Enhancing Luminous Efficacy of White LED Lamp Using $Ca_2MgSi_2O_7:Eu^{2+}$ Phosphor

Doan Quoc Anh NGUYEN[1,*], *Thi Phuong Thao NGUYEN*[2], *Postava KAMIL*[3]

[1]Power System Optimization Research Group, Faculty of Electrical and Electronics Engineering, Ton Duc Thang University, Ho Chi Minh City, Vietnam
[2]Faculty of Electrical and Electronics Engineering, Ton Duc Thang University, Ho Chi Minh City, Vietnam
[3]Nanotechnology Centre, VŠB-Technical University of Ostrava, 17 listopadu 15, 708 33 Ostrava-Poruba, Czech Republic

*Corresponding Author: Nguyen Doan Quoc Anh (email: nguyendoanquocanh@tdt.edu.vn)
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Abstract. *Along with the rapid development of science and technology in today's society, lighting technology, especially white light-emitting diodes (WLEDs), has become a critical and essential element in almost every field of our daily life. Therefore, the enhancement of luminous efficiency have become an important objective in the production of white LEDs to serve the needs of customers. This paper presents the influence of $Ca_2MgSi_2O_7:Eu^{2+}$ green luminescent phosphor on the light quality of WLEDs. In this work, the green-emitting $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor is added into the in-cup phosphor configuration, which leads to the varying of the scattering property of this compounding. This is proved through the scattering coefficient from Mie-theory. Besides, the color quality scale which is called CQS index of WLEDs is also verified. The achieved results indicate that the luminous efficacy increases significantly if the $Ca_2MgSi_2O_7:Eu^{2+}$ concentration is varied. The smaller the $Ca_2MgSi_2O_7:Eu^{2+}$ size is used, the higher the luminous flux is obtained. However, the CQS can decrease if $Ca_2MgSi_2O_7:Eu^{2+}$ concentration is adjusted in another direction. Therefore, it is vital to select a suitable concentration and size of $Ca_2MgSi_2O_7:Eu^{2+}$ for enhancing the luminous efficacy of WLEDs, and the benefit of this phosphor in creating white WLED packages is a potential solution for developing LED illumination technology.*

Keywords

White LEDs, $Ca_2MgSi_2O_7:Eu^{2+}$, luminous efficacy, color quality scale.

1. Introduction

Recently, white light emitting diodes (WLEDs) have been more and more important and indispensable in our daily life because of their potential flexibility as well as dominant functions. With these outstanding advantages, WLEDs can easily be spread in many different illumination applications such as general lighting, medicine, consumer products [1]-[4]. The combination of numerous monochromatic LED chips or the mixture of blue light emitted from LED chip with $YAG:Ce^{3+}$ phosphor is one of the popular methods in white light emitting diode technology to generate white light which is also called phosphor converted LED. Nevertheless, the proccess of light's absorption going back to the package is very harsh, leading to the attained results of lumen output and angular colour uniformity just reach such bad quality. Hence, the purpose of generating WLEDs with high luminous efficiency, excellent colour uniformity, and great CRI has been developing in recent years [5]-[8]. Shi's team has proposed the green-emitting $Ca_7(PO_4)_2(SiO_4)_2$:Eu^{2+} phosphor for improving the optical properties of white light emitting diodes [9]. In 2016, Wang's group has applied perovskite $CsPbBr_3$ quantum dots for enhancing the luminous efficacy of green light-emitting diodes [10]. In 2017, Ma's team added Cr^{3+} to $Y_3Al_5O_{12}$:Ce^{3+} to enhance the color rendering index from 63.2 to 77.9. This study shows that Ce^{3+} and Cr^{3+} co-doped YAG phosphors are suitable for white light-emitting diodes [11]. In 2018, Lu's group has presented the synthesis and the photoluminescence characteristics of the $LiGd_3(MoO_4)_5$:Eu^{3+} red phosphor with high color purity and brightness. The results show that $LiGd_3(MoO_4)_5$:Eu^3 may be a promising red phosphor for white light-emitting diodes [12].

