

Study of GaN-Based LEDs With Hybrid SiO₂ Microsphere/Nanosphere AntiReflection Coating as a Passivation Layer by a Rapid Convection Deposition

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Abstract—A hybrid SiO₂ micro/nanospheres antireflection coating, deposited by a rapid convection deposition, acting as a passivation layer of GaN-based light-emitting diodes (LEDs) is studied in this paper. Since the critical angle could be enlarged by antireflection coating, Fresnel reflection could be reduced. In addition, due to the roughened surface of hybrid SiO₂ microsphere/nanosphere antireflection coating, the scattering effect could be increased. Thus, the light extraction efficiency could be further enhanced. As compared with a conventional LED (device A), at 20 mA, the studied device C exhibits 18.7% enhancement in light output power without any degradation of electrical properties. Reduced leakage current could also be achieved. Therefore, the use of hybrid SiO₂ microsphere/nanosphere antireflection coating could effectively improve the performance of GaN-based LEDs.

Index Terms—Antireflection coating, GaN, light-emitting diode (LED), microsphere/nanosphere, rapid convection deposition (RCD).

I. INTRODUCTION

II–V compound materials have been considered as potential candidates for the application in optoelectronic semiconductor devices [1]–[4]. Due to inherent advantages, GaN-based light-emitting diodes (LEDs) have attracted considerable research attention. As a promising light source, GaN-based LEDs have been extensively applied in many applications [5]. However, there is still room for improvement of external quantum efficiency (EQE) of GaN-based LEDs to apply in

solid-state lighting [6], [7]. Due to the difference in refractive indices between GaN and air, light extraction efficiency (LEE) is limited by total internal reflection [8].

It is known that antireflection coating is an effective method for enhancing LEE [9]. Since antireflection coating could enlarge the critical angle, Fresnel reflection could be reduced [10]. This leads to an improvement of an LEE. The ideal refractive index n_{AR} of the antireflection coating could be calculated by $n_{AR} = (n_s \times n_{air})^{1/2}$, where n_s and n_{air} are refractive indices of semiconductor and air [10]. For GaN-based LEDs, refractive index of antireflection coating should be around 1.58. Thus, SiO₂ ($n = 1.5$) is known as a suitable material. In addition, due to insulating property and high transmittance (up to 90%), an SiO₂ layer is always deposited on the surface of GaN-based LEDs as a passivation layer to protect device and reduce leakage current [11]. In addition, it is known that the roughened surface is also a common method to enhance an LEE by increasing the scattering effect [12]. Previously, a self-assembled SiO₂ nanospheres nanostructure with a close packing arrangement was commonly used to achieve a nominal continuous variation of filling fraction and form the photonic crystal-like structure. This could enhance light scattering effect or reflect photons as a photonic band gap [13], [14]. There are many approaches to obtain a self-assembled SiO₂ nanospheres nanostructure with a close packing arrangement [13], [14]. Rapid convective deposition (RCD) is one of the effective methods to achieve a self-assembled monolayer [15]–[17]. Yet, there is still less study on a roughened antireflection coating or a passivation layer. In this paper, a new hybrid SiO₂ microsphere/nanosphere antireflection coating is deposited on the surface of GaN-based LEDs as a passivation layer by an RCD approach [10]. Experimentally, improved optical performance could be obtained without the degradation of electrical properties.

II. EXPERIMENTS

In this paper, the studied device (denoted as device C) was grown on a c-plane sapphire substrate by a metal organic chemical vapor deposition system. The epitaxial structures consisted of a 2- μm -thick undoped GaN layer, a 2- μm -thick Si-doped n-GaN layer ($n = 1 \times 10^{18} \text{ cm}^{-3}$),

