Band-Gap Tuning of Electron Beam Evaporated Cds Thin Films

Mohamad Javad Eshraghi [1,*], Nima Naderi [2]
[1,2] Semiconductors Department, Materials and Energy Research Center (MERC), P. O. Box 14155-4777, Tehran, IRAN

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ABSTRACT

The effect of evaporation rate on structural, morphological, and optical properties of electron beam evaporated CdS thin films has been investigated. CdS thin film was deposited by electron beam evaporation method in 12nm/min and 60nm/min evaporation rates on glass substrates. X-ray diffraction (XRD), scanning electron microscopy (SEM), UV-Vis-NIR spectroscopy and Atomic Force Microscopy (AFM) were used to characterize the thin films. The x-ray diffraction analysis confirms that the films have polycrystalline hexagonal phase and exhibit preferred orientation along the (002) plane. The crystallite size was calculated and found to increase from 23 to 30 nm by increasing the deposition rate. Results of Atomic Force Microscopy revealed that the RMS roughness values of the CdS films decreased as the deposition rate increased. The relation between deposition rates and optical properties of deposited films was also investigated. It was found that stoichiometric properties and band gap values of the deposited films are correlated to the deposition rates. These dependencies are associated with the Cd/S ratio variation by the deposition rate. The optical band gap values of CdS films increased slightly in a range of 2.32–2.34 eV for deposition rate varied from 12nm/min and 60nm/min.

1. Introduction

Polycrystalline cadmium sulfide thin films have good optical transmittance, wide band-gap and good electrical properties [1-5]. Their high absorption coefficient, electron affinity and low resistivity make them suitable for solar cell applications [3, 6, 7]. Because of these properties, much research is being done on direct band-gap cadmium sulfide thin films [8, 9].

Due to the limited dopability of CdTe, fabrication of high efficiency devices in homojunction configuration based on this material is not so attractive [10]. Therefore, heterogeneous structure with n-type CdS and p-type CdTe is often prescribed to increase efficiency. CdS with 2.42 eV forbidden band [1, 6, 11-13] absorbs photons with wavelengths lower than 510nm. In high-performance superstrate solar cells, usually a thick layer of window layer (CdS) is used between the transparent conducting front electrode (TCE) and the absorber layer (CdTe) to ensure complete coverage of the TCE and avoid contact of the absorber layer and front electrode [14] to maintain consistent open circuit voltage and fill factor. Another advantage of using CdS as window layer is that transparent conductive oxides (TCO used as front electrode) generally form low-resistance ohmic contact by n-type cadmium sulfide window layer and improve the device open circuit voltage [15].

Although thin CdS layers grown by chemical bath deposition (CBD) technique are preferred as window layer because of their low optical absorption losses and good covering properties

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[*] Corresponding author:
E-mail: m.eshraghi@merc.ac.ir, Fax: +98 26 3628 0030

on TCO, pin-holes and irreproducibility are considered for reliable industrial production [12, 16-22]. In addition, although CBD is a simple technique for mass production of high-quality cadmium sulfide thin films, other techniques such as electrodeposition [18, 23, 24], sputtering [1, 3, 25], vacuum evaporation, and spray pyrolysis [26-29] which do not produce toxic byproducts and wastes are environmentally preferable. Therefore, the main purpose of this report is to investigate applicability of cadmium sulfide thin films deposited by electron beam evaporation technique as window layer for photovoltaic applications.

Evaporation of CdS powder is a direct and simple technique for deposition of CdS thin films. Because it is a vacuum-based technique, the incorporation of impurities is relatively low compared with chemical and electrochemical methods. In addition, in electron beam evaporation, there is an opportunity to incorporate oxygen with CdS in order to tune the band gap of deposited CdS thin films [30].

