A new Ca₁₅(PO₄)₂(SiO₄)₆:Eu²⁺ phosphor with green emission for use in n-UV based WLEDs

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ABSTRACT

We have produced a novel phosphor using Eu²⁺, which is green in color coupled to Ca15(PO4)2(SiO4)6 (abbreviated as Ca15P2Si6) by deriving it structurally from α -Ca₂SiO₄ with a typical solid reaction method. Additionally, the decrease of Eu²⁺ concentration as well as heat endurance is being studied. 330 nm is the wavelength at which the substance is stimulated. At 491 nm, it attains the greatest radiation energy when filled with the ideal concentration of Eu^{2+} , which is 0.5 mol%. The primary mechanism of concentration reduction is discovered to be the fast transfer of stimulation through exchanging coupling among stimulated Eu²⁺ ions. The heat activating power boundary has been determined to be 0.244 eV. Additionally, we assess the productivity of this matter by fabricating a lightemitting diodes (LED) with converted phosphor (pc-LED) and integrating a LED with an InGaN-foundation near-ultraviolet and running it at currents between 50 mA and 300 mA. All of the efforts result in an optimal for pc-LED to be able to precede white light-emitting diodes (WLED) implementations.

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1. INTRODUCTION

Since traditional forms of lighting have several errors, such as low durability or poisonous, white light-emitting diodes (WLEDs) have been considered to be a potential alternative [1]. Another reason added to their popularity is their diverse applications. Backdrop illuminations or signaling support are some widely used functions of WLED [2]-[4]. WLEDs are conventionally created using a combination of YAG:Ce³⁺, which has a yellow color, and blue LED [5]. This WLED, though, produces strong chilly white illumination; it may not produce white illumination with warmth due to inadequate red radiation. Besides, owing to two combining, there are a few issues including shift in hue with input stream and poor color rendering indices (CRI). Unfortunately, mixing substances also create hue drawbacks and poor CRI. We can alter it by mixing 3 color together with n-UV LEDs to replace the original strategy [6], because n-UV-phosphor-transformed LEDs are anticipated to have several reasonably practicable implementations owing to the outstanding hue rendering index of them, strong hue tolerance, and excellent transformation effectiveness into visible illumination [7]. To attain the required luminous indices and characteristics, CaMgSi₂O₆:Eu²⁺ and M_2 SiO4:Eu²⁺ can be brought into consideration since their components contain rare components that are

beneficial to WLEDs [8], [9]. In addition, due to the ability to boost photoluminescence (PL), along with a short dissolve period, Eu^{2+} is one of the most popular activators. The radiation of this matter depends on the main grit. It also occurs randomly between the ultraviolet and red regions [10]. Unlike-Ca₂SiO₄, the WLEDs' green-emitting material, both the α and α' are unstable at room temperature when there is a lack of Ca₃(PO₄)₂ [11], [12]. Through observation, it was discovered that with a structure developed from the original α' -Ca₀SiO₄, Ca₁₅(PO₄)₂(SiO₄)₆ was consistent at room temperature. Another substance also capable of being stable is $6Ca_2SiO_4 \cdot 1Ca_3(PO_4)_2$. However, there have not been any results related to the fact that this substance is ideal for WLEDs' phosphor. To identify novel phosphor substances for n-UV based WLEDs, Ca₁₅(PO₄)₂(SiO₄)₆:Eu²⁺ (Ca₁₅P₂Si₆:Eu), along with its aspects like production and illuminating properties, etc are analyzed. This study also includes the examination of phosphor-transformed LEDs, created from n-UV LED chips and the phosphor mentioned above.

2. EXPERIMENTAL DETAILS

2.1. Preparation of green-emitting Ca15(PO4)2(SiO4)6:Eu²⁺phosphor

The main skill applied is called the solid-status reaction. $Ca_{15(1-x)}(PO_4)_2(SiO_4)_6:15xEu^{2+}$ (0.0025 $\leq x \leq 0.015$) powder specimens were equipped. Ingredients include Eu₂O₃, CaCO₃ along with (NH-4)₂(SiO₂)₆. Measuresents of the materials were previously, in a ball mill with the accompanies of ZrO₂ orbs ethanol. Then the mixture was left till dry on heated plates and out in open air [13]-[14]. Then the combination was grated and preheated a crucible made specifically of aluminum. The temperature used is 600 °C and the whole step took 8 hours. After regrounding, we repeated the heating process but at 1300 °C and in a contracting atmoshphere at 500 ml min⁻¹ flow rate (5% H₂/balance N₂) [15].

