Analysis and optimisation of the glass/TCO/MZO stack for thin film CdTe solar cells

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Magnesium-doped Zinc Oxide (MZO) films have recently been proposed as a transparent buffer layer for thin film CdTe solar cells. In this study, the band gap of MZO buffer layers was tuned for CdTe solar cells by increasing the substrate temperature during deposition. Films were deposited by radio-frequency magnetron sputtering. Devices incorporating an optimised MZO buffer layer deposited at 300 °C with a band gap of 3.70 eV yielded a mean efficiency of 12.5% and a highest efficiency of 13.3%. Transmission electron microscopy showed that MZO films are uniformly deposited on the transparent conductive oxide (TCO) layer surface. The favourable band alignment seems to positively counterbalance the low doping level of the MZO layer and its high lattice mismatch with CdTe. Titanium-doped indium oxide, tin-doped indium oxide and aluminium-doped zinc oxide TCOs were also used as alternatives to fluorine-doped tin oxide (FTO), in combination with MZO films. The use of titanium-doped indium oxide and tin-doped indium oxide TCOs did not improve the device efficiency achieved compared with FTO, however using aluminium-doped zinc oxide coupled with a boro-aluminosilicate glass substrate the mean and highest efficiencies were further improved to 12.6% and 13.4% respectively.

1. Introduction

Thin film cadmium telluride (CdTe) photovoltaics (PV) are an extremely promising and scalable PV technology. First Solar, Inc., the largest manufacturer of CdTe PV modules to date, currently provides utility-scale levelised cost of electricity (LCOE) which is competitive with all other renewable and non-renewable energy sources [1]. One of the main drivers for thin film CdTe LCOE reduction is to increase the PV module power conversion efficiency [2]. The efficiency of thin film CdTe modules and laboratory scale cells has been significantly improved in the last decade, achieving record efficiencies of 18.6% and 21.0% respectively [3]. The improvement was mainly a result of increases in the short circuit current density of the solar cell/module, achieved by using a very transparent window layer as well as including a CdSeTe alloy to grade the CdTe band gap and absorb more infrared light [4]. Recently, magnesium-doped zinc oxide (MZO) has been proposed as an effective buffer and alternative to CdS [5]. MZO is a high band gap semiconductor (Eg > 3.3 eV) and transmits a larger fraction of the solar spectrum through to the underlying absorber, as compared with CdS (Eg = 2.45 eV) [6]. There are several other semiconductors with a band gap comparable to MZO; however MZO is effective due to the tuneability of its band alignment with CdTe [7]. This depends upon the Mg concentration in the film and in particular on the MgO/ZnO ratio, with MgO being the high band gap (Eg = 7.8 eV) material that mixed with ZnO causes the energy band structure to shift. The optimal band structure of MZO can be achieved by adding the right amount of Mg to ZnO. In a previous study an alternative method to optimize the MZO energy band gap as a high resistance transparent (HRT) layer for CdTe solar cells was presented, rather than a buffer layer [8]. By using a single target composition, this method enables widening of the energy band gap by increasing the temperature the MZO film is deposited at. In the current study, we have increased the MZO buffer layer deposition temperature to create a favourable MZO/CdTe alignment, thereby maximising the efficiency of CdS-free thin film CdTe solar cells. In the second part of the study, different glass/TCO/MZO combinations were investigated, in an effort to further improve device performance. Devices incorporating aluminium-doped zinc oxide (AZO), titanium-doped indium oxide (ITiO) and tin-doped indium oxide (ITO) TCOs have been compared to the FTO substrates (TEC10, Pilkington NSG) which are typically used when fabricating CdTe solar cells. Finally, soda lime glass
was substituted with boro-aluminosilicate (Eagle XG, Corning) glass to analyse the impact of using a more transparent substrate on the device output.

