

Application of $(\text{Ca}_{1-x}\text{Sr}_x)\text{LaGa}_3\text{S}_6\text{O}:\text{Eu}^{2+}$ phosphor in white light-emitting diode fabrication

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ABSTRACT

An alternating sequence of $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ phosphors was produced using high-thermal and solid-state reactions. It has outstanding excitations at the range between 350 and 500 nm suitable for the close-ultraviolet or GaN-based blue light-emitting diode light emitting diode (LED) chips. These phosphor peaks have a blue shift between 560 and 540 nm when the Sr concentration (x) rises. The Sr content (x) can be modified to produce an illumination with a color between yellow and green. As a result, we can conclude that solid solutions $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ are excellent for use in manufacturing white LEDs. Our data may become valuable for producers in the task of making white light emitting diode (WLED) devices suitable for them.

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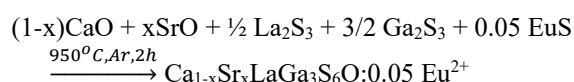
1. INTRODUCTION

White light emitting diodes (WLEDs) with GaN/InGaN based have lately gained more attention. In order to acquire phosphor-transformed white light emitting diodes (LEDs), there are numerous methods. An example is the use of a blue chip in conjunction with a phosphor that emits yellow lighting or the use of an n-UV LED chip with a primary-hue emission phosphor [1]-[3]. A number of researches have been performed to create novel systems of phosphor for white LEDs that can convert light. This makes $\text{Ce}^{3+}/\text{Eu}^{2+}$, two ions with an f-d structure, ideal for use as activators in phosphors due to their ability to create wideband observable light with crystal-field, and nephelauxetic effects. By changing the cations or anion concentrations of the host, it is possible to generate Ce^{3+} or Eu^{2+} ion-doped solidified solutions [4]-[6]. You can respectively replace Sr^{2+} , Mg^{2+} , and Ba^{2+} with Ca^{2+} , and F^- with O^{2-} , Al^{3+} for Ga^{3+} , and Se^{2-} can be swapped with S^{2-} . Compared to an oxide, oxysulfide's elemental sulfur has a lower negative value of electrons. On average, $\text{Ce}^{3+}/\text{Eu}^{2+}$ crystal dispersion should be greater in oxysulfide hosts, thus the 4f-5d absorption may expand to observable wavelengths (400-500 nm). In this case, it's the $\text{Ce}^{3+}/\text{Eu}^{2+}$ doped oxysulfides, such as $\text{Sr}_8\text{Al}_{12}\text{O}_{24}\text{S}_2:\text{Eu}^{2+}$ (yellow-orange), $\text{CaZnOS}:\text{Eu}^{2+}$ (red), and $\text{Ca}_{10}(\text{PO}_4)_6\text{Y}:\text{Ce}^{3+}$ (Y = S, Se) in blue. Therefore, solid-state illumination can easily be performed with these phosphors [7]-[9]. As a result of the substitution of sulfur for oxygen in the $(\text{Sr},\text{Ca})\text{LaGa}_3\text{O}_7$ compound in our group's initial report, $\text{SrLaGa}_3\text{S}_6\text{O}:\text{Ce}^{3+}$, $\text{SrLaGa}_3\text{S}_6\text{O}:\text{Eu}^{2+}$, as well as $\text{CaLaGa}_3\text{Ga}_3\text{S}_6\text{O}:\text{Ce}^{3+}$, Tb^{3+} phosphors came into light for the first time. Being blue and yellow-green phosphors, these chemicals can be used to produce white LEDs. It

was also found that compared to BaMgAl₁₀O₁₇:Eu²⁺ (BAM), CaLaGa₃S₆O:Ce³⁺ has the best absorption band (~400nm). The solid solution of Ca_{1-x}Sr_xLaGa₃S₆O:0.05Eu²⁺, on the other hand, have a limited amount of data on its optical characteristics. The said solution possessing changing Ca/Sr cation proportions will be examined for variations in optical characteristics using the Commission Internationale de l'Eclairage (CIE) chromaticity diagram from 1931. In 2006, the CIE formed a technical committee (TC1-69) to “explore novel methods for assessing the color rendition properties of white-light sources used for illumination, including solid-state light sources, with the purpose of adopting new assessment procedures.”

