Electrochemical and Solid-State Letters, **14** (2) H66-H68 (2011)
1099-0062/2010/14(2)/H66/3/$28.00 © The Electrochemical Society

Enhanced Light Output of Vertical GaN-Based LEDs with Surface Roughened by SiO_2 Nanotube Arrays

Der-Ming Kuo,[a] Shui-Jinn Wang,[a,b,z] Kai-Ming Uang,[c] Tron-Min Chen,[c] Wei-Chi Lee,[a] and Pei-Ren Wang[a]

[a]Department of Electrical Engineering, Institute of Microelectronics, and [b]Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan
[c]Department of Electrical Engineering, WuFeng University, Chiayi 621, Taiwan

The fabrication of SiO_2 nanotube (SiO_2-NT) arrays and their promising application in improving light extraction of vertical structure GaN-based light emitting diodes (LEDs) are proposed. Compared to regular vertical-conducting light emitting dioded (VLEDs), the proposed VLEDs with SiO_2-NT arrays (2–3 μm in length) show increases in light output power (Lop) by 49.8–60.4% at 350 mA. In comparison to VLEDs with ZnO nanowire arrays having the same the dimension, enhancement of 12.3–22.9% in Lop has been achieved from the proposed devices. These improvements could be attributed to the use of SiO_2-NT arrays that not only boost the angular randomization of emitted photons but also enhance light transmission from the waveguiding effect.
 [DOI: 10.1149/1.3516955]

Manuscript submitted August 2, 2010; revised manuscript received October 26, 2010. Published November 19, 2010.

GaN-based light emitting diodes (LEDs) have recently been attracting great interest in the field of displays, traffic signals, and even solid-state lighting.[1,2] Nevertheless, current-crowding and heat-conducting issues due to the insulating sapphire substrate used in the conventional lateral electrode GaN-based LEDs (abbreviated as LLEDs) are the main challenges in further improving the output efficiency and power rating of LEDs. Efforts to enhance the external quantum efficiency (EQE) of the GaN-based LEDs by means of vertical-conducting structures (abbreviated as VLEDs) have been proposed and shown to have a much better EQE as compared to the conventional LLEDs.[3,4] However, the light output efficiency of VLEDs with a flat surface still suffers a considerable loss caused by the total internal reflection (TIR) effect at the interface between GaN and the outer medium.[5]

To alleviate the TIR effect, various surface-roughening approaches such as nanoparticle lithography,[6] nanoimprint technology,[7] wet-etching process,[8,9] and grown nanostructure atop surface of GaN[10,11] have been proposed to roughen the top surface of LEDs. Hence, photons generated within the LEDs could experience multiple scattering at the roughened GaN/air interface and find escape cones to extract more light from the roughened surface. However, most of these techniques involve expensive lithographic patterning, cumbersome fabrication processes, and control difficulty to avoid deterioration of electrical properties, which are not favorable for mass production.

In essence, the use of ZnO nanopillars[12,13] to roughen the LED surface retains simplicity and cost effectiveness. However, the transmittance of the ZnO nanopillar arrays is not good enough for light transmittance in the visible light spectrum. In our previous study,[14] we reported the preparation of controllable SiO_2 nanotube (SiO_2-NT) arrays through the use of a hydrothermal ZnO nanowires (ZnO-NWs) template. The SiO_2-NT arrays exhibited a superior transmittance of 95% compared with that of the ZnO-NWs template, at 82%, in the visible light spectrum. Here, to further improve the light transmittance and light extraction efficiency of VLEDs, we extend our previous work on applying diverse length and density of the SiO_2-NT arrays for n-GaN surface roughening. The waveguiding effect of the SiO_2-NT and enhancement in the light transmittance of the SiO_2-NT arrays were analyzed and investigated. Comparisons of experimental optoelectronic characteristics among VLEDs with prepared SiO_2-NT and ZnO-NW arrays with the same dimensions were also presented and discussed.

[z] E-mail: sjwang@mail.ncku.edu.tw

Experimental

Figure 1a shows schematically the device structure of the proposed VLED with SiO_2-NT arrays (abbreviated as SiO_2-NT VLED) in this work. The GaN LED epitaxial layers were grown on a sapphire substrate in sequence by low-pressure metal organic chemical vapor deposition. For device fabrication, a 5-nm-thick Pt layer was deposited by an electron-beam (E-beam) evaporation onto the p-GaN layer, followed by furnace thermal annealing in N_2 ambient at 525°C (abbreviated as annealed-Pt). After the removal of possible surface oxides, a patterned Al/Pt metal system with a size of 980 μm × 980 μm was deposited to form a highly reflective annealed-Pt/Al/Pt ohmic contact on the p-GaN layer. It was then covered by an adhesive and seed layer comprising of a Cr/Ti/Au metal system using the E-beam evaporation. An 80 μm-thick electroplated nickel layer served as a metal substrate for the LED epitaxial structure after the removal of the sapphire substrate using a patterned laser lift-off (LLO) process.[15] The patterned LLO process was conducted using a quartz mask to define both the size and shape of the KrF excimer laser beam (248 nm) at a reactive energy of 850 mJ/cm^2.

