

# The application of phosphor $\text{Sr}_4\text{La}(\text{PO}_4)_3\text{O}:\text{Ce}^{3+},\text{Tb}^{3+},\text{Mn}^{2+}$ on white light-emitting diodes

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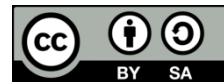
$\text{Sr}_4\text{La}(\text{PO}_4)_3\text{O}$

WLEDs

## ABSTRACT

Using a high temperature solid-state technique, various phosphors  $\text{Sr}_4\text{La}(\text{PO}_4)_3\text{O}:\text{Ce}^{3+}, \text{Tb}^{3+}, \text{Mn}^{2+}$  (SLPO:Ce,Tb,Mn) were produced. In  $\text{Sr}_4\text{La}(\text{PO}_4)_3\text{O}$ , the luminescence characteristics, thermal stability, and energy conversion between  $\text{Ce}^{3+}$  and  $\text{Tb}^{3+}-\text{Mn}^{2+}$  have been thoroughly studied. The sensitizer  $\text{Ce}^{3+}$  particles inclusion could considerably improve the feeble  $\text{Tb}^{3+}$  green radiation and  $\text{Mn}^{2+}$  red radiation via energy transfer. By adjusting the scale of  $\text{Ce}^{3+}/\text{Tb}^{3+}$  over  $\text{Ce}^{3+}/\text{Mn}^{2+}$  atoms, the color of the emission may be changed. White light was produced in the  $\text{Sr}_4\text{La}(\text{PO}_4)_3\text{O}:0.12\text{Ce}^{3+},0.3\text{Mn}^{2+}$  specimen with chromatic coordinates of (0.3326, 0.3298), showing that the phosphors SLPO:Ce,Tb,Mn possess promising application on white light-emitting diodes (WLEDs). The conclusion is supported by Mie-scattering theory and Lambert-Beer's law.

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## 1. INTRODUCTION

Due to various properties such as extended duration, minimal energy usage, compact size, and environmental safety, people utilize white light-emitting diodes (WLEDs) which attracted tremendous potential attention like solid-state illumination industry [1]-[3]. At this time, the producing white color light that is very popular method of is merging the InGaN chip in blue color and the yellow color phosphor  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$  (YAG: Ce) [4], [5]. Nevertheless, owing to the shortage in the red-light component, this kind of white color illumination output has small hue rendered indice ( $\text{CRI}\approx 70-80$ ) as well as great correlating colour temperature ( $\text{CCT}\approx 7500\text{ K}$ ). Many academics have devoted the past few years to the investigation of blue-light-activated red phosphors. Related to these phosphor, there are  $\text{Na}_5\text{Ln}(\text{MoO}_4)_4:\text{Eu}^{3+}$  ( $\text{Ln}=\text{La}, \text{Gd}, \text{Y}$ ),  $\text{CaS}:\text{Eu}^{2+}$ ,  $\text{CaAlSiN}_3:\text{Eu}^{2+}$ , and  $\text{K}_2\text{SiF}_6:\text{Mn}^{4+}$ , etc included [6]-[9]. Low correlated color temperature (CCT) and high color rendering index (CRI) values can be shown by the phosphor converted WLEDs when using the phosphor mentioned above. However, between green and red phosphors were significantly re-absorbed, as well as expensive production costs that cause low blue emission efficiency, is another difficult challenge. As a result, there have been various research derive from the energy transmission (ET) process between the sensitizers and the activators conducted to create efficient, long-lasting, single-stage phosphors of white color light emission featuring the colors of red, green, and blue (RGB) elements [10]-[13].

Co-doping  $\text{Ce}^{3+}$ ,  $\text{Tb}^{3+}$ , or  $\text{Mn}^{2+}$  ions is a popular approach for achieving white color light output by the single-stage phosphors energy transmission process between different kinds of WLED phosphors.  $\text{Ce}^{3+}$

