Scattering regulation using barium sulfate particles to achieve improved colour temperature uniformity of white LEDs

N. T. M. HANH[1], N. L. THAI[2], N. T. P. LOAN[3,*], H. Y. LEE[4]
[1]*Faculty of Mechanical Engineering, Industrial University of Ho Chi Minh City, Ho Chi Minh City, Vietnam*
[2]*Faculty of Engineering and Technology, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam*
[3]*Faculty of Fundamental 2, Posts and Telecommunications Institute of Technology, Ho Chi Minh City, Vietnam*
[4]*Department of Electrical Engineering, National Kaohsiung University of Science and Technology, Kaohsiung, Taiwan*

$BaSO_4$ has showed up to be an ideal candidate for regulating the scattering performance in LEDs owing to its unique properties, thereby improving the LED's efficiency and overall light quality. The high-reflectivity $BaSO_4$ was utilized as the scattering factor in the yellow-phosphor conversion layer in this work. The concentration of $BaSO_4$ was altered in the range of 5 wt% to 50 wt%. The scattering property of $BaSO_4$ particles is investigated using Mie-scattering simulation coupled with LightTools software. The assessed lighting factors of LEDs on varying doping concentrations of $BaSO_4$ in the phosphor layer include corelated color temperature (CCT) uniformity and luminosity. Simulation results exhibited that $BaSO_4$ at high concentration was beneficial for CCT-deviation reduction to enhance the CCT distribution uniformity. Meanwhile, the luminosity declined as a trade-off for the induced scattering coefficients.

(Received April 14, 2024; accepted February 3, 2025)

Keywords: $LiLaO_2:Eu^{3+}$, Dual-layer remote phosphor geometry, WLEDs, Mie-scattering theory, Colour rendering index

1. Introduction

Solid-state lighting (SSL) technology, using semiconductor light-emitting diodes (LEDs), has revolutionized the lighting industry with its numerous advantages [1, 2]. It offers better performance and reliability, and substantially lowers the total cost of ownership compared to conventional incandescent lighting. It moreover is energy-efficient, long-lasting, and durable under hard conditions like shock and vibration [3, 4]. SSL devices can produce light with high efficiency, allowing them to feature in across-aspect lighting applications, such as transportation and traffic lightings, entertainment lighting, and displays. The key element of an SSL device is the LED, indicating that the quality of an SSL device is determined by the quality of the applied LED [5-7]. Typically, a LED is produced using one or multiple blue LED chips and phosphor combination, which is a great alternative to the more expensive three-chip RGB (red, green, blue) LED. Though this conventional LED type offers energy and cost efficiency along with high performance in cold settings, its light color uniformity and rendition are inferior [8]. The shortage in red spectrum and inefficient scattering performance of incident light are the reasons for such drawbacks.

The scattering effect plays a crucial role in the light properties of white LEDs. The inclusion of scattering particles in the resin of LEDs induces multiple scattering and contributes to the color conversion of phosphors and quantum dots [9]. This effect enhances the utilization of blue light emitted from chips and reduces the back-emission loss of light excited from quantum dots [10]. It can also increase the luminous flux of the white LED by over 30% compared with the reference sample without any nanocrystal [11]. According to these findings, regulating the scattering property of the LED phosphor layer can lead to improvements in luminous flux and/or light color conversion and distribution for better chromaticity of white LEDs. In terms of approaches for scattering enhancement, using particles possessing high scattering ability is one of the favorable ones. Among the investigated particles, SiO_2 was the most intensively studied for its unique characteristics. Apart from SiO_2 particles, $BaSO_4$ is a promising alternative offering outstanding optical and chemical characteristics. $BaSO_4$ is a white crystalline solid that is odorless and insoluble in water. It has excellent temperature, acid, and alkali resistance [12], making it suitable for use in LEDs. $BaSO_4$'s high reflectivity, good thermal stability, and chemical inertness are beneficial in solid-state lighting applications where components can generate significant amounts of heat [13]. Its low fluorescence is beneficial in applications like UV LEDs where accurate and traceable measurement is required [14]. Unlike some scattering substances, $BaSO_4$ is not noxious and thus safe for utilization in various scenarios. $BaSO_4$ can also effectively scatter illumination within the UV to visible spectrum and as such, offers significant versatility that suits disparate uses. The many advantages, combined with easy accessibility, make $BaSO_4$ a highly optimal choice as a scattering agent. $YAG:Ce^{3+}$ was chosen as a constituent for phosphor layer as the phosphor is a primary material in contemporary optic apparatuses, including LEDs, exhibiting proficient luminescence, broad excitation spectrum, thermic consistency, compatibility with other substances, and so on.

