# A Study on Epitaxial Lift-Off in InGaP/GaAs Double-Junction Solar Cells via Au-Au Bonding on Pre-Patterned Area

Gwang yeol Park, Chea won Kim, and Hyo jin Kim

***Abstract*—We have investigated the improvement of GaAs substrate reusing and the regrowth possibility of the epi-layer for the flexible InGaP/GaAs double junction solar cell. The presence of residues on the GaAs substrate surface separated by the epitaxial lift-off process is closely related to the regrowth of the epitaxial layer. A pre-patterning process was introduced to investigate the correlation between the lateral etch rate of the sacrificial layer and the generation of residues on the substrate. And the lateral etch rate was adjusted according to the changes in the pre-patterning area. The residues on surface of the GaAs substrate according to the lateral etch rate were observed by SEM/EDX. The structure of the InGaP/GaAs double junction solar cell re-grown on the GaAs substrate was measured by X-ray diffraction. The performance of the re-grown solar cell was measured by the current density-voltage curve at AM1.5G, 1sun with a solar simulator. The GaAs substrate was separated in the cleanest state by the pre-patterning area of 2 x 2mm$^2$, and the conversion efficiency of the re-grown solar cell by reusing this for three times was maintained similar to its conversion efficiency when a solar cell of initial growth.***

***Index Terms*—Flexible solar cell, epitaxial lift-off, pre-patterning, lateral etch rate, substrate reusing.**

## I. INTRODUCTION

THE InGaP/GaAs double (2J) solar cells (SCs) were grown in the reverse direction to fabricate thin-film SCs on (1 0 0) Si-doped GaAs substrates, which are oriented 2° off toward [1 1 1]. Films were deposited using a metalorganic chemical The group III-V compound solar cell can have a multi-junction structure compared to other solar cells, so it can absorb a wide wavelength range of the solar spectrum so that ultra-high efficiency can be obtained [1]. In addition, the III-V compound having a direct transition energy bandgap has low heat loss, so it can maintain its efficiency even with a thin film without a substrate, so it can be applied to flexible solar cells suitable for mobile devices such as automobiles and drones [2]–[4]. However, since the III-V solar cell has the disadvantage of being composed of a high-cost material, many studies are being conducted on a method for reducing the cost [4], [12].

Many groups have been researching substrate recycling, high-speed growth of epi, and the use of inexpensive metals to reduce the manufacturing cost of III-V solar cells [5]–[14]. Among them, the method that plays the biggest role in reducing manufacturing costs is GaAs substrate recycling [6]–[14]. To reuse GaAs substrates, an epitaxial lift-off method (ELO) is required. The epitaxial lift-off method is a method of separating the epitaxial layer from the substrate by growing an AlAs sacrificial layer on a GaAs substrate, growing an epitaxial structure, bonding a flexible substrate, and removing the sacrificial layer with an HF mixture [4]. The manufacturing cost of III-V solar cells is related to how much substrate recycling can be stably performed.

However, it has been reported that it is difficult to obtain conditions capable of growing the epitaxial-layer under the same stable conditions by using the same substrate after recycling the substrate. According to the results of previous studies [6], [7], the reaction residues (such as $As_2O_3$ or $AlF_3$) remained on the substrate surface due to the reaction of GaAs and HF solution during the ELO process, increasing the surface roughness of the GaAs substrate, making it difficult to achieve stable epitaxial growth. In addition, studies have been conducted on the case of regrowth after etching the residue on the surface of the GaAs substrate after ELO through the chemical-mechanical polishing (CMP) [8] process, but this method reduces the thickness of the substrate during substrate recycling, so there is a limit to the number of recycling cycles. K.S. Lee *et al.* [9] reported a study to improve the number of recycling substrates through an HF mixture with a high ELO etching rate. As such, there is still much to study on the stable process operation for GaAs substrate recycling after the ELO process.

