Enhancement in microstructural and optical properties of thermally evaporated CdTe/CdSe heterojunction thin Films

Mehdi Ahmad Dabban^{1*} and Abdel-naser A. M. Alfaqeer²

¹Physics Department, Faculty of Science, University of Aden, Aden, Yemen
²Physcis Department, Faculty of Science and Education, Saba Region University, Marib, Yemen
* Corresponding author: mehdi.ahmed.scie@aden-univ.net
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Abstract

The optimization of microstructural and optical properties of a thin layer is an important step prior device fabrication process, so an enhancement in these properties of thermally evaporated CdTe/CdSe thin films was reported in this work. We choose to research (CdSe) materials as a n-type absorber layer in the CdTe/CdSe heterojunction thin film. The Effect of annealing temperature on the structural and optical properties of CdTe/CdSe Heterojunction thin films was studied, using various techniques such as X-ray diffraction (XRD), scanning electron microscopy (SEM) and Double-beam computer-controlled spectrophotometer in the wavelength between 200 nm and 2500 nm. Diffraction (XRD) patterns showed that the as-prepared films were amorphous nature, whereas the annealed films were polycrystalline. These results were confirmed by scanning electron microscopy investigations. It was found that the crystallite size and degree of crystallinity of the studied films depend on the annealing temperature. Furthermore the optical measurement shows that this treatment shifts the optical absorption edge at low energy and decreases the optical band gap from 1.92 eV, to 1.37eV while the values Urbach energy increase as the annealing temperature increased from 300 K to 433 K. As consequence is that the heat treatment improves the quality of the CdTe/CdSe heterojunction thin films for the potential use in photovoltaic applications.

Keywords: CdTe/CdSe thin films, Thermal evaporation, Microstructural, Optical properties

1. Introduction

Semi-transparent solar cells are a kind of photovoltaic cells in which the thickness of the absorber layer is low enough to provide the desired amount of transparency. These solar cells are used as photovoltaic windows in green buildings. Cadmium Selenide (CdSe) is an n-type, reddish semiconductor material. CdSe thin films have been used in photovoltaic devices because of their suitable direct band gap (about 1.74 eV for bulk CdSe material), high absorption coefficient and electrical conductivity [6,15].

Cadmium telluride (CdTe) is II-VI crystalline compound with a zinc blende crystal structure and has a direct bandgap of 1.44 eV, which is suitable for electronic applications such as photovoltaic devices, light-emitting diodes, solar cells, X-ray and gamma detectors, Field Effect Transistors (FETS), Lasers and non-linear integrated optical devices [21,5,13,7,9,11,14,18,27,3]. CdTe is the only II–VI compound that can be prepared with a high conductivity in both n and p type forms [8]. CdTe thin films are used as an absorber layer in semitransparent thin film solar cells [20]. There are many reports about the usage of the CdSe thin films as a window layer in the CdTe based thin film solar cells, but there is no report about the usage of CdSe thin films as an absorber layer in semitransparent solar cells. The effect of substrate temperature on the performance of the CdSe (window layer)/CdTe thin film solar cell was studied [12]. The usage of oxygenated CdSe thin films as a window layer in the CdTe based thin film solar cells was reported [2]. In this work, the thermal evaporation technique has been chosen to fabricate CdTe/CdSe heterojunction thin films because it can be used to prepare homogeneous thin films with controlled thickness. Also, thermal

Nitrogen-annealing was used to improve the structural and optical properties of the CdTe/CdSe heterojunction thin films.

2. Experimental Details

CdTe/CdSe heterojunction films were prepared by thermal evaporation technique, using a high vacuum coating unit (Edwards E-306) to reduce the contaminants in the film structure. Ingots of CdSe and CdTe were prepared by melting the precise weights of Cadmium (99.99%) and Selenium (99.99%) and Tellurium (99.99%) (Aldrich Chem Co,USA) in a silica tube evacuated from air under 1.33×10^{-3} Pa. The sealed ampoules were kept inside a furnace and heated gradually up to 1173 K and were kept at that temperature for 16hrs. Continuous stirring of the melt was carried out to ensure good homogeneity. The melt was then rapidly quenched in ice-water mixture to obtain the glassy state. After quenching, the solid ingots were removed from the ampoules and were kept in dry atmosphere.

