

The application of (Y,Gd)BO₃:Tb³⁺ and CaGa₂S₄:Mn²⁺ phosphors to remote white light-emitting diodes

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ABSTRACT

The remote phosphor structure is superior to the conformal phosphor and the in-cup phosphor in terms of lighting efficiency; however, managing the color quality of the remote phosphor structure has been a nuisance to the manufacturers. To address this problem, many researches were conducted and the results suggested that using dual-layer phosphor structure and triple-layer phosphor structure could improve the color quality in remote phosphor structures. The purpose of this article is to study which one between the two configurations mentioned above allows multi-chip white LEDs (WLEDs) to reach highest indexes in color rendering index (CRI), color quality scale (CQS), luminous flux (LF), and color uniformity. The color temperature of the WLEDs used for the experiments in this article is 8500 K. The result of this research shows that the triple-layer phosphor configuration has higher CRI, CQS, and LE and also able to reduce color deviation resulting in better color uniformity. This conclusion can be verified by analyzing the scattering features of the phosphor layers using the Mie-theory. Being verifiable increases the reliability of the research result and makes it a valuable reference for producing better quality WLEDs.

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1. INTRODUCTION

Phosphor-converted white light-emitting diodes (WLEDs) with outstanding features such as color consistency, high-energy efficiency, low manufacturing cost, and small size is expected to gain more popularity and replace other conventional lighting sources in the near future [1-4]. The blue light emits from the chip combine with the yellow light from the phosphor complement each other and create the WLEDs [5-7]. WLEDs can be widely used in solid-state lighting should the luminous efficiency be improved [8, 9]. To create white light, the free dispersed coating is the most commonly used method, this method sprays directly a mixture of transparent encapsulated resin and phosphor powder onto the phosphor package. The freely dispersed coating can reduce the manufacturing cost as well as managing the phosphor layer thickness; however, the problem is that this method cannot produce high-quality WLEDs [10].

As a result, the conformal coating method with more consistent color distribution is used as an alternative to create angular homogeneity of correlated color temperature (CCT) [11]. The problem is that the luminous efficiency of the conformal coating method is damaged due to the backscattering effect occurs within the WLEDs. The prior researches have already applied the separating method, isolating the chip from the phosphor layer of remote phosphor structures [12-14]. Improving extraction efficiency by using the internal reflection structure, a polymer hemispherical shell lens with an interior phosphor coating that can boost the extraction efficiency [15-17]. Moreover, attaching empty spaces onto the phosphor structure can benefit the luminous efficiency by directing the reflected light downward [18]. Besides luminous efficacy, other optical properties such as color rendering index (CRI), color quality scale (CQS) and color uniformity are also relevant to evaluate the quality of a WLED.

Therefore, the dual-layer phosphor configuration and triple-layer configuration are the two remote phosphor structures that are used to enhance these optical properties. For the dual-layer phosphor configuration, the structure consists of a yellow phosphor layer underneath a red or green phosphor layer. In the triple-phosphor layer, the yellow phosphor is placed at the bottom with the green phosphor in the middle and the red phosphor layer on top. The concentration of the phosphor layers also affects the luminous efficiency besides the configuration. In particular, when the concentration in the phosphor layers increases, it causes the light lost due to the rise of backscattering events in the phosphor layers. This effect will reduce the luminous efficiency of the WLEDs, especially those at lower CCTs [19, 20]. Therefore, the solution is to limit the unwanted loss of light and increase the amount of blue and yellow light produced.

