Efficiency improvement in InGaN-based solar cells by indium tin oxide nano dots covered with ITO films

Dong-Ju Seo,1 Jae-Phil Shim,1 Sang-Bae Choi,1 Tae Hoon Seo,2 Eun-Kyung Suh,2 and Dong-Seon Lee1,*

1School of Information and Communications, Gwangju Institute of Science and Technology (GIST), 261 Cheomdang-gwagiro, Buk-gu, Gwangju 500-712, South Korea
2School of Semiconductor and Chemical Engineering, Semiconductor Physics Research Center (SPRC), Chonbuk National University, 664-14 1 Ga Deokjin-dong Jeonju-city Jeonbuk 561-756, South Korea
*dslee66@gist.ac.kr

Abstract: InGaN based MQW solar cells have been fabricated with 4 different transparent top electrode structures: (1)- ITO 200 nm, (2)-ITO nano dots only, (3)-ITO nano dots on ITO 50 nm and (4)-ITO nano dots on ITO 100 nm. The solar cell with the ITO 50 nm on ITO nano dots under AM 1.5 conditions showed the best results: 2.3 V for \( V_{oc} \), 0.69 mA/cm\(^2\) for \( J_{sc} \), 41.8% for peak EQE, and 0.91% for conversion efficiency. Efficiency improvement was possible due to the decreased reflectance achieved by the ITO nano dots covered with an ITO film with optimized thickness.

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References and links
13. S. Y. Bae, J. P. Shim, D. S. Lee, S. R. Jeon, and G. Namkoong, “Improved photovoltaic effects of a vertical-type...
1. Introduction

Indium Gallium Nitride (InGaN) material is now widely used in various optoelectronic devices including light-emitting diodes (LEDs) [1] and laser diodes (LDs) [2]. It is also being studied increasingly as a prospective material for solar cells [3–5]. One of the merits for solar cell applications is that the band gap energy can be engineered from 0.7eV for InN to 3.4eV for GaN by varying the indium composition, which covers most of the solar spectrum from UV to IR [6]. Despite this potential advantage, conversion efficiency of InGaN based solar cells is still very low. It is essential for better cell efficiency to improve not only the crystalline quality of the epitaxial layers but also the fabrication of the solar cells. Fabrication includes transparent top electrodes and surface texturing which will improve the carrier extraction. Surface texturing is one of the most employed methods to enhance the extraction efficiency in LED fabrication and can be formed on a p-GaN surface [7] on a N-face of GaN [8], and even on an indium tin oxide (ITO) layer [9–11]. Surface texturing methods have also been adopted in InGaN-based solar cells [12,13] and proved to enhance the efficiency. Because, however, texturing by direct etching of p-GaN was known to induce damage [14] and resulted in degraded electrical properties, texturing has been studied widely on the ITO layers. ITO is used for visible LEDs as a current spreading layer [15] due to its high transparency in the visible spectrum and low electrical resistivity. For similar reasons, the ITO has also been used in InGaN-based solar cells [16]. It is well-known that ITO thickness optimization is important in LED applications since the reflectance is fluctuated by the variation of the ITO thickness resulting in reduced light extraction at the target wavelength [17]. In solar cell applications, the ITO thickness should also be optimized since its variation also causes reflectance fluctuation [18] resulting in reduced absorption of sunlight. Reflectance of the ITO can be further reduced by combining the thickness optimization with the ITO texturing. ITO texturing by wet etching or dry etching, however, was also revealed to increase series resistance of the ITO film. Moreover, those wet- and dry-etched ITO films turned out to be just rough surfaces without any control over film thickness [19,20]. In this work, we report a new way of texturing by deposition of thickness-optimized ITO films on...
the ITO nano dots, so that the films can have minimum reflectance as well as no electrical degradation originated from the ITO etching process.

2. Experiment

The solar cell structures were grown using metal organic chemical vapor deposition (MOCVD). Trimethylgallium, ammonia and trimethylindium were used as Ga, N, and In sources, respectively. Si doped 2.2 um n-GaN was grown at 1040 °C on an undoped GaN/Al₂O₃ template. Multi quantum well (MQW) pairs of In₀.₂₇Ga₀.₇₃N/GaN layers were grown at 810 °C, and a Mg-doped 100 nm p-GaN layer were grown at 1020 °C sequentially. Doping concentration of p-GaN and n-GaN was 10¹⁷ cm⁻³ and 10¹⁸ cm⁻³, respectively. 1 mm × 1 mm solar cell devices with the ITO film on the ITO nano dots were fabricated by a sequential process as shown in Fig. 1(a). ITO 100 nm is deposited by an electron beam evaporator and is annealed in an ambient with a mixture of N₂ and O₂ at 600 °C for 60s in a rapid thermal annealing (RTA) chamber, etched by a diluted HCl (HCl: H₂O = 1: 3) for 5 s to form the ITO nano dots on the p-GaN surface, and annealed at 600 °C for 5 min again. The nano dots formed this way are shown in Fig. 1(b). The ITO nano dots are about 80 nm in diameter and cover about 30% of the surface. Next, the 2nd ITO film is deposited as a current spreading layer on the ITO nano dots to form a network. Then it is annealed at 600 °C for 5 min. ITO 50 nm and 100 nm were deposited for comparison as shown in Fig. 1(c) and 1(d) and we can see that the surfaces are as rough as the ITO nano dots. Finally, Cr/Au (30 nm/300 nm) is deposited for metal contact.

