

Synthesis, Characterization and Luminescence Investigation of Sm³⁺ Doped Na₃Bi(PO₄)₂ Phosphors for White Light Emitting Diodes

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Abstract

The synthesis of the Na₃Bi(PO₄)₂: phosphor that has been activated by Sm³⁺ was accomplished by utilizing a wet-chemical reaction method. Transmission electron microscopy (TEM), Fourier transform infrared spectroscopy (FTIR), UV-Vis diffuse reflectance spectroscopy (DRS), and photoluminescence (PL) were all used to investigate the created Na₃Bi(PO₄)₂:Sm³⁺ phosphors. By utilizing the reflectance spectra of undoped Na₃Bi(PO₄)₂ and doped phosphors Na₃Bi(PO₄)₂:Sm³⁺, researchers have determined the presence of strong energy absorption bands that appear in the region of 200–300 nm. Upon the stimulation of photoluminescence at 404 nm, the doped Na₃Bi(PO₄)₂:Sm³⁺ phosphor exhibits a broad emission in the range of 400-600 nm, with a peak emission wavelength of 598 nm. In contrast, the phosphor Na₃Bi(PO₄)₂ that had been doped with Sm³⁺ had three exceptionally strong emission bands at around 564 nm, 598 nm, and 645 nm, respectively. The transitions that caused the emissions were as follows: ⁴G_{5/2}→⁶H_{5/2} (green emission), ⁴G_{5/2}→⁶H_{7/2} (orange emission), and ⁴G_{5/2}→⁶H_{9/2} (red emission). It has been established that the CIE (Commission Internationale de l'Éclairage) coordinates for the sample are as follows: x = 0.2841 and y = 0.3459. According to these findings, there is the possibility that the Na₃Bi(PO₄)₂:Sm³⁺ phosphors that were synthesized have the potential to be used as a good option for white light emitting diodes and display devices.

Keywords: Na₃Bi(PO₄)₂ phosphors; Wet-chemical method; White light emitting diodes; Display devices.

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1. Introduction

It has been determined that the phosphor, which has an x value that is equal to 0.5 mol%, also has an x value that is equal to 0.2841, a y value that is equal to 0.3459, and is situated in a location that is in close proximity to the color white. Due of this, it could be regarded a promising candidate for near-ultraviolet (NUV) white light-emitting diodes (WLED). This

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is owing to the fact that, throughout the course of the recent past, materials that have been doped with trivalent lanthanides have generated a significant amount of attention among a number of researchers. This is because they are favorable when used in solid-state laser media, fiber amplifiers, and field emission displays [1-4].

The narrow emission bands and the extended radioactive lifetime in the excited state are both characteristics of the species that is referred to as the rare earth ion Sm³⁺. It is challenging to ascertain the energy level of the trivalent samarium ion, which possesses a ⁴f₅ structure. There are a variety of potential transitions between f-energy levels that the Sm³⁺ ion displays. As a consequence of the fact that the transitions are extremely selective, the development of line spectra that are exceptionally different takes place. If the optical materials are doped with appropriate hosts that contain the Sm³⁺ element, they will have the potential to emit white light. Ca₅(PO₄)₃F:Ln (Dy³⁺, Eu³⁺, and Sm³⁺) is one of the many different hosts that have been the subjects of a variety of investigations. A wide range of manufacturing techniques are utilized during the production of WLEDs. The InGaN blue LED chip is responsible for the creation of the yellow phosphor, also known as YAG:Ce³⁺. There is a relationship between the color temperature (CCT) and the color rendering indices (CRI) of white light-emitting diodes (WLEDs) [5,6]. The scientific literature indicates that certain complex oxides, such as silicates, aluminates, and borates, exhibit substantial absorption in the vacuum ultraviolet (VUV) range. Because of their display of pertinent optical features, stoichiometric rare earth phosphates have recently become the subject of intensified inquiry [5-7]. As a consequence of the many condensations of PO₄ groups that occur in the alkali metal–rare earth phosphate system, a number of structural families have been produced, including ML_nP₂O₇, M₃Ln(PO₄)₂, and ML_n(PO₃)₄ (where M represents an alkali metal and L_n represents a rare earth element). Several europium (Eu³⁺)-doped calcium magnesium phosphate (Ca₇Mg₂(PO₄)₆) type phosphors have been synthesized and investigated [8].

