

# Efficient bright green phosphor for white light-emitting diode using in backlight application

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## ABSTRACT

The (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> phosphor with narrow emission band and green emission color peaking at 510 nm is demonstrated in the study. This phosphor is prepared using a solid-state reaction at high temperatures. Based on characterizing data, the increasing concentration of Ca<sup>2+</sup> induces the shrinkage of the unit cell and the narrower spacing, leading to the increase in the photoluminescence of the phosphor. This also cause a red shift in the emitting range from 504 nm to 510 nm. The phosphor shows good absorption peaking at 415 nm, matching the excitation wavelength of the used near-ultraviolet (UV) light emitting diode (LED) chip. When being applied in a white light-emitting diodes (WLED) with 4,000 K, the phosphor could increase the luminous flux as its doping concentration increase. Besides, the WLED doping (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> shows emission regions in blue, green, and red wavelength bands. The data from the study indicates that the phosphor is potential for developing the white-LED for backlight purposes.

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## 1. INTRODUCTION

The materials of phosphor down-conversion for diodes emit white illumination white light-emitting diodes (WLEDs) have been significantly critical to the light emitting diode (LED) industry since the WLED has been an increasingly indispensable part of modern lighting applications. In comparison with other conventional light sources like halogen lamps or fluorescent lamps, the WLED possesses various outstanding features including compactness, power-saving, environmental friendliness, and strong luminosity [1]–[3]. The WLED utilizing phosphor down-transformation to create white illumination requires more studies and development on the phosphor to serve several lighting purposes. For the WLED used in general lighting, the shortage of red emission energy has been the obstacle for it the reach a better rendering performance [4]. On the other hand, if the WLED is used for backlight display, the phosphor must encourage the luminescence of the WLED as well as the emission component in blue, red, and green for the ability to reserve the saturation and brightness of generated white light. So, it is important to get the full spectrum of red, blue, and green to make the phosphor applicable for using in either general lighting or backlight applications [5]–[7].

The backlighting fields, in addition, requires adequate green emission to retain the vividness of the light effectively. Therefore, the utilization of phosphor combination of YAG: Ce<sup>3+</sup> yellow phosphor and (Sr, Ca) <sub>2</sub>Si<sub>5</sub>N<sub>8</sub>: Eu<sup>2+</sup> red phosphor for a blue-pumping LED chip [8]–[10] were not successfully meet this requirement. This combination could not provide efficient green emission spectra. Thus, the phosphor that is suitable for backlighting should have efficient green emission strength. Consequently, several green-phosphor

materials were proposed for WLEDs, such as  $\text{Sr}_2\text{SiO}_4$  [11],  $\beta$ -sialon:  $\text{Eu}^{2+}$  [12], and  $\text{SrGa}_2\text{S}_4$ :  $\text{Eu}^{2+}$  [13], to combine with the mentioned red phosphor (Sr, Ca)  $_2\text{Si}_5\text{N}_8$ :  $\text{Eu}^{2+}$  for the chromatic-gamut enhancement. Nevertheless, the values of the national television system committee (NTSC) standard of these compounds were around 75-83 %, meaning that the improvement of the chromatic gamut is insignificant.

It is possible to obtain a significant enhancement in the luminescence of the green phosphor by accessing the cation substitution, from which the red shift or blue shift can be obtained. The redshift from the shorter to the longer wavelength of the phosphor could be achieved via the site replacement of Ca to Ba in the host of  $\text{Ba}_2\text{MgSi}_2\text{O}_7$ :  $\text{Eu}^{2+}$ , owing to the strong crystal field splitting [14]. Hence, based on this foundation, our study will develop a narrow-emission-band green phosphor providing efficient green emission for keeping the lighting vivid while promoting the luminescence of the WLED.  $\text{Ba}_{1-x}\text{Ca}_x\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  (with x ranging from 0 to 0.12) will be prepared through elevated-heat solid-state reaction. The phosphor characterization will then focus on the crystalline structure and the luminescence of the phosphor influenced by the cation replacement between  $\text{Ca}^{2+}$  and  $\text{Ba}^{2+}$ . Accordingly, the green phosphor will be combined with the red phosphor to simulate the WLED model [15], [16]. The concentration of (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  green phosphor will be varied for the investigation of WLED's properties. The phosphor exhibits good excitation and strong, bright green emission with a redshift, indicating that it is promising for backlights.

