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Green-emitting $Gd_2O_2S:Tb^{3+}$ and red-emitting $Y_3Al_5O_{12}:Cr^{3+}$ phosphors: a suitable selection for enhancing color quality of remote phosphor structure

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ABSTRACT

This article demonstrates green-emitting phosphor $Gd_2O_2S:Tb^{3+}$ and red-emitting phosphor $Y_3Al_5O_{12}:Cr^{3+}$ application in the triple-layer phosphor WLED to enhance optical performance. The arrangement of phosphor layers in the WLED is red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ on top, green phosphor $Gd_2O_2S:Tb^{3+}$ in the middle, and yellow phosphor $YAG:Ce^{3+}$ at the bottom. The principal to utilize these phosphor materials is the exploitation of additional red light component and green light component from the green and red phosphor to enrich the color rendering index (CRI) and luminous efficacy. The influences of green phosphor and red phosphor are also estimated with a new quality indicator, the color quality index (CQS). The results show red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ enable CRI when its concentration increases while green phosphor exhibits a contrast reaction. Regarding the CQS, the optimal red phosphor concentration for CQS is from 10% to 14%, disregard the concentration in green phosphor. The improvement that applying these two phosphor materials brought comes from limiting the light loss from back-scattering and strengthen chromatic performance through addition red and green light components. These findings can support manufacturers in adapting to modern lighting requirements by improving CRI, CQS and particularly luminous efficiency to more than 40%.

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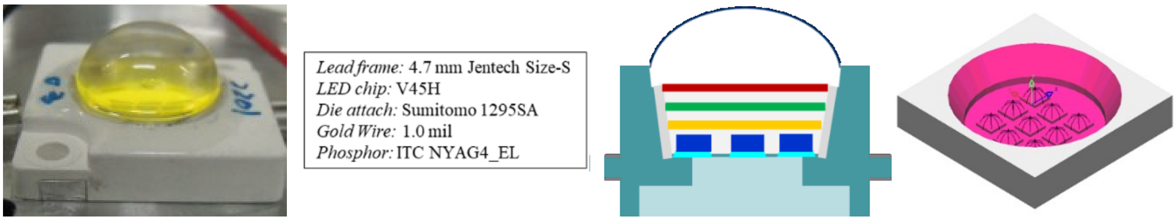
1. INTRODUCTION

The modern lighting solution, white light-emitting diodes (WLEDs), with many remarkable features being compactness, power efficiency, cheap manufacturing cost, and color stability, are receiving more attention and becoming an irreplaceable light source [1-4]. The white light is created from merging the colors from different sources such as blue light that discharged out of blue chip and then blended with the light from the yellow phosphor which gives out yellow light to form the white light [5]. WLEDs are a promising light source in solid-state lighting, however, their inferior luminous flux is an obstacle that prevents them from being widely used. The white light is usually the product of a freely dispersed coating process in which a mixture of transparent encapsulated resin and phosphor powder are sprayed directly onto the phosphor package. The advantages of this method are the optimal phosphor thickness and minimal producing expenses, although the products created cannot meet the standards of high-quality WLEDs [6-9]. As a result, the conformal coating method with uniformly color distribution that can obtain the angular homogeneity of correlated color temperature (CCT) is employed as a substitute [10]. The only problem with the conformal phosphor structure is the damaged luminous efficiency due to the backscattering effect. Many solutions were suggested in previous research to limit the light loss such as detach the chip from the phosphor layer of the remote phosphor structure [11-13]. Employing the internal reflection structure in which a polymer semispherical lens is coated with phosphor on the inside to boost internal light extraction [14]. Moreover, empty cavities are put on the structure to reflect light downward and improve luminous efficiency [15]. Although the package arrangement is usually mentioned as the main factor that decides the lumen output, however, the concentration of each phosphor layer within the WLED model also affects the quality of luminous flux.

