The Impacts of Red-emitting $Mg_2TiO_4:Mn^{4+}$ Phosphor on Color Quality of Dual-layer Remote Phosphor Configuration

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Abstract. *In terms of luminous flux, the remote phosphor structure is better than conformal structure or in-cup phosphor structure, however, this structure often has inferior color quality compared to the others. As a result, many studies have been conducted to find a solution to the drawback mentioned above. In this research, we are after the same goal using WLEDs structure with color temperature of 5600 K and come to the conclusion that dual-layer phosphor structure can improve the color rendering index (CRI) and the color quality scale (CQS). The concept of the research is to place red phosphor layer* $Mg_2TiO_4:Mn^{4+}$ *on a yellow phosphor layer* $YAG:Ce^{3+}$ *and locate the concentration of* $Mg_2TiO_4:Mn^{4+}$ *that allows the color quality to reach the highest value. The result shows that* $Mg_2TiO_4:Mn^{4+}$ *benefits CRI and CQS, more specifically, the addition of* $Mg_2TiO_4:Mn^{4+}$ *in WLEDs boosts the red light component, thus, enhancing CRI and CQS. However, it is demonstrated through the application of Mie-scattering theory and Lambert-Beer law that when the concentration of* $Mg_2TiO_4:Mn^{4+}$ *exceed the limit, it can harm the luminous flux of WLEDs. The result of this research is a valuable contribution to improving the techniques of manufacturing better WLEDs with higher white light quality.*

Keywords

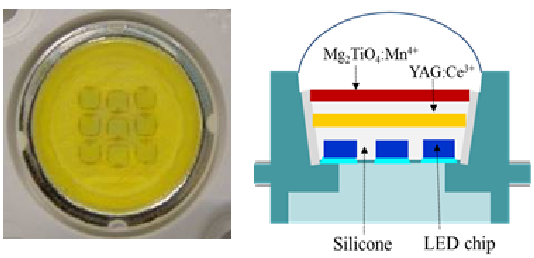
Dual-layer Remote Phosphor Geometry, WLEDs, Mie-scattering Theory, Color Uniformity, Luminous Flux.

1. INTRODUCTION

With many advantages over the previous lighting sources, the phosphor converted white light emitting diodes (pc-WLEDs) are expected to replace the conventional one [1, 2]. Despite the increasing demand of using white light-emitting diodes as a lighting solution in the daily life, from landscape lighting to street lighting and backlighting, etc., its development is still limited by the light extraction efficiency and the angular homogeneity of correlated color temperature [3,4]. Therefore, innovation in color quality and luminous flux has been approached for the breakthrough of pc-WLEDs to fulfill the market's needs [5]. Combining the blue light converted from red phosphor with the yellow light from the LED chip is currently the most common method to obtain white light. Even though the method is important, the configuration of the phosphor layer in the WLEDs is also a significant factor in deciding the luminous efficiency and color rendering index [6]. Dispensing coating and conformal coating are the two frequently used methods in producing LEDs [7]- [9]. The light conversion efficiency of these structures, however, is lowered due to the heat at the contacting point between the phosphor layer and the LED caused by direct exposure of the yellow phosphor layer to the LED. Therefore, the types of structures are unable to generate high color quality WLEDs. The obvious solution is to eradicate the heat at the junction between the phosphor layer and the LED, which can improve the light conversion efficiency of phosphor and prevent irreparable damage to it. Previous studies found out that the remote phosphor structure which provides an adequate space between the LED chip and the phosphor layer can control the temperature increase. The distance in remote phosphor structure also let the LEDs control the backscattering effect and inner light circulation. The benefits of this method have proven it as the ideal arrangement for achieving heat management in WLEDs at the same time with the improvement in the luminous efficiency and the color quality of WLEDs [10, 11]. However, other lighting applications have some requirements different from regular lighting that the remote phosphor structure cannot fulfill. Therefore, advance modifications are in demand to develop the LED adapting those requirements. In regard to this issue, some commonly used remote phosphor structures are applied with the goal of minimizing the backscattering effect from the phosphor against the chip as well as improving other lighting properties such as luminous efficiency. One of the studies suggests using an encapsulated inverted cone lens surrounded by a remote phosphor layer that can deflect the light from the chip on to the LED surface reducing light loss caused by the LED's inner circulation effect [12]- [15]. Another one employs a patterned remote phosphor structure with vacant perimeter area, which means no coating phosphor on the surrounding surface, to obtain high angular-dependent correlated color temperature consistency and chromatic stability [16]. In addition, the application of a patterned sapphire substrate to remote phosphor structure results in superior correlated color temperature uniformity in a far-field patterned compared to a conventional one [17, 18]. To improve the lumen output of the LEDs, the remote phosphor structure with two separated layers of phosphor is suggested. The varieties of research mentioned above all put effort into perfecting the color uniformity and luminous flux of remote phosphor structure WLEDs, however, they cannot be applied to improve optical qualities in WLEDs with high color temperatures as they only emphasis on single-chip WLEDs models at low color temperatures. The dual-layer remote phosphor is the vital point presented in this article as a configuration that can achieve better color quality in WLEDs with the color temperature of 5600K. The article introduces the innovative idea of enhancing CRI and CQS by using the red phosphor layer Mg_2TiO_4:Mn^{4+} to boost red light components in WLEDs and the detailed chemical composition of Mg_2TiO_4:Mn^{4+} that affects the optical qualities of WLEDs. The result of the research shows that the concentration of Mg_2TiO_4:Mn^{4+} positively affects the CRI and CQS yet the management over the concentration needed to be established to avoid damaging the luminous efficiency due to excessive concentration. Coating a red phosphor layer upon the yellow phosphor layer YAG:Ce^{3+} creates two changes. The first one is adding the red light component to increase the red light emission spectrum of the emitted white light, which is the main point in enhancing color quality. The second is that backscattering and inverted light transmission in WLEDs contrast with the concentration of Mg_2TiO_4:Mn^{4+}, thus, changes in lumen output of WLEDs can be obtained through adjusting the concentration of the red phosphor layer.

