

# The Use of Green-Phosphor LuAG:Ce-Al<sub>2</sub>O<sub>3</sub> for High-Luminosity Light Emitting Diode Packages

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## ABSTRACT

The LuAG:Ce-Al<sub>2</sub>O<sub>3</sub> (LAGCA) green phosphor ceramic (GPC) is proposed for high-power white light emitting diodes (LEDs) in this paper. The luminescent properties of the GPC are examined with proper characterizing tools and under laser excitation. Then, LAGCA ceramic layer of 0.6 thickness is applied to fabricate the white LED. The results show LAGCA GPC is promising for high-power LED applications. The phosphor ceramic presents high thermostability and quantum efficacy and intense green emission peaking at nearly 550 nm. In the LED package, the amount of LAGCA in the composite layer is varied. The increasing dosage of LAGCA gives enhancement to the lumen output of the LED. However, the correlated color temperature stability and chromatic rendition declines. Thus, further improvements in LAGCA ceramic need to be carried out in the future works. Besides, with the intense green emission, the LAGCA ceramics can be combined with red luminescent materials to increase the color performance of the LED lighting.

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## 1. INTRODUCTION

Artificial lighting applications utilizing the solid-state illumination technology has quickly emerged in the lighting industry in recent decades and caught considerable attention for the development of new lighting generations. This type of light source presents many benefits, including being compact and friendly with nature, high light brightness and efficiency, long service life, and robustness [1-3]. Among commercial phosphor materials, the YAG:Ce<sup>3+</sup> yellow phosphor is the most applied material. Yellow-emission YAG:Ce<sup>3+</sup> phosphors have been applied extensively because they exhibit good blue-light absorption and lumen efficacy [4, 5]. Traditionally, they are mixed with resin composites, but the efficiency drops gradually after extended service time, especially in the high-power light LED in which the generated heat is significant. This indicates that the traditional YAG:Ce/resin composite is prone to aging and yellowing problems. Many advancements have been made to improve the durability of the YAG:Ce<sup>3+</sup> materials. For example, the YAG:Ce<sup>3+</sup> phosphor-in-glass [6] and phosphor ceramics [7] were suggested as high thermal-conductivity materials. Notably, the phosphor ceramic presents more significant thermo-stability and conductivity, higher homogeneity and robustness, and shorter preparation time. Hence, it can effectively provide high-power white light for LED applications under high density excitations like laser, ultraviolet, and infrared radiations.

The luminescent properties of the phosphor ceramic can be influenced by many factors, such as the doping dosage of the ion activator, the secondary phase (SP), and the particle size [8-10]. The SPs often utilized to improve the transmission of the incident blue light, resulting in higher blue-light absorbing efficiency. Some of the most used SPs include ZrO<sub>2</sub>, MgAl<sub>2</sub>O<sub>4</sub>, and Al<sub>2</sub>O<sub>3</sub>. The Al<sub>2</sub>O<sub>3</sub> SP has been doped into the YAG:Ce<sup>3+</sup> phosphor ceramic and results in higher luminescence output for the ceramic. The Al<sub>2</sub>O<sub>3</sub> SPs in the YAG:Ce<sup>3+</sup> ceramic act as effective scattering centers as they possess noticeable thermo-conductivity and well match with

the YAG:Ce<sup>3+</sup> ceramic configuration. Therefore, they efficiently reduce the thermal quenching and increase the luminescent efficacy for the material [11, 12].

Besides the YAG:Ce<sup>3+</sup> ceramic, the LuAG:Ce<sup>3+</sup> ceramic is another potential material for the light converter materials as they present more significant thermo-conductivity and much lower thermo-quenching, which can stimulate their robustness for extended service periods. The ion activator Ce<sup>3+</sup> in the LuAG material can induce the strong green emission. Therefore, on the basis of YAG:Ce<sup>3+</sup>-Al<sub>2</sub>O<sub>3</sub> phosphor ceramic development, the LuAG:Ce<sup>3+</sup>-Al<sub>2</sub>O<sub>3</sub> (LAGCA) green-emission phosphor ceramic (GPC) is suggested for the high-power white LED (HP-WLED) in this work. The prepared LAGCA ceramic exhibits high thermo-stability and strong green emission, and good blue-light absorption, suitable for HP-WLED applications. Nevertheless, the observed CCT stability and color rendition efficiency of the WLED with LAGCA ceramics are not improved, indicating the needs of structure modification for the GPC in the future works [13, 14]. However, the strong green emission of the ceramic makes it suitable for the combination with red converter materials to improve the color rendering index (CRI) of the WLED devices.