The photoluminescence properties of phosphate host materials doped with trivalent samarium ion, have been explored [9,10]. Literature suggests about several reports, namely Sm³⁺ doped Bi₄MgO₄(PO₄)₂. [11], Sm³⁺ doped Ca₁₀(PO₄)₆Cl₂ phosphor [12]. NaBaBi₂(PO₄)₃: xDy³⁺ and NaBaBi₂(PO₄)₃: 0.07Dy³⁺, ySm³⁺ [13], Sr₃Bi(PO₄)₃:Sm³⁺ by wet- chemical method [14]. Similarly, Sm³⁺:A₃(PO₄)₂ (A = Sr, Ca) phosphors [15], (Ca₁₀(PO₄)₆F₂:xLn³⁺ (where x = 0.5 mol for 1-mol FAP and Ln³⁺: Sm³⁺, Eu³⁺, and Dy³⁺) [16], have been reported.

In the course of this inquiry, the wet-chemical method has been employed in order to synthesize and investigate Sm³⁺ activated Na₃Bi(PO₄)₂ phosphors. The chromaticity diagram that was provided by the International Commission on Illumination offers a reflection on the color coordinates and the amount of purity of the color of the Sm³⁺ doped phosphors that were generated.

2. Materials and Methods

2.1. Methodology

The compound $\text{Na}_3\text{Bi}(\text{PO}_4)_2$ undoped and doped with Sm^{3+} ions, at several concentrations of Sm^{3+} : $x = 0.0, 0.3, 0.5,$ and 0.7 mol % were prepared using the modified wet chemical method. First, $\text{Na}(\text{NO}_3)$ (Loba Chemie, India 99.9 % purity), Ammonium phosphate $(\text{NH}_4)_3\text{PO}_4$ (Loba Chemie, India 99.5 % purity) and $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ (Loba Chemie, India 99.5 % purity) in stoichiometric amounts were dissolved in distilled water. Sm_2O_3 (99.9 %, Sigma-Aldrich, USA) was dissolved in concentrated HNO_3 [65 %, Merck, Germany], to obtain samarium nitrate; which was then incorporated according to calculated doping levels. All solutions were prepared using distilled water. Other chemicals of AR grade were employed without additional purification for the preparation of samples. The relevant solution mixtures were stirred with heating at around 110°C , in order to achieve a gel-like reaction product. The product viscosity progressively increased until it transformed into a resinous gel in oven; finally, it was heated in muffle furnace at 700°C .

2.2. Characterization

The Centre of Advance Research, Dr. H.S. Gour, Vishwavidyalaya (a Central University), located in Sagar, Madhya Pradesh, India, was the location where the majority of the devices used for the analysis of the morphological, optical, and structural characteristics of the samples that had been prepared were situated. A wide variety of instrumentations were employed in this analysis. Fourier Transform Infrared (FTIR) spectra were acquired using a Bruker Alpha II spectrometer, which was made by Bruker (Germany), with the intention of determining the existence of functional groups. A UV-3092 spectrophotometer that was produced by LABINDIA, an Indian corporation, was used to get UV-visible diffuse reflectance spectra (DRS) so that the optical band gap could be determined. Pictures were obtained with the use of an FEI Tecnai G2 30 S-TWIN (FEI, USA) to assess particle morphology and size. This was done using transmission electron microscopy (TEM). A Shimadzu RF5301PC spectrofluorophotometer (Shimadzu, Japan) that was outfitted with a Xenon lamp, which acted as the source of excitation, was used to measure the spectra. An analysis was performed on the photoluminescence excitation (PLE) and emission (PL) spectra. The utilization of a Mettler Toledo MS204S analytical balance was necessary in order to achieve precise measurements of the samples. The production of this balance took place in Switzerland. A Memmert UFE 500 oven, which was produced in Germany, was employed for the purpose of drying. Every single piece of glassware and instrument was subjected to a rigorous cleaning process before to being utilized. The chromaticity coordinates of the sample were ascertained according to the CIE system by taking advantage of the Sylvania OSRAM Color calculator, which is renowned for its exceptional accuracy.

