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ScienceDirect
Optical Materials 30 (2008) 1810–1815
www.elsevier.com/locate/optmat

Gd_2O_3:Eu phosphor powders prepared using a size-controllable droplet generator

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Received 3 September 2007; received in revised form 27 November 2007; accepted 28 November 2007
Available online 31 January 2008

Abstract

Gd_2O_3:Eu phosphor powders were prepared by a filter expansion aerosol generator (FEAG) process capable of changing the mean size of droplets. The change in the mean size of the Gd_2O_3:Eu phosphor powders according to the concentrations of polyethylene glycol added to spray solutions was caused by the difference in the mean size of the droplets produced via the FEAG process. The mean sizes of droplets produced by the FEAG process were affected by the surface tension and viscosity of the spray solutions. The mean sizes of the Gd_2O_3:Eu phosphor powders obtained from the spray solutions with the same concentration of metal salts changed from 1.5 to 4.2 μm according to the concentrations of polyethylene glycol and citric acid added to the spray solutions. The maximum photoluminescent intensity of the phosphor powders obtained from the spray solutions with polymeric precursors and boric acid flux was 144% of that of the phosphor powders obtained from the aqueous spray solutions without flux.

PACS: 81.20.Rg; 78.55.Hx; 78.40.Ha; 81.05.Hd; 81.40.Tv

Keywords: Phosphor; Spray pyrolysis; Flux material; Functional ceramic

1. Introduction

Spray pyrolysis is a gas phase reaction method utilizing several micron size droplets as a reaction media [1–5]. Spray pyrolysis has advantages in the preparation of advanced ceramic, metal and glass powders with controlled morphology and stoichiometry [1–13]. The characteristics of the powders prepared by spray pyrolysis are mainly determined by the types of liquid aerosol generators. Ultrasonic spray generators, which have reasonable production rates of several micron droplets, are mainly applied to the preparation of advanced ceramic, metal and glass powders [1–13]. A new liquid aerosol generator named the filter expansion aerosol generator (FEAG) was also applied to the preparation of advanced ceramic, metal and glass powders [14–16]. The FEAG process is a liquid aerosol generator that produces fine-sized droplets under low pressure [17].

Spray pyrolysis is applied to the preparation of multicomponent oxide phosphor powders for application on displays and lamps [8–13]. The FEAG process has also been applied to the preparation of multicomponent phosphor powders [18,19]. Kang et al. prepared Zn_2SiO_4:Mn and $Y_3Al_5O_{12}$:Eu phosphor powders by spray pyrolysis using the FEAG process. The characteristics of phosphor powders prepared via the FEAG process were similar to those prepared by ultrasonic spray pyrolysis. The droplets produced via FEAG had a similar mean size to those of the ultrasonic spray generator.

Recently, a new capacity of the FEAG process was discovered in our laboratory. The mean size of the droplets produced by the FEAG process is controllable by changing the characteristics of the spray solutions. In this study, Gd_2O_3:Eu phosphor powders were prepared by the FEAG

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0925-3467/$ - see front matter
doi:10.1016/j.optmat.2007.11.026

process from spray solutions with polymeric precursors. The mean size of droplets produced by the FEAG process changed from several microns to several tens of microns according to the concentration of the polymeric precursors added to the spray solutions. Therefore, the characteristics of the Gd_2O_3:Eu phosphor powders prepared by the FEAG process were strongly affected by the concentration of the polymeric precursors added to the spray solutions.

2. Experimental

The schematic diagram of the FEAG process used in this study is shown in Fig. 1. The FEAG process consists of a porous glass filter, an ultrasonic spray generator, a vacuum pump and a bag filter. An ultrasonic spray generator was used as a supplying system of spray solution for continuous and uniform quantity. Spray solution was supplied through an ultrasonic spray generator using carrier gas onto a glass filter surface where it forms a thin liquid film. This liquid film is passed through the filter pores by the carrier gas and expanded into a low pressure chamber. The droplet formation mechanism in the FEAG process was investigated in our previous paper [17]. Metal salt solution is atomized into droplets and delivered into hot-wall reactor of 160 torr. As the aerosol stream passes through the reactor, solvent evaporates and metal salt decomposes to form product powders. At the end of the reactor, powder collection filter and vacuum pump of 600 l/min are connected in series. The length and diameter of the quartz reactor were 1200 and 50 mm, respectively.

