



Effect of different Na supply methods on thin Cu(In,Ga)Se₂ solar cells with Al₂O₃ rear passivation layers

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ARTICLE INFO

Keywords:

Alkali
Back contact
CIGS
Passivation
Thin films
Rear contact
Tunneling

ABSTRACT

In this work, rear-contact passivated Cu(In,Ga)Se₂ (CIGS) solar cells were fabricated without any intentional contact openings between the CIGS and Mo layers. The investigated samples were either Na free or one of two Na supply methods was used, i) a NaF precursor on top of the Al₂O₃ rear passivation layer or ii) an in situ post-deposition treatment with NaF after co-evaporation of the CIGS layer. The thickness of the ALD-Al₂O₃ passivation layer was also varied in order to find an optimal combination of Na supply and passivation layer thickness. Our results from electrical characterization show remarkably different solar cell behavior for different Na supplies. For up to 1 nm thick Al₂O₃ layers an electronically good contact could be confirmed independently of Na deposition method and content. When the Al₂O₃ thickness exceeded 1 nm, the current was blocked on all samples except on the samples with the NaF precursor. On these samples the current was not blocked up to an Al₂O₃ layer thickness of about 6 nm, the maximum thickness we could achieve without the CIGS peeling off the Al₂O₃ layer. Transmission electron microscopy reveals a porous passivation layer for the samples with a NaF precursor. An analysis of the dependence of the open circuit voltage on temperature (JVT) indicates that a thicker NaF precursor layer lowers the height of the hole barrier at the rear contact for the passivated cells. This energy barrier is also lower for the passivated sample, compared to an unpassivated sample, when both samples have been post-deposition treated.

1. Introduction

The record efficiency of Cu(In,Ga)Se₂ (CIGS) solar cells has tremendously increased over the last years due to improvements in the bulk quality and the front contact interface [1]. Just as in Si solar cell technology, the thickness of the absorber layer is expected to decrease for commercial CIGS solar cells to save costs and materials. Thus, controlling the recombination and maximizing the light reflection at the CIGS/rear contact interface will be of increasing importance for the solar cell performance.

Inspired by PERC (Passivated Emitter Rear Contact) silicon solar cells, Vermang et al. [2,3] introduced an Al₂O₃ passivation layer between the CIGS absorber layer and the Mo rear contact. To ensure an electrical contact, different kinds of nano-contacts were developed: conducting Mo-nano-spheres embedded in the passivation layer [4] or nano-openings in the passivation layer [2,3]. Whereas thin (< 15 nm) passivation layers mostly increase the open-circuit voltage (V_{OC}) by lowering the recombination rate at the rear contact [2], both, thicker Al₂O₃ layers (30 nm) [3] and Mo nano-spheres [4], also increase the short-circuit current density (J_{SC}) by increased reflection at the rear

contact and/or by phonons between Mo nano-spheres that enhance absorption. The higher V_{OC} has been explained by a field effect [5,6] (electrical passivation) due to negatively charged centers (V_{Al} and/or O_i) in oxygen rich Al₂O₃ layers [7], which lowers the interface recombination rate at the passivated areas. The electrical passivation effect increases strongly from 5 to 50 nm passivation layer thickness. The interface defect density, however, is found to only be slightly reduced compared to an unpassivated area [7].

The rear contact has been associated with a kink and roll-over in current-voltage (JV) curves of CIGS solar cells measured especially at low temperatures [8–17]. The kink has been explained by a hole extraction barrier [10], whereas the roll-over has been explained by a hole injection barrier [8,10,12]. Some studies identified the valence band off-set between CIGS and MoSe₂, that is usually formed between the CIGS and Mo [10,17], as the hole injection barrier. Even a cross-over between the dark and light curve can be explained by a barrier at the rear contact as exemplified for CdTe solar cells [11,18]. However, in the case of CIGS solar cells a conduction band offset between the CIGS and CdS [8] or an acceptor rich layer [10] in the CIGS near the front interface can also explain a kink and cross-over in the JV curve.

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<https://doi.org/10.1016/j.solmat.2018.07.017>

Received 28 May 2018; Received in revised form 12 July 2018; Accepted 17 July 2018

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The rear contact region is strongly affected by the presence of Na. Whereas the soda-lime glass substrate can act as a source of Na during the CIGS co-evaporation, Na can also be added in a thin NaF precursor layer onto the rear contact before CIGS evaporation (pre-deposition treatment, pre-DT) [19] or by evaporating NaF on top of the CIGS layer during an annealing step after its deposition (post-deposition treatment, post-DT) [9]. While there is a general agreement, that Na application or Na diffusion from the soda-lime glass enhances the electrical properties of the rear contact, the exact mechanism is unclear. For example, a roll-over measured on Na-free devices at room temperature can be reduced by a Na pre-DT [10,19]. Pre-deposited Na [19] or Na from the soda-lime glass [20,21] acts as a catalyst and promotes the formation of MoSe₂, which in turn creates an ohmic contact according to references [17,20,21]. In contrast, Yoon et al. [22] doubt that the MoSe₂ layer is the origin of the ohmic nature of the rear contact and suggest that Na reduces the barrier at the CIGS/MoSe₂ interface as it increases the (effective) doping of the CIGS or the MoSe₂ layer. Jarzembowski et al. [23] found that the rear interface recombination rate for samples with an alkali diffusion barrier is lowered by NaF post-DT. They concluded that Na passivates either the MoSe₂/CIGS interface or the MoSe₂/Mo interface.