The luminous efficiency of WLEDs, especially those at lower CCTs, is affected by the backscattering light loss from the increased phosphor concentrations [16-18]. To address this issue, the most apparent solution is to boost the emitted blue and yellow light while limiting the light loss from the back-scattering process. The triple-layer phosphor WLEDs with color temperature of 6000 K are the subjects for simulations in this research. The arrangement for the phosphor layers in this triple-layer remote phosphor structure is red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ at the highest position right under the protective glass hemisphere, yellow phosphor YAG:Ce^{3+} occupied the lowest position among the phosphor layers above the blue chips, and green phosphor $Gd_2O_2S:Tb^{3+}$ placed between the red and yellow phosphor layers. The inclusion of the green phosphor $Gd_2O_2S:Tb^{3+}$ is to extract the green light component from this phosphor onto the light emission process of WLED which can improve the luminous flux. On the other hand, the red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ which possesses the red light component is expected to benefit the color quality of WLEDs. The results of this research confirm that with the balance between the three primary colors, the WLEDs can produce the highest color quality while causing minimal harm to the luminous flux.

2. COMPUTATIONAL SIMULATION

The experiments employed in this research are to test the effectiveness of green phosphor $Gd_2O_2S:Tb^{3+}$ in improving the luminous flux and red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ in enhancing CRI and CQS. The Light Tools 8.1.0 is the primary tool for the construction of the simulated version of WLEDs which has remote phosphor structure at the average correlated color temperature of 6000K. This simulation program is a commercial software that operate using the Monte Carlo ray-tracing method [19-22]. In Figure 1 (a) is the physical model of the WLEDs in charge of illustrating the optical properties of a remote packaging WLEDs. This physical model is a reflector with the measurement of 8 mm in the bottom, 2.07 mm in height and 9.85 mm in top surface. The reflector consists of 0.08mm phosphor layer above 9 LED chips. The LED square chips are placed in dent embedded on the reflector, the size of each chip is 1.14 mm base and 0.15 mm height and the radiant flux at the wavelength of 455 nm is 1.16 W. The concentrations of phosphor particles vary from 2% to 24% but the ACCTs are stabilized by adjusting the yellow phosphor YAG:Ce^{3+} wt. The technical indices of the components inside the WLED can be observed in Figure 1 (b). The cross-section in Figure 1 (c) demonstrates the arrangement of components inside the WLED model. Figure 1 (d) is the results of simulated WLED.


Lead frame: 4.7 mm Jentech Size-S
LED chip: V45H
Die attach: Sumitomo 1295SA
Gold Wire: 1.0 mil
Phosphor: ITC NYAG4_EL

(a) (b) (c) (d)

Figure 1. (a) The physical model applied in experiments, (b) The technical parameters, (c) The cross-section of triple-layer remote phosphor configuration, (d) The simulation of WLEDs

3. RESULTS AND DISCUSSION

The influences of red and green phosphor on the CRI are expressed in Figure 2 with the concentration ranges from 2% to 20%. CRI gradually rises with the increasing concentration of red phosphor and reach the highest index at 20% concentration. On the contrary, the CRI in WLED only go down when there is green phosphor, which suggest that $Gd_2O_2S:Tb^{3+}$ is not suitable to apply in WLED with high CRI demand, especially when the CRI continuously degrade at any average correlated color temperature (ACCT) as the green phosphor concentration increases regardless of changes in red phosphor. From the result of Figure 2, it is obvious that the red light component from the red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ in WLEDs is crucial in improving the CRI. This is due to the green light become the dominant color when the concentration of green phosphor $Gd_2O_2S:Tb^{3+}$ increase, which is detrimental for the development of CRI. The increased amount of green phosphor concentration also suppresses the red light converted. According to the TRP configuration, the light will reach the green phosphor layer before getting to the red phosphor due to the set up that places $Gd_2O_2S:Tb^{3+}$ phosphor closer to the light source than $Y_3Al_5O_{12}:Cr^{3+}$ phosphor. Therefore, choosing the minimal amount for green phosphor concentration is advisable if the target is to improve the CRI. However, CRI is only one of the criteria to evaluate the color quality that can show the ability of the human eyes to reflect true color when the light is shining in the object. Besides, the truthfulness of the color, the preference of the viewer and color coordinates are the aspects that CRI cannot assess. Therefore, CQS with the ability to measure three elements: CRI, viewer comfort, and white light color coordinates is a better alternative. When comparing the CRI and CQS, the CQS stands out as the more important priority and a harder target to reach. So to solve the question of what can influence and how to improve CQS, we can observe Figure 3 where all changes in CQS are expressed in detail. Overall, the CQS also benefits from the appearance of $Y_3Al_5O_{12}:Cr^{3+}$ phosphor, especially when the concentration of this phosphor extend. The difference is the variation of green phosphor $Gd_2O_2S:Tb^{3+}$ cause less effect to CQS than to CRI. Figure 3 shows that not only the red phosphor affects the CQS but also the green phosphor, which demonstrates the principle of improving CQS, that is to balance the three colors: red, yellow and green. Therefore, in cases that red and green phosphor both increase in concentration level, the suitable adjustment for yellow phosphor is decrease so the ACCT remain the same.

