J Opt
https://doi.org/10.1007/s12596-023-01387-4

RESEARCH ARTICLE

Wet chemical synthesis and photoluminescence properties of $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphors for near UV-based w-LEDs

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Received: 12 February 2023 / Accepted: 26 August 2023

Abstract The $NaSrPO_4:Dy^{3+}$and $NaSrPO_4:Eu^{3+}$phosphors were synthesized with success via wet chemical synthesis. XRD and photoluminescence properties of the prepared phosphors were thoroughly investigated. The XRD was used to confirm the hexagonal phase with $P6_3/mmc$ (194) space group. The emission peaks of $NaSrPO_4:Dy^{3+}$ phosphor are observed at 484 and 574 nm under excitation at 350 nm. When $NaSrPO_4:Eu^{3+}$ phosphor was stimulated at 394 nm, the emission bands at 591 and 613 nm are found. The CIE coordinates indicate that the present phosphors have high colour purity. The results indicate that $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphors were blue-yellow and orange-red emitting under n-UV converting w-LEDs.

Keyword Wet chemical synthesis · XRD · Phosphor · Photoluminescence · w-LED

Introduction

It is widely acknowledged that the invention of w-LEDs in this century has resulted in a significant revolution in illumination techniques due to their excellent properties such as luminous quality, energy saving, excellent stability, high efficiency and environmental friendliness [1–4]. Blue, green and red phosphors have been studied for application in w-LEDs [5–8]. Phosphors are an important material in lighting technology and have received a lot of attention in phosphor converted w-LEDs [9, 10]. As a result, it is important to discover new white phosphors with enhanced brightness, which originate from a single phosphor.

The most popular technique for creating w-LEDs was developed by S. Nakamura et al. in 1997 [11], combining both the blue-based InGaN LED (light-emitting diode) chip and the yellow-emitting (yttrium aluminium garnet) $YAG:Ce^{3+}$ phosphors. A poor colour executing index (CRI, Ra 7000 K) brought due to the absence of a red component and significant thermal quenching are two shortcomings of the pc-w-LEDs previously discussed [12, 13]. Extraordinary rare earth-doped inorganic phosphors are entrancing and have been generally investigated through ongoing numerous years. In this particular circumstance, the uncommon superior properties of trivalent ions doped phosphate materials of the type $ABPO_4$, where A and B are monovalent and divalent cations, independently, have drawn a lot of interest [14]. To determine these issues, single-part white light-communicating phosphors have procured pervasiveness due to their high brilliance efficiency, assortment reproducibility and modest collecting costs [15, 16].

The Dy^{3+} ion has two primary emission groups: blue (470–500 nm) because of the magnetic dipole $^4F_{9/2} \rightarrow {}^6H_{15/2}$ transition and yellow (570–600 nm) connected with the touchy electric dipole $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transition [17, 18]. Orthophosphate is regarded as an important host for luminescent materials due to its excellent properties, which include a large band gap and high absorption of PO_4^{3-} in the n-UV region, moderate phonon energy, high chemical stability, exceptional optical damage threshold and low sintering temperature. Numerous investigations have been

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Published online: 16 September 2023

done on phosphate materials reported by $KBaPO_4$ [19], $BiPO_4$ [20], $KSrPO_4$ [21] and $Sr_3Bi(PO_4)_3$ [22]. The wet chemical method is easy to prepare and has low sintering temperature, low-cost methods, eco-friendly and energy saving [23]. In this work, $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphors were synthesized first time prepared by wet chemical synthesis. The XRD, concentration quenching, CIE chromaticity coordinates and photoluminescence properties of $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphor were all thoroughly investigated. Our results showed that $NaSrPO_4:Dy^{3+}$and $NaSrPO_4:Eu^{3+}$ phosphors were blue-yellow, orange-red emitting under n-UV excitation converting w-LEDs.

Experimental

A wet chemical method was used to synthesize the $NaSrPO_4:Dy^{3+}$and $NaSrPO_4:Eu^{3+}$ phosphors. The raw materials were prepared using sodium nitrate (99%, Loba), strontium nitrate (Extra pure98%, Loba), ammonium dihydrogen phosphate (99%, Loba), dysprosium oxide (99.9%, Loba) and europium oxide (99.9%, Loba). Analytical Reagent (AR) grade materials and chemicals are used. Firstly, the stoichiometric calculations were done and then required chemicals were weighed on weighing machine. Then, in a borosil beaker, required quantities of distilled water is taken. Dy_2O_3 and Eu_2O_3ions dopant were weighed and placed in a test tube, and the mixture of diluted (HNO_3) was heated and converted into $Dy(NO_3)_3$ and $Eu(NO_3)_3$ ions. All the samples were collected in a beaker with water and stirred for 30 min. All the sample became transparent, and after 10 h of heating at 100 °C in a Hot Air Oven, a powder product was obtained. All samples were finely ground using a pestle and mortar. Finally, the powder sample was placed in a furnace at 600 °C for three hours. We synthesized the required phosphors and cooled them to room temperature before using them for further characterization.

