

TELKOMNIKA, Vol.17, No.5, October 2019, pp.2643~2649

ISSN: 1693-6930, accredited First Grade by Kemenristekdikti, Decree No: 21/E/KPT/2018

DOI: 10.12928/TELKOMNIKA.v17i5.11038

2643

Improving luminous flux and color homogeneity of dual-layer phosphor sctructure

Tran Thanh Trang¹, Phan Xuan Le², Nguyen Doan Quoc Anh*³

¹Faculty of Engineering and Technology, Van Hien University, Ho Chi Minh City, Vietnam ²National Key Labrolatory of Digital Control and System Engineering, Ho Chi Minh City, Vietnam ³Power System Optimization Research Group, Faculty of Electrical and Electronics Engineering, Ton Duc Thang University, Ho Chi Minh City, Vietnam *Corresponding author, e-mail: nguyendoanquocanh@tdtu.edu.vn

Abstract

In order to clarify the main purpose of the study, we put a green phosphor layer SrBaSiO4:Eu2+ on the yellow phosphorus layer YAG:Ce3+ through using only one WLEDs structure in different color temperatures like 5600 K, 6600 K, 7700 K. Then, we find the suitable SrBaSiO4:Eu²+ concentration in order that the luminous flux could get the highest value. The results show that SrBaSiO4:Eu²+ brings great benefits to increase not only optical gain but also color uniformity. Specifically, the greater the SrBaSiO4:Eu²+ concentration, the greater the output of WLEDs because of the development of green light component in WLEDs. However, only if the SrBaSiO4:Eu²+ concentration exceeds the level, a slight decrease in color rendering index (CRI) can occur, which based on Monte Carlo simulation. In addition, the results of this paper have contributed significantly to the creation of higher-powered WLEDs.

Keywords: color uniformity, dual-layer phosphor, luminous efficacy, Mie-scattering theory, SrBaSiO₄:Eu²⁺

Copyright © 2019 Universitas Ahmad Dahlan. All rights reserved.

1. Introduction

In recent times, LEDs has been considered as a promising product in the future owing to its superior properties for lighting technology such as high illumination, high energy efficiency, and long lifetimes [1-5]. Therefore, MCW-LEDs is extensively applied in a variety of general-purpose illumination applications, which completely replaced all of the traditional light sources [6]. Generally, the luminous output and the correlated color temperature uniformity are regarded as the most important factors that determine the quality of LED performance. With suitable concentration of SiO₂ particles added into phosphor layer, angular CCT distribution can be optimized without considerable affecting luminous output [7-12]. In addition, the lumen output of MCW-LEDs increases significantly with the Ce_{0.67} Tb_{0.33} MgAl₁₁O₁₉:Ce, Tb concentration added to MCW-LEDs phosphor [13]. With the aim of improving the CRI of MCW-LED greater than 90, several former approaches have been introduced to reimburse the red-light of MCW-LEDs such as combining red-phosphors with phosphor layer or injecting red LEDs [14-20]. Won group combined blue LEDs, green (Ba,Sr) 2SiO4:Eu2+ and red CaAlSiN₃:Eu²⁺ phosphors with various packages to propose great CRI value MCW LEDs [21]. Chen group have executed experiments of adding the missing red part in the phosphor converted white light-emitting diodes (pc-WLEDs) [22]. Nevertheless, these researches solely examine individual pc-WLED structures. Furthermore, the researches simply concentrate on single-chip white LED lamps having CCT varying from 2500 K to 7500 K, but the truly improvement of color rendering ability of MCW-LED lamps having higher CCTs has not been deeply researched [23-26].

Eu²+-activated strontium—barium silicate (SrBaSiO₄:Eu²+) phosphor has been applied extensively in lighting technology to contribute red light [27]. Due to the great properties as well as excellent luminescence efficiency, color purity and stability, green phosphor SrBaSiO₄:Eu²+ have met all the requirements for a favorable red-emitting phosphor. However, SrBaSiO₄:Eu²+ has not been widely applied to enhance color homogeneity and luminous flux yet.

