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Photoluminescence study of $Sr_3La_{1-x}(BO_3)_3$: X Mn^{2+} $(0.01\leq X \leq 0.04)$

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ABSTRACT

Color-tunable $Sr_3La_{1-x}(BO_3)_3$:xMn^{2+} phosphors $(0.01\leq X \leq 0.04)$ were prepared via solution combustion synthesis. Phase purity and crystallographic structure were verified by powder X-ray diffraction. X-ray diffraction analysis confirmed the successful incorporation of Mn^{2+} ions into the $Sr_3La(BO_3)_3$ host lattice, maintaining the characteristic borate framework with isolated BO_3^{3-} anionic groups. Steady-state photoluminescence studies performed at room temperature reveal that Mn^{2+}-doped $Sr_3La(BO_3)_3$ exhibits intense green emission under deep-ultraviolet excitation (λ_{ex} = 266 nm), characterized by a broad emission band peaking at 501 nm, originating from the spin-forbidden $^4T_1(^4G) \rightarrow {}^6A_1(^6S)$ d–d transition of tetrahedrally coordinated Mn^{2+} ions. These results demonstrate that $Sr_3La(BO_3)_3$:Mn^{2+} represents a promising rare-earth-free green phosphor candidate with efficient d-UV absorption, pure green emission, and adequate thermal quenching resistance, offering potential applications in phosphor-converted w-LEDs, display backlighting, and plant growth lighting systems. Further optimization through co-doping strategies or host lattice modification could enhance quantum efficiency and color purity for commercial implementation.

Keywords:- Photoluminescence, Borate-phosphor, Solution combustion

1. INTRODUCTION

In the realm of luminescent materials for light-emitting diodes (LEDs), manganese(II) ions (Mn^{2+}) play a pivotal role as activators in borate-based phosphors, particularly for generating efficient green emission under ultraviolet (UV) excitation. Borate hosts, such as compounds like SrB_4O_7 or BaB_2O_4, provide a stable crystal lattice that accommodates Mn^{2+} ions, enabling d-d electronic transitions that result in broad-band green luminescence typically peaking around 500-550 nm. This green light is crucial for achieving high color rendering indices (CRI) in white LEDs, where it complements blue LED chips and red phosphors to produce balanced, natural white light for applications in general lighting, displays, and backlighting systems. The significance of Mn^{2+} lies in its low cost, non-toxicity compared to rare-earth alternatives like Eu^{2+} or Ce^{3+}, and tunable emission properties through host lattice modifications, which enhance quantum efficiency and thermal stability—key factors for energy-efficient, long-lasting LED devices. Furthermore, Mn^{2+}-doped borate phosphors exhibit minimal re-absorption and concentration quenching, making them ideal for UV-pumped systems that avoid the drawbacks of traditional phosphor-converted LEDs, such as Stokes shift losses. As the demand for sustainable and high-performance lighting grows, Mn^{2+} in borates continues to drive innovations in phosphor engineering, contributing to reduced energy consumption and environmental impact in global illumination technologies. Mn^{2+} (manganese(II)) is a d^5 transition metal ion widely used as an activator in phosphors, especially for green emission in UV-excited systems like borate hosts. Mn^{2+} has a high-spin d^5 configuration in most inorganic hosts (all five 3d electrons unpaired).

All d–d transitions are both Spin-forbidden ($\Delta S = 0 \rightarrow \Delta S \neq 0$, since ground state is sextet and excited states are quartet) and Parity-forbidden (Laporte rule: g ↔ g transitions are forbidden in centro-symmetric environments)

