



# Luminescence studies on $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}$ , $\text{Dy}$ phosphor crystals

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## ABSTRACT

Using urea as fuel,  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}$ ,  $\text{Dy}$  phosphor was prepared by a combustion method. Its luminescence properties under ultraviolet (UV) excitation were investigated. Pure  $\text{SrMgAl}_{10}\text{O}_{17}$  phase was formed by urea-nitrate solution combustion synthesis at  $550^\circ\text{C}$ . The results indicated that the emission spectra of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}$ ,  $\text{Dy}$  has one main peak at 460 nm and one shoulder peak near 516 nm, which are ascribed to two different types of luminescent  $\text{Eu}^{2+}$  centers existing in the  $\text{SrMgAl}_{10}\text{O}_{17}$  matrix crystal. The blue luminescence emission of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}$  phosphors was improved under UV excitation by codoping  $\text{Dy}^{3+}$  ions. The  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}$  phosphors showed green afterglow ( $\lambda = 516\text{ nm}$ ) when  $\text{Dy}^{3+}$  ions were doped.  $\text{Dy}^{3+}$  ions not only successfully play the role of sensitizer for energy transfer in the system, but also act as trap levels and capture the free holes in the spinel blocks.

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## 1. Introduction

Synthetic hexagonal alkaline earth aluminates doped by divalent europium ions are efficient luminescence materials. They show a blue emission that is characterized by high quantum efficiency under UV excitation [1]. They are widely used in plasma display panels (PDPs), field emission displays (FEDs) and fluorescence lamps [2,3]. Hexagonal alkaline earth aluminate phosphors are typically produced by the solid-state reaction method. Combustion synthesis is a novel technique that has been applied to phosphor synthesis in the past few years [4]. Combustion synthesis involves an exothermic reaction between metal nitrates and a fuel. This technique produces highly crystalline powders in the as-synthesized state.

In this paper, the  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  codoped  $\text{SrMgAl}_{10}\text{O}_{17}$  samples were synthesized by a simple combustion process. Their emission and excitation spectra have been investigated and a green afterglow observed. The luminescence performance can be improved greatly when phosphors are doped with suitable auxiliary activators [5]. The role of the codoping with  $\text{Dy}^{3+}$  ions in the enhancement of the fluorescence and afterglow from  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}$  was studied in detail.

## 2. Experimental procedure

The starting materials were  $\text{Eu}_2\text{O}_3$  (4N),  $\text{Dy}_2\text{O}_3$  (4N),  $\text{Sr}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  (AR),  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (AR),  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (AR)

and urea (AR).  $\text{Eu}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$  were dissolved in  $\text{HNO}_3$  solution, and then stoichiometrically weighed metal nitrates and urea were added, mixed with vigorous stirring at  $70^\circ\text{C}$  for 30–40 min. The clear solution was transferred into a porcelain crucible. The crucible containing the solution was introduced into a muffle furnace that had been preheated at  $550^\circ\text{C}$ . With boiling, the solution evaporated and became increasingly more viscous. After a few minutes, combustion ignition took place and voluminous white phosphor powders were obtained.

The crystal phases of the prepared particles were analyzed by X-ray diffraction (XRD) pattern measured using a Bruker D8 (Bruker Co. Ltd, German) X-ray diffractometer with graphite monochromatized  $\text{Cu K}\alpha$  irradiation ( $\lambda = 1.5406\text{ \AA}$ ). The emission and excitation spectra of all the samples were obtained using a spectrophotometer (Perkin–Elmer LS-55, Perkin–Elmer Co. Ltd., USA) using a Xe flash lamp. The decay curve of afterglow was measured using a ST-86LA (Peking Normal University, China) brightness meter.

## 3. Results and discussion

Fig. 1 shows XRD patterns of  $\text{SrMgAl}_{10}\text{O}_{17}$  and  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}_{0.04}, \text{Dy}_{0.04}$  particles prepared by the combustion method. There are no observable differences between the two diffraction patterns, indicating that the pure phase of  $\text{SrMgAl}_{10}\text{O}_{17}$  (JCPDS No. 26-0879, space group  $\text{P6}_3/\text{mmc}$ ,  $a = 5.63\text{ \AA}$ ,  $c = 22.47\text{ \AA}$ ) has already formed in the combustion step. Furthermore, the little amount of doped rare-earth ions has almost no effect on the  $\text{SrMgAl}_{10}\text{O}_{17}$  phase composition. In this work, the structure of  $\text{SrMgAl}_{10}\text{O}_{17}$  with space group  $\text{P6}_3/\text{mmc}$  was taken as the starting model for synthetic phosphors.

