, b), RF magnetron sputtering (Jeyadheepan et al. 2015), vapor transport deposition (VTD) (Gu et al. 2017), metal-organic chemical vapor deposition (MOCVD) (Kartopu et al. 2015), electrodeposition (Diso et al. 2016), and the method of chemical transport reactions (Il'chuk et al. 1999).

The method of thermal evaporation by physical vapor deposition technique has advantages for several reasons. In particular, the technological simplicity of the device structure makes this method very cheap and simple, which, finally, optimizes the production technological lines and reduces the cost of photovoltaic cells.

A few number of technological parameters, which are set during deposition, significantly reduce the resulting errors.

Experimental

CdTe thin films were deposited on (100) silicon and glass substrates using the thermal evaporation method by PVD technique. The used installation can be to receive series (5–10 films) in a single cycle for various technological factors: different thickness $d=(0.05\text{–}10)$ µm at a constant temperature of deposition $T_S=(300\text{–}570)$ K; uniform thickness d with different T_S; different temperature of evaporation T_E (600–970) K with constant thickness d or temperature of deposition T_S.

The investigated thin films were obtained with different thicknesses (determined by different deposition time τ) at the same T_S and T_E. The growth temperature T_s was 470 K, and temperature of evaporation of the pre-synthesized compounds CdTe was $T_E=870$ K. The thickness of the thin films was set by deposition time $\tau=(60\text{–}420)$ s. The morphological and structural properties of CdTe thin films were investigated by scanning electron microscope (Vega 3 Tescan) and atomic force microscope (CSM Instruments). Measurements were carried out in the central part of the samples using a number of silicon probes Veeco MNSL-5 with a nominal radius of curvature of the tip up to 5 nm. The film thicknesses d were measured with the 2D Profilometer Bruker Dektak XT.

Optical transmission spectra were measured in the wavelength range of 190–1500 nm using Agilent Technologies Cary Series UV–Vis–NIR Spectrophotometer. The chemical composition of the obtained films was investigated by SEM using the energy-dispersive X-ray spectroscopy (EDS) method.

Results and discussion

The technological properties of obtained CdTe thin films on glass and (100) Si substrates are presented in Table 1. The thin films were obtained in one technological process with different thickness (different deposition time).

Structural properties

The AFM images of morphological properties of CdTe thin films obtained on (100) Si substrates of different thickness can be observed in Fig. 1. Nanoparticles observed on the surface with normal sizes (21–25) nm for the thin films (sample 9), and (22–28) nm for the thick films (sample 6) (Fig. 1). It can be noted that the thickness of films does not significantly affect on the change of morphology and surface nanoparticles.

Also, for the thin films, there is a certain periodicity of film growth along the Ox axis, as it can be seen from the

Table 1 Technological modes of CdTe thin films

Number of samples	Substrates	Temperature of substrate T_S, K	Evaporation temperature T_E, K	Deposition time τ, s	Thickness d, nm
1	Glass	470	770	210	540
2		470	820	210	2930
3		470	820	420	3915
4		470	870	210	1620
6	Silicon (100) glass	470	820	300	2835
					2760
7		470	820	240	1755
					1620
8		470	820	180	810
					675
9		470	820	120	540
					540
10		470	820	60	270
					270

