Green-phosphor Ca₃Sc₂Si₃O₁₂:Ce³⁺ for solid-status illumination: gel-combustion structural and luminous characteristics

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Article Info

Article history:

ABSTRACT

Received Nov 28, 2021 Revised Jul 11, 2022 Accepted Jul 28, 2022

Keywords:

Ca₃Sc₂Si₃O₁₂:Ce³⁺ Color rendering index Lumen efficacy Mie-scattering theory White light-emitting diodes Ca₃Sc₂Si₃O₁₂:Ce³⁺ phosphors with one stage and tiny dimension were effectively produced utilizing the gel-combustion technique at a lower heat (1,100 °C) than the traditional solid-status reaction technique (around 1,500 °C). The phosphors' crystal phase and microstructure, as well as their luminescence, were studied. The particle size is around 1m, which is significantly smaller than what the solid-status process produces. When particles are combined of silicon and deposited on a blue light-emitting diode (LED), finer particles can minimize interior diffusing. A significant green radiation is noticed that is caused by typical Ce³⁺ transition radiations $5d_{-}^{2}F_{5/2}$ and $5d_{-}^{2}F_{7/2}$. The stimulation spectra reveal a wide and high absorptivity at around 460 nm, indicating that it would be an excellent color conversion in white LEDs. The gel-combustion phosphor has a decay period of 54.65 ns.

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1. INTRODUCTION

Diodes that emit white illumination light-emitting diode (LEDs) are extensively utilized because of their numerous benefits, including power efficiency, extended lifetime, and safety [1]–[3]. Because of its cheap prices, easy manufacturing, and mature manufacturing, the coupling of phosphors emit yellow lighting yttrium aluminum garnet (YAG) with a blue-emitting chip has been the most preferred approach to white illumination production to far [4]. This sort of white LED, on the other hand, generates small red illumination and hence has a poor color rendering indicator, making it unsuitable for solid-state illumination. Rather than using YAG:Ce³⁺ phosphor, this issue might be handled by blending green and red phosphors. Combining redemitting and green-emitting phosphors with blue lighting released from a blue-color LED produces this kind of white lighting. Green phosphors described in patents and study document so far have certain drawbacks [5]. As a result, finding new phosphors emit green lighting with excellent quantum effectiveness and thermal steadiness is critical.

Shimomura and Kijima [6] discovered a new $Ca_3Sc_2Si_3O_{12}$: Ce^{3+} phosphorus emits green light that can be stimulated by blue lighting. In compared to traditional YAG and silicate phosphors, the phosphor exhibits a greater temperature stability and photoluminescence (PL) intensity. This green phosphor was recently produced using the solid-status reaction technique [7], [8]. It is well recognized that annealing solidstatus phosphors necessitates quite high temperatures and extended reaction periods. Moreover, using the

solid-status reaction technique to get a clear $Ca_3Sc_2Si_3O_{12}$ crystal phase has found hard [9]. Based on our prior research [10], [11], soft-chemistry approaches might be a good way to solve this issue and accomplish low-temperature production of oxide-based phosphors. The gel-combustion approach, among several soft-chemistry approaches, needs shorter reaction durations and lower heats. This technique is now utilized in both organic and inorganic synthesis [12]. The gel-combustion technique was used to effectively manufacture $Ca_3Sc_2Si_3O_{12}$: Ce^{3+} phosphors with a homogenous stage in this work. The phosphors' stage layout and morphology were investigated in this paper.

2. EXPERIMENTAL

A gel-combustion technique was used to make the phosphors. The ingredients of $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ phosphor were displayed as Table 1. For nearly 30 minutes, the combination was continually mixed. The resultant solution was burned within 10 hours in a dry oven at 65 °C to eliminate excess water before converting into a clear gel for progressive polymerization and drying at 85 °C to generate a xerogel [13]. The xerogel was burned at 700 °C inside a muffle furnace. A self-propagating combustion technique was used to burn the xerogel until a dry sponge specimen (referred to as the forerunner) was generated. Lastly, the forerunner was placed in an alumina crucible and annealed for 2 hours at 1,100 °C in a muffle furnace containing carbon particles to create a lowering atmosphere. The green-emitting phosphor $Ca_{2.9}Sc_2Si_3O_{12}:Ce^{3+}_{0.1}$ was then acquired. When comparing phosphors, a few were created via the standard solid-status reaction process as well. $CaCO_3$ (99.9%), CeO_2 (99.99%), Sc_2O_3 (99.99%), and SiO_2 (99.99%) were used as initial ingredients for the solid-status reaction (99.9%). The reagents were combined for around 15 minutes in an agate mortar to create a powder combination. Ultimately, utilizing carbon powders to create a decreasing atmosphere, the combination was placed in an alumina crucible and annealed for 1,500 °C within 6 hours for getting the results.

