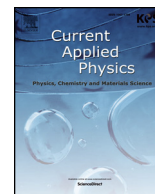




ELSEVIER

Contents lists available at ScienceDirect

Current Applied Physics

journal homepage: www.elsevier.com/locate/cap

Optimization red emission of SrMoO₄: Eu³⁺ via hydro-thermal co-precipitation synthesis using orthogonal experiment

Yongjun Tan^{a,b}, Xuedan Luo^a, Mingfu Mao^c, Dehua Shu^d, Wenfei Shan^a, Guizhi Li^a, Dongcai Guo^{a,b,*}

^a School of Chemistry and Chemical Engineering, State Key Laboratory of Chemo/Biosensing and Chemometrics, College of Chemistry and Chemical Engineering, Hunan University, Changsha, 410082, China

^b Hunan Provincial Key Laboratory for Cost-effective Utilization of Fossil Fuel Aimed at Reducing Carbon-dioxide Emissions, Changsha, 410082, China

^c Zhejiang Qianhe Biotech Lnc., Zhuji, 311800, China

^d China Geo Engineering Corporation, Beijing, 100093, China

ARTICLE INFO

Keywords:

SrMoO₄

Hydro-thermal co-precipitation method

Orthogonal experiment

ABSTRACT

In the present study, the SrMoO₄:Eu³⁺ phosphors has been synthesized through hydro-thermal co-precipitation method, and single factor and orthogonal experiment method was adopted to find optimal synthesis condition. It is interesting to note that hydro-thermal temperature is a prominent effect on the luminescent intensity of SrMoO₄:Eu³⁺ red phosphor, followed by co-precipitation temperature, calcining time, and the doping amount of Eu³⁺. The optimal synthesis conditions were obtained: hydro-thermal temperature is 145 °C, co-precipitation temperature is 35 °C, the calcining time is 2.5 h, and the doping amount of activator Eu³⁺ is 25%. Subsequently, the crystalline particle size, phase composition and morphology of the synthesized phosphors were evaluated by X-ray powder diffraction (XRD) and scanning electron microscopy (SEM). The results show that these phosphors possess a scheelite-type tetragonal structure, and the particle size is about 0.2 μm. Spectroscopic investigations of the synthesized phosphors are carried out with the help of photo-luminescence excitation and emission analysis. The studies reveal that SrMoO₄:Eu³⁺ phosphor efficiently convert radiation of 394 nm–592 and 616 nm for red light, and the luminescence intensity of SrMoO₄:Eu³⁺ phosphors is improved. SrMoO₄:Eu³⁺ phosphors may be a potential application for enhancing the efficiency of white LEDs.

1. Introduction

Rare earth luminescent materials have a very wide range of applications in the light-emitting devices, display, optical fibers [1–6]. In the recent years, rare earth doped molybdates have received considerable attention as luminescent materials due to their highly efficient light emission, excellent physical and chemical stability [7–10]. In these molybdate, Mo⁶⁺ cation is located in the symmetry center and coordinated by four oxygen atoms [11]. This structure allows the O²⁻ → Mo⁶⁺ charge transfer with a intense absorption band in the UV range. Meanwhile, the property of energy transfer from the MoO₄ groups to the dopant ions can act as antenna sensitizer [12]. Molybdates can cut down the non-radiative relaxation due to low photon energy. Thus, molybdates are considered as a promising matrix for doping by RE³⁺ ions due to these excellent properties [13].

The Eu³⁺ doped SrMoO₄, a red luminescent material, is often

prepared by conventional solid-state reactions [14] and wet chemical methods, including combustion method [15], co-precipitation methods [16] or hydro-thermal method [17,18]. The solid-state reaction methods in application to molybdate synthesis exhibit several problems, such as high temperature, long period of calcination time and irregular particle morphology. Although wet chemical methods brought some advantages, including low temperature, short reaction time and good dispersion, all these synthesis methods remain to have some problems, such as presence of organic compounds, surfactant, expensive precursors, poor crystallinity and purity. Further improvement of research methods is needed for SrMoO₄:Eu³⁺ phosphor.

In the present study, the hydro-thermal co-precipitation method [19,20] have been successfully developed to synthesize SrMoO₄:Eu³⁺ phosphor and offer some advantages such as less expensive reactants, low synthesis temperature, uniform composition. Meanwhile, the effects of activator amount, co-precipitation temperature, hydro-thermal

* Corresponding author. School of Chemistry and Chemical Engineering, State Key Laboratory of Chemo/Biosensing and Chemometrics, College of Chemistry and Chemical Engineering, Hunan University, Changsha, 410082, China.