Dresner investigated the crystallinity and electronic properties of electron beam evaporated CdS thin films [31]. Foster observed the preferred orientation with the *c* axis normal to the film plane, in electron beam evaporated CdS thin films. He also provided information required to control the fabrication of shear mode thin film ultrasonic transducers [32]. Tyagi compared the structural and electro-optical properties of electron beam evaporated CdS films in H_2S containing atmosphere [33]. Ashour investigated the effect of the processing conditions (the film thickness, the substrate temperature and the deposition rate) on electrical and optical properties of thermally deposited CdS films. But they didn’t investigate the structural and morphological properties of evaporated films [34]. Pal studied the optical, morphological and structural properties of vacuum evaporated CdS films deposited at different substrate temperatures using scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD) analysis. The optical band gap values of the films varied from 2.38 to 2.41 eV depending on the substrate temperature [35]. Singh investigated the effect of hydrogen sulfide atmosphere on the optical properties of evaporated CdS thin films and determined the optical constants of deposited films over the wavelength range of 350–800 nm [36]. Singh grew stoichiometric CdS thin film by using a low ambient H_2S atmosphere inside the vacuum chamber while thermally evaporating the CdS and studied the effect of ambient H_2S on the physical and optical properties of these films [37]. Yang investigated the effect of substrate temperature on structural and optical properties of highly crystalline and transparent CdS films deposited on glass substrate by electron beam evaporation [38].

There are many other reports regarding the effect of substrate temperature, evaporation atmosphere and various post deposition annealing processes on electrical, structural and optical properties of CdS vacuum evaporated thin films [13, 39-49]. However, to the best of our knowledge there is no report on the effect of deposition rate on structural, morphological and optical properties of electron beam evaporated CdS thin films. Therefore, in the present work, a novel attempt has been made to investigate the role of deposition rate in the structural, morphological and optical properties of CdS thin films deposited by electron beam evaporation. In particular, in this study, the effect of deposition rate on the band gap and absorption spectra of CdS window layer has been investigated. In electron beam technique, the deposition rate is one of the most important parameters influencing the structure, surface morphology, impurity, physical property and especially stoichiometery of the thin films in compound materials. In this research, we show that hexagonal CdS thin films with excellent (002) orientation could be successfully prepared by the electron beam technique. It is well known that hexagonal CdS films invariably grow with columnar structure along the *c*-axis perpendicular to the substrate. This property makes such films very suitable for use in thin film solar cells because there are fewer grain boundaries parallel to the junction which would prevent carrier recombination in the sheet. The morphological, structural and optical properties of CdS films deposited on glass substrates by electron beam evaporation as a function of deposition rate are studied using atomic force microscopy (AFM), X-ray diffractometry (XRD), and UV-Vis transmission spectroscopy.

2. Materials and methods

The deposition was performed using an Edwards Auto 306 vacuum coating unit. CdS powders of 99.99% purity were used as the source materials in electron beam evaporation. The CdS source was formed in 5 mm diameter pellets under 10MPa uniaxial compression. Using the compressed pellets as the evaporation source, deposition rates up to 100 nm per minute were easily attainable. The pellets were transferred to a graphite crucible kept in water-cooled copper hearth of the electron gun. Microscope glass slides were used as substrates. The substrates were first ultrasonically cleaned in alcohol, then rinsed in deionized water, and dried in nitrogen. The glass substrates and their masks were clamped to a stainless steel sheet inserted in a radiation heater with tungsten heating element. The substrate-target distance was 30 cm and the films were deposited on the substrates maintained at 100°C. The surface of CdS pellets were heated through bombardment of confocal electron beam with an accelerating voltage of 4.9 kV. Using a diffusion pump, the vacuum pressure reached below 10^{-5} torr prior to deposition. Deposition rates were controlled by controlling the current of electron beam. Two 700 nm-thick samples with different deposition rates of 12 nm/min and 60 nm/min were obtained. Deposition rates were measured by a quartz crystal monitor. Our results showed that the films with reproducible texture and properties can be obtained by using compressed pellets as the evaporation source, which are cheaper and more available than flow crystals [50].

A Park Scientific Autoprobe CP Atomic Force Microscope was used to quantitatively evaluate the surface topography of the thin films. AFM measured the changes in surface area and root mean square (RMS) roughness. Contact mode was used to capture the height images of each sample. The analyzed field was 2 μm × 2 μm at a scan rate of 1 Hz. ProScan Image Processing software was used to analyze the resultant 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 CdS thin films were obtained using a UV–VIS–NIR spectrophotometer (Perkin Elmer, Lambda 25).

