Enhanced physical properties of e-beam evaporated CdTe thin films for photovoltaic application

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CdTe thin films with 2.8 μm thickness were deposited by electron beam evaporation method. X-ray diffraction, scanning electron microscopy, UV-Vis-NIR spectroscopy and atomic force microscopy (AFM) were used to characterize the films. The results of AFM analysis revealed that the CdTe films have uniform surface. CdTe thin films were heat-treated by $SnCl_2$ solution. Structural analysis using XRD showed that heat treatment by $SnCl_2$ solution improves thin film crystallinity. A solar cell device was fabricated based on electron beam deposited poly-crystalline CdTe and CdS thin film as absorber and window layer on ITO coated soda lime glass as substrate. The electrical characteristics of CdS/CdTe thin film solar cells were investigated under illumination. From the current-voltage characteristics of fabricated device a typical rectifying photovoltaic behavior was obtained.

(Received October 16, 2015; accepted August 9, 2017)

Keywords: CdTe, Absorber Layer, Thin Film, Solar Cell, Electron Beam Evaporation

1. Introduction

Thin film solar cells are the second generation of solar cells which are manufactured by depositing one or several thin-film photovoltaic materials on glass, plastic or metal substrate. Cadmium telluride (CdTe), copper indium gallium selenide (CIGS), copper zinc tin sulfide (CZTS), amorphous and crystalline silicon thin films are the usual materials which are used in manufacturing thin film solar cells.

In the first generation of crystalline silicon- based solar cells, silicon wafers has a thickness of 200 micrometer. But in thin film solar cells, cell thickness can be ranged from a few nanometers to tens of micrometers, which is much thinner than the first generation of silicon-based solar cells. This allows manufacturing of lightweight and flexible solar cells. In addition, this technology makes it possible to integrate the cells with building photovoltaic systems or to cover the glass windows by semi-transparent photovoltaic materials.

Despite the introduction of many new materials for manufacturing thin film solar cells, cadmium telluride thin films are still the best materials. About 5% of global photovoltaic production and more than 50% of thin film solar cells market are fabricated using CdTe. Cadmium telluride has the lowest energy payback time compared to the all the other mass production photovoltaic technologies [1]. In favorable situations, energy payback time of this technology can be as short as eight months. Environmental concerns about the toxicity of cadmium can be fully resolved by recycling cadmium at the end of the period [2].

Although this material can be deposited by various growth techniques [3-6], but all CdS/CdTe thin film solar cells are chemically heat treated in chlorine to increase grain growth and to achieve efficiencies greater than 10% [5, 7-9]. Although chemical treatment is known since 1976 and is widely used for different devices [10], but the effect of this process is not yet completely understood. This thermo-chemical process has a great impact on structural parameters such as doping, crystallization, absorber-window layer connection quality and finally cell efficiency.

The main scope of this research is to investigate applicability of CdTe thin films deposited by electron beam evaporation technique as absorber layer for photovoltaic applications.

Evaporation of CdTe granules is a direct and simple technique for deposition of CdTe thin films. Because it is a vacuum-based technique, the incorporation of impurities is relatively low comparing with chemical and electrochemical methods. In addition, in electron beam evaporation technique, there is an opportunity to incorporate dopant in gaseous phase in order to tune the stoichiometry and bandgap of deposited thin films [11]. In the present work, a novel attempt has been done to deposit CdTe thin films by electron beam evaporation.

In this research, the morphological, structural and optical properties of CdTe films deposited on glass substrates by electron beam evaporation as a function of deposition rate is studied using atomic force microscopy (AFM), X-ray diffractometry (XRD), and UV-Vis transmission spectroscopy. The effect of chemical treated thin films is studied by using X-ray diffraction.

2. Experimental

Indium thin oxide (ITO) disk supplied from Sigma-Aldrich was used as a target material for RF magnetron sputtering of transparent conducting electrodes. Microscope glass slides were used as substrates. The

substrates were first cleaned in alcohol ultrasonically, then rinsed in deionized water, and dried in nitrogen.

