



Effect of Interfacial Properties of p-GaN/Sputter-Deposited NiAg-Based Electrode on Optical Properties of Vertical GaN-Based LEDs

Sunjung Kim^{*,z}

School of Materials Science and Engineering, University of Ulsan, Ulsan 680-749, Korea

The influence of the interfacial adhesion and reflectance of a p-GaN/NiAgNiAu p-electrode on the optical properties of vertical GaN-based light emitting diodes (LEDs) was investigated. The thickness of the sputter-deposited Ni ohmic contact was varied from 2 to 100 nm. The p-electrode with a 2 nm thick Ni layer showed the highest adhesion strength of 160 MPa to the p-GaN and the highest reflectance of 80.79% after annealing because all the Ni atoms participated in the interdiffusion into p-GaN and formed a transparent NiO. The NiAgNiAu p-electrodes with a higher reflectance led to the improved output power of the vertical LEDs regardless of the interfacial adhesion strength of p-GaN/Ni.

© 2009 The Electrochemical Society. [DOI: 10.1149/1.3238468] All rights reserved.

Manuscript submitted July 1, 2009; revised manuscript received August 20, 2009. Published October 5, 2009.

In contrast to conventionally structured GaN-based light emitting diodes (LEDs) that have laterally positioned n- and p-electrodes on the same side, n- and p-electrodes of vertical structure GaN-based LEDs are oppositely placed on the top and bottom of the LED structure, respectively, so that the current can flow through the active well region having a uniform distribution. The vertical LEDs employ a metal substrate on the p-GaN side, which serves as a heat sink and an electrical connector to the packaging components. The metal substrate, which is usually made of Cu or Cu alloy, also functions as a mechanical supporter of the LED structure after removing a sapphire substrate by a laser lift-off (LLO) method.¹ It is created on the p-GaN side of the vertical LEDs usually by electrodeposition. Light extraction can be effectively improved by putting an Al- or Ag-based reflector in the p-electrode structure.^{2,4} A Ag mirror is preferably used for reflective p-electrodes due to its highest reflectance among metals. However, because Ag itself cannot make an ohmic contact with p-GaN, normally a thin Ni layer is introduced between the p-GaN and the Ag mirror to make an ohmic contact with p-GaN forming a transparent NiO under rapid thermal processing.⁵ The Ni layer also serves as an adhesion layer and a diffusion barrier between p-GaN and Ag. The Ni/Ag scheme is an immiscible system in solid state unless a very high temperature over 700°C is applied.^{6,7} However, the reflecting power of the Ag film decreases by its agglomeration under heat-treatment. Therefore, additional NiAu layers are frequently added to protect Ag from thermal degradation. In this study, NiAgNiAu was used as a p-electrode structure in contact with the p-GaN.

The LLO process delivers a large mechanical stress to vertical LED structures. Buffer GaN is dissociated into Ga and N₂ gas at the buffer GaN/sapphire interface by laser irradiation. The explosive force generated by the rapid volume expansion of the N₂ gas is mostly concentrated on the weakly bonded locations of the vertical LED structure, such as p-GaN/p-electrode interface, p-electrode/metal substrate interface, and the sidewall of GaN-based layers. Among those weak locations, the interfacial properties of the p-GaN/p-electrode exercise a serious influence on the operation voltage and light extraction, and at last the output power of vertical LEDs. Partial delamination of the p-GaN/p-electrode interface by laser hit can lead to a decrease in output power. Furthermore, its full delamination leads to device failure of vertical LEDs.⁸ Figure 1a shows the side view of a chip of vertical GaN-based LED during the LLO process to detach a sapphire wafer from the LED structure. Figure 1b and c is the top-view image of the LED structure by optical microscopy and its side-view image by scanning electron microscopy, respectively, showing partial delamination at the p-GaN/p-electrode interface after the LLO process.

In this study, the interfacial adhesion strength between the p-GaN and the first Ni layer of the NiAgNiAu scheme was quantitatively measured using a pull-off test. The NiAgNiAu multilayer structure was deposited by radio-frequency (rf) magnetron sputtering with the expectation that the interfacial adhesion of the p-GaN/NiAgNiAu could be improved by applying sputter deposition rather than other deposition methods. It has been reported that sputter deposition of a film on a substrate contributes to adhesion enhancement between a film and a substrate.^{9,10} The thickness of the first Ni layer was modulated because it could be closely related to the interfacial adhesion of the p-GaN/NiAgNiAu and the reflecting power of the Ag mirror. After all, the output power of vertical LEDs was measured to investigate its dependence on the Ni layer thickness. The result of the optical measurement was discussed considering its relationship with the interfacial adhesion and reflectance of the NiAgNiAu p-electrode.