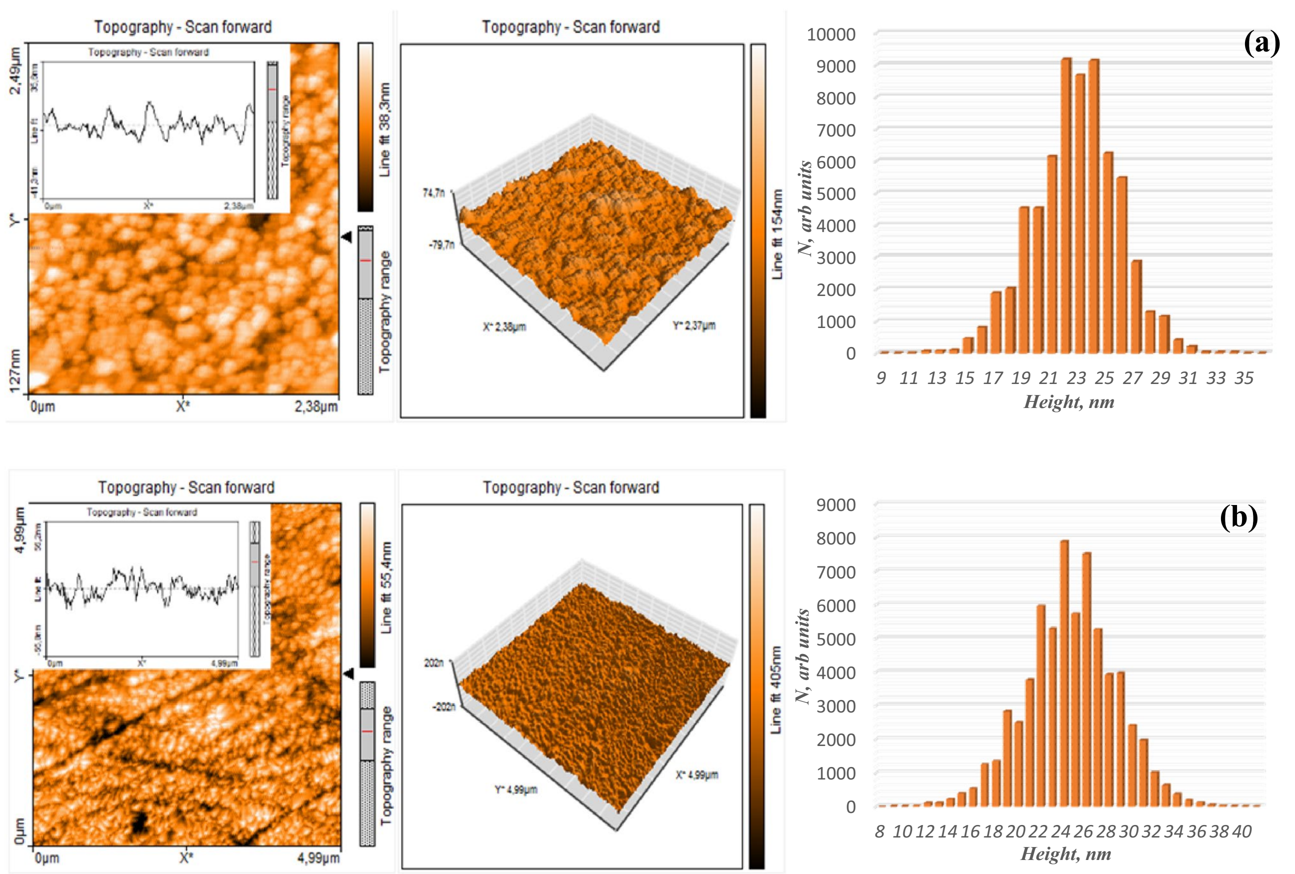


Fig. 1 AFM images of CdTe/(100)Si deposited at the different deposition time τ: **a** sample 9; **b** sample 6

autocorrelation functions (Fig. 2). The analysis of periodicity of grown for these CdTe thin films has been studied in detail using autocorrelation function and Fourier transformation (Saliy et al. 2017a, b; Saliy et al. 2017). This is primarily due to the orientation of growth on the (100) Si substrate (Bera and Saha 2016; Sagan et al. 2005).

