



Effect of an additional Cu-deficient layer deposition on alkali treated Cu(In, Ga)Se₂ solar cells deposited at low temperature

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ABSTRACT

In this work, we found that the deposition of a Cu-deficient layer after alkali post-deposition treatment (PDT) of Cu(In,Ga)Se₂ (CIGS) at low temperature (450 °C) effectively improves the performance of CIGS solar cells. Compared to the CGIS with only the alkali PDT, the deposition of the Cu-deficient layer leads to the formation of larger holes on the surface and results in higher efficiency. The conversion efficiency improved from 11% to 16% after alkali PDT and Cu-deficient layer deposition because of a passivation effect. The formation of a passivation layer after both alkali treatment and Cu-deficient layer deposition was observed by TEM-EDX, showing that the deposition of the Cu-deficient layer could contribute to a low Cu surface composition. Finally, the effect on the CIGS and CdS interface was analyzed using electron-beam-induced current measurement (EBIC).

1. Introduction

Thin-film solar cells with chalcopyrite-based Cu(In,Ga)Se₂ (CIGS) absorbers show high efficiency, above 20%, when using both rigid glass substrates and flexible films [1–4]. The deposition of CIGS on flexible films allows us to reduce the photovoltaic module weight, which can lower the infrastructure cost and give a wider range of applications. The highest most recent efficiency reported for a flexible CIGS solar cell is 20.4% [5]. This was achieved with films deposited on top of a polyimide film at a substrate temperature of 450 °C. There are several approaches in obtaining higher efficiencies in flexible solar cells, for example, band profile arrangement and alkali post-deposition treatment (PDT). The former has been conducted to minimize the open-circuit voltage (V_{OC}) loss, which occurs because of the deep notch in the conduction band that, in turn, rises because of the slow Ga diffusion speed at low temperatures [6,7]. Alkali incorporation has been widely investigated for flexible CIGS solar cells because of the absence of an alkali source in flexible substrates. For rigid soda lime glass (SLG) substrates, it is known that small quantities of alkali metals, which diffuse from the substrate, are crucial for CIGS deposition [8–10]. The presence of an alkali metal such as sodium or potassium contributes to the growth of larger crystals, increasing the carrier concentration [11,12]. However, alkali PDT for CIGS provides beneficial effects different from those reported for alkali diffusion from the substrate [13–15]. Accordingly, PDT has been used for CIGS solar cells on flexible

films and also on rigid glass [16]. Recently, a conversion efficiency of 22.6% has been achieved by adding a heavy alkali metal on a CIGS deposited at high temperature [17]. In addition to alkali PDT, the deposition of a Cu-deficient layer, such as a Cu(In,Ga)₃Se₅ phase, after the CIGS deposition has been reported to reduce the recombination in the CIGS/CdS interface because of a hole repelling effect [18,19]. Cu(In,Ga)₃Se₅ is an ordered vacancy compound (OVC) with a valence band offset of 0.3 eV and a continuous conduction band with CIGS [20–22]. This last method could also increase the efficiency of CIGS solar cells with a single-graded band profile deposited at low temperature [23]. The alkali PDT and Cu-deficient layer deposition methods have been independently investigated; both show promising results by improving the efficiency of CIGS solar cells deposited at high and low temperatures. In this study, we investigate CIGS solar cells deposited at low temperature with sequential alkali PDT and Cu-deficient layer deposition [24]. CIGS with both single- and double-graded band profiles were used to evaluate the influence of Ga composition on the treatment. Further analysis of the effects of alkali PDT and Cu-deficient layer deposition on the surface texture and composition was also conducted to elucidate the phenomena occurring on the CIGS surface. The results indicate that the deposition of the Cu-deficient layer likely enhances the effect of alkali PDT; thus, higher efficiency can be achieved.

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2. Experimental details

Solar cells with an Al/ZnO(B)/i-ZnO/CdS/CIGS/Mo structure were deposited on the top of an SLG. A bilayer Mo back contact was deposited using DC sputtering by changing the Ar pressure on the first and second layers to obtain high adhesiveness and electrical conduction, respectively [25]. CIGS with single- and double-graded band profiles were deposited using a three-stage method at 450 °C with a molecular beam epitaxy apparatus [26]. The single-graded band profile was obtained by supplying only In during the third stage instead of In and Ga. The [Ga]/([Ga] + [In]) (GGI) amounts of the CIGS surface region were 0.02 and 0.24 for the single- and double-graded CIGS, respectively. A low temperature (450 °C) was used considering the durability of polyimide and its application on flexible solar cells. Two types of alkali PDT were conducted. First is the deposition of only NaF on top of the CIGS (NaF-PDT); the second is the deposition of KF after NaF (KF-PDT). The thicknesses of the NaF and KF layers were approximately 10 and 15 nm, respectively. Owing to experimental difficulties, it was not possible to directly measure the NaF and KF thicknesses; thus, a linear approximation of the deposition time and thickness of NaF and KF deposited at room temperature obtained from scanning electron microscopy (SEM) measurements was used. The alkali PDTs were conducted at 400 °C under Se atmosphere. The Se beam flux was approximately 10^{-3} Pa, and the ratio of alkali metal and Se pressure was 1:100 as measured by the beam flux monitor. After the deposition of each alkali metal, the substrate temperature was maintained for 20 min so that it could diffuse into the CIGS. The Cu-deficient layer was deposited at the same temperature (400 °C) after alkali PDT by adjusting the Cu flux to obtain a Cu(In,Ga)₃Se₅ phase layer with a thickness of approximately 10 nm. After the deposition of the Cu-deficient layer, the temperature was immediately decreased. A 50-nm-thick CdS buffer layer and a 1- μ m-thick ZnO transparent contact were deposited by chemical bath deposition (CBD) and metal-organic chemical vapor deposition (MOCVD), respectively. Finally, an Al grid contact was thermally evaporated.