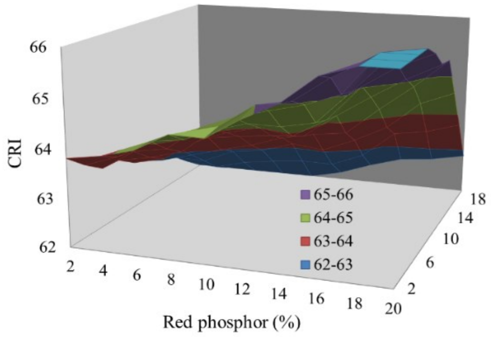


Figure 2. CRI of TRP as a function of red $Y_3Al_5O_{12}:Cr^{3+}$ phosphor và green $Gd_2O_2S:Tb^{3+}$ phosphor

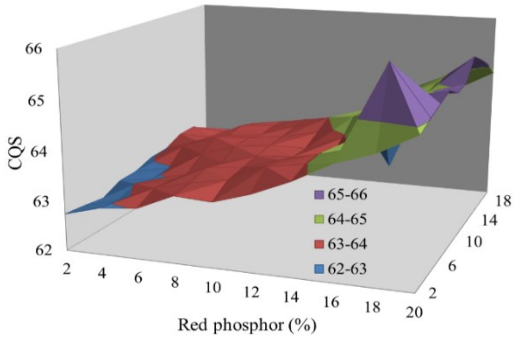


Figure 3. CQS of TRP as a function of red $Y_3Al_5O_{12}:Cr^{3+}$ phosphor và green $Gd_2O_2S:Tb^{3+}$ phosphor

Reducing the concentration of yellow phosphor means the amount of yellow light decline, which can lead to two positive outcomes. First, limiting the amount of light scattered back to the LED hence enhances the luminous flux. Second, lowering the amount of yellow in WLEDs and replacing it with red and green light components, which is good for the balance between these three colors and CQS. When the concentration of green phosphor $Gd_2O_2S:Tb^{3+}$ is rising from 2% to 10% the CQS also rise, the CQS reaches highest indexes when green phosphor concentration is from 10% to 14% and only begin to decrease after that. When the concentration of green phosphor is at low level from 2% to 10%, the CQS cannot achieve the highest index because the amount of yellow light is still dominant, therefore, the back-scattering effect can affect the light energy converted. The CQS in the WLEDs with green phosphor concentration from 10% to 14% only manage to reach the highest point by having an adequate amount of green light component to neutralize the yellow light. However, should the concentration of $Gd_2O_2S:Tb^{3+}$ beyond the 14% mark, it will become abundant and cause an imbalance between the three basic colors: green, red and yellow. Therefore, keeping the concentration of green phosphor at an appropriate level is crucial, as the excessive green light is detrimental to the CQS.

Managing the color quality of the remote phosphor structures is much more complicated than the conformal structure or the in-cup phosphor structure and it is especially harder when the WLEDs is at high ACCTs. However, the results on the CQS of TRP structure show otherwise, the higher the ACCTs the better the CQS in TRP. Moreover, the TRP structure negates the back-scattering effect and boosts the light scattering process occurs inside the WLEDs. This scattering enhancement improves the merging process of light to fabricate better quality white light. The only matter with this increase in scattering is how it affects the converted light energy. This part introduces and illustrates the numerical model of the transmitted blue light and converted yellow light in the dual-layer phosphor structure, from which changes can be made to create immense enhancement to LED efficiency. The blue light transmission and the conversion of yellow light in single layer remote phosphor package with the phosphor layer thickness defined as 2h are computed as follows [23-25]:

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

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

The transmitted blue light and converted yellow light in WLED with dual-layer remote phosphor package that has the phosphor layer thickness of *h* are calculated with the following equation;

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

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

The subscript "1" and "2" depict single layer and double-layer remote phosphor package. β presents the conversion coefficient from blue light to yellow light. γ represent the reflection coefficient of the yellow light. The light intensity from the blue LED are the intensities of blue light (PB) and yellow light (PY), demonstrated by PB_0. α_B; α_Y which are parameters depicting the portion of the blue and yellow lights energy loss in their diffusion process in the phosphor layer.

