



# Preparation and photoluminescence property of a loose powder, $\text{Ca}_3\text{Al}_2\text{O}_6:\text{Eu}^{3+}$ by calcination of a layered double hydroxide precursor

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## ABSTRACT

A novel red light-emitting material,  $\text{Ca}_3\text{Al}_2\text{O}_6:\text{Eu}^{3+}$ , which is the first example found in the  $\text{Ca}_3\text{Al}_2\text{O}_6$  host, was prepared by calcination of a layered double hydroxide precursor at 1350 °C. The precursor,  $[\text{Ca}_{2.9-x}\text{Al}_2\text{Eu}_x(\text{OH})_{9.8}](\text{NO}_3)_{2+x} \cdot 2.5\text{H}_2\text{O}$ , was prepared by coprecipitation of metal nitrates with sodium hydroxide. The material is a loose powder composed of irregular particles formed from aggregation of particles of a few nanometers, as shown in scanning electron microscope (SEM) images. It was found that the photoluminescence intensity reached the maximum when the calcination temperature was 1350 °C and the concentration of  $\text{Eu}^{3+}$  was 1.0%. The material emits bright red emission at 614 nm under a radiation of  $\lambda = 250$  nm.

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## 1. Introduction

Photoluminescent materials have been widely used in display panels, lasers and, also possibly, in window glasses for green houses. Usually, the materials are prepared from ceramic routes (solid state reactions at high temperatures) [1–5], spray pyrolysis of a solution of component elements [6,7], calcination of precursor compounds [8–11], as well as some liquid phase routes [12]. The present work contributes to the precursor method, as it can produce materials in one step without tedious and time-after-time grinding, or special equipments.

Layered double hydroxides (LDHs) have been used as precursors for oxide materials for a long time. They can be expressed by a typical formula  $[\text{M}_{1-x}\text{M}'_x(\text{OH})_2]^{x+}\text{A}^{n-}_{x/n} \cdot m\text{H}_2\text{O}$ , in which  $M$  and  $M'$  could be almost any of the bivalent and trivalent cations, respectively,  $A^{n-}$  could be any of the anions and  $x$  could be any value between 0.2 and 0.33. They are structured with positively charged hydroxide layers and anions in between the layers. [13]. Extended researches on LDHs have shown that their composition can be finely tuned by adjusting  $M$ ,  $M'$ ,  $x$  and  $A$ . And, more importantly, more than two kinds of metal cations can be compounded into a single compound by introducing the cations into the layer cations and/or into the interlayer complex anions, thus producing a LDH precursor with required components of required ratio, and the components are mixed uniformly on an atomic scale [13,14].

For example, calcination of a LDH precursor containing  $M^{2+}$ ,  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  cations ( $M^{2+} = \text{Mg}, \text{Co}, \text{Ni}$ ) with,  $[\text{M}^{2+}]/([\text{Fe}^{2+}]+[\text{Fe}^{3+}]) = 0.5$  produces a pure ferrite spinel,  $\text{MFe}_2\text{O}_4$ , which has higher saturation magnetization than the same material produced by conventional ceramic routes [15]. LDHs have also been used to prepare catalysts for many years [16–19].

We have started a project recently to produce rare earth cation-activated luminescent materials. To the best of our knowledge, there are only a few reports on introducing anions with europium into the interlayer space of LDHs to make luminescent materials [20,21]. But few authors have tried to incorporate rare earth cations into LDH hydroxide layers. Here we report the preparation of a LDH precursor with  $\text{Eu}^{3+}$  in the hydroxide layer by coprecipitation, and calcination of it to produce a loose powder material,  $\text{Ca}_3\text{Al}_2\text{O}_6:\text{Eu}^{3+}$ , in which the activator  $\text{Eu}^{3+}$  is hosted in the lattice of  $\text{Ca}_3\text{Al}_2\text{O}_6$ . It has been known that  $\text{Ca}_3\text{Al}_2\text{O}_6$  has infrared transmission up to  $6\mu\text{m}$  [22,23] and has never been reported as a host for phosphors.

## 2. Experimental section

### 2.1. Reagents

$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ,  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ,  $\text{NaNO}_3$  and acetone were purchased reagents with purity better than 99%,  $\text{NaOH}$  with purity of 96% and  $\text{Eu}_2\text{O}_3$  with purity of 99.99%. Distilled water was boiled to decarbonize before being used.

