Scintillation Properties of Ce:(La, Gd)₂Si₂O₇ at High Temperatures

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Abstract

Temperature dependence of scintillation properties was investigated for (Ce_{0.01}, Gd_{0.90}, La_{0.09})₂Si₂O₇ grown by floating zone method. The light output over 35,000 photons/MeV was found constant in the temperature range from 0 °C to 150°C. In addition, FWHM energy resolution of Ce:La-GPS (roughly 7–8%) at 662 keV remained constant up to 100°C. Thus, this crystal can be applied to oil well logging or other radiation detection application at high temperature conditions.

Keywords: Scintillator, Oil well logging, (Ce_{0.01}, Gd_{0.90}, La_{0.09})₂Si₂O₇

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1. Introduction

Scintillators are used in various fields such as medical imaging, astronomy and so on. Evaluation of temperature dependences of some scintillation parameters such as light output and decay time is important to understand if the scintillation material is suitable for applications in low or high temperature conditions. In oil well logging, scintillator is exposed to temperatures above 100°C in when measuring the subsurface geologic formation properties. Therefore, scintillator with a high light output even at high temperatures is required.

Generally, the light output decreases with increasing temperature and may become quite low for temperatures significantly exceeding the room temperature. However, some scintillators, such as Tl:NaI, Tl:CsI and Ce:Gd₂SiO₅ (Ce:GSO) still have reasonable light outputs of more than 5,000 photons/MeV at high temperature. Thus these scintillators are used in well logging. However, Tl:NaI and Tl:CsI are hygroscopic, therefore oxide scintillator can be more suitable for application in well logging [1-4].

Recently, pyrosilicate scintillation crystals (Ce:Ln₂Si₂O₇, Ln=Gd, Lu) have been reported to have good light output [5-7]; Ce:Lu₂Si₂O₇ showed relatively high light output of 26,300 photons/MeV and good energy resolution of between 9.5% and 12.5% (FWHM) at 662 keV.⁵⁾ Moreover, the scintillation efficiency remains very high even when the temperature increases up to 450 K (177°C) for cerium doped lutetium pyrosilicate Lu₂Si₂O₇ (Ce:LPS) scintillator. On the other hand, decrease of the efficiency below 40% was observed for Lu₂SiO₅ silicate type scintillator when the temperature was raised from 300 to 450K [5]. However, Lu₂Si₂O₇ has intrinsic background signal due to ¹⁷⁶Lu [8], and it is not suitable for oil well logging.

Ce:Gd₂Si₂O₇ (Ce:GPS) had a good light output of 30,000 photons/MeV and FWHM energy resolution of 6.0% at 662 keV at room temperature [6]. The light yield and energy resolution at high temperature have not been reported so far. Additionally, Ce:GPS crystals need to be grown with heavy Ce-doping (approximately, 10 mol%) in order to stabilize the crystal growth process. Such high Ce-concentration would lead to lower light output because of self-absorption or concentration quenching.

Recently, scintillation properties of (Ce_{0.01}, Gd_{0.90}, La_{0.09})₂Si₂O₇ (Ce:La-GPS) have been reported, because the substitution of La instead of Ce can be also applied to stabilize the pyrosilicate phase [9]. The Ce:La-GPS crystal had a light output of approximately 35,000 photons/MeV, FWHM energy resolution of 5.0% at 662 keV. In this paper, we report the light output at high temperature, and we discuss the feasibility of application of Ce:La-GPS in oil well logging.

