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Enhancing light sources color homogeneity in high-power phosphor-based white LED using ZnO particles

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ABSTRACT

Color uniformity is one of the essentials for the on-going development of WLED. To achieve a high color uniformity index, increasing the scattering events within the phosphor layers was reported to be the most efficient method and in this article, ZnO is the chosen material to apply in this method. After analyzing the scattering properties through the scattering cross-section $C_{sca}(D,\lambda)$, scattering coefficient $\mu_{sca}(\lambda)$ and scattering phase function $\rho(\theta,\lambda)$, the which outcomes comfirm that ZnO can enhance the scattered light in the phosphor layers. Moreover, the findings from the study of ZnO concentration from 2% to 26% suggest that color uniformity also depends on the fluctuation of ZnO concentration, therefore, to control color uniformity the focus should be implied on both size and concentration of ZnO. The experimental results from this research show that the luminous flux of WLED is at the peak if the concentration of ZnO is at 6%, and when the concentration of ZnO is at 18% and has 100 nm particles size, the ΔCCT reaches the lowest level. The final choice should be based on the desired characteristic of WLEDs, however, if the WLED need to excel in both luminous flux and ΔCCT then 6% ZnO concentration with particles size from 100 nm-300 nm is the optimal choice.

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1. INTRODUCTION

The luminescence industry, as a constantly developing field, always require innovative lighting sources with higher proficiency to enhance the applications and replace the obsolete ones. To comply with the demands, the lighting device using diodes to emit light (LEDs) was introduced and used widely for the excellent properties such as high durability, energy efficiency, low production costs, and environmental friendliness, is predicted to overcome conventional options and become a trendy lighting solution [1-4]. The most commonly used method to fabricate the white light is merging the chromatic lights of LED chips and phosphor phosphor material in the lighting device together (YAG:Ce) [5, 6]. Even though applying this freely dispersed coating method can give precise control on the thickness of the phosphor layers and lower the manufacturing cost, however, the output light has low chromatic quality that results in the yellow ring effect causing irritation to the viewer's eyes and reducing the quality of the devices is the drawback [7-9]. As a result, many changes in different parts such as the structure configuration [10], reflector [11], exterior phosphor layer [12], and packaging order [13, 14], were made to address the problems. These changes can improve the CCT uniformity but they are not ideal due to the expenses to imply them on industrial manufacturing is too high. Therefore, extensive researches are conducted to find a method that can tune the CCT uniformity and easy to create but has a reasonable cost. The diffuser-load encapsulation method, out of all the reported results, is the most effective one in reaching the goals mentioned above, therefore, some LED that apply the diffuser method with metal oxide diffusers materials, such as TiO_2, ZrO_2 and SiO_2 are already available to improve the angular CCT uniformity [15-18]. In one research, Chen et al. [15] simulated a remote phosphor structure with a 300nm ZrO_2 particle as diffusers in between -70° to 70° and the results show that it can lower the deviated color temperature by 560 K, while in another Lee et al. [18] introduced 320 nm TiO_2 diffusers to cover the phosphor materials and device component, which results in lower angular CCT deviations as diffuser concentration increases. These researches demonstrate the influences of single diffusing particle or the variation of particle concentration on the WLEDs performance but also shows the lack of information on the effects of the nanoparticles' shapes in WLEDs [5, 19, 20]. Therefore, without the assist of thorough analysis and in-depth instruction on the technique, the optimal particle setting for the enhancement of CCT has yet to be decided.

