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ARTICLE

Response Measurement of $Gd_2Si_2O_7$:Ce Scintillator for Alpha Particles

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Gadolinium pyrosilicate ($Gd_2Si_2O_7$, GPS:Ce) scintillators with various cerium concentrations were synthesized using a Top Seeded Solution Growth (TSSG) method. Scintillation properties for 5.5 MeV alpha particles from ^{241}Am are investigated as a part of development of alpha-particle dust monitor for nuclear reprocessing facilities.

Polycrystalline GPS with cerium concentrations of 1.0, 2.5, 10% were fabricated. Orthorhombic GPS with 1–2.5% of cerium has light output 3.5–3.9 times greater than GSO. The triclinic GPS with 10% of cerium is inferior to the orthorhombic GPS in light output. For GPS with 2.5% of cerium, annealing in N_2 atmosphere shows significant improvement in energy resolution.

KEY WORDS: $Gd_2Si_2O_7$, scintillators, crystal structure, decay time, light output

I. Introduction

Contamination control has been conducted with alpha-particle measurements using dust monitors or in-room monitors in facilities where plutonium is handled, e.g., MOX fuel manufacturing facilities and the fuel-reprocessing facilities [1].

For use as a dust monitor, ZnS (Ag) scintillators are widely used because of their durability and large light output. However, for in-room monitors, semiconductor detectors have been used because of their high energy resolution [2].

The radon progeny have a great effect in areas where plutonium is handled because the degree of plutonium contamination is low. The influence of radon progeny has been decreased by pulse height discrimination.

Although the silicon semiconductor detectors used for this purpose have a thicker aluminum electrode than those of conventional silicon semiconductor detectors, corrosion problems arise with the nitric acid atmosphere and/or atmospheric humidity. These devices also present a problem of electric noise vulnerability: accidental alarms have occurred.

To resolve these problems, one of the authors, K. Izaki, developed a highly reliable in-room monitor using a thick-film ZnS(Ag) scintillator and succeeded in discriminating radon progeny for 50% in elimination factor [3]. However, thick-film ZnS(Ag) had only 30 - 40 % of energy resolution for 5.5 MeV alpha-particle. There was room for improvement in terms of the energy discrimination performance.

A gadolinium pyrosilicate ($Gd_2Si_2O_7$:Ce, GPS:Ce) scintillator with high-energy resolution has been developed at Hokkaido University and Hitachi Chemical Co. Ltd. The GPS scintillator had an energy resolution of 5.0% for 662 keV gamma rays, no self-activity and no deliquescence [4,5].

GPS was originally developed as a neutron scintillator for use in neutron science because of the large neutron capture cross-section of gadolinium. Haruna et al. made a scintillator plate by fixing GPS powder on a glass plate with ethylene glycol. They measured a response function for neutrons [6].

For GPS, orthorhombic, triclinic, and other structures appeared depending on the cerium concentration. Two peaks of light output occurred at cerium concentrations of 1.0–2.5% and 10–15% [7]. Improvement of light output by annealing in air or N_2 atmosphere has also been reported [8].

GPS crystals have originally been synthesized in a solid-state reaction or Floating Zone method [9,10]. Only a small quantity of GPS could be obtained by these methods.

A detailed phase diagram around GPS was reported by Kawamura. Thereafter, crystal growth of large diameter GPS was conducted using a Top Seeded Solution Growth (TSSG) method [11,12]. The authors could obtain large amount of crystal, sufficient to develop a large-area scintillator plate using the TSSG method.

Possibilities exist to make an alpha-particle detector with better energy resolution by fixing GPS powder on the plate.

For this study, we synthesized GPS with 1.0, 2.5, 10% of cerium using the TSSG method and measured the response for 5.5 MeV alpha particles from ^{241}Am as a part of this study to develop an alpha-particle dust monitor.

II. Experimental

1. Crystal Growth

Raw materials were powders of Gd_2O_3, SiO_2, and CeO_2 with 4N purity. These were weighed and mixed at compositions of $(Gd_{1-x}Ce_x)_2Si_2O_7$ (x=0.010, 0.025, 0.10). Amounts of SiO_2 in raw materials were slightly larger than those necessary for stoichiometry to obtain silica-rich conditions. The mixed powders were pressed and then sintered at 1650°C for 6 hr in air.

