

LaSiO₃Cl:Ce³⁺, Tb³⁺ and Mg₂TiO₄:Mn⁴⁺: quantum dot phosphors for improving the optical properties of WLEDs

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ABSTRACT

In this research, we focus on the solutions to enhance the lighting properties as well as the heat regulation of the white light-emitting diodes (WLEDs) with conventional phosphor and quantum dots (QDs). Although receiving lots of attention for being an innovative lighting solution with good color rendering index, the potentials of WLEDs conjugated with quantum dots (QDS), especially the QDs-phosphor mixed nanocomposites ones, are restrained due to the lacking performance in the aspects mentioned above. The crucial requirement to produce better WLEDs is finding solutions that improve the lacking aspects, therefore, through observing previous studies and applying advanced technique, this research suggest an effective and unique packaging configuration, in which the nanocomposites QDs-phosphor layer is set horizontally to the WLED. This novel packaging configuration allow WLED performance in terms of lighting and heating to reach it peaks. This is the first time four different types of WLEDs, single-layer phosphor, dual-layer remote phosphor with yellow-red and yellow-green, and triple-layer phosphor, were simulated, utilized and compared in one study to decide the best WLED configuration. The results show that the triple-layer phosphor configurations improve the color rendering ability and lumen output better than the other configurations.

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1. INTRODUCTION

With many advantages, white light-emitting diodes (WLEDs) is the new light source that begins to replace conventional lighting solutions in daily lighting, backlighting and automatic lighting [1-3]. Conjugating GaN LED chip with Y₃Al₅O₁₂:Ce³⁺ (YAG:Ce³⁺) yellow phosphor and let the light emitted from these two sources merge and create white light is the most commonly used technique to fabricate WLEDs. Despite having its advantage, the WLEDs produced from this technique is lacking red light component due to the inability of YAG phosphor to generate red component [4]. This leads to inferior CRI and prevents the QDs-phosphor nanocomposites WLEDs from being widely used. It is obvious that adding more red light component is the solution to develop the color rendering ability, therefore, different types of red phosphors were utilized to combine with the yellow phosphor layer. This method has earned recognition for its ability to enhance the effectiveness of WLEDs, however, the extensive emission wavelength of the deep red phosphors, which is above 650 nm, locates outside the cognitive range of the human eyes causing harm to the luminous efficiency (LE) [5-8]. As a result, colloidal quantum dots (QDs) are being used as

an alternative for normal materials to yield chromatic lights, with desirable features such as small emission spectra and large absorption spectra the QDs are predicted to overcome the issues of the prior material [9-12].

Through extensive research, CdSe, InP, CuInS₂, CH₃NH₃PbBr₃, CsPbBr₃ and several other kinds of QDs have gained many positive interests. CdSe QDs, a semiconductor in group II-VI, is regarded as the most effective ones out of all QDs and has high market value for possessing hyper quantum efficiency over 95%, full width at half maximum (FWHM) from 20-30 nm and can cover the whole cognitive range by customizing the size of particles. Recognizing the advantages of CdSe QDs, researchers were focused on using CdSe QDs to manufacture the earliest QDs WLEDs on the market [13, 14]. Even though the usage of CdSe QDs WLEDs are widespread from common lighting application to advanced purpose of backlighting, its potential is limited as a result of containing Cd, a substance that is harmful to human [15]. Many researches have been conducted to eliminate the Cd component as well as finding the optimal replacement, as a result, InP, CuInS₂ and carbon QDs are the promising contenders for this issue. The issues with InP and CuInS₂ QDs is that they are not as effective and have broader FWHM in comparison to the CdSe QDs, on the other hand, carbon QDs shows inefficiency in converting large wavelength color such as red emitted light [16-18]. Therefore, perovskite QDs with luminescence quality and excellent chromatic performance, especially green, such as CH₃NH₃PbBr₃ and CsPbBr₃ are proposed as a new color converting materials [19]. The perovskite QDs with narrower FWHM and the ability to fit in different emission spectra by adjusting the components is an upgrade in comparison to the CdSe QDs, the problem is perovskite QDs is unstable under airy condition, especially those red and blue perovskite QDs [20].

