Detailed Study on Pulse-Sprayed Conformal Phosphor Configurations for LEDs

Article *in* Journal of Display Technology · June 2013

DOI: 10.1109/JDT.2012.2225019

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Detailed Study on Pulse-Sprayed Conformal Phosphor Configurations for LEDs

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***Abstract*—We report an investigation of the optical performance of a conformal-shaped phosphor coating fabricated using pulsed spray techniques on horizontal LED chips. Both the nitride and YAG:Ce phosphor mixed configuration and the multilayer configuration were studied. Also, 3535-packaged LED devices with the two typical phosphor configurations were prepared and analyzed. The results revealed that the LED devices with multilayer phosphor configurations emitted an average of 5.6% more radiant flux than those with mixed phosphor configurations. However, such LED devices presented an interesting reversed tendency in the luminosity measurements, which were 0.3 lm and 6 lm lower at 10 mA and 350 mA, respectively. Finally, the color rendering index (CRI) and correlated color temperature (CCT) homogeneity of the LED devices were further analyzed. A twofold reduction in CCT variation was observed compared with the conventional phosphor coating methods. It was found that the mixed phosphor LED devices demonstrated outstanding angular CCT distributions for viewing angles ranging from -80° to 0°, and a CCT variation of only 45 K was detected, while the multilayer phosphor coating had higher color rendering capabilities, reaching a $\mathrm{CRI}_{\mathrm{general}}$ value as high as 85.6. A promising guideline found through this work is that the pulse-sprayed conformal phosphor configuration would particularly be able to improve the light quality of LED devices by a significant amount: the mixed phosphor configuration achieves excellent CCT homogeneity, and the multilayer phosphor configuration reveals a novel concept for fabricating a low-CCT and high-CRI LED device with less nitride phosphor. The findings of our research should provide valuable insight to LED industries.**

***Index Terms*—Conformal coating, light-emitting diodes (LEDs), pulse-sprayed phosphor coating, multilayer phosphor.**

I. Introduction

LIGHT-EMITTING diodes (LEDs), especially the III–nitride LEDs, are now accelerating the process of upgrading general lighting to solid-state lighting (SSL) [1]–[3]. The recent progress [4]–[15] has strongly motivated the development of phosphor-based white LEDs. The advances in addressing the internal quantum efficiency and efficiency-droop [4]–[10], material quality [11], [12], and light extraction [13]–[15] issues in InGaN QW LEDs were important for enabling the development of high-performance and practical pump excitation sources in phosphor-based white LEDs. To continue competing with and eventually surpass the performance of conventional lighting systems [16], [17], the design and fabrication of LED devices still has a long way to go. Nowadays, the combination of GaN LED chips with phosphor materials to generate white light dominates most of the SSL market. With the increase in lighting applications, the requirements of LED devices in terms of high luminous efficiency and better light quality are becoming increasingly important. In such types of LED devices, a layer of phosphor materials is coated onto the chip from which light is emitted. Therefore the white light is made up of two components: 1) light that is emitted through the phosphor coating without being absorbed and 2) the converted light, e.g., yellow light in the case of the YAG:Ce phosphor [18] or red light in the case of nitride phosphor [19], [20] and borate phosphor [21]. It has been proved in the literature that the phosphor materials in LED packages did significantly affect the performance of the LED devices. Substantial research in this area has been focused on optimization of phosphor layer thickness [22], [23], concentration [24]–[26], particle size [27]–[30] and locations [31]–[33]. Some other methods have been reported that could further improve the light quality. A combination of different types of phosphors could overcome the color variation induced by the junction temperature rise by counter-balancing red-shifting and blue-shifting phosphors [34]. Also, the photoluminescence properties of $Ca_9Y(PO_4)_7$ doped with Eu and/or Sm phosphor showed a tunable blended emission of blue-green and orange-red light, which has great potential for fabricating LED devices of high color rendering index (CRI) [35].

Among the phosphor coating approaches, researchers found that the conformal-shaped phosphor coating would not only enhance the luminous efficiency [36] but also improve the angular correlated color temperature (CCT) distributions [37]–[39] and benefit the reliabilities [40], [41]. Such a phosphor configuration could be fabricated by using the self-exposure method [36], the capillary microchannel approach [42], and the pulsed spray technique [43]. From a practical point of view, the pulsed spray technique had excellent stability, and more importantly, it was cost effective. Consequently, this method was widely used in the latest CREE® product, X-TE [44]. However, studies of this method were limited, and they mainly focused on vertical [43] or specially shaped LED chips [45]. A demonstration of the application of this method to horizontal LED chips is still lacking in the literature.

