

## Excellent color quality of phosphor converted white light emitting diodes with remote phosphor geometry

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### Article Info

#### Article history:

Received Jul 13, 2019

Revised Apr 6, 2020

Accepted Apr 14, 2020

#### Keywords:

Color quality

Lumen output

Mie-scattering theory

WLEDs

Y<sub>2</sub>O<sub>2</sub>S:Tb<sup>3+</sup>

ZnS:Sn<sup>2+</sup>

### ABSTRACT

The advantage in using a remote phosphor design in a WLED package is the superior luminous flux to that of the conformal and in-cup phosphor geometries. However, the disadvantage is its color quality which is lower than the results from the other two structures. This study suggests using two layers of phosphor for the remote phosphor configuration to improve the light chromaticity, including color rendering index (CRI) and color quality scale (CQS), for WLED packages with color temperature of 8500 K. The main concept of this research is to locate a green Y<sub>2</sub>O<sub>2</sub>S:Tb<sup>3+</sup> or red ZnS:Sn<sup>2+</sup> phosphor layer on the yellow YAG:Ce<sup>3+</sup> phosphor film, and then finding a suitable ZnS:Sn<sup>2+</sup> concentration to match the highest color quality. The results showed that ZnS:Sn<sup>2+</sup> brings great benefits to the increase of CRI and CQS. The greater the ZnS:Sn<sup>2+</sup> concentration is, the better the CRI and CQS become owing to the rise of red-light components in WLED packages. Meanwhile, the green Y<sub>2</sub>O<sub>2</sub>S:Tb<sup>3+</sup> phosphor brings benefits to the lumen output. However, a decrease in the luminescence and chromatic homogeneity appears when the concentrations of ZnS:Sn<sup>2+</sup> or Y<sub>2</sub>O<sub>2</sub>S:Tb<sup>3+</sup> exceed the corresponding level. This finding is verified by applying the Mie-scattering theory and the Lambert-Beer's law. The results of this article are important for the production of high-performance WLEDs in the modern lighting market.

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## 1. INTRODUCTION

As can be seen, phosphor converted white light emitting diodes (pc-WLEDs) are considered as a potential alternative to the conventional light source, and it has a variety of prospects in lighting solution [1]. The application of WLEDs has been used widely in many aspects of life such as advertising, landscape lighting, street lighting, and backlighting. Besides its mentioned benefits, its light extraction efficacy and angular uniformity of correlated color temperature (CCT) are not good enough to meet the rising the demands of illumination markets these days, which places restrictions on its development [2]. Thus, to figure out solutions for this issue, further breakthroughs in luminous efficiency and color quality are essential [3]. Today, one of the most common concepts in this aspect is getting the yellow lights from converse red phosphor combined

with the blue lights from LED chips. This concept though sounds familiar, the importance of LED structure and phosphor films' arrangement in bettering the emitted luminous flux and the color rendering index (CRI) is undeniable [4-8]. Based on it, various phosphor coating methods have been introduced in researches for WLEDs' production, including the two most common approaches: dispensing coating and conformal coating [9, 10]. Nevertheless, these structures do not provide high color quality because the applied phosphors show a degradation in light conversion due to the increased temperature at the interface between the chips and the yellow phosphor film caused by their direct contact. Thus, getting the heat outcome reduced could lead to the enhancement in phosphor performance and also prevent the phosphor from being irreversibly damaged. Many previous pieces of research have determined that the remote phosphor structure, which is designed by creating a gap between the phosphor film and the source of heat (the LED chips), can reduce the effect of heating. With a sufficient gap determined when designing the remote phosphor structure, it is possible to limit the amounts of light rays backscattered and circulated inside the LED package. Therefore, this method is considered as the most effective one in controlling the heat of LED, resulting in the enhancement of the lumen output and the chromatic performance for WLEDs [11-16]. Although the remote phosphor configuration has turned out to be qualified for regular lighting, it could not completely fulfill the advanced requirements from other lighting applications. Hence, it is essential to produce the next WLED generation that can catch up with specifications of state-of-the-art devices. For further development, some innovative remote phosphor structures are proposed for achieving the reduction in the backscattering of the phosphor-emitting lights towards the LED chips and the enrichment of luminous efficiency. A study in 2015 showed that the LED chip-emitting lights are possibly redirected to the WLED's surface with an inverted cone lens encapsulant and a surrounding ring remote phosphor layer, leading to the reduction in the internal light loss occurring due to the light reflection inside the WLED packages [17]. Additionally, the angular-dependent CCT and color stability of LEDs can be accomplished by using a patterned remote phosphor model in which a clear region in the perimeter zone is not coated with phosphors on the surrounding surface [18]. Furthermore, the patterned sapphire substrate in this remote phosphor can bring a WLED package a lot higher CCT uniformity in a far-field pattern than in a conventional pattern [19-22]. Recently, remote phosphor structure with two different phosphor films has been introduced to promote the optical performance of LEDs. Studies have focused on the improvement of the chromatic homogeneity and emitted luminous flux for pc-WLEDs with the remote phosphor configuration. Yet, these articles only concentrated on single-chip LEDs having low CCTs while heightening optical performances for high-CCT WLED package with the remote phosphor design is very complicated. Moreover, there have been no studies comparing the effectiveness of using different dual-layer phosphor structures. Therefore, manufacturers may find it hard to select an appropriate option for accomplishing higher color quality or emitted luminous flux.