It shows extremely weak direct absorption ($\varepsilon \approx 0.1–10\ M^{-1}cm^{-1}$). In practice, Mn^{2+} is almost always sensitized by a co-dopant (Eu^{2+}, Ce^{3+}, Tb^{3+}, etc.) that strongly absorbs UV/blue light and transfers energy. The green (or sometimes yellow/orange) emission of Mn^{2+} originates from the intra-configurational d–d transition: $^4T_1(g) \rightarrow {}^6A_1(s)$. Where Ground state: $^6A_1$ (completely symmetric, highest multiplicity) & First excited quartet state: $^4T_1$ (derived from the free-ion ^4G term). This transition is the lowest-energy spin-allowed (actually spin-forbidden but weakly allowed by spin-orbit coupling and vibronic interactions) emission in most hosts. The emission color of Mn^{2+} is extremely sensitive to the crystal field strength. In Weak crystal field it shows green emission (~500–540 nm). Borates, phosphates, silicates, aluminates having Tetrahedral coordination creates weak crystal field. In Strong crystal field it shows yellow → orange → red (~550–650+ nm) emission. Most borate phosphors (e.g., SrB_4O_7:Mn^{2+}, $Ba_2B_5O_9Cl$:Mn^{2+}, $Li_2B_4O_7$:Mn^{2+}, etc.) place Mn^{2+} in tetrahedral BO_4 units or distorted tetrahedral sites resulting in weak crystal field creating characteristic green emission peaking around 520–535 nm. In borate phosphors designed for UV → green conversion in LEDs, Mn^{2+} is deliberately placed in tetrahedral environments to achieve bright, relatively narrow-band green emission (~FWHM 30–50 nm) with excellent color purity. This combination of fundamental d^5 physics, crystal field tenability, and practical sensitization strategies makes Mn^{2+} one of the most important non-rare-earth activators for modern green-emitting phosphors.

Phosphors that can absorb high-energy UV photons and re-emit lower-energy visible light, often involving lanthanide doped Oxides like Ce^{3+}/Tb^{3+} co-doped phosphors (e.g., $NaBaPO_4:Ce^{3+},Tb^{3+}$) or Bi^{3+}/Eu^{3+} co-doped systems (e.g., $Ba_9Lu_2Si_6O_{24}:Bi^{3+},Eu^{3+}$), utilizing energy transfer from a sensitizer (Bi^{3+} or Ce^{3+}) to an activator (like Eu^{3+} or Tb^{3+}) for tunable green-to-red emission, creating white light or specific colors for plant growth LEDs. Phosphors like $NaBaPO_4:Ce^{3+},Tb^{3+}$ can show green emission around 545 nm when excited by 266 nm, while Bi^{3+}/Eu^{3+} systems can tune from blue to red for white light. In Lanthanide activated materials Sensitizer ions (like Ce^{3+}, Bi^{3+}) absorb the 266 nm UV light and transfer energy to activator ions (like Eu^{3+}, Tb^{3+}) which then emit at specific wavelengths. In Bismuth (Bi^{3+}) Doped Systems $Ba_9Lu_2Si_6O_{24}:Bi^{3+},Eu^{3+}$ efficiently transfer energy from Bi^{3+} (absorbing UV) to Eu^{3+} (red emitter), creating tunable blue, green, or white light. Terbium-doped phosphors, such as $LiYb_{1-x}Tb_x(MoO_4)_2$**,** are notable for being efficiently excited by 266 nm light. They exhibit characteristic green luminescence, often peaking at approximately **545 nm**, but can be tuned toward the 500 nm range depending on the host lattice and co-dopants. Divalent europium (Eu^{2+}) is a common activator for blue-to-green emission. While often excited at 365 nm or 394 nm, certain host matrices like $Na_3La_2(PO_4)_3$ can be excited at 266 nm to produce broad-band blue/cyan emission peaking near 422 nm. Materials like $Ba_9Lu_2Si_6O_{24}:Bi^{3+}$ can exhibit broad emission bands reaching up to 490–500 nm when excited by UV light, serving as tunable components for white-light LEDs. Thermo graphic phosphor, ZnO can be excited by 266 nm (often from a Nd:YAG laser) to avoid spectral overlap with Raman scattering. It emits luminescence that is highly sensitive to temperature, making it useful for fluid diagnostics. In the present work we successfully utilized non rare earth, non toxic activator Mn^{2+} in $Sr_3La(Bo_3)_3$ borate phosphor to produce green light under DUV excitation.

