Y$_2$O$_3$:Ho$^{3+}$ and ZnO:Bi$^{3+}$: a selection for enhancing color quality and luminous flux of WLEDs

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ABSTRACT

As the luminescence industry develops, the white light light-emitting diode (LED) package with a single chip and a single phosphor although produces good luminous flux but has a poor color rendering index (CRI) can no longer fulfill the requirements of modern lighting applications. Therefore, this research is conducted to response to the urgent demands of improving other lighting qualities of WLED while maintaining high luminous efficiency. To achieve this target, we applied the new WLED package, which contains multi-chips and multi-phosphor layers, and have obtained outstanding results in both CRI and luminous efficacy. Two types of phosphor used in the WLED package are Y$_2$O$_3$:Ho$^{3+}$ and ZnO:Bi$^{3+}$. A color configuration model is also developed to adjust the shading of the white-light LED module. The results of this research show that the triple-layer phosphor has the best performance when applied in a white-light LED package, which is demonstrated through better color quality, CRI and luminous efficacy. The manufacturers can rely on this research to produce the optimal-quality WLED, or WLED that is appropriate to their quality demands.

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

INTRODUCTION

High-brightness white-light light-emitting diode (LED) lamps are utilized worldwide as a general light source. LEDs are utilized in various lighting circumstances, for example, in car lighting, low-temperature lighting, and street lighting, in view of their long lifetime, high lighting capacity, durability when exposed to vibration, environmental friendliness, low power utilization, and compact design [1-5]. As of late, their quick switch properties have brought about their utilization in keen lighting, another programmed control innovation. The color rendering index (CRI) is a basic factor for evaluating the light quality of a LED light source in examination with regular daylight or black body radiation. A high CRI indicates the wide outflow range of a phosphor, which is like the persistent range of black body radiation [6-8]. A few strategies are accessible to get white light by blending LEDs and phosphors. The least expensive and earliest method involves a solitary blue chip and a solitary yellow phosphor; notwithstanding, these bundles have poor CRIs and have been enhanced by utilizing one red and one blue chip with a solitary yellow phosphor [9-12]. Because of WLED wide application range and development potential, there are numerous research that study WLED in attempts to enhance it optical properties. While some research show positive results in enhancing one part of the WLED, there are still other drawbacks that had not been attended to. For example, to enhance the luminous efficiency there are research that regard the distance between phosphor layers in flat
dual-remote phosphor (FDRP) structure and concave dual-remote phosphor structure (CDRP), triple-remote phosphor structure, and the influences of phosphor on the controlling the chromatic light output of WLED with conformal phosphor packages. To adjust the shading and CRI, there are numerous reports about blending phosphors and LEDs to streamline and anticipate the range. The examination with experiments and scientific models regarding the decay of the LED and phosphor spectrum are generally used to optimize the light color and CRI [13, 14]. Be that as it may, it is a research to create a white LEDs with changeable correlated color temperatures (CCTs) that can achieve the adequate value of CRI [15]. A blue LED with two phosphors is utilized to accomplish a high CRI, yet the adequacy is low because of the Stokes shift. Blue and red LEDs with one phosphor have a high CRI and high durability, however, come up short on the capacity to tune the shading contrasted in comparison to the two phosphors [16]. As can be seen, there are lots of research focus on the improvement of WLED quality, however, they often demonstrated on one particular quality indicator while neglecting others and there are no comparison between different package configurations, which can cause confusion to the manufacturers. Accordingly, we suggests, for the first time, a white-light LED package with two sorts of LEDs and two phosphors with the expectation to acquire higher CRI, high luminous efficacy while retaining a tunable color. The mechanism of this method is to keep the advantages found in previous studies and discard the unwanted traits by evaluating the performance of WLED with the new phosphor package while adjusting the phosphor arrangement until reaching the highest values in quality indicators. We also based on Beer's law [17–19] to propose an innovative and exclusive shading configuration model that changes to response to the request for an unobtrusive color distinction of white-light LEDs. This new method demonstrates through experimental results and comparison between important optical properties the superior results that can be achieved by changing the phosphor package and phosphor concentration, which is an approach that has not been applied before. The detail of the experiments presented in this paper are divided into 3 parts: the introduction of phosphor materials and the WLED models, the measured results presented in charts, and the discussion on the obtained results. In this occasion, white-light LED modules were manufactured utilizing yttrium aluminum garnet (YAG:Ce$^{3+}$) [20, 21] and nitride-based phosphors in blue and red LEDs with high CRI and luminous efficacy. Different extents of phosphors and different densities of phosphors in silicone with two kinds of red LEDs were utilized in the manufacture of the white LEDs. The outcomes exhibit that the proposed color configuration model can be effectively settled and connected. This research is a condensation of find from previous studies and it results are useful in guiding the manufacturers towards an updated and optimal manufacturing solution to produce a high quality WLED.