This paper proposes two dual-layer remote phosphor structures for the enhancement of the WLED color quality at the CCT of 8500 K. The first configuration uses a green  $Y_2O_2S:Tb^{3+}$  phosphor layer to have the green light component inside WLEDs increased and lead to higher luminescence efficiency. For the second model, a red phosphor layer of  $ZnS:Sn^{2+}$  is added for the rise of the red-light amounts in WLEDs and then leading to increased CRI as well as improved CQS. In addition, the paper also includes a detailed description of  $Y_2O_2S:Tb^{3+}$  and  $ZnS:Sn^{2+}$  chemical compositions which affect the lighting performances of WLEDs. The results of the paper demonstrate the improvement in CRI and CQS when adding phosphor  $ZnS:Sn^{2+}$  into the phosphor layer. However, the concentration of  $Y_2O_2S:Tb^{3+}$  and  $ZnS:Sn^{2+}$  should be chosen appropriately to prevent a steep decrease in chromatic homogeneity or lumen efficacy when blue or red phosphor concentrations increase excessively. There are two main distinguishes when placing a thin film of red or green phosphors above the yellow  $YAG:Ce^{3+}$  phosphor layer: (1) The growth of the blue or red light components for increasing the white-light spectrum, which is a vital element in improving the chromaticity of the generated white lights. (2) The light scattering and light transmission inside WLEDs are inversely proportional to the green or red phosphor concentrations. Thus, determining suitable concentrations for phosphor materials is crucial to the lumen output of WLEDs.

## 2. PREPARATION

$Y_2O_2S:Tb^{3+}$ , a type of yellow-green phosphor, and  $ZnS:Sn^{2+}$  particles have many distinguishing qualities such as high quantum productivity and strength at high temperature, thus they become more and more attractive [23]. Moreover,  $Y_2O_2S:Tb^{3+}$  and  $ZnS:Sn^{2+}$  are particularly utilized for very high-loading and long lifetime fluorescent lamps. Chemical composition greatly affects the optical properties of phosphor. Therefore, when applying to WLEDs, it is necessary to carefully analyze each of their components.  $Y_2O_2S:Tb^{3+}$  glows green at a peak wavelength at 544 nm. The existence of ion  $Tb^{3+}$  increases the luminous efficiency of  $Y_2O_2S:Tb^{3+}$ . Meanwhile,  $ZnS:Sn^{2+}$  emits red light with peak wavelength of 689 nm. For the best application of these phosphors to the structuring process, they must have a spectral range which fits that of the blue lights

