

## The influences of calcium fluoride and silica particles on improving color homogeneity of WLEDs

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### Article Info

#### Article history:

Received Jul 12, 2019

Revised Apr 22, 2020

Accepted May 1, 2020

#### Keywords:

CaF<sub>2</sub>

Luminous flux

Mie-scattering theory

SiO<sub>2</sub>

### ABSTRACT

The LEDs lighting device with phosphor ingredient (pcLEDs) is among the most common lighting methods in recent years and evaluated by chromatic uniformity and lighting capacity. Therefore, we introduce the phosphor particles that can improve the scattering efficiency (SEPs) to apply in pcLEDs at 8500 K correlated color temperature (CCT) with the expectation to produce better pcLEDs by enhancing both quantity and quality of emitted light. Combining various materials such as CaF<sub>2</sub> and SiO<sub>2</sub> with yellow Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce<sup>3+</sup> phosphor composition in the pcLEDs simulation created by the LightTools program is the mechanism of this research. The simulated pcLEDs are tested and the results will be verified with Mie-scattering theory. The observation of the simulation leads to the conclusion about the scattering coefficients of SEPs at 455 nm and 595 nm wavelengths. The calculation showed that CaF<sub>2</sub> is better for color homogeneity yet suffer from luminous flux deficiency as the concentration gets higher. On the other hand, SiO<sub>2</sub> is the scattering enhancement material that can maintain high luminous flux regardless of its concentration.

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## 1. INTRODUCTION

To improve the WLED quality, color homogeneity, luminous flux, and color rendering index are the focus points, despite the fact that these criteria can induce inner scattering of WLED [1-5]. A conventional method to create a pcLEDs is to combine the yellow Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce<sup>3+</sup> phosphor with the silicone glue. The blue light after reaching the coating yellow Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce<sup>3+</sup> phosphor is consumed thus stimulates the yellow light and can be employed to create white light with a color temperature of choice [5-8]. The white LEDs with conformal phosphor configuration similar to the one used in this paper usually has a yellow ring that can cause irritation to the viewer's eyes. The cause of this incident is the imbalance of emitted blue and yellow radiation comes from the light source which resulting in inhomogeneous spatial color distribution [9, 10]. For further explanation, the scattering process weakens the blue light due to it being absorbed by the phosphor layer but boosts the yellow light that is the product of blue light converted from the phosphor layer. The range and

