Exellent Color Quality and Luminous Flux of Wleds Using Triple-Layer Remote Phosphor Configuration

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Abstract. *This study proposed a triple-layer remote phosphor (TRP) structure to improve the color and luminous flux of white LEDs (WLEDs). TRP structure consists of 3 different phosphor layers: yellow YAG:Ce^{3+} layer below, red $CaMgSi_2O_6$:Eu^{2+},Mn^{2+} phosphor on top and green layer $Ba_2Li_2Si_2O_7$:Sn^{2+},Mn^{2+} phosphor in the middle. Using red $CaMgSi_2O_6$:Eu^{2+},Mn^{2+} to control the red light component leads to the increase in color rendering index (CRI). Utilizing the green $CaMgSi_2O_6$:Eu^{2+},Mn^{2+} phosphor to control the green light component results in the increase in luminous efficacy (LE) of WLEDs. Furthermore, when the concentration of these two phosphors increased, yellow layer YAG:Ce^{3+} concentration decreased to maintain average correlated color temperatures (ACCTs) in the range from 6000K to 8500K. Besides CRI and LE, color quality scale (CQS) is also analyzed through the control of green and red phosphors concentrations. The research results show that the higher the concentration of $CaMgSi_2O_6$:Eu^{2+},Mn^{2+} is, the better the CRI becomes. In contrast, CRI decreased significantly when increasing the concentration of $Ba_2Li_2Si_2O_7$:Sn^{2+},Mn^{2+} . Meanwhile, CQS achieved notable enhancement in the concentration range of 10% -14% $CaMgSi_2O_6$:Eu^{2+},Mn^{2+}, regardless of $Ba_2Li_2Si_2O_7$:Sn^{2+},Mn^{2+} concentration. LE, in particular, can also increase by more than 40% along with the improvement of CRI and CQS due to the reduction of the backscattered light and the addition of green light. Research results are a valuable reference for producers who wish to improve the color quality and enhance the luminous flux of WLEDs.*

Keywords

WLEDs, triple-layer structure, remote phosphor package, color quality, luminous flux.

1. Introduction

Phosphor-converted white light-emitting diodes (WLEDs) with many outstanding features such as smallness, energy saving, cost efficiency and cohesion in color have been perceived as a new and improved light source [1]-[4]. The complementary principle of colors is applied in WLEDs as blue light from the blue chip and yellow light from the phosphor layer merge in the configuration [5]. It is expected that WLEDs will be used for solid-state lighting system; however, the luminous efficiency must be improved in order to be used for the aforementioned purpose [6]-[9]. In order to produce the white light, using the freely dispersed coating method is the most well-known one. The transparent encapsulated resin and the phosphor powder are mixed and then dispersed on the phosphor package to fabricate the white light in the process. This procedure may allow better control over phosphor layer thickness and significantly lower the expenses; however, it cannot support the production of high-quality WLEDs [10]-[12]. Therefore, a method that helps to distribute the color homogeneously and has angular homogeneity of correlated color temperature (CCT) such as the conformal coating method is used as a substitution [13]. The luminous efficiency of the conformal phosphor structure, however, decreases due to the backscattering effect this structure has.

The idea of separating the chip and the phosphor layer in remote phosphor structures are presented in previous studies [14]-[16]. The extraction efficiency benefits from the polymer hemispherical shell lens with an interior phosphor coating that enhances the light extraction inside of the reflection structure. Moreover, the luminous efficiency is also improved because the air-gap embedded structure reflects the light downward.

In 2018, Nhan's team used the red-emitting $\alpha-SrO\cdot 3B_2O_3:Sm^{2+}$ for increasing the optical properties of single-remote phosphor. By varying $\alpha-SrO\cdot 3B_2O_3:Sm^{2+}$ concentration from 2% to 24%, the obtained results proposed that color uniformity, color rendering index (CRI), color quality scale (CQS), and luminous efficacy could be improved significantly [17]. With the target of the improvement of CRI and CQS, in 2019, Lee's team has applied the red-emitting $Mg_2TiO_4:Mn^{4+}$ phosphor in the dual-layer remote geometry [18, 19]. However, the luminous flux is a disadvantage in these studies.

The concentration of phosphor, along with the package structure, is also a critical element that affects the luminous flux. When the phosphor concentration increases, it will cause the re-absorption loss in the phosphor layer to rise. The luminous efficiency of the device will be lowered as a result, especially at lower CCTs. Thus, the improvements in the blue and yellow light emission and the reduction in light loss from backscattering and reflection are desirable targets.

