A New TCO/window-buffer Front Stack for CdTe Solar Cells and its Implementation

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A new TCO/window-buffer combination is Abstract presented for use in thin film CdTe solar cells. The TCO laver is cadmium tin oxide (CTO) and the window-buffer layer is tin magnesium oxide (TMO). Zinc magnesium oxide (ZMO), previously utilized by other groups and our group, was also prepared. The layers were produced by hollow cathode sputtering and were characterized by optical and Hall effect measurements. It was found that the carrier concentration of the window (buffer) layer at a fixed Mg concentration could be controlled CdTe solar cells were fabricated on over a wide range. glass/TCO/TMO and other bilayers without use of CdS. Quantum efficiency data confirmed the greatly improved blue response of the cells.

I. INTRODUCTION

The efficiency of solar cells based on thin film CdTe depends strongly on the nature and properties of the transparent conducting oxide (TCO) and buffer front stack layers as well as the doping and lifetime of the CdTe [1-4]. Proper engineering of the front stack offers several opportunities for improvement of J_{sc} , V_{oc} , and FF, namely: i) increased J_{sc} via reduction of TCO optical absorption and reduction of the blue loss stemming from buffer layer absorption; ii) increased FF via reduced TCO sheet resistance; and iii) increased V_{oc} via reduction of dark current and recombination at the absorber-buffer interface. The interface recombination is controlled by the size and sign of the absorber-buffer conduction band offset (CBO).

This work targets a high mobility, low absorption TCO and a wide gap buffer layer offering minimal blue loss and an optimized $\Delta E_c^{abs-buff}$ band offset. Modeling using SCAPS, both at NJIT and elsewhere (e.g. CSU), has shown that a type I heterojunction (straddling, with $\Delta E_c > 0$) is preferable to a type II heterojunction (staggered, with $\Delta E_c < 0$), with ΔE_c preferably lying in the range 0.1 – 0.3 eV. (In this paragraph, $\Delta E_c > 0$ means that the CBM rises in going from the absorber to the buffer.) If $\Delta E_c > 0.4$ eV the collection of the photocurrent would be hindered and temperature dependent. We have explored various combinations of TCO and buffer, including cadmium tin oxide Cd_xSn_yO (CTO) as the TCO, and $Zn_{1-x}Mg_xO$ and $Sn_{1-x}Mg_xO$ as the buffer layer. The use of $Sn_{1-x}Mg_xO$ in this capacity has not previously been reported. The workhorse front end structure has, for several decades, been SnO_2 :F/CdS. Record cells have been reported using the TCO/HRT/buffer combinations $Cd_2SnO_4/Zn_{1-x}Sn_xO/CdS$:O [1] and more recently SnO_2 :F/Z $n_{1-x}Mg_xO$ which contains neither a separate HRT layer nor a CdS layer [5].

II. EXPERIMENTAL

In the work reported here, the metal oxides CTO, ZMO and TMO are deposited by hollow cathode sputtering (HCS) using a proprietary dual-cathode linear source operated in a reactive mode [6]. HCS is a versatile, scalable process for metal oxide deposition featuring low lattice damage in the deposited layers. Low cost metal targets are used and mixed metal oxides can be produced. A photo of the deposition system is shown in Fig. 1.



Fig. 1. Advanced hollow cathode sputtering system at NJIT.

Film thicknesses are determined using a Dektak IIA stylus profilometer. Optical properties are determined using a

Filmetrics F10-RT-UVX measurement system. Resistivity and mobility are determined using an Ecopia 21 HMS-3000 Hall system in a square van der Pauw configuration formed by etching. The magnetic field (0.57 T) was measured using an AlphaLab Hall magnetometer. Solar cell fabrication involves the steps of CdTe deposition on glass/TCO/ZMO by closespaced sublimation ($T_s \leq 610$ °C), CdCl₂ treatment, NP etch, back contact application (including Cu_xO deposition and annealing), and scribing for cell definition. The use of O_2 during CSS [7] was discontinued for these experiments. Solar cells were characterized by J-V curves at AM1.5 100 mW/cm² obtained using a Newport xenon solar simulator and a Keithley 2401 sourcemeter, and by quantum efficiency measurements using a tungsten-halogen source with light dispersed by a grating monochromator and referenced to a calibrated silicon sensor from Gamma Scientific.

III. TCO LAYER PROPERTIES

To prepare cadmium tin oxide (CTO) the cathode was fitted with one pure Cd target and one pure Sn target. The principal variables studied were substrate temperature, target powers, and oxygen flow. The optical properties (spectral *T*, *R*, *A* and T/(1-R)) of a representative as-grown CTO layer are shown in Fig. 2. The properties are comparable to films produced in earlier work [8-10]. In contrast to some other TCOs, the free carrier absorption in CTO only becomes significant for $\lambda >$ 1000 nm and so does not reduce the J_{sc} of a CdTe-based solar cell ($E_g^{CdTe} = 1.5 \text{ eV}$).