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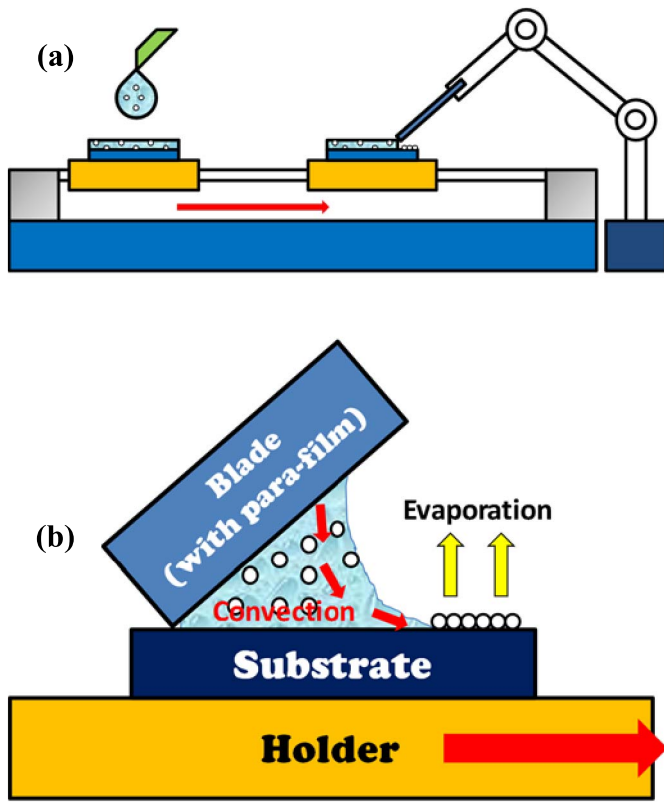


Fig. 1. Schematic of (a) used RCD approach and (b) formation of microscale and nanoscale spheres and particles.

and 15-period InGaN/GaN multiple quantum well as active layers, followed by a 0.3- μm -thick Mg-doped p-GaN layer ($p = 4 \times 10^{17} \text{ cm}^{-3}$). After epitaxial growth, an inductively coupled plasma dry etching process was utilized to define mesa regions. A 70-nm-thick indium-tin-oxide layer was deposited as a current spreading layer and Cr/Pt/Au metals were sequentially deposited as n-p pads by an electron beam evaporator. Then, n-p pads were activated in a nitrogen ambience at 385 °C for 20 min to improve contact characteristics. To form a more uniform hybrid SiO₂ microsphere/nanosphere antireflection coating, hybrid SiO₂ microsphere aqueous suspension was deposited by an RCD approach [13]. Fig. 1(a) shows the used RCD instrument. The suspension was injected by a pipette between the substrate and the glass-based blade. The small droplet of suspension could be controlled by the capillary force between the blade and the substrate wedge. As the substrate was pulled away from the bulk suspension, the meniscus would be stretched out and swept through the substrate, as shown in Fig. 1(b). Spheres from the bulk suspension moved toward the contact line by the liquid flow driven by evaporation and convective flow from the moving substrate [16], [17]. The blade angle was fixed at 45° and the speed of holder was 55 $\mu\text{m/s}$. The hybrid SiO₂ microsphere/nanosphere aqueous suspension was obtained with the composition of 1- μm SiO₂ microspheres : SiO₂ nanoparticles (<30 nm) : sodium dodecyl sulfate (SDS) : ethanol (Eth.) = 1 : 10 : 10 : 10. For comparison, only SiO₂ nanoparticles antireflection coating (SiO₂ nanoparticles (<30 nm) : SDS : Eth. = 10 : 10 : 10) was employed in another LED and denoted as the device B.

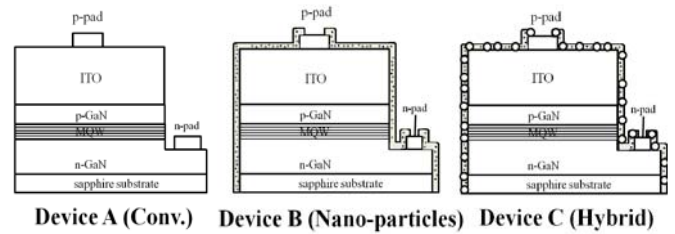


Fig. 2. Schematic cross section of the studied devices.

