ZnO-doped yellow phosphor compound for enhancing phosphor-conversion layer's performance in white LEDs

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This research paper builds upon prior investigations by introducing the utilization of ZnO particles to augment the performance of YAG:Ce phosphor films through the manipulation of tunable scattering factors. Employing Mie-scattering theory, the optical simulation works are performed to demonstrate the performance of a conventional blue-excited while Light-Emitting Diode (LED) upon the varying concentration of ZnO nanoparticles. Specifically, the study carried out the assessments on optical properties, including lumen output, color rendition ability, and light distribution uniformity with ZnO amounts ranging within 0-35 wt%. The scattering simulation of the ZnO is also presented to understand the change in the LED's optical results. Based on previously studies, the simulation inputs were determined, including wavelength of the blue LED chip, particle radii of the phosphor and ZnO, the conversion layer thickness, and the driving current of the LED. The acquired data demonstrate the capability of ZnO to enhance the scattering of lights, the angular color distribution, and lumen efficiencies of white light emitted by the LED by selecting appropriate ZnO concentration. These findings highlight the potential of ZnO for the optimization of LED technologies.

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1. Introduction

In recent years, the demand for high-brightness, highly efficient, and directionally optimized light-emitting diodes (LEDs) has been spurred by their widespread commercial applications in lighting technology and displays [1, 2]. Presently commercialized white LEDs typically pair a semiconductor chip emitting in the ultraviolet or blue wavelength range with one or more phosphor coatings [3], either on-chip or in remote configurations [4-6]. Despite their commendable photometric parameters, contemporary LED-based devices confront challenges related to luminous efficacy and a lack of directional coherence, stemming from the inherent isotropy and incoherence of LED radiation [7, 8].

In this context, zinc oxide ZnO has recently rekindled significant interest owing to a myriad of properties that position it as a superior alternative to existing materials. As a wide band-gap material, ZnO's electronic structure makes it an attractive candidate for luminescent devices. Notably, ZnO has gained prominence for its potential application in low-voltage luminescence technologies, exemplified by field emission displays. In its pure form, ZnO emits a narrow luminescence band in the blue–violet spectral region, and through the incorporation of various dopants, it exhibits emissions spanning from blue to the orange spectral region [9, 10]. The versatility of ZnO-based alloys presents an intriguing possibility as alternatives to GaN-based alloys currently utilized in ultraviolet (UV) optoelectronic devices. Moreover, the customizability of ZnO into nanostructures, coupled with its refractive index that aligns between the refractive indices of GaN-based LEDs and air, positions ZnO as a compelling candidate for enhancing LED efficiency. Beyond its role in efficiency improvement, ZnO also demonstrates proficiency in enhancing light scattering, further underscoring its appeal in the realm of LED technology [11, 12].

Considering findings from previous study, this paper presented the simulated performance of blue-radiation excitable ZnO particles to enhance the YAG:Ce phosphor film's performance via the tunable scattering factors [13-16]. The Mie-scattering theory is the foundation of this simulation work. The MATLAB computation programs is utilized to calculate the performances depending on the input data of the material. Here, the scattering properties are monitored with different ZnO contents, from 0 wt% to 35 wt%. Accordingly, the LED's optical properties, including color distribution uniformity and rendition as well as lumen intensity, are assessed. The obtained data indicates the ability of ZnO in enhancing the LED white light efficiency.

2. Simulation method

The wide-range and profound experiment data from previous works play an essential role in selecting the input parameter for simulation in our paper. The study on ZnO nanoparticles typically show that the material exhibited significant absorption band at around 350-370 nm [13]. This indicates that ZnO is well excited by ultraviolet excitation source. Besides, other studies demonstrated blue-light excitable ZnO particles synthesized with sol-gel combustion technique, showing a narrow band of absorption at around 460 nm, in addition to the 380-430 nm band [14, 15]. Additionally, the particle size of ZnO was various, ranging from about 3-185 nm in the previous studies, depending on the conditions of the synthesis process [16-18]. Some study showed the ZnO size of ~10 nm- 80 nm with spherical morphologies and narrow size distribution [13, 19, 20].

