The impacts of $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+}, \text{Mn}^{2+}$ and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ particles on the optical properties of remote phosphor LED

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As implied in the title, the triple-layer remote phosphor (TRP), constructed with the yellow $\text{YAG}:\text{Ce}^{3+}$ layer at the bottom, the red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor layer on the top, and the green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+}, \text{Mn}^{2+}$ phosphor layer between these two layers, is suggested in this paper to improve the color and luminescence of white LEDs (WLEDs). In order to control the red light for the purpose of increasing the color rendering index (CRI), it is suggested that the red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor should be applied in the TRP structure. Simultaneously, the structure uses the green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+}, \text{Mn}^{2+}$ phosphor layer to control the green light, which increases the luminous efficacy (LE) of WLEDs. In addition, when the concentration of these two phosphors increases, the yellow $\text{YAG}:\text{Ce}^{3+}$ concentration must be reduced to keep the average correlated color temperatures (ACCTs) stable at 6000 K to 8500 K. Besides, appropriate adjusting of CRI, LE, and color quality scale (CQS) is also analyzed by modifying the concentration of the green phosphor and red phosphor. The results show that the CRI can get better values if $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ concentration is higher. In contrast, the CRI decreases dramatically when the concentration of $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+}, \text{Mn}^{2+}$ increases. Meanwhile, CQS can be significantly increased in the range of 10 % to 14 % $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$, regardless of the concentration of $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+}, \text{Mn}^{2+}$. In particular, along with the improvement of CRI and CQS, LE can also be increased by more than 40 % by reducing the scattered light and adding the green light. Obtained results are a valuable reference for manufacturers for improving WLEDs color and luminescence quality to produce a broader range of WLEDs with better quality fulfilling social needs.

Keywords: WLEDs; triple-layer structure; remote phosphor package; color quality; luminous flux

1. Introduction

Phosphor-converted white light-emitting diodes (WLEDs), the potential light source with a variety of outstanding features, such as the compactness, high energy efficiency, low cost, and color stability, have become more and more popular in illumination market [1–3]. The complementary colors principle in which the blue light from a blue chip is combined with the yellow light from the

phosphor is applied for the production of WLEDs [4]. WLEDs will have the capability of being used in solid-state lighting if their luminous efficiency is enhanced [5]. Generally, using freely dispersed coating is the most common method to generate the white light in which the transparent encapsulated resin is combined with phosphor powder and dispersed in the phosphor package. This approach is really beneficial for controlling the thickness of phosphor, and even the price is also cheaper. However, it cannot support high-quality WLEDs. Therefore, the conformal

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coating method can become a better choice because it is capable of distributing colors uniformly, resulting in angular homogeneity of correlated color temperature (CCT) [6]. Nevertheless, it cannot be completely perfect since the backscattering effect reduces luminous efficiency considerably.

The concept of separating the chip and the phosphor layer of remote phosphor structures has been demonstrated in previous studies [7–12]. As can be seen, the enhanced light extraction internal reflection structure which uses a polymer hemispherical shell lens with an interior phosphor coating is known as an effective tool to increase extraction efficiency [13]. Furthermore, an air-gap embedded structure can enhance luminous efficiency by reflecting downward light [14]. Besides, the concentration of the phosphor is as much important as the structure of the package in luminous efficiency. The loss of re-absorption in the phosphor layer is proportional to the phosphor concentration. In other words, when one rises, it leads to the increase of the other. Therefore, the luminous efficiency of the device can be degraded, especially at lower CCTs condition [15–18]. As a result, enhancing the emission of blue and yellow rays as well as reducing the amount of light lost by backscattering and reflection have become essential targets in producing higher quality WLEDs.

From the aforementioned facts, the paper comes up with a proposal of WLEDs triple-layer remote phosphor structure with different color temperatures from 6000 K to 8500 K. The TRP structure includes three different phosphor layers: a green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor layer between the yellow $\text{YAG}:\text{Ce}^{3+}$ phosphor layer and red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor layer [19]. The green phosphor layer extracts green light components and improves the light output. Meanwhile, the red light component is enriched by the red phosphor layer in order to improve the color quality. Obtained results show that the balance among the yellow, green, and red color can make the color quality maximized while the luminous flux of the WLEDs goes down negligibly. This is a positive sign for the WLEDs industry demanding constant improvement.