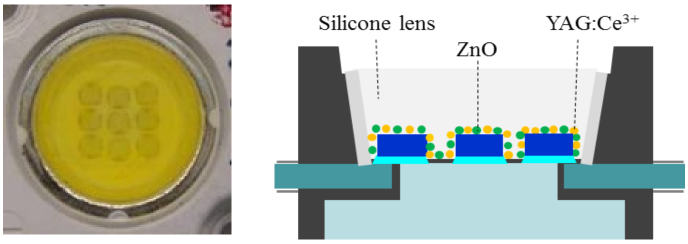
The ZnO nanostructures containing the positive traits of adjustable morphology, easy and cost-saving fabricating procedures are being recognized and used on many occasions [21]. The vital point in using the ZnO structure is to guarantee the requirements for the desired structure are met including temperature, time, and precursor concentration and all others. The ZnO nanostructures with the refraction index of 2 fits between the indices of 2.5 and 1 of GaN and Air respectively, therefore, suitable to be a layer of gradient refractive index. Another usage of the ZnO nanostructures that has been verified by many experimental results is enhancing the light extraction efficiency when applied to LEDs with GaN semiconductor in certain nanostructures. A hybrid ZnO nanostructure with microscopic cylinders and nanorods upon a light-emitting chip can enhance the light-emitting ability of GaN-based LEDs [22]. The result from these GaN blue LEDs with ZnO nanoparticles implanted to planar indium tin oxide (ITO) film shows a stimulation in light emission. A method of using the modification of ZnO nanorods to adjust the light radiation pattern in GaN-based LEDs was reported in reference [23]. These papers all demonstrate the effectiveness of ZnO nanostructures in the aspect of improving the light extraction of LEDs and leave out other potential aspects that can also contribute to the enhancement of WLEDs. Until now, the effects of ZnO at different sizes on lighting characteristics such as correlated color temperature and the consistency of light intensity in light-emitting devices remain undiscovered. So to further the study on WLEDs, future researches should target the identification of ZnO nanostructures advantages in improving the CCT and light intensity uniformity of WLEDs. In this article, the size and concentration of ZnO are analyzed to serve as a guideline for choosing the light output and color uniformity. By calculating the scattering events in the phosphor layers, the impacts of the size and concentration are revealed, thus, assert the application values of ZnO.

2. RESEARCH METHOD

2.1. Physical and optoelectronic characterization

In Figure 1 (a) is the simulated WLED with ZnO that is used in the research and Figure 1 (b) is the cross-section of a WLED with ZnO fused in the yellow phosphor. The WLED consist of GaN-based blue chips placed in the center of a package surrounded with a lead framework, a mixture of yellow phosphor YAG:Ce silicone, ZnO particles, and a semicircular lens that is placed above all the chips as cover and to prevent damage caused by external impacts. The power emission of the blue LED chips is 1 W when the emission wavelength is at 450 nm and the chip is driven by a 350 mA energy source. The compound of yellow phosphor silicone is fabricated by using a vacuum homogenizer to vacuum a yellow phosphor YAG:Ce (D 50%, 13 ± 2 μm) with the other component at a 3 to 8 ratio, 1360 rpm, and 0.2 MPa for 12 minutes. The gel will be then applied on blue chip and heat under 120 °C in the surrounding condition for 3 hours until reaching a solid-state. The ZnO nanoparticles and the adhesive substance are distributed evenly in the mixture by vacuum stirring, degassed. The phosphor layer is ultimately placed between the lens and blue LED chip via a higher pressure injector. Once the fusing process is complete, leave the ZnO particles to dry at 25°C in room atmosphere.

The field emission scanning electron microscope (FE-SEM, Merlin) is the tool to portray the exterior shape of the ZnO particles in this research. The UV-Vis spectrometer (UV-Vis, Agilent Cary 5000) measures the transmittance capacity of ZnO-containing films. The Optoelectronic estimation of ZnO-filled layers and WLEDs devices with ZnO component are both proceed by the Multi Spectrums T-950/930 system that morph spherical structure. These aforementioned evaluations are all performed in the air at a consistent room temperature of 25 °C.