from the LED chip. In other words,  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$  absorption spectra must be consistent with the blue-chip spectrum. The absorption spectrum range of  $\text{ZnS:Sn}^{2+}$ , from 200 nm to 600 nm, is very beneficial to the absorption of emitted light in various bands. The reason is that the blue lights are emitted along with the conversion of the yellow lights from the yellow  $\text{YAG:Ce}^{3+}$  phosphor film. Similar to  $\text{ZnS:Sn}^{2+}$ , the absorption spectrum of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  is also wide, which is from 250 nm to 550 nm. Before performing optical simulation of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$ , their typing parameters, including the concentration, the particle size, and their stimulus, absorption, and emission spectra, need to be accurately determined by experiments. Among those mentioned parameters, the phosphor concentration and particle size are the unknowns to the improvement of color quality and luminous efficiency of WLEDs. Meanwhile, their input spectral values are fixed. According to previous studies, the phosphor grains has a fixed average diameter of 14.5  $\mu\text{m}$  [24, 25]. Meanwhile, the concentration of phosphor  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$  is calibrated to find the optimal value. This is the objective of this study.

In this study, WLEDs having 9 internal chips are used as shown in Figure 1 (a). Each blue chip has 1.6W output and 453 nm peak wavelength. Details of optical parameters of LED configuration are shown in Figure 1 (b). In order to determine the most appropriate concentration of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$ , next, remote phosphor models must be built. This research paper proposes two dual-phosphor structures, including the green-yellow phosphor configuration (GYC) and red-yellow phosphor configuration (RYC). Both structures are comprised of two phosphor films above the nine LED chips, and the yellow phosphor layer is located under the green and red phosphor films. Specifically, in GYC structure, the layer above the yellow  $\text{YAG:Ce}^{3+}$  is green  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  layer, and for the RYC, the green layer is replaced by the red  $\text{ZnS:Sn}^{2+}$  one, as can be seen in Figure 1 (c) and (d). The application of GYC and RYC configurations aims to increase the color and optical quality of WLEDs. This can be achieved by increasing the green scattering and the component of red light in WLEDs. However, concentrations of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$  need to be adjusted accordingly.



(a)

Lead frame: 4.7 mm Jentech Size-S  
LED chip: V45H  
Die attach: Sumitomo 1295SA  
Gold Wire: 1.0 mil  
Phosphor: ITC NYAG4\_EL

(b)



(c)



(d)

Figure 1. (a) The actual MCW-LEDs and (b) its parameters; (c) Illustration of GYC, and (d) RYC

Figure 2 shows the opposite trend between the concentrations of green  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  phosphor, red  $\text{ZnS:Sn}^{2+}$  phosphor and yellow  $\text{YAG:Ce}^{3+}$  phosphor. This change brings the stability to average CCTs of WLEDs and has significant effects on the phosphor films' scattering and absorption. This certainly impacts not only the color performance but also the luminous efficiency of WLEDs. Thus, the selection of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$  determines the color quality of WLEDs. When  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$  turn up from 2% to 20% wt.,  $\text{YAG:Ce}^{3+}$  concentration drops to keep average CCT. This phenomenon occurs equally with WLEDs with color temperature of 8500 K. The most noticeable is the effect of red phosphor concentration  $\text{ZnS:Sn}^{2+}$  on the WLEDs' spectrum, illustrated in Figure 3. Depending on the manufacturer's requirements, the choice is made. WLEDs with high color quality requirements can reduce a small amount of luminous flux. The intensity of the phosphor emission in the spectral regions of 420-480 nm and 500-640 nm increases with  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  concentration. These two regions' spectral emission enhancements demonstrate the increased luminous flux. Also, when the blue-light scattering in WLED develops, it means that the phosphor scattering in WLEDs increases and the result is beneficial for the copper color. This is an important result when using  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$ .

*Excellent color quality of phosphor converted white light emitting diodes with... (Thinh Cong Tran)*

Obviously, the tendency of red light spectrum from 648 nm to 738 nm increases with  $\text{ZnS:Sn}^{2+}$  concentration. However, this is not significant without the spectral increase of the two remaining regions of 420 nm - 480 nm and 500-640 nm. The spectral increase of the two 420-480 nm regions develops the luminous flux of blue light (blue-light scattering). In short, it can be said that as the color temperature rises, the spectral emission increases, and the higher color and optical quality are exhibited as a result. This is an important result when applying  $\text{ZnS:Sn}^{2+}$ , especially the quality control of high-CCT WLED packages is very difficult. This study identifies  $\text{ZnS:Sn}^{2+}$  has ability to yield better color quality for WLEDs having high color temperature (8500 K).

