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Development of Flexible $Cu(In,Ga)Se_2$ Thin Film Solar Cell by Lift-Off Process

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1. Introduction

Clean energy resources as an alternative to fossil fuels has been required. Photovoltaics is the most promising among renewable energy technologies. On the other hand, the cost of the electrical energy generated by the solar cells was higher than that generated by fossil fuels. The cost reduction of the solar cell is therefore required.

Since high-conversion efficiencies have been demonstrated for solar cells using GaAs substrates in 1977 (Kamath et al., 1977; Woodall et al., 1977), a critical problem is how to reduce power generation cost. The characters required to solar cells strongly depend on its applications. In particular, thin film solar cells are promising for terrestrial applications, because thin film solar cells are more advantageous than bulk type solar cells in terms of consumption of raw materials. Konagai et al. fabricated the thin film solar cells on a single crystalline GaAs substrate by the liquid phase epitaxy mehtod, and focused on the reuse of GaAs substrates by detaching these thin film solar cells from the GaAs substrates (Konagai et al., 1978). Konagai et al. named this separation technique the Peeled Film Technology (PFT). This is the invention of the lift-off method in solar cell development. A specific explanation of the PFT is as follows. An $Al_{1-x}Ga_xAs$ layer was introduced between the thin film solar cell and the GaAs substrate as a release layer. The thin film solar cell was separated from the GaAs substrate by etching the $Al_{1-x}Ga_xAs$ layer by the HF solution, because $Al_{1-x}Ga_xAs$ was readily dissolved by the HF solution compared to GaAs. Since a chemical technique was mainly used for the peeling, this method is defined as a chemical lift-off process. Recently, this has been researched as the epitaxial lift-off (ELO) method (Geelen et al., 1997; Schemer et al., 2000, 2005a; Voncken et al., 2002; Yablonovitch et al., 1987).

On the other hand, the cleavage of lateral epitaxial films for transfer (CLEFT) process, where the thin film was mechanically peeled, was developed as a transfer method of a single crystalline GaAs thin film (McClelland et al., 1980). A specific explanation of the CLEFT process is as follows. A photoresist was applied to a surface of a GaAs substrate. The photoresist was patterned with equally-spaced stripe openings by standard photolithographic techniques. Next, a GaAs layer was grown on this patterned substrate surface. In this case, a GaAs layer was grown on only the openings of the photoresist. The lateral growth of a GaAs layer occurs during the GaAs deposition. A single crystalline GaAs layer is therefore formed on the photoresist. Alternative substrate was bonded onto this

surface with epoxy glue. The single crystalline thin film was transferred to the alternative substrate by applying tensile strain. The CLEFT process is theorefore defined as a mechanical lift-off process.

Unfortunately, the conversion efficiency of the GaAs thin film solar cell using the lift-off process was lower than that of the GaAs bulk solar cell (Schermer et al., 2006). Recently, comparable conversion efficiencies have been demonstrated (Bauhuis et al., 2009).

On the other hand, the energy:weight ratio of the photovoltaic module is a very important index for space applications. Integration of high-efficiency III-V solar cells with light weight substrates is required. Schermer et al. developed high-efficiency III-V solar cells with light-weight by the ELO process using the GaAs substrate (Schermer et al., 2005b).

In addition, the lift-off process was applied to reuse Si substrates (Bergmann et al., 2002; Brendel, 2001). Moreover, the lift-off process was applied to fabricate flexible solar cells in the developments of II-VI and I-III-VI_2 semiconductor thin film solar cells (Marrón et al., 2005; Minemoto et al., 2010; Romeo et al., 2006; Tiwari et al., 1999).

Here, we focus on advantages of the lift-off process in fabrication of flexible Cu(In,Ga)Se_2 (CIGS) thin film solar cells. For example, for the fabrication process where CIGS layers were directly grown on flexible substrates, Ti foils (Hartmann et al., 2000; Herz et al., 2003; Ishizuka et al., 2009a; Kapur et al., 2002; Kessler et al., 2005; Yagioka & Nakada, 2009), Cu steel sheets (Herz et al., 2003), Mo foils (Kapur et al., 2002, 2003), stainless steel sheets (Britt et al., 2008; Gedhill et al., 2011; Hashimoto et al., 2003 ; Kessler et al., 2005; Khelifi et al., 2010; Pinarbasi et al., 2010; Satoh et al., 2000, 2003; Shi et al., 2009; Wuerz et al., 2009), Al foils (Brémaud et al., 2007), Fe/Ni alloy foils (Hartmann et al., 2000), ZrO_2 sheets (Ishizuka et al., 2008a, 2008b, 2009b, 2010), and polyimide (PI) films (Brémaud et al., 2005; Caballero et al., 2009; Hartmann et al., 2000; Ishizuka et al., 2008c; Kapur et al., 2003; Kessler et al., 2005; Rudmann et al., 2005; Zachmann et al., 2009;), are used as flexible substrates. Since these materials do not include Na, other processes to introduce Na are required (Caballero et al., 2009; Ishizuka et al., 2008a; Keyes et al., 1997). Since the thermal tolerance temperature of a PI film is ~450˚C, the low temperature growth of a CIGS layer is required. The first of the advantages of the lift-off process is to enable to use a high quality CIGS layer grown on a conventional Mo/soda-lime glass (SLG) substrate in the flexible solar cell fabrication. Consequently, the low temperature growth technology for high quality CIGS layer formation and novel processes for a Na source are not required. The second is to enable to use low thermal tolerance films as the flexible substrate of a CIGS solar cell, because in the CIGS solar cell fabrication process, the highest temperature process is the growth of a CIGS layer and the process temperature after the growth of a CIGS layer is less than 100˚C.

