The impacts of green Cs₂ZnSi₅O₁₂:Eu²⁺ phosphor for white light emitting diode

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ABSTRACT

We created the green-light phosphor Cs₂ZnSi₅O₁₂ (CZSO) with Eu²⁺ replacement for the practical use of ultraviolet white illumination through the solid-state technique. The phosphor displays many different forms and crystallizes in the space group Pbca along with $Pa\overline{3}$, with formation resembling CsAlSi₂O₆, the leucite in cube form. We utilized the X-ray powder diffraction as well as the spectroscopic knowledge to validate that the crystal formation in the phosphor CZSO:Eu²⁺ belongs to the space group $Pa\overline{3}$. CZSO can generate wide green emission with a wavelength of 504 nm when excited by ultraviolet. We enveloped the blend using the translucent silicone resin and the red-light phosphor $Sr_2Si_5N_8$:Eu²⁺ as well as excited it using an ultraviolet light emitting diode (LED) device with a wavelength of 370 nm to generate white illumination that yields remarkable chromatic output. The generation of illumination utilizing the phosphor CZSO benefits from the wide green emission, which does not require the integration of the three phosphors that are critical to the ultraviolet pumping. Such benefit will allow the model of the apparatus to become simpler and simultaneously create a desirable ultraviolet white illumination.

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1. INTRODUCTION

Illumination based on the solid-state method offers many benefits such as its minimal effects on environment, considerable duration of usage as well as great performance; which makes it a suitable substitution for the standard optical sources [1], [2]. Due to the world's demand for less power usage, it is recommended that we begin utilizing light emitting diode (LEDs) for optical display [3]. The LEDs can generate white illumination thanks to the merger between a phosphor and a chip of LED, which involves the down-transmutation of the emission in the LED device into greater wavelengths through the phosphor. The generation of white illumination is usually done through daubing the blue InGaN LED device with a phosphor that emits yellow light [4]. While such technique requires a low cost, it may yield cold blue-white illumination that possesses high CCT (short for correlated color temperature) as well as low CRI (short for color rendering index). In order for us to adopt the newer method of generating illumination, it is necessary to achieve cheaper white illuminations that yield smaller CCT as well as greater CRI output. To achieve such an outcome, it is possible to introduce a red phosphor like $Sr_2Si_5N_8:Eu^{2+}$, $SrAlSiN_3:Eu^{2+}$, or $K_2SiF_6:Mn^{4+}$ [5]. While such method has become a standard and can effectively generate white illuminations, there would be lack of essential photons of the 400-450 nm zone for the resulting illuminations. Another technique involves the merger between the phosphors in blue, red, green colors and the ultraviolet LED. The white illumination resulting from such technique will benefit from a variety of chromas as well as the feature of being adjustable, which is decided by the proportion between the phosphors, along with enveloping the nearultraviolet zone. However, the technique requires the phosphors to have great performance because of the Stokes loss of the emission intensity, created through the transmutation [6]. As such, the development of solid-state illumination requires the advancement of the LED chips and the suitable inorganic phosphors. Such phosphors comprise a host lattice possessing a rare-earth ion (for example, Ce^{3+} or Eu^{2+}) to act as a replacement for the cation. Adding such ion will result in the cleavage of the crystal zone. There, the 5dorbitals in the activation ion undergo a descending transfer, which creates the $4f \leftrightarrow 5d$ electronic shifts for the electromagnetic spectral zone [7]. The phosphor's crystal properties would be essential for the assessment of the down-transmutation's performance, the excitation location, the emission wavelength, the emission band's wideness as well as the heat features. Our research utilizes the phosphor CZSO, which is created via the solid-state method. We replaced Cs of the phosphor using Eu²⁺, which creates an emission in the middle of the green zone within the spectrum when excited by ultraviolet. When mixed with a phosphor in red, CZSO could be the suitable choice to produce an ultraviolet pc-LED (LED based on conversion phosphor) thanks to a wide emission created by the Eu²⁺ spots, in the wavelength range of 400 nm to 700 nm.

