A novel UV pumped yellow-emitting phosphor Ba2YAlO5:Dy3+ for white light-emitting diodes

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Abstract. A series of novel Dy3+ activated aluminate Ba2Y1-xDyxAI O3 (x = 0.01–0.30) red-emitting phosphors were synthesized through a high-temperature solid-state route at 1380°C. The X-ray diffraction patterns revealed that the samples were well crystallized in the space group P21/c (No. 14). The spectrum analysis revealed that under UV light excitation, Ba2YAlO5:Dy3+ phosphor exhibited blue and yellow peaks corresponding to F3/2 → H5/2 transition and F5/2 → H3/2 transition, respectively. The optimum dopant concentration of Dy3+ ions is around 2 mol% and the critical transfer distance of Dy3+ is calculated as 29 Å. The concentration quenching mechanism between Dy3+ has been investigated. Results indicate that Ba2YAlO5:Dy3+ offers the excellent optical properties as a potential yellow-emitting phosphor candidate for n-UV LEDs.

1 Introduction

Dy3+ as the activator can be doped into suitable hosts to obtain white light emission in a single phase. It can present two characteristic emission bands in luminescence spectrum, F3/2 → H5/2 at 488 nm (blue) and F5/2 → H3/2 at 580 nm (yellow)[1, 2]. The feasibility of the phosphor to generate white light is actually indicated by the Yellow and Blue (Y/B) ratios [3].

Recently, white light-emitting diodes (WLEDs) have gained enormous interest from scientists and engineers as a promising light source, which are important candidates for solid-state lighting due to its high luminous efficiency, compactness, long-lifetime, diversity design, fast switching material stability and environmentally friendly [4, 5]. LEDs are regarded as environmentally friendly because they have not utilized the toxic elements such as mercury (excitation light source) and colloidal cadmium selenide quantum dots. The most commercially available W-LEDs are fabricated by a combination of InGaN LED chips with Y3Al5O12:Ce3+ yellow-emitting phosphor [1]. But the above method suffers some major disadvantages in the red spectral region, including correlated color temperature that is higher than 4500 K and color rendering index that is lower than 80. Another effective method is to utilize near-UV LEDs chips coupled with multi-phosphors of red, green and blue phosphor [6, 7].

The aluminates have been extensively explored as hosts for phosphors due to their excellent optical properties [8, 9]. In this paper, we report the red emitting phosphors Ba2Y1-xDyxAI O3 (x = 0.01–0.30) under NUV (Near Ultraviolet) excitation. The phase purity, photoluminescence (PL) properties, influence of doping concentration, and chromaticity coordinate of the Ba2YAlO5:Dy3+ phosphors were investigated.

2 EXPERIMENTAL PROCEDURE

The synthesis of powder samples Ba2Y1-xDyxAI O3 (x = 0.01, 0.02, 0.05, 0.10, 0.15, 0.20, 0.25, and 0.30) phosphors with the concentration of Dy3+ from 1 to 30 mol% was carried out by conventional solid stated reaction in air. Al2O3 (nanopowder, A.R.), BaCO3 (A.R.), Y2O3 (99.99%), and Dy2O3 (99.99%) were completely mixed and ground in an agate mortar. Then the mixtures were put into an alumina crucible and preheated at 600 °C in air for 3 h. After that, the samples were reground completely and calcined in air at 1380 °C for 5 h. The associated reaction equations are as follows:

$$\text{4BaCO}_3 + (1-x)\text{Y}_2\text{O}_3 + \text{Al}_2\text{O}_3 + x\text{Dy}_2\text{O}_3 \rightarrow \text{Ba}_2\text{Y}_{1-x}\text{Dy}_x\text{Al}_2\text{O}_5 + \text{volatile products}$$

X-ray powder diffraction (XRD) was performed with Philips X’Pert MPD (Philips, Netherlands) with Cu Kα radiation (λ = 1.5418 Å) to identify the phase purity of the as-prepared samples. Data collection was carried out in the range of 2θ = 10°–70°. Luminescence properties of the synthesized phosphors were performed using FLS 920 spectrometer (Edinburgh) at room temperature.

