



# Investigation on luminescence properties of emission-tunable $\text{Ba}_9\text{Y}_2\text{Si}_6\text{O}_{24}:\text{Tb}^{3+}, \text{Mn}^{2+}$ phosphors

Songsong An<sup>a</sup>, Jia Zhang<sup>b,\*</sup>

<sup>a</sup>School of Chemistry and Chemical Engineering, Huaiyin Normal University, 111 West Chang Jiang Road, Huai'an 223001, China

<sup>b</sup>School of Physics and Electronic Electrical Engineering, Huaiyin Normal University, 111 West Chang Jiang Road, Huai'an 223001, China

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## Abstract

A series of emission-tunable  $\text{Ba}_9\text{Y}_2\text{Si}_6\text{O}_{24}:\text{Tb}^{3+}, \text{Mn}^{2+}$  (BYS: $\text{Tb}^{3+}, \text{Mn}^{2+}$ ) phosphors were prepared by a solid-state reaction method, and their luminescence properties upon ultraviolet light excitation were studied. The characteristic  $^5\text{D}_4\text{--}^7\text{F}_j$  emissions of  $\text{Tb}^{3+}$  are observed in BYS:0.6 $\text{Tb}^{3+}$  upon 300 nm excitation. By codoping  $\text{Tb}^{3+}\text{--Mn}^{2+}$  into the BYS host, tunable emission is realized in BYS:0.6 $\text{Tb}^{3+}, y\text{Mn}^{2+}$  ( $0 \leq y \leq 0.5$ ), ranging from yellowish green to orange. The energy transfer (ET) from  $\text{Tb}^{3+}$  to  $\text{Mn}^{2+}$  is verified by the decay curves and the ET efficiencies are evaluated from the changing decay lifetimes. By further adjusting the relative concentrations of  $\text{Tb}^{3+}$  and  $\text{Mn}^{2+}$ , nearly white light is obtained finally in the BYS:0.05 $\text{Tb}^{3+}, 0.1\text{Mn}^{2+}$  sample. The above investigations indicate the as-prepared BYS: $\text{Tb}^{3+}, \text{Mn}^{2+}$  phosphors could have potential applications in UV-LEDs.

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**Keywords:** Phosphors; LEDs; Tunable-emission

## 1. Introduction

In recent years, white light-emitting diodes (w-LEDs) have attracted increasing interest because of high luminescence efficiency, fast switching, long lifetime and environmental friendliness [1–3]. For these reasons, they have been widely used as solid-state light sources and components of display devices in our daily life. It is well known that w-LEDs can be obtained by combining blue or ultraviolet (UV) chip with the phosphor materials, known as phosphor converted-LEDs (pc-LEDs) [4]. The most common strategy to achieve w-LEDs is the combination of  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$  (YAG:Ce) phosphor with blue InGaN chip [5]. However, this method suffers the problems of low color rendering index ( $R_a < 80$ ) and unsatisfactory high correlated color temperature ( $T_c > 4500$  K) due to the deficiency of red fluorescent component, which restrict their applications [6,7]. In order to overcome these drawbacks, w-LEDs have been proposed by combining of a UV LED chips with tricolor (red, green, and

blue) phosphors [8]. This type of w-LEDs can produce a high color-rendering index and easily controlled emission colors [9,10]. Moreover, it is also important to design novel multicolor phosphors as well as single-phased white-light-emitting phosphors, which can overcome the shortages of phosphors combination such as different degradation rates and re-absorption between phosphors [11,12]. Based on these points, it is essential to explore a series of novel phosphor which can be effectively excited by UV LED chip at present.

