Optical Materials 45 (2015) 156–160

Contents lists available at ScienceDirect

Optical Materials

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

Preparation of translucent $Gd_2Si_2O_7$:Ce polycrystalline thin plates and their scintillation performance for α-particles

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ARTICLE INFO

Article history:
Received 17 February 2015
Received in revised form 4 March 2015
Accepted 8 March 2015
Available online 30 March 2015

Keywords:
Gadolinium pyrosilicate
Liquid-phase sintering
Translucent polycrystal
α-Particle detection

ABSTRACT

Translucent $Gd_2Si_2O_7$:Ce (GPS:Ce) polycrystalline plates were prepared via liquid-phase sintering using SiO_2 as a self-flux, and their scintillation performances for α-particles were investigated. Dense sintered compacts comprising large grains, some of which were larger than 100 μm in diameter, were successfully prepared by sintering at 1690 °C for 100 h. The best result was obtained with the powder comprising only <40 μm particles. Any combination of powders of <40 μm and <15 μm resulted in inhomogeneous structures with smaller grains of about 50 μm. A translucent GPS:Ce thin plate was fabricated by grinding the sintered compact that contained excess SiO_2 of 8 mol%. Since the plate was composed of large grains, scattering at the grain boundaries was effectively suppressed and many of the grains virtually act as single crystals when the plate thickness was less than 100 μm. Therefore, the decrease in the plate thickness brought increase in the total transmission, and light yield and energy resolution were consequently improved. When the plate thickness was 50 μm, light yield was 82% as compared with that of a GPS:Ce single crystal as a reference, and energy resolution attained to 13%.

1. Introduction

In a nuclear fuel reprocessing plants or mixed plutonium–uranium oxide (MOX) fuel plants, the pollution control has been carried out using dust monitors of α-particles in order to ensure worker safety and prevent the emission of radioactive sources to the outside of the facility [1–3]. In this monitoring, α-particles from plutonium, the energies of which are 5.5 or 5.1 MeV, must be discriminated from the α-particles of radon progeny such as ^{214}Po, which emits an α-particle of 7.7 MeV.

Silicon semiconductors, which have excellent energy resolution, are currently used as an α-particles detection material; however, this material is of poor durability and is also easily affected by external electro-magnetic noises [4]. Izaki et al. attempted to apply a ZnS:Ag scintillator for the α-particles detection [5]. Although they could measure α-particles from Pu in the presence of background from radon progeny by optimizing the thickness of the plate comprising ZnS:Ag powder and binder, there was still room for improvement in energy discrimination ability compared with a silicon semiconductor detector.

Conventional, commercially available scintillators are not suitable as an α-particles detector material, e.g., NaI:Tl is deliquescent and canning is accordingly indispensable and Lu_2SiO_5:Ce is of self-luminous enhancing the background [6]. Although Gd_2SiO_5:Ce (GSO) have no self-luminosity, its scintillation light yield is relatively low [7].

$Gd_2Si_2O_7$:Ce (GPS:Ce) is a promising material for α-particles detection because of appreciably high light yield more than 24000 photon/MeV and energy resolution of 4.6% for 662 keV γ-rays [8–11]. In addition, it has excellent chemical durability. In α-particles dust monitors, scintillation materials are used in a form of a thin plate with a large area (e.g. 50 × 50 mm^2). Single crystal growth of GPS:Ce has been tried by the Czochralski method or top-seeded solution growth (TSSG) method [12,13]; however, it is difficult to grow a large-size single crystal, because GPS melts incongruently. In order to obtain large area scintillators, Galunov et al., prepared composites, which were composed of GPS:Ce single crystalline grains and optically transparent, plastic plate, for neutron detection [14]. Shimaoka et al., also fabricated a plate comprising GPS single crystalline grains of several 10 s to 550 μm in diameter with epoxy resin on a glass substrate, and attained an energy resolution of 9.3% (for 5.5 MeV α-particles) [15]. However, the detection efficiency was about 70% at most because it was difficult to arrange the GPS:Ce grains without interspace. In addition, this technique is rather expensive because single crystals are

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http://dx.doi.org/10.1016/j.optmat.2015.03.026
0925-3467/

necessary, which should be grown by the TSSG method using an iridium crucible.

A sintered compact may be an alternative form of a scintillator. Although GPS belongs to the orthorhombic system, which is optically anisotropic, it is possible to enhance the transmittance if the sintered compact is composed of large grains and the plate is sufficiently thin. Therefore, we focused on fabrication of GPS:Ce sintered compacts, in which grains are large enough to act as single crystals virtually. In this study, translucent GPS:Ce polycrystalline plates were prepared via liquid-phase sintering using SiO_2 as a self-flux, and their scintillation performances for α-particles were investigated.

2. Experimental

2.1. Preparation of sintered compacts

Powders of Gd_2O_3 (99.99%), SiO_2 (99.9%) and CeO_2 (99.9%) were used as starting materials. The Ce-concentration was 2.5 at.% in all rare-earth elements. These raw materials were mixed and ground with a planetary ball mill (P-6, Fritsch) using 500 ml pot and 23 balls 20 mm in diameter made of agate at 390 rpm for 30 min with the aid of ethanol. During the milling process, the powder was contaminated by the milling media made of agate resulting in a SiO_2-rich composition. The amount of the incorporated SiO_2 was determined to be about 8 mol% by measuring the weight loss of the milling media.

