Strong green-emission phosphor $Ca_2SiO_4:Eu^{2+}$ to achieve the color constancy and high luminous efficiency for white LEDs

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ABSTRACT

The study uses the green phosphor of $Ca_2SiO_4:Eu^{2+}$ to achieve the color constancy for the dual-film remote phosphor white-LED model at low as well as big CCTs. The utilized green phosphor $Ca_2SiO_4:Eu^{2+}$ was prepared using the water-soluble-silicon liquid phase precursor method with the doped Eu^{2+} ion concentration of 3 mol%. The $Ca_2SiO_4:Eu^{2+}$ phosphor emits strong green light with emission intensity focused at 502 nm wavelength, and a wide stimulation band of colors of 225 nm – 450 nm. After applying the $Ca_2SiO_4:Eu^{2+}$ green phosphor and modifying its concentration, the modified color and luminous performances can be observed. The better color uniformity and higher luminescence efficiency can be obtained by increasing the percentage of $Ca_2SiO_4:Eu^{2+}$ in the phosphor configuration. Meanwhile, the color rendering metrics tend to reduce slightly when the concentration of $Ca_2SiO_4:Eu^{2+\ is}$ over 10% wt.

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1. INTRODUCTION

The light-emitting diode (LED) has been identified as a promising lighting solution for solid-state lighting applications. The method that has been commonly utilized to produce a white LED package is conformal phosphor coating one [1]-[3]. Though the advantages of this method are simplicity and cost-saving, its lighting efficiency was not satisfied the recent requirement in the lighting market. In addition to that, the phosphor material in the conformal-coated LED devices showed quicker decay due to direct interaction with the heat source, or the LED chip. Thus, the remote phosphor layout was developed to reduce the thermal effects on the phosphor materials, augment the reliability and lifetime of the phosphors as well as the LED lights. Though the LED's emission efficiency has been boosted using the remote phosphor configuration, the color fidelity of this structure would be a persisting downside. This flaw has attracted considerable interest from researchers. Here, for the LED in solid-state lighting solution, besides choosing the suitable packaging configuration, the kind of phosphor also has an impact on the color and luminosity constancy and efficiency of an LED [4], [5]. Thus, a suitable phosphor material for mass production of white light emitting diode (WLED) requires several features, including emitting high lumen intensity when being stimulated by close-UV or blue-color illumination sources and exhibiting high thermal consistency [6], [7]. Several silicate phosphors have become a subject of research owing to the feature of high stability in chemical and thermal performances, as well as providing low production cost. Thus, the rare-earth ions have been widely doped into the silicate host lattice to amplify the performance of these phosphors [8], [9]. 2% concentration of Mn^{2+} ion was doped with Ca_2SiO_4 phosphor and resulted in broad emission bands from 450 to 600 nm, with two emission peaks at 475 nm and 550 nm [10],

[11]. The Ce^{3+} and Eu^{2+} ions have become the focus of many pieces of research. It is reported that the phosphor doped with these two ions have their emission and excitation bands decided by the occupancy of the active ions with the cation sites [12], [13]. The new yellow phosphor was introduced by doping Ce^{3+} ion to the Ca_2SiO_4 composite, exhibiting a radiation range that focused at 565 nm. The $Ca_2SiO_4:Eu^{2+}$ was also reported to possess a broad yellow emission with a light red-shift emission peak when increasing the Eu^{2+} dopant concentration [14], [15].

The rare-earth doped silicate phosphor was usually synthesized using a high-temperature solid-state reaction method. However, this method demonstrated unequal dispensation of the ion inside the phosphor host lattice, the inhomogeneous shape of phosphor particle, leading to the inefficient luminous output of the material [16], [17]. The liquid phase precursor (LPP) approach was developed to address these problems effectively. Yet, the selection of suitable silicon agents that can be homogeneously mixed with water and allow reaction with other chemicals to occur easily is an important note when using the LPP method. The water-soluble silicon compound (WSS) was prepared and presented outstanding characteristics. According to the report, this WSS compound can be mixed with pure water regardless of the ratio and stored for a couple of weeks under an ambient atmosphere [18].