Manuscript received August 26, 2012; revised October 07, 2012; accepted October 07, 2012. Date of publication December 11, 2012; date of current version April 29, 2013. This work was supported by the Key Program of NSFC-Guangdong Joint Funds of China, under Project U0834002; the National Nature Science Foundation of China, under Project 51005079; and Science and Technology Program of Guangdong Province, under Project 2010A080802007 and 2011A081301006.

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Digital Object Identifier 10.1109/JDT.2012.2225019

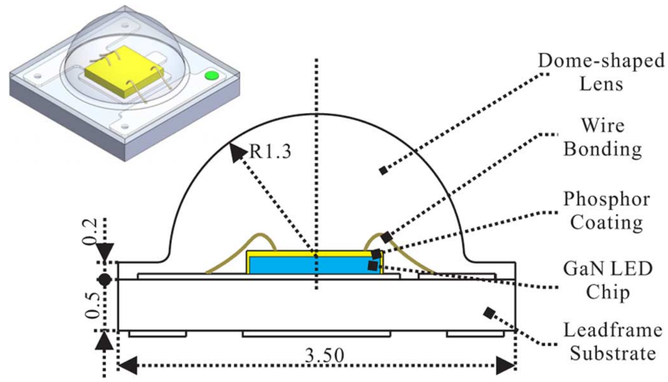

Fig. 1. Schematic of the high-bright white LED package.

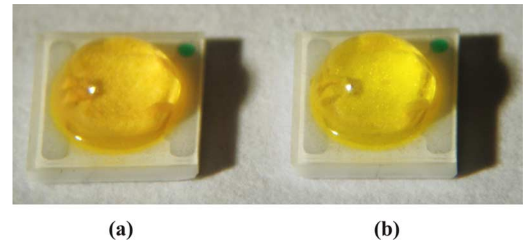

Fig. 2. Experimental LED devices with (a) the YAG:Ce/nitride mixed phosphor configuration and (b) the YAG:Ce multi-layer phosphor configuration.

In addition, the previous work had reported that the multilayer phosphor configuration (consisting of a layer of red phosphor in the immediate vicinity of the LED chips and a yellow phosphor layer covering the red one) was one of the most interesting methods which could achieve a higher radiant flux and higher luminous flux compared with the mixed phosphor method [33]. However, the CCT of those tested samples was about 4700 K, and the ratio of the red phosphor to the yellow phosphor is only 1:21.4; the resulting emission spectra were not significantly influenced by the red phosphor. We believe it would be more interesting if the performance of this method were evaluated on warm-white LED devices (around 2700 K) because more red phosphor would be dispensed to the package, and the red light emitted from the phosphor would significantly influence the performance of LED devices, resulting a color variation of light with different phosphor coating configurations.

In the following, we extend the previous investigations and discuss experimental aspects of conformal-shaped phosphor coatings fabricated on horizontal LED chips via pulsed spray techniques. Both the red and yellow phosphor mixed configuration and the multilayer configuration were studied.