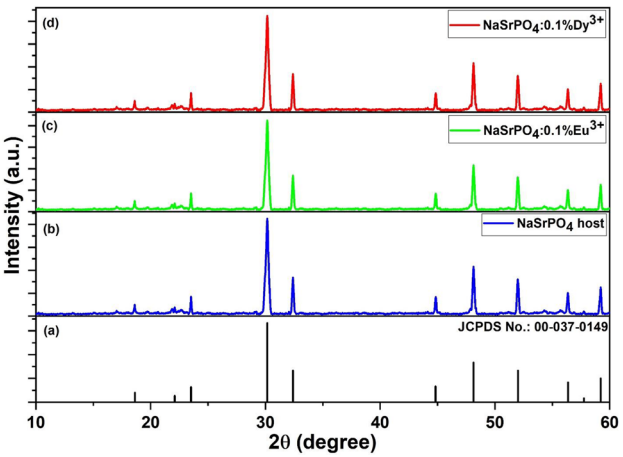

Fig. 1 XRD patterns of $NaSrPO_4$ phosphor

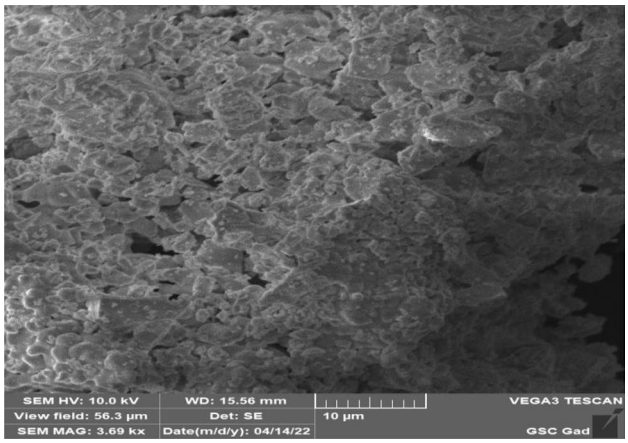

Fig. 2 SEM image of prepared $NaSrPO_4$ phosphor

Results and discussion

XRD pattern of $NaSrPO_4$ phosphor

The XRD was used to identify and confirmed the phase of the phosphor synthesized. Figure 1 (a) represents the JCPDS No. 00-037-0149 (b) $NaSrPO_4$ host (c, d) $NaSrPO_4$:0.1% Eu^{3+} and $NaSrPO_4$:0.1% Dy^{3+} phosphor. The XRD spectra very much matched the JCPDS No. 00-037-0149 pattern. The sample XRD spectra match the JCPDS data file, suggesting that the correct host lattice was synthesized using the wet chemical method. Based on this study, $NaSrPO_4$ phosphor with no impurity phases was produced. The XRD pattern was recorded with a step size of 0.02° in the range $10° \leq 2\theta \leq 60^{o}$ [24]. The XRD confirms hexagonal crystal structure has the space group $P6_3/mmc$ (194) [25].

SEM study of $NaSrPO_4$ phosphor

SEM studies have been carried to further analyse the surface morphology of $NaSrPO_4$ phosphor. Figure 2 shows the sample is SEM micrographs of phosphor showed the aggregation of crystals with the size of the range of 5–10 µm. The particles are irregular in size with rough margins. A closer examination of the SEM images showed high porosity on the surface of the phosphor [26, 27].

