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# Examining the role of ultra-thin atomic layer deposited metal oxide barrier layers on CdTe/ITO interface stability during the fabrication of solution processed nanocrystalline solar cells

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## ABSTRACT

Solution processed CdTe layers are a potentially low-cost alternative for use in thin-film solar cells. We have recently reported the use of such nanocrystalline layers within ITO/CdTe/ZnO/Al device architectures. One key concern with this type of device structure is the possibility of atomic scale interdiffusion between the ITO and CdTe layers, which can result in deleterious n-type doping of the CdTe layer. Rutherford Backscattering has been used to study the chemical composition across the ITO/CdTe interface as a function of thermal annealing temperature. Through these measurements we verify that interdiffusion is observed across the interface for annealing temperatures above 200 °C, and the extent of interdiffusion increases with temperature. Ultra-thin alumina, zirconia and titania layers deposited between the ITO and CdTe layers have been studied for their potential to act as a diffusion barrier. All investigated barriers successfully suppress interdiffusion. The outcomes of these compositional studies are directly compared to solar cells fabricated under analogous processing conditions, demonstrating improved cell performance.

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## 1. Introduction

The limited nature of fossil fuel reserves within the Earth's crust provides the driving force for the advancement of technologies which harness renewable energy sources as the best solution to this problem. Since their invention in Bell Labs in the 1950s [1], silicon based solar cells have emerged as a lucrative technology for capturing fractions of the 107 PW of solar radiation incident upon our planet [2]. Recent years have seen the wide scale adoption of this technology by private households and commercial solar farms. However, the current costs involved in the production, installation and end-of-life disposal of this technology are greater than those of other energy harvesting technologies and significant cost reductions are still imperative for its successful implementation as a viable source for electricity production in the future [3].

To circumvent the high material and processing costs of conventional silicon based solar cells, a variety of thin-film based metal

chalcogenide (CdTe,  $\text{Cu}_2\text{In}_x\text{Ga}_{1-x}\text{S}(\text{Se})$ ,  $\text{Cu}_2\text{ZnSnS}_4$ ) technologies have been developed [4]. Whilst most of these are fabricated through expensive vacuum-based batch processes, a number of examples based on solution-processed, printable alternatives exist [3,5–7]. Cost-benefit analyses have indicated that solution processing may be a vital factor for driving down the cost of solar cell technologies, provided that efficiencies comparable to devices made through vacuum processing can be achieved [8].

With this goal in mind, our work to date has focussed on the development of solution processed CdTe devices [6,9]. The use of solution processing of CdTe based devices was first explored in the mid 1990s by the group of Ginley who used spray deposition of methoxide passivated nanoparticulates within a standard superstrate device configuration [10]. Impressive device efficiencies of up to 12.7% have been reported for devices that have been fabricated using such a deposition technique with micrometre thickness CdTe layers [11]. Subsequent efforts by the groups of Alivisatos [5] and Carter [12], showed that through using nanorod inks, an inverted superstrate configuration of simple CdTe/CdSe bi-layer or CdTe-only Shottky solar cells could be fabricated, with CdTe thicknesses in the hundreds of nanometre range and efficiencies of up to 5%. Building on this bi-layer device architecture,

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we showed that through the use of layer-by-layer deposition, as well as controlled chemical and thermal treatment steps the efficiency of CdTe/ZnO solar cells could be fabricated with increased efficiencies of up to 7.3% [9]. Although this efficiency is still relatively low, significant scope for improvements still exist by further optimizing the device structure and the contact materials.

It has been suggested that for CdTe to be a viable solar cell candidate in the future, device thicknesses must be reduced from the micrometre range, as employed by Ginley and all current commercial CdTe devices, to the hundreds of nanometre range [13]. This latter range is suitable for the CdTe/ZnO devices studied here, which due to the nanocrystalline nature of the CdTe grains can result in less shunting compared to traditional superstrate devices with the required micron sized grains for high performance. While a natural caveat of our CdTe/ZnO device structure is that the p–n junction is spatially located away from the light absorbing interface, which may create charge transport problems for thick devices [14,15], diffusion-based transport dominates within the ultra-thin devices that are of interest here [6]; thus enabling high efficiencies to be maintained in this CdTe thickness range.

The process of manufacturing solution processed CdTe bi-layer solar cells within an inverted superstrate configuration requires a series of deposited layers of nanocrystalline CdTe on ITO, each annealed at temperatures between 300 °C and 350 °C in air. A layer of colloidal n-type ZnO and Al electrode is then, respectively, deposited from solution and thermal evaporation onto the multi-layered p-type CdTe. A key problem with this device architecture, which has been previously postulated by Olson et al. [16], is that any interdiffusion of the ITO and CdTe will result in n-type doping of the CdTe at the ITO/CdTe interface. If this occurs, the p–n type structure of the solar cell system will become an n–p–n structure, thereby reducing the available photocurrent in either direction in the absence of an external potential [17].

