Enhance the chromatic uniformity and luminous efficiency of WLEDs with triple-layer remote phosphor structures

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ABSTRACT

The angular color uniformity (ACU) with the ability to evaluate chromatic performance of WLED has become an important target to achieve in producing higher-quality WLEDs. This paper studies the ACU enhancing effects of novel triple-phosphor configuration in lighting devices with remote phosphor structure. Moreover, the optical influences of remote phosphor structure with three phosphor layers (TL) on WLEDs properties are calculated and compared to the dual-layer (DL) one for reference. The experiments are applied to devices at 5 distinct correlated color temperature ranging from 5600-8500 K. The results presented that DL structure attains better color rendering index (CRI) than the TL one. Meanwhile, in terms of color quality scales (CQS), TL model shows higher values at all ACCTs, compared to the DL. Moreover, the luminous flux of DL configuration is lower than that of TL structure. In addition, the diversion of color temperature depicts as D-CCT in TL structure is much better than the value in DL structure, especially at high ACCT as 8500 K, which means TL is good for chromatic uniformity of high ACCTs WLEDs. These results proved that the triple-layer structure is superior and more effective to apply for acquiring the enhancement of WLEDs package.

Keywords:
Color rendering index
Dual-layer phosphor
Luminous efficacy
Mie-scattering theory
Remote-phosphor
Triple-layer phosphor

1. INTRODUCTION

Due to the efficiency in lighting and energy-saving, the diodes with light radiating ability called LEDs has soon recognized as potential lighting method. The applications of LEDs are spread out in many different fields from general usage of street lighting, back-lighting to more advanced utilization in medical and automobile [1-3]. To produce white light, the most common approach is to apply the combination of chromatic lights which yielded from the blue LED chip and the phosphor that ejects light downward. Such configuration is distinguished based on the appearance of phosphor component and known as phosphor-converted LEDs (pc-LEDs). Although the procedure to apply phosphor on the structure and create pc-LEDs are available which consists of the dispersing and conformal methods, these approaches all have drawbacks that are detrimental to pc-LEDs quality. For instance, the luminous efficiency of WLEDs from these methods are usually poor due to the light loss caused by back-scattering lights [4, 5]. Then, a configuration that separate the phosphor material and blue light source was introduced to enhance light output and known as the remote phosphor (RP) structure. The RP structure creates a gap between the components that helps reduce the re-emission from phosphor, and therefore is able to enhance the optical properties of pc-LEDs [6, 7]. In addition, the RP configuration can reduce the absorption of back-scattered light by the LED chip, thus promoting the reliability of WLEDs and decreasing the junction temperature [8-13].
In recent years, the optical properties of RP LEDs have been significantly improved, especially their luminous efficiency. However, their chromatic uniformity is still in low-quality, especially for planar RP configurations [14-16]. In general, a diffuser sheet is a tool to maintain uniformity in allocation and radiation of correlated color temperature (CCT). However, the results show the energy loss in the process of light transmission through the sheet [17-19]. Though resulting in low light efficiency, these approaches get better color uniformity. Besides, although angular color uniformity (ACU) can be adjusted through many different configurations that have been proposed such as the patterned sapphire substrate (PSS) [19], the DBR structure [20], and the dichroic filter that recycles light [21], but high expenses and complexity when implement on an available illuminating network are the disadvantages of these configuration. Therefore, to create a cost-efficient structure that is easy to integrate and has high color uniformity and luminous output, the remote micro-patterned phosphor (RMPP) layer is proposed as the alternative for the standard flat RP film in WLEDs. The RMPP layer is a structure with multiple layers where the flat RP film is laid in the middle of the micro-patterned (MP) polydimethylsiloxane (PDMS) layers. Adapting this idea, dual-layer remote phosphor structures were proposed and investigated to achieve better optical performance. A dual-layer RP is comprised of a yellow YAG:Ce3+ layer placed under a red or green phosphor one.

