Journal of
Nanoscience and Nanotechnology
Vol. 13, 3696–3699, 2013

# Enhancement of Light Extraction Efficiency of GaN-Based Light-Emitting Diodes by ZnO Nanorods with Different Sizes

Semi Oh[1], Kyung-Sik Shin[2], Sang-Woo Kim[2], Sangbin Lee[3], Hyeongwoo Yu[3], Soohaeng Cho[3], and Kyoung-Kook Kim[1,*]

[1] Department of Nano-Optical Engineering, Korea Polytechnic University, Siheung-Si, Gyeonggi-Do, 429-793, Korea
[2] Department of Advance Materials Science and Engineering, Sungkyunkwan University, Suwon, Gyeonggi-Do, 440-746, Korea
[3] Department of Physics, Yonsei University, Wonju-Si, Gangwon-Do, 220-710, Korea

The improvement of the optical output power of GaN-based light emitting diodes (LEDs) was achieved by employing nano-sized flat-top hexagonal ZnO rods. ZnO nanorods (NRs) with the average diameters of 250, 350, and 580 nm were grown on *p*-GaN top surfaces by a simple wet-chemical method at relatively low temperature (90 °C) to investigate the effect of the diameter of ZnO NRs on the light extraction efficiency. Consequently, the enhancement by the factor of as high as 2.63 in the light output intensity at 20 mA for the LED with 350 nm ZnO NRs was demonstrated without the increase in the operation voltage compared to the reference LED.

**Keywords:** ZnO, Nanorod, Light Extraction Efficiency, Wet Process, Light Emitting Diode.

RESEARCH ARTICLE

## 1. INTRODUCTION

There has been an increased interest in highly efficient light emitting diodes (LEDs) for solid state lightings.[1–6] Various non-planar surface features and fabrication techniques have been proposed to improve light extraction efficiency because the extraction of trapped light is one of the key factors to achieve highly efficient LEDs. Especially, nano-sized surface features have been intensively investigated and have been found to be effective for that matter.[7–11] Among the nano-sized structures applicable to LEDs, the nanorods (NRs) are advantageous compared to other structures as (1) NRs can be easily synthesized by a simple wet process, (2) NRs can be formed on large area surfaces, and (3) the structural dimensions (diameter and length) of NRs can be selectively controlled by the growth parameters.

In view of the previously reported results, the light extraction efficiency of LEDs is seriously influenced by the shape and size of the surface structure. Thus, the research on the dependence of the light extraction efficiency on the NRs' dimension is essential. Nevertheless, most previous researches have been limited to the employment of NRs without the detailed investigations of the relationship between rod's size and light extraction efficiency.[12] Recently, we demonstrated an impressive enhancement in LED's light extraction efficiency by employing randomly sized and distributed ZnO NRs that were grown by a simple wet-chemical growth process at low temperature and it was reported that the electrical property was not deteriorated by a wet-chemical growth technique.[13]

To advance this area of study, we report here the synthesis of ZnO NRs with the controlled diameters on the top surface of LEDs by a wet-chemical growth process at low temperature, and the dependence of light extraction efficiency and electrical property on the shape and size of NRs. A significant enhancement of light output power was demonstrated by the NR-LED with 350 nm ZnO NRs without a noticeable increase in the operating voltage.

## 2. EXPERIMENTAL DETAILS

The LED wafers studied in this work consist of a *p*-GaN (0.25 $\mu$m, $p = 4 \times 10^{17}$ cm$^{-3}$), a five period InGaN multiple-quantum-well (MQW), *n*-GaN (1.5 $\mu$m, $n = 1 \times 10^{18}$ cm$^{-3}$), and undoped GaN/sapphire substrates. Using these wafers, we fabricated InGaN/GaN MQW blue LEDs with conventional planar Indium Tin Oxide (ITO)

*Author to whom correspondence should be addressed.


