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Effects of the Surface Roughness on Optical Properties of CdS Thin Films

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The effects of surface roughness on the optical properties of CdS thin film was investigated using RMS roughness measurements. CdS thin films for a window layer of the CdTe thin film solar cell were grown on ITO glass by a chemical bath deposition method with varying thiourea ($CS(NH_2)_2$) mole concentration. The surface roughness of the CdS thin film was measured by Atomic Force Microscopy (AFM). After obtaining the correlation factor between the RMS roughness and the difference in the diffused transmittance and the spectral transmittance by using the integrating sphere, we investigated the effect of surface morphology on the transmittance in all CdS thin films. It is obtained that the higher RMS value the specimen has, the more difference between the diffused transmittance and the spectral transmittance is measured. Moreover, the difference is evident at the long wavelength region.

Keywords CdS; diffuse; optical property; roughness; spectral; thin films

Introduction

In the thin film solar cell devices, either $Cu(In,Ga)Se_2$ (CIGS) in the group of I-III-VI type quaternary compounds or CdTe in the group of II-VI type binary compounds is considered as one of the most critical light absorbing layers to affect the solar cell efficiency. In CIGS thin film solar cells, CdS has been used as a buffer layer functioning to reduce the effective density of holes at the interface and thereby the recombination of the electron hole pair [1]. In CdTe thin film solar cells, CdS plays an important role of the n-type window layer to form a heterojunction with the p-type CdTe absorber layer. CdS/CdTe heterojunction was first proposed with less than 5% efficiency cell performance [2]. In CdS/CdTe heterojunction solar cell, thicker CdS layer is assured to yield the lower transmittance. In addition, as one

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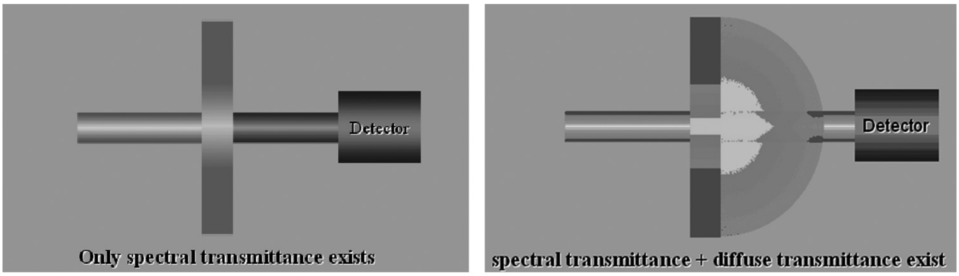


(a) (b)

Figure 1. Schematic diagram comparing (a) spectral transmittance and (b) spectral transmittance + diffused transmittance.

makes thinner CdS films, the probability to make a short circuit between the CdTe layer and the front contact increases. In order to prepare the transparent and highly resistive CdS thin films with a good conformal coverage, there are various deposition techniques such as chemical vapor deposition, electro-deposition [3], spray coating [4], RF sputtering [5] and chemical bath deposition [6–9]. So far, the chemical bath deposition has been known to be the most promising solution-based method to fabricate the CdTe solar cell. Chemical bath deposition mechanism is originated from the decomposition of the thiourea, $CS(NH_2)_2$, in an alkaline solution containing a cadmium salt. This process is based on the controlled precipitation of the cadmium sulphide in the reaction bath. The solubility product of cadmium sulphide is very small, about 10^{-25}, and, therefore, the precipitation has to be controlled by the concentration of free Cd^{2+} in the bath. It can be achieved by using suitable complexing agents, which release small concentrations of ions depending upon the complex ion dissociation equilibrium and the corresponding equilibrium constant [10–11].

The transmittance property of the deposited CdS thin film is one of the critical factors for the n-type window layer of the CdTe solar cell because the light should be passed through the CdS thin film before reaching the CdTe absorber layer. In transmittance, there are a spectral type and a diffuse type as shown in Figure 1. If the surface morphology is quite smooth, there is no diffused light through the specimen. The roughness of surface can contribute to the extent of the diffused light. We investigated the relation between the surface roughness measured by Atomic Force Microscopy (AFM) and the difference in the spectral transmittance and the diffused transmittance measured by UV-VIS-Nir spectrometer.

