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Influences of the Sm^{3+}-Eu^{3+} codoped $Ba_2Gd(BO_3)_2Cl$ phosphors on the commercial white light emitting diodes

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ABSTRACT

The color quality of current commercial white light emitting diodes (w-LEDs) suffers low performance owing to the lack of the red-emission component. Developing quality and stable red-emission phosphors is feasible among various approaches to obtain the red spectral supplement for the w-LEDs in the pursuit of color quality improvement. In this paper, the Sm^{3+}-Eu^{3+} codoped $Ba_2Gd(BO_3)_2Cl$ (BGBC:Sm-Eu) red phosphor was proposed for using in commercial w-LEDs. Its luminescence and influences on w-LED properties were simulated and presented. The solid-phase method was utilized for the fabrication of the phosphor. The results indicated that the phosphor emitted the strong emission in orange-red region with a peak centering at 593 nm. It can be caused by the proficient power shift between Sm3+ and Eu3+. In the w-LED package, the presence of BGBC:Sm-Eu phosphor stimulated the scattering efficiency to promote the blue-light conversion and extraction. The orange emission spectrum of the w-LED increased with the higher BGBC:Sm-Eu doping amount. The luminous strength of the w-LED was enhanced and so was the color temperature uniformity. The color rendering properties declined with high BGBC:Sm-Eu phosphor concentration owing to the red-light dominance over the light spectrum. The BGBC:Sm-Eu phosphor is a promising red phosphor for improving commercial w-LED color-temperature stability and luminosity. It also helps to obtain full-spectrum w-LED with high color rendition when combined with other blue-to-green luminescent materials.

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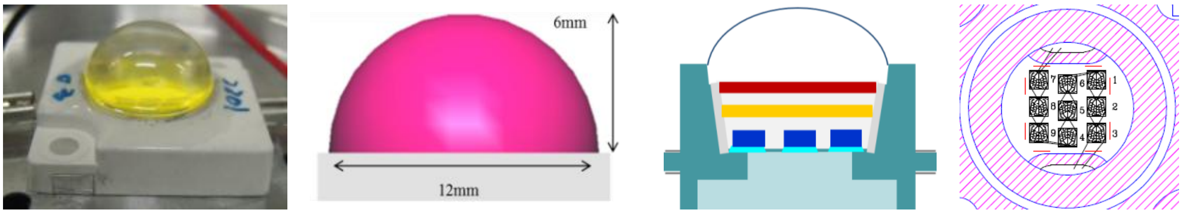
1. INTRODUCTION

Emerging as a promising substantial light source, the solid-state illuminating, especially the light emitting diode (LED), has been developed and applied in many fields, from indoor and outdoor lighting and display [1] to advanced biological sensing [2] and medical treatments [3]. The commercial white LED (w-LED) is usually comprised of ultraviolet/near-ultraviolet or blue LED chips and yellow phosphor compounds to convert the emitted light from the chip [4], [5]. The commonly used yellow phosphor is the YAG:Ce^{3+} phosphor. Though the good luminescence can be realized with the YAG:Ce^{3+} phosphor, the red-emission in the visible spectrum is not observable, resulting in inferior color quality of the w-LED. Therefore, figuring out the phosphor materials to efficiently supplement the orange-red spectra under near-ultraviolet to blue excitation sources is critical to achieve the full-spectrum w-LED with significantly improved color reproduction performance [6]-[8]. Using rare-earth ions as dopants for phosphor compounds has been interested extensively due to their excellent luminescence with sharp emission lines. The Eu^{3+} has been one of the most favorite dopants for producing good red phosphor. Though it can yield eminent red emission, the fluorescent efficacy is relatively low because its f-f transition presents inefficient absorption performance. Thus, to get the luminescence efficiency of the Eu^{3+} improved, a sensitizer ion is essential. The Sm^{3+} ions are recognized as the effective sensitizer for Eu^{3+} in the same host [9]. Besides, the selecting the suitable host advantageous for the ion dopants' excitation and emission spectra is critical. Among many analyzed host matrices, the halo-borates offer excellent functional versatility due to its diverse structural types and the adjustability of cation coordination environment. Moreover, this borate host type possesses high chemical stability and thermostability and excellent luminescent features while can be synthesized under low temperature. Besides, the halo-borate host can yield broad band gap, avoiding absorption of activated ion dopants' emission, leading to strong and high-efficiency luminescence [10], [11]. Therefore, in this work, the halo-borate $Ba_2Gd(BO_3)_2Cl$ is used as a host for co-doping Eu^{3+} and Sm^{3+} ions. The conventional solid-phase method is applied to synthesize the Sm^{3+},Eu^{3+} codoped $Ba_2Gd(BO_3)_2Cl$ (BGBC:Sm-Eu) red phosphors. The power shift between Sm^{3+} and Eu^{3+} would be efficient with the Eu^{3+} emission strengthened. The BGBC:Sm-Eu is utilized with YAG:Ce yellow phosphor and blue LED chip (465 nm) to build a w-LED model. The properties of the w-LED are investigated on the increasing concentration of BGBC:Sm-Eu phosphor. BGBC:Sm-Eu phosphor would be an appropriate phosphor to contribute to obtaining the full-color w-LED when combining with other blue and green luminescence materials. Besides, this phosphor can support the improvement of w-LED luminous intensity and color-temperature uniformity [12], [13].