(a) (b)

Figure 1. (a) Photograph of 9W wLED device,
(b) Schematic cross-sectional view of ZnO-doped wLED devices

2.2. Mie-scattering analysis

The scattering capacity of quantum-dot-converted elements (QDCEs) is similar to other structures scattering property and can be computed through the application of Monte Carlo stimulation [23]. The coefficient of scattering event and it phase function are the two main aspects that will be featured in this part. First, the scattering coefficient μ_{sca} is used to depict the scattering ratio which can be calculated by (1) [24]:

$$\mu_{sca}(\lambda) = \frac{c}{\bar{m}} \int f(D)\, C_{sca}(D, \lambda) dD \tag{1}$$

In (1), $c / \bar{m}$ depicts distributed density of QDs particles, c stands for the amount of Quantum Dots (mg/cm^3), D is magnitude of particles (nm); λ is the wavelength (nm); and $f(D)$ is the QD magnitude of particles distribution function; the weight measured in mg of QD within the QDCE *is* represented by $\bar{m}$ and can be identified through $f(D)$. The scattering cross-section of the QD $C_{sca}(D, \lambda)$ is expressed as follows:

$$C_{sca}(D, \lambda) = \frac{P_{sca}(D,\lambda)}{P_{inc}(\lambda)} = \frac{\int P_{sca}(\theta,D,\lambda) d\theta}{P_{inc}(\lambda)} \tag{2}$$

The incident irradiance of source (W/m^2) is depicted by $P_{inc}(\lambda)$; $P_{sca}(D,\lambda), P_{sca}(\theta,D,\lambda)$ are in turn the scattering power (W) and the scattering power when there is light diffuse through the QDs. The scattering phase function $\rho(\theta,\lambda)$, which is employed as a measurement unit for the scattering energy distribution in normalization conditions [25], can be by this simplified structure:

$$\rho(\theta, \lambda) = \frac{\int f(D) \rho_{sca}(\theta,D,\lambda) / P_{sca}(D,\lambda) C_{sca}(D,\lambda) dD}{\int f(D) C_{sca}(D,\lambda) dD} \tag{3}$$

3. RESULTS AND DISCUSSION

In Figure 2 are the charts demonstrating the scattering cross-section $C_{sca}(D, \lambda)$ of ZnO particles sizes vary from 100 nm – 300 nm. From the result of these charts, we can conclude that the scattering performance is affected by the particles size of ZnO because the scattering cross-section $C_{sca}(D, \lambda)$ increases with the ZnO size, thus, when the ZnO size increases it will result in better scattering effect. While the light with larger sizes particles tends to transmit in a straight direction and benefits the luminous flux, the light in smaller sizes particlesare usually spread evenly in every direction when scattering, therefore, has poor light output but advantageous in color uniformity. The value of $C_{sca}(D, \lambda)$ depends on the variation of wavelength, when the wavelength increases, $C_{sca}(D, \lambda)$ decreases, the wavelength that allows $C_{sca}(D, \lambda)$ to reach the highest index is at about 380 nm.

Similar to the scattering cross-section $C_{sca}(D, \lambda)$, the scattering coefficient $\mu_{sca}(\lambda)$ presented in Figure 3 is also improved accordingly when the size of ZnO rises. Better scattering coefficient indicates that the scattering ability of the ZnO particles are increase and will result in an enhancement in color uniformity. The scattering coefficient $\mu_{sca}(\lambda)$ is also opposed to the changes of wavelength, which means it gets smaller when the wavelength starts to increase and reaches the highest value at 380 nm wavelength. These traits show that ZnO is suitable to use in light quality enhancement. The phenomenon in both $C_{sca}(D,\lambda)$ and $\mu_{sca}(\lambda)$ can be explained by the two following reasons. The first reason is that we need to increase the number of scattering events occur in the phosphor layer so that the emitted lights are integrated perfectly and results in better color uniformity. Second, the scattering properties of ZnO are at the highest point when the wavelength is at 380 nm and decrease gradually until reaching the lowest index at 780 nm. Meanwhile, the emission wavelength of the LED chip is at 453 nm which confirms the suitability of ZnO to enhance the scattering in phosphor.

