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Enhancement of Optical Properties of Cds Thin Films Prepared From Sputtering Technique: Effect of Thermal Annealing

[1]M. A.Islam, [1]M.S. Hossain, [1]K.S. Rahman, [1]F. Haque, [1]M.Y. Sulaiman, [1]K. Sopian, [1,2]Nowshad Amin

[1]*Solar Energy Research Institute (SERI)*
[2]*Universiti Kebangsaan Malaysia, Department of Electrical, Electronic and System Engineering,Faculty of Engineering and Built Environment, 43600 Bangi, Selangor, Malaysia*

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ABSTRACT

Background: The CdS thin films were deposited on ITO coated glasses using sputtering technique at different substrate temperatures and subsequently annealed at 350 °C in an O_2/N_2 ambient with the variation of time. **Objective:** The structural and optical properties of thermally annealed CdS and as-grown CdS thin films were examined from X-ray diffraction lines and UV-Vis spectrometry. **Results:** The X-ray diffraction pattern indicates that the annealed films show the transition tendency from poly crystalline to amorphous or mixed phase. However, the film deposited at room temperature have found completely in an amorphous form after thermal annealing. **Conclusion:** The band gaps of the films were found to be in the range of 2.48 to 3.10 eV. The annealed films also exhibited the blue shift in the direct allowed transition energy band gaps which leads the change of crystalline sizes.

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INTRODUCTION

Cadmium sulphide (CdS) is one of the most promising II-IV compound semi-conductor materials because of its wide range of applications in different kind of hetero-junction solar cells such as cadmium telluride (CdTe), copper indium diselenide/sulphide (CIS) and copper indium gallium diselenide/sulphide (CIGS) solar cells (Dobson, K.D., 2000). It also has applications in various electro-optic devices and infra-red detectors. Typically, CdS has a band gap of 2.42 eV, permit only light with wavelength greater than 510 nm to pass through it to the absorber layer that attributes 20% of the efficiency loss. Moreover, it can absorb 63% of the light incident on it with only one micrometer thick layer (Toshiro Maruyama, Ryota Kitamura, 2001). The efficiency of CdTe and other hetero junction solar cells can effectively be improved by increasing its band gap. It has been reported that the CdS layer band gap is increased by thermal annealing in O_2 environment and enhance its optical and electrical properties (Chu, T.L., 1992). In an attempt to develop an efficient, stable and low-cost CdTe solar cell, it is a crucial need to develop a higher bandgap window material to avoid the above adverse effects on window-absorber hetero-junction.

In this work, the higher band gap CdS thin films are developed for CdTe solar cells by post deposition thermal annealing process. The structural and optical properties of annealed CdS thin films also have determined and compared with the as-deposited CdS using XRD and UV-Vis spectrometry. CdS films were prepared by sputtering on ITO coated glasses at different substrate temperatures and then annealed in O_2/N_2 (partial pressure, 1:99) ambient for different times. This annealing show a conversion trend of polycrystalline CdS to oxygenated amorphous cadmium sulphide (CdS:O) and increase the band gaps which also have been reported by other researchers (Osuwa, J.C., 2009).

Experimental Procedure:

CdS thin films of 200 nm were sputtered on ITO coated glasses by varying the substrate temperatures from room temperature (RT) to 300°C. The prepared CdS thin films were then thermally annealed in a closed furnace by varying the time. The chamber pressure of gas was kept from 250 to 300 Torr during annealing for all samples. At the end of the annealing process, the samples were left in the annealing chamber until their temperatures returned to room temperature. Table 1 illustrates a summary of the sample preparation processes.

Corresponding Author: M.A. Islam, Solar Energy Research Institute (SERI).
Tel: +6-01111223686; E-mail: aminbgm@yahoo.com

Table 1: Annealing processes for the sputtered CdS samples prepared on ITO coated glasses.