2. EXPERIMENTAL

We examined the light attributes of CZSO through placing the multi-crystal materials on a quartz slide, posterior to combining the powders with silicone resin. We acquired the photoluminescent spectra under room temperature using the fluorescence spectrophotometer that possesses a 75-W xenon arc lamp to excite, along with the Janis cryostat to maintain the temperature range of 80 K to 500 K. We utilized diffuse reflectance, determined via the Agilent Technologies Cary 5000 possessing a diffuse reflectance, to identify the bandgap. We determined the inner photoluminescent quantum yield via the technique by de Jia *et al.* [8] as well as an orb daubed with Spectralon under 365-nm exciting wavelength [9]. We identified the duration of luminescence by utilizing the horiba deltaflex lifetime system accompanied by the NanoLED N-360 nm LED with a wavelength of 363 nm. To create the pc-LED, we utilized the phosphor Sr₂Si₅N₈:Eu²⁺, CZSO combined with silicone resin. In order to create the phosphor "cap", we cured the phosphor resin within the standard copper frame, then subjected it to excitation by utilizing a LED with wavelength of 370 nm under the current of 30 milliampere.

As shown in (1) and (2) determines $[CsO_{12}]$ polyhedral capacity, accompanied by the bond distortion index as well as the polyhedral quadratic extension [10], [11].

$$D = \frac{1}{n} \sum_{i=1}^{n} \frac{|l_i - l_{avg.}|}{l_{avg.}} \tag{1}$$

$$\langle \lambda \rangle = \frac{1}{n} \sum_{i=1}^{n} \left(\frac{l_i}{l_{avg.}} \right)^2 \tag{2}$$

D represents the bond length distortion. l_i represents the range separating the ith particle, the particles at the center and $l_{avg.}$ which represents the median bond lengthiness. The polyhedral quadratic extension, represented by $\langle \lambda \rangle$, would be determined in an identical way with l_0 representing the range separating the particle at the center and the peak of polyhedron with $\langle \lambda \rangle$ measured at 1 being the standard of an ideal polyhedron.

For the assessment of energy shift possibility, the critical distance for energy shift had to be identified using the (3). V represents unit cell's capacity. x_c represents the rare-earth concentration. n total amount of Wyckoff locations for the Eu²⁺ spots [12], [13].

$$R_c = 2 \left(\frac{3V}{4\pi x_c n}\right)^{1/3} \tag{3}$$

3. RESULTS AND ANALYSIS

White light, as opposed to red, green, and blue (RGB) light, can be created by combining an only one LED light source including one or many converting phosphors. Most of commercially available LED-based white illumination generators presently employ this technology. Until recently, these were almost entirely based on a blue LED and a phosphor based on YAG:Ce³⁺. There are two approaches that can be distinguished. One can utilize a blue LED and convert a portion of the generated light to longer wavelengths

by using a phosphor material, or one can use phosphors to completely convert the emission from a (near) ultraviolet LED.

What are the benefits of utilizing ultraviolet (UV) pumping LEDs over blue ones? To begin with, if the electrical to optical power conversion in UV LEDs is more efficient than in blue LEDs, switching to UV LEDs can result in a more efficient overall design. Second, it's debatable whether a blue LED and a single conversion phosphor can produce adequate color rendering while maintaining a low color temperature. If two phosphor materials must be utilized, including one with a slight Stokes shift to cover the emission spectrum around 500 nm, the whole phosphor technique with UV pumping LEDs may be considered. This also has the advantage of making the emission spectrum more steady in terms of driving current and LED chip temperature. In this situation, spectral shifts in the pumping LED are not reflected in spectral or intensity variations in the phosphor emission, provided the phosphor excitation spectrum is sufficiently 'flat' around the pumping LED emission. When a blue pumping LED is employed, changes in the LED's emission spectrum cause a color shift in the white LED.