The results from Figure 2 and Figure 3 confirm that when the size of ZnO particles increase. The scattering ability will increase with it. To give the ideal ZnO size for application, the scattering phase function $\rho(\theta,\lambda)$ needs to be considered. The scattering $\rho(\theta,\lambda)$ phase function represents the angular scattering intensity of radiation from a particle. The scattering ability, as well as the scattering angle, are important criteria when choosing a size for the ZnO particles. With large particles size, the luminous flux is enhanced because the light is transmited through the large particles in a straight manner. However, with small particles size, the light often spreads out in different directions when scattering and creates a wider scattering angle. The widened scattering angle usually induces light loss due to backscattering event deflects the light back to the chip. However, employing small size particles is not without advantage, the blue and yellow rays are mixed more thoroughly before forming white light.

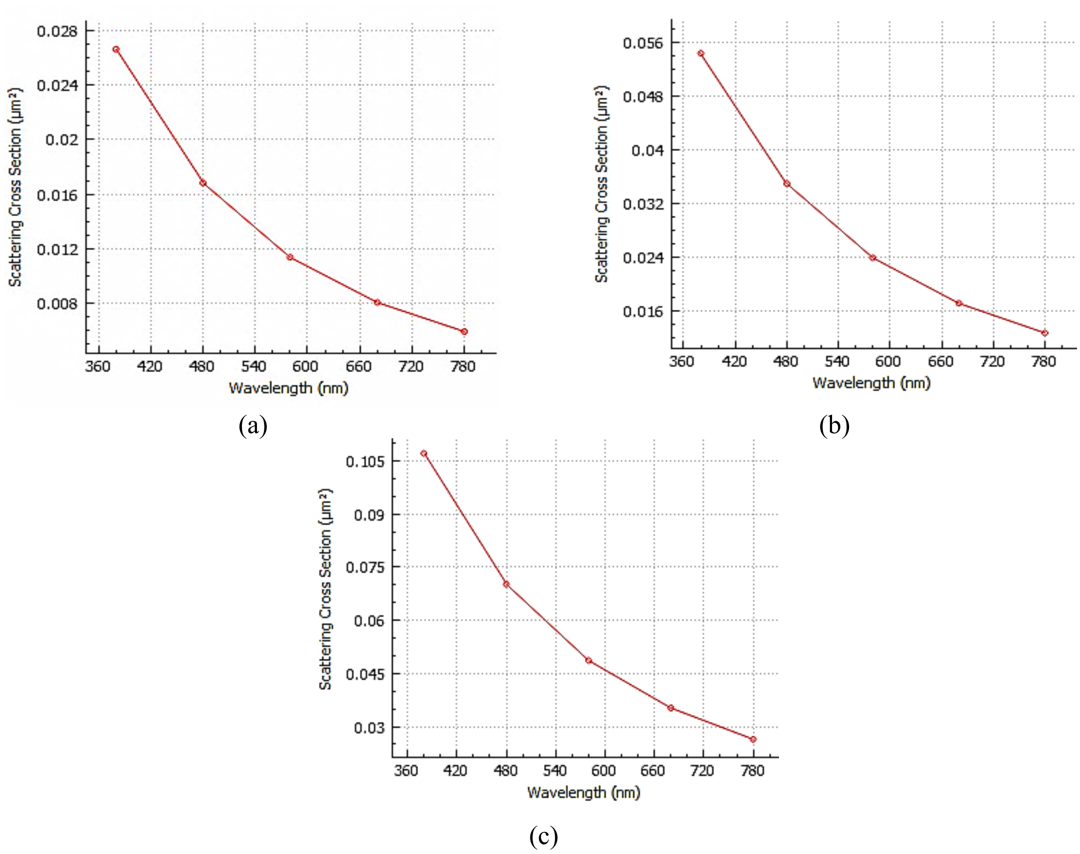


Figure 2. Scattering and cross-sections of ZnO particles with various sizes:
(a) 100 nm, (b) 200 nm, (c) 300 nm

The blue light rays are scattered more with small ZnO particles and these blue lights will appear more in both sides of the LED chip. In this situation, the blue light is merged with the “yellow ring” on the sides to produce white light which lessens the unwanted “yellow ring” effect and boost the color uniformity. Figure 4 shows the value of scattering phase function from the ZnO particles size of 100 nm-300 nm. From the images in Figure 4, we can confirm that the ZnO particles size affects both scattering intensity and scattering angle, particularly, the scattering intensity increases with the particles size while the scattering angle decrease.

