International Journal of Scientific Research in Science and Technology
Print ISSN: 2395-6011 | Online ISSN: 2395-602X (www.ijsrst.com)
© 2021 | IJSRST | Volume 8 - Issue 1

White Light Emission from $La_2(MoO4)_3$: Dy^{3+} Phosphor

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ABSTRACT

White light-emitting Dy3+ activated La2(MoO4)3 phosphor was successfully synthesized by a solid-state reaction method. Their structural, morphological, and luminescence properties were characterized by Photoluminescence techniques. Under ultraviolet (UV) and blue excitation, synthesized phosphor exhibits two emission bandsat blue (484nm) and yellow (575 nm),which correspond to4F9/2→6H15/2and 4F9/2→6H13/2 transitions of Dy3+, respectively.The optimized concentration of Dy3+ ions is 0.7mol% after the concentration quenching takes place. The CIE chromaticity coordinates for the optimized phosphor are (0.329, 0.377), and they lie in the white light region. The above-mentioned results demonstrate thatDy3+ activated La2(MoO4)3 is a potential phosphor for solid-state lighting applications.

Keywords : Solid-state reaction method; Photoluminescence; Concentration quenching; Solid-State lighting application

I. INTRODUCTION

In the recent few years, pc-WLEDs have gained more popularity due to their marvellous advantages. Now, it is commercially used, as like, traffic signals, large outdoor displays, interior and exterior lighting in aircraft, cars, and buses, as bulbs in flashlights and as backlighting for cell phones and liquid-crystal displays[1–3].At present three methods are known for production of WLEDs: first is mixing of Red, Green, Blue (RGB) LEDs, second is the commercial WLEDs, which consists of an InGaN-based blue LED chip and yellow-emitting phosphors (YAG: Ce^{3+}), and another one is near UV chip excited tricolour (RGB) phosphors [4]. Currently, commercial WLEDs have realized some drawbacks such as low color rendering effects, high color temperatures, and the absence of red-emitting components. Because of these shortcomings and weaknesses, researchers, scientists, and industrialists have made several attempts to develop inorganic phosphors for white light generation. So far, various rare-earth activated phosphors have been investigated, but these phosphorus have some drawbacks, such as shorter lifetime and lower quantum efficiency [5].Near UV-LEDs in combination with blue, green, and red-emitting phosphors show superior luminescence properties over the commercialized blue-emitting LED with yellow-emitting phosphors. However, phosphor development for near UV LEDs is a challenging problem and a vibrant area of research. The rare earth (RE) ions, activated phosphors found to be excellent luminescent materials.Trivalent dysprosium (Dy^{3+}) rare earth ions most suitable rare earth for potential single white light center because its emission is very close to white owing to the blue ($^4F_{9/2}\rightarrow{}^6H_{15/2}$) and yellow ($^4F_{9/2}\rightarrow{}^6H_{13/2}$) emission. Moreover its red spectral part comparing to the $^4F_{9/2}\rightarrow{}^6H_{11/2}$transition can improvethe color temperature[6,7]. For solid-state lighting applications, the Molybdanate materials have gained significant attention as self-activated and rare-earth ions doped Molybdanate host materials. Molybdanate based hosts have broadband absorption and emission band in the UV and visible regions due to the electronic charge transfer band. The Molybdanate-based host materials doped with Dy^{3+} phosphors are mostly used in WLEDs because Molybdanate have higher chemical stabilities and absorption in the UV region to the visible region. In the past, Molybdanate host matrices have been widely used in optoelectronic applications due to their many advantages, such as low cost, high stability, easy fabrication, and excellent physical-chemical properties[8,9].

According to the literature, photoluminescence properties never reported for the $La_2(MoO_4)_3:xDy^{3+}$ phosphor. In this study, $La_2(MoO_4)_3:xDy^{3+}$ phosphor was synthesized using Solid State Reaction method and their luminescence properties were analyzed by Photoluminescence Technique.

II. METHODS AND MATERIAL

Synthesis

All sample powder was synthesized by solid-state diffusion method. The $La_2(MoO_4)_3:xDy^{3+}$ phosphors (x = 0, 0.1, 0.2, 0.5, 1 and 2mol%) were preparedby using La_2O_3, $(NH_4)_2MoO_4$ and Dy_2O_3 as starting materials.These materials were weighed according to the designed compositions. The stoichiometric amounts of starting reagents were thoroughly mixed for 1 h using a mortar and pestle. The obtained powder was annealed in a furnace at 400 °C for 2 h using ceramic crucible. The obtained powder was again ground for 1 h using a mortar and pestle and, then, it was heated at 800 °C for 24 h for proper interaction of all reagents via diffusion. After the treatment, the samples were cooled to room temperature and grounded in the final powder products.