In this study, we have investigated the improvement of GaAs substrate reusing and the regrowth possibility of the epi-layer for the flexible InGaP/GaAs double junction solar cells. The lateral etch rate was changed by various pre-patterning areas to investigate the residue on the surface of the GaAs substrate after the ELO process. The residues on the GaAs substrate surface were observed by SEM/EDX. And the grown epitaxial layer structures on the GaAs substrate was investigated by the X-ray

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Manuscript received January 17, 2022; revised March 29, 2022; accepted April 8, 2022. Date of publication April 13, 2022; date of current version May 6, 2022. This work was supported in part by the Technology Development Program to Solve Climate Changes of the National Research Foundation (NRF) funded by the Ministry of Science ICT under Grant 2017M1A2A2048903, in part by the Leading program of the National Research Foundation (NRF) funded by the Ministry of Science under Grant 2021M3H4A1A02051253, and in part by the Ministry of Trade, Industry & Energy (MOTIE) of the Republic of Korea under Grant 70300026. *(Corresponding author: Hyo jin Kim.)*

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Digital Object Identifier 10.1109/JPHOT.2022.3167104

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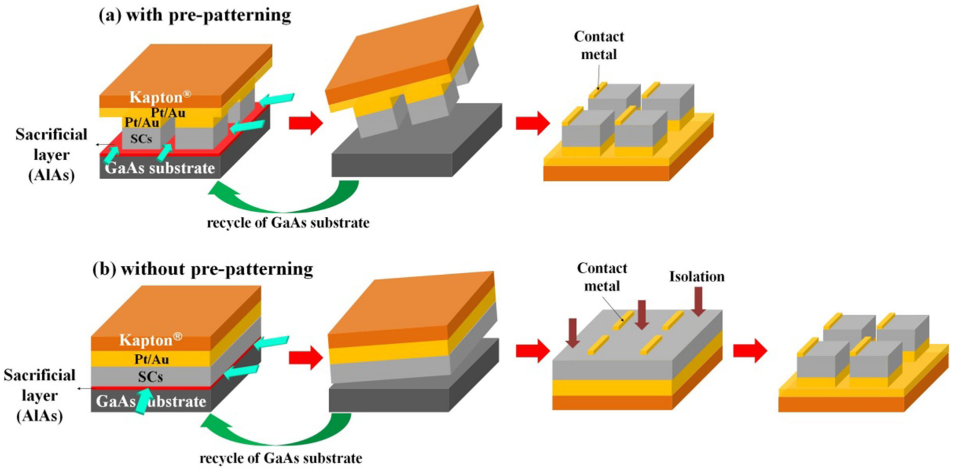

Fig. 1. Schematic of the epitaxial lift-off process for flexible InGaP/GaAs 2J solar cells: (a) With pre-patterning; (b) Without pre-patterning.

diffraction (XRD) peak pattern. The current-voltage curve was investigated with a solar simulator at AM1.5G, 1sun.

## II. EXPERIMENTAL PROCEDURES

The InGaP/GaAs 2 J SCs were grown in the reverse direction to fabricate thin-film SCs on (1 0 0) Si-doped GaAs substrates, which are oriented 2° off toward [1 1 1]. Films were deposited using a metalorganic chemical vapor deposition horizontal reactor (AIX200/4 RF, AIXTRON Inc.). Trimethylgallium, trimethylindium, and trimethylaluminum were used as the group III sources. Meanwhile, arsine and phosphine were employed as group V sources. The substrate temperature and the reactor pressure were kept at 650°C and 100 mbar respectively, while a lower temperature of 550°C was adopted for the tunnel junction growth. The epi-layers started with a 300 nm GaAs buffer layer followed by a selective etching layer of InGaP/GaAs/AlAs/GaAs/InGaP for the ELO process. Then the inverted InGaP/GaAs 2J SCs were grown as follows: a 300 nm n-type GaAs contact layer with an electron concentration of $8.65\times10^{18}$ $cm^{-3}$, InGaP top cell, $p^{+}$-$Al_{0.5}Ga_{0.5}As$/$n^{+}$-GaAs reverse tunneling junction, GaAs bottom cell, p-type GaAs contact layer with a hole concentration of $1.49\times10^{19}$ $cm^{-3}$.