1533-4880/2013/13/3696/004 doi:10.1166/jnn.2013.7327

transparent *p*-contacts. For an *n*-type contact, we partially etched the surfaces (800 nm deep) of the grown LED samples using an inductively coupled plasma etcher with mixed gas of $Cl_2$ and $BCl_3$ until the *n*-type GaN layer was exposed. Next, a Ti/Au (50/200 nm) layer as the *n*-type electrode was deposited by *e*-beam evaporation. To achieve Ohmic contact of the *p*-GaN layer, the *p*-contact transparent copper indium oxide/ITO electrode (3/400 nm) was deposited onto *p*-GaN surfaces by *e*-beam evaporation and was annealed at 600 °C at $O_2$ ambient for 1 minute by an rapid thermal annealing process. The bonding pad electrode of Cr/Au (50/200 nm) on the top surface of the ITO transparent conducting layer without ZnO NRs was deposited by *e*-beam evaporation. The fabricated conventional LED without ZnO NRs (C-LED) will be used as a reference for comparisons.

The ZnO NRs of various diameters were grown on ITO *p*-electrode with the seed layer deposited surface by a wet-chemical growth method using Zinc nitrate solutions with various molar concentrations. Figure 1 shows the schematics of a wet-chemical growth process to realized ZnO NRs along with the C-LED and NR-LED. In order to form ZnO NRs on top of the *p*-electrode layer, the designed ITO area of the *p*-electrode was made etched away by a lift-off process.

Then, a ZnO seed layer on the *p*-GaN layer was deposited by dipping samples into a solution containing 20 mM zinc acetate $[Zn(C_2H_3O_2)_2]$ dissolved in de-ionized (DI) at a low temperature of 90 °C. In order to achieve seed layers with a high seed density, which is essential to produce low standing angle and size distribution of NRs, we optimized the dipping process. Consequently, we observed reasonably high seed density by repeatedly performing the dipping process for three times each for 30 minutes. Following a seed-layer formation, the samples were pre-annealed for 5 minutes at 100 °C prior to the main growth process. ZnO NR arrays on nano-size ZnO seeds dispersed over the ITO electrode were grown by the "dipping-and holding" process of the substrates into a solution consisting of DI water, zinc nitrate hexahydrate $[Zn(NO_3)_2 \cdot 6H_2O$, 25 mM, 50 mM, and 100 mM] respectively and 25 mM hexamethylenetetramine (HMT) $[C_6H_{12}N_4]$ for 180 min at 90 °C to realize the ZnO NRs with distinct diameters.

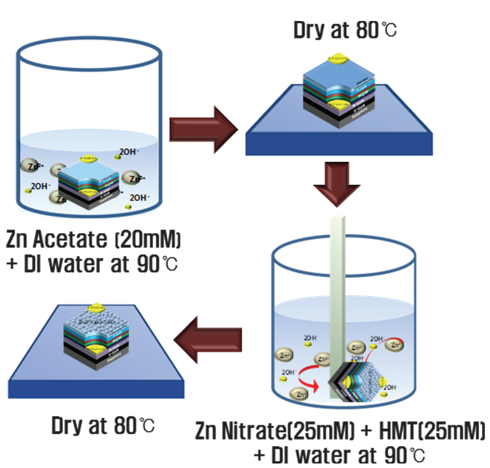


**Fig. 1.** Schematics of a wet-chemical growth method.

Surface morphologies were examined by Field Emission scanning electron microscopy (FE-SEM) (Hitachi, S-4000). Current–voltage ($I$–$V$) measurements were carried out using a parameter analyzer (Keithley-2400 source meter) and light output–current ($L$–$I$) characteristics were measured by a CCD-equipped spectrometer (Ocean optics-USB4000).

