Synthesis and photoluminescence properties in white light-emitting diodes of oxynitride green phosphor $Ba_3Si_6O_{12}N_2$

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ABSTRACT

Utilizing boron-coated Eu_2O_3, highly effective $Ba_3Si_6O_{12}N_2$ green-emitting phosphors were made using a gas reduction nitridation technique under flowing NH3 gas. We found that the synthesized phosphor is a pure phase of $Ba_3Si_6O_{12}N_2$ based on X-ray diffraction patterns. By modifying an alumina boat crystallized, the $Ba_3Si_6O_{12}N_2$ green-emitting phosphors from the result were considerably better and had higher emission intensity. Under stimulation at 405 nm, the radiation spectra revealed a typical wide green radiation band attributable to the $4f^65d \rightarrow 4f^7$ electronic transfer of Eu^{2+} ions. Generally, the green-emitting $Ba_3Si_6O_{12}N_2$ phosphors that were created by that method with boron-coated Eu_2O_3 is a potential phosphor-transformed diodes which emit white illumination white light emitting diodes (pc-WLEDs') element.

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1. INTRODUCTION

Owing to low energy consumption, non-mercury ingredient, long lifespan, and high reliability, phosphor-transformed diodes which emit white illumination white light emitting diodes (pc-WLEDs) are hoped to be potential phosphors for future lighting such as backlightings for light emitting diodes (LED) TV, decorated lamps, and automotive lighting implementations [1]–[4]. Combining a yellow $Y_3Al_{12}O_{15}:Ce^{3+}$ (YAG:Ce^{3+}) phosphor and an InGaN-based blue chip is the most common way for creating white LEDs [5]. Nevertheless, due to the shortage of red radiation, this method of producing WLEDs has a reduced hue rendering index (CRI) and poor heat stability [6]. Therefore, these LEDs are unable to be used for indoor lighting. Another way for tackling this challenge is to combine three distinct techniques. We have YP, RP, GP and BP will be the abbreviations for yellow, red, green, and blue phosphors. The first technique is combining a close-UV chip with RP, GP and BP. The second one is mixing a blue chip with RP and GP. And the last one is combining a blue chip with YP and RP utilizing an oxynitride phosphor [7], [8]. Many oxynitride phosphors have found their way into a variety of study groups owing to their ability to compensate for the shortcomings of prior oxide phosphors while also allowing color adjustments of the phosphor in terms of excitation and emitting wavelengths by adjusting the host lattice and operator. Oxynitride phosphors outperform oxide and sulfide phosphors in terms of heat and chemical stability [9], [10]. As a consequence, oxynitride phosphors have emerged as viable LED phosphors. Still, for the stated oxynitride phosphors that employ high purity nitride as rough ingredient, such as Ca-α-SiAlON: Eu^{2+}, β-SiAlON:Eu^{2+}, $M_2Si_5N_8$: Eu^{2+}

(M=Ca, Sr, Ba), $MSi_2O_2N_2$: Eu^{2+} (M=Ca, Sr, Ba), and $CaAlSiN_3$: Eu^{2+}, a high temperature (over 1500 ºC) and a lengthy soaking time are necessary to synthesis excellent oxynitride phosphors using the solid-status reaction approach [11]–[13]. For the manufacture of oxynitride phosphors, the gas reduction nitridation (GRN) process could be a promising alternative. The GRN approach employing NH_3 gas is advantageous because it provides for better control of the O/N ratio in the synthesis of oxynitride phosphor while also lowering the temperature required for the production of the desired phase [14]. By altering the amount of NH_3 in the synthesis conditions, we can create an oxynitride phosphor that can adjust from blue to red emission.

To aggregate a $Ba_3Si_6O_{12}N_2$ phosphor emits green lighting with strong luminous capabilities; we applied the notion of boron-coated Eu_2O_3 like the operator and used boat adjustment coupled with the GRN approach. To investigate further the crystal layout, luminous characteristics, particle dimension of the $Ba_3Si_6O_{12}N_2$ phosphor (EDX), we will need certain essentials. Those factors are powder X-ray diffraction (XRD), luminescence spectrometry (PL), and energy-diffusive X-ray.