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## 2.2. Preparation of $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$

$[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$  was prepared according to a literature method [24].  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  (17.0 g) and  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (11.7 g) were dissolved in  $\text{CO}_2$ -free distilled water to make a mixed salt solution.  $\text{NaOH}$  (6.25 g) and  $\text{NaNO}_3$  (9.1 g) were dissolved in  $\text{CO}_2$ -free distilled water to make an alkali solution. The two solutions were simultaneously added to a flask and mixed for several minutes under vigorous stirring. After the resulting slurry was aged at  $80^\circ\text{C}$  for 24 h, the solid was filtered out, washed three times with distilled water and acetone, and dried at  $50^\circ\text{C}$  in vacuum for 24 h.

## 2.3. Preparation of Eu-doped LDHs

With a similar method as above, LDH precursors doped with different concentrations of  $\text{Eu}^{3+}$  were prepared by adding certain amount of  $\text{Eu}(\text{NO}_3)_3$ , obtained from dissolution of  $\text{Eu}_2\text{O}_3$  powder in dilute  $\text{HNO}_3$ , into a solution mixture of  $\text{Al}(\text{NO}_3)_3$  and  $\text{Ca}(\text{NO}_3)_2$ . The concentration of  $\text{Eu}^{3+}$  was calculated according to  $[\text{Eu}^{3+}]/([\text{Eu}^{3+}] + [\text{Ca}^{2+}] + [\text{Al}^{3+}])$ . Thus, six samples were synthesized with the  $\text{Eu}^{3+}$  concentrations of 0.005, 0.008, 0.01, 0.012, 0.015 and 0.02, respectively. They will be denoted as samples 1–6 hereafter.

## 2.4. Preparations of the oxides

The  $\text{Eu}^{3+}$ -doped LDHs were calcined at different temperatures for 3 h in a furnace. After calcination the samples were cooled to room temperature in the furnace naturally, then taken out and stored in a vacuum desiccator.

## 2.5. Characterization

All the samples were characterized by X-ray diffraction (XRD) at room temperature, using a  $\text{CuK}\alpha_1$  radiation ( $\lambda = 0.15406$  nm) working at 40 kV, 70 mA. They were step-scanned at a rate of  $0.02^\circ/\text{s}$  from  $3^\circ$  to  $60^\circ$ . Photoluminescence emission and excitation spectra of the samples were taken on an F900 fluorescence spectrometer (Edinburgh Instrument Company Ltd.) with a xenon lamp of 450 W at room temperature. The effect of  $\text{Eu}^{3+}$  concentrations on photoluminescence properties was investigated with a PR-302 relative brightness meter (Zhejiang University Sensing Instruments Company Ltd.). Metal contents of the samples were determined by J-A1100 inductively coupled plasma (ICP) spectrometer (Jarrell-Ash Corporation). The morphology of the materials was observed using a LEO 1530VP scanning electron microscope (SEM, LEO Company, Germany).

## 3. Results and discussion

### 3.1. The structure of the LDH precursors

Fig. 1 shows the XRD patterns of the LDH precursors and the well-known  $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$ . It is obvious that all of them share great similarities; therefore, the samples must have similar structures. All the patterns can be indexed on a trigonal cell ( $P\text{-}3c1$ ), and by which the lattice parameters were obtained. Fig. 2 shows the plots of cell parameters versus concentration of  $\text{Eu}^{3+}$  for samples 1–6. It can be seen that the cell parameter  $a$  linearly increases with the  $\text{Eu}^{3+}$  concentration from samples 1 up to 6, and all the values are larger than that of  $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$ . The basal spacings ( $= c/2$ ) for all the LDHs are about 0.88 nm, which implies that  $\text{Eu}^{3+}$  ions are in the hydroxide layer, because they should be much larger otherwise; and the interlayer anion should be  $\text{NO}_3^-$  as

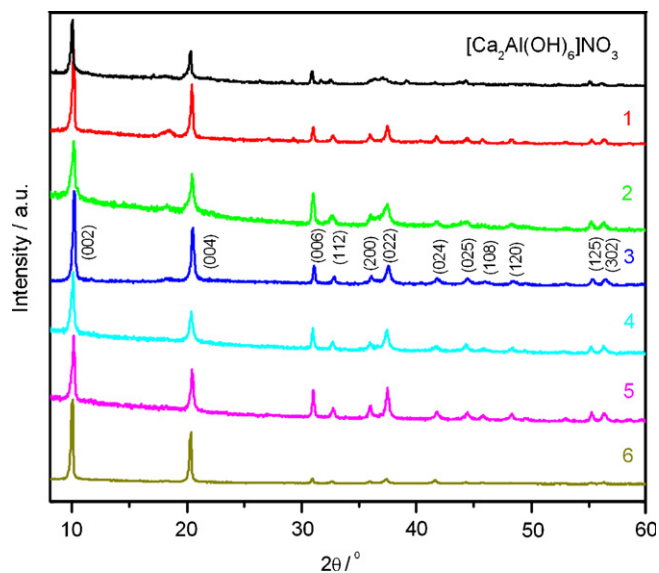


Fig. 1. XRD patterns of  $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$  and LDH precursors 1 through 6.