2. Preparation and simulation

2.1. Preparation of phosphor materials

First, the paper offers the idea of using the green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ layer to increase the green light component in WLEDs, which leads to the increase of output luminous flux. The second idea is to use the red phosphor $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ layer to increase the red light component in WLEDs, resulting in the rise of CRI and CQS. The article also details the effect of chemical composition of these phosphors on the optical properties of WLEDs. The chemical composition of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ is shown in Table 1. The process of making $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ consists of six steps: mixing, drying, firing twice, washing, and drying again. Each step plays an important role during the process and has a close link to the others. All of these steps must be stuck to a certain order to achieve the optimal effect. Firstly, mix the materials by soaking them in methanol with a few cubic centimeters of water. Secondly, dry the materials in the air. After that, fire the dried materials in capped quartz tubes combined with N_2 at 1000 °C for 1 hour. Then continue to fry the powder in capped quartz tubes but with CO instead of N_2 for an hour at a temperature of 1150 °C. Next, collect the product and wash in water several times. Finally, let it dry until the $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ becomes a complete material.

As can be seen from Table 2, the ingredients of $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ consisting of BaCO_3 , Li_2CO_3 , SiO_2 , SnO , MnCO_3 , and NH_4Br are detailed with parameters including the percentage of mole, weight, molar mass, etc. The steps for $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ production process from these components also follow a particular order. First, use the dry grinding or milling method to mix the $\text{BaCO}_3 + \text{Li}_2\text{CO}_3 + \text{SiO}_2$ together. Then fire this compound in the open boats at 850 °C for an hour with the addition of H_2 .

Second, proceed with the fabrication of the $\text{SnO} + \text{MnCO}_3 + \text{NH}_4\text{Br}$ by slurring them in methanol and stirring until the mixture reaches homogeneity. After that, dry it in the air to turn it into powder. Next, fire the material in the capped quartz tubes

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

Ingredient	Mole [%]	By weight [g]	Molar mass [g/mol]	Mole [mol]	Ions	Mole [mol]	Mole [%]
CaO	45.69	150	56.0774	2.67	Ca^{2+}	2.67	0.19
MgO	16.95	40	40.304	0.99	Mg^{2+}	0.99	0.07
SiO ₂	35.82	126	60.08	2.10	Si^{4+}	2.10	0.15
Eu ₂ O ₃	0.17	3.5	351.926	0.01	O^{2-}	8.13	0.58
MnCO ₃	1.37	9.2	114.9469	0.08	Eu^{2+}	0.02	0.0014
NH ₄ Cl	–	5.4	53.49	–	Mn^{2+}	0.08	0.0057

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

Ingredient	Mole [%]	By weight [g]	Molar mass [g/mol]	Mole [mol]	Ions	Mole [mol]	Mole [%]
BaCO ₃	25.04	185	197.34	0.94	Ba^{2+}	0.94	0.082
Li ₂ CO ₃	14.82	41	73.891	0.55	Li^{+}	1.11	0.097
SiO ₂	29.34	66	60.08	1.10	Si^{4+}	1.10	0.096
SnO	13.48	6.8	134.709	0.50	O^{2-}	7.62	0.667
MnCO ₃	3.95	1.7	114.9469	0.15	Sn^{2+}	0.5	0.044
NH ₄ Br	13.36	49	97.94	0.5	Mn^{2+}	0.15	0.013

with N₂ at 850 °C for an hour. The process of creating powder is carried out immediately after the firing process. Then fire the material again in the open quartz boats with CO at 850 °C for a longer time, about 16 hours (overnight). After that powderize as in the previous step. Finally, collect the product and store in a tightly-closed container.

2.2. Simulation of TRP

The actual WLEDs, which present the best optical-thermal stability, were used in our study, as shown in Fig. 1a. We have constructed the setups precisely, to make them identical. Their normalized cross correlations are approximately 99.6 %, meaning that the actual packaging and the simulated packaging are alike. Moreover, the impacts of the factors such as LED wavelength, waveform, light intensity, and operating temperature on the CRI and CCT could be decreased. The commercial software LightTools 8.1.0 based on the Monte Carlo ray-tracing method was used to simulate the WLEDs which have the average CCT of 6000 K, 7000 K, and 8500 K in the remote phosphor structure. The 3-D simulation physical model

