Triple-layer Design of $YVO_4:Eu^{3+}$, $YF_3:Mn^{2+}$, and $YAG:Ce^{3+}$: the Potential for Advancement of Remote Phosphor Structure to Get Better Chromaticity and Luminescence at High Color Temperatures

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Abstract. *The remote phosphor structure is proven to be better than other structures of conformal or in-cup phosphor in terms of luminous efficiency. Nevertheless, its color rendering ability is unstable and difficult to manage at high correlated color temperature (CCT). Hence, improving the color management at high CCT for remote structure is essential. This study will present the triple-layer structures using the yellow phosphor* $YAG:Ce^{3+}$*, the red* $YVO_4:Eu^{3+}$ *and green* $YF_3:Mn^{2+}$ *phosphors to achieve that control over the color and luminous properties of WLED packages. The results of the triple-layer structure are compared with those of the conventional remote phosphor structure (single-layer). The experiments performed on these two structures with the CCT range of 5600–8500 K. The findings in the study show that the triple-layer is more beneficial to the color rendering metric, color quality scale, and also the lumen output, especially at high color temperatures such as 8500 K, compared to the conventional one. Moreover, the higher color uniformity is accomplished with the triple-layer structure as the color deviation in this structure is much smaller than in the single-layer. Thus, the triple-layer structure can be applied as a replacement to the single-layer to attain better control of the color quality and luminous flux for high-CCT WLEDs.*

Keywords

White LED, Lambert-Beer law, color rendering index, luminous efficacy.

1. Introduction

In recent years, the light source of light-emitting diodes (LEDs) is recognized as an effective and promising candidate for the next-generation light source [1, 2]. LEDs are used widely in many aspects of life, including art, transportation, and resident lightings. The broad application of LEDs can be attributed to their excellent properties, such as high lighting efficacy, robustness, small size, and being friendly to the environment as there is no gas or mercury released [3, 4]. In general, there are two common methods that are widely employed to fabricate a LED. The first method is combining red, green, blue chips and the second method is combining a LED chip with phosphor materials [5, 6]. The second method is known as phosphor-converted LED [7]. Usually, in this phosphor-converted LED package, the blue LED chip is combined with yellow $Y_3Al_5O_{12}$:Ce^{3+} (YAG:Ce^{3+}) phosphor because both of them have high lumen efficiency [8]-[10]. However, the performance of this LED package is greatly affected by the applied technique for the combining process. The most common approach is getting the phosphor freely dispersed on the LED chip. The benefit of this technique is the easy control over the phosphor layer thickness, yet the luminous output is not high as expected. Another technique is the conformal phosphor coating on the LED chip. The color rendering of this structure showed improvement, but low light extraction efficiency [11]-[13]. The problem of these two phosphor structures is that the phosphor layer contacts directly with the LED chip, which caused the increase in the thermal generation at their interface, leading to the fast decay of phosphor materials and large amounts of backscattered lights [14]. As the significant backscattering was reported, a considerable proportion of light trapped and lost in the phosphor layer consequently occurred, leading to inferior luminescence. The remote phosphor configuration, which grasps a simple concept of using the separation between the phosphor layer and the LED chip (also the heat source) to limit the thermal influence, has been believed to be an effective solution. The heat generation was reduced and the backscattering was minimized when the remote structure was applied, leading to the enhancement in the light extraction efficiency or the luminous output of LEDs. However, the color quality of the remote phosphor structure is hard to manage, especially in the LED light with high color temperature. The color rendering of this conventional remote structure was not high as other phosphor structures, such as conformal coating or in-cup phosphor. The conventional remote phosphor used only one yellow phosphor layer, thus it lacks the red spectral power in the spectral band of white light. Therefore, the color rendering index of single-layer remote phosphor structure is low, since high CRI requires high red light emission [15]. The double-layer with red phosphor was proposed for the enhancement of the color rendering ability of LEDs. Though this red-yellow double remote phosphor structure performed high CRI, which was 85 in case of 20% red $MgSr_3Si_2O_8$:Eu^{2+}, Mn^{2+}, its luminous was negatively influenced due to the increase of red phosphor concentration [16]. The green-yellow dual-layer structure can enhance the luminous flux yet the color quality is significantly declined. The idea of using three phosphor layers, including yellow, red, and green phosphors, was introduced to fabricate the novel remote phosphor structure. It was reported that the structure showed great control over the color balance with adequate luminous intensity [17].

