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Investigation of Physical Properties of e-Beam Evaporated CdTe Thin Films for Photovoltaic Application

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ABSTRACT

CdTe thin films 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. In this research, we have used $SnCl_2$ heat treatment instead of usual $CdCl_2$ to determine whether this technique can also assist the re-crystallization of the CdTe layers. 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.

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 sulphide (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 are fabricated using CdTe. Cadmium telluride has the lowest energy payback time compared to all other mass production photovoltaic technologies. In favourable 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 [1].

Although this material can be deposited by various growth techniques [2-8], 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% [4, 9-11]. Although chemical treatment is known since 1976 and is widely used for different devices [1], 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.

Evaporation of CdTe granules is a direct and simple technique for deposition of CdTe thin films. In this 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 [12].

There are many attempts regarding characterization of CdTe thin film deposited by electron beam evaporation

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method [13-19]. As to our knowledge, there are few reports regarding the use of electron beam evaporated CdTe thin films for photovoltaic application [14]. The re-crystallization of CdTe thin film using $CdCl_2$ is a necessary process for obtaining high efficiency photovoltaic devices.

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. The main scope of this research is to investigate the applicability of CdTe thin films deposited by electron beam evaporation technique as absorber layer for photovoltaic applications and the efficacy of treatment of deposited film in $SnCl_2$.

2. MATERIALS METHOD

CdS powder and CdTe granules of 99.99% purity (Merck) were used as source materials in electron beam evaporation technique. 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.

ITO thin films prepared by sputtering method on glass substrates have been used as transparent conducting oxide. Sputtering has been performed at 120 W RF power. The thickness and sheet resistance of deposited ITO films were around 700 nm and 0.1 Ω-cm, respectively.

The CdTe and CdS thin film deposition was performed using an Edwards Auto 306 vacuum coating unit. CdTe thin films with thickness of 2.8μm were deposited 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 glass substrates and their masks were clamped to a stainless steel sheet which was 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 was controlled by controlling the current of electron beam. The deposition rate was measured by a quartz crystal monitor. The evaporation rate was kept at 5nm/min. 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 the thickness and backscattered electron imaging was also 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 area 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 regions. The optical transmission and absorption spectra of CdTe thin films were obtained using a UV–VIS–NIR spectrophotometer (Perkin Elmer, Lambda 25).

For improving the crystallinity of deposited CdTe thin films and back contact electrical performance, substrates were immersed in a saturated $SnCl_2$ solution in isopropyl alcohol at 60°C for 5s and subsequently heated under vacuum of ~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 degree/minute giving a step size 0.02 degree.

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

3. RESULT AND DISCUSSION

Figure 1. shows typical AFM micrographs CdTe films evaporated on glass substrates at 120 nm/min. Three-dimensional images show that CdTe films have grown as nanocone arrays. The maximum height and the mean diameter of the nanocones are ~21 nm and ~50nm, respectively. 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.

Figure 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 $2\theta = 23.8^{\circ}$, 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

$2\theta = 39.38^\circ, 46.41^\circ$ and $2\theta = 46.41^\circ$ became stronger in thermo-chemically treated films due to the grain growth.

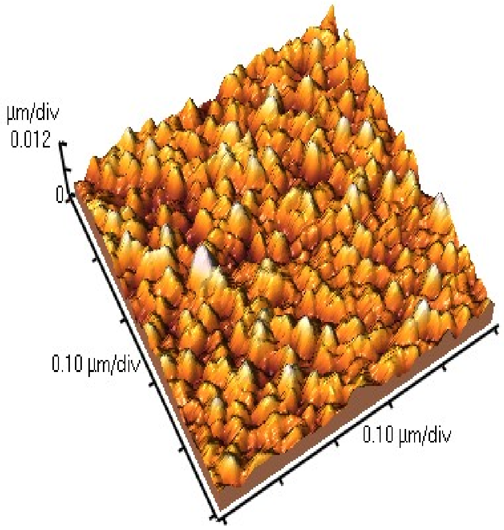

Figure 1. AFM images ((2 × 2) µm2) of 2.8µm thick CdTe films evaporated on soda lime glass substrates.

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.

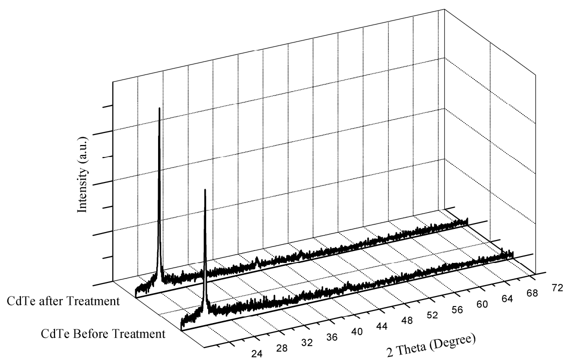

Figure 2. XRD patterns of CdTe thin films before and after thermochemical treatment.