To date, the silicates have proven to be excellent host materials for phosphors due to their good chemical stability and environmental friendliness. It is also reported that tunable emission has been obtained by employing energy transfer (ET) between the luminescent ions in this kind of phosphors, such as  $\text{CaAl}_2\text{Si}_2\text{O}_8:\text{Eu}^{2+}, \text{Mn}^{2+}, \text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Ce}^{3+}, \text{Mn}^{2+}$ , etc. [13,14]. In the previous Refs. [15,16], new  $\text{Ba}_9\text{Y}_2\text{Si}_6\text{O}_{24}$  (BYS) silicate compound was generated by substituting  $\text{Sc}^{3+}$  totally with  $\text{Y}^{3+}$  in  $\text{Ba}_9\text{Sc}_2\text{Si}_6\text{O}_{24}$ , and the ET processes of  $\text{Eu}^{2+}\text{--Mn}^{2+}$  and  $\text{Ce}^{3+}\text{--Tb}^{3+}$  in BYS were also reported. However, the tunable emission of  $\text{Tb}^{3+}\text{--Mn}^{2+}$  by using ET has not been studied in detailed for the time being. In this paper, to develop new emission-tunable

\*Corresponding author.

E-mail address: [zhangjianew@126.com](mailto:zhangjianew@126.com) (J. Zhang).

phosphors for UV LEDs, a series of  $\text{Tb}^{3+}$ – $\text{Mn}^{2+}$  doped BYS samples were synthesized, and their photoluminescence properties were investigated.

## 2. Experimental

Power samples of  $\text{Ba}_{9-y}\text{Y}_{2-x}\text{Si}_6\text{O}_{24}:x\text{Tb}^{3+}, y\text{Mn}^{2+}$  (BYS:  $x\text{Tb}^{3+}, y\text{Mn}^{2+}, 0 \leq x \leq 0.6, 0 \leq y \leq 0.5$ ) were synthesized by a solid-state reaction method. Appropriate amounts of starting materials  $\text{BaCO}_3$  (99%),  $\text{SiO}_2$  (99%),  $\text{Y}_2\text{O}_3$  (99.999%),  $\text{Tb}_4\text{O}_7$  (99.99%),  $\text{MnCO}_3$  (99%), and  $\text{H}_3\text{BO}_3$  (99%) were thoroughly mixed in an agate mortar and pre-fired at  $950^\circ\text{C}$  for 3 h in air. Then the flux  $\text{H}_3\text{BO}_3$  was added and reground, finally heated at  $1130^\circ\text{C}$  for 3 h in a reducing atmosphere (5%  $\text{H}_2$ –95%  $\text{N}_2$ ).

The phase purity was determined by using an ARL X'TRA powder X-ray diffractometer (XRD) with  $\text{Cu K}\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ) operating at 40 kV and 35 mA. Diffuse reflection spectrum (DRS) were obtained by a UV/visible spectrophotometer (UV-3600, SHIMADZU) using  $\text{BaSO}_4$  as a reference in the range of 200–700 nm. The morphology of the as-prepared sample was inspected by field emission scanning electron microscope (FESEM, FEI, Quanta FEG). The luminescence spectra and decay curves were recorded on an FLS-920T fluorescence spectrophotometer. All the spectra measurements were performed at room temperature.

## 3. Results and discussion

Fig. 1(a) shows the XRD patterns of the BYS: $x\text{Tb}^{3+}, y\text{Mn}^{2+}$  ( $0 \leq x \leq 0.6, 0 \leq y \leq 0.5$ ) samples. All the diffraction peaks can be indexed to the hexagonal  $\text{Ba}_9\text{Sc}_2\text{Si}_6\text{O}_{24}$  (JCPDS 82-1119) compound except that the diffraction peaks exhibit a shift toward the low-angle direction. This could be due to the BSY is isostructural with  $\text{Ba}_9\text{Sc}_2\text{Si}_6\text{O}_{24}$  and the ion radius of  $\text{Y}^{3+}$  is larger than that of  $\text{Sc}^{3+}$ . Besides, it also can be found the phase samples can be obtained successfully when the luminescent ions are doped.

Fig. 2 presents the SEM image of the typical BYS host. It can be seen that relatively dispersed particles have been

obtained for the solid-state reaction preparation although the particle sizes are not uniform. The sizes of most particles are within  $20 \mu\text{m}$ . From the enlarged image, it can be found some small particles adhere to the surface of big ones, causing rough particle surface.

