

## Green $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ phosphor: an innovative solution in enhancing the color quality and luminous flux of WLEDs

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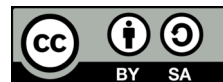
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### ABSTRACT

Light-emitting diodes devices that embedded with multiple chips (multi-chip white LEDs (MCW-LEDs)) are an advanced lighting solution with much potential for improvement in the lighting industry. However, to further the applications and quality of white light emitting diodes (WLEDs) greater achievements must be found, thus, this paper focus on improving the color uniformity and luminous flux with green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ . The results, which were measured through experiments conducted in WLEDs with average correlated color temperature from 6600–7700 K, show enhancements in color uniformity and luminous efficacy. In particular, the growing trend in the concentration of green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  results better color uniformity and luminous flux, although the color quality scale (CQS) suffers a small decline. Therefore, it is confirmed that the  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor is suitable in manufacturing WLEDs that focus on the color uniformity and luminous flux.

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## 1. INTRODUCTION

The light emitting diode (LED) devices using multi-chip white LEDs (MCW-LEDs) with good size, durability, life-time, lighting capacity and many other qualities are excellent answers for the on-going demand of the market. However, the conventional white light emitting diodes (WLEDs) that are made with the common method cannot adapt to the modern lighting requirements, therefore, scientists are searching for a new breakthrough in the production of WLED. The results are expressed through findings regarding the improvements of lumen output and color homogeneity [1]-[5]. Through application of results from previous researches, Anh and his colleagues find the compound consists of YAG: $\text{Ce}^{3+}$  phosphor and  $\text{SiO}_2$  particles yield more noticeable enhancements in lighting performances and promote the quality of WLED to the highest standard [6]. The phosphor compound, which is used to convert yellow light from blue radiation of the chips, is proceeded by integrating YAG: $\text{Ce}^{3+}$  phosphor with the gel made of silicone [7]-[13]. The light emitted will disperse on the phosphor compound of YAG: $\text{Ce}^{3+}$  phosphor and then pass through the phosphor particles to form white light. This scattering process allows the yellow phosphor YAG: $\text{Ce}^{3+}$  to transform the blue light and gradually weaken its light power while strengthening the yellow light emitted [14]-[16]. The result is the slightly blue color at the surface of the LED, however, the outcome light can be tuned to a more

yellow color if the position of the phosphor compound is placed parallel to the LED surface [17]-[19]. This finding implies that adjusting the light intensity distribution can effect the quality of the emitted light.

According to Won research team, the configuration containing chromatic phosphor materials such as  $(\text{Ba,Sr})_2\text{SiO}_4:\text{Eu}^{2+}$  and  $\text{CaAlSiN}_3:\text{Eu}^{2+}$  has strong impact on the luminous flux of high color quality WLEDs [20]. On the other, Oh and his team focus on obtaining the best lumen output and color rendering index (CRI) possible with a multi phosphor layers LED package of green, amber and red [21]. Another reseach emphasizes on the development of color rendering index (CRI) in LED is from the team of Zhang and his partners, the content of the research report proposed using multi-color phosphor to yield the desired amount of CRI [22]. The studies mentioned above concentrate on changing the structures of LED packages and phosphor properties to enhance the lighting performance. The results are all highly valued for their practicality as well as credibility in boosting the CRI and other optical properties, which is concerned by the majority of reseachers. However, it seems that the research range is insufficient as the color uniformity is neglected in those reseaches and only the single-chip LED are studied, thus, making these results become obsolete in WLED with high correlated color temperatures.

Therefore, to perfect the missing point, this paper reports the growth of color uniformity and luminous flux in multi-chips WLED with green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor components. With different parameters of the green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor, the color uniformity and lighting performance will enhance accordingly. The research include building the simualtion of the WLED used in the experiements, measuring the changes that green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor inflicts on the lighting properties of WLED with the mathematical equations and finally analyzing the results to come up with a set up that can achieve optimal efficiency. The summary of the results confirm that using the mixture of green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor and yellow YAG: $\text{Ce}^{3+}$  phosphor benefit the color quality and lumen output of WLED.

## 2. PREPARATION PROCESS AND SIMULATING WLED

### 2.1. Preparation of green $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ phosphor

The phosphor employed in the experiments to enhance the optical properties of WLED is green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ . This phosphor emits a pale yellow-green light color and the emission peak of 2.28eV. The chemical compositions of green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  is presented in Table 1. To create the best quality green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor, the manufacturing process must accur at the exact order as follow: First, mix all the ingredients by grinding or miling untill they mixed well. The ingredients compound is placed on the open quartz boats and fired at approximately 500 °C, the product after firing will be solidified, wait until it cool down and begin to powderize. After that, put the ingredients in a capped quartz tube filled with  $\text{N}_2$  and begin to fire, this time at 1000 °C for an hour, and repeat the powderizing step. Keep putting the product from the previous in capped quartz tube filled with  $\text{N}_2$  and fire one more time, this time at 1200 °C for another hour. The final product should be kept in sealed container until use to prevent contamination.