The triple-layer remote phosphor structure WLEDs with color temperatures from 6000K to 8500K are proposed in this study. The TRP structure consists of three different phosphor layers with green phosphor layer $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ between yellow phosphor layer $YAG:Ce^{3+}$ and red $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor layer. The green phosphor layer adds green light components to improve the luminous flux emitted while the red light component is supplemented by red phosphor layer to improve color quality. The results show that when there is a balance between 3 colors of yellow, green, and red the color quality can reach the highest value, and the luminous flux of WLEDs is reduced only by an insignificant amount.

2. Computational simulation

2.1. Preparation of phosphor materials

The first idea of the study is to use the green phosphor $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ to enrich the green light component in WLEDs and enhance luminous flux. The second idea is to use the red phosphor layer $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ to boost the red light in WLEDs, leading to an increase in CRI and CQS. The paper also details the chemical composition of these phosphor materials that influences the optical properties of WLEDs. The chemical composition of $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ are presented in detail as in Tab. 1.

To produce $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$, a procedure consisting of 6 steps, from mixing, drying, double firing, to washing and then drying again, is required and must be performed following a strict order to achieve the best result. All of

Tab. 1: Composition of red-emitting $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor.

Ingredient	Mole (%)	By weight (g)	Mole (mol)	Ions	Mole (mol)
CaO	45.69	150	2.67	Ca^{2+}	2.67
MgO	16.95	40	0.99	Mg^{2+}	0.99
SiO_2	35.82	126	2.10	Si^{4+}	2.10
Eu_2O_3	0.17	3.5	0.01	O^{2-}	8.13
$MnCO_3$	1.37	9.2	0.08	Eu^{2+}	0.02
NH_4Cl	—	5.4	—	Mn^{2+}	0.08

Tab. 2: Composition of green-emitting $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ phosphor.

Ingredient	Mole (%)	By weight (g)	Mole (mol)	Ions	Mole (mol)
$BaCO_3$	25.04	185	0.94	Ba^{2+}	0.94
Li_2CO_3	14.82	41	0.55	Li^{+}	1.11
SiO_2	29.34	66	1.10	Si^{4+}	1.10
SnO	13.48	6.8	0.50	O^{2-}	7.62
$MnCO_3$	3.95	1.7	0.15	Sn^{2+}	0.5
NH_4Br	13.36	49	0.5	Mn^{2+}	0.15

these steps are critical and based on the step(s) before them. The first step is mixing the materials by dipping into methanol with a few cubic centimeters of water. Second, let it dry in a condition of air. After the materials are dried, fire them in the capped quartz tubes and fused with N_2 at the condition of 1000^0C for 1 hour and then continue to fry the powdery products in capped quartz tubes but with CO instead of N_2 in an hour at a temperature of 1150^0C. The next step is to pick up the product and wash it several times with water. Finally, leave them to dry and we will have the $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$.

Similar to $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ the chemical composition for $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ are presented in Tab. 2. The steps to materialize $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ are demonstrated as follows.

We start by mixing $BaCO_3 + Li_2CO_3 + SiO_2$ using the dry grinding or milling method. After the first step, we continue to fire the mixture in open boats under 850^0C for an hour while adding in H_2. We will then proceed with $SnO + MnCO_3 + NH_4Br$ by soaking them in methanol and stirring the mixture until it reaches uniformity. Then dry the mixture in the condition of air to reduce it to powder form. The powder will be fried with N_2 in capped quartz tubes for 1 hour. After that, turning the product to powder and put it through the firing process one more time in open quartz boats under 850^0C temperature but for about 16 hours (overnight). Once the previous step is done, store the final powder product in a well-closed container.

2.2. Simulation of TRP

The simulation of RP-WLEDs with the average CCT of 8500K, 7700K, 7000K, 6600K, 5600K, and the remote phosphor structure are supported by the commercial software LightTools 8.1.0 that based on the Monte Carlo ray-tracing method. In Fig. 1, there is the 3D stimulated physical model WLEDs used to demonstrate optical simulations of remote package WLEDs. The physical model of WLEDs contains a reflector that is 8 mm of the bottom length, 2.07 mm in height and 9.85 mm for the top surface length. The remote phosphor structure with exactly 0.08 mm thickness for each phosphor film