Table 1. Ingredients of Ca₃Sc₂Si₃O₁₂:Ce³⁺ phosphor

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Materials	By mole (%)
TEOS	99.9%
$Ca(NO_3)_2$	99.9%
$Sc(NO_3)_3$	99.9%
Ce(NO ₃) ₃	99.9%
5/5	

3. RESULTS AND DISCUSSION

The following process is proposed to explain the stage distinction between the phosphors produced by the two techniques. During the hydrolysis of tetraethyl orthosilicate (TEOS), a gel network can be produced using the gel-combustion technique, as (1) [14].

$$nSi(0C2H5)4 + 2nH20 \rightarrow (-0-Si-0-)n + 4nC2H50H.$$
 (1)

The deteriorate curves of the phosphors made of the gel-combustion and solid-status reacting techniques have been studied. A double-exponential decay curve may be successfully matched to the charting of the decay curve that uses the calculation [15].

$$I = A_1 e^{(-t/\tau_1)} + A_2 e^{(-t/\tau_2)},$$
(2)

where τ_1 and τ_2 are the rapid and late luminous lifespan elements, respectively. The equivalent fitting parameters are A_1 and A_2 . τ_1 and τ_2 are 6.91 ns and 57.8 ns, in turn, for the gel-combustion specimen. The durations of the solid-status technique-fabricated specimen are 6.13 ns and 59.48 ns, in turn. The equation [16] could be used to calculate the mean lifespan.

$$\tau = (A_1\tau_1^2 + A_2\tau_2^2)/(A_1\tau_1 + A_2\tau_2).$$
(3)

Figure 1 shows the opposite shift in the concentrations of green phosphorus $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ and yellow phosphorus YAG: Ce^{3+} . The said shift has objectives to maintain the consistency of mean conditional cash transfers (CCTs) and to influence the absorbing and dispersing of White light-emitting diodes (WLEDs) constructed with two-phosphor remote layers. Consequently, the white-light hue features and lighting flux effectiveness of the simulated WLED is modified. Color standards of the WLED is, therefore, determined by the $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ concentration chosen. When $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ grows from 2% to 20% Wt. then the

concentration of YAG: Ce^{3+} must be reduced to preserve the mean CCT. This occurrence displays on the WLEDs that have dissimilar color heats 5600 K - 8500 K [17]–[19].

Figure 2 depicts the influence of the Ca3Sc2Si3O12:Ce3+ weigh amounts on the WLED's transference spectrum. The proper concentration of the phosphor could be determined based on the optical-specification priporty of producers. WLEDs which have excellent color standard can lower illumination by a modest amount. As shown in Figure 2, white light is the spectral region's synthesis. These five figures show 6,000 K spectra. As can be seen eadily, the strength trend grows with concentration Ca3Sc2Si3O12:Ce3+ in two regions of the illuminating spectrum: 420-480 nm and 500-640 nm. This rise of the emitting illumination may be seen in the two-band radiation spectrum. Furthermore, if the scattering of blue light in the WLED is increased, the dispersing in the phosphor layer and in the WLED can be enhanced, allowing the color production to be more uniform [20]–[22]. At a high CCT, the color uniformity easily fluctuates, making it more challenging to keep the stability of color uniformity, easpically in the case of the remote-phosphor layout. Using Ca3Sc2Si3O12:Ce3+ has shown the efficiency for serving the goal of managing adequate color uniformity for the remote-phosphor layout at high CCTs. As a result, this research indicates that Ca3Sc2Si3O12:Ce3+ can make the color quality of WLEDs at both low and high hue temperatures better (at 5,600 K-8,500 K).





