

Critical concentration of Zinc in amorphous silicon solar cells



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ABSTRACT

Transparent conductive oxide (TCO) contacts based on zinc oxide (ZnO) are often used in thin film silicon solar cells and ZnO has a lower contamination potential compared to Tin Oxide or Indium-Tin Oxide. However, in this paper the adverse effects of zinc contamination on solar cell performance in large area systems when using non-optimal processes are demonstrated. Controlled contamination experiments were performed; optical emission spectroscopy allowed us to study the Zn release dynamic; secondary ions mass spectroscopy measurements allowed us to determine the Zn contamination threshold level in a-Si:H solar cells of 10^{18} at/cm³. Higher Zn concentrations lead to loss of cell performance in the initial state and increased light induced degradation compared to cells with lower contamination. Additionally, a simple approach for Zn contamination removal is presented.

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1. Introduction

Thin-film silicon solar cells based on hydrogenated amorphous silicon (a-Si:H) are usually deposited by Plasma-enhanced chemical vapor deposition (PECVD) on a Transparent Conductive Oxide (TCO) front contact. The most commonly used TCOs are Fluorine-doped Tin Oxide (FTO), and Zinc Oxide (ZnO, either Al-doped or B-doped) due to their the good light scattering properties and transparency [1,2], in some cases Indium-doped Tin Oxide (ITO) may be used too. During cell deposition, especially at the initial stage, the TCO surface is exposed to hydrogen radicals which may reduce the oxides to the metals. Literature indicates that ZnO is more resistant to hydrogen plasma and reduction than FTO or ITO [3–6]. No specific studies on Zn contamination in a-Si:H were found, however Zn contamination has been investigated in crystalline silicon revealing that Zn forms deep acceptor levels and other trap levels [7,8].

Recent literature investigated contamination in a-Si:H, especially the influence of oxygen and nitrogen on solar cell performance. Critical impurities concentrations were determined to be approximately between $2 \cdot 10^{19}$ at/cm³ and 10^{20} at/cm³ for oxygen and $4 \cdot 10^{18}$ at/cm³ for nitrogen [9,10]. These results are similar to earlier studies on light induced degradation indicating a threshold for oxygen and nitrogen impurities of approximately 10^{19} at/cm³ [11]. Fluorine contamination was observed to impair performance

of solar cells in production systems starting at concentrations between 10^{18} at/cm³ and 10^{17} at/cm³ [12].

Usually a microcrystalline silicon contact layer is used to achieve a good electrical contact between ZnO and a-Si:H cell [13]. Several attempts were made to implement such a layer in large area systems, however the resulting cell performance was always systematically poorer compared to cells prepared without a microcrystalline silicon contact layer. An alternative contacting method based only on amorphous silicon was found [14,15] to avoid this issue at the cost of lower transparency and consequent power loss. The origin of poor cell performance was eventually found to be Zn contamination. The effects of Zn contamination were then studied more in depth by performing on-purpose contamination leading to the results presented in this publication, additionally from the results of this study further improvement of the ZnO/a-Si:H interface could be made [15].

2. Material and methods

To study the effect of Zn contamination in a-Si:H solar cells experiments were performed by depositing a reference solar cell, then contaminating the deposition system by using a high dilution SiH₄/H₂ plasma suitable for depositing an μ c-Si layer and then depositing several solar cells using the same recipe as the reference cell. Measuring cell performance and Zn concentration in a set of similar experiments allowed to correlate Zn concentration with cell parameters.

Large area (Gen5, 1.43 m²) low pressure chemical vapor deposition (LPCVD) and PECVD systems were used to prepare a-Si:

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H cells as described elsewhere [16,17]. TCO front contacts had 27% Haze at 600 nm and the *i*-layer thickness was 220 nm. All cells used for experiments use an a-Si:H contact *p*-layer deposited using a 1:1:0.5 gas flow ratio of SiH₄:H₂:TMB (2% in H₂), 0.022 W/cm² VHF power density at 40 MHz, 0.5 mbar process pressure and 200 °C substrate temperature [16]. The complete cell deposition was performed in one single chamber as in industrial production lines. Both 1 cm² cells and laminated submodules with 10 series-connected cells and 37.87 cm² active area were used for this study. Each point plotted corresponds to the average of 64 cells or 3 submodules distributed on the glass surface to minimize random scattering effects. A Si wafer piece (single side polished, *n*-doped, ≈ 1 cm², coated with 40 nm a-Si:H:P) was positioned at the center of each glass substrate before each PECVD cell deposition run and later used as sample for Secondary Ions Mass Spectroscopy (SIMS) measurements. The *n*-type a-Si:H coating helps to reduce artefacts at the interface between crystalline Si and hydrogenated amorphous Si thus improving the measurement accuracy at the start of deposition.

Flat samples are necessary for SIMS measurements: the ZnO roughness is comparable with the total a-Si:H cell thickness and such a strong roughness would completely destroy the depth information. The comparatively small SIMS sample surface compared to the glass surface leads to deposition conditions very close to the real device under study, however an underestimation of the Zn content at the very beginning of deposition on ZnO cannot be excluded. Conversely, a direct SIMS measurement on TCO-coated glasses would not determine the trace Zn content in a-Si:H at the interface with ZnO due to the roughness and due to the abrupt change in concentration. More information on SIMS calibration and measurement conditions can be found elsewhere [18]. Although the Zn concentration varies at different positions on the glass, the center position was identified to be representative of the whole glass.