In their first work on CIGS rear surface passivation, Vermang et al. [3] observed that the *JV* curves of solar cells with a 5 nm Al₂O₃ passivation layer with point contact openings and without adding NaF, exhibited a kink and a roll-over. Insufficient Na from the soda-lime glass was suspected and a NaF precursor layer was successfully applied on the Al₂O₃ layer before CIGS evaporation. The passivation layer without openings inhibited the electrical contact for the samples without the NaF treatment. This is in agreement with measurements on Al₂O₃ on Mo with an Hg probe [24]. The sheet resistance as measured with the Hg probe was shown to be negligible for a passivation layer thinner than 3 nm. While direct tunneling dominates for low voltages and thin Al₂O₃ layers (< 3 nm), Fowler-Nordheim tunneling dominates at voltages that are only reached at the rear contact for high forward biases much larger than *V*_{OC} and thicker Al₂O₃ layers. Vermang et al. [3] did not provide any *JV* curves for samples without nano-contacts but with NaF pre-deposition.

In this work, a series of passivated and unpassivated CIGS solar cells was produced. In order to evaluate if thin unpatterned passivation layers can provide a sufficiently strong passivation effect without blocking the current, no point contacts were etched into the passivation layers. Considering the significance of Na outlined above, the Na supply method and the Na concentration were varied in four ways (compare

Table 1

Overview of the produced samples. In column three, the first number gives the number of samples produced for every sample type and the numbers in brackets give the number of cells on every sample. The numbers in the fourth column indicates the corresponding number of ALD cycles. The number of ALD cycles replaces xx in the sample name (e.g. 'post-DT10' is a post-deposition treated sample passivated with a passivation layer deposited by 10 ALD cycles).

Sample type	Sample name	Number of samples (number of cells)	Number of ALD cycles for Al ₂ O ₃ xx
Post-Deposition	post-DTxx	8 (32, 32, 32, 32, 32, 32, 32, 32)	0, 0, 10, 10, 20, 30, 50, 70
7.5 nm pre-Deposition	7.5pre-DTxx	2 (32, 32)	20, 50
15 nm pre-Deposition	15pre-DTxx	7 (32, 12, 12, 32, 32, 32, 32)	0, 10, 20, 30, 30, 50, 70
Diffusion barrier	barrierxx	3 (32, 32, 32)	0, 10, 20
Baseline	baselinexx	1 (32)	0

Fig. 1): i) NaF post-DT, ii) NaF pre-DT, iii) Na supply inhibited by a alkaline diffusion barrier and neither NaF pre-DT nor post-DT and iv) Na supply exclusively from the soda-lime glass substrate (labeled as “baseline”). The passivation layer thickness was varied to optimize it for the different Na supply methods. Current-voltage (*JV*) measurements at a wide range of temperatures were used to further characterize the rear contact for some combinations of passivation layer thickness and Na supply method.

2. Sample processing and characterization

An overview over the whole matrix of samples produced and sample names can be found in Table 1 and the different sample types are further illustrated in Fig. 1. Generally, the samples were produced according to our group's baseline process [25]. The cell area was scribed mechanically to an area of 0.5 cm² and every sample consists of 32 individual cells, except for two samples with 12 cells. In contrast to the baseline process described in reference [25] the co-evaporated CIGS layers were grown with constant evaporation rates, resulting in a flat [Ga]/([Ga] + [In]) ratio throughout the film, i.e. without a built-in electron barrier at the rear of the absorber layer. The Al₂O₃ diffusion barriers, Al₂O₃ passivation layers and NaF post- or pre-DT were added to the baseline process in the following way: For samples with a diffusion barrier, 300 cycles Al₂O₃ were deposited directly on the cleaned

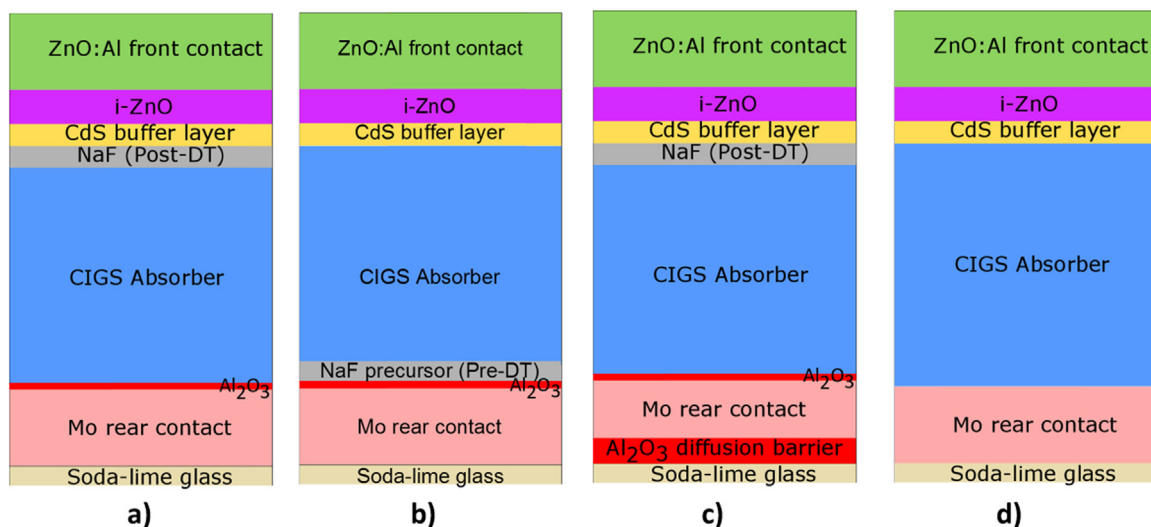


Fig. 1. Schematic representation of the different sample types: a) NaF post-deposition treated (post-DT) b) NaF pre-deposition treated (pre-DT), c) barrier, d) baseline. The passivation layer is very thin compared to all other layers and is indicated as a red line on top of the Mo rear contact.

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