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RESEARCH ARTICLE

Photoluminescence properties of novel $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors for n-UV solid-state lighting prepared by combustion synthesis

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Abstract A series of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors were synthesized using a combustion method. The photoluminescence characteristics and X-ray diffraction of the produced phosphor were studied. X-ray diffraction was used to confirm the hexagonal crystal structure with the space group P63/m (176–1). The photoluminescence characteristics of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors measured under near-UV excitation at 350 nm showed two prominent peaks, blue and yellow emission at 470 nm and 575 nm, which were attributable to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions of Dy^{3+} ions, respectively. The optical dopant concentration for the Dy^{3+} ions was optimized to 0.5 mol%, after which concentration quenching occurred. The photoluminescence results suggest that $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor may be used as a possible material for near-UV-based solid-state lighting.

Keywords XRD · Photoluminescence · Combustion method · Phosphor · Solid state lighting

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Introduction

Phosphate compounds have gained lot of interest from researchers as a prospective host for rare-earth ions. These materials are a novel advancement of solid-state lighting systems that are developing as a substantial non-contaminated source of lighting. Lanthanide-doped phosphate compounds are used in a variety of applications, including solid-state lasers, display systems, w-LEDs, and eco-friendly materials [1–4]. Because of their high efficiency, long lifetime, energy storage, high brightness, good material stability, and fast response, w-LEDs [5–8] are the most likely solid-state lighting technology to replace traditional incandescent and fluorescent bulbs [9–12].

White light-emitting diodes have been regarded as a new generation of light source due to their superior advantages of compact size, energy savings, environmental protection, extended service life, and so on. LEDs have several interesting applications in the field of SSL and display devices owing to their inexpensive cost, high electro-optical conversion efficiency, stability, and reliability [13–16]. LEDs are typically manufactured using a mix of an LED chip and phosphor material. The commercial w-LED, which comprises a blue LED chip and a $Y_3Al_5O_{12}:Ce^{3+}$ yellow-emitting phosphor, cannot produce warm white light with a low color rendering index (CRI $^{>}$ 4500 K) [17–19]. As a result, w-LEDs made with near-ultraviolet LED chips and tricolor (red, green, and blue) phosphors will likely dominate the market in the near future due to their greater color uniformity, stronger CRI value, and outstanding white light lighting [20–23]. Orthophosphates having a generic formula are among them. $A_3RE(PO_4)_2$ (A = alkali metal ion, RE = rare-earth ions) are thought to be the most interesting new type of phosphor material for rare-earth activators because of the tetrahedral PO_4^{3-} group's ability to bond with

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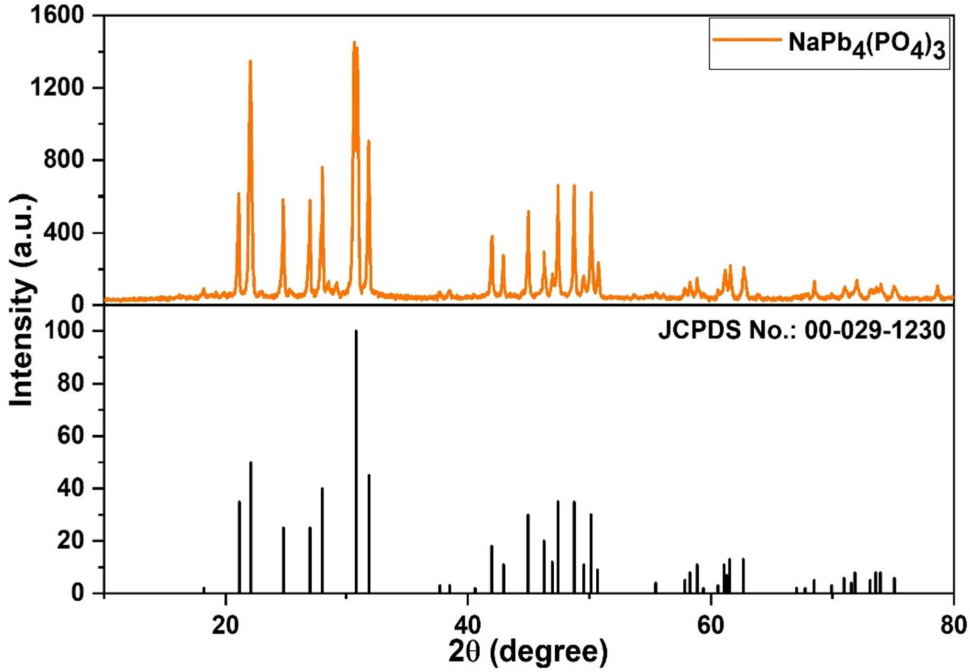


