

# Impact of green-emitting $\text{CaSc}_2\text{O}_4$ : $\text{Ce}^{3+}$ phosphor on the illuminance and color uniformity of white-light-emitting diodes

Le Thi Thuy My<sup>1</sup>, My Hanh Nguyen Thi<sup>2</sup>, Nguyen Le Thai<sup>3</sup>

<sup>1</sup>Faculty of Basic Sciences, Vinh Long University of Technology Education, Vinh Long Province, Vietnam

<sup>2</sup>Faculty of Mechanical Engineering, Industrial University of Ho Chi Minh City, Ho Chi Minh City, Vietnam

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

## Article Info

### Article history:

Received Aug 27, 2022

Revised Jul 16, 2023

Accepted Jul 18, 2023

### Keywords:

$\text{CaSc}_2\text{O}_4$ :  $\text{Ce}^{3+}$

Color homogeneity

Luminous flux

Monte Carlo theory

WLEDs

## ABSTRACT

With blue light stimulation, a novel green phosphor,  $\text{Ce}^{3+}$ -activated  $\text{CaSc}_2\text{O}_4$ , was created, which emits green illumination with a maximum wavelength of 515 nm. Since the brightness performance of  $\text{Ce}^{3+}$ -activated  $\text{CaSc}_2\text{O}_4$  is equivalent to that of commercial phosphors such as  $\text{Y}_3\text{Al}_5\text{O}_{12}$ :  $\text{Ce}^{3+}$ , it may be utilized as a substance for chroma transformation in white-light-emitting diodes (WLEDs) that comprise one blue LED, one green phosphor, as well as one red phosphor. The Ce ion is most likely at an eight-coordinated Ca position in the base crystal for said phosphor, which possesses orthorhombic  $\text{CaFe}_2\text{O}_4$  configuration. The effect of burning temperature and dopant concentration on luminous strength was examined, and it was discovered that the optimal temperature and concentration reached 1,600 °C and 1 mol % Ce replaced in the Ca site, respectively. By substituting Mg for Ca, the luminous maximum wavelength moved towards greater values. Substituting Ca using Sr, on the other hand, and yielded changes towards lesser wavelengths.

This is an open access article under the [CC BY-SA](https://creativecommons.org/licenses/by-sa/4.0/) license.



## Corresponding Author:

My Hanh Nguyen Thi

Faculty of Mechanical Engineering, Industrial University of Ho Chi Minh City

No. 12 Nguyen Van Bao Street, Ho Chi Minh City, Vietnam

Email: [nguyenthimyanh@iuh.edu.vn](mailto:nguyenthimyanh@iuh.edu.vn)

## 1. INTRODUCTION

Light-emitting diodes (LED) devices with a phosphor and an elevated-performance InGaN LED are becoming widely available commercially [1]. The phosphors employed here must absorb blue or UV illumination with a long wavelength. Many phosphors have been suggested for this application, including oxides, sulfides [2], and nitrides [3], [4]. The most common white LED (WLED) comprises one blue InGaN LED and  $\text{Y}_3\text{Al}_5\text{O}_{12}$ : Ce [5], [6]. Said WLED yields very little light of red, thus having undesirable chroma rendition index (CRI). The usage of red and green phosphors rather than yellow phosphors has been recommended as a solution to this issue [7].  $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}$ :  $\text{Ce}^{3+}$ , a green phosphor, was created specifically for this application [8]. The phosphor's host crystal has a garnet structure. The  $\text{Y}_3\text{Al}_5\text{O}_{12}$ :  $\text{Ce}^{3+}$  yellow phosphor, which is the most frequently utilized phosphor for white LEDs, has a configuration that is almost identical to this.  $\text{Lu}_2\text{CaMg}_2\text{Si}_3\text{O}_{12}$ :  $\text{Ce}^{3+}$ , a phosphor with a garnet-kind host crystal and  $\text{Ce}^{3+}$  as an activator, has also been described [9]. As a result,  $\text{Ce}^{3+}$  may effectively display long-wavelength illumination from green to red in a host crystal with a garnet structure, independent of the components present. Calcium sulfide is one of the host crystals in which  $\text{Ce}^{3+}$  ions show long-wavelength illumination, in addition to garnet [10]. Apart from garnet, no oxide host crystal has been identified as a host crystal for long-wavelength  $\text{Ce}^{3+}$  illumination. A new phosphor made up of an oxide host crystal other than garnet and  $\text{Ce}^{3+}$  activator is described in this paper. This phosphor emits a great amount of green light. The connection among the phosphor's crystal structure and its illumination characteristics is also discussed.