Experimental

A 2 in. diameter sapphire substrate was used for the epitaxial growth of GaN-based LED layers, which consisted of a buffer GaN layer, a Si-doped n-GaN layer ($2.5 \times 10^{19} \text{ cm}^{-3}$), InGaN/GaN multiple quantum wells, and a Mg-doped p-GaN layer ($3.5 \times 10^{17} \text{ cm}^{-3}$), by metallorganic chemical vapor deposition. Both sides of the sapphire wafers were polished to be transparent for laser irradiation and reflectance measurement. The chip area of $350 \times 350 \text{ }\mu\text{m}$ was defined by creating trench lines between the chips using inductively coupled plasma reactive ion etching (ICP-RIE). A reflective p-electrode of Ni/Ag (200 nm)/Ni (200 nm)/Au (300 nm) was deposited on p-GaN using rf magnetron sputtering at an rf power of 150 W in an argon environmental chamber. The thickness of the first Ni layer was variable from 2 to 100 nm. The p-GaN/p-electrode contact was annealed at 400°C for 6.5 min in an O₂ condition using a rapid thermal processor (RTP) to form ohmic contact.

For the pull-off adhesion test, blanket sapphire wafers with a multilayer structure of GaN layers and sputter-deposited NiAgNiAu were cut into specimens of $1 \times 1 \text{ cm}$. A stud was attached to the Au surface of the specimen using a commercial epoxy, as shown in Fig. 2a. The curing time of the epoxy was 2 h at 150°C. The stud attached to the wafer specimen was inserted through the hole on the top of a pull-off tester (Quad Group, Romulus) and was held tightly, as shown in Fig. 2b. The load was applied downward to pull down the stud. The reflectance of the RTP-annealed NiAgNiAu contact was measured using a spectrophotometer (Hitachi, U-4001).

Vertical LED chips ($350 \times 350 \text{ }\mu\text{m}$) were fabricated to investigate the variation in the output power according to the first Ni layer thickness. After forming a photoresist (PR, JSR Corp.) passivation structure, a 150 μm thick Cu substrate was electroplated on a continuous seed layer of W/Ni/Au (50/100/400 nm). A KrF excimer laser at 248 nm wavelength was irradiated through the transparent sapphire substrate with an energy density of 0.7 J cm^{-2} for the

* Electrochemical Society Active Member.

^z E-mail: sunjungkim@ulsan.ac.kr

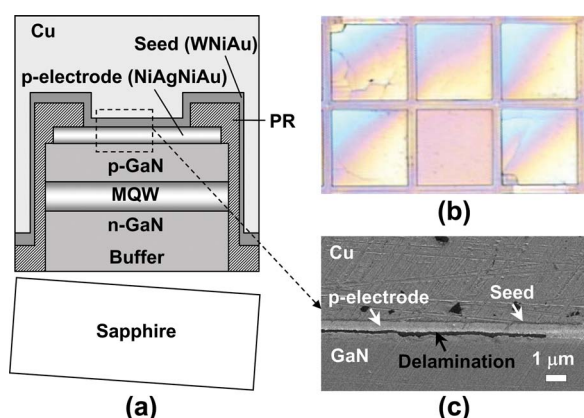


Figure 1. (Color online) (a) Illustration of vertical GaN-based LEDs with NiAgNiAu p-electrode, (b) optical microscope image of vertical LED chips showing partial delamination at the p-GaN/p-electrode interface (top view), and (c) scanning electron microscope image of vertical LED chips showing partial delamination at the p-GaN/p-electrode interface (side view). The chip area was $350 \times 350 \mu\text{m}$.

removal of the sapphire substrate. An n-electrode of Ti/Al/Au (20/20/300 nm) was deposited on n-GaN using rf magnetron sputtering after the n-GaN surface was exposed by ICP-RIE of the buffer GaN layer. Depth profiles of the p-GaN/NiAgNiAu p-electrodes were obtained using an Auger electron spectroscopy (AES, Perkin-Elmer, PHI-4300). A parameter analyzer (HP, 4155A) was used to characterize the current-voltage (I - V) properties. The output power of vertical LEDs was measured from the integrating sphere system using a spectrometer (Optical Precision, OPC-2100).

