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Effect of Bi^{3+} on fluorescence properties of $\text{YPO}_4:\text{Dy}^{3+}$ phosphors synthesized by a modified chemical co-precipitation method

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ABSTRACT

$\text{Y}_{0.99-x}\text{PO}_4:0.01\text{Dy}^{3+}, x\text{Bi}^{3+}$ ($x=0, 0.01, 0.05, 0.10, 0.15, 0.20$ and 0.25) phosphors have been synthesized by a modified chemical co-precipitation method using urea as a pH value regulator. The samples were characterized by X-ray powder diffraction (XRD) and photoluminescence spectroscopy. XRD results show that the samples have only single tetragonal structure when $x \leq 0.15$, but extraneous BiPO_4 phase appears besides major tetragonal phase when $x \geq 0.20$. The crystallinity of the samples is found to improve with increasing Bi^{3+} ion concentration from 0 to 15 mol%, and then decreased for higher concentrations associated with increasing BiPO_4 phase. Photoluminescence excitation spectra results show that the phosphor can be efficiently excited by ultraviolet light from 250 to 400 nm including four peaks at 294, 326, 352 and 365 nm. Emission spectra exhibit strong blue emission (483 nm) and another strong yellow emission (574 nm). When the Bi^{3+} ion concentration is 1 mol%, the intensity of excitation and emission spectra increased evidently. In addition, the yellow-to-blue emission intensity ratio (I_Y/I_B) is strongly related to the excitation wavelength and not to the Bi^{3+} ion concentration.

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1. Introduction

Yttrium orthophosphate is an important host material because of its excellent chemical stabilities, long-wavelength excitation properties, unique electronic structure and numerous transition modes including the 4f shell of Ln^{3+} [1]. Dy^{3+} -doped YPO_4 has been used as a potential white phosphor due to its high absorption and luminescence efficiencies in ultraviolet (UV) region. Its emission color of the luminescence is close to white owing to the blue (${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}$) and yellow (${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$) emissions of Dy^{3+} [2–4].

The luminescence intensity is an important character for phosphor; improved performance of lighting and display requires high quality of phosphors for sufficient brightness. The luminescence properties depend on the activator concentration and crystallinity; so many people have researched the influences of doping content of activator ion, grain size, morphology and agglomeration on luminescence efficiency [5–7]. Further, studies have been done on the influence of Bi^{3+} on fluorescence properties of rare earth vanadate phosphor such as $\text{YVO}_4:\text{Eu}^{3+}$ [6–10], $\text{LaVO}_4:\text{Eu}^{3+}$ [11], $\text{GdVO}_4:\text{Eu}^{3+}$ [11], $\text{YVO}_4:\text{Sm}^{3+}$ [12], etc., because Bi^{3+} can absorb more UV light owing to its $6s^2 \rightarrow 6s6p$ excitations [13] and the charge transfer transition from Bi^{3+} 6s to V^{5+} 3d and then the energy is transferred to Dy^{3+} [8]. However, little attention has been devoted to the effect of Bi^{3+} on photoluminescence of $\text{YPO}_4:\text{Dy}^{3+}$ phosphors. Additionally, previous research has neglected the influence of excitation wavelength on the yellow-to-blue emission

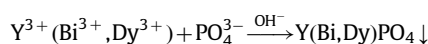
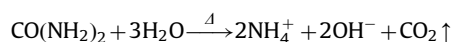
intensity ratio and color temperature of emission of $\text{YPO}_4:\text{Dy}^{3+}$ phosphors, which was studied in our work.

In this paper, $\text{Y}_{0.99-x}\text{PO}_4:0.01\text{Dy}^{3+}, x\text{Bi}^{3+}$ phosphors were synthesized by a modified chemical co-precipitation method and the fluorescence properties induced by various UV excitations were studied. It needs to be emphasized that the concentrations of Dy^{3+} , Bi^{3+} or other ions are those in the starting material and not in the final synthesized powder.