X-ray diffraction patterns of the deposited films were recorded by a Philips PW 3710 X-ray diffractometer using Cu-Kα radiation ($\lambda = 1.5418A^{\circ}$) filtered through nickel filter with the instrument operating at V = 40kV at the interval $24^{\circ} \leq 2\theta \leq 72^{\circ}$ at a scan speed of 2°/minute giving a step size of 0.02.

3. Results and Discussion

Figure 1 shows typical AFM micrographs of 700 nm thick CdS films evaporated on glass substrates at two different deposition rates of 12 nm/min and 60 nm/min.

Three-dimensional AFM images show that the CdS films have grown as nanocone arrays. The mean height of these nanocones decreases from 40 nm to 12 nm as the deposition rate increases from 12 nm/min and 60 nm/min. The mean diameter of nanocones decreases from 80 nm to 50nm. The density of nanocones increases by increasing the depositions rate which results in the growth of a more packed film. Besides, the average roughness of the film surface on 2×2 μm^2 area decreases from 3.4 to 1.4 nm with increasing the deposition rate. These results suggest that the surface quality of the CdS film improves slightly with the increase in deposition rate. It is also well known that the window layer with smooth and continuous surface is highly desired in optoelectronic applications because such properties reduce the reflection loss due to roughness-induced surface scattering and will help minimize the number of conducting paths between the electrode and absorption layer which itself improves open circuit voltage of the device.

Figure 2 demonstrates the X-ray diffraction (XRD) patterns of resultant films prepared using electron beam deposition at different deposition rates of 12 and 60nm/min. X-ray patterns showed a strong preferential orientation with a single main peak at $2\theta = 26.7^{\circ}$, which corresponds either to hexagonal CdS (002) reflection or to (111) reflection of the cubic structure. Based on the CdS film band gap measurements which will be discussed, the deposited films are cubic in nature. Preferential orientations, as observed above, are probably due to the nucleation process associated with the deposition rate of CdS. The weaker intensities of the other planes arise owing to small crystallite size and hence a thorough structural analysis is difficult to make. 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 the films deposited at 12 nm/min and 60 nm/min deposition rates were found to be very close to the standard data given in PCPDF data file No. 01-075-0581 and 01-080-0019 respectively for cubic CdS phase.

The crystallite size was calculated by using Debye-Scherrer equation [7]:

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

Where *d* is crystallite size, λ is the 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 is found to be 23 nm and 30 nm for the films deposited at the deposition rates of 12nm/min and 60nm/min, respectively. The values for the crystallite size were found to be in nanometer scale. It indicates the nano-crystalline nature of resultant films deposited at different deposition rates. It is generally believed that the surface migration of adsorbed particles on substrates increases with increasing the substrate temperature and enhances grain growth and crystalline quality. In comparison to low deposition rate, in high deposition rate, the substrate temperature rises rapidly at the beginning of the deposition process, giving enough energy to the deposited atoms to diffuse and improve the crystallinity.

Figure 3 shows the transmittance (%T) curves for the CdS films deposited at different deposition rates of 12 and 60nm/min. The average transmission for films deposited at different deposition rates was found to be nearly 45%. It is obvious that the CdS films deposited at lower deposition rates show lower transmission. In other words, the transmission was found to increase with accelerating the deposition rate. It might be due to the proper crystallization of pure single phase of CdS at higher deposition rate which is consistent with the XRD results.

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:

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

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 the transmittance data, the absorption coefficient α was calculated (using the following equation [51]) 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 figure 4.