After forming, the mixed powder was fired at 1700 °C for 10 h in air to be the single phase of GPS. The crystalline phase of the calcined powder was identified by X-ray diffraction (RINT2200, Rigaku) using a Cu target. Two kinds of powders with different particle size distributions were prepared from the calcined compact. One is classified with a sieve of 40 μm opening after grinding with an agate mortar, and the other powder was obtained by grinding in a planetary ball milling (P-7, Fritsch) using a pot of 30 ml and 4 balls 10 mm in diameter made of a gate at 800 rpm for 30 min with the aid of ethanol. Since the pot volume and the ball size were relatively small, contamination from the milling media was negligible at this stage. Particle size distribution of each ground powder were measured with a laser diffraction type apparatus (MICROTRAC HRA 9320X-100, NIKKISO).

Before sintering, the two kinds of powders were mixed in several different weight ratios as described later. Taking account of the contamination by ball milling, total excess SiO_2 was set to be 8, 12 and 16 mol%. After forming pellets under a uniaxial pressure of 70 MPa, they were sintered at 1690 °C for 50 or 100 h in air. The sintered compacts were polished to be mirror finish, followed by thermal etching at 1600 °C for 1 h in air. Microstructures of the sintered compacts were observed with a scanning electron microscope (JSM-6390LVS, JEOL).

2.2. Fabrication of GPS:Ce thin plates and evaluation of their scintillation performance for α-particles

A sintered compact was annealed in N_2 at 1200 °C for 12 h to assure the formation of Ce^{3+}, and then one surface of the compact was polished to be mirror finish. The polished surface was adhered to a glass plate with epoxy resin. The specimen was ground to be 500–50 μm in thickness resulting in a translucent thin plate. The total light-transmission of the plate at 400 nm, which corresponds to the emission wavelength of Ce^{3+}, was measured with a spectrophotometer using an integrating sphere (V-630, JASCO). The scintillation properties for α-particles were evaluated using ^{241}Am. A thin copper plate with a hole of 2 mm in diameter was placed between the α-particles source and the specimen to regulate the flux of α-particles. The specimen was fixed on the photomultiplier tube using optical grease. A photomultiplier tube (H1161, Hamamatsu Photonics), a delay line amplifier (460, Ortec), a pulse stretcher (542, Ortec), and a multi-channel analyzer (WE7562, Yokogawa Electric) were used to evaluate the response for α-particles. The integration time constant of the delay line amplifier was set at 250 ns, considering the decay time of the scintillators.

3. Results and discussion

3.1. Characterization of the calcined powder

Fig. 1 shows the observed XRD pattern of the calcined powder and the simulated pattern on the basis of the orthorhombic $Gd_2Si_2O_7$ structure. The observed pattern showed comparable intensity with the simulated pattern and all the peaks of that were assigned to the single phase of GPS:Ce. Since no trace of the second phase was observed, the excess SiO_2 would form glassy phase. Kaneko et al. [16] tried to synthesize GPS:Ce powder via a conventional ceramic process, in which an agate mortar was used for powder mixing, and repeated grinding and calcinations were necessary to obtain single phase of GPS:Ce. In contrast, it was possible in this study to obtain single phase of GPS:Ce with single calcination. Raw powder materials would be effectively activated by planetary ball milling.

Two kinds of powders with different particle size distributions were prepared from the calcined compact. Fig. 2 shows the particle size distribution of the calcined powders. The powder classified with a sieve of 40 μm opening showed a particle size distribution from submicron to 40 μm with a peak size of 15 μm, termed G15. Most of the particles in G15 powder were primary and did not show remarkable aggregation. The other powder ground by planetary ball milling showed bimodal particle size distribution with peak sizes of 1 μm and 5 μm, termed G5. The particles of submicron would form aggregated particles to show the frequency peak of 1 μm.

3.2. Characterization of the sintered compacts

In a typical sintering process, fine base powder is favorable to obtain a compact as dense as possible since such powder is more active than coarse powder. For an optically anisotropic material such as GPS:Ce, however, a transparent or translucent compact is

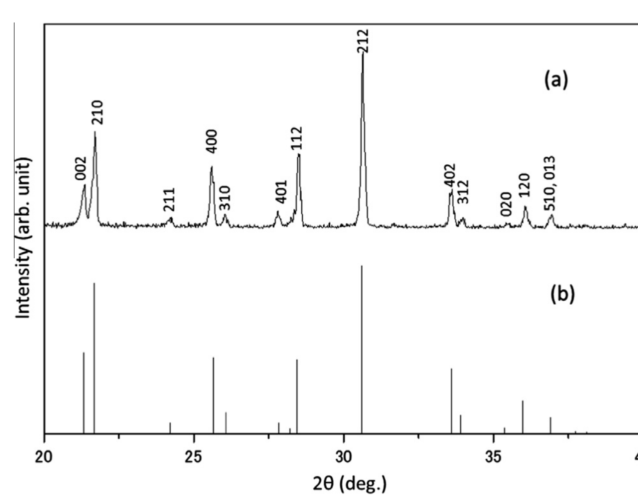

Fig. 1. XRD pattern of (a) the calcined powder and (b) simulation using ICSD #20316.