In this paper, we review the influence of SrBaSiO₄:Eu²⁺ particles in luminous flux and color homogeneity of MCW LEDs in conformal phosphor packages. The pc-WLEDs green-light emitting can be controlled by the presence of SrBaSiO₄:Eu²⁺ phosphor particles. As a result,

2644 ■ ISSN: 1693-6930

the LED light distribution can be adjusted by LED packages to higher luminous flux region. Our study progress is divided into three main steps. First, the precise MCW-LED physical model with average CCTs approximately 5600 K, 6600 K and 7700 K are built by LightTools 8.1.0 program. Then, SrBaSiO₄:Eu²⁺ particles are mixed into the phosphor layers. These particles interact with transmitted light result. Lastly, we examine the impacts of SrBaSiO₄:Eu²⁺ phosphor concentration on MCW-LEDs. The simulation results demonstrated that the proposed approach can increase the luminous flux and color homonegeity substantially.

2. Research Method

2.1. Preparation of green SrBaSiO₄:Eu²⁺ Phosphor

SrBaSiO₄:Eu²⁺ particles, with an emission peak of 2.36 eV and many outstanding characteristics such as high quantum efficiency and stability at high temperature, are known as a type of yellow-green phosphor and become more and more popular. The elements which can directly influence on the luminescence properties of SrBaSiO₄:Eu²⁺ phosphor are their particle sizes and concentration. Its composition includes SrCO₃, BaCO₃, SiO₂, Eu₂O₃, NH₄Cl, and Eu²⁺ ion, all of which are used as raw materials as shown in Table 1. Moreover, SrBaSiO₄:Eu²⁺ is applied particularly for very high-loading and long life-time fluorescent lamps. Therefore, it is one of the most popular commercialized oxide phosphors. On the whole, the fabrication process of SrBaSiO₄:Eu²⁺ is described specifically as the following:

Table 1. Composition of Green-Emitting SrBaSiO₄:Eu²⁺ Phosphor

Ingredient	Mole (%)	By weight (g)	Molar mass	Mole	Ions	Mole	Mole
			(g/mol)	(mol)		(mol)	(%)
SrCO₃	31.28	145	147.63	0.982	Sr ²⁺	0.982	0.088
BaCO ₃	31.79	197	197.34	0.998	Ba ²⁺	0.998	0.090
SiO ₂	33.40	63	60.08	1.049	Si ⁴⁺	1.049	0.094
Eu_2O_3	0.32	3.5	351.926	0.01	O ²⁻	8.068	0.726
NH ₄ CI	3.22	5.4	53.49	0.101	Eu ²⁺	0.02	0.002

Firstly, the materials are slurried in the water to be mixed together. Then they are ball-milled in the water into small particles. After that, the materials will be powdered as soon as they are dried in the air. Secondly, this powder will be fired in capped quartz tubes with CO at 1100°C within an hour and powderized by dry milling. Then 5.4 g NH₄Cl will be added into this mixture and they are mixed together by dry milling. Next, they are fired again in capped quartz tubes with CO at 1100°C within an hour and will be powderized. Finally, the materials are washed in the water for several times (pH ranges from 10 to 12) and dried after that.

2.2. Construction of MC-WLEDs

According to LightTools 8.5.0 program and Monte Carlo method, the phosphor layer of real MCW-LEDs is simulated with flat silicone layer. This modeling process is caried out through two main periods: (1) the mechanical structures and optical properties of MCW-LED lamps must to be set and built (2) Then the optical influences of phosphor compounding through the diversity of SrBaSiO₄:Eu²⁺ concentration are well monitored.

In order to understand the influence of YAG:Ce³+ and SrBaSiO₄:Eu²+ phosphor compounding on the performance of MCW-LED lamps, we must make some comparisons. Among them, the two types of compounding which have average CCTs of 5600 K, 6600 K, and 7700 K, dual-layer remote phosphor, is considered to clarify. In Figure 1 (a), there is a clearly description about MCW-LED lamps with conformal phosphor compounding having average CCT of 8500K. It also indicates the simulation of MCW-LEDs in which the components do not contain SrBaSiO₄:Eu²+. The reflector has a bottom length of 8 mm, a height of 2.07 mm and a length of 9.85 mm at its top surface. The conformal phosphor compounding, whose thickness is fixed at 0.08 mm, covers nine chips. Each LED chip with the square base area of 1.14 mm² and the height of 0.15 mm is attached to the cavity of the reflector. The radiant flux of each blue chip is 1.16 W while 453 nm is the peak wavelength.

