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Luminescence and energy transfer of white emitting phosphor $Ba_3Ce(PO_4)_3$: Dy^{3+}

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ABSTRACT

A series of white emitting phosphors $Ba_3Ce(PO_4)_3$:Dy^{3+} were synthesized by a solid-state method, and the luminescent property and energy transfer were explored in detail. $Ba_3Ce(PO_4)_3$:Dy^{3+} shows three emission peaks, the blue emission band is associated to the $4f^05d^1 \rightarrow 4f^1$ transition of Ce^{3+} ions, and the two narrow peaks are assigned to the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ (490 nm) and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ (580 nm) transitions of Dy^{3+} ions, respectively. There are an efficient energy transfer from Ce^{3+} to Dy^{3+} ions in $Ba_3Ce(PO_4)_3$:Dy^{3+}, which can be proved by the decay curves. Importantly, $Ba_3Ce(PO_4)_3$:Dy^{3+} can produce white emission with the CIE coordinates (0.3294, 0.3376). The results show that $Ba_3Ce(PO_4)_3$:Dy^{3+} may be a potential application in white light emitting diodes.

1. Introduction

In the past years, white light emitting diodes (white LEDs) exhibit the most application value [1–3]. Generally, the white LEDs can be fabricated by combining blue LED with YAG:Ce^{3+}, however, a high correlated color temperature (CCT≈7750 K) and poor color rendering index (CRI≈70-80) are the defect of white LEDs, therefore, the new yellow phosphor or new white LEDs fabricated method are the research focus. For the new yellow phosphor, the nitride and silicate play an important role [4–6]. For the new fabricated method, the white LEDs are obtained by couple of an ultraviolet (UV) or near UV LED excited tri-color phosphors, however, there are the defect of re-absorption and controlling the ratio of the phosphors [7]. Therefore, the single phase white emitting phosphor became the research focus [8–10]. Generally, the $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions of Dy^{3+} ions locate at blue and yellow region, respectively, therefore, Dy^{3+} activated materials can present white light [11–15]. Actually, in order to improve the luminescence performance of Dy^{3+} activated materials, the sensitizer can be introduced [16]. As a sensitizer, Ce^{3+} ions have a wide range of applications, and can influence the emission color of phosphor [17–19]. Dy^{3+} doped phosphor can also be tuned by appropriate adjustment of relative proportion of Ce^{3+}/Dy^{3+} [20–22]. Ce^{3+} acts as activator and sensitizer concurrently in $Ba_3Ce(PO_4)_3$, and the concentration of Ce^{3+} is very large (100% mol^{-1}) in $Ba_3Ce(PO_4)_3$. As a result, Ce^{3+} ion serving as sensitizer can offer a large amount of energy to activator Dy^{3+}. Another key factor as luminescent material, it is important to choose the host compound [23–25]. Because phosphates show high thermal and chemical stability, therefore, they attracted the attention of researchers [26–30]. In this research, a single phase white emitting phosphor $Ba_3Ce(PO_4)_3$:Dy^{3+} was synthesized, and its emission intensity became stronger by doping Ce^{3+} as

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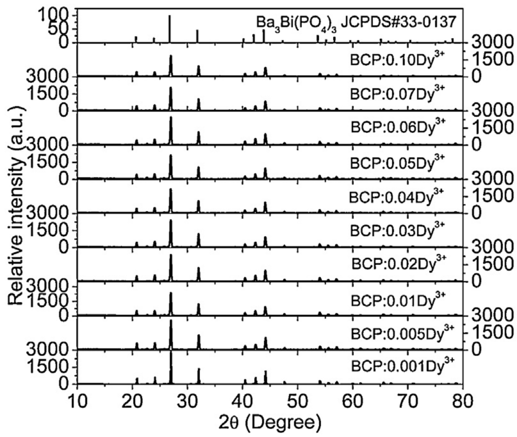

Fig. 1. XRD patterns of $Ba_3Ce_{1-x}(PO_4)_3$:xDy^{3+} (x = 0.001–0.10) with the standard JCPDS card no.33-0137 ($Ba_3Bi(PO_4)_3$).

sensitizer, and the luminescence properties and energy transfer of $Ba_3Ce(PO_4)_3$:Dy^{3+} were investigated. The result may be useful to the development of white LEDs.

2. Experimental

2.1. Samples preparation

A series of $Ba_3Ce_{1-x}(PO_4)_3$:xDy^{3+} (x, molar concentration) samples were synthesized by the high temperature solid-state method. Raw materials were $BaCO_3$ (A.R.), $NH_4H_2PO_4$ (A.R.), CeO_2 (99.99%) and Dy_2O_3 (99.99%). On the basis of stoichiometric proportion, the raw materials were weighed, and the electronic scale was 0.0001 g. According to the stoichiometric ratio, the raw materials were thoroughly mixed, and were ground by an agate mortar and pestle for more than 30 min till they were uniformly distributed. With reducing agent (Toner), the mixtures were heated at 1150 ° for 4 h in an air, and then, the samples were cooled to room temperature and ground again in an agate mortar. Finally, the samples were ground into powder for the sake of measurement.

2.2. Materials characterization

Powder X-ray diffraction (XRD) analysis was used for the phase formation of sample (Bruker AXS D8 advanced automatic diffractometer (Bruker Co., German) with Ni-filtered Cu Kα_1 radiation (λ = 0.15405 nm) operating at 40 kV and 40 mA, and a scan rate of 0.02°/s was applied to record the patterns). Stable and transient spectra of sample were measured by a fluorescence spectrophotometer (F-4600, HITACHI, Japan), and a 450 W Xe lamp was selected as the excitation source. A PMS-80 spectral analysis system is used to record the Commission International de l'Eclairage (CIE) chromaticity coordinates.

3. Results and discussion

3.1. Phase formation

Fig. 1 shows the XRD patterns of $Ba_3Ce_{1-x}(PO_4)_3$:xDy^{3+} (x = 0.001–0.10). All the diffraction peaks match well with that of the cubic $Ba_3Bi(PO_4)_3$ according to the standard reference of JCPDS card no. 33–0137, and no traces of impurity phases are observed. The results indicate there is little change of this crystal structure when Dy^{3+} ions are introduced into $Ba_3Ce(PO_4)_3$. According to ICSD card no.91803 ($Ba_3Bi(PO_4)_3$), $Ba_3Ce(PO_4)_3$ should belong to the eulytite-type compound, which crystallize in the cubic system with space group I-43d, and there is a three- dimensional packing of $[PO_4]^{3-}$ anionic tetrahedra and Ce/Ba octahedra, arranged in a manner to share common apices. As a representative, Fig. 2 shows the Rietveld analysis pattern of $Ba_3Ce_{0.96}(PO_4)_3$:$0.04Dy^{3+}$ by a General Structure Analysis System (GSAS). The black solid lines and red crosses represent calculated patterns and raw experimental patterns, respectively. The olive short vertical lines point out the position of Bragg of $Ba_3Ce(PO_4)_3$, and the differences between the calculated and experimental results are signed with gray bars. All of the observed peaks satisfy the reflection conditions, which illustrates that there is no other impurity phase detected in the phosphor.