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The application of green $YPO_4:Ce^{3+},Tb^{3+}$ and red $LiLaO_2:Eu^{3+}$ layers to remote phosphor LED

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ABSTRACT

Remote phosphor structure is commonly limited in color quality, but has greater luminous flux when comparing to structures with in-cup or conformal coating. From this dilemma, various researches with advance modifications have been proposed to perfect the chromatic performance of remote structure. In this research, we reach higher color quality by obtaining better values in quality indcators such as color rendering index (CRI) and color quality scale (CQS) with the dual-layer phosphor in our remote white light-emitting diodes (WLEDs). The idea is to ultize WLEDs with 7000 K correlated color temperature (CCT) and create dual-layer configuration with yellow phosphor $YAG:Ce^{3+}$ under green phosphor $YPO_4:Ce^{3+},Tb^{3+}$ or red phosphor $LiLaO_2:Eu^{3+}$. After that, we search for suitable concentration of $LiLaO_2:Eu^{3+}$ for addition in order to acquire the finest color quality. The result shows that WLED with $LiLaO_2:Eu^{3+}$ has better CRI and CQS as the higher the concentration of $LiLaO_2:Eu^{3+}$, the larger CRI and CQS due to increased light scattered in WLEDs. Meanwhile, the green phosphor layer $YPO_4:Ce^{3+},Tb^{3+}$ give advantages to luminous flux. However, the reduction in luminous flux and color quality occurs when the concentration of $LiLaO_2:Eu^{3+}$ and $YPO_4:Ce^{3+},Tb^{3+}$ over increase. Results are verified by Mie theory and Beer's Law and can be applied to practical manufacturing of high quality WLEDs.

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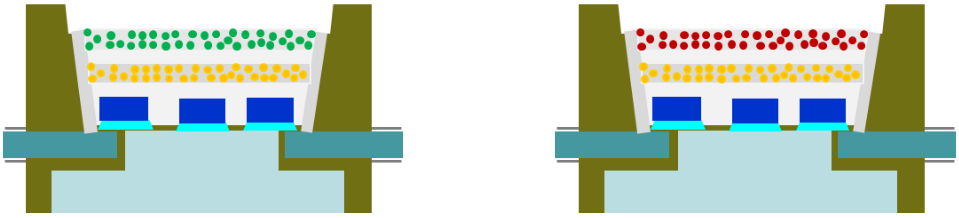
1. INTRODUCTION

As we can see, the most advanced lighting resolution, phosphor converted white light-emitting diodes (pc-WLEDs), with various strong points is a potential replacement for older ones [1]. The application of diodes to discharge light has existed in many different aspects of daily life including landscape, street lighting, backlighting, and more while the light extraction effectiveness and the inconsistent lighting temperature that negatively affects color outcome are boundaries that kept WLEDs from reaching higher efficiency [2]. Regarding the increasing market demand and usage, luminous efficiency and color quality must have further improvement [3]. In present, the most popular approach for white light generation is combining the chromatic lights from the LED chips and phosphor layer in WLED structure together. Even though this method is cheap and easy to apply, it is undeniable that the LEDs structures and phosphor layers arrangement are the deciding factors to WLED performance in light output and redering ability, which this method cannot fully adapt [4-8]. In order to produce LEDs, many common phosphor coating methods have been offered including dispensing coating and conformal coating [9, 10]. However, the temperature rises at the junction of the LED and phosphor layer due to these components are placed together in the structure cause reduced light conversion and results in low color quality. Hence, diminishing the result of heat would enhance the phosphor functioning and refrain the phosphor from being damaged by irreversible conditions. The remote configuration that separates the phosphor layer and the LED chip has been constituted by many previous studies as a solution to lessen the generated heat at the joint. With such a wide distance of phosphor and the LED chip, the light loss from back-scattering event and inner reflection is reduced. This solution does not only solve the problem of poor thermal performance but also raise other optical properties of WLEDs such as light output and chromatic performance [11-16]. Nevertheless, advanced lighting demands cannot be fulfilled with the current state of remote structure lighting devices although these devices are still compatible for standard occasions. This situation shows that higher quality LED is needed to adapt with the growth of luminescence industry. To achive advancement, back-scattering reduction and light output enhancement are still the targets of researchers when remote WLEDs with different structure modifications were proposed in recent studies to improve the mentioned aspects. One of the examinations demonstrated that an inversed core lens capsulation and the phosphor placed as a remote ring directs the chip's emitted light outside the structure and then diminish the reduced amount of light which occurs because of circulation inside LED [17]. Patterned structure of remote WLEDs with clear center within the border region and no phosphor-covered exterior can result in better thermal consistency and color quality output [18]. Furthermore, the sapphire substrate in different patterned remote phosphor structure are more proficient in terms of color temperature homogeneity compared to far-field pattern [19-21]. To improve the light output of LEDs, remote phosphor which has dual layer package is proposed. Many of the studies emphasize on color quality and light output development when it comes to enhancing WLEDs. Nonetheless, these studies only paid attention to low color temperature lighting devices with one LED chip. In reality, improvements in WLEDs with high color temperature are more complicate and desirable at the current point. Moreover, there has not been any study which compared different usage effectiveness of dual-layer phosphor configuration. Therefore, the manufacturer can hardly choose a solution to improve color quality of luminous flux.