## 3. RESULTS AND DISCUSSION

Figure 2 shows the FE-SEM images of ZnO NRs with various diameters. The size distribution of the NRs does not exhibit a Gaussian distribution. The dominant average diameters (the size distributions) of the ZnO NRs of three samples were measured to be 250 nm (146∼337 nm), 350 nm (244∼390 nm), and 580 nm (293∼610 nm), respectively. The size distribution is represented by the diameter range not the standard deviation. The density of ZnO NRs was decreased with increase of diameter of ZnO NR. The diameter of ZnO NRs was controlled with the ratio of mole concentration of Zn nitrate and HMT in a wet-chemical growth process. In this study, the mole concentration ratios of Zn nitrate and HMT solutions are 1:1, 2:1, and 4:1, and the diameter of NRs was observed to increase with the mole fraction of Zn nitrate solution owing to increasing supply of Zn source during ZnO NRs growing process. As mentioned above, this wet-growth method was performed at relatively low temperature (about 90 °C) to suppress thermal damage of *p*-electrode layers, which is also advantageous in terms of fabrication cost.

We investigated the possible effect of the formation and existence of ZnO NRs on the electrical property. Figure 3 shows $I$–$V$ characteristics of the C-LED and NR-LEDs with NRs of various diameters. It was observed that the $I$–$V$ slopes of all samples were almost identical. The operating voltage of NR-LEDs at the injection current of 20 mA ranges from 3.44 V to 3.59 V. Therefore, it can be concluded that the introduction of the ZnO NRs would not result in the significant degradation of the electrical property of LEDs in practice when considering the observed a slight increase in the operation voltage.

Next, the dependence of light extraction efficiency on ZnO NR diameter was investigated. Figure 4 shows the results of light output intensity of NR-LEDs. It was observed that the electroluminescence (EL) intensity from NR-LEDs was generally higher than that of the C-LED in the range of tested injection current. In general, the enhancement of the light extraction efficiency can be qualitatively attributed to the large number of NR sidewalls and a rough surface and edges at which the photons should

RESEARCH ARTICLE

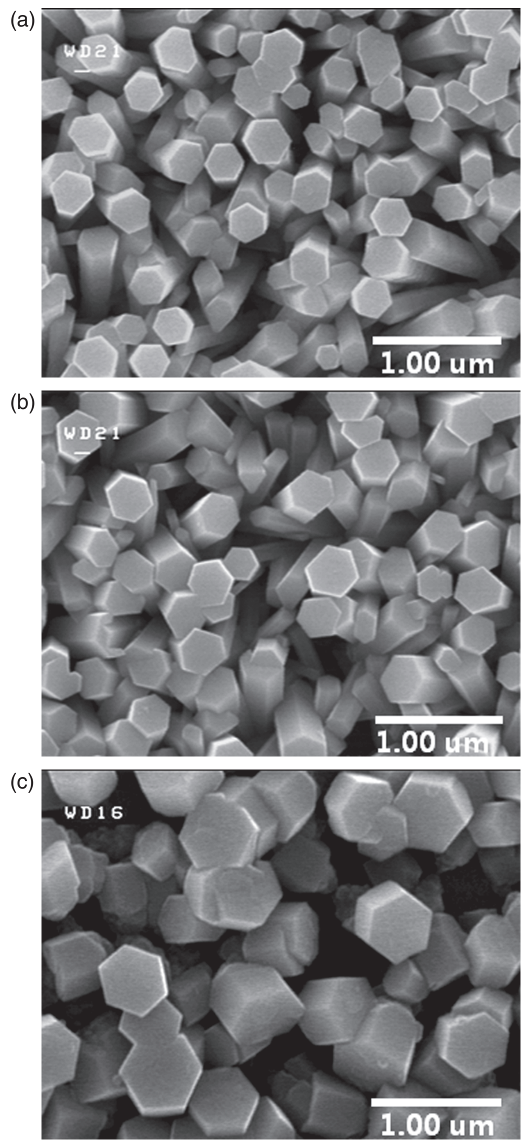


**Fig. 2.** FE-SEM images of ZnO NRs grown on ITO *p*-electrode surfaces. ZnO NRs that have average diameters of about (a) 250 nm (146∼337 nm) (b) 350 nm (244∼390 nm), and (c) 580 nm (293∼610 nm).

experience multiple scattering at the LED surface and can effectively escape from the device.[13] Also, the textured interface between ZnO NRs and the air can also relax the total internal reflection condition and apparently allows extracting more light from the LED chips very effectively.