Using only the particles size to evaluate the lumen output is not enough because lumen out also depends on other aspect such as the concentration of ZnO in the phosphor layer. As a result, Figure 5 responses to this by showing the lumen output correlating to the concentration of ZnO from 2% to 26%. As can be observed from the graphs, when the concentration increases, the lumen output decreases regardless of the particles size, which indicates that ZnO is not adding more ZnO is not ideal for light emission improvement. This phenomenon occurs due to the increased concentration results in more scattering events; the energy is distributed to these scattering events, which eventually affects the light emission energy source. Therefore, choosing the right particles size and concentration for ZnO become the critical requirement when involves in ZnO-Based WLEDs manufacturing. Based on Figure 5, using ZnO 6% with any particles size is the setting to achieve the optimal lumen output. However, the target of this research is not only enhancing the lumen output but also the color uniformity, therefore, we need to discuss the content of Figure 6 which exhibits the color temperature deviation before giving a conclusion.

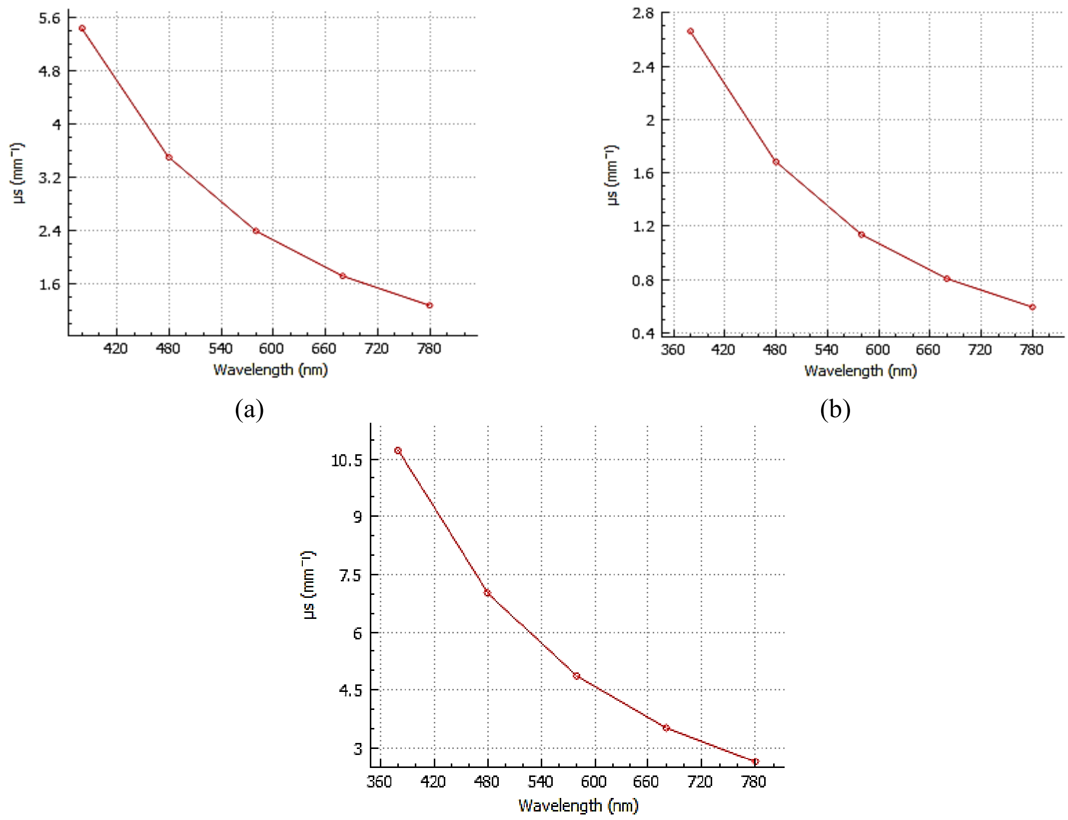


Figure 3. Scattering coefficient of ZnO particles with various sizes:
(a) 100 nm, (b) 200 nm, (c) 300 nm