Results and Discussion

The thickness of the first Ni layer of the NiAgNiAu structure was varied from 2 to 100 nm to measure the interfacial adhesion strength between the p-GaN and the p-electrode using a pull-off tester. All of the specimens were annealed at 400°C for 6.5 min in an O_2 condition using RTP. The annealing process leads to the out-diffusion of Ga atoms toward the p-electrode and, simultaneously, the indiffusion of Ni atoms toward the p-GaN. Ga vacancies are left behind within p-GaN and contribute to the local increase in charge carriers at the p-GaN/Ni for ohmic contact formation. A strong chemical bonding at the p-GaN/Ni interface is created because the Ga and Ni atoms interdiffuse in opposite directions during annealing. The adhesion strength at the annealed p-GaN/Ni interface is quite dependent on the thickness of the Ni layer, as shown in Fig. 3a. As the Ni

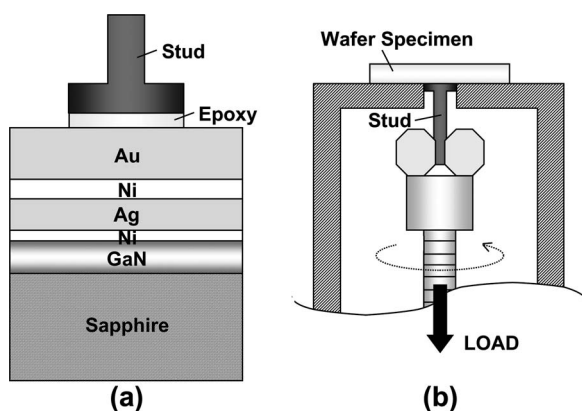


Figure 2. Illustrations of (a) a specimen from the pull-off test to measure the interfacial adhesion strength between p-GaN and NiAgNiAu and (b) a pull-off tester with a specimen mounted.

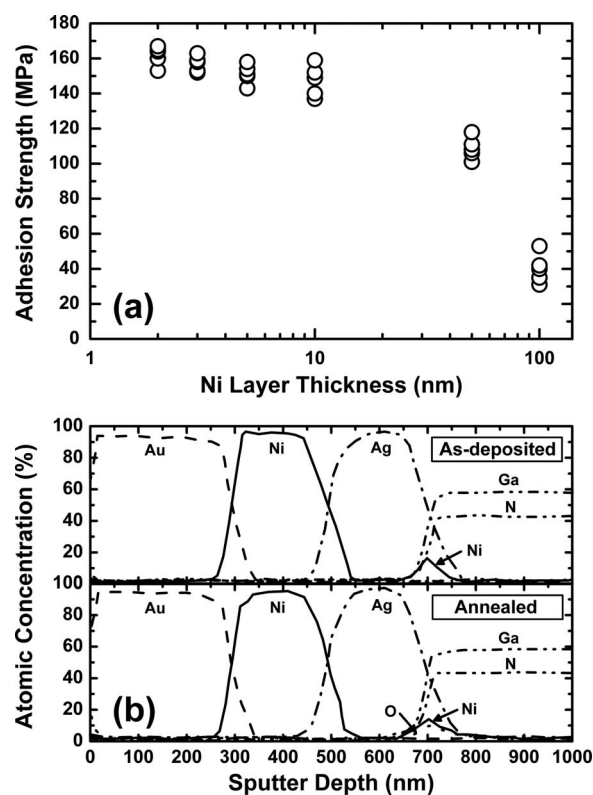


Figure 3. (a) Variation in the adhesion strength of p-GaN/NiAgNiAu interfaces of vertical LEDs as a function of the thickness of the first Ni layer, which was sputter-deposited at an rf power of 150 W. Five measurements were carried out for each Ni layer thickness. (b) AES depth profiles of p-GaN/Ni(10 nm)AgNiAu contact, which were as-deposited and annealed at 400°C for 6.5 min in an O_2 condition using RTP.

