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Sol–gel combustion synthesis and luminescent properties of nanocrystalline YAG:Eu^{3+} phosphors

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Abstract

Eu^{3+}-doped $Y_3Al_5O_{12}$ (YAG:Eu^{3+}) phosphors were synthesized by a facile sol–gel combustion method. In this process, citric acid traps the constituent cations and reduces the diffusion length of the precursors. YAG phase is obtained through sintering at 900 °C for 2 h. There were no intermediate phases such as $YAlO_3$ (YAP) and $Y_4Al_2O_9$ (YAM) observed. The charge transfer band of nanocrystalline phosphors shows a shift toward the high-energy side, compared with that of amorphous ones due to lower covalency of Eu–O bond for nanocrystalline phosphors. The higher concentration quenching in YAG:Eu^{3+} nanophosphors may be caused by the confinement effect on resonant energy transfer of nanocrystalline. It also indicates that the sol–gel combustion synthesis method provides a good distribution of Eu^{3+} activators in YAG host.
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1. Introduction

The development of new types of high-resolution and high-efficiency displays has an urgent need for phosphors with novel and enhanced optical properties. During the past decades, the novel and enhanced properties of nanostructured materials have attracted considerable attention for their interesting chemical and physical properties

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[1,2]. The nanostructured materials may have applications in development of a novel type of luminescent material for display applications. In particular, the Mn-doped ZnS nanocrystal can yield both high luminescent efficiencies and lifetime shortening [1]. This greatly promotes development of rare earth-doped nanophosphors.

YAG:Eu^{3+} is a red phosphor widely used in optical display and lighting applications. Furthermore, YAG:Eu^{3+} can be used as fluorescence thermometry because its fluorescence properties vary with temperature [3]. The conventional routine to synthesize the commercial YAG phosphor is based on solid-state reaction method. It requires high temperature of 1600 °C and prolonged heating to obtain pure phase. In the last few years, various improved wet chemical methods, such as sol–gel [4,5], coprecipitation [6] and solvothermal [7] methods, have been used to synthesize YAG phosphors. These methods have advantages of fine homogeneity, high reactivity of starting materials and lower sintering temperatures. However, they require long time and need strict techniques. In contrast, the combustion method [8] is quite simple, and the combustion reaction lasts only a few seconds. But its reaction is too violent to control, and the powders are too difficult to collect. In the present work, a new sol–gel combustion method was used to synthesize YAG:Eu^{3+} phosphors. This process perfectly combines the advantages of sol–gel process and low-temperature combustion processes. The nanocrystalline YAG:Eu^{3+} phosphors are investigated by X-ray diffraction (XRD), Fourier Transform Infrared (FTIR) spectra, transmission electron microscopy (TEM), and luminescence spectroscopy.

2. Experimental procedure

$Al(NO_3)_3 \cdot 9H_2O$ (99.9%), $Y(NO_3)_3 \cdot 9H_2O$ (99.99%), Eu_2O_3 (99.99%) and $C_6H_8O_7 \cdot H_2O$ (99.9%) were used as starting materials. The nominal compositions are $(Y_{1-x}Eu_x)_3Al_5O_{12}$ with $x = 0$, 0.02, 0.04, 0.06, and 0.08. Aqueous solution of $Eu(NO_3)_3$ was prepared by dissolving high-purity Eu_2O_3 with HNO_3 and some deionized water. $Al(NO_3)_3 \cdot 9H_2O$, $Y(NO_3)_3 \cdot 9H_2O$ and $C_6H_8O_7 \cdot H_2O$ were dissolved in deionized water. The ratio of nitrate to citric acid used in present work was 1:1. Stoichiometric amounts of above solutions were mixed and the solution was initially heated at 60 °C in air and continuously stirred for several hours, and the solution turned to yellowish sol. Then it was heated at 80 °C and stirred continually, to get the sol transformed into transparent sticky gel. The gel was rapidly heated to 200 °C and an auto-combustion process took place yielding a yellowish fluffy precursor. The precursor was calcined at temperatures from 700 to 1100 °C in a muffle furnace in air.

The crystalline evolution was identified using the Rigaku D/MAX-2550-18KW powder diffractometer with CuKα-radiation. The morphology and particle size of the phosphors were analyzed by a JEOL JEM-200CX TEM. FTIR spectra were measured by Nicolet NEXUS 870 FT–IR spectrometer with KBr as a carrier. The excitation and emission spectra were obtained by a JASCO FP-6500 fluorescence spectrophotometer at room temperature.

3. Results and discussion

The XRD spectra of the YAG:Eu^{3+} phosphors sintered at different temperatures are shown in Fig. 1. No diffraction peak appears for the phosphors sintered at 800 °C, indicating that the phosphors are amorphous below 800 °C. The

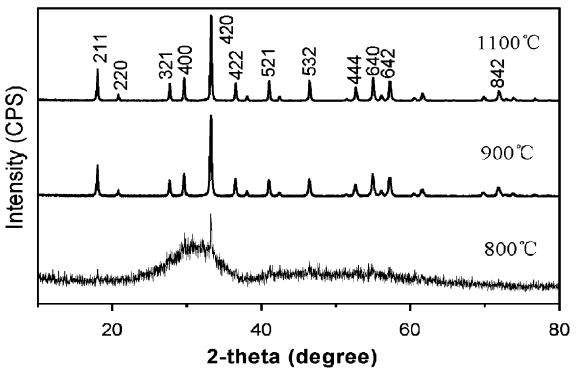

Fig. 1. XRD patterns of YAG:8%Eu^{3+} precursors sintered at different temperatures.

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