Accordingly, the NR-LEDs with the average NR diameters of 250 nm and 350 nm exhibited an impressively increased light output power. Especially, the light output of the NR-LED with 350 nm ZnO NRs is about 2.63 times higher than that of the C-LED at an injection current of 20 mA and 100 mA, respectively. In addition, in case of the NR-LED with 250 nm ZnO NRs, EL intensity of the NR-LED at 100 mA was also higher (∼2.52 times) than that of the C-LED. These enhancement factors are much higher than the best values for the LEDs

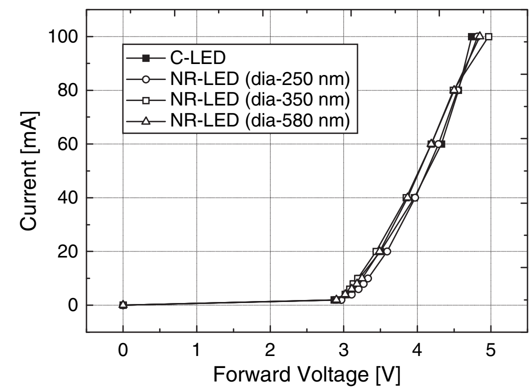


**Fig. 3.** *I–V* characteristics of the C-LED and NR-LEDs.

with the randomly-sized ZnO NRs in our previous work.[13] The comparably higher power-enhancement factors of the NR-LEDs with 250 nm and 350 nm ZnO NRs can be attributed to the higher NR densities than the NR-LED with 580 nm ZnO NRs. Also, it is suspected that the uniformity of the ZnO NR diameter results in the higher light extraction efficiency of the NR-LED with 350 nm NRs compared to the NR-LED with 250 nm NRs.

In contrast, almost no improvement of the light extraction efficiency of the NR-LED with 580 nm NRs can be explained by the energy loss of the energy loss by the intermodal dispersion of ZnO/air wave-guiding effect and also by the smaller total effective surface compared to the LEDs with the narrower NRs.[14, 15] Still, the more detailed simulations with properly measured structural shapes and dimensions should be performed to clearly understand the non-linear dependence of light extraction efficiency on the NR size. This result indicates that the existence of NRs on the LED surface may not guarantee the improvement of light extraction efficiency. In other words, it should be emphasized that the structural dimensions (especially diameter) of ZnO NRs should be carefully designed to optimize optical performance as there would be no monotonically increasing or decreasing tendency of light output power on any structural parameter.

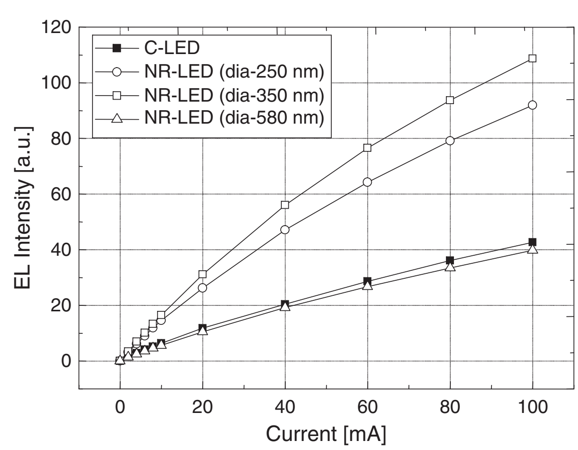


**Fig. 4.** *L–I* characteristics of the C-LED and NR-LEDs.

Nevertheless, the observations of the enhanced light output power of the LED with the size-controlled NRs without any significant degradation in the operation voltage indicate that the ZnO NRs by a simple wet-chemical growth method can be of great use in further development of highly efficiency LEDs.