E-mail address: dguo2001@hnu.edu.cn (D. Guo).

<https://doi.org/10.1016/j.cap.2018.08.005>

Received 16 May 2018; Received in revised form 27 July 2018; Accepted 8 August 2018

1567-1739/ © 2018 Published by Elsevier B.V. on behalf of Korean Physical Society.

temperature, calcination time and pH value on the luminescent properties of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphor were discussed in detail. The magnitudes of factors on the luminescent properties were analyzed by orthogonal experiment [21,22]. The structural analysis of $\text{SrMoO}_4:\text{Eu}^{3+}$ have been carried out using X-ray diffraction (XRD). The morphological investigations have been performed by scanning electron microscopy (SEM). Spectroscopic investigations of the synthesized phosphors are carried out with the help of photo-luminescence excitation and emission analysis.

2. Experimental

2.1. Material

Europium oxide [99.99%, Sinopharm Chemical Reagent Co., Ltd], Polyethylene glycol 200 [CP, Sinopharm Chemical Reagent Co., Ltd], Ammonium bicarbonate [AR, Sinopharm Chemical Reagent Co., Ltd], Ammonia water [AR, Sinopharm Chemical Reagent Co., Ltd], Nitric acid [GR, Sinopharm Chemical Reagent Co., Ltd], Strontium nitrate [AR, Sinopharm Chemical Reagent Co., Ltd], Ammonium molybdate [AR, Sinopharm Chemical Reagent Co., Ltd] and Absolute alcohol [99.7%, Sinopharm Chemical Reagent Co., Ltd] were used as received. Deionized water purified in a Barnstead system was used in all the experiments.

2.2. Synthesis

$\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors were synthesized by hydro-thermal co-precipitation method. At first, Polyethylene glycol were added to deionized water with magnetic stirring for 20 min at 40 °C. $\text{Sr}(\text{NO}_3)_2$ and $\text{Eu}(\text{NO}_3)_3$ were added into above obtained solution and continuous stirring for 30 min. Subsequently, $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ solution was slowly added. The mixed solution pH value was adjusted to 6 by addition of $\text{NH}_4\text{HCO}_3\text{-NH}_3\cdot\text{H}_2\text{O}$ solution and vigorously stirred for 1 h and turned into white turbid liquid. Finally, the reaction solution was transferred into a 100 mL hydro-thermal reactor and maintained at 140 °C for 12 h. After natural cooling to room temperature, the precipitate was separated by centrifugation and washed with ethanol and deionized water. The crude product was further heated at 1000 °C for 2 h, and $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors were obtained.

2.3. Characterization

Powder X-ray diffraction (XRD) pattern were recorded on a Bruker ADVANCED8 Advance X-ray diffractometer with Cu K α radiation at a scanning rate of 4°/min, speed 0.5 s, and angles from 10 to 80°, as well as tube voltage 40 kV and tube current 30 mA. Scanning electron microscopy (SEM) observation was carried out on F20 SEM (model Tecnai G2 F20 S-TWIN, the United States FEI). The preparation of sample was as follows: a trace amount of $\text{SrMoO}_4:\text{Eu}^{3+}$ was dispersed in ethanol and it was evenly mixed. The mixture was dropped with a dropper to the prepared copper grid and dried to conduct experiment. Fluorescence spectrum was measured by HITACHI F-2700 fluorescence spectrometer (operation voltage 400 V, with a slit width of 5 nm).

3. Results and discussion

3.1. Single factor optimization of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors synthesis conditions

The effects of activator amount, co-precipitation temperature, hydro-thermal temperature, calcination time and pH value on the luminescent intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors were studied. The activator amount is expressed by the molar ratio of europium nitrate and strontium nitrate $n_{\text{Eu}}/n_{\text{Sr}}$, co-precipitation temperature is expressed by T_p , hydro-thermal temperature by T_h , hydro-thermal time by

