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2017

Que, M., Que, W., Zhou, T., Shao, J., & Kong, L. (2017). Enhanced photoluminescence property of sulfate ions modified YAG:Ce³⁺ phosphor by co-precipitation method. *Journal of Rare Earths*, 35(3), 217-222.

<https://hdl.handle.net/10356/84089>

[https://doi.org/10.1016/S1002-0721\(17\)60902-5](https://doi.org/10.1016/S1002-0721(17)60902-5)

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Enhanced Photoluminescence Property of Sulfate Ions Modified

YAG: Ce³⁺ Phosphor by Co-Precipitation Method

QUE Meidan (阙美丹)¹, QUE Wenxiu (阙文修)^{1,*}, ZHOU Ting (周婷)¹, SHAO Jinyou (邵金友)¹,
KONG Lingbing (孔令兵)²

¹*Electronic Materials Research Laboratory, International Center for Dielectric Research, Key Laboratory of the Ministry of Education, School of Electronic & Information Engineering, State Key Laboratory for Manufacturing Systems Engineering, Xi'an Jiaotong University, Xi'an 710049, Shaanxi, People's Republic of China*

²*School of Materials Science and Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798, Singapore*

Abstract: The sulfate ions modified YAG: Ce³⁺ phosphors were prepared by a co-precipitation method and characterized by X-ray diffraction, transmission electron microscopy, and photoluminescence. Effects of sulfate ions on the photoluminescence (PL) property of the as-prepared YAG: Ce³⁺ phosphors were studied, with sodium dodecyl sulfate (SDS) being added to R³⁺ (Ce³⁺, Y³⁺, Al³⁺) ions. Results indicate that pure YAG: Ce³⁺ phosphors with different ratios of sulfate ions can be easily obtained by calcining the as-synthesized precursor at 950°C for 2 h, the YAG: Ce³⁺ phosphors with an optimal weight ratio of 3.5 wt.% SDS have a high emission intensity and excellent dispersion behavior, and the fluorescence decay of the as-obtained YAG: Ce³⁺ phosphors is related to the lattice defect, reabsorption and cross correlation. Furthermore, thermal quenching properties of the YAG: Ce³⁺ phosphors and the YAG: Ce³⁺ phosphors with 3.5 wt.% SDS were also discussed, indicating the YAG: Ce³⁺ with SDS phosphors could have potential applications in the daylight LEDs or warm white LEDs.

Keywords: YAG: Ce³⁺ phosphor; sodium dodecyl sulfate; uniform dispersion; luminescence properties

Yttrium aluminum garnet (Y₃Al₅O₁₂) is an attractive inorganic compound because of its excellent

Foundation item: This work was supported by the Research Fund for Doctoral Program of Higher Education of China under the Grant 20120201130004, the Science and Technology Developing Project of Shaanxi Province under Grant No. 2015KW-001, the National Natural Science Foundation of China under Grant No. 61078058, partially by the National Natural Science Foundation of China Major Research Plan on Nanomanufacturing under the Grant No. 91323303.

* **Corresponding author:** QUE Wenxiu (E-mail: wxque@mail.xjtu.edu.cn; Tel: +86-29-83395679)

optical, physical and chemical properties, which has been widely applied as hosts for lasers and phosphors^[1]. Ce^{3+} ion usually serves as a dopant since it has a lower $5d$ state, and electrons at $5d$ state can easily return to ground state with radiative transition, i.e. luminescence process. Hence, Ce^{3+} ions bring new applications, such as tunable solid-state lasers over an effective spectral range^[2, 3]. So far, YAG: Ce^{3+} phosphor has been the best phosphors that can be utilized acceptably in white LED commercial market. Especially, any modification in the luminescence of YAG: Ce^{3+} phosphor is exceedingly valuable to raise the light efficiency for white LED. Thus, much effort has been made to develop technical uses for preparing YAG powders with high crystallinity, small particle size and regular morphology. The conventional method to synthesize the commercial YAG phosphors is based on solid state reaction process, which requires a high-temperature of 1600 °C and aging for a long time^[4] to gain the pure phase. However, the as-obtained particulates are large size and irregular shape, which brings negative influence to luminescent properties.

In recent years, many wet chemical methods, which include spray pyrolysis method, glycothermal method, co-precipitation method and combustion process^[5-10], were used to prepare the YAG precursor. Among them, co-precipitation method combines the advantages (high purity, fine grains and homogenous composition) of chemical methods with low temperature process, convenient process control, cheap precursors and large mass production. However, YAG powders synthesized by wet chemical methods have to face a problem of agglomeration, which usually results from the drying process. Recent studies indicate that the similar problem also happens to the preparation of SnO_2 ^[11], Y_2O_3 ^[12], TiO_2 and mesoporous silica^[13], which, as a matter of fact, can be avoided by an addition of sulfate ions. Actually, sulfate ions have been used in the synthesis of the YAG powders as reported in Ref. [14], but the influence of the sulfate ions on the morphology and luminescence of the YAG: Ce^{3+} phosphors has still not been clarified yet.