$Gd_{1.75}O_3$:$Eu_{0.25}$ phosphor powders were prepared by spray pyrolysis from aqueous and polymeric precursor solutions. The aqueous solution was prepared by dissolving gadolinium oxide and europium oxide using nitric acid in distilled water. The polymeric precursor solution was prepared by dissolving citric acid (CA) and polyethylene glycol (PEG 400) into the above aqueous solution. The concentration of citric acid was changed from 0.02 to 0.3 M. The concentration of polyethylene glycol was changed from 3×10^{-4} to 4.8×10^{-3} M. The precursor powders were prepared by spray pyrolysis at a temperature of 900 °C. The total concentration of metal components was 0.3 M. The content of boric acid used as flux material was 1 wt.% of Gd_2O_3:Eu phosphor powders.

The crystal structures of the powders were studied by X-ray diffraction (XRD, RIGAKU, D/MAX-RB) with Cu Kα radiation ($\lambda = 1.5418$ Å). The morphologies of powders were investigated by scanning electron microscopy (SEM, JEOL, JSM 6060). Photoluminescence measurement was performed with spectrofluorophotometer (SHIMADZU, RF-5301PC) using an Xe lamp excitation source. The surface tension of spray solutions was measured with a thermostated tensiometer (model K10 Krüss GmbH, Hamburg, Germany), using a platinum–iridium ring and the method of Du Noüy. The measurements were carried out at room temperature of 20 °C. The tensiometer was calibrated with distilled water ($\sigma = 72.8$ mN m $^{-1}$ at 20 °C). Viscosity measurements were made using an Ostwald viscometer immersed in water bath, maintained at a temperature of 20 °C.

3. Results and discussion

The morphologies of the precursor powders prepared by the FEAG process from spray solutions with and without polymeric precursors are shown in Fig. 2. The precursor powders obtained from an aqueous spray solution were large in size and had a hollow, porous morphology. The high drying and decomposition rate of the droplets and powders inside the hot-wall reactor maintained at a low pressure produced precursor powders that were large in size and hollow and porous in morphology. The precursor powders obtained from spray solutions with polymeric precursors exhibited differing mean sizes and morphologies according to the concentrations of polyethylene glycol and citric acid added to the spray solution. The concentrations of polyethylene glycol according to the concentrations of citric acid are shown in Table 1. The precursor powders obtained from spray solutions with low concentrations of polyethylene glycol such as 3×10^{-4} and 8×10^{-4} M had a porous, hollow morphology. However, the precursor powders obtained from spray solutions with

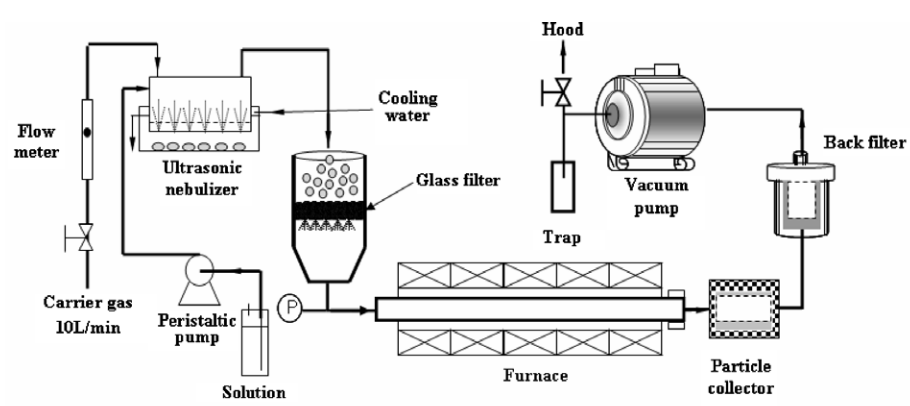

Fig. 1. Schematic diagram of the FEAG process.