2. SIMULATION

The application of LightTools 8.6.0 program and Mie-theory is crucial in simulating a dual-layer phosphor structure. The model can be constructured by breaking down the scattering efficiency of phosphor particles and the effects of $Mg_2TiO_4:Mn^{4+}$ phosphor on the optical properties of 5600 K WLEDs with the help of these tools. The physical model of WLEDs showed in Fig. 1 consists of 9 blue chips, a reflector, a phosphor layer, and a silicon layer. The measurement of the reflector is 2.07 nm in height, the bottom length is 8 nm and the surface length is 9.85 nm. The center beneath the reflector are empty cavities to implant the chips. The radiant energy emitted by each blue chip at a peak wavelength of 453 nm is 1.16 W. The refractive indexes of $Mg_2TiO_4:Mn^{4+}$ and yellow phosphor $YAG:Ce^{3+}$ particles are 1.85 and 1.83 respectively. The $YAG:Ce^{3+}$ phosphor concentration will be adjusted inversely to that of $Mg_2TiO_4:Mn^{4+}$ to maintain average CCT.



(a) (b)

Fig. 1: Photograph of a WLEDs structure: (a) Actual WLEDs, (b) Illustration of pc-WLEDs model with red phosphor layer $Mg_2TiO_4:Mn^{4+}$, and yellow phosphor layer $YAG:Ce^{3+}$.

3. RESULTS AND DISCUSSION

After carrying out various experiments, we obtained significant results that are presented in following illustrations. Fig. 2 shows the opposite trend between red phosphor concentration $Mg_2TiO_4:Mn^{4+}$ and yellow phosphor $YAG:Ce^{3+}$. Two things can be inferred from these opposite phosphor layers: first, it can be utilized to maintain the average CCTs; and second, it can affect the scattering and light absorption processes of phosphor layers in WLEDs which certainly influence the color quality and luminous efficiency of WLEDs. Thus, the selection of $Mg_2TiO_4:Mn^{4+}$ concentration determines the color quality of WLEDs. When the concentration of $Mg_2TiO_4:Mn^{4+}$ increases from 2%-26% wt., $YAG:Ce^{3+}$ concentration decreases to keep average CCTs. This phenomenon is the same for WLEDs with color temperature of 5600 K.

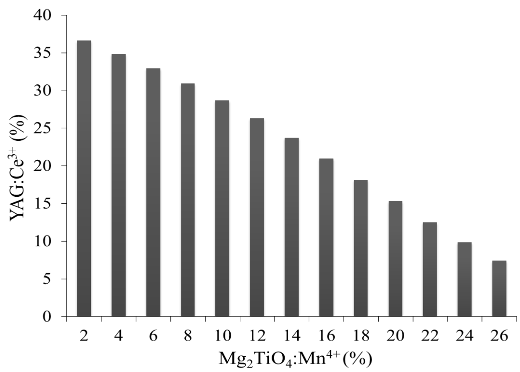


Fig. 2: The change of phosphor concentration for keeping the average CCT.

The influence that the concentration of red phosphor $Mg_2TiO_4:Mn^{4+}$ has on the emission spectrum of WLEDs is most visible as in Fig. 3. Depending on the demand from the manufacturers, a specific level of phosphor concentration can be selected. For WLEDs required high color quality, we can compromise for a smaller luminous flux so that the color quality can reach the highest index.