2. Materials and Experimental Methods

A $(\text{Ce}_{0.01}, \text{Gd}_{0.90}, \text{La}_{0.09})_2\text{Si}_2\text{O}_7$ (Ce:La-GPS) single crystal was grown by the floating zone (FZ) method under argon atmosphere, see the details of the crystal growth process in ref 9. The setup for the light output measurement is shown in Fig. 1(a); the sample with a size of $\sim 2 \text{ mm} \times 2 \text{ mm} \times 1 \text{ mm}$ (thickness) covered with TeflonTM tape as a reflector was coupled to a ruggedized photomultiplier (PMT) (Hamamatsu R1288AH) which can operate up to 200°C. Moreover, this PMT has long service life time of 3,000 and 1,000 hours at 175 and 200°C, respectively, which are 8 and 6 times longer than a conventional ruggedized PMT type at 175 and 200°C, respectively [10]. The PMT was operated at a high voltage of $-1,500 \text{ V}$ and a gain of 0.4×10^6 in the thermostat chamber (Midori HPPC-40-20, Isuzu Co., Ltd). The crystal samples were excited with gamma rays from a ^{137}Cs (662 keV) source. The output signal was preamplified with an integrating time of $5.4 \mu\text{s}$, shaped with a module (572A, ORTEC) using a shaping time of $0.5 \mu\text{s}$, and digitized with a Multichannel Analyzer (MCA8000A, Amptek). After obtaining the pulse height spectra, the ADC channel positions of the photoabsorption peak for 662 keV was determined.

To determine the decay times, decay curves were obtained from the PMT output signal using an oscilloscope (TDS 3052B, Tektronix). The decay curves were fitted with the following equation:

$$I(t) = A_1 \exp[-t/\tau_1] + A_2 \exp[-t/\tau_2] + c, \quad (1)$$

where A_i and τ_i denote the amplitude and decay time ($i = 1, 2$). The term c indicates the background. Each fitting range was 900 ns from the peak. The light output and decay time were tested at 0, 25, 50, 75, 100, 125 and 150 °C.

The temperature dependence of the PMT response was also measured, as shown in Fig. 1 (b). The PMT was irradiated with 400 nm purple light from a light emitting diode (LED) from the outside of the thermostat chamber instead of the Ce:La-GPS scintillation light peaking at 390 nm. The pulse height was then measured under different temperature conditions (0, 25, 50, 75, 100, 125 and 150 °C). Moreover, temperature near the LED was monitored in order to confirm that the temperature and the light output of the LED remain unchanged.

3. Results and discussion

Pulse height spectra of La-GPS excited with gamma rays from a ^{137}Cs source at 0 and 150°C are shown in Fig. 2. The temperature dependence of the light output at 662 keV for Ce:La-GPS is shown in Fig. 3, where the light output at 0°C is normalized to 1.

In order to measure the temperature dependence of the PMT gain, pulse height spectra of the PMT under irradiation with 400 nm light from the LED, were obtained without a Ce:La-GPS scintillator, and each spectrum was fitted by a Gaussian function. The best-fit line for the data points of LED light output is represented as

$$\begin{aligned} & \text{(Relative PMT gain)} \\ & = 1.0 - 0.0034 \times (\text{Temperature } [^{\circ}\text{C}]), \end{aligned} \quad (2)$$

where the light output and peak ADC channel at 0°C were normalized to 1. The temperature coefficient of the dependence of the PMT gain was determined to be approximately $-0.3\%/^{\circ}\text{C}$ for our Hamamatsu R1288AH, which was consistent with other PMTs, for example Hamamatsu R6877A which is also used in oil well logging and which has temperature coefficient of $-0.4\%/^{\circ}\text{C}$ [3]. We corrected the light output of the Ce:La-GPS crystal using the PMT temperature coefficient (Fig 4). These results show that the light output of Ce:La-GPS remained very high when the temperature increases up to 150°C, which is similar as for the Ce:LPS scintillator [5].

Temperature dependence of FWHM Energy resolutions at 662 keV for Ce:La-GPS is shown in Fig. 5. Here, the pulse height spectra at 0 to 125°C showed asymmetric peak consisting of 662-keV absorption peak and Gd-escape peak. These spectra was fitted with “*double Gaussian + linear function*” in order to determine the energy resolution. Generally, the energy resolution gets worse as the temperature increases, while the energy resolutions of Ce:La-GPS did not change significantly up to 100°C (approximately 7-8 %). Since the absorption and escape peak cannot be distinguished in the pulse height spectrum at 150°C, the energy resolution at 150°C changed by approximately 5%-point with respect to the other temperatures.