Experimental

We prepared the CdS thin films on the commercial ITO glass as a window layer of the CdTe thin film solar cell with equipment configuration as shown in Figure 2. We used a commercialized heating mantle to control the temperature of the reaction solution using a programmable PID control via a reading temperature sensor embedded in heat jacket. Additionally, in order to confirm the actual temperature of solutions, we used a thermocouple sensor directly immersed in the reaction solution. DI water, 0.3 mM $CdSO_4$, and 0.1∼0.4 M thiourea were mixed in the deposition bath. As the temperature approaches to a set point for the experiment,

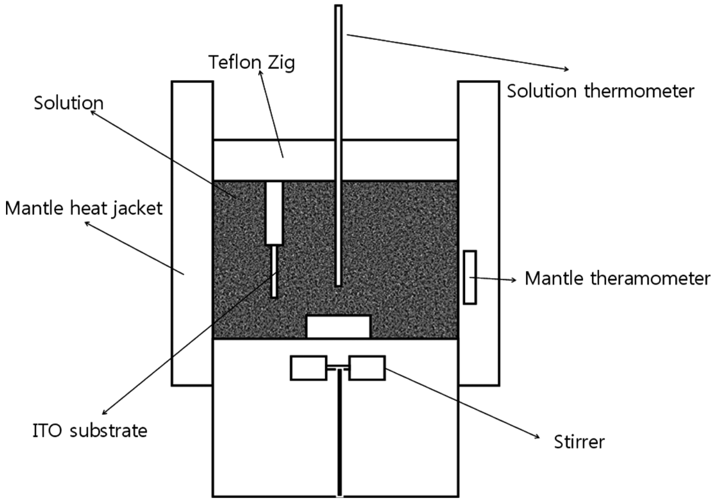


Figure 2. Schematic diagram to describe the configuration of chemical bath deposition method.

ammonia water (25 wt%) was added to the solution to maintain the solution as PH 10–11. PID controlled reaction bath controls the temperature deviation within ±1°C. In the beginning after adding ammonia water, the solution became from opaque liquid to yellow-green color. At this point, ITO substrate attached to teflon zig was immersed into the bulk solution. The exact distance from bottom of beaker and the vertical direction of substrate was maintained parallel with gradient of stirrer's motion within all experiment.

Structural properties of CdS thin films were investigated by using XRD (Philips X pert pro X-ray diffractometer) where the primary wavelength is Cu-Kα1 1.5406A. Optical transmittance measurement was obtained by using the UV-VIS-Nir spectrometer (Perkin Elmer Lambda 950 model) where a 150 mm sphere was equipped for the spectral transmittance. Surface morphology and the surface roughness were measured by using AFM (Digital instruments NanoScope III) with tapping mode. We utilized SEM (Hitachi S-4200 model) to inspect the surface morphology.

Results and Discussion

As shown in Figure 3(a), XRD data shows the structural property of CdS thin film on ITO glass prepared by the chemical bath deposition method with varying thiourea mole concentration. Both hexagonal crystalline phase and cubic phase are observed in the cadmium sulfide thin films prepared using the CBD method. The peak at $2\theta = 26.7°$ is related to the mixture of hexagonal (0 0 2) and cubic (1 1 1) planes. In order to determine the crystalline structures of CdS films in detail, the rectangular shaped region in Figure 3(a), which is in the spectral range of $2\theta = 24°–29°$, was enlarged as shown in Figure 3(b). It was observed that the prepared CdS thin films consist of the hexagonal structure and cubic crystal structure and that the primary peaks are slightly shifted depending on the thiourea mole concentration. And in order to investigate the optical property of specimens under different thiourea