2. Experimental

ITO, AZO, ITIO and MZO thin films were deposited by radio-frequency (RF) magnetron sputtering. 4 mm thick soda lime glass (SLG) and 1 mm thick boro-aluminosilicate glass were used as substrates. The glass was cleaned using a solution composed of 1/3 isopropanol, 1/3 acetone and 1/3 deionised water in an ultrasonic bath at 50 °C for 60 min. Thin films were deposited using an Orion 8 HV magnetron sputtering system (AJA International, USA) equipped with an AJA 600 series RF power supply. All sputtering targets (ITO: 10% SnO2 and 90% In2O3 wt%, AZO: 0.5% Al2O3 and 99.5% ZnO wt%, ITIO: 2% TiO2 and 98% In2O3 wt%, MZO: 11% MgO and 89% ZnO wt%) were 3" in diameter. The glass substrates were rotated at 10 rpm during deposition to enhance the uniformity of the films. The TCO sputtering process was carried out at a constant power density of 3.95 W cm−2 and a pressure of 1 mTorr (0.133 Pa) using pure Ar as the process gas. MZO and ZnO films were sputtered in a 1% O2 in Ar atmosphere at 5 mTorr, and a power density of 3.95 W cm−2. The temperature of the substrates was kept at 450 °C for the deposition of ITO and ITIO and 300 °C for AZO films. The deposition temperature for MZO films was varied between 20 °C and 400 °C. The deposition temperature for ZnO films was 20 °C. The FTO substrates used in this study were NSG TEC™ G10 glass (Pilkington). The optical properties were investigated using a Varian Cary 5000 UV–VIS–NIR spectrophotometer. The composition of the films was measured using an X-ray photoelectron spectrometer (XPS) (Thermo Scientific K-alpha). Hall effect measurements were carried out in the Van der Pauw configuration to measure the resistivity, Hall mobility and carrier concentration of the different layers. Hall effect measurements presented in this work were carried out using an Ecopia HMS-3000 Hall Measurement System. The structural properties of the films were analysed by X-ray diffraction (XRD) using a Brucker D2 Phaser desktop X-ray diffractometer equipped with a Cu-K-alpha X-ray gun. The XRD measurements were obtained using 15 rpm rotation, a 1 mm beam slit and 3 mm anti-scatter plate height. Devices were fabricated in a superstrate configuration on the different TCO/MZO combinations. The CdTe absorber was deposited by close space sublimation (CSS) at a pressure of 1 Torr (133 Pa) in a 6% O2 in Ar atmosphere, with a CdTe source plate temperature of 630 °C and a substrate temperature of 515 °C, for 2 min. All samples have CdTe films deposited with thicknesses in the range of 4–4.7 μm to rule out the effect of absorber thickness variation on device performance. The spacing between substrate and source plate was set to 2 mm. The CdCl2 activation treatment was carried out by thermal evaporation and subsequent annealing. A quartz crucible was loaded with 0.5 g of CdCl2 pellets, which was then evaporated at ~1 × 10−6 Torr for 20 min. The samples were then annealed on a hot plate at a dwell temperature of 425 °C for 3 min. The dwell temperature was reached by using a 22/°min ramping rate bringing the temperature from 25 °C to 425 °C in 18 min, for a total annealing duration of 21 min. Devices were rinsed with DI water to clean the CdTe surface of CdCl2 and completed with 80 nm gold contacts deposited using thermal evaporation. No intentional copper has been added to these devices. The current density–voltage (JV) characteristics of devices were determined using a bespoke solar simulator under a simulated AM1.5G spectrum. External quantum efficiency (EQE) measurements were carried out using a PVE300 EQE system (Bentham Instruments Limited, UK) with a 5 nm resolution. Samples for transmission electron microscopy (TEM) were prepared by focused ion beam milling using a dual beam FEI Nova 600 Nanolab. A standard in situ lift out method was used to prepare cross-sectional samples. An electron beam assisted platinum (e-Pt) over-layer was deposited onto the sample surface above the area to be analysed followed by an ion assisted layer to define the surface and homogenize the final thinning of the samples down to 100 nm. TEM analysis was carried out using a Tecnai F20 operating at 200 kV to investigate the detailed micro-structure of the cell cross sections. Images were obtained using a bright field (BF) detector.