To prepare the heterojunction film, CdTe is deposited on a predeposited CdSe film of thickness 200 nm, up to a thickness of 160 nm, using a tungsten basket as a heating source and oil diffusion pump backed by a rotary pump is used for attaining vacuum of the order of 2.66×10^{-3} Pa.. Deposition is done over the glass substrates. To clean the substrates, they are first washed and put in acetone and then placed in ultrasonic agitators for half an hour. Substrate are taken from the acetone and dried in hot air using a heater. The thermal annealing of the samples was carried out in an electrical furnace under nitrogen flow at (323, 373,423 and 473 K) for 30 min.

To identify the structure of the films, analysis of X-ray diffraction was conducted using Philips X-ray diffractometer 1710 (Netherlands) with monochromatic CuK α radiation ($\lambda = 1.5418$ Å). A computer-controlled spectrophotometer with double-beam (Shimadzu UV–Vis-2101) was employed to research the film's optical properties as transmission (*T*) and reflection (*R*) at 25 °C in the wavelength between 200 nm and 2500 nm. We have also used the scanning electron microscopy (SEM: Jeol (JSM)-T200 type) for the morphological studies.

3. Results and Discussions

3.1 Structural analysis

The microstructure of films is one of the factors that have great effects on film properties. Fig. 1 shows XRD diffraction patterns of the as-prepared and annealed CdTe/CdSe heterojunction thin films at different annealing temperatures. The as-prepared is poor crystalline where two tiny crystalline peaks appeared at $2\theta = 46.2^{\circ}$ and $2\theta = 71.92^{\circ}$ corresponding to orientations (311) and (105), respectively embedded in a hump representing an amorphous matrix. The XRD patterns of annealed CdTe/CdSe heterojunction thin films revealed the polycrystalline nature of these films. Having compared these data with the JCPDS Data files for CdTe (75-2086), CdSe (08-0459) and Se (85-0565), it is inferred that Cubic, hexagonal CdSe and hexagonal Se phases exist in the annealed film. The prominent peak corresponds to (002) plane and the peak in (311) direction was also observed. As annealing temperature increases, the prominent peak intensity of films with the same crystal structure increases greatly, this indicates a better crystalline. The strong X-ray peak at (0 0 2) plane indicates the preferred orientation of the crystalline grains with c-axis perpendicular to the plane of the substrate that makes the (0 0 2) peak relatively stronger than the other peaks.

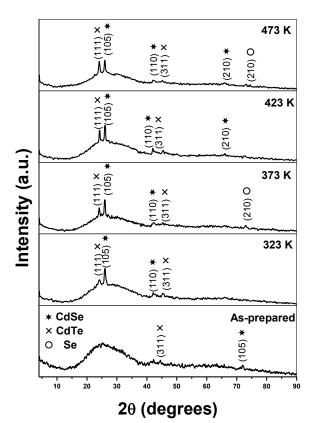


Fig. 1. X-ray diffraction patterns of the as-prepared and annealed CdTe/CdSe heterojunction thin films

X-ray diffraction also allows an estimate of the mean crystallite size D in polycrystalline thin films to be obtained, The crystalline size (D) was calculated for (200) plane, using the Scherrer's equation from the full-width at half-maximum (FWHM) (β) [26]:

$$D = 0.94(\lambda/\beta\cos\theta) \tag{1}$$

where, θ is Bragg's angle, β is the FWHM in radian and $\lambda=1.5418$ Å for the CuK_{α} radiation. Residual strain (ϵ) of annealed films are calculated from the following relation [17,25]:

$$\varepsilon = \left[\frac{\lambda}{D\cos\theta} - \beta\right] \frac{1}{\tan\theta}$$
(2)