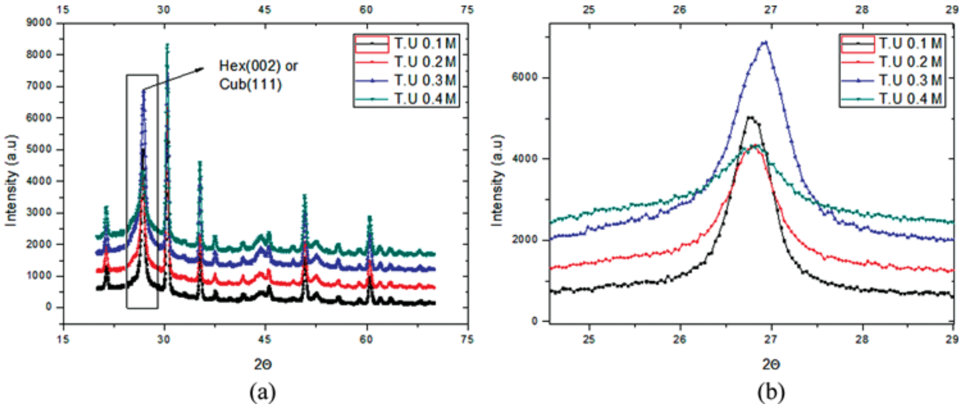


Figure 3. XRD data of CdS thin film depending on thiourea concentration scaled with 2θ (a) 20–70° and (b) 24–29°.

concentrations, UV-VIS-Nir transmittance measurement is done. Figure 4(a) represents the regular transmittance data and Figure 4(b) represents the diffused transmittance using the 150 mm integrating sphere. As shown in Figure 4, the large difference in transmittances between the regular measurement (a) and the integrating sphere measurement (b) at thiourea 0.1 M concentration condition was observed. This fact implies that there were lots of diffused light through CdS thin films which were grown at thiourea 0.1 M concentration. Even if we have no tangible evidences, we inferred that the diffused light is probably originated from the non-uniform surface morphology. Figure 5 shows that AFM data depending on the thiourea mole concentration from 0.1 M to 0.4 M. The RMS value of the specimen grown on the condition thiourea 0.1 M is two times higher than other specimens. Correlation about transmittance difference in the spectral mode and the diffused mode and the surface roughness is introduced in Table 1. The 3rd and 4th columns represent the summation of the transmittance difference within wavelength from 550 to 900 nm and from 200 to 550 respectively. In other words, the 3rd and 4th columns

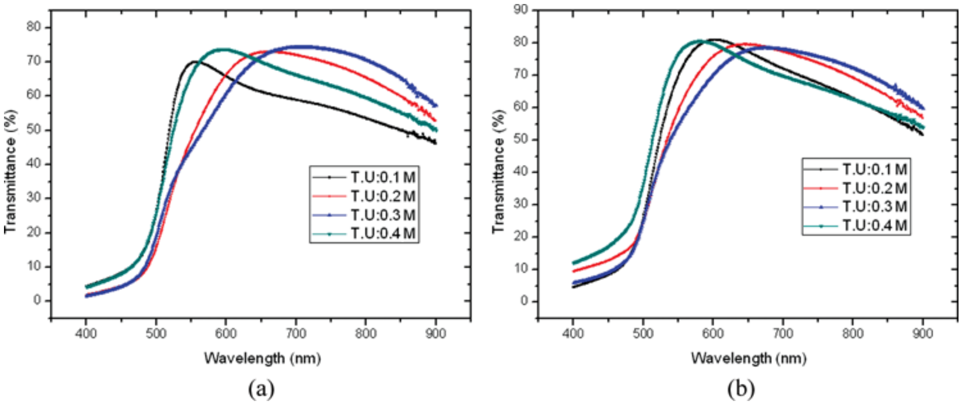
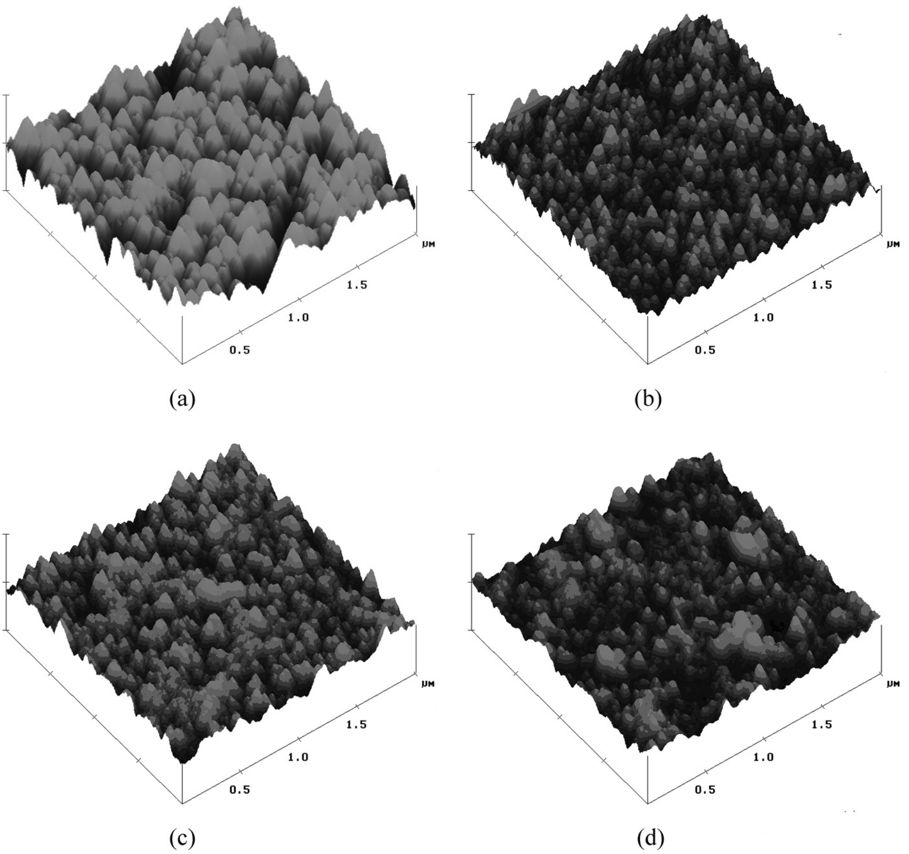