Zn contamination was generated deliberately by exposing ZnO-coated glasses to a high dilution microcrystalline silicon (μ c-Si) plasma deposition regime ($R=[\text{H}_2]/[\text{SiH}_4]=250$, 0.1 W/cm², 2 mbar) for 30 s at 200 °C substrate temperature; these glasses were not used for cell depositions but only for contaminating the deposition system. Optical Emission Spectroscopy (OES) of the plasma was performed using an Ocean Optics S2000 spectrometer, full plasma emission spectra in the range between 344 nm and 1000 nm were recorded with a time resolution of 0.1 s and the Zn emission lines at 472.21 nm, ($3d^{10}4s(^2S)4p:3P^0_1-3d^{10}4s(^2S)5s:3S_1$), 481.05 nm ($3d^{10}4s(^2S)4p:3P^0_2-3d^{10}4s(^2S)5s:3S_1$), and 636.23 nm ($3d^{10}4s(^2S)4p:1P^0_1-3d^{10}4s(^2S)4d:1D_2$) [19] were further analyzed.

Fourier-transform photocurrent spectroscopy (FTPS) measurements were done connecting the sample through a current amplifier (Stanford Research SR570) as an external detector to a Thermo-Scientific Nicolet 8700 FTIR spectrometer. To cope with the large dynamic range the spectrum was split into 3 bands, the first band was measured without filter, the second band was measured through a red/blue color filter and the third band was measured through a Si wafer with antireflective coating. The sample spectra were referenced to the lamp spectrum measured with the internal deuterated triglycine sulfate (DTGS) detector. The corrected partial spectra measured with the filters were then scaled to get a single overlapping curve [20].

More information on current-voltage (*I*/*V*) measurements, external quantum efficiency (EQE) measurements and light induced degradation procedure can be found elsewhere [16,21].

3. Results and discussion

To study the effect of Zn contamination in a-Si:H solar cells experiments were performed exposing ZnO-coated glasses

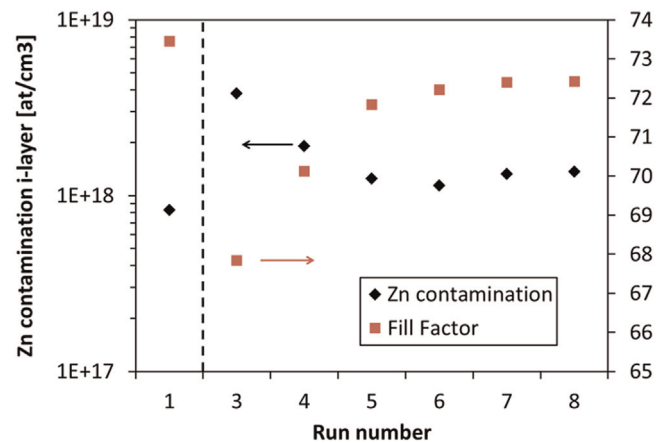


Fig. 1. Run to run evolution of the *i*-layer average Zn concentration (left axis) and fill factor (denoted FF, right axis) during one experiment. Cells in all runs except run 2 were prepared using the same deposition recipe. Run 2 was used to deliberately contaminate the deposition system by exposing a ZnO-coated glass to a high dilution microcrystalline silicon (μ c-Si) plasma deposition regime, the dashed line highlights the deliberate contamination.

deliberately to contamination using a high dilution SiH₄/H₂ plasma suitable for depositing an μ c-Si layer. Reference cells prepared after Zn contamination and without μ c-Si layer were affected by a significant reduction of fill factor (*FF*) from ≈ 73% to ≈ 60% and of open-circuit voltage (*V*_{oc}) from ≈ 880 mV to ≈ 840 mV leading to a reduction of initial conversion efficiency from ≈ 10.4% to ≈ 8.4%. A sequence of nominally identical cells deposited afterwards showed a gradual recovery of performance until the original reference performance was achieved. The evolution of the fill factor after a deliberate Zn contamination in an exemplary sequence of cells is shown in Fig. 1 together with the average *i*-layer Zn contamination. A systematic measurement of the Zn content by SIMS on several samples during performance recovery allowed to determine the effect of Zn content on cell performance.

Fig. 2 shows the conversion efficiency, the *FF* and the *V*_{oc} as a function of the average *i*-layer Zn concentration determined by SIMS in 15 independent runs. Conversion efficiency, *FF* and *V*_{oc} start to decrease when the Zn concentration is larger than 10¹⁸ at/cm³.

Zn contamination SIMS profiles of both the least contaminated and the most contaminated run are shown in Fig. 3 together with the cell parameters of a single representative cell from each run. In the SIMS profiles it was possible to identify three features. First, at a depth of less than 100 nm a *n*-SiO_x layer, then in the middle the *i*-layer and buffer, and finally at depths greater than 250 nm, the *p*-layer at the start of deposition on ZnO with a Zn concentration peak. Slight thickness differences between both samples were related to coating thickness non-uniformities, therefore the profiles were shifted in depth to obtain an overlap at beginning of the deposition (that is, the *p*-layer). The SIMS profiles indicate a strong Zn release and incorporation at the beginning of deposition when the ZnO surface is exposed to the plasma. By comparing the SIMS profiles of Carbon and Zn (data not shown), it is possible to determine that the Zn peak is limited to the SiC:H *p*-layer in regular cells with low contamination (e.g. cells using only an a-Si:H contact layer). However, the peak Zn level in the *p*-layer does not correlate with performance loss. An almost constant Zn level is then present across the *i*-layer and the average *i*-layer contamination correlates with cell performance as shown in Fig. 2. The run to run decrease of Zn contamination in the *i*-layer of a series of depositions as shown in Fig. 1 suggests that Zn contamination is mediated by internal surfaces of the reactor acting as Zn storage and releasing Zn at a constant rate. The almost constant

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