## 2. METHOD

### 2.1. Phosphor preparation

The composition of green phosphor (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  includes key chemical ingredients of  $\text{BaCO}_3$ ,  $\text{BaF}_2$ ,  $\text{Sc}_2\text{O}_3$ ,  $\text{CaCO}_3$ ,  $\text{Bi}_2\text{O}_3$ , and  $\text{K}_2\text{CO}_3$ . All the components have a purity percentage of 99.99 and are purchased from Qi *et al.* [17]. The method used to synthesize the required phosphor is the solid-state reaction at high temperatures. The synthesizing process starts with mixing the ingredients in a stoichiometric ratio and continues with two calcination phases before cooling down and powdering the treated product to get the final one. Particularly, the first calcination took place in a box furnace for about 8 hours at 1,200 degrees Celsius. The second calcination is carried out at 1,100 degrees Celsius for 4 hours in a gas mixture of 10%  $\text{H}_2$ /90%  $\text{N}_2$ . The aim of the second calcination is to obtain the trivalent state for Bi ions. The summary of the composition and synthesis of the phosphor (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  is presented in Table 1 and Table 2, respectively.

Table 1. (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  composition

Components	Purity percentage
$\text{BaCO}_3$	99.99
$\text{BaF}_2$	99.99
$\text{Sc}_2\text{O}_3$	99.99
$\text{CaCO}_3$	99.99
$\text{Bi}_2\text{O}_3$	99.99
$\text{K}_2\text{CO}_3$	99.99

Table 2. (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  synthesizing process

Stages (in order)	Conditions
Mixing	Stoichiometrically
1 <sup>st</sup> calcination	1,200 °C, 8 h
2 <sup>nd</sup> calcination	1,100 °C, 4 h
Cooling down	
Finely powdering	

### 2.2. Phosphor characterization and LED simulation approaches

The characterizing aspects of (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  phosphor powder includes its crystalline and microstructures, absorption, photoluminescence, and stimulation spectra, as well as internal quantum and temperature-dependent luminescence efficiencies (IQE and LE) [18]. The instruments used for analyzing and measuring these characteristics are shown in Table 3.

To assess the performance of WLED models using (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  phosphor, we must simulate and build it using light tools 3-D software. By modeling a WLED package closely similar to the actual model (~99.6%), it is possible to minimize the influences of waveforms, wavelengths, and operating temperature. on the optical properties of the required WLED to get more accurate measurements. The phosphor layer used in addition to yellow YAG: Ce is comprised of the green-phosphor (Ba, Ca)  $\text{ScO}_2\text{F}$ :  $\text{Bi}^{3+}$ ,  $\text{K}^+$  and red-phosphor  $\text{CaAlSiN}_3$ : $\text{Eu}^{2+}$ . The WLED uses near UV chips having 415 nm excitation wavelength. We also

utilize the HSP6000 optoelectronics LED test system (Hangzhou, China) to analyze the optical performance, including color rendition, color uniformity, and LE, of the simulated WLED.

Table 3. Instruments used for characterizing (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> phosphor powder

Phosphor characteristics	Instruments	Notes
Crystalline structure	- TD-3500 X-ray diffractometer (Dandong Tongda, China) - The GSAS (General structure analysis system) software for the refinement of crystal structures.	Operated with 1.5406 Å Cu-K $\alpha$ , 30 kV cathode voltage, and 20 mA driving current.
Microstructures	- Helios NanoLab 600i SEM/FIB DualBeam (FEI, USA) - Oxford Instruments' energy-dispersive spectroscopy (ESD).	
Absorption spectra	Cary 5,000 UV-Vis-NIR spectrophotometer (Varian Inc., USA).	Measured in the range of spectra from 200 nm-800 nm.
Photoluminescence and excitation spectra	Hitachi F-7000 fluorescent spectrophotometer (Hitachi, Japan).	Operated at room temperature with a Xe lamp (1,500 W).
IQE	Fluorescence spectrophotometer Hitachi F-7000	Using a Quanta- $\phi$ integrating sphere and a polytetrafluoroethylene specimen cup.
LE	Edinburgh instruments' FLS980 photoluminescence spectrometer (Edinburgh Instruments, UK)	Using excitation source of a Xe lamp (450 W).

### 3. RESULTS AND DISCUSSION

Depending on specific application lighting aspects, certain properties are focused. For example, if the WLEDs are produced to serve general illuminating purposes, the color rendition and color deviation are crucial factors. On the other hand, when the WLEDs are planned to be used as backlights for display applications like liquid crystal display (LCD), the brightness and saturation of the light are focused. So, the investigation of the phosphor (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> characteristics and WLED's lighting efficiencies are critical to its application and further development.