Though the effectiveness of using a multilayer remote phosphor structure has been drawn, it remains concerned about the appropriate selection of phosphor materials and the range of color temperature for LEDs to get the most satisfying optical improvements. Thus, it is essential to analyze the effects of different phosphor materials at different color temperatures for the best applications. In this paper, we adopt the concept of using three phosphor layers for the LED structure and call it a triple-layer remote phosphor structure. The phosphor materials used in this study are the yellow phosphor YAG:Ce^{3+}, the red YVO_4:Eu^{3+}, and green YF_3:Mn^{2+} phosphors. Triple-layer phosphor structure has been made many times before, but in this article, we try to combine three types of phosphor that have not been mentioned in any previously published paper. The purpose of using this remote structure is to utilize the benefits of the remote phosphor, the red phosphor, and the green phosphor to promote the optical properties of white LEDs (WLEDs). The red phosphor YVO_4:Eu^{3+} was reported to exhibit strong red emission that peaks at 617 nm [18]. Additionally, the green phosphor YF_3:Mn^{2+} will provide more green lights to the white-light generation process. This is a novel study because many researchers previously used YF_3:Er^{3+}, Yb^{3+} instead of YF_3:Mn^{2+} as we used in this paper. We found Mn^{2+} has many advantages over Er^{3+},Yb^{3+} such as better luminous efficiency, easier and safer to use. Moreover, the ion Eu^{3+}

and Mn^{2+} are the popular ions with high efficiency, affordable cost, and have been widely utilized for improving the color rendering index of phosphors. Besides, we fabricate the single-layer structure as a reference for the comparison between the attained results. The phosphor preparation and organization are presented in Section 2. Then, the computation of transmitted and converted light efficiency, together with the discussion on the results, are provided in Section 3. In Section 4, conclusions on the lighting efficiency of WLED with the suitable remote structure are summarized.

2. Phosphor and led model preparation

2.1. Phosphor preparation

This part demonstrates the preparation processes and optical properties of green $YF_3:Mn^{2+}$, red $YVO_4:Eu^{3+}$, and yellow YAG:Cen^{3+} phosphor particles. The composition of each phosphor material is presented in Tab. 1, Tab. 2, and Tab. 3. The red $YVO_4:Eu^{3+}$ and green $YF_3:Mn^{2+}$ phosphors should be prepared first so that the WLED simulation process could be performed more effectively [19]. The red phosphor $YVO_4:Eu^{3+}$ composition includes Y_2O_3, Eu_2O_3, and NH_4VO_3 with the mol percentage and weight described in Tab. 1. To fabricate the red phosphor, all the listed chemical ingredients are mixed by dry grinding or milling. After that, the mixture is fired in an open quartz boat with air, at 1000^{o}C, in one hour, and is powdered subsequently. Then, the powder is fired again in the same container with air, in an hour and at 1200^{o}C. As soon as the firing finishes, the product is powdered again, before being washed in NaOH (or KOH)-water solution. Next, the obtained powder should be re-washed with plain water several times. After drying, the sample is fired in an open quartz boat, at 1200^{o}C, for 1 hour. The attained product is red-emitting $YVO_4:Eu^{3+}$ phosphor, having an emission peak at photon energy of 2 eV.

Tab. 1: Composition of red phosphor $YVO_4:Eu^{3+}$.

Ingredients	*Mole %*	*By weight (g)*
Y_2O_3	95 (of Y)	107
Eu_2O_3	5 (of Eu)	8.8
NH_4VO_3	110	129

The ingredients of green phosphor $YF_3:Mn^{2+}$ are demonstrated in Tab. 2. The characteristics of $YF_3:Mn^{2+}$ can be presented as follows: greenish emission color with emission peaks at 2.38 and 2.60 eV.

Tab. 2: Composition of green phosphor $YF_3:Mn^{2+}$.

