High stability in color chromaticity of warm white emitting diode with dual-hue SrSi₂O₂N₂:Eu²⁺,Yb²⁺ phosphor

My Hanh Nguyen Thi¹, Phan Xuan Le², Nguyen Le Thai³

¹Faculty of Mechanical Engineering, Industrial University of Ho Chi Minh City, Ho Chi Minh City, Vietnam ²Faculty of Mechanical-Electrical and Computer Engineering, School of Engineering and Technology, Van Lang University, Ho Chi Minh City, Vietnam ³Feculty of Engineering and Technology Technology III (Chi Minh City, Vietnam)

³Faculty of Engineering and Technology, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam

Article Info

Article history:

Received Nov 30, 2021 Revised Jun 14, 2022 Accepted Jul 16, 2022

Keywords:

Color rendering index Lumen efficacy Mie-scattering theory SrSi₂O₂N₂:Eu²⁺,Yb²⁺ WLEDs

ABSTRACT

In this study, we analyzed and developed a phosphor with a nitridiosilicate based and emitting color changing from green to orange to apply to our white-light-emitting diodes (WLEDs). Eu²⁺ and Yb²⁺ trap sites for twodoped SrSi₂O₂N₂:Eu²⁺,Yb²⁺ (SrYb) nitridosilicate emit a wide emission in the region between green and orange. By calculating the decompose time of the green-emission power donor, we could determine the converted energy between the active ions. Furthermore, we also analyzed the impact of codoping with varied active ion ratios on the photoluminescence (PL) characteristics. As the concentration of Yb2+ increases, the red emission dominates the green. This was because the obtained phoshor's emission depended on the activator ion components. To generate white light, a combination containing the acquired phosphor and the indium gallium nitride (InGaN) blue LED chip with 450-nm wavelength was used. With only two steps, we could detect the hue balance management. Firstly, by modifying the Eu²⁺ and Yb²⁺ concentrations, we can conduct a green-toorange proportion optimization. Following that, the commission on illumination (CIE) coordinates were transformed to [0.4071; 0.3789] from the original position, which was [0.2805; 0.2014] by enhancing the phosphor powder amount. An environment which has a CRI of around 89 is the optimum condition for conducting white light.

This is an open access article under the <u>CC BY-SA</u> license.



Corresponding Author:

Phan Xuan Le Faculty of Mechanical-Electrical and Computer Engineering, School of Engineering and Technology, Van Lang University Ho Chi Minh City, Vietnam Email: le.px@vlu.edu.vn

1. INTRODUCTION

On account of their many benefits over incandescent lights in being efficient and ecologically friendly while providing notable dependability, portable size, extended life span, etc, InGaN-based white-light-emitting diodes (WLEDs) have been considered to be an alter for next-generation lighting. The integration among InGaN blue LED chip as well as $YAG:Ce^{3+}$ is the most conventional way to generate white light [1]-[3]. Still this method still lies a disadvantage in application due to the hue imbalance on the genuine color rendering index (CRI) when there is a lack of red in the components. Therefore, an light-emitting diodes (LED) with a multi-based phosphor was utilized to improve the red LED. As compared to the YAG:Ce³⁺ blue LED chip method, the multiple-phosphor approach has numerous benefits, such as adequate color rendering (Ra), high efficiency, and luminosity [4]-[6]. Phosphors have varying thermal quenching

temperatures, creating aberrations in color rendering along with raising manufacturing costs for reality applications. As a result of re-absorption, multi-phosphor-based LEDs lose luminous efficiency due to the varied phases of phosphors. Full-color emitting mono phosphor centered in oxide and silicate compounds has been applied to produce steady color rendering. Recently, downconversion nitridosilicate and oxynitridosilicate based have shown remarkable thermal stability and great luminous efficacy. It was reported that both the positive learning environments (PLE) characteristics and configuration of the Eu²⁺ and Yb²⁺ doped SrSiO₂N₂ has wide emission band from green to orange with the peak half maximum 170 nm [7]-[9]. Additionally, external quantum efficacy is about 16% when the excitation is below 450 nm. Yb²⁺ ions can carry energy from to the Eu²⁺ ions. We describe the design and PLE characteristics of a duo-color emitting SrSi₂O₂N₂-xEu²⁺,yYb²⁺. To generate warm white light with high color rendering index (CRI) and stable CIE across a broad range of forward-bias current, the above phosphor was utilized as a single light converter in conjunction with blue-emitting LED chip.