2. EXPERIMENTAL DETAILS

The technique for making boron coated Eu_2O_3 sample is shown in Figure 1. A flask was filled with ten grams of Eu_2O_3, 3.65 g of H_3BO_3, and de-ionized water. The two powders were magnetically swirled in de-ionized water for 3 hours at 60 °C to thoroughly mix them, and then the mixed sample was dried for 24 hours. The well-mixed sample was pulverized in the agate mortar for 30 minutes before being fired at 600 °C for 7 hours. We used a solid-state process to make a boron coated Eu^{2+} phosphor ($Ba_3Si_6O_{12}N_2$) and a non-coated phosphor. The boron coated Eu^{2+} phosphor ($Ba_3Si_6O_{12}N_2$) and the non-coated phosphor ($Ba_3Si_6O_{12}N_2$) shall be referred to as M_1 and M_2, in turn, for the purposes of this paper. $BaCO_3$, SiO_2, Si_3N_4, and boron coated Eu_2O_3 were used as raw materials. These ultra-pure raw materials were combined in an agate mortar for 30 minutes before being fired at 1200 °C for 3 hours under NH_3 gas flow. The samples were re-fired for 7 hours at 1200 °C in a decreasing nitrogen environment containing 5% H_2 gas [15], [16].

3. RESULTS AND ANALYSIS

Various dopant ions are available to make greater converting phosphors, due to the type of emitting spectrum required. The host lattice must also meet certain requirements. To endure the extreme temperatures close the pumping LED chip and produce the extended lifespan that defines LEDs, the host substance must be a heatting and robust composition of chemistry. Also, the substance must be optically transpicous to the generated illumination. Only if a host-dopant transform of energy happens, the host must also be transparent for the pumping LED radiation, restricting substance alternatives to broad band-distance substances. The host substance need to absorb the dopants, which is made simpler if there is no charge or dimension discrepancy among the dopant ion and the replacement host ion. Lastly, the manufacturing of phosphor should be low-cost and ecologically benign in terms of heat input as well as the gases and precursors used [17], [18].

Using a simple combinatorial method to determine the 'best' host-dopant combination would be time consuming, especially as the influence of host substance composition on luminous characteristics is fairly significant for some dopant ions (e.g., wide range generating rare earth ions and Mn^{2+}). Dorenbos, e.g., compiled a list of the luminous characteristics (band width, absorptivity, and emitting energy) of over 300 Eu^{2+}-doped substances, proving effectively that radiations can be adjusted from close UV to deep red. As a result, low dopant concentration, data on previously published Eu^{2+}-doped (oxy) nitrides were also included [19], [20].

However, in several substances, the radiation is missing or shows unusual behavior, such as a protracted decomposition period, widen and red-shifted radiation (anomalous radiation), revealing the host matrix's significant influence once more. Even so, it is able to know the radiation characteristics of certain host-dopant mixtures, like heat extinguishing and the position of the rare earth 4f and 5d levels regardng to the host's band space, by simulating the host's band space impact and the nearby surrounding for rare earth dopants (symmetry, length, and kinds of ions inside the initial coordination shell). Numerous review papers have lately been written on certain host substances types for LED phosphors (like (oxy) nitrides, sulfides), identifying and explaining the numerous host substances and their production approaches. As a consequence, we just discuss the major 'classes' of host substances in general terms below, concentrating on their differentiating characteristics [21], [22].

The requirement for steady and effective RP portion of the emitting bands of color, which oxides and sulfides cannot simply deliver, sparked studies in different of hosts with huge crystal fields and/or substantial centroid change. As expected, according to the six parameters mentioned in this article, can be easily seen that the wide-range generating rare earth ions Eu^{2+} and Ce^{3+} have almost perfect characteristics; the difficulty left is to look for appropriate hosts with a substantial sufficient red-shift of the radiation. This feature was identified in (oxy) nitride phosphors, which prompted much study to this unique group of substances through the previous few years. The luminescent characteristics of them were previously seldom examined, despite the fact that these subtances were investigated for the great heating and chemistry steadiness, power, and hardness, which led to implementations like abrasives and protective coverings [23]. This provided a solid foundation for understanding the crystallographic layouts of numerous (oxy) nitrides, while the system M-S-Al-O-N (with M=Li, Ca, Sr, Ba, La) still need more investigation.