of WLEDs for performing optical simulations of remote package WLEDs is shown in Fig. 1c and Fig. 1d. In this physical model of WLEDs, the size of the reflector is: 8 mm long bottom, the height of 2.07 mm and the length of 9.85 mm at its top surface. The remote phosphor layer with a fixed thickness of 0.08 mm covers the 9 LED chips. Each LED chip with a 1.14 mm square base and height of 0.15 mm is bound in the cavity of the reflector. The radiant flux of each blue chip is 1.16 W at 455 nm wavelength. Furthermore, the spectral values of YAG:Ce³⁺, including absorption spectrum and emission spectrum, are presented in Fig. 1e. Meanwhile, the excitation spectrum and emission spectrum of $CaMgSi_2O_6:Eu^{2+},Mn^{2+}$ are also displayed in Fig. 1f. The concentration of phosphor particles changes continuously from 2 % to 24 %. However, the average CCT values are maintained by controlling the YAG:Ce³⁺ wt.%.

3. Results and discussion

As can be seen from Fig. 2, the change of CRI depends on the variation in red phosphor

Fig. 1. (a) WLEDs structure, (b) its parameters, (c) illustration of triple-layer remote phosphor configuration, (d) simulation of WLEDs, (e) measured spectra of the yellow-emitting YAG:Ce^{3+} phosphor, (f) measured spectra of the red-emitting $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor.

and green phosphor concentrations in the range from 2 % to 20 %. Specifically, when red phosphor concentration goes up, the CRI increases and reaches the maximum value if this concentration is at 20 %. In contrast, the addition of green phosphor is not conducive to CRI, because when the green phosphor increases from 2 % to 20 %, CRI decreases regardless of the increase in red phosphor or change in average correlated color temperature (ACCT). It can be confirmed from the results shown in Fig. 2 that to increase the CRI value, we need to increase the red light component in WLEDs which is generated from red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor layer. When the

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green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor concentration increases, the green light component dominates, but this is detrimental to the CRI. On the other hand, the conversion of energy in the red phosphor layer decreases as the concentration of the green phosphor increases. As known, in the TRP structure, the green phosphor layer is located under the red phosphor, which means that the emitted light reaches the red phosphor after reaching the green layer. Therefore, it is advisable to adjust the green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor concentration to the minimum if the CRI is requested to be improved. However, CRI is just one of the factors to assess the color quality, showing the ability to recognize the true color by the human eye when the light is shining. Meanwhile, in terms of the true color of the object, the viewer's preference and the color coordinates are the two other important criteria, besides the CRI. Color quality scale (CQS), meanwhile, can evaluate the combination of three factors: CRI, the comfort of the viewer and the color coordinates for white light [20]. Thus, comparing with CRI, the value of CQS is more important and difficult to achieve. The question is how to raise the CQS value of WLEDs? Is it just essential to increase the red light component similar to improving the CRI? The answer is in Fig. 3, where the CQS values are presented in detail. Overall, CQS also increases depending on the red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor concentration. However, unlike the CRI deviation, the CQS deviation is small when the concentration of green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor is changed. It can be confirmed from the results shown in Fig. 3 that both green phosphor and red phosphor can increase CQS. The balance among the three colors: yellow, green and red is the key to enhance CQS. As the concentration of red phosphor or green phosphor increases, the concentration of yellow phosphor decreases to stabilize ACCT.

As mentioned above, the key to improve CQS is the balance among the three colors yellow, green, and red. Therefore, controlling CQS is controlling these three color components. Specifically, CQS increases as green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor rises in the range from 2 % to 10 % and then decreases. CQS reaches the highest

Fig. 2. CRI of TRP as a function of red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor and green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor: (a) 6000 K; (b) 7000 K; (c) 8500 K.

Fig. 3. CQS of TRP as a function of red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor and green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor: (a) 6000 K; (b) 7000 K; (c) 8500 K.

value in the range from 10 % to 14 % of $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$. When the concentration of the green phosphor is lower (2 % to 10 %), the composition of yellow light still dominates. The light-transmitting energy is lost by scattering, resulting in the maximum CQS. When the green concentration is between 10 % and 14 %, green light is sufficient to get the highest CQS. However, when the $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ increases over that range, an imbalance among the three primary colors green, red, and yellow occurs due to the excess green light. Hence, as the concentration of green phosphor increases, it results in CQS reduction.