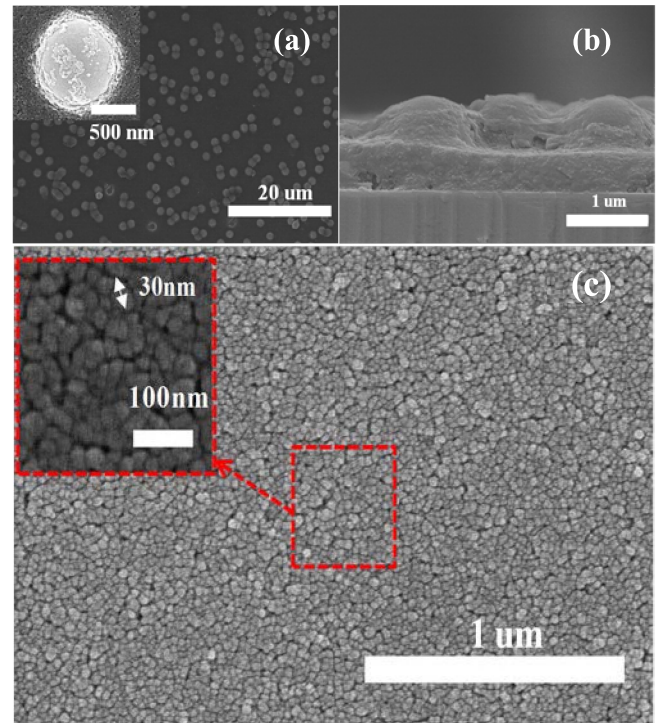


Fig. 3. SEM images on (a) top view and (b) cross-sectional view of hybrid SiO₂ microsphere/nanosphere surface (device C). (c) SEM image on top view of SiO₂ nanoparticles surface (device B).

In addition, a conventional LED without a passivation layer was fabricated and denoted as the device A. To limit the process deviation, all studied devices came from the same uniform LED wafer. All of these samples were diced into individual chips with a dimension of 600 \times 700 μm^2 . Fig. 2 shows schematics of studied devices A, B, and C. These chips were attached and bonded to TO-3 submounts for electrical and optical tests by a semiconductor parameter analyzer (HP-4155C) and an integrated sphere.

III. RESULTS AND DISCUSSION

Fig. 3(a) shows scanning electron microscope (SEM) image on the top view of hybrid SiO₂ microsphere/nanosphere antireflection (device C). A magnified view is shown in the inset. The corresponding cross-sectional view is shown in Fig. 3(b). 1 μm -SiO₂ microsphere surfaces would be covered by SiO₂ nanoparticles (<30 nm). The SEM image on top view of SiO₂ nanoparticles (<30 nm) antireflection coating employed (device B) is shown in Fig. 3(c). A magnified view is shown in the inset. Atomic force microscopy (AFM) images of hybrid SiO₂ microsphere/nanosphere (device C) and nanoparticles antireflection coatings (device B) are shown

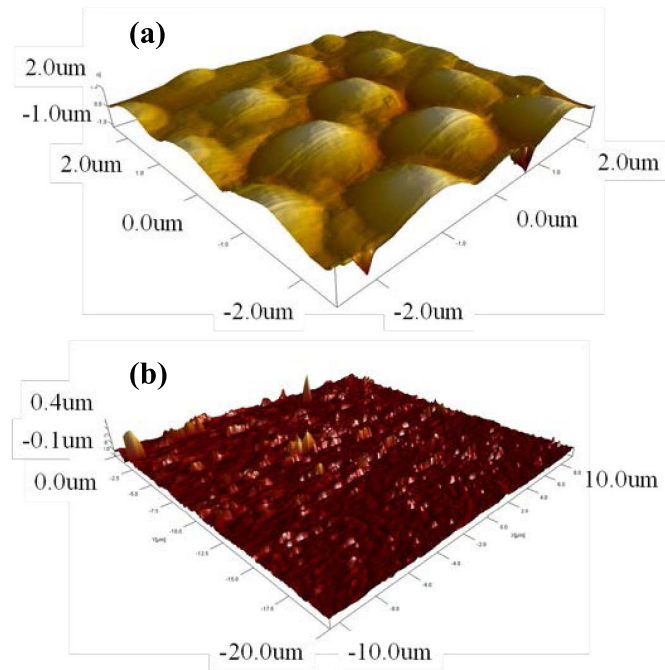


Fig. 4. AFM images of (a) hybrid SiO₂ microsphere/nanosphere surface (device C) and (b) SiO₂ nanoparticles surface (device B).