This research proposed two dual-layer remote phosphor structures to enhance color quality for different WLEDs in color temperature of 7000 K. The first proposal is to use a green phosphor layer $YPO_4:Ce^{3+},Tb^{3+}$ to add green shade to the structure, which leads to an increase in luminous flux. The second proposal is to use a red phosphor layer $LiLaO_2:Eu^{3+}$ as a way to present red light to the structure, which leads to the increase color quality. Detailed compositions of $LiLaO_2:Eu^{3+}$ which affects optical performance of WLEDs are also part of the manuscript. The result of the study shows that color rendering index (CRI) and color quality scale (CQS) has been significantly improved when we add a layer of phosphor $LiLaO_2:Eu^{3+}$. Nevertheless, it is important to choose suitable concentration of $LiLaO_2:Eu^{3+}$ and $YPO_4:Ce^{3+},Tb^{3+}$ to avoid damaging the optical properties of WLEDs when the concentration of green phosphor and red phosphor exceed. Adding a green or red phosphor layer over a yellow phosphor layer YAG: Ce^{3+} will create three differences. First, the component of the green light or red light will increase, which lead to an increase in emission spectrum of white light and the chromatic performance of WLEDs. Second, the amount of scattered light and light conversion in WLEDs oppose to the concentration of added phosphor. Therefore, choosing appropriate phosphor concentration is crucial for preserving luminous flux.

2. PREPARATION AND SIMULATION

In this research, WLEDs with 9 LED chips inside. Every chip discharges 1.16 W when the wavelength is at 453 nm. The study of $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ concentration influences can begin once the simulation of remote phosphor model is completed. Two dual-layer models, green-yellow configuration (GYC) and red-yellow configuration (RYC) are presented as subjects for the experiments in this research. The structure GYC includes 2 phosphor layers put on the blue chip. The phosphor layer $YPO_4:Ce^{3+},Tb^{3+}$ put on the yellow phosphor layer $YAG:Ce^{3+}$ in Figure 1 (a). RYC structure is similar to GYC in terms of configuration but has green phosphor layer $LiLaO_2:Eu^{3+}$ on yellow phosphor layer $YAG:Ce^{3+}$ instead as illustrates in Figure 1 (b). The GYC and RYC are the tools to achieve better results in WLEDs chromatic performance and emission spectrum, which is an achievable outcome from managing the scattered green light and red light in the structure through phosphor concentration. Although advantageous, the concentration of $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ need to be modified appropriately to prevent optical properties damages.