II. Experiments

A. Led Device Preparation

Commercially available 3535-packaged devices structures were applied in our experiments, as shown in Fig. 1. High-brightness GaN-based blue LED chips (HB LED chips) manufactured by Epistar®, with a size of $1143\times1143\times150$ μm and a peak wavelength of 452 nm, emitting an average radiant flux of 400 mW @ 350 mA were used along with two kinds of phosphors: YAG:Ce phosphor and nitride phosphor for converting blue light to yellow-green light and red light, respectively. The white light LED devices were fabricated as follows. 1) The LED chip was attached to the leadframe substrate via the silver-paste die-bonding technique. 2) The anodes and cathodes on the chip were connected to the positive and negative pads on the leadframe substrate by gold-wire bonding. 3) In some cases, a mixture of YAG:Ce phosphor, nitride phosphor, and silicone was conformally sprayed on the chip to form the mixed phosphor configuration; in other cases, a bottom layer of nitride phosphor and silicone mixture and an upper layer of YAG:Ce phosphor silicone mixture was used to form the multilayer phosphor configuration. The overall ratio of YAG:Ce phosphor, nitride phosphor, and silicone are 44.5 wt%, 15.5 wt%, and 40 wt%, respectively. 4) The conformal phosphor coating was cured in an oven at a temperature of 150°C for 1.5 hours. 5) The dome-shaped lens was molded to protect the HB LED chip. The sprayed materials in each component include 0.36 mg YAG:Ce phosphor, 0.12 mg nitride phosphor, and 0.32 mg silicone. In order to ensure that the mass fraction of each material was kept constant, the weights of the phosphor-silicone coatings were measured, and their mass densities were found to be 15.12, 5.04, and 10.08 mg/cm^2 for the YAG:Ce/nitride mixed layer, nitride layer, and YAG:Ce layer, respectively. These two types of LED devices with different phosphor configurations are shown in Fig. 2, in which Fig. 2(a) presents the LED device that was coated with a YAG:Ce/nitride mixed phosphor (mixed Y/N LED), and Fig. 2(b) shows the LED device that had a nitride and YAG:Ce multilayer phosphor configuration (multilayer N-Y LED). As is evident, the multilayer N-Y LED appeared yellowish due to being completely covered on the surface by yellow YAG:Ce phosphor. For both types of LED devices, the phosphor coatings were conformally shaped by using the pulsed spray procedure [43]. Hemispherical lenses were utilized to enhance the light extraction efficiency since the total internal reflection effect would significantly reduce the lumen output [46], [47].

B. Experimental Measurements

The two types of LED devices were spectrally measured with an integrating sphere from Instrument System®. A KESSLY® adjustable DC source was used to provide the drive current. The drive current in our experiments ranged from 10 mA to 350 mA in steps of 10 mA. In order to minimize the effect of the increased junction temperature, LED devices were mounted to the standard metal core printed circuit board (MCPCB) and kept at a temperature of 25 ± 0.5 °C by attaching the MCPCB to a semiconductor refrigerator, as shown in Fig. 3. In addition, there was an interval time of 5000 ms separating one step from the next, which means that the LED devices were measured under pulsed current conditions, which should not influence the junction temperature significantly [48], [49]. The radiant flux, luminous flux, color rendering index (CRI), typical spectrum at a drive current

Fig. 3. Spectral measurements of LED devices.

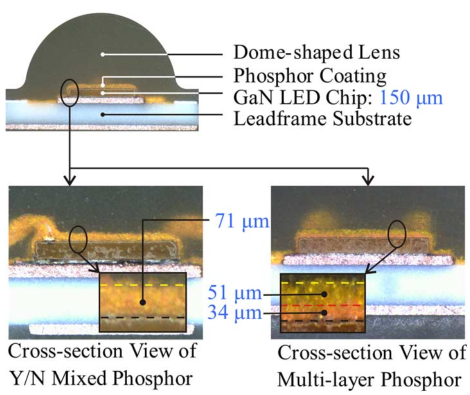

Fig. 4. Cross-section view of different types of phosphor configurations.

