

Enhanced performance of ultra-thin Cu(In,Ga)Se₂ solar cells deposited at low process temperature



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ABSTRACT

To investigate the process temperature on the growth of ultra-thin (≤ 500 nm) Cu(In,Ga)Se₂ (CIGSe) absorbers and the corresponding performance of solar cells, the process temperature was set to 610 °C and 440 °C, respectively. It was found that the low process temperature (440 °C) could reduce the inter-diffusion of Ga–In and thus result in a higher back [Ga]/([Ga]+[In]) ([Ga]/[III]) grading than at the temperature of 610 °C. The higher back [Ga]/[III] grading at 440 °C was evidenced to both electrically and optically contribute to the efficiency enhancement of the solar cells in contrast to the lower back [Ga]/[III] grading at 610 °C. It was also implied that the high back [Ga]/[III] grading was beneficial to the collection of carriers generated from the back-reflected light.

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1. Introduction

Reducing the absorber thickness is critical to realizing the reduction of material consumption (e.g. Indium and Gallium) and resulting cost of Cu(In,Ga)Se₂ (CIGSe) solar cells [1,2]. However, high efficiencies are only achieved with absorbers thicker than 1 μm [3,4]. Further reducing the absorber thickness to 0.5 μm has been experimentally shown to considerably reduce the efficiency. The dominating reason behind the poor performance of ultra-thin solar cells (with absorber thickness below 0.5 μm) is the substantial drop of short current density (J_{sc}) [3–5], which is assumed to mainly originate from the incomplete absorption of the incident solar spectrum and the strong carrier recombination at the back contact (back recombination).

Concerning the back recombination, the risk is much higher for ultra-thin solar cells due to the reduced distance of electrons to the back contact [6]. It is extensively reported that an increasing [Ga]/([Ga]+[In]) ratio ([Ga]/[III]) towards the back contact (the back Ga grading) can serve as an electron back reflector by increasing the conduction band (CB) and thus reduce the carrier recombination [6–11]. Furthermore, Vermang [12] introduced a back passivation structure by depositing an Al₂O₃ film on the nano-sphere coated Mo substrate, which decreased the back recombination as well. In spite of this, there has been no experimental comparison of the

efficacy of back recombination passivation between a higher back Ga grading and a back passivation structure. In the 3-stage co-evaporation processes [13], lowering process temperature (substrate temperature) was proved to be able to reduce the inter-diffusion of Ga–In [14,15] and therefore provides the possibility to create a higher back Ga grading by engineering the deposition sequence of Ga–Se precursor and In–Se. At the same time, the low process temperature can enable the preparation of CIGSe solar cells on flexible polymer substrates [16,17], thus offering a potential to further reduce the manufacturing cost and widen the applications of the solar cells.

However, high-quality CIGSe absorbers are normally deposited above 500 °C. How the low process temperature affects the ultra-thin absorbers and the resulting performances of CIGSe solar cells is still unknown in detail. To identify this, we will investigate solar cells deposited at low process temperature and compare the results to those at high process temperature.

2. Experimental

CIGSe absorbers were fabricated on Mo substrates by the 3-stage co-evaporation process. To create a high intentional back Ga grading, the Ga–Se precursor was deposited before In–Se at 410 °C during the 1st stage of the 3 stages of the evaporation. Cu–Se and In–Ga–Se precursors were evaporated at the substrate temperature T_S during the 2nd and 3rd stage, respectively. T_S is higher than the temperature of 410 °C in the 1st stage and is defined as the process temperature. After reaching [Cu]/([Ga]+[In]) ratio ([Cu]/[III])

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of 1.06 at the end of the 2nd stage, the process transitioned to the 3rd stage and terminated at the [Cu]/[III] ratio of around 0.85–0.9. Laser light scattering (LLS) was used for the in-situ control of the process [18]. The composition ratios mentioned above are the estimated values from LLS signal. To compare the influence of T_S on the absorbers, T_S was set to 610 °C and 440 °C. For evaluating the absorption coefficient α of the absorbers, a reference sample was deposited on glass substrate at each temperature.

The substrate used was soda lime glass (SLG) with a thickness of 2 mm. The 800 nm thick Mo layer was sputtered onto the SLG substrate with a sheet resistance $0.8 \Omega/\square$ for back contact. For the device completion, a 100 nm thick CdS layer was grown by a standard chemical bath deposition (CBD) process after the CIGSe deposition. The CBD process was performed in a solution with 1.1 M ammonia, 0.14 M thiourea and 0.002 M cadmium acetate. A sputtered 130 nm i-ZnO and a 240 nm Al doped ZnO (AZO) layer followed the CdS. The Ni/Al front contact grid was evaporated through a shadow mask with a total thickness of 2500 nm. Finally, 0.5 cm² solar cells were mechanically scribed including the area of the front contact grid.

