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SrBaSiO₄:Eu²⁺ phosphor: a novel application for improving the luminous flux and color quality of multi-chip white LED lamps

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ABSTRACT

This paper described in detail the chromatic homogeneity and luminous flux influences in producing better quality white LED devices with various phosphor layers (MCW-LEDs). The method is to let Eu^{2+} -activated strontium–barium silicate (SrBaSiO4: Eu^{2+}) mixed with their phosphor compounding, which results in notable impact on lighting performance. The increase in concentration of yellow-green-emitting SrBaSiO4: Eu^{2+} phosphor also promotes the color performance and lumen output of WLED devices at high correlated color temperature around 8500K. This is the first time this approach is applied and it results can be utilized for better understanding of optical properties interaction with phosphor materials. Although SrBaSiO4: Eu^{2+} receives many positive responses, we still need to limit it concentration for high SrBaSiO4: Eu^{2+} concentration is detrimental to CQS. The appropriate choice of concentration and size of SrBaSiO4: Eu^{2+} is the principal factor to decide the performance of MCW-LEDs.

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1. INTRODUCTION

Recently, perfecting the optical properties of WLED, especially the luminous efficacy and color-related attributes, is a leading research goal of many scientists, which has been mentioned in previous papers [1-5]. After many experiments, Anh and his colleagues discover the mixture of YAG:Ce³⁺ phosphor compounding with SiO₂ particles could significantly increase the lighting efficiency of white LED lighting devices [6-8]. In addition, the combination of yellow YAG:Ce³⁺ phosphor with the silicone substance could support the blue light and yellow light extraction [9-12] and also generate the white light. This process includes two stages: disintegrating in phosphor compounds and moving through phosphor particles, in which the YAG:Ce³⁺ phosphor layer will metabolize the blue light and simultaneously make it become weaker [13-16]. However, the converted yellow light tends to be more intense after every scattering, which makes the normal extracted light from the WLED to be more blue, while if it is placed near the LED exterior, the color tend to be more yellow [17-19]. Won was in collaboration with his partner to prove the marked effect of phosphor configuration on the lighting efficiency of LED devices with great color quality scale (CQS), where blue LEDs and multiple phosphor materials such as green (Ba,Sr)₂SiO₄:Eu²⁺ and red CaAlSiN₃:Eu²⁺ were applied in various phosphor structures to achieve optimal efficiency [20]. In contrast,

Oh's group proposed the multiple phosphor LED with three primary colors green, yellow, and red, which focus on enabling the luminous flux and color consistency to reach the most satisfactory result [21]. Zhang's team used multi-chromatic materials for better quality of CRI in LED products [22]. The improvement of CRI and optical output is of interest to many researchers, but the increase in color uniformity seems to be ignored. Furthermore, their research subjects is solely white LED with one chip, which gives out low CRI and therefore not really applicable to modern lighting applications that require multiple chips for lighting devices with high CCTs.