In this study, the Ca_2SiO_4 doped with Eu^{2+} at the concentration of 3 mol% via the LPP-WSS method is applied to produce a remote phosphor WLED structure with two layers of phosphor. The dual-layer had superior lumen performance to the single-layer one [19]. The $Ca_2SiO_4:Eu^{2+}$ phosphor is meant to compensate the elements of the green illumination for boosting the luminous flux and color homogeneity. Also, this green phosphor's red shift at 502-nm peak emission can help accomplish the chromatic constancy for the white LED at high color temperatures, such as 8000 K in this research.

2. EXPERIMENTAL DETAILS

2.1. X-ray diffraction analysis for $Ca_2SiO4:Eu^{2+}$ phosphor

The liquid phase precursor method utilizing silicon substance solvable in water (LPP-WSS) can result in a homogenous and single crystalline structure with the fine morphology for the attained $Ca_2SiO4:Eu^{2+}$ phosphor. Moreover, this uniformity in the mono-phase crystalline distribution contributed significantly to strengthening the luminous intensity of $Ca_2SiO4:Eu^{2+}$ green phosphor. To verify this, a comparison between the X-ray diffraction data obtained from the traditional solid-state reaction and the LPP-WSS methods is demonstrated. XRD is taken at two temperatures of 1000^0C and 1200^0C. XRD patterns showed that green phosphor $Ca_2SiO4:Eu^{2+}$ prepared using LPP-WSS had a more uniform structure with the mono-β phase, even the temperature increased to 1200^0C. In terms of the solid-state method, the green phosphor exhibited low crystallinity. When the temperature was lifted to 1200^0C, the β phase of the crystalline structure transformed into the γ phase, owing to the inhomogeneous distribution of doped ion Eu^{2+} in the pure $Ca_2SiO4:Eu^{2+}$ crystal lattice. In particular, the β phase and γ phase of the pure $Ca_2SiO4:Eu^{2+}$ keep their stability at room temperature. When the temperature rose, the synthesis of the γ phase was more likely to occur than that of the β phase because of the thermodynamically steadier crystal structure [20], [21]. In terms of the $Ca_2SiO_4:Eu^{2+}$ phosphor, the crystal stability only existed in the β phase, indicating that the Eu^{2+} ion was the stabilizer for the β phase at high temperature [22]. This means that the LPP-WSS method showed a Eu^{2+} dispensation that more evenly in the host lattice than the solid-state reaction one.

2.2. Impact of Eu^{2+} concentration on the luminescence of the $Ca_2SiO_4:Eu^{2+}$ green phosphor

The preparation of Eu^{2+}-doped Ca_2SiO_4 was carried out at 1100^0C with the approach of LPP-WSS. The Eu^{2+} ion concentration was modified from 0.1 mol% to 5 mol%. The luminous intensity of the Ca_2SiO_4 with three mol% was the highest and gradually declined when the Eu^{2+} concentration continuously climbed to 5 mol%. This result may be proved by connecting the presence of active ions strongly interacting with decreased distance and the increase in non-radiative energy transition among the active ions or among the active ions and the host lattice [23]. Additionally, at three mol% Eu^{2+}, the more significant increase of Gaussian curves that peak at 310 nm and 400 nm were observed, compared to that at 340 nm and 360 nm, and the red shift in the highest excitation and emission bands of the phosphor slightly appeared.

To analyze the effects of $Ca_2SiO4:Eu^{2+}$ on the lighting features of LED, two-layered remote phosphor configuration utilizing green-emitting $Ca_2SiO4:Eu^{2+}$ and yellow-emitting $YAG:Ce^{3+}$ phosphors are structured using 3D design software LightTools 9.0 and Monte Carlo Simulation. The green phosphor layer would be found above the yellow one, which was dispersed on the nine LED chips. Since the remote phosphor layout performs difficulty in yielding good color fidelity metrics for the white-LED at high color temperatures, the simulated models are carried out with low as well as big CCTs of 5700 K and 8000 K, respectively. Moreover, the proportion of $Ca_2SiO4:Eu^{2+}$ amount added into the remoted phosphor packages is modified from 5%wt. to

15%wt. to monitor the light efficiency in the WLED, including chromaticity and luminescence features of the generated white lights.

