

Conformal phosphor coating using pulsed spray to reduce color deviation of white LEDs

Hsin-Tao Huang,^{1,*} Chuang-Chuang Tsai,² and Yi-Pai Huang²

¹Department of Photonics and Institute of Electro-optical Engineering,
National Chiao Tung University, Hsinchu 30010, Taiwan

²Department of Photonics and Display Institute, National Chiao Tung University, Hsinchu 30010, Taiwan
*stevenhuang.eo96g@nctu.edu.tw

Abstract: This work presents a novel “pulsed spray (PS)” process for the coating of yellow YAG:Ce³⁺ phosphor on blue InGaN-based light emitting diodes (LEDs). To coat a phosphor layer of high quality on an LED chip surface, the PS approach is used and studied because of the uniform color distribution, providing a wide range of color temperatures. This PS coating approach applies phosphor by exploiting mechanical principles without risk of chemical pollution. Additionally, it can be applied to wire-bonded LEDs and an array of LED chips on a substrate to fabricate a large-area, planar illumination system of high optical quality, which is easy to manufacture.

©2010 Optical Society of America

OCIS codes: (230.3670) Light-emitting diodes; (350.4600) Optical engineering.

References and links

1. F. M. Steranka, J. Bhat, D. Collins, L. Cook, M. G. Craford, R. Fletcher, N. Gardner, P. Grillot, W. Goetz, M. Keuper, R. Khare, A. Kim, M. Krames, G. Harbers, M. Ludowise, P. S. Martin, M. Misra, G. Mueller, R. Mueller-Mach, S. Rudaz, Y.-C. Shen, D. Steigerwald, S. Stockman, S. Subramanya, T. Trotter, and J. J. Wierer, “High Power LEDs—Technology Status and Market Applications,” *Phys. Stat. Solidi A* **194**(2), 380–388 (2002).
2. M. O. Holcomb, R. Mueller-Mach, G. Mueller, D. Collins, R. M. Fletcher, D. A. Steigerwald, S. Eberle, Y. K. Lim, P. S. Martin, and M. Krames, “The LED Lightbulb: Are we there yet? Progress and Challenges for Solid State Illumination,” *Lasers and Electro-Optics, 2003 CLEO’03 Conference on 1–6 June 2003*.
3. Z. Liu, S. Liu, K. Wang, and X. Luo, “Optical Analysis of Color Distribution in White LEDs with Various Packaging Methods,” *IEEE Photon. Technol. Lett.* **20**(24), 2027–2029 (2008).
4. H. T. Huang, C. C. Tsai, and Y. P. Huang, “A Direct-View Backlight With UV Excited Trichromatic Phosphor Conversion Film,” *J. Display Technol.* **6**(4), 128–134 (2010).
5. J. H. Yum, S. Y. Seo, S. Lee, and Y. E. Sung, “Comparison of Y₃Al₅O₁₂:Ce_{0.05} Phosphor Coating Methods for White Light Emitting Diode on Gallium Nitride,” *Proc. SPIE* **4445**, 60–68 (2001).
6. B. Hou, H. Rao, and J. Li, “Methods of Increasing Luminous Efficiency of Phosphor-Converted LED Realized by Conformal Phosphor Coating,” *J. Display Technol.* **5**(2), 57–60 (2009).
7. N. T. Tran, and F. G. Shi, “Studies of Phosphor Concentration and Thickness for Phosphor-Based White Light-Emitting-Diodes,” *J. Lightwave Technol.* **26**(21), 3556–3559 (2008).