Fig. 1 XRD patterns of $NaPb_4(PO_4)_3$ phosphor

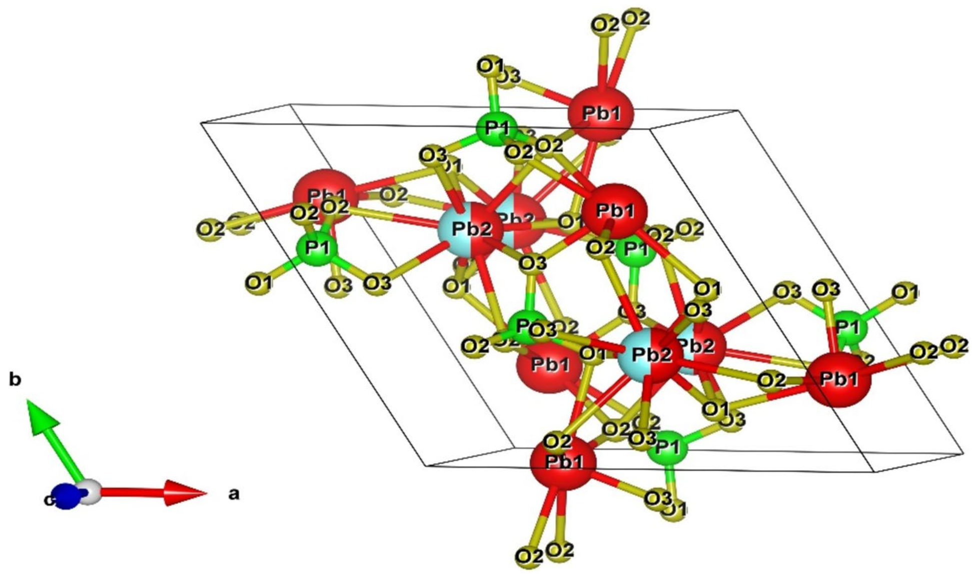


Fig. 2 Crystal structure of $NaPb_4(PO_4)_3$

supplementary structural units, chemical stability, ease of synthesis, low synthesis temperature, and many other properties [24–26].

Phosphate-based phosphors play an important role in luminescence due to their great qualities such as low cost, environmental friendliness, and superior electrochemical performance. The Dy^{3+} ion has two prominent emission bands in the blue (470–500 nm) and yellow (570–600 nm) areas, which correspond to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions, respectively [27, 28]. It is possible to achieve near-white light emission by changing the intensity ratio of yellow emission to blue emission. Previously, Yerojwar et al. [29] investigated Sm^{3+} ions-doped $La_{1.4}Al_{22.6}O_{36}$ phosphors by combustion technique and systematically evaluated their photoluminescence properties. By using a combustion process, Parshuramkar et al. [30] reported $CaMgB_2O_5$: RE^{3+} (RE = Dy, Eu, Sm) environmentally friendly phosphor. Maske et al. [31] also reported $SrAlBO_4$:RE^{3+} ($RE^{3+} = Dy^{3+}$, and Sm^{3+}) phosphor by combustion technique. To the best of our knowledge, the $NaPb_4(PO_4)_3$:Dy^{3+}phosphor was prepared for the first time by using the combustion process. Various