2. Experimental

2.1 Flexible Cu(In,Ga)Se_2 solar cell fabrication procedure

A schematic illustration of the fabrication procedure of our flexible CIGS solar cell using the lift-off process is shown in Fig. 1 (Minemoto et al., 2010). A 0.8-μm-thick Mo layer was deposited on an SLG substrate without intentional substrate heating by the radio frequency (RF) magnetron sputtering method. A 2.5-μm-thick CIGS layer was deposited on the Mo/SLG substrate by the three-stage deposition process at the highest substrate temperature of approximately 550˚C (Contreras et al., 1994a; Negami et al., 2002). From energy dispersive x-ray spectrometry measurements, the Cu, In, Ga, and Se composition ratios of this CIGS layer were approxymately 23, 18, 8, and 51%, respectively. The

[Cu]/[Ga+In] and [Ga]/[Ga+In] ratios of the CIGS layer were therefore calclated to be ~0.88 and ~0.31, respectively. After CIGS surface cleaning by a KCN solution, a 0.2-μm-thick Au layer was deposited on the CIGS surface by a resistive evaporation method as a back electrode. The samples were annealed for 30 min at 250˚C in N_2 ambient. Flexible films were bonded onto support SLG substrates with a silicone adhesion bond for preparation of the alternative substrates. These alternative substrates were also bonded onto the Au/CIGS/Mo/SLG structure with conductive epoxy glue. To dry the conductive epoxy glue, the samples were annealed on a hot plate at 100˚C for 10 min in the atmosphere. Then, the alternative-sub./epoxy/Au/CIGS stacked structures were detached from the primary Mo/SLG substrates by applying tensile strain. In this detachment, the CIGS layer was transferred to the alternative substrate side (Marrón et al., 2005). The lift-off flexible CIGS solar cells were fabricated using this peeled CIGS layer. After cleaning of the CIGS rear surface by a KCN solution, a 0.1-μm-thick CdS layer was deposited on the CIGS rear surface by the chemical bath deposition method. 0.1-μm-thick i-ZnO and 0.1-μm-thick In_2O_3:Sn layers were deposited by the RF magnetron sputtering method. Al/NiCr grids were formed. Finally, the flexible CIGS solar cells using the lift-off process were completed by detaching the flexible films from the support SLG substrates. For comparison, we also prepared a standard solar cell where the lift-off process was not carried out (the Al/NiCr/In_2O_3:Sn/ZnO/CdS/CIGS/Mo/SLG structure). The properties of the films used in this study are summarized in Table 1. Figure 2 shows a photograph of the flexible solar cells using the PI film.

Material	PI	PTFE	Polyester
Thermal tolerance temperature (˚C)	450	260	120
Thickness (μm)	55	120	25

Table 1. Properties of PI, polytetrafluoroethylene (PTFE) and polyester films used in this study are summarized.

2.2 Characterization methods

Current density-voltage (*J-V*) measurements were performed under standard air mass 1.5 global conditions (100 mW/cm²) at 25˚C. External quantum efficiency (EQE) measurements of the AC mode were performed at 25˚C under white light bias (~0.3 sun) conditions. The laser-beam-induced current (LBIC) method using the laser diode (λ: 783 nm, laser power: 0.3 mW) was performed to investigate a spatial distribution of an EQE (Minemoto et al., 2005). In LBIC measurements, a nominal spot size is less than 50 μm and a scan step is 53 μm. The surfaces of the fabricated flexible solar cells were observed with an optical microscope. The *J-V*, EQE, and LBIC measurements were performed after light soaking.

3. Results and discussion

3.1 Characterization of flexible Cu(In,Ga)Se_2 solar cells fabricated using lift-off process

The *J-V* characteristics of the fabricated flexible solar cells are shown in Fig. 3. For comparison, the *J-V* characteristic of the standard solar cell is also shown. Solar cell parameters such as the short-circuit current density (J_{sc}), the open-circuit voltage (V_{oc}), the