3. Theory

Due to the tuneability of the MZO band gap, a MZO buffer layer can enhance the absorber type inversion of thin film CdTe solar cells [7,9]. The type inversion refers to the inversion of the majority/minority carrier densities. Within a thin film solar cell, the CdTe absorber layer is p-type, therefore within this layer holes are majority carriers and electrons are minority carriers. However, when p-type CdTe is contacted with an n-type semiconductor to create a p-n junction the resulting electric field can invert this situation such that electrons become majority carriers and holes become minority carriers in the CdTe layer near the buffer/absorber interface. Simulations of thin film heterostructure solar cells show that increasing this absorber inversion can be beneficial for device performance [7,9]. Changing the buffer layer/CdTe band alignment and the buffer layer n-type doping are two ways to affect this device parameter. Fig. 1 shows the simulated energy band diagram and free carrier distribution for two different situations, using SCAPS-1D software [10]. The first simulation (a) shows the energy band diagram for a CdTe solar cell where the inversion is smaller than in (b). An indicator of the absorber inversion is the energy gap between the CdTe valence band and the Fermi energy at the buffer/CdTe interface (Epa/abs). Epa/abs is 0.83 eV in (a) and 1.27 eV in (b). The higher Epa/abs is due to an increased buffer layer carrier concentration and a change in the conduction band offset from negative (~0.1 eV) to positive (~0.1 eV). From the carrier concentration profile shown below the energy band structure in Fig. 1 it is clear that the increased Epa/abs results in an enhanced absorber inversion where electrons become majority carriers further away from the CdTe/buffer interface. The interface defect density can be very high compared to the bulk, and limiting interface recombination can be crucial for device performance. The simulated situation (b) is more favourable than (a) since interface recombination is limited by the lack of holes for electrons to recombine with.
4. Results

4.1. MZO band gap tuning

The MZO band gap depends upon the MgO/ZnO ratio in the film, with a larger band gap achieved with Mg rich films. The band gap change has been analysed optically [11], by X-ray and UV photoelectron spectroscopy [12], and by density functional theory [13] and it is primarily due to an upward shift in the conduction band minimum. In this study, the heating of the glass substrate during sputtering of the MZO films has been used to increase its band gap, as reported in our previous study [8] and in a study from Hwang et al. [14]. The estimation of the film band gap, done graphically by using the Tauc plot technique, highlights that changing the substrate temperature between 20 °C and 400 °C caused a band gap increase of almost 0.2 eV, from 3.56 eV to 3.73 eV (Fig. 2). X-ray photoelectron spectroscopy was also used to confirm the Zn/Mg ratio of the different films. There is a clear increase in the Mg ratio with deposition temperature, which also corresponds to the band gap increase. (Fig. 2). At 5 mTorr, Zn and Mg form a vapour at ≈ 290 °C and ≈ 380 °C respectively [15], and so during the deposition it is possible that free Zn is lost from the hot substrate surface, reducing the Zn content significantly with higher deposition temperatures. This at least explains qualitatively the significant difference in Mg content and band gap with increasing deposition temperature. Measuring the exact band alignment between the MZO layer and the CdTe layer is complex and different results are presented in the literature. The conduction band offset (CBO) can also vary depending on the films deposition technique and parameters. Rao [12] proposed a CdS/ZnO negative CBO = −0.3 eV. Assuming the CdTe/CdS conduction band alignment to be slightly negative (−0.1 eV [16]) and a ZnO band gap of 3.3 eV, this would translate in a CdTe/ZnO CBO of approximately −0.4 eV. Widening the MZO band gap to 3.56 eV would result in a CdTe/MZO CBO of approximately −0.14 eV. Meanwhile a band gap of 3.73 eV would provide a slightly positive CBO (+0.03 eV). Song et al., however, suggested that a 3.7 eV MZO band gap provides CBO = +0.2 eV with CdTe [7]. Because of this uncertainty it is difficult to precisely estimate the band alignment provided by MZO compositions with different band gaps with CdTe, while it is more effective to analyse the effect of the MZO band gap widening on device performance. The electrical properties of these films could not be analysed because their conductivity was too low to be effectively measured using our Hall effect system.