Understanding the needs for an advanced study in optical properties enhancement and more relevant results to the current WLED structures, this research present experiments in which the chromatic phosphor is employed in a multi-chip LED configuration. Based on previous studies that targeted at the optical properties such as lumen output, CRI, and chromatic quality, we introduce an innovative research method that exploits the advantageous findings while adding in new aspects for research. For example, the application of red phosphor Mg₈Ge₂O₁₁F₂:Mn⁴⁺ in the dual-layer WLED at 6600 K CCT is confirmed to optimize the lumen output and CRI. On the other hand, the remote phosphor configuration is proven to be the optimal set up for WLED among the three configurations including the in-cup phosphor and conformal phosphor after the achieved lumen output of this configuration is demonstrated as better than others [23-25]. However, there are currently a lack of diversity in the applicable types of phosphor for WLED. Therefore, we conducted experiments on a new phosphor, known as the yellow-green phosphor SrBaSiO₄:Eu²⁺, on WLEDs with three different phosphor structures respectively. The approach this research suggests allow comparison between performances of SrBaSiO₄:Eu²⁺ in each phosphor configuration, which assists the identification of the best combination. The yellow-green SrBaSiO₄:Eu²⁺ phosphor plays a supporting role for chromatic homogeneity and luminous efficacy so that the greatest efficiency could be attained from the phosphor materials in MCW-LEDs during the scattering process. The participation of SrBaSiO4:Eu²⁺ particles will decide lighting quality color uniformity. Regarding the remote phosphor structure, there are the remote structure with two flat phosphor layers (FDRP) and the remote structure with two curved phosphor layers (CDRP). However, a preceding study claims that FDRP is the better choice as CDRP yields less lumen while having a more complex fabricating procedure, therefore, the dual-remote WLED in this research is simulated with FDRP arrangement [26-28]. This research has 3 phases: (1) Simulating the MCW-LEDs prototype; (2) Applying SrBaSiO₄:Eu²⁺ as a phosphor material for the MCW-LEDs; (3) Studying the influences of SrBaSiO₄:Eu²⁺ on the optical properties, including the emission spectra and the scattering coefficient. In sum, mixing SrBaSiO₄:Eu²⁺ and YAG:Ce³⁺ phosphors together is considered as an optimal method to maximize quality and lighting efficiency. The conclusions in this manuscript are useful references for luminous flux and chromatic performance development while also effective in supporting the manufacturers in choosing the suitable configuration for quality WLEDs production.

2. PREPARATION AND SIMULATION

2.1. Preparation of yellow-green SrBaSiO4:Eu²⁺ phosphor

Yellow-green SrBaSiO₄:Eu²⁺ phosphor particles contain many superb characteristics, for example, great quantum yield and consistency at high temperature with an emission peak up to 2.36 eV, which makes it become more popular recently [29]. The particle sizes and concentration directly affect the luminescence properties of SrBaSiO₄:Eu²⁺ phosphor. The composition including SrCO₃, BaCO₃, SiO₂, Eu₂O₃, NH₄Cl, and Eu²⁺ ion are used as raw materials as shown in Table 1. Some heavy duty and long lifetime fluorescent lights prefer to use SrBaSiO₄:Eu²⁺. Therefore, it turns into the most well-known commercialized oxide material in the lighting market. The fabrication process of SrBaSiO₄:Eu²⁺ is described specifically as follows:

Firstly, the materials are soaked in water to be dissolved before being crushed into small particles. Then they will be pulverized as soon as being dried in the air. Secondly, the powder will be heated in a closed quartz tube with CO at 1100°C for one hour and crushed into powder by dry grinding. Then 5.4 g NH₄Cl is added and mixed with this compound by dry grinding. Next, they will be reheated in the capped quartz tubes with CO at 1100°C for one hour and will be pulverized. Finally, the materials are washed in water several times (pH ranged from 10 to 12) and dried afterward.

Table 1. Composition of yellow-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_2O_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

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2.2. Construction of MC-WLEDs

Based on the LightTools 8.5.0 software and Monte Carlo method [30-32], the prototype simulation of MCW-LEDs is conducted through two main periods with a flat silicone layer. Setting up and manufacturing the framework and lighting characteristics of MCW-LED at first, then the influences of phosphor compounding with different SrBaSiO₄:Eu²⁺ concentration is analyzed based on the performance of MCW-LED with different phosphor geometries. The realistic simulated product is present in Figure 1(a) with technical measurements can be seen from Figure 1(b). Among them, the two types of compounding which have average CCT of 8500 K, conformal and in-cup phosphor, are picked to analyze. A distinct description for MCW-LEDs with conformal phosphor structure, in-cup structure, and remote structure with the average CCT of 8500 K was shown in Figure 1(c), (d), and (e) respectively. The components of the MCW-LED is an 8x2.07x9.85 mm at bottom, height, and surface reflector, 0.08 mm phosphor layer in phosphor compound, and 9 LED chips. Each chip is 1.14 mm² in acreage and 0.15 mm in height with 1.16 W energy emission for each individual at 453 nm maximum wavelength connected to the base of the structure.