3. Results and Discussion

3.1. FTIR spectra

Using a Bruker Alpha II spectrometer (Bruker, Germany) in the Department of Chemistry at Dr. Harisingh Gour University Sagar, MP, India, the FTIR spectra of doped and undoped samples were acquired over the wave number range of $500\text{--}4000\text{ cm}^{-1}$. The temperature of the environment was at a standard level when the FTIR spectra were recorded.

The IR spectra of $\text{Na}_3\text{Bi}(\text{PO}_4)_2$: (0.0 and 0.7%) Sm^{3+} phosphor have been discussed here to analyse the functional groups. The infrared absorption spectra of the compounds are shown in Fig. 1. The vibrational peak at 611 cm^{-1} indicates the symmetric bending mode of PO_4 group. The peak detected around $745\text{ to }920\text{ cm}^{-1}$ are assigned to symmetric stretching of the P–O–P bonds in PO_4 group. The peak at 1000 cm^{-1} is assignable to asymmetric stretching of PO_4 group [17,18].

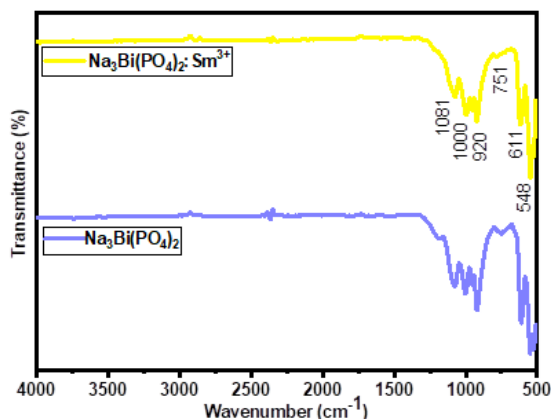


Fig. 1. FTIR spectra of $\text{Na}_3\text{Bi}(\text{PO}_4)_2$ phosphors and doped Sm^{3+} ion (0.7 mol%).

3.2. TEM study of $\text{Na}_3\text{Bi}(\text{PO}_4)_2: \text{Sm}^{3+}$ phosphor

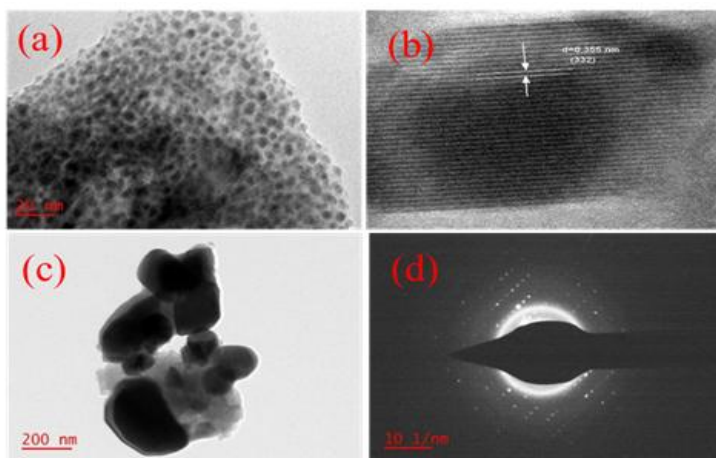


Fig. 2. (a, c) TEM image and (b) HR TEM, and (d) SAED pattern of $\text{Na}_3\text{Bi}(\text{PO}_4)_2: 0.5\text{mol}\% \text{Sm}^{3+}$ doped phosphor.

The TEM image of Na₃Bi(PO₄)₂:0.5 % Sm³⁺ doped phosphor were recorded on FEI Tecnai G2 30 S-TWIN (FEI, USA) at Centre of Advance Research, Dr. Harisingh Gour University Sagar, MP, India. The morphology of the one prepared doped material Na₃Bi(PO₄)₂:0.5 Sm³⁺ phosphor, was analysed by TEM (Fig. 2). The Fig. 2(a,c) represents TEM image; it reflects spongy nature and dumble shape particles [2,19]. The represents, the distance between the lattices have been calculated to be around 0.35 nm (Fig. 2b). The XRD findings indicated orthorhombic crystal system (hkl) value (332), for Na₃Bi(PO₄)₂: 0.5 % Sm³⁺ phosphor. Fig. 2(d) shows SAED pattern of Na₃Bi(PO₄)₂:0.5 Sm³⁺ doped phosphor.