## 2. METHOD

The  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  phosphor was made by a standard solid-status process. Powders of  $\text{CaCO}_3$ ,  $\text{Sc}_2\text{O}_3$ , and  $\text{CeO}_2$  were weighed and combined. Ca: Sc: Ce=0.99:2.00:0.01 was a particular atomic ratio of the elements. The concentration of cerium was increased from 0.05 to 5 mol%. The combined raw resources were put on platinum foil and heat-treated within 180 minutes under 1,400-1,600 °C within 4%  $\text{H}_2+\text{N}_2$  surrounding.  $\text{SrCO}_3$ ,  $\text{MgO}$ , and  $\text{Lu}_2\text{O}_3$  were utilized as raw resources in the research that involved replacing Ca or Sc with some other element. In relation to the Ca content, the Ce concentration was set at 1 mol%. An F4500 fluorescence photometer was used to evaluate luminescence and luminescence stimulation spectra (Hitachi). X-ray diffraction (XRD) was conducted via a Cu-K $\alpha$  X-ray source and the X'Pert MPD (PANalytical) application.  $\text{SrY}_2\text{O}_4:\text{Ce}^{3+}$ ,  $\text{SrY}_2\text{O}_4:\text{Eu}^{3+}$ ,  $\text{SrY}_2\text{S}_4:\text{Ce}^{3+}$ , and  $\text{SrY}_2\text{S}_4:\text{Eu}^{2+}$  are some of the phosphors with the  $\text{CaFe}_2\text{O}_4$  configuration that have been described.  $\text{SrY}_2\text{O}_4:\text{Ce}^{3+}$  is the only one of them with a  $\text{Ce}^{3+}$  activator in an oxide host. Manivannan et al. observed two forms of illumination for the phosphor: 570 nm light from a “ $\text{Ce}^{4+}-\text{O}_2$ ” charge-transfer conversion and 475 nm illumination from the  $\text{Ce}^{3+}$  4f-5d conversion. In our work, nevertheless, the last illumination was not seen at room temperature. Manivannan et al. also claimed a spectrum that was recorded at 10 K. According to the findings from this study,  $\text{Ce}^{3+}$  in the  $\text{SrY}_2\text{O}_4$  host crystal is quenched and does not display effective illumination at normal temperature [11], [12].  $\text{SrY}_2\text{S}_4:\text{Ce}^{3+}$  has been claimed to display illumination by  $\text{Ce}^{3+}$  in sulfides with the  $\text{CaFe}_2\text{O}_4$  configuration. Therefore, it is ineffective as a phosphor.  $\text{CaSc}_2\text{O}_4$  is the first  $\text{CaFe}_2\text{O}_4$ -type host crystal in which  $\text{Ce}^{3+}$  exhibits effective luminescence at room temperature, as seen following. The existence of a Sc ion, which is not usual in phosphor host crystals, creates an adequate crystal field for illumination, albeit the cause for this is unclear. To figure out how the illumination works, more research is needed.

## 3. RESULTS AND DISCUSSION

We can see that there is an opposite change for the dosages for phosphorus  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  as well as phosphorus YAG:  $\text{Ce}^{3+}$  in Figure 1. This change serves two purposes: it maintains average central choroidal thickness (CCTs) [13], [14] while also affecting the absorptivity as well as dispersion in the WLED device containing a pair of phosphor sheets, affecting chroma performance as well as efficacy of the illuminating flux of said device. The  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  concentration used thus affects the chroma output in the device. The YAG:  $\text{Ce}^{3+}$  dosage declined, sustaining the mean CCT values as the  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  scale climbed from 2 percent to 20 percent Wt. WLED apparatuses having chroma heat levels ranging between 5600K and 8500 K are comparable to this as well.