$M_2Si_5N_8$ (M=Ca, Sr, or Ba) and $MAlSiN_3$ (M=Ca, Sr, or Ba) are two nitride compositions doped with Eu^{2+} that exhibit distinct luminescence reaches the maximum point about 600 nm or at further wavelengths. The radiation of Eu^{2+} in oxynitride compositions like $MSi_2O_2N_2$ is frequently found in the green-to-yellow area of the bands of color which can be seen. Mueller-Mach showed a white LED with a constant correlated color temperature (CCT) of 3,200 K and a Ra of 89 in 2005, relying on a blue InGaN=GaN LED and two Eu^{2+}-doped (oxy) nitrides. At fairly high heat extinguishing temperatures, high quantum performances may be achieved in the abovementioned hosts (in mixture with higher doping concentrations for high absorptivity) [24].

In general, one of the key advantages of (oxy) nitride phosphors over other phosphors, such as sulfides, is their chemical and thermal stability. Stability investigations on oxidation or the photo-heating affect of large stimulation fluxes by the pumping LED, on the other hand, are scarcely recorded. Nonetheless, there are some signs that some (oxy) nitride compositions may have stability concerns. For example, $(Ca,Sr)AlSiN_3$ phosphors appeared to be more steady opposed to oxidation than $M_2Si_5N_8$ phosphors, with the latter exhibiting a significant loss in PL strength if burned to 573 K in air, with the steadiness dependent on stoichiometry.

Figure 1 shows the reversal shift in the concentrations of GP $Ba_3Si_6O_{12}N_2$ and YP YAG:Ce^{3+}. The adjustment has two meanings. The initial one is to keep average CCTs the same, and the other is to affect the absorption and diffusing of WLEDs with two phosphor films. This phenomenon, in turn, has an impact on the color standard and illuminating beam performance of WLEDs. The hue standard of WLEDs is thus dependent on the $Ba_3Si_6O_{12}N_2$ concentration chosen. When the $Ba_3Si_6O_{12}N_2$ ratio increased from 2% to 20% Wt., the YAG:Ce^{3+} concentration decreased to keep the average CCTs. This case also occurs for WLEDs with hue heats ranging 5,600-8,500 K.

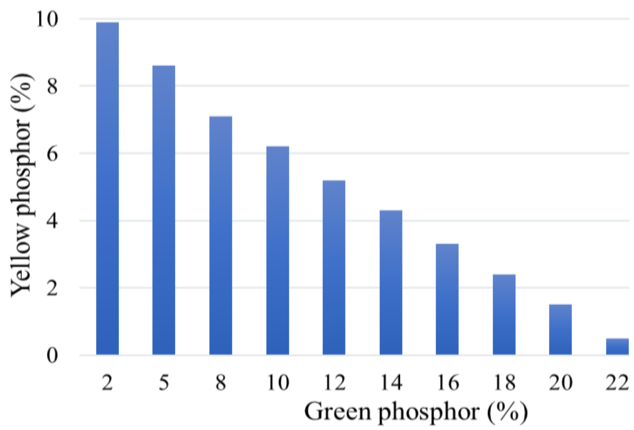

Figure 1. Changing the concentration of phosphor to preserve the average CCT

Figure 2 depicts the influence of the $Ba_3Si_6O_{12}N_2$ green phosphorus concentration on the transmitting spectrum of WLEDs. It is feasible to decide according to the producer's demands. WLEDs demand good colour fidelity can diminish luminous flux by a tiny amount. As shown in Figure 2, white light is the spectral region's synthesis. These five figures show 7,000 K spectra. Obviously, the strength trend grows with concentration $Ba_3Si_6O_{12}N_2$ in two sections of the light spectrum: 420 nm-480 nm and 500 nm-640 nm. The rise in the final luminous flux is obviously illustrated in the two-band emission spectrum. When the blue-light diffusing in WLEDs increases, it will drive the scattering in the phosphoror film and WLEDs either, favouring colour uniformity. When using $Ba_3Si_6O_{12}N_2$, this is a significant outcome. The colour consistency of the elevated heat distant phosphor layout, particularly, is challenging to control. This work found that $Ba_3Si_6O_{12}N_2$, at both poor and elevated colour temperatures (5,600 and 8,500 K), can enhance the colour standard of WLEDs.