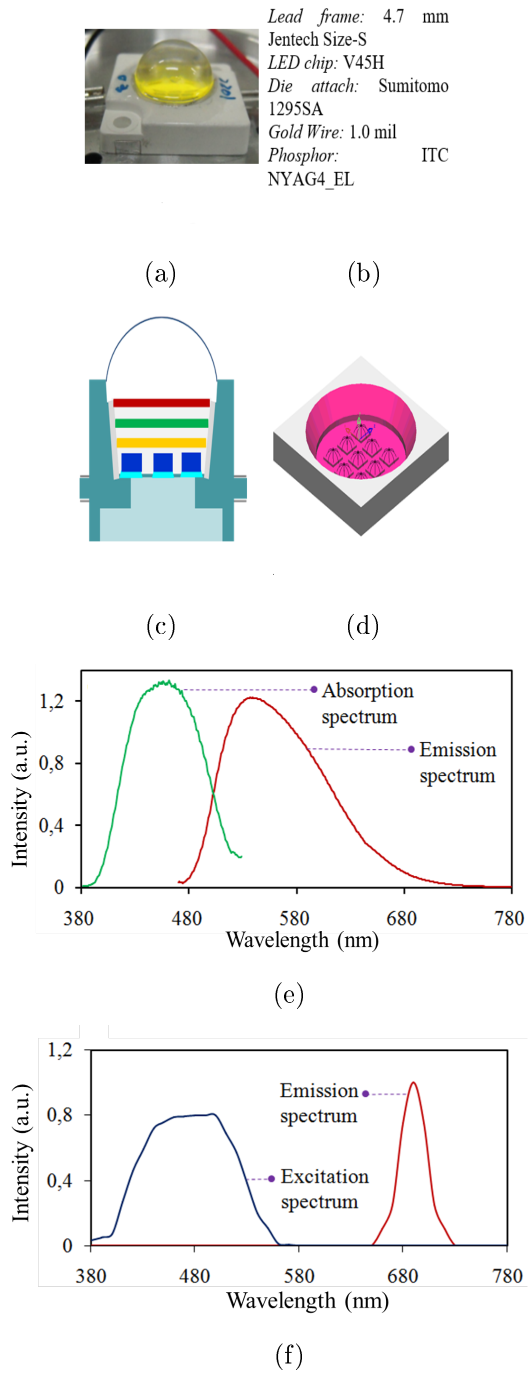


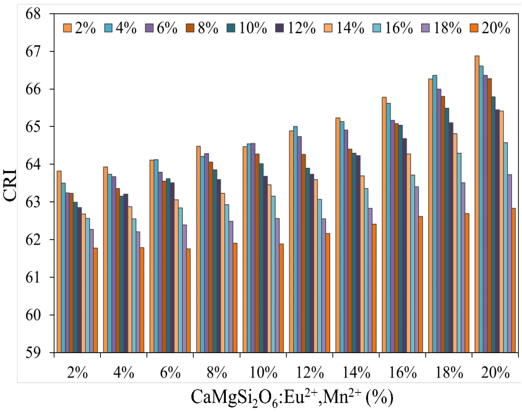
Fig. 1: (a) WLEDs, (b) its parameters, (c) Illustration of triple-layer remote phosphor configuration, (d) the simulation of WLEDs, (e) the measured spectra of the yellow-emitting $YAG:Ce^{3+}$ phosphor, (f) the measured spectra of the red-emitting $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor.

overlays on 9 LED chips with the measurements of 1.14 mm bottom square and 0.15 mm height that are embedded in the gaps on the reflector. These blue chips emit a radiant flux of 1.16 W at 455 nm wavelength. Even though the concentration of phosphor particles are constantly changing from 2% to 24%, the control over $YAG:Ce^{3+}$ wt keeps the average CCT values remain static in their cases. Furthermore, the spectra values of $YAG:Ce^{3+}$ including absorption spectrum and emission spectrum are presented in Fig. 1(e). Meanwhile, the excitation spectrum and emission spectrum of $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ are displayed in Fig. 1(f).

3. Results and discussion

Figure 2 shows the CRI values varying with the concentration of red phosphor and green phosphor from 2% to 20%. The CRI gradually increases with the addition of red phosphor concentration and reaches the maximum value at 20% concentration. On the other hand, the increase in green phosphor does not benefit CRI, due to the fact that when the concentration of green phosphor rises from 2% to 20%, CRI continuously decreases regardless of the improvement in red phosphor or the changes in average correlated color temperature (ACCT). From the results of Fig. 2, it is clear that the red light component in WLEDs, which comes from the red phosphor layer $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$, needs improvement in order to boost the value of CRI.