(a) (b)

Figure 1. (a) Illustration of GYC, and (b) RYC

Figure 2 demonstrated the modification among the phosphor materials to maintain the correlated color temperature (CCT). This modification has two meanings: the first meaning is to maintain the average of CCTs and the second one is that it affects to the scattering and absorbing process of two phosphor layers in WLEDs, which definitely affects the chromatic performance and lumen output of the lighting device. Hence, choosing the correct $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ concentration benefits the color quality of WLEDs. When the $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ concentration respectively increase from 2-20% wt., $YAG:Ce^{3+}$ concentration decreases to maintain the average of CCTs.

In Figure 3, we can observe the effect of the added phosphor concentration on the emission spectrum. The choice depends on the requirement of manufacturer. WLEDs require the high color quality, so it is possible to decrease a litte amount of luminous flux. The intensity tendency at two light spectrum ranges from 420-480 to 500-640 nm tend to increase with $YPO_4:Ce^{3+},Tb^{3+}$ concentration. The increase of spectrum emitted in this range proves for the increase of luminous flux emission. In addition, the emitted blue light in WLEDs increase comes from scattered light enhancement in the phosphor and structure leads to some advantages for the correlated color. This is the important result applying $YPO_4:Ce^{3+},Tb^{3+}$. It is easy to realize that the tendency of red spectrum ranges from 648 to 738 nm increases with $LiLaO_2:Eu^{3+}$ concentration. However, there is not significant meaning if lacking the increase of two spectrum ranges left: 420-480 nm and 500-640 nm. Spectrum expansion at the wavelengths ranges 420-480 nm helps to increase blue-light scattering. The higher temperature expresses the higher spectrum emitted, and the result is the higher color quality and luminous flux. This is an essential result applying $LiLaO_2:Eu^{3+}$. Especially, the regulation of color quality of WLEDs having high teamperature is a challenge. This research claims that $LiLaO_2:Eu^{3+}$ is an addition that benefits chromatic performance of WLEDs at any color temperature.

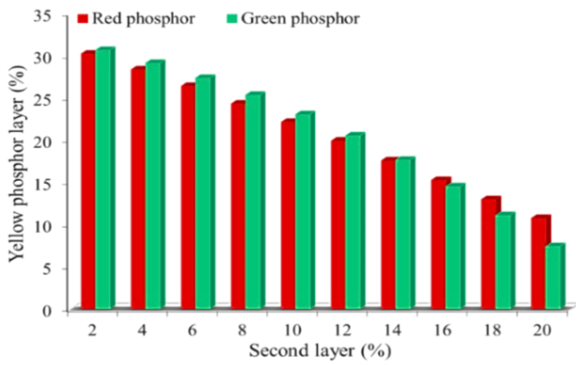


Figure 2. The change of phosphor concentration of GYC (above) and RYC (below) for keeping the average CCT

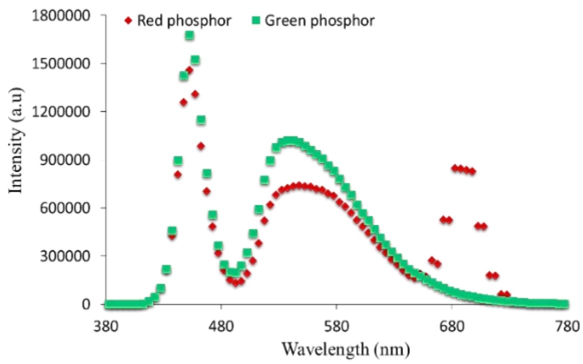


Figure 3. Emission spectra of dual-layer phosphor configurations

Color rendering index truthfully accesses the device's ability to express color of an object when its light illuminates that object. The amount of green-light increase overwhelmingly causes the color imbalance between the primary chromatic lights constitute to the creation of white light. This also affects to the color quality and leads to poor color expression of the lighting devices. In Figure 4, the CRI is shown to be slightly decline when remote phosphor layer of $YPO_4:Ce^{3+},Tb^{3+}$ exists. However, it is acceptable because CRI is just an element of CQS, which means green phosphor is still a suitable option if the WLEDs with green phosphor have sufficient value of CQS. Comparing between CRI and CQS, the color quality scale is more complicate and broader quality indicator, therefore, should be considered above CRI. In Figure 5, CQS retains value if $YPO_4:Ce^{3+},Tb^{3+}$ concentration less than 8%. Therefore, 8% $YPO_4:Ce^{3+},Tb^{3+}$ can be choosen to apply after considering the luminous flux emission.