1. Introduction

White light-emitting diodes (wLEDs) are becoming increasingly important light sources for illumination applications, because they are compact, mercury-free, and energy-efficient. However, conventional wLEDs that are fabricated by dispensing [Fig. 1(a)], settling method, spin coating, self-exposure or electrophoretic deposition (EPD) have problems [1–4]. The dispensing method is the most popular approach in industry, but the settling of the phosphor powder before curing causes disharmony between blue light and yellow light. Additionally, the settling approach is a chemical reaction and requires a flip-chip LED with a flat surface for phosphor coating. Spin coating centrifugally separates phosphor from slurry. Self-exposure or EPD can produce an uniform phosphor coating, but these chemical processes causes Cr ion pollution. Therefore, an environmentally friendly phosphor coating on InGaN chips with uniform thickness was formed to optimize the performance of wLEDs. This study uses a novel approach that applies the pulsed spray (PS) process to ensure that the phosphor coating conforms to the LED chip outline. This coating is called the conformal phosphor coating (CPC), as shown in Fig. 1(b). The PS approach with an interval control is used to feed

phosphor slurry through an air atomizing nozzle to spray and pile up the phosphor layer by layer. It yields a thin, uniform phosphor along the perimeter of the chip. Experimental results show that the PS approach can be used to form LEDs with an accurate color distribution achieving a wide range of color temperatures (T_c from 2500 to 9500 K), for use in a great diversity of applications, including TFT-LCD backlights and general lighting. Additionally, PS can be applied to LED chips of the wire-bonded type. Therefore, this study uses a lighting system with an array of blue LED chips on a board (COB), which are coated with yellow phosphor by the aforementioned PS technique for evaluation. Additionally, as one of the means of phosphor coating, PS is fast and inexpensive, and yields an uniform phosphor layer and reproducible results.

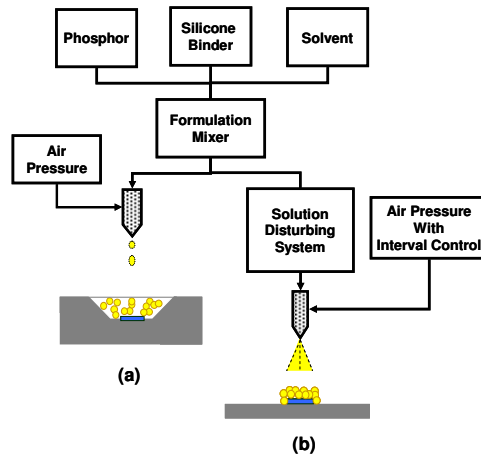


Fig. 1. Schematic diagram of the phosphor coating process. (a) Conventional dispensing; (b) Pulsed Spray (PS).

2. Experiments

Currently, most wLEDs are formed by the two-color light mixing of blue (LEDs excitation) and yellow (YAG:Ce³⁺ phosphor emission) wavelengths. In this study, the conventional dispensing method [Fig. 1(a)] is compared with the PS method [Fig. 1(b)] of forming a phosphor coating. The phosphor powder is blended with a silicone binder and an alkyl-based solvent to form phosphor suspension slurry for use in both described coating processes. The dispensing and PS approaches have different fluid requirements, and therefore different binder/solvent ratios. However, the phosphor powder that was used in this experiment was YAG:Ce³⁺, which has a broadband wavelength distribution (490~680 nm) to ensure favorable color rendering. Ninety percent of the phosphor used in this experiment had a diameter (D_{90}) of less than 13.27 μm [Figs. 2(a) and 2(b)]. An indium gallium nitride (InGaN)-based blue LED, which emits at a wavelength of 457 nm, was used to excite yellow phosphor.

The PS parameters are studied to optimize the coating conditions. The velocity of the spray, the amount of phosphor in the slurry (phosphor loading), the ejected air pressure and the spraying interval (pulsed spraying frequency) were controlled. The suspended slurry was prepared and disturbed mechanically to prevent the phosphor from settling before it was sprayed out. Then, the slurry was poured into the coating system to be atomized, and sprayed at a pulsing frequency of 5-10 Hz onto the surfaces of bare LED chips.

In this study, a lighting system was fabricated using a novel PS approach to yield an uniform CPC on the LEDs surfaces. As shown in Fig. 3(a), blue LED chips were mounted on a 4-inch aluminum alloy substrate using the COB process, and then yellow phosphor was coated on them to cover the whole perimeter of the LED with an uniform thickness layer. The advantages of CPC using the PS approach will be discussed below.

