RED-EMITTING Ca₂Si₅N₈Eu²⁺ PHOSPHOR: A NEW RECOMMENDATION FOR IMPROVING COLOR UNIFORMITY AND COLOR QUALITY SCALE OF THE CONFORMAL PACKAGING MULTI-CHIP WHITE LEDs

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Light emitting diodes (LEDs) are commonly used in general lighting applications due to their rapidly improving efficiency, long-life, compact, low power consumption, and high climate resistance. In this paper, the Red-Emitting $Ca_2Si_5N_8Eu^{2+}$ Phosphor was investigated like an innovative approach for improvement on the color uniformity and Color Quality Scale (CQS) of 8500 K, 7000K, 5600 K multi-chip white LED lamps (MCW-LEDs). The influence of the Red-Emitting $Ca_2Si_5N_8Eu^{2+}$ Phosphor concentration on the color uniformity and the CQS is simulated, analyzed with using Mat lab and Light Tools software. The research results indicated that the color uniformity and the CQS were crucially affected by the Red-Emitting $Ca_2Si_5N_8Eu^{2+}$ Phosphor concentration. This paper provided an essential recommendation for selecting and developing the phosphor materials for MCW-LEDs manufacturing.

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1. Introduction

The last four decades have seen a growing trend towards in rapid development of light emitting diodes (LEDs) with compound semiconductor technology. Since the first red LEDs that was invented by Holonyak and Bevacqua in 1962 [1], considerable efforts have been put into the study to obtain brighter LEDs. In the decades that followed, LEDs were used extensively in digital displays and signaling applications. However, only around 1995 high brightness and blue LEDs were developed, which made it possible to use LEDs for general lighting. Nowadays, MCW-LEDs have attracted considerable attention from both general lightings manufacturers and consumers due to its excellent properties for display technology, including high brightness, low power consumption, long lifetime, fast response as well as climate impact resistance [2]. LEDs have a narrow emission spectrum. Generally, there are three different approaches which can be used for generating white light based on LEDs: (1) by mixing reds, greens, and blues, i.e. red–green–blue (RGB) LEDs, (2) by using an ultraviolet (UV) LED to stimulate RGB phosphors, and (3) by using a blue-emitting diode that excites a yellow-emitting phosphor embedded in the epoxy dome; the combination of blue and yellow light makes a white-emitting LED. The last method is commonly used in general lighting applications because of its simple procedure [3-7, 24].

Nowadays, the improvement of optical properties of WLEDs is central direction research in optoelectronics. Up to date, many studies focused on improving optical properties of WLEDs by controlling geometry, thickness, concentration, and packaging of the yellow-emitting phosphor in the phosphor compounding of WLEDs [8-14]. In another way, some researchers just concentrated on adding SiO2 to control and enhance the optical properties of WLEDs [15,16]. Previous studies

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showed that in this way the optical properties of WLEDs could be improved significantly. These studies just focused on single chip WLEDs with the low correlated temperature or only control one of three optical properties of WLEDs (the color correlated temperature deviation (D-CCT), CRI and luminous efficacy). However, not much attention has been paid to improving the optical properties of the high color temperature MCW-LEDs yet. This research could fill the remaining gap.

In this paper, the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor [17,18] is proposed to improve the optical properties (color quality scale (CQS) and color uniformity) of the conformal package multi-chip WLEDs with CCT 5600K, 7000K, and 8500K. The research results, using the simulation package Light Tool and Mat lab software based on the Lambert-Beer law and Mie theory, demonstrated that concentration of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor had a significant impact on the color uniformity (D-CCT) and CQS of the conformal package MCW-LEDs. The overall structure of the study takes the form of three segments. Firstly, the 8500 K, 7000 K, and 5600 K MCW-LEDs physical model is conducted by the commercial Light Tools software. Secondly, by varying concentration of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor particles, the D-CCT and CQS calculated, analyzed and investigated. Finally, according to the results and the Beer-Lambert law, Mie theory, effect of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor particles on CQS and D-CCT of MCW-LEDs is discussed. From the results, the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor could be a prospective approach for MCW-LEDs improvement shortly.

