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Scintillation properties of the $Gd_3Al_2Ga_3O_{12}$: Ce crystal

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We investigated luminescence and scintillation properties of the $Gd_3Al_2Ga_3O_{12}$: Ce (GAGG) crystal grown in TPS(Total Polishing Solution) Co.. The GAGG crystal has the highest light yield among oxide crystal at room temperature and fast decay time for the detection of radioactivity and in nuclear and particle physics experiments and medical imaging. The scintillation properties of the GAGG crystal were studied by comparing the GAGG crystals grown by two companies. We measured an X-ray induced emission spectra, an energy resolution, an absolute light yield and a decay time. A pulse shape discrimination capability for a and g-ray was also demonstrated.

Key words: GAGG, Crystal scintillator, Energy resolution, Czochralski, Decay time, Luminescence.

Introduction

Scintillation crystals are often and widely used for detecting various particles and radiations in the fields of nuclear, particle and astro-physics, radiation detection as well as medical imaging. Nowadays having different scintillation properties various kind of the scintillation crystals ware developed. Having a high light yield, a fast decay time without signals pileup, a high effective Z number for an efficient radiation or particles detection and a good energy resolution could make scintillators promising [1-4]. A $Gd_3Al_2Ga_3O_{12}$: Ce (GAGG) crystal was first developed by Tohoku group which showed the high light yield (22-46,000), the fast decay time of main component (68-92 ns) and excellent energy resolution (4.8-11.9%) [5-7] due to the 5d-4f emission of Ce^{3+} at the peak emission around 520 nm. It has rather high density of 6.63 g/cm^3, effective atomic number of 54.4 and pulse shape separation capability [8].

We grew GAGG crystal by Czochalski method and the purpose of the present study is to present the luminescence and scintillation properties of our grown crystal and compare with other groups. Luminescence and scintillation properties include the measurements of X-ray excitation emission spectrum, decay time, energy resolution, scintillation light yield and α/β ratio as well as pulse shape discrimination by mean time method.

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Crystal Growth

The Czochralski crystal growth process is still one of the leading crystal growth methods. The set of Ce-doped $Gd_3(Ga, Al)_5O_{12}$ single crystals were grown by Czochralski technique. Concentration of Ce was 1%, except for $Gd_3Ga_3Al_2O_{12}$ host where 1% and 2% Ce-doped crystals were prepared.

Ce : $Gd_3(Ga, Al)_5O_{12}$ garnet is a multi-component garnet type crystal that demonstrates very high light yield with moderately fast scintillation response. In the current, the Ce-doped GAGG attracted much attention as a scintillating material. The GAGG crystals were produced in Ar + 1 ~ 2%O_2 atmosphere from inductively heated Iridium crucible with diameter and height of 100 mm each. The growth atmosphere was selected to prevent oxidation of the crucible and to reduce evaporation of Ga_2O_3 from the surface of the melt at the same time. The growth was performed with automatic diameter control.

Starting materials were prepared from the stoichiometric mixture of 4N ~ 5N purity Gd_2O_3, Ga_2O_3 and Al_2O_3 powders with respect to ideal $Gd_3(Ga, Al)_5O_{12}$ composition. The crucible was heated inductively at a frequency of 15 kHz. This was necessary to perform the process from the Gd-enriched melt corresponding to congruently melting composition of GAGG.

The steady state pulling rate for the step of constant diameter growth was 1 ~ 2 mm/h with crystal rotation rate of 10 ~ 20 rpm that established at solid-liquid interface. At faster rotation, the interface became convex resulting undesired crystal faceting. In addition, it was detected that the crack formation was more intense when the crystals were faceted that was result

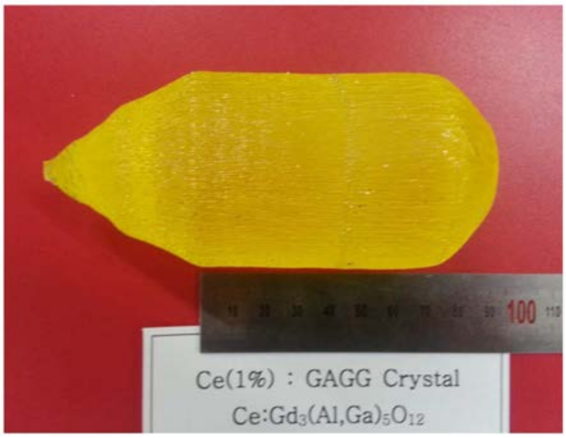

Fig. 1. A GAGG single crystal grown by TPS used in this study.

of convex shape of the solid-liquid interface.

The crystal obtained from the melt was about 50 mm in diameter and 100 mm long, and it demonstrated reasonable optical quality and had no visible defects as shown in Fig. 1.

