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Template synthesis and luminescent properties of nano-sized YAG:Tb phosphors

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Abstract

Terbium-activated $Y_3Al_5O_{12}$ (YAG:Tb) phosphor nanoparticles with homogeneous grain size and crystallinity have been prepared at 700 °C by a novel gel-network coprecipitation route using gelatin as a template. The products were characterized by using TG-DTA, XRD and TEM techniques. The diffraction profile of YAG:Tb nanoparticles could be indexed as a garnet structure and exhibited peak broadening phenomenon, as revealed by X-ray diffraction (XRD) data. The products appeared to be regular spherical or elliptical and sizes ranged from 40 to 55 nm. Furthermore, the photoluminescence spectra of the YAG:Tb nanoparticles were investigated to determine the energy level of electron transition related to luminescence processes. The effects of Tb concentration and the average grain size on the PL spectra were also studied.

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Keywords: YAG:Tb; Phosphor nanoparticle; Photoluminescence; Template synthesis

1. Introduction

Three compounds exist in the system Y_2O_3–Al_2O_3:$Y_4Al_2O_9$, $YAlO_3$, and $Y_3Al_5O_{12}$ [1]. $Y_3Al_5O_{12}$ (YAG), and the most important of these compositions exists in the cubic form with a garnet structure. It is an important crystal for fluorescence and solid-state lasers. Tb-activated YAG phosphor has luminescence properties fairly insensitive to temperature variation and shows little tendency to saturate at high current excitations. Furthermore, YAG:Tb is a characteristic narrow-band phosphor suitable for contrast-enhanced display applications in high ambient illumination conditions [2]. Hence, YAG:Tb is one of the promising phosphors candidates which may be used in projection CRT's, FED and scintillation

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and EL applications [3]. For enhancing the brightness and resolution of these displays, it is important to develop phosphors with high quantum efficiency, controlled morphology and small particle sizes. It has been found that the luminescence efficiency of YAG:Tb phosphor depends on the synthesis method, which can lend itself to the formation of single-phase YAG.

Traditionally, YAG phosphor powders are synthesized via a solid-state reaction at high temperatures for a long time. Such conditions lead normally to powders of relatively large and varied grain sizes (5–20 μm) and varying impurity contents. There are often problems obtaining phase-pure material because of the intermediate formation of other phase, such as $Y_4Al_2O_9$ and $YAlO_3$ [4]. To achieve the desired phase purity and required particle size, high-temperature treatment($>1600\,^{\circ}C$) and extensive ball milling, which generally introduce additional impurities and defects and greatly reduce luminescence efficiency, are essential. For phosphor applications, it is desirable to have a fine particle size for high resolution and chemical purity for optimum chromaticity and brightness [5]. For overcoming the drawbacks of the solid-state reaction process, several wet chemical synthesis techniques have also been used to prepare YAG powders [6–8]. Although YAG can be obtained at low temperatures via these methods, further annealing at high temperatures ($\sim$1400 °C) is required to improve the luminescent properties of the powders. Thus, hard aggregation of powers easily arises.

In this study, we report for the first time the preparation of nano-sized YAG:Tb phosphors via a gel-network-coprecipitation method using gelatin as template at relatively low temperatures (700 °C). The phase development, particle size and morphology, as well as the optical properties of the resulting powders were evaluated. The relation between the photoluminescent properties of the nano-size particles and the Tb concentration was discussed.

2. Experimental

The yttrium and terbium sources for YAG:Tb synthesis were yttrium oxide (99.99%) and terbium oxide (Tb_4O_7 99.99% pure), respectively. All solvents and other reagents were of analytical grade. Aluminum nitrate was dissolved in distilled water. RE $(NO_3)_3$ salts were freshly prepared by a reaction of RE_2O_3 with dilute nitric acid.

YAG:Tb nanoparticles were synthesized via a procedure similar to that reported previously for YAG:Bi,Eu nanoparticles [9]. The typical process is as follows: 25 ml of 0.2 M aluminum nitrate solution was mixed uniformly with 14 ml of 0.2 M yttrium nitrate and 2 ml of 0.1 M terbium nitrate solution under stirring and then 6 g gelatin was dissolved in above mixed nitrate solution with violent stirring in an 80 °C water bath .The mixture was cooled to 4 °C, turning to a yellowish gel. The gel was cut into small pieces and soaked in excessive 1:1 $NH_3 \cdot H_2O$ solution for 24 h at 4 °C. The hydroxides were coprecipitated in the gel-network. Washed gel with cooled distilled water and ethanol and dried in a vacuum chamber at 110 °C. The dried gel (was termed the ''precursor'') was preheated at 400 °C for 2 h. The obtained powders were subsequently calcined at various temperatures from 500 to 1000 °C for 3 h, producing fine phosphor powders.

TG-DTA measurements of the precursor were performed on Shimadzu DT-40 thermal analyzer. The XRD of the products was recorded using a Bruker D8 Advance X-ray diffractometer (CuKα radiation). The particle size and morphology of the powders were examined using a 200CX transmission electronic microscope (Jeol 200CX, Japan). The photoluminescence spectra of phosphors were measured on a Hitachi F-4500 fluorescence spectrophotometer at room temperature.

3. Results and discussion

The TG–DTA curves of the precursor are shown in Fig. 1. The endothermic reaction up to 230 °C resulted from the removal of absorbed moisture and dehydration. The exothermic reaction took place between 230 and 430 °C accompanied with a most significant weight loss, which was attributed to the decomposition of nitrate in the precursor. The strong exothermic behavior could be clearly seen at about 539 °C, the maximum