2. The MCW-LEDs physical model

In the commercial software Light Tools, the real-world model of MCW-LEDs was used (Fig. 1(a)). In this simulation stage, the depth, the inner and outer radius of the reflector were set at 2.07 mm, 8 mm and 9.85 mm, respectively. Nine LED chips are covered with a fixed thickness of 0.08 mm and 2.07 mm. The dimension of the blue chip is 1.14 mm by 0.15mm, the radiant flux of 1.16 W, and the peak wavelength of 453 nm. Fig. 1(b) shows that the phosphor layer of the conformal phosphor packaging is coated conformally on 9 LEDs chips. These phosphor layers consist of the yellow-emitting YAG:Ce and the redemitting $Ca_2Si_5N_8Eu^{2+}$ phosphors particles and the silicone glue, which respectively have the refractive indices of 1.83, 1.93 and 1.50. Also, the average radius of YAG:Ce phosphor particles are set to 7.25 µm for all packages, a value of real particle size. In order to keep the color of MCW-LEDs the same while the concentration of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor writes, the yellow-emitting YAG:Ce phosphor concentration should be inversely changed to provide same CCT value. The weight percentage of the MCW-LEDs phosphor layers can be expressed as:

$$\sum W_{pl} = W_{Yellow \ phosphor} + W_{silicone} + W_{Red \ phosphor} = 100\%$$

Here the $W_{silicone}$, $W_{Yellow phosphor}$ and $W_{Red phosphor}$ are in turn the weight percentage of the silicone glue, the yellow-emitting YAG: Ce phosphor and the red-emitting CeTb phosphors.



Fig. 1. (a) The MCW-LED product, (b), Illustration of MCW-LEDs with the conformal phosphor package.

3. Results and discussion

In this segment, the color correlated temperature deviation (D-CCT) and the CQS of the 8500K, 7000 K, and 5600 K MCW-LEDs are calculated and obtained by using the commercial software Light Tools. In this simulation, the concentration of the red-emitting Ca₂Si₅N₈Eu²⁺ phosphor varied continuously from 0% to 14%. From the simulation results, the corresponding values of D-CCT are calculated and indicated in Fig. 2. As shown in Fig, 2, the D-CCT had a slight decrease when the concentration of the red-emitting $Ca_2Si_5N_8Eu^{24}$ phosphor varied from 0% to 8%. After that, the D-CCT of the MCW-LEDs increased while the red-emitting Ca₂Si₅N₈Eu²⁺ phosphor changed to 14%. In this figure, the lowest value of the D-CCT is obtained at 8-10 % concentration of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor. On the other way, Fid. 3 shows the CQS of the conformal packaging MCW-LEDs while the concentration of the red phosphor changed from 0% to 14%. It can be indicated that the CQS grows with the concentration red phosphor in the range from 0% to nearly 14%. The highest CQS is obtained with the red weight vary from 10% to 12%. In Fig. 3, the CQS could be obtained highest value near 80. From the results, the CQS of high CCT MCW-LEDs can be controlled by adding red-emitting phosphor in phosphor compound and varying its concentration. The best value of the D-CCT and CQS can be obtained when the concentration of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor is nearly 8-10%.

The influence of the concentration of the red-emitting Ca₂Si₅N₈Eu²⁺ phosphor on the color uniformity (D-CCT) and the CQS can be demonstrated by using Mie theory [19]. Here, we can apply Mie-scattering theory [20,21], the scattering coefficient $\mu_{sca}(\lambda)$, anisotropy factor $g(\lambda)$, and reduced scattering coefficient $\delta_{sca}(\lambda)$ can be computed by the below expressions (1), (2), and (3):

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

$$g(\lambda) = 2\pi \int_{-1}^{1} p(\theta, \lambda, r) f(r) \cos \theta d \cos \theta dr$$
(2)

$$\delta_{sca} = \mu_{sca}(1-g) \tag{3}$$

In these equations, N(r) indicates the distribution density of diffusional particles (mm³). C_{sca} is the scattering cross sections (mm²), $p(\theta, \lambda, r)$ is the phase function, λ is the light wavelength (nm), r is the radius of diffusional particles (µm), and θ is the scattering angle (°), and f(r)) is the size distribution function of the diffuser in the phosphorous layer. Moreover, f(r) and N(r) can be calculated by:

$$f(r) = f_{dif}(r) + f_{phos}(r)$$
(4)

$$N(r) = N_{dif}(r) + N_{phos}(r) = K_N [f_{dif}(r) + f_{phos}(r)]$$
(5)

N(r) is composed of the diffusive particle number density $N_{dif}(r)$ and the phosphor particle number density $N_{phos}(r)$. In these equations, $f_{dif}(r)$ and $f_{phos}(r)$ are the size distribution function data of the diffusor and phosphor particle. Here K_N is the number of the unit diffusor for one diffuser concentration and can be calculated by the following equation:

$$c = K_N \int M(r) dr \tag{6}$$

Where M(r) is the mass distribution of the unit diffuser and can be proposed by the below equation:

$$M(r) = \frac{4}{3}\pi r^{3} [\rho_{dif} f_{dif}(r) + \rho_{phos} f_{phos}(r)]$$
(7)