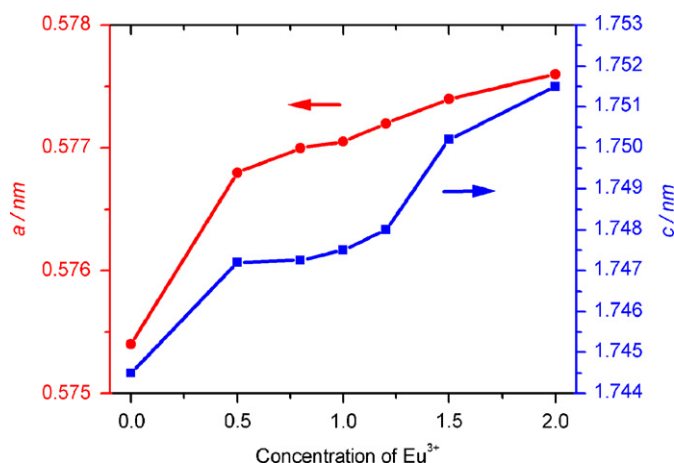


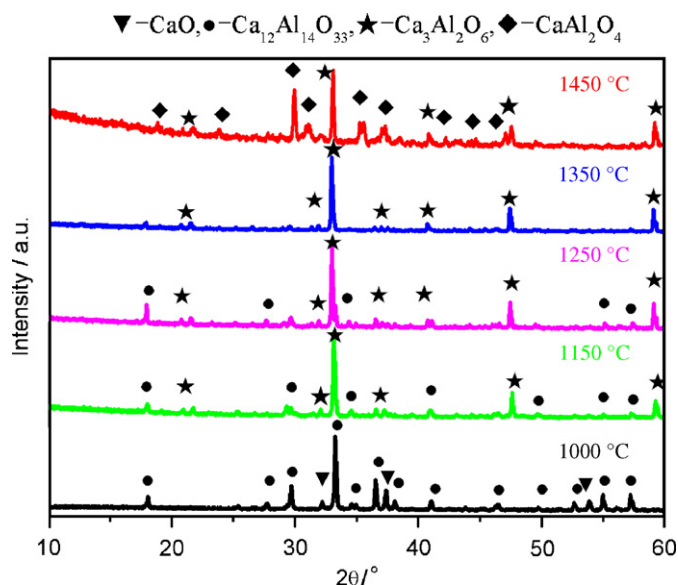
Fig. 2. The lattice parameters ( $a$  and  $c$ ) of  $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$  and LDH precursors 1 through 6 obtained from indexing on a trigonal cell ( $P\text{-}3c1$ ).

it is in  $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{NO}_3$ . No  $\text{Eu}(\text{OH})_3$  phase was found in the samples by any means used in this study.

The radii of  $\text{Eu}^{3+}$ ,  $\text{Al}^{3+}$  and  $\text{Ca}^{2+}$  are 0.95, 0.54 and  $1.00 \text{ \AA}$ , respectively, when their coordination numbers are all six. As it is generally believed that impurity ions always take place of the ions of similar radii, it is possible that small amount of  $\text{Eu}^{3+}$  should occupy some of the  $\text{Ca}^{2+}$  sites. An ICP analysis of the sample 3 showed that contents of Ca, Al and Eu were 22.11%, 10.49% and 1.54%, while those calculated from  $[\text{Ca}_{2.85}\text{Al}_2\text{Eu}_{0.05}(\text{OH})_{9.8}](\text{NO}_3)_{2.05} \cdot 2.5\text{H}_2\text{O}$  were 22.20%, 10.49% and 1.48%, respectively.

### 3.2. The characterization of calcined products of the LDH precursors

To reduce the amount and cost of the work sample 3 was arbitrarily selected to be calcined at 1000, 1150, 1250, 1350 and  $1450^\circ\text{C}$ , respectively, to find the best calcination temperature. The XRD patterns of the resulted materials are shown in Fig. 3. It is found that the diffraction peaks of the calcined product at  $1000^\circ\text{C}$  can be indexed partially with lattice parameters of CaO (JCPDS file no. 48-1467) and partially with those of  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  (JCPDS file no. 48-1882). As the calcination temperature increases, the CaO



**Fig. 3.** XRD patterns of calcined products of LDH precursor 3 at temperatures indicated on the right ends of the curves.