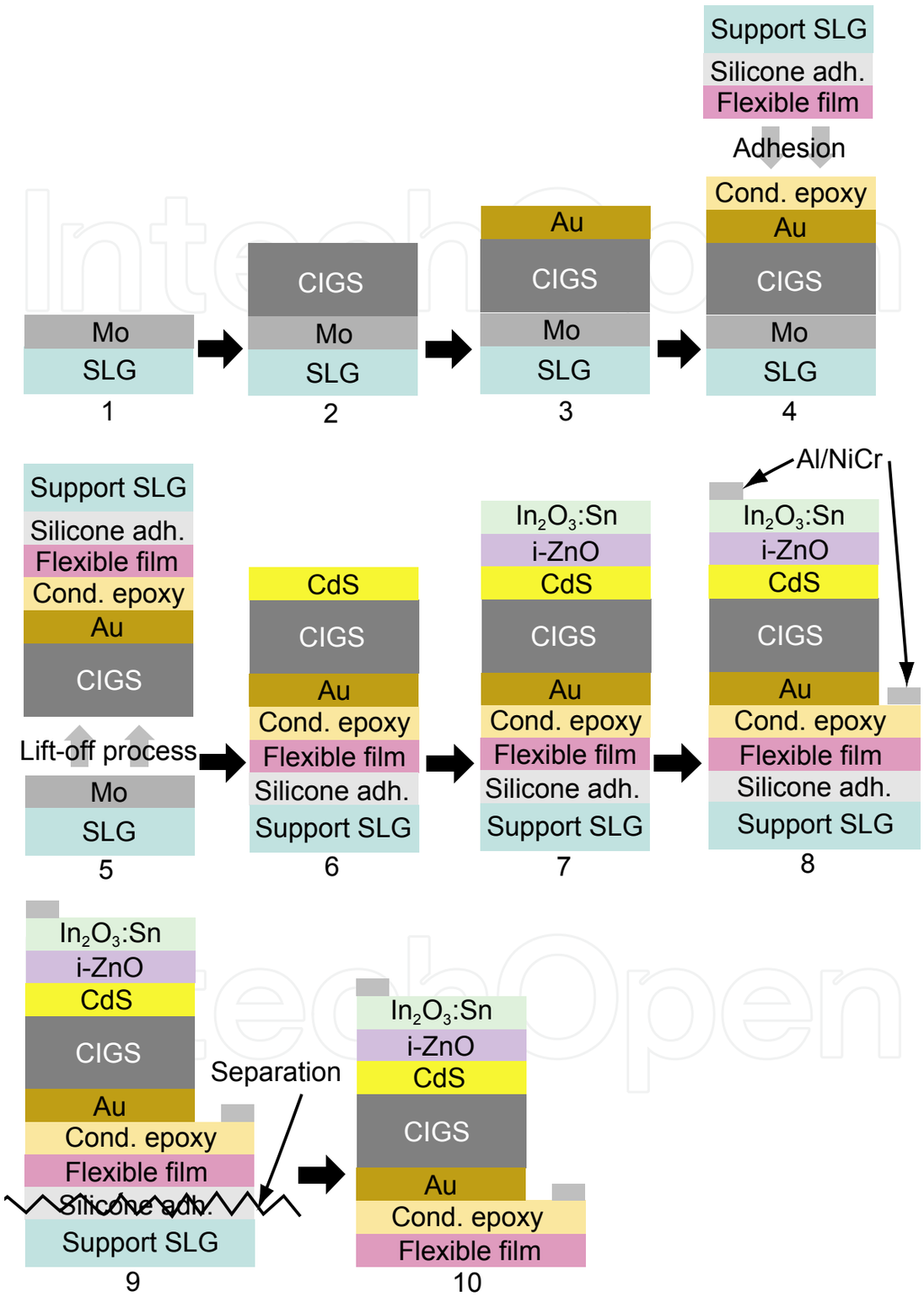

Fig. 1. Schematic illustration of fabrication procedure of flexible CIGS solar cell using lift-off process.

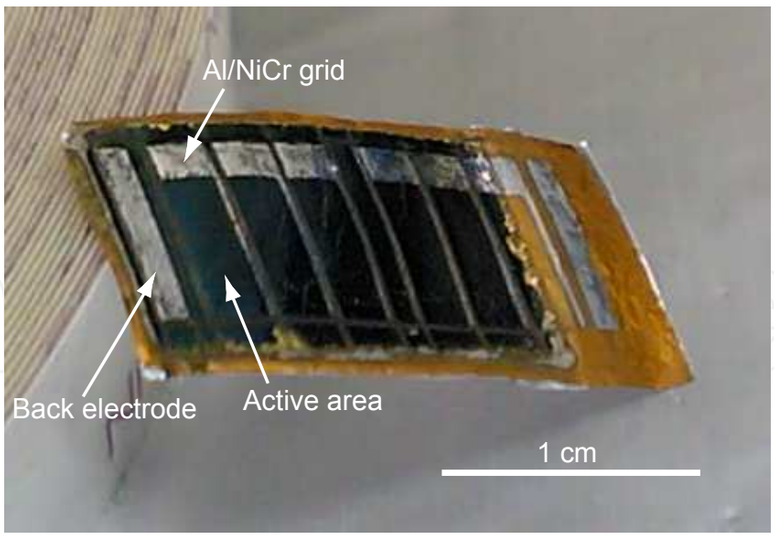

Fig. 2. Photograph of flexible CIGS solar cells using PI film.