3 RESULTS AND DISCUSSION

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The phase purity of the Ba$_2$YAlO$_5$:0.02Dy$^{3+}$ phosphors are identified by XRD and depicted in Fig. 1. All the diffraction peaks of the sample were consistent with the standard card (No.37-0292) for the Ba$_2$YAlO$_5$. The lattice constants of Ba$_2$YAlO$_5$:0.02Dy$^{3+}$ are calculated to be $a = 13.1702$ Å, $b = 7.4539$ Å, $c = 5.7102$ Å and $V = 527.22$ Å$^3$, respectively. It suggests that the Dy$^{3+}$ ions substituted the Y$^{3+}$ sites in Ba$_2$YAlO$_5$ due to their similar ionic radii of Dy$^{3+}$ ($r = 0.912$ Å) and Y$^{3+}$ (0.900 Å) when coordination number = 6 [10].

The emission spectrum of Ba$_2$YAlO$_5$:0.02Dy$^{3+}$ excited at 357 nm is shown in Fig. 3. The PL spectrum consists of four sharp lines in 450-750 nm attributing to the $^4$P$_{0,2}$, $^4$H$_{15/2}$, $^4$H$_{11/2}$, $^4$G$_{11/2}$ and $^4$H$_{15/2}$ → $^4$I$_{15/2}$ transition, respectively [14]. The peak at 357 nm is the strongest one which results from $^4$H$_{15/2}$ → $^4$P$_{0,2}$ transition. Thus, the Ba$_2$YAlO$_5$:Dy$^{3+}$ phosphors are suitable for InGaN-chip based w-LEDs.

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The critical energy transfer distance ($R_c$) between Dy$^{3+}$ ions in Ba$_2$Y$_{1-x}$Dy$_x$AlO$_5$ phosphors can be evaluated by the following formula given by Blasse [17]:

$$R_c = 2\left(\frac{3V}{4\pi x_N}\right)^{1/3}$$

where $N$ means the formula units per unit cell, $x$ represents the critical concentration of Dy$^{3+}$, and $V$ is the volume of the unit cell. Taking the appropriate values of $N$, $V$, and $x$ (2, 526.48 Å$^3$, and 0.02, respectively) for the Ba$_2$YAlO$_5$:0.02Dy$^{3+}$ phosphors, we estimate $R_c$ to be about 29 Å, which is far greater than 5 Å. Thus, the concentration quenching mechanism of Dy$^{3+}$ ions can be mainly ascribed to the multipole–multipole interaction.

Figure 4 showed the concentration influences to the PL intensities of Ba$_2$Y$_{1-x}$Dy$_x$AlO$_5$ phosphors ($x = 0.01, 0.02, 0.05, 0.10, 0.15, 0.20, 0.25, and 0.30$).
In summary, we have synthesized a series of aluminate yellow-emitting $\text{Ba}_2\text{YAlO}_5:\text{Dy}^{3+}$ phosphors by a solid-state reaction. The XRD examined results confirmed that all the compounds crystallize in the space group $P2_1/c$ (14). The synthesized phosphors $\text{Ba}_2\text{YAlO}_5:\text{Dy}^{3+}$ was excited by 357 nm, and showed a strong yellow-emitting emission at 580 nm which was assigned to the $^4\text{F}_{9/2} \rightarrow ^4\text{H}_{13/2}$ transition of $\text{Dy}^{3+}$ ions. When the doping concentration was over 2 mol%, the phosphor appeared concentration quenching phenomenon. The electric dipole-dipole interaction is the concentration quenching mechanism of $\text{Ba}_2\text{YAlO}_5:\text{Dy}^{3+}$ phosphors. All the above properties indicated the $\text{Dy}^{3+}$-activated $\text{Ba}_2\text{YAlO}_5$ phosphors could be a new yellow-emitting candidate for color mixing in the white light-emitting diode.

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