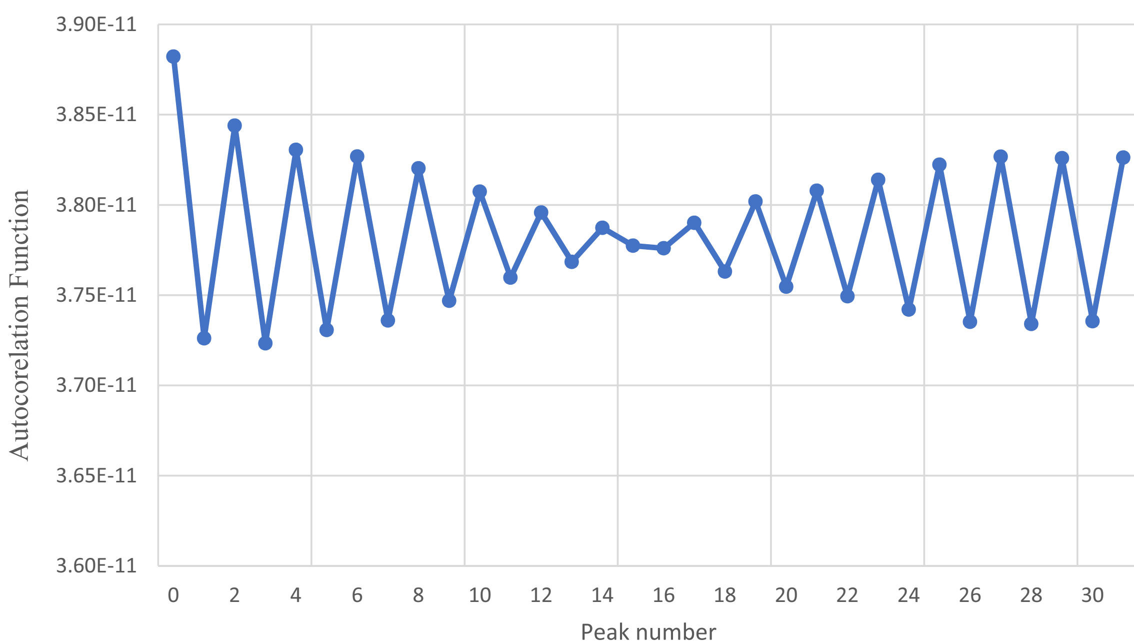


Fig. 2 The values of two-dimensional autocorrelation function along coordinate axes: $x=$var, $y=$const [sample 6/(100)Si]

From the cross-sections (Fig. 3), we see that the surface is smooth, and the growth comes with the formation of nanowire inside the film. Such nanowires make a significant contribution to the absorption of light in photovoltaic elements.

For CdTe/(100)Si sample, the chemical composition analysis was provided on SEM (Bruker Quantax) using the energy-dispersive X-ray spectroscopy (EDS) method. The assayed EDS analysis was carried out in the low-vacuum mode. In particular, for the sample 7 the atomic weight percent consists of Cd 47.63% and for Te 49.29%. Si 3.08% as basic element of the substrate was obviously observed in the spectrum (Fig. 4). From the sequential experiments, the conditions can be determined for the deposition of stoichiometric CdTe semiconductor films with a small tellurium excess using the vapor-phase condensation method.

Optical properties

The spectral distribution of optical transparence was carried out for the identification of CdTe thin films. The region of