## 2. METHOD

The ingredients and process of synthesizing the LAGCA ceramic are shown in Table 1. The solid-phase reaction and vacuum sintering are applied to synthesize the required ceramic. The doping concentration Ce<sup>3+</sup> ion in the phosphor composite ceramic is fixed at 0.3 wt%. The amount of Al<sub>2</sub>O<sub>3</sub> is determined at 40 wt%. The tetraethyl orthosilicate is added as sintering additive with an amount of 0.5 wt%. The ingredients are weighed stoichiometrically before starting the synthesis of LAGCA ceramic [15, 16]. The characterization methods of the fabricated LAGCA ceramics are demonstrated in Table 2. The WLED using the as-prepared LAGCA ceramic disks is depicted in Figure 1. The depicted WLED structure features nine disks separated into two symmetrical columns flanking a non-symmetrical column which each column containing an equal number of three disks. The LAGCA amounts used for examining WLED light properties varies from 5 wt% to 45 wt%.

Table 1. The ingredients and synthesizing process of the LAGCA ceramic

Ingredients	Synthesizing process
Lu <sub>2</sub> O <sub>3</sub>	1. Mixing the ingredients by ball-milling for 12 hours; ethanol is used as a dispersant. 2. Drying the mixture at 70 degrees Celsius for 2 hours (in air). 3. Sieving the mixture through a 200-mesh screen. 4. Calcinating the product at 600 degrees Celsius for 4 hours (in air). 5. 20-MPa pressing the obtained mixture into disks with 18-mm diameter. 6. Get the disks compacted using 250-MPa cold-isostatic-pressing process for 3 minutes. 7. Vacuum sintering the disks for 10 hours at 1750 degrees Celsius. 8. Annealing the disks for 10 hours at 1450 degrees Celsius. 9. Polishing the ceramic disks' surfaces to obtain the thickness of 0.6 mm for further examination.
Al <sub>2</sub> O <sub>3</sub>	
CeO <sub>2</sub>	

Table 2. The characterization for the LAGCA ceramic

Characteristics	Determining methods
Morphology	Hitachi SU8220 field-emission scanning electron microscope, coupled with an energy-dispersive X-ray spectroscope
Total transmission	Varian Carry 5000 UV-Vis and NIR spectrophotometer
Internal quantum yield	Spectroradiometer equipped with a blue laser diode (450 nm) and an integrating sphere
Emission and excitation spectra	Edinburgh FLS1000 fluorescence spectrophotometer
Temperature-dependent luminescence spectra	Hitachi F-4600 fluorescence spectrophotometer
Lighting properties of WLED with LAGCA ceramic	HASS-2000 spectroradiometer equipped with an integrating sphere