3. Theory

The thickness of the ITO film is very important and is controlled by adjusting the deposition time. Reflectance can be calculated and minimized at the target wavelength by varying the ITO thickness according to the following reflectance equation at normal incidence.

\[ R = \frac{n_i^2(n_0 - n_t)^2 \cos^2 \delta + (n_0n_t - n_i^2)^2 \sin^2 \delta}{n_t^2(n_0 + n_1)^2 \cos^2 \delta + (n_0n_t + n_1^2)^2 \sin^2 \delta} \]  

\[ \delta = \frac{2\pi}{\lambda_0} (n_1 t) \]
$n_s$, $n_l$, and $n_0$ are the refractive indices of each material as shown in the inset in Fig. 2(a). $\delta$ is the phase difference between the light reflected on the $n_s$ material (red arrows in the inset of Fig. 2(a)) and the light reflected on the $n_l$ material (blue arrows). $t$ is thickness of the $n_l$ film and $\lambda_0$ is the wavelength of light at normal incidence. In this case, constructive or destructive interference occurs between lights with different phases. That makes reflectance fluctuation at the thin film dependent on the thickness of the sandwiched film between the $n_s$ and $n_0$ material. This phenomenon is caused by the interference between the reflected lights from the ITO and the GaN surface. Figure 2(a) shows the reflectance of the ITO on GaN. The lines with filled markers are measured data, and unfilled markers are simulated data by finite-difference time-domain (FDTD) method using Eq. (1). Simulated reflectance fluctuation tendency is very close to the measured data and the small deviation is thought to come from the surface morphology, absorption and thickness variation by the E-beam evaporator. Contour plot in Fig. 2(b) was obtained as a result and we can see that the 50 nm ITO has a lower reflectance than the 100 nm ITO on GaN.

![Fig. 2.](image)

Based on the previous simulation, structures having ITO nano dots covered by ITO films were made and reflectance was measured on them at normal incidence. Results are shown in Fig. 3 for 4 different structures and are drawn on the right side. In the same plot, the ITO 50 nm and 100 nm without nano dots are also plotted for comparison. It clearly shows that 3)-ITO 50 nm on the ITO nano dots has the lowest reflectance and 4)-ITO 100 nm on the ITO nano dots has the next lowest reflectance over the entire wavelength range. Not only the lower reflectance of the ITO 50 nm, but also the surface roughening due to the nano dots [21] contribute to the lowest reflectance of the ITO 50 nm on the ITO nano dots.

![Fig. 3.](image)

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4. Results and discussion

Then, in order to verify the enhancement through reducing the reflectance, solar cells were fabricated with the (1)-200 nm ITO film, (2)-ITO nano dots only, (3)-ITO 50 nm on the ITO nano dots and (4)-ITO 100 nm on the ITO nano dots, respectively. Figure 4(a) shows current density versus voltage ($J-V$) characteristics of the samples under AM 1.5 illumination. The obtained open-circuit voltages ($V_{oc}$) are approximately 2.3 V for all samples and a large $V_{oc}$ value had identical characteristics as the InGaN solar cells that were in previous publications [22,23]. Fill factors (F.F.) are 58.2%, 58.5%, 58.1% and 58.1%, corresponding to (1)-200 nm ITO film, (2)-ITO nano dots only, (3)-ITO 50 nm on the ITO nano dots and (4)-ITO 100 nm on the ITO nano dots, respectively. F.F. of these samples are very similar in spite of the degraded curvature in the $J-V$ curve, which is known to come from doping variation and/or polarization in the InGaN MQW structure [24]. While the $V_{oc}$ and the F.F. are almost the same for all samples, we can see a considerable difference in the short-circuit current density ($J_{sc}$) which is influenced by the light absorption. $J_{sc}$ values were 0.56 mA/cm$^2$ for (1)-ITO 200 nm (ref.), 0.53 mA/cm$^2$ for (2)-ITO nano dots only, 0.69 mA/cm$^2$ for (3)-ITO 50 nm on the ITO nano dots and 0.63 mA/cm$^2$ for (4)-ITO 100 nm on the ITO nano dots. The sample with the ITO 100 nm on the ITO nano dots showed ~12% improvement in $J_{sc}$ compared to the ref. and ITO 50 nm on the ITO nano dots showed a 22% improvement as shown in Fig. 4(a). It indicates that the samples with the ITO film on the ITO nano dots have higher sunlight absorption.