Figure 1. Illustration of phosphor-converted MCW-LEDs as doping SrBaSiO₄:Eu²⁺:
(a) the actual MCW-LEDs and (b) its parameters; (c) conformal phosphor geometry (CPG);
(d) in-cup phosphor geometry (IPG); and (e) remote phosphor geometry (RPG)

As can be seen from Figure 1(c), the chips are covered with phosphor particles diffused by conformal coating. Conversely, separate methods are used for the structure with in-cup phosphor and for mixture with silicone lens. The scattering analysis of these two structures was carried out based on Mie-theory. The measurement for particles in this research is 14.5 μ m in size, similar to the real parameter. The phosphor composite is a combination of SrBaSiO₄:Eu²⁺, YAG:Ce³⁺, and the silicone gel with the same sizes as the real parameters and own the refractive indexes respectively of 1.85, 1.83, and 1.52. Once the refractive index and diameter of phosphor particles are set, we can study the emission spectra of research subjects. Figure 2 (top) described the emission spectra of the conformal structure WLED at different SrBaSiO₄:Eu²⁺ concentration fluctuating from 0% toward 24%. Figure 2 (bottom) expresses in details the emission spectra of in-cup structure WLED with different ranges of SrBaSiO₄:Eu²⁺ concentration which can reach 1.4%. According to this chart, the lighting efficiency of MCW-LEDs will improve after SrBaSiO₄:Eu²⁺ phosphor are added to the structure.



Figure 2. Emission spectra of multiple phosphors: (a) CPG; (b) IPG; and (c) RPG

3. COMPUTATION AND DISCUSSION

Based on the Mie theory [23-35], the scattering coefficient μ_{sca} are computed to verify the optical properties of phosphor compounding. The relationship among the scattering coefficient (SC), the wavelength, and diameter of SrBaSiO₄:Eu²⁺ particles is expressed in the formula below:

$$\mu_{sca}(\lambda) = \frac{c}{\bar{m}}\bar{C}_{sca}(\lambda) \tag{1}$$

$$\bar{C}_{sca}(\lambda) = \frac{\int C_{sca,D}(\lambda) f(D) dD}{\int f(D) dD}$$
(2)

$$\bar{m} = \frac{\int m_i(D) f(D) dD}{\int f(D) dD}$$
(3)

$$C_{sca}(\lambda) = \frac{P_{sca}(\lambda)}{I_{inc}(\lambda)}$$
(4)

where, f(D) is the function of diameter distribution, c is the distribution density of phosphor (g/cm³), $C_{sca,D}$ is the scattering cross-section of the phosphor having particle diameter D. While $\overline{C}_{sca}(\lambda)$ and \overline{m} are the scattering cross-section and the particle mass of the phosphor integrated over f(D), respectively. $P_{sca}(\lambda)$ and $I_{inc}(\lambda)$ are the scattered power by phosphor particles and the irradiance intensity in turn.

When SrBaSiO₄:Eu²⁺ phosphor involves in the scattering, the scattering coefficient (SC) of the phosphor layer is indicated in Figure 3. Concentrations and other agents of SrBaSiO₄:Eu²⁺ will be the catalyst that makes SC of phosphorus mixture significantly change. This also verifies that previous concentrations and stimulation of SrBaSiO₄:Eu²⁺ affect the color quality of both CPG and IPG structures. When SrBaSiO₄:Eu²⁺ concentration increases, SC also tends to develop regardless of SrBaSiO₄:Eu²⁺ particle size. With a size of about 1 μ m, SC can achieve optimum value and making color uniformity better regardless of the larger sizes. Therefore, if the target is CQS, SrBaSiO₄:Eu²⁺ size can be approximately 1 μ m. When it is approximately 7 μ m, the SC value of phosphorus is more stable regardless of the increase in SrBaSiO₄:Eu²⁺ concentration, which brings color quality (CQS) of LED lights a lot of benefits. As a result, if the target is CQS, size less than 7 m can be chosen. Apparently, the SC value depends on the amount of SrBaSiO₄:Eu²⁺ particles and also their diameter, which is why SrBaSiO₄:Eu²⁺ can be used to increase luminous efficiency and color uniformity of the LEDs. The appearance of SrBaSiO₄:Eu²⁺ is one of the factors affecting the color deviation result of MCW-LED as illustrated in Figure 4.