$La_2O_3 + 3(NH_4)_2MoO_4 \rightarrow La_2(MoO_4)_3 + 6.NH_3\uparrow + 3.H_2O\uparrow$

III. RESULTS AND DISCUSSION

3.1 XRD and SEM measurement

The phase confirmation and the crystallinity of synthesized phosphors were analyzed by XRD measurement and particle size, morphological behavior was observed by scanning electron microscope. XRD pattern and SEM images of $La_2(MoO_4)_3$:1mol%Eu^{3+} phosphor phosphors are already recently published [10] . The XRD pattern of synthesized phosphor shows a good crystalline nature and XRD patterns well matched with that of ICSD Ref. Code 98-2634. The phase of the phosphors is identified as monoclinic structure in space group C2/c. The SEM images show that the phosphor consists of spherical shape. The particle size of the synthesized phosphor is around 1μm.

3.2 Photoluminescence study

The excitation spectrum of synthesized Dy^{3+} activated $La_2(MoO_4)_3$ phosphor monitored under 482nm emission wavelength in the range of 300nm to 460nm. The excitation spectrum depicts fivehighly intense excitation peaks in the UV and visible region as shown in Figure 1. The Excitation peaks are situated around at 324nm, 350nm, 365nm, 386nm and 454nm, which are ascribed due to $^6H_{15/2} \rightarrow {}^6P_{3/2}$, $^6H_{15/2} \rightarrow {}^6P_{7/2}$, $^6H_{15/2} \rightarrow {}^6P_{5/2}$, $^6H_{15/2} \rightarrow {}^4I_{13/2}$, and$^6H_{15/2} \rightarrow {}^4I_{15/2}$ respectively [11].All the excitation peaks situated in 320nm to 460nm, which is indicating that Dy^{3+} ions may be used as efficient activators for white LEDs[11].

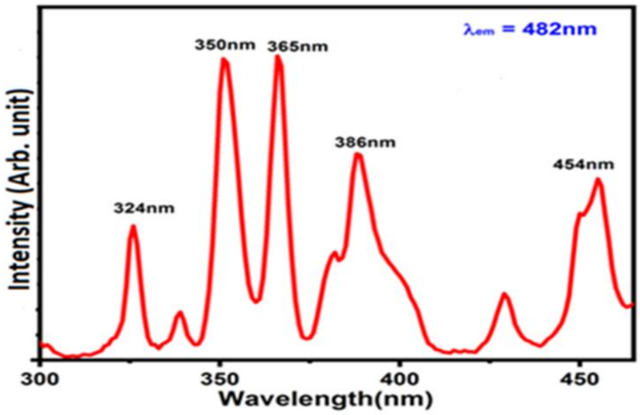

Figure 1: PL excitation spectrum of Dy^{3+} activated $La_2(MoO_4)_3$phosphor under 482nm emission wavelength

Figure 2 (A) represents PL emission spectra of $La_2(MoO_4)_3$:xDy^{3+}(x = 0.1, 0.2, 0.5, 0.7, 1.0mol%)phosphors under 350nm excitation wavelength. The emission spectra depicts two emission peak around at 481nm, 491 (blue region) and 573nm (Yellow region), corresponding to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions, respectively.The $^4F_{9/2} \rightarrow {}^6H_{15/2}$ transition can be stated as a magnetic dipole transition, whereas the $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transition can be attributed to a forced electric dipole transition[12].However, the peak intensity is found to increase when the dopant concentration varies from x=0.05 to 0.7mol%. After 0.7mol% concentration of Dysprosium ions PL emission intensity suddenly decrease because of concentration quenching effect.Figure 2(b)shows concentration quenching spectra of $La_2(MoO_4)_3$:Dy^{3+} phosphor.