Figure 2 depicts the luminescence as well as luminescence stimulation spectra of the  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  phosphor.  $\text{Ce}^{3+}$  has a 5d-4f conversion, which causes luminescence. The luminescence maximum was found around 515 nm, with a shoulder on the longer wavelength side due to the splitting of the  $\text{Ce}^{3+}$  ion's 4f base state. The luminescence stimulation spectrum has a maximum wavelength of 450 nm. This is a substance that may be used to convert the color of a white LED into a blue InGaN LED. The luminous performance was around 90% of  $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Ce}^{3+}$ , which also includes Sc, and was larger than that of a commercial yttrium aluminum garnet (YAG) phosphor (P46-Y3 by Kasei Optonix). Figure 3 provides a comparative analysis of luminescence spectra with stimulation at 455 nm. The following is a comparison among  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  and  $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Ce}^{3+}$ . Raising the synthesis temperature improved the light strength, and the phosphor produced at 1,600°C had the highest effective illumination. Higher synthesis temperature enhanced crystallinity, which likely increased illumination effectiveness. Figure 2 also illustrates the influence of the  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  dosage upon the transference spectrum in the WLED apparatus. When using  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$ , this is a noteworthy result. As a consequence, our research suggests that  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  can enhance WLED color quality at low hue temperatures (5600K) and high hue temperatures (8500K) [15], [16].

As a result, the research proved the efficiency for the generated illumination of this two-layer remote setting. Figure 3 displays the increase of the  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  concentration from 2% wt. to 20% wt., resulting in a considerable improvement in illumination. In Figure 4, the color deviation was considerably decreased regardless of CCT when concentrations of  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  were used. The green illumination presence in the WLED device increases when  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  is added. Based on this, the chroma consistency improves. The pricier the WLED white illumination, the greater the color uniformity indicator [17]–[19]. Therefore, one of the most significant WLED characteristics right now is color uniformity.  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$ , on the other hand, is quite cheap. As a result,  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  has recently been employed in a variety of applications [20]–[22].

Chroma uniformity would be among the many factors utilized for appraising the chroma performance in WLED apparatuses, therefore a high chroma uniformity indicator cannot be used for indicating color quality excellence. Instead, the researchers created a color rendering indicator as well as chroma quality scale. The chroma rendition index assesses the real hue in a thing. When green light presence in the primary chromas (yellow, green, blue) is excessive, chroma disparity occurs [23], decreasing hue fidelity, which has a significant

influence on WLED's hue standard. In Figure 5, when the phosphors layer is remote from  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$ , the CRI decreases slightly. CRI, on the other hand, are permitted since they fill a gap in the CQS, which would be a more important and difficult indicator to attain [24]–[26]. The hue rendering indicator, the selection of the viewers, and the hue coordinate are all part of the CQS index. CQS is used as a general indicator of color quality. CQS increases in the presence of a distant  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  phosphor layer, as seen in Figure 6. Additionally, when the concentration of  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  was below 10% wt, CQS did not change substantially as the  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  content was increased. When the concentration of  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  exceeds 10% wt., CRI as well as CQS would be considerably reduced, caused by severe hue penalty caused by the green color of  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  taking up too much space. In general, while utilizing green phosphor  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$ , it is important to choose a suitable concentration carefully.

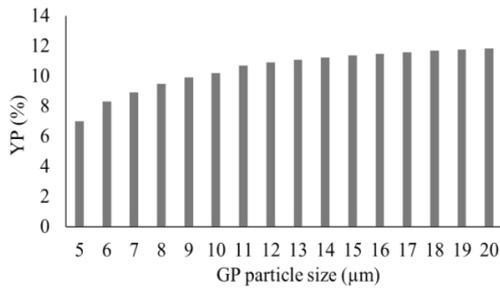


Figure 1. Adjusting the phosphor concentration to maintain the median CCT

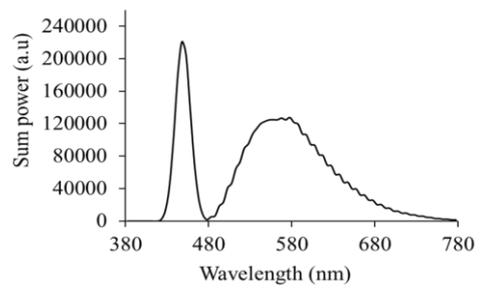


Figure 2. The emission spectra of 5000K WLEDs as a function of  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  size