When green phosphor $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ concentration increases, the green light component prevails, and that is a disadvantage for CRI because the energy conversion in red phosphorous layer decreases as the concentration of green phosphor increases. According to TRP structure, the green phosphor layer is below the red phosphor layer, which means the light reaches the green phosphor layer first, before going through the red layer. So, green phosphor $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ concentration should be reduced as much as possible, if the target is CRI. CRI is the only factor to evaluate color quality as it has the ability to reflect the color



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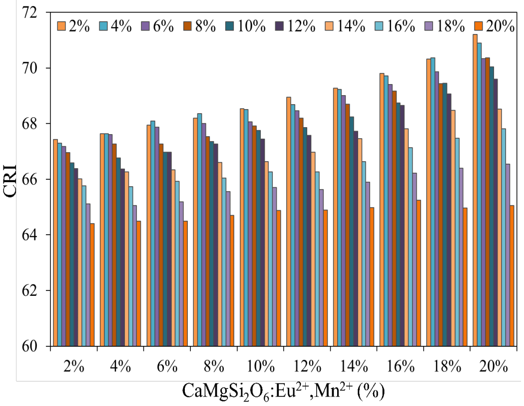

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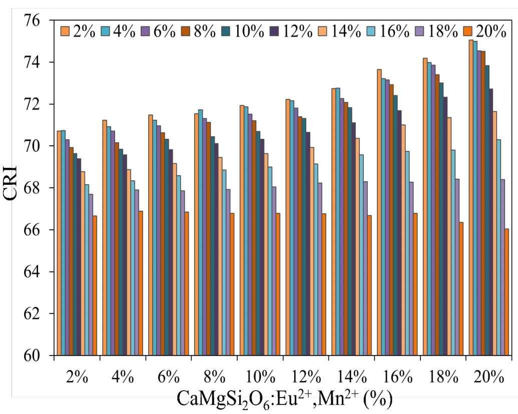
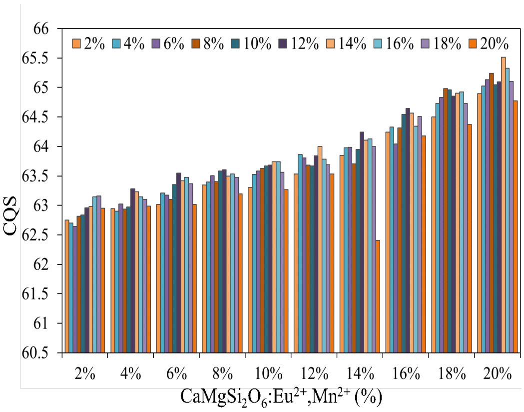


(c)

Fig. 2: CRI of TRP as a function of red $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor and green $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ phosphor: (a) 6000K; (b) 7000K; (c) 8500K



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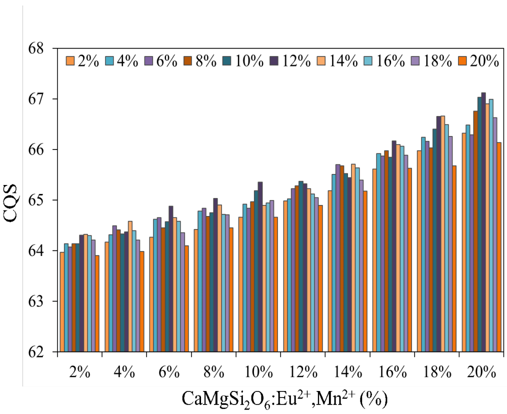

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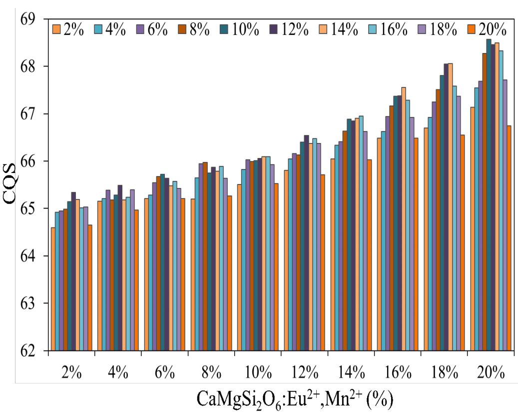