In 2017, Nhan and his partners reported that structure with red phosphor SrO.3B2O3:Sm2+ in the dual remote phosphor can enhance light output by 17% in comparison to the conventional phosphor configuration. Besides, in the stable concentration of red phosphor SrO.3B2O3:Sm2+, the structure can achieve 5% higher color rendering index (CRI) than the mixed phosphor WLEDs [22, 23]. However, this study just showed the better results of CRI which is one of elements using to evaluate the color quality. Thus, it cannot completely assure the benefit of this structure to color quality of WLEDs, though the luminous flux is better. Another value called color quality scales (CQS) which includes CRI, viewer’s preference and color coordinates is consider a more powerful index to evaluate the color quality of WLEDs. Thus, Lee et al. introduced the two packages of dual-layer remote phosphor structures to enhance the lumen output, CRI and CQS of WLEDs. One of them uses SrBaSiO2:Eu2+ green phosphor layer while the other uses red phosphor Sr2F2B2O7: Eu2+,Sm2+ layer to place above the yellow YAG:Ce3+ layer. The results showed that the green-yellow structure can increase the green light component to better the luminous flux but it leads to a large decline in color quality. Meanwhile, the red-yellow model has better rendering ability that expresses through CRI, CQS and resulting in better color quality, although inferior in lumen efficacy. Besides, Anh’s team also researched on the different shapes of phosphor layers in the structure which are the flat dual-remote phosphor (FDRP) with planar layer and the concave dual-remote phosphor (CDRP) with indented layer. They finally figured out that compared to CDRP structure, the FDRP can accomplished better luminous flux as the light go through two phosphor layers more easily. They also suggested the suitable distances which are d1 = 0.08 mm or d2 = 0.63 mm to be the gap among the two phosphor layers and the led chip’s surface to achieve highest luminous flux. However, they also proposed a significant reduction in the color uniformity in both cases. In general, dual-layer remote phosphor can enhance the optical properties of WLEDs, but it hard to attain better luminous flux and color quality at the same time. Therefore, a triple-layer remote phosphor is proposed to improve those aspects in LED production. A triple-layer remote phosphor structure consists of three different phosphor layers: the yellow YAG:Ce3+ is placed at the bottom, covering the nine LED chips, while the red phosphor film is located at the top and the green phosphor layer is the middle. That this structure includes green and red phosphor, not only the green but also the red-light components are increases, leading to the possibility of achieving both higher lumen output and color uniformity simultaneously. Hence, this study will investigate the use of triple-layer remote phosphor to enhance the performance of WLEDs. Moreover, comparisons between the effects of DL structure and TL structure on luminous flux, CRI and CQS will be demonstrated to assured the benefits of the using three phosphor layers inside the LED packages. Thus, we believe that the results from this paper are valuable information for manufacturers to produce WLEDs having better performance.

The phosphor structure in this study is a triple remote phosphor with yellow YAG:Ce3+, green Ca2La3BO6:5Pb2+, and red Mg2Ge2O7F2: Mn4+ phosphor layers. The arrangement of studied contents is preparation process of green and red phosphor and the organization of these phosphor layers will be presented in section 2. Section 3 includes the mathematic system used to analyze the performance of the remote phosphor layer, and discusses the attained results. Finally, the conclusion is demonstrated in section 4.

2. RESEARCH METHOD
2.1. Preparation of phosphor materials

The green phosphor Ca2La3BO6:5Pb2+ is utilized to increase the internal green light component of WLEDs, and thus enhancing the luminous output and color uniformity. Besides, the red-light component inside WLEDs can be improved by using the red Mg2Ge2O7F2:Mn4+ phosphor, leading to higher values of CRI and CQS. The preparations of these two phosphors are demonstrated as follows.
To prepare Ca$_2$La$_2$BO$_{6.5}$:Pb$^{2+}$ phosphor, the steps must be put into a strict order. Initially, the materials are mixed by using dry grinding or milling method. Next, they are fired in open quartz boats with air flows, at about 500°C. After that, the mixture goes through powderizing process. Then, it is put into capped quartz tubes with N$_2$, and fired for the second time under 1000°C, in an hour. In the next step, this material will be powderized again. Finally, it will go through the third firing process at 1200°C with the presence of N$_2$ flows, for 1 hour.

