Thin Ni film on graphene current spreading layer for GaN-based blue and ultra-violet light-emitting diodes

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We introduced a thin nickel (Ni) film onto graphene as a current spreading layer for GaN-based blue and ultraviolet (UV) light emitting diodes (LEDs). The thin Ni film was confirmed to improve the electrical properties of the graphene by reducing the sheet and contact resistances. The advantages of Ni on graphene were more remarkable in UV LEDs, in which the operation voltage was reduced from 13.2 V for graphene alone to 7.1 V. As a result, UV LEDs with Ni on graphene showed a uniform and reliable light emission, at ~83% of electroluminescence of indium tin oxide.

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revealed. Then, the Ni mask was etched in HCl. For the Ni on GR sample, we deposited 3 nm of Ni onto the graphene. Then, both the GR and Ni on the GR sample were annealed in a rapid thermal annealing (RTA) machine under a N2 ambient at 500°C. Finally, Cr/Au (30 nm/300 nm) was deposited on all samples as p- and n-type electrodes.

We subsequently investigated the optical, morphological, and electrical properties of the GR, Ni on GR, and ITO transparent layers. In Fig. 2(a), GR has over 90% transmittance for the measured wavelength region; Ni on GR has approximately 75% transmittance from 365 nm to 700 nm due to the presence of the 3 nm Ni film; and ITO shows a significant absorption near 380 nm and transmittance fluctuation due to the effects of interference. As a result, the transmittances of Ni on GR and ITO were 74% and 70% at 380 nm and 75% and 86% at 460 nm, respectively. Figure 2(b) presents the sheet resistance of each transparent layer; resistances of 1250 ± 50 Ω/□, 690 ± 50 Ω/□, and 43 ± 5 Ω/□ were obtained for GR, Ni on GR, and ITO, respectively, using a Hall measurement system. Although the thin Ni (3 nm) reduced the transmittance in the Ni on GR, the conductivity was nearly twice that of the GR. Figure 2(c) shows the optical microscope images of the fabricated device having GR (top figures), Ni on GR (bottom figures), and their top surface images measured using scanning electron microscopy (SEM). In the top SEM figure in Fig. 2(c), randomly formed Ni residue that was not completely removed during the Ni mask wet-etching process was observed on the GR surface. The Ni residue was in the shape of separated nano-islands that covered a very small fraction of the surface and did not seem to affect the optical transmittance or sheet resistance of the graphene. However, as shown in the bottom SEM image, the 3 nm deposition of a Ni film on the graphene formed a mesh-like structure, which enabled a higher conductivity than the GR sample. In Fig. 2(d), a Raman spectrum was used to investigate positions “A,” “B,” and “C” to verify whether or not graphene remained after fabrication was completed. Two main peaks of graphene corresponding to the G peak (~1596 cm⁻¹) and the 2D peak (~2705 cm⁻¹) were observed at points “A” and “C,” which are in the GR layer and Ni on GR spreading layer regions, respectively; however, no G and 2D peaks were found at point “B,” which is the n-type GaN region. Here, the broadening of the G and 2D peaks of the GR and Ni on GR regions than immediately transferred graphene on GaN was due to the interaction of graphene with the Ni nano-island residue and thin Ni film. Note that a increased D peak (~1360 cm⁻¹) was seen in both the GR and Ni on GR regions due to defects caused by Ni atoms that were incorporated into the graphene.

Figure 3 shows the I–V characteristics and electroluminescence (EL) intensity of the fabricated blue LEDs under different current spreading layers, including GR, Ni on GR, and ITO. In Fig. 3(a), the forward voltages at a 20 mA current injection were measured to be 3.5 V, 4.8 V, and 6.2 V for blue LEDs with ITO, Ni on GR, and GR transparent.
layers, respectively. The comparatively higher operation voltage and series resistance of GR than for the ITO sample were due to the higher sheet and contact resistances of the graphene on p-GaN than for ITO.\textsuperscript{6,15} However, the deposition of a thin Ni film on graphene reduced the sheet resistance of graphene by almost half. Diffusion of Ni atoms, which have a larger workfunction ($\frac{4.21}{eV}$) than graphene ($\frac{4.1}{eV}$), to graphene/p-GaN during the annealing process further reduced the contact resistance by lowering the Schottky barrier height.\textsuperscript{10,11,18} As a result, by reducing the operation voltage and series resistance, the Ni on GR sample led to a better electrical performance than the GR sample. Among the three samples, the ITO sample displayed twice as strong EL intensity than either the GR or Ni on GR samples. This result is comparable to the performance of blue LEDs having graphene electrodes reported by another group.\textsuperscript{11} Note, however, that although graphene showed over 92% transmittance at 460 nm, it still did not exceed the electrical properties of ITO, such as the sheet and series resistances. In addition, the Ni on GR sample had a nearly identical EL intensity as the GR sample since the electrical properties of the graphene were enhanced by combining it with a thin Ni layer, which compensated for the optical transmission loss.

