



Effects of deposition temperature and CdCl₂ annealing on the CdS thin films prepared by pulsed laser deposition



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ABSTRACT

Pulsed laser deposition (PLD) technique is suitable for the deposition of high-quality compound semiconductor thin films, and has been widely developed in recent years. However, pulsed laser deposition of CdS films has rarely been reported. In this work, we prepared CdS thin films using PLD. The effects of growth temperature on the PLD-CdS thin films were studied towards high-performance CdS/CdTe thin film solar cells. Results showed that the CdS film prepared at 400 °C has the best crystallinity and optical transmittance, while 200 °C is more suitable for the window layer of CdTe solar cells due to the highest energy conversion efficiency and the best short-wavelength response. CdCl₂ annealing treatment was also employed on the 200 °C-deposited and 400 °C-deposited PLD-CdS layer. Annealing treatment further enhanced crystallinity, and obviously enlarged the grain size. Optical transmittance spectra showed that the band gap of the CdS films increased after annealing. Fermi level of CdS films shifted closer to the conduction band from XPS analysis. CdTe solar cells with annealed windows obtained further improved performance, including higher short-circuit current, open-circuit voltage and energy conversion efficiency.

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1. Introduction

With a direct band gap of 1.46 eV and high absorption coefficient ($>10^5 \text{ cm}^{-1}$), CdTe is a promising semiconductor material for high-performance thin film solar cell [1]. The highest energy conversion efficiency of CdTe solar cell is predicted to be 28% [2]. N-type CdS is commonly utilized as window layer for p-type CdTe solar cell due to its wide band gap (~2.4 eV) [3,4]. Generally, CdS polycrystalline thin film is deposited using chemical bath deposition (CBD) [5–8]. However, the CBD method has several disadvantages. For example, the deposition process is very slow, and it produces considerable liquid waste that needs to be recycled [9]. In this paper, we attempt to prepare CdS polycrystalline thin films using pulsed laser deposition (PLD).

PLD is a physical vapor deposition technique which has been widely developed in recent years. During the PLD process, a pulsed laser beam is focused to strike the target of a specific material, and

the material is vaporized from the target (in a plasma plume) and deposited as thin film on a substrate. PLD technique has numbers of advantages. It doesn't generate waste by-product, compared to the CBD method. Another advantage of PLD for the growth of thin films is an extremely smooth film surface compared to films grown by close-space sublimation (CSS) and RF magnetron sputtering. In addition, the relatively higher kinetic energy of atoms in PLD generated plasma plume renders them higher mobility on substrate surface, enabling lateral growth at relatively lower substrate temperatures than other physical vapor deposition methods [10]. Also, the film thickness can be controlled to a single atomic layer by fine adjusting the pulse number. Those unique merits make PLD suitable for the deposition of various high-quality thin films [11]. However, PLD technique has rarely been reported on the preparation of CdS films. So we first investigated the influence of different deposition temperatures on the physical properties of PLD-CdS thin films, since the optical and structural properties of the thin films prepared by PLD have a strong correlation to the deposition temperature [12].

However, the as-deposited CdS polycrystalline thin films prepared by PLD were composed of nano-scaled grains, which severely

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affect the performance of CdS/CdTe solar cells [13]. A post anneal process in CdCl_2 was then applied to improve the crystalline quality through a recrystallization process. In fact, improved physical properties of CBD-CdS films were reported previously after the CdCl_2 annealing [13,14]. A similar trend is observed on PLD-CdS films, and the annealed CdS film has a much lower density of defects, better crystalline and larger grain size. In this paper, we have studied the influence of annealing treatment on the properties of PLD-CdS films and the performances of CdTe solar cells with annealed CdS windows.

2. Experiment

CdS thin films were deposited in a vacuum chamber using 248 nm KrF excimer laser (Lambda Physic COMPEX201). A CdS target (purity 99.99%) was mounted on a rotating target stage. The substrates were 50 mm $\times$ 30 mm commercial fluorine-doped tin oxide (FTO) glass.

Four CdS thin film samples were fabricated at room temperature, 200 °C, 300 °C and 400 °C, respectively. The vacuum chamber pressure was maintained $\sim 10^{-6}$ Torr during the entire deposition process. The laser pulse energy and repetition rate were 90 mJ and 6 Hz, respectively. The thickness of these four samples was all 100 nm. X-ray diffraction (XRD) patterns were recorded with Dandong Fangyuan DX-2600 diffractometer using $\text{CuK}\alpha$ radiation and optical transmittance spectra were measured using an UV/Vis spectrophotometer (Perkin Elmer-Lambda 950).

CdTe solar cells based on these four PLD-CdS layers were then fabricated by the following process. First the CdTe layers were prepared onto CdS window layers by close-space sublimation (CSS) under the mixture gases of Ar and O_2 at 500–600 °C. During this process, the pressure (Ar + O_2) was maintained at 3 kPa. About 2 μm -thick CdTe polycrystalline layer was deposited. Then these four samples were annealed in CdCl_2 atmosphere at 385 °C for 30 min and subsequently etched in bromine-methanol solution. The cells were completed with the evaporated Cu-doped ZnTe back contact layer and Au electrodes before characterization of the solar cell performance.