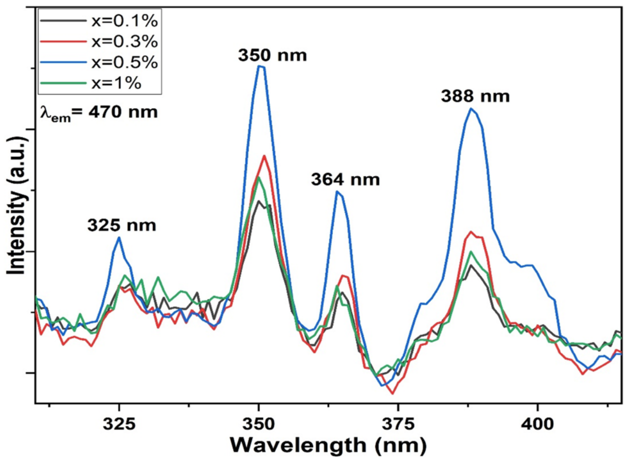


Fig. 3 Excitation spectrum of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor observed at 470 nm emission

phosphate-based phosphors like $Ca_{10}(PO_4)_6Cl_2$, $LaPO_4$, $LiSrPO_4$, $Ba_3(PO_4)_2$, $Sr3Y(PO_4)_3$, $NaCaPO_4$, etc., have been already studied [32–37]. Phosphate-based phosphors have excellent applications in white LEDs. In this paper, a series of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors were synthesized by combustion method and their X-ray diffraction and photoluminescence characteristics, concentration quenching, and CIE chromaticity diagram were thoroughly investigated. The results show that the $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor sample has great potential for n-UV-based solid-state lighting.

Experimental

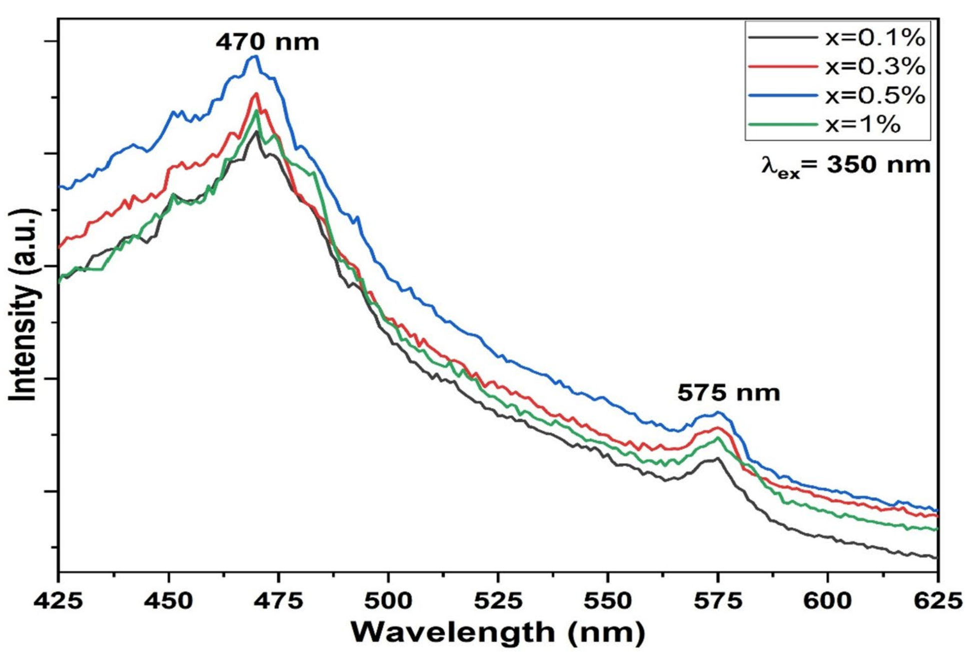
A series of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors were synthesized utilizing a combustion synthesis. The fuel was urea, and the oxidation-to-fuel ratio was set at 1:1 for complete combustion. The sample was prepared using sodium nitrate, lead nitrate, ammonium dihydrogen phosphate, dysprosium oxide, and urea. For the synthesis purpose, AR grade materials and chemicals were used. In this synthesis, materials were weighed in stoichiometric proportions and crushed for 30 min with a mortar and pestle. Dopants of Dy_2O_3 ions were then weighed and placed in a test tube, and the combination of diluted HNO_3 was heated and converted into $Dy(NO_3)_3$ ions. In a mortar, the solution was mixed with the dopant. The prepared clear solution was then transferred to a crucible; the furnace was kept at 550 °C and burnt with a flame, yielding abundant powders of components. This crystalline powder was crushed into a powder with a pestle and mortar. The powders were annealed for three hours at 600 °C. The sintered samples were removed from the furnace and allowed to achieve room temperature, then slightly crushed to fine powder. The powder samples were utilized for further characterization.