layer gets thicker from 2 to 100 nm, the decrease in the interfacial adhesion strength from 160 to 152 MPa, which are the mean values, is not considerable. However, with a 50 and 100 nm thick Ni, a severe reduction in the interfacial adhesion strength to 109 and 37 MPa, respectively, is found. As stated earlier, all of the specimens underwent the RTP process to form ohmic contact. This means that a strong interfacial bonding by interdiffusion of the Ni and Ga atoms exists at the p-GaN/Ni interface regardless of the Ni layer thickness, as shown in Fig. 3b. Figure 3b shows the AES depth profiles indicating the interdiffusion of Ni and Ga and the formation of NiO by RTP annealing. Nevertheless, in Fig. 3a, the decline of the adhesion strength is seen as the Ni layer gets thicker. This is because the amount of Ni atoms attending the indiffusion toward the p-GaN is limited during the RTP process. Ni atoms of the 2 nm thick Ni layer may fully attend the indiffusion and form transparent NiO. For the 100 nm thick Ni layer, only a small portion of Ni atoms takes part in the indiffusion, but most Ni atoms remain within the Ni layer. The RTP process gives rise to a large thermal stress at the p-GaN/Ni interface because the difference in thermal expansion coefficients between the p-GaN and Ni would be rather large compared to their differences between metal layers of NiAgNiAu. Thermal residual stress, which could be stored within the Ni layer, can be released by diffusing Ni atoms into the p-GaN and rearranging them. Thus, a 2 nm thick Ni layer, which has fully transformed into NiO, could minimize the possibility of residual-stress-induced delamination. However, because most Ni atoms of a 100 nm thick Ni layer do not diffuse into p-GaN, it still contains a large residual stress, which is mostly tensile stress that may easily initiate an interfacial crack propagation and finally cause full delamination. In fact, vertical

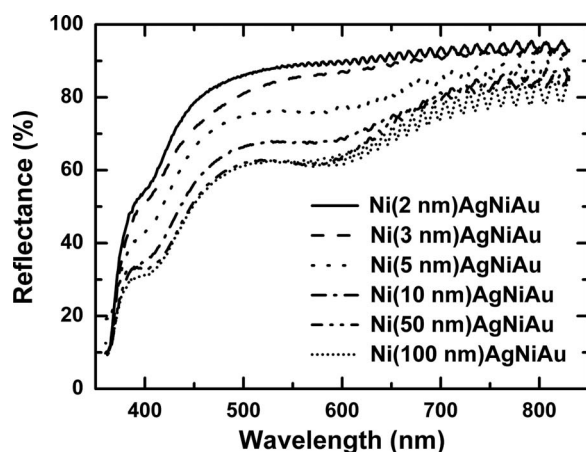


Figure 4. Variation in the reflectance of NiAgNiAu p-electrodes, which were annealed at 400°C for 6.5 min in an O₂ condition using RTP.

LED chips having a 100 nm thick first Ni layer in the p-electrode showed a full delamination during the LLO process, which led to the device failure of the vertical LEDs.

Figure 4 shows the reflectance variation in the annealed NiAgNiAu p-electrodes as a function of the wavelength of the incident light. Light emitted from a halogen lamp was incident through a polished back side of a sapphire wafer and reflected from the p-electrodes. It is clearly shown in Fig. 4 that as the Ni layer gets thicker, reflectance drops significantly. The reflectance of the 2 nm thick Ni/Ag scheme was 80.79% at 460 nm. It is quite high considering a reflective electrode of flip-chip LEDs and vertical LEDs, although it is lower than 96.64% of pure Ag. The reflectances of the 50 nm thick Ni/Ag scheme and the 100 nm thick Ni/Ag scheme were 54.78 and 54.07% at 460 nm, respectively. These values are very close to the reflectance of pure Ni, which is 50.96%. This implies that for a thick first Ni layer, incident light rarely has a chance to reach the Ag mirror; instead, it is reflected from the Ni layer. From this result, the previously suggested issue that only a small portion of the thick Ni layer may participate in the indiffusion to form a transparent NiO, but its most part may remain as a steady Ni layer, can be reconfirmed.