In present work, YAG: Ce^{3+} phosphors were prepared by the co-precipitation method, where ammonium hydrogen carbonate (AHC) was used as precipitant and sodium dodecyl sulfate (SDS) was used as anionic surfactant to reduce agglomeration of the powders. In addition, effects of sulfate ions on the morphology and photoluminescence of the YAG: Ce^{3+} phosphors are also studied.

1 Experimental

1.1 Synthesis

YAG: Ce^{3+} phosphor was prepared by the reverse-strike co-precipitation method. Experimental

process is shown in Fig. 1. Y_2O_3 , $Al(NO_3)_3 \cdot 9H_2O$ and $Ce(NO_3)_3 \cdot 6H_2O$ were used as initial materials, which were precisely weighted according to the formula $Y_{2.94}Ce_{0.06}Al_5O_{12}$ with stoichiometric ratio ^[15]. Yttrium nitrate solution was prepared by dissolving Y_2O_3 in distilled water with a certain amount of HNO_3 . The obtained nitrate salt solution was mixed with cerium nitrate solution and aluminum nitrate solution. Then, the nitrate salts solution was instilled into the solution of ammonium hydrogen carbonate (AHC) in distilled water and ethanol. Different amounts of SDS were added into the NH_4HCO_3 solution in order to alleviate agglomeration phenomenon. The weight of SDS addition was well-controlled to vary in a range from 0 to 10.5 wt.% in different samples as listed in Table 1. The concentration ratio of NH_4HCO_3 to R^{3+} was 2.5. During the titration process, pH value of the solution was kept at 7.2-7.8 and the procedure was conducted at room temperature with vigorous stirring. When this process was finished, the final solution was aged at room temperature for 24 h, so as to allow Ce^{3+} entering into the YAG lattice adequately. After that, the precipitation was collected by centrifuging and washed with distilled water and ethanol three times respectively to remove impurities and make a better dispersion. Finally, the precursor powders were obtained by drying in oven at 75°C.

1.2 Characterization

Phase composition of the as-prepared phosphor powders was examined by X-ray diffraction (XRD) analysis, with Cu K α radiation at 40 kV and 100 mA. Scanning speed was 15° min⁻¹ at step of 0.02°. Thermal behaviors of the precursor powders were characterized by differential scanning calorimetry and thermogravimetry (DSC/TG) analysis using a thermal analyzer (NETZSCH STA 449C, Germany) at a heating rate of 10 °C min⁻¹ in air. Transmission electron microscopy (TEM, JEM-200CX, JEOL Inc., Japan) was used to observe the morphological properties of YAG: Ce phosphors. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra of the as-prepared phosphors were also measured by using a FLS-920T fluorescence spectrophotometer (Edinburgh, UK) equipped with a 450 W Xe light source and double excitation monochromators.

2 Results and discussion

The YAG: Ce^{3+} phosphor was prepared by the reverse-strike co-precipitation process with AHC as precipitant, which leads to the hydrolysis reaction processes as below ^[16],

It is well known that Al^{3+} precipitates as $AlOOH$ or $NH_4Al(OH)_2CO_3$, whereas Y^{3+} precipitates as $[Y_2(CO_3)_3 \cdot nH_2O (n=2-3)]$ or $[Y(OH)CO_3]$.

Figure 2a shows the XRD patterns of the YAG: Ce^{3+} phosphors with different ratios of sulfate ions, which displays the cubic garnet phase after being calcined at 950 °C for 2 h. There are no any diffraction peaks that are consistent with starting materials or impurities, implying that a pure crystalline compound is obtained. The fact that no other phase or impurity was detected, suggesting that Ce^{3+} ions were adequately dissolved in YAG matrix without inducing significant changes of the crystal structure. The obtained diffraction pattern was well parallel and consistent with the standard JCPDS card no.33-40 ^[17]. As seen in Fig. 2b, TG-DTA curves of the precursors show the exothermic peak up to about 1000°C and a total weight loss of about 50%. Weight loss of the precursor below 400°C is from the evaporation of the ammonia and molecular water, as well as partial decomposition of CO_3^{2-} , while further weight loss at higher temperature is mainly due to the further decomposition of the carbonate species ^[18]. There is an obvious endothermic peak at 187°C, which corresponds to the evaporation of water. CO_2 and NH_3 are released during the decomposition process of the precipitates, leading to a broad exothermic band between 200°C and 800°C. A significant exothermic peak at 927°C should be attributed to the formation of YAG phase, as evidenced by XRD results in Fig. 2a. These results indicate that pure YAG phase can be obtained after the precursor is calcined at 950°C.