In this study, $BaSO_4$ with different concentrations is applied to regulate scattering performance of the phosphor composite in the conventional LED. The scattering of $BaSO_4$ and its effects on lighting properties of the white LED are simulated using Mie-scattering theory and software LightTools. Since the concentration of $BaSO_4$ varies from 0-50 wt%, using Mie theory is preferable since it is highly suitable for analyzing scattering in particles of various size ranges, hence its versatility. The Mie theory can provide a more precise analysis, yielding more detailed data on particle scattering compared to other theories. From the results of this study, it can be concluded that the use of $BaSO_4$ in LEDs presents a promising avenue for enhancing the performance of solid-state lighting.

2. Preparation and simulation

In this work, the high-reflectivity $BaSO_4$ was prepared using the method demonstrated in [15]. According to this study, the synthesized $BaSO_4$ possesses greater reflectivity than the commercial one in the market, enhancing its applicability. The optimal stirring rate and temperature were determined at 250 r/min and 55^0C for the even distribution. The particle size of the prepared $BaSO_4$ was about 1 μm.

To prepare the phosphor layer, the $BaSO_4$ was mixed with $YAG:Ce^{3+}$ yellow phosphor and silicone. The concentration of the $BaSO_4$ particles in the layer was adjusted from 5 wt% to 50 wt%. On the increasing concentration of $BaSO_4$, the amount of $YAG:Ce^{3+}$ in the conversion layer decreases, as shown in Fig. 1.

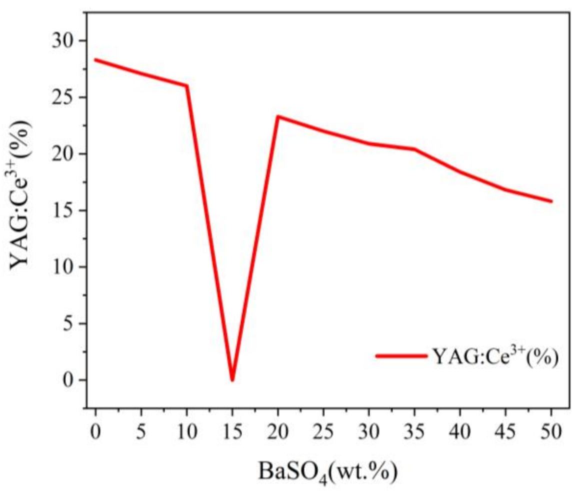


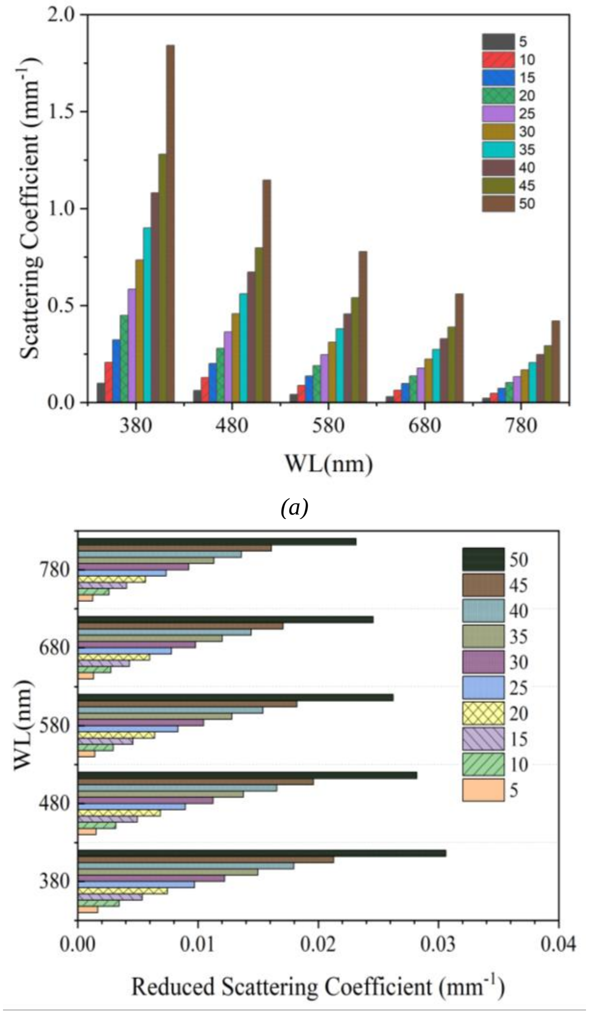
Fig. 1. $YAG:Ce^{3+}$ amounts varying corresponding to $BaSO_4$ concentrations (colour online)