2. Experimental

2.1. Sample synthesis

It is well known that Y^{3+} (Bi^{3+} , Dy^{3+}) and PO_4^{3-} can coexist in a strong acid solution in the form of ionic state and no deposition appears, which means pH value of solution is the crucial condition. Urea is soluble in water and can be decomposed into CO_2 , NH_4^+ and OH^- when the solution is heated at a high temperature. So urea can be used as a pH value regulator in a chemical co-precipitation process. The mechanism is presented by the following reaction equations:



In this paper, $\text{Y}_{0.99-x}\text{PO}_4:0.01\text{Dy}^{3+}, x\text{Bi}^{3+}$ ($x=0, 0.01, 0.05, 0.10, 0.15, 0.20$ and 0.25) phosphors were synthesized by a modified chemical co-precipitation method, using Y_2O_3 (99.99%),

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Dy₂O₃ (99.99%), Bi₂O₃ (99.99%) and (NH₄)₂HPO₄ (analytical reagent) as starting materials and using CO(NH₂)₂ (analytical reagent) as a pH value regulator. The specific procedure is described as follows. Firstly, stoichiometric amounts of Y₂O₃, Dy₂O₃, Bi₂O₃, (NH₄)₂HPO₄ and an appropriate amount of CO(NH₂)₂ were dissolved into concentrated nitric acid including distilled water. Subsequently, the solution was heated at 85 °C until its pH value reached 7–8; the precursor was attained after the course of filtering, washing and drying. Finally, the phosphor was obtained when the precursor was heat-treated at 800 ° for 4 h.

2.2. Sample characterization

All the samples were characterized by an X-ray diffractometer (Cu K_α, λ=1.5406 Å, XRD-7000, SHIMADZU). The excitation and emission spectra were measured by a spectrophotometer (entrance slit width is 3 nm, receiving slit width is 3 nm, RF-5301PC, SHIMADZU). All the measurements were performed at room temperature.

3. Results and discussion

3.1. Crystal structural

Fig. 1(a) shows the XRD pattern of Y_{0.99-x}PO₄:0.01Dy³⁺, xBi³⁺ (x=0, 0.01, 0.05, 0.10, 0.15, 0.20 and 0.25) phosphor and the standard JCPDS card of YPO₄ (No. 11-0254). Fig. 1(b) shows the XRD pattern comparison of Y_{0.99-x}PO₄:0.01Dy³⁺, xBi³⁺ (x=0.15, 0.20 and 0.25) around 2theta of 19.5°. Fig. 1(c) shows the XRD pattern comparison of Y_{0.99-x}PO₄:0.01Dy³⁺, xBi³⁺ (x=0.05, 0.15 and 0.25) around 2theta of 25.95°.

It can be seen from Fig. 1(a) and (b) that when x ≤ 0.15, the observed diffraction peaks are well-matched with the theoretical

data of YPO₄ and the XRD pattern of every sample is successfully consistent with the tetragonal zircon structure known from bulk YPO₄ without any extraneous phases. Further, it can be observed from Fig. 1(c) and (b) that the diffraction peak positions are slightly shifted towards lower angle direction with increasing Bi³⁺ ion concentration from 5 to 25 mol%. It is attributed to lattice augmentation owing to the substitution of smaller sized Y³⁺ (ionic radius=0.090 nm) sites by the larger sized Bi³⁺ (ionic radius=0.096 nm) in the Y_{0.99-x}PO₄:0.01Dy³⁺ phosphor. It indicates that Dy³⁺ and Bi³⁺ ions are substituted into the Y³⁺ sites without the formation of any additional phase at lower concentrations. However, for Bi³⁺ ion concentration beyond 15 mol%, additional peaks (represented by ‘♦’ mark) relating to BiPO₄ phase (JCPDS No. 43-0637) are observed, but the major phase still has tetragonal zircon structure. Moreover, the crystallinity of the samples is found to improve with increasing Bi³⁺ ion concentration from 0 to 15 mol%, and then decreases for higher concentrations associated with increasing BiPO₄ phase.