Finding the optimal solution to enhance the optical features in WLED is a troubling task for manufacturers because there are so many different options as mentioned above, each with distinct advantages and disadvantages. Therefore, this research is dedicated to finding the best choice possible to improve the quality of WLEDs. The conclusions from this article will demonstrate how to develop each specific optical property to the manufacturers

2. EXPERIMENT AND SIMULATION DETAILS

The first experiment in this research is adding the green phosphor layer $(Y,Gd)BO_3:Tb^{3+}$ to add the green light component to the WLEDs, thus leading to an increase in luminous flux. The second experiment utilizes the red phosphor layer $CaGa_2S_4:Mn^{2+}$ to add more red light component and enhance the CRI and CQS. The physical model of the WLED used in these experiments is shown in Figure 1 (a). The WLED has 9 LED chips placed in the air gaps, each chip has a lighting capacity of 1.16 W at the peak wavelength of 453 nm. Any further information about the optical indexes of the LED configuration are listed in Figure 1 (b). In Figure 1 (c) there is the single-layer remote phosphor structure (Y) consisting of a yellow phosphor layer $YAG:Ce^{3+}$ covering the surface of LED chips. Figure 1 (d) demonstrates the dual-layer remote phosphor structure (YG) with a red phosphor layer $CaGa_2S_4:Mn^{2+}$ above a yellow phosphor layer $YAG:Ce^{3+}$.

Figure 1 (e) also demonstrates the dual-layer remote phosphor structure but with a green phosphor layer $(Y,Gd)BO_3:Tb^{3+}$ on top of a yellow phosphor layer $YAG:Ce^{3+}$ that covers the LED chips. The triple-layer remote phosphor structure is presented in Figure 1 (f) with the green phosphor layer $(Y,Gd)BO_3:Tb^{3+}$ between the red and yellow phosphor layers which are placed on top at the bottom, respectively. Each phosphor layer has a fixed thickness of 0.08 mm. When the concentrations of green and red phosphor fluctuate, the concentration of $YAG:Ce^{3+}$ needs to adapt to that change in order to maintain the average correlated temperature color (ACCT). At 8500 K ACCT in different phosphor structures, the amount of $YAG:Ce^{3+}$ concentration is also changed. This feature creates the distinctiveness in scattering properties of LED resulting in the diversity of optical characteristics.

From Figure 2, it is easy to identify that the concentration of yellow-emitting $YAG:Ce^{3+}$ phosphor is highest in structure Y and lowest in structure YRG. Judging at the same ACCT for all remote phosphor structure, the higher the concentration of $YAG:Ce^{3+}$, the greater the possibility of backscattering which can damage the luminous flux. On the other hand, the color quality of WLEDs is also degraded, the reason is due to the imbalance between the three basic colors fabricating white light, yellow, green and red caused by the high concentration of $YAG:Ce^{3+}$. Therefore, to solve the problems with luminous flux and color quality of WLEDs, limiting the backscattering effect and maintaining the balance of the three colors, red, yellow and green are advisable. As we know, the color rendering index can be manipulated using red light components from the red phosphor layer. Furthermore, the green light components can control the color uniformity and luminous flux. According to the statements above, the triple-layer phosphor seems to be the most convenient structure in adjusting optical properties. However, the emission spectrum is another important index relating to the remote phosphor structures that needed to be reviewed in order to confirm this theory.

The significant differences between the emission spectra of remote phosphor structures can be observed in Figure 3. The emission spectrum of Y structure has the lowest intensity compared to three other remote phosphor structures, which means the luminous flux of Y structure is the smallest. On the contrary, YRG structure obtains the highest emission spectrum intensity in wavelength from 380 nm to 780 nm. In the wavelength from 400 nm to 500 nm, the spectrum intensity of YG structure is higher than that of YR structure, therefore, the luminous flux of YG structure can be bigger than the luminous flux YR in this wavelength band. However, in the wavelength of 660 nm and 750 nm, that the intensity of emission spectrum produced by YR structure is greater than YG structure allows YR structure to achieve better color rendering index in this situation. In order to conclude the aforementioned findings, all the achieved results are evaluated in section 3.