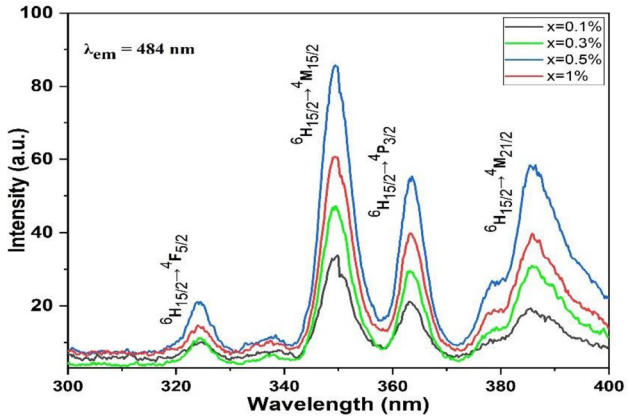

Fig. 3 Excitation bands of $NaSrPO_4:Dy^{3+}$ phosphors monitored at 484 nm emission

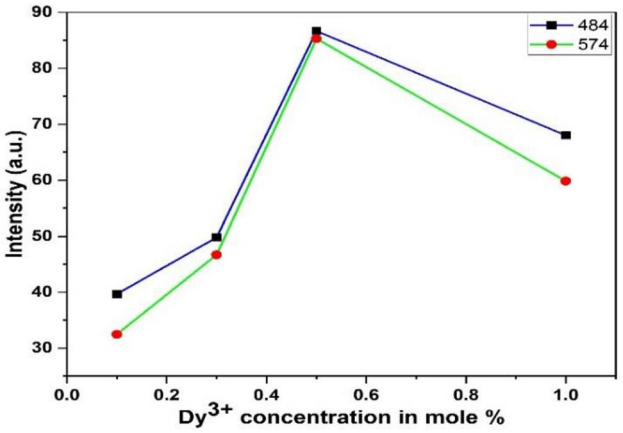

Fig. 5 Variation in 484 nm and 574 nm emission intensity as a function of $NaSrPO_4:Dy^{3+}$ phosphor

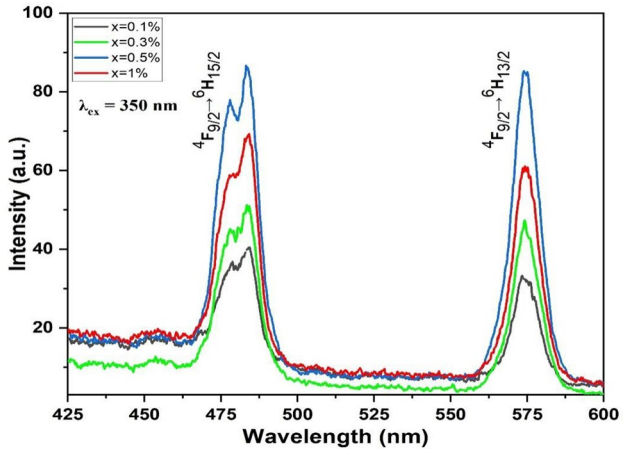

Fig. 4 Photoluminescence emission bands of $NaSrPO_4:Dy^{3+}$phosphor when excited at 350 nm

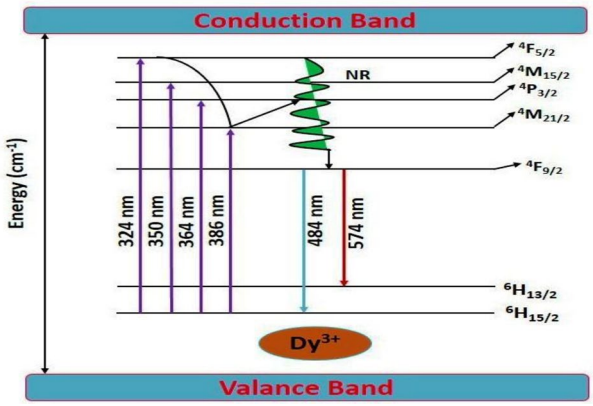

Fig. 6 Schematic energy level diagram of Dy^{3+} ion

Photoluminescence properties of $NaSrPO_4:Dy^{3+}$ phosphor

Figure 3 depicts the excitation spectra of $NaSrPO_4:xDy^{3+}$ ($x = 0.1$, 0.3, 0.5, and 1 mol%) phosphor in the wavelength range of 300–400 nm, with a monitored emission band at 484 nm. The photoluminescence excitation peaks are detected at 324 nm, 350 nm, 364 nm and 386 nm corresponding to the electronic transitions $^6H_{15/2} \rightarrow {}^4F_{5/2}$, $^6H_{15/2} \rightarrow {}^4M_{15/2}$, $^6H_{15/2} \rightarrow {}^4P_{3/2}$ and $^6H_{15/2} \rightarrow {}^4M_{21/2}$, respectively [28, 29]. The strong excitation peaks at 350, indicating that this phosphor is excellent for improving near-UV-based w-LEDs