In this paper, Mie-theory are used for all simulation processes. Thus, all the luminescent materials, including the phosphor and ZnO, are considered spherical. The fixed input parameters for our simulation are the particle sizes of the phosphor and ZnO, the conversion layer thickness, and the driving current of the LED package. The variables include the concentrations of the materials and the tested light wavelengths for scattering. All of the input data are shown in Table 1.

Table 1. Input data for simulation

ZnO particle size	30 nm [19, 20]
Yellow particle size	6 µm [21, 22]
Conversion layer thickness	0.8 mm
Current	30 mV
ZnO concentrations	5 wt% - 35 wt%
Light wavelength for scattering test	380 nm – 780 nm

Besides, the scattering property of the ZnO with different concentrations is expressed with Scatteri Coefficient (μ_{sca}). The SC parameter can be computed as follows:

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

$$N(r) = N_{df}(r) + N_{pp}(r) \tag{2}$$

where $N(r)$ represents the distribution density of diffusional particles (mm^3), C_{sca} shows the scattering cross sections (mm^2), λ indicates the radiation wavelength (nm), and r defines the radius of diffusional particles (μm), the $N_{df}(r)$ and $N_{pp}(r)$ show the density of diffusional and phosphor spheres, respectively.

The concentration of ZnO varies in the range of 5-35 wt%, inducing the change in YAG:Ce phosphor concentration, as demonstrated in Fig. 1. When the amount of ZnO added into the luminescent-converted layer was higher, the amount of YAG:Ce showed a drop in general. However, when initially added 5 wt% ZnO, we noted a small increase in YAG:Ce concentration. The variation of YAG:Ce phosphor amount is essential to keep the correlated color temperatures (CCT) stable as well as regulating the scattering properties of the film. Thus, it can be implied that by adjusting ZnO amounts, the optical performance of the prepared LED package could be improved [23, 24].

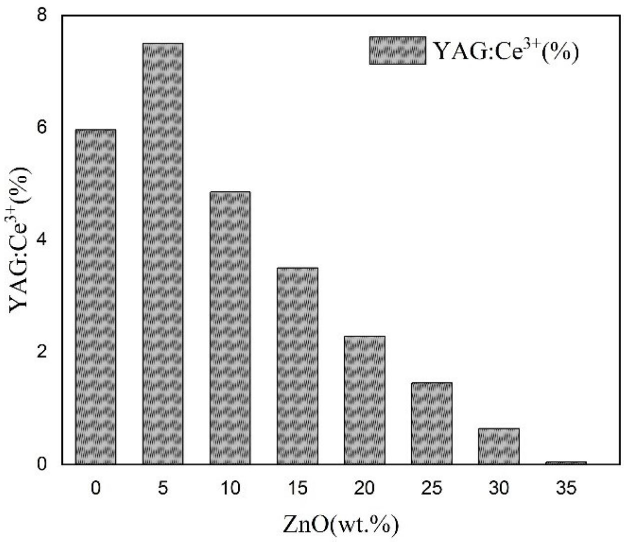


Fig. 1. YAG:Ce concentrations with varying ZnO amounts

3. Computation and discussion

The scattering parameter of the ZnO particles in the phosphor sheet was calculated with Mie-scattering theory and displayed in Fig. 2. The scattering coefficients were monitored with different wavelengths from 380 nm to 780 nm. The greater ZnO concentration, the higher the values of scattering coefficients, regardless of the light wavelengths. Particularly, the scattering is most notable with 480 nm and 580 nm wavelengths, indicating that the light in the visible light region of light blue to yellow. The obtained scattering results also imply that the LED chip blue radiation and phosphor layer’s emitted yellow-green light will be distributed in a wider angle, contributing to enhancing the uniformity of light distribution on the chroma scale [25].

