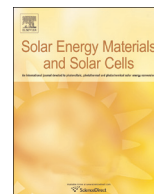




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Temperature dependent current–voltage and admittance spectroscopy on heat–light soaking effects of Cu(In,Ga)Se₂ solar cells with ALD-Zn(O,S) and CBD-ZnS(O,OH) buffer layers



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ABSTRACT

Heat-light soaking effects on Cu(In,Ga)Se₂ (CIGS) solar cells with atomic layer deposition (ALD)-Zn(O,S) and chemical bath deposition (CBD)-ZnS(O,OH) buffer layers were investigated using temperature-dependent current–voltage and admittance spectroscopy measurements. Both CBD-ZnS(O,OH)/CIGS and ALD-Zn(O,S)/CIGS solar cells showed a significant increase in the cell performance after the combined heat and light soaking (HLS) post-treatment. Temperature-dependent current–voltage measurements showed a reduced roll-over of current density–voltage curve after the HLS post-treatment, suggesting a reduced recombination at the absorber/buffer interface. Admittance spectroscopy measurement revealed a remarkable shift towards shallower energy positions for the defect N₁ after HLS post-treatment in both CIGS solar cells fabricated using CBD and ALD methods. By optimizing the HLS and the deposition conditions for each buffer layer, CBD-ZnS(O,OH)/CIGS and ALD-Zn(O,S)/CIGS solar cells yielded total efficiencies of 18.8 and 18.7%, respectively.

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1. Introduction

Cu(In,Ga)Se₂ (CIGS) solar cells with a zinc-compound buffer are well-known as one of the most promising candidate for high efficiency Cd-free CIGS solar cells [1]. Among the alternative possible buffers, a ZnS(O,OH) buffer layer deposited by chemical bath deposition (CBD) [1–11] and a Zn(O,S) buffer layer deposited by atomic layer deposition (ALD) [12–17] are typically used for high efficiency CIGS solar cells. An efficiency of 20.9% has recently been obtained on small solar cells, while 30 × 30 cm² sub-module reached 17.8% [6,8]. In general, high-efficiency CIGS solar cells with Zn-compound buffer layers are achieved after various types of post-treatments such as air-annealing, light-soaking (LS) and their combination [5,10,11,16,18,19]. We have reported the beneficial effects of a combined heat and light soaking (HLS) post-treatment on zinc-compound buffer/CIGS solar cells [11,19]. CIGS solar cells showed a drastic and irreversible increase in the cell performance after this HLS post-treatment [19,20]. X-ray photoelectron spectroscopy (XPS) measurement suggested that the increase in *J*_{sc} and FF of CBD-ZnS(O,OH)/CIGS device was caused by the re-

arrangement of the conduction band offset (CBO) due to the decreased S/(S+O) atomic ratio of CBD-ZnS(O,OH) layer after the HLS [19]. Recently, an efficiency of 18.3% for sputter-deposited Zn(O,S)/CIGS solar cells has been reported without any post-treatment such as annealing and light soaking [21]. In our previous experiments, high-efficiency zinc-compound buffer/CIGS solar cells were never obtained without an additional HLS and/or LS post-treatment, as no sputtering process was used for the formation of buffer and/or window layers [11,17,19,20,22,23]. Since the sputtering process itself provides the light illumination and the thermal radiation for the device during the film growth, it is suggested that an as-fabricated device with sputter-deposited window layer already receives a HLS-like treatment in the course of its fabrication. It has been reported that the HLS, even at temperatures of around 80 °C, beneficially affected the performances of CIGS device and/or ZnO:B films [24,25]. On the other hand, ALD-Zn(O,S)/CIGS devices showed stably high *J*_{sc} even before the HLS, because the HLS causes almost no compositional change in ALD-Zn(O,S) buffer layers resulting in a good alignment of CBO at buffer/CIGS interface regardless of the HLS. It was also noted that both devices with CBD-ZnS(O,OH) and ALD-Zn(O,S) buffer layers showed a significant increase in *V*_{oc} and FF [17]. However, the reason for the increased *V*_{oc} and FF of both devices has not been sufficiently clarified yet. Post-treatments such as annealing and/or

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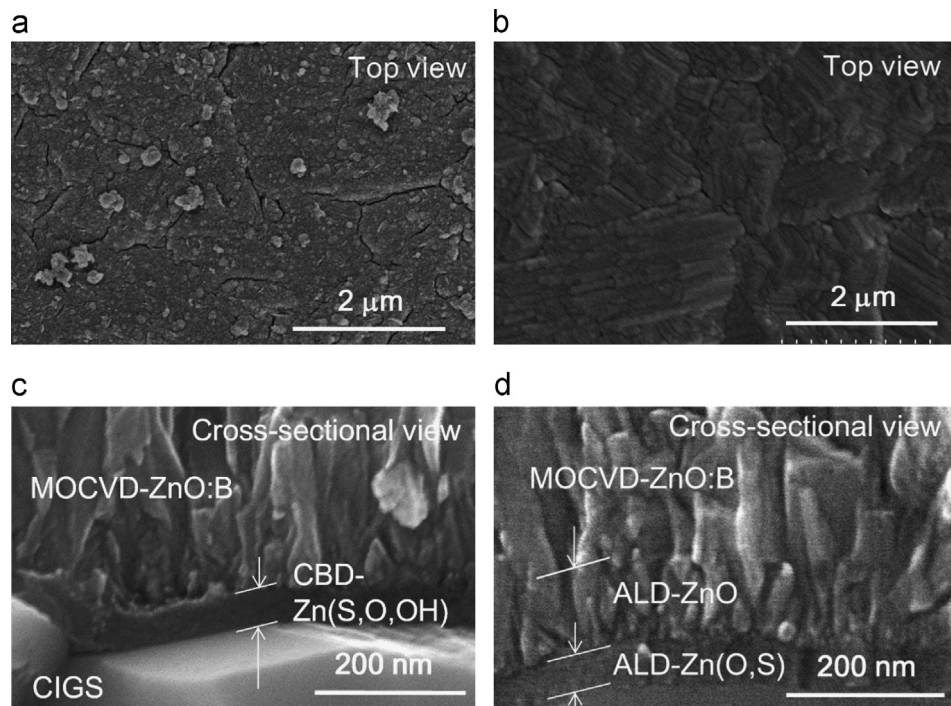