2. EXPERIMENTAL DETAILS

SrYb was synthesized using a combination of Si₃N₄, SrCO₃, Eu₂O₃ and Yb₂O₃. We acquired all of our components from Aldrich Company and utilized them without any additional processing. It was combined well in an agate mortar with ethanol and dried for 2 hours in a 120 °C oven. Alumina-cruciblepackaged powder combinations were burned at 1400 °C for 8 hours in a horizontal tube furnace made also of the same material as the crucible using N2/H2 (5%) gas. Sr²⁺ ion concentration was maintained at 2%, whereas Yb²⁺ concentration was changed from 0% to 8%. After fired, a slightly compacted sample was obtained, which was then ground into a powder form. To create white light, phosphor powder was used. For 2 hours at 120 °C, the phosphor powder was combined with silicon resin (EG6301A and EG6301A from the Dow Chemical Company), then placed on top of an the indium gallium nitride (InGaN) blue-emitting chip. X-ray diffractometer (Cu K (=1.542 A) at RT was used to determine phase compositions. A scanning microscope was used to examine the powder's microstructure (S-5000, Hitachi Ltd., Tokyo, Japan). An F-700 Fluorescent Spectrophotometer from Hitachi Ltd. in Tokyo, Japan, was used to record the PLE spectra at room temperature using a 150 W Xe lamp. Streak camera C4334 observed decay time (Hamamashu, Japan). Using the integrated sphere technique, the quantum efficiency was measured. When it comes to thermal quenching, PTE-VUVD2L-100 was used to test it. Two-exponential decay curve (1) is a good approximation of the luminescence decay curves [10]-[12].

$$I = I_0 + A_1 e^{-\frac{t}{\tau_1}} + A_2 e^{-\frac{t}{\tau_2}}$$
(1)

For example, I is the illuminating strength and t is the period. Both A_1 and A_2 are constant factor. The lifespans for exponential compositions are accordingly τ_1 and τ_2 . On the basis of the estimated values for all of the above factors, as shown in (2) may be used to compute the average decay time (τ^*) [13]-[15].

$$\tau^* = (A_1\tau_1^2 + A_2\tau_2^2)/(A_1\tau_1 + A_2\tau_2) \tag{2}$$

3. RESULTS AND ANALYSIS

Figure 1 shows that SrYb (green) and YAG:Ce³⁺ (yellow) As a result of this modification, the average correlated color temperature (CCT) values will be preserved, however, the white-light-emitting diodes (WLEDs) absorption and scattering of two layers will be affected, and concentrations decrease reversely [16]-[18]. This would alter the hue output as well as light flux performance for WLEDs. Consequently, the WLEDs hue standard is determined by the concentration of SrSi₂O₂N₂:Eu²⁺,Yb²⁺. For example, when YAG:Ce³⁺ concentration increased from 2 to 30% Wt., the SrYb concentration decreased to keep the mean CCTs. Similarly, WLEDs with a hue heat range of 5,600 K to 8,500 K provide the same effect.

We can see from Figure 2 that SrYb concentration has a direct influence on the transmittance spectrum of WLEDs. Decisions can be made based on the manufacturer's needs. High hue quality WLEDs could cause descending light flux quality. As seen in Figure 2, white-light emission is a synthesis of the spectral range. The figure shows emission data at 3,000 K. SrYb concentration rises the intensity of two spectrum scopes, 420-480 nm as well as 500-640 nm. Due to an intensity growth in the two-band emission spectrum, the output luminance has increased. Blue-light scattering WLEDs have a greater ability to disperse blue light. This means that WLEDs have better color consistency. This is an essential outcome from applying $SrSi_2O_2N_2$:Eu²⁺,Yb²⁺. Especially challenging is the color consistency of a distant structure at high temperatures. Researchers found that $SrSi_2O_2N_2$, including low (5,600 K) and high (8,500 K) hue heats, may improve the WLEDs' color quality [19]-[21].

