Materials Research Bulletin 68 (2015) 295–301

Contents lists available at ScienceDirect

Materials Research Bulletin

journal homepage: www.elsevier.com/locate/matresbu

Effect of ultrasound treatment on the morpho-structural and luminescent characteristics of cerium doped yttrium silicate phosphors

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ARTICLE INFO

Article history:
Received 31 October 2014
Received in revised form 25 March 2015
Accepted 27 March 2015
Available online 28 March 2015

Keywords:
Y_2SiO_5:Ce
Phosphor
Luminescence
Combustion
Fuel

ABSTRACT

Cerium activated yttrium silicate (Y_2SiO_5:Ce) phosphors were prepared by gel-combustion, using yttrium–cerium nitrate as oxidizer, aspartic acid as fuel and TEOS as source of silicon. Two modalities for samples preparation were approached namely: the classical gel-combustion and sonication gel-combustion. The ultrasound treatment during the gelling stage has a positive effect on the structural and luminescent characteristics of the final product. Therefore, a well crystallized single X2–Y_2SiO_5 phase phosphor was obtained at 1200 °C. Based on FT-IR and XRD investigations, conversion of X1 to X2–Y_2SiO_5 phases is observed as the firing temperature is varied (1100 °C, 1200 °C, 1300 °C 1400 °C). The ultrasound treatment leads to smaller particle size and enhances the luminescent performances up to 151% in comparison with samples prepared by classical way.

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1. Introduction

The capacity of luminescent materials (phosphors) to transform different exciting energies (UV light, X-rays, electrons, α or β radiations) into visible radiation make possible their use in lightening, displays and optical communications fields [1–4]. Phosphors are materials composed from a crystalline host lattice in which activator ions (rare earth or transitional) are incorporated [1].

Accommodation of cerium ions in yttrium silicate host lattice (Y_2SiO_5) leads to an efficient blue emitting phosphor widely studied due to its applications especially in FED [5,6].

Depending on the synthesis conditions, Y_2SiO_5 can be found in two different monoclinic structures named: X1–Y_2SiO_5, the low temperature phase and X2–Y_2SiO_5, high temperature phase. In both X1 and X2 structures there are two crystallographic non-equivalent low symmetry Y^{3+} sites which differ in the oxygen coordination: 7 and 9 for X1 type; 6 and 7 for X2 type respectively [7–10]. Because the X2 structure is more suitable for reaching high emission intensities than X1, researches are focused mainly to obtain pure X2 phase [11]. The usual synthesis route for Y_2SiO_5:Ce powders is the conventional solid state reaction. In this process, Y_2O_3 and SiO_2 are mixed with cerium nitrate and fired at high temperature for long sintering time [12]. When the reactants are heated below 1500 °C, a single phase cannot be obtained.

New methods were developed, for the fabrication of Y_2SiO_5 based phosphors to overcome the specific disadvantages of the classical method that include: sol–gel, melting salt synthesis, hydrothermal, spray pyrolysis or combustion [8,13–16]. Use of ultrasound treatment, also, has been proven to be a useful for the synthesis of large variety of materials [17–19].

The chemical effects of ultrasound arise from acoustic cavitation involving the formation, growth and collapse of bubbles in liquid, which generates localized hot spots having a temperature of roughly 5000 K, pressure of about 20 MPa, and very high cooling rates of about 107 K s^{-1} [20,21].

Ultrasound irradiation was used for the synthesis of different materials namely: fluorides [22]; aluminates [23]; oxides [24] or sulphides [25,26]. Sonication can be also adopted in order to get fine particle size, and avoid the possible particle agglomerations [27].

However, the literature survey brought no information regarding the utilization of ultrasonic treatment during the preparation of Y_2SiO_5:Ce phosphors and its effect on the phosphor characteristics.

The novelty of our work brings together the advantages of both ultrasound irradiation during the gelling stage and combustion method in order to bring improvements on the structural and

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http://dx.doi.org/10.1016/j.materresbull.2015.03.063
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luminescent characteristics of Y_2SiO_5:Ce^{3+} phosphor. This work is a continuation of our previous researches [28] and is focused to investigate the influence of sonication treatment on the morphological, structural and luminescent characteristics of Y_2SiO_5:Ce. In this respect, X2-single phase Y_2SiO_5:Ce phosphors were prepared by gel-combustion from a mixture containing yttrium–cerium nitrates; aspartic acid and TEOS using the ultrasound irradiation during the gelation process.

2. Experimental part

2.1. Materials

Cerium activated yttrium silicate phosphors were prepared by solution combustion from yttrium–cerium nitrates as oxidizer and aspartic acid as fuel. Commercial yttrium oxide–Y_2O_3 (99.9%, Alfa Aesar), cerium nitrate–$Ce(NO_3)_3 \cdot 6H_2O$ (extra pure, Merck), aspartic acid-L-$C_4H_7NO_4$ (99.0%, Alfa Aesar), TEOS–$Si(OC_2H_5)_4$ (98% Alfa Aesar) and ethanol–C_2H_5OH (94–96% Alfa Aesar) were used as starting materials for the phosphor synthesis.

2.2. Sample preparation

A description of synthesis was presented in our previous work [28]. In principle, the combustion method consists in addition of aspartic acid, TEOS and ethanol to a Y–Ce nitrate solution that was kept at 80 °C under continuous stirring until a viscous gel is formed. In this study, two modalities for samples preparation were approached namely: *the classical gel-combustion and *sonication gel-combustion.

