Transfer of CuInS₂ layer by lift-off process and its solar cell applications

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According to the PV 2030+ of new energy and industrial technology development organization (NEDO), Japan, a conversion efficiency of 25% is required for CuInSe₂-based chalcopyrite semiconductor thin film solar cells in 2017 [1]. Since a conversion efficiency of over 30% was achieved in a tandem structure solar cell using III-V based semiconductors [2], a tandem structure solar cell is one of promising candidates even in CuInSe₂-based chalcopyrite semiconductor thin film solar cells [3-6]. Unfortunately, conversion efficiencies of the tandem solar cells are still lower than those of the single junction solar cells [3-6]. In particular, several groups indicated that the fabrication of the high efficiency top solar cells with the wide band gap absorber layers such as CuGaSe₂ and Cu(In,Ga)S₂ on the transparent back electrodes was difficult due to the low temperature growth [3,6]. We think that mechanical stack using a lift-off process is attractive as the preparation method of the top solar cell, because the high quality absorber layer, which is grown on a conventional Mo/soda-lime glass (SLG) substrate at the appropriate growth temperature, can be prepared. Establishment of the lift-off process for the CuInS₂-based thin films which suit the absorber layer of the top solar cell [7] is therefore required.

In this study, we investigated the peeling of a $CuInS_2$ layer grown on a Mo/SLG substrate. In addition, we fabricated the superstrate-type $CuInS_2$ solar cell by transcribing a substrate-type $CuInS_2$ solar cell. In this paper, we report on the peeling of the $CuInS_2$ layer and characteristics of the superstrate-type $CuInS_2$ solar cell.

A schematic illustration of a transfer sample is shown in Fig. 1(a). The transfer sample of a CuInS₂ layer was fabricated as follows. An 800-nm-thick Mo layer was deposited on an SLG substrate by the radio frequency (RF) magnetron sputtering method without intentional substrate heating. A 3.5- μ m-thick CuInS₂ layer was grown on the Mo/SLG substrate at 550°C by the vacuum coevaporation method [8]. An alternative SLG substrate was bonded onto the

CuInS₂ surface with cyanoacrylate glue. The alternative SLG substrate was vertically separated from the primary SLG substrate by tensile strain [9].

The schematic illustrations of the superstrate-type and substrate-type $CuInS_2$ solar cells are shown in Figs. 1(b) and 1(c), respectively. The fabrication procedure of the superstrate-type $CuInS_2$ solar cell is as follows [10]. After the



Fig. 1. Schematic illustrations of fabricated samples. (a) Lift-off sample, (b) substrate-type $CuInS_2$ solar cell, and (c) superstrate-type $CuInS_2$ solar cell.

2-µm-thick CuInS₂/Mo/SLG structure was fabricated through the above procedure, the samples were dipped into a KCN solution to remove Cu sulfide compounds. A 100-nm-thick CdS layer was deposited by the chemical bath deposition method. 100-nm-thick i-ZnO/200-nm-thick In₂O₃:Sn layers were deposited by the RF magnetron sputtering method. An Al/NiCr grid was formed by heat resistance deposition method using a shadow mask. An alternative SLG substrate was bonded onto this substrate-type solar cell surface with ultraviolet glue with light transparency. The lift-off process was performed. After a graphite paste was applied as a back electrode, the sample was annealed at 120°C for 1 h in the atmosphere to dry the graphite back electrode.

Grazing incident x-ray diffraction (GIXRD) and x-ray absorption fine structure (XAFS) spectroscopy measurements were performed to identify the part separated by the lift-off process. XAFS spectroscopy measurements were performed at BL-10 of the SR center at Ritsumeikan University, Japan [11]. S K-edge x-ray absorption near edge structure (XANES) spectra were measured at room temperature under vacuum conditions. In this experiment, a double crystal Ge(111) monochromator was utilized. We used the total electron yield (TEY) method for the surface-sensitive analysis. Current density-voltage (*J-V*) measurements were performed under standard air mass 1.5 global conditions (100 mW/cm²) at 25°C to investigate performance of the fabricated solar cells. For comparison, the substrate-type CuInS₂ solar cells were also measured before the lift-off process.

Figure 2 shows GIXRD patterns of the $CuInS_2$ rear and Mo sides. Peaks of $CuInS_2$ only are detected for the $CuInS_2$ rear side. On the other hand, peaks of Mo only are detected for the Mo side. We therefore have to interpret that this peeling occurred at the Mo/CuInS₂ interface from this CIXRD result.

Figure 3 shows S K-edge XANES spectra of the CuInS₂ rear and Mo sides. The result of a high purity single crystalline MoS_2 reagent is also shown for comparison. The XANES spectrum of the CuInS₂ rear side is remarkably different from that of the Mo side. On the other hand, the XANES spectrum of the Mo side is very similar to that of the MoS_2 reagent. We found that MoS_2 formed at the surface of the Mo side. Moreover, we found that the XANES spectrum of the Mo side hardly included the component of CuInS₂. On the other hand, if an ultrathin MoS_2 layer exists on the surface of the CuInS₂ rear side, the MoS_2 component can not be recognized because of relatively high intensity of the CuInS₂ spectrum compared to the MoS_2 component. This result therefore indicates that peeling of the CuInS₂ layer at the CuInS₂/MoS₂ interface or through breaking of the interfacial MoS_2 layer at the CuInS₂/MoS the CuInS₂ layer does not occur in the transfer



Fig. 2. GIXRD patterns of $CuInS_2$ rear and Mo sides.



Fig. 3. S K-edge XANES spectra of Mo and $CuInS_2$ rear sides. MoS_2 reagent result is also shown for comparison.

of the CuInS₂ layer, it is easy to control the eventual thickness of the transferred CuInS₂ layer.

Figure 4 shows the *J*-V characteristic of the superstrate-type $CuInS_2$ solar cell fabricated by transcribing the substrate-type $CuInS_2$ solar cell. The *J*-V characteristic of the substrate-type $CuInS_2$ solar cell before the lift-off process is also shown in Fig. 4 for comparison. A conversion efficiency of the substrate-type solar cell is very close to that of the superstrate-type solar cell. We therefore established the lift-off process without remarkable degradation of a conversion efficiency for a $CuInS_2$ thin film solar cell.

We investigated how the CuInS₂ layer was peeled by the lift-off process in the transfer of the CuInS₂ layer grown on the Mo/SLG substrate. Our result indicates that the peeling CuInS₂ layer occurred of the at the CuInS₂/MoS₂ interface or through breaking of the MoS₂ layer. Moreover, we investigated the solar cell performance of the superstrate-type CuInS₂ solar cell fabricated by transcribing the substrate-type CuInS₂ solar cell. We found that the conversion efficiency of the substrate-type solar cell is very close to that of the superstrate-type solar cell. We therefore established the lift-off process without degradation of a conversion remarkable efficiency for the CuInS₂ thin film solar cell.



Fig. 4. *J-V* characteristics of superstrate-type and substrate-type CuInS₂ solar cells.

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