To serve the purpose of ameliorating and limiting the loss backscattered light inside LED chip, an opinion based on a structure with remote phosphor configuration designed in which the phosphor layer separated from chip is put forward [13]. Although the remote phosphor structure can give the better luminous efficiency than the dispensing and conformal structure, it is not easy to achieve the concave surface of this structure which leads to a non-uniform phosphor thickness in fabrication technology. Thus, it is really hard for luminous efficiency and uniform colour distribution of WLEDs to meet the demand of different cases in illumination applications. During the process of development, the optimization of remote phosphor structure is created via designing the concentric green, red phosphor rings to focus on lessening the problems of backscattering. With this propose, the extraction efficiency and the colour rendering property of WLEDs are more excellent than these of WLEDs with a mixed phosphor layer [14]-[18]. A different phosphor conformation with an innovative double remote micro-patterned phosphor film is used to enrich the colour homogeneity of WLEDs. On the other hand, the configuration of LED's lens need to be researched in many aspects to keep balance in optimization of uniform illumination and high light efficiency. Therefore, the luminescent material is concerned as a significant factor that supports the performance of multi-chip WLEDs, and hence its main function is to overwhelm the condition of poor light extraction and colour uniformity.

Although almost above approaches aim to improving the performance of WLEDs, the luminous efficiency and the colour quality of white LEDs are not completely gratified at many dissimilar illumination applications as well as extensive demands in the commercial LED lighting market [19]-[25]. In addition, there is only single-chip white LED lamps with low colour correlated temperature, which is chiefly concentrated. Moreover, in order to satisfy the competitiveness in lighting market and offer the better light quality, advanced studies might have been extensively conducted to find more optimal configuration of LEDs or a phosphor material that help the luminescent intensity and colour quality increase at such high temperatures.

This paper proposes a method in which the $Ca_2MgSi_2O_7$:Eu^{2+} phosphor compounding is applied to mix with the YAG:Ce^{3+} yellow phosphor compounding of the WLEDs. The main purpose of this combination is to optimize its light extraction, colour quality at high colour correlated temperature. In the process of simulation, the weight proportion and the size of phosphor particle need to be varied in order that it is exact to realize its influence on the optical performance of WLEDs. The optical properties of $Ca_2MgSi_2O_7$:Eu^{2+} and YAG:Ce^{3+} phosphors compounding are surveyed via analysing light absorption and scattering as well as light conversion in LEDs at high correlated colour temperature. In other words, simulations and calculations are fully taken advantage so that they can create a chance which find the positive factors of concentration and size of $Ca_2MgSi_2O_7$:Eu^{2+} green luminescent phosphor on luminous flux, colour quality. According to the obtained results, we recognize that $Ca_2MgSi_2O_7$:Eu^{2+} green phosphor does not only improve the luminescent properties but also can verify colour quality of the in-cup phosphor configuration.

2. PREPARATION AND SIMULATION

2.1. Material preparation

Green-emitting $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor is frequently employed for very high loading and long life-time fluorescent lamps due to the high quantum efficiency, chemical properties, and thermal stability of this material. $Ca_2MgSi_2O_7:Eu^{2+}$ composition comes from chemical processes among different materials such asCaO, MgO, SiO_2, Eu_2O_3, and NH_4Cl. This phosphor compounding can be considered as an efficient host candidate for good luminous efficacy of WLEDs [26]. The detail composition

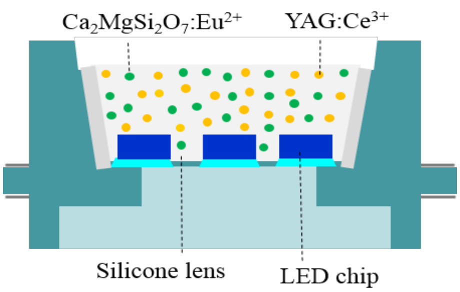


Fig. 1: (top) In-cup phosphor configuration; and (bottom) its 2-D illustration.

of green-emitting $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor is calculated in Table 2. Besides, the preparation of the material is described as three following steps: Firstly, we start to mix the material together by slurrying them in methanol plus a few cubic centimeters of water. Next, dry it in the air. When the mixture gets dry, we powderize it. Lastly, fire this powder in capped quartz tubes with N_2 at 1000°C for an hour. Then powderize it, and fire the material again in capped quartz tubes with CO, at 1150°C for an hour.