High stability in color chromaticity of warm White emitting Diode with Dual-Hue ... (My Hanh Nguyen Thi)

The efficiency of the light flux emitted by this phosphor layer has been demonstrated in the article. Outcomes shown in Figure 3 indicate that the light flux radiated by SrYb rose considerably 2-20% wt. Figure 4 indicates the hue dispersion in the three mean CCTs was considerably reduced when the SrYb concentration was higher. Red phosphor layer absorption may be to blame. They collect the blue illumination in the LED chip and turn it to green illumination. Aside from the blue illumination in the LED chip, the SrYb particles absorb yellow, and the blue illumination created by the LED chip. On the other hand, according to the material's absorptivity characteristics, the blue illumination absorptivity is larger compared to the others. When SrYb is added to WLEDs, the green content increases and the color uniformity index improves. The color uniformity of contemporary WLEDs is one of the most important characteristics. Naturally, WLED white light cost can be high and related to the hue homogeneous [22]-[24]. However, SrYb has the benefit of being inexpensive. As a result, SrYb is extensively applicable.



Figure 1. Retaining the mean CCT via altering phosphor content



Figure 2. The emitting bands of color of 3,000 K WLEDs as a function of SrYb concentration



Figure 3. The WLEDs luminous flux as a function of SrYb concentration



Figure 4. Correlation between WLEDs chroma deviation and SrYb content

In the evaluation of WLED color quality, color uniformity is just one factor. It's impossible to say that color quality is good when the color homogeneity index is high. A recent study has developed the CRI and the colour quality scale (CQS) as a result of this research. Light lits color rendering index and it determines the real color of a subject's hue and saturation. The overwhelming quantity of green light leads to an unbalanced color scheme between the primary colors: blue, yellow and green. As a result, WLEDs' color fidelity is degraded. There is a small drop in CRI when the distant phosphor SrYb layer is presented, as seen in Figure 5. In spite of the fact that they are acceptable, CQS only has a problem when it comes to CRI. It's clear that when comparing the importance of the CQS to the CRI, the CQS comes out on top. Colour Quality Scale is a three-factor indices that takes into account the hue rendering index, viewer choice, and hue coordinate. On the basis of these three variables, CQS is virtually an accurate measure of color quality [25]-[27]. A layer of SrYb can enhance CQS. When SrYb concentration is raised, CQS does not change substantially at concentrations below 10% wt, see Figure 6. Not only does CQS decrease but also CRI is lowered when the SrYb concentration is more than 10% wt. because of the extreme color loss when green dominates. SrYb Phosphorus must thus be applied at the correct concentration.



Figure 5. Correlation between WLEDs chroma rendering index and SrYb content



Figure 6. Correlation between hue standard scale in WLED device as a function of SrYb concentration

High stability in color chromaticity of warm White emitting Diode with Dual-Hue ... (My Hanh Nguyen Thi)

4. CONCLUSION

SrYb phosphor oxynitridosilicate has been created for the first time to be utilized in a warm white LED. For Eu^{2+} ions, the phosphor produced by using SrYb has broad band emission centering at around 540 to 612 nm. There is a correlation between the relative concentrations of activator ions in green and orange phosphors. That Eu^{2+} ion's decay period was so short, it proved that energy was being transferred from it's source to the Yb²⁺ trapping sites. InGaN emitting white LED with SrYb phosphor emits warm white light, has a high hue rendering index and outstanding hue steadiness against supply energy, according to a newlyfabricated white LED.

ACKNOWLEDGEMENTS

This study was financially supported by Van Lang University, Vietnam.