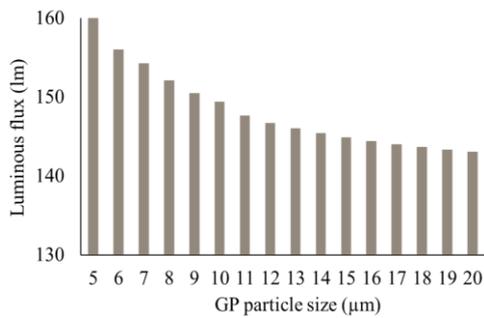


Figure 3. Relation between the lighting flux from the WLED device and  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  size

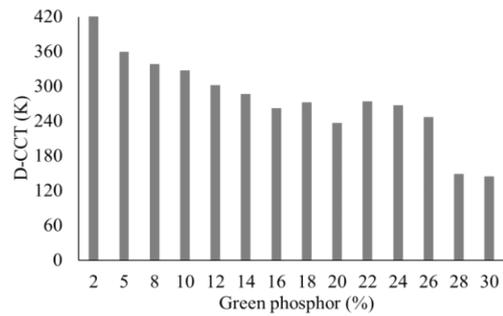


Figure 4. Relation between the hue aberration from the WLED device and  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  size

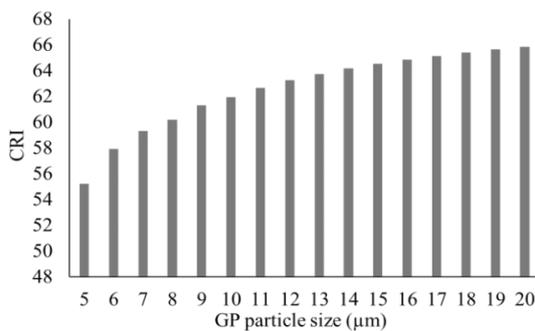


Figure 5. Relation between CRI from the WLED device and  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  size

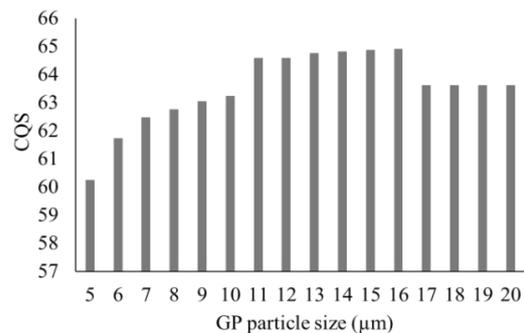


Figure 6. Relation between CQS from the WLED device and  $\text{CaSc}_2\text{O}_4:\text{Ce}^{3+}$  size

#### 4. CONCLUSION

A novel green phosphor,  $\text{CaSc}_2\text{O}_4: \text{Ce}^{3+}$ , has been created, consisting of an oxide host crystal with a  $\text{CaFe}_2\text{O}_4$  structure and a  $\text{Ce}^{3+}$  activator. This phosphor is intended to be used as a hue transformation substance for white LEDs with good hue rendering. The crystal field of  $\text{Ce}^{3+}$  and its luminous properties were addressed in this paper in contrast to  $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}: \text{Ce}^{3+}$ . For luminescent characteristics, the influences of heating temperature and activator concentration were investigated. The illumination maximum shift and reduction in illumination strength were detected when the component elements of the host crystal were substituted.