2.2. LED chip fabrication

To create LEDs, we utilize green phosphors with near-ultraviolet light-emitting diodes (n-UV LEDs). This n-UV LED essentially requires a InGaN base. Additionally, we mix together $Ca_{14.925}(PO_4)_2(SiO_4)_6:0.075Eu^{2+}$ in about 0.015 g, and $Ba_2SiO_4:Eu^{2+}$ in 0.0025g 1 g of hardener or resin. Above the n-UV LED, 0.7 g of the mixtures was placed. Following defoaming and curing, the phosphor-transformed LEDs were finished. Furthermore, UV LEDs were created using the similar method, but the phosphor was different. The real physical model of the WLED with n-UV LED is shown in Figure 1.



Figure 1. Photograph of WLEDs

According to Dexter's theory of energy transfer mechanism between ions in a phosphor matrix, the energy transfer between Eu^{2+} can be ascribed to the multipole-multipole interaction. Accordingly, such an interaction could be defined using the emission-intensity differences connected with the emitting level with multipolar interaction. Dexter developed a formula [16], [17] to investigate the kind of nonradiative power transmission, which Ozawa and Jaffe adjusted to a more practical version [18], which is expressed as (1):

$$\frac{l}{r} = \left[1 + \beta'(x)^{\theta/3}\right]^{-1} \tag{1}$$

I represents the radiation strength, *x* indicates concentration of activator ions, and β' represents the identical host crystal's constant under the similar stimulation environment. Evaluating the constant θ from this formula allows us to establish the sort of non-radiative power transmission. The data obtained are adjusted using the Arrhenius equation [18] to explore the thermal power obstacle [19]-[21].

$$I(T) \approx \frac{I_0}{1+c \exp\left(-\frac{E_a}{kT}\right)}$$

3. **RESULTS AND ANALYSIS**

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The inverse change in concentration of green phosphorus $Ca_{15}P_2Si_6$:Eu and phosphorus YAG:Ce³⁺ is denoted in Figure 2. This shift has two leads to two outcomes: The first is to maintain correlated chromaticity temperatures (CCTs), and the second is to affect the absorptivity and diffusing in WLEDs with two phosphor films. This eventually influences the hue standard and iluuminating beam productivity of WLEDs. Consequently, the phosphor's concentration chosen determines the hue standard of WLEDs. When the concentration increased to 20% wt, the YAG:Ce³⁺ concentration decreased to conserve the mean CCTs. This is also true for WLEDs with hue temperatures ranging 5600-8500 K.

-10% $\pm 15\%$ 8 (%) 6 4 2 0 5 10 15 20 GP particle size (µm)

-5%

Figure 2. Making change of the concentration of phosphor to keep the mean CCT

Figure 3 depicts the effect of Ca₁₅P₂Si₆:Eu green phosphorus concentration to the WLEDs transmission spectrum. It is able to achieve a selection relying on the requirements of the producers. WLEDs demand significant hue standard can decrease illuminating beam by a tiny portion. As shown in Figure 3, white light is made of different spectral regions. The spectrum of 5000 K is depicted in this Figure. It can be seen easily, the strength trend rises with the phosphor's concentration in two locations of the lighting spectrum: 420-480 nm and 500-640 nm. This shift of the two-range radiation spectrum indicates a rise in the illuminating beam productivity. Furthermore, blue-light diffusion in WLED is raised, implying that diffusion in the phosphorous film and in WLEDs has risen, preferring hue homogeneity. This is a significant outcome when green phosphor is used. Controlling the hue homogeneity of the extreme temperatures distant phosphor configuration, in specific, is challenging. These results proved that Ca₁₅P₂Si₆:Eu, at both poor (5600 K) and strong color temperatures (8500 K), can enhance the hue standard of WLEDs [22], [23].