3.3. UV-DRS spectra

Fig. 3 shows UV-DRS spectra of Na₃Bi(PO₄)₂: xSm³⁺ (x = 0.0 and 0.7 mol%) single-doped phosphors. The UV-DRS spectra have been recorded in the range of 200-500 nm wavelength. The observed peaks appeared in near UV region 200 to 300 nm. The Na₃Bi(PO₄)₂:Sm³⁺ single-doped phosphors show the optical properties in near UV-visible region [20]. Using UV-DRS spectra, the band gaps of the prepared samples, have been determined using Kubelka–Munk function 2:

$$(F(R)hv)^2 = C1(hv - Eg)^2(F(R)h\nu)^2 = C1(h\nu - Eg)^n \dots \dots \dots (1)$$

Where C1 is a proportionality, *hν* is the photon energy, *E_g* is the band gap energy and *n* is a constant associated with different type of electronic transitions. The optical band gaps Na₃Bi(PO₄)₂ phosphor has also been estimated corresponding to these transitions. The calculated band gap value is 4.37 eV.

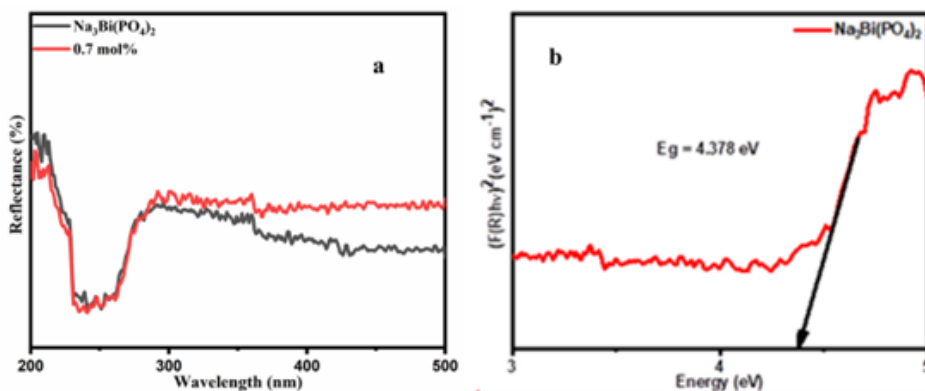


Fig. 3. (a) DRS spectra of undoped Na₃Bi(PO₄)₂ and Na₃Bi(PO₄)₂:0.7 mol% phosphor, (b) Na₃Bi(PO₄)₂ phosphor.

3.4. Photoluminescence properties of $\text{Na}_3\text{Bi}(\text{PO}_4)_2:\text{Sm}^{3+}$ phosphor

The PLE excitation spectrum of selected doped sample, has been analysed at room temperature conditions (Fig. 4). The $\text{Na}_3\text{Bi}(\text{PO}_4)_2: 0.7 \text{ Sm}^{3+}$ sample excited around 598 nm wavelength ($\lambda_{\text{em}} = 598 \text{ nm}$), gives several peaks at 344 nm (${}^6\text{H}_{5/2} \rightarrow {}^4\text{H}_{9/2}$), 361 nm (${}^6\text{H}_{5/2} \rightarrow {}^4\text{D}_{3/2}$), 375 nm (${}^6\text{H}_{5/2} \rightarrow {}^4\text{D}_{1/2}$), 404 nm (${}^6\text{H}_{5/2} \rightarrow {}^4\text{F}_{7/2}$) and 417 nm (${}^6\text{H}_{5/2} \rightarrow {}^6\text{P}_{5/2}$), 461 (${}^6\text{H}_{5/2} - {}^4\text{I}_{13/2}$), and 478 nm (${}^6\text{H}_{5/2} - {}^4\text{I}_{11/2}$). These are attributable to the f–f forbidden transitions of trivalent samarium ion. The emission intensity of the f–f transition at 403 nm is significant than other transitions; thus, this transition can be chosen for the emission spectra of $\text{Na}_3\text{Bi}(\text{PO}_4)_2: 0.7 \text{ Sm}^{3+}$ phosphor. The findings indicates that $\text{Na}_3\text{Bi}(\text{PO}_4)_2: 0.7 \text{ Sm}^{3+}$ phosphor may be excellent for near-UV excitation for white light emitting diodes [21,22].