The sintered material was set into an iridium crucible in a RF type Czochralski furnace. Sintered GPS rods with 5mm diameter were chosen as crystal seeds, because there was no crystal that would be large enough for seed. The growth rate and rotation ratio was set respectively at 0.5 mm/h and 20

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rpm. Growth procedures were taken in N_2 atmosphere with 100 – 1000 ppm of O_2.

Figure 1 presents an example of a GPS crystal. Polycrystalline GPS with large grains was obtained. Some crystals were annealed in N_2 atmosphere for 12 hr to reduce coloration. Coloration observed in some crystals was reduced by annealing in N_2.

For scintillation measurements, grown crystals were cut and polished mechanically into approximately 5×5×1 mm.

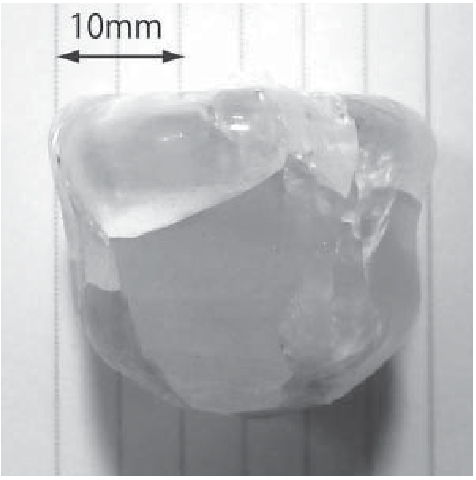

Fig. 1 Sample image of GPS (Ce 2.5%) crystal. Large transparent grains were obtained.

2. Response Measurement for Alpha Particles

For response measurements, 5.5 MeV alpha particles from ^{241}Am were used.

Figure 2 presents a schematic diagram showing the measurement setup. The distance between the checking source and GPS crystals was less than 1 mm to reduce the attenuation of energy by air.

The GPS crystals were set on photomultiplier tube (H1161; Hamamatsu Photonics K.K.) using optical grease. The bias voltage of the photomultiplier tube was set at -1600 V. Pulse height spectra were measured using a multi-channel analyzer (WE7562; Yokogawa Analytical Systems, Inc.). The output cable from the photomultiplier tube was connected to a delay line amplifier (460; Ortec) and linear gate and stretcher (542; Ortec).

For light output measurements, it was difficult to secure sufficient linearity in one amplifier because of a difference of decay time.

To avoid any influence by the amplifier, signals from the dynode of photomultiplier tube were measured directly using an oscilloscope with digital storage (LT584; Lecroy Corp.). An output signal for which the pulse height was located at the peak on a multi-channel analyzer was entered to the single-channel analyzer. It generated a trigger pulse to stop the oscilloscope with digital storage.

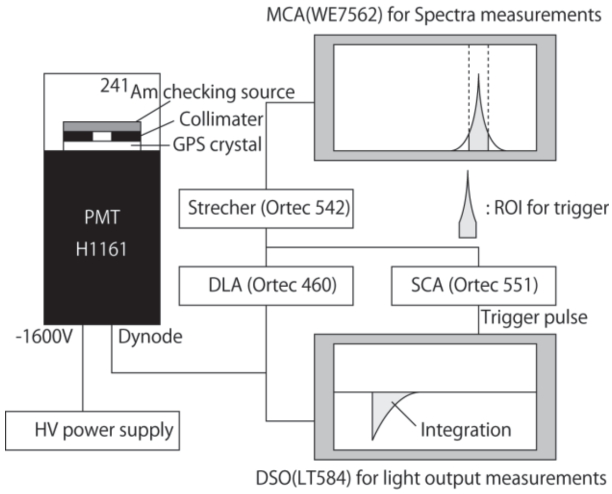

Fig. 2 Response measurement setup for alpha particles. Signals from PMT were recorded directly using an oscilloscope with digital storage.