2. METHOD

The synthesis of the BGBC:Sm-Eu phosphor was performed via the solid-phase reaction. The concentration of Sm^{3+} was constant at 4 mol% while that of the Eu^{3+} varied in the range of 1-7 mol%. The details of starting ingredients (with purity >99.5%) and synthesizing process of the phosphor are shown in Table 1. After the required product was obtained, the luminescence characterization for the phosphor was carried out with the Hitachi F-7000 fluorescent spectrophotometer coupling with an excitation source from one xenon light under 150 W. The assessment was carried out under room temperature. The modelling of a w-LED with BGBC:Sm-Eu phosphor, yellow phosphor YAG:Ce, and blue-pumped LED chips (465 nm) are depicted by Figure 1. Figure 1(a) displays the real photo of the built w-LED package, while Figure 1(b) shows the 3D simulated model of the package created by the LightTools software [14], [15]. Figure 1(c) and 1d present the internal details of the w-LED via the cross-section and chip-bonding diagrams, respectively.



(a) (b) (c) (d)

Figure 1. w-LED package in the work: (a) real photo of the w-LED, (b) w-LED 3D simulation by LightTools software, and (c) w-LED cross-section details, and (d) chip cluster of the w-LED

Table 1. BGBC:Sm-Eu starting ingredients and creating stages

Ingredients	Producing steps
$BaCO_3$	1. Weigh the ingredients in a stoichiometric ratio and get them
$BaCl_2{\cdot}2H_2O$	well mixed in an agate mortar.
H_3BO_3	2. Blend the mixture with 5 mol% H_3BO_3 flux and evenly ground
Gd_2O_3	in an agate mortar.
Eu_2O_3	3. Put the mixture in a corundum crucible and calcinate for 6
Sm_2O_3	hours at 1652 ^{0}F in a tube furnace.

3. RESULTS AND DISCUSSION

3.1. Computational luminescence for BGBC:Sm-Eu

The BGBC:Sm-Eu phosphor exhibited strong and broad absorption spectra with multiple peaks in 365-415 nm and at 465 nm. The most intense peak was noticed at 393 nm. Such numbers are matched with the ultraviolet and blue wavelength of the LED chips. In other words, this BGBC:Sm-Eu phosphor can be utilized in ultraviolet/blue-pumped w-LEDs [16]-[19]. The emission spectra of the BGBC:Sm-Eu phosphor with 4 mol% of Sm^{3+} and 4 mol% of Eu^{3+} showed multiple peaks with three eminent centers at 593 nm, 612 nm, as well as 623 nm. Among these peaks, the most significant peak was at 593 nm, which is generated from the shifts between $^5D_0$ and $^7F_1$ for Eu^{3+}. Besides, the other lower peaks at 612 and 623 nm are induced by the shifts between $^5D_0$ and $^7F_2$ for Eu^{3+}. Such findings denoted that the emission peaks originated from Sm^{3+} transitions are not included in the emission spectrum of the phosphor. This means the increasing doping dosage of the Eu^{3+} ion had the Sm^{3+} emission declined, suggesting power shift betweeb Sm^{3+} ion and Eu^{3+}. The $^4G_{5/2}$ status energy for ion Sm^{3+} would be adjacent to the Eu^{3+} ion's $^5D_0$ state energy [20]. As a result, the power shift between Sm^{3+} and Eu^{3+} would be allowed and effective.