2. EXPERIMENTAL DETAILS

Both La₂O₃ and Eu₂O₃ were used at 99.99% and with CS₂-reducing pressure at 1250°C to create β-La₂S₃ and EuS. This procedure was 3 hours long. 950°C for 3 hours along with H₂S was the condition used to produce Ga₂S₃ from Ga₂O₃ (A.R.). The grating was used to completely mix the CaO, SrO, -La₂S₃, Ga₂S₃, and EuS combination, and the mixture was then sintered in furnaces with a horizontal tube at 950°C for 2 hours in Ar condition. A collection of Ca_{1-x}Sr_xLaGa₃S₆O:0.05Eu²⁺ specimens s a result of the following reaction [10], [11].



With a Rigaku D/max vpc X-ray diffractometer and Cu Kα radiation under 40kV as well as 30mA, the final structure was determined. As part of the research, we examined the PL as well as photoluminescence excitation (PLE) spectrum of this phosphor with the use of Fluorolog-3 and double excitation monochromators. Prior to scanning electron microscopy (SEM) inspection, we utilized gold-coated substances for the task of evaluating the particle size and morphology.

Ca_{1-x}Sr_xLaGa₃S₆O:0.05Eu²⁺ are predicted to have a peak wavelength shift and luminous features using an initial formula generated via a linear fitting in the graph [12]:

$$y = -20.57x + 561.61(0.0 \leq x \leq 1.0) \quad (1)$$

this corresponds to y being the maximum wavelength along with x being the independent fluctuating constituent indice. The below equation can be used to calculate the crystal field strength [13]:

$$D_q \propto \frac{1}{R^5} \quad (2)$$

the M-S(O) bond length among the core metal as well as the ligand ions, while the crystal field energy and the bond length are respectively represented as D_q and R. Sr²⁺ and Ca²⁺ have ionic radius of 1.26 Å and 1.12,24 Å, correspondingly.

3. RESULTS AND ANALYSIS

Sulfide hosts, treated via Eu²⁺, attracted attention back in the beginning times of LED devices based on conversion phosphor as the discharge would be generally red-shifted, unlike oxide, a result of a higher centroid transfer. The discharge lines for Eu²⁺ of SrS as well as CaS display broadband discharge with peaks around 620 as well as 660 nm, with an full width at half maximum (FWHM) reaching 70 nm, making them appropriate if used in the form of a red element for WLED devices. It is possible to adjust the discharge from orange-red to saturated red by utilizing solid solutions Ca_{1-x}Sr_xS:Eu²⁺. Despite its use for said devices, these substances have significant limitations, including relatively strong concentration quenching and weak stability when exposed to moisture. Heat abatement would also be relatively potent, notably in mixtures with high Sr concentrations as well as superior concentrations of Eu. Thermal quenching is already considerable at room temperature. Solid-state processes for sulfides frequently need poisonous H₂S; however solvothermal synthesis techniques under small temperature that generate sub-micron-sized independent crystals are also accessible. Some mixtures of ternary sulfide came into consideration in the form of LED phosphors. Thiogallates gained particular interest as they have substantially higher reliability which surpasses that of thioaluminates (in the case of SrGa₂S₄:Eu²⁺). By changing the ion of alkaline earth (Ca, Sr, Ba) as well as the doping ion (Ce, Eu), it is possible to adjust the emission of thiosilicate phosphors M₂SiS₄ from deep blue (Ba₂SiS₄:Ce) to saturated red (Ca₂SiS₄:Eu). The comparatively small heat abatement temperature, usually between 400 and 450 K, is a disadvantage.

Various sulfide compounds mentioned exhibit considerable reactivity with air chemicals (moisture, carbon dioxide), degrading the phosphor as well as reducing luminescence. Encasing sulfides that have inactive as well as translucent layers was considered capable of significantly improving reliability using certain approaches like non-water sol-gel method or atomic sheet deposition. In theory, similar procedures can be used for substances responsive to moisture. When considering the lengthy lifetime of an LED device, one would prefer to pick a stable substance like the majority of oxides or nitrides.

In the recent times, phosphors made of MZnOS ($M = \text{Ca, Ba}$) with decent heat as well as chemical reliability were found. CaZnOS:Eu^{2+} emits red light at 650 nm and has significantly better thermal quenching, surpassing SrS:Eu^{2+} . CaZnOS:Mn^{2+} emits at a short wavelength of 614 nm with FWHM reaching 50 nm) as well as exhibits prominent Mn^{2+} excitation bands between 350 and 500 nm, an unusual result for luminescence of Mn^{2+} .

