

Application of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ and $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ in improving the lighting quality of phosphor-in-glass based white light-emitting diodes with the dual-convex design

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ABSTRACT

Although the luminous of the remote phosphor structure tends to be better than that of the in-cup or conformal phosphor structures, the poor light quality prevents this lighting method from being widely used. It is recognized through experiments that the two-layer remote phosphorus structure should be used to improve color rendering index (CRI) and color quality ratio (CQS) for WLEDs. In the experiments, WLED structures containing green $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ or red $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ phosphor on the yellow YAG:Ce³⁺ phosphor were performed at 8500 K. After that, $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ and $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ concentrations in each WLED structure is adjusted until chromatic performance reached the finest quality. As a result, $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ is proved to bring great benefits to the increase of CRI and CQS. Specifically, the greater the concentration of $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$, the better CRI and CQS because of the additional red-light material from this phosphor. The other phosphor material, green $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ phosphor, is beneficial for the expansion of luminous flux. However, if the concentration of $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ or $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ is over the limit, the decrease in lumen output and chromatic quality will occur. While doing the experiment, Mie-scattering theory and the Beer's law are great tools to verify the accuracy of results. The results of this article can serve the purpose of improving WLEDs fabrication to produce higher quality product.

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1. INTRODUCTION

Phosphor converted white light emitting diodes (pc-WLEDs) has the noble mission of replacing conventional light sources. This fourth-generation light source is widely used in many areas of our life, from indoor to outdoor lighting, for examples, household lighting, street lighting, and backlighting [1-3]. However, it has some existing issues in light extraction efficacy and uniformity of angular color temperatures (CCT) [4, 5]. Because people always demand the best quality in products, the luminous efficiency and color quality should be improved to match this expectation. Today's most common approach to produce white light device is to apply the combination of blue light from converse red phosphor and the converted yellow light from LED chip. This familiar concept has proven itself in the lighting industry; moreover, the organization of

elements to form the LED package, especially the phosphor arrangement, have enormous effects on the lumen output and the color rendering index (CRI) of the LED products [6-8].

Dispensing coating and conformal coating are two of the several common phosphorus coating methods that have been proposed for LED lighting. However, these structures do not guarantee good color quality because of the degradation in light conversion of phosphorus materials caused by the increase of heat at the surface where the LED directly contacts the phosphor layer [9-11]. Therefore, if the generated temperature can be controlled, the efficiency of phosphor layer will improve and also prevent irreversible damage to the device. Many previous pieces of research have determined that distancing the phosphor from the LED chip (the heat source) can achieve this goal; and this design is also known as remote-phosphor structure [12-15]. When the distance is large enough, LED lights can absolutely minimize the scattering and circulation of light inside so that the luminous efficiency and the color quality of LEDs could be enhanced. Though the remote packaging design is qualified for regular lighting, it is unable to fulfill all the requirements of modern illuminating applications, leading to the need of fabricating the next generation of LED with better performance [16, 17].

For further development, particularly the enhancement in LED luminescence, the backscattering reduction inside the LED package is the focus, and there have been noticeable structures suggested to achieve this objective. According to the information gathered from several of researches, it is found that inverted cone lens encapsulant and a surrounding ring remote phosphor layer are perfectly capable of transferring the emitted blue light from the chip to the LED surface and reducing the light loss caused by the internal reflection. In addition, the LED can attain high angular CCT homogeneity and color stability when a patterned remote phosphor structure is applied. Moreover, it is noticed that this patterned remote structure has a clear zone in the perimeter area which is not coated by the phosphor on the surrounding surface [18]. Besides, it is reported that applying the patterned sapphire substrate for the patterned remote phosphor structure can result in much more excellent CCT uniformity in a far field pattern than in a conventional pattern [19-22]. With the aim of advancing the efficiency of this remote phosphor package, the remote phosphor comprised of two layers of phosphor is proposed. The researchers conducted on this new dual-layer structure focus on achieving both better color uniformity and higher emitted output at the same time. However, only single chip WLEDs with low color temperatures were studied due to the complexity of improving optical performance in multi-chip WLEDs at high CCT. Moreover, there has not been any study comparing the effectiveness of using different dual-layer remote phosphor modules. Therefore, manufacturers are still facing difficulties in choosing a suitable option to improve the chromatic homogeneity and luminous flux for their LEDs.