Figure 3 shows small angle XRD patterns of the YAG: Ce^{3+} phosphors calcined at 950° C for 2 h and with different ratios of sulfate ions, including 0 wt.%, 3.5 wt.%, 7 wt.%, 10.5 wt.%. It can be observed that the diffraction peak location of all the samples is basically identical, but the diffraction intensity of the 3.5 wt.% sample is the strongest and followed by 0 wt.%, 7 wt.% and 10.5 wt.% samples, respectively.

Figure 4 displays TEM images of the YAG: Ce^{3+} phosphors with different ratios of sulfate ions and calcined at 950°C for 2 h. The phosphor powders without SDS have an obvious agglomeration, which is improved as an addition of 3.5 wt. % SDS (Sample 2) is introduced as shown in Fig. 4b. However, as the addition content of SDS increases to 7 wt. % (Sample 3), agglomeration appears again. In addition, the morphology of Sample 2 looks better than those of other samples. When the sulfate ions are added, the electronegative property of the precursor increases ^[16], thus, giving rise to the repulsive force among the precursor particles. An increase of the repulsive force would largely reduce

collision caused by Brownian motion among the particles, which contributes to a good dispersion behavior. Hence, agglomeration phenomenon is alleviated. According to the literature as reported in Ref. 19, sulfate ions have a higher decomposition temperature and can absorb the surface precipitants, which also contributes to a better dispersion of the powders. On the other hand, excessive addition of sulfate ions may lead to an opposite effect, so that YAG: Ce³⁺ particle agglomeration emerges again as shown in Fig. 4 c-d. That is to say, to get optimal morphology and dispersion behavior of the YAG: Ce³⁺ phosphors, the ratio of sulfate ion should be well controlled.

The YAG: Ce³⁺ phosphors show a broad emission band at the range of 500-700 nm under an excitation of 460 nm as shown in Fig. 5a. The emission spectra can be ascribed to two energy level transition, that is, ⁵D₀-⁷F₁ at the maximum position (550 nm) and ⁵D₀-⁷F₂ [20]. The excitation spectrum for Ce³⁺ includes two strong band peaks at 342 and 460 nm [21], respectively. These excitation bands induce the electron transitions from the ground state of Ce³⁺ (²F_{2/5}) to different crystal field splitting components of the excited 5d state. The relative intensity of the two bands is evolved with increasing the content of the sulfate ions due to the surface absorption. Obviously, the excitation and emission intensities of Sample 2 show the maximum value in all measured samples, which can be attributed to the better dispersion behavior of Sample 2. These results are in accordance with the analysis of TEM images. Furthermore, the corresponding CIE chromaticity diagram and the chromaticity coordinate values of the YAG: Ce³⁺ without doping S-ions are also shown in Fig. 6 and Table II, respectively. It should be mentioned here that since the locations of the emission peaks of different samples are almost close, thus, only the chromaticity coordinates of Sample 1 and Sample 2 are marked in Fig. 6 (i.e., a and b points).

A typical result of the decay fluorescence lifetime of Sample 2 is presented in Fig. 7. Theoretically, Ce³⁺ ion only occupies the dodecahedral lattice site, so the decay curve of perfect crystal structure can be fitted by single exponential components. However, a portion of sulfur or other elements might replace a part of oxygen element, which causes a new lattice site in a small range. Furthermore, it is unavoidable to introduce defects during the prepared process. These factors may induce the deviation between the single exponential fitting and the experimental results. Based on the above analysis, we fit the decay curve of Sample 2 in our work by single-, two-, three-exponential components, among which the three-exponential decay is the most satisfactory one. Therefore, we here propose the assumption of a three-exponential decay. Thus, this experiment was conducted under 460 nm excitation and

monitored at 550 nm. The black line in Fig. 7 indicates that the experimental intensity depends on the amplitude modulation, and the red line depicts the best fitting result. A satisfactory fit is achieved under assumption of a three-exponential decay and based on the following equation ^[22, 23],

$$I = A_1 \exp(-t / \tau_1) + A_2 \exp(-t / \tau_2) + A_3 \exp(-t / \tau_3) \quad (5)$$

where I represents luminescence intensity, A_1 , A_2 and A_3 are constants, t is the time, τ_1 , τ_2 , and τ_3 are the respective short and long lifetimes for the exponential components. Based on these parameters, the average decay time can be obtained using the following formula ^[23].

$$\tau^* = (A_1 \tau_1^2 + A_2 \tau_2^2 + A_3 \tau_3^2) / (A_1 \tau_1 + A_2 \tau_2 + A_3 \tau_3) \quad (6)$$

Average decay time is summarized in Fig. 7. The decay constant τ_1 (7.12 ns) maintains a fast decay due to tiny defects of the host lattice ^[24]. τ_2 (36.78 ns) can be attributed to the reabsorption of photon followed by nonradiative relaxation under excitation as shown in Fig. 5b. τ_3 (106.85 ns) is owing to the nonradiative decay through cross correlation because of the energy exchange between two neighboring activator ions, the migration under the excitation from one ion to another throughout the crystal (the hopping mechanism) ^[25], and the clustering of Ce^{3+} . The average Ce^{3+} lifetime detected at 550 nm is determined to be 68.72 ns as presented, which is consistent with that as reported in Ref. [26].