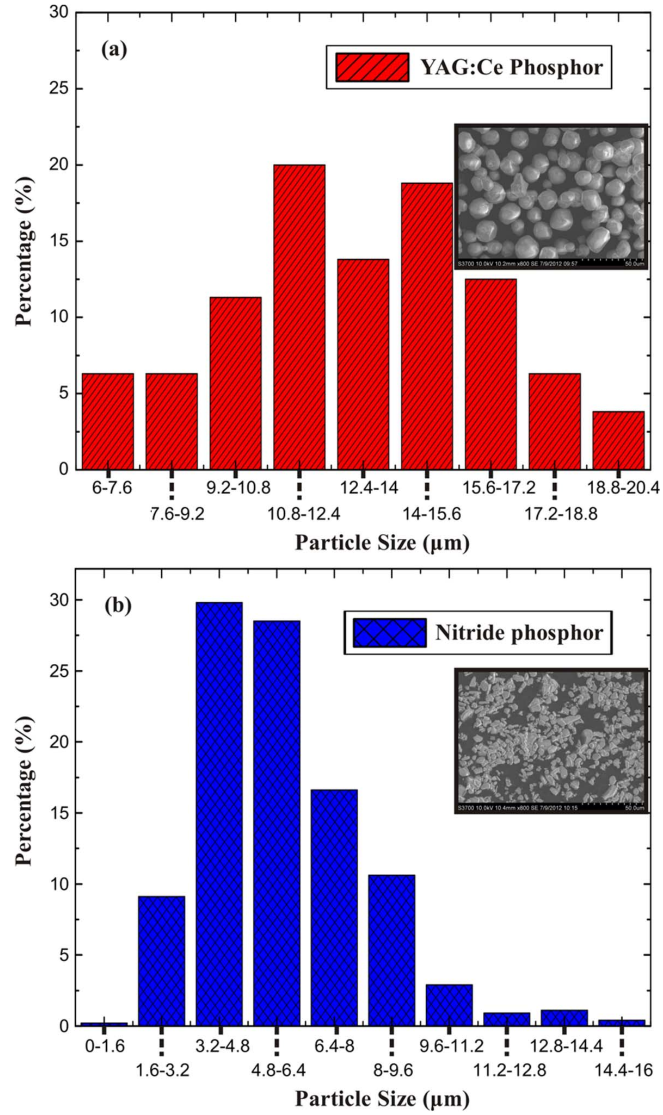

Fig. 5. Particle size distribution of: (a) YAG:Ce phosphor and (b) nitride phosphor.

of 350 mA, and the CCT, as well as its angular distributions, were measured and analyzed.

III. Results and Discussion

Fig. 4 reveals the cross sections of different phosphor configurations. The phosphor coating was basically fabricated as we designed it. The total thicknesses of the mixed phosphor and of the multilayer phosphor are 71 μm and 85 μm, respectively. The surfaces of the LED chips are coated with a layer of phosphor material with a constant thickness, and the bonded wires are inevitably covered by phosphor powders. Interestingly, we find that the sidewalls of the chips are only coated with a very thin phosphor layer. This could be reasonably explained by the decrease in the adhesive forces when the phosphor silicone mixture was sprayed onto these walls. In the multilayer configuration, the YAG:Ce phosphor layer occupies more than half of the total thickness, and the boundary between the YAG:Ce phosphor and the nitride phosphor is blurred. Fig. 5 shows the particle size distributions of the different kinds of phosphors that are used in our experiments. It is obvious that the particle size of the YAG:Ce phosphor is much larger than that of the nitride phosphor. Taking both the phosphor coating thickness and the particle size into consideration, we may infer that the stacking concentrations are in the following order: $C_{\mathrm{nitride}} > C_{\mathrm{YAG:Ce/nitride}} \gg C_{\mathrm{YAG:Ce}}$. This is because the space between two YAG:Ce phosphor particles is possibly filled by the small nitride phosphor particles.

The radiant flux and luminous flux versus the drive current are shown in Fig. 6. The radiant flux results indicate that the LED devices with multilayer phosphor configurations emit more light (energy) than those based on the mixed phosphor coating. The former have a higher radiant flux of 4.9% more than the latter at 10 mA, and they increase slightly as the current rises. Finally, the maximum enhancement of the multilayer N-Y LED reached 5.9% at a current of 350 mA, exhibiting an average increase of 5.6%. However, the luminous flux has a contradictory tendency.

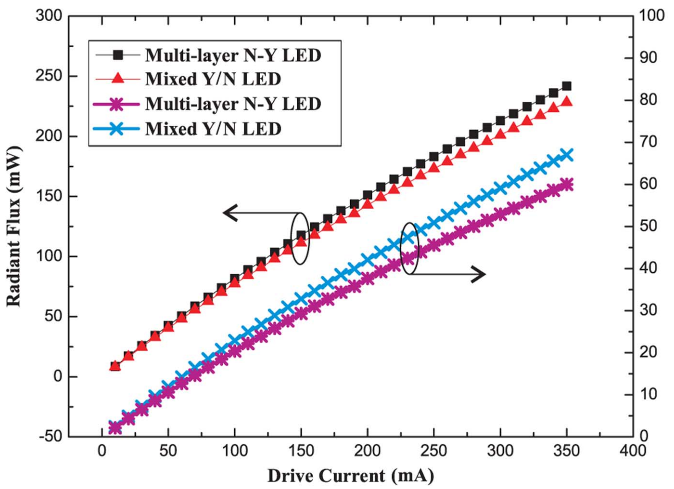

Fig. 6. Radiant flux and luminous flux of the multi-layer N-Y LED and the mixed Y/N LED.

