# Effective optimization and analysis of white LED properties by using nano-honeycomb patterned phosphor film

HUANG-YU LIN,<sup>1</sup> SHENG-WEN WANG,<sup>1</sup> CHIEN-CHUNG LIN,<sup>1,2,3</sup> ZONG-YI TU,<sup>1</sup> PO-TSUNG LEE,<sup>1</sup> HUANG-MING CHEN,<sup>1</sup> AND HAO-CHUNG KUO<sup>1,\*</sup>

**Abstract:** This study presents an approach for patterning a polydimethylsiloxane (PDMS) phosphor film with a photonic crystal nano-honeycomb structure on a blue chip package. A phosphor film with a nano-honeycomb structure was patterned and transferred using a nanosphere and used for fabricating remote white light-emitting diodes (w-LEDs). The angular correlated color temperature deviation of the remote phosphor LED could be improved by varying nano-honeycomb structure pitches (450, 750, and 1150 nm). In particular, w-LED samples with excellent color uniformity (ΔCCT ranging from 940 to 440 K) were fabricated from 750-nm w-LED samples with nano-honeycomb-patterned tops.

©2016 Optical Society of America

OCIS codes: (230.3670) Light-emitting diodes; (230.2090) Electro-optical devices; (050.5298) Photonic crystals.

### References and links

- S. Nakamura, T. Mukai, and M. Senoh, "Candela-class high-brightness InGan/AlGan double-heterostructure blue-light-emitting diodes," Appl. Phys. Lett. 64(13), 1687–1689 (1994).
- T. Franzl, D. Koktysh, T. Klar, A. Rogach, J. Feldmann, and N. Gaponik, "Fast energy transfer in layer-by-layer assembled CdTe nanocrystal bilayers," Appl. Phys. Lett. 84(15), 2904–2906 (2004).
- 3. E. F. Schubert and J. K. Kim, "Solid-state light sources getting smart," Science 308(5726), 1274–1278 (2005).
- D. A. Steigerwald, J. C. Bhat, D. Collins, R. M. Fletcher, M. O. Holcomb, M. J. Ludowise, P. S. Martin, and S. L. Rudaz, "Illumination with solid state lighting technology," IEEE J. Sel. Top. Quantum Electron. 8(2), 310–320 (2002).
- J. K. Kim, H. Luo, E. F. Schubert, J. Cho, C. Sone, and Y. Park, "Strongly enhanced phosphor efficiency in GaInN white light-emitting diodes using remote phosphor configuration and diffuse reflector cup," Jpn. J. Appl. Phys. 44(21), L649–L651 (2005).
- H.-C. Chen, K.-J. Chen, C.-C. Lin, C.-H. Wang, H.-V. Han, H.-H. Tsai, H.-T. Kuo, S.-H. Chien, M.-H. Shih, and H.-C. Kuo, "Improvement in uniformity of emission by ZrO<sub>2</sub> nano-particles for white LEDs," Nanotechnology 23(26), 265201 (2012).
- K.-J. Chen, H.-V. Han, H.-C. Chen, C.-C. Lin, S.-H. Chien, C.-C. Huang, T.-M. Chen, M.-H. Shih, and H.-C. Kuo, "White light emitting diodes with enhanced CCT uniformity and luminous flux using ZrO2 nanoparticles," Nanoscale 6(10), 5378–5383 (2014).
- 8. Y.-K. Su, P.-C. Wang, C.-L. Lin, G.-S. Huang, and C.-M. Wei, "Enhanced Light Extraction Using Blue LED Package Consisting of-Doped Silicone Layer and Silicone Lens," IEEE Electron Dev. Lett. **35**, 575–577 (2014).
- H. Sugawara, K. Itaya, and G.-i. Hatakoshi, "Hybrid-type InGaAIP/GaAs distributed Bragg reflectors for InGaAIP light-emitting diodes," Jpn. J. Appl. Phys. 33(11), 6195–6198 (1994).
- H.-Y. Lin, K.-J. Chen, S.-W. Wang, C.-C. Lin, K.-Y. Wang, J.-R. Li, P.-T. Lee, M.-H. Shih, X. Li, H.-M. Chen, and H. C. Kuo, "Improvement of light quality by DBR structure in white LED," Opt. Express 23(3), A27–A33 (2015).
- L. Yang, S. Wang, Z. Lv, and S. Liu, "Color deviation controlling of phosphor conformal coating by advanced spray painting technology for white LEDs," Appl. Opt. 52(10), 2075–2079 (2013).
  Y. Shuai, Y. He, N. T. Tran, and F. G. Shi, "Angular CCT uniformity of phosphor converted white LEDs:
- 12. Y. Shuai, Y. He, N. T. Tran, and F. G. Shi, "Angular CCT uniformity of phosphor converted white LEDs: Effects of phosphor materials and packaging structures," IEEE Photonics Technol. Lett. **23**(3), 137–139 (2011).
- 13. Z. Liu, S. Liu, K. Wang, and X. Luo, "Optical analysis of color distribution in white LEDs with various packaging methods," IEEE Photonics Technol. Lett. 20(24), 2027–2029 (2008).