REFERENCES

- Y. Liu et al., "Preparation and reliability of flexible phosphor film for warm white LED," Materials Technology, vol. 33, no. 1, [1] pp. 22-28, Jan. 2018, doi: 10.1080/10667857.2017.1376787.
- [2] H. E. Dillon, C. Ross, and R. Dzombak, "Environmental and energy improvements of LED lamps over time: A comparative life cycle assessment," LEUKOS - Journal of Illuminating Engineering Society of North America, vol. 16, no. 3, pp. 229-237, Jul. 2020, doi: 10.1080/15502724.2018.1541748.
- Y. Luo et al., "White-light emitting diodes' spectrum effect on photosynthesis and nutrient use efficiency in Podocarpus [3] macrophyllus seedlings," Journal of Plant Nutrition, vol. 43, no. 19, pp. 2876-2884, Nov. 2020. doi: 10.1080/01904167.2020.1798999.
- J. K. Kim, M. E. Han, J. Y. Lee, S. H. Kong, and D. Jung, "LED with a zener chip coated with a highly reflective material," [4] Molecular Crystals and Liquid Crystals, vol. 676, no. 1, pp. 114-122, Nov. 2018, doi: 10.1080/15421406.2019.1596213.
- R. Malik, K. Ray, and S. Mazumdar, "A Low-Cost, Wide-Range, CCT-Tunable, Variable-Illuminance LED Lighting System," [5] LEUKOS - Journal of Illuminating Engineering Society of North America, vol. 16, no. 2, pp. 157-176, Apr. 2020, doi: 10.1080/15502724.2018.1541747.
- J. Hou et al., "White-light-emitting from single-phased (Ca,Eu,Mn)9Al(PO4)7 phosphor with blue-white-yellow tunable [6] luminescence properties for UV-based LEDs," Materials Technology, vol. 34, no. 3, pp. 135-142, Feb. 2019, doi: 10.1080/10667857.2018.1540331.
- M. Rajendran, K. Singh, and S. Vaidvanathan, "A novel Sm3+-activated Li3BaSrLn3(MO4)8 [Ln = La, Gd, and Y; M = Mo and [7] W] deep red-emitting phosphors for plant cultivation and white LEDs," Journal of Information Display, vol. 22, no. 2, pp. 63-81, Apr. 2021, doi: 10.1080/15980316.2020.1831630.
- G. M. House, E. B. Sobotik, J. R. Nelson, and G. S. Archer, "A comparison of white/red and white/blue LED light fixtures on [8] Pekin duck production, stress and behaviour," British Poultry Science, vol. 62, no. 4, pp. 467-473, Jul. 2021, doi: 10.1080/00071668.2021.1891524.
- [9] G. S. Archer, "Effect of two different commercially available white light LED fixtures on broiler hatchability and chick quality," British Poultry Science, vol. 59, no. 3, pp. 251–255, May 2018, doi: 10.1080/00071668.2018.1436160.
- [10] H. Zhang, Q. Su, and S. Chen, "Recent progress in the device architecture of white quantum-dot light-emitting diodes," Journal of Information Display, vol. 20, no. 4, pp. 169-180, Oct. 2019, doi: 10.1080/15980316.2019.1650129.
- [11] M. Niknejad and A. H. Karimy, "Lead white or lead whites? reconsideration of Methods of sefidab-i-sorb Production in Iran," Studies in Conservation, vol. 64, no. 1, pp. 1–9, Jan. 2019, doi: 10.1080/00393630.2018.1457290. D. Zhou, G. Yan, W. Zhang, J. Lin, and K. Qiu, "Synthesis and luminescence properties of Zn3B2O6:Eu3+, Li+ red-emitting
- [12] phosphor for white LEDs," Ferroelectrics, vol. 528, no. 1, pp. 114-121, May 2018, doi: 10.1080/00150193.2018.1448654.
- E. C. Navarre, "Characterization of 'cool' and 'warm' white light-emitting diodes for low-cost spectrophotometry," Spectroscopy [13] Letters, vol. 54, no. 8, pp. 581-589, Sep. 2021, doi: 10.1080/00387010.2021.1963781.
- [14] J. Song, "Manufacture of Quantum Dot LED using Hybrid sol," Molecular Crystals and Liquid Crystals, vol. 677, no. 1, pp. 91-95, Dec. 2018, doi: 10.1080/15421406.2019.1597515.
- C. Orhan et al., "Allyl isothiocyanate attenuates LED light-induced retinal damage in rats: exploration for the potential molecular [15] mechanisms," Cutaneous and Ocular Toxicology, vol. 40, no. 4, pp. 376–386, Oct. 2021, doi: 10.1080/15569527.2021.1978478.
- [16] Y.-P. Chang et al., "New scheme of LiDAR-embedded smart laser headlight for autonomous vehicles," Optics Express, vol. 27, no. 20, p. A1481, Sep. 2019, doi: 10.1364/oe.27.0a1481.
- [17] J. X. Yang, D. S. Li, G. Li, E. Y. B. Pun, and H. Lin, "Photon quantification in Ho 3+ /Yb 3+ co-doped opto-thermal sensitive fluotellurite glass phosphor," Applied Optics, vol. 59, no. 19, p. 5752, Jul. 2020, doi: 10.1364/ao.396393.
- [18] H. Yuce, T. Guner, S. Balci, and M. M. Demir, "Phosphor-based white LED by various glassy particles: control over luminous efficiency," Optics Letters, vol. 44, no. 3, p. 479, Feb. 2019, doi: 10.1364/ol.44.000479.
- Y. Chu et al., "Perception enhancement using importance-driven hybrid rendering for augmented reality based endoscopic [19] surgical navigation," Biomedical Optics Express, vol. 9, no. 11, p. 5205, Nov. 2018, doi: 10.1364/boe.9.005205.
- [20] Q. Guo et al., "Characterization of YAG:Ce phosphor dosimeter by the co-precipitation method for radiotherapy," Applied Optics, vol. 60, no. 11, p. 3044, Apr. 2021, doi: 10.1364/ao.419800.
- [21] Y. Tang, Z. Li, G. Liang, Z. Li, J. Li, and B. Yu, "Enhancement of luminous efficacy for LED lamps by introducing polyacrylonitrile electrospinning nanofiber film," *Optics Express*, vol. 26, no. 21, p. 27716, Oct. 2018, doi: 10.1364/oe.26.027716.
- [22] J. O. Kim, H. S. Jo, and U. C. Ryu, "Improving CRI and scotopic-to-photopic ratio simultaneously by spectral combinations of cct-tunable led lighting composed of multi-chip leds," Current Optics and Photonics, vol. 4, no. 3, pp. 247-252, 2020, doi: 10.3807/COPP.2020.4.3.247.
- L. Yang, Q. Zhang, F. Li, A. Xie, L. Mao, and J. Ma, "Thermally stable lead-free phosphor in glass enhancement performance of [23] light emitting diodes application," Applied Optics, vol. 58, no. 15, p. 4099, May 2019, doi: 10.1364/AO.58.004099.