300000 Sum power (a.u) 240000 180000 120000 60000 0 380 480 580 680 780 Wavelength (nm)

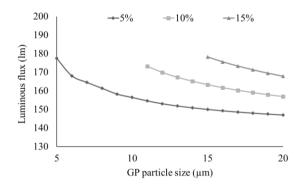
Figure 3. The radiation spectra of 5000 K WLEDs as a function of Ca₁₅P₂Si₆:Eu concentration

As a result, the paper proved the efficacy of this double-film far remote phosphor configuration's discharged illumination beam. The results in Figure 4 show that the illumination beam emitted increases dramatically as the concentration grows 2% wt-20% wt. The hue deviation was considerably lowered with

360000

the concentration in all three mean CCTs, as can be seen in the outcomes of Figure 5. This can be analyzed using the red phosphor film's absorptivity. When this phosphor imbibes the illumination of blue from the LED chip, these blue particles change it to green illumination. The particles imbibe yellow illumination additionally to LED chip's blue color. Nevertheless, due to the absorbing characteristics of the substance, the blue illumination absorptivity from the LED chip is higher in comparison to these two absorbs. As a consequence of the additament of $Ca_{15}P_2Si_6$:Eu, the green illumination substance in WLEDs grows, resulting in an enhancement in the hue homogeneity index. Hue homogeneity is an important element between many contemporary WLED light parameters. Evidently, the greater the hue homogeneity indicator, the more costly WLED. The advantage of utilizing $Ca_{15}P_2Si_6$:Eu is its cost effective. This substance can therefore be broadly utilized.

Hue homogeneity is only one element to think of carefully when evaluating the hue standard of WLEDs. Hue standard may not be considered to be excellent with just a great hue uniformity indicator [24], [25]. As a result, latest researches include a hue rendering indicator and a hue standard ratio. When a beam glows on the hue rendering indicator, it determines the real hue of an item. The hue imbalance is largely made up of green illumination between the main hues: green, blue, yellow. There is an impact on the hue standard of WLEDs, resulting in a decrease in WLED hue fidelity. The results in Figure 6 show a small decrease in CRI when there is the distant $Ca_{15}P_2Si_6$:Eu film. Nevertheless, these are reasonable since CRI is just a weak point of colour quality scale (CQS). Once trying to compare CRI and CQS, the second is more essential, also harder to obtain. CQS is a three-element value that is defined by three elements: the hue rendering indicator of hue standard. Figure 7 depicts the optimization of CQS when there is the $Ca_{15}P_2Si_6$:Eu film. Besides that, when its concentration is continued to increase, CQS does not shift dramatically with $Ca_{15}P_2Si_6$:Eu concentrations below 10% wt. When the $Ca_{15}P_2Si_6$:Eu concentration is more than 10% wt., both CRI-CQS are lessened dramatically owing to significant hue losing once green is prominent. Correspondingly, once utilizing green phosphor, proper concentration choosing is critical.



 $\begin{array}{c} 480 \\ \hline 9 \\ \hline 9 \\ \hline 0 \\ \hline 0 \\ \hline 120 \\ 0 \\ \hline 0 \\ \hline 5 \\ \hline 10 \\ \hline 15 \\ \hline 20 \\ \hline 25 \\ \hline 30 \\ \hline 0 \\$

Figure 4. The illuminating beam of WLEDs as a function of $Ca_{15}P_2Si_6$:Eu concentration

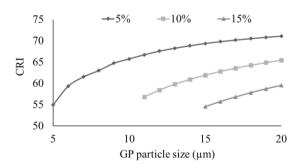


Figure 6. The hue rendering indicator of WLEDs as a function of $Ca_{15}P_2Si_6$:Eu concentration

Figure 5. The hue deviation of WLEDs as a function of $Ca_{15}P_2Si_6$:Eu concentration

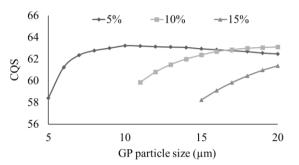


Figure 7. The hue standard ratio of WLEDs as a function of $Ca_{15}P_2Si_6$:Eu concentration

4. CONCLUSION

The article depicts the effect of $Ca_{15}P_2Si_6$:Eu on the optic properties of a dual-film phosphorus configuration. The results proved that the phosphor is a reasonable selection for improving hue homogeneity utilizing Monte Carlo computational simulations. This applies not just to WLEDs with a minimum hue temperature of 5000 K, but to ones with a maximum hue heat of 8500 K. The result of this research has indeed achieved the goal of improving hue standard and illuminating beam, which is very complex due to the distant structure of phosphorus. However, there is one small drawback for CRI and CQS. When the concentration is raised overly, the CRI and CQS drop rapidly. As a result, relying on the producer's goals, the appropriate concentration must be chosen. The article has provided a wealth of useful data for reference in generating better hue homogeneity and lighting beam WLEDs.

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