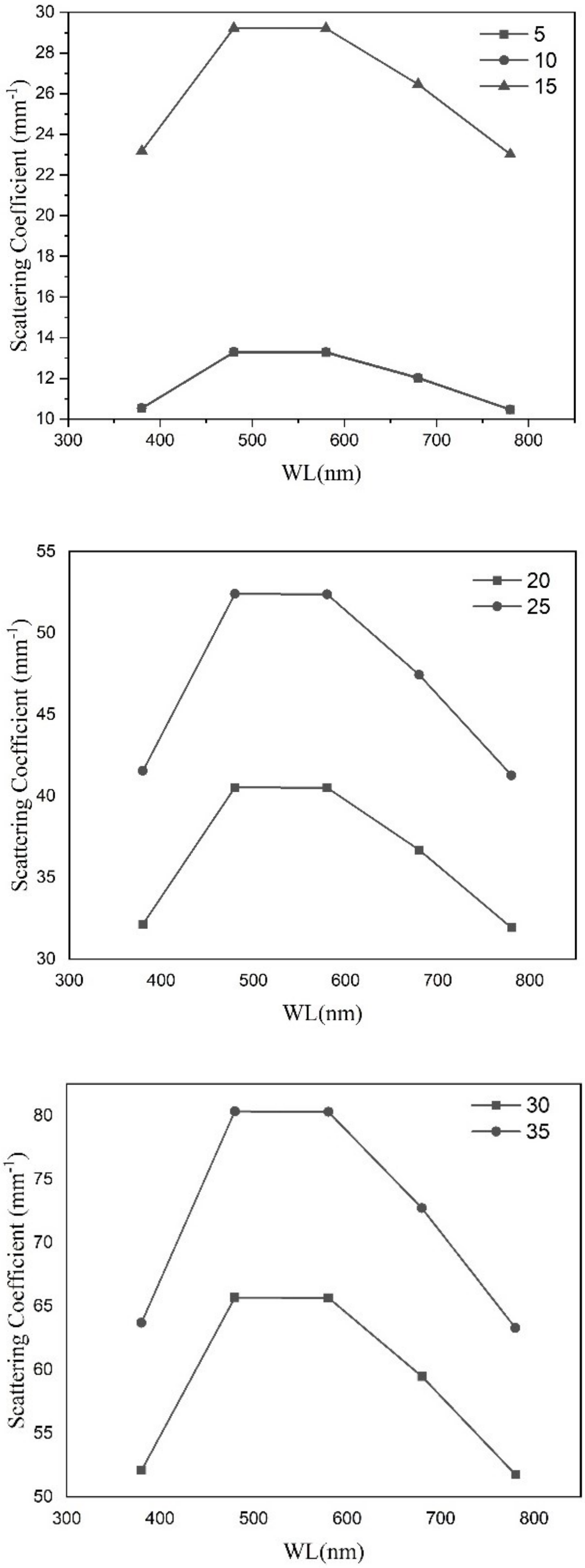


Fig. 2. Scattering of light with varying ZnO amounts

The transmission power of the LED on varying ZnO concentration is shown in Fig. 3, displaying two notable emission bands, matching with the high-scattering light regions in Fig. 2. In particular, with ZnO in the package, the transmission power exhibits two notable bands: a narrow band in blue region (430-480 nm) and a broader band in greenish-yellow region (500-590 nm). Despite the increasing of ZnO amount, the transmission peaks' positions do not change much. In the case of 5-15 wt% ZnO, the sharp peaks centered at ~460 nm and ~545 nm are recorded. Beyond 15 wt% ZnO, another peak in the greenish-yellow region appears, centered at about 570 nm. Besides, as the ZnO concentration increases, the 545 nm is more notable than the others, showing that the green radiation is extracted well and could have larger proportion on the chroma scale. The reported ZnO emission wavelengths also presented that the defects in ZnO allowed the material to emit light from blue to red, whose intensity depended on the excitation source and the driving current strength [26-29]. Hence, under the blue-light excitation and a fixed current at 30 mV, the green-to-yellow peaks in transmission power of the LED are probably induced by the ZnO, in combination with the yellow phosphor [19]. On the other hand, the intensity of the whole transmission band declines in response to the increasing ZnO amount in the conversion film. These findings demonstrate a certain effect of using high ZnO concentration: it could harm the chromatic rendition and luminosity of the LED output [30].

