

Synthesis, characterization and photoluminescence properties of $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ sub-microphosphor by homogeneous precipitation method

Jingbao Lian^a, Xudong Sun^{b,*}, Ji-Guang Li^b, Xiaodong Li^b

^a School of Mechanical Engineering, Liaoning Shihua University, Fushun 113001, PR China

^b Key Laboratory for Anisotropy and Texture of Materials (Ministry of Education), Northeastern University, Shenyang 110004, PR China

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ABSTRACT

Homogeneous precipitation method for synthesizing $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ sub-microphosphor was developed, using the commercially available Gd_2O_3 , Pr_6O_{11} , H_2SO_4 and $(\text{NH}_2)_2\text{CO}$ (urea) as the starting materials. It was found that the as-synthesized precursor is mainly composed of $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2(\text{OH})_2(\text{CO}_3)(\text{SO}_4) \cdot n\text{H}_2\text{O}$. Pure quasi-spherical shaped $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ particles can be synthesized by calcining the precursor at a temperature higher than 700°C for 1 h in flowing hydrogen. The $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ particles have a narrow size distribution with a mean grain size of about 300–400 nm. Photoluminescence spectra of $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ under 303 nm UV excitation show a green emission at 515 nm as the most prominent peak, which corresponds to the $^3\text{P}_0 \rightarrow ^3\text{H}_4$ transition of Pr^{3+} ions. Decay study reveals that the $^3\text{P}_0 \rightarrow ^3\text{H}_4$ transition of Pr^{3+} ions in $\text{Gd}_2\text{O}_2\text{S}$ host lattice has a single exponential decay behavior.

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1. Introduction

Rare earth oxysulfides are wide-gap (4.6–4.8 eV) semiconductors, and often exhibit favorable properties such as excellent chemical stability, innocuity and high luminescent efficiency [1,2]. Therefore, they have been widely utilized as luminescent host materials for a variety of applications, such as high efficiency red phosphors for color television picture tube [3], X-ray computed tomography [4], laser materials [5]. Among these oxysulfides, Gadolinium oxysulfide ($\text{Gd}_2\text{O}_2\text{S}$) is a promising host material, because of its high density (7.34 g/cm^3) and high effective atomic number of Gd (60), which leads to a high stopping power for X-ray radiation. Especially, trivalent praseodymium doped $\text{Gd}_2\text{O}_2\text{S}$ phosphor is known to be an important imaging system extensively applied in X-ray microscope, soft X-ray phosphor screen for water window, and so on. It provides an advantageous high intrinsic X-ray to light conversion efficiency, 1.8 times greater light output than CaWO_4 and reasonably long decay time. George Fern's study [6] shows the $\text{Gd}_2\text{O}_2\text{S}:\text{Pr}^{3+}$ sub-microphosphor has higher detection efficiency, higher quality image and lower structure noise under soft X-rays exposure conditions than that of the much larger commercially available phosphor, resulting from higher effective packing fraction, thinner phosphor layer and lower scattering of radiation.

A number of synthesis routes have been developed for processing $\text{Gd}_2\text{O}_2\text{S}$ phosphors, including solid-state reaction (SSR) method [7], reduction method [8], combustion synthesis method [9],

emulsion liquid membrane (ELM) method [10] and gas sulfuration method [11]. Among these synthesis methods, the first two methods are the most typical and common ones because of their high reliability, low cost, and high luminescent properties. However, the two methods have the disadvantage of unable to achieve ultra-fine phosphor and difficult to control the particle morphology. Thus, the first two methods are not easily to meet the requirements for increasing high resolution of display due to the coarser particle. The combustion synthesis and ELM methods also have some disadvantages, e.g., the former is a high cost synthesis route due to the use of expensive dithioamide or other organic solvent as fuels, the latter is a complicated and time-consuming synthesis process. Although the gas sulfuration method can synthesize ultra-fine phosphor, this method involves the use of hydrogen sulfide (H_2S), carbon disulfide (CS_2) or sulfur vapor, which is harmful to the environment.

