Study on Design, Synthesis and Controlled Color Emission of (Gd,Y)AG:Ce³⁺ Based Phosphors through Replacing Al³⁺ Partly

Shiyu Ma,¹ Bin Liu,¹ Bingqiang Cao,¹ Yizhong Lu,¹ Zongming Liu^{1,*} and Jinkai Li^{1,*}

Abstract

Owing to its excellent yellow light emission, the (Gd,Y)AG:Ce³⁺ based phosphors have been widely studied. In this paper, the (Gd,Y)₃(Al_{1-x}Ga_x)₅O₁₂:Ce³⁺ (x=0.1-0.8) and (Gd,Y)₃(Al_{1-2y}Mg_ySi_y)₅O₁₂:Ce³⁺ (y=0.05-0.4) phosphors were successfully prepared by coprecipitation and high temperature solid phase method, respectively. The lattice stabilization, microstructure and fluorescence properties were systematically investigated. With increasing the concentration of Ga³⁺, the emission peaks of (Gd,Y)₃Al₅O₁₂:Ga³⁺/Ce³⁺ phosphors displayed a blue-shift from ~575 nm (yellow) to ~549 nm (yellow-green), and the decay time was decreased gradually. However, the emission peaks of (Gd,Y)AG:Mg²⁺/Si⁴⁺/Ce³⁺ phosphors red shifted from ~575 nm (yellow) to ~592 nm (orange-red) with increasing the concentration of Mg²⁺/Si⁴⁺, and the decay time change was similar to the former. The underlying mechanisms were thoroughly investigated and elucidated. In addition, the effect of temperature on the fluorescence properties of (Gd,Y)AG:Ce³⁺ based phosphors was investigated in detail. The (Gd,Y)AG:Ce³⁺ based phosphors show promising potential in various lighting and optical display applications.

Keywords: (Gd, Y) AG:Ce³⁺ phosphors; Synthesis; Luminescent property.

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1. Introduction

The garnet phase (Ln₃Al₅O₁₂) as a very important solid luminescent material is widely used in phosphors and scintillators owing to its excellent fluorescence properties.^[1-2] The YAG:Ce³⁺ phosphor is widely used in fluorescent lamps and white light emitting diode (w-LED) due to its excellent yellow emission at ~550 nm (5d-4f transition).^[3] With the blue-ray gallium nitride (InGaN) chip excited, this phosphor emits white light emission, which is widely used in field emission displays (FED), w-LED, cathode-ray tubes (CRT), electroluminescent (EL), plasma display panels (PDP), and so forth.^[4-8]

The Ga³⁺/Ce³⁺ or Mg²⁺/Si⁴⁺/Ce³⁺ co-doped (Gd, Y)AG solid solution is chosen in this study due to the following main reasons: (1) the large radius of Gd³⁺ ions (1.053 Å) causes the GdAG phase to demonstrate a high temperature instability.^[9-12] A smaller radius of Y³⁺ ions (1.019 Å) doping in GdAG phase achieves the lattice stability; (2) the yellow phosphors need to be further improved. The Ga³⁺/Ce³⁺ or Mg²⁺/Si⁴⁺/Ce³⁺ co-

doped (Gd, Y)AG phosphors with controllable color emission effectively reduced the production blue light composition in white LEDs is too high, and its properties of blue light components,^[13-16] and its fluorescence properties can be significantly improved; (3) replacing Al³⁺ with Ga³⁺ or Mg²⁺/Si⁴⁺ ions in (Gd, Y)AG results in the changes of crystal coordination field, which realizes the adjustable fluorescence performance of (Gd, Y)AG based phosphors.^[17,18] The double substitution of [Mg²⁺]-[Si⁴⁺] for [Al³⁺]-[Al³⁺] leads to the lattice constant increasing and achieves the color emission fine-tuning. As a result, The Ga³⁺/Ce³⁺ or Mg²⁺/Si⁴⁺/Ce³⁺ co-doped (Gd,Y)AG phosphors are investigated in this work.