Through consideration of all the materials mentioned above, it seems that CdSe QDs remain as an acceptable option to improve the WLEDs color rendering ability. Regarding the phosphor configuration, most WLEDs with QDs components work well with remote phosphor setup that creates a gap between the QDs layer and the LED chip, this feature allows the WLEDs to achieve better quantum particles, higher efficiency and consistent heating performance. This conclusion is made based on the results of previous studies, which involves studying about the distance between phosphor layers in flat dual-remote phosphor (FDRP) structure and concave dual-remote phosphor structure (CDRP), analyzing distances between phosphor layers in triple-remote phosphor structure, and calculating the impacts of SrBaSiO₄:Eu²⁺ particles in controlling WLEDs green light output with conformal phosphor packages. However, these studies focus on the effect of distance on the lighting efficiency while neglecting other quality indicators and the solutions to improve them. Therefore, an overall assessment of optical performances from single-layer to triple-layer structure is needed.

In this research, we conducted that assessment with the results of each phosphor structure is compared to the others for optical quality evaluation and development study [21, 22]. In fact, the light conversion layer that consists of phosphor layers and QDs usually has issues with the energy transfer efficiency and show inadequate lighting efficiency and thermal performance due to the back-scattering occurs between the phosphor and QDs [23]. To address this problem and improve WLEDs optical and thermal performance, the phosphor layers and the QDs plate are placed separately in a vertical position, this is the vertically packaging structure that is applied in the mixed-type WLEDs [18-21]. The position of the phosphor layer has direct influences on the performance of WLEDs as it shows through the improvement in quantum yield in phosphor compared to QDs when putting the phosphor layer beside the LED chip, furthermore, the effectiveness of the color converting process is also determined by the layer next to the LED chip [24, 25]. The WLED with QDs-on-phosphor configuration perform excellently and shows incredible capability in LE, CRI and controlling thermal performance compared to the mixed-type or phosphor-on-QDs type in both occasions which promote QDs-In-phosphor as the correct choice to create high quality WLEDs. This packaging structure is good at many aspects yet unable to eradicate reabsorption as the red QDs still absorb the green and yellow light from the phosphor layers when they pass through it. Another concern is that LED has a Lambertian intensity distribution, which means the energy transfer efficiency is not consistent and the intensity focus on the center and gradually falling towards the side. These are important issues that need immediate attention, therefore, we renovated the packaging structure and decided to arrange the phosphor layers horizontally as a solution to the problem of reabsorption and inconsistent energy conversion. This is the first time the horizontally layered QDs phosphor nanocomposite ability to improve the lighting features of WLEDs is applied in a structure and achieved positive results. Four distinct types of WLEDs including single-layer phosphor, dual-layer remote phosphor with yellow-red and yellow-green, and triple-layer phosphor configuration were assembled and applied in the experiments, the phosphor fabrication processes, mathematic equations and detailed results are presented respectively throughout the article. The results are informative instruction for the selection of the suitable packaging configuration to fulfill the manufacturing requirements.

2. EXPERIMENT AND SIMULATION DETAILS

2.1. Preparation of phosphor materials

The green phosphor was utilized $\text{LaSiO}_3\text{Cl}:\text{Ce}^{3+},\text{Tb}^{3+}$ to stimulate the green light component in WLEDs leading to increased luminous flux and color uniformity. The red phosphor $\text{Mg}_2\text{TiO}_4:\text{Mn}^{4+}$ with red emission ability is an essential element in enhancing WLEDs color rendering index (CRI) and color quality scale (CQS). The procedures to fabricate these two phosphors are expressed in the following instructions:

