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## The activation of thin film CdTe solar cells using alternative chlorine containing compounds

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### ABSTRACT

The re-crystallisation of thin film cadmium telluride (CdTe) using cadmium chloride (CdCl<sub>2</sub>) is a vital process for obtaining high efficiency photovoltaic devices. However, the precise micro-structural mechanisms involved are not well understood. In this study, we have used alternative chlorine-containing compounds to determine if these can also assist the re-crystallisation of the CdTe layer and to understand the separate roles of cadmium and chlorine during the activation. The compounds used were: tellurium tetrachloride (TeCl<sub>4</sub>), cadmium acetate (Cd(CH<sub>3</sub>CO<sub>2</sub>)<sub>2</sub>), hydrochloric acid (HCl) and zinc chloride (ZnCl<sub>2</sub>). TeCl<sub>4</sub> was used to assess the role of Cl and the formation of a Te-rich outer layer which may assist the formation of the back contact. Cd(CH<sub>3</sub>CO<sub>2</sub>)<sub>2</sub> and HCl were used to distinguish between the roles of cadmium and chlorine in the process. Finally, ZnCl<sub>2</sub> was employed as an alternative to CdCl<sub>2</sub>. We report on the efficacy of using these alternative Cl-containing compounds to remove the high density of planar defects present in untreated CdTe.

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### 1. Introduction

The leading thin film photovoltaic technology as measured by annual production is thin film cadmium telluride (CdTe). Thin film CdTe modules are manufactured in high volume on an industrial scale and have the lowest production cost of any solar module technology. The module efficiency is currently lower than crystalline silicon [1], but the cell record efficiency has recently increased markedly. The record for a laboratory cell efficiency is 20.4% [1] and a commercial module record efficiency of above 17.0% has been reported by First Solar Inc.

As deposited thin film CdTe solar cells do not perform well and it is necessary to use a cadmium chloride (CdCl<sub>2</sub>) activation process to obtain high efficiencies. Although this process is used in module manufacture, the precise mechanisms involved in the cell activation process are not fully understood. In this paper, we report on the use of different chlorine containing compounds to treat the CdTe/CdS stacks. The effects of the treatments on the electrical performance and the material properties have been analysed. The compounds used include: tellurium tetrachloride (TeCl<sub>4</sub>), cadmium acetate (Cd(CH<sub>3</sub>CO<sub>2</sub>)<sub>2</sub>), hydrochloric acid (HCl) and zinc chloride (ZnCl<sub>2</sub>). TeCl<sub>4</sub> was used to assess the role of chlorine and the formation of Te(0) on the surface, which might assist the back contact. Cd(CH<sub>3</sub>CO<sub>2</sub>)<sub>2</sub> and HCl were used to investigate the separate roles of Cd and Cl. Also, HCl does not contain any elements present in the CdTe/CdS stack. Finally, ZnCl<sub>2</sub> was studied as a possible

alternative to CdCl<sub>2</sub>, since Zn has the same electronic configuration and oxidation state as cadmium when bonded with chlorine.

### 2. Methodology

Many alternative methods of application can be used for the activation process of CdTe [2–5]. This paper is focused on the use of a wet treatment to analyse the effect of various chlorine containing compounds on CdTe solar cells. Close-space sublimation (CSS) was used to deposit both the CdS and the CdTe layers. The cells were deposited in superstrate configuration on TEC 10 fluorine doped tin oxide coated on 3 mm soda lime glass. The superstrate was passed through the deposition chambers at 0.1 Pa (mixture of N<sub>2</sub> and O<sub>2</sub>) held a few millimetres above graphite boats. The boats were heated to 650 °C and the films were sublimed on to the superstrate surface. A thin n-type CdS layer was deposited followed by ~3 μm layer of CdTe.

The variable process parameters used in the wet activation treatment were as follows: the concentration of the solution, deposition time, annealing temperature and annealing time. All the compounds used are soluble in methanol, so the temperature of the solution was not a critical parameter. The annealing temperature was varied between 300 °C and 425 °C. The concentration of the solutions was then varied to obtain the best result. Percentages were used to identify concentrations where 1% = 0.054 M, based on molecular molar mass. The range chosen depended on the compound used, but was generally between 0.1% and 2.5% (higher concentrations were used for HCl). The annealing and deposition times were varied between 5 min and 25 min. All the compounds were in powder form (Sigma Aldrich®), except HCl which

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was already in solution. The powders were dissolved in methanol using stirring. Samples were immersed in the solution for a specified time and then transferred to a ventilated oven for the annealing procedure.

The back contact was deposited on the cells following the wet treatment to allow the electrical performance of the device to be measured. A thickness of ~50 nm of sputtered gold was used for the back contact. No optimisation of the back contact was carried out. No copper was added and no etching procedure was performed so that only the effect of the activation treatment could be determined.