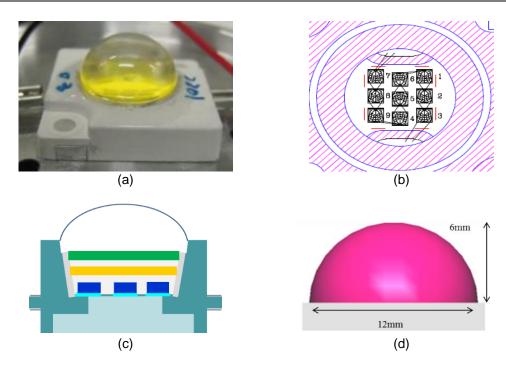


Figure 1. Photograph of WLEDs structure: (a) actual WLEDs, (b) bonding diagram, (c) illustration of pc-WLEDs model, (d) simulation of WLEDs using light tools commercial software

3. Results and Analysis

Figure 2 shows the antipodal change between the SrBaSiO₄:Eu²⁺ green phosphorus concentration and the yellow phosphorus YAG:Ce³+. This change hiddenes two meanings: one is maintaining average CCTs and the other is this change directly affects the scattering and absorption of two phosphoric layers in WLEDs. This inevitably affects the color quality and luminescence generated by WLEDs. Thus, the choice of concentration SrBaSiO₄:Eu²⁺ determines the color quality of WLEDs. When SrBaSiO₄:Eu²⁺ concentration increased from 2-20% wt, YAG:Ce³⁺ concentration decreased to keep average CCTs. This phenomenon is the same for WLEDs with all the condition of different color temperatures of 5600 K, 6600 K, and 7700 K.

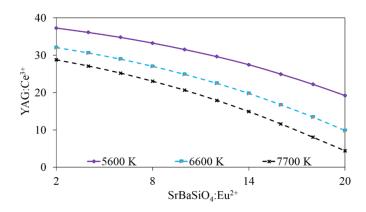


Figure 2. The change of phosphor concentration for keeping the average CCTs

It is not difficult to be seen the most noticeable effect of the SrBaSiO₄:Eu²⁺ green phosphorus concentration on the emission spectrum of WLEDs, which is shown in Figures 3-5. Besides, a choice will be made according to the requirements of manufacturer. In

2646 ■ ISSN: 1693-6930

the case of the requirement of WLEDs with high color, a small amount of flux can be considered to reduce. White light is known as the synthesis of the spectral region as shown in Figures 3, 4 and 5. These three represent emission spectra of 5600 K, 6600 K and 7700 K, respectively. The trend of the red light spectrum from 648 nm to 738 nm was found to increase with SrBaSiO₄:Eu²⁺ concentration. However, this is not significant unless there is an increase in the spectrum of the two regions 420-480 nm and 500-640 nm. The two-zone 420-480 nm of the spectrum enhancement helps blue-light scattering. The higher the color temperature, the higher the emission spectra as well as the higher the color and luminosity. This is a very important result when applying SrBaSiO₄:Eu²⁺. Mostly, it is very difficult to control the color quality of WLEDs. This study implies that SrBaSiO₄:Eu²⁺ can improve color quality of WLEDs including low color temperature (5600 K) and high (7700 K).

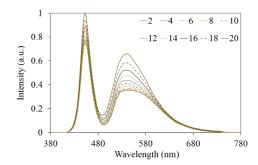


Figure 3. The emission spectra of 5600 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

Figure 4. The emission spectra of 6600 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

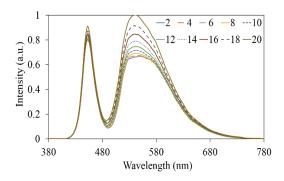


Figure 5. The emission spectra of 7700 K WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