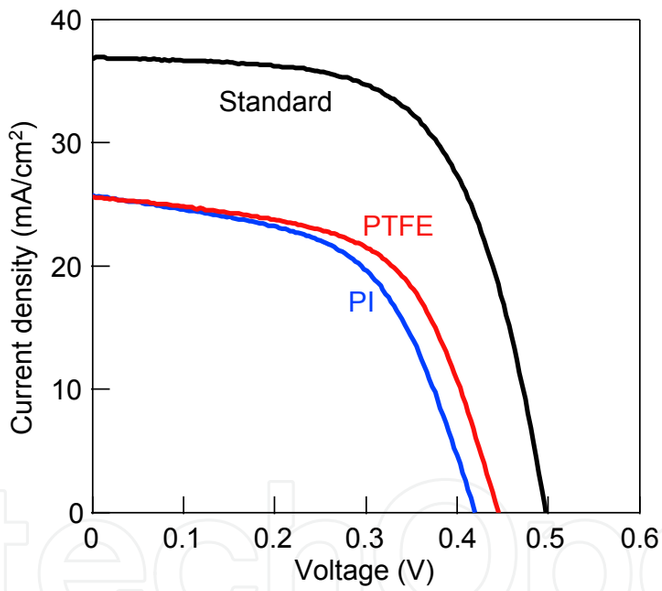

Fig. 3. Photo *J-V* curves of flexible solar cells using PTFE (red) and PI (blue) films. Photo *J-V* curve of standard solar cell without lift-off process (brack) is also shown for comparison.

Sample structure	*Eff.* (%)	J_{sc} (mA/cm²)	V_{oc} (V)	*FF* (%)
PI flexible	5.9	25.7	0.420	54.9
PTFE flexible	6.6	25.6	0.445	57.9
Standard	11.4	36.9	0.497	62.4

Table 2. Solar cell parameters obtaind from flexible solar cells using PI and PTFE films. Solar cell parameters of standard solar cell are also shown for comparison.

conversion efficiency (*Eff.*), and the fill factor (*FF*) are summarized in Table 2. The conversion efficiencies of the flexible solar cells are an approximately half conversion efficiency of the standard solar cell. EQE spectra of these solar cells are shown in Fig. 4. EQEs of the flexible solar cells remarkably decrease in the long wavelength region from 700 to 1200 nm compared to the standard solar cell. We discuss this cause as below.

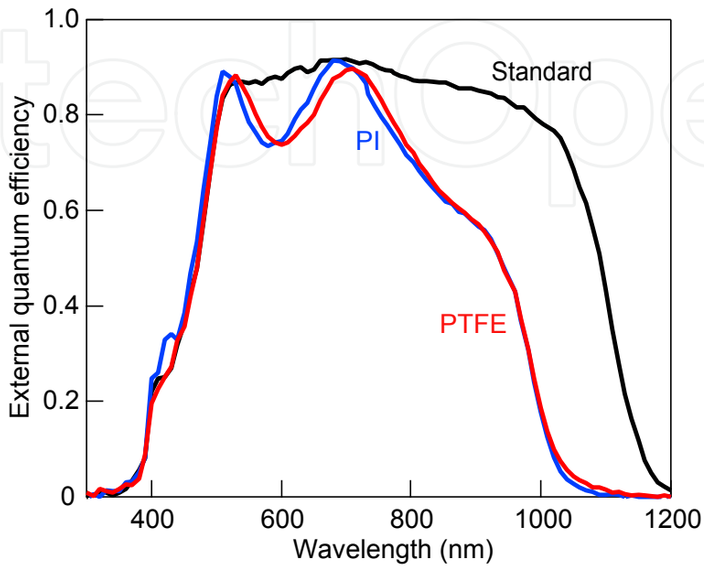

Fig. 4. EQE spectra of flexible solar cells using PTFE (red) and PI films (blue). EQE spectrum of standard solar cell without lift-off process (black) is also shown for comparison. EQE spectra of flexible solar cells are similar irrespective of substrate materials.

As shown in Fig. 5(a), the band gap profile of the standard solar cell consists of the graded band gap structure because of the three-stage deposition process. The diffusion length of electrons generated by the long wavelength light near the back electrode is improved due to the quasi-electric field in which the CIGS layer forms (Contreras et al., 1994b). The graded band gap structure is therefore beneficial for collecting the photogenerated carriers. On the other hand, as shown in Fig. 5(b), the band gap profile of the CIGS layer is inverted due to

