The Use of $Y_3Al_5O_{12}:Ce^{3+}$ and $CaTiO_3:Pr^{3+}$ in a Dual-layer Remote Phosphor Configuration Improves The Optical Efficiencies of a Phosphor-in-glass White Ligh-emitting Diode

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Abstract. *Although distant phosphor structure is not the best choice for color fidelity of white-light-emitting diode (W-LED) sources, it benefits the luminous flux of LED more than conformal or in-cup phosphor models. After recognizing the potential of remote phosphor structures, numerous studies have been done to overcome their drawbacks of light chromaticity. In this study, a phosphor package with two phosphor films is suggested for the W-LED. The two-layer remote phosphor structure is recognized to offer a great probability of heightened chromatic rendering parameters, including the color rending index CRI and the color quality scale CQS. The color temperature of the W-LEDs employed in this investigation is 8500 K. Placing a layer of green* $Y_3Al_5O_{12}:Ce^{3+}$ *or red* $CaTiO_3:Pr^{3+}$ *phosphor on top of the yellow phosphor* $YAG:Ce^{3+}$ *layer will create the desired two-layered phosphor structure's form. The added phosphor* $CaTiO_3:Pr^{3+}$ *concentration will then be adjusted to achieve the best color quality. The existence of* $CaTiO_3:Pr^{3+}$ *increased CRI and CQS, implying that it has a significant impact on these two features. The concentration of* $CaTiO_3:Pr^{3+}$ *parallel with the CRI and CQS as the red spectral component is promoted within the W-LED. In the meantime, the green phosphor* $Y_3Al_5O_{12}:Ce^{3+}$ *advances the luminous flux. However, if the red* $CaTiO_3:Pr^{3+}$ *and green* $Y_3Al_5O_{12}:Ce^{3+}$ *phosphor concentrations are above the respective levels, it will diminish the luminous flux and color quality. According to Mie-scattering theory and Lambert- Beer's law to describe the outcome of this research. The conclusions of this study are crucial guidelines for producing better white light quality in W-LEDs.*

Keywords

W-LEDs, $CaTiO_3:Pr^{3+}$***,*** $Y_3Al_5O_{12}:Ce^{3+}$***, lumen output, color quality, Mie-scattering theory.***

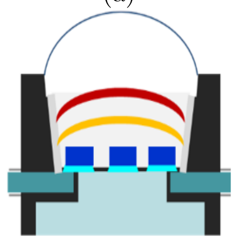
1. Introduction

Because of their low power consumption compared to traditional incandescent light bulbs, white light-emitting diodes (W-LEDs) have gained significant recognition for their tremendous energy efficiency and long life. LEDs produce white light by mixing the chip-emitted blue light with yellow light from the phosphor that is excited by the LED chip [1]. A white LED that contains blue-LED has an average color rendering index (CRI) of 80. Because of a minor red spectral proportion, the light's spectrum differs from blackbody emission [2]. Higher CRI could be accomplished with an optimized combination of the purple LED die, near-ultraviolet light (n-UV) whose wavelength is about 400 nm, and phosphor compound of blue, yellow, and red phosphor particles [3, 4]. However, as the CRI rises, the energy loss since the Stokes shift increase, and the luminous efficiency falls.

Lately, people use remote phosphor (RP) techniques as a solution for this decreasing lumen efficacy of W-LEDs [5]. The phosphor's position is usually near the LED chip inside a white LED's construction. Throughout the light emission process, the heat that both the LED chip and the phosphor generated causes significant temperature quenching to the package, thus lowering the white LED's emission efficiency. Increasing the distance between the LED chip and the phosphor could avoid the high level of heat generation and limit the temperature quenching to improve luminous efficiency. A combination of the flat plate and a phosphor film that was put on the LED chip [6] was sealed with an encapsulant [7] in an LED module using RP technology. White LED bulbs that use RP technology are also available in the market. An LED packaging structure with a phosphor coating usually includes a heat sink in white LED bulbs, and a resin globe then covers the module. Instead of phosphor, a resin ball mixed with phosphor granules shields the LED module in white LED bulbs with the RP method. Yet, kneading the phosphor powders into the resin for a W-LED package pumped with purple LED is problematic because it requires different types and higher concentrations of phosphors than the blue-LED-pumped one. Li et al. [8] demonstrated how to produce a layer of phosphor to place on the internal surface of the globe by crossing a slurry of certain phosphor powder and silicone resin into the distance between an injection molding mold and a hemispherical resin globe. The chromaticity of the W-LED lights, on the other hand, is determined by the phosphor-film thickness and its concentration. As stated by Liu et al., if we employ a mold, we must satisfy two criteria to create a uniform chromatic distribution: the great dimensional precision of the hemispherical globes, and the high-alignment accuracy between the hemispherical globe and mold. Furthermore, the entrance in the resin globe of a white LED light bulb is typically narrower than the globe's maximum diameter, and therefore bringing about the impossibility of inserting the mold to develop a phosphor layer.