Here $\rho_{diff}(r)$ and $\rho_{phos}(r)$ are the density of diffuser and phosphor crystal. In Mie theory, C_{sca} can be obtained by the following expression:

$$C_{sca} = \frac{2\pi}{k^2} \sum_{0}^{\infty} (2n-1)(|a_n|^2 + |b_n|^2)$$
(8)

where $k = 2\pi/\lambda$, and a_n and b_n are calculated by:

$$a_{n}(x,m) = \frac{\psi_{n}(mx)\psi_{n}(x) - m\psi_{n}(mx)\psi_{n}(x)}{\psi_{n}(mx)\xi_{n}(x) - m\psi_{n}(mx)\xi_{n}(x)}$$
(9)

$$b_{n}(x,m) = \frac{m\psi_{n}(mx)\psi_{n}(x) - \psi_{n}(mx)\psi_{n}(x)}{m\psi_{n}(mx)\xi_{n}(x) - \psi_{n}(mx)\xi_{n}(x)}$$
(10)

Where x = k.r, *m* is the refractive index, and $\psi_n(x)$, $\xi_n(x)$ are the Riccati - Bessel function [24].

The extinction coefficient values of the red-emitting phosphor are verified at wavelengths, 555 nm, and 453 nm, respectively. The variation of the mentioned parameters on the red phosphor concentration according to the above equations are displayed in Fig.4 (a)-(c) for 8500K, 7000 K and 5600 K respectively.

As displayed in Fig. 5, the scattering coefficients grew with increasing red phosphor concentration. It means that the white-light quality can be enhanced by controlling red phosphor concentration. The reduced scattering coefficient of red phosphor with wavelengths 453nm, 555nm and 680nm are approximate with each other (Fig 6). It indicated that the scattering stability of red phosphor is used for controlling the color quality of MCW-LEDs. The results indicated that red phosphor particles had a significant advantage in producing blue light. The more the blue light emitted, the more the yellow ring phenomenon reduced. These calculations and analysis can use to proving the results in Fig. 2 and Fig. 3.



Fig. 2. The color quality scale (CQS) at average CCTs of 8500 K, 7000K, and 5600 K

328



Fig. 3. The color quality scale (CQS) at average CCTs of 8500 K, 7000K, and 5600 K



Fig. 4. The extinction coefficients of MCW-LEDs with CCT 8500K (a), 7000K (b), 5600K (c).



Fig. 5. Scattering coefficients of $Ca_2Si_5N_8Eu^{2+}$ of 453nm, 555nm, and 680nm



Fig. 6 Reduced scattering coefficient of $Ca_2Si_5N_8Eu^{2+}$ of 453nm, 555nm, and 680nm

4. Conclusions

The purpose of the current paper was to determine the influence of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor concentration on the color uniformity and the CQS of the MCW-LEDs. The results of this investigation show that the concentration of the red phosphor particles crucial influenced on the optical properties of the conformal packaging MCW-LEDs.

The both D-CCT and CQS can obtain the best value at the 8-10% concentration of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor. Further works must concentrate to validate the influence of the red-emitting $Ca_2Si_5N_8Eu^{2+}$ phosphor size on the optical properties of the conformal packaging MCW-LEDs.