The lifetime decay curves of the Sm^{3+} luminescence can be used to examine the power shift between Sm^{3+} and Eu^{3+}. We can determine the fitted dual-exponential degradation curves for BGBC:Sm-Eu luminescence as the expressions (1), (2):

$$L_I = B_1\, exp(-t/\tau_1) + B_2\, exp(-t/\tau_2) \quad (1)$$

$$\tau_a = \frac{(B_1\tau_1{}^2 + B_2\tau_2{}^2)}{(B_1\tau_1 + B_2\tau_2)} \quad (2)$$

where L_I means the luminescence intensity, and τ_a denotes the average decay time. B_1 and B_2 are the fitting constants. τ_1 and τ_2 represent the rapid and steady degradation times. With the increase in Eu^{3+} doping amount, the τ_a value decreases, implying the effective energy transfer with non-radiation mechanism from Sm^{3+} to Eu^{3+}. Besides, the decreasing Sm^{3+} luminescence lifetimes with increasing Eu^{3+} concentration is also induced by the concentration quenching effect [21]-[23]. The relation between the concentration quenching and the power shift between Sm^{3+} and Eu^{3+} can be demonstrated via the critical range (D) between these two ions. The critical distance can be determined using the Blasse (3):

$$D = 2\left(\frac{3V}{4\pi CZ}\right)^{1/3} \quad (3)$$

V represents the unit-cellule volume; C represents the optimal concentrations of Eu^{3+} and Sm^{3+} ions; Z represents the number of units in BGBC:Sm-Eu, which equals to 2. By estimating the D between Sm^{3+} and Eu^{3+}, it is possible to determine how the power shift between Sm^{3+} and Eu^{3+} works. As the calculated D would be larger than 5 Å, the multipolar interaction is accountable for the power shift rather than the exchange interaction. The theory of Dexter can be used to express the multipolar interaction for this energy-transfer process (4):

$$\frac{\eta_{Sm0}}{\eta_{Sm}} \propto S^{n/3} \quad (4)$$

where η_{Sm0} and η_{Sm} represent the luminescent quantum efficacies for Sm^{3+} accompanied and not accompanied by Eu^{3+}. S indicates the Eu^{3+} and Sm^{3+} amounts combined. n is the constant indicating the multipolar-interaction types. As n equals to 8, the best fitted result was obtained, indicating that the interaction type for $Sm^{3+} \rightarrow Eu^{3+}$ energy transfer is dipole – quadrupole.

According to the obtain computation results, the BGBC:Sm-Eu phosphor is excitable under ultraviolet (UV) to visible blue wavelength range. This means the red phosphor can be used for either blue-pumped (465 nm) or UV-pumped (365 nm) LEDs, indicating that the phosphor is applicable in this study's simulation settings. The emission of BGBC:Sm-Eu phosphor is orange-red, enabling the production warm-light white LED, considering the lighting devices' manufacturing purposes and applying contexts. Moreover, 4 mol% was found to be the most proper concentration for both ion dopants Sm^{3+} and Eu^{3+}. More than such a concentration level, the concentration quenching effects occurs, resulting in declining emission power of the phosphor.