Fig. 1(a) and (b) show that the schematics diagram for the fabrication procedures of the flexible III-V solar cells with the pre-patterning area and without it before the bonding the epi-layer on flexible substrate. The pre-patterning area [14] defined as a pre-patterning mesa before the bonding process, resulting in process simplification by using the pre-patterning mesa for device isolation. The GaAs substrate was used a quarter piece of 2-inch. The pre-patterning area for lateral etch rate change was prepared by a photolithography process with the following surface dimensions: 2 × 2 $mm^2$, 3 × 3 $mm^2$, 5 × 5 $mm^2$. The interval between each pre-patterning area was equally patterned at 600 $\mu$m intervals to reduce the lateral etch parameters. As a control sample, a quarter of 2 inches was prepared without pre-patterning (Fig. 1(b)). The pre-patterning area was deposited with 20 nm of platinum and 70 nm of gold by an e-beam evaporator and was selectively isolation etched by HCl and $NH_4OH/H_2O_2$ mixture solution until AlAs sacrificial layer was exposed as shown Fig. 1(a). Then, Au-Au bonding was performed with a 150um-thick flexible sheet (Kapton) on which Pt and Au were deposited.

The Au-Au bonding between the epi-layer of solar cell and the flexible sheet was performed with during 1.5 hour at 200 °C by high-vacuum bonding system. Before the bonding, the surface of the epi-layer and the flexible sheet was activated using the plasma-activated method [15], [16] in a reactive ion etching (RIE) chamber. RIE was operated at the working pressure of 30mTorr and the radio frequency power of 100 W during 1 min 30 sec.

The lateral etch rate of the AlAs sacrificial layer was calculated by measuring the separation time with different pre-patterning area. The residual elements on the surface of GaAs substrate after ELO were investigated by the scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX). Then, the epi layer for InGaP/GaAs 2J SCs was re-grown on the same substrate. The epitaxial layer structure of re-grown solar cell was measured by X-Ray diffraction (XRD). The current density-voltage curve of InGaP/GaAs 2J SC was measured at AM1.5G, 1sun with Solar Simulator.

## III. RESULTS AND DISCUSSION

Fig. 2 shows the changes in the lateral etch rates corresponding to the pre-patterning surface areas of 2 × 2 $mm^2$, 3 × 3 $mm^2$, 5 × 5 $mm^2$, and the control area (without pre-patterning). The lateral etch rate is a value obtained by measuring the time until the epitaxial layer of the 2J SC and the GaAs substrate are separated during the ELO process. According to our results, the lateral etch rate increased as the pre-patterning area decreased, as shown in Fig. 2. The lateral etch rate during the ELO process is associated with an increase in the surface area exposed to the etchant, as the pre-patterning area decreases. In addition, the lateral etch rate of the pre-patterning samples resulted in a higher value compared to that of the sample without pre-patterning. In a previous study, the isolated space between the pre-patterning area created by device isolation served as a release passage for $H_2$ bubbles generated

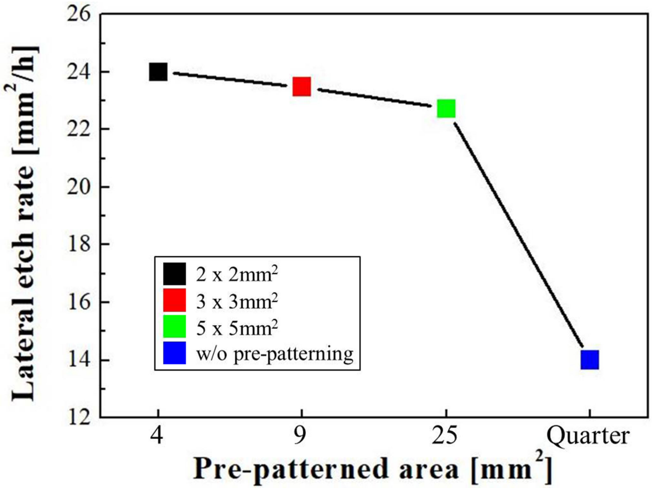

Fig. 2. Lateral etch rate of AlAs sacrificial layer under different pre-patterning area surface.