The output power of the vertical LEDs is determined by various factors such as light generation efficiency, device structure, surface morphology for light extraction, reflective electrode, and electrical efficiency, that is, operation voltage. In this study, it has been stated that the interfacial adhesion at the p-GaN/Ni is related to the operation and reliability of the vertical LEDs. Even if a partial delamination occurs at the p-GaN/Ni interface, it leads to an increase in contact resistance; in other words, the operation voltage of the vertical LEDs increases. If the interfacial adhesion at the p-GaN/Ni is not strong enough though no delamination occurs after the LLO process, interfacial cracks can be initiated and propagated under repetitive thermal cycles during device operation. After all, a lack of adhesion in the p-GaN/p-electrode interface gives a negative effect on the output power and operating stability of vertical LEDs. Figure 5a shows the output power variation in vertical LED devices as a function of the applied current according to the thickness of the first Ni layer in the annealed NiAgNiAu. As the Ni layer becomes thinner, the output power of the vertical LEDs is improved. However, it is hard to directly relate this result to the interfacial adhesion strength of p-GaN/Ni because none of the p-GaN/Ni interface with a Ni layer in the thickness range of 2–50 nm showed delamination during the LLO process. It means that the interfacial adhesion is not a factor determining the output power of vertical LEDs, as shown in Fig. 5a. The output power of the vertical LEDs with a 100 nm thick Ni layer in the p-electrode could not be measured because poor interfacial adhesion at the p-GaN/Ni led to interfacial rupture during the LLO process, and thus it was not possible to construct an LED

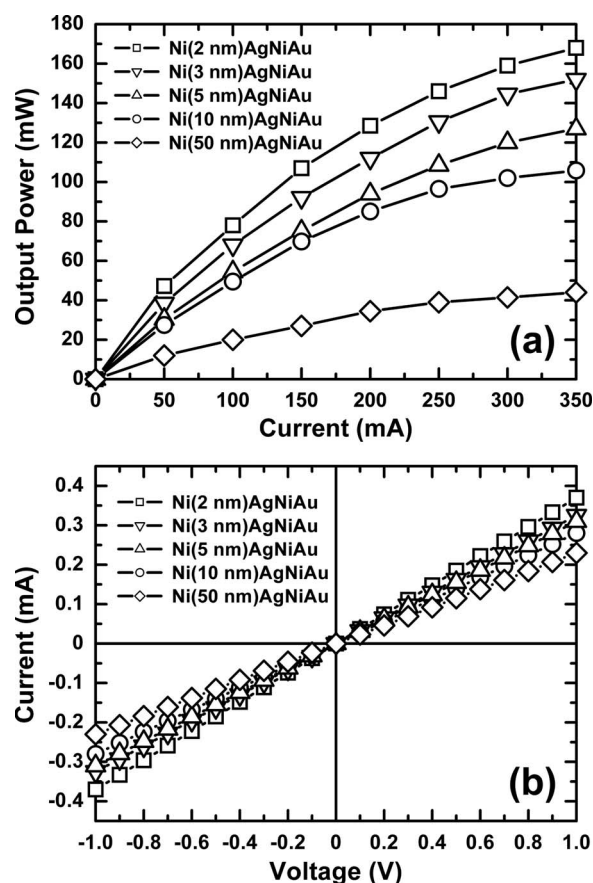


Figure 5. (a) Variation in the output power of vertical GaN-based LED devices with reflective NiAgNiAu p-electrodes, which were annealed at 400°C for 6.5 min in an O₂ condition using RTP and (b) *I*-*V* curves of RTP-annealed p-GaN/NiAgNiAu contacts.

device structure. The main factor that determines the output power variation shown in Fig. 5a is the reflectance of the NiAgNiAu p-electrode. Figure 5b shows the *I*-*V* characteristics of the RTP-annealed NiAgNiAu p-contacts to p-GaN of the vertical LEDs. The *I*-*V* curves of the NiAgNiAu p-contacts with different thicknesses of the first Ni layer indicate a good ohmic behavior with a low specific contact resistance on the order of $10^{-5} \Omega \text{ cm}^2$. Thus, supposing the specific contact resistance of the p-GaN/Ni contact is similar among p-electrodes regardless of the Ni layer thickness, the highly reflective p-electrode such as the 2 nm thick Ni/Ag scheme would lead to a high output power of the vertical LEDs because the light generated from the active well region can penetrate through the transparent NiO and mostly reflect from the Ag mirror. The influence of the rf input power on the interfacial adhesion strength of p-GaN/NiAgNiAu was also studied. As shown in Fig. 6, the improvement of the interfacial adhesion strength with increasing rf power from 150 to 350 W is not significant even though a slight increase in interfacial adhesion strength can be observed. Compared to the first Ni layer thickness, the effect of rf power on the interfacial adhesion strength of the p-GaN/NiAgNiAu is negligible.

