Synthesis and characterization of novel oxygenated CdSe window layer for CdTe thin film solar cells

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ABSTRACT

Significant increases in the solar conversion performance of thin film CdS/CdTe solar cells have been realized in recent years by the inclusion of wider gap materials, notably CdS:O, in place of the CdS window layer. Similarly, use of CdSe in combination with CdTe has given yet further increases in current density due to the formation of CdTe_{1-x}Se_x and the resulting enhanced long wavelength collection. Here we report the synthesis and properties of oxygenated CdSe films and their influence on device performance when included into CdTe solar cells. The CdSe:O films were made by pulsed laser deposition in oxygen at pressures of 0.01 and 0.03 Pa. Optical transmission measurements indicated that oxygenation acts to increase transparency in the 400–750 nm range, and that increased oxygen content acts to increase the apparent absorption threshold slightly. The higher transmission acts to increase the spectral response of CdTe solar cells in the range 350–500 nm compared to CdSe- and CdS-containing control devices. Moreover, the already known advantage of enhanced long wavelength response (850–910 nm) over that of CdS/CdTe is retained for both CdSe and CdSe:O window layers. Hence the use of CdSe:O as a window layer is demonstrated to increase the current harvesting of these solar cells by increasing both their long and short wavelength performances. These gains in the short circuit current outweigh small losses in fill factor and open circuit voltage making CdSe:O a valuable possible means to increase the performance of CdTe solar cells.

1. Introduction

The development of low-cost polycrystalline thin film solar energy materials is important for future energy generation. Cadmium telluride thin film absorbers for photovoltaics are well-known to have the advantages of an ideal direct band gap E_g (1.45 eV), and a high absorption coefficient. Its best known p-n junction partner, the n-CdS window layer, belongs to II-VI group of compound semiconductor materials, has a band gap E_g of approximately 2.4 eV and good band line ups with CdTe. Currently, the reported power conversion efficiency of typical CdS/CdTe solar cell has exceeded 20% [1]. In addition, it is also reported that the open circuit voltage of CdS/CdTe devices has broken the 1 V barrier without substantial decreases in current through doping the CdTe with a Group V element [2]. However, the CdS/CdTe solar cell has a relatively low current density, but this deficit has been corrected by replacing the CdS with wider gap alternatives, notably oxygenated CdS, i.e. CdS:O. Also, Paudel's group [3] noticed that CdSe/CdTe solar cells present a higher current density and a higher response

from 860 nm to 910 nm. In contrast to CdS, CdSe has also been recognized as an attractive partner layer material for CdTe because of its lower lattice mismatch of around 6% with CdTe (compared to CdS) and its higher solubility in the CdTe absorber layer. An efficiency of about 15% of CdSe/CdTe thin film photovoltaic device with an area of 0.08 cm² has already been demonstrated by University of Toledo [3]. Thus, replacing CdS with CdSe has great potential to fabricate high-efficiency CdTe-based solar cells. Nevertheless, CdSe thin film shows a comparatively lower band gap E_g of 1.7 eV [4] compared to CdS, which may be disadvantageous. Many researchers improve the short wavelength response of CdTe solar cells through the conventional method of reducing the thickness of CdS window layer, but the enhanced short circuit current density is at the cost of losses in voltage and fill factor [5]. Here, a novel attempt to incorporate oxygen in the CdSe thin film is made to improve its properties, and further enhance the photoelectric response of CdSe/CdTe solar cell in the short wavelength region (in addition to the already demonstrated long wavelength enhancement). It was found that the photocurrent collection could indeed be increased

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Fig. 1. Schematic diagram of the pulsed laser deposition system.

both in the short and long wavelength ranges compared to standard structures and it is concluded that the use of CdSe:O is viable for CdTe-based photovoltaic devices.

It is well-known that there are a large number of techniques available for the formation of CdSe thin films, including chemical bath deposition [6,7], molecular beam epitaxy (MBE) [9], magnetron sputtering [3,8] and close space sublimation (CSS) [10]. In this work, we utilized pulsed laser deposition (PLD) to fabricate both CdSe window layers and oxygenated CdSe window layers. Compared to other methods, pulsed laser deposition can replicate the composition of the targets in the samples easily, and a variety of parameters can be accommodated to optimize the process of manufacture. This is achievable since PLD is able to yield a rapid rise in temperature ($> 10^{11} \text{ K s}^{-1}$), which results in a rapid evaporation of target elements and compounds [11]. In addition, the high surface ablation temperature causes particles arrive on the substrate with an elevated kinetic energy of 1–100 eV, which has the potential to increase the surface mobility of adatoms and is beneficial to manufacture high quality thin films at low substrate temperatures. The instrument also affords accurate computer aided thickness control. It is shown in Fig. 1.