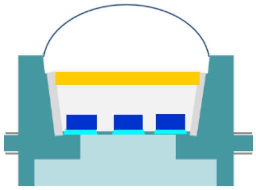
Ingredients	*Purity %*	*Purchased from*
Y_2O_3	> 99.99	Sinopharm Chemical Reagent Co. Ltd.
Eu_2O_3	> 99.99	
Tb_4O_7	> 99.99	
NH_4F	> 96.0	
C_2H_5OH	> 95.0	
$C_6H_8O_7{\cdot}H2O$	> 99.8	China Bei Jing Chemical Reagent Co. Ltd.
H_2NCONH_2	> 99.0	

Tab. 3: The composition of yellow phosphor $Y_3Al_5O_{12}:Ce^{3+}$.

Ingredients	*Mole %*	*By weight (g)*
Y_2O_3	100	10
Eu_2O_3	200	9.03
$Ce(NO_3)_3{\cdot}4H_2O$	2	0.555
B_2O_3	2	0.062

2.2. WLED simulation

The simulation of the triple-layer structure is created with the LightTools 9.0 software. The model of WLED with remote phosphor structure used in this study is fabricated based on the actual multi-chip WLED, as illustrated in Fig. 1. Specifically, the nine LED chips, having 1.6 W radiant power and 453-nm peak wavelength, are attached to the lead frame by die bonding. The usual remote phosphor utilizes a layer of yellow phosphor YAG:Ce^{3+}, while the proposed multi-layer remote phosphor is prepared with three different layers of phosphor, including the yellow phosphor YAG:Ce^{3+}, the



(a)

(b)

Fig. 1: Simulation of remote phosphor structure models: (a) Single-layer structure (SL), (b) Triple-layer structure (TL).

red phosphor $YVO_4:Eu^{3+}$, and the green phosphor $YF_3:Mn^{2+}$, as can be seen in Fig. 1-(a) and Fig. 1-(b), respectively. In the triple-layer (TL) structure, the phosphor layers are arranged following this color order: yellow-green-red, with yellow being the closest to the LED chips. In addition, each phosphor layer of the LED model is 0.8 mm in its thickness, and the concentration of used phosphors is varies depending on specific optical experiments and measurements. In particular, when adding the red and green phosphor to achieve better lighting performances, the concentration of yellow phosphor must be lowered as the concentrations of the others increase. This is to keep the stable color temperature of the LED packages, which is described in Fig. 2.

In Fig. 2, the concentration of the yellow phosphor layer decreases when the color temperature of WLEDs becomes higher. Moreover, the triple-layer phosphor structure shows a much lower $YAG:Ce^{3+}$ concentration than the single phosphor one, at all CCTs. The lower the yellow phosphor concentration, the lower the thermal generation in the WLED package. It leads to better optical efficiency [18]. In other words, the higher $YAG:Ce^{3+}$ concentration the inefficiency in light scattering, absorption, and extraction of the package, and thus degrading the lighting performance of LEDs. This may imply that the triple-layer could be more beneficial to the LED lighting efficiency than the single-layer.

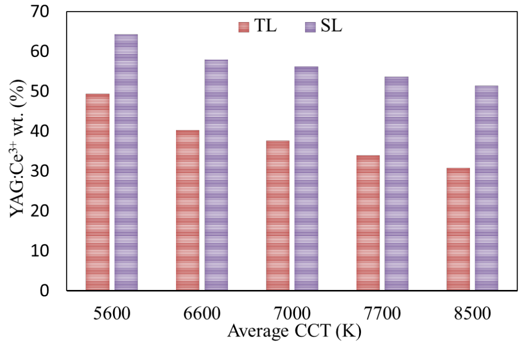


Fig. 2: The concentration of yellow $YAG:Ce^{3+}$ phosphor in each remote phosphor structure.