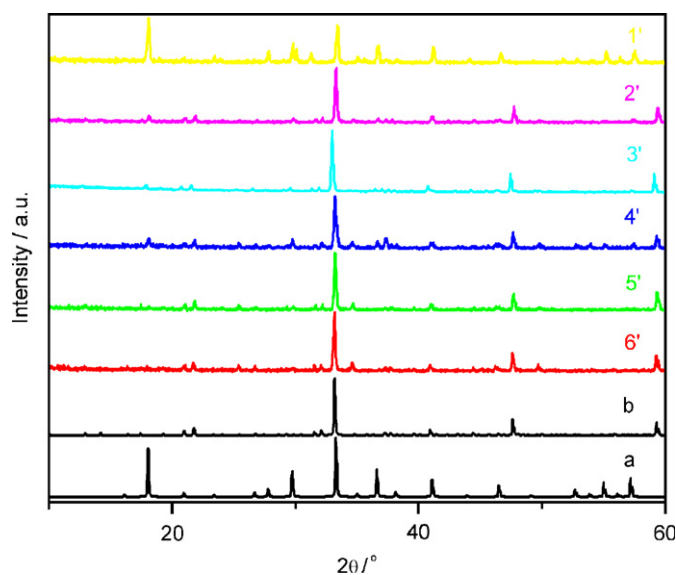
phase disappears and a new phase,  $\text{Ca}_3\text{Al}_2\text{O}_6$  (JCPDS file no. 38-1429), forms instead. At 1350 °C, only  $\text{Ca}_3\text{Al}_2\text{O}_6$  can be identified. Finally, at 1450 °C the calcined product contains  $\text{CaAl}_2\text{O}_4$  (JCPDS file no. 70-0134) together with  $\text{Ca}_3\text{Al}_2\text{O}_6$ . This is slightly different from what the phase diagram [23] of  $\text{CaO}-\text{Al}_2\text{O}_3$  predicts; since the phase diagram tells that at the composition,  $\text{Ca}_3\text{Al}_2\text{O}_6$  could coexist with  $\text{CaO}$  and  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  but not with  $\text{CaAl}_2\text{O}_4$  under 1700 °C. Therefore, the precursor method could supply phases without thermodynamic stability at the investigated conditions.

All the five samples were viewed under a UV light of 254 nm, which showed that the sample obtained at 1350 °C emitted the brightest red light. Therefore, all the LDH precursors were calcined at 1350 °C. The samples as-prepared are denoted as 1' through 6' hereafter according to the sample numbers of the precursors. The sample 3' was also analyzed with an ICP spectrometer, which gave Ca: 42.10%, Al: 20.13%, Eu: 2.92%; those calculated from  $\text{Ca}_{2.85}\text{Al}_2\text{Eu}_{0.05}\text{O}_{5.925}$  were 42.21%, 19.94% and 2.81%, respectively. It can be seen that the molar ratio of metallic cations is exactly the same as the precursor 3.

Fig. 4 shows the XRD patterns of samples 1' through 6'. It can be identified that each of the samples, except sample 3', contains not only  $\text{Ca}_3\text{Al}_2\text{O}_6$  but also  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  more or less. All the peaks in the patterns are indexed either on cubic cells of symmetry group  $Pa\bar{3}$  for  $\text{Ca}_3\text{Al}_2\text{O}_6$ , or  $I-43d$  for  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ . The lattice parameters obtained are shown in Table 1. It can be seen that  $a$  increases as the concentration of  $\text{Eu}^{3+}$  reaches 1.0% (in sample 3') and then decreases. It is also noticed that  $a$  of  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  changes with the concentration of  $\text{Eu}^{3+}$  in a different way, and all the  $a$  values are less than that of  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ .

The samples 1' through 6' were also viewed under the UV light of 254 nm, which showed that sample 3' emitted the brightest red light. Therefore, this work only studied sample 3' in more detail.