- [24] A. D. Corbett et al., "Microscope calibration using laser written fluorescence," Optics Express, vol. 26, no. 17, p. 21887, Aug. 2018, doi: 10.1364/oe.26.021887.
- [25] Y. Zhang, X. Zhu, A. Liu, Y. Weng, Z. Shen, and B. Wang, "Modeling and optimizing the chromatic holographic waveguide display system," *Applied Optics*, vol. 58, no. 34, p. G84, Dec. 2019, doi: 10.1364/ao.58.000g84.
- [26] X. Shi, J. Liu, J. Xiao, and J. Han, "Design of a compact waveguide eyeglass with high efficiency by joining freeform surfaces and volume holographic gratings," *Journal of the Optical Society of America A*, vol. 38, no. 2, p. A19, Feb. 2021, doi: 10.1364/josaa.404280.
- [27] L. Li and M. J. Escuti, "Super achromatic wide-angle quarter-wave plates using multi-twist retarders," *Optics Express*, vol. 29, no. 5, p. 7464, Mar. 2021, doi: 10.1364/oe.418197.

BIOGRAPHIES OF AUTHORS



My Hanh Nguyen Thi D X S P received a Bachelor of Physics from An Giang University, VietNam, Master of Theoretical Physics And Mathematical Physics, Hanoi National University of Education, VietNam. Currently, she is a lecturer at the Faculty of Mechanical Engineering, Industrial University of Ho Chi Minh City, Viet Nam. Her research interests are Theoretical Physics and Mathematical Physics. She can be contacted at email: nguyenthimyhanh@iuh.edu.vn.



Phan Xuan Le D \fbox{M} \fbox{M} received a Ph.D. in Mechanical and Electrical Engineering from Kunming University of Science and Technology, Kunming city, Yunnan province, China. Currently, He is a lecturer at the Faculty of Engineering, Van Lang University, Ho Chi Minh City, Viet Nam. His research interests are Optoelectronics (LED), Power transmission and Automation equipment. He can be contacted at email: le.px@vlu.edu.vn.



Nguyen Le Thai D E The P received his BS in Electronic engineering from Danang University of Science and Technology, Vietnam, in 2003, MS in Electronic Engineering from Posts and Telecommunications Institute of Technology, Ho Chi Minh, Vietnam, in 2011 and PhD degree of Mechatronics Engineering from Kunming University of Science and Technology, China, in 2016. He is a currently with the Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam. His research interests include the renewable energy, optimisation techniques, robust adaptive control and signal processing. He can be contacted at email: nlthai@nttu.edu.vn.

High stability in color chromaticity of warm White emitting Diode with Dual-Hue ... (My Hanh Nguyen Thi)