The pc-LEDs lighting efficiency is significantly improved with the dual-layer phosphor structure compared to the results acquired in the single layer structure:

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

The scattering of phosphor particles was studied using the Mie-theory. Moreover, by applying the Mie theory, the scattering cross section C_{sca} for spherical particles can be calculated with the following expression. The Lambert-Beer law can be used to calculate the transmitted light power.

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

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

Through the application of (5), it is obvious that the luminous flux of WLEDs is better if the number of phosphor layers increases. The Figure 4, which expresses the luminous flux measured from the TRP, is the explanation for this statement, the red and green phosphor concentration increase corresponding to the expansion of the luminous flux, therefore, forcing the concentration of yellow phosphor to decline to stabilize the ACCT. The vital point is lowering the concentration of yellow phosphor to reduce the light loss in the back-scattering process. Moreover, according to the Lambert-Beer law in (6), as the concentration of yellow phosphor decreases the light emission energy grows. Therefore, the increase in red and green phosphor concentration will support the extension of luminous flux. However, the exceeding amount of red and green light components in WLEDs might cause a disproportion between the three colors, thus leave a negative impact on the CQS. As the content of Figure 5 suggests, the green phosphor $Gd_2O_2S:Tb^{3+}$ that increase the green light component within WLED reduces the backscattering which is favorable for the lighting efficacy (LE) and can enhanced it up to 40% regardless of the red phosphor

$Y_3Al_5O_{12}:Cr^{3+}$ concentration. So if the target is to achieve high index in CQS and LE, the concentration of $Gd_2O_2S:Tb^{3+}$ to choose should be from 10% to 14% while having the concentration of $Y_3Al_5O_{12}:Cr^{3+}$ at 20%. LE in WLEDs with the average correlated color temperature of 6000K increase slightly with the concentration of $Y_3Al_5O_{12}:Cr^{3+}$. When the concentration of $Y_3Al_5O_{12}:Cr^{3+}$ is in the range from 2% to 14%, LE almost remain unchanged and only start to deteriorate when the concentration of green phosphor rise to 20%. These results are useful information that can be utilized by the manufacturers to choose the right amount of phosphor concentration for the desired goal.

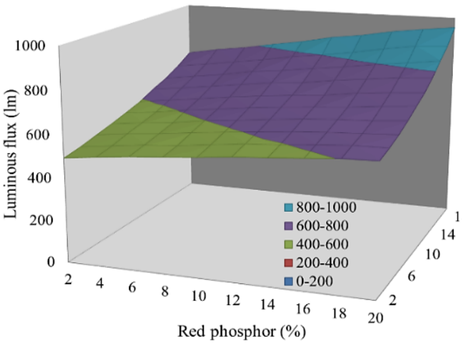


Figure 4. Luminous flux of TRP as a function of red $Y_3Al_5O_{12}:Cr^{3+}$ phosphor và green $Gd_2O_2S:Tb^{3+}$ phosphor

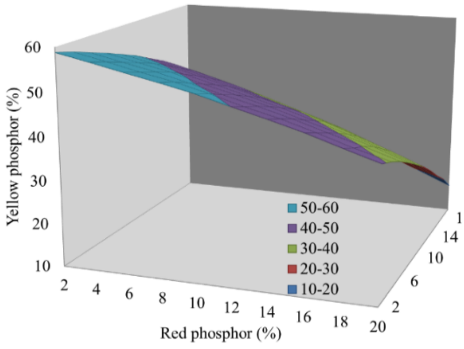


Figure 5. Luminous flux of TRP as a function of red $Y_3Al_5O_{12}:Cr^{3+}$ phosphor and green $Gd_2O_2S:Tb^{3+}$ phosphor