The green-phosphor (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> concentration varying from 5-10 wt%. When utilized in the phosphor layer, so that we can know the differences and how the phosphor influences the lighting outcome of the WLED model. Accordingly, when the concentration of (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> changes, the concentration of the YAG: Ce also changes in the inverse proportion to serve the color-temperature stability of the WLED, which is shown in Figure 1. Consequently, optic features like scattering, absorption, and emission properties of the entire package could get significant influences. The emission strength of the WLED at 4,000 K, particularly, shows differences between the 5 wt% and 10 wt% concentrations of (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> phosphor, as can be observed in Figure 2. Overall, the emission of 4,000-K WLED is significant in the wavelength range from 550 nm to 600 nm, which means it covers the green and bright red regions. In other words, the green and red emissions are included in the emitted white light. Especially, in the case of 5 wt% (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup>, the emission of blue is observable. These three emission parts are important to the WLED for the backlighting purpose because they could help retain the saturation of the light color. At this point, (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> could be suitable for a backlighting WLED.

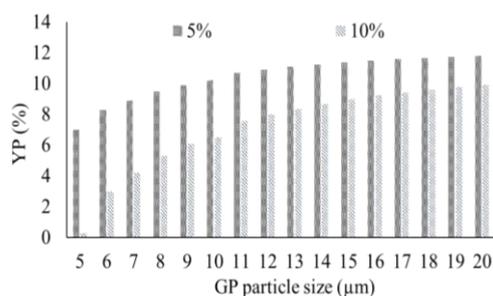


Figure 1. Changing the concentration of phosphor to preserve the average CCT

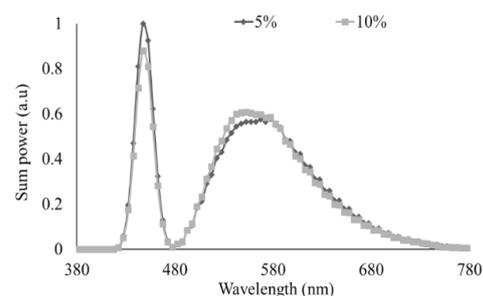


Figure 2. Spectral emission of a WLED at 4,000 K with different (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup> concentrations

For further demonstration of WLED emission power when applying the green-phosphor (Ba, Ca) ScO<sub>2</sub>F: Bi<sup>3+</sup>, K<sup>+</sup>, the analysis of the phosphor's photoluminescence and crystal field should be presented. When the concentration of Ca<sup>2+</sup> increases, the Ba<sup>2+</sup> is replaced by the smaller Ca<sup>2+</sup>, according to Bragg law (1) [19]:

$$2d \sin \theta = n\lambda \quad (1)$$

where  $d$ ,  $\lambda$ , and  $\theta$  indicate the interplanar spacing, X-ray wavelength, and diffraction angle. The increase in  $\text{Ca}^{2+}$  concentration decreases the cell parameters and volume; in other words, the unit cell shrinks and the spacing decreases. As a result, the bond lengthiness  $d(\text{Ba/Ca/Bi/K-O})$  become shorter, leading to the  $[\text{BiO}_{12}]$  polyhedral distortion and changes in  $\text{Bi}^{3+}$  local crystal field strength. The polyhedral distortion can be calculated using (2) [20];

$$D = 1/n \sum_{i=1}^n (|d_i - d_{av}|) / d_{av} \quad (2)$$

in which  $D$  represents the index of polyhedral distortion,  $d_i$  and  $d_{av}$  show the space between the central and  $i$ -th coordinating atoms and the mean lengthiness of all the bonds, respectively. Subsequently, (3) is utilized to demonstrate the replacement relation of the  $\text{Ca}^{2+}$  in the phosphor lattice [21];

$$D_r = 100 \times [R_m(\text{CN}) - R_d(\text{CN})] / [R_m(\text{CN})] \quad (3)$$

in which  $D_r$  is the diameter percentage dissimilarity,  $R_m$  is the radius of the host cation, and  $R_d$  is the radius of the integrated ion. Additionally, CN is the number of coordination. As both  $\text{Ca}^{2+}$  and  $\text{Ba}^{2+}$  have the similar ionic radius ( $\text{Ca}^{2+}$  and  $\text{Ba}^{2+}$  have the same CN of 12, and their radii are 1.34 Å and 1.61 Å, respectively), the  $D_r$  can be computed to be 16.6%, lower than the limiting point, leading to the substitution of  $\text{Ca}^{2+}$  to  $\text{Ba}^{2+}$  in the  $\text{BaScO}_2\text{F}$  host.