Results and discussion

XRD pattern of $NaPb_4(PO_4)_3$ phosphor

The X-ray diffraction data are shown in Fig. 1. All of the diffraction peaks in the samples were well indexed to the

Fig. 4 Emission spectra of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors ($\lambda_{ex}=350$ nm)



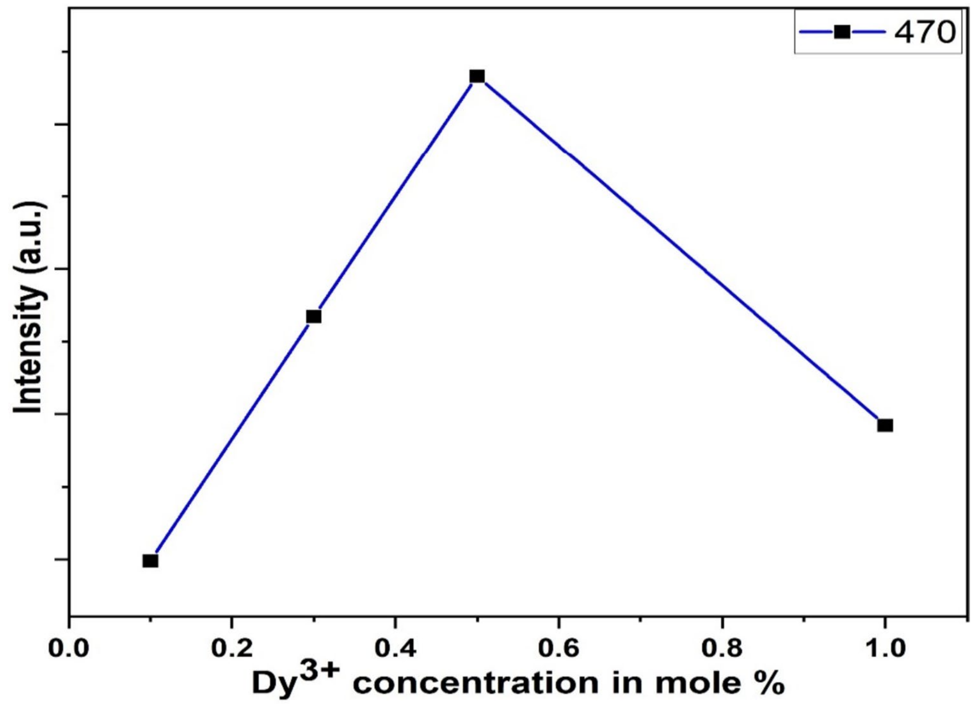


Fig. 5 Variation in 470 nm emission intensity as a function of Dy^{3+} ion concentration in $NaPb_4(PO_4)_3$ phosphor