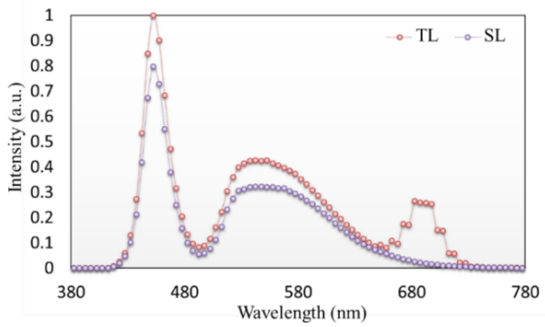


Fig. 3: The emission spectra of phosphor in each remote phosphor structure.

Besides, the color properties of WLED could be managed by optimizing the color balance among yellow, green, and red light components. Also, the color rendering index (CRI) is probably enhanced with the increase of red-light spectral energy while that of the green-light energy can contribute greatly to the higher lumen output of the LED structures. Therefore, it is essential to measure and compare the emission spectra of the SL and the TL. Fig. 3 clearly displays the experimental results of emission spectra from each remote phosphor structure. As can be seen, the TL has stronger light emission at the wavelength of blue, green, and especially red lights than the SL structure. This indicates that compared to the single-layer remote model, the triple-layer successfully preserves the color balance and is able to boost the color rendering index and luminous efficiency for WLED lamps. To verify this state, the computation of light scattering and incident light power is applied with the use of Mie-scattering theory and Beer's law [19, 20], which is demonstrated specifically in Section 3.

3. Results and discussion

The color rendering indexes (CRIs) of both remote phosphor structures are described in Fig. 4. Obviously, the triple-layer phosphor structure shows higher CRI than the single-layer structure, at all color temperatures. It is noticed that the CRI of both structures increases with higher CCTs and peaks at 8500 K, which can be attributed to the decrease in yellow phosphor concentration at high color temperatures. However, the improvements in the CRI of TL are not much. Therefore, to examine the effectiveness of the TL on the color quality of LEDs, another metric is taken into consideration.

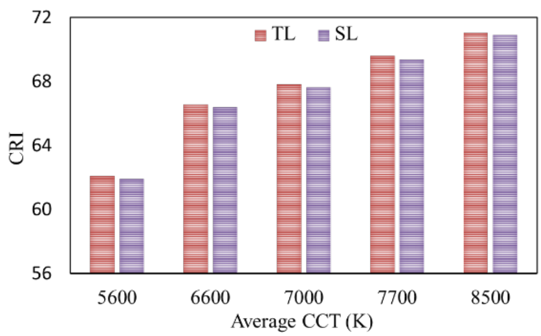


Fig. 4: Color rendering indices each remote phosphor structure at different average CCTs.

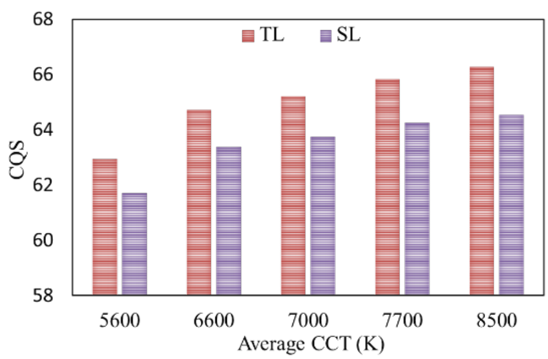


Fig. 5: Color quality scale each remote phosphor structure at different average CCTs.

The color quality scale (CQS), which is proved to be more powerful than the CRI, is investigated. The CQS specifically is an index evaluating the color performances of the white-light via three different aspects: the CRI, the color coordinates, and the preferences of the viewers. Thus, attaining the enhancement in CQS is somehow more challenging than getting the CRI boosted. As can be seen in Fig. 5 that illustrates the color quality scale of the TL and SL, significant improvements can be accomplished with the TL structure. In particular, the CQS of the TL configuration is much higher than that of the SL. Similar to the CRI, the higher the CCTs, the higher the CQS. Note that, the better the CQS is, the greater the color quality gets. This enhancement can be attributed to the color balance among yellow, red, and green lights when applying the three-layered structure. In addition to that, the yellow concentration noticeably declines with the presence of the two layers of green and red phosphor, leading to more scattering events inside the package, causing the lighting components to be mixed more and distributed more evenly. Also, the lower yellow concentration in TL structure is beneficial to the blue-light conversion owing to the enhancement in scattering and absorption properties of the phosphor layers, helping to diminish the yellow ring problem for white LED lights. Therefore, the TL is better than the SL in terms of color uniformity and efficiency.

