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Synthesis of YAG:Ce phosphor via different aluminum sources and precipitation processes

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Abstract

Ce^{3+}-doped yttrium aluminum garnet (YAG:Ce) phosphors were synthesized by the four different precipitating processes, in which aluminum nitrate or aluminum ammonium sulfate was used as the aluminum source. Pure YAG:Ce powder can be obtained by using aluminum nitrate combine normal strike precipitation method as calcined at 850 °C for 2 h. The property of YAG powder is affected by the cation homogeneity of precursor powder. The product formed by aluminum nitrate combine normal strike precipitation method has the highest emission peak at 535 nm after excitation at 470 nm.

Keywords: YAG:Ce; Phosphor; Cation homogeneity; Luminescent properties; Co-precipitation; White light emitting diode

1. Introduction

Yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) has many applications due to the optical properties and pyroceram structure [1–4]. Moreover most of the material for applications was doped with a small amount of impurity ions such as Eu, Cr, Tb, Ce to be substituted into the dodecahedral site of the YAG lattices to have different kinds of luminescent properties [5–8].

YAG:Ce phosphor was used in most of white light emitting diode (WLED) [9]. The broad emission band from 4f to 5d levels extends from 500 to 650 nm excited by blue light diode. There are many technologies used for preparation of YAG powder. Although the traditional solid state method is easy to form YAG powder, it needs to calcine at a higher temperature and age for a long time [10] to complete the solid state reaction. In recent years, several wet chemical techniques such as glycothermal method, spray pyrolysis method, combustion process and co-precipitation method [11–17] were used to prepare the YAG precursor.

In the previous reports, the homogeneity of Y^{3+} and Al^{3+} in precursor for synthesis of the pure YAG phase at temperatures below 1000 °C [18–20] was discussed. In this study, two different precipitation processes were used: one is the normal strike process (NS) and the other is the reverse strike process (RS) [16,21,22], with two different aluminum sources of: $Al(NO_3)_3$ and NH_4AlSO_4. The products obtained were determined by X-ray diffraction (XRD) for the phase formation, energy dispersion spectrometer (EDS) for the composition, transmission electron microscopy (TEM) for the observation of powder morphology and photoluminescence spectrometer for the phosphors properties.

2. Experimental

The precipitates were formed by mixing the two reactants of the ions solution and the 0.2 M ammonium hydrogen carbonate (AHC) solution together. The ions solutions used including solution "n" consisting of 0.01485 M

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Table 1
Products of the precipitation processes used

	Precipitation process	Aluminum source
NSYAGs	Normal strike	NH_4AlSO_4
NSYAGn	Normal strike	$Al(NO_3)_3$
RSYAGs	Reversal strike	NH_4AlSO_4
RSYAGn	Reversal strike	$Al(NO_3)_3$

$Y(NO_3)_3$ 0.00015 M $CeCl_3$ and 0.025 M $Al(NO_3)_3$ and solution "s" consisting of 0.01485 M $Y(NO_3)_3$ 0.00015 M $CeCl_3$ and 0.025 M NH_4AlSO_4. Table 1 shows the assembly of parameters of the processes used in this study.

The reverse strike (RS) process was performed by titrating 200 ml of the ions solution into 400 ml AHC solution. The titration rate was kept at about 3.4 ml/min. On the other hand, the normal strike (NS) process was performed by titrating 400 ml of AHC solution into the 200 ml of the ion solution. The "n" and "s" in Table 1 represent the aluminum ion sources of nitrate and sulfate, respectively. The final pH range after titration was in the range of 6.5–7. The products were then centrifuged and washed four times with distilled water and dried in the oven at 100 °C for 24 h. The precursor was calcined at the desired temperature in the range of 800–1300 °C for 2 h. X-ray diffraction measurement of the powders was performed by using Rigaku MultiFlex, Tokyo, Japan. The scan range was from 15° to 65° with a scan step of 0.02° and scan rate of 4° min^{-1}. Particle morphology and EDS analysis were carried out by field emission transmission electron microscope (HITACHI FE2000). The photoluminescence spectra of the samples were analyzed with MFS230 fluoresce spectrometer.