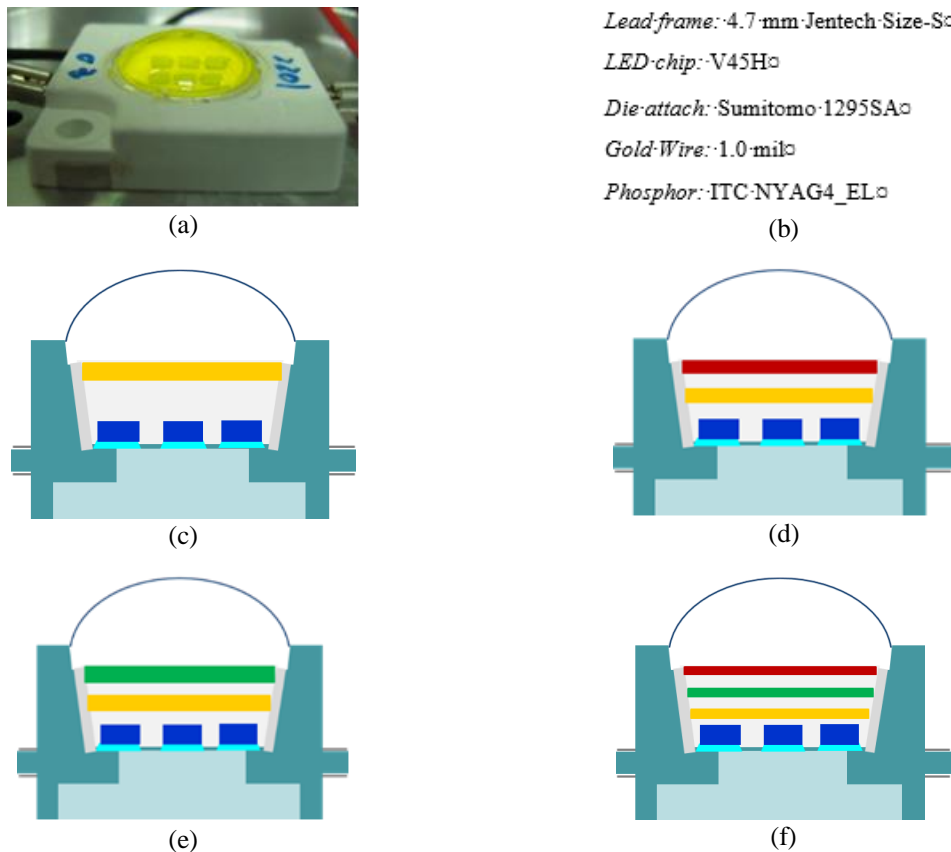


Figure 1. Illustration of multi-layer phosphor structures of white LEDs: (a) The actual MCW-LEDs, (b) its parameters, (c) Single-layer phosphor, (d) Dual-layer remote phosphor with YR, (e) YG, and (f) triple-layer phosphor