The inverse relationship between $BaSO_4$ and $YAG:Ce^{3+}$ concentration is to keep the optimal preset corelated color temperature (CCT) during the simulation and measurement process [16-17]. Besides, it influences the scattering parameters of the conversion layer, resulting in changes in the LED output quality. Here, a significant drop in $YAG:Ce^{3+}$ concentration is noticed when $BaSO_4$ amount reaches 15 wt%. As the amount of $BaSO_4$ continues to increase to 20 wt%, the $YAG:Ce^{3+}$ amount begins to rise but showing a downward slope in response to higher $BaSO_4$ concentrations.

The decline in $YAG:Ce^{3+}$ concentration can contribute to improving the luminescence of the phosphor layer under blue-LED excitation [20]. Yet, the increasing in of $BaSO_4$ also influences the scattering events in the layer, initiating notable changes in LED output. Thus, the simulation investigation on luminosity and color parameters of LED related to the varying $BaSO_4$ concentration was carried out with Mie-scattering simulation combined with LightTools software. The discussion on simulation results will provide basis understanding on $BaSO_4$ utilization as scattering particles for LED fabrication.

3. Results and analysis

The scattering efficiency of the $BaSO_4$ film sample with different concentrations is shown in Fig. 2. Overall, the increase of $BaSO_4$ results in the increase in scattering coefficients, indicating that higher $BaSO_4$ concentrations can enhance the scattering events of the phosphor layer. However, under ranging wavelengths from 380 nm to 780 nm, the scattering factors are significantly different. The strongest scattering factor is noticed at 380 nm, especially at high $BaSO_4$ concentration, e.g. 50 wt%. This implies that the $BaSO_4$ can be the best suit for near-ultraviolet LED. At blue wavelength of about 480 nm, the scattering coefficients of the material decline by nearly a half of that at 380 nm but higher than the values at longer wavelengths (580-780 nm). In other words, the scattering coefficients of the $BaSO_4$ material decline with longer light wavelengths. Such an observation indicates that the blue light from the LED chip is scattered most, supporting greater light absorption and re-emission by phosphors. Meanwhile, the re-emitted light or converted light by phosphor with longer wavelength is less scattered, allowing them to escape from the LED package easily. Yet, the high blue light utilization can reduce the LED luminous flux since the longer wavelength often exhibits lower spectral energy than the short wavelengths [18].



(a)

(b)

Fig. 2. The scattering coefficients of $BaSO_4$ at different concentrations (colour online)