2.2. Simulation

It is vital to apply the LightTools 8.1.0 program and Mie-theory into this work. It helps WLEDs with in-cup phosphor structure be easily simulated through analyzing the scattering of phosphor particles and supports the process of investigating the influence of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor on the performance of the WLEDs at the high correlated temperature of 8500 K. In order to prepare for the process of the in-cup phosphor configuration of WLEDs, we blend the $Ca_2MgSi_2O_7:Eu^{2+}$ and $YAG:Ce^{3+}$ phosphor compounding as expressed in Fig. 1.

Consequently, the phosphor layer of WLEDs contains $Ca_2MgSi_2O_7:Eu^{2+}$ green phosphor, the yellow phosphor $YAG:Ce^{3+}$, and the silicone glues. The constituents of simulated WLEDs expressed in the model are blue chips, a reflector cup, a phosphor layer, and a silicone layer. A reflector with a 2.07 mm depth, a bottom length of 8 mm and a length of 9.85 mm at its top surface is bonded with these chips. The radiant power of each nice blue chip was designed with 1.16 W, a peak wavelength of 453 nm, as shown in Table 1. The radius of the green $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor particles were changed from 1 μm to 10 μm. to achieve the optimization of color uniformity and lumen output efficiency. The $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor particle density can be changed from 0 to 1.4% in simulation process. The refractive index of phosphor particle is set to be 1.85 and 1.83 for $Ca_2MgSi_2O_7:Eu^{2+}$ and $YAG:Ce^{3+}$ respectively. To maintain the average CCT of white LEDs at 8500 K, the $YAG:Ce^{3+}$ phosphor concentration need to change appropriately to the concentration of $Ca_2MgSi_2O_7:Eu^{2+}$. The optical simulation is performed along with the change in $Ca_2MgSi_2O_7:Eu^{2+}$ particle size and distribution so that the the effect of $Ca_2MgSi_2O_7:Eu^{2+}$ green phosphor on WLEDs could be accurately defined. The scattering of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor particle was analyzed by using the Mie-theory [14]. In addition, the scattering cross section Csca for spherical particles can be computed by the following expression through ap-

Talbe 1. The actual parameters of LED chip.

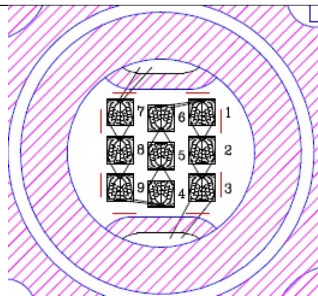
LED vender	Epistar
LED chip	V45H
Voltage (V)	$3.5 \sim 3.6$
Peak Wavelength (nm)	453
Power (mW)	$320 \sim 340$
Lead frame	4.7mm Jentech Size-S
Die attach	Sumitomo 1295SA
Bonding diagram	

Table 2. Composition of green-emitting $Ca_2MgSi_2O_7$:Eu^{2+} phosphor.

Ingredient	Mole (%)	By weight (g)	Molar mass (g/mol)	Mole (mol)	Ions	Mole (mol)	Mole (%)
CaO	7.4	112	56.0774	0.28	Ca^{2+}	0.56	0.045
MgO	26.2	40	40.304	0.99	Mg^{2+}	1.98	0.162
SiO_2	55.5	126	60.08	2.09	Si^{4+}	4.19	0.342
Eu_2O_3	0.26	3.5	351.926	0.01	O^{2-}	5.49	0.448
NH_4Cl	10.6	21.4	53.49	0.4	Eu^{2+}	0.02	0.003

plying the Mie theory:

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

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

where $k = \pi D/\lambda$ is particle size parameter, r is the particle radius, λ is the wavelength, $\psi_n(x)$ and $\xi_n(x)$ are the Riccati-Bessel functions, $\mu_{sca}(\lambda)$ is the scattering coefficient, and a_n, b_n are calculated by:

$$a_n(x,m) = \frac{\psi_n'(mx)\psi_n(x) - m\psi_n(mx)\psi_n'(x)}{\psi_n'(mx)\xi_n(x) - m\psi_n(mx)\xi_n{}'(x)} \tag{3}$$

$$b_n(x,m) = \frac{m\psi_n'(mx)\psi_n(x) - \psi_n(mx)\psi_n'(x)}{m\psi_n'(mx)\xi_n(x) - \psi_n(mx)\xi_n{}'(x)} \tag{4}$$

