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The roles of ZnTe buffer layers on CdTe solar cell performance

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ABSTRACT

The use of ZnTe buffer layers at the back contact of CdTe solar cells has been credited with contributing to recent improvements in both champion cell efficiency and module stability. To better understand the controlling physical and chemical phenomena, high resolution transmission electron microscopy (HR-TEM) and atom probe tomography (APT) were used to study the evolution of the back contact region during rapid thermal processing (RTP) of this layer. After activation the ZnTe layer, initially nanocrystalline and homogenous, transforms into a bilayer structure consisting of a disordered region in contact with CdTe characterized by significant Cd–Zn interdiffusion, and a nanocrystalline layer that shows evidence of grain growth and twin formation. Copper, co-evaporated uniformly within ZnTe, is found to dramatically segregate and aggregate after RTP, either collecting near the ZnTe/Au interface or forming Cu_xTe clusters in the CdTe layer at defects or grain boundaries near the interface. Analysis of TEM images revealed that Zn accumulates at the edge of these clusters, and three-dimensional APT images confirmed that these are core–shell nanostructures consisting of Cu_{1.4}Te clusters encased in Zn. These changes in morphology and composition are related to cell performance and stability.

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1. Introduction

With an ideal band gap (~ 1.45 eV) and large absorption coefficient ($> 10^4$ cm⁻¹) CdTe has emerged as the leading thin-film photovoltaic (PV) technology with record device efficiency currently at 21.5% [1]. Most recent advances have been due to improvements in short circuit current (J_{sc}) and fill factor (FF), whereas the open circuit voltage (V_{oc}) values have remained largely unchanged [2]. A requirement for high V_{oc} is good ohmic contact with CdTe, which is challenging due to its low doping and high work function. Consequences of such barriers include loss of open circuit voltage (V_{oc}) and fill factor (FF), which are often manifested by the presence of roll over behavior in J – V curves [3]. A common strategy to address this problem is through the insertion of a thin interfacial layer between the CdTe and metal contact [4]. Copper-doped zinc telluride (ZnTe:Cu) has been widely adopted for this role [5]. Copper degenerately dopes this layer, which narrows the barrier width and permits electron tunneling,

creating a quasi-ohmic contact [6]. Copper has been associated with several defect states located within the CdTe band gap [7], some of which have been implicated to contributing to reduced carrier lifetime and/or problems with stability [8]. First Solar Inc. recently reported that they have integrated ZnTe buffer layers into their commercial modules [9]. Incorporation of ZnTe has been credited with improving champion device efficiency as well as enhancing both the stability and temperature sensitivity of their modules. In this paper we provide nanoscale characterization of the back contact region that provides new insights into the mechanism(s) that may contribute to these improvements.

We recently introduced a back contact procedure in which ZnTe:Cu is co-evaporated at low temperature followed by activation using rapid thermal processing (RTP) [10,11]. RTP offers several advantages for this activation step including improved control, low thermal budget, and high throughput. This technique has been used in the fabrication of CdTe solar cells on flexible glass with certified efficiencies of 16.4% [12]. Previously the macroscopic distribution of Cu was characterized using secondary ion mass spectrometry (SIMS) both before and after RTP processing [10], and quantitative analysis of this data raised a number of important questions. First, the optimal dose of Cu in the as-deposited ZnTe:Cu is $Q_0 \sim 10^{17}$ cm⁻², which is about two orders of magnitude

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greater than the optimal amount required for doping CdTe as reported by Kranz et al. [13], and about an order of magnitude greater than the few nms of copper that is typically used to form Cu_xTe -based contacts [14,15]. In fitting the Cu diffusion profiles in CdTe as a function of RTP temperature the modeled dose was reduced to 20% of the experimental value in order to obtain good agreement with the SIMS data [10], which implies that the majority of the Cu supplied during co-evaporation does not enter the absorber layer. Lastly, the effective diffusivity of Cu extracted from the SIMS profiles was just $\sim 10^{-12} \text{ cm}^2/\text{s}$, which is three to five orders of magnitude less than values expected at the temperatures involved [16,17].

One limitation of SIMS is that it provides averaged one-dimensional profiles, and does not account for any lateral variations that may be present. In polycrystalline CdTe, it is well known that impurities such as Cu are preferentially transported and accumulated along grain boundaries [18]. Understanding the nanoscale distribution of these elements is critical to advancing the fundamental understanding of this material system. To this end we employ atom probe tomography (APT) and high-resolution transmission electron microscopy (HR-TEM) to quantify the changes in structure and composition in the back contact region of ZnTe:Cu contacted solar cells during RTP activation.