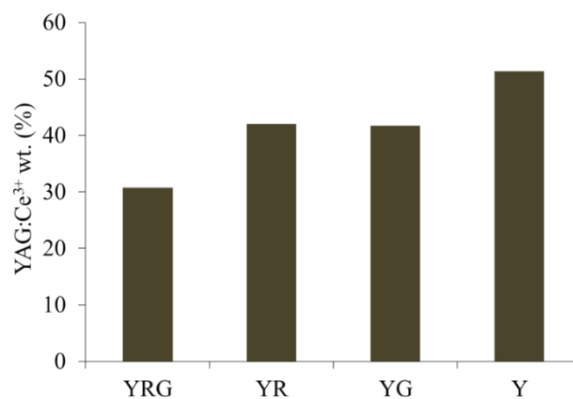


Figure 2. Concentration of yellow-emitting phosphor YAG:Ce³⁺ in

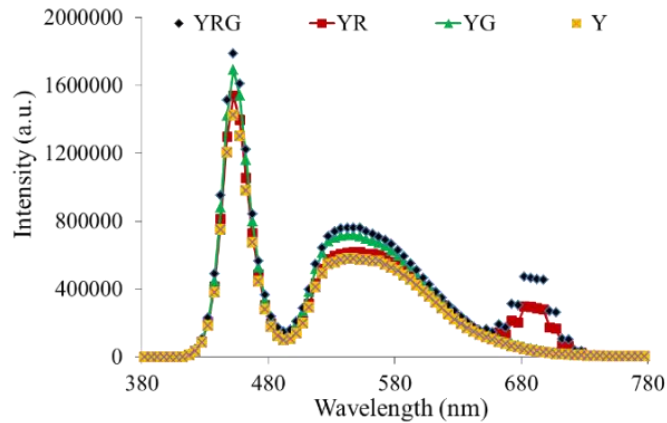


Figure 3. Emission spectra of remote phosphor configurations

3. RESULTS AND DISCUSSION

According to Figure 4 which compares the CRI of the remote phosphor structures, the YR structure stands out with the highest CRI achieved regardless of the ACCT. Controlling CRI at high ACCT (above 6600 K) is a significantly hard task to perform, yet the YR structure can benefit the CRI by adding red light component through the red phosphor layer $\text{CaGa}_2\text{S}_4:\text{Mn}^{2+}$. This result marks the progress of improving CRI in remote phosphor structure. Following the YR structure is the YRG structure with the second highest achievable CRI. Out of the 4 remote phosphor structures, the YG structure is the one having the lowest CRI. From these results, we can assume that choosing YR structure for mass-producing when having CRI as a goal is the best choice. However, CRI is only able to cover limited features of WLEDs and the quality of WLEDs cannot be judged base on CRI alone.

Therefore, a more thorough and in-depth measurement such as CQS, which is a combination of CRI, viewer's preference and color coordinate, is the main target for other research in recent years. In this part, the CQS is applied to all 4 remote phosphor structures and the results are shown in Figure 5. The result regarding the CQS in Figure 5 is different from the result in Figure 3 as YRG has the highest CQS among all remote phosphor structure instead of YR.

This is due to the better balance between the three basic colors: yellow, red and green that benefit the color quality and make the CQS rise. On the other hand, the lack of green and red light components in Y structure makes the color quality in this structure hard to control. As a result, the CQS in Y structure is the lowest despite the good luminous flux. Nevertheless, having lower color quality leads the Y structure to other advantages which are low manufacturing cost and easier to make compared to other structures.

From the content of Figure 5, the YRG structure is the most suitable option for manufacturing WLEDs with high color quality requirements. Using the YRG structure can enhance the color quality but the effect it has on the luminous flux is still unknown. To answer this question, the research group did an experiment to compare the luminous flux between single-layer and double-layer.

The purpose of this part is to present the mathematical model to compute the transmitted blue light and converted yellow light in the double-layer phosphor structure, and further developments for lighting efficiency can be proposed from this part. The transmitted blue light and converted yellow light for single layer remote phosphor package with the phosphor layer thickness of $2h$ are expressed as follows:

$$PB_1 = PB_0 x e^{-2\alpha_{N_1}k} \quad (1)$$

$$PY_1 = \frac{1}{2} \frac{\beta_1 x PB_0}{\alpha_{B_1} - \alpha_{Y_1}} (e^{-2\alpha_{Y_1}h} - e^{-2\alpha_{N_1}h}) \quad (2)$$

the transmitted blue light and converted yellow light for double layer remote phosphor package with the phosphor layer thickness of h are defined as:

$$PB_2 = PB_0 x e^{-2\alpha_{N_2}k} \quad (3)$$

$$PY_2 = \frac{1}{2} \frac{\beta_2 x PB_0}{\alpha_{B_2} - \alpha_{Y_2}} (e^{-2\alpha_{N_2}k}) \quad (4)$$

where h is the thickness of each phosphor layer. The subscripts “1” and “2” indicate single layer and double-layer remote phosphor package. β is the conversion coefficient for blue light converting to yellow light. γ stands for the reflection coefficient of the yellow light. The intensities of blue light (PB) and yellow light (PY) are the light intensity from blue LED, indicated by PB_0 . α_β ; α_γ are parameters illustrating the fractions of the loss blue and yellow lights during their scattering events in the phosphor layer respectively.