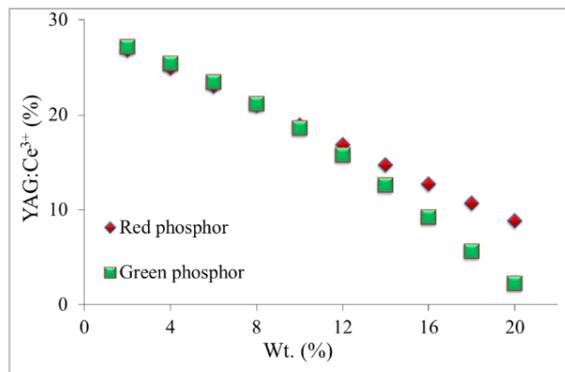


Figure 2. The change of phosphor concentration of GYC and RYC for keeping the average CCT

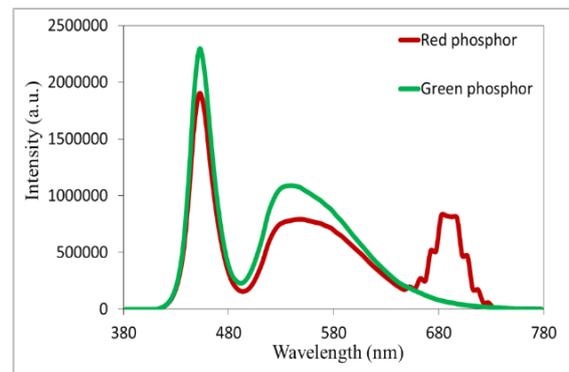


Figure 3. Emission spectra of GYC and RYC

### 3. COMPUTATION AND DISCUSSION

The color rendering index (CRI) evaluates how a light source expose the true tone of color of an object it illuminates. The increased green-light amount causes the color imbalance between the three dominant colors composing the white light: blue, yellow and green. This imbalance probably has negative influences on the color integrity of WLEDs. The graph in Figure 4 presents the degradation of CRI with the growth of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  in the structure. But this is acceptable because CRI is only a good factor of CQS. When comparing between CRI and CQS, CQS is a more complicated, crucial, and strenuous value to obtain. In Figure 5, CQS is constant as  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  concentration is less than 8%. Thus, 8%  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  can be selected to apply after considering emitted luminous flux. As shown in Figure 4, the color rendering index increased with  $\text{ZnS:Sn}^{2+}$  concentration. This can be explained by the absorption of the red phosphor layer. When phosphor  $\text{ZnS:Sn}^{2+}$  absorbs the LED chip-emitting blue light components, it turns these blue lights into red lights. In addition to blue lights from LED chips,  $\text{ZnS:Sn}^{2+}$  still absorbs yellow lights. Yet, when drawing a comparison between the yellow-light absorption and the blue-light absorption, the latter absorption happens more strongly owing to the absorption characteristics of the red phosphors. And so, the red-light amount in WLEDs grows with the higher contents of  $\text{ZnS:Sn}^{2+}$ , and this leads to increased color rendering index (CRI). When choosing a modern WLED product, CRI becomes one of the most vital parameters. Thus, the WLED having high CRI will cost more than the others with lower CRI. Nevertheless, the benefits of using  $\text{ZnS:Sn}^{2+}$  are low cost. Therefore,  $\text{ZnS:Sn}^{2+}$  can be widely used. However, as mentioned above, CRI is an element for judging the color quality included in a more overall parameter CQS. So, it is impossible to say good color quality when the CRI is high. CQS is an index comprised of three factors: the first is the CRI, the second is the preference of the viewer, and the third is the color coordinate. Because of covering three vital elements, CQS is considered as a true overall color quality index. The enhancement of CQS in accordance with the  $\text{ZnS:Sn}^{2+}$  concentration is illustrated in Figure 5. And when increasing the phosphor concentration for  $\text{ZnS:Sn}^{2+}$ , CQS also increased significantly. Clearly, using  $\text{ZnS:Sn}^{2+}$  can increase the white-light color quality for WLEDs when applying dual-layer phosphor design. This is a crucial result of research with the goal of better color quality. However, it is impossible not to consider the disadvantages of  $\text{ZnS:Sn}^{2+}$  to emitted luminous flux.