To obtain a light source with high efficiency and reasonable emission color characteristics, a proper combination of high-performance phosphors is definitely necessary. The excitability of phosphors, or how closely their stimulation spectra fit the radiation of the pumping LEDs, is a second critical criteria for their usefulness. Actually, this is the major reason why fluorescent lamp phosphors, even when fine-tuned, are frequently useless to be used in LEDs. The emitting line of mercury at 254 nm predominantly excites these phosphors. For effective energy considerations, it is not desirable to make pumping LEDs with radiation in this wavelength range. As a consequence, great excitability phosphors in the close-UV to blue area of the spectrum are required. Additionally, the stimulation spectrum should be wide enough to offset for variations in the emitting spectra of the pumping LED caused by variations in driving current and/or junction temperature. It is ideal to have a slightly flat stimulation spectrum for the phosphor around the LED's maximum radiation to preserve hue steadiness over the entire wLED.

The development of the phosphors to be utilized with the LED devices to generate solid-state illumination has to involve the emission consistency for the temperature. The longer the pc-LEDs operates, the higher the temperature of the apparatuses. Such outcome deteriorates the photoluminescent the phosphors' quantum yield as well as their chromatic consistency. As such, we must examine the phosphors with high heat potency before utilizing them for the pc-LEDs. For the assessment of heat consistency, it is possible to examine T_{50} (the temperature where emission reaches fifty percent of the emission under room temperature) [14]. In Figure 1, we can see the emission intensity for the phosphor CSZO: Eu^{2+} with the respective temperature range of 80 K to 500 K. Starting at 80 K, as the temperature goes up to reach 260 K, the emission intensity slightly goes up, which is the possible result of trap states occurrence caused by the inconsistency concerning the size of rare-earth and the alternative Eu²⁺ spot as well as their aliovalent replacement. However, the intensity of emission continues to be consistent till 360 K. But when it exceeds 360 K, the intensity considerably goes down until it becomes about fully abated under the temperature of 500 K. T₅₀ happens under the temperature of 392 K, noticeably lower compared to 423 K, the working temperature for the majority of LED devices. The considerable heat abatement appears to be unexpected as the phosphor CsAlSi₂O₆:Eu²⁺ solidifies to form a cube shape similar to leucite and preserves seventy-five percent of the normal temperature emitting intensity under 425 K [15]. Moreover, the phosphor Cs_{1.97}Eu_{0.03}MgSi₅O₁₂ with similar formation displayed no noticeable heat abatement under the temperature of 423 K. The insufficient heat consistency of the phosphor CZSO:Eu²⁺ is probably caused by a d^{10} Zn²⁺ ion. The ion could be reason behind the thin bandgap that lets the photoionization, which advances the photons into the transmission band, manifest and abate the emission as a result. Figure 2 demonstrates that the green phosphor CZSO:Eu²⁺ concentration is inversely proportional to the yellow phosphor YAG:Ce³⁺ concentration, indicating two things: first, that the average CCT levels are maintained; second, that the absorbing and dispersing of the two phosphor films of WLEDs are affected. Consequently, the chromatic efficiency and luminous production of WLEDs may be compromised. As a result, the concentration of CZSO:Eu²⁺ used influences the chromatic efficiency of WLEDs [16]-[18]. As the concentration of YAG:Ce³⁺ increased from 2% to 20% Wt., the concentration of YAG:Ce³⁺ decreased to retain the average CCT values. This is also true for WLEDs with CCTs range from 5600 K to 8500 K.

Figure 1 shows how the concentration of the green phosphor CZSO:Eu²⁺ affects the transmitting spectrum of WLEDs. The selection may be determined by the company's demands. WLEDs with a high chromatic efficiency demand may have a minor drop in luminous production. As seen in Figure 1, the mixture of the spectral zones results in white illumination. These five data depict the spectrum at 8000 K CCT. The two regions of the optic band of colors with wavelengths spanning from 420 nm to 480 nm and 500 nm to 640 nm clearly suggest that their brightness increase in proportion to the concentration of CZSO:Eu²⁺. An increase in the two-band emitting spectra suggests an increase in brightness. Furthermore, the dispersion of blue light in WLED exhibits increased activity indicates greater activity of the dispersion in

the phosphor film and in WLED, which improves chromatic uniformity. Such a result might be critical for the usage of CZSO:Eu²⁺. Manipulating the chromatic uniformity in a distant phosphor package at high temperatures, particularly, is a difficult task. Our findings confirmed CZSO:Eu²⁺'s capacity to improve the chromatic efficiency of WLEDs at low and high color temperatures (5600 K and 8500 K) [19]-[21].