Experimental Setup

The GAGG crystals ware cut for the study of scintillation properties. The size of sample grown by TPS is $2 \times 1 \times 1$ cm^3 and grown by Russia is $0.5 \times 0.5 \times 0.5$ cm^3 as shown in Fig. 2. Every experiment was performed at room temperature.

The X-ray luminescence characteristics of the GAGG crystals were measured with X-ray tube from a DRGEM Co. having a W anode. The power settings used for measurement were 90 kV and 1 mA. The emission spectra was measured with QE65000 fiber optic spectrometer made by Ocean Optics.

The pulse height spectra under 662 keV γ-ray from a ^{137}Cs radio-active source were recorded with direct coupling of the sample crystals with the 3 inch bi-alkali photomultiplier tube (PMT) (R6233, Hamamatsu Co) with index matching optical grease and value of high voltage was –900V. To minimize the light losses, prior to the coupling with the PMT, all faces of the sample crystals except the one facing PMT are wrapped in several layers of 0.1-mm-thick UV reflecting Teflon tape. The signal from the PMT was amplified and shaped into a Gaussian distribution with a shaping amplifier (ORTEC 571). A 25 MHz flash analog-to-digital converter (FADC) (NOTICE Co.) was used to digitize the output signal. A software threshold setting was applied to trigger an event by using a self-trigger algorithm on the field programmable gate array chip on the FADC board. By using a USB2 connection, the FADC output data was recorded into a computer and the data were analyzed with a C++ data analysis program [9]. From this, we could obtain an energy spectra and the energy resolution with the full width at half maximum [1].

For the study of an absolute light yield, a 16 mm windowed large area avalanche photodiode (LAAPD) (630-70-74-501, Advanced Photonix Inc.) was used. Under the 662 keV γ-ray, the GAGG crystal was attached to the LAAPD with the optical grease and high voltage applied was −1800V. The signal from the LAAPD went through a low noise preamplifier (TC175B, TENNELEC) and then into the shaping amplifier [10]. Then, with the 25 MHz FADC and the C++ data analysis program, the energy spectra for measuring the absolute light yield was taken.

To determine a decay time and an α/β ratio, ^{137}Cs for 662 keV γ-ray and ^{241}Am for 5486 keV α-ray were used. The signal from the PMT was fed into a 400 MHz FADC (NOTICE Co.). The home-made FADC module is designed to sample the pulse every 2.5 ns for duration up to 32 μs so that one can fully reconstruct each photoelectron pulse. With a C++ based data analysis program, we gathered the data, analyzed it and obtained the decay time and the energy spectrum for the α/β ratio [11]. For analyzing a pulse shape under γ-ray and α-ray, ^{60}Co (1173 and 1333 keV) radio-active source which makes the signal in same channel with 5486 keV α-ray was additionally used.

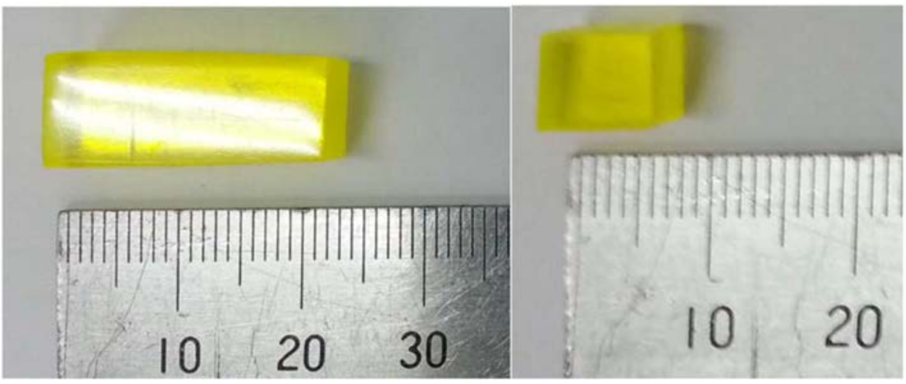

Fig. 2. GAGG samples grown by TPS (left) and Russia (right) used in this study.

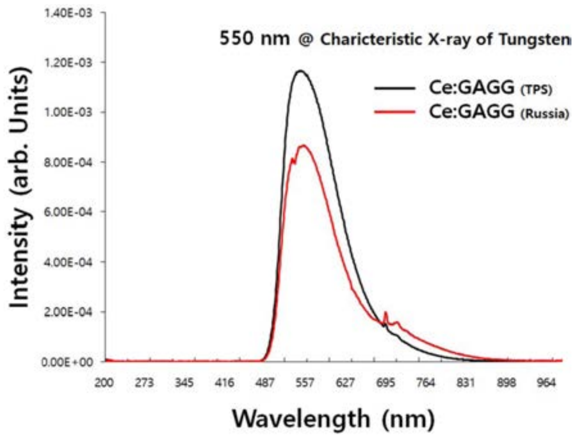

Fig. 3. X-ray induced emission spectra of the GAGG crystal.