2. EXPERIMENTAL DETAILS

Powder sample of $Sr_3La_{1-x}(BO_3)_4$:-xMn^{2+} inorganic borate phosphors were prepared by the solution combustion synthesis technique [8,9]. Stoichiometric amounts of high purity starting materials, $SrNO_3$ (A.R.), $Mn(NO_3)_2$ (high purity 99.9%), H_3BO_3 (A.R.), $CO(NH_2)_2$ (A.R.), ($La(NO_3)_3\cdot 6H_2O$) (A.R.) are used for phosphor preparation. The starting materials with little amount of double distilled water were mixed thoroughly in agate mortar to obtain a homogeneous solution. Excess water was removed by heating the samples at temperature 100 ^{O}C for about 30 min and the paste was then transferred directly to a pre-heated Muffle furnace, maintained at temperature 680^{O}C, for combustion. Following the combustion, the resulting foamy samples was crushed to obtain fine particles and then annealed in activated charcoal for 3 h at temperature 950^{o}C. As prepared Borate phosphor material was characterized by powder XRD, PL and FT-IR techniques.

TABLE 1: Molar concentration of ingredients

$3[Sr(NO_3)_2]$+ x[$(Mn(NO_3)_2)$] + (1-x) $(La(NO_3)_3\cdot 6H_2O)$ + $3H_3BO_3$ + $CO(NH_2)_2$ $\xrightarrow{680\ ^{o}C}$ $Sr_3La_{1-x}(BO_3)_4$:-xMn^{2+} + Gaseous products				
$Sr(NO_3)_2$	$Mn(NO_3)_2$	$La(NO_3)_3$	$3H_3BO_3$	$CO(NH_2)_2$
6.3486	0.0669	1.13	1.855	4.80

3. RESULTS AND DISCUSSION

"The phosphors materials were studied by powder XRD, PL and FT-IR techniques. Powder X-ray diffraction measurements were taken on a Rigaku-Miniflex II X-ray Diffractometer and compared with ICSD files. PL and PLE measurements at room temperature were performed on a Hitachi F-7000 spectro- flurometer with spectral resolution of 2.5 nm. FTIR of sample was done on F.T. Infra-Red Spectrophotometer Model RZX (Perkin Elmer)".

3.1 XRD STUDY OF $Sr_3La(BO_3)_3$

According to Lamentable the JCPDS card and the Inorganic Crystal Structure Database (ICSD) of $Sr_3La(BO_3)_3$ is not available. But all of the diffraction peaks of samples could be read between the lines to pure structure of $Ba_3Dy(BO_3)_3$ (JCPDS NO.50-0098) [23]. Some diffraction peaks of samples had a little shift to high diffraction degrees compared with JCPDS NO.50-0098. The reason may be the difference of ionic radius between Sr and Ba ($R_{Sr}{}^{2+} < R_{Ba}{}^{2+}$). The ionic radius of La is close to Ln^{3+} (Ln = Dy, Eu, Tb). Previous research suggests that, Ln^{3+} might replace the site of La^{3+} in the host structure [24].

K. Denault, Z. Cheng, J. Brgoch, et al., reported the crystal structure of $Sr_6HoSc(BO_3)_6$ which had similar crystal structure with $Sr_3La(BO_3)_3$. The crystal structure of $Sr_3La(BO_3)_3$ was shown in Figure (1), it belongs to the rhombohedral space group $R\bar{3}$ (no. 148). There were three crystalographically independent sites for the Sr^{2+}

and La^{3+} cations and therefore three potential sites for Ln^{3+} substitution. The first site is Wyckoff position 18f is coordinated by 8 oxygen atoms, corresponding to the coordination polyhedron of SrO_8. And the other two sites are Wyckoff positions 3a and 3b in which distorted octahedra coordinated by 6 oxygen atoms formed, corresponding to the coordination polyhedron of LaO_6. The three sites were connected by BO_3 triangle.

