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# A single-phased warm-white-emitting $K_3Y(PO_4)_2$ :Dy<sup>3+</sup>,Sm<sup>3+</sup> phosphor with tuneable photoluminescence for near-UV-excited white LEDs



PIGMENTS

Balaji Devakumar<sup>a</sup>, Heng Guo<sup>a</sup>, Yu-Jia Zeng<sup>b</sup>, Xiaoyong Huang<sup>a,\*</sup>

<sup>a</sup> Key Lab of Advanced Transducers and Intelligent Control System, Ministry of Education and Shanxi Province, College of Physics and Optoelectronics, Taiyuan University of Technology, Taiyuan 030024, PR China

<sup>b</sup> Shenzhen Key Laboratory of Laser Engineering, College of Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, PR China

ARTICLE INFO	A B S T R A C T		
Keywords:	Single-phased warm-white-emitting $Dy^{3+}/Sm^{3+}$ codoped $K_3Y(PO_4)_2$ phosphors with tuneable emissions,		
Photoluminescence	tuneable color tone and correlated color temperature (CCT) were synthesized through conventional high tem-		
WLEDs	perature solid state method. The crystal structure, morphology, and photoluminescence properties of the		
Crystal structure	phosphors were investigated in detail. Under 351 nm excitation, the $K_{2}(PO_{4}) \sim 0.03 Dv^{3+} 0.01 Sm^{3+}$ sample		
Phosphors	emitted warm-white light with CIF coordinates of (0.433, 0.406) low CCT (308.8K) and excellent color ren-		
Rare earth ions White emissions	dering index of 95.31. The energy transfer mechanism from the Dy <sup>3+</sup> to Sm <sup>3+</sup> was determined to be electric dipole_dipole interaction. The results indicate that the Dy <sup>3+</sup> (Sm <sup>3+</sup> codoped K <sub>2</sub> V(PO <sub>2</sub> ) <sub>2</sub> phosphors will find their		
	suitable applications in the near-ultraviolet-pumped white light-emitting diodes.		

#### 1. Introduction

The present world is moving towards the scarcity of energy, which is resolved by the development of modern solid-state lighting (SSL) and the photovoltaic devices [1–7]. Nowadays, white light-emitting diodes (LEDs) are playing a major role in the SSL and displays. In particular, rare-earth (RE) ions doped luminescent materials mixed with blue and UV LEDs are fabricated to produce white light [8–14]. Unfortunately, the white light produced by the above does not meet the requirements such as high luminous efficiency, excellent color rendering index (CRI), good thermal and chromatic stability [15–17]. Hence, it is necessary to find the suitable luminescent materials that should meet the abovementioned conditions.

Many researchers believe the single component white light emitting phosphors excited by the blue and UV LEDs will find their application in the SSL. The energy transfer from the sensitizer to the activator is one of the main processes for generating white light in single phase compounds [1,18]. As an evidence of energy transfer method, a number of phosphors such as  $Y_2SiO_5:Ce^{3+},Tb^{3+},Eu^{3+}$  [19], LiGd(WO<sub>4</sub>)<sub>2</sub>:RE<sup>3+</sup> (RE = Tm, Tb, Dy, Eu) [20], NaGd(WO<sub>4</sub>)<sub>2</sub>:Tm<sup>3+</sup>,Dy<sup>3+</sup>,Eu<sup>3+</sup> [21], CaNb<sub>2</sub>O<sub>6</sub>:RE<sup>3+</sup> (RE<sup>3+</sup> = Eu<sup>3+</sup>/Tb<sup>3+</sup>/Dy<sup>3+</sup>/Sm<sup>3+</sup>) [22], LaB-WO<sub>6</sub>:Tb<sup>3+</sup>,Eu<sup>3+</sup> [23], and Ba<sub>2</sub>Y(BO<sub>3</sub>)<sub>2</sub>Cl:Bi<sup>3+</sup>,Eu<sup>3+</sup> [24] have been prepared and investigated. Tuneable, multicolor and white emissions were obtained in these single-phased compounds. Still, the energy transfer was not being investigated in many hosts.

Among phosphates, alkali trivalent RE double phosphate with general formula  $M_3^{I}RE^{III}(PO_4)_2$  (M = K, Na; RE = Y, La-Nd, Sm-Lu) have been studied widely for their excellent photoluminescence properties with good thermal and chemical stability [25]. Recently, Eu<sup>3+</sup>, Tb<sup>3+</sup>, Sm<sup>3+</sup> and Ce<sup>3+</sup> ions doped K<sub>3</sub>Gd(PO<sub>4</sub>)<sub>2</sub> phosphors [26–29] and tuneable emission from Eu<sup>3+</sup>, Tb<sup>3+</sup>, Dy<sup>3+</sup> and Sm<sup>3+</sup> in Na<sub>3</sub>Bi(PO<sub>4</sub>)<sub>2</sub> were reported recently and these phosphors were found to have good luminescence intensity with excellent thermal stability [30–32]. However, the photoluminescence properties of Dy<sup>3+</sup> and Sm<sup>3+</sup> codoped K<sub>3</sub>Y(PO<sub>4</sub>)<sub>2</sub> (KYP) phosphors were not reported until now. The Dy<sup>3+</sup> and Sm<sup>3+</sup> ions were chosen because white emission could be obtained by adjusting the blue/yellow intensity in the Dy<sup>3+</sup> ions, and moreover, warm white could be obtained if Sm<sup>3+</sup> was codoped with Dy<sup>3+</sup> ions [33–35].