For the preparation of Mg$_2$Ge$_2$O$_7$:Pb$^{2+}$, the process is simpler as it consists of fewer steps than that of Ca$_2$La$_2$BO$_{6.5}$:Pb$^{2+}$ phosphor. Firstly, the materials used in this process are mixed by dry ball-milling. Secondly, the mixture is fired with air flows in capped quartz tubes at 1200°C for 2 hours. Thirdly, the dry ball-milling method is used to grind the fired substance into powder. Finally, the powder is re-fired in open quartz boats at the same temperature 1200°C for approximately 16 hours (overnight).

Before simulating Ca$_2$La$_2$BO$_{6.5}$:Pb$^{2+}$ and Mg$_2$Ge$_2$O$_7$:Pb$^{2+}$ phosphor, measurements of the materials properties such as concentration, particle size, stimulation spectrum, absorption spectrum, and emission spectrum must be ensured by experiments. However, among the measurements, we need to identify the suitable concentration of phosphor and particle size to achieve the best index in chromatic performance and lighting output. Meanwhile, the parameters of the phosphor spectrum are constants. Based on the results from previous researches, the particles of phosphor have a width of 14.5 µm.

2.2. Simulation process

The cross-sectional schematics in Figure 1 are the illustration of LEDs packages used in the research. Figure 1(a), 1(b), respectively provides visual presentation on the arrangement of WLEDs package with glass cover, phosphor layers, a half-dome, and package with conformal coating. Specifically, the blue areas indicate lighting-emitting chips, the chromatic lines represent the phosphor of the corresponding color, and the empty spaces are the silicone matrix lenses. The substrate for the structure is predetermined as aluminum nitride and the default yellow phosphor is YAG:Ce$^{3+}$.

![Figure 1. Illustration of multi-layer phosphor structures of white LEDs, (a) dual-layer phosphor (DL) and (b) triple-layer phosphor (TL)](image)

This density of the phosphors layers is predetermined at 0.08 mm. Besides, the concentration of YAG:Ce$^{3+}$ changes following the variation of the red or green or red phosphor concentration to maintain the average correlated color temperatures (ACCTs). In addition, at each different ACCT of each phosphor structure, the YAG:Ce$^{3+}$ concentrations are diverse, which creates the diversity of scattering characteristic inside WLEDs. Moreover, this discrepancy leads to the differences in optical properties.

It can be seen from Figure 2 that at all ACCTs, the yellow YAG:Ce$^{3+}$ phosphor concentration in DL structure higher than in TL structure. Considering the same ACCT in all structures, if the YAG:Ce$^{3+}$ concentration is higher, there will be more back-scattering events, and the reduced emitted luminous flux is higher. On the other hand, when the concentration of YAG:Ce$^{3+}$ is raised, the imbalance among the three primary colors that produce white lights, including yellow, red and green, will appear, leading to the reduction in color quality. Hence, to advance the luminous flux and the chromatic quality of WLEDs, the back-scattering needs to be reduced by increasing the red-light component. Besides, the chromatic performance as well as the light output also under influences of green light. According to the mentioned information, it seems that the triple-layer phosphor structure is the most advantageous one, in terms of managing optical properties. In order to prove this assumption, the research team continues to provide other essential references to the remote phosphor structures. In Figure 3 is the measured emission spectra in both dual and triple-layer configurations. From the results, the emission spectrum of TL structure is higher than DL structure at several wavelength ranges which suggests that pc-LEDs with three phosphor layers has better lighting efficiency regardless of color performance.
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The emitted blue light and yellow light from light conversion for remote phosphor structure containing one phosphor layer with thickness described as \(2h\) are expressed as follows [24, 25]:

\[
P_{B2} = P_{B0}e^{-\alpha B2h} = P_{B0}e^{-2\alpha B2h}  
\]

\[
PY_2 = \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B2 - \alpha Y_2}[e^{-\alpha Y_2 h} - e^{-\alpha B2 h}] + \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B2 - \alpha Y_2}[e^{-\alpha Y_2 h} - e^{-\alpha B2 h}] 
\]

\[
= \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B2 - \alpha Y_2}[e^{-2\alpha Y_2 h} - e^{-2\alpha B2 h}] 
\]