The physical design of the commercial WLED with conformal-coating yellow phosphor film is displayed in Figure 1. The green phosphor $Ca_2SiO4:Eu^{2+}$ is not present in this form. The LED model includes the main parts of a reflector, two layers of phosphor, and nine LED chips attached to the lead frame. The reflector dimension is commonly set at 9.85 mm, 8 mm, 2.07 mm in top length, bottom length, and height. The phosphor layers are thin and flat, measuring 0.08 mm in thickness. The blue LED chips are 1.14 mm^2 x 0.15 mm (square base x height) and exhibits 1.16 W illuminating flux with a maximum wavelength at 453 nm.

Figure 1. Picture of WLEDs form

3. RESULTS AND ANALYSIS

The opposite trend in the concentrations of both yellow $YAG:Ce^{3+}$ and green $Ca_2SiO4:Eu^{2+}$ phosphors can be observed in Figure 2(a). When the concentration of the green phosphor $Ca_2SiO4:Eu^{2+}$ tends to increase, that of the yellow phosphor probably decreases to keep the CCTs' consistency of the white-LED models, regardless of the pre-set color temperatures, see Figure 2(b). In addition, the reduction of yellow phosphor content has significant effects on the absorption, scattering, and transmission properties of lights in the remote phosphor packages, which also change the chromatic and luminous performance of the LEDs. Therefore, it is essential to determine how much $Ca_2SiO4:Eu^{2+}$ concentration can be increased to achieve the most satisfying WLED optical properties.

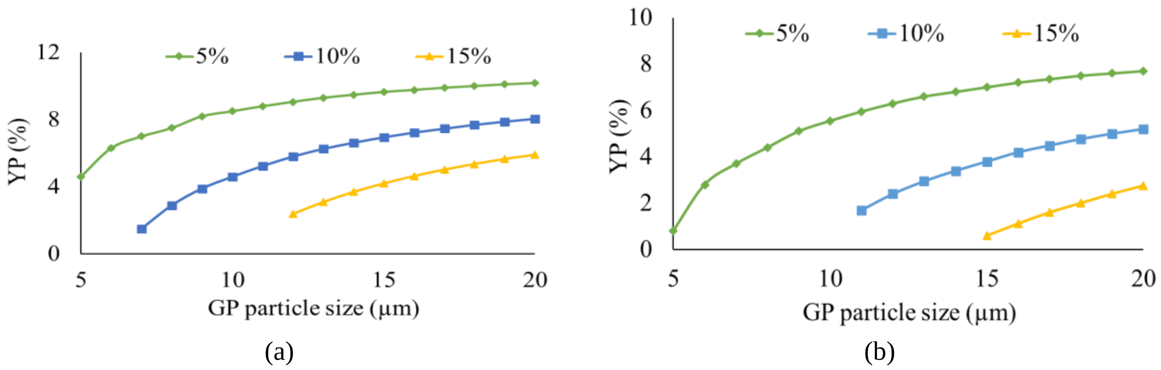
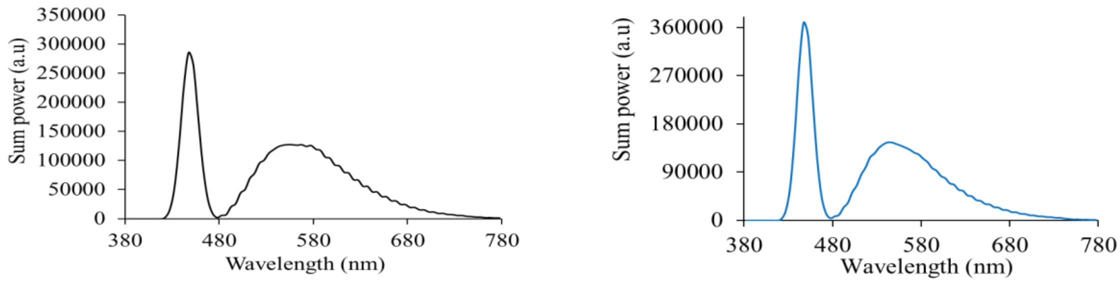