properties in the phosphor layer are distinct from the wavelengths, therefore, upon understanding the concept we instantly adapt it to change color homogeneity of pcLEDs. Wang's group has the purpose of reducing color deviation from 761K to 171K at the average CCT of 600K. Their study subjects are pcLEDs with chromatic phosphor  $\text{SiO}_2$ ,  $\text{B}_2\text{O}_3$ ,  $\text{PbO}$ ,  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$  particles merged with silicone adhesive and placed in glass composite [11]. Besides, Lin's group is in charge of assembling the  $\text{HfO}_2/\text{SiO}_2$  DBR film to adjust the color deviation of pcLEDs at approximately 5000 K from 1758 K to 280 K [12]. Moreover, Yu's group tests the remote micro-patterned phosphor film on pcLEDs at 5537 K and study its effectiveness in reducing the color deviation to 441 K [13]. The results confirm that applying these phosphor configurations is beneficial for the quality of spatial color homogeneity. Even though the benefits are undoubtedly valuable but they could not be widely used due to the high producing cost and difficult manufacturing requirements. Therefore, SEPs such as  $\text{TiO}_2$ ,  $\text{ZrO}_2$ , microspheres and  $\text{SiO}_2$  are more practical materials that can mix with yellow phosphor to create new phosphor compositions are being used [14-18]. The research conduct by Lee with his partner in 2010 which dispersed  $\text{TiO}_2$  on pcLEDs to exam the possibility that color homogeneity can benefit from adding 0.1%  $\text{TiO}_2$  to the encapsulated phosphor component. As a result of many attempts to enhance color homogeneity, numerous findings have been announced, such as Yang's group demonstrated that using  $\text{CaCO}_3$  can boost the scattering features of pcLEDs, specifically the spatial color homogeneity is greatly increased when adding 10% of  $\text{CaCO}_3$  [19]. Similarly, Anh's group discovered that adding the SEP  $\text{SiO}_2$  in the phosphor composition of pcLEDs can result in positive change relate to spatial color homogeneity. Besides, other aspects of  $\text{SiO}_2$  can also affect pcLEDs, the positioning within pcLEDs influence the color quality, likewise the chromatic performance is also influenced by the magnitude of  $\text{SiO}_2$  molecule [20]. From the results of other research, SEPs are good for the overall improvement of pcLEDs, however, an optimal SEP that can yield the biggest development is still undiscovered. The SEPs usage does not end there, previous research confirms that pcLEDs with one chip and emitted yellowish light can benefits from the improvements of color deviation and lumen output if the structure employs SEPs. Besides the type of SEPs, choosing the concentration and size for the SEP particle is important as it could enhance lighting performance and color quality with a suitable setting. This research aims to testify the influences of the  $\text{CaF}_2$  and  $\text{SiO}_2$  particles nominated above on optical properties of pcLEDs as well as measures their particular effect on enhancing the pcLEDs performance. The focus is to find optimal SEP material for different types of pcLEDs with distinct demands and explain how the SEPs improve color homogeneity and luminous flux using Mie theory along with Monte Carlo simulation. The contents of the article from this point onward are arranged into 3 sections with section 2 analyzes the inner circulation of light inside pcLEDs and contributes basic information for further experiments mentioned in section 3. Besides, section 3 also discuss the results about optical characteristics from the experiments. In section 4, we summarize the paper and give conclusions on the topic.

## 2. ANALYZATION ON THE SCATTERING EFFECT

Light scattering effect is a phenomenon caused by SEPs when it is in the pcLEDs with conformal phosphor structure according to the Mie-scattering theory and will be calculated with the help of MATLAB [21-25]. The following equations are the instruments for calculating the scattering coefficient  $\mu_{sca}(\lambda)$ , the anisotropy factor  $g(\lambda)$ , the decreased scattering coefficient  $\delta_{sca}(\lambda)$  and the scattering amplitude functions  $S_1(\theta)$  and  $S_2(\theta)$ :

$$\mu_{sca}(\lambda) = \int N(r) C_{sca}(\lambda, r) dr \quad (1)$$

In (1), the  $N(r)$  is the amount of diffusive molecule ( $\text{mm}^3$ ), also known as diffusion density distribution.  $C_{sca}$  stands for scattering cross section ( $\text{mm}^2$ ).  $\lambda$  is the wavelength in nanometers and the radius of the SEPs particles are presented as  $r$  (mm).

The scattering coefficients of  $\text{CaF}_2$  and  $\text{SiO}_2$  are computed and shown in Figure 1. From 380 nm to 780 nm, these scattering coefficients are different, which means there's a difference in bright scattering. It is easy to see that the scattering coefficient of  $\text{CaF}_2$  larger than  $\text{SiO}_2$ . Therefore, the color quality in the case of using  $\text{CaF}_2$  would be better than  $\text{SiO}_2$ . However, it is necessary to identify the luminous flux that obtains when using these particles. The larger the scattering coefficient will benefit for color homogeneity, but not beneficial for luminous flux. The larger the scattering coefficient means the greater the scattering process, the light rays are mixed more times before the outside and result in a low color deviation. The scattering coefficient can be used to evaluate the level of scattering in pcLEDs. And this is the key point to controlling color homogeneity and luminous flux. For  $\text{CaF}_2$  and  $\text{SiO}_2$ , the selection of appropriate concentration is important. Besides the size of  $\text{CaF}_2$  and  $\text{SiO}_2$  particles must also be of interest.