The dislocation density (δ), defined as the length of dislocation lines per unit volume, was evaluated using the relation [16]:

$$\delta = \frac{n}{D^2} \tag{3}$$

where n is a factor, which equals unity giving minimum dislocation density. The number of crystallites per unit surface area N is also obtained by using the relation [19]

$$N = \frac{d}{D^3} \tag{4}$$

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where d is the thickness of the films. The calculated values of crystallite size, strain and dislocation density of the annealed thin films are included in Table 1 and replotted in Fig. 2. The crystallite size indicates that the films are noncrystalline nature. The increase in crystallite size values for the annealed samples from 21.83 nm to 47.29 nm shows the improvement in the crystallinity of the films. Since the dislocation density and strain are manifestation of dislocation network in the films, the decrease in the strain and dislocation density indicates the formation of high quality films at higher annealed temperature.

 Table 1: Structural and optical parameters of the as-prepared and annealed CdTe/CdSe

 heterojunction thin films

| Temp. (K) | X _c (%) | D(nm) | Е (×10 ⁻³) | $\frac{\delta}{(\times 10^{11})}$ | $(\times 10^{11})$ | E _g (eV) | E _U (eV) |
|-------------|-----------------------|-------|---------------------------|-----------------------------------|-------------------------|------------------------|---------------------|
| A 1 | 1 201 | | | (lin/cm²) | (Crys/cm ²) | 1.02 | 0.1(1 |
| As-prepared | 1.291 | - | - | - | - | 1.92 | 0.161 |
| 323 | 9.800 | 21.83 | 1.95 | 21.00 | 34.61 | 1.86 | 0.242 |
| 373 | 10.285 | 26.60 | 1.6 | 14.10 | 19.10 | 1.63 | 0.370 |
| 423 | 11.289 | 38.69 | 1.1 | 6.68 | 6.22 | 1.48 | 0.442 |
| 473 | 11.716 | 47.29 | 0.089 | 4.47 | 3.40 | 1.37 | 0.623 |

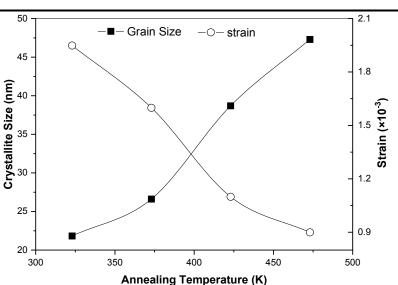


Fig. 2: Values of the crystallite size (D), Strain (ε) as a function of annealing temperature for CdTe/CdSe heterojunction thin films

Fig. 3 shows the crystalline fraction that can be obtained by integrating the areas under the crystalline A_c , and amorphous A_a of the diffraction patterns, according to:

$$X_c = \frac{A_C}{A_c + A_a} \tag{5}$$

The estimated degree of crystallinity, X_c values of the as-prepared and annealed CdTe/CdSe heterojunction thin films, shown in Table 1 and plotted in Fig. 4, increase with the annealing temperature. This is an indication for increasing the crystallinity of the studied films with the thermal annealing. The dependence of X_c on the annealing temperature T can be fitted to a third-degree polynomial function of the form:

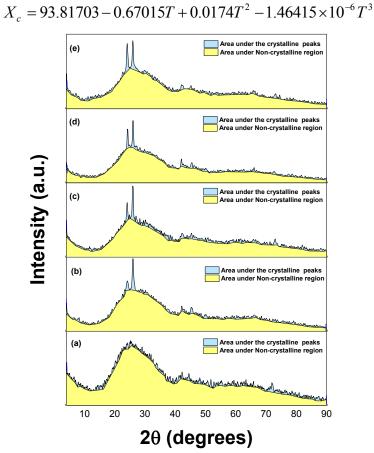


Fig. 3. The crystalline fraction of the as-prepared and annealed CdTe/CdSe heterojunction thin films