Fig. 2. Specular spectral transmittance and reflectance of Cd_xSn_yO produced by HCS.

We may extract the optical absorption coefficient $\alpha(\lambda)$ using the equation

$$\alpha(\lambda) = -\frac{1}{t} \ln \left(\frac{T(\lambda)}{1 - R(\lambda)} \right) \tag{1}$$

where *t* is the film thickness. Since the fundamental absorption is both direct and allowed the band gap can be found from the usual $(\alpha E)^2$ versus *E* (Tauc) plot. The bandgap for CTO film B155is $E_g = 3.41$ eV, with values typically lying between 3.25 - 3.41 eV.

Table I shows the electrical parameters of three of the more interesting CTO films. Film B265, with resistivity $\rho = 3.78 \text{ x}$ 10⁻⁴ Ω cm, had an excellent mobility $\mu = 46.5 \text{ cm}^2/\text{Vs}$, and a sheet resistance of 6.5 Ω/sq . We note that films with high carrier concentrations (e.g. B166, with $n_e = 1.05 \text{ x} 10^{21} \text{ cm}^{-3}$) tend to have lower mobilities because of increased ionized impurity scattering [6, 11].

 $TABLE\ I$ Electrical properties of $CD_xSn_vO,\ Zn_{1-x}MG_xO,\ Sn_{1-x}MG_xO$ and $Zn_xSn_vMG_zO$ layers prepared by hollow cathode

Run #	Matl.	<i>R_{sh}</i> Ω/sq	Carrier conc. <i>n</i> e (/cm³)	Mobility µ (cm²/Vs)	Resistivity ρ (Ω cm)
B265	СТО	6.5	3.55 x 10 ²⁰	46.5	3.78 x 10 ⁻⁴
B160	СТО	7.9	4.08 x 10 ²⁰	41.9	3.65 x 10⁻⁴
B166	СТО	4.3	1.05 x 10 ²¹	30.5	1.95 x 10⁻⁴
B184	ZMO	4.5k	1.05 x 10 ¹⁹	8.64	6.89 x 10 ⁻²
B189	ZMO	17M	5.52 x 10 ¹⁵	1.94	5.83 x 10 ²
B274	тмо	157M	1.84 x 10 ¹⁴	6.0	5.63 x 10 ³
B322	ZTMO	1.59M	1.16 x 10 ¹⁷	5.2	1.04 x 10 ¹

Scrutiny of the deposition conditions for these and other CTO films suggests that the Cd/Sn ratio in the film plays a strong role in determining carrier concentration n_e , resistivity ρ , and also optical properties. A temperature-dependent Cd desorption process at the film surface appears to exist, and film mass measurements support this hypothesis. The Cd loss could be mitigated by increasing the oxygen partial pressure. We concluded that n_e declined with decreasing Cd/Sn ratio. Mamazza *et al.* co-sputtered CdO and SnO₂ at RT to produce amorphous CTO films that became polycrystalline after annealing above 580 °C [9]. In contrast, we have produced CTO films with $\mu > 40$ cm²/Vs in the as-deposited state. Meng *et al.* also produced CTO by RF magnetron sputtering but the films required annealing to attain a low resistivity [10].

Hall effect data confirms the n-type conductivity of all CTO films made to date. The nature of the donors was in the past commonly ascribed to point defects such as oxygen vacancies or interstitial Cd [12] but more recent work indicates that the oxygen vacancy is a very deep donor with transition energy at $E_{\text{CBM}} - 0.66$ eV while the Sn_{Cd2} anti-site defect is a shallow donor at $E_{\text{CBM}} - 0.05$ eV with a low formation energy [13].

Figure 3 shows an uncalibrated depth profile of a CTO film on soda-lime glass. There would appear to be a slight gradient in the Cd/Sn ratio. (We plan to investigate this using optical emission spectroscopy conducted on the HC discharge.)



Fig. 3. Depth profile obtained using RF GD-OES of Cd_xSn_yO film produced by HCS.

IV. WINDOW LAYER PROPERTIES

To prepare $Zn_{1-x}Mg_xO$ films two $Zn_{0.85}Mg_{0.15}$ metal alloy targets were fitted to the cathode; $Sn_{1-x}Mg_xO$ films were prepared using two $Sn_{0.85}Mg_{0.15}$ targets; $Zn_xSn_yMg_zO$ films were prepared using one of each of the two target types.

