

Enhancing color homogeneity and luminous flux of WLEDs with double-layer phosphor configuration

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The applications of white light-emitting diodes (WLEDs) have increasingly been popular in today's society, yet color inhomogeneity and poor luminous flux are restricting their development. In this paper, a structure of double-layer phosphor package of white LED is proposed to improve color uniformity and luminous flux, in which the green phosphor layer SrBaSiO₄:Eu²⁺ was placed above the yellow phosphor layer YAG:Ce³⁺. The SrBaSiO₄:Eu²⁺ concentration added must be modified until the highest luminous flux and chromatic homogeneity are achieved. Five similar WLEDs with different color temperatures ranging from 5600 K - 8500 K were used in this study. The results show that the greater the SrBaSiO₄:Eu²⁺ concentration, the higher the luminous flux due to the increased green light component in WLEDs. However, a slight decrease in color rendering index and color quality index occurred when the SrBaSiO₄:Eu²⁺ concentration increased excessively, according to the simulations and Monte Carlo calculations. This paper is a vital reference to the fabrication of WLEDs with higher illumination efficiency and color uniformity of the phosphor structures.

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1. Introduction

In fact, phosphor-converted white light-emitting diodes (WLEDs), the fourth potential generation light source used to replace the conventional one, has been widely applied in many different aspects of our daily life such as landscape, street lighting, backlighting, etc. but light extraction efficiency and the angular homogeneity of the correlated color temperature of white LED are limiting its development [1, 2]. Due to the continued rise in demand of the lighting market, significant progress should be made to improve luminous efficiency and color quality [3]. Nowadays, one of the most common approaches for white light generation is combining the blue light from converse red phosphor with the yellow light from the LED chip. This concept was mentioned in previous studies but it cannot be denied that the structure of WLEDs, as well as the arrangement of phosphor layers, plays a vital role in determining the luminous efficiency, especially the color rendering index [4-8]. Several common phosphor coating methods used to produce WLEDs such as dispensing coating and conformal coating have been proposed in previous pieces of research [9, 10]. Nevertheless, these structures do not provide high color quality because of the degradation in the light conversion of phosphor material, which occurs when yellow emitting phosphor contacts directly with the LED chip, causing the junction point of the LED and phosphor layer gets hotter. Therefore, reducing the outcome of heat would improve the phosphor performance and avoid the unexpected damage to the

phosphor. It has been established in many previous studies that the remote phosphor structure in which phosphor is placed far apart from the heat source (LED chip) can reduce the effect of heating. With such a sufficiently large distance of phosphor and the LED chip, WLEDs could limit the backscattering and circulation of the light inside. As a result, the heat of LED is managed optimally and thus the luminous efficiency, as well as the color quality of WLEDs, can be enhanced considerably [11-13]. However, the remote phosphor structure has enough quality for regular lighting but does not meet other requirements of many other illumination applications, which requires the creation of next generation of LED. In 2016, Peng's team have proposed an optimized packaging structure, which is a combined structure of a patterned PiG with a crater-type lens, to improve the optical performance of multi-component phosphor-in-glass (PiG) based WLEDs [14-16].

For further development, some novel structures of the remote phosphor are proposed as a tool to minimize the backward scattering of the phosphor towards the chip and increase the luminous efficiency. Another study has shown that it is possible to redirect the light from the WLED chip to the surface of the LED and then reduce the loss energy caused by WLED's internal reflection using an inverted cone lens encapsulant and a surrounding ring remote phosphor layer [17]. A patterned remote phosphor structure with a noticeable region in the perimeter area without coating phosphor on the surface surrounding could bring out high uniformity of angular-dependent correlated

color temperature and chromatic stability [18]. Moreover, the patterned sapphire substrate applied in the remote phosphor could deliver the uniformity of the correlated color temperature in a far field pattern more effectively than a conventional pattern [19-20]. The remote phosphor with dual layer package is proposed to improve the light output of WLEDs. The above-mentioned studies focus on improving the color uniformity and photon emission of WLEDs of the phosphor remote structure. However, only single-chip WLEDs with low color temperatures were used while the improvement of optical parameters for WLEDs with high color temperature is very complex.