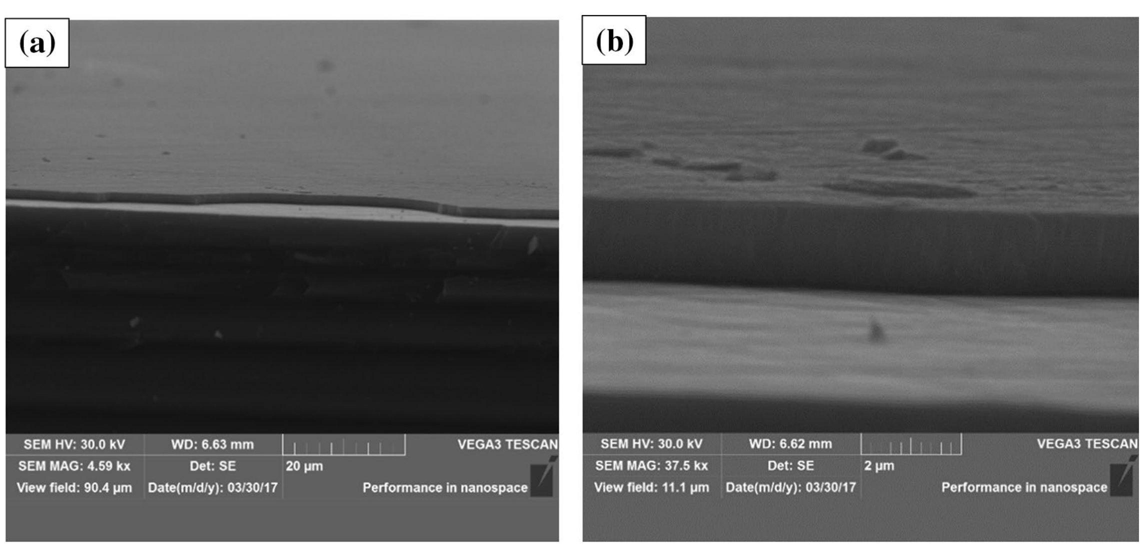
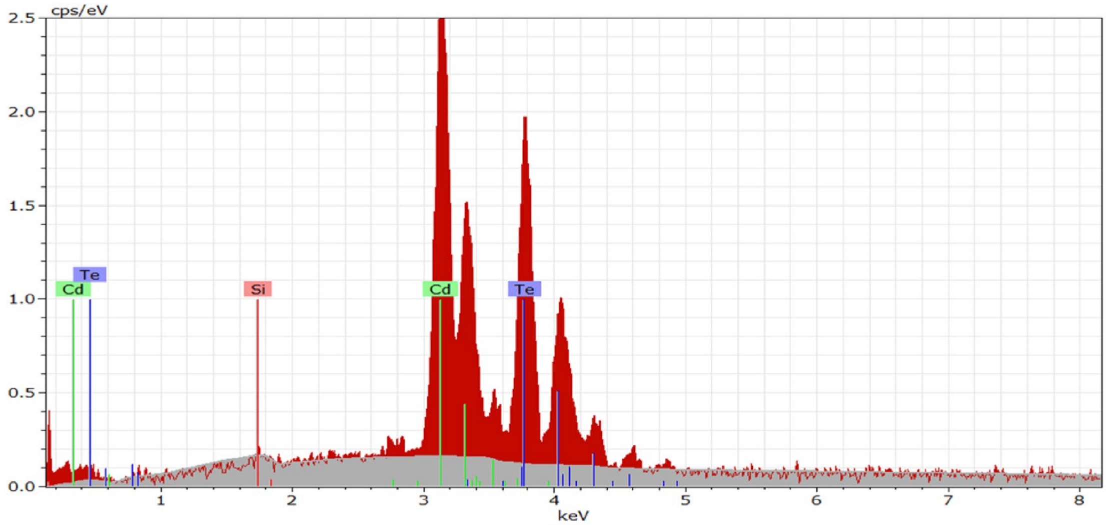


Fig. 3 The cross-sectional images of CdTe/(100) Si thin films (sample 7): **a** in scale 20 µm; **b** in ×10 zoom



Element	AN	series	Net	[wt.%]	[norm. wt.%]	[norm. at.%]	[norm. wt.%]	Error in wt.% (3 Sigma)
Silicon	14	K-series	134	0,65292	0,73646	3,075935	0,73646	0,29678
Cadmium	48	L-series	10936	40,46918	45,64712	47,63420	45,64712	4,08691
Tellurium	52	L-series	8353	47,53450	53,61642	49,28986	53,61642	4,54436
			Sum:	88,65661	100	100		

Fig. 4 The EDS spectrum of CdTe/(100) thin films (sample 7)