White light is a combination between different emission spectra as displayed in Fig. 3. The Figure illustrates the luminous flux of WLEDs with color temperature of 5600 K. The change of the red light spectrum following the concentration of $Mg_2TiO_4:Mn^{4+}$ can be easily observed in the wavelength from 648 nm to 738 nm. However, a more significant factor here is the increase in the emission spectrum at 420 nm – 480 nm and 500 nm – 640 nm, as the emission spectrum enhancement in these two regions also benefits the blue-light scattering effect. The higher the color temperature, the higher the spectral emission, leading to the better color quality and luminous efficiency.

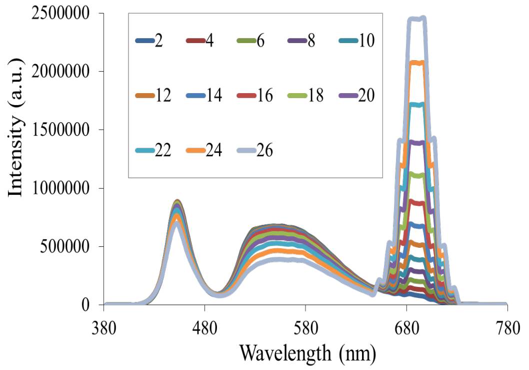


Fig. 3: The emission spectra of 5600 K WLEDs as a function of Mg_2TiO_4:Mn^{4+} concentration.

This is an important feature in applying Mg_2TiO_4:Mn^{4+}, especially when managing color quality is more difficult with WLEDs at high temperatures. This research verified that WLEDs color quality in low temperature (5600K) can be improved by using Mg_2TiO_4:Mn^{4+}. According to Fig. 4, the color rendering index increases with the upward trend of Mg_2TiO_4:Mn^{4+} concentration. This can be explained by the absorption feature of the red phosphor layer because when red phosphor Mg_2TiO_4:Mn^{4+} absorbs blue light from the LED chips, these red phosphor particles will transform blue lights into red lights, resulting in the improvement of the color rendering index. Besides, Mg_2TiO_4:Mn^{4+} particles also take in yellow light aside from blue light from the chip. However, a comparison between these two processes shows that the blue light absorbed from the chip is more significant due to the absorption attribute of the material. Therefore, when adding Mg_2TiO_4:Mn^{4+}, that the red light components in WLEDs rise benefits the color rendering index (CRI). In modern criteria for choosing a WLED, color rendering index is an important parameter obviously due to the fact that the higher color rendering index leads to the higher the price of WLEDs. The advantage of Mg_2TiO_4:Mn^{4+} is the low manufacturing cost which allows it to be widely used. Although color rendering index is important, it is just one out of many required aspects to evaluate a WLED's quality. Thus, a further examination is crucial before concluding that the color quality of WLEDs is good and that utilizing Mg_2TiO_4:Mn^{4+} is beneficial to lighting quality. Therefore, the color quality scale (CQS), a combination of color rendering index, viewer's preference and color coordinate, is the appropriate overall measurement for color quality. Figure 5

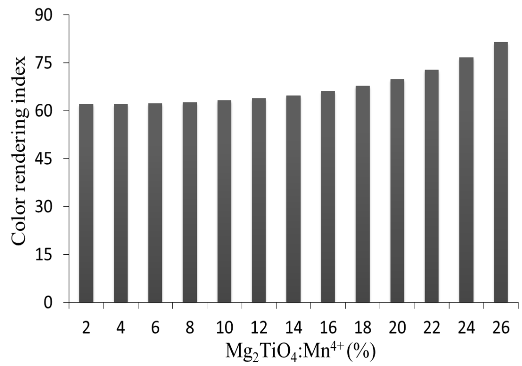


Fig. 4: The color rendering index of WLEDs as a function of Mg_2TiO_4:Mn^{4+} concentration.

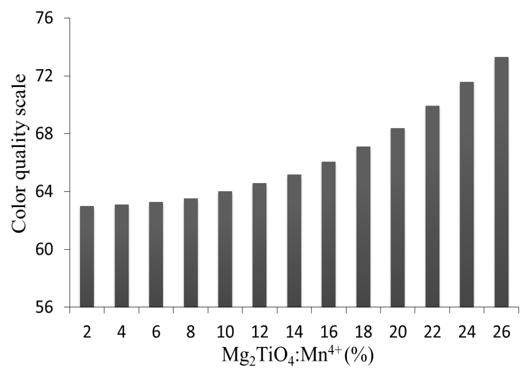


Fig. 5: The color quality scale of WLEDs as a function of Mg_2TiO_4:Mn^{4+} concentration.

shows that CQS is clearly improved when adding in the remote phosphor Mg_2TiO_4:Mn^{4+}. In addition to this, the CQS also has notable development once the concentration of Mg_2TiO_4:Mn^{4+} rises. WLEDs with dual-layer phosphor structure obviously benefit from the Mg_2TiO_4:Mn^{4+} in terms of emitted white light quality. On the other hand, the disadvantage that the excessive concentration of Mg_2TiO_4:Mn^{4+} causes to the luminous cannot be neglected. This result can be a valuable reference for further advancement in color quality.