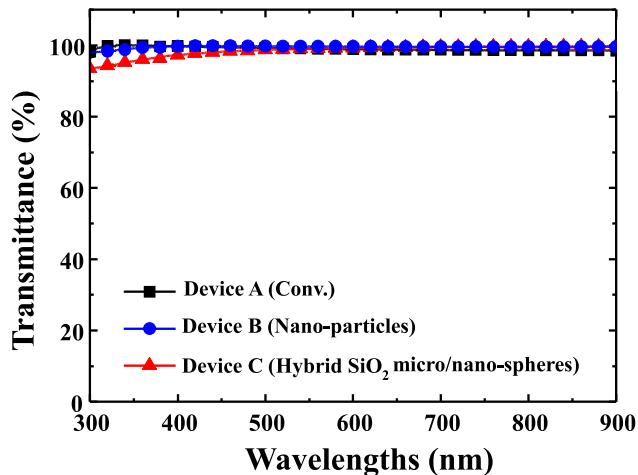


Fig. 5. Transmittances of hybrid SiO₂ microsphere/nanosphere, SiO₂ nanoparticles, and an RF sputtered SiO₂ thin film antireflection coatings.

in Fig. 4(a) and (b), respectively. The root-mean-square value of roughness Rq in an AFM analysis of hybrid SiO₂ microsphere/nanosphere (nanoparticles) antireflection coating is 164 (23.7) nm. These results indicate that the hybrid SiO₂ microsphere/nanosphere (nanoparticles) antireflection coating could be deposited in a large area with a uniform arrangement by using the RCD approach. Moreover, the roughened surface of hybrid SiO₂ microsphere/nanosphere antireflection coating means that photons have more opportunities to be scattered and redirected [18]. Fig. 5 shows transmittances of hybrid SiO₂ microsphere/nanosphere, nanoparticles (<30 nm) antireflection coatings, and a sputtered SiO₂ thin film. Wavelengths of emitted photons are ranged from 300 to 900 nm. Due to the high transmittances up to 97.5% of all samples in blue light region, photons emitted within devices could be

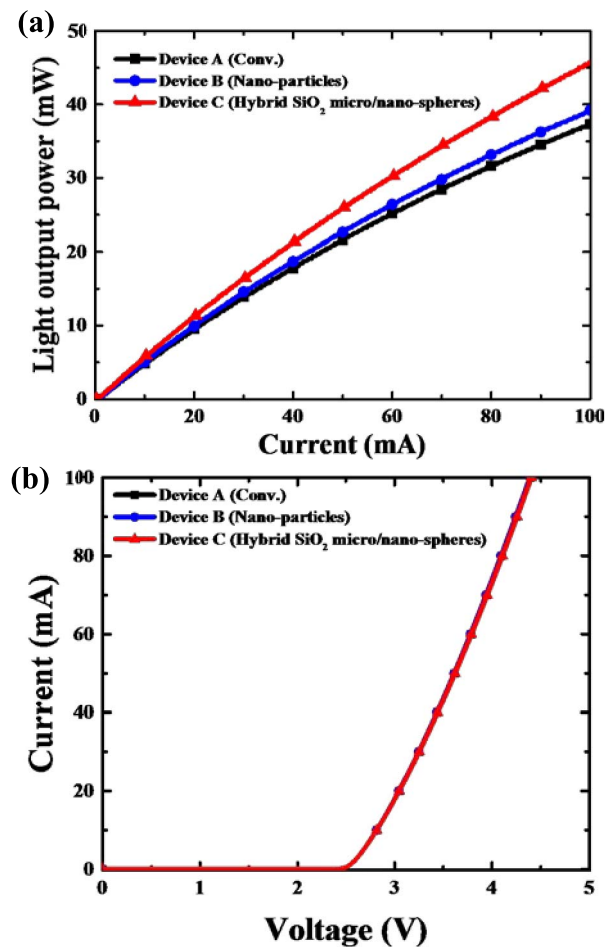


Fig. 6. (a) LOPs. (b) Current-voltage ($I-V$) characteristics of studied devices.

mostly transmit through hybrid SiO₂ microsphere/nanosphere and nanoparticles antireflection coatings.