The ultrasound treatment was performed by keeping the synthesis mixture in a ultrasound bath (model: ELMA Transsonic T660/H instrument Frequency: 35 kHz; Rated output power: 360 W) for 2.5 h at 80 °C until the gel was formed.

In both cases, the resultant gel is ignited in a heating mantle at 350 °C, resulting precursors with a foamy appearance. Next, the precursors were thermal treated at different temperatures (1100 °C, 1200 °C, 1300 °C and 1400 °C) for 4 h in air. The conditioning stage consists in soft milling with acetone and sieving the fired powders.

The emission intensity of above prepared phosphors was compared with an internal standard, considered to have the luminescent emission of 100%. The synthesis of standard as a single phase X2–Y_2SiO_5:Ce was done by solid-state reaction by mixing proper amounts of Y_2O_3 (Aldrich), $Ce(NO_3)_3$ (Aldrich) and SiO_2 (fumed – Aldrich) followed by thermal treatment at 1400 °C for 4 h in air.

2.3. Sample characterisation

The characterization of samples was done by: Fourier-Transform Infrared Absorption spectroscopy (Thermo Scientific Nicolet 6700 FT-IR Spectrometer; KBr pellet technique); Scanning Electron Microscopy (JEOL–JSM 5510LV Microscope; Au-coated powders); X-ray diffraction (6000 Shimadzu diffractometer, Ni filter, radiation $\lambda_{CuK\alpha}$ = 1.5418 Å, voltage 40 kV, current 30 mA); Surface area measurements (TriStar II3020 – Micromeritics, nitrogen adsorption at 77 K; sample decontamination at 150 °C for 24 h in nitrogen flow) and Photoluminescent measurements (JASCO FP-6500 Spectrofluorimeter Wavel; Glass filter WG 320).

3. Results and discussion

3.1. Structural characterization

Investigations regarding the structure of phosphors were made by X-Ray diffraction. Diffractograms depicted in Fig. 1, show that all samples are well crystallized but the phase composition strongly depends on the preparative conditions.

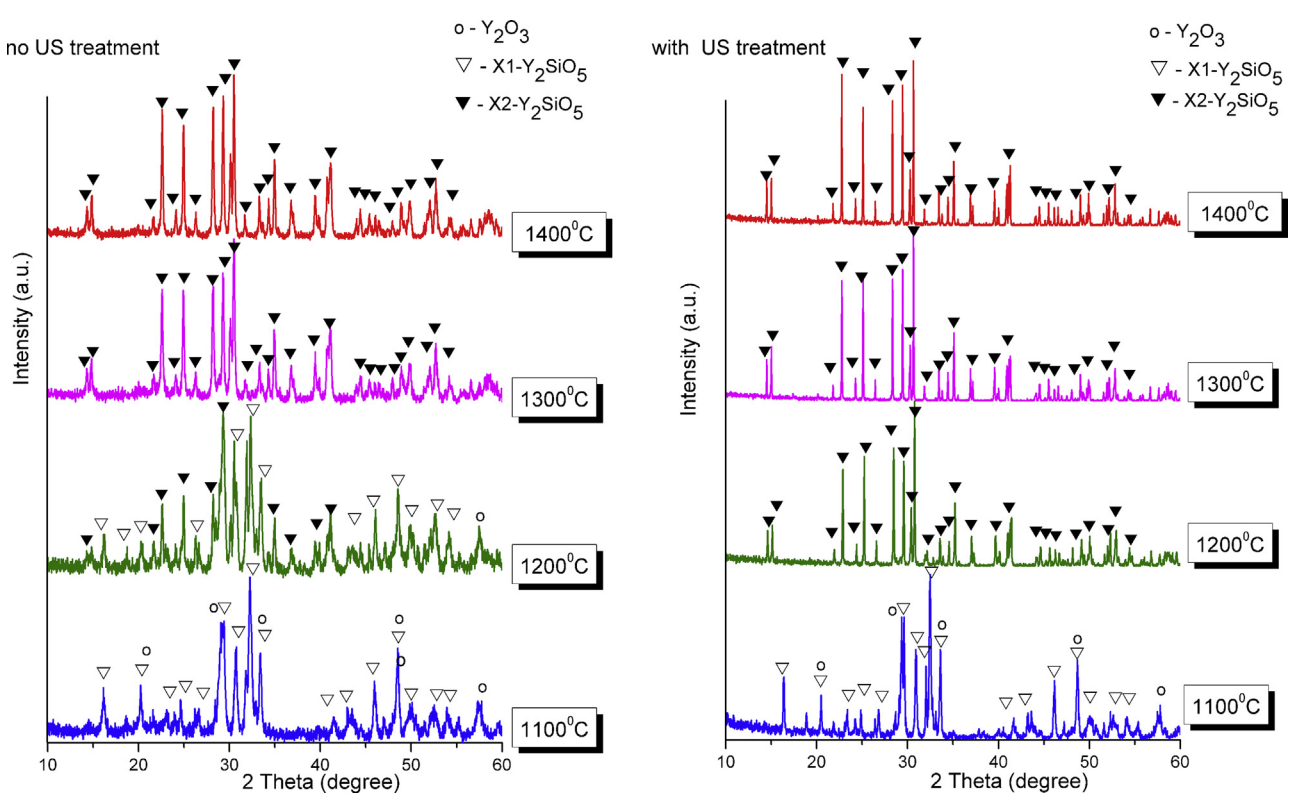

Fig. 1. Normalized X-ray diffractograms of Y_2SiO_5:Ce phosphors (λ_{Cu} = 1.540598 Å).

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