has a ground state of  $4f^15d^0$  and an excited state of  $4f^05d^1$  and so exhibits normal equality permitted 5d-4f electronic transitions. Emission from the 5d-4f transition can occur throughout a wide range of wavelength due to the high dependence of  $Ce^{3+}$  energy levels' distribution on the host lattice [14]-[16]. We have extensively employed both green  $Tb^{3+}$  and red  $Mn^{2+}$  emission components particles. Nonetheless, due to prohibited 4f-4f transition of  $Tb^{3+}$  as well as  ${}^4T_1-{}^6A_1$  conversion of  $Mn^{2+}$ , Between the ultra-violet and detectable range, the excitation bands of  $Tb^{3+}$  and  $Mn^{2+}$  particles are exceedingly weak. To increase the absorption of  $Tb^{3+}$  and  $Mn^{2+}$  in the ultraviolet (UV) zone, an efficient sensitizer such as  $Ce^{3+}$  is commonly employed, and the highly absorbed stimulation power may be transmitted between  $Ce^{3+}$  5d level and  $Tb^{3+}$   ${}^5D_{3,4}$  level or  $Mn^{2+}$  4G level. This method yields emission-tunable single-phase phosphors, creating benefits such as decreased production costs, superior color uniformity, along with greater luminous efficacy [17]-[19]. Because of their excellent physical and chemical stability, apatites were suitable phosphor anchor substances as of now. The compound  $Sr_4La(PO_4)_3O$  (SLPO) is isomorphic to apatite and has two of cationic groups, a nine-time coordinate of 4f point accompanied by  $C_3$  point asymmetry as well as a seven-time coordinate of 6h site alongside the  $C_s$  point asymmetry [20], [21]. The SLPO:  $Eu^{3+}/Tb^{3+}/Ce^{3+}$  phosphors were investigated in our earlier work.  $Tb^{3+}$  or  $Mn^{2+}$  were added into SLPO:  $Ce^{3+}$  in this study. ET may be used for  $Tb^{3+}$  to gain the 539nm peak of green output light and the approximately 605 nm peak of  $Mn^{2+}$  red output band. We thoroughly analyzed the photoluminescence characteristics and ET behaviors of  $Ce^{3+}$ ,  $Tb^{3+}$ , and  $Mn^{2+}$ . Adjusting the  $Tb^{3+}$  or  $Mn^{2+}$  dopant concentration resulted in tunable emitting colors ranging between blue and green and warm white, indicating a potential usage in UV-pumped WLEDs.

## 2. COMPUTATIONAL SIMULATION

### 2.1. Characterization of the $Sr_4La(PO_4)_3O:Ce^{3+},Tb^{3+},Mn^{2+}$

Typical high temperature solid-state processes were used to create  $Sr_4La_{1-x-y}(PO_4)_3O: xCe^{3+}, yTb^{3+}$  and  $Sr_{4-z}La_{1-x}(PO_4)_3O: xCe^{3+}, zMn^{2+}$  phosphors with various elements.  $SrCO_3$  (99.9%),  $(NH_4)_2HPO_4$  (99.9%),  $La_2O_3$  (99.99%),  $Tb_4O_7$  (99.99%),  $MnCO_3$  (99.9%), along with  $CeO_2$  (99.9%) were applied for starting compositions. As flux, 2wt%  $Li_2CO_3$  (99%) was injected [22], [23]. With an agate mortar, the stoichiometric initial chemicals were completely combined and ground. First, we prefire the whole mixture with natural atmosphere in 3 hours at 600 °C, then pulverized it and carbonized it in 5 hours in reducing environment (N<sub>2</sub>:H<sub>2</sub>=95:5). An X-ray diffractometer (XRD) using Cu K radiation was used to measure phase clarity (=1.5406) activated at 40 kV and 35 mA. A UV/visible spectrophotometer and  $BaSO_4$  as guiding element were used to obtain diffuse reflection spectra (DRS) in the 200-700 nm range. An area emission scanning electronmicroscope was applied to study the morphologies of the as-prepared specimens. The photoluminance stimulation (PLE) and photoluminance (PL) spectrum were obtained using a fluorescence spectrophotometer and a 150 W xenon luminous supply. With a FS5 spectrofluorometer device, the thermal based magnitude was conducted. A heating unit that can be raised from 25 to 200 °C in 1 °C increments was used on the samples. Photoluminescence externals quantum values of the samples were achieved using an integrated spheroid within the FS5 spectrophotometer. Fluorescence duration on the Edinburgh FLS 920 Fluorescence Spectrophotometer was monitored.

### 2.2. Luminous characteristics and energy transmission in SLPO: $Ce^{3+},Tb^{3+}$

The Kubelka-Munk (K-M) equation was used to calculate the absorption spectra for SLPO from its scattered spectra in order to experimentally determine the SLPO compound optic bandspac magnitude [24], which is expressed as (1). The Tauc connection links the bandgap  $E_g$  and  $\alpha$  with the straight absorption coefficient of a substance [25], as used in (2).