According to the received results shown in Fig. 2, the growth of scattering coefficient

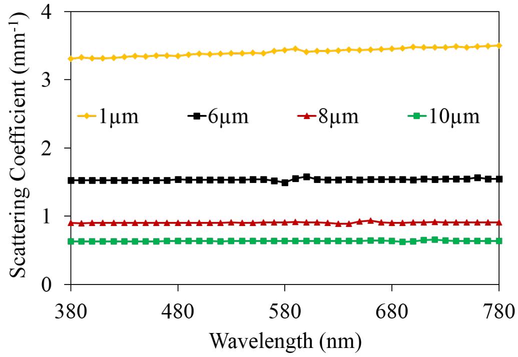


Fig. 2: The computed scattering coefficients of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor as a function of size.

exits parallel with the decrease of the size of $Ca_2MgSi_2O_7:Eu^{2+}$ particle, which proves the truth that the smaller the phosphor particles are, the stronger the abilities of scattering of incident light are and the more color uniform they achieve. At the size of 1 μm, the best luminous

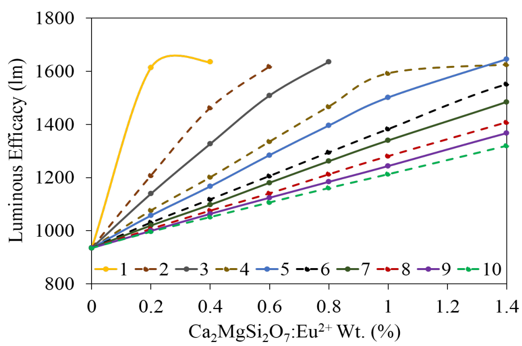


Fig. 3: The luminous efficacy of WLEDs as a function of the size and concentration of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor.

efficacy can be reached. Generally, the scattering coefficient is the same as the wavelengths varied from 380 nm to 780 nm. The scattering coefficient slightly increases with the changes of the wavelength at the size of 1 μm. Moreover, the scattering is slightly sensitive to the incident wavelength. The longer wavelength can lead to the stronger scattering. It implies that the scattering of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor benefits considerably to color quality.

3. RESULTS AND DISCUSSION

As demonstrated in Fig. 3, the obtained results show that $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor has a positive effect on the luminous efficacy of WLEDs and its luminescent properties are completely influenced by concentration and particle dimension. $Ca_2MgSi_2O_7:Eu^{2+}$ green luminescent phosphor is considered as a good element for green light enhancement and thermal condensation. Therefore, we have an idea that adding these particles to the YAG:Ce^{3+} compounding will help the LED achieves higher brightness at high temperature of 8500 K.

To obtain accurate results on the optical properties of $Ca_2MgSi_2O_7:Eu^{2+}$, the simulations with concentrations ranging from 0 to 1.4% corresponding to each size within $1 - 10$ μm are performed. The lines in the flow chart show that the luminous efficacy increases so fast and it can reach the maximum values in the concentration range of 0% to 0.6% and the size from 1 to 5 μm as depicted in the Fig. 3. Normally, the luminescence flux can be improved at all particle sizes when adding $Ca_2MgSi_2O_7:Eu^{2+}$ to the phosphor compound in concentration.