Figure 4 shows the emission bands of $NaSrPO_4:Dy^{3+}$phosphor with 395 nm excitation wavelength. The two emission peaks are observed at 484 and 574 nm corresponding to the electronic transitions $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ of Dy^{3+} ions, respectively [30, 31]. The two emission peaks blue and yellow results in white light emission in this matrix. The relative emission intensity as a function of Dy^{3+} ions concentration is shown in Fig. 5. The emission band's luminous intensity first increases as the concentration of Dy^{3+} ions increases from 0.1 to 0.5 mol%, but then decrease. When x exceeds 0.5 mol% due to the concentration quenching effect. The inset depicts the fluctuation of the blue and yellow emission lines as a result of Dy^{3+} ions concentration [32].

Multiple electronic transitions of the $NaSrPO_4:Dy^{3+}$ phosphor are presented in the schematic energy level diagram in Fig. 6. The higher energy ($^4F_{5/2}$, $^4M_{15/2}$, $^4P_{3/2}$ and $^4M_{21/2}$) of the Dy^{3+} ions are occupied at different excitation wavelengths (324 nm, 350 nm, 364 nm and 386 nm). The ions relaxed to the exciting level's ground state ($^4F_{9/2}$) because the energy difference between the state was insufficient to allow for radiative transmission. Finally, it decays to two ground states ($^6H_{15/2}$ and $^6H_{13/2}$) to produce blue and yellow photons [33].

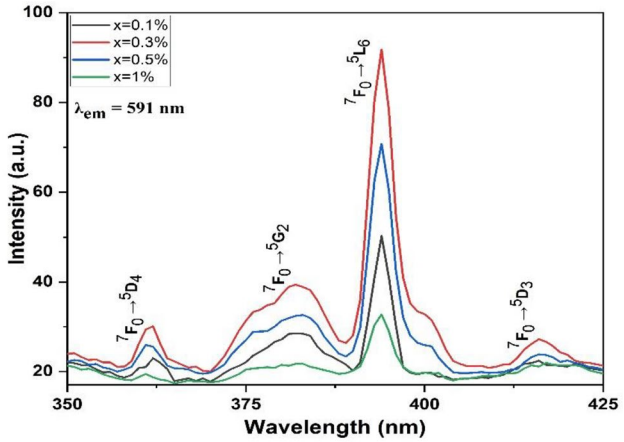

Fig. 7 Excitation bands of $NaSrPO_4:Eu^{3+}$ phosphor monitored at 591 nm emission

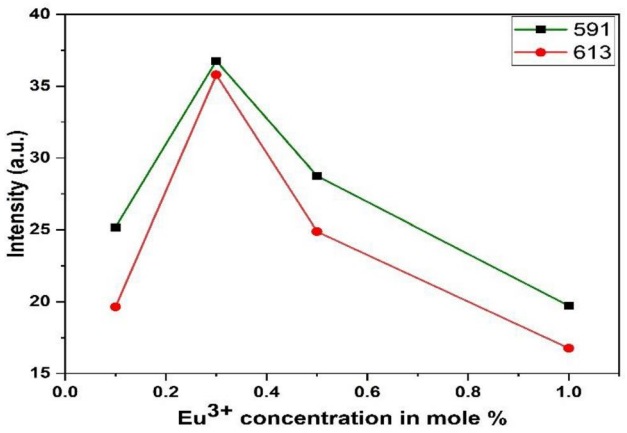

Fig. 9 Variation in 591 nm and 613 nm emission intensity as a function of $NaSrPO_4:Eu^{3+}$ phosphor

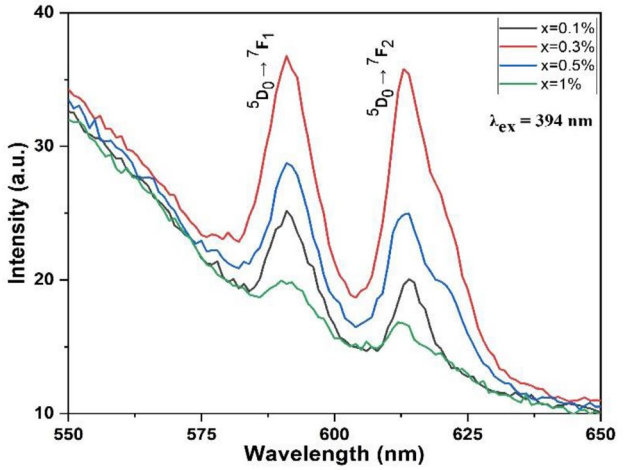

Fig. 8 Photoluminescence emission spectrum of $NaSrPO_4:Eu^{3+}$ when stimulated at 394 nm