Table 1. The chemical composition of green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$

Ingredient	Mole %	By weight (g)
$\text{CaCO}_3$	198	198
$\text{La}_2\text{O}_3$	200 (of La)	326
$\text{PbO}$	2	4.6
$\text{H}_3\text{BO}_3$	105	65

### 2.2. MC-WLEDs simulation

Figure 1 (a) demonstrate the WLED physical model that are made for the experiments in this research. Figure 1 (b) lists in detail all the information relate to technical parameters of the WLED, and Figure 1 (c) shows the cross-section of the dual-remote phosphor WLED diagram. Using the Monte Carlo method and optical engineering software LightTools 8.1.0, we constructed the WLED simulation. The WLED have reflectors measuring 8x2.07x9.85 mm in the bottom, side, and top, respectively. The reflectors hold 9 chips that are embedded into the gaps on the reflectors, each chip is 0.15 mm in height, 1.14 mm<sup>2</sup> base area and achieves highest intensity of 1.16 W at 453 nm. Above the chips is a 0.08 mm thick phosphor layer containing phosphor particles with an average of 14.5 μm in diameter.

The simulation of the MCW-LED begins with mixing the phosphors with the flat silicone layers and then applied to the WLED structure and begin the manufacturing process. After the manufacturing process, proceed to evaluate WLEDs quality by comparing their performances when distinct color temperatures and different levels of phosphor concentration. In this research, the green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  and the average correlated color temperatures of 6600 K and 7700 K are chosen as specimens. The results from this

experiment should reveal the impacts of the green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  on the optical properties of WLED and serve as a adjustments The results from this experiment should reveal the impacts of the green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  on the optical properties of WLED and serve as a adjustments guideline for better lighting quality.

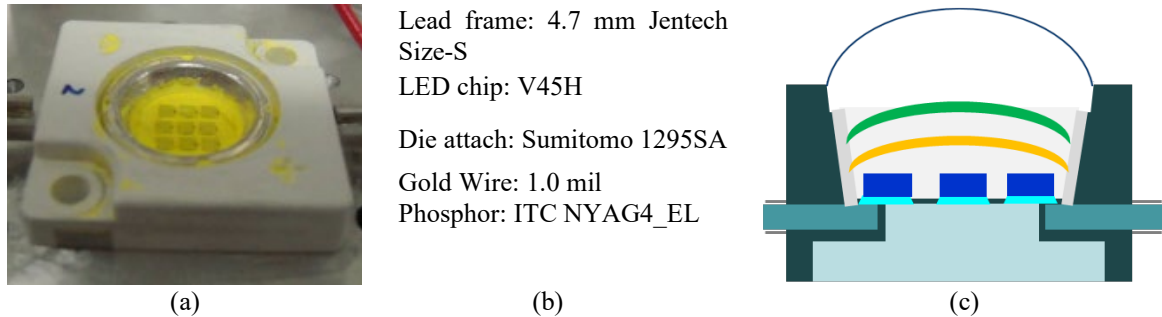


Figure 1. Illustration of phosphor-converted MCW-LEDs as doping  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ : (a) the actual MCW-LEDs, (b) its parameters, and (c) dual-remote phosphor geometry

### 3. COMPUTATION AND DISCUSSION

First, we analyze the emission spectra of MCW-LEDs with the presence of green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor with the results expressed in Figure 2. These results are measured from WLEDs that employed the phosphor compound of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ , YAG: $\text{Ce}^{3+}$  particles, mixed in the silicone glue. The parameters of phosphor particles must match the predetermined values mentioned on the previous part before the experiments. Once the measurements are correct and the refractive index of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  particles, YAG: $\text{Ce}^{3+}$  particles, and the silicone glue are exactly 1.85, 1.83, and 1.52 respectively, then the calculation of emission spectra in WLEDs can proceed. From Figure 2, which the emission spectra of the dual-remote phosphor compounding with 10%  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  at 6600 K and 7700 K ACCTs, the spectrum intensity increases in several regions. In particular, at both 6600 K and 7700 K, the emission spectra at 430–480 nm wavelength and 500–630 nm wavelength increases. This finding entails that the luminous flux of MCW-LEDs can be enhance by using  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor in the phosphor compound.