In our previous work [9], the FWHM energy resolution of Ce:La-GPS was 5.0% at 662 keV at room temperature, while this resolution was roughly 7.6% at 25°C for the crystal in this work. Here, the quantum efficiency (QE) of the ruggedized PMT (Hamamatsu R1288AH) of

roughly 15% at 390 nm is lower than that of R7600-200 we used in the previous work (QE of 40%). That is why the energy resolutions were different even the same sample was measured.

Figure 6 shows the temperature dependence of the decay time for the Ce:La-GPS under gamma ray excitation from a ^{137}Cs source. The decay curves consisted of two components: primary (shorter decay time) and secondary (longer decay time). The corresponding decay times were roughly 60 – 40 and 440 – 370 ns, respectively. Additionally, the fraction of total scintillation light for the primary one, defined as $A_1/(A_1 + A_2)$ in (1), was between 80–90% in the studied temperature range. The fraction of total scintillation light for the primary component increased with increasing temperature (Fig. 7).

Both Ce:LPS and Ce:La-GPS belong to the pyrosilicate group, and a maximum emission wavelengths are almost the same around 400 nm, where a PMT had maximum quantum efficiency in this region. Ce:LPS had a good light output of 26,300 photons/MeV, which remain practically constant or slightly increased by approximately 10% around 177°C compared with the light output at room temperature [5]. While the FWHM energy resolution at 662 keV were 9.5 – 12.5% at room temperature, Ce:LPS has intrinsic background. On the other hand, Ce:La-GPS had good energy resolution of roughly 7–8% from room temperature to 100°C. Moreover, Ce:La-GPS had good light output of over ~35,000 photons/MeV from room temperature to 150°C, and value of its intrinsic background is smaller than Ce:LPS by approximately two orders of magnitude. This background can be negligible for the oil well logging.

Although Ce:GSO scintillator has also no intrinsic background, the light output and energy resolution are worse when compared to those of Ce:La-GPS at room temperature. Moreover, the relative light output and energy resolution of Ce:GSO are degraded over 100°C [6]. From the above results, Ce:La-GPS is expected to be more suitable for application in well logging.

4. Conclusions

We investigated the temperature dependence of the light output and decay times for $(\text{Ce}_{0.01}, \text{Gd}_{0.90}, \text{La}_{0.09})_2\text{Si}_2\text{O}_7$ to confirm its potential applications in oil well logging. Ce:La-GPS was found to keep a good light output at even 150°C, and good FWHM energy resolution at 662 keV remained constant around 7–8% up to 100°C. Moreover, Ce:La-GPS has small intrinsic background and no hygroscopic nature. Therefore, this material can be applied to oil well logging or other radiation detection application at high temperature conditions.

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Figure caption:

Fig.1 Schematic view of setup for temperature dependence of light output for the scintillator (a) and that of photomultiplier response (b).

Fig.2 Pulse height spectra of Ce:La-GPS irradiated with gamma rays from a ^{137}Cs source at 150°C (solid line) and 0°C (dotted line).

Fig. 3 Light output of Ce:La-GPS irradiated gamma-rays from a ^{137}Cs source as a function of temperature. The light outputs at 0°C were normalized to 1.

Fig. 4 Corrected light output of Ce:La-GPS irradiated gamma-rays from a ^{137}Cs source as a function of temperature. The light outputs at 0°C were normalized to 1.

Fig. 5 FWHM energy resolution at 662 keV as a function of temperature.

Fig. 6 Decay time as a function of temperature. Close and open circles denote primary and secondary components.

Fig. 7 Fraction of scintillation decay time between primary and secondary components as a function of temperature.

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