This article recommended dual-layer remote phosphor structure used to improve color quality for WLEDs at different color temperatures from 5600 K to 8500 K, in which the green phosphor $\text{SrBaSiO}_4:\text{Eu}^{2+}$ was used to increase the green light in the WLEDs, resulting in increased luminous flux and color homogeneity. The article also described how the chemical composition of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ affects the optical properties of WLEDs. According to the researched results, luminous flux and color homogeneity were significantly improved due to the addition of the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor layer. However, in order to avoid a sharp decline in color rendering index (CRI) and color quality scale (CQS) when green phosphor levels increase excessively, a suitable $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration should be chosen. There were two obvious changes when adding green phosphor to the $\text{YAG}:\text{Ce}^{3+}$ phosphor layer. First, the green light component increases when the spectral area of light emitted by the white light increases. Second, the scattering and light transmission in WLEDs with $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration are inversely proportional. Hence, choosing the appropriate $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration is a very important factor for the optical balance and color quality of WLEDs.

2. Preparation and simulation

2.1. Preparation of green-emitting $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor

$\text{SrBaSiO}_4:\text{Eu}^{2+}$ particles, a type of yellow-green phosphor with an emission peak of 2.36 eV and many remarkable characteristics such as high quantum efficiency and stability at high temperature, have become more and more popular [21]. It is particle sizes and concentration which can directly influence on the luminescence properties of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor. Besides, its composition including SrCO_3 , BaCO_3 , SiO_2 , Eu_2O_3 , NH_4Cl , and Eu^{2+} ion are used as raw materials as shown in Table 1. Particularly, $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor, which is applied for high-loading and long lifetime fluorescent lamps, becomes one of the most popular commercialized oxide phosphors. Following is the specific description of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor's fabrication process.