The preparation procedure of $\text{LaSiO}_3\text{Cl}:\text{Ce}^{3+},\text{Tb}^{3+}$ compound consists of 7 steps that needed to be applied in the exact order of mixing, drying, firing, re-firing twice, washing, and drying again, to achieve the highest quality phosphor. First of all, mixing the materials thoroughly in water added with NH_3 until forming a uniform slurry. After the first step, leave the materials to dry in airy place and then powderize when they are completely dry. In the first firing process, place the materials in a capped quartz tube filled with N_2 and put it in the furnace at approximately 500°C for 1 hour, after the product cooled down powderize it to obtain the desirable fine powdery state. Re-firing the materials in capped quartz tubes filled with N_2 for an hour but this time at 1200°C before powderizing. Repeat the firing process one last time in open quartz boats filled with CO at 1200°C for one hour and finish by powderizing the product. After the firing processes, washing the mixture in water for several times to remove unwanted residues. Finally, leave the product until dry and store in concealed container. The fabrication process of $\text{Mg}_2\text{TiO}_4:\text{Mn}^{4+}$ also have to be in a specific order to yield the best result as each prior step is related to the subsequent ones. To create $\text{Mg}_2\text{TiO}_4:\text{Mn}^{4+}$, the first step is to blend the starting materials into water or methanol until the mixture reaches a uniform state. Drying the mixture in airy condition before powderize the dried materials. Then proceed to fire the materials in a quartz boat that is left open to let the air in, the temperature should be at about 1300°C and the length of this process is one hour, powderize the product by grinding or milling it once the firing process is done and the product is back to normal temperature. Firing the materials one more time in an open quartz boat but with O_2 at 570°C overnight for 16 hours. The final product is stored in concealed container. Before conducting the optical simulation of $\text{LaSiO}_3\text{Cl}:\text{Ce}^{3+},\text{Tb}^{3+}$ and $\text{Mg}_2\text{TiO}_4:\text{Mn}^{4+}$ particles, the input data such as phosphor concentration, phosphor particles sizes, excitation spectra, absorption spectra, and the emission spectra of the phosphor needed to be properly examined through experiments to ensure authenticity. Among the aforementioned parameters, parameters relating to the spectra are constant numbers while phosphor concentrations and phosphor particle sizes are the unknown values needed to enhance the color quality and luminous flux of WLEDs. Based on the results of prior researches, the diameter of each phosphor particle is fixed at an average of $14.5\ \mu\text{m}$.

2.2. Simulation process

In Figure 1(a) is an image of the physical model simulated to use in the experiments, in which contains 9 blue chip, phosphor layers and dome-like lens. Figure 1(b) contains the detailed measurements of this WLED and Figure 1(c), 1(d), 1(e) and 1(f) shows this remote phosphor WLEDs with different types of phosphor from single-layer phosphor, Dual-layer remote phosphor with YR structure and YG structure to triple-layer phosphor structure. The substrate in the simulation is aluminum nitride and the phosphor is $\text{YAG}:\text{Ce}^{3+}$, the CCTs are 6600 K and 7700 K for packages to ensure accuracy and the experiments are observed through the vertical axis.

The measurement of the remote phosphor layers is fixed at 0.08 mm. To maintain the average color correlated temperature (ACCT), it is crucial that the concentration of $\text{YAG}:\text{Ce}^{3+}$ must adjust accordingly to the changes occur in the concentration of green or red phosphor. At different ACCTs of each phosphor configuration the concentration of $\text{YAG}:\text{Ce}^{3+}$ also varies, this lead to distinct light scattering effects in each WLED resulting in diverse lighting properties. By analyzing the content of Figure 2, it can be concluded that the concentration of the yellow emitting phosphor $\text{YAG}:\text{Ce}^{3+}$ is at the highest level in the Y structure and struggle in YRG structure at all ACCTs. When examining the same ACCT in all remote phosphor structures, the higher the concentration of $\text{YAG}:\text{Ce}^{3+}$ get, the greater the reabsorption losses become causing the luminous flux to decline. Moreover, the high concentration of $\text{YAG}:\text{Ce}^{3+}$ also induce the imbalance between the three colors constituting white light, red, yellow and green, which is detrimental to the chromatic quality of WLEDs. Therefore, keeping the balance between the white light constituent colors as well as limiting the light loss from back-scattering effect are the fundamental requirements in enhancing luminous flux and color quality of WLEDs. To achieve these goals, the red emissive phosphor with red light component can be employed to boost the color rendering index while the green light component from the green phosphor can be useful in managing color uniformity and luminous flux. Possessing both red and green phosphor, the triple remote phosphor stands out as the most beneficial phosphor configuration for improving the lighting performance of WLEDs. However, to fully determine this statement, we need to consider another important parameter of the remote phosphor structure that is the emission spectrum. As can be seen from the graphs of Figure 3, the differences between the emission