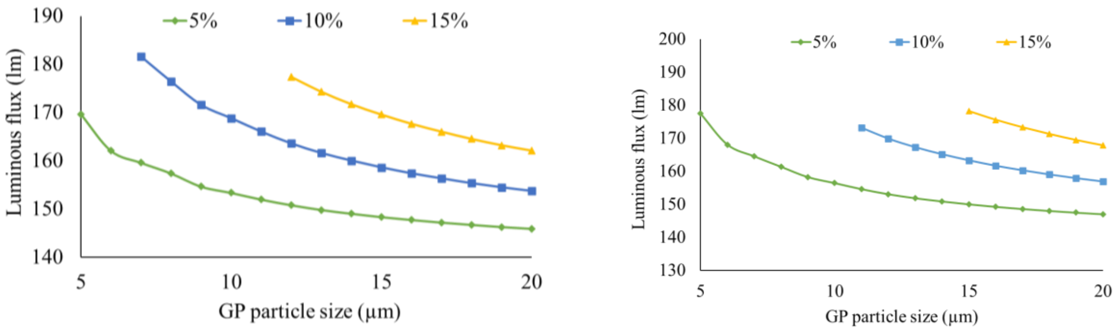
Figure 2. Changing the concentration of phosphor to preserve the average CCT, (a) 5700 K and (b) 8000K

The emission intensities of the dual-layer WLED models with $Ca_2SiO4:Eu^{2+}$ in the package are exhibited in Figure 3(a). The spectral energies of the WLED are enhanced significantly with the addition of this green phosphor. Moreover, the emission spectra at 8000 K are much stronger than that at 5700 K, which means the phosphor emits green light could enhance the luminescence of the WLED at not only low CCTs but also high CCTs, see Figure 3(b). Particularly, the LED's blue (420 – 480 nm) and green (500 – 640 nm) spectral regions are enhanced, indicating that the transmittance of blue lights is promoted and the green light component to achieve better color balance increase. This also implies that the light scattering in the phosphor layers is boosted, leading to higher color fidelity. In comparison with the WLED having 5700 K CCT, the LED phosphor package with 8000 K has improved the red emission intensity since the total spectral energy is enhanced with the rise in $Ca_2SiO4:Eu^{2+}$ concentration, resulting in the improvement of the chromatic rendering index. Thus, it is possible to conclude that $Ca_2SiO4:Eu^{2+}$ could heighten both color balance and rendering ability and luminous efficiency of the dual-layer white LED, especially at high CCTs (from 7000 K). The luminescence efficiency of the LED model with green phosphor layer $Ca_2SiO4:Eu^{2+}$ is demonstrated in Figure 4(a), in which the concentration of the phosphor emits green light is modified from 5% to 15%wt. As the concentration of $Ca_2SiO4:Eu^{2+}$ becomes higher, the increase in luminous flux is observed, regardless of the CCTs and particle sizes, see Figure 4(b). This result has confirmed the significant contribution of green phosphor $Ca_2SiO4:Eu^{2+}$ in enhancing the optical extrication performance of the double-layer remote phosphor design. The hue homogeneity is usually evaluated using the color deviation parameter. The lower the chromatic deviation is, the higher the color constancy becomes. Figure 5(a) indicates that the increase of $Ca_2SiO4:Eu^{2+}$ greatly benefits the color uniformity since it gets the color variance significantly declined, especially at the CCT of 8000 K. This decrease in chromatic deviation when $Ca_2SiO4:Eu^{2+}$ is added in the dual-layer structure results from the compensation of the green light components, leading to the higher color balance of the white light, see Figure 5(b). Such event is indicated via the altered absorption properties of the phosphor package of LEDs when there is $Ca_2SiO4:Eu^{2+}$ phosphor in the dual-layer structure. Specifically, when the $Ca_2SiO4:Eu^{2+}$ phosphor is in the LED, the blue-color illuminations from LED chips and yellow-color illuminations from the $YAG:Ce^{3+}$ layer are absorbed by this phosphor and subsequently are converted into green lights. As a result, the color homogeneity property is heightened. Besides the enhancement of luminous and chromatic performance attained from utilizing $Ca_2SiO4:Eu^{2+}$, the fabrication cost of this green phosphor is relatively low. Thus it can be a suitable phosphor material for WLED bulk production.