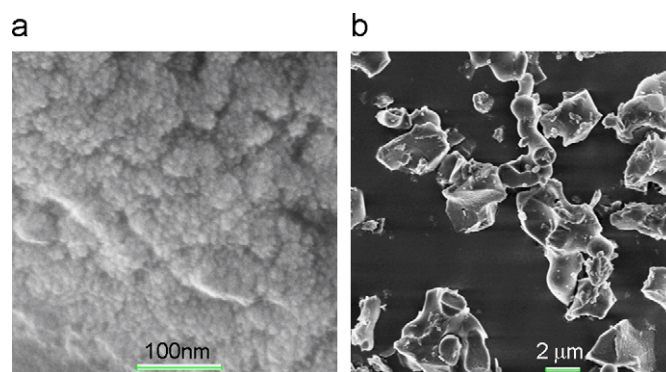
Fig. 5 shows representative SEM images of the  $\text{Ca}_3\text{Al}_2\text{O}_6:\text{Eu}^{3+}$  powder. It can be seen that the powder is composed of irregular particles of a few  $\mu\text{m}$ , which are actually formed from agglomeration of much smaller particles of a few nanometers. This explains why the materials are very easily pulverized. This easiness of pulverizing is very important, since it has been found that the performance of some materials deteriorated while they are pulverized.



**Fig. 4.** XRD patterns of (a)  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  (JCPDS file no. 48-1882), (b)  $\text{Ca}_3\text{Al}_2\text{O}_6$  (JCPDS file no. 38-1429) and samples 1' through 6', which were obtained from calcination of the LDH precursors 1 through 6 at 1350 °C.

**Table 1**  
The lattice parameters of samples 1' through 6'

Sample numbers	$\text{Eu}^{3+}$ concentrations (%)	Cell parameter $a$ (nm)	
		$\text{Ca}_3\text{Al}_2\text{O}_6$	$\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$
1'	0.5	1.52356	1.19934
2'	0.8	1.52400	1.19884
3'	1.0	1.53620	–
4'	1.2	1.52911	1.19500
5'	1.5	1.52760	1.19700
6'	2.0	1.52680	1.19724



**Fig. 5.** SEM images of 3' at indicated scale (a) 100 nm and (b) 2  $\mu\text{m}$ .

### 3.3. Excitation and emission spectra of the samples

The excitation and emission spectra of the calcined LDH precursor 3 at different temperatures (1150, 1250, 1350 and 1450 °C) were measured and are shown in Figs. 6 and 7. The excitation spectra of the calcined products (Fig. 6) consist of a strong broad band at 250 nm and several other weak but sharp lines. The broad absorption peak is due to the charge transfer transition from oxygen to  $\text{Eu}^{3+}$  center, which is generally believed that the  $\text{Eu}^{3+}$  ion ( $4f^6$  configuration) tends to capture one electron to achieve a more stable half-filled shell state ( $4f^7$  configuration). It can also be seen that the peak of the charge-transfer band shifts to a longer wavelength when the calcination temperature rises.

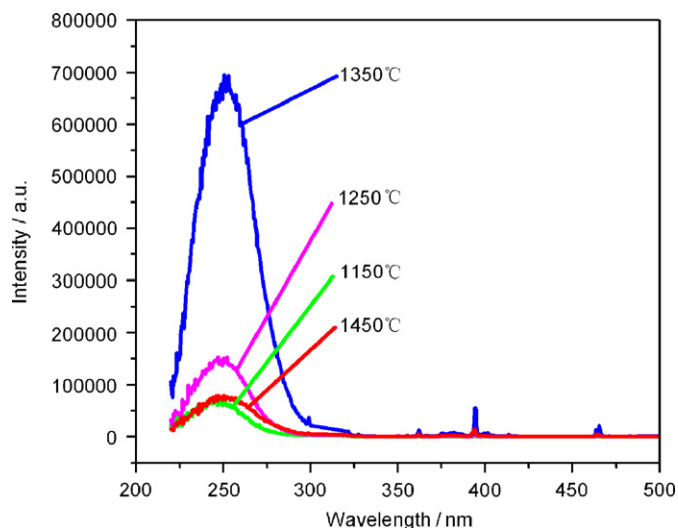


Fig. 6. Excitation spectra of 3' obtained at indicated temperatures (monitored at 614 nm).