This paper proposes two dual-layer remote phosphor structures to enhance the color quality of WLEDs at color temperature of 8500 K. Specifically, green light component in WLEDs will be improved by using the green $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ phosphor layer so that the luminous could be better. Meanwhile, the use of red $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ phosphor layer aims to enrich the red-light amount in WLEDs and leads to increased CRI and CQS. Additionally, the chemical composition of $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ and $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ which affect the optical properties of WLEDs are also described in details. It is found that the participation of $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ and $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ is useful for the improvement of WLEDs in general. However, the concentration of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ and $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ need to be selected appropriately, otherwise, the quality of white light color or luminous efficiency will go down as a result from excessive amount of phosphor. Setting additional green phosphor or red phosphor layer over the $\text{YAG}:\text{Ce}^{3+}$ yellow phosphor layer will create some differences in lighting performance. Firstly, green and red-light components expand the emission spectrum of white light, which is the main factor that contributes to the quality of light. Secondly, the direction of scattering and the transmission of light in WLEDs are opposite to the phosphor concentration, therefore, the suitable phosphor concentration should be carefully determined to keep high luminous flux of WLEDs.

2. PREPARATION

In this paper, we need to prepare two compounds: $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ and $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$. The following descriptions are guides on how to make these two substances, with the first one is $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ and then $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$. The first step of preparation is understanding that $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ is a combination of 4 grassroots: 282 g of MgO (with a very large majority), 62 g of MgF_2 , 9.2 g of MgCO_3 and 201 g of GeO_2 . Secondly is the process of creating $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$. The ingredients need to be mixed at the start by dry milling or balling. Next, fire the combination in capped quartz tubes in the air and at a temperature of 1200 degrees Celsius for 2 hours, after that milling it to powder. After that, put the outcome of previous step into open quartz boats and fire it in normal air condition, at 1200 degrees Celsius within 16 hours. After the firing finishes, we will get the final phosphor product.

The process of creating $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ is seen as successful when the combination attains these following optical properties. During the emission, the compound has a deep red color. In addition, its peak emission is 1.88 eV, while its indexes of excitation efficiency by UV are $-(4.88 \text{ eV})$ and $-(3.40 \text{ eV})$. The process of creating $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ is simpler than $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$. A good preparation of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ means it meets the following conditions. The excitation efficiency by UV and e-beam are weak red emissions. Next, the excitation efficiency by infrared light is also weak red emission. The optical properties are divided into two parts: emission color and peak. Color must be green and peak belongs to 550 nm. In order to meet these requires, we need 78 percent of YF_3 , 20 percent of YbF_3 , 2 percent of ErF_3 , 100 percent of BaF_2 and 10 percent of ZnF_2 in the ingredient. Next, mix thoroughly the combination in ethanol and then fire it in a capped alumina crucible in inert or fluorizing atmosphere at a temperature above 1100 degrees Celsius. Finally, cleanse with water after letting cool down to remove residue.

The green-yellow double-layer phosphor configuration (GYC) and red-yellow double-layer phosphor configuration (RYC) designed with convex shape are applied for better color quality and emission capacity, a result achieved by the increased green and red-light components in WLEDs. As in Figures 1(a) and 1(b), the settings in GYC and RYC are presented in details. In Figure 1(a) is the illustration of the GYC, in which the two phosphor layers are above blue chips; particularly, the $\text{YAG}:\text{Ce}^{3+}$ yellow phosphor layer is below the phosphor $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$. The RYC structure in Figure 1(b) also has similar arrangement, red phosphor $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ is placed above $\text{YAG}:\text{Ce}^{3+}$ yellow phosphor layer. However, the concentrations of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ and $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ need to be suitably adjusted to achieve the desired results.