In fact, controlling the color quality of the remote phosphor structure is more complex than that of the conformal phosphor or in-cup phosphor structures. Specifically, it is more difficult to control the color quality with WLEDs having ACCTs in the range of 7000 K to 8500 K. However, the results show that for the TRP structure, the higher the ACCTs, the greater the CQS. Besides, reducing the amount of scattered light, the TRP structure also increases light scattering inside WLEDs, which increases the mixing of light components, resulting in the formation of high-quality white light. However, does this increase in scattering cause a decrease in light energy?

This part will present and demonstrate the mathematical model of the transmitted blue light and converted yellow light in the double-layer phosphor structure, from which a huge improvement of LED efficiency can be obtained. The transmitted blue light and converted yellow light for a single layer remote phosphor package with the phosphor layer thickness of $2h$ are expressed as follows [21]:

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

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

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

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

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

where h is the thickness of each phosphor layer. The subscripts 1 and 2 are used to describe single layer and double-layer remote phosphor package. β presents the conversion coefficient for blue light converting to yellow light. γ is 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_B and α_Y are parameters describing the fractions of the energy loss of blue and yellow lights during their propagation in the phosphor layer, respectively.

The lighting efficiency of WLEDs 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 scattering of phosphor particles was analyzed by using the Mie-theory. In addition, the scattering cross section C_{sca} for spherical particles can be computed by the following expression through applying the Mie theory. The transmitted light power can be calculated by the Lambert-Beer law [22]:

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

In this formula, I_0 is the incident light power, L is the phosphor layer thickness (mm) and μ_{ext} is known to be the extinction coefficient, which can be expressed as: $\mu_{\text{ext}} = N_r \cdot C_{\text{ext}}$, 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.

Equation 5 demonstrates that the addition of a phosphor layer enhances the luminous emission of WLEDs. The increase of luminous emission leads to the increased red phosphor and green phosphor concentrations. To keep the ACCTs stable as the concentration of red phosphor and green phosphor increase, the concentration of yellow phosphor must be decreased. The

purpose of reducing the concentration of yellow phosphor is to reduce the light energy caused by scattering. Moreover, the concentration of yellow phosphor decreases as the light transmission energy is larger, according to the Lambert-Beer law in equation 6. Thus, the larger the concentration of the phosphoric $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ layer or $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$, the higher the output luminous flux. However, this is detrimental to CQS because the excessive degree of red or green light causes color imbalance, resulting in the decreased CQS.

As can be seen from Fig. 4, the $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor layer helps luminous emission (LE) to increase up to 40 % due to the increase in green light and the light scattering regardless of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration. The higher the ACCT, the larger the luminous flux is obtained, as displayed in Fig. 5. The obtained result is an important reference for manufacturers to choose the appropriate concentration of these two phosphors. Specifically, in order that CQS and LE could receive high-value range, the $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ concentration must range from 10 % to 14 %, and the concentration of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ must be at 20 %. Meanwhile, LE also increases slightly depending on the concentration of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ with ACCTs of 6000 K and 7000 K. At ACCT 8500 K, LE is almost unchanged in the range of 2 % to 14 % of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$. Then LE decreases slightly when the green phosphor concentration increases to 20 %.

4. Conclusions

In conclusion, the TRP structure with two phosphor layers $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ was proposed as a promising material for enhancing the CRI, CQS, and LE of WLEDs. As a result, not only can TRP structure improve the color quality, but it also improved LE, which is seen as a new achievement never appeared before. To gain this result, there is a need to balance the yellow, green, and red light in these phosphor layers by controlling the concentration of $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ and

(a)

(b)

(c)

Fig. 4. Luminous flux of TRP as a function of red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor and green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor: (a) 6000 K; (b) 7000 K; (c) 8500 K.

$\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$. In order to operate the green $\text{Ba}_2\text{Li}_2\text{Si}_2\text{O}_7:\text{Sn}^{2+},\text{Mn}^{2+}$ phosphor, it is necessary to manage the green light component in WLEDs first so that the flux could increase.

Fig. 5. The emission spectra of WLEDs with varying TRP conditions.

Furthermore, the use of multiple layers of the phosphor is more conducive to flux than using just a single layer. Besides, when controlling the red $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ layer concentration, the red light component in WLEDs needs to be controlled to make CRI increase. Obtained results show that the balance among yellow, green, and red color, together with the scattering of yellow $\text{YAG}:\text{Ce}^{3+}$ will provide the highest color quality and luminescence for the new WLEDs generation with remarkable features, completely meeting the needs of users.

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