After being dipped in a 6 mol KOH solution at 60°C for 90 s to roughen the n-GaN surface with hexagonal cones and further improve its contact properties, the samples were then placed into a mixed solution of 0.07 M $Zn(NO_3)_2 \cdot 6H_2O$ and 0.09 M $C_6H_{12}N_4$ at 90°C to grow the ZnO-NW arrays with three different hydrothermal growth (HTG) times (60, 80, and 100 min). After that a 1-μm-thick SiO_2 layer was deposited onto the surface of the ZnO-NWs arrays using plasma enhance chemical vapor deposition. To unveil the top portion of the ZnO-NWs for synthesizing the SiO_2-NT arrays, the samples were loaded into an inductively coupled plasma chamber to go through a SiO_2 dry-etching process. After the residual ZnO-NWs removed completely, the vertically aligned SiO_2-NT arrays with length and density determined by the HTG time and tube wall thickness determined by the deposition of SiO_2 layer were obtained. Finally, a metal pad and a passivation layer were deposited on the exposed n-GaN layer surface and vertical sidewalls of the whole epitaxial structure, respectively. For comparison, two other types of samples were also prepared as shown in Fig. 1b, including VLEDs with the ZnO-NW arrays atop the KOH-etched n-GaN surface (named as ZnO-NW VLED) and VLEDs with no nanostructures atop the KOH-etched n-GaN surface (named as regular VLED). Figure 1c shows the scanning electron microscopy (SEM) images of the surface morphology of the prepared VLEDs. Note that the HTG time for the SiO_2-NT and ZnO-NW VLEDs is 60 min. It is seen that all the surfaces of the top GaN layer of these three VLEDs were roughened with hexagonal cones.

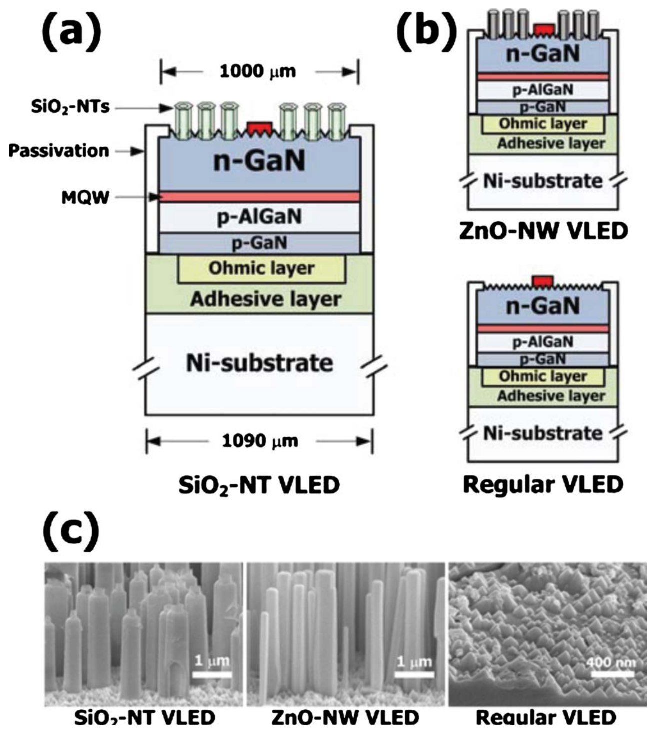

Figure 1. (Color online) Schematic diagrams of GaN-based VLED structures prepared in this work. (a) SiO_2-NT VLED, (b) ZnO-NW VLED and regular VLED, and (c) SEM images of the surface morphology of the prepared VLEDs. The HTG time for the SiO_2-NT and ZnO-NW VLEDs is 60 min. Note that all devices are with the same chip size of 1 mm × 1 mm.