#### REFERENCES

- [1] W. Zhong, J. Liu, D. Hua, S. Guo, K. Yan, and C. Zhang, "White LED light source radar system for multi-wavelength remote sensing measurement of atmospheric aerosols," *Applied Optics*, vol. 58, no. 31, p. 8542, Nov. 2019, doi: 10.1364/ao.58.008542.
- [2] 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.
- [3] Y. Zhou *et al.*, "Comparison of nonlinear equalizers for high-speed visible light communication utilizing silicon substrate phosphorescent white LED," *Optics Express*, vol. 28, no. 2, p. 2302, Jan. 2020, doi: 10.1364/oe.383775.
- [4] H.-Y. Lee, P. X. Le, and D. Q. A. Nguyen, "Selecting a suitable remote phosphor configuration for improving color quality of white led," *Journal of Advanced Engineering and Computation*, vol. 3, no. 4, p. 503, Dec. 2019, doi: 10.25073/jaec.201934.249.
- [5] L. Wu *et al.*, "Hybrid warm-white organic light-emitting device based on tandem structure," *Optics Express*, vol. 26, no. 26, p. A996, Dec. 2018, doi: 10.1364/oe.26.00a996.
- [6] G. Tan, Y. Huang, M.-C. Li, S.-L. Lee, and S.-T. Wu, "High dynamic range liquid crystal displays with a mini-LED backlight," *Optics Express*, vol. 26, no. 13, p. 16572, Jun. 2018, doi: 10.1364/oe.26.016572.
- [7] H. Gu, M. Chen, Q. Wang, and Q. Tan, "Design of two-dimensional diffractive optical elements for beam shaping of multicolor light-emitting diodes," *Applied Optics*, vol. 57, no. 10, p. 2653, Apr. 2018, doi: 10.1364/ao.57.002653.
- [8] O. H. Kwon, J. S. Kim, J. W. Jang, and Y. S. Cho, "Simple prismatic patterning approach for nearly room-temperature processed planar remote phosphor layers for enhanced white luminescence efficiency," *Optical Materials Express*, vol. 8, no. 10, p. 3230, Oct. 2018, doi: 10.1364/ome.8.003230.
- [9] A. Ferrero, J. L. Velázquez, A. Pons, and J. Campos, "Index for the evaluation of the general photometric performance of photometers," *Optics Express*, vol. 26, no. 14, p. 18633, Jul. 2018, doi: 10.1364/oe.26.018633.
- [10] N. Fujimoto, M. Kifune, T. Hara, and N. Akizawa, "The longest transmission experiment of 200 m SI-plastic optical fibre using a high-luminous green LED with a new equalizing and carrier sweep out circuit," in *Optics InfoBase Conference Papers*, 2018, vol. Part F98-BGPPM 2018, p. JTU5A.57, doi: 10.1364/BGPPM.2018.JTU5A.57.
- [11] W. J. Kim *et al.*, "Improved angular color uniformity and hydrothermal reliability of phosphor-converted white light-emitting diodes by using phosphor sedimentation," *Optics Express*, vol. 26, no. 22, p. 28634, Oct. 2018, doi: 10.1364/oe.26.028634.
- [12] H. Yang *et al.*, "'Giant' quantum dots encapsulated inside a freeform lens," *Applied Optics*, vol. 57, no. 35, p. 10317, Dec. 2018, doi: 10.1364/ao.57.010317.
- [13] X. Li, B. Hussain, L. Wang, J. Jiang, and C. P. Yue, "Design of a 2.2-mW 24-Mb/s CMOS VLC Receiver SoC with ambient light rejection and post-equalization for Li-Fi applications," *Journal of Lightwave Technology*, vol. 36, no. 12, pp. 2366–2375, Jun. 2018, doi: 10.1109/JLT.2018.2813302.
- [14] X. Ding *et al.*, "Improving the optical performance of multi-chip LEDs by using patterned phosphor configurations," *Optics Express*, vol. 26, no. 6, p. A283, Mar. 2018, doi: 10.1364/oe.26.00a283.
- [15] H.-Y. Yu *et al.*, "Solar spectrum matching with white OLED and monochromatic LEDs," *Applied Optics*, vol. 57, no. 10, p. 2659, Apr. 2018, doi: 10.1364/ao.57.002659.
- [16] 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.