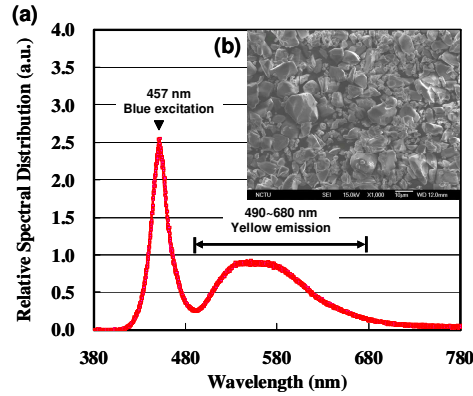


Fig. 2. (a) The spectral properties of conformal phosphor coating using pulsed spray; (b) SEM morphology of YAG:Ce³⁺ phosphor.

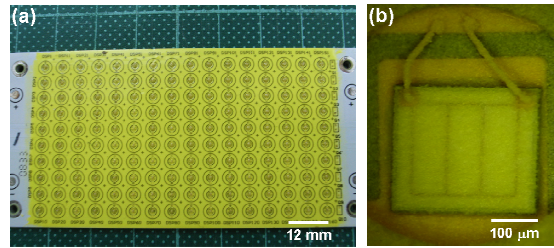


Fig. 3. Phosphor coating using PS technique. (a) An array of blue LED chips on board (COB); (b) the magnification of individual LED.

3. Results and discussion

The phosphor coating using dispensing method and PS method are both uncomplicated and involves no chemical reaction. In the following, the color distribution, the color temperature and the other optical performance parameters will be evaluated for these two methods and compared.

3.1 Color distribution

The color deviation of a lighting system, $\Delta u'v'$ [Eq. (3)], is evaluated as follows:

$$u'_i = 4x_i / (-2x_i + 12y_i + 3), \Delta u'_{i=1} - \Delta u'_{i=2,3,\dots} \quad (1)$$

$$v'_i = 9y_i / (-2x_i + 12y_i + 3), \Delta v'_{i=1} - \Delta v'_{i=2,3,\dots} \quad (2)$$

$$\Delta u'v' = \left((\Delta u')^2 + (\Delta v')^2 \right)^{1/2} \quad (3)$$

where x_i, y_i denote the chromaticity coordinates in the CIE 1931 diagram and u'_i, v'_i denote the chromaticity coordinates in the CIE 1976 diagram [Eqs. (1) and (2)]. For dispensing method, the suspended phosphor is easily delaminated by gravity before the silicone binder is cured, causing color deviation at various viewing angles [Fig. 4(a)]. LED packages must therefore be sorted and when necessary binned before use, resulting in high production costs. The experimental $\Delta u'v'$ value reveals that the PS technique yields a much greater spatial color uniformity [Fig. 4(b), $\Delta u'v'$ of 0.07] than does the dispensing method ($\Delta u'v'$ of 0.23) when the light is emitted at angles from -60° to $+60^\circ$. Therefore, PS technique allows the accurate

control of color distribution for high-quality lighting applications. Table 1 summarizes the comparison of optical data regarding conventional dispensing method and CPC by PS technique. When controlling T_c of experiment around 9500 K, CPC by PS reveals accurate color distribution and higher luminous efficacy (8% improvement).

Table 1. Optical comparison between conventional dispensing and CPC by PS^a

	C_x	C_y	Y (lm)	$\Delta u' v'$	T_c (K)
Conventional dispensing	0.290	0.276	48.80	0.23	9533
CPC by PS	0.284	0.290	52.70	0.07	9519

^aPower consumption: 0.5 W (forward voltage (V_F): 3.5 V, forward current (I_F): 150 mA)

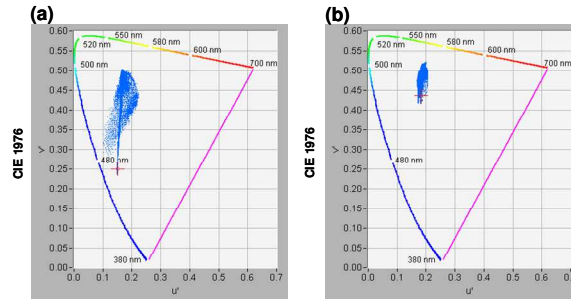


Fig. 4. CIE 1976 chromaticity indices versus angular distribution of LED when the light is emitted at angles from -60° to $+60^\circ$. (a) Conventional dispensing; (b) conformal coating using PS.