Fig. 6(a) and (b) shows light output powers (LOPs) and current-voltage ($I-V$) curves of devices A, B, and C, respectively. At 20 mA, similar forward voltages of all studied devices are around 3.05 V. Because the used antireflection coatings are not located in the current pathway, the related electrical properties are not influenced. At 20 mA, LOPs of devices A, B, and C are 9.50, 9.91, and 11.28 mW, respectively. As compared with the device A, devices B and C exhibit 4.3% and 18.7% improvements in LOPs. Since the critical angle could be enlarged by antireflection coating, Fresnel reflection could be reduced [19]. In addition, due to the roughened surface of hybrid SiO₂ microsphere/nanosphere antireflection coating, the scattering effect could also be increased [20]. Thus, LEE could be further enhanced. Fig. 7 shows the reverse-biased $I-V$ curves of studied devices. Under -5 V, reverse-biased leakage currents of devices A, B, and C are 4, 2, and 2 nA, respectively. The reduced leakage currents could be attributed to higher barrier gap and insulation properties of SiO₂ passivation layer [19].

The EQE and luminous flux as a function of operating current are shown in Fig. 8(a) and (b). Highest luminous efficacy is obtained for a monochromatic emission at 555 nm, at which human eye is most sensitive [21]. Since high-power

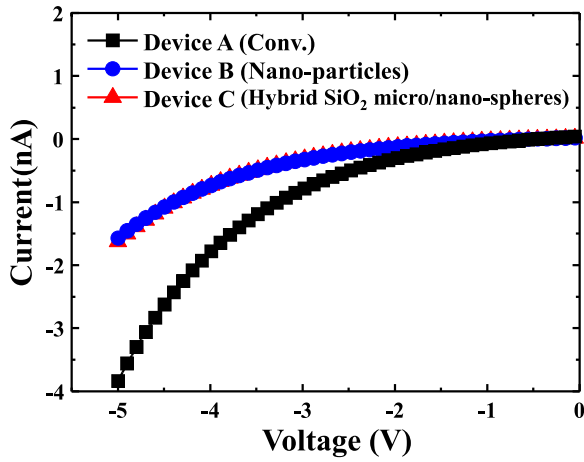


Fig. 7. Reverse-biased *I-V* characteristics of studied devices.

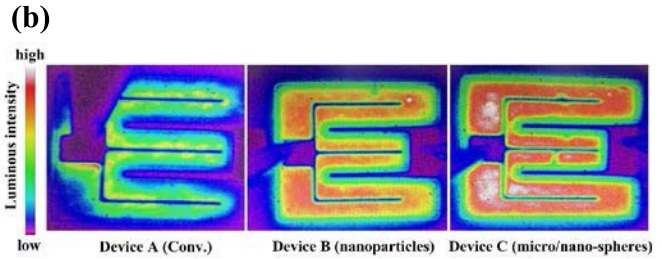
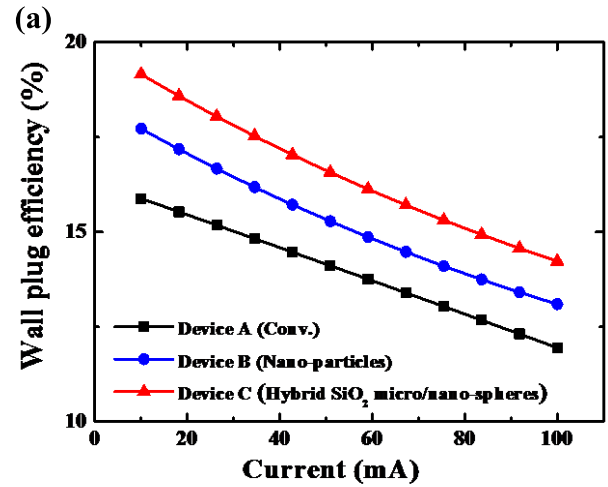


Fig. 9. (a) WPE as a function of operating current. (b) Light emission mapping images of studied devices.