For white LED light bulbs, we suggest a new coating technique to generate a phosphor layer performing homogeneous thickness on the interior surface of a resin globe [9]. The proposed coating system is depicted in Fig. 1. The combination of phosphorus powder and silicone is added to the globe of a W-LED bulb. Then, the centrifugal force generated by the rotation is used to coat the dispensed slurry mix in the middle of the globe, and use the rotation–revolution force to coat the opening. This research work is going to describe the optimizing approach for the phosphor film inside the silicone globe. It also demonstrates characteristics of a W-LED pumped with a purple LED by using a phosphor-resin globe.

(a)



(b)

Fig. 1: Schematic illustrations of the dual-layer structure of (a) YC, and (b) YR.

2. Essential preparation and simulation

2.1. Phosphor preparation process

Phosphor compositions of red $CaTiO_3:Pr^{3+}$ and yellow-green $Y_3Al_5O_{12}:Ce^{3+}$ are critical components of this study. So, in order to have the best phosphor layers that can deliver precise results, the phosphor material must be properly prepared [10].

The composition of phosphor $Y_3Al_5O_{12}:Ce^{3+}$ includes 4 different ingredients, as listed in Tab. 1. The preparation process of this cubic (garnet) structure phosphor composition consists of 3 stages of burning and followed by powdering. Initially, it is essential to mix the chemical elements together by dry grinding or milling. Then, the process of firing and powderizing will start. The combination will be burned in capped quartz boats at 1300°C with CO for the first time in 1 hour. Add another 2.7 g of NH_4Cl before moving on to the next stage, then mill the mixture to combine it. The second firing starts with the same tubes and temperature as the first, but this step lasts 2 hours. This time, however, the finished product does not need to be powderized. It should also include the basic optical properties of phosphor $Y_3Al_5O_{12}:Ce^{3+}$.

For phosphor $CaTiO_3:Pr^{3+}$ composition, its ingredients are detailed in Tab. 2. The process to get this red phosphor is a 2-stage-firing process. Before starting the first stage, all the ingredients must be mixed by dry milling or grinding to create a homogeneous mixture. Then, press this mixture into pellets. First, fire this mixture in the air at 1000°C for an hour. After 1 hour, remove the mixture from the tube and powder it using the milling process. We next crush it and squeeze it into pellets once again. The firing process has progressed to the second stage. Within 3 hours and at a temperature of 1300°C, burn the powdered product in open quartz boats. The final properties should be red emitting, with a peak emission wavelength of 612 nm. Too, the UV excitation efficiency should be +(3.40 eV), +(4.88 eV).

Tab. 1: Yellow-green phosphor $Y_3Al_5O_{12}:Ce^{3+}$ composition.