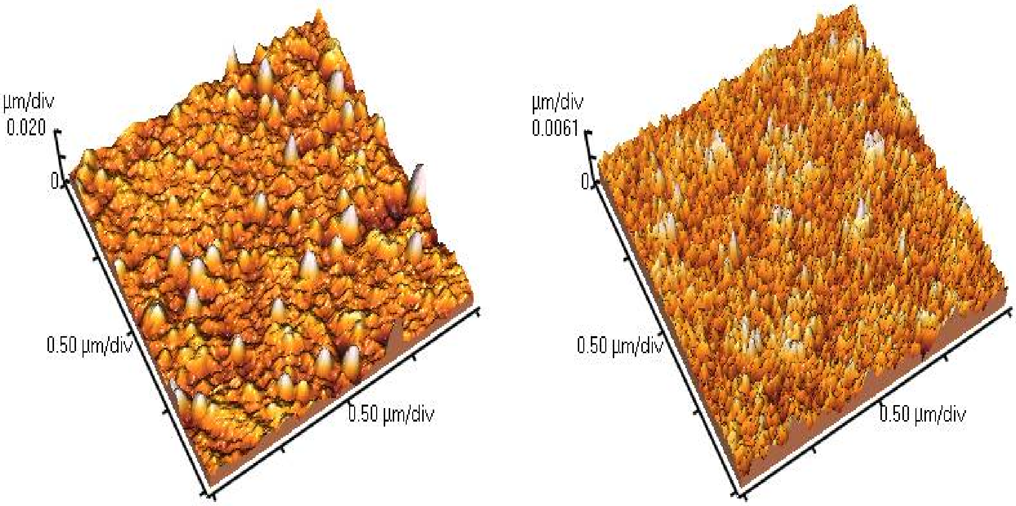

Figure 1. AFM images of 700 nm thick CdS films evaporated on soda lime glass substrates at 12 nm/min (right) and 60 nm/min (left) deposition rates in the contact mode. The scan size is 2 × 2 µm.

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

where t is the film thickness and T is the transmittance (%) of the films.

The energy band gap is 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 values of band gaps are found to be 2.32 and 2.34 eV for the films deposited at the deposition rates of 12 and 60nm/min, respectively. It is well known that CdS exists in either of two different structural phases, namely, a highly stable hexagonal phase and a metastable cubic phase. Cardona et al. have demonstrated that such phases have a band-gap energy differing by less than 0.1eV [52]. Therefore, our calculated values for band gaps are consistent with the band gap of cubic CdS.

Bandgap correlation to deposition rate could be attributed to the correlation of composition [50] to the deposition rate. Wilson et al. showed that at higher evaporation rates, there will be a greater tendency to form excess cadmium so that lower resistivity films whould result [50]. Thus, the CdS film deposited at a higher deposition rate has a composition with a lager Cd/S ratio. It was reported that the rise in the Cd/S ratio leads to a decrease in the band gap of CdS film [38, 53]

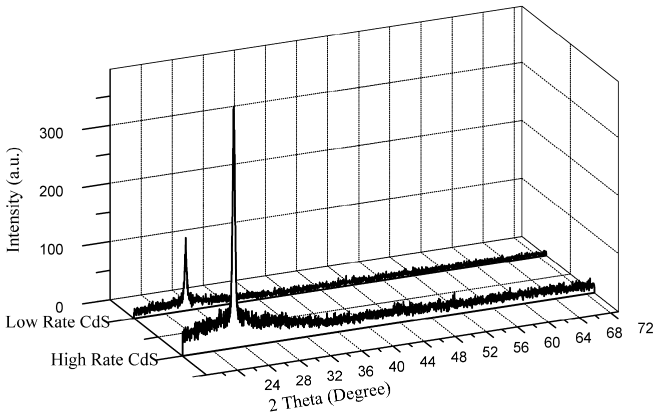

Figure 2. XRD pattern of CdS thin films deposited at high and low deposition rates.

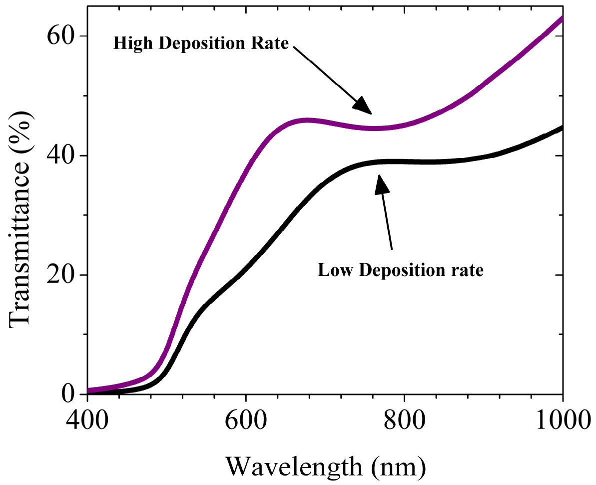

Figure 3. Optical transmittance spectra of the films deposited at two different deposition rates.