$$F(R) = \frac{(1-R)^2}{2R} = K/S \quad (1)$$

$$ahv \propto (hv - E_g)^{n/2} \quad (2)$$

In which, the coefficients of backscattering, absorption, and diffusion are denoted by R, K, and S, respectively. Photonic power is illustrated by v and n=1, 2, 3, 4, and 6 are direct transfers permitted, non-metallic substances, immediate transfers prohibited, mediate transfers permitted and mediate transfers prohibited. The absorbance K reaches  $2\alpha$  in case the substance is totally diffusely distributed. Because the dispersing number S could be considered as consistent for the wavelength, we can derive the following expression using (1) and (2).

$$[F(R)hv]^2 \propto (hv - E_g)^n \quad (3)$$

We can calculate from the graph of  $[F(R)hv]^2$ ,  $[F(R)hv]$ ,  $[F(R)hv]^{2/3}$ ,  $[F(R)hv]^{1/2}$ , and  $[F(R)hv]^{1/3}$  from  $hv$  photon power the optimum linear matching at the absorbing edge of  $n=2$ . When the  $[F(R)hv]$  is 0, the SLPO optic bandgap (3.85 eV) value may be achieved via extrapolating the linear matching. Actual multi-chip white LEDs (MCW-LEDs) phosphorus layer of is replicated with flat silicone layers based on the LightTools 9.0 Program as well as the Monte Carlo approach. There are two primary periods in this simulation method: (1) the definition and construction of MCW-LED lamp design and optical properties is crucial (2). The SLPO:Ce,Tb,Mn concentration variation also affects the phosphorous compounding optical effects. We have to create some disparities to see the effect of phosphorous composites YAG:Ce<sup>3+</sup> and SLPO:Ce,Tb,Mn on the output of the MCW-LED lights. The two types of CCTs with averages of 3000 K, 4000 K and 5000 K are necessary described as double-layer remote phosphorus for clarification. The MCW-LED light of protective-coating phosphorous composition with 8500 K mean CCT of is fully described in Figure 1. It is also recommended to simulate MCW-LEDs when SLPO:Ce,Tb,Mn does not feature in the components. The reflectors are 8 mm in beneath lengthiness, 2,07 mm in altitude along with 9,85 mm in outside-cover's lengthiness. Nine chips, with a thickness of 0.08 mm, are coated with the conformal phosphorus compound. Each LED chip has an area of quadratic base at 1.14 mm<sup>2</sup> and a 0.15 mm high, which is linked to the reflector gap. The radiant flux is at 1.16 W for each blue chip while the peak wavelength is 453 nm.

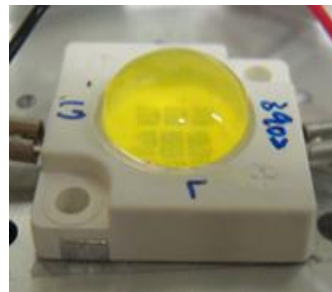


Figure 1. Photograph of WLEDs

**3. RESULTS AND ANALYSIS**

Figure 2 depicts the inverse relationship involving SLPO:Ce,Tb,Mn as well as YAG:Ce<sup>3+</sup>, which is critical to preserve the pre-set values of WLED's CCTs. Moreover, as the increasing SLPO:Ce,Tb,Mn concentration initiates the decrease in YAG:Ce<sup>3+</sup> concentration, the absorption and scattering features of WLEDs' double phosphor layers are also affected. As a result, the colour standard, as well as the efficiency of WLEDs' luminous flux, are eventually influenced. Thus, the color quality of WLEDs is defined by choices of SLPO:Ce,Tb,Mn. The YAG:Ce<sup>3+</sup> concentration decreases to maintain the average CCTs, as the SLPO:Ce,Tb,Mn rose from 5% to 15% Wt. The WLEDs with a huge temperature ranging from 5600 K to 8500 K are the same.

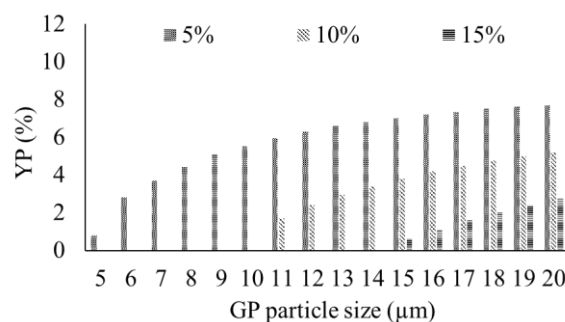


Figure 2. Varying the phosphor concentration to keep the average CCT

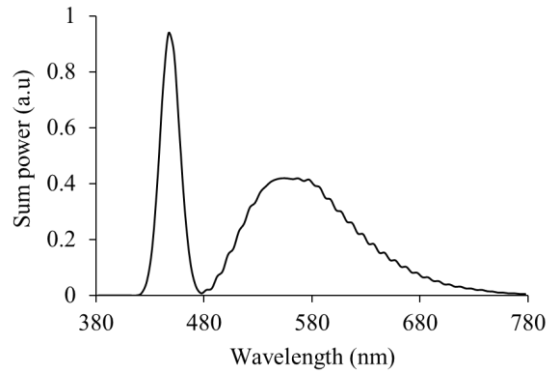


Figure 3. SLPO:Ce,Tb,Mn concentration functions as the 5000 K emitting spectrum of WLEDs