3.2. Influences of BGBC:Sm-Eu phosphor dosage on w-LED performances

When the BGBC:Sm-Eu phosphor used in combination with the YAG:Ce yellow phosphor compound, it is imperative to adjust the YAG:Ce concentration in response to the variation in BGBC:Sm-Eu phosphor dosage. According to Figure 2, the concentration of YAG:Ce phosphor is inversely proportional to that of the BGBC:Sm-Eu phosphor. This is essential for changing the scattering efficiency and stabilizing the correlated color temperature (CCT). Subsequently, Figures 3 and 4 depict the calculated scattering coefficients (SCs) and reduced scattering coefficients (RSCs) of light in the w-LED with the BGBC:Sm-Eu phosphor compound. Significantly, both figures show an enhancement of SCs and RSCs with increasing doping dosage of the red phosphor. This enhancement provides a higher probability of forward light propagation and blue-light absorption, thereby leading to better conversion efficiency, improved color uniformity, and increased luminous intensity of the w-LED light [24].

The orange-red transmission power is intensified due to enhanced scattering performance, as illustrated in Figure 5. When a higher concentration of BGBC:Sm-Eu phosphor is used, the emission peak positions remain unchanged, but the orange-red emission peak intensity increases while that of the blue region emission decreases correspondingly. Such a phenomenon indicates better absorption of blue light and effective light conversion by the BGBC:Sm-Eu red phosphor. Essentially, under blue-chip excitation, the BGBC:Sm-Eu particles emit orange-red light and absorb some blue light, converting it into orange-red light, resulting in a more intense emission line in the orange-red spectrum with a peak at 593 nm. Additionally, in the presence of BGBC:Sm-Eu phosphor, the intensity of the blue-emission peak (465 nm) decreases but remains noticeable, indicating efficient and sufficient extraction of blue light for good luminosity of the w-LED. Notably, the benefit of the BGBC:Sm-Eu phosphor to the lumen output of the w-LED can be reinforced by the data in Figure 6. It particularly illustrates an increase in lumen output of the w-LED with the increase in BGBC:Sm-Eu doping amount, which denotes the benefits of improved forward scattering [25] and the ability to enhance the luminescence efficiency of the BGBC:Sm-Eu phosphor.