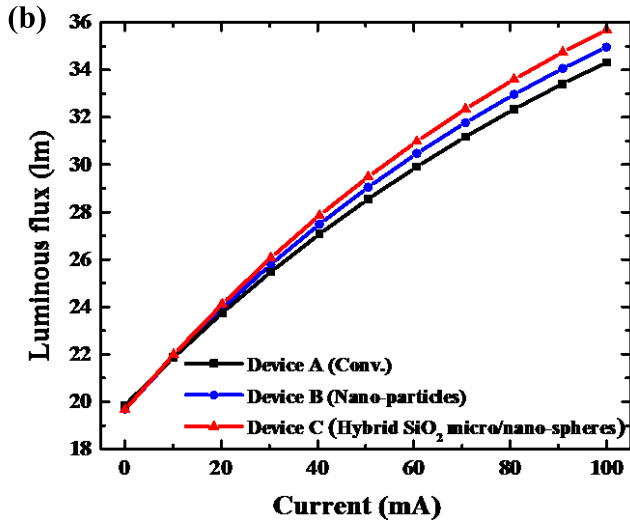
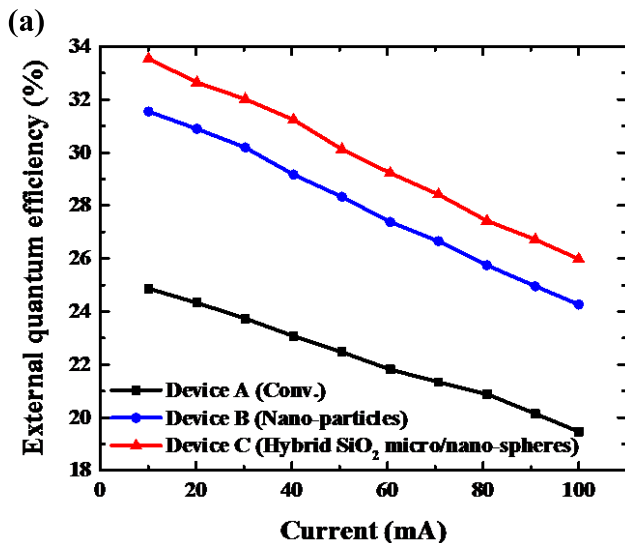


Fig. 8. (a) EQE. (b) Luminous flux as a function of operating current of studied devices.

GaN-based LEDs have been used as a promising light source for solid-state lighting, luminous efficacy should be seriously considered. Hence, although LOP of LED is an important

factor, a high luminous flux is also required. Similarly, devices B and C show higher luminous flux and EQE values than device A. Experimentally, under an injection current of 20 mA, the luminous fluxes are 0.67, 0.78, and 0.86 lm for devices A, B, and C, respectively. As compared with the device A, devices B and C exhibit 16.4% and 28.4% improvements in luminous fluxes [22]. The corresponding EQEs, at an operating current of 20 mA, are 24.3%, 28%, and 32.7% of devices A, B, and C, respectively. Clearly, devices B and C show 26.7% and 34.6% improvements in EQE, respectively, as compared with the device A. These improvements of optical properties are mainly caused by roughened micro (nano) scale SiO₂ surfaces. Certainly, the device C shows the best optical performance due to the most roughened surface of antireflection coating.

The wall-plug efficiencies (WPEs) as a function of operating current of studied devices are revealed in Fig. 9(a). Obviously, devices B and C show higher WPEs than the device A. Under an injection current of 20 mA, WPEs are 15.6%, 17.1%, and 18.5% for devices A, B, and C, respectively. As compared with the device A, devices B and C exhibit 9.6% and 18.6% improvements in WPE, respectively. Fig. 9(b) shows light emission mapping images of devices A to C under 20-mA current injection. As compared with the device A, it can be seen that the light emission intensity of devices B and C are obviously improved. In addition, the light emission intensity of device C is higher than that of device B. It could be attributed to the roughened surface of hybrid SiO₂ microsphere/nanosphere. This has once again confirmed that the use of hybrid SiO₂ microsphere/nanosphere antireflection coating could effectively improve LEE, leading to significantly enhanced

performance of GaN-based LEDs [23]–[26]. Therefore, to compete with traditional light sources in applications of next-generation solid-state lighting, high-power GaN LEDs could be expected [27].