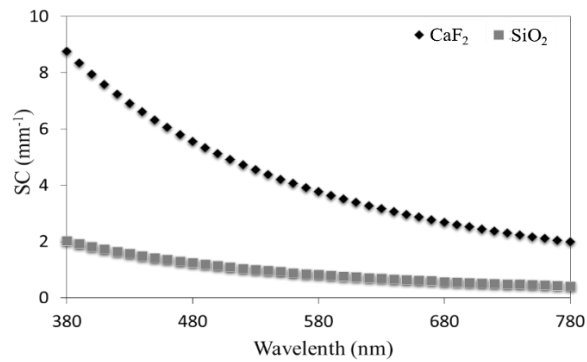


Figure 1. Computation of scattering coefficient the SEPs

### 3. RESULTS AND DISCUSSION

This part shows the optical properties of SEPs pcLEDs mentioned above simulated by LightTools 8.1.0 program. The physical model is presented in Figure 2 (a) and components details are in Figure 2 (b). The measurements of the model reflector are 2.1 mm in depth, 8 mm inner and 10 mm on the surface. The arrangement within the pcLEDs can be observed from the cross-section Figure 2 (c). Finally, Figure 2 (d) illustrate the result of simulated LED device.

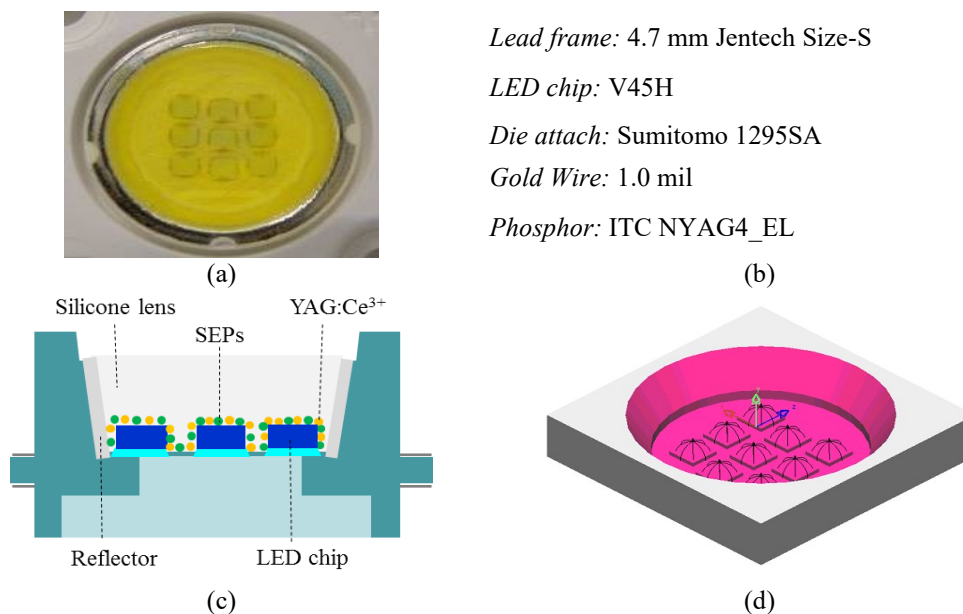


Figure 2. (a) Photograph of WLEDs sample, (b) Manufacturing parameter of WLEDs, (c) Illustration of 2D WLEDs model, (d) the simulated WLEDs model

The structure have 9 LED chips placed at the base under the phosphor materials. The density of the phosphor layer is fixed at 0.08 mm. SEPs are deemed as  $0.5 \mu\text{m}$  spherical with the refractive indexes for  $\text{CaF}_2$  is 1.44,  $\text{SiO}_2$  is 1.54. Phosphor particles radius is  $7.25 \mu\text{m}$  on average with 1.83 refractive index disregard to position in the visible spectrum. The silicone glue used in the experiment has its refractive index unchanged at 1.5. The distribution of particle density can change depends on the requirements for CCT uniformity and lighting efficacy.