spectra are apparent. Regarding the width of the emission spectra, the Y structure has the lowest intensity at all ACCTs when compared to others structures, this confirms that the luminous flux obtains from this structure is the smallest while in YRG structure, the emission spectrum intensity in the wavelength ranging from 380–780 nm is the highest. When comparing the YG and the YR structure, we can see that the luminous flux of YG is higher than that of YR owing to YG structure higher spectrum intensity in the wavelength band from 400–500 nm. On the contrary, the spectrum intensity of YG is lower than YR in the wavelength band from 650–750 nm which proving that YR has better color rendering index than YG. These are important information in choosing the suitable structure for WLEDs, however, the results in part 3 also need consideration before concluding.

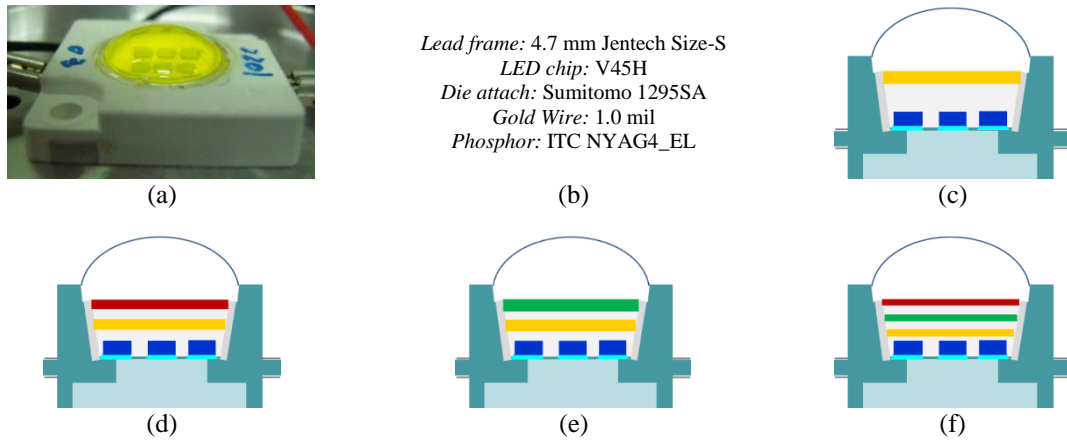


Figure 1. Illustration of multi-layer phosphor structures of white LEDs: (a) the actual MCW-LEDs and (b) its parameters; (c) single-layer phosphor, dual-layer remote phosphor with YR (d) and YG (e), and (f) triple-layer phosphor

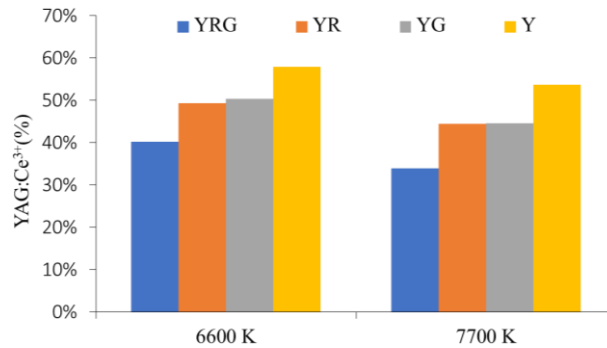


Figure 2. The concentration of yellow-emitting YAG:Ce³⁺ phosphor correlating to each remote phosphor structures at different ACCTs