The deposition was performed using an Edwards Auto 306 vacuum coating unit. CdS powder and CdTe granules of 99.99% purity were used as source materials in electron beam evaporation technique. CdTe thin films with thickness of 2.8μm were deposited as absorber layer by electron beam evaporation technique from CdTe granules as evaporation source. The CdTe granules were transferred into graphite crucible kept in water-cooled copper hearth of the electron gun. The surface of CdTe granules were heated through bombardment of confocal electron beam with an accelerating voltage of 4.9 kV. The glass substrates and their masks were clamped to a stainless steel sheet which inserted in a radiation heater with tungsten heating element. The substrate-target distance was 30 cm and the substrate temperature was maintained at 100°C. The chamber pressure reached below 10^{-5} torr prior to deposition by using a diffusion pump. Deposition rate were controlled by controlling the current of electron beam. The deposition rate was measured by a quartz crystal monitor. The evaporation rate kept at 5nm/min. Our results showed that films with reproducible texture and properties can be obtained.

Molybdenum thin film was deposited by electron beam deposition method as cell back contact.

The SEM images were taken using Cambridge 360. SEM cross section in secondary electron mode was used for estimation of thickness and backscattered electron imaging was used for elemental composition evaluation. Cross-sectional SEM was prepared using breaking the samples.

A Park Scientific Autoprobe CP Atomic Force Microscope was used to quantitatively evaluate the surface topography of the thin films. Using AFM, changes in surface area and root mean square (rms) roughness was measured. Contact mode was used to capture the height images of each sample. The analyzed field was 2μm × 2μm at a scan rate of 1 Hz. A ProScan Image Processing software was used to analyze the resulting images. The surface area and rms roughness (nm) were obtained, averaged, and recorded in two random and arbitrary fields. The Optical transmission and absorption spectra of CdTe thin films were obtained using a UV–VIS–NIR spectrophotometer (Perkin Elmer, Lambda 25).

After deposition, CdTe thin films were immersed for 5s in a saturated $SnCl_2$ solution in isopropyl alcohol 60°C and subsequently heated under vacuum~10^{-3} mbar at 400°C for 10 min. The films immersed for more than 5s, showed physical disintegration.

X-ray diffraction patterns of deposited films and chemically treated films were recorded by X-ray diffractometer model Philips PW 3710 using copper Kα radiation (λ = 1.5418A˚) filtered through Nickel filter in the instrument operating at V = 40kV in the interval 24˚ ≤ 2θ ≤ 72˚ at a scan speed of 2˚/minute giving a step size 0.02.

The current–voltage analysis of CdS/CdTe heterojunction was performed using Autolab potentiostat under AM 1.5 illumination.

3. Results and discussion

Fig. 1 shows typical AFM micrographs of 700 nm thick CdTe films evaporated on glass substrates at 120 nm/min.

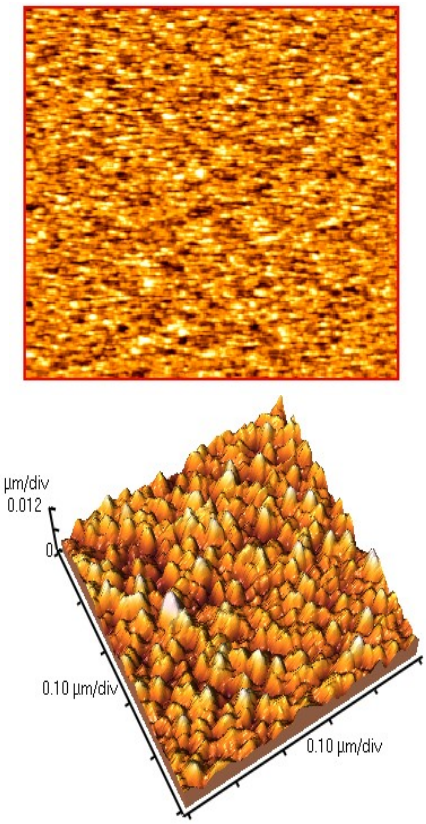

Fig. 1. AFM images ((2 × 2) μm^2) of 2.8μm thick CdTe films evaporated on soda lime glass substrates

Three-dimensional AFM images show that CdTe films have grown as nanocone arrays. The maximum height of nanocones is ~21 nm. The mean diameter of nanocones is ~50nm. Besides, the average roughness of the film surface on area of 2×2 μm2 is ~2.28 nm. These results suggest that CdTe thin films grown by electron beam deposition technique are closely packed and has smooth surface which is suitable for photovoltaic applications.