In this work, a homogeneous precipitation method that uses commercially available Gd_2O_3 , Pr_6O_{11} , H_2SO_4 and $(\text{NH}_2)_2\text{CO}$ as the starting materials was developed for the synthesis of $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ sub-microphosphor. This method has the advantage of simplicity in processing, low cost, and not involving toxic carbon disulfide (CS_2) or hydrogen sulfide (H_2S).

2. Experimental procedure

2.1. Synthesis

Gd_2O_3 powder (99.99% purity, Shanghai New Materials Yuelong Co. Ltd., Shanghai, China), Pr_6O_{11} powder (99.99%, Nanfang Rare

* Corresponding author. Tel.: +86 24 83687787; fax: +86 24 23906316.

E-mail address: xdsun@mail.neu.edu.cn (X. Sun).

Earth High Technology Co. Ltd., Ganzhou, China), H_2SO_4 (GR, Shenyang Xinhua Reagent Factory, Shenyang, China) and $(\text{NH}_2)_2\text{CO}$ (AR, Sinopharm Chemical Reagent Co. Ltd., Shanghai, China) were used as the starting materials. Previous studies [6,12] indicated that a 1.0 mol.% Pr^{3+} concentration in the $\text{Gd}_2\text{O}_3\text{S}$ host lattice gave the highest luminescence intensity values. So, the Pr^{3+} activator concentration was set at 1.0 mol.% in this study. First, $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2(\text{SO}_4)_3$ solution (0.01 mol/L) was prepared by dissolving a stoichiometric amount of Gd_2O_3 and Pr_6O_{11} powders in dilute H_2SO_4 solution. Urea solution (0.2 mol/L) was obtained by dissolving $(\text{NH}_2)_2\text{CO}$ in deionized water and was used as the precipitant. Uniform mixture solutions were formed by mixing a $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2(\text{SO}_4)_3$ solution (500 mL) and a urea solution (500 mL) under vigorous stirring. Next, the mixture solutions were heated at 90 °C for 2 h in a water bath. The resulted white precipitation was separated centrifugally, washed several times with deionized water and dried at 80 °C for 8 h to obtain the precursor. Finally, the precursor was calcined at 600, 700, 800 and 900 °C, respectively for 1 h in flowing hydrogen atmosphere to synthesize the resulted phosphor.

2.2. Characterization

Fourier transform infrared spectra (FT-IR) were recorded in the region of 4000–400 cm^{-1} using a Perkin–Elmer FT-IR spectrophotometer with KBr pellets. Thermal analysis, i.e. thermogravimetry (TG), derivative thermogravimetry (DTG) and differential thermal analysis (DTA) were performed using a simultaneous differential thermal and thermo-gravimetric analyzer (SDT 2960) with a heating rate of 10 °C/min in flowing hydrogen atmosphere. Phase analysis of the synthesized phosphors was performed by X-ray diffractometry (XRD, X'pert Pro MPD, Mode PW3040/60 diffractometer, Philips, Netherlands) with $\text{Cu K}\alpha$ (1.5406 Å) radiation and graphite monochromator in the 2θ range from 10° to 90°, operating at 40 kV and 40 mA. The scanning step was 0.0334° and the scanning rate was 3° (2θ)/min. The particle morphology and size