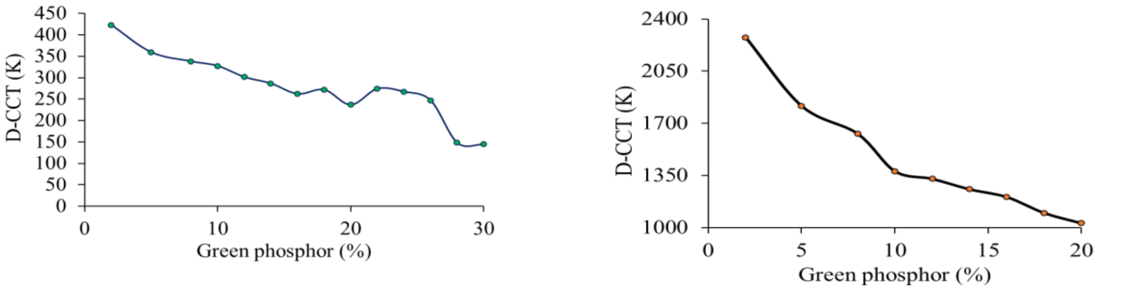
(a) (b)

Figure 3. Radiation spectra as a function of $Ca_2SiO4:Eu^{2+}$ concentration, (a) 5700 K and (b) 8000K



(a) (b)

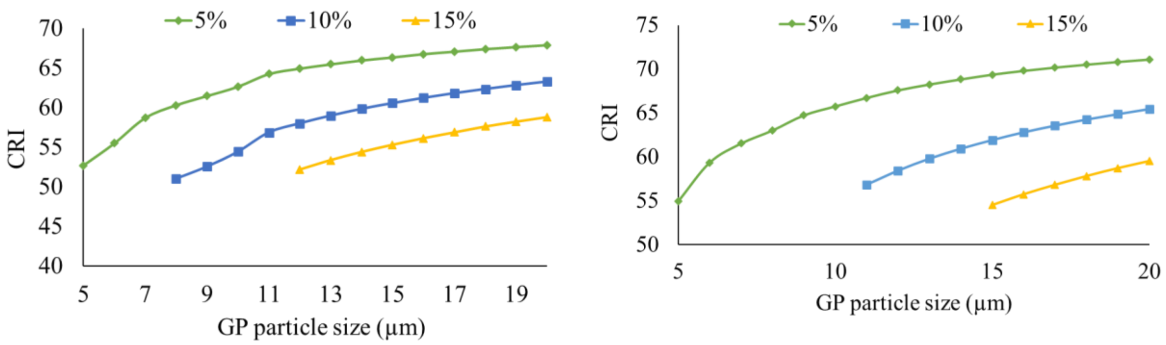
Figure 4. Luminous fluxes as a function of $Ca_2SiO4:Eu^{2+}$ concentration, (a) 5700 K and (b) 8000K



(a) (b)

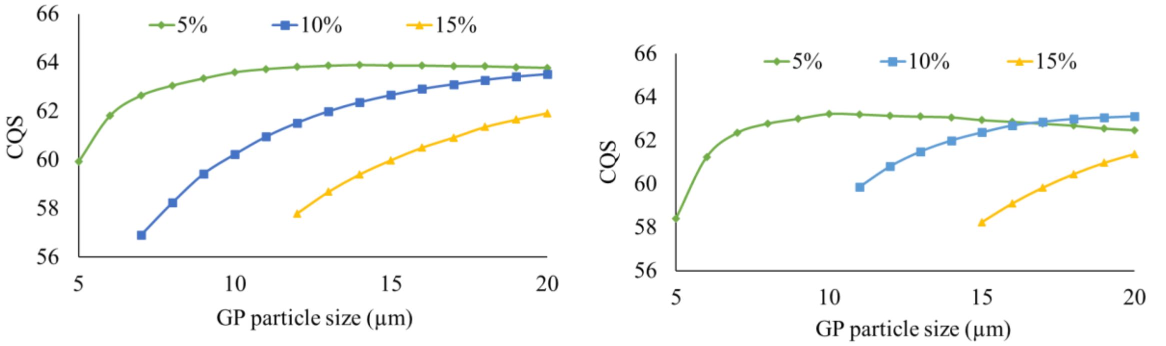
Figure 5. The hue deviation as a function of $Ca_2SiO4:Eu^{2+}$ concentration, (a) 5700 K and (b) 8000K