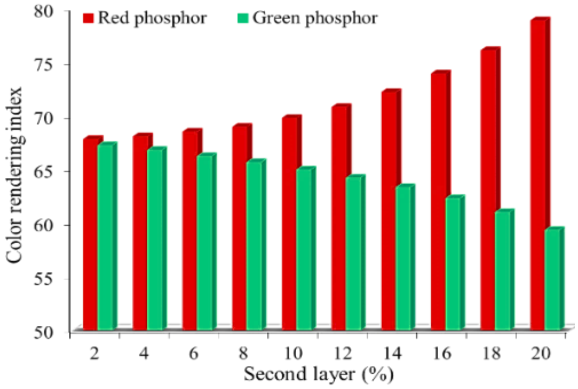


Figure 4. The color rendering index as a function of the concentration of $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ phosphors

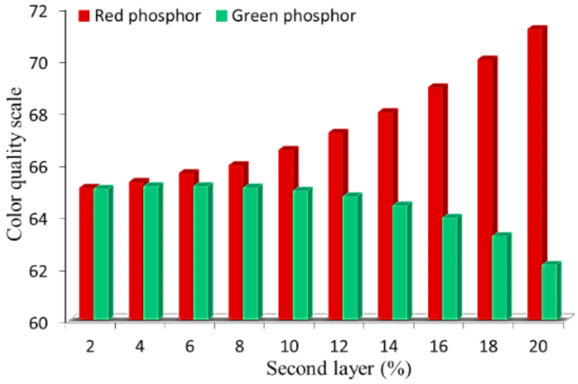


Figure 5. The color quality scale as a function of the concentration of $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ phosphors

The mathematical equation to calculate the blue and yellow light yielded from WLEDs with two phosphor layers is presented in this part. The results provide valuable information for better understanding of WLEDs mechanism and enhancing methods. The amount of emitted blue light and modified yellow light in single phosphor package with $2h$ phosphor density are shown in,

$$PB_1 = PB_0 \times e^{-2\alpha_{B1}h} \tag{1}$$

$$PY_1 = \frac{1}{2}\frac{\beta_1 \times PB_0}{\alpha_{B1} - \alpha_{Y1}}\left(e^{-2\alpha_{Y1}h} - e^{-2\alpha_{B1}h}\right) \tag{2}$$

In dual-phosphor remote WLEDs with an h phosphor layer density, the amount of emitted blue light and modified yellow light are expressed as:

$$PB_2 = PB_0 \times e^{-2\alpha_{B2}h} \tag{3}$$

$$PY_2 = \frac{1}{2}\frac{\beta_2 \times PB_0}{\alpha_{B2} - \alpha_{Y2}}\left(e^{-2\alpha_{Y2}h} - e^{-2\alpha_{B2}h}\right) \tag{4}$$

The h denotes phosphor density (mm). The subscript "1" indicates WLEDs with single phosphor structure and "2" indicates WLEDs with dual-phosphor remote structure. The β stands for the rate of internal conversion from blue transforms to yellow light. The reflection coefficient of the yellow light is illustrated by γ. The radiation intensities of LED chip including the blue light intensity (PB) and yellow light intensity (PY) are represented by PB_0. The blue and yellow light energy lost during the spreading process in the phosphor layer are indicated by α_B; α_Y respectively. The WLEDs in double-layer phosphor structures have their luminous efficacy improved significantly in comparison with the single layer structure ones:

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

The Mie-scattering theory was applied to study the scattering of $LiLaO_2:Eu^{3+}$ [22, 23]. Besides, the Mie theory also contributes to the calculation of scattering cross section C_{sca} for spherical particles. The Lambert-Beer law measures the light power emitted [24, 25]:

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

According to the formula above, I_0 is the optical power of incident light and L is the density of phosphor coating (mm). μ_{ext} indicates the extinction coefficient that is computed by this equation: $\mu_{ext} = N_r$.