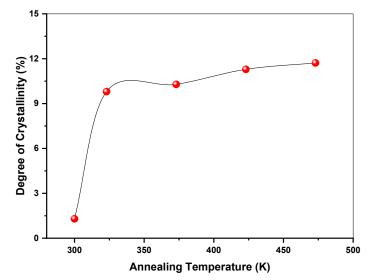
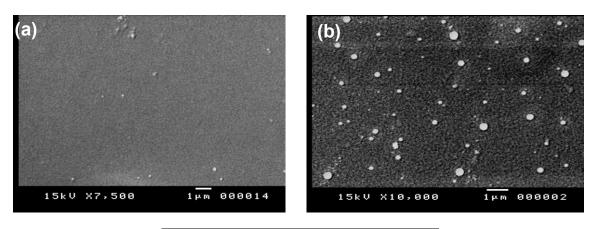


Fig. 4: Values of degree of crystallinity, X_c (%) as a function of annealing temperature for CdTe/CdSe heterojunction thin films.

SEM micrographs of (Fig. 5a-c) show the surface morphology as-prepared and annealed CdTe/CdSe heterojunction thin films at 10,000 magnification. The scale bar length is 1µm. The as-

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prepared film exhibits the amorphous state with some small crystallites in the amorphous matrix (Comparing the SEM and XRD results, suggests that these small crystallites might be for Cubic – CdTe and Hexagonal-CdSe phases On the other hand, the surface morphology of the films annealed at 373 K for 30 min in N₂ atmosphere (Fig.5b) shows small crystallites that are uniformly distributed over amorphous homogenous background. Increasing the annealed temperatures in films leads to the increase of the size with distinguishable agglomerations, as shown in (Fig.5c). Comparing again the SEM and XRD results of the annealed films suggests that the large crystallites are formed.



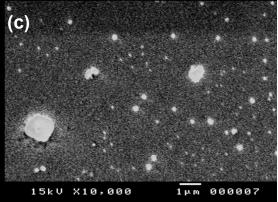


Fig. 5. SEM photograph of as-prepared and annealed CdTe/CdSe heterojunction thin films

3.2 Optical properties analysis

The spectral distribution of the transmittance T and reflectance R of the as-prepared and annealed CdTe/CdSe heterojunction thin films as a function of the wavelength of the incident light is shown in Fig. 6. In general, the transmittance decreases, while variable behaviour of the reflectance with increasing the annealing temperature (323–473 K). Moreover, the changes in the optical transmission of the annealed films show the shifts of the high energy absorption edges to higher wavelengths with increasing the annealing temperature. The change in T and R indicates a change in the structure of the studied samples during the thermal annealing. The change in structure may lead to a change in the electronic properties.

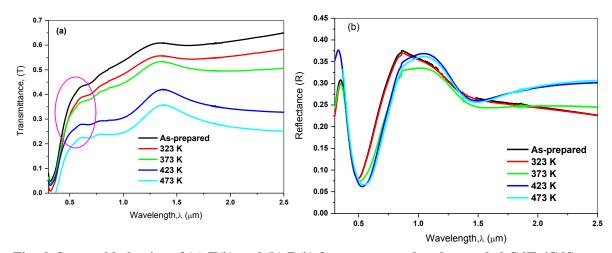


Fig. 6: Spectral behavior of (a) $T(\lambda)$ and (b) $R(\lambda)$ for as-prepared and annealed CdTe/CdSe heterojunction thin films.

The absorption coefficient can be estimated from the experimentally measured values of $T(\lambda)$ and $R(\lambda)$, using the following relationship [1]:

$$\alpha = \frac{1}{d} \ln \left[\frac{(1-R)^2}{2T} + \left\{ \left(\frac{(1-R)^2}{2T} \right)^2 - R^2 \right\}^{\frac{1}{2}} \right]$$
(6)

where *d* is the film thickness, according to Tauc [22], it is possible to isolate three distinct regions in the absorption edge spectrum in amorphous semiconductors : The first one (where $\alpha < 1 \text{ cm}^{-1}$) is the weak absorption tail which originates from defects and impurities, The second one where ($1 \le \alpha \le 10^4 \text{ cm}^{-1}$) is the exponential region, which is strongly related to the structural randomness of the system. The width of the band tails of the localized states near the band edges can be calculated, using Urbach's empirical relation [24]:

$$\alpha(v) = \alpha_o \exp(h v / E_U)$$

where h is the Plank's constant, hu is the photon energy, α_o is constant, EU, is often called the Urbach energy which reflects the width of the tails of the localized states in the band gap, and The last one, is the high absorption region, where ($\alpha > 104$ cm-1) the absorption coefficient has the following frequency dependence [23]:

$$(\alpha hv) = A(hv - E_g)^r \tag{8}$$

where A is constant depends on the transition probability and E_g is the optical band gap of the specimen and r takes the value 1/2 for the direct allowed transition, the value 3/2 for direct forbidden, the value 2 and 3 for the indirect allowed and indirect forbidden transitions respectively.

Fig. 7(a) shows the dependence of the absorption coefficient (α) on the incident photon energy for the as-prepared and annealed films. The value of (α), estimated using Eq. (6), was found to increase and shifts the optical absorption edge at low energy with the increase of the annealing temperature. Also, from Fig. 7 (a), the absorption spectra can be divided into two regions: the low absorption region ($\alpha < 10^4$ cm⁻¹) and the high absorption region ($\alpha > 10^4$ cm⁻¹). In the region of low absorption ($\alpha \le 10^4$ cm⁻¹), the absorption coefficient (α) shows an exponential dependence on the photon energy, hv, and obeys Urbach's relation given in Eq. (7). Thus plotting of ln(α) versus hv, Fig. 7 (b), should give straight line with the inverse of its slope equals E_U. The calculated values for E_U values for different films are listed in Table 1. The higher values of the absorption coefficients, $\alpha \ge 10^4$ cm⁻¹, correspond to transitions between extended states in both valence and conduction bands, where the power behavior of Tauc in Eq. (8) is valid. Fig. 7 (c) shows the linear

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relationship between $(\alpha hv)^2$ and the photon energy (hv) of the as-prepared and annealed thin films at the absorption edge. This result confirms the direct band gap transition in CdTe/CdSe heterojunction thin films. The optical energy gap of the direct transition is obtained by extrapolating the linear portion of the curves shown in Fig. 7 (c) to zero absorption. The obtained values of band gap energy Eg are listed in Table 1. Fig. 8 shows the variation of the estimated Eg and E_U values for different annealing temperatures. It can be noted that the values of the energy gap decrease from 1.92 eV to 1.37eV as the annealing temperature increases from 323 K to 473 K) which is confirmed in Fig. 7 (a) as a shift of the absorption coefficient, α , towards lower energies. The decrease in the optical energy gap E_g and the increase of localized states tails (Urbach energy) E_U with the annealing temperature can be interpreted based on the production of surface dangling bonds around the crystallites during the process of crystallization [4]. It has been suggested by many authors that nearly ideal amorphous solids crystallize under the thermal annealing and accordingly the dangling bonds are produced around the surface of the crystallites [10]. Thus, the concentration of localized states in the band structure increases gradually as a result of increasing the dangling bonds in the annealed films. This could lead to an increase in the width of localized states, indicating a decrease in the optical energy gap of the investigated films.

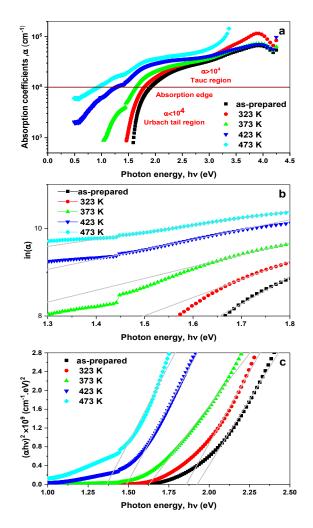


Fig. 7: Plots of (a) Absorption coefficient, α (b) $\ln(\alpha)$ and (c) $(\alpha hv)^2$ versus photon energy (hv) for as-prepared and annealed CdTe/CdSe heterojunction thin films.