The small particles usually provide less luminous flux than bigger ones due to the unwanted backward scattering inside WLEDs. It means that the light is more likely to be trapped inside a package and there will be less light from the LEDs when the particles are applied. The intensity of light emission decreases and hardly reaches the maximum for small particles at a color temperature of 8500 K. Thus, the luminous efficacy of WLEDs using $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor with the size of 1 μm can decrease with further increase of concentration. This phenomenon occurs at 0.4% wt. of $Ca_2MgSi_2O_7:Eu^{2+}$. For the size of $2 - 3$ μm, the reduction of luminous efficacy occurs at 0.6% and 0.8% wt. of $Ca_2MgSi_2O_7:Eu^{2+}$, respectively. The increase of luminous efficacy grows with the size of $Ca_2MgSi_2O_7:Eu^{2+}$ particles, from 4 μm to 6 μm regardless the $Ca_2MgSi_2O_7:Eu^{2+}$ concentration. With the size of $6 - 10$ μm, the tendency of light propagates more vigorously in the forward direction and weakens in the opposite direction of the LED chip, so that large particle sizes show advantages for luminous efficacy. The main purpose of this study is not only to enhance luminous flux but also to verify the color quality scale (CQS). CQS index which can evaluate the overall color quality of WLEDs about color fidelity, chromatic discrimination, and observer preferences shows slight decrement around particle sizes $6-10$ μm as Fig. 4. The CQS values slightly decrease within the size of 6 to 10 μm. The $Ca_2MgSi_2O_7:Eu^{2+}$ green luminescent phosphor can lead to green light supplementation and deficiency of red components in the spectrum of WLEDs, causing the reduction of CQS.

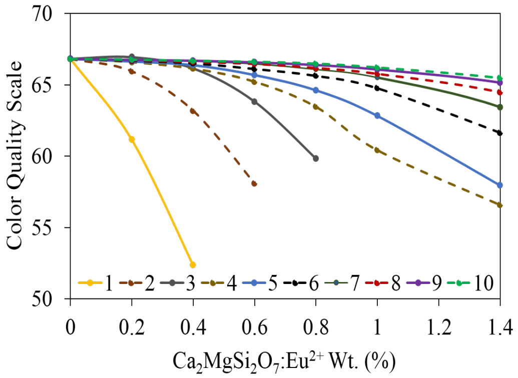


Fig. 4: The color quality scale of WLEDs as a function of the size and concentration of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor.

4. CONCLUSIONS

In this article, the influence of $Ca_2MgSi_2O_7:Eu^{2+}$ green luminescent phosphor on WLEDs is presented and demonstrated, which helps us be aware of the possibility of improvement both luminous flux and color uniformity at high color correlated temperature basing on the optical simulation. We analyzed the impact of phosphor particle size and concentration of $Ca_2MgSi_2O_7:Eu^{2+}$ on luminous flux and CQS. The highest luminous flux accompanies with insignificant decrease in CQS in this scope. As a result, by adding $Ca_2MgSi_2O_7:Eu^{2+}$ green phosphor with the concentration range from $0-1.4\%$ and the particle size from $6-8$ μm into phosphor compounding, we can obtain better optical performance of white LEDs. Hence, the employment of $Ca_2MgSi_2O_7:Eu^{2+}$ phosphor to white WLED packages is a promising solution for developing LED illumination technology in the future.

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About Authors

Doan Quoc Anh NGUYEN was born in Khanh Hoa province, Vietnam. He has been working at the Faculty of Electrical and Electronics Engineering, Ton Duc Thang University. Quoc Anh received his PhD degree from National Kaohsiung University of Applied Sciences, Taiwan in 2014. His research interest is optoelectronics (such as Multi-chip white light LEDs, free-form lens, optical material).

Thi Phuong Thao NGUYEN was born in Quang Ngai province, Vietnam. She graduated from the University of Technology, Ho Chi Minh City, Viet Nam. She received her B.Eng and M.Eng degrees in automatic control engineering in 2006 and 2009, respectively. Now, she is teaching at the department of electrical and electronics engineering, Ton Duc Thang University, Ho Chi Minh city, Vietnam. Her research interests include an automatic control system and optoelectronics.

Postava KAMIL (born in 1970) is an associate professor with the Department of Nanotechnology Centre, Technical University of Ostrava, Czech Republic. He received his Ph.D degree in optical in 1997 at the Institute National des Sciences Appliquees Toulouse, France and Palacky University Olomouc, Czech Rep. Topics of his research interests are magneto-optical ellipsometry, linear and quadratic magneto-optic effects, magneto-optic vector magnetometry, magneto-optic microscopy, magneto-optic waveguides and isolators, spin-lasers, optics of thin films, light propagation in anisotropic layered media.