2. Materials and methods

2.1. Device fabrication

Fig. 1 displays a schematic cross-section of the superstrate architecture employed by the devices examined in this work. The

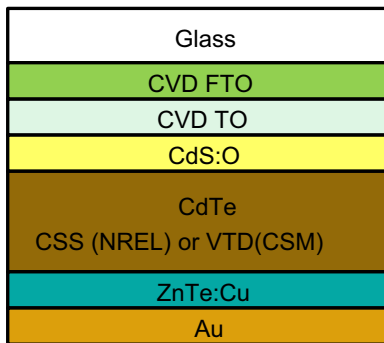


Fig. 1. Schematic cross-section of the device structures considered in this work. NREL absorbers were deposited by CSS at 600 °C while CSM absorbers were deposited by VTD at 450 °C.

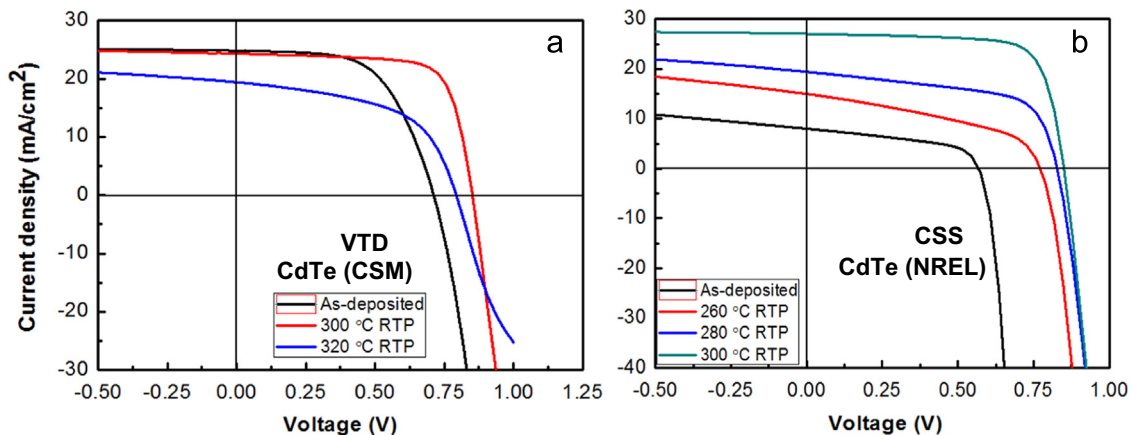


Fig. 2. Evolution of J - V performance as a function of RTP treatment temperature for devices employing (a) CSM VTD CdTe and (b) NREL CSS CdTe.

front contacts employ a tin oxide bilayer deposited on Corning 7059 glass by thermal chemical vapor deposition followed by an oxygenated CdS:O window layer deposited by reactive sputtering as described in the literature [19]. The only difference in the two sets of devices examined was the nature of the CdTe absorber layer. One set of devices employed CdTe deposited by close space sublimation (CSS) at 600 °C at the National Renewable Energy Laboratory (NREL) [19]. The second set of devices was fabricated at Colorado School of Mines (CSM) by vapor transport deposition (VTD) at 450 °C [20]. In both cases the devices were completed at CSM. Back contact preparation involved a 10 s dip in a 0.05 vol% bromine methanol solution to remove oxides and surface contaminants introduced during vapor CdCl_2 treatment. Afterwards $\sim 165 \text{ nm}$ of ZnTe:Cu ($\sim 4 \text{ wt}\%$ Cu) was evaporated by co-evaporation, followed by evaporation of $\sim 100 \text{ nm}$ of Au which served as the metallization layer. Activation of the back contact was accomplished by annealing the devices in an RTP furnace for 30 s at the desired temperature set-point, which was measured by a thermocouple in contact with the AlN susceptor as described previously [11].

2.2. Device performance

The solar cell performance was measured under simulated AM1.5 radiation using a commercial tool that is calibrated using a certified silicon standard (PV Measurements). Fig. 2 displays the evolution of the J - V behavior of these devices as a function of RTP annealing temperature. The CSS devices have very low efficiency ($< 2\%$) after application of the back contact. RTP activation leads to monotonic increases in all three contributors to efficiency (V_{oc} , FF, and J_{sc}) up until the optimal temperature which was 300 °C. In contrast, the devices fabricated at CSM display strong current collection prior to RTP activation. Application of an optimal RTP treatment increases V_{oc} and FF without altering J_{sc} . The application of excessive heat treatment reduced efficiency through a decline in all secondary parameters, and was correlated with an increased density of Cu-related defects [7].

The reasons for the strikingly different J - V evolution profiles is not fully understood. Impurities are one possibility, though the CdTe source materials used in both VTD and CSS processes had nominally identical purity ($> 99.999\%$). Another factor might be the different temperatures used, but the most plausible explanation is the presence of oxygen (6.25%) used during CSS deposition. It has been shown that oxygen can oxidize grain boundaries and inhibit interlayer diffusion [21,22]. It is postulated that the oxidized grain boundaries may limit the effective conductivity of CSS-deposited CdTe prior to RTP activation. Despite the differences prior to RTP activation, the J - V characteristics of optimally

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