In this paper, the (Gd,Y)AG:Ga³⁺/Ce³⁺ phosphors were synthesized by a co-precipitation method using ammonium bicarbonate as the precipitant, and the (Gd,Y)AG:Mg²⁺/Si⁴⁺/Ce³⁺ solid solutions were obtained via a high temperature solid phase reaction. The detailed properties of (Gd,Y)AG:Ga³⁺/Ce³⁺ and (Gd,Y)AG:Mg²⁺/Si⁴⁺/Ce³⁺ phosphors were characterized via XRD, FE-SEM, PLE/PL, decay behavior and quantum efficiency analysis. Herein, we report the synthesis, micro-morphology, PLE/PL spectra, and decay behavior of (Gd,Y)AG:Ga³⁺/Ce³⁺ and (Gd,Y)AG:Mg²⁺/Si⁴⁺/Ce³⁺ phosphors.

2. Experimental procedure

¹ School of Materials Science and Engineering, University of Jinan, Jinan, Shandong 250022, China.

*Email: mse_lijk@ujn.edu.cn (J. Li); liuzm@ujn.edu.cn (Z. Liu)

2.1 Materials

Rare earth oxides Gd_2O_3 , Y_2O_3 and CeO_2 99.99% were acquired from Huizhou Ruier Rare Chemical Hi-Tech Co. Ltd., Huizhou, China. Gallium nitrate ($\text{Ga}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$, 99.9%), magnesium oxide (MgO , 99%), silica (SiO_2 , 99%), aluminum nitrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, AR), aluminum oxide (Al_2O_3 , 99%) and nitric acid (HNO_3 , AR) were provided by Sinopharm Chemical Reagent Co. Ltd., Shanghai. Ammonium bicarbonate (NH_4HCO_3 , AR) and alcohol ($\text{C}_2\text{H}_5\text{OH}$, AR) were from Fuyu Fine Chemical Co., Ltd., Tianjin. All reagents were directly used without further purification.

2.2 Preparation

2.2.1 Synthesis of $(\text{Gd,Y})\text{AG}:\text{Ga}^{3+}/\text{Ce}^{3+}$ phosphors

Rare earth nitrates $\text{RE}(\text{NO}_3)_3$ ($\text{RE} = \text{Gd}, \text{Y}$ or Ce) were obtained by dissolving rare earth oxides (Gd_2O_3 , Y_2O_3 and CeO_2) in nitric acid. These rare earth nitrates $\text{RE}(\text{NO}_3)_3$ were then mixed with gallium nitrate to acquire the mother solution according to the chemical formula $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$. The mother solution was titrated into ammonium bicarbonate solution, and then aged for 30 min. After aging, the turbid liquid was centrifuged and washed repeatedly with distilled water and alcohol to remove by-products. The precipitate was dried at 80 °C for 24 h to obtain the precursor. Finally, the precursor was calcined at 1500 °C in a reducing atmosphere (20% H_2 in N_2) to obtain the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors.

2.2.2 Synthesis of $(\text{Gd,Y})\text{AG}:\text{Mg}^{2+}/\text{Si}^{4+}/\text{Ce}^{3+}$ phosphors

Rare earth oxides (Gd_2O_3 , Y_2O_3 and CeO_2), aluminum oxide, magnesium oxide and silica were mixed with alcohol according to the chemical formula $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}$. Then, the mixed solution was transferred to a grinding tank and agitated for 10 h. After ball milling, the mixture was placed in a drying box at 80 °C for 8 h to remove ethanol. The dried product was further ground to 200 mesh, and calcined at 1500 °C for 4 h under a reducing atmosphere (20% H_2 in N_2).

2.3. Characterization

The XRD patterns for phase analysis were collected at room temperature using nickel-filtered $\text{CuK}\alpha$ radiation in the 2 θ range of 10–50° at a scan speed of 4.0° 2 θ /min (Model D8 ADVANCE, BRUKER Co., Germany). The morphology of the precursors and resultant products were collected via FE-SEM (QUANTA FEG 250, FEI Co., America) with an acceleration voltage of 10 kV. The PLE and PL spectra were obtained using a Fluorescence Spectrophotometer (FP-6500, JASCO Co., Japan) at room temperature equipped with a Φ 60-mm integrating sphere (ISF-513, JASCO, Tokyo, Japan) and a 150-W Xe-lamp as the excitation source. The optical performances for all samples were conducted under identical

conditions with the slit breadth of 5 nm. The phosphor powder was excited with a selected wavelength and the intensity of the intended emission was recorded as a function of elapsed time after the excitation light was automatically cut-off using a shutter.