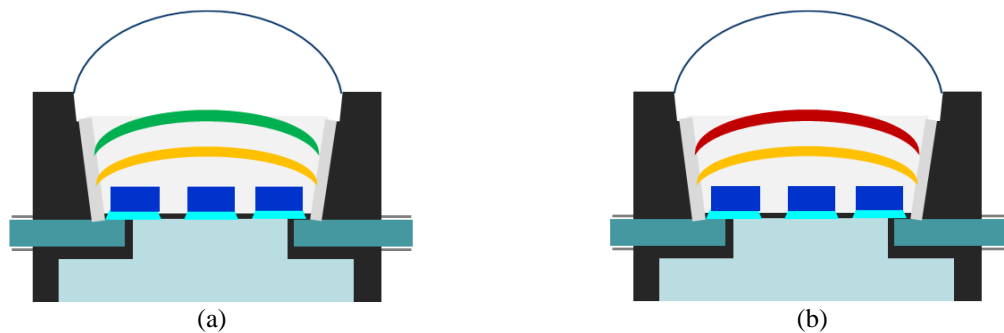


Figure 1. Settings in GYC and RYC: (a) Illustration of GYC, (b) RYC

Figure 2 simultaneously shows the changes in the concentrations of three phosphors: green $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ phosphor, red $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ phosphor and yellow $\text{YAG}:\text{Ce}^{3+}$ phosphor. This implies two functions of phosphor concentration: retaining the stability of the average CCTs and creating the influence on the phosphor scattering and absorption in WLEDs. Therefore, it certainly affects the color quality and lumen output of WLEDs. Thus, the selection of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ and $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ determines the color quality of WLEDs. When $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ and $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ turn up from 2%-20% wt., $\text{YAG}:\text{Ce}^{3+}$ concentration drops to keep average CCT. This phenomenon occurs in WLEDs to keep the condition of color temperature at 8500 K. In Figure 3, we need to observe carefully the effect of red phosphor concentration $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ on the spectrum of WLEDs to conclude the impact of this phosphor on optical properties. WLEDs with high color quality can reduce a small amount of luminous flux. $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ concentration supports the development of intensity in the spectral regions of 420-480 nm and 500-640 nm. These two regions' spectral emission enhancements demonstrate the increased luminous flux. The scattering of the blue rays inside the WLED increases, which means the phosphor scattering in WLED increases, and thus this is beneficial to the copper color. This finding is important for the application of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ in the phosphor package. Besides, the increasing tendency of the red spectrum from 648 nm to 738 nm with concentration $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ is obviously presented. However, this is not significant without the spectral increases of the two remaining regions of 420-480 nm and 500-640 nm. The spectral increase of these two regions increases the luminous flux of blue light (blue-light scattering). Accordingly, the higher the color temperature, the higher the spectral emission. Thus, the color and optical quality become much better. This is a noticeable success when applying $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$, especially controlling the color quality of WLEDs with high temperature is very difficult. This study identifies that $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ could upgrade the chromatic performance of WLEDs with high CCT (8500 K).

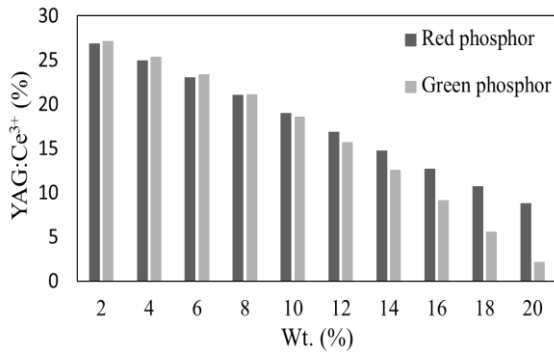


Figure 2. The change of phosphor concentration for keeping the average CCT in GYC and RYC