Ingredient	*Mole %*	*By weight (g)*
Y_2O_3	35.5 (of Y)	40
Al_2O_3	62.5 (of Al)	32
CeO_2	2	3.44
NH_4Cl	5	2.7

Tab. 2: Red phosphor $CaTiO_3:Pr^{3+}$ composition

Ingredient	*Mole %*	*By weight (g)*
$CaCO_3$	100	10.01
TiO_2	100	7.98
$Pr(NO_3)_3.6H_2O$	0.5	0.313
B_2O_3	3	0.209

2.2. Simulation process

The yellow-green structure (YG) and the yellow-red structure (YR) are dual-layer phosphor configurations simulated and demonstrated in this study. These YG and YR packages are made up of two phosphor films placed on blue chips. As illustrated in Fig. 1-(a), the phosphor $Y_3Al_5O_{12}:Ce^{3+}$ layer is above the $YAG:Ce^{3+}$ yellow phosphor layer in the YG structure. Meanwhile, in Fig. 1-(b), the $CaTiO_3:Pr^{3+}$ phosphor film is above the yellow $YAG:Ce^{3+}$ layer in the YR structure. The aim is to acquire the enhancement of both the color rendition and uniformity and the efficiency of luminous output for W-LEDs. This is achievable by packing the green scattering and red-light components into the W-LED modules. However, the concentration levels of $Y_3Al_5O_{12}:Ce^{3+}$ and $CaTiO_3:Pr^{3+}$ phosphors must properly change to accomplish this.

The concentrations of green $Y_3Al_5O_{12}:Ce^{3+}$ and red $CaTiO_3:Pr^{3+}$ phosphors break on the contrary path of yellow $YAG:Ce^{3+}$ phosphor, as seen in Fig. 2. This difference will maintain average CCTs and influence the scattering and absorption of phosphor layers in W-LEDs. Obviously, this will impact the LED lamps' color quality and luminous flux significantly. Consequently, the concentration of these phosphors, $Y_3Al_5O_{12}:Ce^{3+}$ and $CaTiO_3:Pr^{3+}$, is a critical factor in improving the color quality of the W-LED. Even though the W-LED is set with a

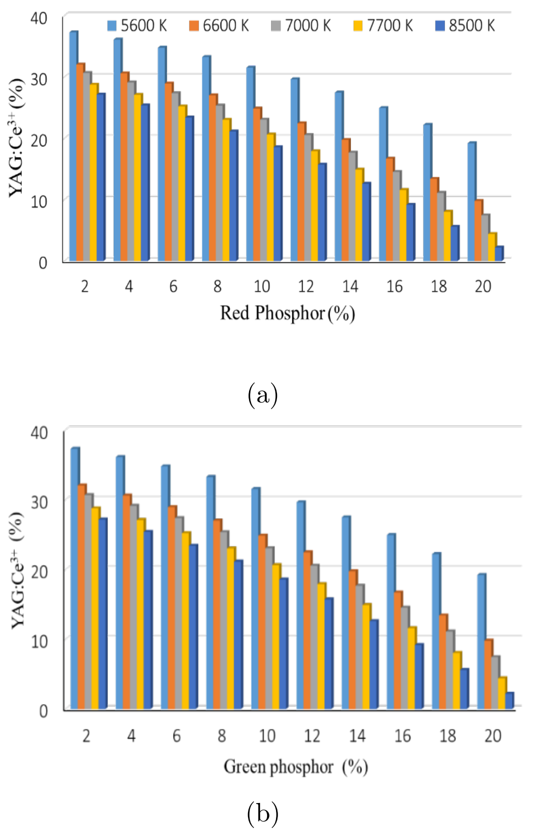


Fig. 2: The change of phosphor concentration of YR (a) and YC (b) for keeping the average CCT.