$\text{Y}_2\text{O}_2\text{S:Eu}^{3+}$ is also a popular oxysulfide that has been used as a cathode ray phosphor as it offers excellent red discharge. However, the substance is not stimulatable between near-ultraviolet and blue since the charge shift lines seem to be located lower than 360 nm. In addition, the inner 4f-4f absorptions for Eu^{3+} around 395 or 465 nm appear to be feeble. As a result, using the substance in the form of a benchmark phosphor to evaluate the efficiency for different transmutation phosphors when activated within the near-ultraviolet or observable is very unfair.

The concentration of green $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ and yellow YAG:Ce^{3+} are in reverse, as shown in Figure 1. As a result of this modification, the mean CCT values will be maintained, however, the absorption, as well as scattering in the lights of two layers will be influenced. This affects the hue output as well as optical ray in WLEDs. WLEDs' color quality is determined by this phosphor's concentration. YAG:Ce^{3+} concentration decreased when $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ concentration increased from 2 to 20% Wt to maintain the mean CCTs. Similarly, WLEDs with a color temperature ranging between 5600 K and 8500 K provide the same effect.

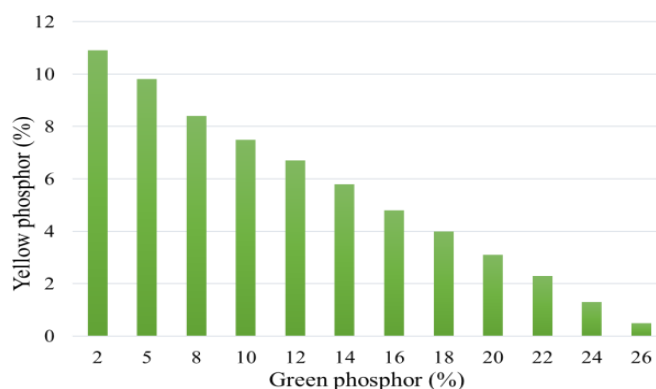


Figure 1. Altering phosphor concentration for the task of sustaining mean CCT

We can see from Figure 2, the concentration of $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ has a significant impact on the transmittance spectrum. Decisions are made based on the conditions of the manufacturer. Color-sensitive WLEDs can lower their luminous flux by a tiny amount if they demand good color fidelity. As seen in Figure 2, white illumination is a mixture of spectral areas. Each of these graphs shows a spectrum at 6000 K. Clearly, $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ ascends the intensity of the two light spectrums, 420-480 nm as well as 500-640 nm. As the spectrum with two bands widens, the luminous flux increases as a result. Additionally, WLEDs have a greater ability to disperse blue light. This means that WLEDs have better color consistency. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ produces this crucial finding. Especially challenging is to manage the color consistency of a distant phosphor structure at high temperatures. According to the results, poor and elevated color temperatures (5600 K and 8500 K) of $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ may improve the color quality [14], [15]. A double-layer's efficacy in emitting light has been demonstrated in the study. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ with concentrations between 2-20% wt. results in a substantial increase in the luminous flux output, as indicated in Figure 3. Figure 4 indicates the significant drop in the hue deviation when the $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ concentration was enhanced in all three mean CCTs. Red phosphor layer absorption may be to blame. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$ phosphor particles turn blue light into green light when they were absorbed. Red phosphor layer absorption may be to blame. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O:0.05Eu}^{2+}$

phosphor particles turn blue light into green light when they were absorbed. Although the phosphor's particles absorb the blue light, they also absorb yellow light. In comparison to these two absorbers, the LED chip's absorption of blue is larger because of the absorption qualities. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ increases the green light content of WLEDs, improving the color uniformity index. The color uniformity of contemporary lamps is one of the most important characteristics. Naturally, WLED costs more as the color uniformity index increases. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$, on the other hand, has the benefit of being inexpensive. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ could be utilized in a variety of applications.

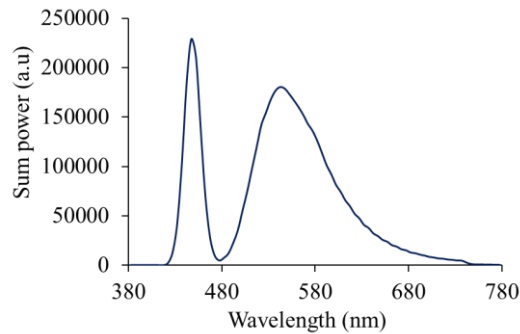


Figure 2. The radiation spectra in 6000 K WLED device along with $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ concentration