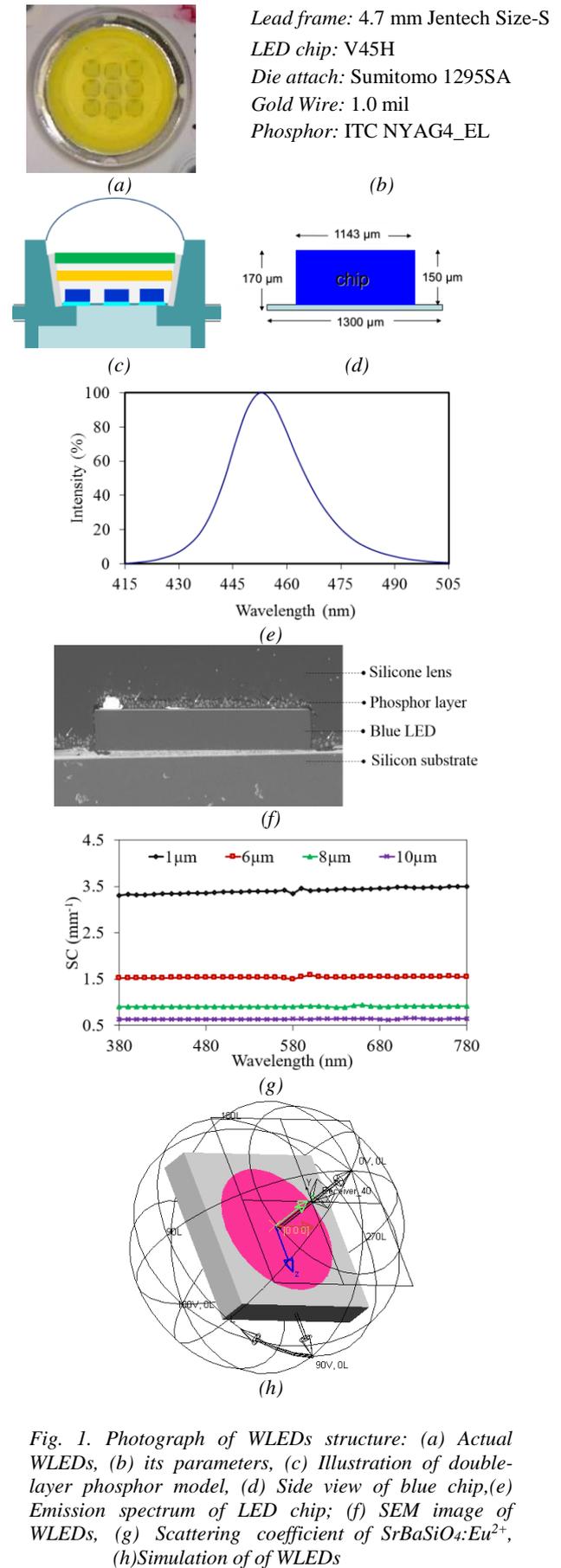


Fig. 1. Photograph of WLEDs structure: (a) Actual WLEDs, (b) its parameters, (c) Illustration of double-layer phosphor model, (d) Side view of blue chip, (e) Emission spectrum of LED chip; (f) SEM image of WLEDs, (g) Scattering coefficient of $\text{SrBaSiO}_4:\text{Eu}^{2+}$, (h) Simulation of WLEDs

Firstly, the materials are mixed together by slurring in water. Then they are ball-milled in the water into small particles. After that, the materials will be powdered as soon as they are dried in the air. Secondly, this powder will be fired in capped quartz tubes with CO at 1100°C within an hour and powdered by dry milling. Then 5.4 g NH₄Cl will be added and mixed with this mixture by dry milling. Next, they are fired again in capped quartz tubes with CO at 1100°C within an hour and powdered. Finally, the materials are washed in the water for several times (with pH ranges from 10 to 12) and dried after that.

2.2. Construction of WLEDs

In this part, the phosphor layer of real WLEDs is simulated with flat silicone layer based on the LightTools 8.5.0 program and Monte Carlo method, as shown in Figure 1(h). This modeling process includes two chief periods: (1) setting and building the mechanical structures and optical properties of WLED lamps, (2) monitoring the optical influences of phosphor compounding through the diversity of SrBaSiO₄:Eu²⁺ concentration.

In order to study the influence of YAG:Ce³⁺ and SrBaSiO₄:Eu²⁺ phosphor compounding on the performance of WLED lamps, some comparisons are compulsory. For instance, the comparison between the two types of compounding which have average CCTs of 5600 K, 6600K, 7000 K, 7700 K and 8500 K, dual-layer remote phosphor, must be clarified. As can be seen in Figure 1(a), there is an obvious description about WLED lamps with conformal phosphor compounding having average CCT of 8500 K, see Figure 1(b). The figure also indicates the simulation of WLEDs which does not contain SrBaSiO₄:Eu²⁺ in the component. The reflector has a bottom length of 8 mm, a height of 2.07 mm and a length of 9.85 mm at its top surface as displayed in Fig. 1 (c). The

conformal phosphor compounding, whose thickness is fixed at 0.08 mm, covers nine chips, as demonstrated in Fig. 1 (f). Each LED chip with the square base area of 1.14 mm² and the height of 0.15 mm is attached to the cavity of the reflector, see Fig. 1 (d). The radiant flux of each blue chip is 1.16 W while 453 nm is the peak wavelength, see Fig. 1 (e).

Without doubt, the combination of the SrBaSiO₄:Eu²⁺ particles and the YAG:Ce³⁺ phosphor layer will produce a scattering enhancement in WLEDs.