<sup>&</sup>lt;sup>1</sup>Department of Photonics & Institute of Electro-Optical Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan

<sup>&</sup>lt;sup>2</sup>Institute of Photonic System, National Chiao Tung University, Tainan 711, Taiwan

<sup>&</sup>lt;sup>3</sup>chienchunglin@faculty.nctu.edu.tw

<sup>\*</sup>hckuo@faculty.nctu.edu.tw

- L. Yang, Z. Lv, Y. Jiaojiao, and S. Liu, "Effects of melamine formaldehyde resin and CaCO3 diffuser-loaded encapsulation on correlated color temperature uniformity of phosphor-converted LEDs," Appl. Opt. 52(22), 5539–5544 (2013).
- C.-Y. Liu, K.-J. Chen, D.-W. Lin, C.-Y. Lee, C.-C. Lin, S.-H. Chien, M.-H. Shih, G.-C. Chi, C.-Y. Chang, and H.-C. Kuo, "Improvement of emission uniformity by using micro-cone patterned PDMS film," Opt. Express 22(4), 4516–4522 (2014).
- L.-C. Chen, C.-K. Wang, J.-B. Huang, and L.-S. Hong, "A nanoporous AlN layer patterned by anodic aluminum oxide and its application as a buffer layer in a GaN-based light-emitting diode," Nanotechnology 20(8), 085303 (2009).
- 17. Y. Li, S. You, M. Zhu, L. Zhao, W. Hou, T. Detchprohm, Y. Taniguchi, N. Tamura, S. Tanaka, and C. Wetzel, "Defect-reduced green GaInN/GaN light-emitting diode on nanopatterned sapphire," Appl. Phys. Lett. **98**(15), 151102 (2011).
- J.-K. Huang, D.-W. Lin, M.-H. Shih, K.-Y. Lee, J.-R. Chen, H.-W. Huang, S.-Y. Kuo, C.-H. Lin, P.-T. Lee, and G.-C. Chi, "Investigation and comparison of the GaN-based light-emitting diodes grown on high aspect ratio nano-cone and general micro-cone patterned sapphire substrate," J. Disp. Technol. 9(12), 947–952 (2013).
- Y.-J. Lee, C.-H. Chiu, C. C. Ke, P. C. Lin, T.-C. Lu, H.-C. Kuo, and S.-C. Wang, "Study of the excitation power dependent internal quantum efficiency in InGaN/GaN LEDs grown on patterned sapphire substrate," IEEE J. Sel. Top. Quantum Electron. 15(4), 1137–1143 (2009).
- C.-H. Chiu, C.-C. Lin, H.-V. Han, C.-Y. Liu, Y.-H. Chen, Y.-P. Lan, P. Yu, H.-C. Kuo, T.-C. Lu, S.-C. Wang, and C. Y. Chang, "High efficiency GaN-based light-emitting diodes with embedded air voids/SiO2 nanomasks," Nanotechnology 23(4), 045303 (2012).
- M. Ali, A. Romanov, S. Suihkonen, O. Svensk, S. Sintonen, M. Sopanen, H. Lipsanen, V. Nevedomsky, N. Bert, M. Odnoblyudov, and V. E. Bougrov, "Analysis of threading dislocations in void shape controlled GaN re-grown on hexagonally patterned mask-less GaN," J. Cryst. Growth 344(1), 59–64 (2012).
- 22. J. Zhao, A. Wang, M. A. Green, and F. Ferrazza, "19.8% efficient "honeycomb" textured multicrystalline and 24.4% monocrystalline silicon solar cells," Appl. Phys. Lett. **73**(14), 1991–1993 (1998).
- G. Widawski, M. Rawiso, and B. Francois, "Self-organized honeycomb morphology of star-polymer polystyrene films," Nature 369(6479), 387–389 (1994).
- L. Li, Y. Zhong, C. Ma, J. Li, C. Chen, A. Zhang, D. Tang, S. Xie, and Z. Ma, "Honeycomb-patterned hybrid films and their template applications via a tunable amphiphilic block polymer/inorganic precursor system," Chem. Mater. 21(20), 4977–4983 (2009).
- 25. L. A. Connal and G. G. Qiao, "Preparation of Porous Poly (dimethylsiloxane)-Based Honeycomb Materials with Hierarchal Surface Features and Their Use as Soft-Lithography Templates," Adv. Mater. 18(22), 3024–3028 (2006).
- H. Li and N. Wu, "A large-area nanoscale gold hemisphere pattern as a nanoelectrode array," Nanotechnology 19(27), 275301 (2008).
- X.-B. Shi, M. Qian, Z.-K. Wang, and L.-S. Liao, "Nano-honeycomb structured transparent electrode for enhanced light extraction from organic light-emitting diodes," Appl. Phys. Lett. 106(22), 223301 (2015).
- C.-C. Lin, W.-L. Liu, and C.-Y. Hsieh, "Scalar scattering model of highly textured transparent conducting oxide," J. Appl. Phys. 109(1), 014508 (2011).
- H. T. Chiu, C. Y. Chang, C. L. Chen, T. Y. Chiang, and M. T. Guo, "Preparation and characterization of UV-curable organic/inorganic hybrid composites for NIR cutoff and antistatic coatings," J. Appl. Polym. Sci. 120(1), 202–211 (2011).