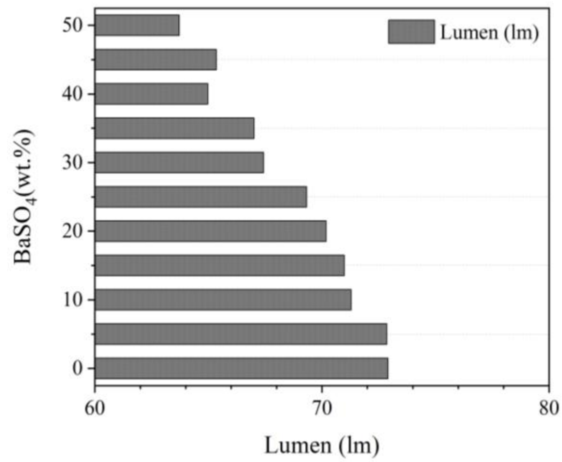


Fig. 3. Lumen corresponding to $BaSO_4$ concentrations

Fig. 3 demonstrates the luminous flux intensity of the LED with 5-50 wt% of $BaSO_4$ concentration in the phosphor luminescent layer. The decline in luminous flux of the LED with higher $BaSO_4$ concentrations is exhibited clearly in the figure. Obviously, the highest simulated luminous flux is presented with 5 wt% $BaSO_4$, while the lowest result is with 50 wt% $BaSO_4$. The YAG:Ce^{3+} phosphor amount declines in the presence of the $BaSO_4$, which can be an advantage in inducing the quantum efficiency of the LED; however, the higher concentration of $BaSO_4$ stimulates the scattering of incident LED-chip blue light and also converted light, reducing the energy of transmission light. In addition, it was demonstrated that the YAG:Ce^{3+} phosphor particles exhibit stronger scattering ability than its absorption ability, implying that the converted light is more likely to be re-scattered and reabsorbed by the substrate of the LED design [19]. Consequently, the total transmission power of the LED decreases. Therefore, high scattering events and low-absorption performance of the luminescent phosphor materials are not optimal for high-luminosity LED packages.

On the other hand, by utilizing scattering events of the materials in the LED package, the improvement in angular color uniformity is achievable [21]. The angular CCT of the LED package is shown in Fig. 4. The CCT distribution was collected across viewing angle range of ± 60 degrees with the concentration of $BaSO_4$ adjusted from 5 wt% to 50 wt%. The CCT distribution of the LED with 0 wt% was also obtained for comparison.

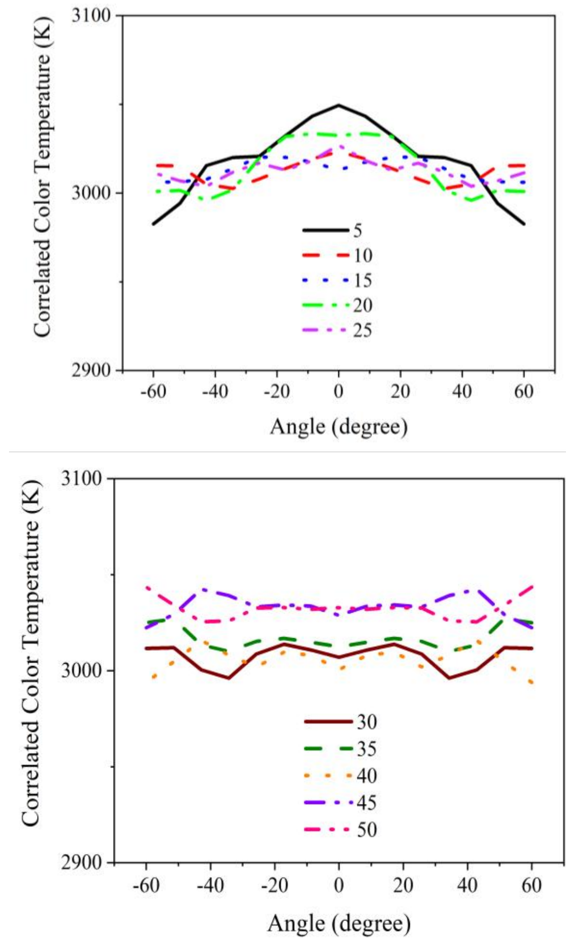


Fig. 4. Colour-temperature ranges corresponding to $BaSO_4$ concentrations (colour online)

Overall, the increasing $BaSO_4$ concentration results in the decline of light intensity at the center (0 degree viewing angle) and light-intensity variation at the wider angles. Particularly, the light is concentrated at the center in the cases without and with 5 wt% $BaSO_4$. Moreover, the CCT line with 5 wt% $BaSO_4$ is relatively higher than that of 0 wt% $BaSO_4$. However, the CCT distribution at the center decreases with higher concentrations of $BaSO_4$. The CCT line shape at the center is flattened in the cases of 20 wt% and 50 wt%, or becomes a downward-facing cone when $BaSO_4$ concentration reaches 15 wt% and 30-45 wt%. Meanwhile, the CCT distribution at the wider angles shows significant changes with the higher concentrations of $BaSO_4$. Considering the viewing angle of ± 40 degrees, for examples, the CCT spot is at higher values than that at the center when the amount of $BaSO_4$ reaches 35-40 wt%.