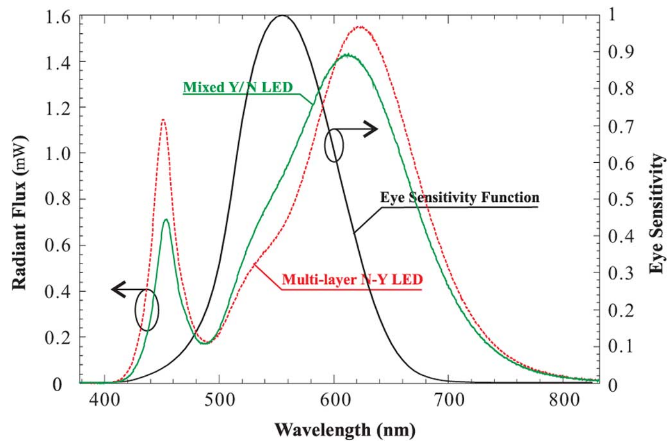

Fig. 7. Typical spectra of different types of LED devices and the eye sensitivity function.

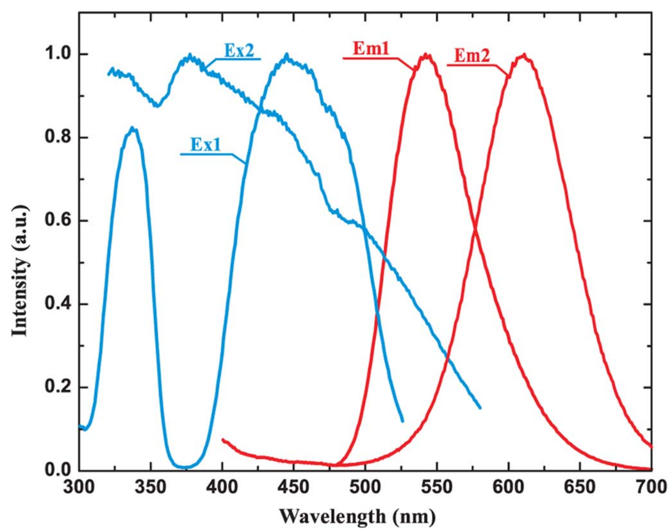

Fig. 8. Excitation spectra and emission spectra of the YAG:Ce phosphor (Ex1 and Em1) and of the nitride phosphor (Ex2 and Em2).

The mixed Y/N LED gives out 2.4 lm and 67 lm at 10 mA and 350 mA respectively, while the multilayer N-Y LED emits 0.3 lm and 6 lm less than the mixed Y/N LED, respectively, which is considerably different from the results of the current report [33].

Fig. 7 reveals typical spectra of different types of LEDs at 350 mA. The results show that a significantly higher radiant power is measured around the blue peak wavelength of the LED chips (452 nm) with the multilayer phosphor configuration. We believe that the decrease in radiant energy of the mixed Y/N LEDs is mainly due to the high concentration caused by the compact stacking properties of the large and small phosphor particles [26]. We also investigated the spectrum characteristics of the phosphors, as shown in Fig. 8. The YAG:Ce phosphor absorbs blue light at the peak wavelength of 450 nm and down-converts the photons to 533 nm yellow-green light automatically. However, the absorption of the nitride phosphor stretches from blue light to yellow-green light. This means that the emitted light from the YAG:Ce phosphor may be significantly reabsorbed by the nitride phosphor, which will induce energy loss [33], and such a phenomenon becomes notable when the YAG:Ce phosphor particles are surrounded by nitride phosphor in mixed configurations. Considering the discussion above, we can properly explain the radiant flux enhancements of multilayer N-Y LEDs by using the combined influence of the phosphor concentration, the particle size effect, and the reabsorption phenomenon.