The color rendition and luminosity of the LED light output are depicted in Figs. 4 and 5, respectively. The color rendition assessment in this work includes the color rendering index (Fig. 4a) and color quality scale (Fig. 4b). Overall, the color rendering index and quality scale decline when we increase the amount of ZnO addition. However, with 5 wt% of ZnO in use, we noticed a small increase in both color quality scale and rendering index. More than 5wt% of ZnO, these two parameters start to decrease gradually. In Fig. 5, the lumen intensity of the LED light output showcases the similar trend, in which 5wt% ZnO results in the increase, which is also a peak of measured luminous intensity. The luminosity also declines when the concentration of ZnO is beyond 5wt%. These results align with the data of transmission power in Fig. 3. The transmission bands recorded with 5 wt% is relatively broader and the strongest, facilitating the color rendition and luminous performance of the LED output. Besides, the scattering of light in the case of 5 wt% is not too intense to hinder the extraction of blue light, resulting in the increase of luminosity as seen in Fig. 5.

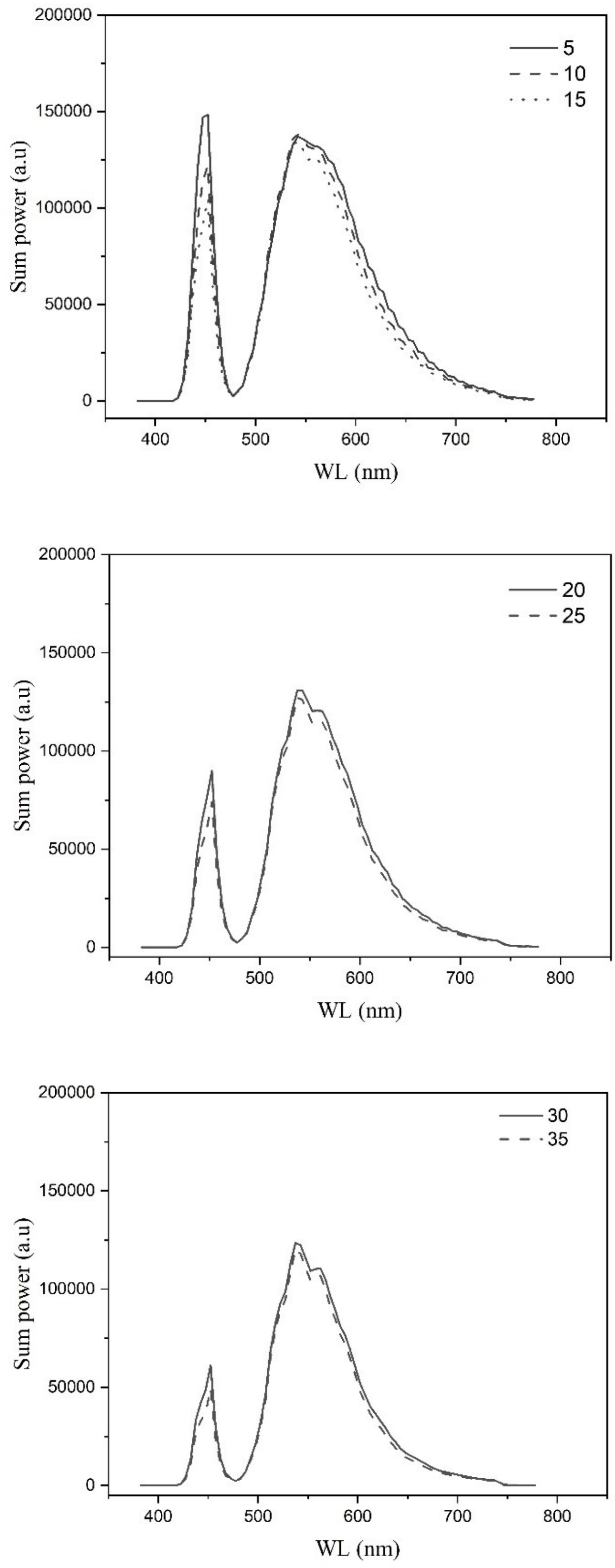
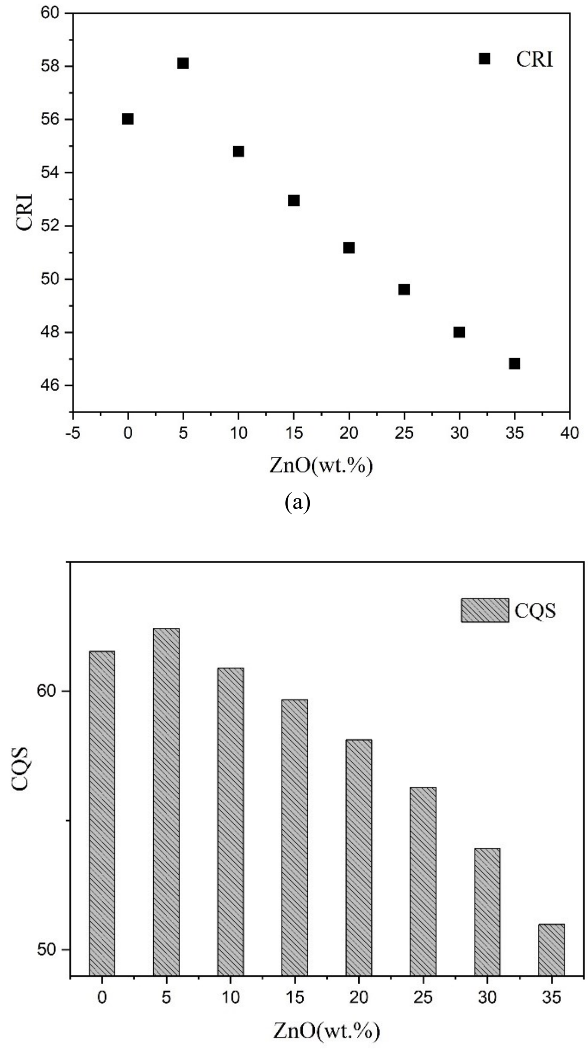