Such changes in CCT distributions can be attributed to the scattering events of the $BaSO_4$ material. The increase in $BaSO_4$ doping concentrations results in the greater scattering events, as depicted in Fig. 2 (a). The improved scattering can result in the wider light propagation area, giving chances for light being distributed at the peripheral areas. Therefore, the light distribution can be improved. However, the difference between the CCT spot at the center and periphery should not be significant to obtain the optimal CCT uniformity. Fig. 2 (b) shows the correlation between scattering coefficients and wavelengths. As can be seen, the correlation appears to be an inverse mechanism. As the wavelength increases, the scattering exhibits a gradual and consistent decline. Apparently, great scattering events occur at smaller wavelengths. While $BaSO_4$ can enhance scattering, it may also contribute to a slight increase in LED temperature, depending on its concentration, with higher concentration leading to temperature surge. The additional particles can also absorb some light and energy, causing a greater temperature.

The CCT deviation (D-CCT) is a critical parameter in evaluating the CCT distribution uniformity. The lower CCT deviation leads to the more even distribution of CCT value. D-CCT plays a critical role in determining the quality of white light color, depending on certain requirements. Greater levels in color temperature influence our mood and comfort. Warmer temperatures are often associated with relaxation, whereas cooler temperatures can enhance alertness and productivity. D-CCT can improve the overall visual experience by mimicking natural lighting conditions throughout the day, making spaces feel more inviting. With the increasing concentration of $BaSO_4$ from 5 wt% to 50 wt%, the CCT deviation exhibits a notable fluctuation, as illustrated in Fig. 5. It can be described as follows. The CCT deviation (CCT-D) slightly increases when $BaSO_4$ increases from 5 wt% to 10 wt%. At 15 wt% of $BaSO_4$, the CCT-D shows a drop, staying at about 49 K. As the concentration of $BaSO_4$ reaches 20-25 wt%, the CCT-D surges to approx. 120-160 K, three and four times greater than the value at 15 wt% $BaSO_4$. Greater $BaSO_4$ concentration demonstrates the notable decrease in CCT-D, and the bottomed out CCT-D value is about 40 K with 50 wt% $BaSO_4$ concentration. This result indicates that the greatest CCT distribution uniformity is obtained with 50 wt% $BaSO_4$. Another option of $BaSO_4$ concentration for improved CCT distribution uniformity is 15 wt%.

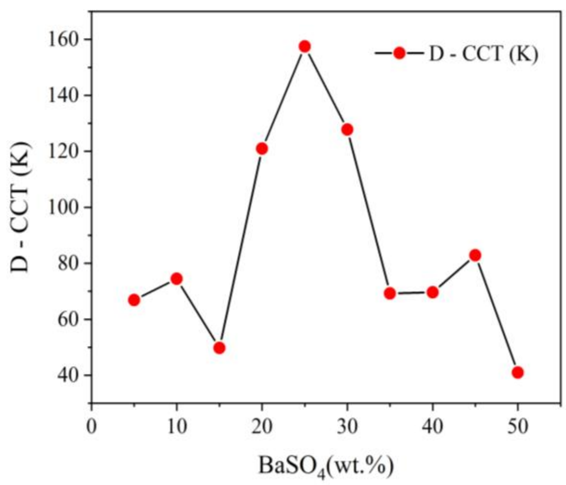


Fig. 5. Colour-temperature deviation corresponding to $BaSO_4$ concentrations (colour online)