The crystallite size was calculated by using Debye-Scherrer equation [5]:

$$d = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

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 which indicates the nano-crystalline nature of resultant films.

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

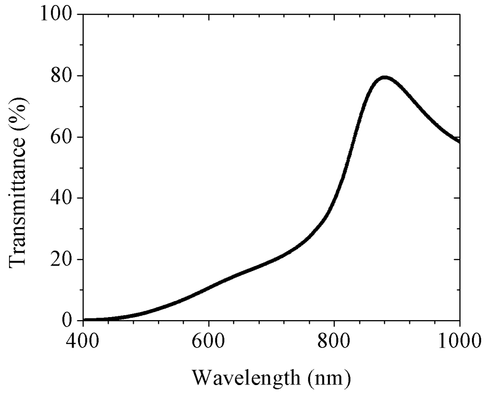

Figure 3. Optical transmittance spectra of CdTe thin films.

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:

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

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 [8]) and was then used to determine the band gap for the deposited films as shown in Figure 4.

$$\alpha = -\left(\frac{2.303}{t}\right)\log_{10}\left(\frac{1}{T}\right) \quad (3)$$

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

Figure 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).

Next, the solar cell application of fabricated CdS and CdTe thin films was studied. The current–voltage curves shown in Figure 6 are recorded under AM 1.5 global spectrum at room temperature. The device showed a typical rectifying property and photovoltaic effect.

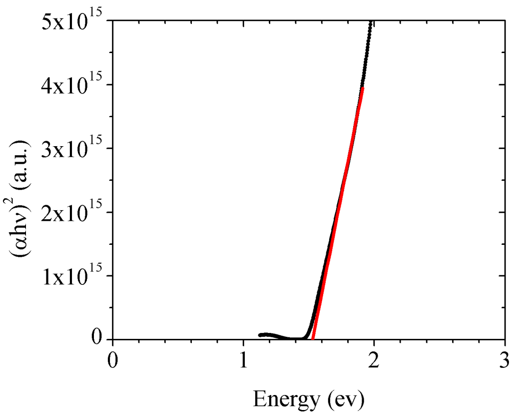

Figure 4. Tauc Plot of versus for CdTe films.

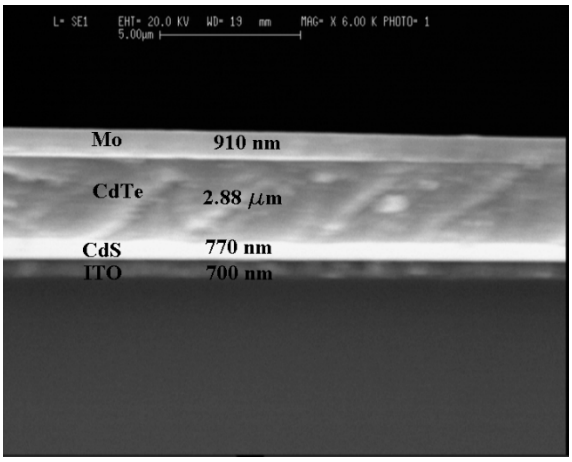

Figure 5. Cross-sectional SEM micrograph obtained from the secondary electrons for the fabricated CdS/CdTe solar cell using fracturing the glass substrate.

The open-circuit voltage, short-circuit current density, and fill factor of fabricated cell were 200 mV, 100 $\mu A/cm^2$, and 25%, 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. CONCLUSION

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. In addition, characteristic I-V curve of the fabricated solar cell was also investigated.

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.

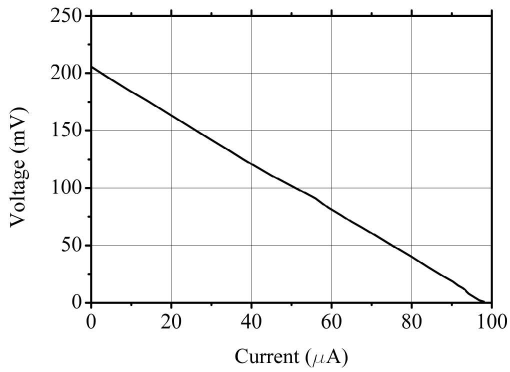

Figure 6. The I–V measurement of CdS/CdTe fabricated cell under AM 1.5 illumination.

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.

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