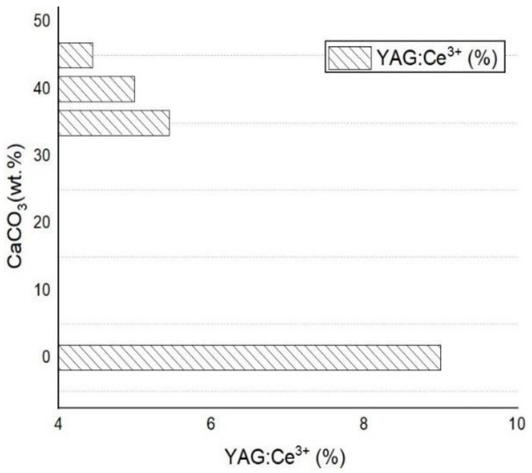


Figure 2. YAG:Ce concentration in the presence of BGBC:Sm-Eu phosphor

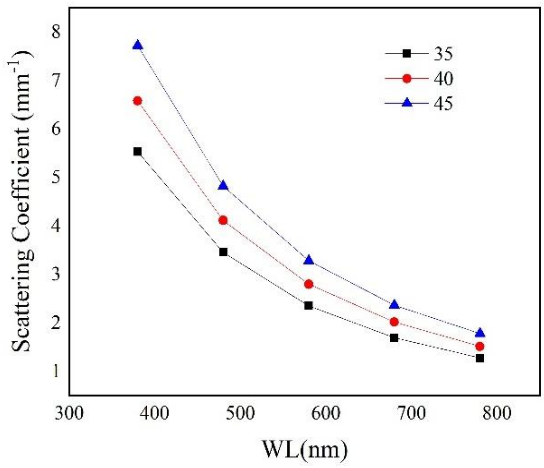


Figure 3. Scattering coefficients in the presence of BGBC:Sm-Eu phosphor

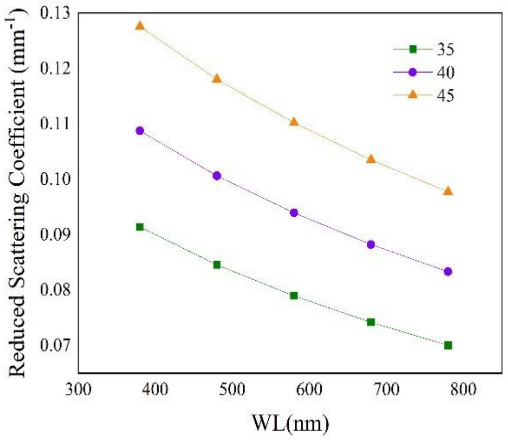


Figure 4. Reduced scattering coefficients in the presence of BGBC:Sm-Eu phosphor

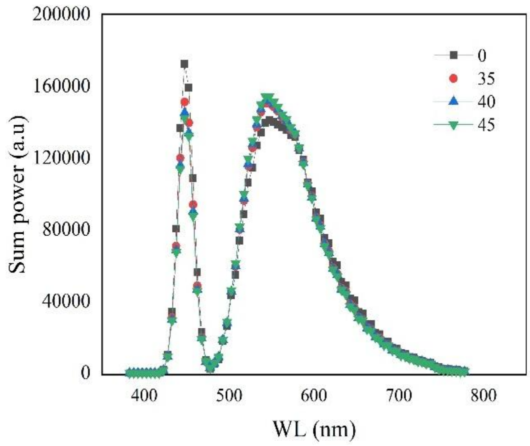


Figure 5. The transmission spectra of the w-LED in the presence of BGBC:Sm-Eu phosphor

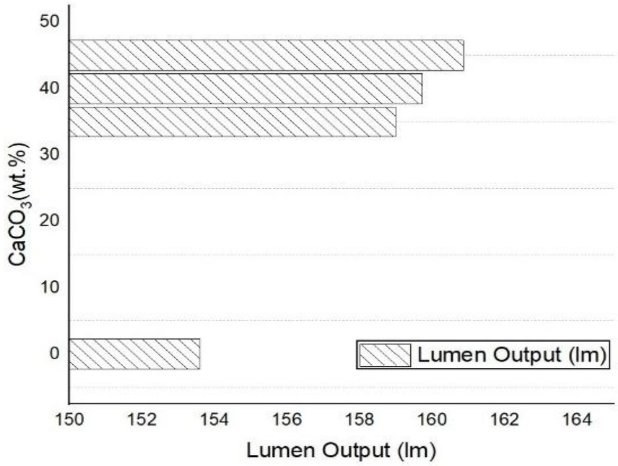


Figure 6. The lumen output of the w-LED in the presence of BGBC:Sm-Eu phosphor

As presented, the lumen output is promoted with the increasing BGBC:Sm-Eu phosphor amount. The increase in luminosity often induces a decrease in color quality and a rise in CCT of the white LED. Thus, the following simulation examination are performed on the CCT uniformity and color rendition assessments involving BGBC:Sm-Eu concentration. The changes in CCT values of the w-LED light with increasing BGBC:Sm-Eu phosphor amounts is then depicted in Figure 7. The CCT distribution uniformity assessment includes the angle-dependent CCT as shown in Figure 7(a) and deviation of CCT (d-CCT) as shown in Figure 7(b). These data will serve as insights into the impact of BGBC:Sm-Eu phosphor on the color consistency performance of white light. Specifically, on the presence of BGBC:Sm-Eu phosphor, the reduction in CCT intensity observed at the center is revealed. In other words, the CCT level is reduced when adding the red-phosphor BGBC:Sm-Eu to the layer. Furthermore, the lower deviation of CCT is observed as the BGBC:Sm-Eu phosphor is introduced to the w-LED package, compared to the case without using the red phosphor. Such results are potentially attributable to a higher red-emission intensity in the visible spectrum. The lowest d-CCT value is attained with 35 wt% of BGBC:Sm-Eu. Beyond this concentration, an increase in d-CCT is observed, albeit significantly lower than in the absence of BGBC:Sm-Eu phosphor. Hence, to obtain the highest CCT uniformity with relatively increased lumen intensity, 35 wt% of BGBC:Sm-Eu phosphor is advisable.