In addition, PLD-CdS thin films prepared at 200 °C and 400 °C were annealed with CdCl_2 to extract the effects of the CdCl_2 annealing on the physical properties of the window layer before deposition of the CdTe absorber layer. CdCl_2 was ultrasonic-sprayed onto the surface of CdS layer initially. Then CdS layer was annealed in a tube furnace at 400 °C for 30 min with a flow of 20 sccm N_2 and 20 sccm O_2 . The annealed CdS film samples were characterized by XRD, optical transmittance spectra, SEM and XPS. CdTe solar cells were then prepared on the annealed CdS window layer, followed with characterization of their current density–voltage (J–V) curves.

3. Results and discussion

1. Effects of deposition temperature

The XRD spectra of four window layer samples made at different temperatures were depicted in Fig. 1 together with that for FTO for comparison. All the CdS layers are in the hexagonal phase, which is desirable for the window layer of high efficiency CdS/CdTe solar cells. On CdS samples deposited at room temperature and 200 °C, the three peaks located at 26.5°, 43.6° and 47.7° are respectively associated with the (002), (110), and (103) planes, and the others belong to the FTO substrate. When the temperature increases to 300 °C and 400 °C, the (110) peak is disappeared. Also, as the deposition temperature rises, the diffraction peaks become more intense and sharper, indicates better crystallinity in the CdS film. In

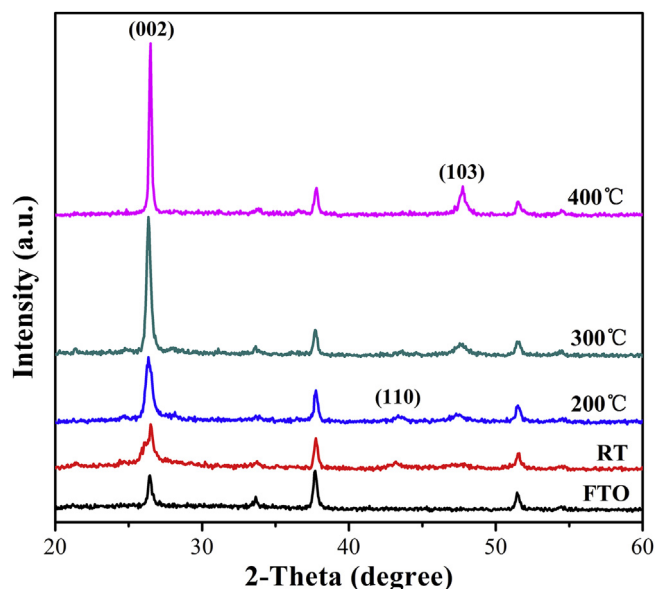


Fig. 1. X-ray diffraction pattern of PLD-CdS films prepared at various temperatures together with the spectrum of FTO.

addition, the volume portion of the (002) phase is much higher with the CdS growth temperature since the (002) peak intensity increases much more than other XRD peaks at higher deposition temperature.

Fig. 2 shows the optical transmittance spectra of these four CdS samples which have the same thickness of 100 nm. The transmittance of CdS deposited at room temperature is poor ($\sim 60\%$) within the visible range. With increasing deposition temperature, the optical transmittance increases considerably, which is accompanied by the blue shift of the absorption edge, indicative of widened band gap. Among these four samples, the CdS prepared at 400 °C hence has the best optical property as the window layer for a solar cell.

The performance of the CdTe solar cells was tested using a Solar Cell Tester (G solar XJCM-9) under AM1.5. Fig. 3 shows the box charts comparing the results of energy conversion efficiency, fill factor (FF), short-circuit current (J_{sc}) and open-circuit voltage (V_{oc})

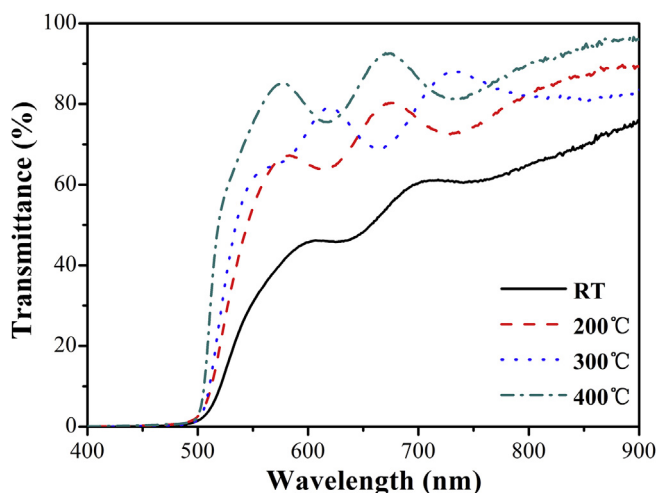


Fig. 2. Optical transmittance spectra of PLD-CdS films prepared at various temperatures.

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