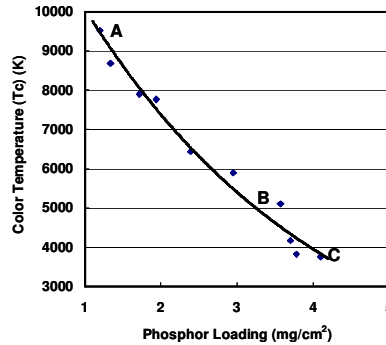


Fig. 5. Relationship between phosphor loading (mg/cm^2) and color temperature (T_c).

3.2 Range of color temperature

Different applications have different T_c requirements. TFT-LCD TV applications require illumination at a high color temperature ($T_c > 9000$ K). However, general lighting has a T_c range of 3000 K (warm white) to 5000 K (cool white). This work studies the dependence of T_c for white light on phosphor loading (mg/cm^2) (Fig. 5). The experimental results reveal that wLEDs based on the PS technique have a T_c value that is inversely proportional to the phosphor loading. Whereas a low T_c is achieved at high phosphor loading (point C), a high T_c is achieved at low phosphor loading (point A), and is associated with strongly non-uniform color distribution [7]. Therefore, the color distribution of wLEDs at high T_c was explored. As shown in Fig. 6(a), the conventional dispensing approach cannot easily ensure color uniformity with a T_c at 9533 K. The chromaticity indices display a severe variation with the viewing angle from -60° to $+60^\circ$. When the PS approach is used at a phosphor loading of $1.20 \text{ mg}/\text{cm}^2$, the resulted T_c is 9519 K [Fig. 6(b)] with a harmonic chromaticity distribution.

When the spraying interval in PS is varied to control the amount of deposited phosphor, a thin and uniform phosphor layer can be coated on the surface of the LED chips. In this case, when a higher percentage of yellow light (complementary wavelength) is mixed in, the Tc is shifted toward the yellow side (low Tc). Figures 5 and 7 reveal that a Tc of 5100 K (cool white, point B), a Tc of 4174 K (warm white, point C) and a Tc of 9519 K (high color temperature, point A) can be achieved using the PS approach.

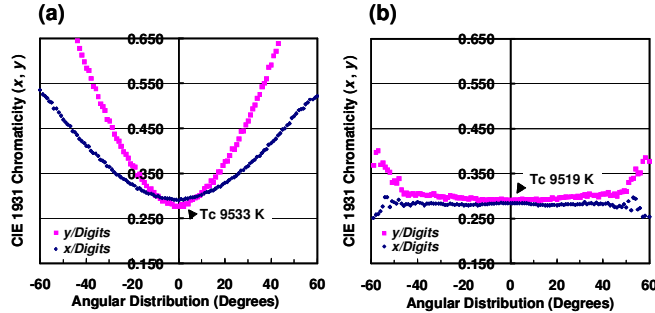


Fig. 6. CIE 1931 chromaticity indices versus angular distribution of LED at Tc centre around 9500 K. (a) Conventional dispensing; (b) conformal coating using PS.

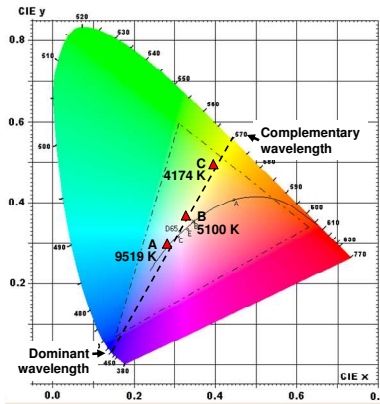


Fig. 7. The CIE 1931 chromaticity diagram. Red triangle denotes the white point with different color temperature (Tc).