3. Results and discussion

Fig. 1a shows the XRD patterns of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors which were obtained by calcination at 1500 °C. It can be seen that the phosphors basically maintain the XRD diffraction behavior of GdAG garnet (JCPDS:73-1371) with the Ga^{3+} content increasing from 10 at.% to 40 at.%, but the crystal faces of (321) and (420) belong to the GdGG phase (JCPDS:71-0701). This phenomenon is attributed to the substitution of Ga^{3+} for Al^{3+} , and the detailed conversion phase process is displayed in Fig. 1b. Fig. 1c and 1d show the FE-SEM images of the $(\text{Gd,Y})_3(\text{Al}_{0.9}\text{Ga}_{0.1})_5\text{O}_{12}:\text{Ce}^{3+}$ precursor and its phosphors calcined at 1500 °C, respectively. From which can be seen that the precursor exhibits a soft agglomeration state with the particle size of ~200 nm (Fig. 1c). After calcined at 1500 °C, the particles of the phosphor obviously grow with the size of 0.5 to 1 μm in diameter, and the phosphor particles also are uniformly dispersed.

Fig. 2a shows PLE spectra of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ ($x = 0.1-0.8$) phosphor. Monitored at the wavelength of 575 nm, the PLE spectra of the $(\text{Gd,Y})\text{AG}:\text{Ce}^{3+}$ phosphor mainly contains two excitation peaks, among which the main excitation peak at ~469 nm (the strongest) and ~330 nm belong to the $^2\text{F}_{5/2} \rightarrow \text{E}_{2g}$ transition of Ce^{3+} and $^2\text{F}_{5/2} \rightarrow \text{T}_{2g}$ transition of the $(\text{Gd,Y})\text{AG}:\text{Ga}^{3+}/\text{Ce}^{3+}$ phosphors, respectively. However, the position of the main excitation peak shifted from ~469 nm to ~442 nm with the concentration of Ga^{3+} increasing to 80 at.%. The PL spectra of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ ($x = 0.1-0.8$) phosphors under the optimal excitation wavelength are depicted in Fig. 2b.^[19] The $(\text{Gd,Y})\text{AG}:\text{Ce}^{3+}$ phosphor displays yellow emission at the wavelength of 575 nm under optimal excitation wavelength of 469 nm, which comes from the $5d-4f$ transition of Ce^{3+} .^[20] The emission intensities of the $(\text{Gd,Y})\text{AG}:\text{Ga}^{3+}/\text{Ce}^{3+}$ phosphors decrease as the concentration of Ga^{3+} increases. Compared to the $(\text{Gd,Y})\text{AG}:\text{Ce}^{3+}$ phosphor, the emission peaks of the $(\text{Gd,Y})\text{AG}:\text{Ga}^{3+}/\text{Ce}^{3+}$ phosphors blue shift from ~567 nm to ~549 nm when monitored at optimal excitation wavelengths of these phosphors with the concentration of Ga^{3+} increased from 10 at.% to 80 at.%.^[21] The phenomenon is consistent with the results of the excitation spectrum of Fig. 2a. The blue shift of the Ce^{3+} $5d-4f$ originates from the decrease in crystal field splitting with the substitution of Al^{3+} by the larger Ga^{3+} ions, and this phenomenon is wellreported for garnets.^[22-25]

Fig. 1 (a) XRD patterns of the $(\text{Gd},\text{Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ phosphors calcined at 1500 °C as a function of Ga^{3+} concentration ($x = 0.1-0.4$). (b) Schematic diagram of crystal structure changed of the $(\text{Gd},\text{Y})\text{AG}:\text{Ga}^{3+}/\text{Ce}^{3+}$ phosphors before and after the substitution of Al^{3+} with Ga^{3+} ions. FE-SEM images of the $(\text{Gd},\text{Y})_3(\text{Al}_{0.9}\text{Ga}_{0.1})_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ precursor (c) and its phosphor calcined at 1500 °C (d).

Fig. 2 PLE (a) and PL (b) spectrum of the $(\text{Gd},\text{Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ phosphors ($x = 0-0.8$) calcined at 1500 °C.