Fig. 2. Band gap variation of MZO films deposited with increasing substrate temperature, with the Tauc plot presented in the insert. In blue, the Mg/Zn atomic ratios of the MZO films deposited at different substrate temperatures.

Fig. 3. Box Plots of the main performance parameters of CdTe solar cells including MZO buffer layers deposited at increasing substrate temperature. On the left hand side of each graph the first box plot (in black) refers to a CdTe solar cells incorporating a ZnO buffer layer instead of MZO.
4.2. The effect of MZO band gap on CdTe solar cell performance

Thin film CdTe solar cells have been fabricated with ZnO buffer layers and MZO buffer layers deposited at substrate temperatures in the range between 20 and 400 °C and their performance is shown in Fig. 3. The performance of devices using ZnO buffers has been included to highlight the influence of Mg. All device parameters clearly improve when Mg is added to the buffer layers. The device efficiency further improves with increasing deposition temperature up to 300 °C. There is a clear trend between the substrate deposition temperature, the MZO film band gap and the device Voc. The Voc is a strong indicator of recombination, suggesting that the increase of the MZO band gap towards a flat or slightly positive CBO with the CdTe layer reduces the interface recombination. At 400 °C the device Voc and the efficiency slightly degrade compared with samples with an MZO buffer deposited at 300 °C. With the exception of devices incorporating MZO deposited at room temperature, the current density output decreases with increasing deposition temperature of the MZO layer. The higher Jsc of the MZO device deposited at 100 °C, compared to room temperature, is due
to a higher EQE response across all wavelengths (Fig. 4). Further increase of the MZO deposition temperature reduces the EQE response at wavelengths below 600 nm (200 °C). Subsequently, at higher deposition temperatures (300 °C and 400 °C) the decrease occurs across the whole spectrum. The degradation of photo-generated carrier collection over the full active spectrum of the solar cell could be related to the MZO/CdTe band alignment. The increasingly positive CBO due to the increase of the MZO band gap can act as a barrier for the electrons flowing from the CdTe to the MZO layer [17,18]. There is a slight shift in the high energy absorption edge of the EQE, corresponding to the MZO band gap variation seen in Fig. 2. A more significant shift is showed when ZnO is used as a buffer, since it has a lower band gap compared to MZO. The lower EQE response in the ultra-violet range of the solar spectrum of samples incorporating ZnO buffers is the cause of the low Jsc. The J-V characteristics indicate that increasing the MZO layer band gap has a positive impact on device Voc and efficiency, up to a band gap of approximately 3.7 eV, in agreement with previous work [5]. Increasing the substrate temperature during deposition was found to be an effective method to tune the MZO band gap. Although it was not possible to estimate the n-type doping of the material, increasing the free carrier concentration of MZO films also has the potential to increase the absorber inversion and device performance.

4.3. TEM and XRD analysis of the MZO films

TEM cross section imaging of devices shows a thin (100 nm) but uniform MZO layer deposited at 300 °C, separating FTO and CdTe (Fig. 5). The buffer layer uniformity is considered to be an important aspect to achieve high efficiencies, as interruptions or particularly thin buffer layer areas may result in weak diodes, with the consequent degradation of device Voc, FF and efficiency [19]. The crystal structure of MZO films grown on top of the FTO TCO has been investigated by XRD in the 2θ angular range 20–70°. All films exhibited an MZO (002) peak at 34.6° ± 0.2° (Fig. 6). All other peaks observed in Fig. 6 are attributed to the FTO film. This indicates that MZO films grow with a single phase with a wurtzite structure typical for ZnO, avoiding secondary phases related to the MgO cubic structure (peak 200 and 002) which can appear at higher MgO/ZnO atomic ratios [20]. The MZO peak is slightly shifted when compared to that of intrinsic ZnO films, also deposited on FTO [21] (ICDD 00-003-0752). This slight peak shift corresponds to a slightly smaller MZO lattice constant (5.18 Å) compared to ZnO films (5.21 Å). The buffer/absorber lattice mismatch can be an indicator of the quality of the junction that will form between the two, as a larger lattice mismatch can lead to a large number of dislocations and defects at their interface [22,23]. MZO has a large lattice mismatch with the CdTe zinc blende (cubic) structure which has a lattice constant c of 6.48 Å. The mismatch is even larger considering the MZO lattice constant a (3.17 Å, c/a = (8/3)1/2 = 1.633, [24]). The lattice mismatch is higher than that between CdS and CdTe (the wurzite CdS lattice constants are a = 4.136 Å, c = 6.713 Å, ICDD 00-006-0314). Considering this large lattice mismatch, it appears that a favourable band alignment can also mitigate the negative effect of a high defect density at the buffer/absorber interface.