Figure 3. Scattering coefficients of phosphor compounding at 453 nm as a function of the concentration and size of SrBaSiO₄:Eu²⁺: (a) CPG; (b) IPG



Figure 4. The CCT peak-valley deviation as a function of the concentration and size of SrBaSiO₄:Eu²⁺: (a) CPG; (b) IPG; and (c) RPG

The CCT peak deviation decreases significantly when $SrBaSiO_4:Eu^{2+}$ is involved and keep going as the phosphor concentration increases. That means the spatial distribution of MCW-LED with $SrBaSiO_4:Eu^{2+}$ is much better than when the $SrBaSiO_4:Eu^{2+}$ is absence. There must be a balance between performance factors and optimization issues because if we only focus on optimizing one element, optical systems can be weak in other aspects. In addition, the greatest CQS and the luminous efficiency of white LED packages cannot be achieved at the same time. The solution is that the spectrum and wide source efficiency must be maximized in monochromatic radiation of 555 nm wavelength to better CRI. Therefore, the remaining optical properties that need optimization are CQS, luminous flux, and CCT P-V deviation values. Below are the Figure 5 that illustrates the luminous efficacy of WLED with $SrBaSiO_4:Eu^{2+}$ and Figure 6 that shows the color quality scale (CQS) in WLED with SrBaSiO₄:Eu²⁺. It can be found from the simulation results these figures that the luminous yield grows along with SrBaSiO₄:Eu²⁺ concentration. However, if SrBaSiO₄:Eu²⁺ concentration continues to grow higher, the luminous flux will still follow, even though CQS will decrease. Therefore, for WLEDs that focus on the CQS values, it is advisable to limit the phosphor concentration under permitted level to prevent damaging CQS. Moreover, SrBaSiO₄:Eu²⁺ concentration develops backward but not significantly, it will contribute to better CCT stability and greater luminous efficiency.



Figure 5. Luminous efficacy as a function of the concentration and size of $SrBaSiO_4:Eu^{2+}$: (a) CPG; (b) IPG; and (c) RPG



Figure 6. Color quality scale as a function of the concentration and size of $SrBaSiO_4:Eu^{2+}$: (a) CPG; (b) IPG; and (c) RPG

4. CONCLUSION

In summary, the effectiveness of the yellow-green SrBaSiO₄:Eu²⁺ in improving color quality and light output of WLEDs devices was verified through some references in the article. The experimental results from this research are also viable to conclude about the optical properties influences as follows. First, no matter what the average CCT value, color uniformity can be very diverse according to Mie scattering theory. This is the effect of light loss from scattering in white LED packages. Second, through Monte Carlo method, the luminous efficacy is confirmed to increase with SrBaSiO₄:Eu²⁺ concentration. Particularly, the emitted light will grow as long as SrBaSiO₄:Eu²⁺ concentration keep increasing at any phosphor geometry. Finally, selecting SrBaSiO₄:Eu²⁺ phosphor is the appropriate solution for developing phosphorus materials as well as producing white LED packages. Although SrBaSiO₄:Eu²⁺ phosphor is an effective enhancement material, the concentration of this phosphor must be monitored in application as any amount of SrBaSiO₄:Eu²⁺ is good for lumen output and CCT deviation, however, CQS can be damaged if the SrBaSiO₄:Eu²⁺ concentration is too high. This research not only presented a new approach by applying the yellow-green phosphor to WLED but also compared the performance based on the type of phosphor geometry and phosphor concentration, which opens more options for manufacturers to develop their products and leaves valuable references for future studies.

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