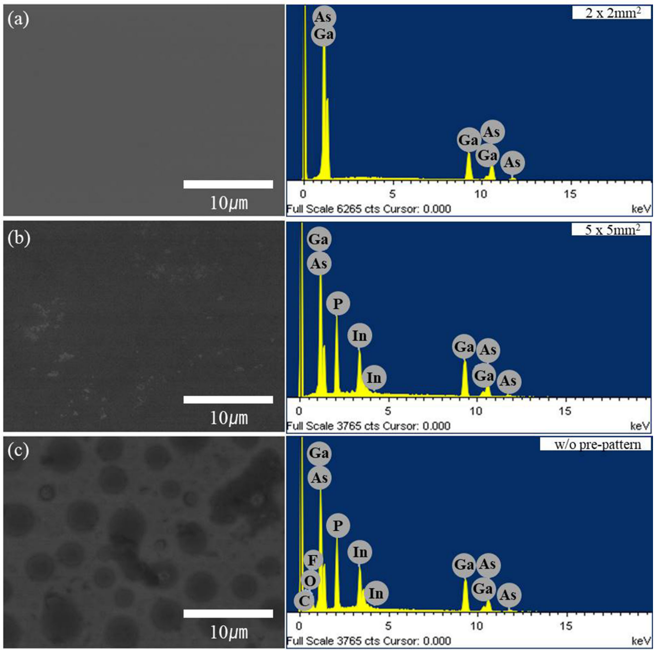

Fig. 3. SEM images and EDX spectra of the GaAs substrate surface with different pre-patterning surface after the ELO process: (a) 2 x 2 mm$^2$ (b) 5 x 5 mm$^2$ (c) Without pre-patterning area.

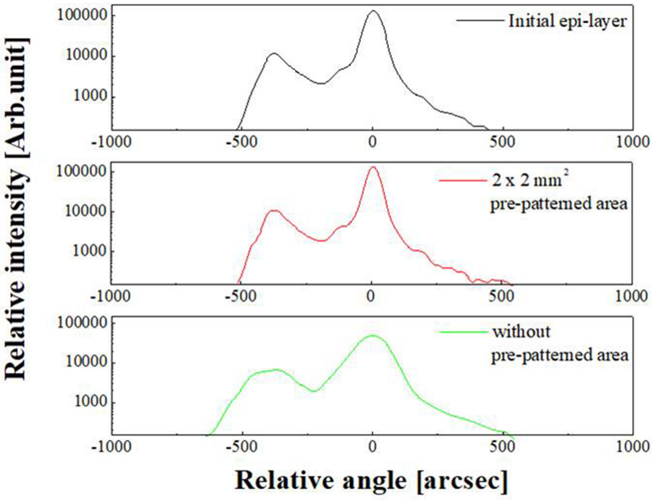

Fig. 4. XRD spectra of the epitaxial-layer structure: (Black line) Initial grown of InGaP/GaAs 2J SCs on GaAs substrate,; (Red line) 3rd regrowth of InGaP/GaAs 2J SCs on GaAs substrate using a 2 x 2 mm$^2$ pre-patterning area,; (Green line) 3rd regrowth of InGaP/GaAs 2J SCs on GaAs substrate used without pre-patterning.

during the ELO process, which allowed the etchant to contact sufficiently with the sacrificial layer [17]. As a result, the lateral etch rate under pre-patterning conditions was faster than that without pre-patterning.