To have a better understanding of the scattering ability for the phosphor compounding, the SCs are presented at four typical SrBaSiO₄:Eu²⁺ sizes: 1 μm, 6 μm, 8 μm, 10 μm. From Fig. 1 (g), it can be seen that the SCs decrease with the increasing of SrBaSiO₄:Eu²⁺ size. This implies that the scattering ability for larger SrBaSiO₄:Eu²⁺ particles also reduces inside the phosphor compounding. This is of much benefit to the color homogeneity but loss of the luminous efficacy. Therefore, the selection of a suitable SrBaSiO₄:Eu²⁺ size become important before adding it to the phosphor compounding. Moreover, the SCs for in the wavelength range of vision are almost the same. Consequently, the higher scattering uniformity can be achieved when mixing SrBaSiO₄:Eu²⁺ phosphor, resulting in better color quality.

3. Results and analysis

Fig. 2 depicts the opposite change between the SrBaSiO₄:Eu²⁺ green phosphor concentration and the yellow phosphor YAG:Ce³⁺ concentration. This change leads to two outcomes: maintaining average CCTs, and affecting the scattering and absorption of two phosphoric layers in WLEDs. This inevitably affects the color quality and luminescence generated by WLEDs.

Table 1. Composition of green-emitting SrBaSiO₄:Eu²⁺ phosphor

Ingredient	Mole (%)	By weight (g)	Molar mass (g/mol)	Mole (mol)	Ions	Mole (mol)	Mole (%)
SrCO ₃	31.28	145	147.63	0.982	Sr ²⁺	0.982	0.088
BaCO ₃	31.79	197	197.34	0.998	Ba ²⁺	0.998	0.090
SiO ₂	33.40	63	60.08	1.049	Si ⁴⁺	1.049	0.094
Eu ₂ O ₃	0.32	3.5	351.926	0.01	O ²⁻	8.068	0.726
NH ₄ Cl	3.22	5.4	53.49	0.101	Eu ²⁺	0.02	0.002

Thus, the choice of concentration SrBaSiO₄:Eu²⁺ is critical to the color quality of WLEDs. When SrBaSiO₄:Eu²⁺ concentration increases from 2% -20% wt, YAG:Ce³⁺ concentration must decrease to keep average CCTs unchanged.

This phenomenon is the same for all WLEDs with different color temperatures of 5600 K-8500 K.

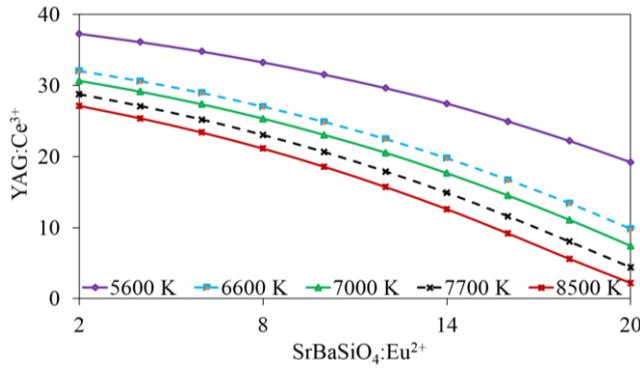


Fig. 2. The change of phosphor concentration for keeping the average CCT

The most noticeable point is the effect of green phosphor concentration SrBaSiO₄:Eu²⁺ on the transmitter spectrum of WLEDs as shown in Figs. 3-7. The choice is made based on the manufacturer's requirements. WLEDs with high color requirements can reduce the amount of light involved. Actually, the white light is the synthesis of the spectral region as shown in Figs. 3-7, which represent the spectral range of 5600 K, 6600 K 7000 K, 7700 K, and 8500 K, respectively. It is easy to recognize that the intensity at two the two light spectrums 420 nm - 480 nm and 500 nm - 640 nm tend to increase with the SrBaSiO₄:Eu²⁺ concentration. This rise in two-zone spectra demonstrates the increase in the emission of the photon. In addition, blue-light scattering in WLED, or the scattering in the phosphor layer and in WLEDs increases will result in color uniformity. This shows the importance of applying SrBaSiO₄:Eu²⁺ in producing better WLEDs, especially when the control of the color of the high-temperature phosphoric structure is a challenge. This study proves that SrBaSiO₄:Eu²⁺ can improve the color quality of WLEDs regardless of low color temperature (5600 K) or high color temperature (8500 K).