$$W_{\text{phosphor}} + W_{\text{silicone}} + W_{\text{SEP}} = 100\% \quad (2)$$

In (2) is the percentage composition of a pcLEDs, where  $W_{silicone}$ ,  $W_{phosphor}$  and  $W_{SEP}$ , in turn, demonstrate the weight proportion of the silicone, phosphor and SEP in the phosphor composition of the structure. The balance between proportions of phosphor and SEP will result in CCT stability that keep it at 8500K. The color deviation is an important feature that indicate the quality of the lighting device. In lighting applications that use light-emitting diodes (LEDs), if the variation of CCT is high at different angles means that the yellow ring phenomenon will occur together with inhomogeneous white light causing the lighting performance of pcLEDs to decline. To calculate the CCT deviation the following expression can be applied:

$$\Delta CCT = CCT(Max) - CCT(Min) \quad (3)$$

In this equation,  $CCT_{(Max)}$  and  $CCT_{(Min)}$  in turns, denote the highest and lowest CCT at 0- and 90-degrees point of view. These features fluctuate because of unequal particles emitting capacity. To settle this issue, the emitted blue light needs improvement to match with the scattered light from other sources, hence effectively reduces the CCT deviation. The CCT deviation in the pcLEDs are measured and expressed in Figure 3.

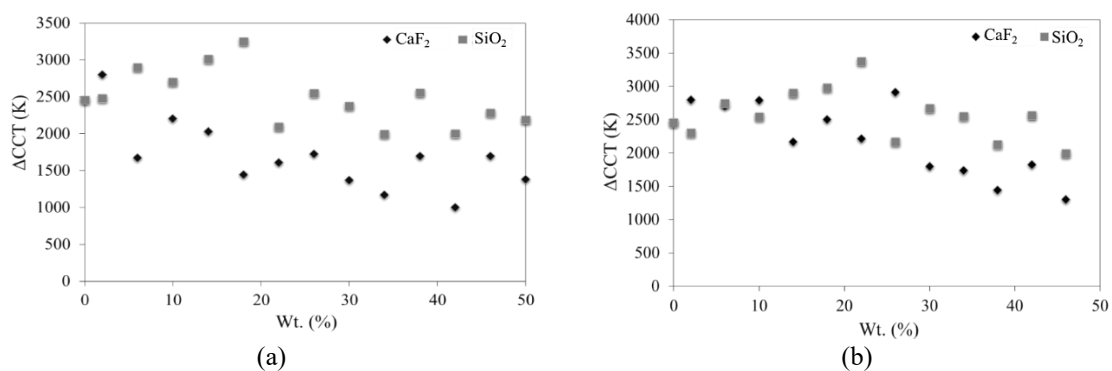


Figure 3. Comparison of CCT deviation of pcLEDs using different SEPs at the size of (a) 400 nm and (b) 800 nm

According to Figure 3,  $CaF_2$  particles at 400 nm and 800 nm achieve the lower deviation of angular scattering amplitude in comparison with the case of  $SiO_2$ . This indicates that using  $CaF_2$  is the most effective SEP to limit the variation in radiant intensity distributions. The angular scattering amplitude of  $CaF_2$ , similar to other SEPs, is higher with particles at 455 nm than 595 nm. The combination of blue and yellow light fabricates white light but will also induce the “yellow ring” if the discrepancy between these chromatic lights are high enough, therefore, having enough blue light makes CCT deviation more manageable and eradicates yellow ring, an either redundant or insufficient amount of scattered blue light in pcLEDs can widen the CCT deviation. As Figure 1 suggests, the angular scattering amplitudes of  $CaF_2$  and  $SiO_2$  with 455 nm particles almost triple the results measured with 595 nm particles which is a huge improvement for the chromatic performance and lumen output of pcLEDs. The content of Figure 3 demonstrates all the aforementioned arguments that adding  $CaF_2$  and  $SiO_2$  reduce the CCT deviation. The results of CCT deviations in pcLEDs with and without  $CaF_2$  and  $SiO_2$  SEPs at 2670 K show that applying SEPs in the structure can reduce the color deviation down by 1800 K. On the contrary, the concentration of  $CaF_2$  and  $SiO_2$  particles in their own separate case causes the CCT deviation to increase accordingly to them. Besides CCT deviation, the light output of pcLEDs are also changed because of  $CaF_2$  and  $SiO_2$  and is recorded in Figure 4.