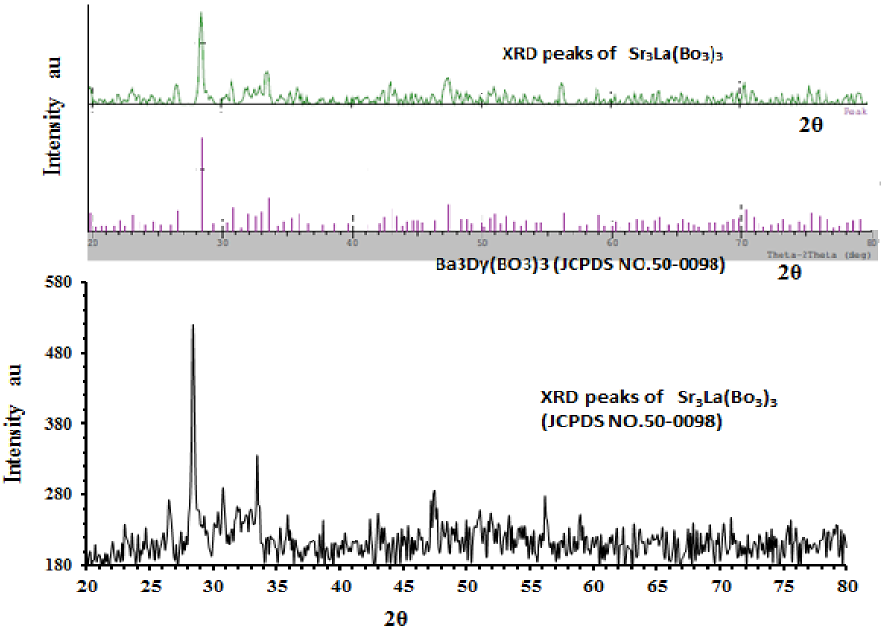

Figure-1. XRD OF $Sr_3La(Bo_3)_3$

3.2 FTIR

FTIR of sample was done on F.T. Infra-Red Spectrophotometer Model RZX (Perkin Elmer).The coordination environment of B–O in $Sr_3La(BO_3)_3$ structure is confirm using, the FTIR spectra measured at room temperature; which is shown in Figure(2). The IR absorption at wave numbers smaller than 500 cm^{-1} mainly originates from the lattice dynamic modes. The strong bands observed above 1100 cm^{-1} should be assigned to the B–O stretching mode of the triangular $[BO_3]^-$ groups, while the bands with maxima at about 700–800 cm^{-1} should be attributed to the B–O out of plane bending, which confirms the existence of the $[BO_3]^-$ groups(12). The absence of peaks in 1500–2000 cm^{-1} indicates the complete removal of nitrate and organic matter. The peak at 3300–3500 cm^{-1} corresponds the stretching mode of O–H are also absent.

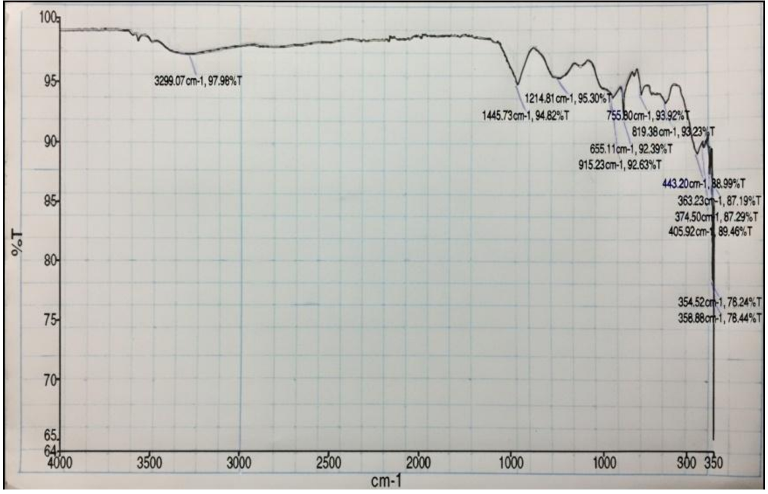

Figure -1 FTIR of $Sr_3La(BO_3)_3$

3.3 PL AND PLE STUDY

Mn^{2+} activated phosphors are important for lighting and display devices. Here Mn^{2+} doped $Sr_3La(BO_3)_3$ green phosphor was synthesized by an conventional combustion method and annealed in reducing atmosphere using charcoal. Upon 266nm excitation green photoluminescence band centered at 501nm is present in PL spectra. The manganese ion activated phosphors have been widely used over years and years. Mn^{2+} phosphors are used in illumination, display and optical imaging. Among various valence Mn ions, Mn^{2+} and Mn^{4+} ions are most used as luminescence activators. The Mn^{2+} ions with $^3d_5$ electron configuration produces wavelength tunable emission ranging from green to red region depending upon host crystal field strength. The Mn^{4+} with $^3d_3$ electron configuration usually shows strong absorption in the ultraviolet blue region and gives sharp emission in the red region (620-680nm). Photoluminescence spectra of $Sr_3La_{0.97}(BO_3)_3$: 0.03 Mn^{2+}phosphor is shown in figure(3).