Figure 1. Modifying the concentration of phosphor to keep the mean CCT

Figure 2. The emitting bands of white light from a 6000 K WLED as a function of $Ca_3Sc_2Si_3O_{12}$:Ce³⁺ concentration

Consequently, this paper demonstrated the effectiveness of the produced lighting beam of this double-film distant phosphor layer, as displayed in Figure 3. From the preseted data, it can affirm that the increasing $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ concentration (2%-20% wt.) leads to a dramatically improved illumination strength of the white light. In Figure 4, when using increasing concentrations for $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$, the color variations were notably declined, regardless of CCTs. The absorptivity of the green-phosphor $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ sheet is can be used to explain this phenomenon. The blue-LED emitted light is transformed to green illumination when it is absorbed by the $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ phosphor. In addition to the blue illumination generated by the LED chip, the yellow illumination emitted by the $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ particles will be absorbed. Owing to the material's absorptivity, the blue illumination absorption from the LED chip is more than these two absorbs. As a result of the addition of $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$, the green light percentage in WLEDs increases, improving the color uniformity indicator. Hue homogeneity is one of the imperative factors required for an excellent-performance WLED device. This leads to the fact that the WLED that can introduce high hue homogeneity will have higher price in the martketplace. Hence, the cheap price of phosphor materials, such as $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$, in WLED production is advantageous. $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ can therefore be used in a variety of applications.

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Figure 3. The luminous flux of WLEDs as a function of $Ca_3Sc_2Si_3O_{12}$: Ce³⁺ concentration

Figure 4. The color deviation of WLEDs as a function of Ca3Sc2Si3O12:Ce3+ concentration

Color uniformity is simply one element to consider when assessing WLED color standard. Color standard cannot be stated to be excellent with a high color uniformity indicator. As a result, subsequent studies have developed a color rendering indicator and a color standard scale. When the color rendering indicator is lightened by a light, it assesses the true color of an item. The excess quantity of green illumination over the other two primary colors of blue and yellow can break the color balance, leading to lowering the chromatic fidelity. In the presence of the remote phosphor $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ layer, the data in Figure 5 show a small decrease in common representative intermediates (CRI). However, this small reduction of CRI is allowed since it is just a part that comprises the composite quality score (CQS), indicating that CQS is as a more necessary and more challenging indicator to obtain. Particularly, CQS carries the three important optical elements, including CRI, viewer's visual inclination, and the coordination of color, for WLED's quality evaluation. CQS is said to be an overall indicator of color standard [10], [23]-[25]. Figure 6 illustrates that CQS enhances with the use of green-phosphor Ca₃Sc₂Si₃O₁₂:Ce³⁺ in the remote phosphor layout. Furthermore, when the Ca₃Sc₂Si₃O₁₂:Ce³⁺ concentration is increased, CQS does not change considerably when the Ca₃Sc₂Si₃O₁₂:Ce³⁺ concentration is lower than 10% wt. CRI and CQS are dramatically reduced when Ca₃Sc₂Si₃O₁₂:Ce³⁺ concentrations are more than 10% wt. owing to severe color loss when there is more green color proportion. As a result, when using green phosphor $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$, proper concentration choice is important.

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Figure 5. The color rendering index of WLEDs as a function of $Ca_3Sc_2Si_3O_{12}$: Ce^{3+} concentration

Figure 6. The color quality scale of WLEDs as a function of $Ca_3Sc_2Si_3O_{12}$:Ce³⁺ concentration

4. CONCLUSION

All of the foregoing leads to the conclusion that, a gel-combustion technique was used to efficiently manufacture the single stage of the $Ca_3Sc_2Si_3O_{12}:Ce^{3+}$ phosphor for solid-status illuminating implementations. A solid-status technique for manufacturing commercial phosphors was also provided for compare and contrast. The gel-combustion technique produces particles that are around 1m in size, which is significantly littler than the solid-status reaction. The distinctive 5d–4f transfer of Ce^{3+} is responsible for the

light green radiation found in all specimens. The study also demonstrates that the luminous strength of the gel-combustion phosphor annealed in decreasing ambience is better than the solid-status reacting phosphor. We believe that the phosphors emit green lighting $Ca_3Sc_2Si_3O_{12}$:Ce³⁺ are potential for white LEDs based on their superior size and the gel-combustion produced phosphors' luminous characteristics. In comparison to the other technique, the gel-combustion process offers numerous notable benefits, including significantly reduced preparation process, lower annealing temperature, improved resource savings, and the resultant tiny-sized particles and good chemical uniformity.

ACKNOWLEDGEMENTS

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

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