In Figure 4 are the resulting luminous flux of pcLEDs with  $CaF_2$  and  $SiO_2$  that have the materials concentration varies 0-50% and the size from 100nm to 1000nm. In  $SiO_2$  cases, the results show a certainty that the luminous flux is proportional to the concentration and size of the particles. For  $CaF_2$ , it provides improvement in luminous flux with all particles size at the concentration range from 0-20% but tends to create the opposite effect if the concentration of  $CaF_2$  continues to rise. Apparently, the particles size have a deciding role in controlling the back-scattering effect of the phosphor layers which can be utilized for better luminous efficacy in WLEDs. To examine the decrease caused by the exceeding concentration of SEPs, we apply the Lambert-Beer law and the Mie-scattering theory in the calculation.

The Mie-theory is employed for analyzing the scattering of SEPs, specifically the scattering cross section  $C_{sca}$  for spherical particles while the transmitted light power is analyzed using the Lambert-Beer law:

$$I = I_0 \exp(-\mu_{ext}L) \quad (4)$$

According to the formula above,  $I_0$  is the optical power of incident light and  $L$  is the density of phosphor coating (mm).  $\mu_{ext}$  indicates the extinction coefficient that is computed by this equation:  $\mu_{ext} = N_r \cdot C_{ext}$ , where  $N_r$  stands for the number density distribution of particles ( $\text{mm}^{-3}$ ) and  $C_{ext}$  ( $\text{mm}^2$ ) is the extinction cross-section of phosphor particles. Based on (4), a conclusion can be made that the higher the concentration of SEPs the lower the luminous flux of WLEDs. This incident is due to the emission energy being damaged by the increase of light scattering inside of the phosphor layers and high concentration of SEPs that cause back-scattering effect.

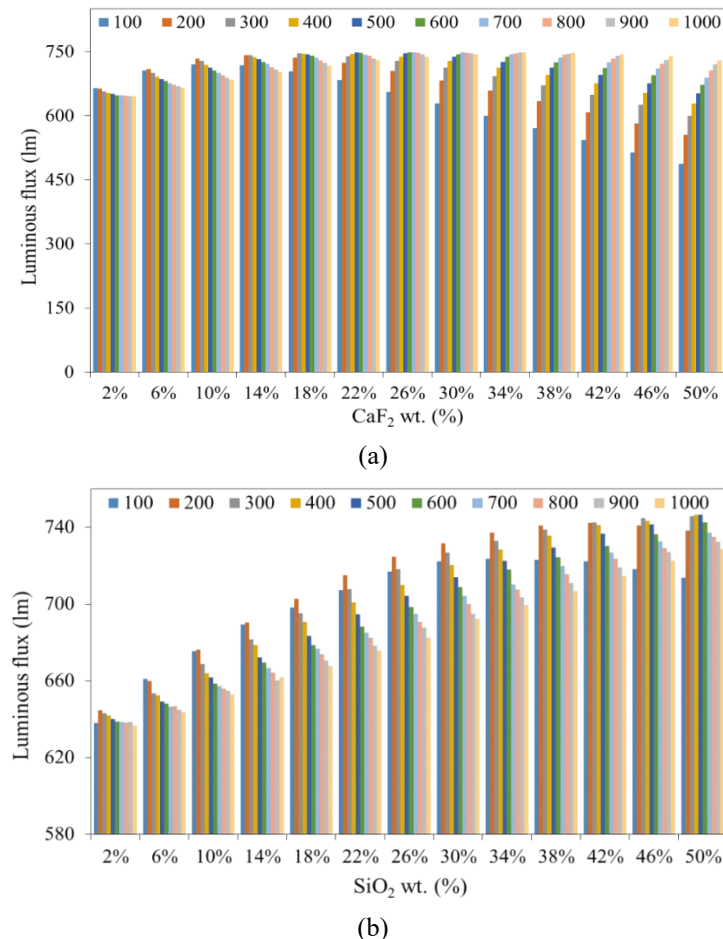


Figure 4. Comparison of luminous flux of pcLEDs adding CaF<sub>2</sub> (a) and SiO<sub>2</sub> (b)