The CIE color coordinates diagrams of the $(\text{Gd},\text{Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors were plotted via the PL spectra of these phosphors calculated, as shown in Fig. 3a. The $(\text{Gd},\text{Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors were analyzed to have the color coordinates (x , y) and color temperatures of (0.49, 0.50), ~2918 K, (0.48, 0.51), ~3090 K, (0.47, 0.52), ~3264 K, (0.45, 0.54), ~3611 K, (0.43, 0.55), ~3920 K and (0.41, 0.55), ~4205 K when the x was set as 0.1, 0.2, 0.4, 0.6 and 0.8, respectively. With the replacement of Al^{3+} by Ga^{3+} , this work demonstrates the luminescent color of these phosphors changing from yellow to yellow-green and this result is consistent with the PL spectra of Fig. 2b.

In order to further elucidate the fluorescence attenuation behavior of these phosphors, the fluorescence decay kinetics

was studied. It is found that the decay data can be well fitted with the following double exponential equation in each case:

$$I = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + B \quad (1)$$

$$\tau = \frac{A_1 \tau_1^2 + A_2 \tau_2^2}{A_1 \tau_1 + A_2 \tau_2} \quad (2)$$

where I is the luminescence intensity at a certain moment, t is the time, A_1 , A_2 and B are constant, τ_1 and τ_2 show the time with the luminescence intensity rapidly and slowly weakened, respectively. Among them, the A_1 , A_2 , B , τ_1 and τ_2 are obtained from the double exponential fitting curve, and the effective decay time (τ) is calculated by the following formula (2). Fig. 3b shows the decay curves of the $(\text{Gd},\text{Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ ($x = 0.1-0.8$) phosphors.

Fig. 3 (a) CIE chromaticity coordinate of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ ($x = 0.1-0.8$) phosphors. (b) Decay curves of Ce^{3+} emission monitored for the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ ($x = 0.1-0.8$) phosphors, and the inset of Fig. 3b is the variation of fluorescence lifetimes of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors with the concentration of Ga^{3+} changed from 0.1 to 0.8.

Fig. 4 The temperature-dependence behavior of $(\text{Gd,Y})_3(\text{Al}_{0.9}\text{Ga}_{0.1})_5\text{O}_{12}:\text{Ce}^{3+}$ phosphor under 459 nm excitation (a) as function of temperatures and the relative intensity with temperature (b). (c) the relationship between $\ln[(I_0/I)-1]$ and $1/kt$ for thermal quenching of the $(\text{Gd,Y})_3(\text{Al}_{0.9}\text{Ga}_{0.1})_5\text{O}_{12}:\text{Ce}^{3+}$ phosphor.

The curves can be fitted with the single exponential decay model presented. The variation of fluorescence lifetimes of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ ($x = 0.1-0.8$) phosphors are shown in the inset of Fig. 3b. From that the figure, the lifetime of phosphors decreases from 27.7019 ns to 4.9389 ns as the amount of Ga^{3+} ions increased from 10 at.% to 80 at.%. The main reason for this phenomenon is that a resonance energy transfer process between Ce^{3+} is formed with the increasing concentrations of Ga^{3+} , and part of the energy generating by the 5d-4f transition of Ce^{3+} is transferred from the resonance energy network to the surface of phosphors in the form of non-radiation.

The temperature-dependence behavior of the $(\text{Gd,Y})_3(\text{Al}_{0.9}\text{Ga}_{0.1})_5\text{O}_{12}:\text{Ce}^{3+}$ phosphor under 459 nm excitation as function of temperatures is shown in Fig. 4. It can be seen from Fig. 4a that the emission intensity of the $(\text{Gd,Y})_3\text{Al}_4\text{Ga}_1\text{O}_{12}:\text{Ce}^{3+}$ phosphor at 563 nm decreased with increasing the temperature from 50 K to 450 K, which are attributed to the thermal quenching. Moreover, considering the

practical application of phosphors in LEDs, the emission intensity of Ce^{3+} can still maintain $\sim 59.80\%$ at room temperature at 373 K (Fig. 4b).

In order to further explore the impact of temperature on thermal quenching, the explanation can be acquired by the Arrhenius equation:^[26]

$$\ln\left(\frac{I_0}{I} - 1\right) = \ln A - \frac{E_a}{kT} \quad (3)$$

where E_a , T , A and k refer to activation energy, temperature (K), constant and Boltzmann constant, respectively. The I_0 and I represent the emission intensity at room temperature and operating temperature, respectively. The relationship between $\ln(I_0/I - 1)$ and $(kT)^{-1}$ of the phosphor is drawn in Fig. 4c. The slope of the fitted linear function is -0.35, thus the activation energy E_a is approximately 0.35 eV. The relatively high activation energy achieved in this work indicates that the phosphor possesses good thermal stability, which is an excellent candidate for application in LEDs.^[27-29]

Fig. 5 PLE (a) and PL spectrum (b) of $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+0.01}$ ($y = 0-0.4$) phosphors calcined at 1500 °C.