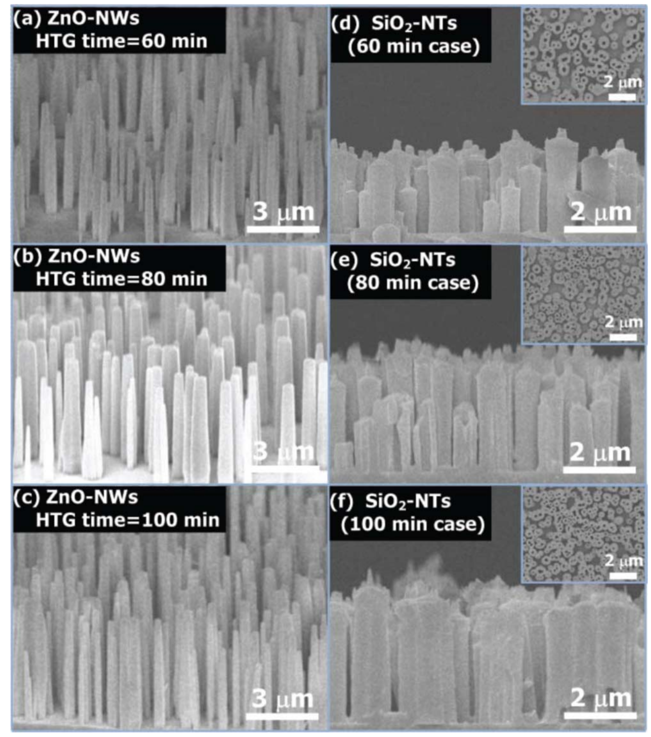

Figure 2. (Color online) SEM images of synthesized ZnO-NW and SiO_2-NT arrays on ITO glass substrate for transmittance measurement. As-grown ZnO-NWs via (a) 60, (b) 80, and (c) 100 min of HTG process. (d)–(f) Side-view and top-view (inset) images of the prepared SiO_2-NT arrays using the corresponding ZnO-NWs templates shown in (a)–(c).

Results and Discussion

Figure 2 shows the SEM images of surface morphology of the samples with ZnO-NW arrays or SiO_2-NT arrays grown for different HTG times atop indium tin oxide (ITO) glass substrates. As shown in Figs. 2a through 2c, it can be seen that the well-ordered and vertically aligned ZnO-NWs were grown on the GaN layer. For the cases with 60, 80, and 100 min HTG times, the grown ZnO-NWs are of an average length of about 3, 3.5, and 4 μm, respectively. According to Figs. 2d through 2f, the average length of the prepared SiO_2-NTs was estimated to be about 2, 2.5, and 3 μm, respectively. Note that the 100-min case of the SiO_2-NT arrays has the best uniformity in tube length among all the prepared samples. The insets of Figs. 2d through 2f show the top-view of the prepared SiO_2-NTs, which reveal that the densities of the SiO_2-NT arrays were about 5.8×10^8, 6.9×10^8, and 7.2×10^8 cm^{-2}, respectively, indicating a weak HTG time dependence. Note that after the removal of ZnO-NWs, the SiO_2-NTs have a uniform tube wall thickness of about 150 nm. These results suggest that SiO_2-NTs with controllable length and inner/outer diameter could be easily obtained through a suitable control of HTG of ZnO-NWs and deposition of SiO_2 layer.

Light transmittances of the SiO_2-NT and ZnO-NW arrays (with three different lengths of 2, 2.5, and 3 μm) prepared on the ITO glass substrate were compared and shown in Fig. 3. It is noted that the ZnO-NW arrays shown in Fig. 2 were subjected to an etching process to shorten the wire length for fair comparisons. One can observe that the transmittances of SiO_2-NT and ZnO-NW arrays are about 95–96% and 71–82% within the visible light spectrum range of 400–700 nm, respectively. The excellent transmittance of the SiO_2-NT arrays could be mainly attributed to their relatively high energy bandgap[16] (∼ 9 eV) as compared to that of ZnO (Ref. 17) (3.3–3.7 eV) and the waveguiding effect associated with the SiO_2-NT arrays. In essential, the incident light at the n-GaN/SiO_2-NT interface will travel in the SiO_2 tubular wall (150 nm in thickness) in single-mode with virtually no loss because the SiO_2-NT creates a step-index waveguide,[18] as shown in the inset of Fig. 3. As a result, it leads to a much better transmission as compared to ZnO-NW.