## 1. Introduction

Recently, white light-emitting diodes (w-LEDs), which are manufactured using green technology, have replaced conventional incandescent light bulbs [1–3]. In general, many types of phosphor package, such as phosphor-dispensing, conformal phosphor [4], and remote phosphor packages [5], are used for fabricating w-LEDs. A remote phosphor light-emitting diode (LED) is a type of high-performance, high-illumination-flux w-LED. However, this LED has disadvantages such as poor color uniformity [6]. Various studies have attempted to overcome this drawback. Many techniques such as using zirconia and TiO<sub>2</sub> nanoparticles in the fabrication of w-LEDs can be employed to improve the color uniformity and optimize the properties of w-LEDs [6–8]. TiO<sub>2</sub> and zirconia nanoparticles are used as scattering particles for improving the uniformity and luminous efficiency of w-LEDs. Other approaches include using a distributed Bragg reflector (DBR) structure, which serves as an optical mirror and offers an alternative ordering that achieves high reflectivity [9].

The design of the DBR structure can improve the w-LED's color uniformity [10]. In addition to using nanoparticles and a DBR structure, many other approaches facilitate optimizing the uniformity and luminous efficiency of w-LEDs [11–14]. Among them,

patterned structures such as surface-textured structures and nanostructures are suitable for LED packages and improve light extraction [15]. A textured polydimethylsiloxane (PDMS) pattern was transferred using a patterned sapphire structure. To enhance the quantum efficiency of LEDs through epitaxial lateral overgrowth, a patterned sapphire substrate has been developed for the LED industry; this substrate also improves light extraction efficiency because of its high-scattering ability [16–19]. Many patterning techniques have been investigated, with one approach patterning sapphire by inserting voids in it, which increased light scattering at the substrate interface [20,21].