The lighting efficiency of pc-LEDs with the double-layer phosphor structure enhances considerably compared to a single layer structure:

$$\frac{(PB_2 + PY_2) - (PB_1 + PY_1)}{PB_1 + PY_1} > 0 \quad (5)$$

the Mie-theory is used to calculate the scattering effect of the phosphor components. In addition, the Mie theory [21-23] can also be applied to the following expression to compute the scattering cross section C_{sca} of spherical particles. The transmitted light power can be calculated by the Lambert-Beer law [24, 25]:

$$I = I_0 \exp(\mu_{ext}L) \quad (6)$$

in this formula, I_0 is the incident light power, L is the thickness of the phosphor layer thickness (mm) and μ_{ext} is the extinction coefficient, which can be expressed as: $\mu_{ext} = N_r \cdot C_{ext}$, where N_r is as the number density distribution of particles (mm^{-3}). C_{ext} (mm^2) is the extinction cross-section of phosphor particles. As Shown in (5) verified that using multiple phosphor layers is more beneficial to the luminous flux than employing a single-layer. This is also true regarding the results in Figure 6 in which the Y structure with only one phosphor layer has the lowest luminous flux. On the contrary, the highest luminous flux belongs to the YRG structure, which confirms that besides having the highest color quality the YRG structure is also favorable to luminous flux. The YG structure with green phosphor layer $(Y,Gd)BO_3:Tb^{3+}$ that helps to add green light component and to expand the emission spectrum at 500 nm and 600 nm wavelengths has the second highest record on the CQS.

Comparing the emission spectra of YG, YR and Y structure in this wavelength, the emission intensity of YG is higher than others, thus, resulting in better color quality. In YRG structure, the concentration of the yellow phosphor layer $YAG:Ce^{3+}$ is lower than in other structures to maintain the ACCT. This lower the back-scattering effect caused by the function of $YAG:Ce^{3+}$ which are allowing the blue light from the chips to easily pass through the yellow phosphor layer and reaching other layers at the same time. In other words, applying the YRG structure means the blue light from the LED chips can be transmitted more efficiently. The result of this is the YRG highest spectrum intensity among all other remote phosphor structures in the same white light wavelength band. The luminous flux of YRG structure, therefore, is also the greatest.

It is apparent that YRG is not only good for improving CQS but also LF due to its superior characteristics. However, color uniformity is also an important feature that needs to be considered when discussing color quality. Color uniformity is a feature that can be improved by various methods from adding scattering enhancement particles to employing the conformal phosphor configuration. Although these methods improve the color uniformity of WLEDs but can also damage the luminous flux, thus negatively affecting the lighting performance. Meanwhile, using green phosphor $(Y,Gd)BO_3:Tb^{3+}$ and red phosphor $CaGa_2S_4:Mn^{2+}$ in remote phosphor structure not only enhance the scattering properties of the WLEDs but also add red and green light components, which allows better white light to be created. Remote phosphor structure with suitable adjustments on the concentration of the phosphor layers to achieve the highest emission energy can limit the back-scattering effect, resulting in better luminous flux. The Lambert-Beer law in (6) can examine the result of this statement.

The comparison of color deviations between the remote phosphor structures is shown in Figure 7. From this, we can see that the remote phosphor structure with the higher color uniformity has lower color deviation. The YRG structure with 3 phosphor layers that boosts the amount of light scattering inside WLEDs before forming white light results in better color uniformity. Therefore, it is obvious that Figure 7 shows the color deviation index of YRG structure as the smallest. Of course, the decline in luminous flux can occur when there are too many scattering events appear in the WLEDs; however, these changes are not comparable to the advantage of lowering the back-scattering effect that the YRG structure provides. In fact, the YRG structure still obtains the best color uniformity and achieves the highest luminous flux. The Y structure, on the other hand, has the highest color deviation according to the results in Figure 7.