Characteristics of both the epitaxial layer and graphene should be considered when graphene is to be used as the current spreading layer for LEDs. Epitaxial structures containing a highly resistive p-GaN or p-AlGaN layer further increase the series resistance. And, the resistance of the neutral region, such as AlGaN-based MQWs, could also affect the carrier transport.\textsuperscript{19–21} As a result, having an electrically stable graphene current spreading layer is critical for UV LEDs, which have difficulties in carrier transport, reducing series resistance, and lowering the turn-on voltage (operation power). In Fig. 3(c), the I-V characteristics of UV LEDs with various current spreading layers are seen to follow a similar trend as the blue LEDs. The turn-on voltages and series resistance of UV LEDs increased in the order of ITO, Ni on GR, and GR samples. However, the operation voltages of UV LEDs having the same current spreading layer were generally larger than those of the blue LEDs due to carrier transport problems resulting from the highly resistive $p$-GaN/$p$-Al$_{0.25}$Ga$_{0.75}$N layer. Of particular note, the operation voltage of the UV LEDs with GR at 20 mA current injections was 13.2 V, whereas the Ni on GR reduced the operation voltage to 7.1 V. By applying a thin Ni film onto the graphene, the turn-on voltage and series resistance were rapidly reduced compared to GR—even in UV LEDs—than for blue LEDs. This film application significantly improved the electrical properties, compared to results previously reported by Kim et al.\textsuperscript{12,13} ($\sim$9 V turn-on voltage at only 3 mA current injection). As shown in Fig. 3(d), the EL intensities of 380 nm emissions in UV LEDs were similar for all samples under a 5 mA low current injection, due to the higher transmittances of GR and Ni on GR than ITO near the UV region. In spite of the high transmittance (91%) of GR at 380 nm, its EL intensity was almost the same due to it having the worst electrical characteristics among the three samples. Note, however, that the EL intensity was not measured for the GR sample at a 20 mA current injection since it died after 30 s of operation, due to graphene burning resulting from the high power consumption of the GR sample. At a 20 mA injection, an extreme amount of power accumulates under the $p$-metal pads in the GR sample due to the large forward voltages, and the graphene is burned by joule heat. However, the Ni on GR displayed stable light emission at a 20 mA current injection for over 30 s of operation, even in UV LEDs, and showed 83% of the EL intensity of the ITO sample.

The main point of using Ni on GR can be better observed in Fig. 4, which compares the light emission images of the blue and UV LEDs under three different spreading layers. A bright and uniform light was emitted from the entire area of the top surface for all blue LEDs. While the light emission was uniform in the blue LEDs for all three transparent layers, as shown in the left column of Fig. 4, uniform light emission was only observed in UV LEDs for Ni on GR and ITO. In particular, the UV LED
with GR had light emitting only near the p-metal region due to incomplete current spreading. The more resistive p-GaN/p-Al$_{0.25}$Ga$_{0.75}$N layers of UV LEDs compared to the p-GaN layers of blue LEDs had an additional adverse influence on the current spreading of UV-LEDs with graphene alone.\textsuperscript{21,22} However, the application of a thin Ni film on graphene, which reduced sheet resistance and contact resistance, significantly alleviated the total resistance on the p-GaN/p-Al$_{0.25}$Ga$_{0.75}$N layers—even in UV LEDs—and allowed for uniform current spreading. These results indicate that a reliable and uniform light emission could be confirmed in GaN-based LEDs having a thin Ni film on the graphene current spreading layer. Furthermore, if the transmittance of Ni on GR in the UV region is further improved by optimizing the micro-mesh structure, the Ni on GR could be a more useful current spreading layer for highly resistive p-GaN- or p-AlGaN-containing UV LEDs.

In conclusion, we fabricated blue and UV LEDs having current spreading layers including GR, Ni on GR, and ITO. We demonstrated that not only the transmittance but also the electrical properties of the graphene are important for LED devices. Here, the Ni on GR layer reduced the sheet resistance and diffusion of Ni atoms to graphene/p-GaN, which subsequently reduced the contact resistance such that the I-V characteristics of blue and UV LEDs were much improved compared to those having only a GR current spreading layer. Furthermore, advantages of a thin Ni film on graphene in UV-LED applications included a higher transparency than ITO in the near UV region, significantly reduced operation voltage, and uniform light emission, obtained by alleviating the high resistance incurred due to the very resistive p-GaN/p-AlGaN layer of UV-LEDs. As a result, UV-LEDs having a Ni on GR layer were confirmed to operate well for a long operation time and showed $\sim$83\% of the ITO EL intensity.

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\begin{figure}[h]
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\includegraphics[width=\textwidth]{leds.png}
\caption{Optical images of light emission of blue and UV LEDs under different current spreading layers (GR, Ni on GR, and ITO) at 10 mA and 20 mA current injections.}
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