To reveal the effectiveness of the SiO_2-NT arrays in enhancing the light extraction efficiency, comparisons of current–voltage (*I–V*) and light output power–(Lop–) current (*L–I*) characteristics for the fabricated VLEDs with different surface texturing designs are shown

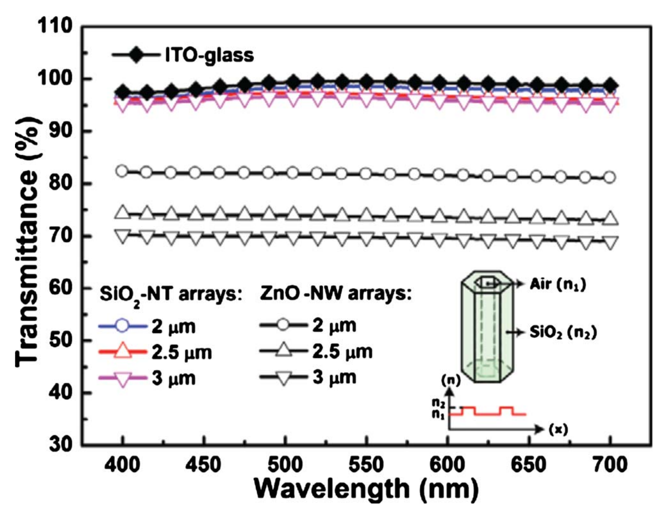

Figure 3. (Color online) The transmittances of the prepared SiO_2-NT and ZnO-NW arrays on ITO glass substrate. The inset shows the schematic of a single SiO_2 nanotube exhibiting as an air-clad step-index waveguide.

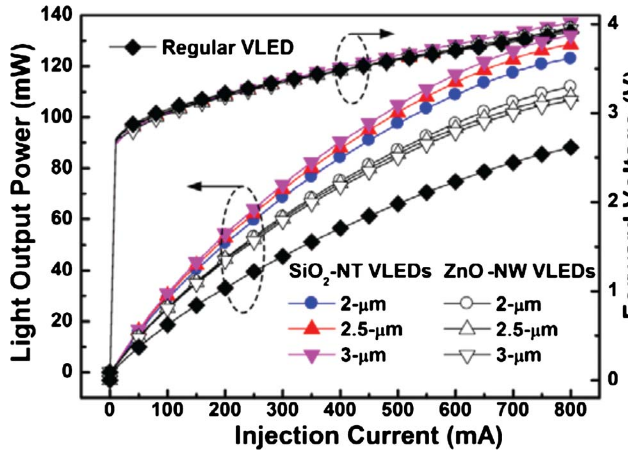

Figure 4. (Color online) Comparison of measured forward *I–V* and *L–I* characteristics among the fabricated SiO_2-NT VLEDs, ZnO-NW VLEDs, and regular VLED. Note that both the SiO_2-NTs and ZnO-NWs have three different tube/wire lengths of 2, 2.5, and 3 μm.

in Fig. 4. Here, both SiO_2-NT and ZnO-NW VLEDs have three different tube/wire lengths of 2, 2.5, and 3 μm. No obvious difference was observed from their *I–V* characteristics, which indicate that the proposed technology did not degrade the device electrical performance. Based on the *L–I* characteristics, the effectiveness of the SiO_2-NT arrays in releasing the TIR effect can be reflected by the amounts of Lop enhancement as compared to that of regular VLEDs. It is found that VLEDs with 2, 2.5, and 3 μm in length of SiO_2-NT arrays show gains of 49.8, 56.7, and 60.4% in Lop at 350 mA, respectively, as compared to regular VLEDs. Also note that the measured values of refractive index for the 2, 2.5, and 3 μm in length of SiO_2-NT arrays are 1.45, 1.468, and 1.48, respectively, indicating that the refractive index of the SiO_2-NT arrays increases slightly with increasing tube length/density. Based on the reason, the SiO_2-NT VLED with 3 μm tube length has the most significant amount of improvement in Lop, which could be attributed to the fact that it has a better refractive index (n) of 1.48 to maximize the alleviation of the TIR issue. In our experience, though the SiO_2-NT arrays with an average length greater than 3 μm has a higher refractive index, it is not suitable for Lop improvement because of poor transmittance.

Note that the improvement in Lop of the SiO_2-NT VLEDs as compared to that of the ZnO-NW VLEDs with 2, 2.5, and 3 μm tube/wire length are 12.3, 18.7, and 22.9%, respectively. Such improvements are in good agreement with the gains (14, 20.5, and 24%, respectively) obtained from the transmittance of the SiO_2-NT arrays compared to those of the ZnO-NW arrays. This result confirms that the predominance of the SiO_2-NT VLEDs over the ZnO-NW VLEDs in light output power could be attributed to the considerable improvement in the light transmittance of the proposed SiO_2-NT arrays.