C_{ext}, where N_r stands for the number density distribution of particles (mm^{-3}) and C_{ext} (mm^2) is the extinction cross-section of phosphor particles.

From (5), it is comfirmed that more phosphor layers means better WLEDs luminous efficiency. Thus, the paper has proven that applying two phosphor layers in the remote structure results in higher lumen output in comparison to single-layer structure. Figure 6 shows that luminous flux increased significantly whenever $YPO_4:Ce^{3+},Tb^{3+}$ concentration rises. On the other hand, luminous flux of RYC structure does not reponse the same to the concentration of phosphor $LiLaO_2:Eu^{3+}$. This can be explained through Beer's Law that since the reduction factor μ_{ext} increases with the concentration of $LiLaO_2:Eu^{3+}$ but the light transmission proficiency decreases when red phosphor concentration increases.

Therefore, when the layer thickness of phosphor materials in remote structure is fixed, the emitted light may decrease when the concentration of $LiLaO_2:Eu^{3+}$ increases. When concentration $LiLaO_2:Eu^{3+}$ at 20% wt optics decreased significantly. However, the advantages of the red phosphor class $LiLaO_2:Eu^{3+}$ to other aspects of WLEDs, especially the values of color quality indicators, is very considerable. Besides, light emission in RYC remote phosphor structure is better compared to the common remote structure that does not have $LiLaO_2:Eu^{3+}$ phosphor. Therefore, a small reduction in luminous flux is still tolerable if WLEDs require high color quality. The final decision relies on the manufacturers to choose the appropriate type of phosphor and concentration to fulfill the production demands.

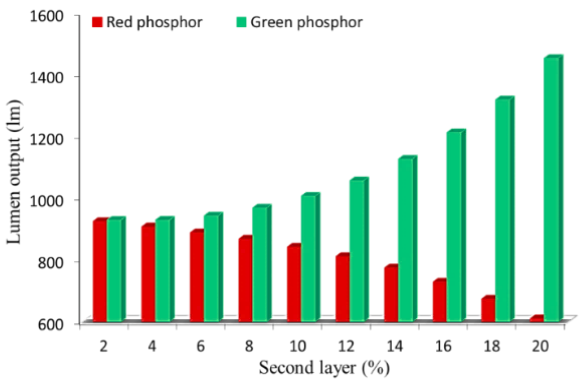


Figure 6. The luminous flux as a function of the concentration of $YPO_4:Ce^{3+},Tb^{3+}$ and $LiLaO_2:Eu^{3+}$ phosphors

3. CONCLUSION

The paper presents the effect of phosphor green $YPO_4:Ce^{3+},Tb^{3+}$ and phosphor red $LiLaO_2:Eu^{3+}$ on optical properties development in WLED structure with two phosphor layers. The Mie scattering theory and Beer's Law are the tools to confirm the effectiveness of $LiLaO_2:Eu^{3+}$ in improving color quality. Meanwhile, $YPO_4:Ce^{3+},Tb^{3+}$ is the choice to improve the luminous flux of WLEDs. The results are applicable for remote phosphor structure at any color temperature. Thus, the conclusions in this manuscript have presented an efficient method to enhance chromatic performance of remote structure WLEDs, a difficult to obtain and desirable objective for many research. Although there could be a small drawback if the concentrations of $YPO_4:Ce^{3+},Tb^{3+}$ or $LiLaO_2:Eu^{3+}$ become excessive, color quality or luminous flux significantly reduced. Therefore, choosing the optimal concentration for phosphor material is crucial to satisfy production demands while not damaging other aspects. And the article has provided much important information for reference in producing better quality WLEDs.

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