4.4. TCO properties

In this section different TCO materials and glass substrates have been used to improve the SLG/FTO/MZO interface. The electrical properties of the investigated TCOs are summarised in Table 1. Each film was deposited to a thickness which produces a sheet resistance, R_sheet, of lower than or equal to 10 Ω/sq. ITO had the lowest resistivity due to both a high carrier concentration and mobility. AZO exhibited the highest resistivity, although this slightly improves when the AZO film is deposited on BSG. ITiO achieves a low resistivity due to a very high mobility [25]. The transmission (T%) and the 100 − A(%) spectra of each TCO are shown in Fig. 7(a) and (b). The 100 − A(%) spectrum was calculated by:

\[
100 - A(\%) = T(\%) + R(\%)
\]

where R% is the reflectance of the layers stack. The 100 − A(%)
data present the light available through the TCO assuming no reflection losses at the air/glass and TCO/air interface during the measurement. Using this data as a comparison is useful because the TCO transmission and reflection spectra are affected by the films subsequently deposited on top of the TCO (in this case the MZO and CdTe), due to differing refractive indices of these layers; this means that the solar cell structure can be optimised to maximise T% and minimise R%, while the absorption is a characteristic of the glass/TCO only and is an effective measure of the optical quality of the film. The transmittance drop in the near-infrared (NIR) is strongest for ITO, due to the high free carrier absorption caused by the high free carrier density of the film. ITiO does not show free carrier absorption within the wavelength range analysed due to the low free carrier concentration and thickness. Below the absorption onset of CdTe at 826 nm, ITiO exhibits very high 100 − A(%). The AZO deposited on BSG also has high 100 − A(%) within this range in the visible and near infrared, but a higher absorption in the UV range caused by the lower band gap of ZnO based TCOs compared to In$_2$O$_3$ based TCOs. By comparing the optical properties of AZO films deposited on the different glass substrates it is clear that BSG glass strongly improves the transparency of the stack, due to a lower iron content than SLG [26]. Also, because of the higher conductivity of the film deposited on BSG, a thinner AZO film is sufficient to obtain an equivalent sheet resistance than AZO on SLG. FTO represents a compromise in electrical and optical characteristics compared to the other TCOs in this study.

Fig. 9. The EQE spectra of CdTe solar cells deposited on FTO on SLG (a), ITO on SLG (b), AZO on SLG (c), AZO on BSG (d) and ITiO on BSG (e). For comparison the 100 − Absorption(%) spectra and the Transmittance spectra of the TCOs has been added to each graph.
4.5. Performance of the TCO/MZO combinations