As shown in Figure 6, the color deviation decreases with the SrBaSiO₄:Eu²⁺ phosphorus concentration in all three CCTs. This can be explained by the absorption of red phosphorus. When SrBaSiO₄:Eu²⁺ phosphor absorbs blue light from the LED chip, the blue phosphor particles turn blue light into green light. Except the blue light from the LED chip, SrBaSiO₄:Eu²⁺ particles still absorb the yellow light. However, if it is compared to these two absorptions, the blue light absorbed by the LED chip is stronger due to the absorption properties of the material. Thus, the green light composition in WLEDs increases with the addition of SrBaSiO₄:Eu²⁺, which leads to an increase in colorimetric index. In the selection of modern WLED lamps, color uniformity is one of the important parameters. Obviously, the higher the color rendering index, the higher the price of white light WLED. However, the benefit of using SrBaSiO₄:Eu²⁺ is low cost. Thus, SrBaSiO₄:Eu²⁺ can be widely applied. Obviously, the using of SrBaSiO₄:Eu²⁺ can increase the color quality of white light of WLEDs with dual-layer phosphor structure. This is an important result of the study with the aim of improving color quality. However, it cannot be ignored the second advantage of the SrBaSiO₄:Eu²⁺ to the emission of FPGAs is shown in Figure 7. It is easy to see that the rise in luminous flux

increases significantly as SrBaSiO₄:Eu²⁺ concentrations increase in all CCTs. To prove this result, we consider expressions from (1) to (5).

This part will present and demonstrate the mathematical model of the transmitted blue light and converted yellow light in the double-layer phosphor structure, from which a huge improvement of LED efficiency can be obtained. The transmitted blue light and converted yellow light for single layer remote phosphor package with the phosphor layer thickness of 2*h* are expressed as follows:

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

$$PY_{1} = \frac{1}{2} \frac{\beta_{1} \times PB_{0}}{\alpha_{B1} - \alpha_{Y1}} (e^{-2\alpha_{Y1}h} - e^{-2\alpha_{B1}h})$$
(2)

The transmitted blue light and converted yellow light for double layer remote phosphor package with the phosphor layer thickness of *h* are defined as:

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

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

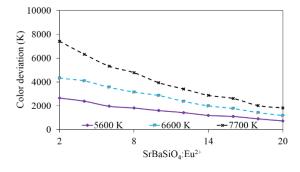
where h is the thickness of each phosphor layer. The subscript "1" and "2" are used to describe single layer and double-layer remote phosphor package. β presents the conversion coefficient for blue light converting to yellow light. γ is the reflection coefficient of the yellow light. The intensities of blue light (PB) and yellow light (PY) are the light intensity from blue LED, indicated by PB_0 . α_B ; α_Y are parameters describing the fractions of the energy loss of blue and yellow lights during their propagation in the phosphor layer respectively.

The lighting efficiency of pc-LEDs with the double-layer phosphor structure enhances considerably compared to a single layer structure:

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

from (5) we can see that the light output of WLEDs with the dual-layer remote phosphor is greater than the single-layer phosphor. Thus, the paper demonstrates the effective output of the dual-layer remote phosphor layer.

However, this color homogeneity is only one factor that evaluates the color quality of WLEDs. This cannot say good color quality only if high color uniformity index. Thus, some current studies provide a color rendering index (CRI), which can evaluate the true color of the object when the light is shining. The amount of green light goes up, which causes the color imbalance between the three main colors: blue, yellow and green. This affects the color quality of WLEDs, which decreases the color accuracy of WLEDs. Specifically, Figure 8 shows the mitigation of CRI in the presence of the SrBaSiO₄:Eu²⁺ remote phosphor layer.



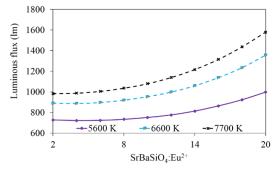


Figure 6. The color deviation of WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

Figure 7. The luminous flux of WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

2648 ■ ISSN: 1693-6930

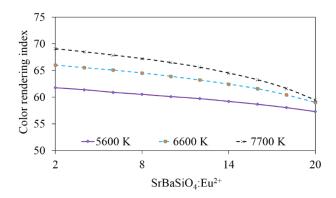


Figure 8. The color rendering index of WLEDs as a function of SrBaSiO₄:Eu²⁺ concentration

4. Conclusion

This paper presents the effect of green phosphor SrBaSiO₄:Eu²⁺ on the optical properties of the dual-layer phosphor structure. Based on Monte Carlo simulation simulations, the study demonstrated that SrBaSiO₄:Eu²⁺ is the appropriate choice for enhancing color uniformity. This is not only true for WLEDs with a low color temperature of 5600 K but also for color temperatures above 7700 K. Thus, this research has achieved a goal of improving color and luminous flux. However, there is still a small disadvantage for CRI that is when the concentration of SrBaSiO₄:Eu²⁺ exceeds, CRI decreases dramatically. Therefore, the choice of suitable concentration becomes important, depending on the manufacturer's goal and the article provided important information for reference in the production of color-coordinated WLEDs and better luminous flux.