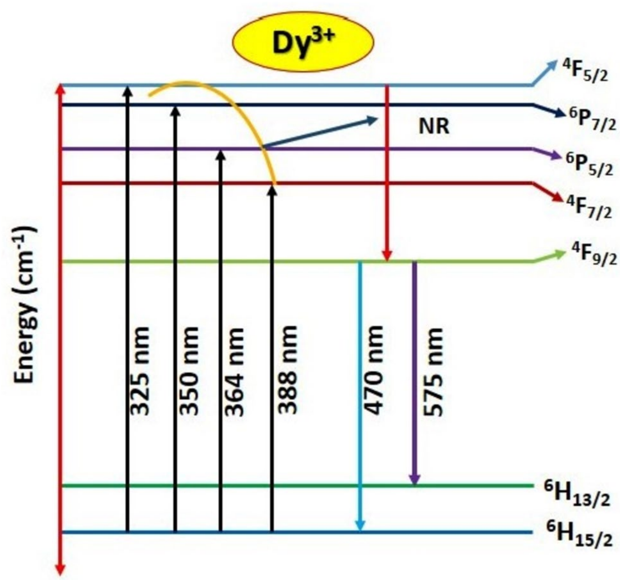


Fig. 6 Schematic energy level diagram of the Dy^{3+} ions

JCPDS 00-029-1230 standard card, and no impurity peak was observed in the experimental range. The X-ray diffraction pattern may be indexed in the hexagonal structure of $NaPb_4(PO_4)_3$ phosphor (sp. gr. P63/m) [38, 39].

The hexagonal crystal structure of $NaPb_4(PO_4)_3$ with a space group of P63/m (No. 176–1) was drawn using VESTA software. The lattice parameter values are as follows: $a=b=9.72490$ Å, $c=7.19000$ Å, $\alpha=\beta=90°$, $\gamma=120°$, $Z=2$ and $V=588.8841$ Å^3. The Pb (1) site has six oxygen neighbors, whereas the Pb (2) site has nine oxygen neighbors, and the Na (1)) sites coordinated nine oxygen atoms as neighbors [40] (Fig. 2).

Photoluminescence properties of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor

As shown in Fig. 3, the $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor showed wavelengths ranging from 310 to 410 nm. The excitation spectrum was detected at 470 nm emission wavelength. This photoluminescence excitation spectrum exhibits various excitation peaks at 325 nm, 350 nm, 364 nm, and 388 nm, which can be attributed to the transitions $^6H_{15/2}\rightarrow{}^4F_{5/2}$, $^6H_{15/2}\rightarrow{}^6P_{7/2}$, $^6H_{15/2}\rightarrow{}^6P_{5/2}$, and $^6H_{15/2}\rightarrow{}^4F_{7/2,}$ respectively [41, 42]. The strong excitation spectrum has a wavelength of 350 nm. As a result, these excitations may be used to generate photoluminescence emission spectra.

The photoluminescence emission spectra of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor in the spectral region 425–625 nm under 350 nm excitation are shown in Fig. 4. These emission bands of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor display two peaks at 470 nm and 575 nm, which correspond to