To this extent, in the early work on sintered nanocrystal cells by Alivisatos and co-workers, an ultrathin layer of alumina ( $\text{Al}_2\text{O}_3$ ) was deposited on top of the ITO via atomic layer deposition (ALD). In these devices it was found that the PV performance was highly dependent on the thickness of the insulating alumina layer. Optimal performance was obtained with an alumina thickness of 0.2 nm, with lower thicknesses leading to device shorting and higher thicknesses leading to blocking effects [18]. Using a layer-by-layer approach device shorting is less of a concern; however, it is possible that an ultra-thin interfacial layer of alumina may serve as a barrier to prevent undesirable indium diffusion [19,20].

The aim of this work is to investigate the evolution of the compositional nature across the interface between the ITO and CdTe layers during the processing conditions used for making solution processed CdTe bi-layer solar cells. Rutherford backscattering (RBS), because of its suitability for depth profiling thin films of light materials that contain heavy elements, has been used to determine the compositional depth profile. In addition, we explore the use of atomic layer deposited metal oxide thin films between the ITO and the CdTe as potential diffusion buffer layers. The findings from these studies are compared directly to solar cell devices, thus giving insight into the role that the ITO/CdTe interface has on the performance of our simple solar cell devices.

## 2. Materials and methods

### 2.1. RBS sample preparation

RBS analysis was conducted on glass/ITO (Kintec 5  $\Omega$ /sq.)/CdTe samples that were prepared through a layer-by-layer assembly of colloidal CdTe nanoparticles in a process that is described in detail elsewhere [6]. For this study, we investigated the influence of

different annealing temperatures between each of the four nanocrystalline CdTe layers deposited in the assembly process. The final CdTe layer thickness was fixed at  $\sim 350$  nm for all samples. In order to determine whether the composition and structure of the ITO has an influence on the diffusion, a number of glass/ITO substrates purchased from different suppliers were examined.

### 2.2. Atomic layer deposition

Atomic layer deposition was performed on pre-cleaned glass/ITO (Kintec) substrates using a Savannah thermal ALD system. The precursor used for alumina ( $\text{Al}_2\text{O}_3$ ) deposition was trimethylaluminum (TMA). For titania ( $\text{TiO}_2$ ) and zirconia ( $\text{ZrO}_2$ ) deposition, the precursors consisted of titanium tetrachloride and zirconium tetrachloride respectively. Deposition was performed at a temperature of 150 °C through alternating pulses of the metal precursor and water, with a 30 s nitrogen purge following every pulse.

### 2.3. Solar cell preparation and characterization

A detailed description of CdTe and ZnO nanocrystal syntheses, solar cell preparation and device characterization has been reported elsewhere [6]. Briefly, ITO coated glass was cleaned through a sequential process, that included sonication in distilled water, acetone and iso-propanol for 10 min. The substrates were then cleaned by exposing to UV-ozone (at RT) for 10 min. Pyridine coated CdTe nanocrystals dispersed in 1:1 (*v/v*) *n*-propanol:pyridine were spin-cast onto ITO-coated glass slides at 800 rpm for 30 s. These samples were placed on a hot plate at 150 °C for 2 min, then immediately dipped into a solution of saturated  $\text{CdCl}_2$  in methanol at 60 °C, then rinsed gently with 1-propanol and finally dried under a nitrogen stream. The substrates were then placed onto a hot plate at elevated temperatures between 200 °C and 350 °C for 30 s. This process was repeated four times giving CdTe thicknesses of 350 nm. A 60 nm thick ZnO layer was spin-cast on the CdTe then heated at 300 °C for 2 min. All film deposition and annealing steps were performed in air. 100 nm thick aluminium contacts were deposited via thermal evaporation through a shadow mask at pressures below  $2 \times 10^{-6}$  Torr. The total measured device areas were  $\sim 0.21$  cm<sup>2</sup>. A connection point for the ITO electrode was made by manually scratching off a small area of the active layers. The completed devices were then encapsulated with glass and a UV-cured epoxy (Summers Optical, Lens Bond type J-91) by exposing to 365 nm UV-light inside a glove box ( $\text{H}_2\text{O}$  and  $\text{O}_2$  levels both  $< 1$  ppm) for 10 min.

The cells were tested with an Oriel solar simulator fitted with a 1000 W Xe lamp filtered to give an output of 100 mW/cm<sup>2</sup> at AM1.5. The lamp was calibrated using a standard Si cell from Peccell Limited which was subsequently cross-calibrated with a KG5 filtered reference cell traceable to the National Renewable Energy Laboratory. The devices were tested using a Keithley 2400 Sourcemeter controlled by LabVIEW Software. All current-voltage measurements were corrected for overestimation of the current density due to the spectral mismatch; the error was on the order of 2%.

### 2.4. Rutherford backscattering setup

RBS was performed at the STAR accelerator at the Australian Nuclear Science and Technology Organisation (ANSTO, Lucas Heights). In this investigation helium ( $\text{He}^+ 2$  MeV) and carbon beams ( $\text{C}^{++} 6$  MeV) were used. The backscattered ions detected provide a concentration depth profile that can be used to determine any interdiffusion between the solution-processed CdTe layers and the ITO substrates. It is important to note that use of He projectiles did not allow for the complete separation of the Cd,

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