Acknowledgements

This research was supported by National Key Labrolatory of Digital Control and System Engineering (DCSELAB), HCMUT, VNU-HCM.

References

- [1] Anh NDQ, Lee HY. Improving the Angular Color Uniformity and the Lumen Output for White LED Lamps by Green Ce_{0.67}Tb_{0.33}MgAl₁₁O₁₉:Ce,Tb Phosphor. *Journal of the Chinese Institute of Engineers*. 2016; 39(7): 871-875.
- [2] Myoung SJ, Yong HC, Shulu W, Tae GL, Jae SY. Material properties of the Ce³⁺-doped garnet phosphor for a white LED application. *Journal of Information Display*. 2016; 17(3): 117-123.
- [3] Katja MR, Marta KG, Grega B, Matej BK. White LED compared with other light sources: age-dependent photobiological effects and parameters for evaluation. *International Journal of Occupational Safety and Ergonomics*. 2015; 21(3): 391-398.
- [4] Anh NDQ, Lee HY, Tran TP, Nguyen HKN, Tran HQM, Truong HL. Y₂O₃:Eu³⁺ phosphor: a novel solution for an increase in color rendering index of multi-chip white LED packages. *Journal of the Chinese Institute of Engineers*. 2017; 40(3): 228-234.
- [5] Anh NDQ, Thuong TN, Lee HY. Selection of scattering enhancement particles for improving color homogeneity and luminous flux of phosphor-converted LEDs. *Journal of the Chinese Institute of Engineers*. 2017: 40(4): 307-312.
- [6] Guan RF, Hou GH, Li QQ. Synthesis and characterisation of CaMoO₄:Eu³⁺, Eu³⁺/Sm³⁺, Eu³⁺/Sm³⁺/Li⁺ red phosphor for white LED. *Materials Technology*. 2014; 29(3): 152-158.
- [7] Markus S, Tobias R, Oliver O, Wolfgang S. Highly Efficient pc-LED Phosphors Sr_{1-x} Ba_xSi₂O₂N₂:Eu²⁺_(0x1) Crystal Structures and Luminescence Properties Revisited. Critical *Reviews in Solid State and Materials Sciences*. 2014; 39(3): 215-229.
- [8] Santa C, Tapashree R, Kanishka M, Ashish Y. Red enhanced YAG:Ce, Pr nanophosphor for white LEDs. *Journal of Experimental Nanoscience*. 2012; 9(8): 776-784.
- [9] Jia Z, Cheng J. Photoluminescence properties of emission-tunable Ca₈MgLa(PO₄)₇:Eu²⁺, Mn²⁺ phosphors for white LEDs. *Optical Materials Express*. 2014; 4: 2102-2107.
- [10] Heleen FS, Reinert V, Jonas JJ, Dirk P, Philippe FS. K₂SiF₆:Mn⁴⁺ as a red phosphor for displays and warm-white LEDs: a review of properties and perspectives. *Optical Materials Express*. 2017; 7: 3332-3365.