IV. CONCLUSION

A hybrid SiO₂ microsphere/nanosphere antireflection coating, deposited on the surface of GaN-based LEDs, acting as a passivation layer is studied in this paper. The SiO₂ microsphere/nanosphere antireflection coating was uniformly deposited by the RCD method. The diameters of used microspheres and nanoparticles are 1 μm and <30 nm. Since the critical angle could be enlarged by antireflection coating, Fresnel reflection could be reduced. In addition, due to the roughened surface of hybrid SiO₂ microsphere/nanosphere antireflection coating, the scattering effect could also be increased. Thus, LEE could be further enhanced. As compared with a conventional LED (device A), at 20 mA, the studied device C exhibits 18.7% and 34.6% enhancements in an LOP and an EQE without any degradation of electrical properties. Reduced leakage current could also be obtained. These significant improved properties show that high-performance GaN-based LEDs could be achieved by the use of hybrid SiO₂ microsphere/nanosphere antireflection coating.

REFERENCES

- [1] W.-C. Yang, C. Lo, C.-Y. Wei, and W.-S. Lour, "Cell-temperature determination in InGaP–(In)GaAs–Ge triple-junction solar cells," *IEEE Electron Device Lett.*, vol. 32, no. 10, pp. 1412–1414, Oct. 2011.
- [2] H.-C. Yu *et al.*, "Performance improvement of InGaAsN/GaAs quantum well lasers by using trimethylantimony preflow," *Appl. Phys. Exp.*, vol. 4, p. 012103, Dec. 2011.
- [3] D. Steigerwald, S. Rudaz, H. Liu, R. S. Kern, W. Götz, and R. Fletcher, "III–V nitride semiconductors for high-performance blue and green light-emitting devices," *JOM*, vol. 49, pp. 18–23, Sep. 1997.
- [4] N. Y. Pacella, M. T. Bulsara, C. Drazek, E. Guiot, and E. A. Fitzgerald, "Fabrication and thermal budget considerations of advanced Ge and InP SOLES substrates," *ECS J. Solid State Sci. Technol.*, vol. 4, pp. P258–P264, May 2015.
- [5] Y.-L. Chou *et al.*, "Improvement of surface emission for GaN-based light-emitting diodes with a metal-via-hole structure embedded in a reflector," *IEEE Photon. Technol. Lett.*, vol. 23, no. 7, pp. 393–395, Apr. 1, 2011.
- [6] Y.-C. Chang, J.-K. Liou, and W.-C. Liu, "Improved light extraction efficiency of a high-power GaN-based light-emitting diode with a three-dimensional-phonic crystal (3-D-PhC) backside reflector," *IEEE Electron Device Lett.*, vol. 34, no. 6, pp. 777–779, Jun. 2013.
- [7] M. Liu, K. Li, F.-M. Kong, J. Zhao, Q.-A. Ding, and M.-Y. Zhang, "Electrical-optical analysis of photonic crystals GaN-based high power light emitting diodes," *Opt. Quantum Electron.*, vol. 48, pp. 274–288, May 2016.
- [8] J.-K. Liou *et al.*, "Effects of the use of an aluminum reflecting and an SiO₂ insulating layers (RIL) on the performance of a GaN-based light-emitting diode with the naturally textured p-GaN surface," *IEEE Trans. Electron Devices*, vol. 60, no. 7, pp. 2282–2289, Jul. 2013.
- [9] J. K. Kim *et al.*, "Light-extraction enhancement of GaInN light-emitting diodes by graded-refractive-index indium tin oxide anti-reflection contact," *Adv. Mater.*, vol. 20, pp. 801–804, Jan. 2008.
- [10] S.-J. So and C.-B. Park, "Improvement of brightness with Al₂O₃ passivation layers on the surface of InGaN/GaN-based light-emitting diode chips," *Thin Solid Films*, vol. 516, pp. 2031–2034, Feb. 2008.
- [11] C.-M. Yang, D.-S. Kim, Y. S. Park, J.-H. Lee, Y. S. Lee, and J.-H. Lee, "Enhancement in light extraction efficiency of GaN-based light-emitting diodes using double dielectric surface passivation," *Opt. Photon. J.*, vol. 2, pp. 185–192, Sep. 2012.