Fig. 2 demonstrates the X-ray diffraction (XRD) patterns of resultant films prepared using electron beam deposition technique before and after chemical treatment by $SnCl_2$. X-ray patterns shows a strong preferential orientation with a single main peak at $2\theta = 23.8$, which corresponds to cubic CdTe reflection. Preferential

orientations, as observed above, are probably due to the nucleation process associated with the deposition rate of CdTe. Obviously, peaks at $2\theta = 39.38$,46.41 became stronger in thermo-chemically treatmented films due to the grain growth. There is not any significant peak related to impurity in the XRD pattern which shows that there is no incorporation of impurities due to this treatment. By using the Bragg's diffraction condition inter-planar spacing (*d*) values were calculated for different planes of the XRD patterns. The observed *d* values of films were found to be very close to the standard data given in PCPDF data file No. 00-015-0770 cubic CdTe phase.

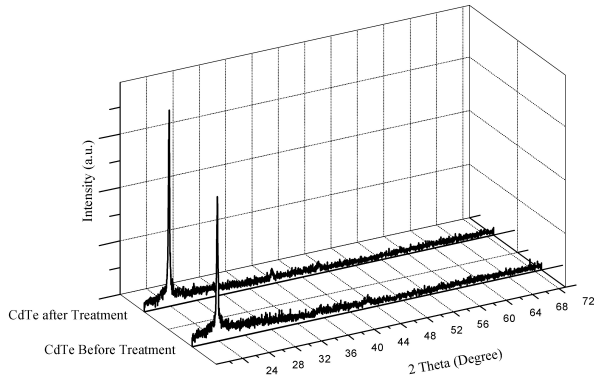

Fig. 2. XRD pattern of CdTe thin films before and after thermochemical treatment

The crystallite size was calculated by using Debye-Scherrer equation [12]:

$$d = \frac{0.9\lambda}{\beta \cos\theta}$$

Where, *d* is crystallite size, λ is wavelength of X-rays, β is angular full width at the half - maximum (FWHM) of intensity and θ is Bragg's diffraction angle. No correction has been made for instrument line broadening. The average crystallite size of as-deposited and thermo-chemically treated films was found to be 41 nm and 59 nm, respectively. The values for the crystallite size were found to be in nanometer scale. It indicates the nano-crystalline nature of resultant films.

Fig. 3 shows the transmittance (%T) curves for the CdTe films. The average transmission for films was found to be less than nearly 20%.

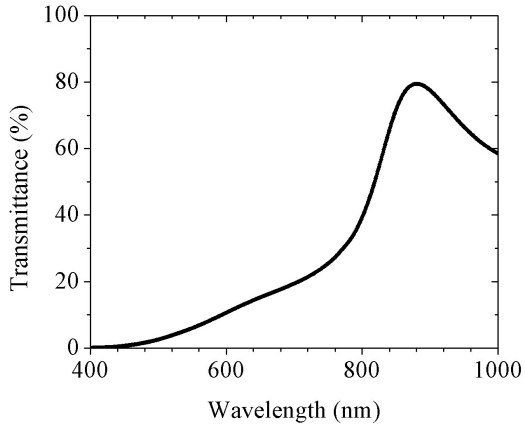

Fig. 3. Optical transmittance spectra of CdTe thin films

For a direct transition semiconductor, the optical band gap (E_g) can be calculated using the experimentally determined data of absorption coefficient α from the following equation:

$$(\alpha h\upsilon)^2 = A(h\upsilon - E_g)$$

where *A* is a constant, α is the absorption coefficient, *h* is the Planck's constant and $h\upsilon$ is the energy of incident photon. Using transmittance data, the absorption coefficient α was calculated (using following equation [6]) and was then used to determine the band gap for the films deposited at different deposition rates of 12 and 60nm/min., as shown in Fig. 4.

$$\alpha = -\left(\frac{2.303}{t}\right)\log_{10}\left(\frac{1}{T}\right)$$

Here, *t* is the film thickness and *T* is the transmittance (%) of the films.

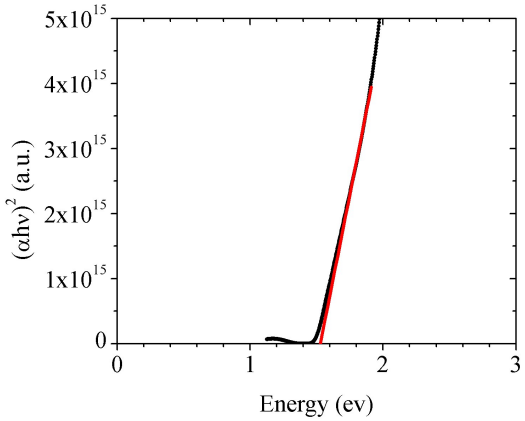

Fig. 4. Tauc Plot of $(\alpha h\upsilon)^2$ *versus* $h\upsilon$ *for CdTe films*

The energy band gap was estimated from the Tauc plot of $(\alpha h\upsilon)^2$ versus the photon energy ($h\upsilon$), through extrapolating the linear portion of each curve back to the energy axis in Figure 4. The value of band gap was found to be 1.55 eV which was 0.05 eV higher than the reported values. This slight difference can be correlated to nano-size of crystals in deposited film.