When the phosphor is monitored under 506 nm wavelength, the peak absorption spectra of the phosphor is recorded at 360 nm and 415 nm, which could be demonstrated by the energy transition of the  $\text{Bi}^{3+}$ :  $^1\text{S}_0 \rightarrow ^1\text{P}_1$  and  $^1\text{S}_0 \rightarrow ^3\text{P}_1$  (ground state  $\rightarrow$  excited state). Then, with the increasing concentration of the  $\text{Ca}^{2+}$ , the photoluminescence strength of the phosphor increases and peaks at 510 nm before gradually decreasing. This can be attributed to the substitution of smaller  $\text{Ca}^{2+}$  to larger  $\text{Ba}^{2+}$  in the host. The unit cell's shrinkage improves the luminosity of the phosphor as it encourages the process of non-radiative relaxation and the structure's rigidity of  $\text{BaScO}_2\text{F}$  host. However, as there are more  $\text{Ba}^{2+}$  replaced by  $\text{Ca}^{2+}$ , the luminosity is reduced because of the defect formation energy. Moreover, such an increase in the volume of substituted  $\text{Ba}^{2+}$  leads to the occurrence of red shift as observed from 504 nm to 510 nm. The crystal field splitting caused by shrinking unit cells and decreasing cell volume can be utilized for demonstrating this occurrence. The (4) is applied to investigate crystal field splitting energy ( $D_q$ ) [22];

$$D_q = (Ze^2r^4) / (6R^5) \quad (4)$$

in which  $Z$ ,  $e$ ,  $r$ , and  $R$  indicate the following parameters, respectively: anionic and electron charges, radius of the d-wave function, and the luminescent cation-ligand bond lengthiness. It can be inferred from (4) that the energy of the field splitting depends significantly on the cation-ligand bond length. Subsequently, based on double exponential function, the corresponding decay times ( $\tau_1$  and  $\tau_2$ ) of phosphor luminescence are measured using (5) [23];

$$I(T) = I_0 + A_1 \exp(-T/\tau_1) + A_2 \exp(-T/\tau_2) \quad (5)$$

in which  $T$  indicates the time, and  $I(T)$  show the illumination strength of the phosphor at the time  $T$ . The fitting constants are represented by  $A_1$  and  $A_2$ .

Shortly, the luminosity of the phosphor improves when the concentration of  $\text{Ca}^{2+}$  increases from 0 to 0.06 and decreases after the concentration of  $\text{Ca}^{2+}$  exceeds 0.06. From this, when fabricating the phosphor for WLED simulation, the doping concentration of the  $\text{Ca}^{2+}$  should be chosen at 0.06 for the maximal luminescence strength. Subsequently, the influence of this green-phosphor ( $\text{Ba, Ca ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  doping concentration should be investigated on the overall luminous efficiency of the WLED model. Figure 3 shows the luminosity of the WLED recorded at 4,000 K. Obviously, the increasing concentration of the phosphor increase the luminous flux. As the improve luminosity can be obtained by increasing the green emission part of the white light, the phosphor ( $\text{Ba, Ca ScO}_2\text{F: Bi}^{3+}, \text{K}^+$ ) can serve this. Moreover, the phosphor has an adequate excitation efficiency (two absorption peaks at 360 nm and 415 nm) in the near UV-LED-chip wavelength. This attributes to the conversion efficiency of the whole WLED's phosphor package.

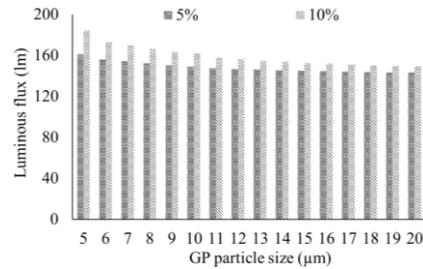


Figure 3. Luminescence of the WLED with (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  at 4,000 K

As can be inferred from the above discussion, (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  is adequate for WLED to serve backlight purpose since it provides the three important emission parts of blue, red, and green in the spectrum band to retain the light saturation and brightness. Besides, the phosphor could improve the luminosity of the entire WLED package when increasing its doping concentration. If the manufacturer focuses on achieving high-luminescence WLEDs for backlighting application, the color deviation and color rendition could be not too important. However, if they want the WLEDs for general lighting applications, the effects of (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  phosphor concentration on the color rendering intent, color divergence, and color reproduction of the WLED should be considered. Moreover, the eye sensitiveness of the human users needs to be included in the assessment. It is reported that the human eyes prefer bright and saturated color and are sensitive to greenish (555 nm) light. Hence, for a comprehensive analysis of light color performance, the three criteria-hue rendering index (CRI), human eyes' response, and hue coordinates, should be included. Considering these factors, the hue standard scale (CQS) can be applied to access the desirable color evaluation [24], [25].