![Fig. 4. (a) J-V curves and (b) EQE characteristics for Solar Cells with planar ITO 200 nm (ref.), ITO nano dots only, ITO 50 nm on the ITO nano dots and ITO 100 nm on the ITO nano dots for transparent current spreading layer, respectively.](image)

External quantum efficiency (EQE) curves in Fig. 4(b) also surely show an enhancement of carrier extraction due to increased sunlight absorption. The peak intensity of the EQE is approximately 5.19% higher in the sample with ITO 100 nm on the ITO nano dots, and 14.05% higher in the sample with ITO 50 nm on the ITO nano dots than the conventional ITO film sample. It is due to the reduced reflectance coming from the surface roughness and is consistent with the result in Fig. 3 that ITO 50 nm has approximately a 10% lower reflectance than ITO 100 nm on GaN. All the measured solar cell parameters are summarized and compared in Table 1. Cell efficiency is highest, 0.91%, in the sample with ITO 50 nm on the ITO nano dots and 21.76% higher than the reference.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>$V_{oc}$ (V)</th>
<th>F.F. (%)</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>Peak EQE (%)</th>
<th>Efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1)-ITO 200 nm (ref.)</td>
<td>2.3</td>
<td>58.2</td>
<td>0.56</td>
<td>36.7</td>
<td>0.74</td>
</tr>
<tr>
<td>(2)-ITO nano dots</td>
<td>2.3</td>
<td>58.5</td>
<td>0.53</td>
<td>32.6</td>
<td>0.71</td>
</tr>
<tr>
<td>(3)-ITO 50 nm on ITO nano dots</td>
<td>2.3</td>
<td>58.1</td>
<td>0.69</td>
<td>41.8</td>
<td>0.91</td>
</tr>
<tr>
<td>(4)-ITO 100 nm on ITO nano dots</td>
<td>2.3</td>
<td>58.1</td>
<td>0.63</td>
<td>38.6</td>
<td>0.82</td>
</tr>
</tbody>
</table>
Figure 5 shows dark current versus voltage ($I-V$) curves of the 4 samples and the inset shows electroluminescence (EL) at 20 mA. We can see that the sample with nano dots only has higher series resistance than the others and this is due to lack of current spreading. This also resulted in highly reduced EL intensity as in the Fig. 5 inset. While the voltage at 20mA was 3.2 V for the ITO dots only sample, all the other samples having ITO films showed almost the same value, 2.9 V. This indicates that the electrical properties such as resistivity were recovered to the same level as the ITO 200 nm after the 2nd deposition of the ITO film. EL spectra intensity at $I = 20$ mA was enhanced by 18% in ITO 100 nm on the ITO nano dots, and 40% in ITO 50 nm on the ITO nano dots compared to the planar ITO 200 nm sample. The EL enhancement comes from the surface roughening effect by the ITO nano dots resulting in better light extraction, and at the same time results in the current spreading being maintained by the 2nd ITO deposition. This result shows that the ITO 50 nm on the ITO nano dots can also act very nicely as a top electrode with better extraction efficiency in LEDs.

![Fig. 5. Dark I-V curve for Solar Cells with (1)-200 nm ITO film only, (2)-ITO nano dots only, (3)-ITO 50 nm on the ITO nano dots and (4)-ITO 100 nm on the ITO nano dots, respectively. The inset shows typical EL spectra at 20 mA.](image)

5. Conclusion

In conclusion, InGaN-based MQW solar cells have been fabricated with 4 different transparent top electrode structures to enhance the power conversion efficiency by reducing reflectance. The solar cell with the ITO 50 nm on the ITO nano dots under the AM 1.5 condition showed the following superior results: 2.3 V for $V_{oc}$, 0.69 A/cm$^2$ for $J_{sc}$, 41.8% for peak EQE, and 0.91% for conversion efficiency. Efficiency improvement was possible due to the decreased reflectance achieved by the ITO nano dots covered with a thickness optimized ITO film. This study indicates that the thickness optimized ITO film on the ITO nano dots can improve the power conversion efficiency without electrical degradations in solar cells.

Acknowledgments

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