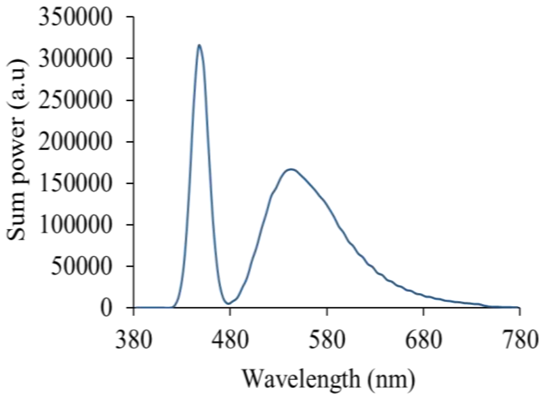

Figure 2. The emitting spectra of 7000 K WLEDs as a function of $Ba_3Si_6O_{12}N_2$ concentration

In the paper, we further demonstrate the efficiency of the emitted light flux of this double-film distant phosphor. The results in Figure 3 show when the concentration of $Ba_3Si_6O_{12}N_2$ rises from 2% wt. to 20% wt., the illuminating flux emitted increases dramatically. In all three average CCTs, the colour divergence reduces significantly with the phosphor $Ba_3Si_6O_{12}N_2$ concentration, as shown in Figure 4. The paternity of this circumstance is the absorptivity of the red phosphor film. When the $Ba_3Si_6O_{12}N_2$ absorb the blue illumination from the LED chip, it turns into green light. The addition of blue light also contributes to the yellow light absorption by the $Ba_3Si_6O_{12}N_2$ particles from the LED chip. The blue lighting absorption from the LED chip, though, is the strongest in two absorbs due to the material's absorption qualities. As a consequence of the addition of $Ba_3Si_6O_{12}N_2$, the green illumination element in WLEDs rises, improving the colour homogeneity index. Colour homogeneity is one of the most important factors among current WLED light parameters. The higher the colour uniformity index, the more costly WLED is. However, the low cost of $Ba_3Si_6O_{12}N_2$ is an advantage. $Ba_3Si_6O_{12}N_2$ can thus be used in a variety of applications.

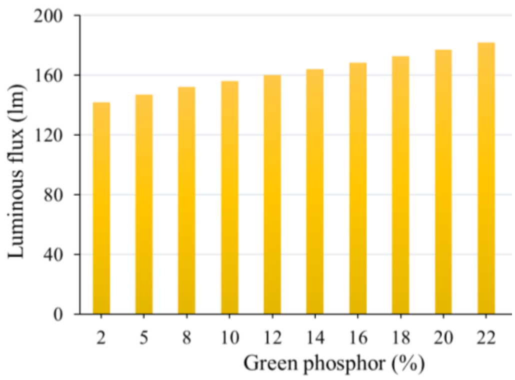

Figure 3. The illuminating flux of WLEDs as a function of $Ba_3Si_6O_{12}N_2$ concentration

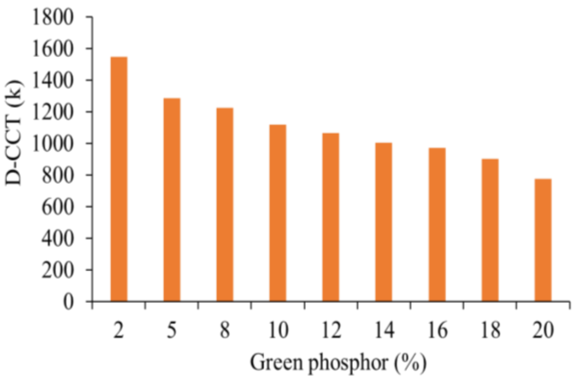

Figure 4. The color deviation of WLEDs as a function of $Ba_3Si_6O_{12}N_2$ concentration