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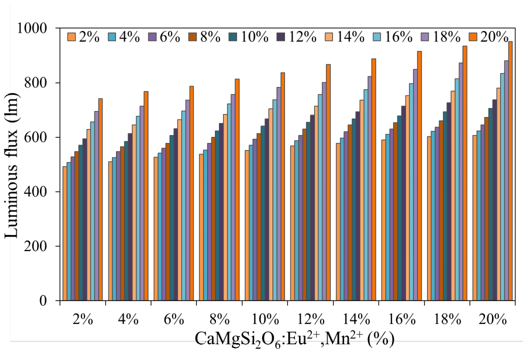
Fig. 3: CQS of TRP as a function of red $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor and green $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ phosphor: (a) 6000K; (b) 7000K; (c) 8500K



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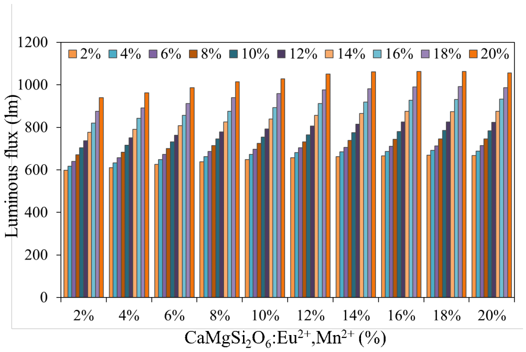

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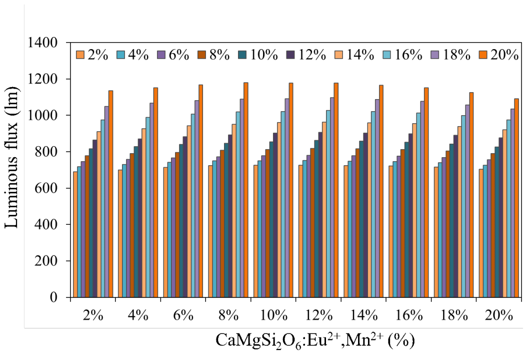

(c)

Fig. 4: Luminous flux of TRP as a function of red $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor and green $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ phosphor: (a) 6000K; (b) 7000K; (c) 8500K

more correctly in the human eyes when there is a lighting effect.

However, besides the true color of objects, the preference of the viewers and the color coordinates are two important criteria that CRI does not have access to.

However, color quality scale (CQS) can evaluate the combination of three factor: CRI, the preference of the viewer and the color coordinates for white light. Hence, in a comparison between CRI and CQS, CQS value stands out as a more important and difficult target to achieve. The remaining question is how to improve the CQS value of WLEDs? Does it only require the enhancement in the red light component to improve the CRI? To find answers to these questions, CQS values are also presented in Fig. 3. In general, CQS increases with red $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ phosphor.

However, unlike the CRI, the CQS experiences a small change when the concentration of the green phosphor layer $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ varies. From the results shown in Fig. 3, it is possible to confirm that both the green phosphor and the red phosphor contribute to the improvement of CQS. The balance between 3 colors: yellow, green and red is the key to enhance CQS. When the concentration of red phosphor or green phosphor increases, yellow phosphor concentration decreases to maintain the ACCT.

The reduced yellow phosphor concentration causes the yellow light component to decrease, and this has two benefits. The first one is reducing the amount of backscattered light to the LED chip so that the luminous flux improves significantly. Another benefit of reducing yellow phosphor concentration is to lower the yellow light component and replace the yellow light component with the red and green light components. Gaining control over CQS is the key to manage these 3 color components. CQS increases gradually when the green phosphor $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ concentration moves from 2% to 10% and then gradually decreases. The highest CQS values are obtained when $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ is from 10% to 14%. When the green phosphor concentration is low (2% to 10%), the yellow light component still dominates, therefore, the light transmission energy is lost due to backscattering, which leads to CQS not reaching its maximum. When the green phosphor concentration is between 10% and 14%, the green light component is enough for CQS to reach the highest level. However, if the concentration of $Ba_2Li_2Si_2O_7$:Sn^{2+},Mn^{2+} keep going up, the green light components become excessive, causing a color imbalance among the 3 primary colors green, red and yellow. Therefore, the increase in green phosphor concentration from that point onward will cause the CQS to decrease.