Thermal quenching behavior is an important factor in evaluating the luminescent properties of the phosphor, and a number of studies have discussed the thermal quenching behaviors ^[27-31]. The temperature-dependent luminescent properties of Sample 1 and Sample 2 are measured at temperature range of 293-533 K, and the results are shown in Fig. 8. The thermostability of Sample 1 is almost equal to that of commercial YAG: Ce^{3+} before 400 K, however, the emission intensity of Sample 1 is lower than that of commercial YAG: Ce^{3+} . It can be also seen that the emission intensity of Sample 2 decreases more rapidly than that of commercial YAG: Ce^{3+} with increasing measurement temperature, indicating that the thermostability of Sample 2 is inferior to that of commercial YAG: Ce^{3+} . Furthermore, the relative emission intensity of Sample 1 is the lowest, followed by Sample 2 and commercial YAG: Ce^{3+} at 533 K.

3 Conclusions

The YAG: Ce^{3+} phosphors have been successfully prepared by the co-precipitation process, where AHC is used as precipitant and SDS as dispersing promoter. The as-synthesized precursor can be converted to single phase YAG by calcining at 950°C for 2 h. Results indicate that sulfate ion has significant influence on the dispersion behavior of the YAG: Ce^{3+} phosphors. The morphology and

dispersion behavior of the YAG: Ce³⁺ phosphors have an important effect on the luminescent property. Among different samples with different weight ratios of SDS, it is found that the YAG: Ce³⁺ phosphors with an addition of an optimal weight ratio of 3.5 wt.% SDS show the best luminescence property in all measured samples. These results indicate that the introduction of a proper amount of the sulfate ions has positive effect on controlling the morphology and enhancing the emission intensity of the samples. Besides, the lattice defect, reabsorption and cross correlation all have influence on the decay fluorescence time.

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Figure Captions

Fig. 1 Flow chart of YAG: Ce³⁺ phosphors prepared by reverse-strike co-precipitation method

Fig. 2 a) XRD patterns of YAG: Ce³⁺ phosphors with different ratios of sulfate ions including 0 wt.%, 3.5 wt.%, 7 wt.%, 10.5 wt.%. b) DSC-TG curves of the as-prepared YAG precursors

Fig. 3 Small angle XRD patterns of YAG: Ce³⁺ phosphors calcined at 950°C for 2 h with different ratios of sulfate ions, (a) 0 wt.%, (b) 3.5 wt.%, (c) 7 wt.%, (d) 10.5 wt.%

Fig. 4 TEM images of YAG: Ce³⁺ phosphors calcined at 950°C for 2 h with different ratios of sulfate ions, (a) 0 wt.%, (b) 3.5 wt.%, (c) 7 wt.%, (d) 10.5 wt.%

Fig. 5 a) PLE ($\lambda_{\text{em}} = 550 \text{ nm}$) and PL ($\lambda_{\text{ex}} = 460 \text{ nm}$) spectra of Sample 1, 2, 3 and 4, b) Energy-level diagram of YAG: Ce³⁺ phosphors

Fig. 6 CIE chromaticity diagram for Sample 1 (a) and Sample 2 (b)

Fig. 7 PL spectrum decay results of Sample 2 and the fitting result

Fig. 8 Temperature-dependent emission intensities of a) Sample 1 and b) Sample 2, c) temperature-dependent relative emission intensities of Sample 1, Sample 2 and commercial YAG: Ce³⁺

Table I Weight ratios of (NH₄)₂SO₄ and SDS to R³⁺ (Y³⁺, Ce³⁺, Al³⁺)

NO.	Sample 1	Sample 2	Sample 3	Sample 4
Content of SDS	0 wt. %	3.5 wt. %	7 wt. %	10.5 wt. %

Table II The chromaticity coordinates and peaks' location of samples

	x	y	Peak (nm)
Sample 1	0.4213	0.5444	543
Sample 2	0.4272	0.5417	553
Sample 3	0.4285	0.5415	553
Sample 4	0.4186	0.5484	543

Fig. 1

Fig. 2

Fig. 3

*Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8