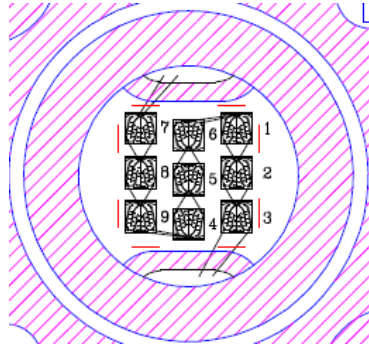


Figure 1. Structural depiction of WLED package used in the paper

### 3. RESULTS AND DISCUSSION

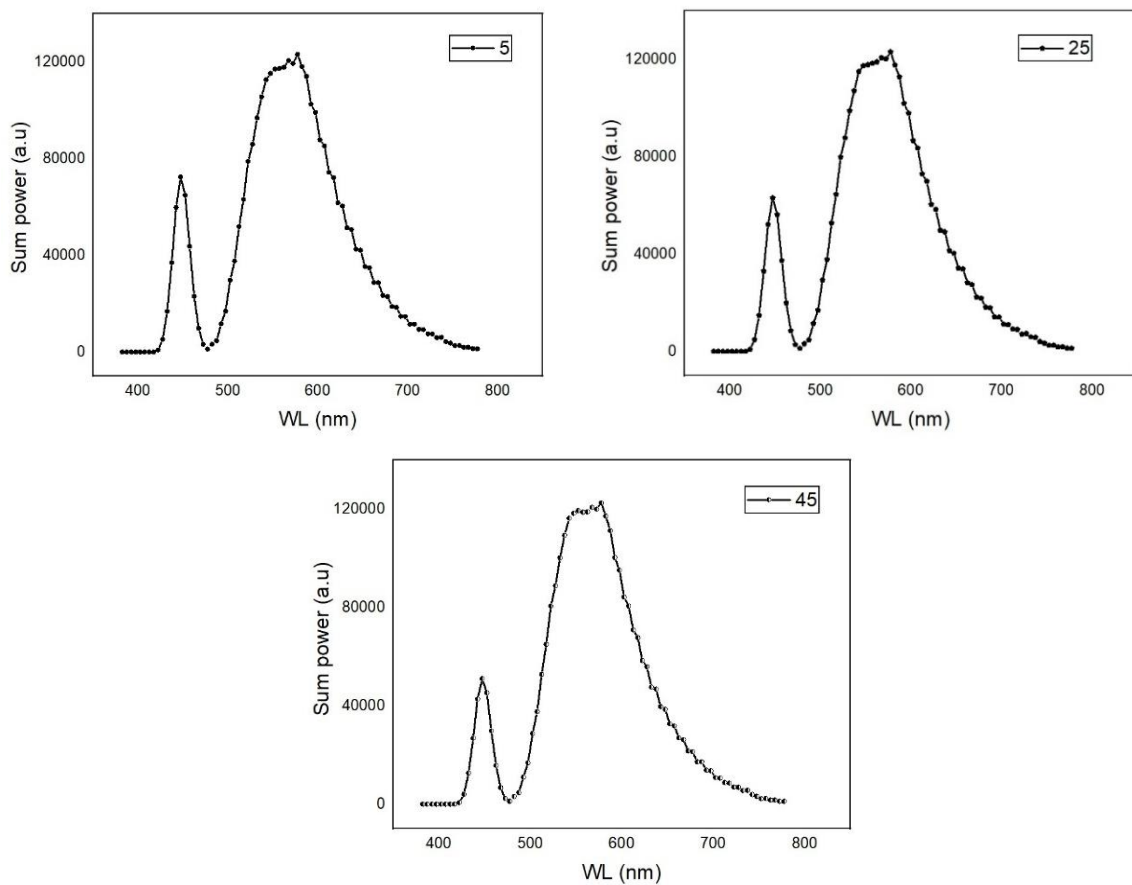


Figure 2. The total transmission power of the WLED in the presence of LAGCA phosphor ceramics

The total transmission power of the WLED in the presence of the LAGCA ceramic is shown Figure 2. The spectral bands in 450 nm and 535-600 nm are observed, which are probably induced by the addition of LAGCA ceramic. The ceramic is well excited under the 450-nm blue light excitation, giving the absorption peaks at around 450 nm due to the transition  $4f-5d_1$  of the ion dopant  $Ce^{3+}$ . Additionally, the intensity of spectra in the green region of 545-595 nm are more noticeable than the other, indicating that the blue-light conversion efficiency is enhanced, making it appropriate for high intensity green illumination [17, 18]. The enhancement in blue light conversion can be attributed to the enhanced scattering efficiency of emitted blue light in the WLED package when adding the GPC. Besides, as the LAGCA ceramic is used in addition to the  $YAG:Ce^{3+}$  phosphor layer in the WLED model, the increase in LAGCA amount induces the reduction of  $YAG:Ce^{3+}$  dosage. The decrease of doping concentration of  $YAG:Ce^{3+}$  yellow phosphor with the higher LAGCA amount

is illustrated in Figure 3. Such decrease of YAG:Ce<sup>3+</sup> yellow phosphor dosage also supports the scattering effectiveness of the incident blue light to serve the luminous improvement.