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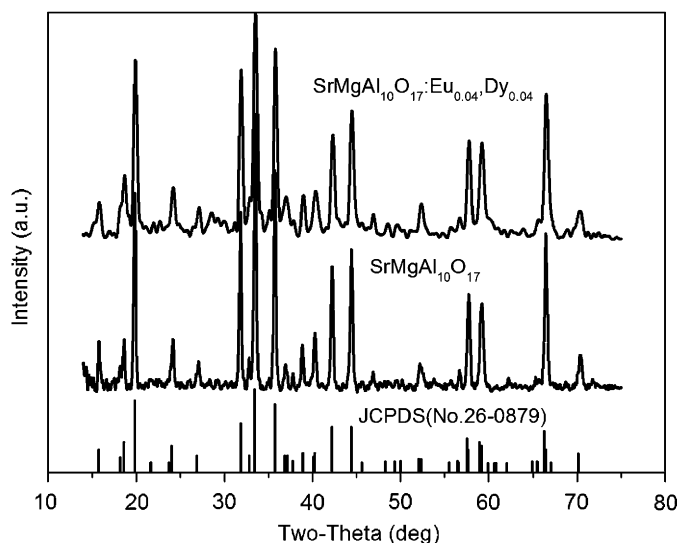


Fig. 1. Powder XRD patterns of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  phosphor.

The luminescence excitation and emission spectra of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  specimens prepared with different dopant contents are shown in Fig. 2. The emission intensity of phosphors was monitored under UV ( $\lambda = 314 \text{ nm}$ ) excitation with varying rare-earth ions concentration and ratio. Meanwhile, the excitation intensity of phosphors was monitored at  $\lambda = 460 \text{ nm}$ . The prepared phosphors exhibit a blue luminescence emission. No emission beyond  $600 \text{ nm}$  was observed, indicating that the  $\text{Eu}^{3+}$  ions have been effectively reduced to  $\text{Eu}^{2+}$  ions during the combustion process [6]. The emission spectrum excited by UV ( $\lambda = 314 \text{ nm}$ ) consists of a wide band with a peak at about  $460 \text{ nm}$ , which corresponds to the  $5d-4f$  transitions of  $\text{Eu}^{2+}$  ions. The excitation spectrum of the blue fluorescence (monitored at  $\lambda = 460 \text{ nm}$ ) shows two wide bands with their peaks at  $250$  and  $314 \text{ nm}$ , respectively, which are due to the crystal field splitting of the  $\text{Eu}^{2+}$   $d$ -orbital. Excitation spectra monitored at  $460 \text{ nm}$  show an optimal excitation band centered at  $314 \text{ nm}$ .

When  $\text{Dy}^{3+}$  is doped in the phosphors, the shape of the excitation and emission spectra remains, but the excitation intensity increases in comparison with the undoped case. The luminescent intensity excited by UV increases with the amount of  $\text{Dy}^{3+}$  increasing, as shown in Fig. 2a, which gives the luminescent spectra of  $\text{SrMgAl}_{10}\text{O}_{17}:0.04\text{Eu}^{2+}, x\text{Dy}^{3+}$  phosphors when  $x$  is changed. With increasing  $\text{Dy}^{3+}$  content at fixed  $\text{Eu}^{2+}$  content ( $0.04 \text{ mol}$ ) in  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}, \text{Dy}^{3+}$  phosphor, the emission intensity increases. But when the value of  $\text{Dy}/\text{Eu}$  is over 1, the emission intensity decreases.

In general, the excess concentration of activators quenches the photoluminescence. So, it is necessary to experimentally find the critical concentration of  $\text{Eu}^{2+}$  or  $\text{Dy}^{3+}$  in terms of optimizing the luminous efficiency of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}_x\text{Dy}_x$  under the UV excitation. The concentration quenching was observed (Fig. 2b) when the  $\text{Eu}$  content ( $x$ ) was  $0.04$ . At this critical concentration, the highest luminescent intensity was observed.

In addition to the main peak at  $460 \text{ nm}$ , a shoulder emission at  $516 \text{ nm}$  was also seen in the  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  phosphors. This shoulder emission was found to be the origin of a green color of afterglow from  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  crystals based on afterglow measurements.