Sample's ID	Substrate Temperature [^oC]	Annealing temperature [^oC]	Time of Annealing
1	RT	350	15 min.
2	100	350	15 min.
3	150	350	15 min
4	200	350	15 min
5	300	350	05 min
6	300	350	15 min
7	300	350	25 min
8	300	as deposited	-

The structural properties of the films were investigated using X-ray diffraction (XRD) spectroscopy. The XRD patterns were recorded in the 2θ range 20^o to 60^o using Cu Kα radiation of wave length λ = 1.5408 Å. The grain sizes, dislocation densities and micro-strains of the films were observed from the XRD spectra. The optical properties of CdS layer were measured by using UV-Visible spectrometer. A similar glass slide was used as a reference to get the actual optical absorption. The optical band gaps were calculated from their absorption edge extrapolated from the absorption spectra.

RESULT AND DISCUSSION

XRD Spectra:

X-ray diffractograms (XRD) of the CdS thin films are taken on a 'BRUKER aXS-D8 Advance Cu-Kα'diffractometer. The used radiation wave length, λ= 1.52 Å and a Nickel filter was used to block Kβ. The XRD spectra found from diffractometer are shown in Fig. 1(a) and Fig. 1(b). The spectra of Fig. 1(a) are for the samples sputtered at substrate temperatures of RT, 100^oC, 150^oC, 200^oC and were annealed at 350^oC for 15 minutes. In Fig. 1(b), the spectra are for the samples that were sputtered at substrate temperature of 300^oC and were annealed at 350^oC for 5, 15 and 25 minutes, respectively and also shown the as-grown sample. The spectrum of the as-grown film shows a broad peak at 2θ = 26.65^o attributed from the (002) crystalline planes of the hexagonal CdS. It has been observed that there are some additional peaks in the spectra at 28.2^o, 35.5^o, 43.8^o, 47.8^o and 52.2^o corresponding to the (101), (102), (110) crystalline planes, respectively. The peaks at 2θ = 30.5^o and 50.5^o do not belong to CdS but, they are attributed to ITO. There is no peak have found for the film deposited at RT which means it is completely in an amorphous form. In Fig. 1(b), the most prominent peak decreases as the annealing time is increased, indicating that the decrease of the crystalline size and gradual conversion to amorphous form (Jayakrishnan, R., 1996)]. Therefore, the effect of thermal annealing is clearly observed from the spectrum of the different annealed CdS thin films. In Fig. 1(a), the change in the physical structure of the CdS is more pronounced for samples of lower substrate temperature. By examining whole patterns of Fig. 1(a) and 1(b), it can be concluded that the annealed films contained both cubic and hexagonal structures as a mixture and these crystallites are surrounded by amorphous phases.

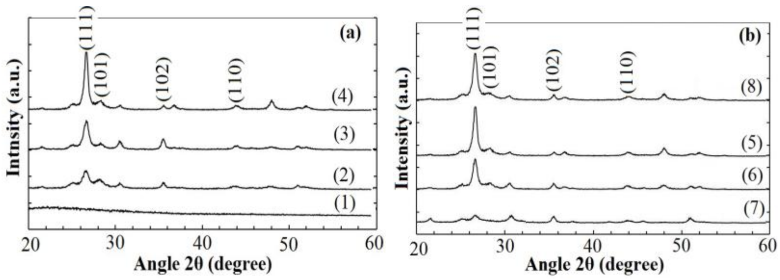

Fig. 1: XRD spectra of CdS thin films (a) prepared at different substrate temperature and annealed for 15 min, (b) prepared at 300 ^oC and annealed for different duration of time.

The average crystalline size was calculated from the recorded XRD patterns by using Debye-Scherrer formula [6] and micro strains (ε) and dislocation densities (δ) (Islam *et al*. 2013) were also calculated for the films. The calculated values are putted in Table 2. It has been found that the dislocation densities are increased with the increase of substrate temperature as well as increase of the duration of thermal annealing time.

Optical Studies:

The energy band gaps of both as-grown and annealed films were studied by optical transmission and absorption using 'Lambda 900 UV/Vis/NIR' spectrometer. The absorption spectra are shown in Fig. 2(a) and 2(b) which was recorded for wavelengths ranging from 300 to 800 nm. It has been observed that the increase of the film's transmittance does not depend only on the substrate temperature but also on the annealing time. Moreover, the films show a blue shift in the wavelength range from 450 nm to 550 nm as indicating by arrows in the Fig. 2.