The SEM image in Fig. 3(a) shows that the GaAs substrate surface of the pre-patterning sample with pre-patterning area of 2 × 2 mm$^2$ remained intact, with no traces of residues after the ELO process. This was verified by the EDX results, where only the elements constituting the GaAs substrate (Ga, As) were observed. Fig. 3(b) shows the presence of some residual elements on the surface of the pre-patterning GaAs substrate with surface dimensions of 5 × 5 mm$^2$. Indium (In) and phosphide (P) also appeared in the EDX graph, which were theoretically attributed to the part of the InGaP surface that remained unetched by the remaining AlAs layer after the ELO process. On the other hand, residues were observed on the GaAs substrate surface of the sample without pre-patterning process, as shown in Fig. 3(c). it should be noted that O and F peaks were also observed in the EDX spectra, in addition to the elements constituting the GaAs substrate and the unetched part of the etch stop layer. A previous study [18] has confirmed that these elements stem from reaction residues ($As_2O_3$ or $AlF_3$) that are generated when the AlAs layer is etched by the HF solution. From these results, a correlation was inferred between the pre-patterning area and the residues on the separated GaAs substrate surface. The surface of the GaAs substrate separated by the ELO process was more intact, as the pre-patterning area was smaller.

Fig. 4 shows the XRD graph of the structure of InGaP/GaAs 2J SCs were grown inverted on a GaAs substrate. The peaks of the epitaxial layer of InGaP SCs is located between -650 and -250 arcsec, while the peaks of the GaAs substrate and GaAs SC are located at near 0 arcsec. Each peak was expressed on a log scale of relative intensity. The sample with pre-patterning area of 2 × 2 mm$^2$ was grown in a structure similar to the structure grown on the initial substrate when SCs were re-grown by reusing the separated GaAs substrate three times. The InGaP and GaAs peaks of regrown of SCs three times without using pre-patterning had widened full width at half maximum (FWHM) and reduced relative intensity. From the results of previous studies [12], it was known that the presence of residual materials, such as oxides, on the GaAs substrate can act as defects affecting the growth of SCs. According to the SEM/EDX graph in Fig. 3, it could observe that the residues on the surface of the GaAs substrate by the presence or absence of pre-patterning were different. It could observe that the surface of the substrate applied by pre-patterning was clean, whereas the surface of the substrate without pre-patterning had a lot of residues. In conclusion, the residual material on the GaAs

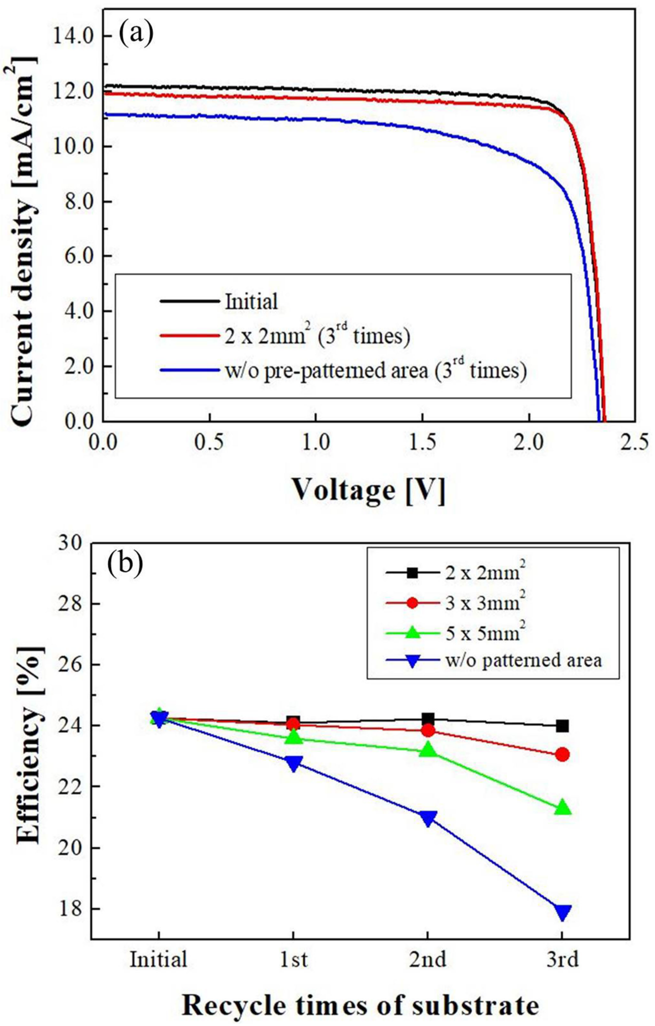

Fig. 5. Characteristics of flexible inverted InGaP/GaAs 2J SCs: (a) Current density-voltage curves of the SC grown on initial substrate and three times reused GaAs substrate, under 1 sun(AM1.5G) of solar illumination; (b) Conversion efficiency of the flexible inverted InGaP/GaAs 2J SCs vs number of GaAs substrate-recycle per pre-patterning area.

