

Luminescence properties and energy transfer of $\text{CdWO}_4\text{:Sm}^{3+},\text{Bi}^{3+}$, M^+ ($\text{M}=\text{Li}, \text{Na}, \text{K}$) phosphors for white LEDs

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Abstract

A series of $\text{Sm}^{3+}\text{--Bi}^{3+}$ co-doped CdWO_4 phosphors with wolframite structure have been prepared via a high temperature solid state reaction process and their structural and luminescence properties were thoroughly investigated. The color-tunable emission from blue to yellow can be realized by $\text{WO}_6\text{--Sm}^{3+}$ or $\text{WO}_6\text{--Bi}^{3+}\text{--Sm}^{3+}$ energy transfer. A new distinct excitation bands from the $\text{CdWO}_4\text{:Sm}^{3+},\text{Bi}^{3+}$ phosphors are clearly observed at about 361 nm which comes from the $^1\text{S}_0\rightarrow^3\text{P}_1$ transitions of Bi^{3+} . The Fluorescent Resonance Energy Transfer between Bi^{3+} and Sm^{3+} were considered to be the primary mechanisms and it was proved from their crystal structure. Based on the results in this study, $\text{CdWO}_4\text{:Sm}^{3+},\text{Bi}^{3+},\text{K}^+$ sample is considered as an efficient phosphor with excellent color tunability for white UV-LEDs.

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1. Introduction

White LED with advantages such as high efficiency, long lifetime and environment-friendly characteristics is very likely to be the next generation of illumination technology in the near future [1,2]. Currently, the commercial phosphor-converted white LEDs (pc-WLEDs) made by combining yellow YAG: Ce^{3+} phosphor with the blue LED chip has some disadvantages such as low chromatic stability under different driving currents, low efficiency and low color rendering index [3,4]. White UV-LEDs fabricated by UV-LED chips coated with tri-color phosphors might conquer the aforementioned pitfalls owing to the invisible emission of the LED chips [2]. However, this new type of white LED has low luminescence efficiency because of the reabsorption of blue emission by red and green phosphors [5]. And the red phosphors still involve some problems because of their low brightness and chemical instability [6]. So UV-LED chips coated

with white light-emitting single-phased phosphors is a very potential approach to generate white UV-LEDs. At present, two or more activators simultaneously substitute one host lattice to form single-phase and multicolor-emitting phosphors is a research hotspot [7,8].

Cadmium tungstate CdWO_4 with wolframite structure has attracted much interest due to its potential application in luminescent lamp coatings [9,10] and solid state lasers [11]. It emits blue–green lights themselves under ultraviolet excitation [9]. Sm^{3+} -doped materials have been used as the orange–red emitting phosphors due to their intense $^4\text{G}_{5/2}\text{--}^6\text{H}_7$ emission in the 550–670 nm spectral region, and show potential applications in optical displays [12,13]. Energy transfer between $\text{O}^{2-}\text{--W}^{6+}$ CT state and Sm^{3+} ions was identified by the investigation of photoluminescence properties. It is well known that the Bi^{3+} ion can be used as an activator as well as a sensitizer of luminescence [14]. Luitel et.al [15] reported that there is an efficient energy transfer from Bi^{3+} to Sm^{3+} emission center. Jinsheng Shi [16] has predicted and confirmed that the $^1\text{S}_0\text{--}^3\text{P}_1$ transition of Bi^{3+} is located around 350 nm in CdWO_4 . Herein, the possible effective excitation light wavelength of Sm^{3+} and Bi^{3+} co-doped CdWO_4

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is at about 350 nm. And the doping of Bi^{3+} is expected to strengthen the absorption of near-UV radiation, and further enhance the emission of the luminescent center (Sm^{3+}). Moreover, in Sm^{3+} and Bi^{3+} co-doped CdWO_4 phosphor, WO_6 polyhedron may effectively transfer energy to Bi^{3+} and Sm^{3+} ions to generate strong green and red emissions. Combined with W-O blue-green emission, it may be a potential single-phase phosphor candidate for applications in white LED.

As far as our knowledge is concerned, there exists no detailed report on the photoluminescence properties of Sm^{3+} and Bi^{3+} co-doped CdWO_4 phosphor. In order to develop new series of optical materials, it is necessary to further investigate the luminescence of Sm^{3+} and Bi^{3+} activated CdWO_4 phosphor.

In this research, we reported our recent investigation results on the luminescence and color/chromaticity of $\text{CdWO}_4:\text{Sm}^{3+}$, Bi^{3+} , M^+ ($\text{M}^+ = \text{Li}^+, \text{Na}^+, \text{K}^+$). By varying the relative

concentrations of Sm^{3+} and Bi^{3+} , white light with tunable color emission was demonstrated. The efficient energy transfer from Bi^{3+} to Sm^{3+} was attributed to their close distance. Furthermore, the influence of the alkali metal ions M^+ ($\text{Li}^+, \text{Na}^+, \text{K}^+$) which were added into the host as the charge compensation agent on the luminescent intensity of Sm^{3+} and Bi^{3+} co-doped CdWO_4 phosphors was also discussed.