- [17] V. M. Igba *et al.*, "Structural elucidation and optical properties of  $\text{LiZrO}_2$ – $\text{LiBaZrO}_3$  nanocomposite doped with  $\text{Mn}^{2+}$  ions," *Optical Materials Express*, vol. 10, no. 11, p. 2877, Nov. 2020, doi: 10.1364/ome.402111.
- [18] H.-K. Shih, C.-N. Liu, W.-C. Cheng, and W.-H. Cheng, "High color rendering index of 94 in white LEDs employing novel  $\text{CaAlSiN}_3: \text{Eu}^{2+}$  and  $\text{Lu}_3\text{Al}_5\text{O}_{12}: \text{Ce}^{3+}$  co-doped phosphor-in-glass," *Optics Express*, vol. 28, no. 19, p. 28218, Sep. 2020, doi: 10.1364/oe.403410.
- [19] A. Zhang *et al.*, "Tunable white light emission of a large area film-forming macromolecular complex with a high color rendering index," *Optical Materials Express*, vol. 8, no. 12, p. 3635, Dec. 2018, doi: 10.1364/ome.8.003635.
- [20] X. Yang, C. Chai, J. Chen, S. Zheng, and C. Chen, "Single 395 nm excitation warm WLED with a luminous efficiency of 10486 lm/W and a color rendering index of 907," *Optical Materials Express*, vol. 9, no. 11, p. 4273, Nov. 2019, doi: 10.1364/ome.9.004273.
- [21] C. Wu *et al.*, "Phosphor-converted laser-diode-based white lighting module with high luminous flux and color rendering index," *Optics Express*, vol. 28, no. 13, p. 19085, Jun. 2020, doi: 10.1364/oe.393310.
- [22] H. Lee *et al.*, "Color-tunable organic light-emitting diodes with vertically stacked blue, green, and red colors for lighting and display applications," *Optics Express*, vol. 26, no. 14, p. 18351, Jul. 2018, doi: 10.1364/oe.26.018351.
- [23] H. S. El-Ghoroury, Y. Nakajima, M. Yeh, E. Liang, C.-L. Chuang, and J. C. Chen, "Color temperature tunable white light based on monolithic color-tunable light emitting diodes," *Optics Express*, vol. 28, no. 2, p. 1206, Jan. 2020, doi: 10.1364/oe.375320.
- [24] Y. Wang *et al.*, "Tunable white light emission of an anti-ultraviolet rare-earth polysiloxane phosphors based on near UV chips," *Optics Express*, vol. 29, no. 6, p. 8997, Mar. 2021, doi: 10.1364/oe.410154.
- [25] M. Gupta, A. K. Dubey, V. Kumar, and D. S. Mehta, "Indoor daylighting using Fresnel lens solar-concentrator-based hybrid cylindrical luminaire for illumination and water heating," *Applied Optics*, vol. 59, no. 18, p. 5358, Jun. 2020, doi: 10.1364/ao.389044.
- [26] A. Ali *et al.*, "Blue-laser-diode-based high CRI lighting and high-speed visible light communication using narrowband green-/red-emitting composite phosphor film," *Applied Optics*, vol. 59, no. 17, p. 5197, Jun. 2020, doi: 10.1364/ao.392340.

**BIOGRAPHIES OF AUTHORS**

**Le Thi Thuy My**    received the Master degree in physics from Can Tho University, Vietnam. She is working as a lecturer at the Faculty of Basic Sciences, Vinh Long University of Technology Education, Vietnam. Her research interests focus on developing the patterned substrate with micro- and nano-scale to apply for physical and chemical devices such as solar cells, OLED, photoanode, and theory physics. She can be contacted at email: myltht@vlute.edu.vn.



**My Hanh Nguyen Thi**    received a Bachelor of Physics from A 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, Vietnam. Her research interests are theoretical physics and mathematical physics. She can be contacted at email: nguyenthimyhanh@iuh.edu.vn.



**Nguyen Le Thai**    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 Ph.D. degree of Mechatronics Engineering from Kunming University of Science and Technology, China, in 2016. He is 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.