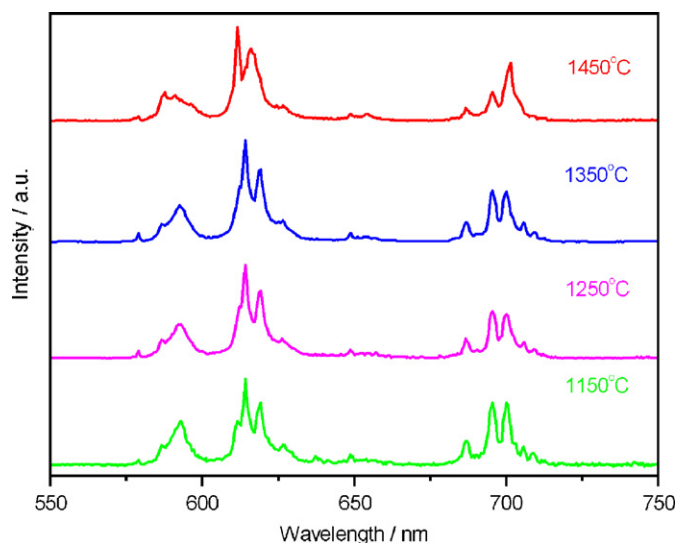


Fig. 7. Emission spectra of 3' obtained at different temperatures (excited with a UV light of  $\lambda = 250$  nm).

The emission spectrum of  $\text{Ca}_3\text{Al}_2\text{O}_6:\text{Eu}^{3+}$  (Fig. 7) is composed of groups of several sharp lines, which belong to the characteristic transitions between  $^5\text{D}_j$  ( $J = 0$ ) and  $^7\text{F}_j$  ( $J = 0-4$ ) of  $\text{Eu}^{3+}$ . The peak near 579 nm is based on the transition of  $^5\text{D}_0 \rightarrow ^7\text{F}_0$ . The peak in the vicinity of 593 nm is ascribed to the  $\text{Eu}^{3+}$  magnetic dipole transition  $^5\text{D}_0 \rightarrow ^7\text{F}_1$ , and the main peaks around 610–620 nm (614 and 619 nm) are assigned to the  $\text{Eu}^{3+}$  electric dipole transition of  $^5\text{D}_0 \rightarrow ^7\text{F}_2$ . The peak at about 649 nm is due to the transition of  $^5\text{D}_0 \rightarrow ^7\text{F}_3$ . And the emission lines near 700 nm are based on the transition of  $^5\text{D}_0 \rightarrow ^7\text{F}_4$  [25,26].

It is well known that emission spectrum of  $\text{Eu}^{3+}$  strongly depends on the site symmetry which  $\text{Eu}^{3+}$  occupies. If it occupies a site with an inversion symmetry, only magnetic-dipole transition  $^5\text{D}_0 \rightarrow ^7\text{F}_1$  can be observed, but if there is no inversion symmetry at the site of the  $\text{Eu}^{3+}$  ion, the electric-dipole transitions  $^5\text{D}_0 \rightarrow ^7\text{F}_2$  can be observed, also, the transition is sensitive to the coordination environment. Therefore, there are  $\text{Eu}^{3+}$  ions that occupy sites without inversion symmetry since the electric dipole  $^5\text{D}_0 \rightarrow ^7\text{F}_2$  transitions at 614 and 619 nm are stronger than

that of the magnetic-dipole  $^5\text{D}_0 \rightarrow ^7\text{F}_1$  transition around 593 nm in Fig. 7.

As it is known from the crystal structure of  $\text{Ca}_3\text{Al}_2\text{O}_6$  [27], all the  $\text{Al}^{3+}$  in the lattice are in  $[\text{AlO}_4]$  tetrahedrons and every six  $[\text{AlO}_4]$  tetrahedrons form a ring by apex sharing; and there are six types of  $\text{Ca}^{2+}$ . Two of them at the (000) and  $(\frac{1}{2}00)$  are octahedrally coordinated to six oxygen atoms with symmetry  $\text{S}_3$ , another two with symmetry  $\text{C}_3$ , and yet another two are asymmetrically coordinated to six or even eight oxygen atoms. It is known that  $\text{Eu}^{3+}$  ions occupy the asymmetrical site from photoluminescence spectrum, and there are four of such  $\text{Ca}^{2+}$  sites in the lattice of  $\text{Ca}_3\text{Al}_2\text{O}_6$ . It is difficult to take the  $\text{Al}^{3+}$  sites for  $\text{Eu}^{3+}$ , as the difference of radii is very big; also,  $\text{Eu}^{3+}$  may break the ring structure of  $[\text{AlO}_3]_6$ . There are many such examples. Yang [28] reported that for the emitting phosphor  $\text{Ca}_2\text{SnO}_4:\text{Eu}^{3+}$ , the  $\text{Eu}^{3+}$  ions substituted for the  $\text{Ca}^{2+}$  sites but not the  $\text{Sn}^{4+}$  sites. Hu [29] also reported a novel red phosphor,  $\text{CaMoO}_4:\text{Eu}^{3+}$ , in which  $\text{Eu}^{3+}$  ion was expected to occupy the  $\text{Ca}^{2+}$  site. In addition a phosphor  $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3:\text{Eu}^{3+}$  was prepared by Samantaray [30], who also suggested the same thing.