PL of **$Sr_3La_{1-x}(Bo_3)_3$: X Mn^{2+} (0.01≤ X ≤ 0.04)** phosphor monitored at 266nm is shown in figure (3). It is recorded on F-7000 FL spectrophotometer with scan speed 240 nm/min, excitation-emission slit width 1nm. PLE of $Sr_3La_{0.97}(BO_3)_3$: 0.03 Mn^{2+}phosphor shows narrow absorption band at 266nm and low intensity peak at 363nm. PL spectra of phosphor shows broad emission band in range 400nm to 700nm with maximum intensity at 501nm. PLE monitored for 501nm emission line indicates 266nm absorption band. Mn^{2+} activated green emitting phosphor synthesized at different molar concentration from 1 mole % to 4 mole % . Emission intensity at 3 mole % of Mn^{2+} is maximum. Concentration quenching takes place as we increase the doping concentration beyond the 4 mole% of Mn^{2+}.

For different concentration of Mn^{2+}photoluminescence monitored at 266nm line is a shown in figure(4). For 3mole % concentration of Mn^{2+} PL intensity is maximum and then decreases.

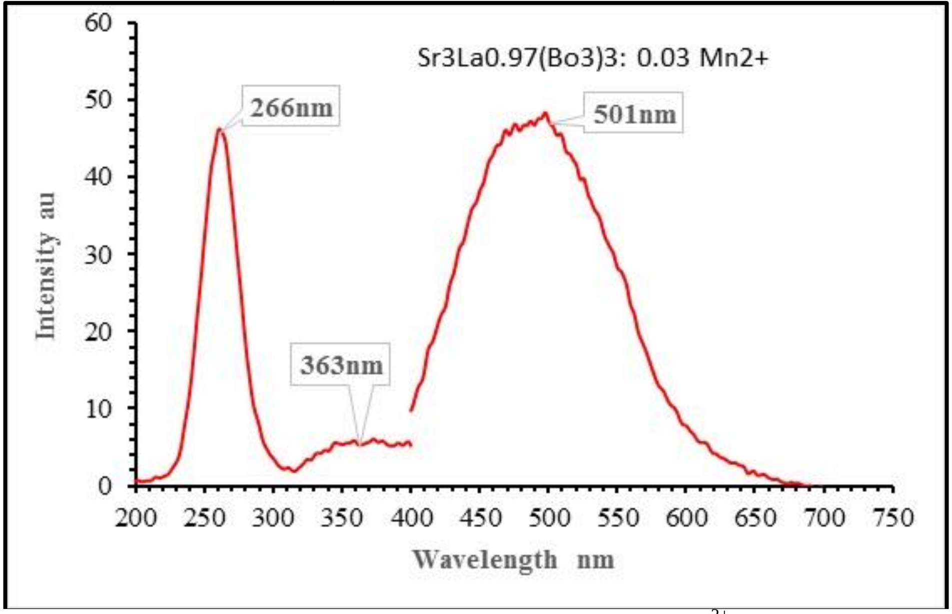

Figure-3 PL-PLE of $Sr_3La_{0.97}(BO_3)_3$: 0.03 Mn^{2+}

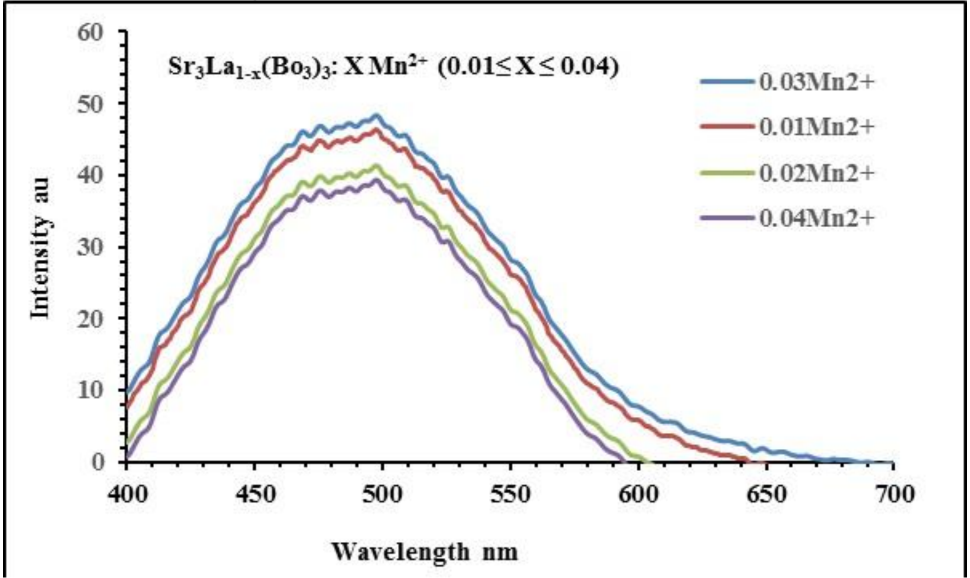

Figure-4 PL of $Sr_3La_{1-x}(BO_3)_3$: X Mn^{2+} (0.01≤ X ≤ 0.04)

3.4 CIE-CHROMATICITY DIAGRAM:

Figure (5) shows the “Commission International del Eclairage (CIE) chromaticity coordinates diagram” of the $Sr_3La_{0.97}(BO_3)_3$: 0.03 Mn^{2+}phosphor when excited at 266nm. The chromaticity coordinates of the phosphor $Sr_3La_{0.97}(BO_3)_3$: 0.03 $Mn^{2+}$for fixed 3 mole % weight concentration of Mn^{2+} when excited at 266 nm are X:0.277 Y:0.441. It comes in green region of CIE Chromaticity diagram and is indicated by black circle in figure (5). It is UV excited green emitting phosphor.