- [12] I. Schnitzer, E. Yablonoitch, C. Caneau, T. J. Gmitter, and A. Scherer, "30% external quantum efficiency from surface textured, thin-film light-emitting diodes," *Appl. Phys. Lett.*, vol. 63, no. 16, pp. 2174–2176, Oct. 1998.
- [13] K. M. Huang, H. J. Chang, C. L. Ho, and M. C. Wu, "Enhanced light extraction efficiency of GaN-based LEDs with 3-D colloidal-phonic-crystal bottom reflector," *IEEE Photon. Technol. Lett.*, vol. 24, no. 15, pp. 1298–1300, Aug. 1, 2012.
- [14] A. David, H. Benisty, and C. Weisbuch, "Optimization of light-diffracting photonic-crystals for high extraction efficiency LEDs," *J. Display Technol.*, vol. 3, no. 2, pp. 133–148, Jun. 2007.
- [15] P. Kumnorkaew, Y.-K. Ee, N. Tansu, and J. F. Gilchrist, "Investigation of the deposition of microsphere monolayers for fabrication of microlens arrays," *Langmuir*, vol. 24, no. 21, pp. 12150–12157, Jun. 2008.
- [16] P. Kumnorkaew, A. L. Weldon, and J. F. Gilchrist, "Matching constituent fluxes for convective deposition of binary suspensions," *Langmuir*, vol. 26, pp. 2401–2405, Feb. 2010.
- [17] R. D. Deegan, O. Bakajin, T. F. Dupont, G. Huber, S. R. Nagel, and T. A. Witten, "Capillary flow as the cause of ring stains from dried liquid drops," *Nature*, vol. 389, pp. 827–829, Oct. 1997.
- [18] P. Zhu, G. Liu, J. Zhang, and N. Tansu, "FDTD analysis on extraction efficiency of GaN light-emitting diodes with microsphere arrays," *J. Display Technol.*, vol. 9, no. 5, pp. 317–323, May 2013.
- [19] A. N. Noemaun *et al.*, "Optically functional surface composed of patterned graded-refractive-index coatings to enhance light-extraction of GaInN light-emitting diodes," *J. Appl. Phys.*, vol. 110, p. 054510, Sep. 2011.
- [20] N. Dhiman, B. P. Singh, and A. K. Gathania, "Synthesis and characterization of dye-doped TiO₂-SiO₂ core-shell composite microspheres," *J. Nanophoton.*, vol. 6, pp. 063511-1–063511-10, Jul. 2012.
- [21] M. Fabretto *et al.*, "Faradaic charge corrected colouration efficiency measurements for electrochromic devices," *Electrochim. Acta*, vol. 53, pp. 2250–2257, Jul. 2008.
- [22] K. Huang, Y. Gan, Q. Wang, and X. Jiang, "Enhanced light extraction efficiency of integrated LEDs devices with the taper holes microstructures arrays," *Opt. Laser Technol.*, vol. 72, pp. 134–138, Sep. 2015.
- [23] N.-C. Chen, C.-M. Lin, Y.-K. Yang, C. Shen, T.-W. Wang, and M.-C. Wu, "Measurement of junction temperature in a nitride light-emitting diode," *Jpn. J. Appl. Phys.*, vol. 47, no. 12R, pp. 8779–8782, 2008.
- [24] Y. Xi and E. F. Schubert, "Junction-temperature measurement in GaN ultraviolet light-emitting diodes using diode forward voltage method," *Appl. Phys. Lett.*, vol. 85, no. 12, pp. 2163–2165, 2004.
- [25] L. B. Hoch *et al.*, "Nanostructured indium oxide coated silicon nanowire arrays: A hybrid photothermal/photochemical approach to solar fuels," *ACS Nano*, vol. 10, pp. 9017–9025, Sep. 2016.
- [26] M. Duzyol, O. Sagsoz, N. P. Sagsoz, N. Akgul, and M. Yildiz, "The effect of surface treatments on the bond strength between CAD/CAM blocks and composite resin," *J. Prosthodontics*, vol. 25, pp. 466–471, Aug. 2016.
- [27] M. R. Krames *et al.*, "Status and future of high-power light-emitting diodes for solid-state lighting," *J. Display Technol.*, vol. 3, no. 2, pp. 160–175, Jun. 2007.



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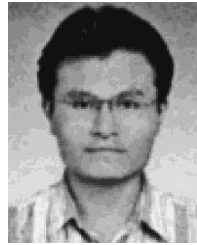
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