It is noticed that  $\text{Eu}^{3+}$  ion has one more positive charge than  $\text{Ca}^{2+}$  ion; therefore, one-by-one replacement of  $\text{Ca}^{2+}$  by  $\text{Eu}^{3+}$  makes accumulation of positive charges. In order to balance the charges some vacancies at  $\text{Ca}^{2+}$  sites should occur in the lattice, or some  $\text{O}^{2-}$  anions should occupy the  $\text{Ca}^{2+}$  sites. In either case, it will degrade the symmetry of the coordination environment of the  $\text{Eu}^{3+}$  ion, which enhances the electric dipole transition of  $^5\text{D}_0 \rightarrow ^7\text{F}_2$ . However, further studies are needed to reveal the details of the mechanism.

A careful observation of Fig. 7 reveals the difference of the sample obtained at 1450 °C from the others, in which the position of maximal peak shifts to a shorter wavelength. As shown in Fig. 3 the sample obtained at 1450 °C is composed of both  $\text{Ca}_3\text{Al}_2\text{O}_6$  and  $\text{CaAl}_2\text{O}_4$ , therefore the blue shift should be caused by  $\text{Eu}^{3+}$  in the  $\text{CaAl}_2\text{O}_4$  host.  $\text{CaAl}_2\text{O}_4:\text{Eu}^{3+}$  would not be a good material for red light emitting, which may be confirmed by no reports found on this material in literatures, although there are many researches on  $\text{CaAl}_2\text{O}_4:\text{Eu}^{2+}$  which is an excellent material for green light emitting [31].

### 3.4. The effect of calcination temperatures on photoluminescence properties

The emission intensities at 614 nm in Fig. 6 were drawn versus temperatures at which the materials were prepared, which are shown in Fig. 8.

The experimental result shows that the calcined product has no photoluminescence property until the calcination temperature is higher than 1150 °C. With the increase of temperature, the photoluminescence intensity increases. At 1350 °C, the intensity attains the maximum. However, when the temperature increases to 1450 °C the sample agglomerates and becomes glassily rigid, and the photoluminescence intensity decreases.

The phase compositions as shown in Fig. 3 indicate that the sample prepared at 1000 °C is composed of CaO and  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ ; the samples prepared at 1150, 1250 and 1350 °C have larger and larger amount of  $\text{Ca}_3\text{Al}_2\text{O}_6$ , although there may be some  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  in them; and the sample prepared at 1450 °C is composed of  $\text{CaAl}_2\text{O}_4$  and  $\text{Ca}_3\text{Al}_2\text{O}_6$ . It seems that the photoluminescent intensity increases when the amount of  $\text{Ca}_3\text{Al}_2\text{O}_6$  increases, therefore, only  $\text{Ca}_3\text{Al}_2\text{O}_6$  is a good host for the activator  $\text{Eu}^{3+}$ , others including CaO,  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ ,  $\text{CaAl}_2\text{O}_4$  are all poor hosts. In fact, searches from EI and SCI databases show that  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$  [32] and  $\text{CaAl}_2\text{O}_4$  [31] are hosts for  $\text{Eu}^{2+}$ -

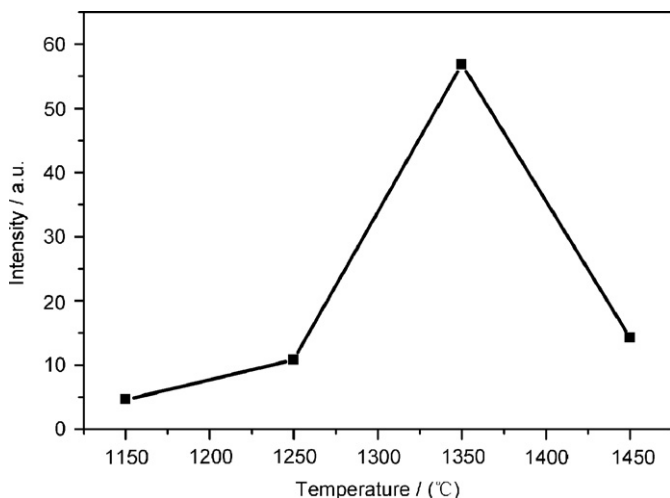


Fig. 8. The effect of calcination temperatures on photoluminescence properties.