4. CONCLUSION

Proposing the application of TRP configuration together with the green phosphor $Gd_2O_2S:Tb^{3+}$, and red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ to improve CRI, CQS and LE of WLEDs. The TRP was proven efficient in both enhancing color quality and LE, which is a useful result that has not been discovered before. To achieve this result, the balance between yellow, red and green lights is the prerequisite requirement which is obtainable by adjusting the concentration of $Gd_2O_2S:Tb^{3+}$ and $Y_3Al_5O_{12}:Cr^{3+}$. Managing the concentration of the green phosphor is controlling the green light component in WLED thus lead to better luminous flux. The red phosphor $Y_3Al_5O_{12}:Cr^{3+}$ with the additional red light component, on the other hand, is a suitable material to induce the growth in CRI. Moreover, employing multiple layers of phosphor is more beneficial for the luminous flux than using a single one. From the results of this research, reducing the back-scattering effect from the yellow phosphor $YAG:Ce^{3+}$ and maintaining the balance of the three colors yellow, red and green are the vital point to get the optimal color quality and highest luminous flux.

REFERENCES

[1] J. P. You, N. T. Tran, and F. G. Shi. “Light extraction enhanced white light-emitting diodes with multi-layered phosphor configuration,” *Optics Express*, vol. 18, no. 5, pp. 5055-5060, 2010.
[2] N. D. Q. Anh and H. Y. Lee. “Improving the angular color uniformity and the lumen output for white multi-chip LED lamps by green $Ce_{0.67}Tb_{0.33}MgAl_{11}O_{19}$:Ce,Tb phosphor,” *J. of the Chin. Ins. of Engineers,* vol. 39, no. 7, pp. 871-875, 2016.
[3] S. J. Myoung, H. C. Yong, W. Shulu, G. L. Tae, and S. Y. Jae, “Material properties of the Ce^{3+}-doped garnet phosphor for a white LED application,” *Journal of Information Display*, vol. 17, no. 3, pp. 117-123, 2016.
[4] M. R. Katja, K. G. Marta, B. Grega, and B. K. Matej, “White LED compared with other light sources: Age-dependent photobiological effects and parameters for evaluation,” *Int. J. of Occup. Saf. and Erg.*, vol. 21, no. 3, pp. 391-398, 2015.
[5] N. D. Q. Anh, P. X. Le, and H. Y. Lee. “Selection of a remote phosphor configuration to enhance the color quality of white LEDs,” *Current Optics and Photonics*, vol. 3, no. 1, pp. 78-85, 2019.
[6] Z. T. Li, H. Y. Wang, B. H. Yu, X. R. Ding, and Y. Tang, “High-efficiency LED COB device combined diced V-shaped pattern and remote phosphor,” *Chinese Optics Letters*, vol. 15, no. 4, pp. 42301-42304 , 2017.
[7] N. Q. Li, Y. Zhang, Y. W. Quan, L. Li, S. H. Ye, Q. L. Fan, and W. Huang. “High-efficiency solution-processed WOLEDs with very high color rendering index based on a macrospirocyclic oligomer matrix host,” *Optical Materials Express*, vol. 8, no. 10, pp. 3208-3219, 2018.
[8] Y. Y. Li, J. J. Chen, H. H. Feng, H. Chen, and Q. M. Wang, “Rugate filters used in slit-lamp delivery to improve color rendering of illumination for retinal photocoagulation,” *Applied Optics*, vol. 53, no. 15, pp. 3361-3369, 2014.
[9] K. A. G. Smet and P. Hanselaer, “Impact of cross-regional differences on color rendition evaluation of white light sources,” *Optics Express,* vol. 23, no. 23, pp. 30216-30226, 2015.

[10] J. H. Oh, S. J. Yang, Y. G. Sung, and Y. R. Do, "Excellent color rendering indexes of multi-package white LEDs," *Optics Express*, vol. 20, no. 18, pp. 20276-20285, 2012.

[11] H. K. Park, J. H. Oh, and Y. R. Do, "Toward scatter-free phosphors in white phosphor-converted light-emitting diodes," *Optics Express*, vol. 20, no. 9, pp. 10218-10228, 2012.