Fig. 5 shows a cross-sectional SEM micrograph obtained from the secondary electrons for the fabricated CdTe solar cell using fracturing the glass substrate. This image clearly shows the presence of ITO (~700 nm thick) on glass, CdS layer (770 nm thick), CdTe (~2.8 μm thick) layer and molybdenum back contact (~910 nm thick).

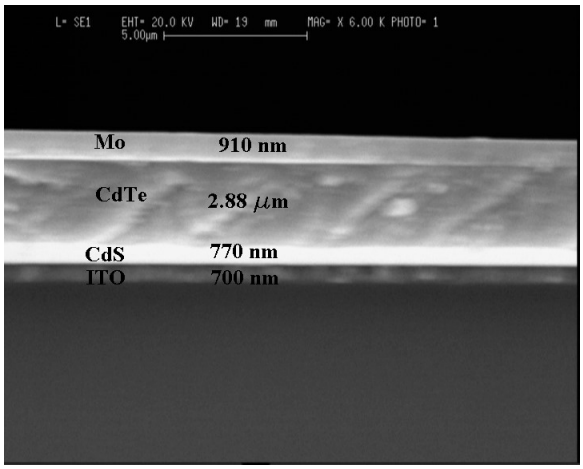

Fig. 5. Cross-sectional SEM micrograph obtained from the secondary electrons for the fabricated CdS/CdTe solar cell using fracturing the glass substrate

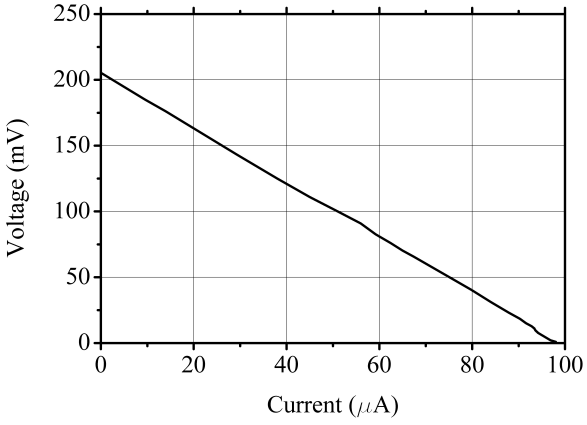

Fig. 6. The I–V measurement of CdS/CdTe fabricated cell under AM 1.5 illumination

Next, the solar cell application of fabricated CdS and CdTe thin films was studied. The current–voltage curves shown in Fig. 6 are recorded under AM 1.5 global spectrum at room temperature. The device showed a typical rectifying property and photovoltaic effect. The open-circuit voltage, short-circuit current density, and fill factor of fabricated cell were 200 mV, 100 μA/cm^2, and 50%, respectively. It led to an efficiency of 0.5%. In further studies, characteristics of fabricated solar cells can be improved by implementing grain growth technique such as thermo-chemical treatment, engineering the layer sequence and designing better masks for deposition.

4. Conclusions

The structural, morphological and optical properties of CdTe films deposited on glass substrates by electron beam evaporation were investigated by XRD, AFM, UV-VIS Spectra.

XRD studies showed that films had a strong preferential orientation with a single main peak. In addition X-ray patterns showed that peaks sharpens after thermo-chemical treatment by $SnCl_2$ which is the result of crystallite growth in deposited films.

The optical properties showed that deposited films were smooth, free from pin holes with transmittance less than ~20%. The optical band gap of as-deposited film was found to be 1.55 eV.

In conclusion, we have shown that thermo-chemical treatment by $SnCl_2$ increases the crystallite size and incorporates no impurity in the films.

The photovoltaic behavior of CdS/CdTe thin film solar cell showed that the electron beam evaporation technique is an efficient technique for green production and large area coating of CdTe for solar cell applications.

Acknowledgments

The authors would like to thank Mr. A. Soleimani for supporting electron beam depositions and Ms. S. Ghofrani for supporting SEM observations. The authors wish to thank Materials and Energy Research Center for the financial support to the project No. 421392015.

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