Table 2: FWHM, crystalline size, dislocation density and strain of thermally annealed CdS thin films.

Sample's ID	FWHM (radian)	Crystalline size [d](nm)	Ratio of hkl (111)/(110)	Dislocation density [ε] (x 10^{-3})	Microstrain [δ] (x 10^{-3})
1	-	-	-	-	-
2	0.00960	14.667	10.00	9.34	1.64
3	0.01048	13.435	15.28	9.73	1.86
4	0.01135	12.406	21.79	10.83	2.25
5	0.00960	14.667	25.07	10.34	1.82
6	0.00909	15.505	16.26	8.84	1.47
7	0.00956	14.677	5.00	9.34	1.64
8	0.00908	15.507	18.09	8.84	1.46

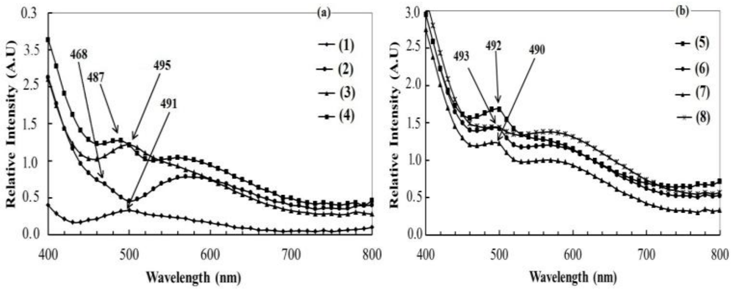

Fig. 2: UV-Visible absorption spectrum of annealed CdS thin films (a) prepared at different substrate temperature and annealed for 15 min, (b) prepared at 300 ^{o}C and annealed for different duration of time.

The values of the optical band gap energies (Eg) were obtained by extrapolating the lines to the hγ-axis at $(\alpha h\gamma)^2 = 0$ (Hossain, M.S., 2012) and found to be between 2.48 and 3.10eV. These results are summarized in Table 3. It is noted that the band gap increases with the increase of annealing time. In the case of fixed annealing time, the band gap increases with the decrease of the substrate temperature during the RF-sputtering.

Table 3: Band gaps of thermally annealed CdS thin films found from UV-Vis.

Sample's ID	Sub. Temperature [^{o}C]	Annealing time	Band gap [eV]	Shift in Band gap [eV]
1	RT	15 min.	3.1	0.68
2	100	15 min.	2.89	0.47
3	150	15 min.	2.88	0.46
4	200	15 min.	2.84	0.42
5	300	05 min.	2.73	0.31
6	300	15 min.	2.80	0.38
7	300	25 min.	3.03	0.61
8	300	as grown	2.48	0.06

By examining the α-hγ values, it has been found that the films deposited at the higher temperature (300 ^{o}C) have the higher 'α' values at the forbidden band gap region than the film deposited at the lower temperature. However, It has been seen that Urbach tailing seems little steeper for the films deposited at the low temperature. The films deposited at the higher temperature have a higher 'α' value in the band to band absorption region, although the films show a larger band gap. The absorption increased at the band to band region may be explained by proposing that the crystallite size of the films become smaller as it was thermally annealed as observed in Table 2.

Conclusion:

The post deposition thermal annealing effects on sputtered CdS thin films are elaborately explained in this work. The XRD spectroscopy shows that the resulting films show structural transition trends from polycrystalline to amorphous whereas the film deposited at RT have found in an amorphous form. This transition trend is increases by the increase of annealing time and with the decrease of substrate temperature.

Thermal annealing increases the band gap of annealed CdS as it was the main objective of this work. Finally, the band gap has been found for annealed CdS thin films are 3.1eV and 3.03 eV for deposition temperature RT and 300 ^{o}C, respectively with annealed at a fixed temperature 350 ^{o}C. As the band gap of the films is increased, the prepared films would be suitable as a window material for higher efficiency solar cells.

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