#### 4. CONCLUSION

The target of this research is to study the influences that SEPs might have on two quality-deciding properties of white LEDs devices, which are chromatic quality and luminous flux. Through applying the mechanism of Mie-scattering and Monte Carlo in the verification process, the results are approved and certain that with different types of particle the enhancements occur in pcLEDs are distinct. Correspondingly, this encourages the discovery of an optimal SEPs and concentration level for a specific occasion that benefits the optical performance of pcLEDs the most. Our findings in this particular article can serve as a guideline to manufacture WLEDs with predetermined requirements effectively or base knowledge for further development. Specifically, the CCT deviation opposes to the concentration of CaF<sub>2</sub> and SiO<sub>2</sub> which is a characteristic useful for CCT management, therefore, to reduce color deviation to the lowest possible using CaF<sub>2</sub> would be the suitable choice. With that being said, control over CaF<sub>2</sub> concentration is desirable as it prevents damage to luminous output caused by an excessive concentration. On another note, SiO<sub>2</sub> is the material that benefit the growth of luminous flux in pcLEDs with SiO<sub>2</sub> being the material that provides the lumen output.

## REFERENCES

- [1] Cheng J., Zhang J., Zhang H. C., Maryam S., Bian X. T., Shen Z. H., Ni X. W., Lu J., "Synthesis and photoluminescence properties of Sr<sub>4</sub>La(PO<sub>4</sub>)<sub>3</sub>O:RE<sup>3+</sup>(RE=Eu/Tb/Ce) phosphors," *Chinese Optics Letters*, vol. 15, no. 12, pp. 121602, 2017.
- [2] Sharma S., Brahme N., Bisen D. P., Dewangan P., "Cool white light emission from Dy<sup>3+</sup> activated alkaline alumino silicate phosphors," *Optics Express*, vol. 26, no. 22, pp. 29495-29508, 2018.
- [3] Min K. T., Choi Y. K., Jeon H. S., "Model calculations for enhanced fluorescence in photonic crystal phosphor," *Optics Express*, vol. 20, no. 3, pp. 2452-2459, 2012.
- [4] Tang Y., Zhi Li Z., Liang G. W., Li Z., L. JSi, Yu B. H., "Enhancement of luminous efficacy for LED lamps by introducing polyacrylonitrile electrospinning nanofiber film," *Optics Express*, vol. 26, no. 21, pp. 27716-27725, 2018.
- [5] 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.
- [6] Peng Y., Wang S. M., Li R. X., Li H., Cheng H., Chen M. X., Liu S., "Luminous efficacy enhancement of ultraviolet-excited white light-emitting diodes through multilayered phosphor-in-glass," *Applied Optics*, vol. 55, no. 18, pp. 4933-4938, 2016.
- [7] Tang Y. R., Zhou S. M., Yi X. Z., Lin H., Zhang S., Hao D. M., "Microstructure optimization of the composite phase ceramic phosphor for white LEDs with excellent luminous efficacy," *Optics Letters*, vol. 40, no. 23, pp. 5479-5481, 2015.
- [8] Siao C. B., Wang K. W., Chen H. S., Su Y. S., Chung S. R., "Ultra high luminous efficacy of white ZnxCd1-xS quantum dots-based white light emitting diodes," *Optical Materials Express*, vol. 6, no. 3, pp. 749-758, 2016.
- [9] Kim S. H., Song Y. H., Jeon S. R., T. J., Kim J. Y., Ha J. S., Kim W. H., Baek J. H., Yang G. M., Park H. J., "Enhanced luminous efficacy in phosphor-converted white vertical light-emitting diodes using low index layer," *Optics Express*, vol. 21, no. 5, pp. 6353-6359, 2013.