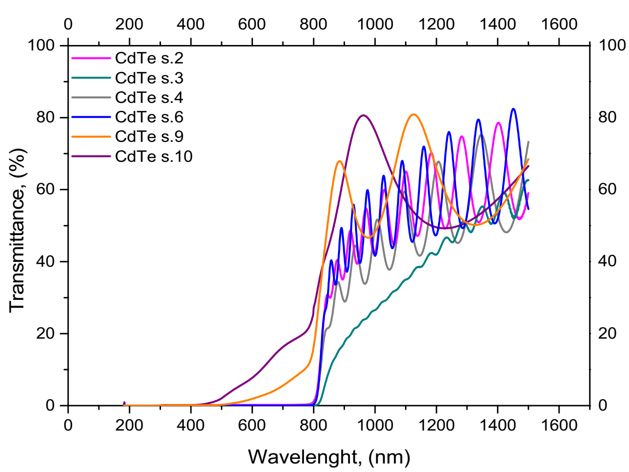


Fig. 5 Optical transmission of the CdTe/glass thin films

fundamental absorption was observed in transmission spectra. The transmission spectra of CdTe thin films obtained on glass substrates with different thickness were measured in the wavelength range from 180 to 1500 nm and shown in Fig. 5. It can be observed that the films are highly transparent in the near infrared region and the average transmittance varies from 57 to 80%. Absorption edge is about 800 nm for all samples. In addition, the observed interference patterns in the optical transmission spectra are the indication for the thickness homogeneity of deposited films. These results are also confirmed by structural analysis. This could be explained by the fact that there is a difference between the refractive indexes of the film and substrate and also due to the interference of multiple light reflections (Moshfegh et al. 2005; Punitha et al. 2014). One very useful method that uses these interference fringes to determine the optical properties of the material is the Swanepoel method (Swanepoel 1983; Shaaban et al. 2012).

To obtain the value of band gap, the absorption coefficient was calculated from the transmission data using the following relation (Tauc 2012):

$$\alpha = \frac{\ln(1/T)}{d}, \tag{1}$$

where ''d'' is the film thickness and ''T'' is the transmission. For the direct transition, the optical band gap energy of the CdTe thin film was determined using the equation:

$$(\alpha h\nu)^2 = A(h\nu - E_g), \tag{2}$$

where A is a constant, $h\nu$ photon energy, and E_g is the optical energy gap.

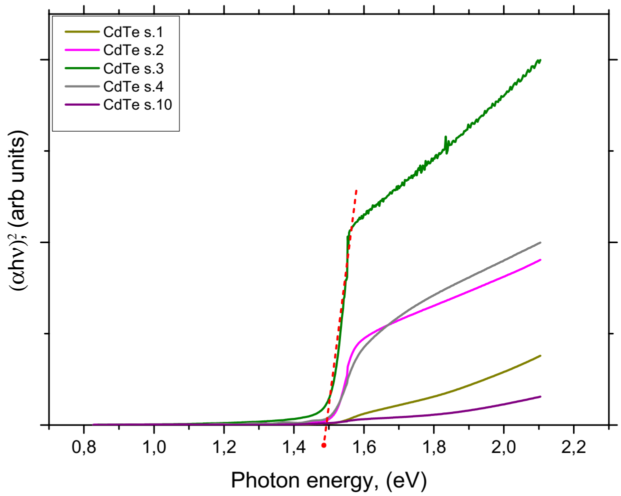


Fig. 6 Optical band gap of the CdTe thin films

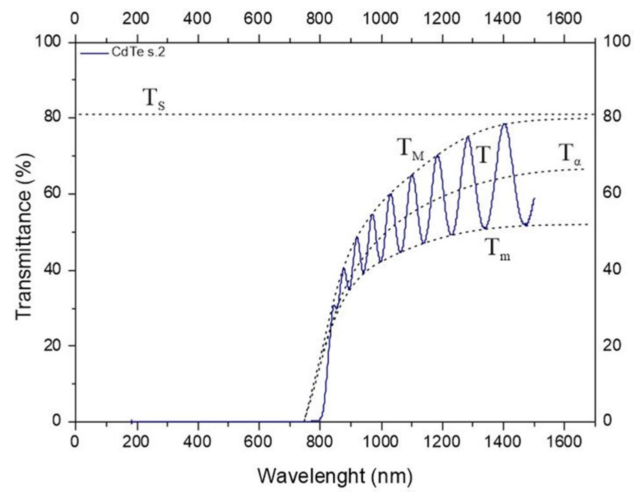


Fig. 7 The optical transmission spectra of the as-deposited CdTe thin film with determined T_M and T_m

Figure 6 shows five graphs of $(\alpha h\nu)^2$ versus $h\nu$ derived from a Tauc formula (Akbarnejad et al. 2016). The direct energy band gap is estimated by extrapolation from the intersection of the linear part at $\alpha = 0$. The optical band gap energy for the freshly grown CdTe thin film was 1.52 eV. Depending on the technological parameters of the obtaining, the extracted optical band gaps of the samples decrease to values between 1.54 and 1.43 eV. These values completely coincide with other works on CdTe band gap (Punitha et al. 2014; Smith and Nie 2010; Schulman and Mcgill 1979).