In this present work,  $Dy^{3+}$  and  $Sm^{3+}$  ions singly-doped and codoped  $K_3Y(PO_4)_2$  (KYP) phosphors were prepared using traditional solid state method. Crystal structure, morphology, luminescence properties, decay lifetimes, energy-transfer were investigated in detail. The energy transfer from  $Dy^{3+}$  to  $Sm^{3+}$  in the KYP host has been demonstrated to be the dipole-dipole interaction. Under 351 nm excitation,  $Dy^{3+}/Sm^{3+}$  codoped  $K_3Y(PO_4)_2$  phosphors showed tuneable emissions. Importantly, the KYP:0.03Dy<sup>3+</sup>,0.01 Sm<sup>3+</sup> phosphor sample gave rise to warm white emission with CIE coordinates of (0.433, 0.406), low correlated color temperature (CCT = 3088 K) and excellent CRI of 95.31. Therefore, the KYP:Dy<sup>3+</sup>,Sm<sup>3+</sup> phosphors would have

\* Corresponding author.

E-mail address: huangxy04@126.com (X. Huang).

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promising application in the UV-excited WLEDs.

#### 2. Experimental

The powder samples of KYP:xDy<sup>3+</sup> (x = 0.01-0.05), KYP:xSm<sup>3+</sup> (x = 0.01-0.05) and KYP:0.03Dy<sup>3+</sup>,xSm<sup>3+</sup> (x = 0.005-0.03) were prepared using conventional high temperature solid-state reaction method. K<sub>2</sub>CO<sub>3</sub> (99.0%), Y<sub>2</sub>O<sub>3</sub> (99.99%), Dy<sub>2</sub>O<sub>3</sub> (99.99%), Sm<sub>2</sub>O<sub>3</sub> (99.99%) and (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> (99.0%) were weighed in stoichiometric ratio and ground thoroughly in the agate mortar. The mixture was annealed at 950 °C for 6 h in air to obtain the phosphor samples. The final products were obtained by cooling down to room temperature in the furnace.

The phase identification of the obtained phosphors was characterized using Bruker D8 Advance with Cu K $\alpha$  radiation (1.5406 Å) operated at 40 kV and 40 mA. Structure refinements were performed using Fullprof. Surface morphology and elemental mapping of the obtained phosphors were measured using field-emission scanning electron microscopy system (FE-SEM, MAIA3 TESCAN). The photoluminescence emission and excitation were investigated using an Edinburgh FS5 spectrometer with a 150 W continuous-wave Xenon lamp as light source. Decay curves were recorded in Edinburgh FS5 spectrometer with a pulsed Xenon lamp as light source. The temperature-dependent emission spectra of the studied samples in the temperature range of 303–483 K were measured by using Edinburgh FS5 and the temperature was adjusted by a home-made temperature controlling system.

#### 3. Results and discussion

The powder XRD patterns of the KYP:xDy<sup>3+</sup> and KYP:xSm<sup>3+</sup> (x = 0.01-0.05) phosphors were recorded from 5 to 60° with a step rate of 0.02° at room temperature (Fig. 1(a–c)). The diffraction patterns were in good agreement with the standard JCPDS file no.049–1085. The sharp and single diffraction peak indicated the formation of single phase compound. No peaks of other phases were observed in the experimental range, showing the substitution of Dy<sup>3+</sup> and Sm<sup>3+</sup> ions did not influence the phase and structure of KYP.

To obtain the detailed crystal structure information for the obtained phosphors, Rietveld refinement was performed by using Fullprof program. The structural refinement results of KYP, KYP:0.03Dy<sup>3+</sup> and KYP:0.03Sm<sup>3+</sup> were illustrated in Fig. 2(a–c) and the refined crystal lographic parameters were presented in Table 1. The results indicated the good agreement between the observed and calculated XRD patterns. The crystal structure of KYP (Fig. 3) was modelled using the Vesta software on the basis atomic coordinates refinements in Table S1 [36]. The KYP consisted of PO<sub>4</sub> tetrahedra and YO<sub>7</sub> monocapped prism,



Fig. 1. Powder X-ray diffraction patterns of (a) KYP, (b) KYP:0.03Dy  $^{3+}$ , and (c) KYP:0.03 Sm  $^{3+}.$ 



Fig. 2. Rietveld refinement patterns of (a) KYP, (b) KYP:0.03Dy  $^{3+},$  and (c) KYP:0.03  $\rm Sm^{3+}.$ 

Table 1Refined Crystallographic parameters of KYP, KYP:0.03Dy<sup>3+</sup> and KYP:0.03 Sm<sup>3+</sup>.

Compounds	КҮР	KYP:0.03Dy <sup>3+</sup>	KYP:0.03 Sm <sup>3+</sup>
Crystal System Space Group	Monoclinic $P2_1/m$ (No. 11)	Monoclinic $P2_1/m$ (No. 11)	Monoclinic <i>P2<sub>1</sub>/m</i> (No. 11)
Lattice Parameters			
а	7.38258 (13) Å	7.38838 (14) Å	7.3878 (3) Å
b	5.60588 (16) Å	5.6095 (3) Å	5.6089 (3) Å
с	9.3502 (3)Å	9.3582 (6) Å	9.3583 (5) Å
Unit Cell Volume V	386.919 (18) Å <sup>3</sup>	387.80 (3) Å <sup>3</sup>	387.74 (3) Å <sup>3</sup>
β	90.903 (2) °	90.918 (5)°	90.886 (5) °
Rp %	3.69	6.56	3.90
R <sub>wp</sub> %	5.92	9.73	6.76

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