Fig. 6 (a) CIE coordinates of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+0.01}$ ($y = 0.05-0.4$) phosphors. (b) Fluorescence attenuation curves of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+0.01}$ ($y = 0.05-0.4$) phosphors, and the inset is the decay lifetimes of $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+0.01}$ phosphors with the concentration of $\text{Mg}^{2+}/\text{Si}^{4+}$ increased from 0.05 to 0.4.

Fig. 5a shows the PLE spectra of $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors at the optimal emission wavelength. It can be seen that when emitted at 575 nm wavelength, the PLE spectrum of $(\text{Gd,Y})\text{AG}:\text{Ce}^{3+}_{0.01}$ phosphor mainly contains a strong excitation peak at ~ 469 nm (${}^2\text{F}_{5/2} \rightarrow \text{E}_{2g}$ transition of Ce^{3+}) and a secondary peak at ~ 330 nm (${}^2\text{F}_{5/2} \rightarrow \text{T}_{2g}$ transition of Ce^{3+}), which are consistent with the result of Fig. 2a. The $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0.05-0.4$) phosphors were synthesized under the same experimental conditions, and the fluorescence excitation spectrum of the series of phosphors was tested.^[30-32] It can be seen that with the incorporated amount of $\text{Mg}^{2+}/\text{Si}^{4+}$ increasing, the shape of the two excitation peaks of Ce^{3+} has basically not changed. Nevertheless, the main excitation peak has red-shifted from ~ 469 nm [$(\text{Gd,Y})\text{AG}:\text{Ce}^{3+}_{0.01}$] to ~ 487 nm [$(\text{Gd,Y})_3(\text{Al}_{0.2}\text{Mg}_{0.4}\text{Si}_{0.4})_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$].

Fig. 5b shows the PL spectra of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors under the optimal excitation wavelength.^[33-35] Under the optimal excitation wavelength at 469 nm, the optimal emission wavelength of the $(\text{Gd,Y})\text{AG}:\text{Ce}^{3+}_{0.01}$ phosphor displays a broadband emission at ~ 575 nm, which originates from the $5d-4f$ transition of Ce^{3+} . Similarly, monitoring at the optimal excitation wavelengths of these phosphors, the emission intensities of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors decrease as the concentration of $\text{Mg}^{2+}/\text{Si}^{4+}$ increasing. Furthermore, the emission wavelengths of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors red shift from ~ 576 nm to ~ 592 nm with the concentration of $\text{Mg}^{2+}/\text{Si}^{4+}$ increasing to 40 at.%, which is consistent with the results of the PLE spectra in Fig. 5a. The main reason for this phenomenon is that the Ce-O bond distance becomes shorter in aluminum oxide octahedron (AlO_8) and longer in aluminum oxide dodecahedron (AlO_{12}) with the $\text{Mg}^{2+}/\text{Si}^{4+}$ replacing Al^{3+} gradually. Furthermore, the distortion of (CeO_8) octahedron is increased, which enhances the energy level cleavage of Ce^{3+} , resulting in the reduction of $5d-4f$ energy level transition, so the phosphor exhibits a red shift in emission.

In order to further study the fluorescence properties, the Commission International de L'Eclairage (CIE) chromaticity coordinates for the emission of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors are shown in Fig. 6a. It can be seen that the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors are analyzed to have color coordinates (x, y) and color temperatures of $(\sim 0.49, \sim 0.50)$, ~ 2918 K, $(\sim 0.50, \sim 0.49)$, ~ 2749 K, $(\sim 0.52, \sim 0.48)$, ~ 2486 K, $(\sim 0.53, \sim 0.47)$, ~ 2334 K, $(\sim 0.54, \sim 0.46)$, ~ 2250 K and $(\sim 0.55, \sim 0.45)$, ~ 2058 K for $y = 0.05, 0.1, 0.2, 0.3$ and 0.4 , respectively. With the doping of $\text{Mg}^{2+}/\text{Si}^{4+}$, the luminescent color of this series of phosphors changes from yellow to orange red, which further confirms the result in Fig. 5b.