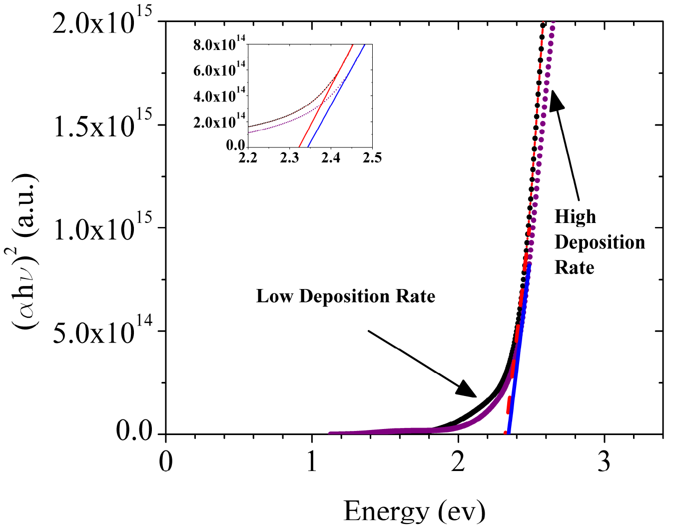

Figure 4. Tauc Plot of $(\alpha h\upsilon)^2$ versus $h\upsilon$ for CdS films deposited at two different deposition rates

4. Conclusions

CdS films deposited by electron beam evaporation were shown to have a polycrystalline hexagonal phase and exhibited preferred orientation along the (002) plane. Due to the enhancement of the nucleation rate and migration ability of adsorbed CdS particles as the substrate temperature increased, the crystalline quality of the CdS films improved at first. However, further increasing the substrate temperature reduced the film thickness, leading to a lower crystalline quality. So, it is important to control the growth temperature and film thickness to an appropriate value for the preparation of high quality thin films. It was found that the optical band gap of the CdS films exhibited fluctuation changes with increasing the substrate temperature and was attributed to the variation in Cd-composition and density of the disorder structure in the samples.

The structural, morphological and optical properties of CdS films deposited on glass substrates by electron beam evaporation as a function of deposition rate were investigated by XRD, AFM, UV-VIS Spectra. XRD studies showed that the films had a strong preferential orientation with a single main peak at 26.7°. Based on the XRD studies, the average crystallite size of the thin films deposited at 12 nm/min and 60 nm/min deposition rates are 23 nm and 30 nm, respectively. The larger crystallite size of the films deposited at higher deposition rate could be due to the temperature rise of the films at the beginning of the deposition process under the influence of the substrate intensive bombardment by particles.

The optical properties showed that the films were smooth, free from pin holes with transmittance of ~45%. Optical studies showed that the films deposited at higher deposition rate were more transparent than those deposited at lower deposition rate. The more transparency of films deposited at the higher rate could be the result of larger grain size of the films. During light passage through films with larger grain size, it encounters less grain boundary and therefore the scattering diminishes with respect to films with smaller grain sizes. The optical band gap increased due to the change in Cd/S ratio by increasing the deposition rate. The rise in the Cd/S ratio led to a decrease in the band gap of CdS film. The band gap of deposited films was consistent with the band gap of cubic CdS.

It was found that the RMS roughness of CdS films decreases with increasing the deposition rate. The film produced at a higher rate exhibited narrower nanocones protruding out from the film surface. The density of the nanocones increased with an increase in the deposition rate leading to the fabrication of a more uniform film. In conclusion, we have shown that increase in the deposition rate promotes the smoothness of films and improves the crystallinity and optical transparency of the CdS films.

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