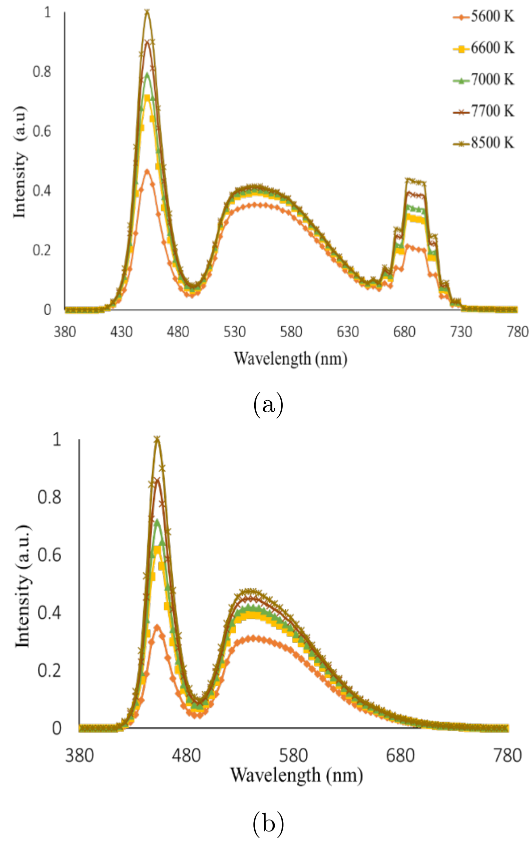


Fig. 3: Emission spectra of YR (a) and YC (b).

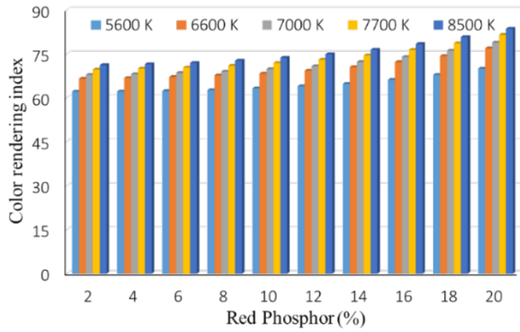
high correlated color temperature of 8500 K, YAG:Ce^{3+} concentration drops when the concentrations of $Y_3Al_5O_{12}$:Ce^{3+} and $CaTiO_3$:Pr^{3+} increase from 2% to 20% wt. to preserve the average CCT.

Furthermore, as shown in Fig. 3, the concentrations of red $CaTiO_3$:Pr^{3+} have a noticeable influence on the emission spectra of the W-LEDs. The red-yellow arrangement displays an increase in spectrum emission in three separate locations, compared to the green phosphor $Y_3Al_5O_{12}$:Ce^{3+} structure. When the $Y_3Al_5O_{12}$:Ce^{3+} concentration rises, the spectral bands of 420 nm - 480 nm and 500 nm - 640 nm also demonstrate their increasing intensity. To put it another way, an increase in emission spectra in these two sections indicates a higher luminous flux value. Besides, it seems to appear more internal blue-light scattering events when there is a $Y_3Al_5O_{12}$:Ce^{3+} presence. It means phosphor scattering increases inside the LEDs, profiting the copper color. Meanwhile, the emission spectra in the range of 648 nm to 738 nm increase as the $CaTiO_3$:Pr^{3+} concentration rises. Since the increased intensity of two 420-480 nm and 500-640 nm spectra zones would benefit the luminescence flux of the blue light (blue-light scattering), this development is negligible without enhanced emission spectra in these two comparable regions. Another finding in Fig. 3-(a) is the connection between the enhanced emission spectra and the preset CCT increase. To put it another way, the more the preset CCT rises, the stronger the emitted spectral intensity is. In that case, greater chromaticity and optical performance are achievable. This finding is a valuable reference for the LED production using the $CaTiO_3$:Pr^{3+} phosphor, especially when dealing with the quality of W-LEDs at high temperatures. In summary, the study discovered that the red phosphor $CaTiO_3$:Pr^{3+} has the ability to stimulate the color fidelity of W-LEDs with high color temperatures (8500 K). The structure can be used alternatively based on the needs and goals of the manufacturer. A modest drop in luminous flux is acceptable if they wish to make W-LEDs with excellent color fidelity.

3. Computation and discussion

CRI is a metric that assesses how well a light source reveals the color correctness of the tested models compared to natural light. According to the three primary colors: red, yellow, and green, the color imbalance arises when the green light component surpasses the others. For this reason, W-LEDs' color quality suffers, and their color fidelity suffers as a result. The graphs in Fig. 4 show the drop in CRI when we combine green phosphor $Y_3Al_5O_{12}$:Ce^{3+}. CRI falls steadily as the concentration of this phosphor is increased from 2% to 20%.