Color uniformity is only one criterion to consider when assessing WLED color quality. With a high color homogeneity indicator, the color standard is not good enough. Accordingly, recent studies have developed a hue rendering index and a hue standard scale. When light shines above the hue rendering index, it determines the genuinely color of an object. The excessive abundance of green illuination between the three colors: blue, yellow, and green, causes the color imbalance. This imbalance has an effect on the color quality of WLEDs, resulting in a decrease in color fidelity. The results in Figure 5 show a minor decline in CRI in the presence of the remote phosphor $Ba_3Si_6O_{12}N_2$ layer. Though, because CRI is simply a flaw in CQS, these are allowed. When comparing CRI with CQS, the CQS is more essential and harder to attain. CQS is a three-element index, with the first being the color rendering indicator, the second being the viewer's preference, and the third being the hue coordinate [25]. For these three key factors, CQS is nearly a genuine overall assessment of hue standard. Figure 6 shows the increase of CQS in the existence of the distant phosphor $Ba_3Si_6O_{12}N_2$ configuration. When the $Ba_3Si_6O_{12}N_2$ concentration advances, CQS does not change considerably when the $Ba_3Si_6O_{12}N_2$ concentration is under 10% wt. Both CRI and CQS are dramatically diminished when $Ba_3Si_6O_{12}N_2$ concentrations are larger than 10% wt. owing to severe hue losing when green is dominant. As a result, employing green phosphor $Ba_3Si_6O_{12}N_2$ needs proper concentration choice.

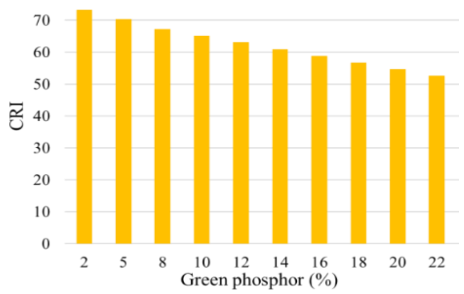

Figure 5. The hue rendering indicator of WLEDs as a function of $Ba_3Si_6O_{12}N_2$ concentration

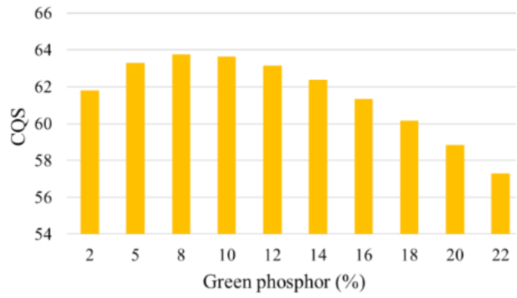

Figure 6. The hue standard scale of WLEDs as a function of $Ba_3Si_6O_{12}N_2$ concentration

4. CONCLUSION

When the criteria of excellent excitability in the close-UV to blue region of the bands of color, a quick decomposition period, and the ability to adjust the emit spectra are taken into account, the rare earth ions Eu^{2+} and Ce^{3+} obviously outperform the competition. The next stage is to choose the appropriate host, which will dictate not only the ions' stimulation and emitting spectra, but also their heat extinguishing behavior and (chemical and photo-heating) steadiness. The hosts of selection for phosphors generating in the red area of the bands of color are unquestionably oxynitrides and nitrides. This is completely a novel type of bright substances, and study is currently underway. Under flowing NH_3 gas, a series of $Ba_3Si_6O_{12}N_2$ green-emitting oxynitride phosphors were generated utilizing boron-coated Eu_2O_3 as an operator on the boron nitride plate. At a concentration of 0.3 mol of boron coated Eu_2O_3, attain the greatest emission intensity. When activated by illumination in the UV to the blue portion of the bands of color, the synthesized phosphor emits a usually broad emission band. The radiation strength of the manufactured phosphor treated with acid washing was higher than that of the commercial specimen of P46-Y3 and that of the phosphor before acid washing. These findings point to the new concept's possible applicability in the production of white LEDs, as well as an alternate way for the synthesis of other phosphors.

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