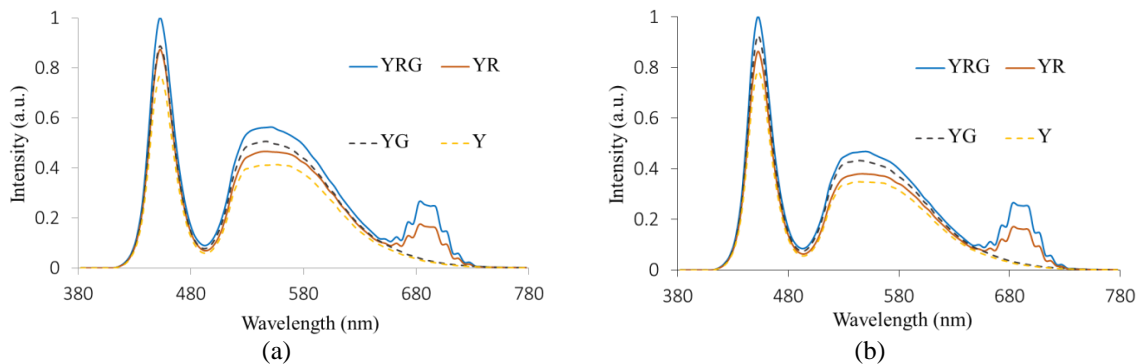


Figure 3. Emission spectra of phosphor configurations: (a) 6600 K and (b) 7700 K

As can be seen from the graphs, the differences between the emission spectra are apparent. Regarding the width of the emission spectra, the Y structure has the lowest intensity at all ACCTs when compared to others structures, this confirms that the luminous flux obtains from this structure is the smallest while in YRG structure, the emission spectrum intensity in the wavelength ranging from 380-780 nm is the highest. When comparing the YG and the YR structure, we can see that the luminous flux of YG is higher than that of YR owing to YG structure higher spectrum intensity in the wavelength band from 400-500 nm. On the contrary, the spectrum intensity of YG is lower than YR in the wavelength band from 650-750 nm which proving that YR has better color rendering index than YG. These are important information in choosing the suitable structure for WLEDs, however, the results in part 3 also need consideration before concluding.

3. RESULTS AND ANALYSIS

Figure 4 demonstrated the overall review of the CRI between the remote phosphor structures. From this graph, the YR is the most favorable structure for the growth of CRI which result in the highest CRI at all ACCT. The graphs also show another important finding relating to the improvement of CRI in remote phosphor layer that is the CRI increases with ACCT and reach the highest index at 8500 K. Even though controlling the color rendering ability of WLEDs at the ACCT above 7000 K is a difficult task, the YR structure is still able to benefit the CRI by adding red component from the red phosphor $\text{Sr}_w\text{F}_x\text{B}_y\text{O}_z:\text{Eu}^{2+},\text{Sm}^{2+}$. The YRG is placed second regarding the achievable CRI and CRI in YG is the lowest among all remote phosphor structures. From this result, it is confirmed that YR is the optimal structure for mass production if the goal is to achieve the highest CRI possible. However, CRI can only assess a few aspects of WLEDs optical characteristics while there are broader and harder to obtain parameter such as CQS that can fully describe the performance of WLED. By assembling three important factors including CRI, viewer preference, and color coordinate, the color quality scale (CQS) has become a research goal for many studies in recent years and is frequently used as a fundamental index to evaluate the chromatic performance of WLEDs as the higher the CQS the better the color quality. The CQSs of the remote phosphor utilized in this research are measured and expressed in Figure 5. Due to the balance between the three colors red, yellow and green provided by the three phosphor layers, the CQS achieved in YRG is the highest among the 4 remote phosphor structures. The lowest CQS belongs to structure Y, despite having an advantage in luminous flux, the color quality of the Y structure is unable to progress due to the lack of red and green lights, the two important ingredients for balancing the primary colors and improving the color quality. The color quality of Y structure might not be ideal, however, the strong points that this structure has over other options are its easy-to-handle producing process and small manufacturing cost.