Many research groups have developed nano-honeycomb structures, introduced approaches for fabricating them, and determined applications for these structures [22,23]. How to apply the thermal stability of polymer nano-honeycomb films and their resistance to organic solvents under harsh conditions should be examined [24]. The transfer printing method is widely used for fabricating nano-honeycomb structures; for example, a nano-honeycomb film can be cast from a dilute benzene solution of a star polymer onto a glass surface [25]. The current study fabricated a patterned phosphor PDMS film with a nano-honeycomb structure by using PDMS and polystyrene (PS) nano-spheres. PS nano-spheres have been used as a mask in the lithography process for fabricating nano-honeycomb films or patterned ITO structures for use in organic LEDs [26,27]. These products are always with the characteristic of the photonic crystal by the use of the (PS) nano-spheres. Using a nano-honeycomb structure in w-LEDs reduces the large color temperature deviation of the remote phosphor LED. To optimize LED devices, this study selected a convenient method for patterning phosphor film, using simulation and experiment results to demonstrate the advantages of fabricating phosphor film with a nano-honeycomb structure.

### 2. Experimental methods

In an experiment, w-LEDs were fabricated by using nano-honeycomb -patterned phosphor films and the remote phosphor method. Figure 1(a) shows the flow of the nano-honeycomb structure of the phosphor film in the experiment. The following procedure was used: First, PS nano-spheres with dimensions of 1.5  $\mu$ m, 1  $\mu$ m, and 600 nm were prepared. Second, 12 wt% YAG phosphor with the particle size of 10  $\mu$ m was blended in the silicone glue to form a phosphor suspension slurry. Third, the phosphor slurry was cast onto a glass substrate coated with the PS nanospheres. After the glass substrate was baked at 70 °C for 10 min, a PDMS film with a nano-honeycomb structure was obtained, and the film was transferred using the nano-spheres. Films with dimensions of 1150nm, 750nm, and 450nm and respective impression depths in the nano-honeycomb structures of approximately 180nm, 120nm, and 100 nm, were fabricated using this procedure.

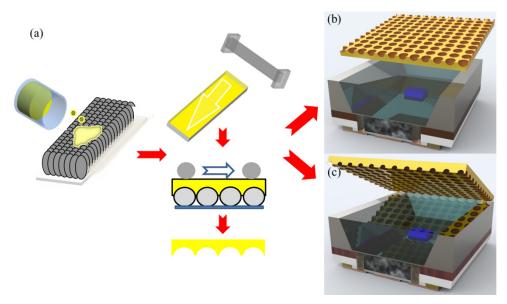


Fig. 1. (a) Flow of the honeycomb structure in the experiment and honeycomb structures patterned at the (a) top and (b) bottom covering a remote phosphor LED.

Scanning electron microscopy (SEM) images of top views of the honeycomb -patterned PDMS films with the dimensions of 450nm, 750nm, and 1150nm are shown in Fig. 2, and the inset shows a cross-sectional image of the nano-honeycomb film. The depth of impression for the honeycomb structures are about approximately 180nm, 120nm, and 100 nm, respectively. Subsequently, LED 5070 packages were prepared, and silicone was dispensed on the packages. Finally, the LED 5070 packages were covered with the patterned phosphor PDMS films to obtain remote phosphor w-LED samples of two types: one with the phosphor patterned on the top and the other with the phosphor patterned on the bottom.

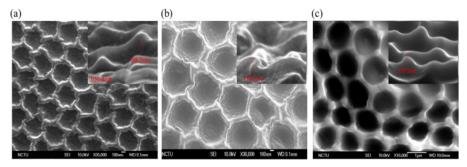


Fig. 2. SEM images of the honeycomb-patterned PDMS films with the dimensions of (a) 450nm, (b) 750nm, and (c) 1150 nm 3. Results and discussion.

A ray-tracing simulation was designed to examine the effect of the different dimensions of the nano-honeycomb structure on the color uniformity of the w-LEDs. LightTools software was used to obtain the simulation spectrum of the angular correlated color temperature (CCT) deviations from the three dimensions and two sample types of nano-honeycomb -patterned w-LEDs were as shown in Fig. 3(a)-3(b). The simulation results showed optimal color uniformity for nano-honeycomb -structured samples with a dimension of 750 nm and a patterned top.