Device performance parameters are presented in Fig. 8. The performance of the different glass/TCO/MZO combinations does not depend on the properties of the TCO only, but also on the interface properties of the window layer stack. AZO TCOs deposited on SLG substrates in combination with MZO buffers produce efficiencies comparable to the FTO baseline. When combined with a thinner BSG substrate, the efficiency is improved largely through an increased Jsc. Devices incorporating ITO TCOs yield lower efficiencies, although the TCO has a high conductivity and high transparency. The low efficiencies are primarily a consequence of the low Voc and FF of the devices, while the Jsc is relatively high due to the high transparency of the material. Devices incorporating ITO yield relatively low efficiencies due to poor FF and Jsc. The low current output is due to the high free carrier absorption in the VIS-NIR wavelength range. The EQE of CdTe devices incorporating the different glass/TCO/MZO combinations are shown in Fig. 9 in comparison with the Transmission and 100% Absorption(%) spectra. The UV absorption edge of devices incorporating the high band gap TCOs (ITO: E_g = 3.85 eV, FTO: E_g = 3.85 eV, ITO: E_g = 3.93 eV) is shifted to longer wavelengths, presumably by the smaller band gap of the MZO layer, whilst this is not observed when using an AZO TCO, which has a lower band gap than MZO (AZO: E_g = 3.30 eV). Generally, the transmittance spectrum is lower than the EQE. This suggests that there are more photons reaching the absorber layer than implied by the transmittance curves of the glass/TCO combination only, and that the further addition of MZO and CdTe is reducing the interfacial reflectance. The 100% Absorption(%) data, on the other hand, is the maximum transmission limit. Reducing the gap between this limit and the EQE can be achieved by maximising photo-generation and extraction efficiencies of charge carriers. This gap can be visualised in Fig. 9 as the red area between the 100% Absorption(%) curve and the EQE curve. Following this approach, the SLG/AZO devices are the most effective in converting the available light, converting roughly 90% of the available, non-absorbed, photons. This percentage was calculated by dividing the value of maximum ideal Jsc (Jsc(max)), and the value of device Jsc estimated from the EQE data (Jsc(EQE)). Jsc(max) was calculated by

\[ J_{sc(max)} = q \int \frac{1}{\lambda} \phi(\lambda) d\lambda \]

where q is the electron charge, A(\lambda) is the wavelength dependent film absorption and \( \phi(\lambda) \) is the wavelength dependent photon flux of the AM1.5G spectrum. This calculation assumes that all electrons not absorbed by the glass substrate and the TCO will be converted into current by the cell. Jsc(EQE) is calculated by

\[ J_{sc(EQE)} = q \int \frac{EQE(\lambda)}{\lambda} \phi(\lambda) d\lambda \]

where EQE(\lambda) is the wavelength dependent EQE response of the solar cell. Devices with AZO deposited on BSG, although having a higher Jsc and a larger number of available photons, convert a lower fraction of the available light, converting roughly 90% of the available, non-absorbed, photons. This percentage was calculated by dividing the value of maximum ideal Jsc (Jsc(max)), and the value of device Jsc estimated from the EQE data (Jsc(EQE)). Jsc(max) was calculated by

\[ J_{sc(max)} = q \int \frac{1}{\lambda} \phi(\lambda) d\lambda \]

where q is the electron charge, A(\lambda) is the wavelength dependent film absorption and \( \phi(\lambda) \) is the wavelength dependent photon flux of the AM1.5G spectrum. This calculation assumes that all electrons not absorbed by the glass substrate and the TCO will be converted into current by the cell. Jsc(EQE) is calculated by

\[ J_{sc(EQE)} = q \int \frac{EQE(\lambda)}{\lambda} \phi(\lambda) d\lambda \]

5. Conclusions

This study focused on the analysis and improvement of the window layer for a thin film CdTe solar cell, including the glass substrate. The band gap of MZO films was widened by increasing the deposition temperature during sputtering. Results suggest that the band gap increase helps create a favourable band alignment between the MZO layer and the CdTe absorber. The MZO films are uniformly deposited on the TCO surface. However, MZO films have a low doping density and a larger lattice mismatch factor with CdTe if compared with CdS, which can introduce a high number of interface defects. Results suggest that a favourable buffer/absorber band alignment positively dominates the negative effects due to the large MZO/CdTe interface lattice mismatch and the low MZO doping density. A number of TCOs were also examined as partners for MZO and as alternatives to FTO. AZO TCOs, when deposited on borosilicate glass, yielded better opto-electronic properties and yielded the highest Jsc and overall efficiencies. ITIO TCOs showed exceptional opto-electronic properties thanks to the high free carrier mobility and relatively low carrier concentration in the films. However, in this study, indium oxide based TCOs (ITO and ITIO) did not yield efficiencies as high as those of devices including AZO and PTO in combination with MZO. The TCO/buffer interface chemistry and/or the band alignment may play a key role in the functioning of these devices and might explain the different TCO behaviour, however further investigation is required to improve our understanding on the mechanism occurring within the window structure of these samples.

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