As shown in Figure 5, the YRG structure is the most suitable configuration if the target of the manufacturers is to produce WLED with the best color quality. However, to determine whether this structure going to have a negative effect on the luminescence efficiency or not, a comparison between the luminous flux of single-layer structure and dual-layer structure is needed. Figure 6 illustrate the luminescence efficiency (LE) between the remote phosphor structures, which shows that the YRG structure excels at all ACCTs while Y structure is at the bottom. This confirmed that along with the high color quality the YRG structure is also able to yield the largest luminous flux. This can be explained with the appearance of both red and green phosphor in the YRG structure forcing a decrease in the concentration of yellow phosphor $\text{YAG}:\text{Ce}^{3+}$ and as the same time reduce the light loss due to re-absorption. With the concentration of $\text{YAG}:\text{Ce}^{3+}$ decreases, the energy conversion efficiency increases because the blue light from the chips can now easily pass through the yellow phosphor layer and reach other phosphor layers. As a result, the intensity of the emission spectrum in the YRG structure is the highest compared to other structures in the white light wavelength and the luminous flux of this structure is also the highest accordingly. Moreover, in the wavelength band from 500 nm – 600nm, the additional green light from the green phosphor layer $\text{SrBaSiO}_4:\text{Eu}^{2+}$ enhance the emission spectrum of YG structure making it higher than YR and Y structure, therefore, the second-highest luminous flux belongs to the YG structure. According to the results above, the WLEDs with YRG will contain good optical quality in both CQS and LE, this proves that YRG is an outstanding structure to choose when manufacturing WLEDs. Beside other features, color uniformity is also an essential target when discussing color quality, however, the two most commonly used method to improve color uniformity are employing scattered-enhancing particles and using the conformal phosphor, which can severely damage the luminous flux of WLEDs. Therefore, using the red phosphor $\text{Sr}_w\text{F}_x\text{B}_y\text{O}_z:\text{Eu}^{2+},\text{Sm}^{2+}$, green phosphor $\text{SrBaSiO}_4:\text{Eu}^{2+}$ and the remote phosphor structure is the solution for this problem as the phosphor layers are able to boost the scattering properties and improved the white light output quality while the remote structure layer is very efficient in enhancing the luminous flux by limiting the amount of light reflected to the LED chip. Figure 7 expressed the color deviation between the structures. In this figure, the structure with the smallest color deviation index has the highest color uniformity, therefore,

the color uniformity of the YRG structure is the best correlating to the lowest color deviation illustrated in Figure 7. This is the result of the scattering events occur inside the LED package before forming white light, the color uniformity of WLEDs increases with the amount of scattering events occurred. Although having too many scattering events also impair the luminous flux, this drawback is compensated with the reduced back-scattering effect. As a result, the YRG still has the highest color uniformity while maintaining the best luminous flux. The Y structure, on the other, is the structure with the highest color deviation measurement at all ACCTs.

