



Fabrication of high efficiency sputtered CdS:O/CdTe thin film solar cells from window/absorber layer growth optimization in magnetron sputtering



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ARTICLE INFO

Keywords:

Thin film solar cell, CdTe, Sputtering

CdS:O thin films

XPS

PL

Conversion efficiency

ABSTRACT

In this study, CdTe (up to 2.0 μm thick) and oxygenated CdS (CdS:O, up to 100 nm thick) films were deposited by magnetron sputtering and optimum conditions of film growth were investigated for CdS:O/CdTe solar cells. Favourable TeO_2 has been confirmed in XRD after the CdCl_2 heat treatment of the CdTe films. Moreover, improved structural, optical and electrical properties are observed in the CdCl_2 heat treated films. A detailed quantitative study has also been executed using XPS that finds sulfide, sulfate and an intermediate oxide as a function of oxygen content. In many cases, CdS contribution remains predominant, however, the CdSO_x contribution increases with the increase of oxygen's partial pressure and decrease of growth rate. The complete solar cell device was fabricated of various CdTe thin films with different growth rates in sputtering. A highly resistive transparent (HRT) buffer layer ZnO:Sn was placed in between the FTO and CdS:O to avoid the forward leakage problem and screen printed C:Cu/Ag is used as the back contact for low cost fabrication. The J-V characteristics and external quantum efficiency (EQE) were measured for the solar cells under the illumination of AM 1.5G and the highest efficiency of 10.3% was achieved for the optimized CdTe growth rate of 5.4 $\text{\AA}/\text{s}$, while CdS:O growth rate was 0.25 $\text{\AA}/\text{s}$.

1. Introduction

CdTe is the most promising and leading material among all the thin film solar cells' absorber materials. The conversion efficiency as well as the commercial implication of CdTe photovoltaic (PV) technology have been growing rapidly over the past decade. A multi-giga-watt/year production has already been achieved by CdTe thin film photovoltaic manufacturers attributed to achieved high efficiency and lower fabrication cost. Cadmium Telluride (CdTe), owing to its direct optical bandgap ($E_g = 1.5$ eV) and higher absorption coefficient of around $5 \times 10^5/\text{cm}$, has already been recognized as the most promising compound semiconductor for solar PV application [1]. CdTe thin film solar cell efficiency has already reached to 22.1% in laboratory [2] and 17.5% in module scale [3]. However, to be a sustainable photovoltaic material, the cost should be reduced further possibly through the reduction of material usage, cell processing and balance of system costs. Therefore, reducing the thickness (≤ 2 μm) of CdTe absorber layer to its theoretical limit of optical absorption without compromising the higher

conversion efficiency is highly desirable. Needless to mention that among the other cost related parameters, a large portion of installed solar panel cost is directly related to the material usage [4]. However, the ultra-thin layer, i.e. thickness near to theoretical limit of solar spectrum absorption, provokes several problems like pinhole, shorting the front and back contact pinching through the window layer etc. This local shunting surely reduces the performance of the solar cell and increases the deep penetration loss. In the case of ultra-thin CdTe absorber layer, the cell performance is also affected by full or partial depletion of the layer and/or by interference between the p-n and/or contact junctions. Even though, the efficiency of CdTe-based solar cells is increasing rapidly, yet the conversion efficiency for a specific deposition technique, structural, optical and electrical properties are an open-ended problem for the ultra-thin CdTe solar cells. Any deposition process (e.g. sputtering) and post-deposition parameters such as growth rate and CdCl_2 wet-treatment are to be extensively investigated to get optimized ultra-thin CdTe layered solar cells.

A number of techniques were employed by the researchers

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worldwide to prepare CdTe films and they extensively studied structural and opto-electronic properties [5–7]. All these techniques have their own advantages and disadvantages. Among the processes, RF-sputtering technique uses a low temperature for which the layer underneath of CdTe are not affected and the stoichiometry of underneath material is retained during the deposition [8]. Moreover, the sputtering process allowed depositing films with precise, uniform and ion damage free films over a large area [9]. The best efficiency reported for sputtered-CdTe with FTO as a front contact and Au as a back contact is 12.6% [10]. In contrast, we have investigated the CdTe solar cell with the lowest possible thickness of around 2.0 μm with screen printed C:Cu/Ag back contact in this study. However, it is well recognized that the CdCl₂ activation heat treatment is a vital requirement in fabricating high efficiency CdTe-based solar cells. During the CdCl₂ heat treatment, CdTe partially melts and increase grain size, recrystallization occurs and bulk and interface defects level begins to be reduced [11]. It is also well-known that the CdTe properties are also influenced by the growth rate. Improving the quality of the ultra-thin CdTe films by optimizing the key processing steps (e.g., growth parameters, cadmium chloride recrystallization treatment, and back electrode) is one of the vital issues for next generation CdTe based solar cells.