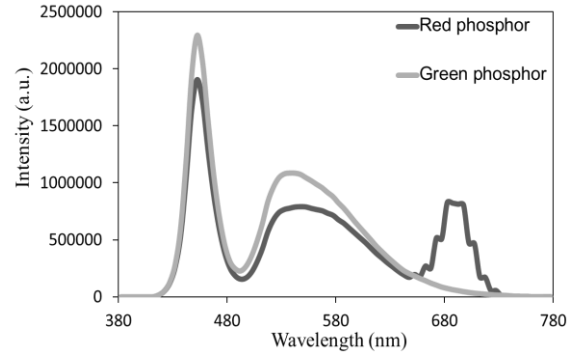


Figure 3. Emission spectra of GYC and RYC

3. COMPUTATION AND DISCUSSION

The color rendering index (CRI) is known as a tool to assess how the color of an object is exposed when a light source illuminate it. When there is too much blue light, it will lead to an imbalance between three colors: blue, yellow, and green. This directly affects the color quality of WLEDs and color integrity of WLEDs. In Figure 4, as soon as the remote phosphor $BaY_2F_8:Er^{3+},Yb^{3+}$ appears, CRI tends to fall down. After all, it is still acceptable if CRI decreases but acquire high index in CQS. Between CRI and CQS, the CQS index is much more important and harder to acquire. In Figure 5, CQS stays stable when $BaY_2F_8:Er^{3+},Yb^{3+}$ concentration is less than 8%. Thus, 8% $BaY_2F_8:Er^{3+},Yb^{3+}$ can be selected to apply after considering emitted luminous flux. As can be seen from Figure 4, the color rendering index has an upward trend with $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ concentration. This can be explained by the absorption of the red phosphor layer. After absorbing the blue rays emitted from the blue chips, $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ phosphors convert these blue lights into the red lights. Apart from the blue beams, $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ still absorbs the yellow light components. However, because of the absorption properties of the material that has stronger blue light absorption. Consequently, the red-light component in WLEDs increases when adding $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ and leads to increased CRI. In fact, CRI is one of the key parameters for selecting modern WLED lamps. The WLED devices that own higher color rendering index undoubtedly cost more to fabricate. However, the benefits of using $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ is low cost, which probably handle this issue. Therefore, $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ can be widely used. However, CRI is just a factor that evaluates an aspect of WLED color quality. It is impossible to conclude that WLEDs have good color quality just because of high CRI. Recent studies provide a new parameter for white-light color evaluation, color quality scale (CQS), is more efficient. CQS is a synthetic index of three factors: the first is the CRI, the second is the audiences' preference, and the third is the color coordinate. As including three crucial factors in color evaluating, CQS is almost a true overall index of color quality. Figure 5 illustrates the enhanced CQS when $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ exists in the WLED remote structure. Moreover, when increasing concentration of $Mg_8Ge_2O_{11}F_2:Mn^{4+}$, CQS also increases significantly. Clearly, using $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ can increase the quality of light color for WLEDs with convex-dual-layer remote phosphor structure. This outcome is valuable for manufactures whose aim is to improve the color quality. However, it is impossible to ignore the disadvantages of $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ caused to emitted luminous flux.

In the next part, there is the demonstration of scientific model in relation to the transmitted blue light and the converted yellow light of a dual-layer remote phosphor structure. The calculation in this part will contribute greatly to the significant enhancement in WLED performance. In particular, the transmitted blue light and the converted yellow light in a single-layer remote structure in which the phosphor layer is $2h$ thick are computed by [23-25]:

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

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

Meanwhile, for the dual-layer structure with each phosphor thickness of h , the expressions of its transmitted blue light and converted yellow light are:

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

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

In which, h mentions the thickness of each phosphor film in the WLED structure. The subscripts “1” and “2” express the single-layer and the dual-layer remote phosphor structure, respectively. β indicates the conversion coefficient for blue light converting to yellow light, while γ describes the yellow-light reflection coefficient. The intensities of blue light (PB) and yellow light (PY) define the light intensity from blue LED whose mathematical symbol is PB_0 . α_B ; α_Y are parameters characterizing the fractions of the energy loss of the blue ray and yellow ray during their multiplication in the phosphor class, separately.

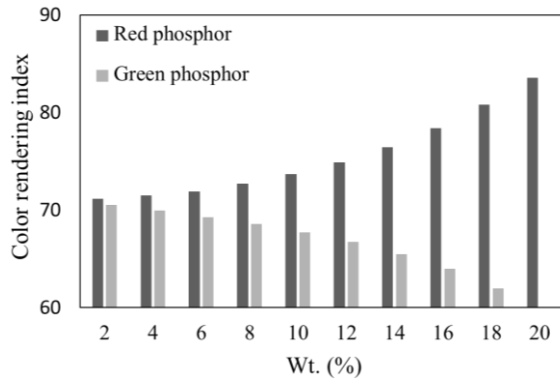


Figure 4. The color rendering index as a function of the concentration of $BaY_2F_8:Er^{3+}, Yb^{3+}$ and $Mg_8Ge_2O_{11}F_2:Mn^{4+}$