When comparing CRI to CQS, however, CRI is not the best metric because it cannot assess all aspects of light quality. As a result, the CRI reductions while utilizing $Y_3Al_5O_{12}$:Ce^{3+} are tolerable. CQS is a mixed parameter of three crucial elements comprising of the chromatic coordinates, CRI, and human preferences. Thus, CQS is a more critical goal to achieve. CQS maintains its high values when the concentration of $Y_3Al_5O_{12}$:Ce^{3+} does not exceed 8%, see Fig. 5. When the $Y_3Al_5O_{12}$:Ce^{3+} concentration increases from 8% to 20%, however, the CQS reduces dramatically. After accounting for emitted luminous flux, if the goal is to improve color quality, an adequate concentration of $Y_3Al_5O_{12}$:Ce^{3+} should be determined to range from 2% to 8%. Meanwhile, when the red phosphor $CaTiO_3$:Pr^{3+} is present, the CRI increases slightly. In other words, as seen in Fig. 4-(a), CRI also grows along with the $CaTiO_3$:Pr^{3+} concentration. The absorption property of the red phosphor $CaTiO_3$:Pr^{3+} in the remote phosphor configuration is the driving factor behind this increased trend in CRI. This red phosphor may absorb both the blue and yellow light emitted by the LED chip. Furthermore, when $CaTiO_3$:Pr^{3+} absorbs the blue light, it converts it to red light. The absorption performance of the blue-light case is more intense than that of the yellow-light one, despite the fact that these red $CaTiO_3$:Pr^{3+} particles absorb both types of emitted lights. In effect of adding $CaTiO_3$:Pr^{3+}, the red light component has risen, resulting in a greater CRI. Because the color rendering index is an important criterion for an efficient modern W-LED light, an LED package with a high CRI will be more expensive than others. The usage of $CaTiO_3$:Pr^{3+}, on the other hand, allows for lower production costs, which has led to its widespread use in LED creation. However, as previously said, researchers have concentrated on CQS to offer a more precise valuation to the color properties of W-LEDs.



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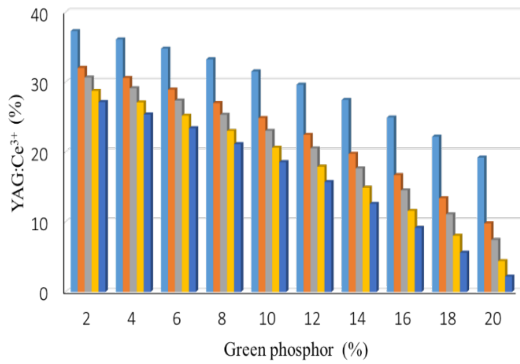
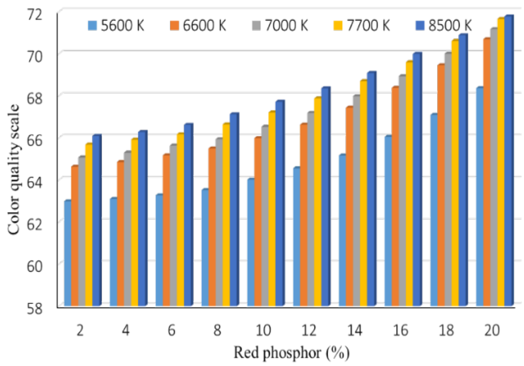


(b)

Fig. 4: CRI of W-LEDs as a function of (a) $CaTiO_3$:Pr^{3+} and (b) $Y_3Al_5O_{12}$:Ce^{3+} concentrations.

As with the CRI, the red phosphor $CaTiO_3$:Pr^{3+} in Fig. 5 can improve CQS



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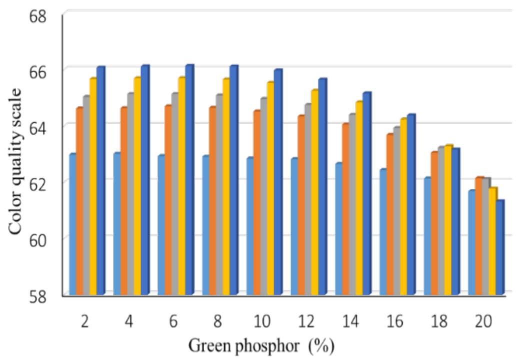