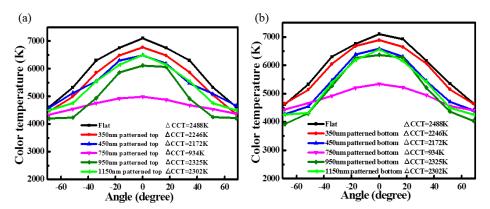


Fig. 3. Simulation spectrum of angular CCT for w-LEDs with (a) a nano-honeycomb covering with a patterned top and (b) a nano-honeycomb covering with a patterned bottom.

Figure 4(a) shows the experimental spectrum of the angular CCT deviations for the nanohoneycomb -structured samples with dimensions of 1150nm, 750nm, and 450 nm at a current of 120 mA. The experimental and simulation results showed that samples with patterned tops had greater color uniformity than those with patterned bottoms and that those with a dimension of 750 nm showed superior results. The CCT deviations of the LEDs were caused by anisotropic diffraction at the various optical layers in the structure. In this study, color uniformity was optimized by introducing extra nano-honeycomb layers, which diffused omnidirectional photons and showed excellent diffusion ability as shown in Fig. 4(b). The haze value represents light scattering capability and is defined as the ratio of nonspecular photons to all-diffracted photons, was calculated by using the following eq [28,29].

$$Haze(\%) = T_{diffrac} \times T_{total}^{-1} \times 100\%. \tag{1}$$

where  $T_{diffraction}$  is the diffractive transmittance (the deviation of total transmittance and specular diffraction), and  $T_{total}$  is the total transmittance. As shown in Fig. 4(b), the nanohoneycomb-structured PDMS layers in the samples with dimensions of 450nm and 1150 nm had a higher haze value than a flat layer and scattered over 85% of the incident light.

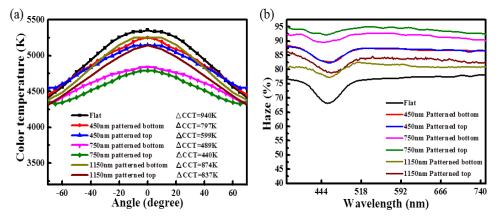


Fig. 4. (a) Spectrum of angular CCT at 120 mA obtained in the experiment. (b) The haze intensity of the nano-honeycomb structured PDMS films with dimensions of 450nm, 750nm, and 1150 nm.

To investigate the effect of the nano-honeycomb structure on luminous efficiency, a simulation was performed that entailed w-LEDs with nano-honeycomb -structured PDMS films having dimensions from 350 to 1150 nm. Figure 5(a) and 5(b) show a comparison of

simulation spectra for a reference sample of flat phosphor film and samples with nanohoneycomb structures of various sizes and patterned tops or bottoms used as coverings for remote phosphor w-LEDs. The intensities of the w-LEDs with the flat and nano-honeycomb structured phosphor films at a wavelength of 560 nm (yellow light peaks) were plotted in Fig. 6. The results showed that the nano-honeycomb structure could improve the luminous efficiency of w-LEDs and that the 750-nm samples with patterned tops showed the highest luminous efficiency.

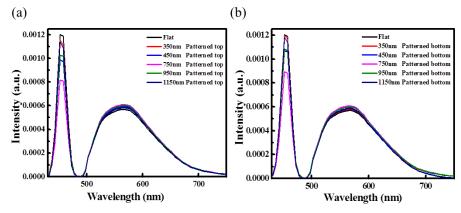


Fig. 5. Simulation spectrum of w-LEDs covered with nano-honeycomb structures patterned at the (a) top and (b) bottom.

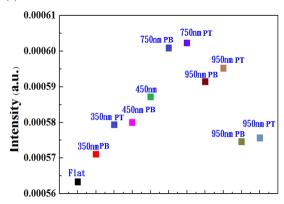


Fig. 6. Flow plot of the spectra (obtained from Figs. 5(a) and 5(b)) for a wavelength of 550nm.