- [10] 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 AlGaIn cap layer," *Optics Express*, vol. 24, no. 16, pp. 17868-17873, 2016.
- [11] Zhang F. Z., Xu H. S., Wang Z., "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.
- [12] Cantore M., Pfaff N., Farrell R. M., Speck J. S., Nakamura S., DenBaars S. P., "High luminous flux from single crystal phosphor-converted laser-based white lighting system," *Optics Express*, vol. 24, no. 2, pp. A215-A221, 2016.
- [13] Lee T. X., Chou C. F., "Ideal luminous efficacy and color spatial uniformity of package-free LED based on a packaging phosphor-coated geometry," *Applied Optics*, vol. 55, no. 27, pp. 7688-7693, 2016.
- [14] Lee S. M., Sock Choi C. S., Choi K. C., "Effects of Auxiliary Electrode Width in AC Plasma Display Panels With Auxiliary Electrodes," *Journal of Display Technology*, vol. 6, no. 12, pp. 607-613, 2010.
- [15] Zhang Q., Zheng R., Ding J. Y., Wei W., "Excellent luminous efficiency and high thermal stability of glass-in-LuAG ceramic for laser-diode-pumped green-emitting phosphor," *Optics Letters*, vol. 43, no. 15, pp. 3566-3569, 2012.
- [16] Oh J. H., Rok Oh J. R., Park H. K., Sung Y. G., Do Y. R., "Highly-efficient, tunable green, phosphor-converted LEDs using a long-pass dichroic filter and a series of orthosilicate phosphors for tri-color white LEDs," *Optics Express*, vol. 20, no. S1, pp. A1-A12, Yukio Narukawa, 2012;
- [17] Erdem T., Nizamoglu S., Demir H. V., "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.
- [18] Bol'shukhin V. A., Ilyasov V. S., Soshchin N. P., Ulasyuk V. N., "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.
- [19] Tran N. T., Shi F. G., "Studies of Phosphor Concentration and Thickness for Phosphor-Based White Light-Emitting-Diodes," *Journal of Lightwave Technology*, vol. 26, no. 21, pp. 3556-3559, 2008.
- [20] He G. H., Zheng L. H., "Color temperature tunable white-light light-emitting diode clusters with high color rendering index," *Applied Optics*, vol. 49, no. 24, pp. 4670-4676, 2010.
- [21] Deng X., Wu Y., Khalid A. M., Long X., Linnartz J. P. M. G., "LED power consumption in joint illumination and communication system," *Optics Express*, vol. 25, no. 16, pp. 18990-19003, 2017.
- [22] Oh J. H., Yang S. J., Sung Y. G., Do Y. R., "Improved color coordinates of green monochromatic pc-LED capped with a band-pass filter," *Optics Express*, vol. 21, no. 4, pp. 4539-4550, 2013.
- [23] Kim S. N., Kim B. N., Kim H. S., "Optical properties of densified phosphor-in-glass LED encapsulants by spark plasma sintering," *Optical Materials Express*, vol. 7, no. 12, pp. 4304-4315, 2017.
- [24] Oh J. R., Cho S. H., Oh J. H., Kim Y. K., Lee Y. H., Kim W., Do Y. R., "The realization of a whole palette of colors in a green gap by monochromatic phosphor-converted light-emitting diodes," *Optics Express*, vol. 19, no. 5, pp. 4188-4198, 2011.
- [25] Smet K., Ryckaert W. R., Pointer M. R., Deconinck G., Hanselaer P., "Optimal colour quality of LED clusters based on memory colours," *Optics Express*, vol. 19, no. 7, pp. 6903-6912, 2011.