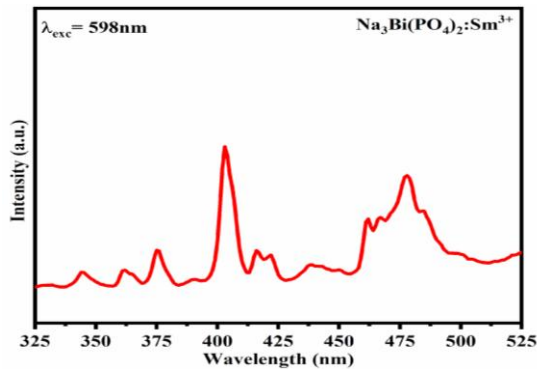


Fig. 4. PL excitation spectra $\text{Na}_3\text{Bi}(\text{PO}_4)_2:0.7 \text{ Sm}^{3+}$ phosphors monitored by 598 nm.

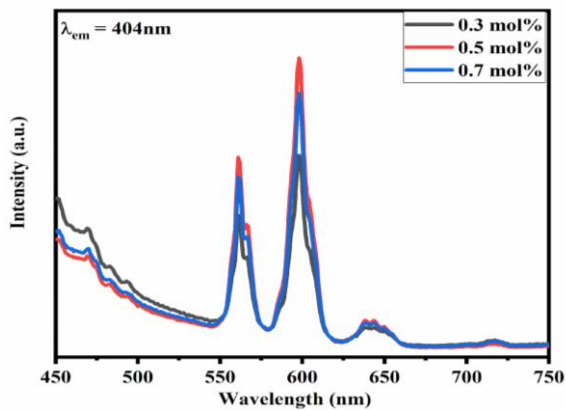


Fig. 5. PL emission spectra of Sm^{3+} (0.3, 0.5 and 0.7 mol%) activated $\text{Na}_3\text{Bi}(\text{PO}_4)_2$ phosphors.

Fig. 5. shows the emission spectra of Sm^{3+} doped $\text{Na}_3\text{Bi}(\text{PO}_4)_2$ phosphors excited at ($\lambda_{\text{exc}} = 404 \text{ nm}$). The results show that the whole emission spectrum from 550 nm to 625 nm is constituted of dominant peaks at 564 nm, 598 nm and 645nm, respectively; these are

ascribed to the ${}^4G_{5/2} \rightarrow {}^6H_{5/2}$ (green), ${}^4G_{5/2} \rightarrow {}^6H_{7/2}$ (orange) and ${}^4G_{5/2} \rightarrow {}^6H_{9/2}$ (red) transitions of Sm^{3+} ions. The weaker emission band appears at 645 nm. The intensity of the emission increases initially, reaching a maximum at 0.5 mole % Sm^{3+} ion concentration, and then drops as the concentration enhances further more. The distinctive emission bands of Sm^{3+} attributable to the ${}^4G_{5/2} \rightarrow {}^6H_{5/2}$ (564 nm) and ${}^4G_{5/2} \rightarrow {}^6H_{7/2}$ (598 nm) transitions have been observed, for this sample. Emission spectra of $\text{Na}_3\text{Bi}(\text{PO}_4)_2:\text{xSm}^{3+}$ for different Sm^{3+} concentrations: $x = 0.3, 0.5$ and 0.7% , have been recorded. The most prominent peak (intense) was observed at 598 nm as compared to the other emission peaks [23,24]. The peak at 564 nm has been ascribed to ${}^4G_{5/2} \rightarrow {}^6H_{5/2}$ emission transition due to magnetic dipole transition. Literature suggested that the emission peak at 598 nm (${}^4G_{5/2} \rightarrow {}^6H_{7/2}$) is due to partially magnetic dipole (MD) transition and partially in quality. Previous literature suggested that MD transition is less sensitive to the host environment. In this investigation concentration quenching has been observed up to 0.5 mol% Sm^{3+} ions concentration [5,25,26]. The ED transition is hypersensitive to the local environment of host lattice. When both MD and ED transition is present in the spectrum then Sm^{3+} ion occupies both symmetric and antisymmetric site in the host matrix [27].