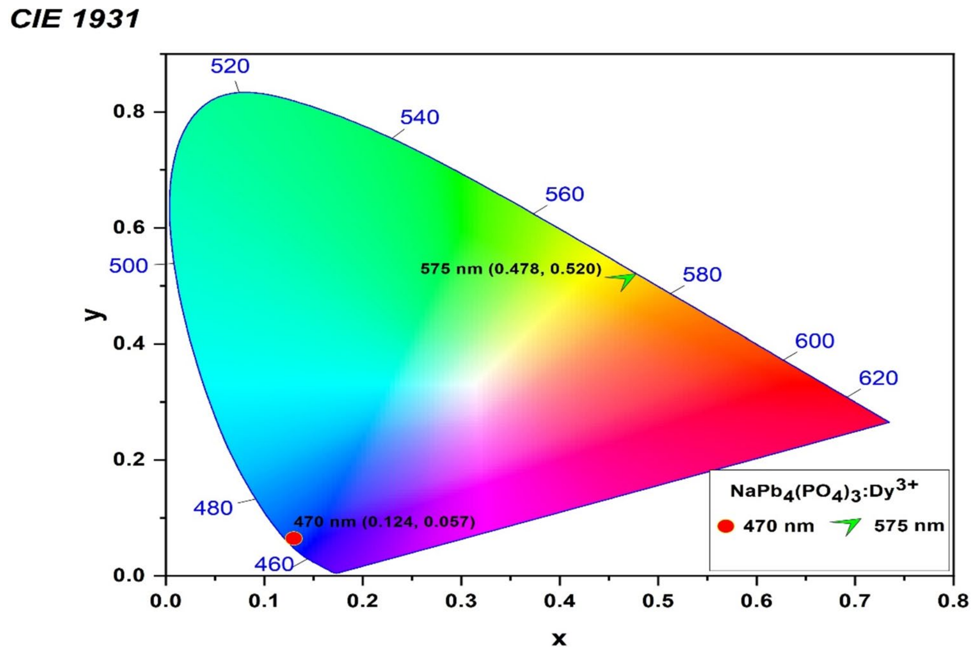


Fig. 7 CIE color coordinates of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor

the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$, respectively [43, 44]. Both emission bands at 470 nm (blue) and 575 nm (yellow) correspond to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ owing to magnetic and electric dipole transitions [45].

To investigate the effect of varied Dy^{3+} ion concentrations on luminescence intensity. Figure 5 depicts the emission spectra of phosphor materials with varying Dy^{3+} ion concentrations. As can be observed, there was no obvious change in the locations of the emission peaks for all samples when excited at 350 nm; nevertheless, the strength of the emission gradually varied with increasing concentrations of Dy^{3+} ion. The image shows that the emission intensity first increased and reached a maximum at 0.5 mol%, which was chosen as the concentration, and subsequently decreased due to the typical concentration quenching effect [46, 47].

The energy level diagram of $NaPb_4(PO_4)_3:Dy^{3+}$phosphor with some typical excitation and emission transitions according to the determined data of energy levels by Carnall, etc. [48, 49] is presented in Fig. 6. The excited Dy^{3+} ions in the $^6P_{7/2}$ state relaxed to the $^4F_{9/2}$ state by cascade non-radiative relaxation. Finally, the Dy^{3+} ions in the $^4F_{9/2}$ level relaxed radiatively to the $^6H_{15/2}$ and $^6H_{13/2}$ states, resulting in increased blue (470 nm) and yellow (575 nm) emissions, respectively.

CIE coordinates of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor

The CIE diagram is used to determine the color emission of synthesized phosphors. Based on the Commission Internationale de L'Eclairage (CIE) functions [50, 51], the color of any light may be represented as a (x, y) coordinate. The phosphor samples have been calculated using the emission spectra, and the results are shown in Fig. 7. The $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor's CIE chromaticity coordinates for blue and yellow emission are ($x = 0.124$, $y = 0.057$) and ($x = 0.478$, $y = 0.520$), respectively [52]. The current findings show that $NaPb_4(PO_4)_3:Dy^{3+}$ phosphors produced via combustion synthesis might emit blue and yellow emissions when activated by an n-UV, making them one of the best possibilities for white LED applications.

Conclusion

The combustion method was used to successfully produce varied concentrations of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor, and their photoluminescence characteristics and phase purity were investigated. The phase purity of the synthesized phosphors has been verified by XRD analysis. Under n-UV excitation, the photoluminescence emission peaks at 470 and 575 nm. The blue emission band at 470 nm and yellow emission band at 575 nm are attributed to $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions of Dy^{3+} ions, respectively. The strongest emission intensity of $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor was measured at 0.5 mol% concentration. According to the photoluminescence results, the $NaPb_4(PO_4)_3:Dy^{3+}$ phosphor might be useful in the fields of near UV-excited solid-state lighting applications.

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Data availability Data will be made available on request.

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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