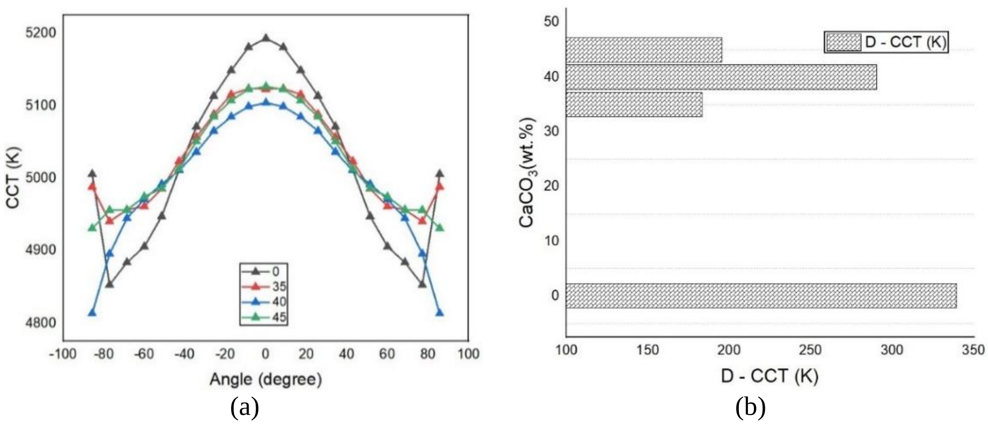


Figure 7. CCT values of the w-LED with BGBC:Sm-Eu phosphor (a) angular CCTs and (b) D-CCT

As anticipated at the onset of the study, the enhancement of red-light through the use of BGBC:Sm-Eu phosphor has the potential to improve the color-reproduction efficiency of the w-LED. To investigate this, we took into consideration the color rendition index (CRI) and color quality scale (CQS), as presented in Figures 8 and 9, respectively. However, the incremental concentration of BGBC:Sm-Eu phosphor leads to a gradual decrease in both CQS and CRI. With an escalating BGBC:Sm-Eu concentration, the orangish-red region is amplified, while green emission is not observed, and blue emission gradually diminishes. Nonetheless, superior CRI and CQS necessitate a broad spectrum that encompasses at least three wavelength regions of critical colors: blue, green, and red [26]-[28]. Therefore, this dominance of red-light results in a significant color imbalance and hampers chromatic rendition performance. Despite the decreasing value of both chromatic rendering parameters, the CQS shows a higher level of rendering than the CRI. Specifically, the CQS values are consistently above 60, while those of CRI are below 60 at all BGBC:Sm-Eu concentrations. The CQS is considered more complex to control than the CRI because it evaluates not only the eight color samples of the CRI, but also a wider range of vivid colors with saturation. It also considers color coordinating performance and visual preferences from the users. This result implies that the BGBC:Sm-Eu phosphor holds the potential to improve the overall color quality of the w-LED. Additionally, it is possible to address the color rendering performance issue by supplementing the deficient emission colors. For example, using the BGBC:Sm-Eu phosphor in conjunction with other green and blue luminescence phosphors could potentially yield a full-color spectrum.