3.5. CIE Diagram

Fig. 6 shows the CIE (Commission International de l'Eclairage – 1931) chromaticity diagrams. These were used to analyse coordinate values of emissions colour in $\text{Na}_3\text{Bi}(\text{PO}_4)_2: (0.5 \text{ mol}\%) \text{Sm}^{3+}$ excited with $\lambda_{\text{ex}} = 404 \text{ nm}$ for the dopant (calculated). CIE coordinate of $\text{Na}_3\text{Bi}(\text{PO}_4)_2: \text{xSm}^{3+}$ ($x = 0.5 \text{ mol}\%$) was found in the Near white color region, its coordinate was ($x = 0.2841, y = 0.3459$). The results indicated that $\text{Na}_3\text{Bi}(\text{PO}_4)_2:\text{Sm}^{3+}$ ($x = 0.5 \text{ mol}\%$) emits Near white light under 404 nm excitation [28,29].

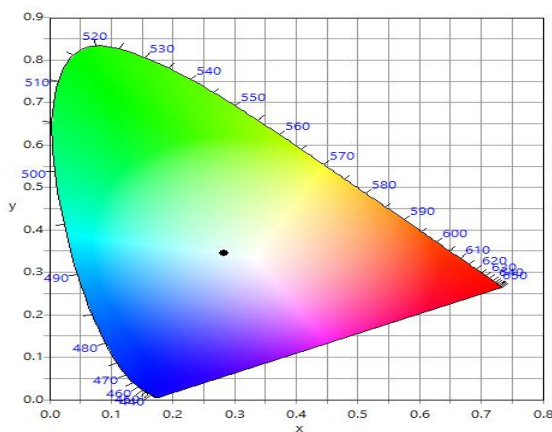


Fig. 6. Chromaticity coordinates of $\text{Na}_3\text{Bi}(\text{PO}_4)_2:0.5 \text{ Sm}^{3+}$ phosphor in the CIE 1931 chromaticity diagram.

4. Conclusion

By utilizing a wet chemical approach, phosphors were synthesized that had the chemical composition of Na₃Bi(PO₄)₂:xSm³⁺ (where x is equal to 0, 0.3, 0.5, or 0.7 mol%). The synthesized materials were studied by UV-DRS spectra, transmission electron microscopy (TEM), and Fourier-transform infrared spectroscopy (FT-IR). Studies of the PL were carried out as well. According to the findings of the FTIR research, the PO₄ group (P-O-P) and the Sm–O bonds are responsible for absorption. An absorption band with a broad range and a relatively low intensity absorption band that is typical in nature show up at around 920 cm⁻¹ and 548 cm⁻¹, respectively. In addition, the produced phosphors' morphology was investigated using transmission electron microscopy (TEM), with a size range of micrometers; this has an impact on the properties of PL as well. All of the results that were obtained demonstrate that Na₃Bi(PO₄)₂:xSm³⁺ (x = 0.3, 0.5, 0.7 mol%) phosphor has the potential to be used as a promising orange-red emitting material for converted near-ultraviolet wide-LEDs. When the concentration is increased, the PL intensity first rises and then falls when the concentration is raised even further (0.7 mol% Sm³⁺). The PL absorption spectra excitation lies at 404 nm. The CIE chromaticity coordinate of Na₃Bi(PO₄)₂:xSm³⁺ (x = 0.5 mol%) phosphor has been calculated to be x = 0.2841, y = 0.3459 located in the near white region. Hence, this may be considered as a favourable candidate in NUV white light-emitting diodes (WLED).

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