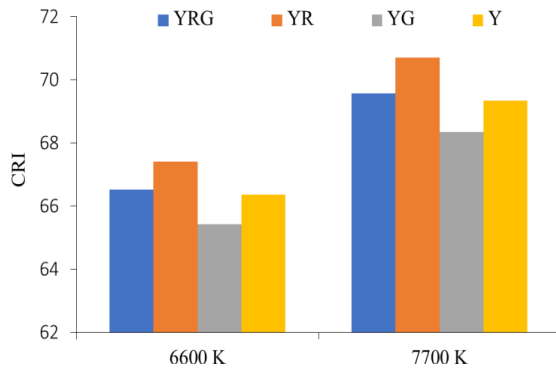


Figure 4. Color rendering indexes of phosphor configurations corresponding to ACCTs

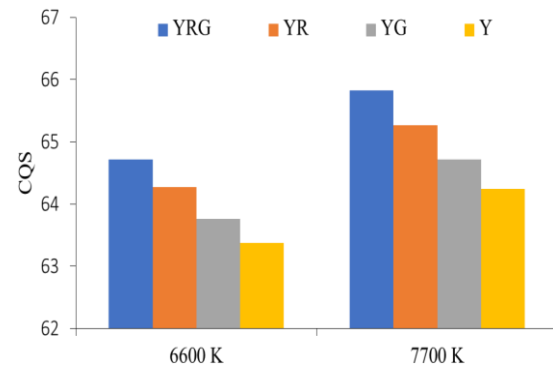


Figure 5. Color quality scale of phosphor configurations corresponding to ACCTs

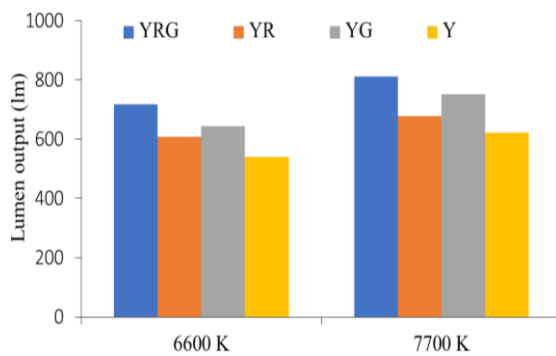


Figure 6. Luminous efficacy of phosphor configurations corresponding to ACCTs

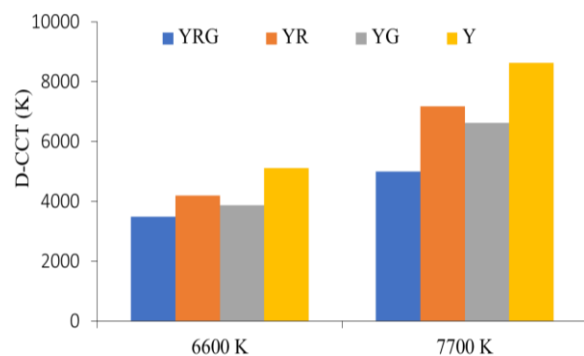


Figure 7. Correlated color temperature deviation (D-CCT) of remote phosphor configurations corresponding to ACCTs

4. CONCLUSION

In conclusion, the YRG offers a balance between the three primary colors, reduced back-scattering effect and low color deviation that results in highest color quality, color uniformity, and luminous flux. These results confirm that YRG is the optimal WLED structure for the performance enhancing ability and superior values in every optical properties compared to other structures, therefore, this is an important reference for manufacturers while choosing set up for their product. The YG and YR structures have the second-highest optical indices which vary depend on the type of optical property, in particular, the YG structure has better luminous flux and color uniformity than the YR structure because the extra green component from the green phosphor $\text{SrBaSiO}_4:\text{Eu}^{2+}$ boosts the luminescence efficiency and color uniformity, while the CRI and CQS in the YR structure is better than YG structure because they are enhanced by the red light component. The Y structure, although not excelling in terms of quality, is still a very possible choice because of the low manufacturing cost, simple producing process, and high luminous efficiency. These results are from thorough experiments that compared the optical properties between the most frequently used structures, Y, YG, YR, and YRG, and then verified using the Mie theory and the Lambert-Beer law, therefore, the relevancy and reliability is high and can be applied in practical situations. The article does not only provide information of the best structure for WLED but also demonstrates the most suitable ones for specific

occasions, therefore, these are valuable references that can support manufacturers in choosing the appropriate structure for the production of WLEDs with any particular demand while ensuring the quality.

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