- [11] Zhijun W, Shuqin L, Panlai L. Single phase tunable warm white-light-emitting Sr₃La(PO₄)₃:Eu²⁺, Sm³⁺ phosphor for white LEDs. *Opt. Mater. Express.* 2016; 6: 114-124.
- [12] Ji WM, Bong GM, Jin SK, Myoung SJ, Kang MO, Kwan YH, Jae SY. Optical characteristics and longevity of the line-emitting K₂SiF₆:Mn⁴⁺ phosphor for LED application. *Optical Materials Express*. 2016; 6: 782-792.
- [13] Shuyun Q, Yanlin H, Taiju T, Wei H, Hyo JS, Versatile luminescence of Eu^{2+,3+}-activated fluorosilicate apatites M₂Y₃[SiO₄]3F (M = Sr, Ba) suitable for white light emitting diodes. *Optical Materials Express*. 2014; 4: 396-402.
- [14] Jia Z, Baowei J, Zhenghe H. Investigations on the luminescence of Ba₂Mg(PO₄)₂:Eu²⁺,Mn²⁺ phosphors for LEDs. Optical Materials Express. 2016; 6: 3470-3475.
- [15] Yurong S, Ge Z, Masayoshi M, Yasuo S, Yuhua W, Photoluminescence of green-emitting Ca7(PO4)2(SiO4)2:Eu2+ phosphor for white light emitting diodes. Optical Materials Express. 2014; 4: 280-287
- [16] Qiaosong C, Fanliang Z, Ning Y, Haiyuan X, Stefania B, Alessia C, Mauro F, Guorong C. Enhanced and shortened Mn²⁺ emissions by Cu⁺ co-doping in borosilicate glasses for W-LEDs. Optical Materials Express. 2015; 5: 51-58.
- [17] Jia Z, Zhenghe H, Shizheng W. Generation of tunable-emission in Li₄Ca_{1-x}Sr_{0.96 +x}(SiO4)₂:0.04Eu²⁺ phosphors for LEDs application. *Optical Materials Express*. 2015; 5: 1704-1714
- [18] Ge Z, Zhipeng C, Qian W, Yurong S, Yuhua W. Full-color emission generation from single phased phosphor Sr₁₀[(PO₄)_{5.5}(BO₄)_{0.5}](BO₂):Ce₃₊, Mn²⁺,Tb³⁺ for white light emitting diodes. *Optical Materials Express*. 2013; 3: 1810-1819.
- [19] Ran M, Chaoyang M, Jiantao Z, Jiaqi L, Zicheng W, Xuanyi Y, Yongge C. Energy transfer properties and enhanced color rendering index of chromaticity tunable green-yellow-red-emitting Y₃Al₅O₁₂:Ce³⁺,Cr³⁺ phosphors for white light-emitting diodes. *Optical Materials Express*. 2017; 7: 454-467.
- [20] Jie H, Xinghong G, Jianhua H, Yujin C, Yanfu L, Zundu L, Yidong H. Near ultraviolet excited Eu³⁺ doped Li₃Ba₂La₃(WO₄)₈ red phosphors for white light emitting diodes. *Optical Materials Express*. 2016; 6: 181-190.
- [21] Luting W, Shuanglong Y, Yunxia Y, Francois C, Franck T, Guorong C. Luminescent properties of novel red-emitting phosphor: Gd₂O₂CN₂:Eu³⁺. Optical Materials Express. 2015; 5: 2616-2624.
- [22] Ching TL, Yu TS, Hsin YL. Nanorod-structured flip-chip GaN-based white light-emitting diodes. Proc. SPIE 9003, Light-Emitting Diodes: Materials, Devices, and Applications for Solid State Lighting XVIII. 2014; 90030I.
- [23] Su JY, Ji HO, Keyong NL, Young RD. Realization of quantum dot-based polarized white LEDs using short-wavelength pass dichroic filters and reflective polarizer films. Proc. SPIE 9190, Thirteenth International Conference on Solid State Lighting. 2014; 919012.
- [24] Zetian M, Hieu PTN, Shaofei Z, Ashfiqua TC, Md GK, Qi W, Ishiang S. Phosphor-free InGaN/GaN/AIGaN core-shell dot-in-a-wire white light-emitting diodes. Proc. SPIE 9003, Light-Emitting Diodes: Materials, Devices, and Applications for Solid State Lighting XVIII. 2014; 900306
- [25] Ashok KL, Saroj KP, Sandeep K, Sumitra S, Suchandan P, Chenna D. Theoretical analysis of blue to white down conversion for light-emitting diode light with yttrium aluminum garnet phosphor. *Journal of Photonics for Energy*. 2014; 4(1): 043596.
- [26] Cosmo MDS, Artur SGN, Luciano AB. Novel samarium/erbium and samarium/terbium codoped glass phosphor for application in warm white light-emitting-diode. Proc. SPIE 9003, Light-Emitting Diodes: Materials, Devices, and Applications for Solid State Lighting XVIII. 2014; 90031G.
- [27] Yen WM, Weber MJ. Inorganic Phosphors: Compositions, Preparation and Optical Properties. CRC Press, LLC, 2000 N.W. Corporate Blvd., Boca Raton, Florida 33431. 2004.