Fig. 1. Top view SEM photographs of 50 nm-thick (a) CBD-ZnS(O,OH) and (b) ALD-Zn(O,S) buffer layers on CIGS films, and cross-sectional SEM photographs of (c) MOCVD-ZnO:B/CBD-ZnS(O,OH)/CIGS and (d) MOCVD-ZnO:B/ALD-Zn(O,S)/CIGS stacked layers [10].

light-soaking are also investigated for various types of thin film solar cells including perovskite-type solar cells, and even organic-type solar cells [26–28].

In this work, the recombination paths of CIGS thin-film solar cells with CBD-ZnS(O,OH) and ALD-Zn(O,S) buffer layers are investigated through temperature-dependent current–voltage and capacitance–frequency measurements on devices before and after HLS post-treatment. Plasma damage-less metal organic chemical vapor deposition (MOCVD) which does not contain light-illumination during the fabrication process was used for deposition of the boron-doped ZnO (ZnO:B) window layer. The improved cell performance after the HLS is firstly discussed in connection with the changed J – V roll over and a remarkable shift of energy positions for the defect N_1 after the HLS. The cell performance is also discussed in connection with the compositional changes after HLS in both buffer layers. The recombination paths and their origins were investigated through temperature-dependent current–voltage and capacitance–frequency measurements on devices before and after HLS post-treatment.

2. Experimental

2.1. Fabrication of CBD-ZnS(O,OH)/CIGS solar cells

CBD-ZnS(O,OH)/CIGS solar cells were fabricated as follows: a 2.6 μm -thick CIGS absorber layer was deposited by three stage process at a maximum substrate temperature of 540 °C. The average Ga/(In+Ga) and Cu/(In +Ga) atomic ratios measured by inductively coupled plasma (ICP) spectroscopy for the CIGS film were 0.29 and 0.81, respectively. 50 nm-thick ZnS(O,OH) buffer layers were deposited onto the CIGS absorber layers by two consecutive chemical bath deposition (CBD) runs with 0.03 M-thioacetamide (CH_3CSNH_2), 0.16 M- ZnSO_4 , and 7.5 M-ammonia (NH_4OH) aqueous solution at RT–67 °C [23], as our baseline process for CBD-ZnS(O,OH) buffer layer [23]. The total duration time for the CBD process was 16 min. Undoped highly resistive ZnO is

commonly deposited before the growth of the transparent conducting window layer in order to make the pn-junction less sensitive to shunts and material fluctuations of the CIGS absorber, and to protect junction region against sputter damage by energetic ions [1,29,30]. Since no significant change in the cell efficiency was observed between CBD-ZnS(O,OH)/CIGS solar cells without and with 100 nm-thick MOCVD-ZnO layers in our preliminary experiments, the ZnO layer was not used for the CBD-ZnS(O,OH)/CIGS device in this work.

The films were air-annealed at 200 °C for 10 min immediately after the CBD process. 0.9 μm -thick MOCVD-ZnO:B films were then deposited on ZnS(O,OH)/CIGS layers at a substrate temperature of 185 °C for 15 min using diethylzinc (DEZ, $(\text{C}_2\text{H}_5)_2\text{Zn}$), triethylboron (TEB, $(\text{CH}_3\text{CH}_2)_3\text{B}$) and water vapor (H_2O) as zinc, boron, and oxygen sources [25]. The solar cells were completed by vacuum evaporation of a standard Al/Ni front electrode.

2.2. Fabrication of ALD-Zn(O,S)/CIGS solar cells

50 nm-thick Zn(O,S) buffer layers were deposited onto CIGS films at a substrate temperature of 125 °C by an ALD system. DEZ was used as a zinc precursor, while hydrogen sulfide (H_2S) and H_2O were used as group VI precursors, respectively. Nitrogen was introduced at a flow rate of 20 sccm as a carrier gas. The precursor gases were introduced into the nitrogen stream by means of fast switching valves. The amount of precursor introduced into the chamber during thin film deposition was controlled by the opening time of the switching valves. The $\text{H}_2\text{S}/(\text{H}_2\text{O} + \text{H}_2\text{S})$ pulse ratio was maintained at 0.27, which corresponds to S/(S+O) atomic ratio of 0.33. Subsequently, a high resistive 100 nm-thick ZnO layer was deposited by an ALD system using DEZ and H_2O at the same substrate temperature. The ZnO:B window layer and the Al/Ni front electrode were deposited by the above-mentioned process. The average Ga/(In+Ga) and Cu/(In+Ga) atomic ratios measured by ICP spectroscopy for the CIGS film were 0.37 and 0.81, respectively.

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