In the following part, the article will present and demonstrate the mathematical model of the transmitted blue light and converted yellow light in the double-layer phosphor structure, from which a huge improvement of LED efficiency can be obtained. The transmitted blue light and converted yellow light for single layer remote phosphor package with the phosphor layer thickness of $2h$ are expressed as follows [19]- [21]:

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

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

The transmitted blue light and converted yellow light for double layer remote phosphor package with the phosphor layer thickness of h are defined as:

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

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

Where h is the thickness of each phosphor layer. The subscripts "1" and "2" are used to describe single layer and double-layer remote phosphor package. β presents the conversion coefficient for blue light converting to yellow light. γ is the reflection coefficient of the yellow light. The intensities of blue light (PB) and yellow light (PY) are the light intensity from blue LED, indicated by PB_0. $\alpha_B; \alpha_Y$ are parameters describing the fractions of the energy loss of blue and yellow lights during their propagation in the phosphor layer respectively.

The lighting efficiency of pc-LEDs with the double-layer phosphor structure enhances considerably compared to a single layer structure:

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

The scattering of $Mg_2TiO_4:Mn^{4+}$ phosphor particle was analyzed by using the Mie-theory. In addition, the scattering cross section C_{sca} for spherical particles can be computed by the following expression through applying the Mie theory. The transmitted light power can be calculated by the Lambert-Beer law:

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

In this formula, I_0 is the incident light power, L is the phosphor layer thickness (mm) and μ_{ext} is known to be the extinction coefficient, which can be expressed as: $\mu_{ext} = N_r C_{ext}$, where N_r is as the number density distribution of particles (mm^{-3}). C_{ext} (mm^2) is the extinction cross-section of phosphor particles.

Expression (5) shows the lighting efficiency of WLEDs with dual-layer remote phosphor is better than those with single-layer phosphor. Therefore, this article has achieved the goal of proving the effectiveness of the dual-layer remote phosphor in enhancing luminous flux for WLEDs. However, the concentration of the $Mg_2TiO_4:Mn^{4+}$ phosphor layer negatively affects the luminous flux in the dual-layer remote phosphor configuration. The Lambert-Beer law stated that the extinction coefficient μ_{ext} increase is in connection with the concentration of $Mg_2TiO_4:Mn^{4+}$ but contrasts with the light emitting power. Therefore, if the thickness of the phosphor layers in WLEDs is unchanged, the lumen output could decline. The results in Fig. 6 verify this assumption by showing a decrease in luminous flux which is especially considerable when the concentration of $Mg_2TiO_4:Mn^{4+}$ is at 26% wt. However, this minor defect is acceptable regarding $Mg_2TiO_4:Mn^{4+}$ achievements in enhancing CRI and CQS along with the fact that the luminous flux in the dual-layer phosphor structure is still better than that of single-layer (without the red phosphor layer). The specific concentration of $Mg_2TiO_4:Mn^{4+}$ used in mass production of WLEDs depends on the quality requirements of the manufacturers.

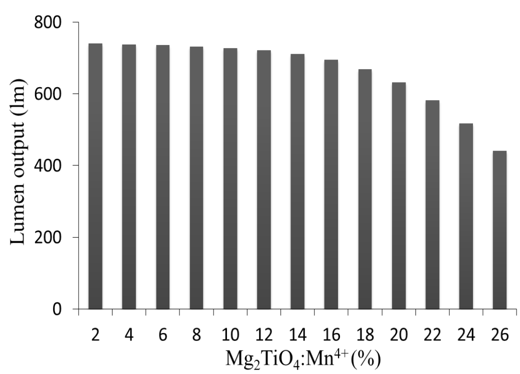


Fig. 6: The luminous flux of WLEDs as a function of $Mg_2TiO_4:Mn^{4+}$ concentration.

4. CONCLUSIONS

The goal of this article is to demonstrate the influence of red phosphor $Mg_2TiO_4:Mn^{4+}$ on CRI, CQS and luminous flux of dual-layer phosphor structure. Based on the Mie scattering theory and the Lambert-Beer law, the research comes to the conclusion that using $Mg_2TiO_4:Mn^{4+}$ is an optimal choice to improve color quality. The only drawback is that if the concentration of $Mg_2TiO_4:Mn^{4+}$ is too high, the luminous flux of WLEDs is reduced. Therefore, the concentration of phosphor layers becomes the main concern for many manufacturers because based on it or the important information provided in this article for reference, they can produce the WLEDs with the quality that accomplish their requirements.

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