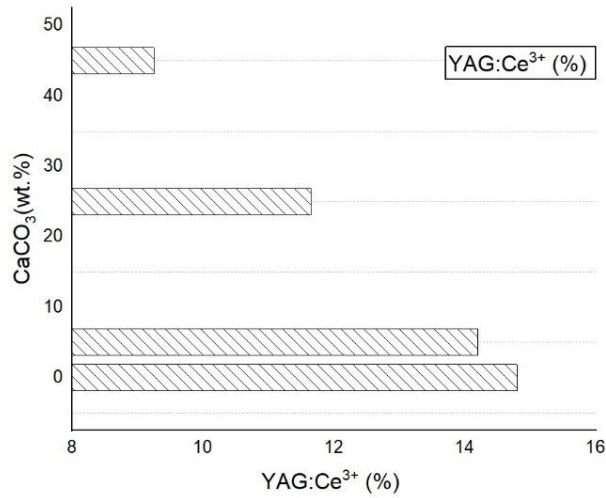


Figure 3. Doping concentration of YAG:Ce<sup>3+</sup> changes with higher LAGCA amounts

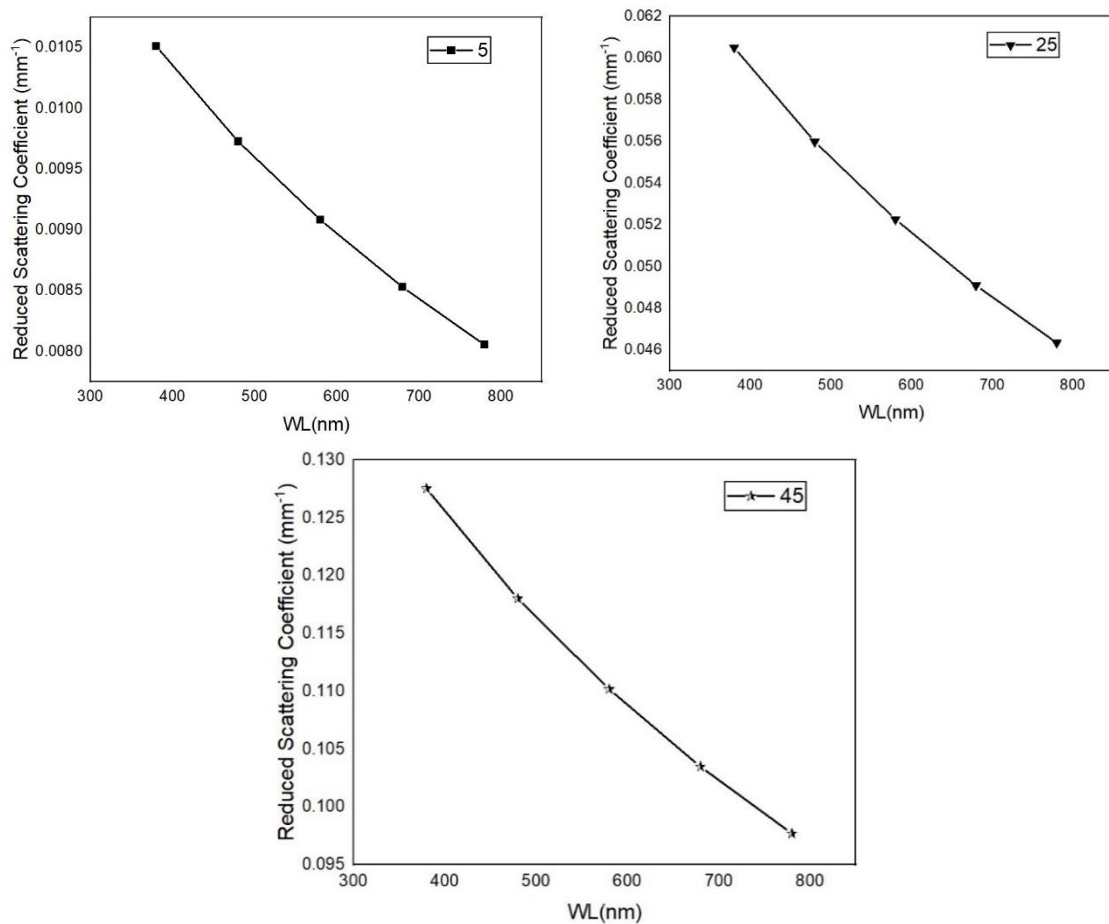


Figure 4. Reduce scattering coefficients of light in WLED in the presence of LAGCA phosphor ceramics

The scattering performance of the WLED increases as the concentration of LAGCA becomes larger, which is depicted in Figure 4. The ability to enhance the scattering performance of blue light could be ascribed

to the structure of the LAGCA ceramic. In the GPC, there are some pores on the boundaries of the phosphor particles, which plays an important role as scattering centers to redirect the incident light path so that the absorption effectiveness can be improved. In addition, the addition of  $Al_2O_3$  considerably stimulate the multiple blue-light scattering as the propagation of light is supported. As a result, the blue-light conversion is more effective, leading to the higher quantum yield as well as higher emission intensity. Thus, the presence of LAGCA ceramic can promote the luminosity strength of the WLED package [19, 20].