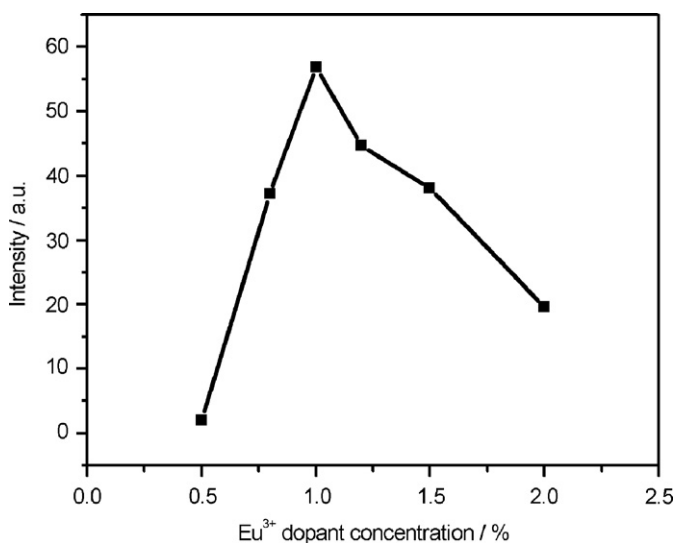


Fig. 9. The effect of Eu<sup>3+</sup> concentrations on photoluminescence properties.

activated phosphors, no literatures were found for Eu<sup>3+</sup>-activated phosphors.

### 3.5. The effect of Eu<sup>3+</sup> concentrations on photoluminescence properties

Fig. 9 shows the plot of relative intensities of samples 1' through 6' versus concentrations of Eu<sup>3+</sup>, which were measured on a PR-302 meter. It can be seen in Fig. 9 that the photoluminescence intensity is very low when the concentration of Eu<sup>3+</sup> = 0.5%. With the increase of Eu<sup>3+</sup> concentration, the intensity reaches a maximum when the concentration equals 1%. This means that the concentration quenching effect exists in the system. Consequently, the optimal concentration is 1%, which corresponds to the sample 3', Ca<sub>2.85</sub>Al<sub>2</sub>Eu<sub>0.05</sub>O<sub>5.925</sub>.

In the unit cell of Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>, there are 72 Ca<sup>2+</sup> ions and 48 Al<sup>3+</sup> ions; therefore, 1% of Eu<sup>3+</sup> means that only 1 Eu<sup>3+</sup> ion will be in the unit cell. Since the unit cell is cubic, and  $a = 1.5263$  nm, the interaction of Eu<sup>3+</sup> ions can be neglected; however, when the concentration of Eu<sup>3+</sup> increases the distances between Eu<sup>3+</sup> will be shortened, which dramatically increases the interaction and causes serious concentration quenching.

## 4. Conclusions

LDH precursors containing Ca<sup>2+</sup>, Al<sup>3+</sup> and Eu<sup>3+</sup> were prepared by the co-precipitation method, and Eu<sup>3+</sup> was incorporated successfully into the hydroxide layer. Calcination of the precursors at 1350 °C directly produces a loose and uniform powder in one step, which is in contrast to that of the high-temperature solid state reaction routes.

It is shown that the concentration of Eu<sup>3+</sup> in the mother solution for the LDH precursor should be 1% for the best results, and the precursor will produce a red light-emitting material with a composition like Ca<sub>2.85</sub>Al<sub>2</sub>Eu<sub>0.05</sub>O<sub>5.925</sub> when it is fired at 1350 °C. The phosphor can be excited by UV light of 210–300 nm with the peak at around 250 nm and emit a bright red light mainly at 614 nm.

It is also shown that the precursor turns out to be mainly the Ca<sub>12</sub>Al<sub>14</sub>O<sub>33</sub> phase without photoluminescence property if calcinated at 1000 °C; Ca<sub>12</sub>Al<sub>14</sub>O<sub>33</sub> and Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> phases at 1150 °C; only pure Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> phase at 1350 °C; and CaAl<sub>2</sub>O<sub>4</sub> as well as Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> at 1450 °C. It seems that only Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> is the best host for the Eu<sup>3+</sup>-activated phosphor, CaO, Ca<sub>12</sub>Al<sub>14</sub>O<sub>33</sub> and CaAl<sub>2</sub>O<sub>4</sub> are not good host for Eu<sup>3+</sup>-activated photoluminescent materials.

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