As mentioned above, the lower yellow phosphor concentration of the TL not only enhances the scattering properties but also increases the light extraction efficiency. However, there will be a decline in luminous flux when the scattering ability is improved. This leads to a question in the ability of the triple-layer structure to perform high luminous flux while obtaining better color uniformity. Hence, the study presents an analysis of the lighting efficiency of the single-layer structure and the multi-layer structure.

The computation carried out for the transmitted blue light and converted yellow light in the multi-layer remote structure (we used the dual-layer to simplify the process) is expressed as the following equations. It is also noted that from this mathematic model, the efficiency in lumen out of WLED can be achieved with the multi-layer remote phosphor structure. In the single-layer remote model, the phosphor-layer thickness is $2h$, the transmitted blue light and converted yellow light are calculated by [20]:

$$IB_2 = IB_0 \times e^{-2\alpha_{B2}h} \quad (1)$$

$$IY_1 = \frac{1}{2}\frac{\beta_1 \times IB_0}{\alpha_{B1} - \alpha_{Y1}}(e^{-2\alpha_{Y1}h} - e^{-2\alpha_{B1}h}) \quad (2)$$

In the dual-layer remote configuration, those lights are expressed as follows. Here, the thickness of each phosphor layer is h.

$$IB_2 = IB_0 \times e^{-2\alpha_{B2}h} \quad (3)$$

$$IY_2 = \frac{1}{2}\frac{\beta_2 \times IB_0}{\alpha_{B2} - \alpha_{Y2}}(e^{-2\alpha_{Y2}h} - e^{-2\alpha_{B2}h}) \quad (4)$$

Specifically, in the presented mathematic expressions, h indicates the thickness of each phosphor layer. The subscripts *"1"* and *"2"* show the single- and double-layer remote phosphor structures. β and γ present the blue-light conversion coefficient for converting to yellow light, and the yellow-light reflection coefficient, respectively. The blue-light and yellow-light intensities, presented by IB and IY respectively, are considered as the blue-LED light intensity, described as IB_0. α_B; α_Y demonstrate the fractions of the energy loss of blue and yellow lights during their propagation in the phosphor film respectively.

The dual-layer phosphor structure can yield better luminous performance than the single-layer does:

$$\frac{(IB_2 + IY_2) - (IB_1 + IY_1)}{IB_1 + IY_1} > 0 \quad (5)$$

In addition, the transmitted light power is calculated using Lambert-Beer law:

$$P = P_0 exp(-\mu_{extH}) \quad (6)$$

In Eq. (6), P_0 indicates the incident light power, H shows the thickness of the phosphor film (mm), and μ_{ext} is the extinction coefficient. Here, $\mu_{ext} = N_r \cdot C_{ext}$, where N_r shows the number density distribution of the phosphor particles (mm^{-3}), while C_{ext} (mm^2) indicates the extinction cross-section of phosphor particles. According to Eq. (5), the multi-layer remote phosphor structure is more advantageous

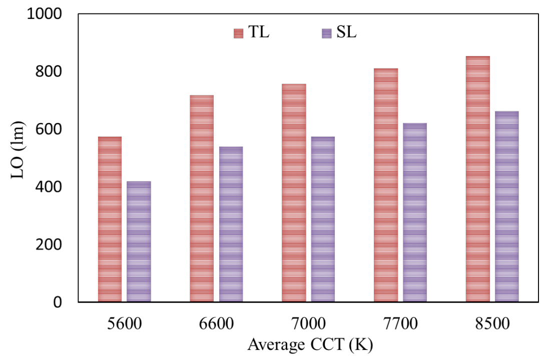


Fig. 6: Luminous flux of each remote phosphor structure at different average CCTs.

to the luminous efficiency than the single-layer structure. This means the triple-layer one can promote much better lumen output for WLED packages. The luminous flux of each structure is also demonstrated in Fig. 6 to prove the efficiency of using the triple-layer structure.