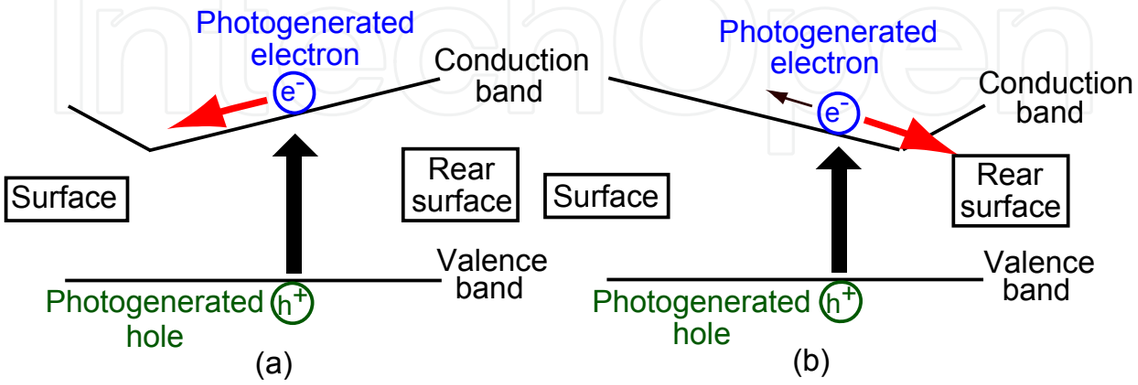

Fig. 5. Schematic illustrations of band gap profiles of CIGS layers. CIGS absorber layers with (a) double geraded band gap and (b) inverted double graded band gap structures are shown.

the lift-off process for the flexible solar cells. We speculate that the band gap profile of the inverted graded band gap structure is not beneficial for collecting the photogenerated carriers by long wavelength light. We conclude that the EQE reductions observed for the flexible solar cells are attributed to the influence of the inverted graded band gap structure.
We describe an interesting point of our flexible solar cells as below. Different materials with different thermal tolerance temperatures are used as the flexible substrates of these flexible solar cells, as shown in Table 1. These flexible solar cells, however, show the similar characteristics irrespective of the flexible film materials from Fig. 3 and Fig. 4.
LBIC and optical microscope images of the flexible solar cell using the PTFE film are shown in Figs. 6(a) and 6(b), respectively. There is a low EQE region on the lower side of the solar cell from Fig. 6(a). This low EQE region corresponds approximately to the flexurelike region from a comparison between Figs. 6(a) and 6(b). This result therefore suggests that this flexure cause reduction of an EQE. LBIC and optical microscope images of the standard solar cell are shown in Figs. 6(c) and 6(d), respectively. In contrast, the LBIC and optical microscope images are uniform for the standard solar cell.

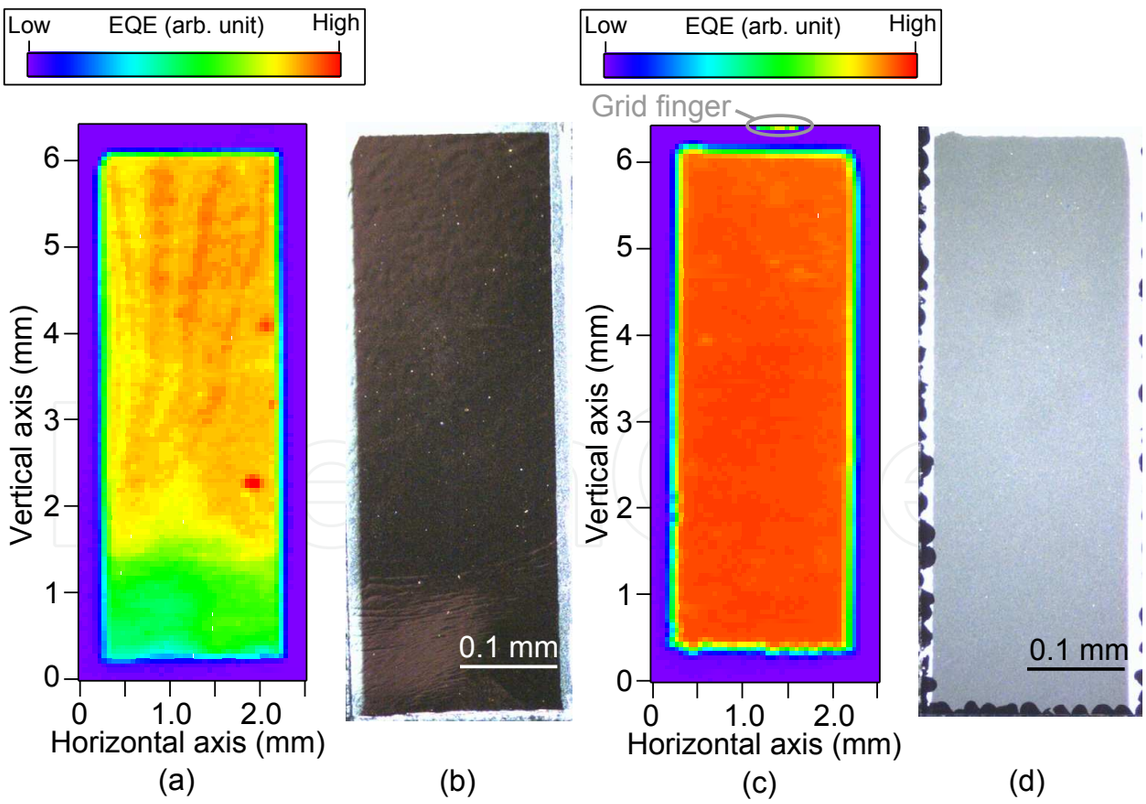

Fig. 6. (a) LBIC and (b) optical microscope images of flexible solar cell using PI film. (c) LBIC and (d) optical microscope images of standard solar cell. Indicators of EQE intensity are shown next to LBIC images.