The emitted blue light and yellow light from light conversion for remote phosphor package containing two phosphor layers with the layer thickness of \(h\) are measured with:

\[
P_{B3} = P_{B0}e^{-\alpha B2h}e^{-\alpha B3h} = P_{B0}e^{-2\alpha B3h} 
\]

\[
PY'_3 = \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B3 - \alpha Y_3}[e^{-\alpha Y_3 h} - e^{-\alpha B3 h}] + \frac{1}{2} \frac{\beta_3 P_{B0}e^{-\alpha B3h}}{\alpha B2 - \alpha Y_2}[e^{-\alpha Y_2 h} - e^{-\alpha B2 h}] 
\]

\[
= \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B3 - \alpha Y_3}[e^{-2\alpha Y_3 h} - e^{-2\alpha B3 h}] 
\]

\[
PY_3 = PY'_3, e^{-\alpha Y_3 h} + P_{B0}e^{-2\alpha Y_3 h} \frac{\beta_3}{\alpha B2 - \alpha Y_2}[e^{-\alpha Y_2 h} - e^{-\alpha B2 h}] 
\]

\[
= \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B3 - \alpha Y_3}[e^{-\alpha Y_3 h} - e^{-\alpha B3 h}] + \frac{1}{2} \frac{\beta_3 P_{B0}e^{-\alpha B3h}}{\alpha B2 - \alpha Y_2}[e^{-\alpha Y_2 h} - e^{-\alpha B2 h}] 
\]

\[
= \frac{1}{2} \frac{\beta_3 P_{B0}}{\alpha B3 - \alpha Y_3}[e^{-2\alpha Y_3 h} - e^{-2\alpha B3 h}] 
\]

The \(h\) depicts the phosphor layer density. The subscript “1” and “2” note the single layer and double-layer remote phosphor configurations. \(\beta\) is the conversion coefficient for blue light converting to yellow light. \(\gamma\) is the reflection coefficient of the yellow light. The intensities of blue light \((PB)\) and yellow light \((PY)\) are results obtained from light intensity of blue LED, indicated by \(PB_0\). \(\alpha B\), \(\alpha Y\) are indices describing the loss of transmitted energy in blue and yellow lights during their scattering process in their respective phosphor layer. Besides, in subscript “4”, \(PY_3\) is the amount of yellow light that has passed two phosphor materials.

In TL remote structure pc-LEDs, there is a notable improvement in lighting efficiency when compared to a dual-layer one that can be seen from the following:

\[
\frac{(PB_B-2PB_Y)-(PB_2+PY_2)}{PB_B+PY_2} > \frac{e^{-2\alpha B2 h} - e^{-2\alpha Y_2 h}}{e^{-2\alpha Y_3 h} - e^{-2\alpha B3 h}} > 0 
\]

The Mie-theory is used in the analysis of scattering of phosphor particles. Moreover, through the following expression that applied the Mie theory, the scattering cross section \(C_{ext}\) for circular particles is obtained. In addition, the efficiency of light power transmission can be defined with Beer’s law [25]:

\[
I = I_0 \exp(-\mu_{ext}L) 
\]
phosphor concentration to keep the ACCT. At that time, the TL structure can reduce the internal back-scattering lights, leading to the blue light from LED chips easily transmit through YAG:Ce\textsuperscript{3+} layer to other layers. In other words, TL structure helps the blue light power from LED chips to be converted effectively. Thus, compared to DL model, TL structure achieves higher emission spectrum intensity in the wavelength band of white light, resulting in better performance in its luminous flux. Hence, the TL structure can be chosen due to its superior in optical properties of WLEDs, including CQS and LE.