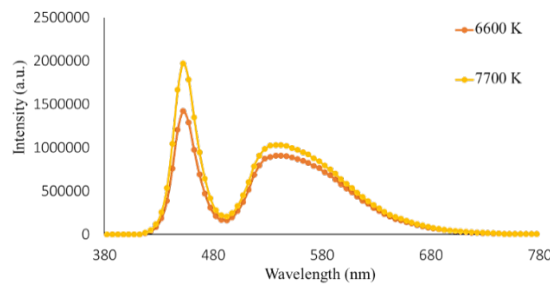


Figure 2. Emission spectra of MCW-LEDs at 6600 K and 7700 K CCTs WLED

Next, measuring other optical properties of the phosphor compound in MCW-LED would requires the value of scattering coefficient  $\mu_{sca}$ . Therefore, we calculated the scattering coefficient (SC)  $\mu_{sca}$ , the wavelength, and  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  particles size by applying Mie-theory in the (1) [23]-[25].

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

$$\bar{c}_{sca}(\lambda) = \frac{\int^{\lambda} c_{sca,D}(\lambda) f(D) dD}{\int^{\lambda} f(D) dD} \quad (2)$$

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

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

In these equation,  $f(D)$  is the function for size distribution, and  $c$  is the density of phosphor in an area ( $\text{g}/\text{cm}^3$ ).  $\bar{C}_{sca}(\lambda)$  is the scattering cross section and  $C_{sca,D}$  is scattering cross-section of the particle with the size of  $D$ .  $\bar{m}$  is the phosphor particle mass being used in  $f(D)$ .  $P_{sca}(\lambda)$  illustrates the scattering power and  $I_{inc}(\lambda)$  is emission intensity.

Figure 3 demonstrates the scattering coefficient (SC) of WLED when there is presence of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor. According to this figure,  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor concentration is the main factor that cause fluctuation in the scattering coefficient and that it can also improve the color quality aspect in IPG and CPG structures. In particular, the scattering coefficient always increase when the concentration of phosphor rises, this trend is stronger when at 680 nm wavelength and above. Regardless of the particles size of phosphor particles remain at 1  $\mu\text{m}$  throughout the experiment, the scattering coefficient achieved is consistently higher with more phosphor concentration and results in better color uniformity. On the other hand, when the phosphor particles is at 7  $\mu\text{m}$ , the SC is more stable regardless of the increase in  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  concentration. This phenomenon benefits the color quality scale (CQS), therefore, 7  $\mu\text{m}$  phosphor particles is the suitable choice for WLED that focus on improving the color quality. These results confirms that WLEDs lighting efficiency is influences by both the concentration and particles diameter of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor. These properties of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor can be freely altered by the manufacturers implies that  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  is effective in enhancing luminous efficiency and color uniformity of WLEDs.

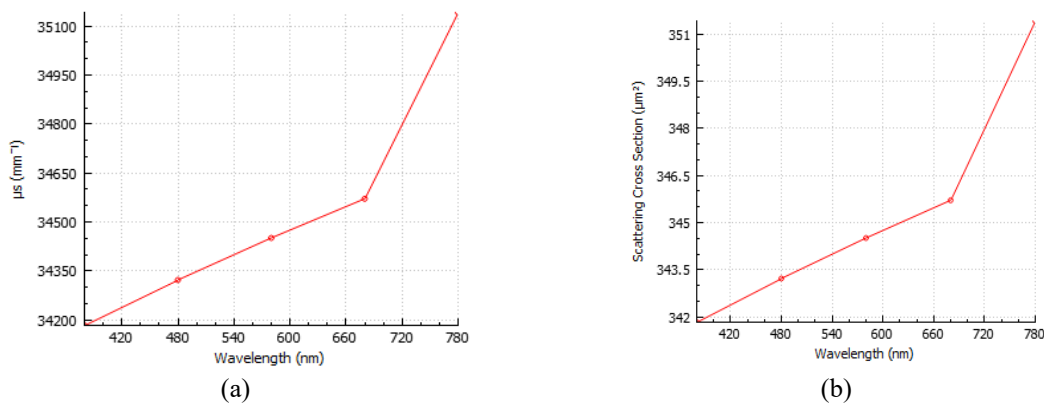


Figure 3. The computed values of (a) scattering coefficients and (b) scattering cross-section of phosphor compounding

To ensure the consistency of the research, the average correlated color temperatures must be kept in 6600 K and 7700 K. By doing this, we can focus on finding the solution suitable for upgrading the lighting properties for a specific type of WLED. Therefore, yellow  $\text{YAG}:\text{Ce}^{3+}$  phosphor and green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor concentrations must correspond to each other to maintain the ACCTs at a suitable index. This result of phosphor concentrations balance in WLED is achievable and demonstrated in (5).

$$\sum W_{pl} = W_{yellow\ phosphor} + W_{silicone} + W_{yellow-green\ phosphor} = 100\% \quad (5)$$

With  $W_{silicone}$ ,  $W_{yellow\ phosphor}$ , and  $W_{yellow-green\ phosphor}$  are proportion of weight in the phosphor compound take up by the silicone glue, the yellow  $\text{YAG}:\text{Ce}^{3+}$  phosphor, and green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor. In this formular, the amount of yellow phosphor must decline if more green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  is added into the compound to maintain the original mass count expressed as 100%.