The mathematic framework of the transmitted blue light and converted yellow light in the dual-layer phosphor structure is presented in this part. Then, based on the attained results, we can develop the LED efficiency much better. For calculating the transmitted blue light and the converted yellow light in the remote phosphor configuration with a layer of phosphor whose thickness is  $2h$ , it is possible to apply the following expression:

$$PB_1 = PB_0 \times e^{-2\alpha_{B1}h} \quad (1)$$

$$PY_1 = \frac{1}{2} \frac{\beta_1 \times PB_0}{\alpha_{B1} - \alpha_{Y1}} (e^{-2\alpha_{Y1}h} - e^{-2\alpha_{B1}h}) \quad (2)$$

Meanwhile, the computation of these two aforementioned lights in the dual-layer phosphor packaging structure in which the phosphor film has  $h$  thickness can be expressed as:

$$PB_2 = PB_0 \times e^{-2\alpha_{B2}h} \quad (3)$$

$$PY_2 = \frac{1}{2} \frac{\beta_2 \times PB_0}{\alpha_{B2} - \alpha_{Y2}} (e^{-2\alpha_{Y2}h} - e^{-2\alpha_{B2}h}) \quad (4)$$

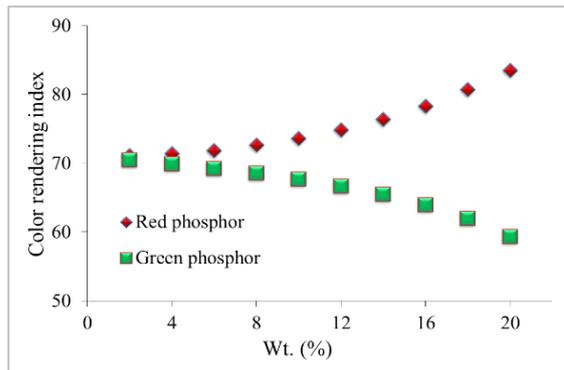


Figure 4. The color rendering index as a function of the concentration of  $Y_2O_2S:Tb^{3+}$  and  $ZnS:Sn^{2+}$

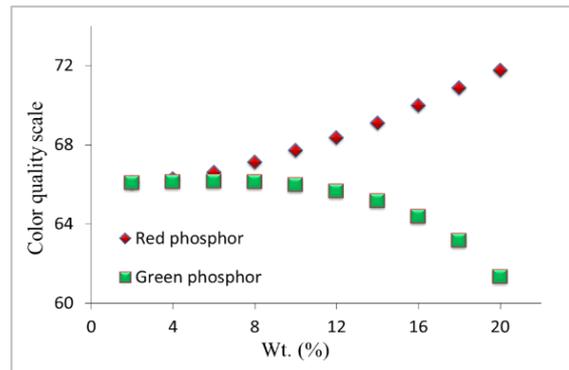


Figure 5. The color quality scale as a function of the concentration of  $Y_2O_2S:Tb^{3+}$  and  $ZnS:Sn^{2+}$

In these expressions,  $h$  symbolizes the thickness of each phosphor film in the remote structure. The single and dual-layer remote phosphor structures are expressed by the subscripts “1” and “2”, respectively. Indicated by  $\beta$  is the conversion coefficient for the blue light converting to the yellow light while  $\gamma$  represents the yellow light’s reflection coefficient.  $PB_0$  is the light intensity from the blue LED chip which is comprised of blue ( $PB$ ) and yellow ( $PY$ ) light intensities.  $\alpha_B$  and  $\alpha_Y$  characterize the fractions of the energy loss of blue and yellow lights during their multiplication in the phosphor layer separately.