Aside from color constancy, CRI is another important metric in the color measurements of WLED lights. CRI determines an item's hue when affected by a tested illumination in relation to its look when affected by a reference one. Yet, the increase in $Ca_2SiO4:Eu^{2+}$ concentration leads to the green-component redundancy, causing the inefficient color distribution, see Figure 6(a). Therefore, it is short of red color components that are necessary for high CRI. According to Figure 6(b), the increase in $Ca_2SiO4:Eu^{2+}$ concentration reduces the CRI slightly. However, it is not enough to evaluate an illumination's color quality using a single metric of CRI because the CRI merely tests eight color samples, and there are more related aspects that need a color evaluation of a light source. Thus, CRI cannot evaluate the entire gamut of a visual LED light. CQS proposed by NIST alternatively is a more sufficient index for measuring the chromaticity of the LED. CQS measures three main aspects of the light source, including the preference of human observers, the chromatic coordinates, and even the CRI [24], [25]. Therefore, achieving high CQS is more complicated and complex than obtaining high CRI. Adding a layer of green phosphor $Ca_2SiO4:Eu^{2+}$ is beneficial to the development of CQS of white LED lights. When the phosphor concentration that emits green lighting in the film increases to 10%, the CQS slightly goes upward. However, when there is more than 10%wt. $Ca_2SiO4:Eu^{2+}$, the CQS starts to decrease. This might result from the reduction of CRI due to the color imbalance and the decrease of red spectral emission caused by the green-light redundancy. Hence, to get the best color performance along with high luminescence efficiency, the appropriate concentration of $Ca_2SiO4:Eu^{2+}$ should be determined. Here, 10%wt of $Ca_2SiO4:Eu^{2+}$ might be suitable for applying in white LEDs with double-layer remote phosphor packages. Figures 7(a) and (b) shows the color quality scale connected with $Ca_2SiO_4:Eu^{2+}$ concentration.



(a) (b)

Figure 6. CRI connected with $Ca_2SiO_4:Eu^{2+}$ concentration, (a) 5700 K and (b) 8000K



(a) (b)

Figure 7. The color quality scale connected with $Ca_2SiO_4:Eu^{2+}$ concentration, (a) 5700 K and (b) 8000K

4. CONCLUSION

According to this article, the green phosphor $Ca_2SiO_4:Eu^{2+}$ is used to enhance the light efficiency in the WLED fabricated with distant phosphor packets, including two different phosphor films. The Eu^{2+} doped into the phosphor composition of Ca_2SiO_4 resulted in the deep green emission spectra that peaks at 502 nm and exhibited high luminescence intensity and uniformity of phase distribution. When applied to the dual-layer structure, the green-emitting $Ca_2SiO_4:Eu^{2+}$ has considerable effects on the absorption, extraction efficiency, conversion, and transmission of lights in the LED models. Regardless of the color temperature, the green phosphor proves to help reduce the color deviation to better the color constancy. The luminous intensity also benefits from the increased $Ca_2SiO_4:Eu^{2+}$ phosphor concentration. Nevertheless, the rise in $Ca_2SiO_4:Eu^{2+}$ percentage leads to the excessive green-light amount, causing tremendous color loss and leading to the reduction in color rendering metrics, CRI and CQS. It is observed that, more than 10% wt. of $Ca_2SiO_4:Eu^{2+}$ can decrease the CRI and CQS; therefore, keeping the concentration of this green phosphor below 10% wt. could be suitable for fabricating WLED lights with constant CRI and CQS and strengthened luminescence.

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