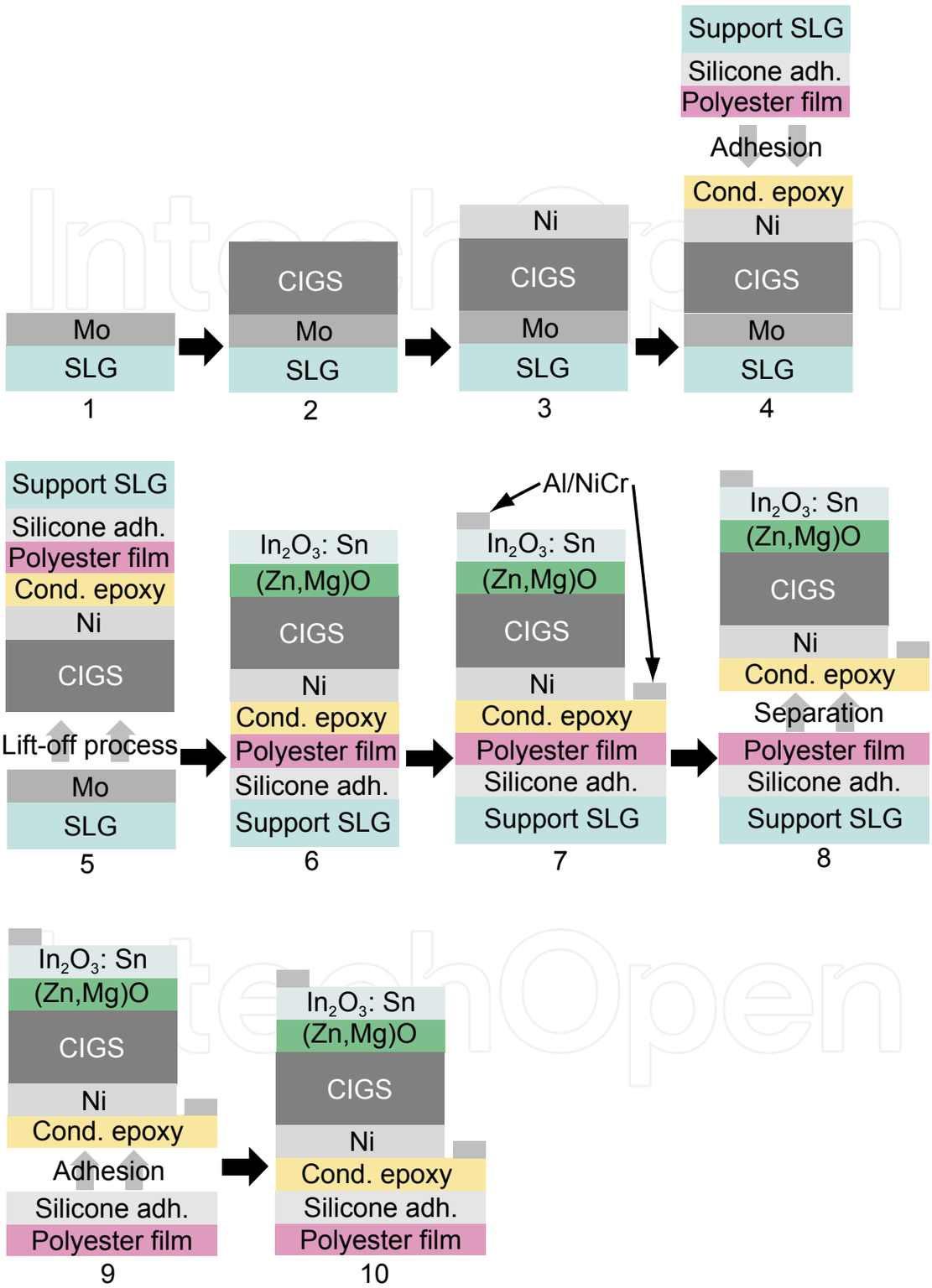

Fig. 7. Schematic illustration of fabrication procedure of flexible CIGS solar cell using $(Zn_{0.83},Mg_{0.17})O$ window layer and lift-off process.