The dual-layer remote phosphor structure shows the considerable advancement in the lighting efficacy of WLEDs, compared to the single-layer one:

$$\frac{(PB_2 + PY_2) - (PB_1 + PY_1)}{PB_1 + PY_1} > 0 \quad (5)$$

Mie-scattering theory is used to analysed the scattering of the phosphor particles, and calculate the scattering cross section  $C_{sca}$  for spherical particles. Meanwhile, the Lambert-Beer law is applied to measure the transmitted light power:

$$I = I_0 \exp(-\mu_{ext} L) \quad (6)$$

where,  $I_0$ ,  $L$ , and  $\mu_{ext}$  indicate the incident light power, the thickness of the phosphor layer (mm), and the extinction coefficient, in turn. In addition, the computation of the extinction coefficient  $\mu_{ext}$  can be defined as:  $\mu_{ext} = N_r \cdot C_{ext}$ , in which  $N_r$  is the number density distribution of particles ( $mm^{-3}$ ), while  $C_{ext}$  ( $mm^2$ ) presents the particles’ extinction cross-section.

From (5), we can see the luminous efficiency of WLEDs built with two separated phosphor films is higher than the packages having one phosphor layer. Thus, the paper has demonstrated the efficiency of emitting luminous flux of this dual-layer remote phosphor layer. Figure 6 shows that luminous flux increased significantly with the growth in  $Y_2O_2S:Tb^{3+}$  concentration from 2% wt. to 20% wt.. Nevertheless,  $ZnS:Sn^{2+}$  concentration has negative impact on the emitted. Clearly, based on Lambert-Beer law, the reduction factor  $\mu_{ext}$  is in direct proportion to  $ZnS:Sn^{2+}$  concentration but in inverse proportion to the light transmission energy. Therefore, the thicknesses of both phosphor films need to be fixed. Photoluminescence emitted may decrease

when the concentration of  $\text{ZnS:Sn}^{2+}$  increases. And indeed, Figure 6 shows a decrease in luminous flux. When concentration  $\text{ZnS:Sn}^{2+}$  at 20% wt., luminous flux significantly reduced. However, consider the advantages of the red phosphor layer  $\text{ZnS:Sn}^{2+}$  improve CRI and CQS. Moreover, the dual-layer remote phosphor structure results in higher lumen output, compared to the single-layer one (without the red phosphor film). Thus, it is possible to accept this reduction in the luminous flux when using  $\text{ZnS:Sn}^{2+}$  layer in WLED packages. The last problem is the requirement of manufacturers, depending on which an appropriate concentration of  $\text{ZnS:Sn}^{2+}$  is offered to produce these WLEDs in bulk.

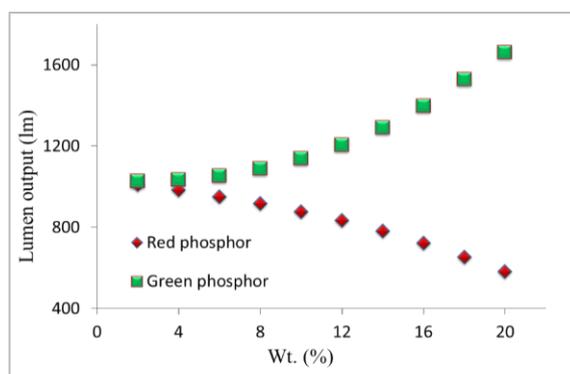


Figure 6. The lumen output as a function of the concentration of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  and  $\text{ZnS:Sn}^{2+}$

#### 4. CONCLUSION

The effects of green  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  phosphor and red  $\text{ZnS:Sn}^{2+}$  phosphor on CRI, CQS, and lumen efficacy of double-layer phosphor models are demonstrated in this research paper. By applying the Mie-scattering theory in combination with the Lambert-Beer rule, this article has successfully assured that the right phosphor material to enhance the color quality is  $\text{ZnS:Sn}^{2+}$ , while  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  is the suitable choice for the better lumen output of WLEDs. This result is true for the WLEDs having either low or high color temperatures, especially the one with color temperature higher than 8500 K. Thus, this study has accomplished its objective, enhancing the white light color performance, which is considered as one of the most difficult tasks for remote-phosphor structure. However, this structure still has a small drawback to the luminous flux. When increased concentrations of  $\text{Y}_2\text{O}_2\text{S:Tb}^{3+}$  or  $\text{ZnS:Sn}^{2+}$  are excessive, the inferior color quality or lumen efficacy can be occurred. Therefore, according to the goal of manufacturers, the importance in WLED production is to determine an appropriate concentration for the phosphor materials. And the article has provided much important information for reference in producing better quality WLEDs.

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