(b)

Fig. 5: CQS of W-LEDs as a function of (a) $CaTiO_3:Pr^{3+}$ and (b) $Y_3Al_5O_{12}:Ce^{3+}$ concentrations .

values. The first graph in Fig. 5 points that CQS levels climb in lockstep with the concentration of $CaTiO_3:Pr^{3+}$. Thereupon, it is possible to conclude that using a $CaTiO_3:Pr^{3+}$ phosphor layer improves the color performance of LED white lights utilizing dual-layer phosphor structures. This finding is essential for advanced WLED models, and the W-LED producers can consider these to be applied to their high-chromaticity LED products. However, the fact that $CaTiO_3:Pr^{3+}$ causes a negative impact on W-LED lumen output should not be underestimated.

This section will demonstrate a scientific model of blue light transmission and yellow light conversion in a two-layer phosphor structure. Furthermore, this model may lead to a significant improvement in LED efficiency.

Initially, the computing model of the transmitted blue light and the converted yellow light of a one-layer RP geometry is presented with two following expressions [11]-[13]. Note that the thickness of the phosphor film, in this case, is $2h$.

$$PB_1 = PB_0 \times e^{-2\alpha_{B_1} h} \tag{1}$$

$$PY_1 = \frac{1}{2} \frac{\beta_1 \times PB_0}{\alpha_{B_1} - \alpha_{Y_1}} \left(e^{-2\alpha_{Y_1} h} - e^{-2\alpha_{B_1} h}\right) \tag{2}$$

Subsequently, this model in the case of two-layer RP structure using a phosphor thickness of h is performed as:

$$PB_2 = PB_0 \times e^{-2\alpha_{B_2} h} \tag{3}$$

$$PY_2 = \frac{1}{2} \frac{\beta_2 \times PB_0}{\alpha_{B_2} - \alpha_{Y_2}} \left(e^{-2\alpha_{Y_2} h} - e^{-2\alpha_{B_2} h}\right) \tag{4}$$

in which h indicates the thickness of a phosphor layer in the structure. The subscripts "1" and "2" express one-layer and two-layer RP structures. β is the conversion coefficient of blue light to yellow light while γ is the reflection coefficient of the yellow light. The sum of blue-light and yellow-light intensities, PB and PY, respectively, is the LED-emitted light intensity, presented by PB_0. Besides, $\alpha_B; \alpha_Y$ show the parameters of the energy-loss fractions of blue light and yellow light during their multiplication in the phosphor layer separately.

With the two-layer RP structure, the illuminating performance of the W-LED is more outstanding than that of the one-layer RP one:

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

With the support of Mie-scattering theory [11, 12], the phosphor material's scattering was specified, and the scattering cross section C_{sca} of phosphor spheres can be calculated by the following expression. In addition, the transmitted light power is computed utilizing the law of Lambert Beer [14]-[16]:

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

where, I_0, L, and μ_{ext} are the incident light power, the thickness of the phosphorus layer (mm), and the extinction coefficient, in turn. Besides, μ_{ext} is expressed as: $\mu_{ext} = N_r.C_{ext}$, in which N_r defines the number density distribution of particles (mm^{-3}), and C_{ext} shows extinction cross-section of phosphor particles (mm^2).

The luminous efficiency of dual-layer remote phosphor pc-LEDs is higher than that of single-layer phosphor pc-LEDs, as shown in expression (5). As a result of this study, the lumen efficacy of the dual-layer remote phosphor layer used in LED packages has been demonstrated.