The optical data for the ZMO film B189 is shown in Fig. 4. A sharp absorption edge is seen, blue shifted from that of ZnO.



Fig. 4. Spectral transmittance and reflectance of $Zn_{1-x}Mg_xO$ produced by HCS.

Tauc plots for ZnO, ZMO film B240 (prepared using one Zn and one Zn_{0.85}Mg_{0.15} target), and ZMO film 189 are shown in Fig. 5. The band gaps are 3.30, 3.38, and 3.505 eV, respectively. Also shown in Fig. 4 is data for TMO film B270. Both layers exhibit almost zero absorption for $\lambda > 400$ nm. The bandgap of the TMO film was found to be 4.24 eV, one of the largest values we have measured for this class of material.



Fig. 5. Determination of band gap of $Zn_{1-x}Mg_xO$ from a plot of $(\alpha E)^2$ vs. photon energy *E*.

Regarding ZMO, the ionic radius of Mg²⁺ (0.57 Å) is close to that of Zn²⁺ (0.60 Å) [14] and allows it to substitute without change in the wurzite structure, at least for x < 0.4. The variation of bandgap of with x is found to be approximately linear and we deduce from a figure given in [15] that $E_g(x)$ is given by

$$E_a(x) = 3.296 + 2.19x \text{ eV}$$
 (2)

Based on this equation, the Mg contents in the two ZMO films B240 and B189 are x = 0.04 and 0.10, respectively. For larger values of x phase separation may occur with some MgO (rocksalt structure) occurring. We found that the ZMO films prepared using two Zn_{0.85}Mg_{0.15} targets exhibited exceptionally bright photoluminescence even at room temperature [16].

The as-deposited ZMO films were n-type and, for a given Mg content, it was found that the carrier concentration could be varied over a wide range $(10^{15} - 10^{19} \text{ cm}^{-3})$ through control of the oxygen flow rate during deposition (see Fig. 6).



Fig. 6. Control of ZMO carrier density via flow rate of O_2 during deposition by HCS.

Detailed electrical properties for ZMO, TMO, and ZTMO films are given in Table I above.

V. DEVICE PROPERTIES

The superstrate devices being investigated in this program have the general structure: glass/TCO/window/CdTe/ohmic contact. With ZMO replacing CdS as the window layer, the band diagram of this type 1 (straddling) heterojunction (HJ) device becomes that shown in Fig. 7. The relevant material parameters needed to construct such a diagram are shown in Table II.



Fig. 7. Schematic band diagram for the ZMO/CdTe straddling heterojunction interface region showing a $\Delta E_c > 0$ band offset (several features not to scale).

TABLE II FILM PARAMETERS (BANDGAP AND ELECTRON AFFINITY, IN EV) NEEDED TO CONSTRUCT THE BAND DIAGRAM

Property	FTO	СТО	CdS	ZnO	ZMO	CdTe
<i>Е</i> _g (300 К)	≈4.2	3.3– 3.7*	2.4	3.3	3.4– 3.7	1.5
χ	4.8– 5.3	4.2	4.5	4.4– 4.5	4.0– 4.2	4.3- 4.4

From Anderson's rule, the CBO ΔE_c^{And} between the absorber and the window layers is defined as (and = - ΔE_c of page 1):

$$\Delta E_c^{And} = E_c^{abs} - E_c^{window} = \chi_{window} - \chi_{abs}$$
(3)

By raising E_c of the Zn_{1-x}Mg_xO window layer by introducing Mg, the magnitude of ΔE_c^{And} can be brought into the favorable range of 0.1 – 0.3 eV (spike rather than cliff offset). The introduction of ZMO into thin-film solar cells was first studied in the context of CIGS [17-21] and later in CdTe [4,5].

Hollow cathode sputtering (HCS) was successfully used to prepare an entire front stack on glass suitable for CdTe solar cells that performed the roles of TCO, high resistance layer, window layer, and emitter. Using such stacks, viable CdTe solar cells were fabricated. The *J-V* curve of such a cell prepared on glass coated with CTO and overcoated with a ZMO window layer is shown in Fig. 8. This preliminary experiment yielded a very good J_{sc} of 25.8 mA/cm². The reason for the poor V_{oc} is not known at present but could be due either to the use of CTO or ZMO, or to sub-optimal cell processing. It was decided after these early experiments that the number of uncertainties should be reduced and subsequent ZMO development was conducted on commercial FTO.



Fig. 8. Light J-V curve for CdS-free CdTe solar cell on HCS CTO as TCO and HCS ZMO as wide-gap window layer.