Figure 4. Transmittance of CdS thin films by (a) regular measurement and (b) integrating sphere.



(a) (b)

(c) (d)

Figure 5. AFM images of the CdS thin film in the solution with (a) [thiourea]: 0.1 M, rms:14.138 (b) 0.2 M, rms:7.814 (c) 0.3 M, RMS: 7.576 and (d) 0.4 M, rms:7.867.

represents the summation of the quantity of the net diffused light ($T_d - T_s$) in the long and short wavelength range respectively, where T_d and T_s is the transmittance using integrating sphere and using spectral measurement mode. Interestingly according to Table 1, the difference of transmittance in the spectral mode and the diffused mode is a minimum at short wavelength range in the specimen deposited under

Table 1. Correlation about the transmittance difference between the spectral mode and the diffused mode and the surface roughness where T_d is the diffused transmittance and T_s is the spectral transmittance respectively

Thiourea mole concentration (mole)	Surface roughness (nm)	$\sum_{\lambda=550}^{900} T_d - T_s$	$\sum_{\lambda=200}^{549} T_d - T_s$
0.1	14.14	3863	−131
0.2	7.81	2072	1374
0.3	7.58	1596	898
0.4	7.87	1511	1468

thiourea 0.1 M condition. However at the long wavelength region, the difference in the spectral mode and the diffused mode is the maximum value in the specimen deposited under thiourea 0.1 M condition.

We also obtained the optical bandgap information from the transmittance measurement. From these spectral data optical absorption coefficient α, was calculated using Lambert's law [12]

$$\ln\left(\frac{I_0}{I}\right) = 2.303A = \alpha d \tag{1}$$

where I_o and I are the intensity of incident and transmitted light, respectively, A is the optical absorbance, and d is the film thickness. As CdS is a direct band-gap semiconductor, the band gap energy can be calculated from the well-known relation [13],