The electrical performance was studied using current density–voltage (J–V) curves, measured with the solar cell exposed under 1000 W/m<sup>2</sup> illumination and using an AM 1.5 filter. The values presented are averaged from a range of treated cells. Additional analysis was performed using the spectral response technique to investigate the external quantum efficiency (EQE) of a representative cell from each category [6]. Band gap values were calculated from the EQE measurements [7]. Surface chemical analysis was performed by X-ray Photoelectron Spectroscopy (XPS) using a Thermo Scientific K-Alpha Surface Analysis system. The samples were etched with argon ions for 30 s at 1 keV prior to analysis, this was done to remove any surface contamination. A flood gun was used to minimise peak shifting from charging of the sample during analysis, the area of analysis was 200 μm using monochromated copper X-rays. The composition was calculated using sensitivity factors and the peaks were fitted using Avantage software. Scanning Electron Microscopy (SEM) images were obtained using a FEI Nova 600 Nanolab using an acceleration voltage of 10 kV, which was also equipped with a Focussed Ion Beam (FIB), to prepare TEM samples using a standard in-situ lift-out method. Transmission Electron Microscopy (TEM) images were obtained with a Jeol JEM 2000FX operating at 200 kV. Chemical point analysis was carried out using an FEI Tecnai F20 Scanning TEM ((S)TEM) operating at 200 kV, equipped with an Oxford Instruments X-max 80 Energy Dispersive X-ray (EDX) detector.

### 3. Results and discussion

#### 3.1. CdCl<sub>2</sub>

As a reference, TEM images of untreated and standard CdCl<sub>2</sub> wet treated CdS/CdTe are presented in Fig. 1. After the treatment, the CdTe has large grains, without a region of smaller grains near the CdS interface. The grain boundary volume was reduced and the planar defect density inside the grains decreased following the treatment. Defects present in the untreated cells were identified as stacking faults and twins, using atomic scale high resolution-TEM [8]. The stacking faults are completely removed by the activation treatment [9]. The CdS layer was also affected by the treatment with grain enlargement and less sharp grain boundaries.

#### 3.2. TeCl<sub>4</sub>

The process parameters for the TeCl<sub>4</sub> assisted activation treatment were tested and the best conditions were found to be annealing at a temperature of 350 °C for 15 min using a solution concentration of 2% for 15 min dipping time. The best performing cell produced open circuit voltage (V<sub>oc</sub>) = 594 mV, short circuit current (J<sub>sc</sub>) = 17.91 mA/cm<sup>2</sup>, and Fill Factor (FF) = 0.43 with a cell efficiency of 4.58%. These optimised process parameters differ from those of the standard CdCl<sub>2</sub> treatment. TeCl<sub>4</sub> is a volatile compared with CdCl<sub>2</sub> and it has a lower melting point (224 °C) and a lower boiling point (380 °C). This might lead to additional reactions during the annealing procedure. It is possible that the process may lead to the formation of a surface layer that could affect the contact and lower the Fill Factor (FF). XPS analysis showed the presence of CdCl<sub>2</sub>, TeO<sub>2</sub> and Te(0) on the surface. An improvement in the V<sub>oc</sub> and J<sub>sc</sub> was observed compared with the performance of the untreated cell. Recrystallization of the CdTe layer resulted in larger grains with reduced intragranular defect density. The grains were approximately uniform in size unlike the untreated samples

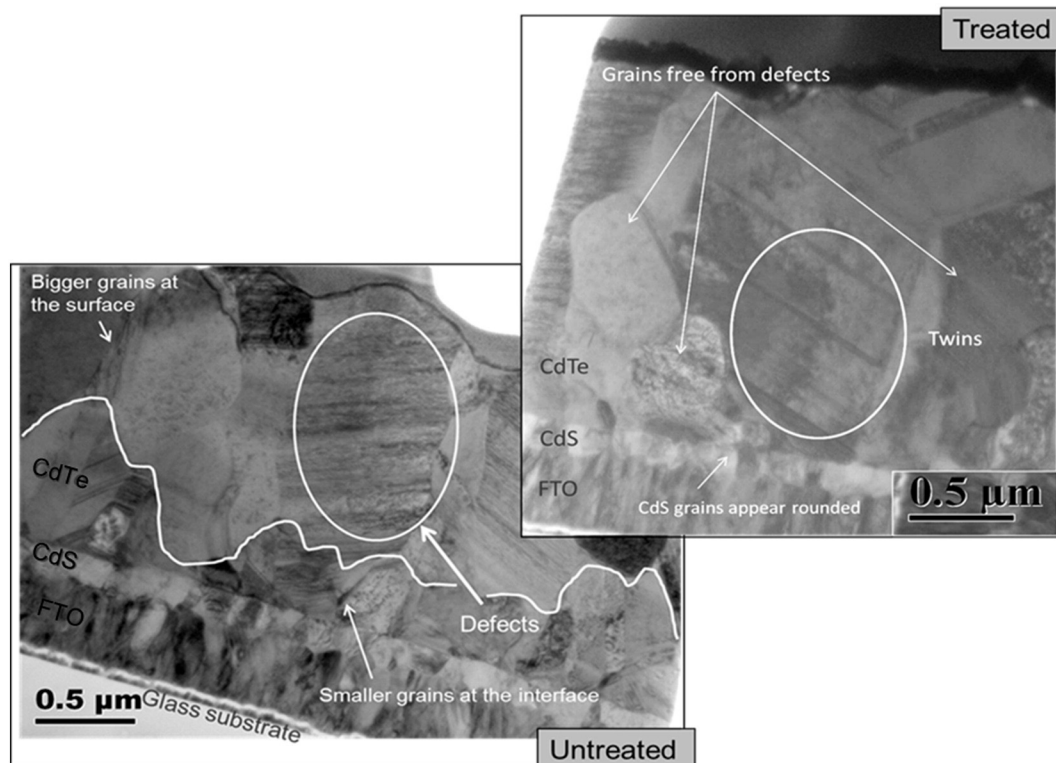


Fig. 1. TEM images of untreated and standard CdCl<sub>2</sub> treated CdS/CdTe.

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