The luminous flux, $\Phi_{\rm lum}$ could be calculated from the radiant light power using (1) [50]

$$\Phi_{\rm lum} = 683 \frac{\rm lm}{\rm W} \int_{\lambda} V(\lambda) P(\lambda) d\lambda \tag{1}$$

where $P(\lambda)$ is the power spectral density (light power per unit wavelength), and $V(\lambda)$ is the eye sensitivity function, which is also plotted in Fig. 7. The results indicate that within the wavelength range from 510 nm to 610 nm, where the human eye is the most sensitive (the normalized eye sensitivity is larger than 0.5), the mixed Y/N LEDs emits 12.93 mW more radiant power than the multilayer N-Y LEDs and emits 8.08 lm more than the multilayer LEDs in luminous flux measurements. However, for wavelengths below 510 nm and above 610 nm, even though the radiant power of the multilayer N-Y LEDs is 25.09 mW higher, the luminous flux is only 1.47 lm brighter. More interestingly, we find that the second peak wavelength around 610 nm is shifted from 612 to 623 nm if we change the mixed YAG:Ce/nitride phosphor configuration to the multilayer configuration. The above discussions clearly demonstrate that when the nitride phosphor is stacked close to the LED chip more red light will be emitted resulting an considerable red-shifted phosphor spectrum of multilayer N-Y LEDs. And the spectrum is far away from the visual sensitive zone (510 to 610 nm), so the luminous flux is decreased. Such a red-shifting phenomenon would become notable in the cases in our study where there is a high ratio of nitride phosphor (which is 7.3 times higher than that in [33]). This is the reason why we obtained a result contradictory to what was reported in [33].

We also investigated the phosphor efficiency under different drive currents. Theoretically, the efficiency of the phosphor coating should be defined as the emitted radiant power divided by the absorbed radiant power of the phosphor layer [51].

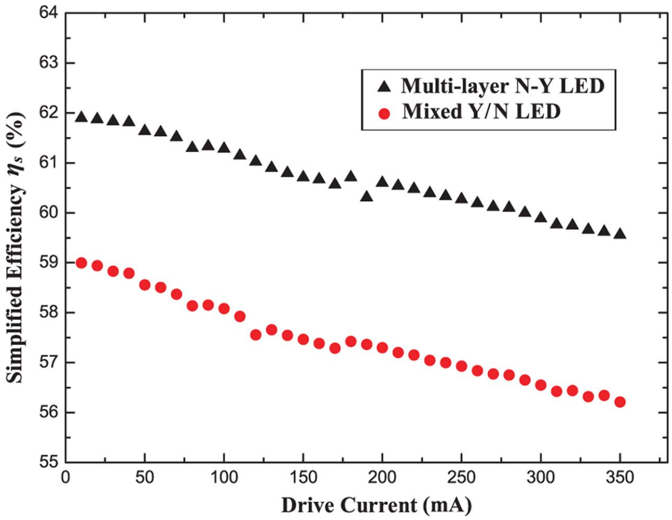

Fig. 9. Simplified efficiency of the multi-layer N-Y LED and the mixed Y/N LED.

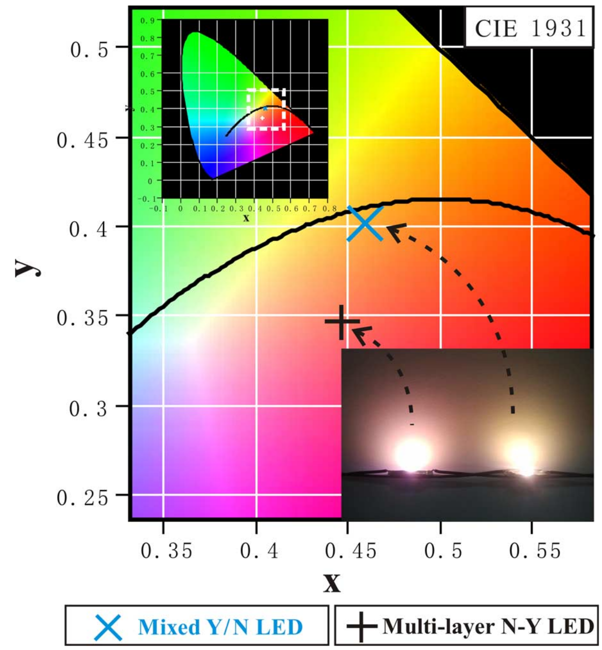

Fig. 10. Chromaticity coordinates of the multi-layer N-Y LED and the mixed Y/N LED in the CIE 1931 chromaticity diagram.