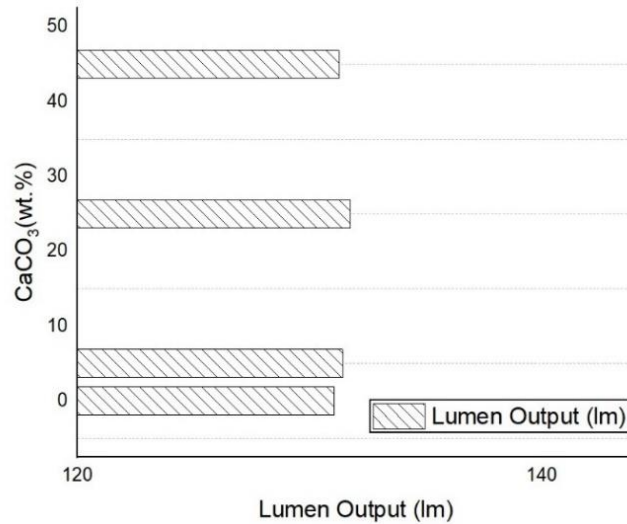


Figure 5. Lumen output changes with higher LAGCA amounts

Figure 5 illustrates the lumen output of the WLED with different amount of LAGCA. The intensity of lumen output increases as the amount of LAGCA increases from 0 wt% to 25 wt%. When the LAGCA amount continues increasing to 45 wt%, the luminosity starts to decline. It could be because of the reabsorption of the blue light as the LAGCA amount is significantly high. Besides, the increase in temperature during the operation of the WLED could contribute to induce the shorter-wavelength reabsorbing. Therefore, the emission peaks in spectra composition will move to the longer-wavelength region, resulting in the decrease in luminosity. However, the decrease of the WLED lumen output with high LAGCA amount is not too significant as the GPC has good thermostability.

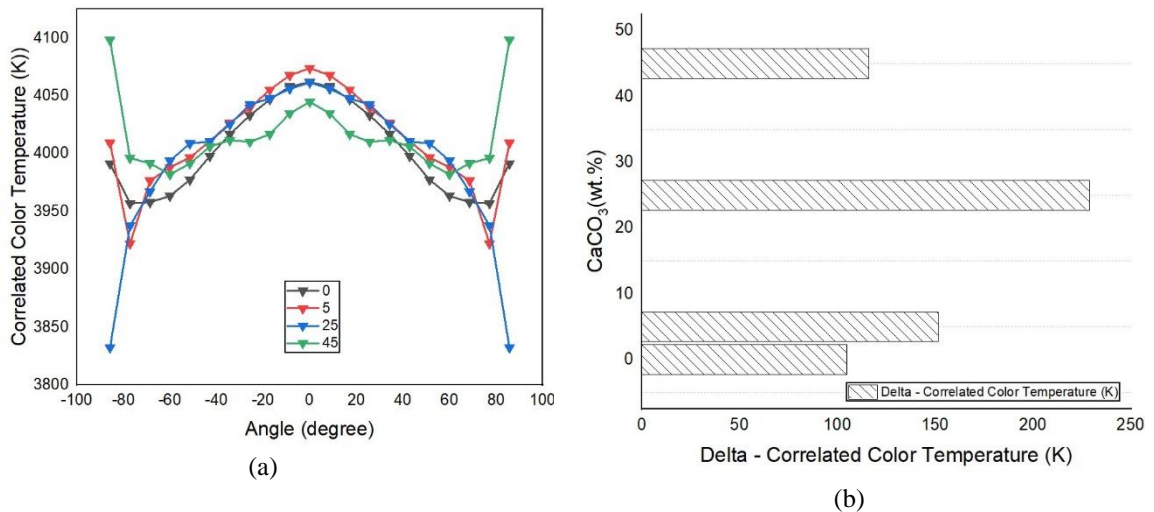


Figure 6. Color temperature (CT) properties of the WLED with different LAGCA amounts: (a) Angular CT and (b) CT deviation values