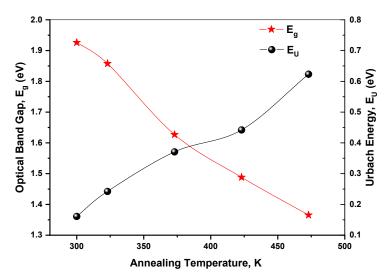


Fig. 8. The values of E_g and E_U as a function of annealing temperature of CdTe/CdSe heterojunction thin films

4. Conclusions

In this paper, the properties of CdTe/CdSe heterojunction thin films with different annealing temperatures were studied by XRD, SEM and UV–VIS spectrometer. The major conclusions drawn from this work can be summarized as follows:

- 1. The structure of CdTe/CdSe heterojunction thin films is related to annealing temperatures. As annealing temperature rises, it changes from the amorphous state to polycrystalline structure.
- 2. Direct electronic transition is presented in CdTe/CdSe heterojunction thin films.
- 3. Increasing the annealing temperature leads to the decrease in the optical band gap E_g and increase in the width of localized states (Urbach energy, E_U).
- 4. The reflectance (at transparent region), absorption coefficient are increased with the increase annealing temperature.
- 5. The thermal annealing temperature is an important factor which can control the optical parameters of CdTe/CdSe heterojunction thin films and help in developing these materials for the potential use in technological applications.

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تحسين الخواص التركيبية والبصرية للأغشية الرقيقة غير المتجانسة CdTe/CdSe

المطرة بتقنية التبخير الحراري

مهدي أحمد دبان¹ وعبدالناصر عبدالرحمن محمد الفقير² ¹ قسم الفيزياء، كلية العلوم، جامعة عدن، عدن، اليمن ² قسم الفيزياء، كلية العلوم والتربية، جامعة إقليم سبأ، مأرب، اليمن ³ المؤلف المقابل: mehdi.ahmed.scie@aden-univ.net * DOI: <u>https://doi.org/10.47372/uajnas.2022.n2.a14</u>

المُلخّص

يعد تحسين الخصائص التركيبية والبصرية للأغشية الرقيقة خطوة مهمة قبل عملية تصنيع الأجهزة الإلكترونية للتطبيقات الكهر وضوئية، لذلك في هذا البحث عملنا على تحسين هذه الخصائص للأغشية الرقيقة غير المتجانسة CdTe/CdSe المحضرة بتقنية التبخير الحراري. واستخدمت (n-CdSe) كطبقة ماصة في تلك الأغشية الرقيقة.

تمت دراسة تأثير درجة حرارة التلدين على الخصائص التركيبية والبصرية لتلك الأغشية باستخدام تقنيات مختلفة مثل حيود الأشعة السينية (XRD) والميكروسكوب الإلكتروني (SEM)، ومقياس الطيف الضوئي المزدوج الشعاع عند الطول الموجي بين 200 نانومتر و2500 نانومتر. أكدت نتائج XRD أن الأفلام المحضرة لها طبيعة غير متبلورة، وبزيادة درجة حرارة تلدين الغشاء الرقيق تحدث التحولات من الحالة الأمورفية إلى الحالة البلورية. وجد أن الحجم البلوري ودرجة التبلور للأغشية الرقيقة تعتمد على درجة حرارة التلدين.

تُم تأكيد هذه النتائج عن طريق دراسة مورفولوجيا السطح بواسطة المجهر الإلكتروني الماسح. علاوة على ذلك، وضحت القياسات الضوئية أن المعالجة الحرارية بزيادة درجة حرارة التلدين من 300 كلفن إلى 433 كلفن تزيح حافة الامتصاص البصري إلى الطاقات المنخفضة، وهذا يقلل فجوة الطاقة من 1.92 إلكترون فولت إلى 1.37 إلكترون فولت بينما تزداد قيم طاقة أورباخ.

أثبتت الدراسة الحالية أن المعالجة الحرارية تعمل على تحسين جودة الأغشية الرقيقة غير المتجانسة CdTe/CdSe للاستخدامات في التطبيقات الكهروضوئية.

الكلمات المفتاحية: الأغشية الرقيقة، تقنية التبخير الحراري، التركيب الدقيق، الخواص البصرية.