Swanepoel method

The application of this method entails, as a first step, the calculation of the maximum T_M (λ) and minimum T_m (λ) transmission envelope curves by parabolic interpolation (Swanepoel 1983) to the experimentally determined positions of peaks and valleys (Fig. 7).

$$T = T(\lambda, s, n, d, \alpha), \tag{3}$$

where λ—wavelength, s—substrate refractive index, n—refractive index of CdTe, d—thickness, and α—coefficient of absorption.

The transmission T for the normal incidence resulted from the interference of the wave transmitted from three interfaces can be written as (Swanepoel 1983)

$$T = \frac{Ax}{B - Cx\,\cos\varphi + Dx^2}, \tag{4}$$

where

$$A = 16n^2 s, \tag{5}$$

$$B = (n+1)^3\left(n + s^2\right), \tag{6}$$

$$C = 2\left(n^2 - 1\right)\left(n^2 - s^2\right), \tag{7}$$

$$D = (n - 1)^3\left(n - s^2\right), \tag{8}$$

$$\varphi = 4\pi nd/\lambda, \tag{9}$$

$$x = \exp(-\alpha d). \tag{10}$$

The extremes of the interference fringes can be written as

$$T_M = \frac{Ax}{B - Cx + Dx^2}, \tag{11}$$

$$T_m = \frac{Ax}{B + Cx + Dx^2}. \tag{12}$$

T_s is the maximum value of the transmission of substrate (Fig. 7). Thereafter, the substrate refractive index s can be given by the following equation:

$$s = \frac{1}{T_s} + \left(\frac{1}{T_s^2} - 1\right)^{1/2}, \tag{13}$$

$$T_m = \frac{4n^2 s}{n^4 + n^2\left(s^2 + 1\right) + s^2}. \tag{14}$$

In the region of weak and medium absorption received $\alpha \neq 0$ and $x < 1$—Subtracting the reciprocal of Eq. (11) from the reciprocal of Eq. (12) yields an expression that is independent of x:

$$\frac{1}{T_m} - \frac{1}{T_M} = \frac{2C}{A}. \tag{15}$$

From Eqs. (5–10) and Eq. (15) the refractive index is:

$$n = \left[N + (N^2 - s^2)^{1/2}\right]^{1/2}, \quad (16)$$

where

$$N = \frac{2s(T_M - T_m)}{T_M T_m} + \frac{(s^2+1)}{2}. \quad (17)$$

Using the Eqs. 16 and 17, and taking $s = 1.46$, the refractive index of CdTe sample in the range of fitted curves (Fig. 7) can be obtained which is presented in Fig. 8. This equation can be only use for interference zone. With increasing wavelength, the refractive index of CdTe thin film is increased from 2.45 (for the sample 4) to 2.95 (the sample 2). Averaging the calculated values we will get 2.55 (Table 2), can be seen that they are completely consistent with the literary ones that matter for CdTe thin films to 2.7 (Madelung et al. 1999).

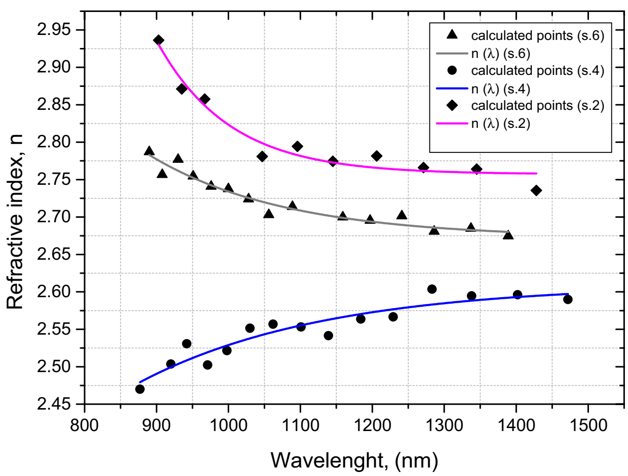


Fig. 8 Refractive index (n) of the CdTe thin films as a function of the wavelength (λ) for the samples 2, 4, and 6