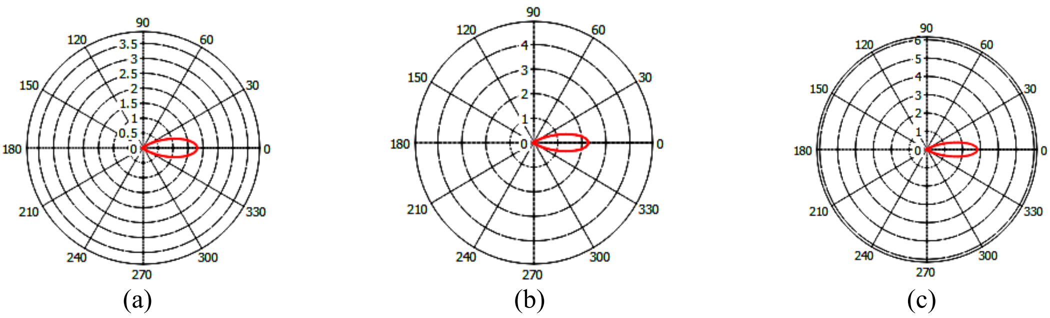


Figure 4. Scattering phase distributions of ZnO particles with various sizes:
(a) 100 nm, (b) 200 nm, (c) 300 nm

With the concentration of ZnO at 6%, the ΔCCT is not at the lowest point but still exhibits considerable decrease. ΔCCT reaches the lowest level at 18% ZnO concentration, the ZnO particles size of 100 nm generate the lowest ΔCCT at this concentration percentage. This can be explained by the excessive scattering events occur in the phosphor layers that make the scattered blue light redundant, especially with smaller size particles. The low ΔCCT low index and the balance between the proportions of blue and yellow light intensity are the requirements to achieve color uniformity. These requirements are targets when adjusting the size and concentration of ZnO particles. When the "yellow ring" phenomenon occurs that means the yellow light is in control and creates warm white light, in this case, the scattering of blue light needs enhancement to be equal with the yellow light. With the ZnO scattering examination conducted based on the scattering cross-section $C_{sca}(D,\lambda)$, the scattering coefficient $\mu_{sca}(\lambda)$, and the scattering phase function $\rho(\theta,\lambda)$, we can choose a ZnO particle with suitable indices to implement. If the requirement from the manufacturer focuses on the lumen output then the ZnO 6% should be added in the phosphor layer. The ZnO 18% is ideal for color uniformity development, therefore, it the optimal choice to go for if color uniformity is the more important feature of WLED. However, in cases that the objectives are both lumen output and color uniformity then the ZnO 6% is the most appropriate option.

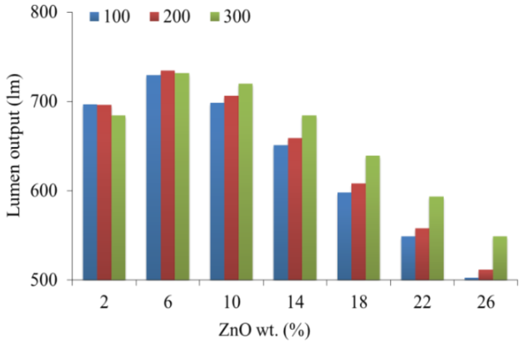


Figure 5. Lumen output of wLED as a function of the size and concentration of ZnO particles