Figure 3 shows the influence of the green phosphorus SLPO:Ce,Tb,Mn concentration on the WLEDs transmission spectrum band. A decision on SLPO:Ce,Tb,Mn concentration can be made according to the manufacturer's criteria. WLEDs with great hue standards can reduce luminance with a tiny amount. The synthesis of the spectral area like Figure 3 is white light. The spectrum of this figure is 5000 K. In these light spectra zones, 420-480 nm and 500-640 nm, the intensity tendency increases with the concentration of SLPO:Ce,Tb,Mn. This expansion in the two-range emitting spectrum shows a growth in the light productivity flow. Furthermore, WLEDs have increased blue-colour light dispersion, implying a rise in the dispersion inside the phosphorous film and in the WLEDs, which promotes hue homogeneity. This is a significant outcome for SLPO:Ce,Tb,Mn applications. Especially in the high-temperature remote phosphorus arrangement, it is difficult to regulate the color consistency. This work has proven the possibility of enhancing the color quality of the WLEDs by SLPO:Ce,Tb,Mn, including low color (5600 K) along with great colour heat (8500 K).

Thus, this article showed the effectiveness of this dual-remote layers of phosphorus released from the luminous flux. Results in Figure 4 demonstrate, in particular, that luminous flux radiated considerably increased from 2% wt to 20% wt by SLPO:Ce,Tb,Mn concentration rise. The color deviation values within three median CCTs has been significantly reduced by the concentration of SLPO:Ce,Tb,Mn phosphorus, as shown in Figure 5. The absorbing of the red phosphorous layer supports this. These blue phosphorus-particles capture the LED chip's blue emitting light and convert it to green emitting light by absorbing it with SLPO:Ce,Tb,Mn phosphorus. SLPO:Ce,Tb,Mn particles absorb yellow light. However, LED chip blue light's absorbing an is higher compared to these two absorbers because of the material's absorption characteristics. Thus, with the addition of SLPO:Ce,Tb,Mn, the WLEDs green illumination concentration is increasing, leading to an improved hue homogeneity index. Hue homogeneity among modern light parameters is one of the major criteria. The greater the color homogeneity index, of course, the more expensive the cost of white WLED light. The advantage of using SLPO:Ce,Tb,Mn is that it is inexpensive which enables for a greater use.