2. Experimental details

Four different window layer materials were prepared i.e. CdS, CdSe and CdSe:O having two different amounts of oxygen. For comparative purposes, the thicknesses of all four was kept at $\sim 100 \text{ nm}$, the temperature of substrates was 150°C and they were all synthesized by PLD technique. The substrate temperature of 150°C was chosen since it had given us the best outcomes in earlier tests in making CdSe/CdTe cells. When manufacturing CdSe:O thin films, the dynamic oxygen atmosphere was modified from 0 to 0.03 Pa. Photovoltaic devices were synthesized on commercial $\text{SnO}_2\text{:F/SnO}_2$ coated soda lime glass substrates (FTO). And FTO glass substrates were cleaned by

a series of processes, including with detergent, de-ionized water and isopropyl alcohol. The cleaned FTO glasses were fixed on a heated substrate holder (max. temperature 800°C). Before conducting an experiment, the growth chamber was evacuated to $3.0 \times 10^{-3} \text{ Pa}$ to eliminate air. The KrF excimer laser ($\lambda=248 \text{ nm}$, pulse duration 20 ns, variable pulse energy from 60 to 150 mJ) was used at its optimal energy of 90 mJ and pulse repetition frequency of 6 Hz [12–14].

The single thin films were characterized by UV–Vis–NIR spectrophotometer (PerkinElmer Lambda 950), and X-ray diffraction (XRD) (Dan Dong Hao Yuan DX-2700) with using Cu ($K\alpha$) $1.54 \text{ \AA}$ X-rays. The cross-sectional structure of CdS/CdTe and CdSe/CdTe double layers were measured by scanning electron microscopy (SEM) (Hitachi S-4800). The composition of the oxygenated CdSe films was examined by energy dispersive spectrometry (EDS) (Hitachi S-4800). In order to eliminate interferences from the substrate, the compositional analysis was done on films that were specially deposited onto silicon substrates. The surface morphologies of the pure CdSe and the CdSe:O were also investigated by scanning electron microscopy (SEM) (Hitachi S-4800).

For the device fabrication, the CdTe layers were fabricated using the industrially compatible method of close space sublimation (CSS) [15]. CSS employed high-purity CdTe powders as the source material, and the distance of source to substrate was approximately 5 mm. During deposition, the substrate temperature was maintained at $550\text{--}580^\circ\text{C}$ and the source temperature was $80\text{--}100^\circ\text{C}$ higher. Although the base chamber pressure was 0.03 KPa, we could introduce a mixed gas $\text{Ar}+\text{O}_2$ ambient ($\text{Ar } 90\%, \text{O}_2 \text{ } 10\%$) at 1.6 KPa. Oxygen is thought to increase the acceptor density of CdTe. A CdTe absorber thickness of $5 \mu\text{m}$ was used in the devices. Subsequently, the completed CdTe layer underwent heat treatment at 390°C for 35 min in a tube furnace with CdCl_2 , which is thought to enable grain growth and passivation hence reducing carrier recombination. After heat treatment the CdTe surface was cleaned by etching, which is thought to form a tellurium-rich layer to enhance the p-conductivity of the CdTe layer for contacting [16,17]. A back contact of ZnTe:Cu ($\sim 100 \text{ nm}$) was then deposited by PLD at 180°C . Then the structure was in-situ annealed under a protective pressure of $2 \times 10^4 \text{ Pa}$ of N_2 at 200°C in order to facilitate the Cu diffusion. Finally, Au electrodes ($\sim 80 \text{ nm}$ thick) were applied by thermal evaporation. The finished device comprised many small cells each having an area of 0.08 cm^2 . Fig. 2 shows the schematic illustration of the CdTe solar cell device manufactured in this work.

3. Results and discussion

3.1. Optical transmittance and band gaps

As shown in Fig. 3(a), it could be seen that three window layers displayed different optical transmittance curves. However, both the CdSe and the oxygenated samples show $> 80\%$ transmittance in the long wavelength region. Nevertheless, below 700 nm the pure CdSe film shows low optical transmittance as expected from its band gap of 1.7 eV. Despite this, it is known that when CdSe is included in CdTe photovoltaic devices, dissolution of the CdSe in the CdTe - and the resulting downward bandgap bowing, cause enhanced long wavelength solar cell response, rather than the short wavelength response (or

Fig. 2. Schematic illustration of CdTe solar cell device manufactured in this work.