For the glass/FTO/ZMO/CdTe cells we observed that the linear dark J - V curves maintained a conventional diode shape with low series resistance and high shunt resistance in the dark. Interestingly, the curves exhibited an increased turn-on voltage relative to that obtained using CdS, as shown in Fig. 9. The magnitude of the J-V voltage shift was found to correlate with ZMO thickness, with O₂ flow also playing a role.



Fig. 9. Dark J-V characteristics for CdS/CdTe and two ZMO/CdTe solar cells on FTO revealing the substantial voltage shift obtainable with ZMO.

Regarding the light *J*-*V* characteristics for 1-sun illumination we have found that a large dark *J*-*V* voltage shift correlates with kinky light *J*-*V* curves showing photocurrent suppression and an inflection as V_{oc} is approached. This type of *J*-*V* curve is not uncommon in experimental devices. It has been observed in both CIGS and CdTe solar cells containing $Zn_{1-x}Mg_xO$ buffer layers with a Mg content greater than the optimal value [22, 23]. This is shown in Fig. 10 for a ZMO/CdTe cell on FTO that has an impressively high V_{oc} of 878 mV but only a 4.2% efficiency. Use of a higher CdTe deposition temperature and a thinner ZMO layer enabled the kink to be largely eliminated and a respectable efficiency of 10.5% to be achieved [16].



Fig. 10. Light and dark J-V curves for ZMO/CdTe solar cells with different ZMO thickness.

Since carriers have to surmount the ΔE_c barrier by thermionic emission (TE) it is clear that a sufficiently large ΔE_c offset will affect the transport properties of these HJ devices. For barriers > 0.4 eV, TE cannot support a J_{ph} of 25 mA cm⁻² and so reduced collection will occur. If the dark current is modulated by a factor exp($-\Delta E_c/kT$) this can account for the observed shift in the dark J-V curve. We have conducted simulations of TCO/ZMO/CdTe devices using SCAPS and can qualitatively reproduce the photocurrent suppression if the doping of the ZMO is too low (Fig. 11).



Fig. 11. SCAPS simulation of J-V curves of CdTe solar cells as a function of ZMO doping ($\Delta E_c = 0.4$ eV, TCO doping 10¹⁸ cm⁻³). Green, light blue (overlapped), pink and red indicate ZMO doping of 0, 10¹⁴, 10¹⁶ and 10¹⁸ cm⁻³, respectively.

Using ZMO and TMO window layers in devices fully fabricated at NJIT with the structure glass/FTO/ZMO/CdTe, glass/CTO/ZMO/CdTe or glass/FTO/TMO/CdTe has enabled us to almost completely eliminate the blue loss previously caused by optical absorption in the dead CdS layer. The dramatic improvement in external quantum efficiency (*EQE*) for $\lambda < 550$ nm exhibited by devices using ZMO or TMO as a window layer is shown in Fig. 12. At 400 nm, for example, the *EQE* can be doubled. The short-circuit current density implied by integration of the measured *EQE* multiplied by the 100 mW cm⁻² AM1.5 solar photon flux distribution is 25.8 mA cm⁻² for a device with ZMO, and 25.4 mA cm⁻² for TMO, compared to 20.4 mA cm⁻² for a standard device with CdS.



Fig. 12. External quantum efficiency versus wavelength for conventional CdTe cells on FTO/CdS and the new cells on FTO/ZMO or FTO/TMO.



Fig. 13. Dark and light J-V curves for CdS-free CdTe solar cell with HCS $Sn_{1-x}Mg_xO$ (TMO) as a wide-gap window layer.

The use of TMO as the window layer in CdTe solar cells eliminated the photocurrent suppression often seen with ZMO. Thus, the dark and light *J-V* curves for a TMO/CdTe cell are shown in Fig. 13. A good J_{sc} of 24 mA/cm² was achieved and the cell efficiency was 10.7%. The somewhat disappointing V_{oc} of 760 mV suggests that ΔE_c remains to be more favorably adjusted. While ZTMO has been prepared, results for cells using this type of window layer are not yet available.

VI. CONCLUSIONS

A custom-built hollow cathode sputtering (HCS) system was used to prepare high quality CTO, ZMO, TMO, and ZTMO layers for use in CdTe solar cells. CTO layers with electron mobilities up to 46.5 cm²/Vs were demonstrated. Carrier concentrations in ZMO could be controlled over the range 10¹⁵ – 10¹⁹ /cm³. High J_{sc} 's up to 26 mA/cm² demonstrate that ZMO and TMO provide a means to obtain competitive CdTe solar cells with improved blue response due to elimination of CdS. With ZMO, V_{oc} 's up to 878 mV were achieved. The flexibility of HCS to produce almost any type of metal oxide offers a route for comprehensive optimization of the entire front side (TCO/window layer) of thin film CdTe solar cells including control of ΔE_c at the absorber-buffer interface.

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