However, it is complicated to separate these two components because of the complexity of the band filling and band gap variation under different drive currents [50]. Therefore, we assessed the phosphor efficiency by using a simplified approximate approach, wherein the efficiency was calculated as follows:

$$\eta_s = \frac{P_B + P_C}{P_{B0}} = \frac{P_W}{P_{B0}} \tag{2}$$

where P_{B0} is the radiant power of devices without a phosphor coating and P_W is the radiant power with a phosphor coating comprising a component of unabsorbed blue light (P_B) and a component of converted light emitted by phosphors (P_C). The η_s values of different phosphor configurations are shown in Fig. 9. Under low drive-current conditions, the η_s values are 61.9% and 58.9% for multilayer N-Y LEDs and mixed Y/N LEDs respectively. These values decrease as the drive current increases, falling to 59.6% and 56.2% when the drive current rises to 350 mA, resulting in average η_s values of 60.7% and 57.4%, respectively. These results reveal that the efficiency of the multilayer phosphor coating is a little higher than that of the mixed one, confirming our discussion about the integral radiant flux. Moreover we found a drop in η_s for different phosphor configurations, and we think this is perhaps because of the partial saturation effects induced by the phosphor close to the emitting surface of the chip [52].

The color rendering capabilities of the LED devices ($\mathrm{CRI}_{\mathrm{general}}$, CIE 1995) are 82.8 and 85.6 for mixed Y/N LEDs and multilayer N-Y LEDs, respectively. The higher value of $\mathrm{CRI}_{\mathrm{general}}$ the multilayer N-Y LEDs have, the better color displaying properties it exhibits, which is a characteristic that would be preferable in exhibition lighting applications. Meanwhile, six supplemental test-color samples (referred to as CRI_i for integer values of i ranging from 9 to 14) are used to further assess the color rendering properties. The six CRI_i values of the multilayer N-Y LEDs are 56.5, 98.4, 83.2, 73.3, 97.1, and 94.8, respectively; which are also much higher than those of the mixed Y/N LEDs (where $[\mathrm{CRI}_9, \mathrm{CRI}_{10}, \ldots \mathrm{CRI}_{14}] = 25.9, 75.9, 74.6, 67.0, 83.8,$ and 96.7) especially for CRI_9. The CRI_9 value of the multilayer N-Y LEDs is 2.2 times as high as that of the mixed Y/N LEDs. This means that the multilayer N-Y LEDs are particularly suitable for displaying strong red (CIE 1995).

Finally, we studied the CCT of LED devices, as shown in Fig. 10. The mixed Y/N LEDs emit much more yellow-green light and less strong red light than the multilayer N-Y LEDs. As a result, the overall CCT of the multilayer N-Y LEDs is much lower than that of mixed Y/N LEDs, and their CCT values are 2369 K and 2653 K, respectively. Obviously, the chromaticity coordinates of the mixed Y/N LEDs are much closer to the Planckian locus, and the color appears to be tending toward yellow, while the multilayer Y-N LEDs have a much lower y coordinate and appear reddish in the CIE 1931 chromaticity diagram. Consequently, we can easily tell the difference between these two types of LEDs just by examining the colors of their light, as can be seen by inspection in Fig. 10. Furthermore, Fig. 11 shows the angular CCT distributions. As the devices present good symmetrical CCT distributions in both the X and Y directions, the angular CCT distribution was tested for viewing angles from $-90°$ to $0°$. The results indicate that the average CCT values of the multilayer N-Y LEDs in the X and Y directions are 2223 K and 2235 K, respectively, while the same values of the mixed Y/N LEDs are 2542 and 2514 K, respectively, which are about 100 K lower than the overall CCT values. Between the central area and the fringe, the CCT values are quite homogeneous, and the maximum CCT changes are only 257 K and 225 K for the multilayer N-Y LEDs and the mixed Y/N LEDs, respectively. We can infer that the insufficient thickness of the phosphor materials on the chips' sidewall is the main cause of the variation of the CCT values. Such