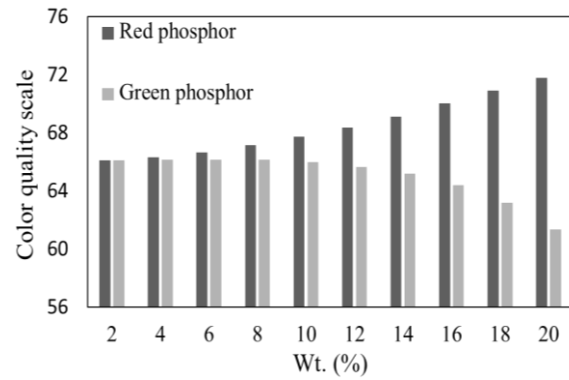


Figure 5. The color quality scale as a function of the concentration of $BaY_2F_8:Er^{3+}, Yb^{3+}$ and $Mg_8Ge_2O_{11}F_2:Mn^{4+}$

The LED illumination efficacy with the dual-layer remote phosphor structure shows a remarkable enhancement when it is compared to the performance of the single-layer one:

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

With the application of Mie’s theory, the scattering ability and the scattering cross section C_{sca} for spherical phosphor particles can be determined and computed, respectively. Meanwhile, thanks to the Beer’s law, the calculation of transmitted light power is carried out:

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

I_0 here is the power of incident light, L represents the thickness of each phosphor class (mm), and μ_{ext} is the extinction coefficient that is calculated by: $\mu_{ext} = N_r \cdot C_{ext}$, in which N_r expresses the number density distribution of particles (mm^{-3}), and C_{ext} (mm^2) means the phosphor extinction cross-section.

From (5) we can see the luminous efficiency of dual-layer remote phosphor is larger in comparison to that of a single-layer structure. Thus, the paper has proven that the double-layer remote phosphor package is more beneficial to light output than the traditional package. Figure 6 shows that luminous flux increases significantly when $BaY_2F_8:Er^{3+}, Yb^{3+}$ concentration goes up to 20% from 2% wt. However, the luminous flux of the dual-layer WLED package is influenced by the phosphor content of $Mg_8Ge_2O_{11}F_2:Mn^{4+}$. Clearly from the Lambert-Beer’s law, the reduction factor μ_{ext} is in the direct proportion to the $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ concentration but inversely proportional to the light transmission energy. Therefore, if we keep the thickness of two phosphor layers constant, the photoluminescence emitted may decrease when the concentration of $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ increases. And indeed, Figure 6 shows a decrease in luminous flux. When concentration $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ stays at 20% wt., luminous flux significantly reduced. However, this degradation of flux is completely acceptable. The reasons are that the advantages of the red phosphor layer $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ is proven in obtaining CRI and CQS improvement, and the dual-layer remote phosphor has higher luminescence than the single-layer that do not use a layer of red phosphor. The last problem is the intentions of the manufacturers, based on which the appropriate concentration $Mg_8Ge_2O_{11}F_2:Mn^{4+}$ can be decided for producing these WLEDs in bulk.

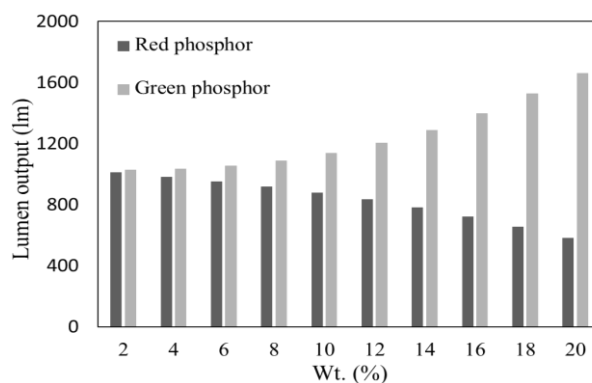


Figure 6. The lumen output as a function of the concentration of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ and $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$

4. CONCLUSION

Through the Mie-theory and the Beer's rule, this article has shown the advantages of each additional phosphor. $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ is seen as a compound supporting color quality; meanwhile, $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ is the choice to improve the luminous flux of WLEDs. The color temperature of WLEDs must be kept at 8500K by balancing the concentrations of phosphor in WLEDs to ensure the performance of red and green phosphor. The results are only applied to WLEDs that employ remote phosphor design. In addition, the luminous still faces a small trouble which is the reduction of luminous flux when the concentrations of $\text{BaY}_2\text{F}_8:\text{Er}^{3+}, \text{Yb}^{3+}$ or $\text{Mg}_8\text{Ge}_2\text{O}_{11}\text{F}_2:\text{Mn}^{4+}$ increase excessively. Therefore, the article provide the information on phosphor application and the manufacturers should decide on the amount of phosphor needed to improve their products.

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