Figure 1. The emission spectra in 8000 K WLED devices as a function of CZSO:Eu²⁺ concentration

Figure 2. Maintaining the median CCT by altering the phosphor concentration

As a result, the luminous effectiveness in the two-sheet distant phosphor film has been proven in this study. Figure 3 shows that when the CZSO:Eu²⁺ concentration increases from 2% wt. to 20% wt., the luminosity created increases significantly. Figure 4 demonstrates that the hue divergence decreased significantly with increasing phosphor CZSO:Eu²⁺ concentration at three mean CCT levels. The absorptivity in the film of red-colored phosphor might shed light on such an incident. As the blue illumination emitted by the LED chip is absorbed by the granules of blue-color phosphor, it is converted into green illumination. Aside from the blue light, the granules of CZSO:Eu²⁺ absorb yellow illumination as well. Owing to the substance's absorption properties, the absorption of blue illumination created by the LED chip has a higher efficacy among the aforementioned absorptions. As a consequence, when CZSO:Eu²⁺ is added, the green factor of WLEDs is increased, which increases chromatic uniformity. Regarding today's WLED lamp characteristics, chromatic homogeneity is regarded as critical [22]-[24]. It is obvious that increasing the chromatic uniformity can boost the price of the WLED. However, the usage of CZSO:Eu²⁺ can be cost-effective, and as such, it may find broad utilization.



Figure 3. The lumen in the WLED devices with respective CZSO:Eu²⁺ concentrations

Figure 4. The chromatic aberration in the WLED devices with respective CZSO:Eu²⁺ concentrations

When it comes to assessing the chromatic efficiency of WLEDs, chromatic uniformity is the only factor to consider. A high level of chromatic homogeneity does not ensure good chromatic efficiency. As a result, previous research has proposed a parameter to evaluate hue production and chromatic quality. When a light is placed on the hue rendering index, the index reveals the true hue of the item. The absence of chromatic homogeneity is caused by the existence of green-light being overabundant among the three primary hues, which are blue, yellow, and green. This has an effect on the WLED's chromatic efficiency,

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which can reduce chromatic uniformity. Figure 5 shows that when a layer of distant phosphor CZSO:Eu2+ is applied, the CRI lowers somewhat. However, such disadvantages are minor because CRI is only a disadvantage of a CQS. When comparing CRI and CQS, the CQS is more difficult to get, thus the CQS should be preferred over CRI. The CQS parameter considers three factors: CRI, beholder preference, and color coordination. CQS might be regarded the efficient and generic parameter defining chromatic effectiveness with such key aspects [25]. Figure 6 shows the rise in CQS with the distant phosphor film CZSO:Eu²⁺. Moreover, when the phosphor CZSO:Eu²⁺ concentration grows, the CQS shows no discernible change when the concentration is less than 10% wt. When the concentration approaches 10% wt., CRI and CQS all suffer a significant drop due to the huge loss of color produced by the preponderance of green color. As a result, we have to select a suitable concentration of green phosphor CZSO:Eu²⁺.

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Figure 5. The CRI in the WLED devices with the respective CZSO:Eu²⁺ concentrations



Figure 6. The CQS in the WLED devices with respective CZSO:Eu²⁺ concentrations

4. CONCLUSION

We created the phosphor BaE using the solid-state technique and examined the phosphor's luminescence features. We also integrated the phosphor $Ba_{1.95}Si_3O_8:Eu_{0.05}$ with an n-UV LED to create and examine the pc-LED device. The pc-LED device yielded a low CRI output measured at 60 because the red spectrum was not adequate. We created the orange ZnCdSe/ZnSe QD that possessed the diameter measured at roughly 7,5 nm as well as a quantum yield measured at roughly 75%. The QD was incorporated into the pc-LED device possessing a distinctive sheet layout: the QD-PMMA blend followed by the phosphor-resin blend, for the generation of white illumination that yields greater CRI output. The acquired WLED device produced white illumination yielding greater CRI from 85 to 86, which is the result of QD emission as well as the consistent CIE color coordinates under the currents measured at 10 to 40 milliampere.

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