The color uniformity and CRI of the WLED doping (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  monitored at 4,000 K are shown in Figure 4 and Figure 5, in turn. The CCT does not deviate significantly from 4,000 K when using 10 wt% (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  concentration. The CCT is also lower with (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  doped at 10 wt%. This indicates that the increasing concentration of (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  is not only benefits the luminosity but also the color uniformity of the WLED. The CRI, in contrast, decreases when the concentration of (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  becomes higher. A small reduction in CRI values of the WLED can be recorded when increasing the (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  weight percentage from 5 to 10 wt%, as depicted in Figure 5. As there is the contrast in the two critical parameter CRI and color coordination, the performance of CQS might be affected. Figure 6 demonstrates the CQS results of the 4,000-K WLED. The CQS does decrease as the concentration of the phosphor increase. However, when the particle size of the phosphor increase, the CQS with 10% (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  concentration also gradually increases and eventually is equal to that with 5 wt% (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$ . This can be attributed to the diffusing impact of the phosphor. When the particle size of the phosphor is smaller, the space distribution tends to be larger, which could cause scattering loss and non-uniform color distribution. However, reduced scattering effect is advantageous to the luminosity since it can limit the loss of absorption for better conversion-meaning more green lights are generated, and increase the escaping light from the WLED package. As the particle size increase, the space distribution is narrower, inducing the scattering for better color coordination but higher absorption loss. In other words, the improved scattering promotes the color blending and distributing on the chroma scale, as well as decrease the generated green light by higher absorption loss. Hence, the increasing particle size of the (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  green phosphor could stimulate the color uniformity, color rendition, and the reduction in strong brightness for better CQS.

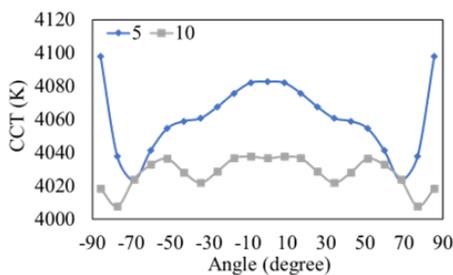


Figure 4. CCT of WLED with (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$

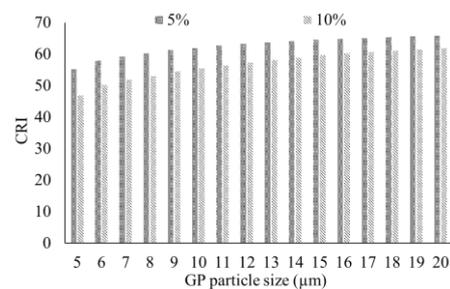


Figure 5. The hue rendering index of WLED of the WLED with (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$

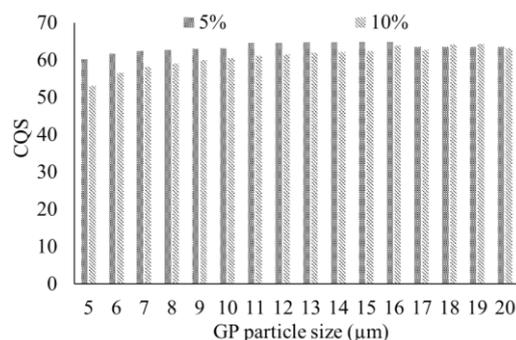


Figure 6. The hue quality scale of WLED with (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$

#### 4. CONCLUSION

In this study, the green-phosphor (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  is demonstrated to be suitable for WLED to serve backlighting purposes. The phosphor shows green emitting peak at 510 nm at excitation of 415 nm. The replacement of smaller  $\text{Ca}^{2+}$  to larger  $\text{Ba}^{2+}$  could be ascribed to the change in luminous strength of the phosphor when increasing the doping dosage of  $\text{Ca}^{2+}$ . When the phosphor is utilized for the WLED at 4,000 K, the WLED exhibits the emission parts in blue, green, and bright red wavelengths. The luminous strength of the entire package displays enhancement with increasing concentration of the phosphor. The color properties-CQS and CRI, on the other hand, are decreased with increasing concentration of the (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$ . However, by using larger particle size of the (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  phosphor, the CQS can increase owing to the limited scattering loss, yet the luminosity is reduced as a compensation. Hence, the phosphor (Ba, Ca)  $\text{ScO}_2\text{F: Bi}^{3+}, \text{K}^+$  could be also applied in general lighting after considering the suitable concentration and particle size.

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