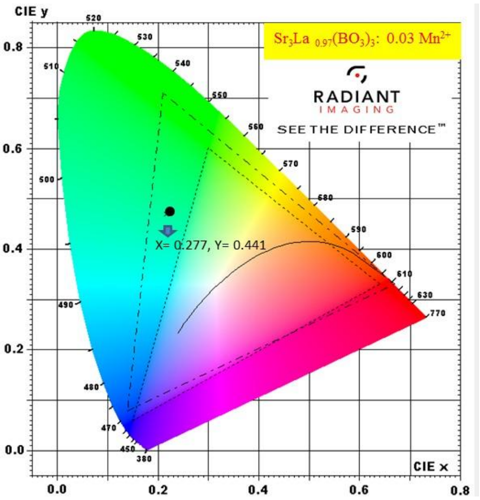

Figure-5 CIE-Chromaticity diagram of $Sr_3La_{0.97}(BO_3)_3$: 0.03Mn^{2+}

4. CONCLUSION

The $Sr_3La_{1-x}(Bo_3)_3$: X Mn^{2+} (0.01≤ X ≤ 0.04) polycrystalline phosphor was synthesized by solution combustion method using urea as a fuel and nitrates as a oxidizer. Using XRD formation of compound is confirmed and it matches with standard JCPDS NO.50-0098. Using FTIR presence of $[BO^3]^-$ groups, coordination environment of B-O in structure and Complete combustion of nitrates and organic matter were confirmed. Phosphor shows narrow excitation band from 200 to 290 nm with prominent peak at 266 nm. PLE for characteristic emission wavelength 501 nm was found to be 266 nm. At 266 nm light excitation $Sr_3La_{1-x}(Bo_3)_3$: X Mn^{2+} phosphor emits green light. As Mn^{2+} is sensitive to surrounding crystal field environment it shows more absorption peak at 363nm. Hence $Sr_3La_{1-x}(Bo_3)_3$: X Mn^{2+} is new DUV excited green emitting phosphor useful for DUV based solid-state lighting (SSL) for white LEDs (by combining with blue/red), plant growth lighting (horticulture) for tailored PAR spectra, biological imaging for deepa-tissue visibility (often NIR, but green is useful), security/anti-counterfeiting (UV excitation, visible emission), and visible light communication (VLC)**.**

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