Conclusions

The interface between the p-GaN and reflective NiAgNiAu p-electrode of vertical GaN-based LEDs supported by an electroplated metal substrate is important considering the optical performance, mechanical stability, and operation reliability of vertical LEDs. The thickness of the first Ni layer of NiAgNiAu, which was prepared by rf magnetron sputtering deposition, was critical to determine the interfacial adhesion strength of the p-GaN/Ni and the

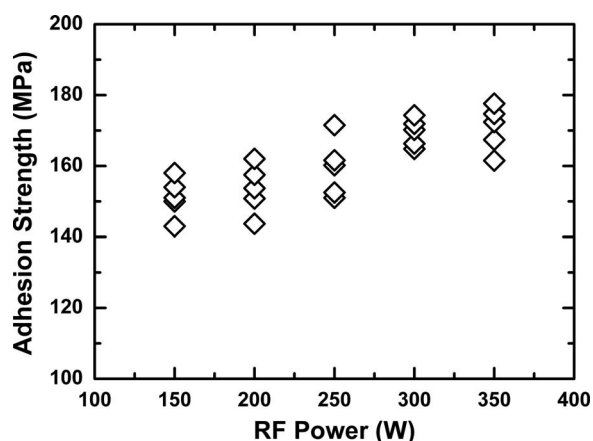


Figure 6. Variation in the adhesion strength of p-GaN/Ni(5 nm)AgNiAu interfaces of vertical LEDs as a function of the rf power of the sputter deposition system. Five measurements were carried out for each rf power.

reflectance of Ni/Ag scheme. Once an ohmic contact is formed at the p-GaN/Ni interface, the interfacial adhesion influences the structural stability and the operation reliability of the vertical LEDs. The reflectance of the Ni/Ag scheme is directly related to the output

power of the vertical LEDs. The vertical LEDs with a NiAgNiAu electrode of higher reflectance showed a higher output power. The rf input power of the sputter deposition did not make a meaningful improvement on the interfacial adhesion strength of p-GaN/NiAgNiAu.

Acknowledgments

This work was supported by the 2008 Research Fund of University of Ulsan.

University of Ulsan assisted in meeting the publication costs of this article.

References

1. W. S. Wong, T. Sands, and N. W. Cheung, *Appl. Phys. Lett.*, **72**, 599 (1998).
2. J.-Y. Kim, S.-I. Na, G.-Y. Ha, M.-K. Kwon, I.-K. Park, J. H. Lim, and S. J. Park, *Appl. Phys. Lett.*, **88**, 043507 (2006).
3. J.-O. Song, W. K. Hong, Y. Park, J. S. Kwak, and T.-Y. Seong, *Appl. Phys. Lett.*, **86**, 133503 (2005).
4. R.-H. Horng, Y.-K. Wang, S.-Y. Huang, and D. S. Wu, *IEEE Photonics Technol. Lett.*, **18**, 457 (2006).
5. W. S. Chen, S. C. Shei, S. J. Chang, Y. K. Su, W. C. Lai, C. H. Kuo, Y. C. Lin, C. S. Chang, T. K. Ko, Y. P. Hsu, et al., *IEEE Trans. Electron Devices*, **53**, 32 (2006).
6. A. Wehr and A. Barcz, *J. Mater. Sci. Lett.*, **12**, 1920 (1993).
7. N. Tanovic, L. Tanovic, and J. Fine, *Vacuum*, **43**, 1177 (1992).
8. S. Kim, D.-K. Bae, J.-H. Choi, J.-H. Jang, and J.-S. Lee, *Electrochem. Solid-State Lett.*, **10**, H334 (2007).
9. S. Abidin, A. Huber, G. Morillot, and C. Val, *Electrocomponent Sci. Technol.*, **7**, 159 (1980).
10. T. Yoneyama, I. Kondo, O. Takenaka, and M. Yamaoka, *Thin Solid Films*, **193-194**, 1056 (1990).