The color temperature (CT) properties of the WLED with different LAGCA amounts are shown in Figure 6, in which Figure 6a shows the angular CT values, and Figure 6b shows the deviation of CT values with increasing amount of LAGCA. The deviation of CT can support to determine the color uniformity of the WLED; the lower CT deviation indicates the higher color uniformity. It can be interpreted from the graphs that the CT increases as the LAGCA is introduced into the package (0-25 wt%). Increasing the LAGCA amounts above 25 wt% leads to the significant decrease in CT deviation. This can be the result of blue-light reflection on the ceramic surfaces. Though the CT at 45 wt% of LAGCA amount is still slightly higher than the one at 0 wt% LAGCA, it indicates the possibility in enhancing the CT stability and color uniformity with LAGCA ceramic.

The CRI and color quality scale (CQS) of the WLED light with different amounts of LAGCA are used to examine the performance of color rendition capability, which are shown in Figures 7 and 8, respectively. In both figures, it is easy to observe the decline in CRI and CQS [21, 22]. Such phenomena could be ascribed to the reflection of blue light and color imbalance in the spectra composition. As the LAGCA is introduced, the scattering is enhanced due to the higher absorption efficiency of the incident blue light, leading to more converted green lights. This also means the green light will become dominant, which is not favorable to the color rendition. The high color rendition requires broad light spectrum including at least three key colors: blue, green, and red to reproduce the best color fidelity of the illuminated objects. Thus, to acquire the higher color rendition performance, the red components should be supplemented by using luminescent phosphors or red LEDs [23-25].

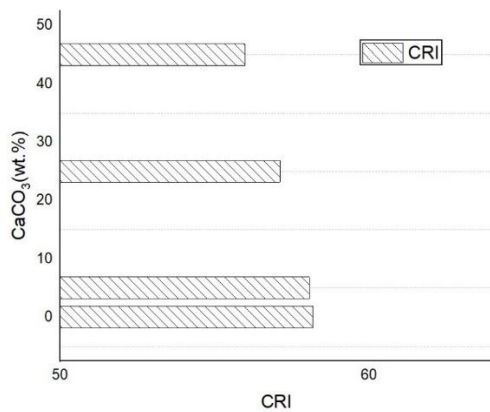


Figure 7. Color rendering index of the WLED with different LAGCA amounts

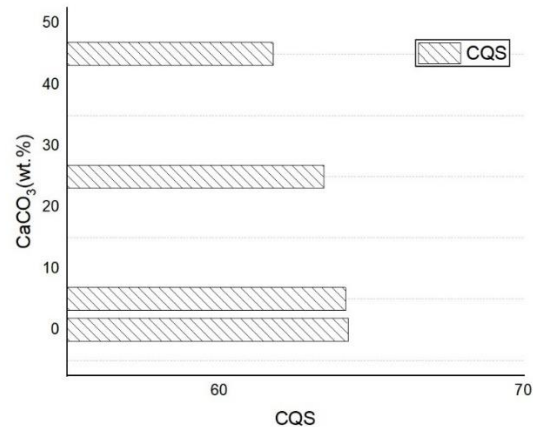


Figure 8. Color quality scale of the WLED with different LAGCA amounts

#### 4. CONCLUSION





The LAGCA green-emission phosphor ceramic was synthesized through the solid-phase reaction route and the vacuum sintering. The obtained LAGCA ceramic has good thermostability and high emission of green color (545 nm), making it suitable for high-power WLEDs. The scattering of blue light in the WLED is significantly induced in the presence of the LAGCA ceramic owing to the efficient scattering centers and the secondary phase Al<sub>2</sub>O<sub>3</sub> in the ceramic. The improvement of incident blue light leads to the higher blue-light absorbing and converting efficiencies, which stimulate the luminous intensity of the WLED. More than 25 wt% LAGCA can slightly reduce the luminosity because of the rise in blue-light reabsorption. The CT stability, CRI, and CQS of the WLED light shows the downward slopes in the presence of LAGCA owing to the reflection of blue light on the ceramic surfaces and the color imbalance in the total light spectrum. The LAGCA ceramic can be used with red-luminescent materials to increase the color quality of the high-power WLED lighting.

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