2. RESEARCH METHOD

2.1. Preparation of phosphor materials

Green phosphor Y$_2$O$_3$:Ho$^{3+}$ is used to increase green light component in WLEDs, leading to increased luminous flux and color uniformity. Red phosphor ZnO:Bi$^{3+}$ is used to increase red light component in WLEDs, leading to increased CRI and CQS [22, 23]. The process of preparing these two phosphor types is as follows: To create green phosphor Y$_2$O$_3$:Ho$^{3+}$ particles, first we need to mix by slurrying in water or methanol, then dry in air and collect the powder when it dry. Continue to fire in capped quartz tubes with stagnant air at 1300°C for 1 hour, resulting in powder. The preparation process to produce red phosphor ZnO:Bi$^{3+}$ is mixing by dry grinding. There are two ways: the first is to fire in open air at 800°C within 1 hour; the second is to fire in capped quartz tubes with stagnant air at 1120°C for 2 hours. Finally, store the final product in well-sealed containers. Before performing the optical simulation of particles Y$_2$O$_3$:Ho$^{3+}$ and ZnO:Bi$^{3+}$, the input parameters such as phosphor concentration, phosphor particle size, excitation spectrum, absorption spectrum, and phosphor emission spectrum, should be accurately determined by experiments. Out of the five parameters above, concentration and size of phosphor are the unknown measurements that we need to achieve the highest color and luminous flux quality of the LED. Spectral parameters are constant. Based on the results of the previous study, the diameter of phosphor particles is fixed at an average of 14.5 µm.

2.2. Simulation process

The schematic cross-segment of conformal-coating pc-WLEDs are shown with a remote-arch, a remote-plate, a half-vault, in Figures 1(a), 1(b), 1(c) and 1(d), separately. The blue spots are the LED chips, the yellow concealed areas are the phosphor and the clear areas are the silicone mixture joining focal points. Figure 1(f) records the normal parameters of these pc-WLEDs while Figure 1(e) illustrate their physical model [24, 25]. The substrate is assumed to be aluminum nitride. The phosphor is assumed to be YAG:Ce$^{3+}$. To maintain the consistency of the examination, the CCTs of each type of WLED are 5600 K and 8500 K when seeing along the z-axis.
These remote phosphor layers are 0.08 mm thick. In order to maintain the average correlated colors (ACCTs), YAG:Ce³⁺ concentrations change with the changes of yellow or red phosphor concentration. At each different ACCT for each phosphor structure, YAG:Ce³⁺ concentrations also different. This makes the difference in scattering properties in LED, resulting in differences in optical properties. It is easy to see in Figure 2, the concentration of yellow-emitting YAG:Ce³⁺ phosphor is the highest in the YL structure and lowest in the RGYL structure at all ACCTs. Consider the same ACCT in the remote phosphor structures, if the YAG:Ce³⁺ concentration is higher, the higher the scattering capacity increases, resulting in less luminous flux. On the other hand, there will be an imbalance between the three primary colors that produce white yellow, red and green light if the YAG:Ce³⁺ concentration is high, causing a decrease in color quality of WLEDs. Therefore, to improve the luminous flux and color quality of WLEDs, reduce the backscattering and keep the three basic colors yellow, red and green balanced are the essential requirements. The color rendering index can be controlled by increasing the red light component. Therefore, luminous flux and color quality can be controlled by green and red light component. If so, does it seem that the triple-layer phosphor structure is the most convenient in controlling optical properties? To answer this question, the team continues to give other important information of remote phosphor structures, emission spectrum. There are significant differences in spectral emission in remote phosphor structures. The YL emission spectrum has the smallest intensity compared to the other three remote phosphor structures in all 5 ACCTs. This confirms that the YL structure can achieve the smallest luminous flux. In contrast, the RGYL structure has the largest spectral intensity in the wavelength range of 380-780 nm. In the range of 400-500 nm, GYL structure has a higher spectral intensity than RYL structure so GYL’s luminous flux can be higher than RYL in this wavelength range. However, RYL’s spectral emission intensity is higher than GYL in the range of 650-750 nm, which helps RYL to have a higher color rendering index than GYL. However, to confirm the above mentioned, it is necessary to consider the results achieved in section 3.
3. RESULTS AND ANALYSIS

Figure 3 shows the emission spectra of phosphor configurations, while Figure 4 shows the CRI comparison between remote phosphor structures. It is easy to see that the RYL structure achieves the highest CRI regardless of the ACCT. In particular, CRI increases with ACCT, therefore, is highest at 8500 K. This is an important result for improving CRI for remote phosphor structures. While CRI control is difficult to control high ACCT (greater than 7000 K), RYL class can do this as the CRI of RYL structure is benefited from the red light component added from the red phosphor layer ZnO:Bi$^{3+}$. The second position is RGYL structure regarding the CRI value achieved. Meanwhile, CRI is lowest in GYL structure. Once again, it can be affirmed, with the goal of CRI, it is advisable to choose RYL structure to produce WLEDs in bulk. However, CRI is just one of the color quality indicators. In recent years, CQS has become the target of research. CQS is a combination of three elements: CRI, the viewer's preference and color coordinates. With this three-factor coverage, CQS becomes a big target and "seems" to be the most important indicator to assess color quality. In this study, the CQS of the remote phosphor structures are compared in Figure 5. If the RYL reaches the highest CRI, the RGYL reaches the highest CQS. This can be explained by the balance of 3 basic colors yellow, red and green. The higher the CQS value, the higher the color quality. The CQS is the lowest in the YL structure. In general, the Y structure has a high luminous flux, but it is difficult to control the color quality because there are no additional red and green light components in this structure. Despite the disadvantage of color quality, YL structure has advantages in production. The production procedure will be simpler than the rest, and the production costs are also lower.