It is well recognized that a major conversion loss in CdTe solar cell and the module is owing to absorption loss in the CdS window layer. Since the bandgap of CdS is around 2.41 eV, a considerable absorption occurs in the blue region and light absorbed in this layer is not collected attributing to the efficiency loss of about 20%. Numerous approaches have been studied to reduce this loss. Once, a very thin CdS (≤ 100 nm) has been proposed; however, too thin CdS reduces the open circuit voltage (Voc) and fill factor (FF) via pinholes as created by missing deposition and/or 100% diffusion of CdS to CdTe [12]. The minimum CdS thickness can be used for optimum cell efficiency via the addition of a high resistive transparent (HRT) layer [13] underneath of the CdS window layer. This configuration has been reported using SnO₂ HRT layer [14]. However, the solar cells showed poor performance and need to be optimized in terms of current collection and other device parameters for higher efficiency. A higher bandgap window layer (Cd, Zn)S has also been approached and cell suffered from the low Voc and phase segregation issues as reported in literatures [15,16].

Another very different approach is luminescence downshifting using a polymer dye on the front surface of the cell [17]. The dye converts high energy photon to low energy which can pass through CdS, however, this process has only improved blue response by 16% [18]. Also, this process increases the cost of cell or module and it has stability issue of the polymer film. Therefore, the increase of transmission and reduced diffusion are considered to be the best way to improve efficiency in CdTe based solar cells. Reactively sputtered CdS in the presence of oxygen may have the potential to meet the above-required properties. Wu et al. [19] first prepared this material, named oxygenated cadmium sulfide (CdS:O) and reported structural, electrical and electrical properties. Later on, CdS:O film has been prepared by employing CSS followed by the post-deposition thermal annealing in oxygen ambient [20,21]. The presence of oxygen into the CdS:O films reduce film crystallinity, increase bandgap, reduce inter-diffusion and lattice

mismatch at the junction region [19]. Many groups have been fabricating CdTe based solar cells, which have somehow similar configurations. However, in this study, the complete solar cell has also been fabricated with the novel structure of ‘glass/FTO/ZnO:Sn/CdS:O/CdTe/C:Cu/Ag’, where the back contact of the cell has been done by screen printing technique for its low cost potential, but it requires rigorous optimization.

2. Experimental procedure

At first, both CdTe and CdS:O films were separately deposited on top of bare soda lime glass substrates for characterization purposes and then optimized films were stacked for complete solar cell fabrication. CdTe thin films of about 2.0 μm were deposited on top of the soda lime glass substrate by RF sputtering at different growth rate ranging from 1.8 $\text{\AA}/\text{s}$ to 5.4 $\text{\AA}/\text{s}$ by controlling RF power from 1.0 W/cm^2 to 3.0 W/cm^2 . The growth rates of the films were observed during the deposition by the thickness monitoring system (FTM-2000, TORR International, Inc.) embedded to the sputtering system. Table 1 shows the growth parameters including ambient pressure and temperature during the deposition. The glass substrates used in this study were cleaned by the several sequential steps in the ultra-sonication tub using acetone, ethanol and deionized water as the clean solution. The duration of each cleaning step was 10 min and finally samples were dried by dry N₂. As-deposited CdTe thin films have undergone the CdCl₂ heat treatment. A schematic diagram of the CdCl₂ process employed in this study is shown in Fig. 1. The samples were immersed in cadmium chloride solution of 0.3 M for 10 s. After the dipping process, they were dried naturally for 10 min and then, heated in an evacuated furnace of N₂/O₂ ambient for 15 min at 66.66 Pa pressure and at a temperature of 390 °C. After the completion of the heat treatment, the samples were washed in warm water for several times.

At an optimized RF power of 2.0 W/cm^2 , CdS:O thin films of thickness around 100 nm were deposited on bare microscopic glass substrate at room temperature. The growth rate of the films was varied from 0.15 $\text{\AA}/\text{s}$ to 0.45 $\text{\AA}/\text{s}$. The O₂ partial pressure was varied from 11 mPa to 24 mPa during the sputtering by changing the pressure of argon:oxygen (99:1) gas mixture inserted to the chamber. It has been found that the deposition rate inversely changes with the increase of ambient pressure as expected. Therefore, to maintain the required film thickness, the deposition time was increased suitably with the increase of ambient pressure.

The structural properties and surface morphology were investigated using X-ray diffraction (XRD) spectroscopy, atomic force microscopy (AFM) and scanning electron microscopy (SEM). XRD patterns were taken in the 2 θ ranging from 20° to 70° using Cu K α radiation wavelength of 1.5408 \AA using “BRUKER aXS-D8 Advance Cu-K α ” diffractometer. The average crystallite size (D), microstrain (ϵ), and dislocation density (δ) have been determined by using the formula, $D = 0.89 \lambda / \beta \cos\theta$ [22], $\epsilon = \beta / 4\tan\theta$ [23] and $\delta = 1 / D^2$ [24], respectively, where, θ is the Bragg's or diffraction angle of the x-rays, β is the full-width half maximum (FWHM) of the reflection peak that has the same maximum intensity in the diffraction pattern, λ is the

Table 1
Growth parameters of CdTe and CdS:O thin films.

CdTe (300 °C)				CdS:O (RT)			
Dep. power (W/cm^2)	Flow rate (Ar)	Ambient pressure	Growth rate ($\text{\AA}/\text{s}$)	Dep. power (W/cm^2)	Flow rate (Ar:O = 99:1)	Oxygen partial pressure (mT)	Growth rate ($\text{\AA}/\text{s}$)
1.0,	8 sccm	8 mT	1.8	2.0	2 sccm	0.08	0.45
1.5			2.5		4 sccm	0.12	0.32
2.0			3.6		8 sccm	0.15	0.25
2.5			4.6		12 sccm	0.18	0.15

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