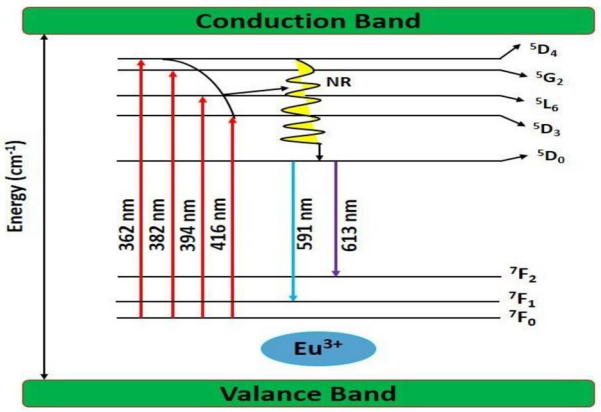

Fig. 10 Schematic energy level diagram of Eu^{3+} ion

Photoluminescence properties of $NaSrPO_4:Eu^{3+}$ phosphor

Figure 7 shows the excitation bands of $NaSrPO_4:Eu^{3+}$ phosphor in the wavelength range 350–425 nm, with a monitoring emission peak at 591 nm. The excitation peaks at 362, 382, 394and 416 nm corresponding to the transitions $^7F_0 \rightarrow {}^5D_4$, $^7F_0 \rightarrow {}^5G_2$, $^7F_0 \rightarrow {}^5L_6$ and $^7F_0 \rightarrow {}^5D_3$ of Eu^{3+} ions, respectively [34, 35]. The $^7F_0 \rightarrow {}^5L_6$ transition maximum excitation band at 394 nm. As a consequence, the $NaSrPO_4:Eu^{3+}$ phosphor performed n-UV-based w-LEDs

Figure 8 shows the emission spectra $NaSrPO_4:Eu^{3+}$ phosphor with wavelength range in the 550–650 nm, with an excitation band of 394 nm. The emission spectra of $NaSrPO_4:Eu^{3+}$ phosphor consist of two emission bands located at 591 nm (orange) and 613 nm (red) are attributed to the transitions $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$of the Eu^{3+} ions [36, 37]. As a result, the $NaSrPO_4:Eu^{3+}$ phosphor is proven excellent for usage in near-UV exciting w-LEDs. To determine the influence of Eu^{3+} ion concentrations on the photoluminescence emission intensity of $NaSrPO_4:Eu^{3+}$ phosphor as shown in Fig. 9. A series of were prepared samples with various Eu^{3+} ion concentrations ranging from 0.1 to 1 mol% while maintaining all other processing conditions constant. The luminescence emission intensity increases due to the concentration of Eu^{3+} ions, reaching a maximum at $x = 0.3$ mol%, and then decrease for further clearly showing that concentration quenching occurs [38, 39].

Figure 10 depicts the schematic energy level diagram of $NaSrPO_4:Eu^{3+}$ phosphor. Under excitation wavelength (362 nm, 382 nm, 394 nm and 416 nm), the electrons absorb energy and corresponding to the excited states ($^5D_4$, $^5G_2$, $^5L_6$ and $^5D_3$) from the ground state ($^7F_0$). Then, the electrons relax to the low $^5D_0$ energy level via a non-radiative