Fig. 3 presents the DRS of BYS from 200 to 700 nm. It shows a high reflectance in the visible range (400–700 nm), which is in agreement with the white daylight color of the BYS matrix. To determine the optical bandgap value of the BYS compound experimentally, the absorption spectrum of BYS (see the inset of Fig. 3) was obtained from its reflection spectrum by using the Kubelka–Munk ( $K$ – $M$ ) function [17]

$$F(R) = (1 - R)^2 / 2R = K/S \quad (1)$$

where  $R$ ,  $K$  and  $S$  are the reflection, absorption and scattering coefficient, respectively. By extrapolating the  $K$ – $M$  function to  $K/S=0$ , the optical bandgap was determined to be about 3.55 eV.

Fig. 4 shows the normalized excitation ( $\lambda_{\text{em}} = 543 \text{ nm}$ ) and emission ( $\lambda_{\text{ex}} = 300 \text{ nm}$ ) spectra of the typical BYS: $0.6\text{Tb}^{3+}$  phosphor. From the enlarged part (320–420 nm) of the excitation spectrum, it can be found several narrow excitation peaks exist, which are assigned to the 4f–4f transitions of  $\text{Tb}^{3+}$  and the strongest one at 379 nm belongs to the  ${}^7\text{F}_6$ – ${}^5\text{D}_3$  transition [18]. The strong excitation bands from 230 to

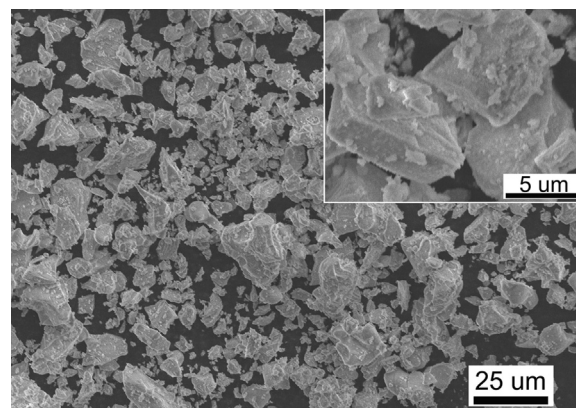


Fig. 2. SEM image of BYS.

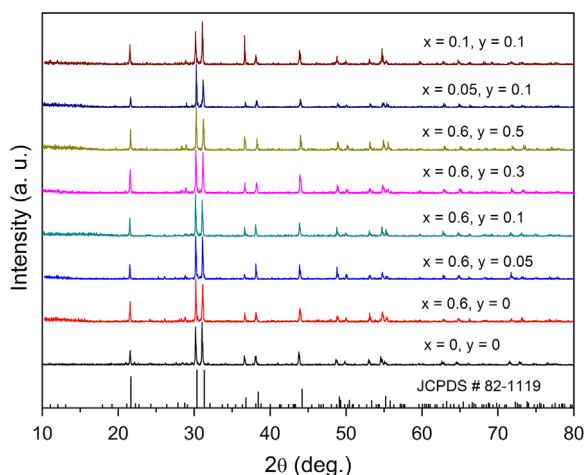


Fig. 1. XRD patterns of BYS: $x\text{Tb}^{3+}, y\text{Mn}^{2+}$  ( $0 \leq x \leq 0.6, 0 \leq y \leq 0.5$ ).

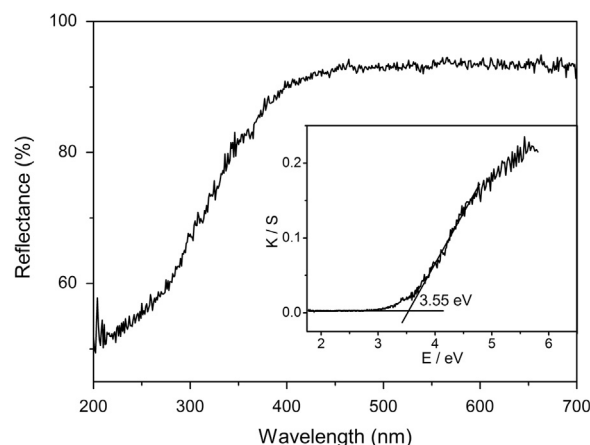


Fig. 3. DRS of BSY, inset shows the absorption spectrum of BYS obtained from its reflection spectrum using the  $K$ – $M$  function.

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