Fig. 3. (a) The optical transmittance spectra of the three thin film window layers: CdSe, CdSe:O (0.01 Pa), CdSe:O (0.03 Pa). (b) Tauc plots of the optical absorption data of window layers comprising CdSe, CdSe:O (0.01 Pa) and CdSe:O (0.03 Pa). Oxygen causes a shift to higher energy absorption thresholds but the plots do not indicate classical direct-gap behaviour.

parasitic absorption) that might be expected. This interdiffusion occurs readily during the growth of the CdTe part of the solar cell and its subsequent processing.

However, for the case of oxygenated CdSe:O films [18,19], transmission in the range 400–750 nm is increased progressively over that for CdSe upon inclusion of higher levels of oxygen from zero to 0.03 Pa, as shown in Fig. 3(a). Moreover, the increase in oxygen in the growth apparatus was accompanied by an apparent blue-shift of the absorption edge i.e. oxygen appears to increase the threshold for optical absorption.

To investigate this further, we made Tauc plots for the three films as shown in Fig. 3(b). While there is a general trend of a shift to higher energy for the onset of absorption with increasing oxygen content, the plots are not linear, and the fits with their intercepts of 1.652, 1.862 and 1.889 eV are made. To conclude that the shift is due to simple modification of the bandgap to higher levels would therefore be tentative. An alternative explanation – that the material is a mixture rather than a solid solution – is also plausible.

3.2. Crystal structure

Fig. 4 shows the XRD patterns of the CdSe and CdSe:O thin films deposited at vacuum, 0.01 Pa oxygen pressure, and 0.03 Pa O_2 pressures. All the films had a clear (111) CdSe peak occurring at $2\theta=25.35^\circ$, 25.45° and 25.40° respectively, this being identified by comparison with JCPDS data. Oxygen appears to have had the effect of increasing

Fig. 4. X-ray diffraction patterns of CdSe and CdSe:O thin films grown under the three conditions.

the Bragg angle of the (111) peak, which supports the idea of a solid solution. For all samples, there were weaker peaks that were identified as (220), (311), (400) indicating that all the completed films were polycrystalline regardless of their oxygen content. For films fabricated with the highest level of oxygen (0.03 Pa) an additional peak that was consistent with being from (200) tetragonal SeO_2 , was present. It is therefore possible that CdSe:O comprises a mixture of SeO_2 and a CdSe-rich phase. Finally, since the intensity of the primary (CdSe 111) diffraction peak gradually decreased with the increase of oxygen pressure, it is possible that oxygen suppresses crystallinity, as reported previously in the literature for the case of CdS:O [20,21].

3.3. Surface morphology and cross-sectional structure

Scanning electron microscopy (SEM) was utilized to further give more surface morphology information about the CdSe thin films and oxygenated CdSe thin films. CdSe and CdSe:O thin films were fabricated by pulsed laser deposition. The surface of these films was quite compact, and the crystalline size was so small that it was very difficult to distinguish the grain boundary like Fig. 5(a). Therefore, for further investigating the surface morphology of these films, three thin films fabricated were annealed in dry air ambient at 385°C for 30 min. The grains underwent regrowth and became larger than before as shown in Fig. 5b-d. However, from these images, we found there existed several voids, which might form the recombination center to lower the performance of solar cells. Compared to CdSe thin film, the voids of the oxygenated CdSe thin films were relatively fewer, and that of CdSe thin film with 0.01 Pa oxygen were distinctly fewer than CdSe thin film with 0.03 Pa oxygen. These studies describe that oxygen plays a significant role in thin films or solar cells.

Next, the microstructure of CdTe in combination with both CdS and CdSe are compared by cross sectional SEM. Care was taken to deposit 100 nm of each in order to allow meaningful comparison.

It was found that after the deposition of CdTe layer and heat treatment on top of the CdS and CdSe layers, it was clear that the residual CdS layer thickness was greater than that of the CdSe layer. Moreover for the case of CdSe/CdTe double layers, the interface was not clearly visible. To some extent, this confirms that Se has a stronger solubility in CdTe than S. When Se diffuses into the CdTe layer it is expected that the ternary compound $\text{CdTe}_{1-x}\text{Se}_x$ forms, and that owing to the bandgap bowing effect, the band gap of the ternary is reduced below that of CdTe, while maintaining a lattice match. These factors are expected to result in an excellent response of CdSe/CdTe solar cell in the region of 860–910 nm, realizing the effective collection of photon-generated current in long wavelength [8,22,23] (Fig. 6).

Fig. 5. SEM surface morphologies of CdSe films grown under the ambient of: a) vacuum (before annealing); b) vacuum (after annealing); c) oxygen pressure of 0.01 Pa (after annealing); d) oxygen pressure of 0.03 Pa (after annealing).