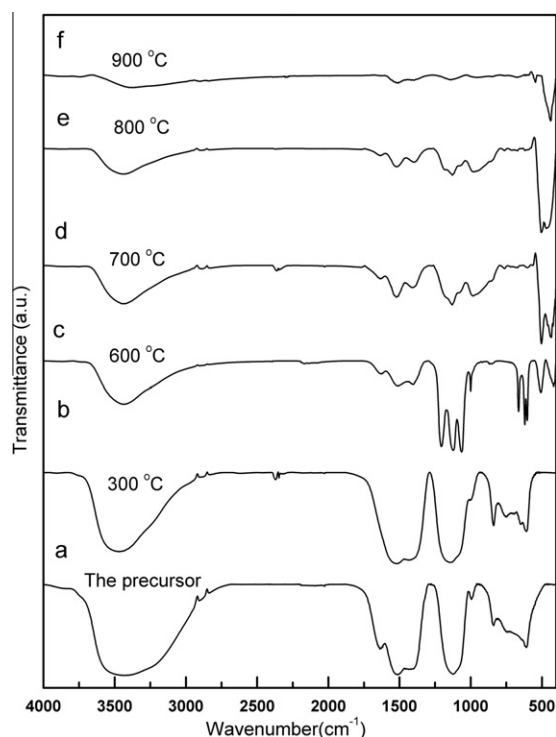


Fig. 1. FT-IR spectra of the precursor and its calcination products at different temperatures for 1 h in flowing hydrogen atmosphere.

distribution were observed by using a JEOL JSM-7001F Field Emission Scanning Electron Microscope (FESEM). Photoluminescence (PL) spectra and decay time of $(\text{Gd}_{0.99}\text{Pr}_{0.01})_2\text{O}_2\text{S}$ phosphors were performed on a Perkin–Elmer LS55 fluorescent spectrophotometer equipped with a xenon lamp as an excitation source. All of the measurements were performed at room temperature.

3. Results and discussion

3.1. Formation mechanism and transformation of the precursor

The chemical reaction for synthesizing the precursor can be shown as below:

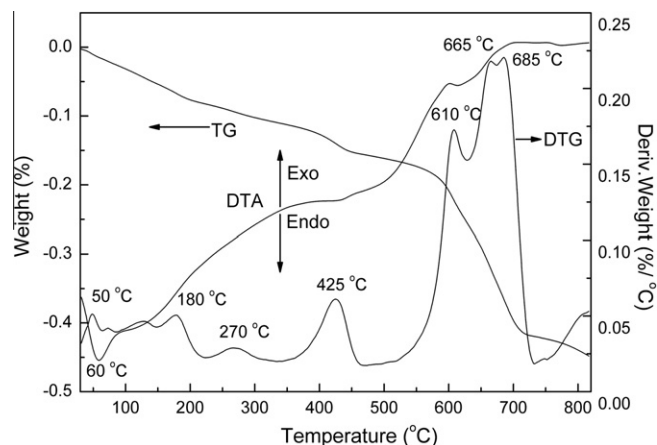


Fig. 2. DTA–TG–DTG curves of the precursor in flowing hydrogen atmosphere.

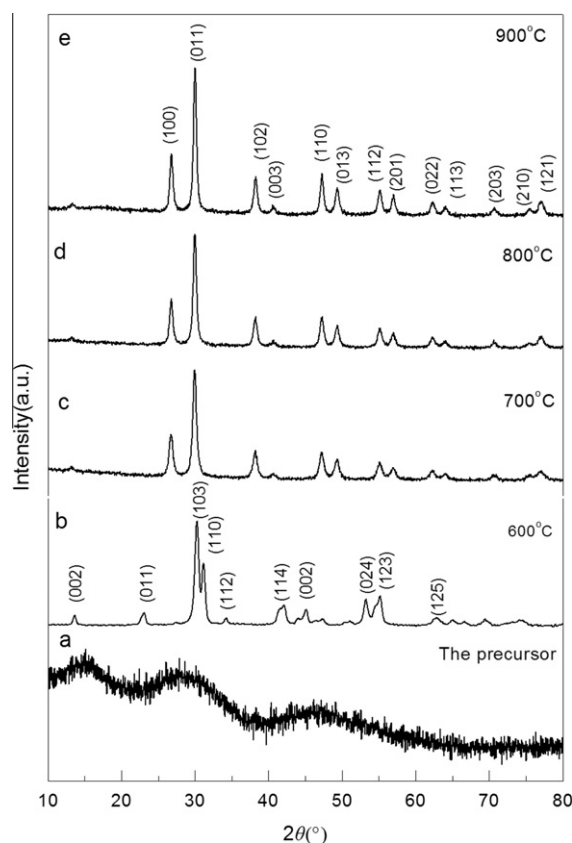


Fig. 3. XRD patterns of the precursor and its calcination products at 700, 800 and 900 °C.

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