Figure 7(a) shows the emission spectra at a current of 120 mA. The measured spectra show the suppression of blue rays and enhancement of yellow rays; these effects are strongest for the 750-nm samples with patterned tops. The current-dependent luminous flux of w-LEDs with different PDMS films and patterned tops or bottoms was measured for currents from 100 to 800 mA as shown in Fig. 7(b). The luminous efficiency values of the samples were compared with that of a sample without a nano-honeycomb pattern and of the samples with different dimension patterns as shown in Table 1. The experimental and simulation results showed that w-LED samples possessing a 750-nm phosphor film with a patterned top showed excellent luminous efficiency, with values that were respectively 6.4% and 9.5% greater than that of the sample with a flat phosphor film. This result was expected because the nano-honeycomb structure improves phosphor excitation, thus increasing the number of yellow photons and leading to lumen enhancement.

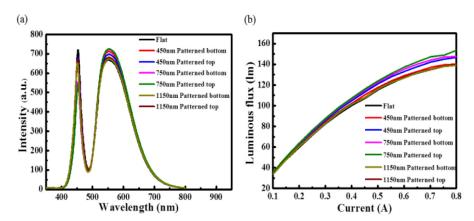


Fig. 7. (a) Emission spectra at a current of 120 mA and (b) luminous flux for the 450nm, 750nm, and 1150nm PDMS films with patterned tops or bottoms that covered the w-LEDs.

Table 1. Luminous Efficiency Values of Samples with Flat Phosphor Layer and Patterned Layers

Luminous efficiency(lm/W)	Flat	450nm down	450nm Up	750nm Down	750nm Up	1150nm Down	1150nm Up
Simulation	90.8	93.6	94.4	95.5	96.6	92.7	93.3
Experiment	101.4	104.4	105.8	110	111.1	103.2	105.2

Figure 8 shows the CCT deviation and the figure of merit (FOM) from the result of the simulation and experiment, which was defined as the following Eq [7].

$$FOM = (Lumen_{patterned} - Lumen_{non-patterned})\Delta CCT^{-1}.$$
 (2)

where Lumen<sub>pattern</sub>-Lumen<sub>non-pattern</sub> is defined as the luminous efficiency deviation between the flat and nano-honeycomb patterned samples. According to the result of the FOM, increasing size of the honeycomb pattern can optimize the color uniformity and luminous. However, if the size improvement of the honeycomb structure trend to larger than the suitable dimension, it also caused to luminous efficiency reduction and color non-uniform. In this study, from the result of FOM, the suitable design is the remote LED with 750nm patterned tops phosphor film covered.

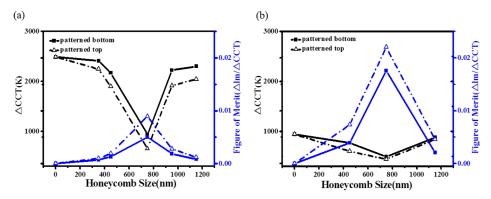


Fig. 8. The CCT deviation and the figure of merit of different size of honeycomb phosphor layer with patterned bottom or top covered for remote LEDs by the result of (a) simulation and (b) experiment.

In this study, the main focus is on the variation of honeycomb structure and its effect on the CCT and luminous efficiency enhancement. Therefore, the concentration of the phosphors

in the film and the main thickness of the film were controlled at the same level to have similar yellow photons output. The different experimental results shows clearly what different honeycomb layouts can provide. The important factors in the wLED such as the control on the CCT and overall luminous flux can be further optimized by varying the thickness and phosphor concentration, which will not be covered in the scope of this study.

### 4. Conclusion

In conclusion, using a nano-honeycomb -structured phosphor covering on a chip used for fabricating remote w-LEDs can improve the color uniformity and luminous efficiency of the LEDs. Experimental and simulation results showed that the 750-nm w-LED samples with patterned tops exhibited excellent luminous efficiency (9.5% higher than that of flat samples) and color uniformity (ΔCCT ranging from 940 to 440 K). This study demonstrates a convenient approach for designing nano-honeycomb structures of various dimensions, which is a potentially convenient design for optimizing w-LEDs.

# Acknowledgment

The authors express their gratitude to Epistar for their technical support. This research was funded by the Ministry of Science and Technology of Taiwan for financial support through the grant number: MOST 101-2221-E-009-046-MY3, MOST104-3113-E-009-002-CC2, and MOST102-2221-E-009-131-MY3.