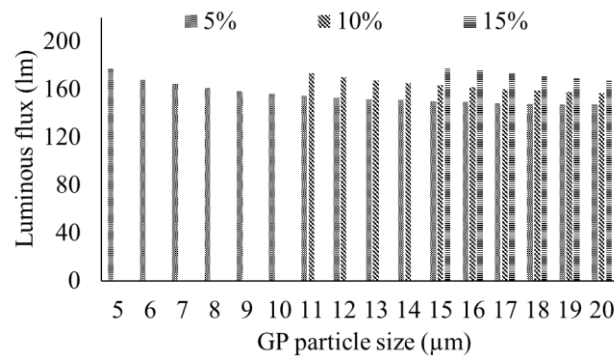


Figure 4. SLPO:Ce,Tb,Mn concentration functions as WLEDs flux of light

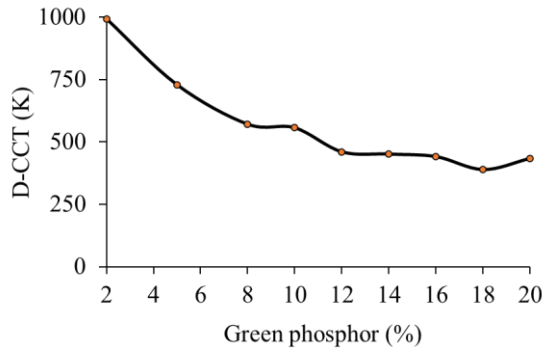


Figure 5. SLPO:Ce,Tb,Mn concentration functions as WLEDs colour deviation

Uniformity of color distribution is just a single factor to take into account while evaluating the color standard for WLED light. It cannot be claimed that color quality is better with a high color homogeneity index. Hence, a color rendition score and color quality scale is offered in recent research. The real color of an item is evaluated if the color rendering index is lit by a light. The dominating green light that lies among these main blue, yellow, and green colours causes color imbalance. WLED's colour standard has been affected by this result, leading to a decrease in WLED's color accuracy. Figure 6 shows a modest decrease in CRI when there is the remote phosphorus SLPO:Ce,Tb,Mn layer. However, they are allowed as CRI is just a color quality scale (CQS's) disadvantage. The CQS is more essential and difficult to attain compared to the CQS. CQS is an index determined based on three elements: the color rendering indice, user's inclination, and colour coordinate. For these three important elements, CQS is virtually a genuine all color quality index. More than this, at a SLPO:Ce,Tb,Mn concentration of less than 10% wt, CQS does not alter appreciably when the concentration is raised, as demonstrated in Figure 7. CRI and CQS are lowered substantially owing to the severe loss in color when green is dominant with over 10% wt of SLPO:Ce,Tb,Mn concentration. Applying SLPO:Ce,Tb,Mn green phosphorus thus requires an appropriate concentration selection.

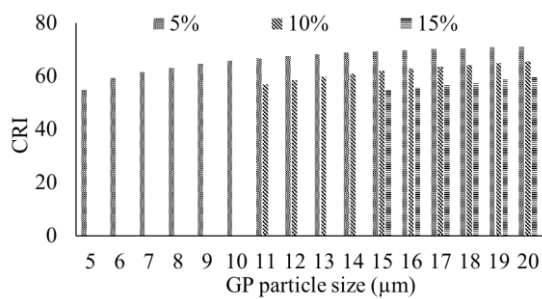


Figure 6. SLPO:Ce,Tb,Mn concentration functions as WLEDs colour rendering indice

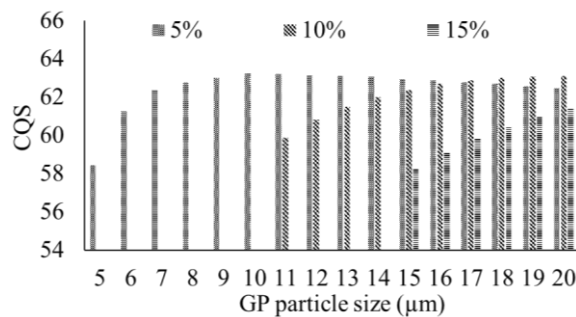


Figure 7. SLPO:Ce,Tb,Mn concentration functions as WLEDs colour quality scale

#### 4. CONCLUSION

The influence of the double-layer of SLPO:Ce,Tb,Mn green phosphorus structure on optic characteristics is described in this paper. The study showed that SLPO:Ce,Tb,Mn is the optimal choice to increase the colour uniformity based on computational simulations by Monte Carlo. This is not only true of WLEDs of 5,000 K, but also of WLEDs with an overall color temperature of more than 8,500 K. The results of this research have therefore achieved an aim of improving the quality of hue and illuminating beam, which have the distant phosphorus design is highly complex. But for CRI and CQS, there is a slight disadvantage. CRI decreases substantially when the concentration of SLPO:Ce,Tb,Mn is elevated. The correct concentration must thus be selected, based on the manufacturer's aims. The study provided a large number of critical facts for reference in order to produce higher color uniformity and luminous flux WLEDs. The following sets of SLPO's are synthesized using conventional solid-state reactions:  $0.12\text{Ce}^{3+}$ ,  $y\text{Tb}^{3+}$  as well as SLPO:  $0.12\text{Ce}^{3+}$ , with  $z\text{Mn}^{2+}$ . Detailed study was carried out on ETs among  $\text{Ce}^{3+}$  and  $\text{Tb}^{3+}$ ; among  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$ . Dipolar dipole interaction was prevailing within the processes of the two ETs.  $\text{Tb}^{3+}$  or  $\text{Mn}^{2+}$  ion concentrations can be adjusted to produce a variety of emission hues ranging between greenish blue to green and warm white. In SLPO:  $0.12\text{Ce}^{3+}$ ,  $0.3\text{Mn}^{2+}$  phosphor under 310 nm UV-light stimulation, white light emission of (0.3326, 0.3298) CIE value may be obtained. The chips may be manufactured with varied emitting wavelengths for phosphorus manufacturing in LED, indicating the SLPO:  $\text{Ce}^{3+}/\text{Tb}^{3+}/\text{Mn}^{2+}$  is a prospect of one-composition phosphorus with white LEDs.

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