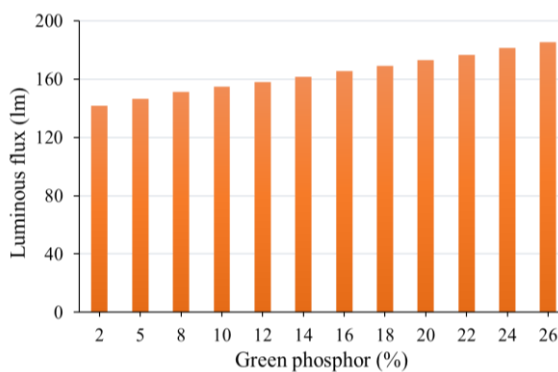


Figure 3. The illuminating beam of WLEDs as a function of $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ concentration

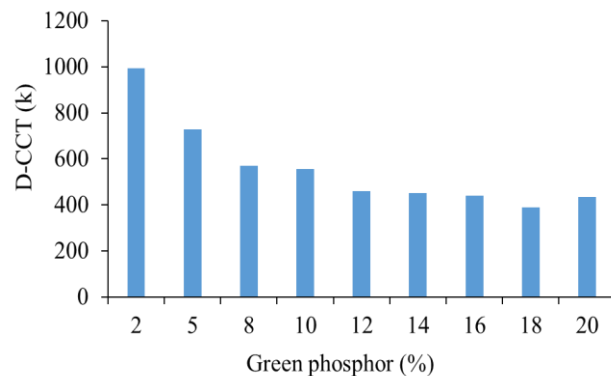


Figure 4. The chroma deviation of WLEDs along with $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ concentration

In the evaluation of color quality, color uniformity is just one criterion. There can be no such thing as good colour quality with a high index of color uniformity. Recent research has developed an indicator for color rendering and a color quality scale. An object's real color is assessed when the colour-rendering index (CRI) is lit. There is too much green light between the three primary hues, resulting in an unbalanced color scheme. As a result, WLEDs' color fidelity is degraded. $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ is a distant phosphor, and the results in Figure 5 suggest a small drop in CRI. In spite of the fact that they are appropriate, colour quality scale (CQS) only has a problem when it comes to CRI. It's clear that compared to the importance of the CQS to the CRI, the CQS comes out on top [16]-[18]. CQS is a three-factor index that takes into account the color rendering index, viewer choice, and hue coordinate. On the basis of these variables, CQS is nearly an accurate measure [19]-[26]. The phosphor is shown to improve CQS in Figure 6. As a result, the CQS does not alter considerably with $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ concentrations less than 10% wt. CQS and CRI are considerably diminished when the phosphor's concentrations are above 10% wt. due to the severe color loss and the domination of green. Applying $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ (green) necessitates a careful selection of concentration.

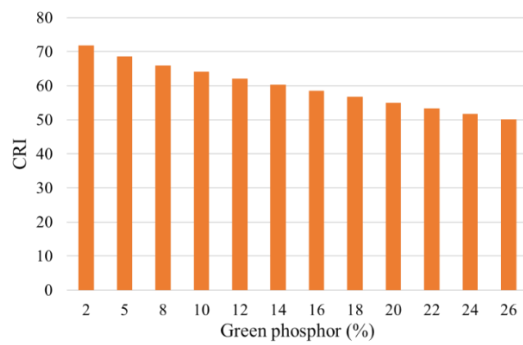


Figure 5. The hue generation indicator in the WLED device along with $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ concentration

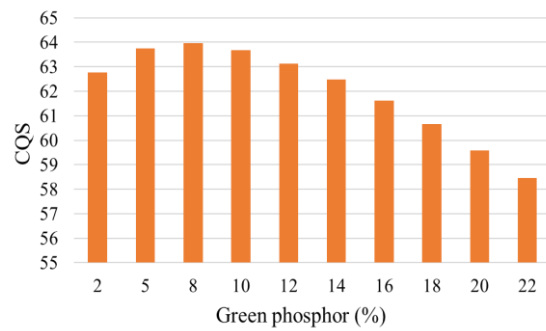


Figure 6. The hue standard scale in WLEDs as a function of $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ concentration

4. CONCLUSION

$\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ solid solutions made through a solid-state reaction at high thermal. Researchers looked at the crystal design and optical characteristics of the materials. Sr concentration in $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ phosphors were modified to produce an appropriate peak location of emission in the range of 560-540 nm. The absorptivity for the 350-500 nm regions precisely fits the close-ultraviolet or LED chip that generates blue lighting, which is GaN-based, according to the researchers. WLEDs can benefit from the use of $\text{Ca}_{1-x}\text{Sr}_x\text{LaGa}_3\text{S}_6\text{O}:0.05\text{Eu}^{2+}$ based solution.

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