Besides the CCT distribution uniformity, the color rendering fidelity is considered to evaluate the quality of LED light. The color rendering index (abbreviated as CRI) and color quality scale (abbreviated as CQS) are simulated for assessment in this paper, illustrated in Figs. 6 and 7, respectively. The high-fidelity LED light requires the wide spectrum of emission colors, especially including the red, green, blue to reproduce the color of projected objects. The light source with high red or blue emission color could give the good CRI parameter but discomfort to human eyes [23, 24]. Hence, to ensure the good-quality LED light, both CRI and human visual inclination should be evaluated together. The CQS can addressed this problem since its evaluation elements includes these two aspects. As such, CQS is an essential parameter for gauging the quality of white light [25]. It is shown in the figures that the CRI and CQS both decrease with increasing $BaSO_4$ concentration. This means the scattering of $BaSO_4$ does not contributes to enhancing the color rendition performance of the LED light. The addition of $BaSO_4$ just enhances the scattering of blue light and part of the converted yellow light but does not compensate for the shortage of red and green emission colors. In addition, the intensive scattering can degrade the color balance between the mentioned three colors, leading to the imperfections in rendered colors. Due to the lack of this balance caused by greater $BaSO_4$ concentrations, CRI and CQS, which are parameters that evaluate quality of light based on the spectrum of emission colors, show a tendency to decline.

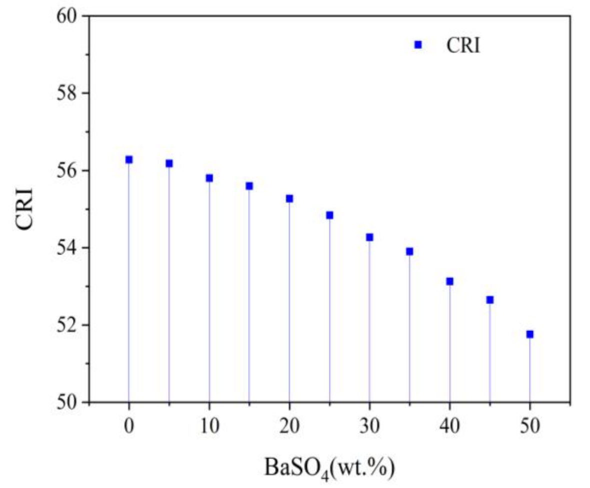


Fig. 6. CRI corresponding to $BaSO_4$ concentrations (colour online)

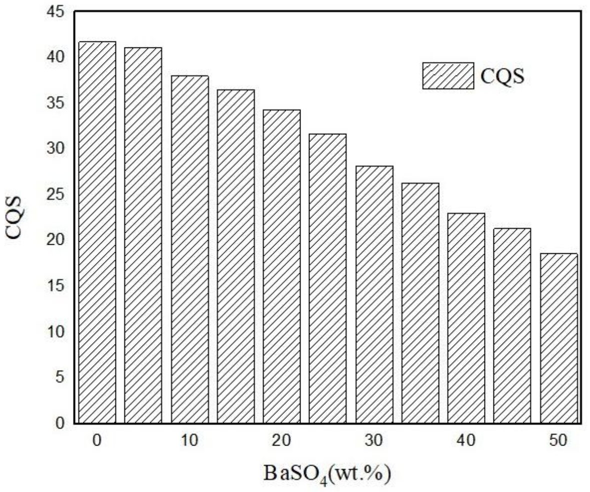


Fig. 7. CQS corresponding to $BaSO_4$ concentrations

4. Conclusions

The high-reflectivity $BaSO_4$ was integrated into the phosphor layer for influencing the scattering of the incident and converted light rays. The Mie-scattering theory and LightTools software were applied for simulation of the scattering performance of $BaSO_4$ with different concentrations, ranging from 5 wt% to 50 wt%, in the layer. The data from simulation demonstrated that increasing $BaSO_4$ amount increased the scattering coefficients, especially in the cases of short wavelengths. The CCT distribution at wider angles was improved with high $BaSO_4$ concentrations, as a result of strong scattering events. At 50 wt%, the CCT deviation is the lowest, meaning that utilizing $BaSO_4$ to regulate scattering events contributes to the better uniformity of CCT distribution. Another concentration of $BaSO_4$ showing the nearly equivalent CCT-distribution result was 15 wt%. However, the luminous intensity of the LED decreased gradually with increasing concentration of $BaSO_4$ due to the scattering hindering the transmission efficiency of the LED. The color rendition parameters (CQS and CRI) were also not benefited from scattering improvement by $BaSO_4$ since the red and green emission colors were not supplemented. Therefore, to achieve improvement in luminous flux and color rendering performance, the intensive scattering is not advisable and using additional or emission-tuneable phosphor materials should be considered in future research of $BaSO_4$ utilization in LED design.

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*Corresponding author: ntploan@ptithcm.edu.vn