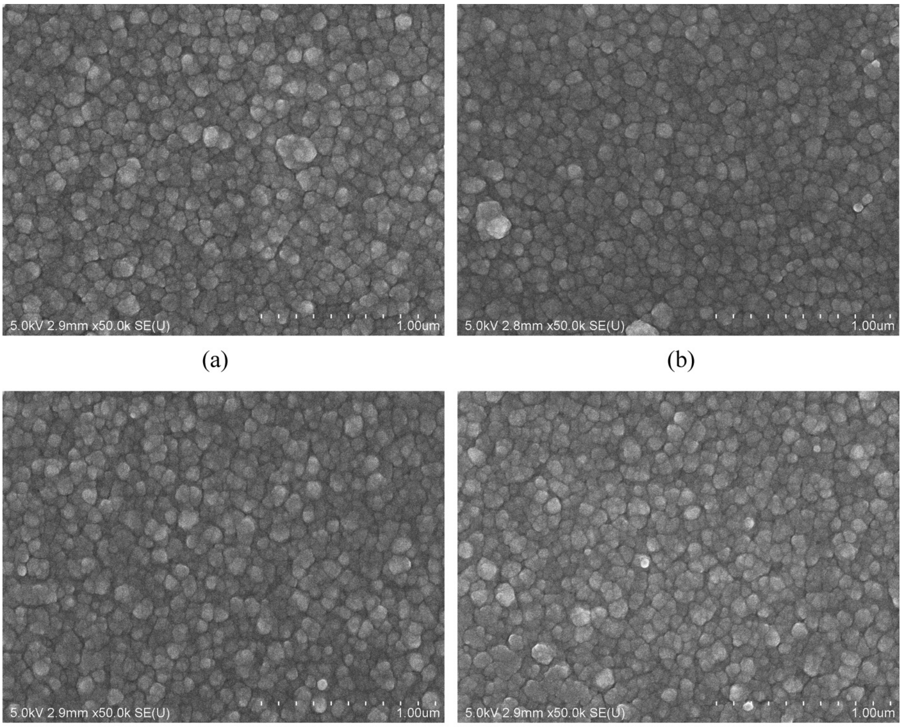
$$\alpha h\upsilon = [k(h\upsilon - E_g)^{\frac{1}{2}}] \tag{2}$$

where k is a constant, $h\upsilon$ is the photon energy, and E_g is the band gap energy. For $h\upsilon > 2.4\,\mathrm{eV}$, the plot of $(\alpha h\upsilon)^2$ as a function of $h\upsilon$ follows a straight line (fundamental absorption). Therefore, in this range the absorption spectrum can be described by Eq. (1). The least-square fit to the linear part gives the optical band gap E_g. The non-linear steps in the energy range $h\upsilon < 2.4\,\mathrm{eV}$ (residual absorption) can be due to multiple-reflections that take place at the surface and the backside of the film. The value of the energy bandgap estimated from the plots for film deposited at varying thiourea mole concentration is from 2.38 to 2.41 eV as shown in Table 2. Also the energy bandgap difference between the spectral mode and the diffused mode is shown in Table 2. The minimum energy bandgap difference is acquired from the maximum surface roughness specimen of thiourea 0.1 M concentration. Although their estimated optical band gap differences between the spectral mode and the diffused mode are too small to confirm their results, this result is in good agreement with the transmittance difference between the spectral mode and the diffused mode at short wavelength (<550 nm) region as shown in Table 1.

We also obtained the SEM images to discover the particular morphology property to supplement our subject which is the correlation between the specimen's roughness and the transmittance property. Figure 6 represents the SEM image of the specimen. As shown in Figure 6, there is no peculiar image to be connected with our discussion.

Table 2. Bandgap energies estimated from diffuse and spectral mode under different thiourea concentrations

Thiourea mole concentration (mole)	Bandgap energy from diffuse mode (eV)	Bandgap energy from spectral mode (eV)	Surface roughness (nm)
0.1	2.40	2.41	14.14
0.2	2.35	2.38	7.81
0.3	2.39	2.41	7.58
0.4	2.39	2.41	7.87



(a) (b)

(c) (d)

Figure 6. SEM image of the CdS thin film in the solution with (a) [thiourea]: 0.1 M (b) 0.2 M (c) 0.3 M and (d) 0.4 M.

Conclusions

We investigated the relationship between surface roughness measured by Atomic Force Microscopy (AFM) and the difference in the spectral transmittance and the diffused transmittance measured by UV-Vis-Nir equipment. Although the quantity of light spectrally transmitted through CdS thin film is small because of large surface roughness, it can hardly affect the quantity of light to reach at CdTe absorber layer. Therefore the reasonable measurement method of transmittance of CdS thin film is to use the integrating sphere to be able to measure the diffused light as well as the spectrally transmitted light. However, in order to fabricate high efficienct CdTe solar cell, we should consider the interface property which can be affected by the surface morphology of CdS thin films. We are studying the dependency of RMS value of CdS thin films to have an effect on the efficiency of CdTe solar cell.

Acknowledgments

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