In addition, the XRD results also indicate that this modified chemical co-precipitation method can be applied to prepare YPO₄-based phosphors and the pure single-phase YPO₄:Dy³⁺ phosphors can be developed at lower heat treatment temperature through our preparation procedure.

3.2. Luminescence properties

Fig. 2(a) shows the excitation spectra of Y_{0.99}PO₄:0.01Dy³⁺ phosphors via monitoring of 574 and 483 nm emissions. It can be seen that there is a broad strong absorption band ranging from 250 to 330 nm, which includes a strong peak at 294 nm and another peak at 326 nm. It is noteworthy that the broad excitation band around 294 nm cannot be attributed to host band absorption and the charge transfer band (CTB) for Dy³⁺ because the bandgap energy of YPO₄ is around 144 nm [14–17] and the

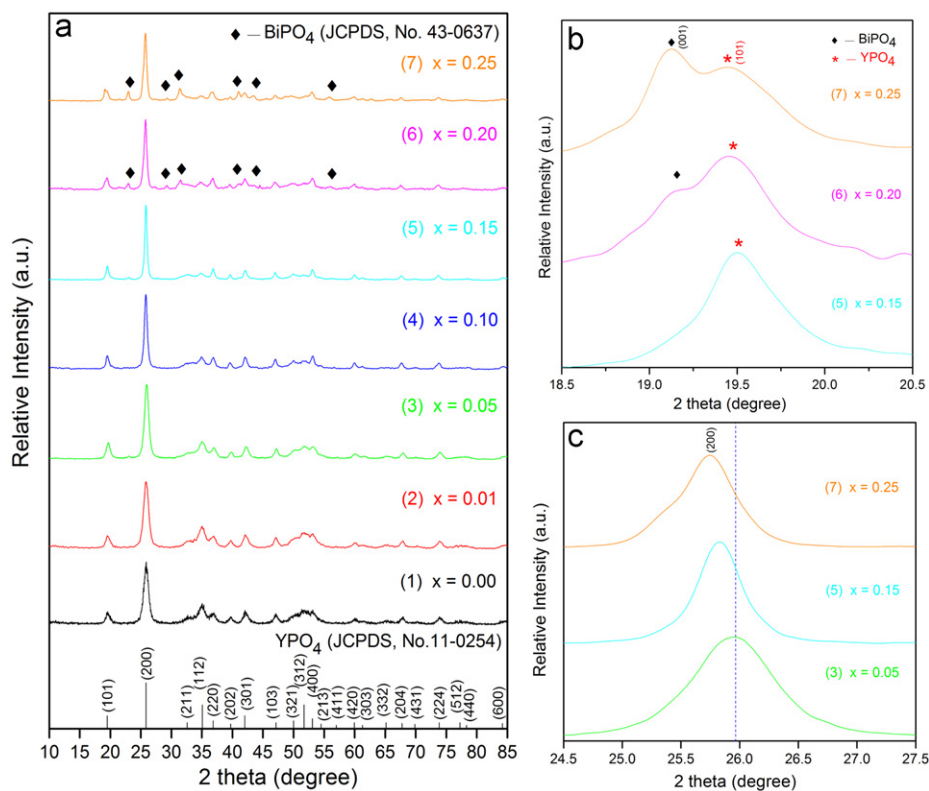


Fig. 1. (a) XRD pattern of Y_{0.99-x}PO₄:0.01Dy³⁺, xBi³⁺ and JCPDS card of YPO₄. (b) Comparison of Y_{0.99-x}PO₄:0.01Dy³⁺, xBi³⁺ (x=0.15, 0.20 and 0.25) around 2theta of 19.5°. (c) Comparison of Y_{0.99-x}PO₄:0.01Dy³⁺, xBi³⁺ (x=0.05, 0.15 and 0.25) around 2theta of 25.95°.

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