3.4. Thin film composition analysis

The EDS technique was used to confirm the composition of the oxygenated CdSe window layers as shown in Fig. 7(a)–b. For the analysis, special samples were grown on silicon wafers in order to avoid interference from the substrate. Due to the intrinsic error of EDS, plus little content of oxygen, the data of the images is not very accurate. However, at least, it indicates that oxygen has been successfully incorporated into the CdSe thin films.

3.5. Solar cell characteristics

Following on from the studies of the window layers themselves, a comparative study of CdTe-based solar cells made with the three window layers (CdS, CdSe, and CdSe:O having the two oxygen concentrations) was undertaken to understand their effects on devices. The device structure was: window layer (CdS, CdSe or CdSe:O)(PLD)/

absorber CdTe (CSS)/contact ZnTe:Cu(PLD)/electrode Au (there was no anti-reflection coating). Current - voltage (J - V) measurements were obtained under AM1.5 illumination yielding the efficiency (η), fill factor (FF), short circuit current density (J_{sc}), and open voltage (V_{oc}) values listed in Table 1, with the best performance for each device being selected for presentation. It could be clearly seen that CdS/CdTe solar cell had the highest open circuit voltage of 713 mV. However, it also had the lowest current density (20.1 mA/cm^2) as might be expected from parasitic absorption in the CdS. With a CdSe window layer, an increase of current density (21.8 mA/cm^2) was realized, meanwhile, but at the expense of a much lower voltage of 621 mV compared to that of the CdS/CdTe device (i.e. 92 mV lower). This is presumed to be due to the low bandgap of CdSe (1.7 eV) compared to CdS and the possibility that the built-in potential value is small. However, the inclusion of oxygen in the CdSe acted to overcome the limitations of the undoped binary CdSe window layers: When oxygen pressure was 0.01 Pa, the overall efficiency rose to 8.9%, this being driven by an

Fig. 6. The cross-sectional SEM images of CdS/CdTe and CdSe/CdTe double layers: a) CdS/CdTe; b) CdSe/CdTe.

Fig. 7. EDS spectra of oxygenated CdSe:O thin films deposited on silicon wafers: a) CdSe:O (0.01 Pa); b) CdSe:O (0.03 Pa).

Table 1

The best performance of CdS/CdTe solar cells and CdSe/CdTe solar cells with the CdSe window layers prepared by pulsed laser deposition under different deposition temperatures.

Window layer	Efficiency (%)	FF (%)	J_{sc} (mA/cm ²)	V_{oc} (mV)
CdS	8.56	60.12	20.1	713
CdSe	7.52	55.40	21.8	621
CdSe:O(0.01 Pa)	8.91	58.37	22.5	678
CdSe:O(0.03 Pa)	7.60	55.01	21.5	642

Fig. 8. J - V curves of CdTe-based solar cells with the CdS, CdSe and two CdSe:O window layers.

increased current density (J_{sc} =22.5 mA/cm²), and open circuit voltage (V_{oc} =678 mV). Nevertheless, further improvements from incorporating more oxygen could not be obtained, and increasing the oxygen pressure during growth to 0.03 Pa acted to decrease all of the working parameters of the solar cells, including the J_{sc} . Hence it was found that there is an optimum level of oxygen incorporation in CdSe:O that gives a maximum in the solar cell performance (Fig. 8).

To further understand the underlying mechanism of this variation, external quantum efficiency (EQE) measurement were made and are shown Fig. 9 for the cells with CdS, CdSe and the two compositions of CdS:O. Firstly the long wavelength response is discussed: While the cell with a CdS window has an EQE threshold at about 850 nm, all of the cells containing either CdSe or CdSe:O showed identical enhanced long wavelength response extending to about 910 nm. This response is consistent with the literature interpretation that CdSe diffuses into the CdTe (during growth and processing), and that bowing [3] causes the bandgap to dip to 910 nm, this being below the intrinsic bulk CdTe

Fig. 9. Measured quantum efficiency spectra of CdTe-based solar cells with different window layers.

value of 860 nm and hence extending the near infra-red response of the cells. From the coincidence of the lines at the long wavelength side of Fig. 9, this diffusion effect appears to take place identically regardless of the level of oxygenation of the CdSe, and all gave an increased long wavelength response over that of CdS/CdTe.