substrate affected the re-growth of the solar cell, which could be observed as a change in the XRD peak.

Fig. 5(a) shows the current density-voltage (I-V) curves of the initial flexible InGaP/GaAs 2J SCs, as well as the flexible InGaP/GaAs 2J SCs regrown on the GaAs substrates that were reused three times. The flexible InGaP/GaAs 2J SCs applied with the pre-patterning process had similar performance to the initial flexible SCs, even after re-growth three times. On the other hand, the I-V curve of the flexible InGaP/GaAs 2J SCs re-grown applied without a pre-patterning process decreased as reusing of the GaAs substrate. The cause of this result as shown in SEM/EDX that a residue was left on the surface of the GaAs substrate after the ELO process. These residues harmed to re-growth of solar cells as shown from the XRD graph in Fig. 4 which was caused the degradation of the performance of the fabricated solar cells. Fig. 5(b) illustrates the conversion efficiency ($\eta$) of the flexible III-V SCs plotted against the number of GaAs substrate recycling per pre-patterning area. The $\eta$ of the SCs using the pre-patterning area of $2 \times 2$ mm$^2$ was similar to that of the initial one, even after being reused three times. However, the $\eta$ of the SCs using the pre-patterning area of $3 \times 3$ mm$^2$ started to decrease from the 2nd reuse. The $\eta$ of the SCs using a pre-patterning area of $5 \times 5$ mm$^2$ decreased when reused for a 2nd and 3rd time, respectively. The $\eta$ of the SCs without a pre-patterning area started to rapidly decrease from 1st reuse. Thus, it could be confirmed that the $\eta$ value changed according to the pre-patterning area and the number of substrates recycles; specifically, the generated residue by the ELO process was reduced as the pre-patterning area was smaller, and an epitaxial structure similar to the initial growth could be grown, thereby was remaining the efficiency of the solar cell. On the other hand, more residues were generated in the absence of a pre-pattering area or as the pre-patterning area became larger. These residues continuously were accumulated whenever the GaAs substrate was reused. As a result, the conversion efficiency continued to decrease as harmed the re-growth of the InGaP/GaAs 2J SCs.

## IV. CONCLUSION

In conclusion, by coupling the ELO technique with a pre-patterning process, we were able to effectively recycle GaAs substrates for the fabrication of flexible InGaP/GaAs 2J SCs. The pre-patterning process increased the contact surface area between the AlAs sacrificial layer and the HF etchant, improved the lateral etch rate in the process, and served as a release passage for the $H_2$ gas bubbles generated during the ELO process. The lateral etch rate of the AlAs sacrificial layer increased as the pre-patterning area became smaller. As the lateral etch rate increased, the GaAs substrate could be separated in a more cleaned state by the ELO process. The epitaxial layer of the InGaP/GaAs 2J SCs was re-grown by reusing the same substrate. Notably, the structure of the re-grown epitaxial layer remained similar to the structure of the initially grown layer even after the substrate recycles three times. In addition, the flexible InGaP/GaAs 2J SCs fabricated with the regrown epitaxial layer preserved almost the same of their initial performance after three times recycles while retaining their similar current density-voltage characteristics. Overall, it was experimentally confirmed that the coupling of the ELO technique with a simple pre-patterning process allows for the effective reuse of GaAs substrates in the fabrication process of III-V solar cells. This strategy induces relatively zero performance downgrades and is expected to reduce the fabrication costs of III-V solar cells and improve price competitiveness.

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