3. Results and discussion

3.1. Results of XRD and EDS analysis

Fig. 1 shows the XRD pattern of the products, which were calcined at different temperatures. The XRD pattern of precursor of NSYAGs calcined at 800–1100 °C is shown in Fig. 1(a). The powders of NSYAGs heated at 800 °C are almost amorphous with a small amount of cubic Y_2O_3. When the precursor is calcined at 850–900 °C, the intermediate phases appear which are identified as YAP ($YAlO_3$) and YAM ($Y_4Al_2O_9$). The precursor almost convert to YAG phase but still has some intermediate peaks when calcined at 1000 °C. Pure YAG phase powder is obtained as the calcination temperature above 1100 °C as shown in Fig. 1(a). The samples of RSYAGs (Fig. 1(b)), NSYAGn (Fig. 1(c)) and RSYAGn (Fig. 1(d)) are amorphous as heated at the temperatures below 850 °C. The precursors of RSYAGs and RSYAGn are almost convert to pure YAG phase as the heating temperature is raised up to 850 °C, but still have a small amount of YAP phase remaining. The sample of NSYAGn has

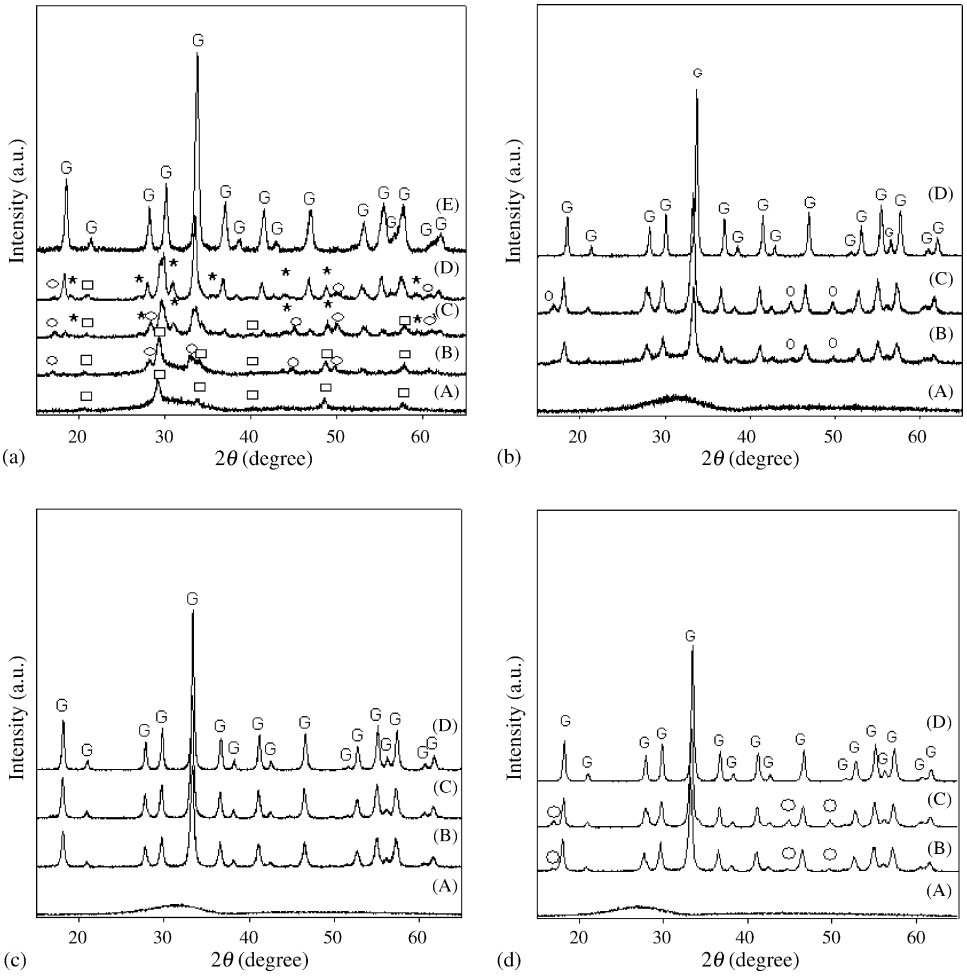

Fig. 1. XRD spectra of YAG:Ce precursor calcined at different temperatures for: (a) NSYAGs, (b) RSYAGs, (c) RSYAGn and (d) NSYAGs (G: YAG, *: YAM, ○: YAP, □: Y_2O_3 at (A) 800 °C, (B) 850 °C, (C) 900 °C, (D) 1000 °C and (E) 1100 °C).