2. Experimental

2.1. Sample synthesis

A series of CdWO_4 phosphors with various concentrations of Sm^{3+} , Bi^{3+} and alkali metal ions were synthesized with the solid state reaction method in air atmosphere. The raw materials are cadmium oxide (CdO , 99%), samarium oxide (Sm_2O_3 , 99.9%), bismuth oxide (Bi_2O_3 , 99.9%), lithium carbonate (Li_2CO_3 , 99%), sodium carbonate (Na_2CO_3 , 99%), potassium carbonate (K_2CO_3 , 99%) and tungsten oxide (WO_3 , 99%). The starting materials were weighed according to the desired stoichiometric ratio and well mixed in an agate mortar. Each batch of the mixtures was put into an alumina crucible and calcined in a muffle furnace at 1000 °C for 8 h, and then the resultant powder with white body color was obtained.

2.2. Sample characterization

The structures of CdWO_4 samples were determined by using a Bruker D8 Advance X-ray diffractometer ($\text{Cu K}\alpha_1$ radiation, $\lambda = 0.15406$ nm) with radiation at a 0.02° (2θ)/0.05 s scanning step. The photoluminescence (PL) emission and excitation spectra were recorded with a Hitachi F-4600 spectrophotometer equipped with a 150 W xenon lamp as an excitation source.

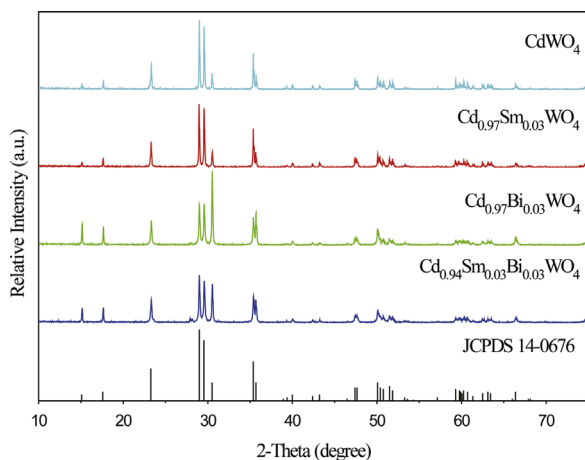


Fig. 1. XRD patterns of the CdWO_4 , $\text{Cd}_{0.97}\text{Sm}_{0.03}\text{WO}_4$, $\text{Cd}_{0.97}\text{Bi}_{0.03}\text{WO}_4$ and $\text{Cd}_{0.94}\text{Sm}_{0.03}\text{Bi}_{0.03}\text{WO}_4$ phosphors sintered at 1000 °C for 8 h.

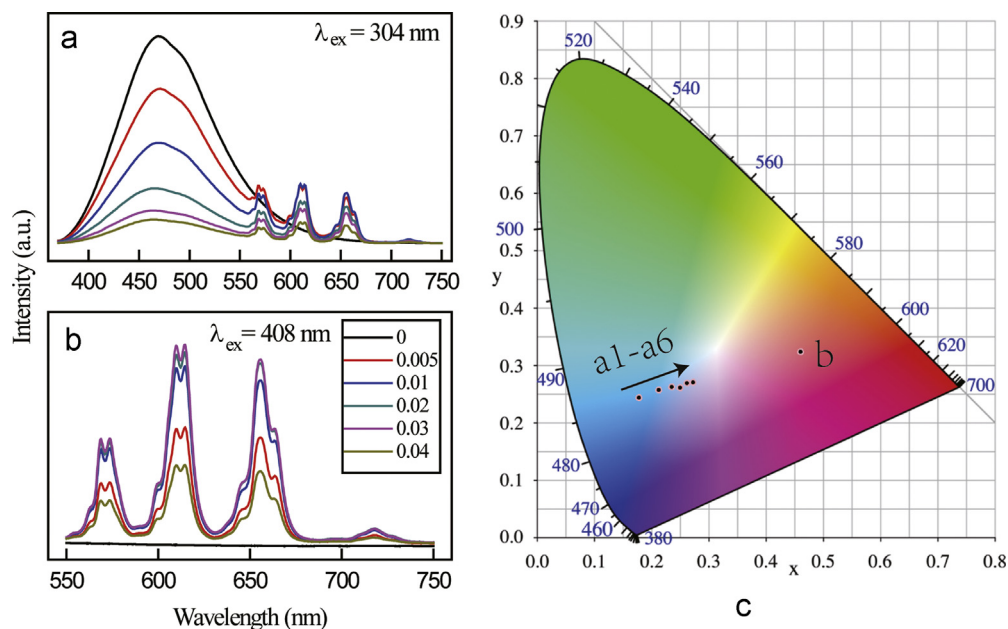


Fig. 2. Excitation (a), emission (b) spectra and chromaticity coordinates (c) of $\text{Cd}_{1-x}\text{Sm}_x\text{WO}_4$ with different concentrations.

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