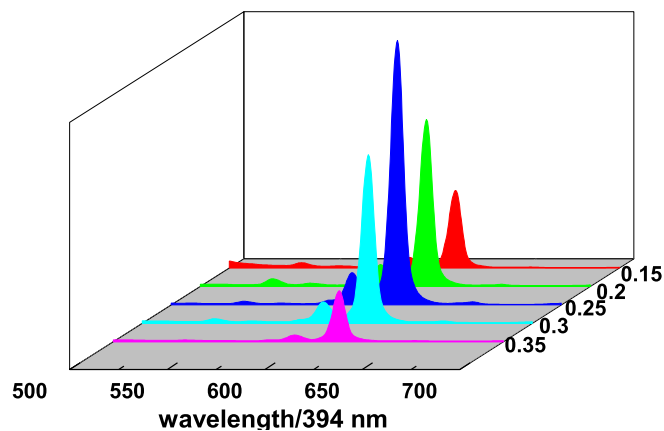


Fig. 1. The emission spectra of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors prepared under different activator amount ($n_{\text{Eu}}/n_{\text{Sr}}$) ($\lambda_{\text{ex}} = 394 \text{ nm}$).

t_h , calcination time by t_c , calcination temperature by T_c .

3.1.1. Effect of activator amount on the luminescent intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors

Fixed $T_p = 50 \text{ }^\circ\text{C}$, $T_h = 140 \text{ }^\circ\text{C}$, $\text{pH} = 5$, $t_h = 8 \text{ h}$, $t_c = 2 \text{ h}$, $T_c = 1000 \text{ }^\circ\text{C}$, the effect of $n_{\text{Eu}}/n_{\text{Sr}}$ on the luminescent intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ was explored by emission spectra analysis. The emission spectra are shown in Fig. 1.

It is observed from Fig. 1 that the luminescent intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors increased with the increase of $n_{\text{Eu}}/n_{\text{Sr}}$ from 0.15 to 0.25 and obtained the maximum at $n_{\text{Eu}}/n_{\text{Sr}} = 0.25$. Further increase of $n_{\text{Eu}}/n_{\text{Sr}}$ resulted in quenching emissions of $\text{SrMoO}_4:\text{Eu}^{3+}$ phosphors. The results imply that the increase of $n_{\text{Eu}}/n_{\text{Sr}}$ has both positive and negative effects. This is ascribed to concentration quenching. The increase of $n_{\text{Eu}}/n_{\text{Sr}}$ from 0.15 to 0.25 can raise activator amount. Moreover, nonradiative transition would not exist due to the weaker interaction between the $n_{\text{Eu}}/n_{\text{Sr}}$ in the range of 0.1 from 0.25. Therefore, good results have been obtained with the increase of $n_{\text{Eu}}/n_{\text{Sr}}$ from 0.15 to 0.25. While $n_{\text{Eu}}/n_{\text{Sr}}$ was greater than 0.25, the resonance between activators has occurred, which cause the migration of energy between activator ions. Thus, the luminescent intensity decreased in the range from 0.25 to 0.35. So the optimal activator amount is $n_{\text{Eu}}/n_{\text{Sr}} = 0.25$.

3.1.2. Effect of co-precipitation temperature

Fixed $n_{\text{Eu}}/n_{\text{Sr}} = 0.25$, $\text{pH} = 5$, $T_h = 140 \text{ }^\circ\text{C}$, $t_h = 8 \text{ h}$, $t_c = 2 \text{ h}$, $T_c = 1000 \text{ }^\circ\text{C}$, the effect of co-precipitation temperature on luminescent intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ was explored by emission spectra analysis. The emission spectra are shown in Fig. 2.

It can be seen from Fig. 2 that there is an increase in the luminescence intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ with co-precipitation temperature from 30 °C to 40 °C. The optimal co-precipitation temperature is 40 °C. A regular decrease in the luminescent intensity of $\text{SrMoO}_4:\text{Eu}^{3+}$ was observed with co-precipitation temperature from 50 °C to 70 °C. The result is attributed to different ion diffusion rate at different co-precipitation temperatures. At co-precipitation temperature 30 °C, the slow ion diffusion rate in co-precipitation reactions causes incomplete grains, which eventually led to weaker luminescent intensity. On the contrary, a higher diffusion of $\text{Sr}(\text{NO}_3)_2$ and $\text{Eu}(\text{NO}_3)_3$ exists at the co-precipitation temperature 40 °C and hinder local precipitation, which get the best luminescence performance. Greater ion diffusion rate exists at higher co-precipitation temperatures, but the luminescent intensity gradually reduces at the range from 40 °C to 70 °C, which might be an effect from the formation of bad crystallinity under the greater ion diffusion rate. Thus, the best co-precipitation temperature is 40 °C.

Download English Version:

<https://daneshyari.com/en/article/11008846>

Download Persian Version:

<https://daneshyari.com/article/11008846>

[Daneshyari.com](https://daneshyari.com)