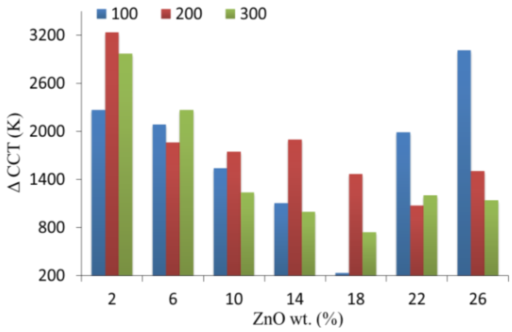


Figure 6. ΔCCT of WLED as a function of the size and concentration of ZnO particles

4. CONCLUSION

This article presents the influences of the ZnO particles on the optical properties of WLED. When adding ZnO particles with the sizes from 100 nm-300 nm into the yellow phosphor layer YAG:Ce, the mixture between ZnO and YAG:Ce enhance the scattered light obtained from the phosphor materials. Enhanced color uniformity can be achieved by boosting the light scatterd from the light sources, however, controlling the concentration of ZnO is essential to prevent damage to the lumen output. Through the study of the scattering cross-section $C_{sca}(D,\lambda)$, the scattering coefficient $\mu_{sca}(\lambda)$, and the scattering phase function $\rho(\theta,\lambda)$, this research is able to demonstrate the scattering of ZnO associating with particles sizes in details. Besides the particles size, the concentration of ZnO is another element mentioned to provide manufacturers with a more general view on the subject of applying ZnO. From the results relating to the ZnO concentration and particles size, it is verified that the lumen output reaches the highest index with ZnO at 6% concentration, while with ZnO 18% and 100 nm particles size, the obtained ΔCCT is lowest. It depends on the intention of the manufacturer to choose the suitable configuration for the ZnO particles size and concentration. However, if the focus implies on lumen output as well as ΔCCT then ZnO 6% is suitable for particles with the size from 100-00 nm.

REFERENCES

[1] Guan A. X., Mo F. W., Chen P. C., Geng Y., Chen Q., Zhou L. Y., "Photoluminescence Properties and Energy Transfer of $Eu3+$, $Bi3+$ Co-Doped $Ca9Y(PO4)7$ Phosphors," *Journal of Display Technology*, vol. 12, no. 2, pp. 136-142, 2016.
[2] Ding X. R., Chen Q., Tang Y., Li J. S., "Deepak Talwar DP, Binhai Yu BH, and Zongtao Li ZT. Improving the optical performance of multi-chip LEDs by using patterned phosphor configurations," *Optics Express*, vol. 26, no. 6, pp. A283-A292, 2018.
[3] Huang X. Y., Wang S. Y., Li B., Sun Q., Guo H., "High-brightness and high-color purity red-emitting Ca3Lu(AlO)3(BO3)4:Eu3+ phosphors with internal quantum efficiency close to unity for near-ultraviolet-based white-light-emitting diodes," *Optics Letters*, vol. 43, no. 6, pp. 1307-1310, 2018.

[4] Cho H. S., Joo C. W., Lee J. H., Lee H. K., Moon J. H., Lee J. I., Lee J. Y., Kang Y. J., Cho N. S., “Design and fabrication of two-stack tandem-type all-phosphorescent white organic light-emitting diode for achieving high color rendering index and luminous efficacy,” *Optics Express*, vol. 24, no. 21, pp. 24161-24168, 2016.

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

[6] He G. X., Zheng L H. “White-light LED clusters with high color rendering,” *Optics Letters*, vol. 35, no. 17, pp. 2955-2957, 2010.

[7] Choi S. I., “New Type of White-light LED Lighting for Illumination and Optical Wireless Communication under Obstacles,” *Journal of the Optical Society of Korea,* vol. 16, no. 3, pp. 203-209, 2012.

[8] Kim S. T., Kim J. S., Kim H. T., Kim Y. K., “Effects of Current Modulation Conditions on the Chromaticity of Phosphor Converted (PC) White LEDs,” *Journal of the Optical Society of Korea*, vol. 16, no. 4, pp. 449-456. 2012.

[9] Joo B. Y., Ko J. H., “Analysis of Color Uniformity of White LED Lens Packages for Direct-lit LCD Backlight Applications,” *Journal of the Optical Society of Korea*, vol. 17, no. 6, pp. 506-512, 2013.