## 4. CONCLUSION

We investigated the dependence of light output intensity of the LEDs on the diameter of ZnO NRs that were grown on top of the LED surface as a light extracting structure by a low temperature wet-chemical growth method. The optoelectronic properties of the LEDs covered by ZnO NRs with the average diameters of 250 nm, 350 nm, and 580 nm were compared with the reference LED. Among the prepared samples, the LED with 350 nm ZnO NRs demonstrated a 2.63-fold enhancement of light output power at 20 mA compared with the reference LED. No significant increase in the operational voltage was observed for the LEDs with ZnO NRs indicating that no damage on the top *p*-GaN surfaces was induced. These results point out that the existence of ZnO NRs with the controlled size can be an effective solution to increase light extraction efficiency of conventional planar surface LEDs without any deteriorative effect on electrical properties.

**Acknowledgment:** This research was supported by the MKE (The Ministry of Knowledge Economy), Korea, under the ITRC (Information Technology Research Center) support program supervised by the NIPA (National IT Industry Promotion Agency, NIPA-2012-H0301-12-1010).

## References and Notes

1. R. H. Horng, C. C. Yang, J. Y. Wu, S. H. Huang, C. E. Lee, and D. S. Wuu, *Appl. Phys. Lett*. 86, 221101 (**2005**).
2. C. H. Kuo, C. M. Chen, C. W. Kuo, C. J. Tun, C. J. Pan, B. J. Pong, and C. Chi, *Appl. Phys. Lett*. 89, 201104 (**2004**).
3. J. K. Sheu, Y. S. Lu, M.-L. Lee, W. C. Lai, C. H. Kuo, and C.-J. Tun, *Appl. Phys. Lett*. 90, 263511 (**2007**).
4. S. J. Chang, C. F. Sheu, W. S. Chen, C. T. Kuo, T. K. Ko, S. C. Shei, and J. K. Sheu, *Appl. Phys. Lett*. 91, 013504 (**2007**).
5. J. H. Park, H. K. Park, J. Jeong, W. Kim, B. K. Min, and Y. R. Do, *J. Electrochem. Soc.* 158, K131 (**2011**).
6. H. Kim, K.-K. Kim, K.-K. Choi, H. Kim, J.-O. Song, J. Cho, K. H. Baik, C. Sone, Y. Park, and T.-Y. Seong, *Appl. Pyhs. Lett*. 91, 023510 (**2007**).
7. H. Q. Le, S. J. Chua, Y. W. Koh, K. P. Loh, and Z. Chen, *Appl. Phys. Lett*. 87, 101908 (**2005**).
8. M. H. Mamat, Z. Khusaimi, M. M. Zahidi, S. A. Bakar, Y. M. Siran, S. A. M. Rejab, A. J. Asis, S. A. Bakar, Y. M. Siran, S. A. M. Rejab, A. J. Asis, S. Tahiruddin, S. Abdullah, and M. R. Mahomood, *Jpn. J. Appl. Phys*. 50, 06GH04 (**2011**).
9. K. Kim, Y. H. Jeong, C. Kim. Y. Lee, J. Cho, and J. Paik, *Jpn. J. Appl. Phys*. 50, 055003 (**2001**).
10. X. Wang, C. J. Summers, and Z. L. Wang, *Nano Lett*. 4, 423 (**2004**).
11. C.-S. Kee, D.-H. Ko, and J. Lee, *J. Phys. D: Appl. Phys*. 38, 3850 (**2005**).
12. C.-W. Chen, K.-H. Chen, C.-H. Shen, A. Ganguly, and L.-C. Chen, *Appl. Phys. Lett.* 88, 241905 (**2006**).
13. K.-K. Kim, S.-D. Lee, H. Kim, J.-C. Park, S.-N. Lee, Y. Park, S.-J. Park, and S.-W. Kim, *Appl. Phys. Lett*. 94, 071118 (**2009**).
14. K. S. Kim, S. M. Kim, H. Jeong, M. S. Jeong, and G. Y. Jung, *Adv. Funct. Mater*. 20, 1076 (**2010**).
15. R. Olshansky, *Rev. Mod. Phys*. 51, 341 (**1979**).

Received: 27 January 2012. Accepted: 3 June 2012.

RESEARCH ARTICLE