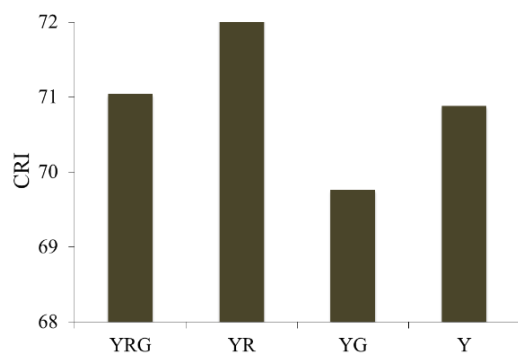


Figure 4. Color rendering indexes of remote phosphor configurations

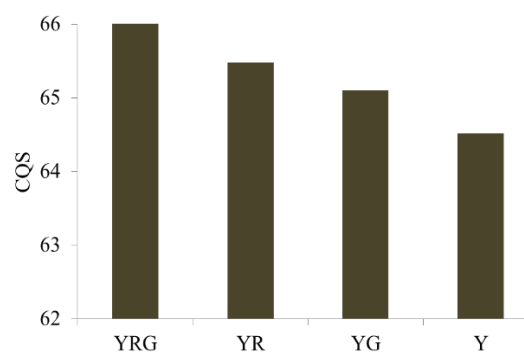


Figure 5. Color quality scale of remote phosphor configurations corresponding

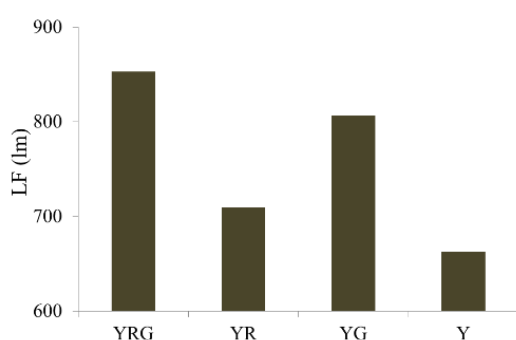


Figure 6. Luminous flux of remote phosphor configurations

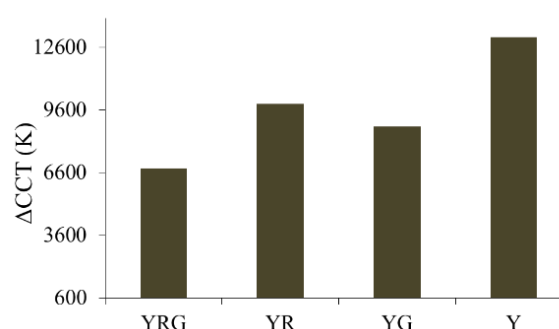


Figure 7. Correlated color temperature deviation (Δ CCT) of remote phosphor configurations

3. CONCLUSION

This article compares the optical properties of 4 remote phosphor structures: Y, YG, YR and YRG. The green phosphor $(Y,Gd)BO_3:Tb^{3+}$ and the red phosphor $CaGa_2S_4:Mn^{2+}$ are the components used to stimulate the results in the experiments and the accuracy of the results is verified by the Mie-theory and the Lambert-Beer law. According to the simulation results, using the green phosphor $(Y,Gd)BO_3:Tb^{3+}$ benefits the color uniformity and the luminous flux, it can be seen that these features of YG structure are higher than the YR structure. The red phosphor $CaGa_2S_4:Mn^{2+}$ is suitable for improving the CRI and CQS due to the addition of red light component from this phosphor layer. As a result, the CRI and CQS measurements from the YR are higher than YG. The color quality in YRG structure is highest among all structures; this can be explained by the ability to balance the three basic color, red, yellow and green of YRG structure, which is an important requirement for color quality. Moreover, the YRG structure also achieves the highest luminous flux as the back-scattering effect is reduced significantly in this structure giving it a considerable enhancement in luminous flux. The result of this research can be served as a valuable reference for the manufacturers in choosing the optimal structure to enhance their WLEDs quality.

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