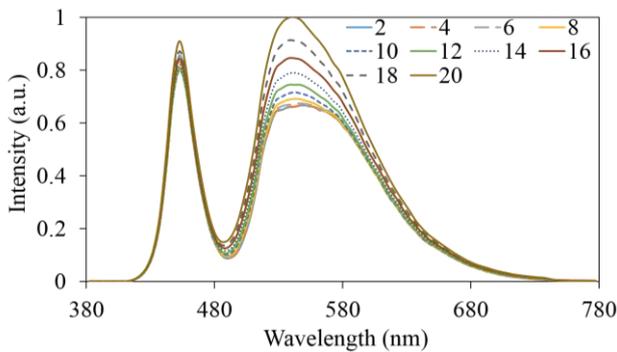


Fig. 3. The emission spectra of 5600 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

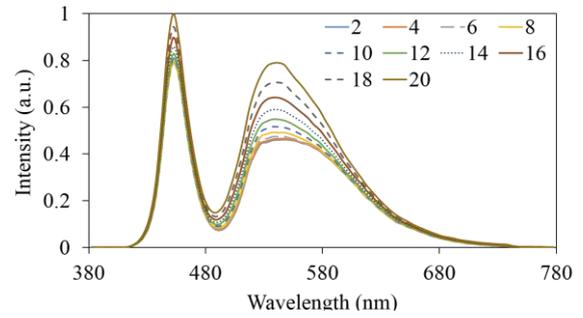


Fig. 4. The emission spectra of 6600 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

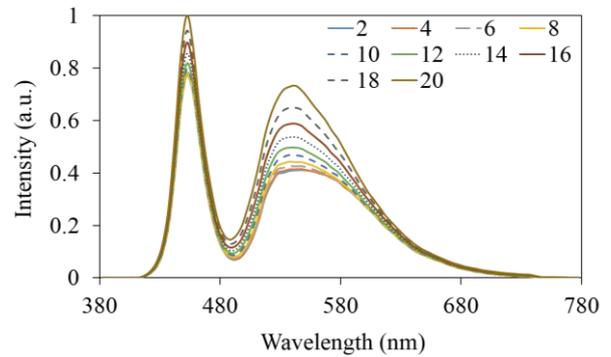


Fig. 5. The emission spectra of 7000 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

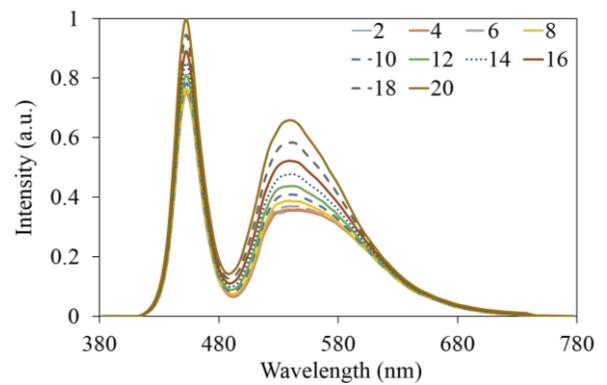


Fig. 6. The emission spectra of 7700 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

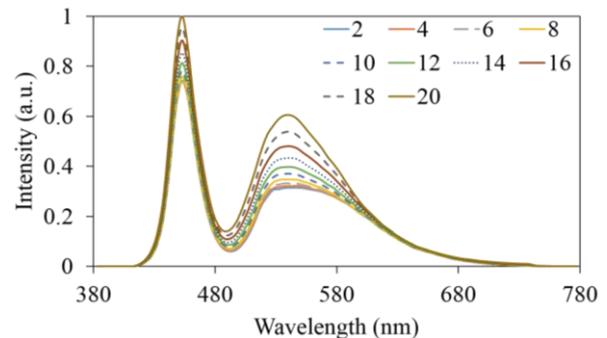


Fig. 7. The emission spectra of 8500 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