3.2 Development of Cd-free flexible Cu(In,Ga)Se_2 solar cells

We developed a new Cd-free flexible CIGS solar cell using a (Zn,Mg)O window layer. The fabrication procedure is shown in Fig. 7. This process is basically similar to Fig. 1. We deposited a 0.1-μm-thick ($Zn_{0.83}$,$Mg_{0.17}$)O window layer in stead of the ZnO window/CdS buffer layers. The RF magnetron cosputtering method using ZnO and MgO targets was used as the deposition technique (Minemoto et al., 2000, 2001). We also deposited a 0.2-μm-thick Ni layer by the resistive evaporation method as the back electrode in stead of the Au layer. In this subsection, a 55-μm-thick polyester film was used as a flexible substrate. Interestingly, when the flexible solar cell using the polyester film was separated from the support SLG substrate, the detachment occurred not at the support SLG/polyester interface but at the polyester/epoxy interface due to the weaker adhesion at the polyester/epoxy interface. After the substrate-free structure was once, the polyester film was therefore bonded onto the rear surface of the solar cell with a silicone adhesion bond. The photograph of the flexible solar cells fabricated via the above procedure is shown in Fig. 8. We also prepared not only the flexible solar cells using the conventional ZnO window/CdS buffer layers but also the solar cells without the lift-off process for comparison.

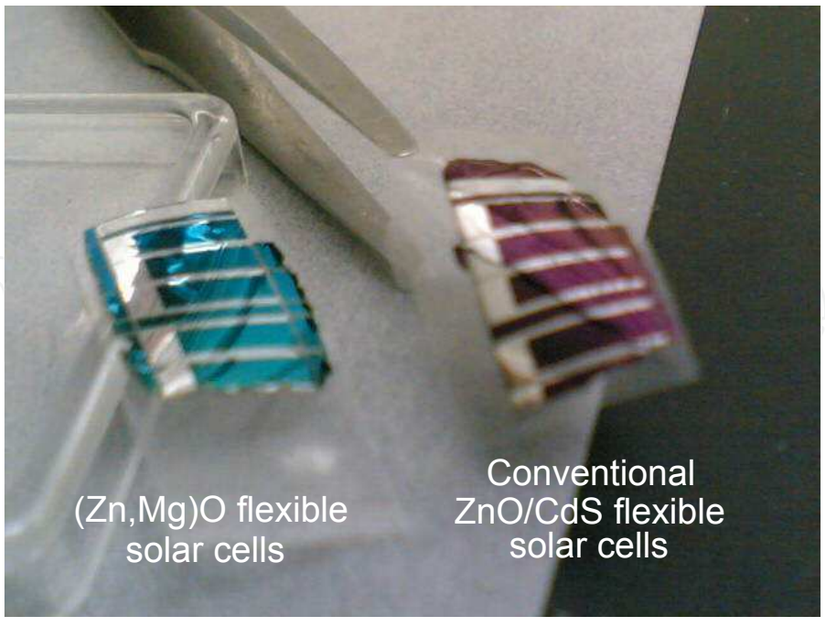

Fig. 8. Photograph of flexible solar cells using polyester film. Left solar cells are Cd-free solar cells using (Zn,Mg)O window layer. Right solar cells consist of conventional ZnO window/CdS buffer layers structure.

The *J-V* characteristics of the flexible solar cells are shown in Fig. 9. The results of the standard solar cells without the lift-off process are also shown in Fig. 9. Solar cell parameters obtained from the *J-V* characteristics are summarized in Table 3. All parameters of the ZnO/CdS solar cell is higher than those of the (Zn,Mg)O solar cell for the standard solar cells. On the other hand, although there are the differences in the window layer/ buffer layer structures for the flexible solar cells, these flexible solar cells show the similar properties.

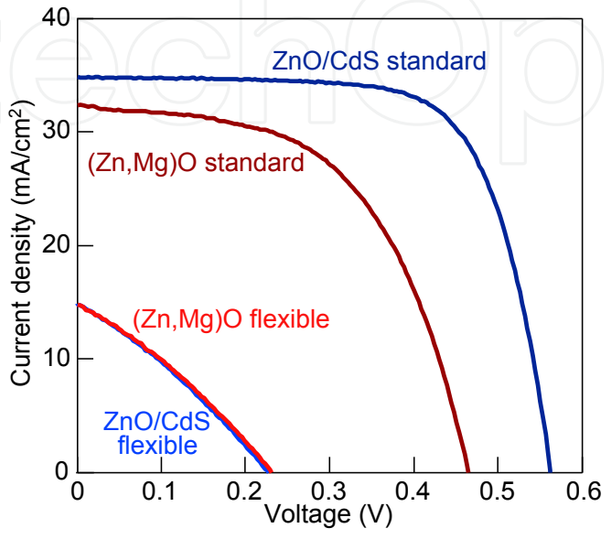

Fig. 9. Photo *J-V* curves of flexible solar cells using (Zn,Mg)O window layer and conventional ZnO window/CdS buffer layers. Photo *J-V* curves of standard solar cells without lift-off process are also shown for comparison.

EQE spectra of these solar cells are shown in Fig. 9. EQEs of the (Zn,Mg)O standard solar cell are higher than those of the ZnO/CdS standard solar cell in the region from 300 to 480 nm, because the band gap of $(Zn_{0.83},Mg_{0.17})O$ is higher than those of CdS and ZnO (Minemoto et al., 2000). These high EQEs in this region is therefore attributed to a low transmission loss of the short wavelength light. Moreover, the tendency of this result is also observed for the flexible solar cells. We found that the (Zn,Mg)O window layer structure was effective for reducing a transmission loss of the short wavelength light even in our flexible solar cells.