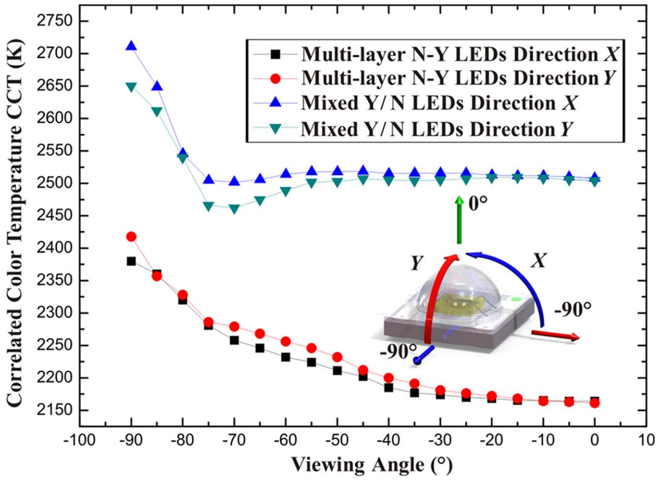

Fig. 11. Angular distribution of CCT values of the multi-layer N-Y LED and the mixed Y/N LED.

variations are more notable in multi-layer phosphor configurations because a longer operation time is necessary for preparation of samples, which had led to more phosphor particle losses due to the influence of gravity. However, compared with the conventional phosphor coating method reported by Steranka *et al.* [37]–[39] and Zheng *et al.* [42], although the angular CCT distribution is not perfect, an approximate twofold reduction in CCT variation is observed, reducing it to the point that it is not easily detectable by the human eyes. In particular, the mixed Y/N LEDs present an excellent angular distribution for viewing angles ranging from -80° to 0°, and a CCT variation of only 45 K is observed (the light at angles exceeding -80° is difficult to utilize, and it only contains less than 5% of the total energy).

Based on the analysis of the CCT and CRI values, promising guidelines have been obtained. First, the mixed Y/N LEDs produce a much better CCT homogeneity in a larger range of viewing angles, which is preferable for high-end lighting applications. Second, the multilayer phosphor configuration results in emission spectra consisting of more red light. Therefore, from the mass production point of view, it is possible to fabricate a low-CCT and high-CRI LED device with less nitride phosphor, which will lead to a lower cost. This is because nitride phosphor is extremely difficult to produce [53], [54], and its price is 10–30 times higher than the price of the YAG:Ce phosphor.

IV. Conclusion

The present study has experimentally investigated conformal-shaped phosphor coatings fabricated on horizontal LED chips via pulsed spray techniques. 3535-packaged LED devices with mixed nitride and YAG:Ce phosphor configurations and multilayer phosphor configurations were measured and discussed. Our results indicate that the multilayer N-Y LEDs emit an average of 5.6% more radiant flux than the mixed Y/N LEDs. Interestingly, the luminous flux reveals a reversed tendency, and the outputs of the multilayer N-Y LEDs are 0.3 lm and 6 lm lower at 10 mA and 350 mA, respectively. These findings could be attributed to the combined influence of the following factors: 1) the compact stacking properties of small nitride phosphor particles and large YAG:Ce phosphor particles; 2) the reabsorption phenomenon between nitride phosphors and YAG:Ce phosphors; and 3) the emission spectrum shift induced by different phosphor configurations. Moreover, the efficiencies of these two kinds of LED devices are discussed via a simplified approach, which shows that there is about a 2% efficiency drop in either of the configurations. Also, the chromaticity properties demonstrate that the pulse-sprayed conformal phosphor configuration would improve the light quality of the LEDs, especially the angular CCT distributions. The maximum CCT variations are only 257 K and 225 K for the multilayer N-Y LEDs and the mixed Y/N LEDs, respectively, which are about twofold lower compared with that of the conventional phosphor methods. Finally, a promising guideline was obtained: The mixed phosphor is recommended for high-end lighting applications because of the excellent CCT homogeneity, while the multilayer phosphor is preferable for its potential for cost reduction in LED mass productions by reducing the amount of nitride phosphor, as the higher phosphor conversion efficiency (more red light) is observed in such configurations. There are still shortcomings of our studies, e.g., we are still not able to precisely control the thickness of the phosphor layer on the chip's side walls, which need further study. We hope this paper will arouse the readers' interest in working on the conformal phosphor coating methods for horizontal LED chips, which are urgently required in the LED industry.

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