3.3 Environmentally friendly process

In contrast with conventional approaches, PS yields an environmentally friendly and uniform phosphor coating. After PS process, the binder (silicone based) in slurry will be cured (100° C heating for 4 hours) and the solvent (alkyl based) will be evaporated. However, the aforementioned EPD or self-exposure process uses ammonium dichromate ((NH₄)₂Cr₂O₇) ADC solution as a photosensitizer, producing Cr ions that can reduce the intensity of emitted light [5,6]. Additionally, ionic Cr is a hazardous substance in electrical and electronic equipments, which is restricted by the Restriction of Hazardous Substances (RoHS) directive. Since EPD is performed using a water soluble photoresist in exposure reactions, polyvinyl alcohol ([-CH₂-CH(OH)-]_n) PVA and ADC in slurry can be introduced into the phosphor coating. When the photochemical oxidation of the PVA and the reduction of the ADC occur, cross-links are formed between the PVA chains, and the Cr³⁺ ions are generated by the reduction of the Cr⁶⁺ ion. Cr ions that are in the phosphor layer not only contaminate the irradiative recombination sites of phosphor, but also cause the light intensity to be low, since Cr⁶⁺ ions absorb light. However, the PS approach applies phosphor by exploiting mechanical principles without pollution risk of chemical reaction.

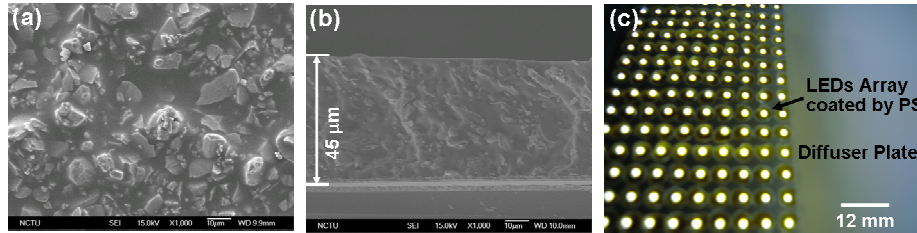


Fig. 8. SEM micrographs of phosphor coating using PS. (a) Top view; (b) cross-sectional view; (c) illuminated lighting module with phosphor coated by PS.

By EPD or self-exposure approaches, LED chips must be flip-chip mounted with a flat emitting surface [6]; the top of the LED chips must be free of wire bond pads. In contrast, LED chips that are coated by PS need not have a flat emitting surface, as shown in Fig. 3(b). Although the surface of the diode is still connected using gold wires to an electrode pad, PS can be applied uniformly to the surface of LEDs. Figure 8(a) presents a top view of a PS coated LED with phosphor particles in the silicone binder. Figure 8(b) presents a cross-sectional view with a uniform mean coating thickness of 45 μm and the left side of Fig. 8(c) is an illuminated demonstration of LEDs array on board coated by PS technique. The right side of Fig. 8(c) is CPC covered with another diffuser plate to get a planar illumination.

4. Conclusions

This study proposes a novel pulsed spray (PS) method of conformal phosphor coating (CPC) for LED illumination applications. The phosphor is applied directly to LED chips of the wire-bonded type. This approach is also applied to an array of blue LED chips as an inexpensive method with a high manufacturing throughput. The PS yields LEDs with superior optical characteristics including low color deviation ($\Delta u'v'$ of 0.07) when observed from various illumination angles (-60° to $+60^\circ$). Additionally, PS can yield a wide range of color temperatures (T_c) from 2500 K to 9500 K with high color accuracy, supporting a diversity of TFT-LCD backlights and general lighting applications. Meanwhile, unlike other coating techniques, such as dispensing, the settling method, spin coating, self-exposure and electrophoretic deposition (EPD), PS is a mechanical spray-based, environmentally friendly method that does not cause harmful ion pollution by chemical reactions. This study successfully demonstrates CPC by PS for future wLEDs lighting systems.

Acknowledgments

The authors would like to thank the KISmart Corporation (Hsinchu, Taiwan) for assisting in the conformal coating by pulsed spray (PS) technique and preparing the samples.