Sample structure	*Eff.* (%)	J_{sc} (mA/cm²)	V_{oc} (V)	*FF* (%)
(Zn,Mg)O flexible	1.0	14.8	0.231	30.5
ZnO/CdS flexible	1.0	14.8	0.227	30.2
(Zn,Mg)O standard	8.3	32.4	0.465	54.9
ZnO/CdS standard	13.7	34.9	0.562	70.0

Table 3. Summary of solar cell parameters obtained from flexible solar cells using (Zn,Mg)O window layer and conventinal ZnO window/CdS buffer layers. For comparison, solar cell parametaers otained from standard solar cells using (Zn,Mg)O window layer and ZnO window/CdS buffer layers are also summarized.

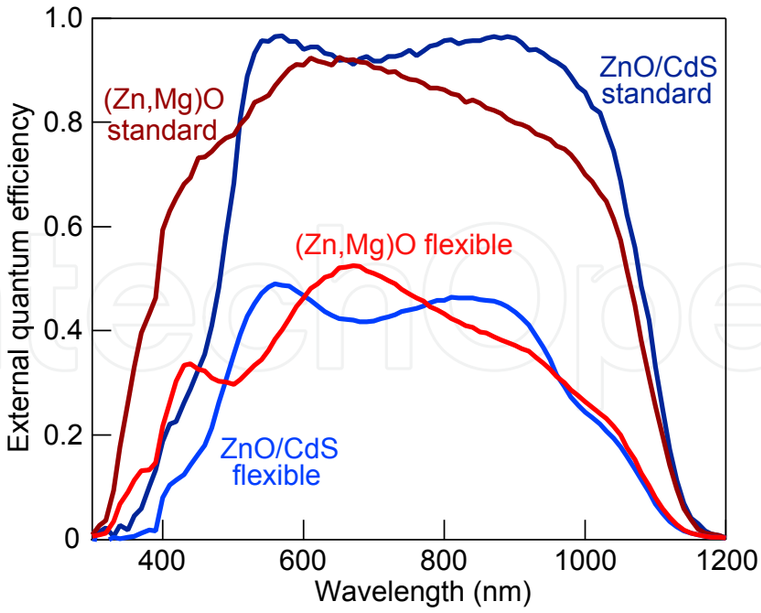

Fig. 10. EQE spectra of flexible solar cells using (Zn,Mg)O window layer (red) and conventional ZnO window/CdS buffer layers (blue). EQE spectra of standard solar cells using (Zn,Mg)O window layer (dark red) and ZnO window/CdS buffer layers (dark blue) are also shown for comparison.

Here, we discuss why these flexible solar cells showed the similar solar cell parameters. In this subsection, we used Ni in stead of Au as a back electrode material. In subsection 3.1, the ZnO/CdS flexible solar cells with the Au back electrode showed a conversion efficiency of ~6%. We think that the Ni back electrode may limit performance of these solar cells. We therefore speculate that the Ni atoms, which diffused into the CIGS layer from the back side due to the low temperature annealing, behave as recombination centers for electrons.

4. Conclusion

After we described the review of the lift-off process, we also described the advantages of the lift-off process in the flexible CIGS solar cell fabrication. We developed the fabrication procedure of the flexible CIGS solar cells using the lift-off process. The characteristics of the flexible solar cells were shown compared to the standard solar cell. Although the conversion efficiencies of the flexible solar cells using the lift-off process are an approximately half conversion efficiency of the standard solar cell, the flexible solar cells showed the similar characteristics irrespective of the substrate materials. Moreover, we attempted the concept of a Cd-free solar cell. We found that the choice of back electrode materials is a crucial problem rather than the window layer/buffer layer structure. We expect that the lift-off process further advances through our results.

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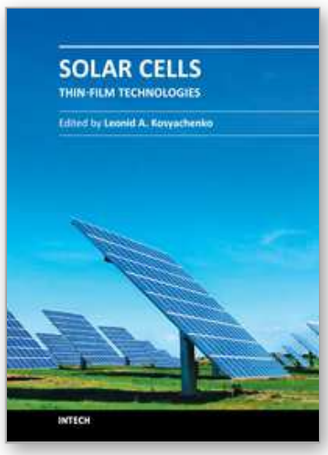

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The first book of this four-volume edition is dedicated to one of the most promising areas of photovoltaics, which has already reached a large-scale production of the second-generation thin-film solar modules and has resulted in building the powerful solar plants in several countries around the world. Thin-film technologies using direct-gap semiconductors such as CIGS and CdTe offer the lowest manufacturing costs and are becoming more prevalent in the industry allowing to improve manufacturability of the production at significantly larger scales than for wafer or ribbon Si modules. It is only a matter of time before thin films like CIGS and CdTe will replace wafer-based silicon solar cells as the dominant photovoltaic technology. Photoelectric efficiency of thin-film solar modules is still far from the theoretical limit. The scientific and technological problems of increasing this key parameter of the solar cell are discussed in several chapters of this volume.

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