[10] Kim I. I., Chung K. Y., “Wide Color Gamut Backlight from Three-band White LED. *Journal of the Optical Society of Korea*, vol. 11, no. 2, pp. 67-70, 2007.

[11] Anous N., Ramadan T., Abdallah M., Qaraqe K., Khalil D., “Impact of blue filtering on effective modulation bandwidth and wide-angle operation in white LED-based VLC systems,” *OSA Continuum*, vol. 1, no. 3, pp. 910-929, 2018.

[12] Sun C. C., Chang Y. Y., Wang Y. H., Chen C. Y., Cheng H. H., “Precise Spatial-Color Optical Modeling in Phosphor-Converted White LEDs,” *Journal of Display Technology*, vol. 11, no. 3, pp. 261-265, 2015.

[13] Jesús M., Quintero J. M., Antoni Sudrià A., Charles E., Hunt C.E., and Josep Carreras J., “Color rendering map: a graphical metric for assessment of illumination,” *Optics Express*, vol. 20, no. 5, pp. 4939-4956, 2012.

[14] Linhares J. M. M., Pinto P. D. A., Nascimento S. M. C., “Color rendering of art paintings under CIE illuminants for normal and color deficient observers,” *Journal of the Optical Society of America*, vol. 26, no. 7, pp. 1668-1677, 2009.

[15] Lin K. C. L., “Approach for optimization of the color rendering index of light mixtures,” *Journal of the Optical Society of America*, vol. 27, no. 7, pp. 1510-1520, 2009.

[16] Cheng G., Mazzeo M., D'Agostino S., Sala F. D., Carallo S., Gigli G., “Pure white hybrid light-emitting device with color rendering index higher than 90,” *Optics Letters,* vol. 35, no. 5, pp. 616-618, 2010.

[17] Mirhosseini R., Schubert M. F., Chhajed S. C., Cho J., Kim J. K., Schubert E. F., “Improved color rendering and luminous efficacy in phosphor-converted white light-emitting diodes by use of dual-blue emitting active regions,” *Optics Express*, vol. 17, no. 13, pp. 10806-10813, 2009.

[18] Nizamoglu S., Erdem T., Sun X. W., Demir H. V., “Warm-white light-emitting diodes integrated with colloidal quantum dots for high luminous efficacy and color rendering: reply to comment,” *Optics Letters*, vol. 36, no. 15, pp. 2852-2852, 2011.

[19] Thornton W. A., “Luminosity and Color-Rendering Capability of White Light,” *Journal of the Optical Society of America*, vol. 61, no. 9, pp. 1155-1163, 1971.

[20] Sahu I. P., Bisen D. P., Tamrakar R. K., “Dysprosium-Doped Strontium Magnesium Silicate White Light Emitting Phosphor Prepared by Solid State Reaction Method,” *Journal of Display Technology*, vol. 12, no. 11, pp. 1478-1487, 2016.

[21] Singh V. K., Tripathi S., Mishra M. K., Tiwari R., Dubey V., Tiwari N., “Optical Studies of Erbium and Ytterbium Doped Gd2Zr2O7 Phosphor for Display and Optical Communication Applications,” *Journal of Display Technology*, vol. 12, no. 10, pp. 1224-1228, 2016;

[22] Chen L. Y., Chang J. K., Wu Y. R., Cheng W. C., Chen J. H., Tsai C. C., Cheng W. H., “Optical Model for Novel Glass-Based Phosphor-Converted White Light-Emitting Diodes,” *Journal of Display Technology*, vol. 9, no. 6, pp. 441-446, 2013.

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

[24] Oh J. H., Kang H. J., Ko M. J., Do Y. R., “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.

[25] Ying S. P., Fu H. K., Tu H. Z., “Curved remote phosphor structure for phosphor-converted white LEDs,” *Applied Optics*, vol. 53, no. 29, pp. H160-H164, 2014.