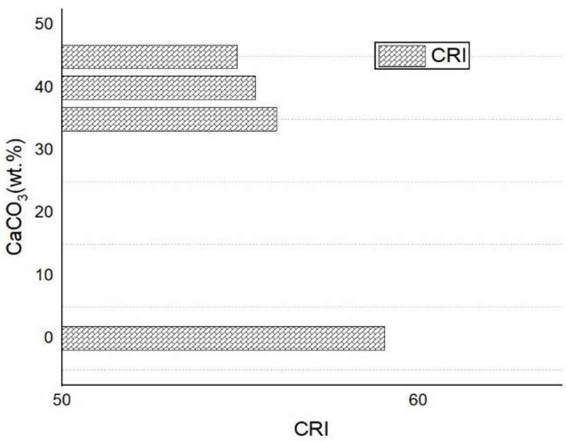


Figure 8. CRI of the w-LED with BGBC:Sm-Eu phosphor

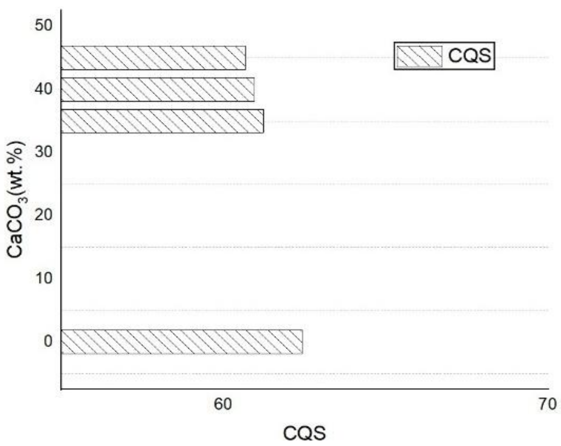


Figure 9. CQS of the w-LED with BGBC:Sm-Eu phosphor

4. CONCLUSION

The w-LED is well-known for its high luminous efficiency, but there is enormous room for improvements in terms of color fildety and uniformity. Therefore, developing the phosphor to supplement the emission color shortage is the critical act in the development in the road to achieve the high-quality and performance w-LED devices. The red emission from BGBC:Sm-Eu phosphors can contribute addressing low color uniformity and reproduction persistent in general w-LED devices. The computational results indicated that the power shift between Sm^{3+} and Eu^{3+} would be efficient, and the phosphor emitted strong orangish-red with at peak centred at 593 nm. Results demonstrated in this work show considers the scattering factor, which is a critical factor influence the light transmission efficiency in the w-LED. This work evaluates the rendering factor with both CRI and CQS for a more comprehensive evaluation, by which the potential of using red phosphor to reach higher color production w-LEDs is emphasized. Particularly, the increasing integrating concentration of BGBC:Sm-Eu phosphor led to the decrease in YAG:Ce dosage to improve the scattering efficiency of light. The emission spectrum of the w-LED contained two eminent peaks located at 465 nm (blue) and 593 nm (orange). Such findings showed that the forward blue-light scattering and conversion were all bettered with the addition of BGBC:Sm-Eu phosphor. Moreover, the CCT uniformity and luminous efficiency of the w-LED were stimulated with the growth of BGBC:Sm-Eu amount. However, the CRI and CQS declines as BGBC:Sm-Eu amount became higher. Though the CQS and CRI declines, the color rendering can be promoted with this red phosphor as the CQS values are higher than the CRI ones. Therefore, the BGBC:Sm-Eu can be utilized for enhancing the luminosity and color fidelity and uniformity of the w-LED. Furthermore, when aiming to achieve greater chromatic rendition for the device, the green and blue phosphors can be applied to combine with the BGBC:Sm-Eu phosphor to obtain the full-spectrum white light. Besdies, as the BGBC:Sm-Eu phosphor has great absorption ability in the ultraviolet (UV) region, the UV LED chip can be applied to present the high-power w-LED with greatly improve lumen ouput.

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AUTHOR CONTRIBUTIONS STATEMENT

Name of Author	C	M	So	Va	Fo	I	R	D	O	E	Vi	Su	P	Fu
Luu Hong Quan		✓	✓	✓	✓	✓			✓	✓				
My Hanh Nguyen Thi	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	

C : **C**onceptualization
M : **M**ethodology
So : **So**ftware
Va : **Va**lidation
Fo : **Fo**rmal analysis
I : **I**nvestigation
R : **R**esources
D : **D**ata Curation
O : Writing - **O**riginal Draft
E : Writing - Review & **E**diting
Vi : **Vi**sualization
Su : **Su**pervision
P : **P**roject administration
Fu : **Fu**nding acquisition

CONFLICT OF INTEREST STATEMENT

Authors state no conflict of interest.

DATA AVAILABILITY

The data that support the findings of this study are available in Materials Science Poland at https:// doi/10.2478/msp-2022-0008, reference number [2]; https://doi/10.2478/msp-2022-0010, reference number [3]; https:// doi/10.2478/msp-2022-0050, reference number [4].

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