The film thickness can be calculated from the refractive index, using the following equation, where $n(\lambda_1)$ and $n(\lambda_2)$ are the refractive index at two adjacent maxima or minima at λ_1 and λ_2, respectively (Swanepoel 1983):

$$d = \frac{\lambda_1\lambda_2}{2\left(\lambda_1 n(\lambda_2) - \lambda_2 n(\lambda_1)\right)}. \quad (18)$$

The thicknesses of the CdTe thin films for the sample 2 are presented as d_1 in the Table 2. For the samples 4 and 6, the calculated values of thicknesses are 1898 and 3039 nm, respectively. The measured values of the thickness were received using the profilometer method (Table 1). Comparing the experimental and calculated values of the thickness, a small difference in value associated with the error of the thickness calculation can be noted according to Eq. 18.

As it can be seen from Fig. 8, the values of the refractive index $n(\lambda)$ of CdTe thin films (the samples 2 and 6) calculated from the Eq. (16) decrease with the increasing of wavelength. A sharp increase in the refractive index at wavelengths < 1000 nm is due to decrease in the transmission near the edge of own absorption of cadmium telluride thin films. It can be seen that the refractive index depends considerably on the film thickness; for the thin films (sample 4), the refractive index is within 2.47–2.58.

The order of interference is:

$$m = \left[\frac{\lambda_2}{\lambda_1 - \lambda_2}\right], \quad (19)$$

Table 2 Optical parameters (experimental) and calculated results of CdTe thin films for the sample 2

λ, nm	T_M	T_m	n	d_1, nm	m	α, cm^{-1}	σ_{opt}, 10^{11}
1472	0.789	0.518	2.590				
1402	0.786	0.515	2.596				
1338	0.771	0.509	2.595		9.5	507.399	3.100
1283	0.748	0.497	2.604	2817.367	10.5	517.897	3.170
1229	0.720	0.492	2.567	3351.731	11	593.012	3.580
1184	0.702	0.484	2.564	3677.039	11.5	1390.962	8.300
1139	0.668	0.472	2.542	3496.162	12.5	698.014	4.170
1101	0.651	0.461	2.553	3252.864	13	1452.531	8.730
1062	0.625	0.447	2.557	2837.066	13.5	1179.906	7.100
1030	0.600	0.435	2.551	3161.608	14.5	1100.359	6.610
998	0.569	0.423	2.522	3201.433	15.5	1111.593	6.600
971	0.548	0.414	2.502	3001.171	16	2361.357	13.900
942	0.520	0.394	2.531	3125.911	16.5	2147.537	12.800
920	0.487	0.378	2.504	3469.052	17.5	2854.082	16.800
893	0.450	0.351	2.545	3050.406	18	3275.103	19.600
877	0.404	0.329	2.470			4684.632	27.200
				⟨d⟩ 3203.484			

where $\lambda_1$and λ_2 are the wavelengths of two adjacent transmission maxima.

Knowing the following values of the thickness (d) of the CdTe thin films, the refractive index (n), the refractive index substrate (s), and the absorption coefficient of the CdTe material deposited on the glass substrate (α) can be calculated:

$$\alpha = \frac{1}{d}\ln\left[\frac{16n^2 s}{(n+1)^3(n+s^2)T_0}\right]. \tag{20}$$

If $T_0 = Ax/B$, Eq. (20) can be shown:

$$\alpha = \frac{1}{d}\ln\frac{(n-1)^3(n-s^2)}{E_{\mathrm{m}} - \left[E_{\mathrm{m}}^2 - \left(n^2-1\right)^3(n^2-s^4)\right]^{1/2}}, \tag{21}$$

where

$$E_{\mathrm{m}} = \left(8n^2 s/T_{\mathrm{m}}\right) - \left(n^2-1\right)\left(n^2-s^2\right). \tag{22}$$

Absorption coefficient of CdTe thin films is presented in Fig. 9. We can see that the largest value of the absorption coefficient is appropriate for the sample 2 and it is $4.8 \cdot 10^3$ (cm^{-1}).