Conclusions

In summary, the effectiveness of preparing size-controllable SiO_2-NT arrays atop VLEDs to improve Lop has been investigated. With the tube length in the range of 2–3 μm, considerable improvements in the Lop of SiO_2-NT VLEDs by 49.8–60.4% at 350 mA as compared to that of regular VLEDs have been obtained, which reveal the effectiveness of the SiO_2-NT arrays in enhancing the light extraction on releasing the TIR effect at the n-GaN surface. In addition, the wider energy bandgap and waveguiding effect of the SiO_2-NT arrays exhibit excellent transmittances of 95–96% in the visible light spectrum as compared to 71–82% of the ZnO-NW arrays with the same dimensions. These result in the predominance of the SiO_2-NT VLEDs in enhancing the light extraction by 12.3–22.9% over the ZnO-NW VLEDs. It is expected that the SiO_2-NTs prepared with the method proposed in this work could offer potential applications in electronics and photonics in the near future.

Acknowledgment

This work was supported by the National Science Council (NSC) of Taiwan, under grants NSC-97-2221-E-274-013 and NSC-99-2221-E-006-150.

National Cheng Kung University assisted in meeting the publication costs of this article.

References

1. Y. C. Lin, S. J. Chang, Y. K. Su, C. S. Chang, S. C. Shei, J. C. Ke, H. M. Lo, S. C. Chen, and C. W. Kuo, *Solid-State Electron.*, **47**, 1565 (2003).
2. H. X. Jiang, S. X. Jin, J. Li, J. Shakya, and J. Y. Lin, *Appl. Phys. Lett.*, **78**, 1303 (2001).
3. S. J. Wang, K. M. Uang, S. L. Chen, Y. C. Yang, S. C. Chang, T. M. Chen, C. H. Chen, and B. W. Liou, *Appl. Phys. Lett.*, **87**, 011111 (2005).
4. J. S. Ha, S. W. Lee, H. J. Lee, H. J. Lee, S. H. Lee, H. Goto, T. Kato, K. Fujii, M. W. Cho, and T. Yao, *IEEE Photon. Technol. Lett.*, **20**, 175 (2008).
5. C. H. Liu, R. W. Chuang, S. J. Chang, Y. K. Su, L. W. Wu, and C. C. Lin, *Mater. Sci. Eng., B*, **112**, 10 (2004).
6. M. A. Tsai, P. Yu, C. L. Chao, C. H. Chiu, H. C. Kuo, S. H. Lin, J. J. Huang, T. C. Lu, and S. C. Wang, *IEEE Photon. Technol. Lett.*, **21**, 257 (2009).
7. S. J. Chang, C. F. Shen, W. S. Chen, C. T. Kuo, T. K. Ko, S. C. Shei, and J. K. Sheu, *Appl. Phys. Lett.*, **91**, 013504 (2007).
8. S. L. Qi, Z. Z. Chen, H. Fang, Y. J. Sun, L. W. Sang, X. L. Yang, L. B. Zhao, P. F. Tian, J. J. Deng, Y. B. Tao, et al., *Appl. Phys. Lett.*, **95**, 071114 (2009).
9. C. F. Lin, C. M. Lin, K. T. Chen, W. C. Huang, M. S. Lin, J. J. Dai, R. H. Jiang, Y. C. Huang, and C. Y. Chang, *Appl. Phys. Lett.*, **95**, 201102 (2009).
10. W. C. Lai, P. H. Chen, L. C. Chang, C. H. Kuo, J. K. Sheu, C. J. Tun, and S. C. Shei, *IEEE Photon. Technol. Lett.*, **21**, 1293 (2009).
11. C. H. Chiu, P. Yu, C. H. Chang, C. S. Yang, M. H. Hsu, H. C. Kuo, and M. A. Tsai, *Opt. Express*, **17**, 21250 (2009).
12. S. J. An, J. H. Chae, G. C. Yi, and G. H. Park, *Appl. Phys. Lett.*, **92**, 121108 (2008).
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. D. M. Kuo, S. J. Wang, K. M. Uang, T. M. Chen, W. C. Tsai, W. I. Hsu, W. C. Lee, P. R. Wang, and C. R. Tseng, *Jpn. J. Appl. Phys.*, **49**, 04DN10 (2010).
15. S. L. Chen, S. J. Wang, K. M. Uang, T. M. Chen, W. C. Lee, and B. W. Liou, *IEEE Photon. Technol. Lett.*, **19**, 351 (2007).
16. F. Gracia, F. Yubero, J. P. Holgado, J. P. Espinos, A. R. Elipe, and T. Girardeau, *Thin Solid Films*, **500**, 19 (2006).
17. Q. Li, V. Kumar, Y. Li, H. Zhang, T. J. Marks, and P. H. Chang, *Chem. Mater.*, **17**, 1001 (2005).
18. L. Tong, J. Lou, and E. Mazur, *Opt. Express*, **12**, 1025 (2004).