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Original Research Paper

Synthesis and luminescence properties of YAG:Ce nanopowder prepared by the Pechini method

S.A. Hassanzadeh-Tabrizi

Department of Materials Engineering, Islamic Azad University, Najafabad Branch, Isfahan, Iran

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ABSTRACT

Cerium-doped yttrium aluminum garnet (YAG:Ce) powder was synthesized by the Pechini method with aluminum nitride, yttrium nitride, citric acid and ethylene glycol as the starting materials. Structure, morphology and luminescence spectra were investigated by using X-ray diffraction, thermogravimetric and differential thermal analysis, scanning electron microscopy, Fourier transform infrared spectroscopy and photoluminescence spectroscopy measurements. The pure YAG phase was formed after heat treatment at 800 °C for 3 h and no intermediate phase was observed. The average size of the particles was about 70 nm. The photoluminescence spectrum of the crystalline YAG:Ce phosphors showed the green-yellow emission with 5d → 4f transition as the most prominent group.

The increase of the ethylene glycol:citric acid molar ratio, resulted in a powder with smaller particle size and better luminescence properties.

1. Introduction

Yttrium aluminum garnet $Y_3Al_5O_{12}$ (YAG) has a cubic garnet structure with variety of good optical properties as well as excellent chemical stability and creep resistance [1,2]. Doping with other rare earth ions, such as Nd^{3+} and Ce^{3+}, into YAG makes it an ideal material for phosphors and solid-state lasers, which have attracted both technological and industrial interest [3,4]. Generally, YAG is prepared at high temperatures (>1500 °C) for an extended time period via a solid state reaction process. Such traditional processes normally lead to powders with large grain sizes. Moreover, several intermediate phases such as $Y_4Al_2O_9$ (YAM) and $YAlO_3$ (YAP) are observed in the products [5,6]. For correcting these drawbacks of the solid-state reaction process, many chemical methods have been developed and successfully used for powder processing in recent years. These methods include molten salt method [7,8], sol–gel processing [9,10], precipitation [11], co-precipitation [12], spray pyrolysis [13], combustion [14] solvo-thermal [15], polyacrylamide gel [16], hydrothermal synthesis [17] and so on. These wet chemical processes achieve intimate mixing of reactant cations on the atomic level, leading to an increase in reaction rate and lowering synthesis temperature. The Pechini method [18] is an alternative to the conventional sol–gel method. The objective of this method is the immobilization of metallic ions in a rigid polymer network. The metallic ions were dispersed in the polymer network at the atomic scale without precipitation and phase segregation. This process allows a complete control over the product stoichiometry, even for more complex oxide powders [19].

In the present paper, YAG:Ce in the nanometer scale has been synthesized by the Pechini method. The luminescence properties of this material have been studied. The influence of ethylene glycol:citric acid ratio on powder morphology and luminescence properties was also investigated.

2. Experimental procedure

Pure $Al(NO_3)_3{\cdot}9H_2O$ (Merck), $Y(NO_3)_3{\cdot}6H_2O$ (Aldrich) and $Ce(NO_3)_3{\cdot}6H_2O$ (Merck) were used as the cations' source. The nitrate salts were dissolved in 80 ml of deionized water and citric acid (CA) was then added (molar ratio CA:total cations = 1). These metal salts were dissolved at a molar ratio, Y:Al, of 3:5, respectively. The concentration of Ce varied from 0.1 to 2.0 at.%. The mixture was magnetically stirred until a clear solution was obtained. Ethylene glycol (EG) was added into solution at EG:CA molar ratios of 1 and 2. The solution was continuously stirred at 80 °C in order to facilitate the evaporation of the excess water and accelerate the polyesterification reaction. During the polyesterification process no turbidity or precipitation was observed. The gel was then heated at 100 °C in an oven for 24 h. The gel was ground and calcined at various temperatures for 3 h.

Differential thermal analyses (DTA) and thermogravimetric (TG) were used in the range of 100–975 °C at a rate of 10 °C min^{-1} with the STA equipment (PL Thermal Sciences STA 1500). Fourier transformation infrared spectroscopy analysis (FTIR) of powders was

E-mail addresses: tabrizi1980@gmail.com, hassanzadeh@pmt.iaun.ac.ir

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