When concerning the lighting performance of WLED, the angle color deviation is also a deciding factor. In Figure 4, the values fluctuate when there is green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  in MCW-LED, more specifically, the growth of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  reduces the amount of color deviation. This proves that MCW-LEDs with  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  has better spatial color distribution compares to when there is none.

When optimizing WLED, the development of the optical properties must be considered side by side to maintain balance. Because not all optical properties increase together and can reach the highest index, for example, CQS and luminous efficiency of WLED growth is opposite to each other. Therefore, when optimizing CQS we also need to consider the fluctuation trend in luminous flux to prevent one aspect is too

high and becomes disproportion with others. In this study, we will study CQS and luminous flux and regard their relation while finding the optimal set up for high quality WLED. As Figure 5 suggest, luminous efficacy is gradually increase when the concentration of green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor move from 2-18%. This trend happens in both 6600 K and 7700 K ACCT, which confirms the effectiveness of green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  in boosting the lumnuous efficiency in WLEDs.

The color quality scale (CQS) expressed in Figure 6, on the other hand, exhibits the opposite trends to that of luminous flux in Figure 6. The color quality scale dose not increases with green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  concentration, in particular, CQS remains the same when the phosphor concentration is from 2%-8% then begin to continuously decrease at 10% phosphor concentration onwards. So the presence of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$ .phosphor is unfavourable for the growth of CQS, however, the reduction is insignificant and considering the benefits toward color uniformity and luminous flux we can set the concentration at a suitable to achieve good results in all elements.

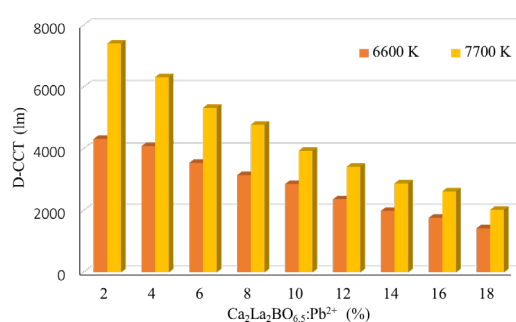


Figure 4. The CCT peak-valley deviation as a function of the concentration of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$

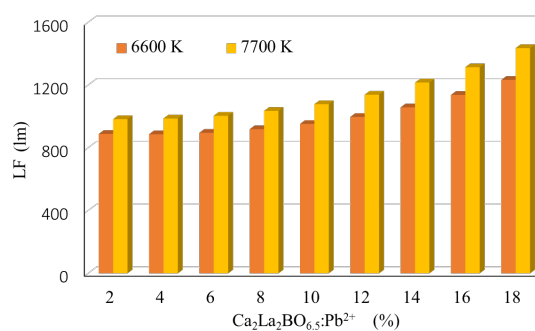


Figure 5. Luminous flux as a function of the concentration of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$

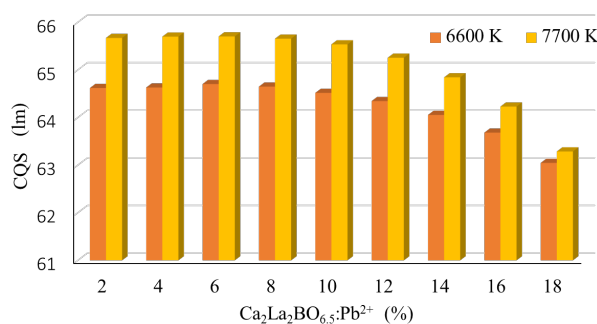


Figure 6. Color quality scale as a function of the concentration of  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$

#### 4. CONCLUSION

With the results from this research the positive effect of green phosphor  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  on WLED color uniformity and the lumen output is proven. Through experiemnts based on the Mie-scattering theory, the color uniformity can vary due to the compensation of scattered light to WLED regardless of correlated color temperature. In addition, the WLED simulation based Monte Carlo method confirms the green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor benefits the luminous efficiency, in particular, the higher the phosphor concentration is the better luminous efficiency becomes regardless of the ACCTs. In conclusion, using the green  $\text{Ca}_2\text{La}_2\text{BO}_{6.5}:\text{Pb}^{2+}$  phosphor is good for the development of WLED lighting properties.

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