III. Results and Discussion

Table 1 presents overall results including light output, energy resolution, and decay time. The orthorhombic GPS with 2.5% of cerium shows 3.9 times light output greater than GSO. However, the triclinic GPS with 10% of cerium is inferior to the orthorhombic GPS in light output. It had faster decay time than that of the orthorhombic GPS.

Examples of decay curves are presented in **Figure 3**. The decay time became shorter with increasing cerium concentration. Triclinic GPS with 10% of cerium shows fast decay time of 78 ns. Further investigation is necessary to

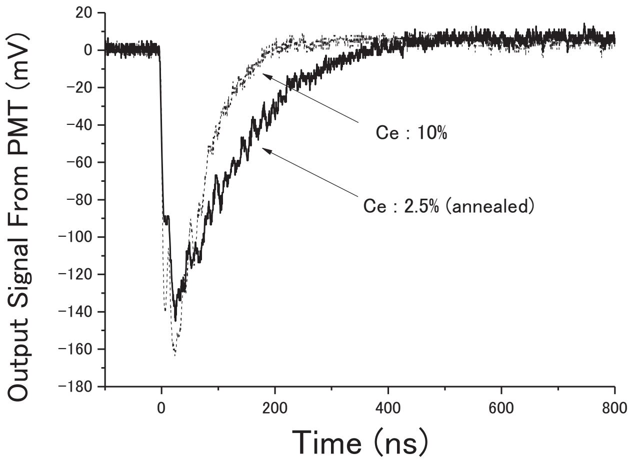

Fig. 3 Example of decay curve of GPS (Ce 2.5 and 10%) for alpha particles from ^{241}Am.

Table 1 Scintillation properties of GPS for 5.5 MeV alpha particles from ^{241}Am.

Sample	Structure	Light output (GSO=1)	Energy resolution (%)	Decay time(ns)
GPS Ce 1.0 %	Orthorhombic	3.5	7.8	195
GPS Ce 2.5%	Orthorhombic	3.7	7.2	141
GPS:Ce 2.5% (annealed)	Orthorhombic	3.9	5.7	150
GPS:Ce 10%	Triclinic	2.4	6.7	78

determine which is the dominant factor producing the effect of fast decay: crystal structure or cerium concentration.

Figure 4 presents an example of response spectra to alpha particles. As-grown GPS crystal had 6.7–7.8% energy resolution. The GPS annealed in N_2 showed better energy resolution of 5.7%. This improvement might result from the increased uniformity of crystal, which would engender reduction of defects and residual stress. Although annealing in N_2 atmosphere will reduce excess oxygen, cerium ions will engender Ce^{3+} from Ce^{4+}. For GPS synthesized using the TSSG method, the annealing treatment enhances the luminescent efficiency.

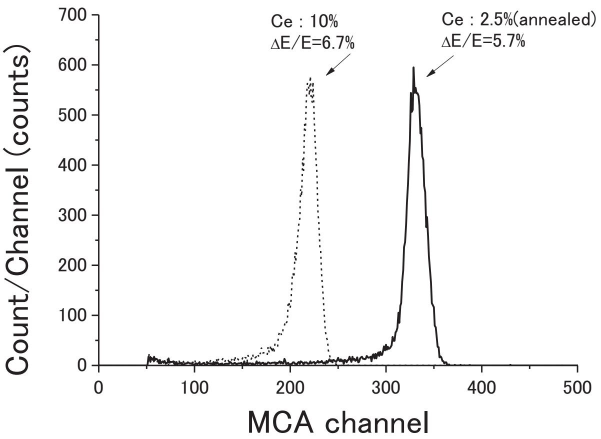

Fig. 4 Comparison of pulse height spectra. GPS with 2.5% of cerium shows 5.7% of energy resolution for 5.5 MeV alpha-particles.

For an alpha-particle dust monitor, GPS with 2.5% of cerium is suitable from the aspect of light output and energy resolution.

IV. Conclusion and Future Works

GPS crystals with 1.0, 2.5, and 10% of cerium were synthesized using a TSSG method. The response for alpha particles was measured. Annealed GPS with 2.5% of cerium showed the highest light output and energy resolution.

Development of a scintillator plate with large diameter using crystal obtained using a TSSG method is underway.

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