For characterization of the absorber morphology, scanning electron microscopy (SEM) was used. The element composition of the absorbers was measured by X-ray fluorescence analysis (XRF). The overall [Ga]/[III] and [Cu]/[III] ratios are 0.4 ± 0.01 and 0.86 ± 0.01 , respectively. The thickness of the absorbers was evaluated by both XRF and optical simulation of the CIGSe samples on glass substrates [19]. The thickness in both cases is 460 ± 5 nm. The phase structure was characterized by X-ray diffraction (XRD). Glow discharge optical emission spectrometry (GDOES) was used to evaluate the [Ga]/[III] depth profile across the CIGSe absorber. The GDOES measurement was performed by a spectrometer (GDA 650 from Spectrumba) with 2.5 mm anode and Ar discharge gas [20,21]. The I - V curve was measured under standard test conditions (AM 1.5; 100 mW/cm²; 25 °C) by a home-made system with a sun-simulator (WXS-140S-SUPER from WACOM), which consists of both a xenon and a halogen lamp. The AM 1.5 condition is calibrated by a certified crystalline Si solar cell from the Fraunhofer Institute for Solar Energy Systems (ISE). The external quantum efficiency (EQE) was measured with a two-source illumination system of a xenon and a halogen lamp, using calibrated Si and Ge diodes as references.

3. Results and discussion

3.1. Morphology and [Ga]/[III] depth profile

Fig. 1 displays the morphology of CIGSe layers in cross section at different process temperatures T_S . During the 3-stage process

deposition, the maximum temperature T_S dominates the growth of the absorbers, which can be also observed in Fig. 1. At high temperature $T_S=610$ °C, the grains are closely packed and extend through the entire thickness. At $T_S=440$ °C, there still exists large grains but with fine grains near the Mo back contact, which was correlated to high [Ga]/[III] CIGSe phases by Kaufmann [15]. Fig. 2(a) shows the (112) XRD diffraction peaks of the absorbers at the two process temperatures. In contrast to the single and sharp peak for the absorber deposited at $T_S=610$ °C, the (112) diffraction peak evolves into a broad double-peak when the process temperature T_S drops to 440 °C. Besides, the two sub-peaks locate at each side of the single peak corresponding to $T_S=610$ °C. Because of a larger atomic radius of In than Ga, as the [Ga]/[III] ratio increases, the peak position evolves from CIGSe with the lowest 2 theta value to CGSe with the highest. Since the two absorbers have the same overall [Ga]/[III] ratio,

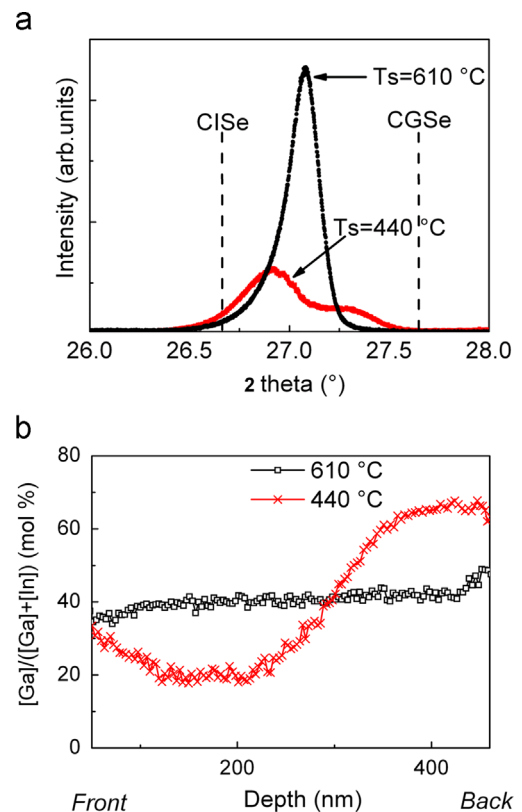


Fig. 2. (a) (112) XRD diffraction peaks and (b) the corresponding [Ga]/[III] depth profiles evaluated by GDOES at both temperatures.

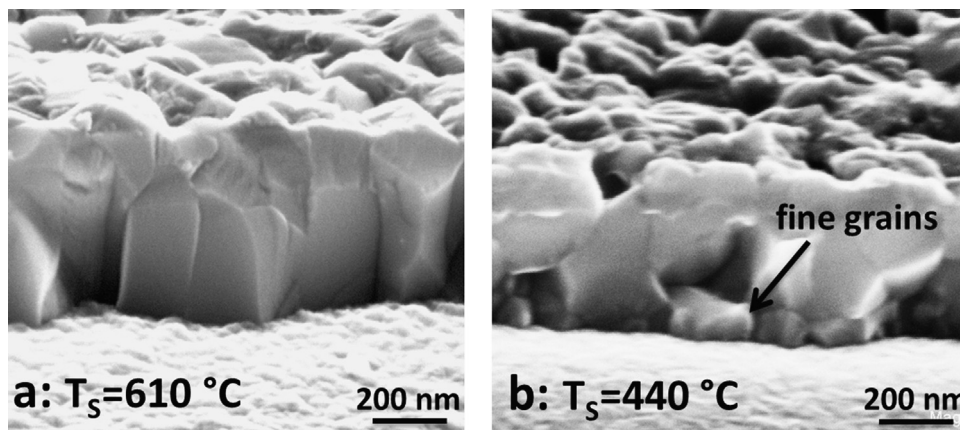


Fig. 1. Cross sections of the CIGSe layers on Mo substrate at process temperatures of (a) $T_S=610$ °C and (b) $T_S=440$ °C.

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