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 single layer remote phosphor package with the phosphor layer thickness of $2h$ are expressed 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 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 subscript “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 , 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 pc-LEDs with the double-layer phosphor structure enhances considerably compared to a single layer structure:

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

It can be drawn from Equation (5) that the illumination efficiency of WLEDs with dual-layer remote phosphor is greater than the single-layer phosphor's. Hence, the efficiency of the luminous flux output of this dual-layer remote phosphor is demonstrated in this paper. Specifically, Fig. 8 shows that the emitted photon increased significantly when $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration increased from 2 wt%. to 20% wt.

As shown in Fig. 9, the color deviation was significantly reduced by the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor concentration at all three CCTs, which is caused by the absorption of green phosphor. Specifically, when $\text{SrBaSiO}_4:\text{Eu}^{2+}$ phosphor absorbs the blue light from the LED chip, the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ particles will turn the blue light into green light. Beside the blue light from the LED

chip, $\text{SrBaSiO}_4:\text{Eu}^{2+}$ particles also absorb the yellow light. However, the blue light absorbed by the LED chip is stronger than the yellow light absorbed by $\text{SrBaSiO}_4:\text{Eu}^{2+}$ particles due to the distinguish absorption properties of these materials. Therefore, the green light composition in WLEDs increases with the addition of $\text{SrBaSiO}_4:\text{Eu}^{2+}$, which results in an increase in the colorimetric index. Color uniformity is one of the most important parameters in modern WLED lighting options. Obviously, the higher the color uniformity, the higher the price of white light WLED. However, when $\text{SrBaSiO}_4:\text{Eu}^{2+}$ was used to produce WLEDs, the cost would be much lower but the quality requirements are still met totally. That is the reason why $\text{SrBaSiO}_4:\text{Eu}^{2+}$ become a powerful factor which is widely used in the lighting market today.

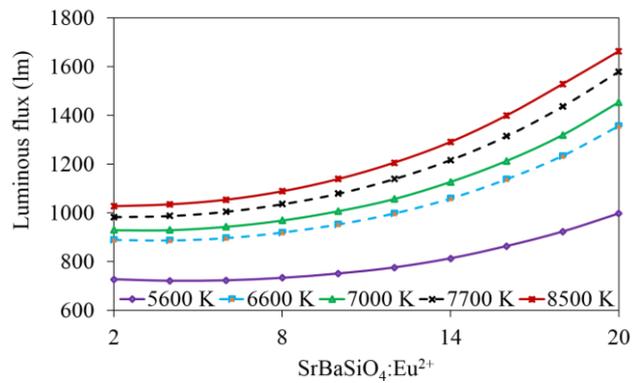


Fig. 8. The luminous flux of WLEDs as a function of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration

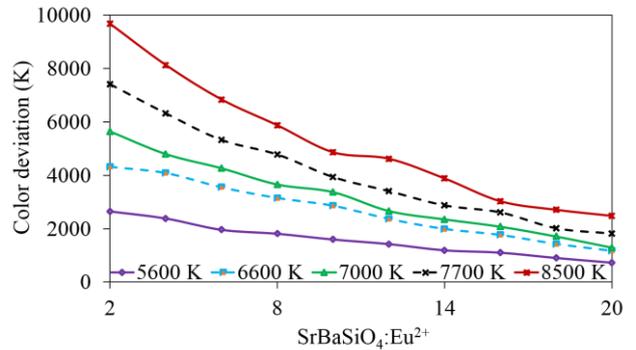


Fig. 9. The color deviation of WLEDs as a function of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration

Color uniformity is an important index but it cannot totally evaluate the color quality of WLEDs. Thus, the current study provided a correlation between the color rendering index and the color quality ratio. Color rendering index evaluates the true color of an object when the light is shining. However, excess green light is responsible for the color imbalance between the three main colors: blue, yellow, and green, which affects the color quality of WLEDs, reducing the color accuracy of WLEDs. In Fig. 10, the CCT of 8500 K was dramatically reduced with the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration of the 14 wt%. The expression in Figure 10 shows that the CRI

tends to decrease slightly with the presence of the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ remote phosphor layer. This can be acceptable because CRI is just one element of CQS while CQS is true overall color quality index because it covers three factors: color rendering index, viewer preference, and color coordinates. Fig. 11 showed the increase in CQS with the presence of the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ remote phosphor layer. In particular, when the $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration increased no more than 10% wt, the change in CQS was negligible.

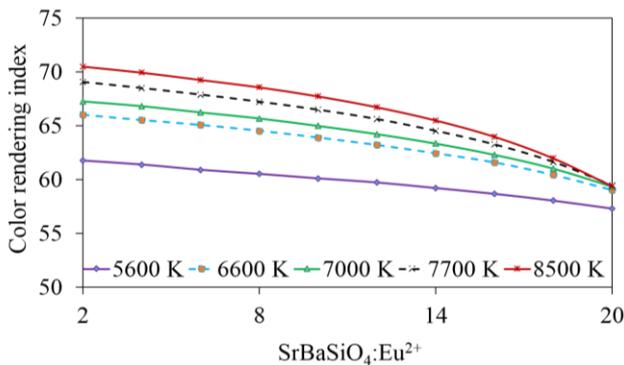


Fig. 10. The color rendering index of WLEDs as a function of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration

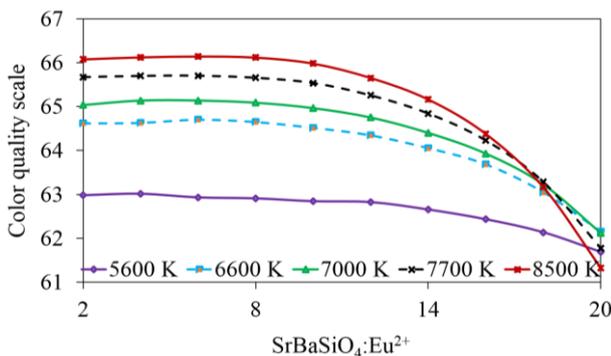


Fig. 11. The color quality scale of WLEDs as a function of $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentration

When $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentrations are greater than 10% wt, both CRI and CQS are significantly reduced due to a severe color imbalance since green is dominant at this time. Therefore, the choice of suitable phosphor concentration levels becomes very necessary in the application of green phosphor $\text{SrBaSiO}_4:\text{Eu}^{2+}$.

4. Conclusions

In conclusion, this paper presented the effect of green phosphor $\text{SrBaSiO}_4:\text{Eu}^{2+}$ on the optical properties of the dual-layer phosphor structure. Studies based on Monte Carlo simulations and calculations have proven that $\text{SrBaSiO}_4:\text{Eu}^{2+}$ is the wise choice to improve color uniformity. This does not only work on WLEDs with a low color temperature of 5600 K but also on color temperatures above 8500 K. Thus, the results of this study

have achieved the goal of improving color and luminosity while using the remote-phosphor structure will encounter many difficulties. However, there is still a limitation on CRI and CQS. Specifically, when $\text{SrBaSiO}_4:\text{Eu}^{2+}$ concentrations increase excessively, CRI and CQS decrease sharply. Therefore, it is necessary to carefully consider and select the appropriate concentration to meet the requirements of the customer as well as the target of the manufacturer. The information provided in this article is extremely important and helpful for reference to the production of WLEDs with superior color homogeneity and luminous flux.

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