[12] J. H. Oh, H. J. Kang, M. J. Ko, and Y. R. Do, "Analysis of wide color gamut of green/red bilayered freestanding phosphor film-capped white LEDs for LCD backlight," *Optics Express*, vol. 23, no. 15, pp. A791-A804, 2015.

[13] G. X. He and H. F. Yan, "Optimal spectra of the phosphor-coated white LEDs with excellent color rendering property and high luminous efficacy of radiation," *Optics Express*, vol. 19, no. 3, pp. 2519-2529, 2011.

[14] C. Hu, Y. Shi, X. Q. Feng, and Y. B. Pan, "YAG:Ce/(Gd,Y)AG:Ce dual-layered composite structure ceramic phosphors designed for bright white light-emitting diodes with various CCT," *Optics Express,* vol. 23, no. 14, pp. 18243-18255, 2015.

[15] O. H. Kwon, J. S. Kim, J. W. Jang, and Y. S. Cho, "Simple prismatic patterning approach for nearly room-temperature processed planar remote phosphor layers for enhanced white luminescence efficiency," *Optical Materials Express,* vol. 8, no. 10, pp. 3230-3237, 2018.

[16] Y. H. Won, H. S. Jang, K. W. Cho, Y. S. Song, D. Y. Jeon, and H. K. Kwon, "Effect of phosphor geometry on the luminous efficiency of high-power white light-emitting diodes with excellent color rendering property," *Optics Letters*, vol. 34, no. 1, pp. 1-3, 2009.

[17] A. I. Alhassan, R. M. Farrell, B. S. Saifaddin, A. Mughal, F. Wu, S. P. DenBaars, S. Nakamura, and J. S. Speck, "High luminous efficacy green light-emitting diodes with AlGaN cap layer," *Optics Express,* vol. 24, no. 16, pp. 17868-17873, 2016.

[18] F. Z. Zhang, H. S. Xu, and Z. Wang, "Optimizing spectral compositions of multichannel LED light sources by IES color fidelity index and luminous efficacy of radiation," *Applied Optics*, vol. 56, no. 7, pp. 1962-1971, 2017.

[19] T. Erdem, S. Nizamoglu, and H. V. Demir, "Computational study of power conversion and luminous efficiency performance for semiconductor quantum dot nanophosphors on light-emitting diodes," *Optics Express*, vol. 20, no. 3, pp. 3275-3295, 2012.

[20] V. A. Bol'shukhin, V. S. Ilyasov, N. P. Soshchin, and V. N. Ulasyuk, "Illuminators based on composite LEDs for multifunctional high-luminance active-matrix liquid-crystal displays," *Journal of Optical Technology,* vol. 78, no. 7, pp. 444-448, 2011.

[21] W. J. Gao, K. Ding, G. X. He, and P. Zhong, "Color temperature tunable phosphor-coated white LEDs with excellent photometric and colorimetric performances," *Applied Optics*, vol. 57, no. 31, pp. 9322-9327, 2018.

[22] J. H. Oh, J. R. Oh, H. K. Park, Y. G. Sung, and Y. R. Do, "New paradigm of multi-chip white LEDs: Combination of an InGaN blue LED and full down-converted phosphor-converted LEDs," *Optics Express*, vol. 19, no. S-3, pp. A270-A279, 2011.

[23] Y. Ma, M. Wang, J. Sun, R. Hu, and X. B. Luo, "Phosphor modeling based on fluorescent radiative transfer equation," *Optics Express*, vol. 26, no. 13, pp. 16442-16455, 2018.

[24] Z. Wen, C. Y. Ma, C. Zhao, F. Tang, Z. Cao, Z. Q. Cao, X. Y. Yuan, and Y. G. Cao, "Fabrication and optical properties of Pr^{3+}-doped Ba (Sn, Zr, Mg, Ta)O_3 transparent ceramic phosphor," *Optics Letters*, vol. 43, no. 11, pp. 2438-2441, 2018.

[25] X. L. Song, Z. Liu, Y. J. Xiang, and K. Aydin, "Biaxial hyperbolic metamaterials using anisotropic few-layer black phosphorus," *Optics Express*, vol. 26, no. 5, pp. 5469-5477, 2018.