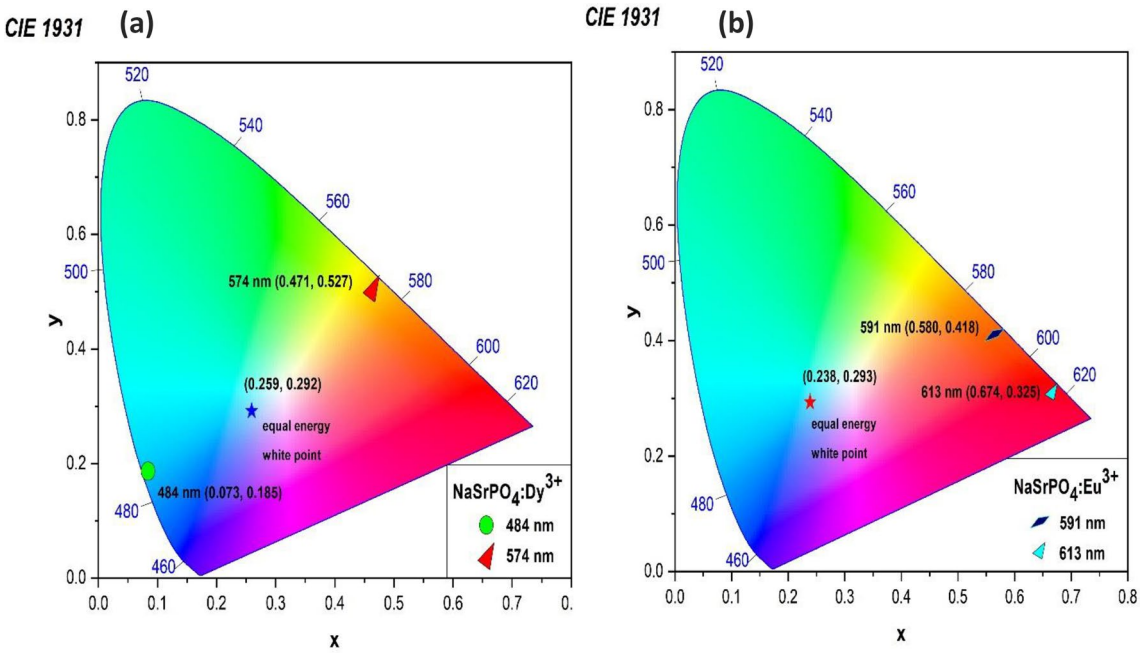

Fig. 11 **a**, **b** CIE diagram showing the CIE Coordinates of $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ Phosphors

transition. Then, it decays to two ground states ($^7F_1$ and $^7F_2$) that corresponding to the Eu^{3+} ions, by releasing rising emissions peak at 591 (orange) and 613 nm (red) [40]

CIE coordinates of $NaSrPO_4:Dy^{3+}$and $NaSrPO_4:Eu^{3+}$ phosphors

The chromaticity diagram produced by the CIE demonstrates the importance of coordinates in determining the precise emission colour purity of a sample. Figure 11a shows the CIE coordinates for $NaSrPO_4:Dy^{3+}$ phosphor were obtained using photoluminescence spectrum excitation peak at 350 nm. The $NaSrPO_4:Dy^{3+}$ phosphor has chromaticity coordinates (x, y) of (0.073, 0.185) and (0.471, 0.527), indicating it emits blue and yellow light [41, 42]. As a result, by connecting only two places, such as the $NaSrPO_4:Dy^{3+}$ phosphor, white emission with CIE coordinates ($x = 0.259$, $y = 0.292$) may be achieved. Figure 11b the $NaSrPO_4:Eu^{3+}$ phosphor has CIE coordinates of (0.580, 0.418) at 591 nm (orange) and (0.674, 0.325) at 613 nm (red) under excitation at 394 nm [43, 44]. As a result, by connecting only two places, such as the $NaSrPO_4:Eu^{3+}$ phosphor [45], white emission with CIE coordinates ($x = 0.238$, $y = 0.293$) may be achieved. The observed CIE coordinates suggest that the prepared phosphors have high colour purity. It also implies that $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphor were blue-yellow and orange-red emitting for n-UV based w-LEDs are possible.

Conclusion

A wet chemical method was used to successfully synthesize $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphors. The X-ray diffraction, photoluminescence properties and chromaticity property of were investigated systematically. The hexagonal phase was confirmed using the XRD. The $NaSrPO_4:Dy^{3+}$ phosphors emission bands at 484 and 574 nm under excitation peak at 350 nm. The two separate emission peaks corresponding to the Dy^{3+} ion $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions. The $NaSrPO_4:Eu^{3+}$ phosphor yields orange (591 nm) and red (614 nm) emissions, with excitations peak at 394 nm. The CIE Coordinates show that the current phosphors have high colour purity. As a result of its exceptional luminous qualities of $NaSrPO_4:Dy^{3+}$ and $NaSrPO_4:Eu^{3+}$ phosphor is a possible phosphor candidate for under near-UV converting w-LEDs.

Acknowledgements Author Mr. Chandrahasya M. Nandanwar is thankful for financial support from the MJPRF-2021, Nagpur (MS), India, fellowship (Outward No. MAHAJYOTI/Nag./Fellowship/2021-22/1042 (142) Dated 17/01/2022).

Data availability No data were used for the research described in the article.

Declarations

Conflict of interest The authors declare that they do not have any known competing financial interests or personal relationships that could appear to have influenced the work reported in this paper.

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