![Figure 3. Emission spectra of phosphor configurations](image1)

![Figure 4. Color rendering indexes of phosphor configurations corresponding to ACCTs](image2)

![Figure 5. Color quality scale of phosphor configurations corresponding to ACCTs](image3)

Based on the result of Figure 5, it can be confirmed that if the manufacturer's goal is color quality, it is recommended to select RGYL structure. However, if the color quality is better, will the luminous flux be affected? To answer this question, the team compared the luminous flux emitted between 1 layer and 2 layers structures. The luminous flux when using multiple phosphor layers is definitely higher than when using a single layer. The evidence is illustrated in the results of Figure 6, the structure Y reaches the lowest luminous flux of the four structures in all ACCTs. In contrast, the highest luminous flux is achieved in the RGYL structure. So there is no more doubt about RGYL’s optical gain even when the RGYL structure also has the best color quality. Standing second in optical gain is GYL structure thanks to the green phosphor $Y_2O_3:Ho^{3+}$ and ZnO:Bi$^{3+}$: a selection for enhancing color quality... (Nguyen Thi Phuong Loan)
Y₂O₃:Ho³⁺. Green phosphor Y₂O₃:Ho³⁺ helps to increase green light composition and increase spectra in the wavelength range of 500 nm - 600 nm. Clearly in this wavelength range, GYL’s intensity is greater than GYL and YL. Due to the smallest YAG:Ce³⁺ phosphor concentration in the RGYL structure to keep ACCT. Then, the RGYL structure reduces the amount of scattered light after the YAG:Ce³⁺ concentration decreases. Blue light rays from LED chips are easily transmitted straight through the YAG:Ce³⁺ layer through other layers. In other words, the RGYL structure helps blue light energy from the LED chip to convert efficiently. Therefore, the RGYL spectral intensity is highest compared to other remote phosphor structures in the white light wavelength range. Therefore, the luminous flux of the RGYL structure is also highest.

Thus, the RGYL structure can be selected due to the superior optical properties of WLEDs including CQS and LE. However, color uniformity is also a considerable factor when it comes to color quality factor. There are many methods to improve color uniformity, including using advanced scattering particles or using conformal phosphor configuration. Although it improved color uniformity, luminous flux can be significantly reduced if the two methods are to be used. The green Y₂O₃:Ho³⁺ phosphor and red ZnO:Bi³⁺ phosphor can both create more scattering events and add the green or red light component inside WLEDs to produce more white light. The use of the remote phosphor structure enhances the luminous flux emitted by reducing the back reflection of the LED chip. However, it is necessary to control the phosphor layer concentration to achieve the highest transfer energy. Figure 7 shows the comparison of color deviation between structures. The smaller the color deviation, the higher the color uniformity. It is clear that the color deviation of RGYL is the smallest. This can be explained by scattering inside the LED package before forming white light. The more phosphor layers, the more scattering events. The result is increased color uniformity of WLEDs. Having many scattering events is not always a positive trait as it can reduce the luminous flux. However, this reduction is negligible compared to the benefit obtained when the backscattering is reduced. Therefore, the RGYL structure with the most phosphor layers achieves the best color uniformity and the highest luminous flux. In contrast, the highest color deviation is expressed in the YL structure at all ACTTs.

4. CONCLUSION

This paper introduces a new research method by comparing the optical performance of four structures YL, GYL, RYL and RGYL at different ACCTs. The results has fulfill the demand for an overall assessment of phosphor configurations and show great potential for WLED optical improvements. The Green Y₂O₃:Ho³⁺ phosphor and red ZnO:Bi³⁺ phosphor are used in the simulation process to create WLEDs and the results were verified by the Mie theory and the Lambert-Beer law to ensure credibility of the study. Accordingly, adding green Y₂O₃:Ho³⁺ phosphor to enhance green light benefits the color uniformity and luminous flux. Therefore, the GYL structure achieves better luminous flux and color uniformity than the RYL structure. CRI and CQS can be improved when increasing the red light component through the red ZnO:Bi³⁺ phosphor. As a result, RYL structure achieves higher CRI and CQS than GYL. The color quality depends on the balance between the three primary colors: yellow, green and red. RGYL structure, which contains all 3 colors, can control the colors balance by adding or removing when needed. In addition, the low backward reflection rate of RGYL is also a notable advantage that can lead to significant increase in the luminous flux. The evidence is that the highest luminous flux is also achieved in the RGYL structure. With these results, it is certain that manufacturers should employ the RGYL configuration to create WLEDs.
with highest quality possible for it superior performance, while other options might be considered for cheaper production cost and specific advantages mentioned in the paper. These are important references as they satisfy the needs of improvement method for the LED lighting solution and provide the manufacturers a suitable structure in distinct cases to match the production demands of their WLEDs.

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