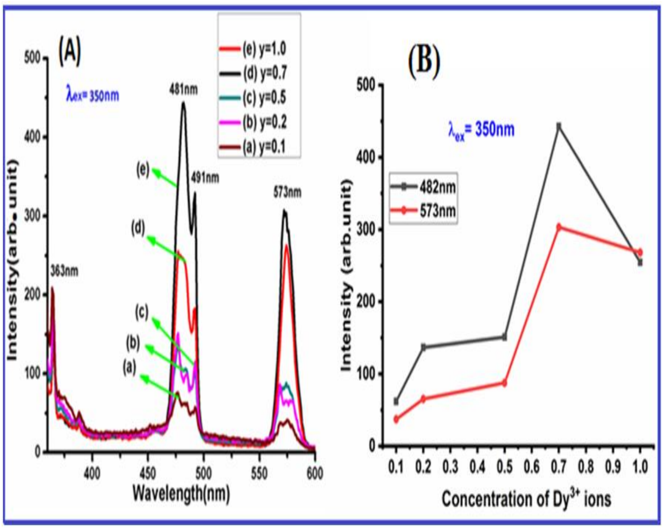

Figure 2 (A) PL emission spectrum of $La_2(MoO_4)_3$:xDy^{3+} (x 0.1, 0.2, 0.5, 0.7, 1.0mol%)phosphor under 350nm excitation wavelength and **Figure 2 (B)**Variation in PL emission intensity with concentration of Eu^{3+} ions

3.3 Photometric Characterization

In 1931, the Commission International de l'Eclairage (CIE) co-ordinates were used to study color emission of the synthesized phosphor in the visible spectrum. Figure 5 shows CIE chromaticity coordinate of synthesized 0.7mol% Dy^{3+} activated $La_2(MoO_4)_3$ phosphor, CIE coordinate were calculated by using OSRAM SILVANIYA Color calculator and PL emission intensity under 350nm, 365nm, 386nm and 454nm excitation wavelength. The calculated coordinate is represented in table 1.

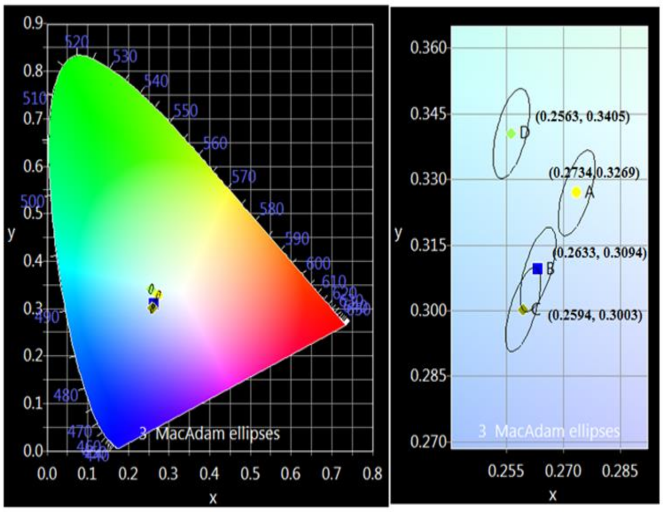

Figure 5:CIE chromaticity Diagram of $La_2(MoO_4)_3$:0.7mol%Dy^{3+} phosphors at different excitation wavelength

Table: 1: CIE Chromaticity diagram and Color Purity of$La_2(MoO_4)_3$:0.7mol%Dy^{3+}phosphors

Sr. No	Compound Name	Excitation wavelength	CIE Chromaticity Coordinates	Color Purity
1.	$La_2(MoO_4)_3$ 0.7mol%Dy^{3+}	350nm	(0.2734, 0.3269)	79%
2.	$La_2(MoO_4)_3$ 0.7mol%Dy^{3+}	365nm	(0.2633, 0.3094)	76%
3.	$La_2(MoO_4)_3$ 0.7mol%Dy^{3+}	386nm	(0.2594, 0.33003)	75%
4.	$La_2(MoO_4)_3$ 0.7mol%Dy^{3+}	454nm	(0.2563, 0.3405)	77%

IV. CONCLUSION

In the present work, the Dy^{3+} activated $La_2(MoO_4)_3$ phosphor were synthesized by solid state reaction method. Under 350nm excitation wavelength, PL emission spectra observed. It shows blue and yellow color emission around 482nm and 573nm transition, which ascribed $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions of Dy^{3+} ions. PL investigation shows PL Emission spectra shows highest intensity under 350nm excitation wavelength. The concentration quenching (CQ) spectra shows highest PL emission intensity observed at 0.7mol% Dy^{3+} concentration. The CIE coordinates for synthesized phosphor were also calculated by using OSRAM SILVANIA color calculator and PL emission intensity. All these results shows that Dy^{3+} activated $La_2(MoO_4)_3$phosphor may be a potential phosphor for solid-state lighting applications.

ACKNOWLEDGEMENT

One of the authors Yatish R. Parauha is thankful to Department of Science and Technology (DST), India for financial support through INSPIRE fellowship (INSPIRE Code – IF180284).One more authors SJD is thankful to Department of Science and Technology (DST), India(Nano Mission) (Sanction Project Ref. No. DST/NM/NS/2018/38(G), dt.16/01/2019) forfinancial assistance.

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