Controlling the color quality of remote phosphor structures is more complex than that of conformal phosphor or in-cup phosphor structures. It is even more difficult with WLEDs that have ACCTs from 7000K - 8500K. Nonetheless, the results showed that with the TRP structure the higher the ACCTs, the greater the CQS. In addition to reducing the amount of backscattered light, the TRP structure also supports the scattering of light inside WLEDs. This enhancement in scattering is beneficial to the mixing of light components, resulting in a high-quality white light. However, does this enhancement in the scattering process reduces the light transmission energy?

The focus of the next part is the mathematical model used to calculate the transmitted blue light and converted yellow light in the double-layer phosphor structure, which is an area that can generate important changes for the LED efficiency. The formulas for transmitted blue light and converted yellow light in single layer remote phosphor package with the phosphor layer thickness of 2h are as follows:

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

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

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

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

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

The h is the thickness of each phosphor layer while the subscripts "1" and "2" indicate the number of layers, single layer or double-layer remote phosphor package. The conversion coefficient for blue light converting to yellow light is illustrated as β, and γ is the reflection coefficient of the yellow light. The intensities of blue light (PB) and yellow light (PY) are the light intensities from the blue LED, indicated by PB_0. αB; αY are parameters which indicate the proportions of blue and yellow lights' energy loss during the scattering process in the phosphor layer.

The lighting effect of pc-LEDs with the double-layer phosphor structure improved significantly in comparison with a single layer structure:

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

By using the Mie-theory [20], the scattering of phosphor particles was studied, and the scattering cross section C_{sca} for spherical particles is also computed. The Lambert-Beer law [21] can calculate the transmitted light power:

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

I_0 is the incident light power, L is the phosphor layer thickness (mm), and μ_{ext} is the extinction coefficient which can be expressed as $\mu_{ext} = N_r C_{ext}$, where N_r is the number density distribution of particles (mm^{-3}). Cext (mm^2) is the extinction cross-section of phosphor particles.

Equation (5) certifies that the use of additional phosphor layers enhances the luminous emission of WLEDs. The increase in luminous emission affects red phosphor and green phosphor concentrations, causing them to rise. To preserve the ACCTs when the concentrations of red phosphor and green phosphor increase, the yellow phosphor concentration decreases. The vital point in reducing the yellow phosphor concentration is to prevent light loss due to the backscattering characteristic. Furthermore, a reduced yellow phosphor concentration makes light transmission energy become stronger, according to Lambert-Beer's Law in Equation (6). Therefore, the higher the concentrations of the phosphor layer $Ba_2Li_2Si_2O_7$:Sn^{2+},Mn^{2+} or $CaMgSi_2O_6$:Eu^{2+},Mn^{2+} are, the more powerful the luminous flux emitted. However, this is unfavorable for CQS as that red or green light components exceed a certain limit as this will cause color imbalance, which reduces the obtained CQS.

According to results available in Fig. 4, phosphor layer $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ allows luminous emission (LE) to rise up to more than 40%, regardless of the phosphor concentration $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ due to the increase in green light component and the reduction of backscattering effect. The obtained results are important references, paving the way for manufacturers to choose the appropriate level of concentration for these two phosphor types to reach their goals. Specifically, if the target is to achieve high value in CQS and LE, it is optimal to keep the concentrations of $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ from 10% to 14%, and $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ at 20%. Moreover, LE also increased slightly with $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ concentrations at 6000K and 7000K ACCTs. At ACCT 8500 K, LE is almost unchanged in the range of 2% - 14% ACCTs. Then, if the green phosphor concentration reaches 20%, LE decreases slightly.

4. Conclusions

In conclusion, the TRP structure with two phosphor layers $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ and $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ was proposed to improve the CRI, CQS, and LE of WLEDs. As a result, not only can TRP structure improve the color quality, it also improved LE, which is a new achievement that has never obtained before. In order to have those results happened, balancing the yellow, green, and red light in these phosphor layers by controlling the concentration of $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$ and $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ is required. The key for controlling the green light component in WLEDs, which can benefit the luminous flux, is to manage the green phosphor $Ba_2Li_2Si_2O_7:Sn^{2+},Mn^{2+}$. Furthermore, the use of multiple phosphor layers is more favorable for the luminous flux than using one single layer. In the meantime, if the control over the concentration of red phosphor layer $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ is gained, it means the red light component in WLEDs can be manipulated to improve CRI. Researched results show that the balance of the three colors yellow, green, and red together with the reduction of backscattering from the yellow $YAG:Ce^{3+}$ will provide the highest color quality and luminous flux.

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