Divalent Europium ions show a rather short decay time in the range  $1-10 \mu\text{s}$  since the  $5d-4f$  emission of  $\text{Eu}^{2+}$  is spin- and parity-allowed [7]. However, luminescent materials activated by  $\text{Eu}^{2+}$  can show afterglow, especially if additional dopants, e.g.  $\text{Dy}^{3+}$ , are

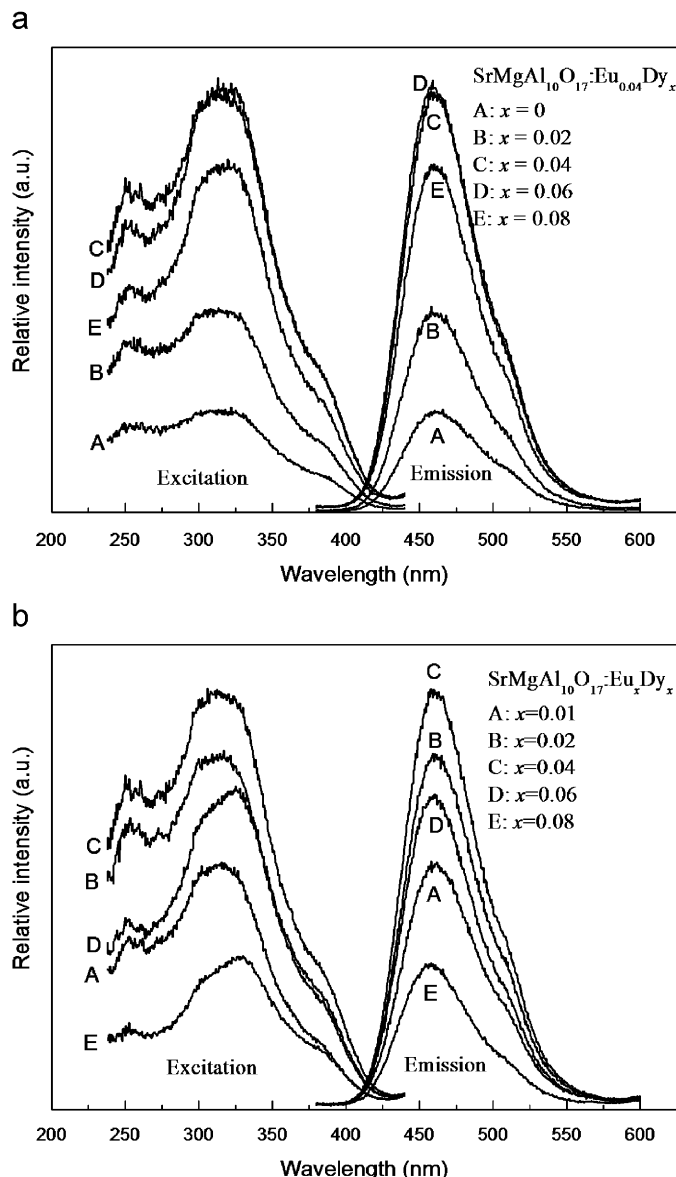


Fig. 2. Excitation spectra ( $\lambda_{\text{em}} = 460 \text{ nm}$ ) and emission spectra ( $\lambda_{\text{ex}} = 314 \text{ nm}$ ) of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  phosphor.

incorporated into the host lattice. Interestingly, the  $\text{SrMgAl}_{10}\text{O}_{17}$  phosphor doped only with  $\text{Eu}^{2+}$  shows a small but certain amount of afterglow.

The afterglow spectra of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  with different compositions are shown in Fig. 3. The afterglow intensity of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}, \text{Dy}$  phosphors was measured under UV ( $\lambda = 334 \text{ nm}$ ) excitation. Meanwhile, the excitation spectra of phosphors were monitored under  $\lambda = 516 \text{ nm}$ . The afterglow spectra of  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}, \text{Dy}^{3+}$  were measured after the excitation source was switched off at  $1 \text{ ms}$ . Under UV excitation all specimens yielded a green afterglow with the peak wavelength at  $516 \text{ nm}$ . The optimal concentration of  $\text{Eu}^{2+}$  is  $0.04$  and the optimal ratio of  $\text{Eu}$  to  $\text{Dy}$  is 1 for maximum intensity of afterglow (Fig. 3a).

An important result of the present work is that we observed green afterglow in the  $\text{SrMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors. Fig. 4 shows the decay curves of afterglow of the specimens. These specimens were irradiated by the UV light for  $10 \text{ min}$ . After the light source was removed, all of the specimens showed a rapid decay and subsequently an afterglow. When the value of  $\text{Eu}/\text{Dy}$  is

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