There are various methods to improve the color uniformity, which is an important element for color quality, such as scattering enhancement particles SiO\textsubscript{2}, CaCO\textsubscript{3}, etc., or conformal phosphor structure. When applying the green Ca\textsubscript{2}La\textsubscript{2}BO\textsubscript{6}:Pb\textsuperscript{2+} phosphor and red Mg\textsubscript{8}Ge\textsubscript{2}O\textsubscript{11}F\textsubscript{2}:Mn\textsuperscript{4+} phosphor, the scattering properties and chromatic light components inside WLEDs are increased, leading to generating better white light. Additionally, utilizing the remote phosphor structure can promote the emitted luminous flux due to the reduction in the back-scattering of light to the LED chip. Although these two approaches can improve the color uniformity, the luminous flux seems to decrease. Therefore, the phosphor concentration needs to be adjusted to an appropriate amount to achieve the highest power transmission, which can be proven by the Lambert-Beer law in (7). In Figure 7 is the comparison of color deviation between DL and TL structures. As can be seen, the color deviation of TL structure is much smaller than that of DL structure, especially, at high ACCTs such as 8500 K. The fact is that the smaller the deviation is, the higher the color uniformity becomes; and this statement can be demonstrated by the internal scattering of WLEDs: when more phosphor layers are applied, more scattering events occur. As a result, the chromatic uniformity of WLEDs increases. However, the rise in scattering event can lead to a decline in the luminous output. Nevertheless, this decrease is insignificant compared to the advantages given to WLEDs when the back-scattering is reduced. Therefore, the TL structure can attain either the best color uniformity or the highest luminous flux.

4. CONCLUSION

This article compares the optical efficiency of the structures DL and TL at five ACCTs. The green Ca\textsubscript{2}La\textsubscript{2}BO\textsubscript{6}:Pb\textsuperscript{2+} and red Mg\textsubscript{8}Ge\textsubscript{2}O\textsubscript{11}F\textsubscript{2}:Mn\textsuperscript{4+} phosphors are applied during the simulating process. Besides, the research results are verified by the Mie theory and the Lambert-Beer law. According to the results, adding the green Ca\textsubscript{2}La\textsubscript{2}BO\textsubscript{6}:Pb\textsuperscript{2+} phosphor layer increases the green light component to get the color uniformity, and luminous output enhanced. Thus, the TL structure results in better luminous flux and chromatic uniformity than the DL one. Moreover, color rendering index and color quality scale, two most important color quality indicators are improved with the presence of red Mg\textsubscript{8}Ge\textsubscript{2}O\textsubscript{11}F\textsubscript{2}:Mn\textsuperscript{4+} phosphor in the configuration. The result shows that the TL structure has lower CRI but better and CQS than the DL. It can be easily seen that the chromaticity depends on the balance among the three primary colors yellow, red, and green. Therefore, the TL turns out to be the best selection in controlling these three colors. Besides, the reduction in back-scattering of TL structure causes its luminous flux to increase noticeably. The proof is that TL presents the highest value of luminous efficiency. In other words, TL structure can increase the luminous efficiency and color quality of WLEDs at the same time, which could not be accomplished by using DL structure. Based on the results of this study, manufacturers can easily choose the most appropriate structure to enhance the quality of WLEDs.
REFERENCES