Fig. 3. Light transmission power of prepared LED with varying ZnO amounts



(b)

Fig. 4. Color rendition of LED with varying ZnO amounts: (a) color rendering index and (b) color quality scale

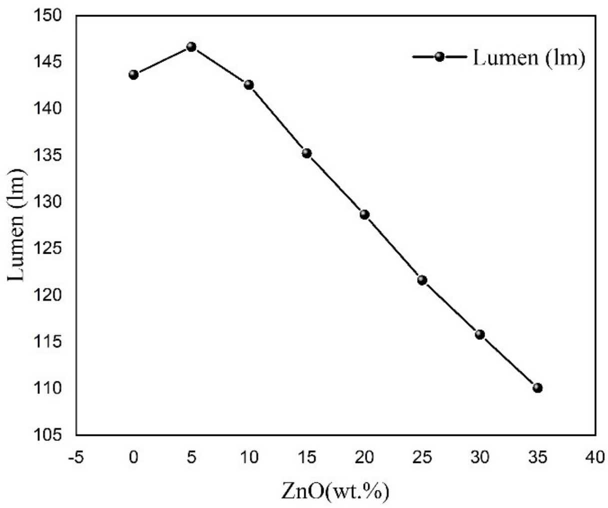


Fig. 5. Luminosity of LED light output with varying ZnO amounts

Though the scattering effects could reduce the color reproduction and lumen output of the white LED, it can induce the uniformity light distribution on the chroma scale. The analysis presented in Fig. 6 provides a compelling insight into the angular CCT ranges of LED output as influenced by varying concentrations of ZnO. We observed a notable reduction in the variation of CCT values across diverse viewing angles with increasing ZnO concentration. Particularly, the CCT-range is gradually stabilized around 5000 - 5050 K as ZnO concentration escalates around 10-20 wt%. Fig. 7 further substantiates these observations by illustrating the delta CCT values, underscoring the efficacy of higher ZnO concentrations in enhancing the uniformity of light dispersion from the LED. The data are well matched with the CCT ranges in Fig. 6, showing that the notable decline of delta-CCT level is when using 10-20 wt%. Moreover, the smallest delta CCT value is accomplished with 20 wt% ZnO concentration, indicating an optimal point for achieving the highest CCT uniformity in this study. Beyond this concentration, delta CCT values rise, albeit remaining lower compared to the 5 wt% ZnO case. This can be attributed to the improvement in scattering behavior, particularly with higher concentrations of ZnO. The greater scattering facilitates the dispersion of blue and longer-wavelength lights towards the edges of the LED package. This strategic light distribution mitigates the persistent yellow-ring issue prevalent in conventional LED models, as it encourages the amalgamation of multiple light components, reducing the prominence of a yellow-only emission. These findings underscore the pivotal role of ZnO concentration in not only improving CCT uniformity but also addressing practical challenges in LED models, offering promising avenues for advancing LED technology.