Optical conductivity

We can analyze the value of the optical conductivity σ_{opt} as follows (Pankove 1975) using the calculated values of the absorption coefficient by the Swanepoel method

$$\sigma_{\mathrm{opt}} = \frac{\alpha n c}{4\pi}, \tag{23}$$

where α is the absorption coefficient and c is the velocity of light.

Figure 10 shows the variation of optical conductivity σ_{opt} as a function of photon energy $h\nu$. The optical conductivity of thin films increases with the growth of photon energy and with the thickness. The values of optical conductivity for sample 2 are shown in Table 2. This infers that the contribution of electron transition increases with dopant level, which may be due to the reduction in energy band gap (Punitha et al. 2014).

Conclusion

The thin films of CdTe were obtained by vapor-phase condensation on the (100) Si and glass substrates. Based on structural studies, it has been found that the films have a certain growth periodicity along the axis Ox, which is evidently observed on silicon substrates. Arranged on the surface of the film nanoparticles are characterized by Stranski–Krastanov mechanism of growth, which allows to control smoothly the photoelectric properties. The autocorrelation function well describes the distribution of these nanoparticles, and their sizes were (25–28) nm, both on glass and silicon substrates. It is established that the increase of the film thickness does not greatly affect on the size of the nanoparticles. The edge of the absorption band in the films is appropriate for a short infrared wavelength. For larger wavelengths, the light begins to interfere with the surface of the film, forming an interference pattern, which further confirmed the presence of a homogeneous smooth surface. Using the Swanepoel method, the optical characteristics of thin films (such as refractive index, film thickness, absorption coefficient and optical conductivity) were calculated. They are consistent with the experimental data.

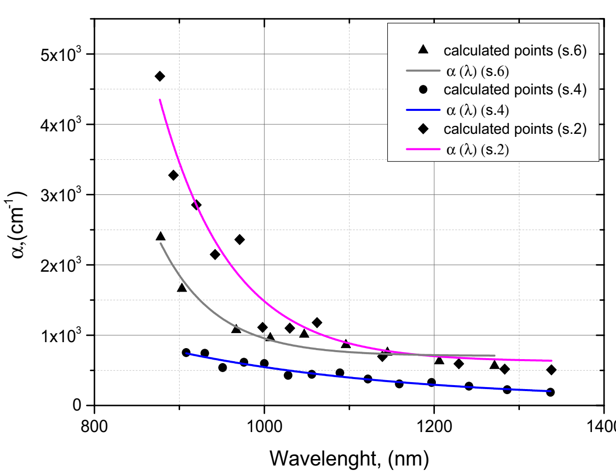


Fig. 9 Absorption coefficient of CdTe thin film calculated using Swanepoel method

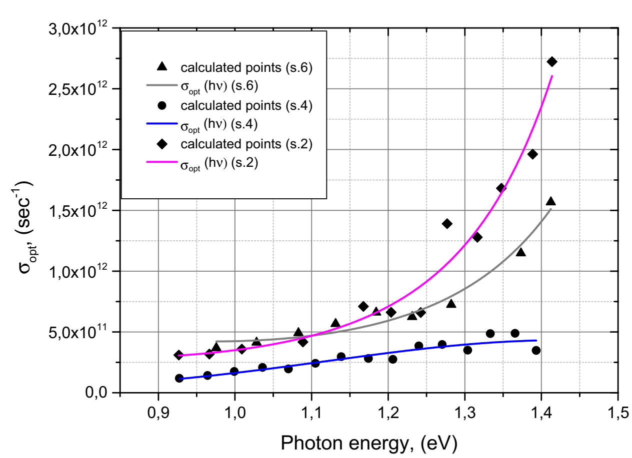


Fig. 10 Dependence of optical conductivity versus photon energy for CdTe thin films

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Author contributions RY grew the thin films, calculated the optical properties, interpreted the results and wrote the manuscript. LN performed and determined the problem, conducted the experiments, and performed the characterization. GW has defined the methods and subjects of investigation. PP has conducted research on the optical properties of thin films. SA has conducted research of structural properties on the AFM. SzG performed SEM, cross-section images and EDX analysis. All authors have read and approved the final version of the manuscript.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no competing interests.

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