[1] S. Pan, et al., "Image restoration and color fusion of digital microscopes," Appl. Opt., vol. 58, pp. 2183-2189, 2019.
[2] O. Kunieda and K. Matsushima, "High-quality full-parallax full-color three-dimensional image reconstructed by stacking large-scale computer-generated volume holograms," Applied Optics, vol. 58, pp. G104-G111, 2019.
[3] Q. Zaman, et al., "Two-color surface plasmon resonance nanosizer for gold nanoparticles," Optics Express, vol. 27, pp. 3200-3216, 2019.
[4] D. Durmus and W. Davis, "Blur perception and visual clarity in light projection systems," Optics Express, vol. 27, pp. A216-A223, 2019.
[5] R. Hirayama, et al., "Projection of multiple directional images on a volume structure with refractive surfaces," Optics Express, vol. 27, pp. 27637-27648, 2019.
[6] T. Z. Wu, et al., "Analyses of multi-color plant-growth light sources in achieving maximum photosynthesis efficiencies with enhanced color qualities," Optics Express, vol. 26, pp. 4135-4147, 2018.
[7] Y. L. Piao, et al., "Chromatic-dispersion-corrected full-color holographic display using directional-view image scaling method," Appl. Opt., vol. 58, pp. A120-A127, 2019.
[8] P. Zhu, et al., "Design of circadian white light-emitting diodes with tunable color temperature and nearly perfect color rendition," OSA Continuum, vol. 2, pp. 2413-2427, 2019.
[9] D. Lin, et al., "Silicon solar cells efficiency enhanced in NIR band by coating plasmonics ITO- and UC phosphor-particles layers on back-side surface using spin-on film deposition," CLEO: Applications and Technology Optical Society of America, 2019.
[10] T. Li, et al., "Efficient X-ray excited short-wavelength infrared phosphor," Optics Express, vol. 27, pp. 13240-13251, 2019.
[11] Q. T. Fouliard, et al., "Modeling luminescence behavior for phosphor thermometry applied to doped thermal barrier coating configurations," Applied Optics, vol. 58, 2019.
[12] Z. J. Zhang, et al., "Tunable photoluminescence in Ba1-xSrxi3Si3O4N2: Eu2+/ Ce3+, Li+ solid solution phosphors induced by linear structural evolution," Optical Materials Express, vol. 9, pp. 1922-1932, 2019.
[13] A. K. Dubey, et al., "Laser-line-driven phosphor-converted extended white light source with uniform illumination," Applied Optics, vol. 58, pp. 2402-2407, 2019.
[14] G. Zhang, et al., "Spectral optimization of color temperature tunable white LEDs with red LEDs instead of phosphor for an excellent IES color fidelity index," OSA Continuum, vol. 2, pp. 1056-1060, 2019.
[15] B. Wang, et al., "Eu3+ doped high-brightness fluorophosphate laser-driven glass phosphors," Optical Materials Express, vol. 9, pp. 1749-1762, 2019.
[16] F. Steudel, et al., "Pixelated phosphors for high-resolution and high-contrast white light sources: erratum," Optics Express, vol. 27, pp. 9097-9098, 2019.
[17] B. Fond, et al., "Investigation of the tin-doped phosphor (Sr,Mg)3(PO4)2:Sn2+ for fluid temperature measurements," Optical Materials Express, vol. 9, pp. 802-818, 2019.
[18] H. Yuce, et al., "Phosphor-based white LED by various glassy particles: control over luminous efficiency," Optics Letters, vol. 44, pp. 479-482, 2019.
[19] W. Wang, et al., "Red photoluminescent Eu3+-doped Y2O3 nanospheres for LED-phosphor applications: Synthesis and characterization," Optics Express, vol. 26, pp. 34820-34829, 2018.
[20] T. Wei, et al., "Single Pr3+-activated high-color-stability fluoride white-light phosphor for white-light-emitting diodes," Optical Materials Express, vol. 9, pp. 223-233, 2019.
[21] A. Dwivedi, et al., "Monochromatic NIR UC emission in Tm3+/Yb3+ co-doped GdVO4 phosphor: the effect of the Bi3+ ion concentration and pump power of a diode laser," Optics Letters, vol. 43, pp. 5785-5788, 2018.
[22] J. S. Li, et al., "High efficiency solid-liquid hybrid-state quantum dot light-emitting diodes," Photonics Research, vol. 6, pp. 1107-1115, 2018.
[23] A. Zhang, et al., "Tunable white light emission of a large area film-forming macromolecular complex with a high color rendering index," Optical Materials Express, vol. 8, pp. 3635-3652, 2018.
[24] B. Li, et al., "High-efficiency cubic-phased blue-emitting Ba3Lu2B6O15:Ce3+ phosphors for ultraviolet-excited white-light-emitting diodes," Optics Letters, vol. 43, pp. 5138-5141, 2018.
[25] X. Wang, et al., "Broadband multicolor upconversion from Yb3+-Mn2+ co-doped fluorosilicate glasses and transparent glass ceramics," Optics Letters, vol. 43, pp. 5013-5016, 2018.