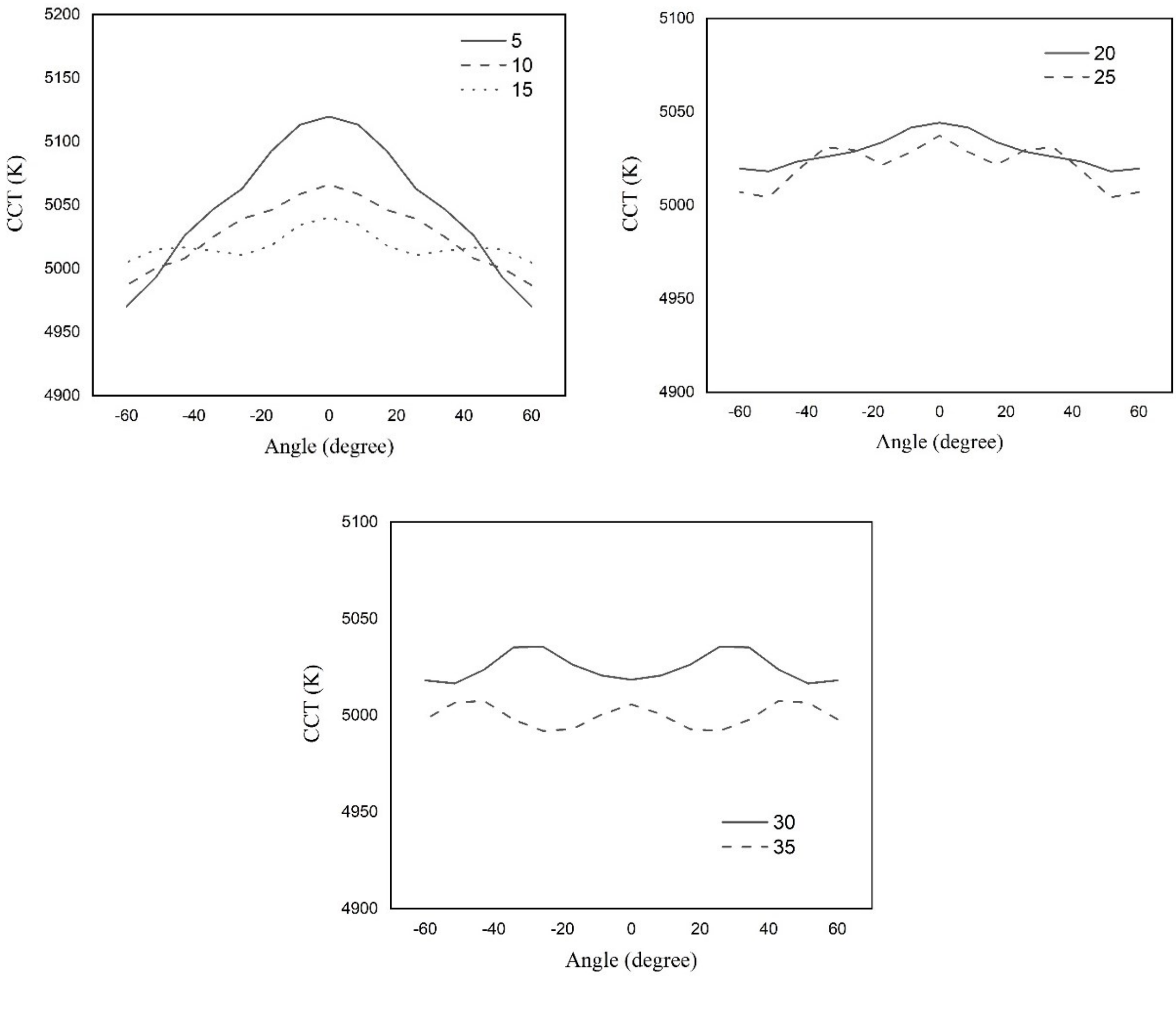


Fig. 6. CCT ranges of LEDs with varying ZnO amounts

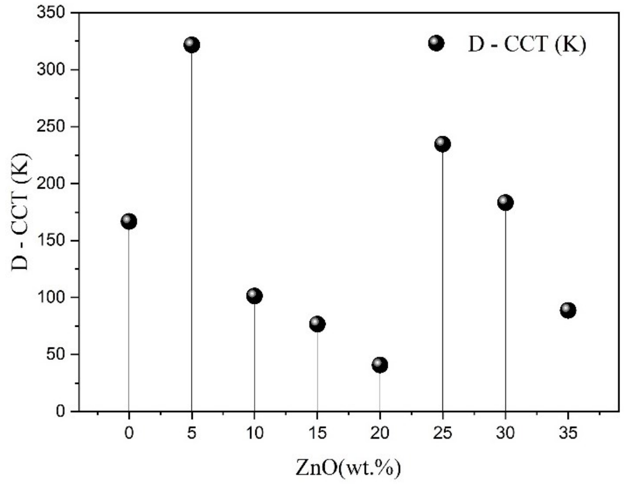


Fig. 7. Delta-CCT of LED light output with varying ZnO amounts

4. Conclusions

In summary, this paper used Mie-scattering theory-based simulation to demonstrate the effects of ZnO varying concentration on blue-excited LED. The input parameters were selected based on previous experimental studies. The obtained results of this work act as the essential supplementing reference to enrich the database of ZnO utilization in white-LED development. In particular, the significant impact of ZnO concentration in enhancing light scattering for improved LED lighting performance was presented. The YAG:Ce film was simulated with the addition of ZnO nanoparticles (30 nm), and the ZnO concentration ranges from 5 wt% to 35 wt%. The investigation reveals that using ZnO concentration of 5 wt% not only lead to improvements in lumen output but also color reproduction efficiencies of the prepared white LED. Yet, continuously increasing ZnO concentration more than 5 wt%, the LED's luminosity and color rendering efficiency declines substantially. The observed reduction in luminous energy and a deficiency in blue and red-light emission with increasing ZnO concentration elucidate the intricacies of light dispersion within the LED.

However, the positive outcome emerges in the form of enhanced scattering, manifesting in improved color uniformity, as indicated by the diminishing correlated color temperature difference (Delta-CCT). Remarkably, within the concentration range of 10-20 wt%, a pronounced decline in Delta-CCT is evident, reaching an optimum at 20 wt% ZnO concentration. This optimal concentration becomes pivotal in achieving the highest color uniformity, underscoring the significance of meticulous ZnO concentration selection for warm white LED lighting. As we navigate the intricate balance between ZnO concentration and LED performance, this study contributes substantively to the ongoing efforts in optimizing LED technologies for the future of efficient and uniform warm white LED lighting solutions.

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