References

- [1] J. N. Holonyak, S. F. Bevacqua, Applied Physics Letters 1(4), 82 (1962).
- [2] E. Fred. Schubert, "History of Light-Emitting Diodes." Light-Emitting Diodes, 1–26. doi:10.1017/cbo9780511790546.002.
- [3] Fundamentals of Solid-State Lighting. 2014. "White Inorganic LEDs," February, 261–276. doi:10.1201/b17076-18.
- [4] S. Liu, X. B. Luo, LED Packaging for Lighting Applications: Design, Manufacturing and Testing, Chemical Industry Press and John Wiley & Sons, Beijing, 2011.
- [5] Narendran Nadarajah, Lei Deng, Richard M. Pysar, Yimin Gu, Hua Yu. "Performance Characteristics of High-Power Light-Emitting Diodes." Third International Conference on Solid State Lighting. doi:10.1117/12.515647, 2004.
- [6] H. Yamamoto, T. Yamamoto. "Phosphors for White LEDs." Nitride Semiconductor Light-Emitting Diodes (LEDs), 144–180. doi:10.1533/9780857099303.1.144. 2014.
- [7] S. Ye, F. Xiao, Y. Pan, Y. Ma, Q. Zhang. Materials Science and Engineering: R: Reports 71(1), 1 (2010).
- [8] Sun Ching Cherng, Yu-Yu Chang, G. Hsun Yang. "Exploring Packaging Efficiency in Phosphor-Converted White LEDs." SPIE Newsroom, April. doi:10.1117/2.1201502.005739. 2015..
- [9] Liu Zong Yuan, Sheng Liu, Kai Wang, Xiao Bing Luo, IEEE Transactions on Components Packaging Technologies 33(4), 680 (2010).
- [10] Yu Renyong, Shangzhong Jin, Songyuan Cen, and Pei Liang. IEEE Photonics Technology Letters 22(23), 1765 (2010).
- [11] You Jiun Pyng, Yeong-Her Lin, Nguyen T. Tran, Frank G. Shi, Journal of Electronic Packaging 132(3): 031010. (2010).
- [12] Hu Run, Xiaobing Luo, Han Feng, and Sheng Liu, Journal of Luminescence 132(5), 1252 (2012). doi:10.1016/j.jlumin.2011.12.059.
- [13] Smet Philippe F., Anthony B. Parmentier, Dirk Poelman. Journal of The Electrochemical

Society 158(6) (2011). doi:10.1149/1.3568524.

- [14] Liu Zongyuan, Sheng Liu, Kai Wang, Xiaobing Luo IEEE Transactions on Device and Materials Reliability 9(1), 65 (2009). doi:10.1109/tdmr.2008.2010250.
- [15] Quoc Anh, N. D., M. F. Lai, H. Y. Ma, H. Y. Lee, Journal of the Chinese Institute of Engineers 38(3), 299 (2014). doi: 10.1080/02533839.2014.981214.
- [16] M. F. Lai, N. D. Quoc Anh, H. Y. Ma, H. Y. Lee. "Scattering Effect of Sio2 Particles on Correlated Color Temperature Uniformity of Multi-Chip White Light LEDs." Journal of the Chinese Institute of Engineers 39. 2014. doi: 10.1080/02533839.2015.1117950.
- [17] Soni, Abhishek Kumar, and Vineet Kumar Rai. 2016. "Optical Studies in Er3 Doped BaMoO4 Down converting Phosphor for Blue LEDs." doi:10.1063/1.4945122.
- [18] Liao Junyuan, Haibo Rao, Wei Wang, Xianlong Wan, Linsong Zhou, Da Zhou, Xuemei Wang, Qiaolin Lei. Journal of Semiconductors 34(5), 053008 (2013). doi:10.1088/1674-4926/34/5/053008.
- [19] Ryckaert Jana, Sven Leyre, Peter Hanselaer, Youri Meuret, "Simulation of White LEDs with a Planar Luminescent Layer Using the Extended Adding-Doubling Method." Optical Systems Design 2015: Illumination Optics IV. doi:10.1117/12.2191165. 2015.
- [20] J. Zhong, M. Xie, Z. Ou, R. Zhang, M. Huang, F. Zhao, Mie Theory Simulation of the Effect on Light Extraction by 2-D Nanostructure Fabrication, In Proceedings of 2011 Symposium on Photonics and Optoelectronics (SOPO), Wuhan, 16-18 May 2011: 1-4. New York: IEEE. 2011,
- [21] O. Tapaninen, Optical and thermal simulation chain for LED package, 17th International Conference on Thermal, Mechanical and Multi-Physics Simulation and Experiments in Microelectronics and Microsystems (EuroSimE), Montpellier. doi:10.1109/EuroSimE.2016.7463349. 2016,
- [22] Minh Tran Hoang Quang, Nguyen Huu Khanh Nhan, Nguyen Doan Quoc Anh, Hsiao-Yi Lee. Journal of the Chinese Institute of Engineers 40(4), 313 (2017). doi:10.1080/02533839.2017.1318720.
- [23] Y. Q. Li, J.E.J. Van Steen, J.W.H. Van Krevel, G. Botty, A.C.A. Delsing, F.J. Disalvo, G. De With, H.T. Hintzen Journal of Alloys and Compounds 417(1-2), 273 (2006) doi:10.1016/j.jallcom.2005.09.041.
- [24] T. H. Minh, N. H. Nhan, N. D. Anh, International Journal of Electrical Engineering and Informatics, **9**(1), 139 (2017). doi:10.15676/ijeei.2017.9.1.9