Fig. 6b displays the fluorescence attenuation curves of $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0.05-0.4$) monitoring at 576 nm. The fluorescence decay curve of $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0.05-0.4$) phosphors is obtained by

double exponential equation of (1) and (2). The embedded figure in Fig. 6b is a line diagram of the fluorescence life of $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}_{0.01}$ ($y = 0-0.4$) phosphors with the changing content of $\text{Mg}^{2+}/\text{Si}^{4+}$ ions. From which can be seen that the fluorescence life of Ce^{3+} decreases from 60 ns to 24 ns as the content of $\text{Mg}^{2+}/\text{Si}^{4+}$ ions increases from 0 to 40 at. %. The main reason can be explained that part of the energy generated by the $5d-4f$ transition of Ce^{3+} is transferred to the phosphor surface by the resonance energy network in the form of non-radiation with the increase of the content of $\text{Mg}^{2+}/\text{Si}^{4+}$.

4. Conclusions

In general, the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ and $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors were successfully prepared by co-precipitation and high temperature solid phase reaction, respectively. The combination techniques of XRD, FE-SEM, PLE/PL and fluorescence decay on the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ and $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors were performed. The $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors show good crystallinity and uniform size morphology with the concentration of Ga^{3+} changed. The emission peaks of the $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors blue shift from ~ 575 nm (yellow) to ~ 549 nm (yellow green) with the concentration of Ga^{3+} changes from 10 at.% to 80 at.%, which are attributed to lattice expansion with the Ga^{3+} ions doping. The decay time decreases gradually owing to the probability of non-radiative transitions rose. Moreover, these phosphors exhibit good high temperature stability, with the emission intensity remaining $\sim 59.80\%$ at 373 K compared to room temperature. Similarly, the emission of the $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors red shift from ~ 575 nm to ~ 592 nm with the increase of the incorporation of $\text{Mg}^{2+}/\text{Si}^{4+}$, which are derived from the increased distortion of (CeO_8) octahedron. The $(\text{Gd,Y})_3(\text{Al}_{1-x}\text{Ga}_x)_5\text{O}_{12}:\text{Ce}^{3+}$ and $(\text{Gd,Y})_3(\text{Al}_{1-2y}\text{Mg}_y\text{Si}_y)_5\text{O}_{12}:\text{Ce}^{3+}$ phosphors with good luminescent properties are expected to be widely used in lighting and display areas.

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Supporting information

Not applicable

Conflict of interest

There are no conflicts to declare.

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Author information

Shiyu Ma is the doctoral student of University of Jinan. His research field included Energy materials, Nano friction and lubrication. More than 10 papers have been published.

Bin Liu is a doctor at School of Materials Science and Engineering in University of Jinan, China. He is mainly engaged in the synthesis of perovskite nanocrystals and the construction of devices.

Bingqiang Cao received Ph.D. degree from the Institute of Solid State Physics, Chinese Academy of Sciences (CAS) in 2006. After that, he worked as a postdoctoral researcher at Universitaet Leipzig, Germany, from 2006 to 2008, and then a JSPS fellow at Kyushu University, Japan, from 2008 to 2009. He then joined the University of Jinan, China, as a principle investigator and Taishan Scholar Chair Professor. He is the author of five invited book chapters and over 100 journal papers. His group is focusing on thin films, nanostructures, heterostructures, and related semiconductor devices, especially for solar cells and light-emitting devices.

Yizhong Lu received his Ph.D. from Changchun Institute of Applied Chemistry, Chinese Academy of Sciences (2015). He then worked as a research fellow at Nanyang Technological University & Cambridge Center for Advanced Research in Energy Efficiency in Singapore. Currently, he is a full professor in the School of Materials Science and Engineering, University of Jinan, Shandong. His research interests focused on size-controlled synthesis, characterization, and size-dependent property study of metal nanoclusters, nanobiosensing and electrochemistry.

Zongming Liu joined the University of Jinan as a principle professor in 1985 and is the Young and middle-aged expert with outstanding contributions in Shandong Province. He is the author of over 60 journal papers. His group is focusing on nanomaterials, powder engineering, and thermal engineering.

Jinkai Li received Ph.D. degree from the Northeastern University, Shenyang city, China in 2014. After that, he worked as a vice professor at University of Jinan. He is the author of over 30 journal papers. He is focusing on inorganic luminescent materials, and related devices, especially for light-emitting devices.

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