The effect of the alkali treatment and deposition of Cu-deficient layer on the solar cell performance was evaluated by measuring the current density–voltage (J - V) characteristics of the cells with a total area of approximately 0.18 cm² (with 10% of shadowed area) under AM1.5 G (100 mW/cm²) illumination. SEM was used to observe the CIGS surface after each treatment. The average composition of the CIGS from the surface to approximately 500 nm deep was measured using an JED-2300 energy-dispersive X-ray spectrometer (EDX) that was attached to the SEM equipment (SEM-EDX). Beam with a 15 kV accelerated voltage and 6 nA emission current were used in the measurement. The cross-section of the CIGS was also observed by JEM-ARM200F transmission electron microscopy (TEM), and material composition was simultaneously evaluated by EDX. Unlike the SEM-EDX experiments, the high resolution of the TEM-EDX allowed us to measure the local area near the CIGS surface. For preparation of the TEM specimen, we adopted a FIB (Focused Ion Beam) method using a Ga ion beam, and the specimen thickness is approximately 40 nm with thickness fluctuation was 10%. The EDX spectrum result were calibrate using single-crystalline CIGS that the composition already known with error estimation of 1 at%. This also allowed us to de-convoluted Indium and Potassium signals in the EDX spectrum. Electron-beam-induced current (EBIC) was also performed to evaluate the correlation between the treatment and carrier collection in the CIGS. The samples for the EBIC measurements were prepared by polishing the CIGS cross-section using an Ar ion beam with a 4 kV accelerating voltage. The light material, i.e., Ar, and low accelerating voltage were used to reduce the damage on the CIGS and prevent the formation of an amorphous surface layer.

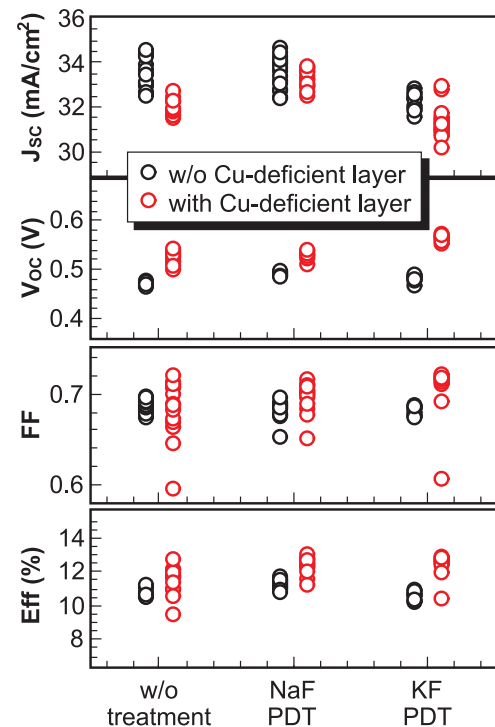


Fig. 1. Characteristics of CIGS solar cells with single-graded band profiles. From left: as-deposited sample, CIGS with NaF-PDT, and CIGS with KF-PDT. Open red circles show the Cu-deficient layer deposition before and after alkali PDT. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3. Results and discussion

Fig. 1 shows the effects of NaF-PDT and KF-PDT without (open black circles) or with (open red circles) the additional Cu-deficient layer on the characteristics of single-graded CIGS solar cells. The conversion efficiency slightly increased with the NaF-PDT because the V_{OC} improved. The diffusion of Na into the CIGS is known to increase the carrier concentration and improve the V_{OC} [27,28]. Moreover, the deposition of the Cu-deficient layer shows an improvement on both the V_{OC} and fill factor (FF) with or without the NaF-PDT. This indicates that, similarly to previous experiments with no alkali PDT [23], the deposition of a Cu-deficient layer can improve the performance of single-graded CIGS solar cells even when deposited at low temperature. Contrastingly, when using KF after NaF-PDT, there was no improvement in the single-graded band profile CIGS, even when the additional Cu-deficient layer was deposited. These results suggest that KF-PDT is not effective in improving the performance of CIGS solar cells with single-graded band profile. It is likely that the low GGI in the surface causes KF-PDT ineffectiveness. The SEM-EDX measurements show that the [Cu]/([In] + [Ga]) (CGI) amount remained at approximately 0.86 for all of the treatments, indicating that the average composition of the CIGS did not change. The effects of the Cu-deficient layer deposition after KF-PDT on CIGS with double-graded band profile are shown in Fig. 2. Contrary to KF-PDT on single-graded CIGS, the V_{OC} and FF significantly improved in this case. This contrasting alkali PDT effect could be caused by a different GGI ratio on the surface, which was 0.02 for single-graded CIGS and 0.24 for double-graded ones. Handick et al. suggested that KF-PDT forms KInSe₂ or In₂Se₃ on the surface, which increases the band gap and acts as a passivation layer [29]. Therefore, increasing the In amount in the surface would ease the formation of such passivation layer, contradicting the obtained results. There are two possible explanations for this behavior. First, the absence of Ga in the surface could impede the formation of the passivation layer; second, the

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