In Fig. 6, the higher the color temperature gets, the better the luminous flux is presented. Moreover, the lumen output of the TL structure shows higher values than that of the SL, especially at high CCTs. For instance, at 8500 K, it is possible for the TL to achieve more than 800 lm in luminous flux, which is approximately 200 lm enhancement compared to the lumen output of the SL structure. This enhancement can be attributed to the back-scattering reduction when the yellow phosphor concentration is lower in the TL. Since the backscattering is degraded, the light extraction efficiency is enhanced. In other words, the trapped lights between the phosphor layer and LED chip are reduced, and the blue lights could easily pass through the yellow phosphor film to reach the green and red phosphor layers, and be converted effectively. Therefore, the TL has higher luminous intensity than the SL one.

As the TL shows better CQS, it is appropriate to enhance the color quality of WLED. Besides the CQS, the color uniformity, which can be examined via the color temperature deviation (D-CCT), is a crucial part of the color-quality evaluation. Previous studies show that the scattering enhancement can result in smaller color deviation. Thus, the scattering enhancement par-

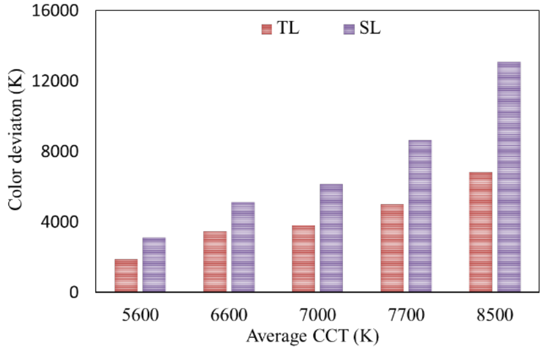


Fig. 7: Correlated color temperature deviation (D-CCT) of each remote phosphor structure at different average CCTs.

ticles (SiO_2, $CaCO_3$, TiO_2, ...) are applied. Though the application of SEPs resulted in the improvement of D-CCT, the luminous flux is significantly reduced. The triple-layer structure can enhance the scattering properties owing to the multi-layer design, and balance the color distribution due to the addition of green and red spectra to the white-light spectral band while enhancing the light extraction efficiency of the WLED package. In Fig. 7, it is easy to observe the considerable enhancement in the color uniformity, especially at the high CCT of 8500 K. In particular, the TL has smaller color-deviation values than the SL. Moreover, this difference between D-CCTs of the SL and the TL becomes larger as the color temperature rises. Therefore, the triple-layer is an appropriate alternative to the conventional remote phosphor structure (single-layer) since it can promote higher CRI, CQS, color uniformity, and luminous output. However, it is noted that the reduction in luminescence is unavoidable when the scattering is improved. Yet, the benefits getting from the backscattering reduction of the TL can outweigh this insignificant luminous flux decrease.

4. Conclusion

This work presents the effects of green $Y_3Al_5O_{12}:Ce^{3+}$ and red $CaTiO_3:Pr^{3+}$ phosphors on lumen efficacy and the color rendition properties of the two-layer RP W-LED. The Mie-scattering theory and Lambert-Beer's law were used to establish that $CaTiO_3:Pr^{3+}$ is a good phosphor material for the stimulation of high-chromatic-rendering W-LED with RP packages. On the other hand, $Y_3Al_5O_{12}:Ce^{3+}$ has turned out to be a good choice for increasing the luminous flux of W-LEDs. The attained results moreover are affirmative in the cases of low and high preset CCTs (5600-8500 K). As a result, our research has succeeded in increasing the color fidelity of white light, which is a challenge for RP structures. Nevertheless, there is one minor disadvantage in the optical output of the package regarding usage of the phosphors, which is that too high $Y_3Al_5O_{12}:Ce^{3+}$ or $CaTiO_3:Pr^{3+}$ concentrations might get the color rendition or lumen intensity suffered. As a result, determining an optimum concentration of those phosphors, based on the purpose of the W-LED manufacturer, is critical. Furthermore, the research paper's findings can be a vital reference for a potential and practical development path of future higher-quality W-LED packages.

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