Secondly the EQE response in the range 350–500 nm is discussed. This is the region above the band gap of CdS, and which in the CdS/CdTe device is subject to the parasitic absorption that gives current loss in this wavelength range – unless the CdS is very thin. Indeed, the curve for the device with the CdS window layer shows a step at ~500 nm that is typical for a CdS-containing device. On the contrary, the devices with CdSe and CdSe:O all had higher EQE in the range 350–500 nm than the CdS-containing device. For the case of CdSe compared to CdSe:O (0.01 Pa O₂ in deposition), the apparent increase in the band gap of the CdSe:O shown in Fig. 3 gave the expected result that the EQE of the cell with the oxygenated CdSe:O had a higher EQE. However, this expected trend did not continue for the most highly oxygenated window layer tested, namely the CdSe:O (0.03 Pa O₂ in deposition), for which the EQE in the range 350–500 nm was the lowest of all those in the CdSe and CdSe:O series – and was only just above that of CdS. So paradoxically, even though the CdSe:O sample grown under the most oxygen-rich conditions exhibited the most advantageous optical transmission when grown as a single layer, it gave inferior solar cell performance. We speculate that it is possible that high levels of oxygen incorporation in CdSe introduces high levels of point or cluster defects, either within the CdSe:O films, or at the interfaces of the CdSe:O/CdTe structures. This might act to cause

degradation of the electrical junction performance or junction position that would degrade the EQE in the region 350–500 nm.

Lastly we note that the substitution of CdS with CdSe or any of the CdSe:O compositions tried acts to degrade the fill factor from about 60 to about 55%, except in the case of CdSe:O (0.01 Pa O₂) for which the degradation was less severe. Fill factor loss is normally associated with current leakage paths or incomplete film coverage, although that was not investigated here.

Overall though, the inclusion of CdSe, and CdSe:O as a replacement window layer for CdS in CdTe solar cells – especially at the composition obtained with 0.01 Pa of O₂ during growth – acted to increase the solar conversion efficiency. Although there was a slight loss of V_{oc} and FF, CdSe:O acted to increase the EQE response in both a) the low (350–500 nm) and b) the high (850–910 nm) ranges over and above that achieved with CdS. This is considered to be due to a) the increase in short wavelength optical transmission in CdSe:O and b) the increase in long wavelength absorption by CdTe_{1-x}Se_x. These increase in optical performance outweighed the voltage and fill factor decreases. Nevertheless, since excess oxygen reverses the device benefits, in our later work, we would continue to search for the most optimum content of oxygen incorporation and optimize all kinds of preparation conditions to further improve the performance of CdSe/CdTe solar cells. Moreover, the problem of the reduced fill factor that is present in all CdSe/CdTe and CdSe:O/CdTe solar cells remains, and this might be related to the intrinsic properties of semiconductor materials – hence it may become the focus of subsequent research. All in all, we demonstrated the effectiveness of PLD in a totally novel study of deposition CdSe:O window layer by ablation in an oxygen ambient. We believe that this effective method and window layer chemistry can help promote the development of CdTe-based solar cells.

4. Conclusions

In this study we have exploited the reproducibility and reliability of pulsed laser deposition in a novel study of both CdSe:O films and CdTe photovoltaic devices made using them as window layers – as alternatives to CdS and plain CdSe. After annealing, on the surface the PLD CdSe:O films showed fewer voids than that of pure CdSe thin films. The higher levels of oxygen caused the XRD peaks of CdSe to become less strong, presumably due to amorphisation caused by the oxygen. The most highly oxygenated samples showed evidence of SeO₂ in their XRD spectra. Increasing the oxygen pressure during growth also acted to increase the short wavelength optical transmission and shifted the apparent absorption edge to higher energies in the approximate range 1.65–1.89 eV. The result that CdSe interdiffuses into CdTe faster than does CdS was confirmed in a comparative diffusion experiment. Like-for-like solar cell devices containing window layers of CdS, CdSe, CdSe (grown under 0.01 Pa O₂) and CdSe (grown under 0.03 Pa O₂) demonstrated that: a) the long wavelength EQE increases that are ascribed to bandgap-bowed interdiffused layers of CdTe_{1-x}Se_x, are obtained for both plain CdSe and all compositions of CdSe:O tested, and b) the increase in short wavelength optical transmission observed for CdSe:O (over that of CdSe) is responsible for increased EQE response in solar cells having CdSe:O window layers over and above those with CdS or plain CdSe. There is an optimum for the level of oxygen in the CdSe:O that gives the best cells and this acts to increase the EQE in both the low (350–500 nm) and long (850–910 nm) wavelength ranges. Overall this acts to increase the solar cell efficiency, despite some losses in (V_{oc}) and fill factor. Further work will focus on identifying the optimum composition for CdSe:O more exactly and

investigating the reasons why higher levels of oxygen cause the cell performance to decline.

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