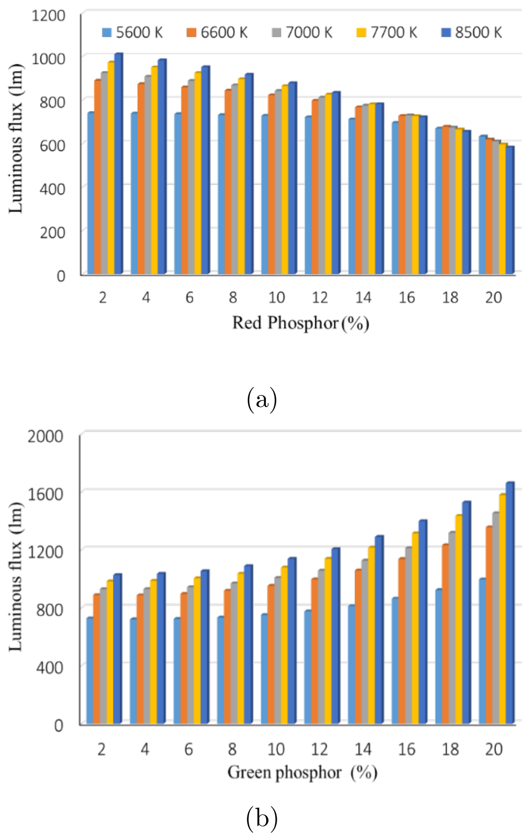

Fig. 6: The lumen output as a function of (a) $CaTiO_3:Pr^{3+}$ and (b) $Y_3Al_5O_{12}:Ce^{3+}$ concentrations

The link between the concentration of the phosphor materials and the luminous flux of two YR and YG configurations is also examined and demonstrated in Fig. 6. Particularly, in Fig. 6-(a), the light flux of the YR structure decreases as the concentration of red $CaTiO_3:Pr^{3+}$ increases in the range of 2%-20% wt. In contrast, Fig. 6-(b) shows the improvement of the luminous flux in direct proportion to the growing concentration of green $Y_3Al_5O_{12}:Ce^{3+}$ from 2% to 20% wt., regardless of the CCT. To demonstrate this contrast when using these two different phosphors, it is essential to employ the Beer's law. The Lambert-Beer law states that the reduction factor μ_{ext} is proportional to the $CaTiO_3:Pr^{3+}$ concentration and inversely proportional to the lighting transmission power. As a result, if the thicknesses of the two phosphors are kept constant, the photoluminescence output may decrease as the concentration of $CaTiO_3:Pr^{3+}$ grows. The light flux produced by the two-layer RP structure with a red phosphor layer of $CaTiO_3:Pr^{3+}$ is superior to that produced by the one-layer RP structure (not using red phosphor).

Though the luminous flux is reduced in the case of high-concentration red $CaTiO_3:Pr^{3+}$ layer, both CRI and CQS get benefit from this stronger red spectral energy [17, 18]. As a result of the benefits gained from utilizing $CaTiO_3:Pr^{3+}$, the decrease in W-LED luminescence is totally acceptable. Meanwhile, the increase in the green phosphor $Y_3Al_5O_{12}:Ce^{3+}$ concentration can elevate the lumen output but might not be completely favorable to the color rendition properties of RP structures. Hence, depending on the market and industry needs and demands, LED-production companies can choose the suitable concentration of $CaTiO_3:Pr^{3+}$ to use in their W-LEDs fabrication.

4. Conclusion

This work presents the effects of green $Y_3Al_5O_{12}:Ce^{3+}$ and red $CaTiO_3:Pr^{3+}$ phosphors on lumen efficacy and the color rendition properties of the two-layer RP W-LED. The Mie-scattering theory and Lambert-Beer's law were used to establish that $CaTiO_3:Pr^{3+}$ is a good phosphor material for the stimulation of high-chromatic-rendering W-LED with RP packages. On the other hand, $Y_3Al_5O_{12}:Ce^{3+}$ has turned out to be a good choice for increasing the luminous flux of W-LEDs. The attained results moreover are affirmative in the cases of low and high preset CCTs (5600-8500 K). As a result, our research has succeeded in increasing the color fidelity of white light, which is a challenge for RP structures. Nevertheless, there is one minor disadvantage in the optical output of the package regarding usage of the phosphors, which is that too high $Y_3Al_5O_{12}:Ce^{3+}$ or $CaTiO_3:Pr^{3+}$ concentrations might get the color rendition or lumen intensity suffered. As a result, determining an optimum concentration of those phosphors, based on the purpose of the W-LED manufacturer, is critical. Furthermore, the research paper's findings can be a vital reference for a potential and practical development path of future higher-quality W-LED packages.

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