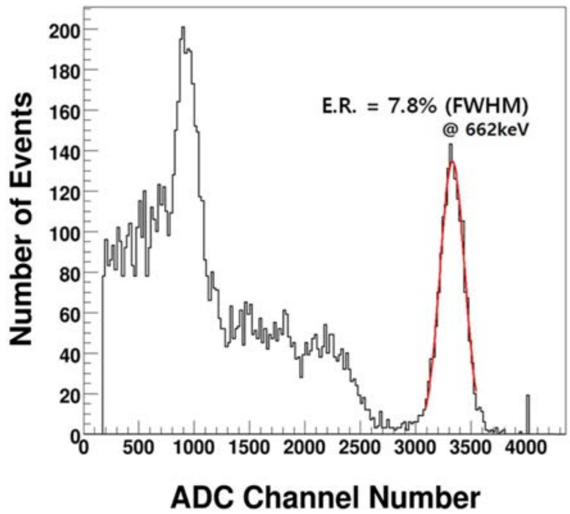

Fig. 4. The pulse height spectra of the GAGG (TPS) crystal.

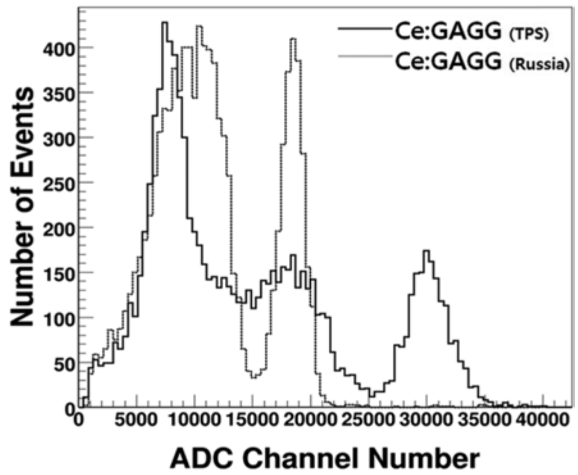

Fig. 5. The pulse height spectra of the GAGG (Russia and TPS) crystals.

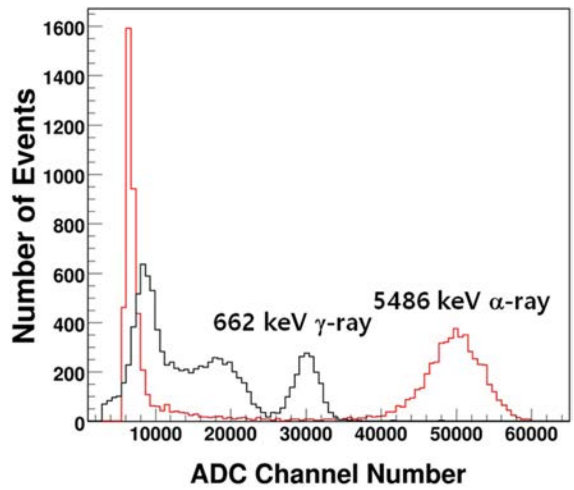

Fig. 6. The pulse height spectra of the GAGG (TPS) crystal with ^{137}Cs, ^{60}Co and ^{241}Am radio-active source.

Results and Discussion

The X-ray induced emission spectra of the GAGG crystals (TPS and Russia) at room temperature is shown in Fig. 3. The emission spectra is located between 480-730 nm peaking at around 550 nm, which is ascribed to the 5d-4f emission of the Ce^{3+}.

The pulse height spectra of the GAGG crystal in shown in Fig. 4. Under the 662 keV γ-ray, the GAGG crystal (TPS) has a 7.8% energy resolution and the GAGG crystal (Russia) has a 9.9% energy resolution with bi-alkali PMT.

To estimate an absolute light yield of the GAGG crystal (Russia), we used a calibrated crystal CsI : Tl having an absolute light yield of 52,000 ph/MeV. The reason for using the CsI : Tl as a reference is an almost same emission wavelength peaking at 550 nm with the GAGG crystal. The absolute light yield of the CsI:Tl crystal was measured with a windowless LAAPD and the ^{137}Cs. Under 5.9 keV X-ray from ^{55}Fe, the number of e-h pairs per channel were calculated and then the absolute light yield considering the quantum efficiency of the CsI : Tl under the 662 keV γ-ray was obtained [10]. The absolute light yield of the GAGG crystal was obtained by comparing the responses of the CsI : Tl and the GAGG (Russia) from the windowed LAAPD, and it corresponds to 46,000 ± 4,600 ph/MeV which shows consistent result compare with Ref. 8. The absolute light yield of the GAGG crystal (TPS) was calculated from the pulse height spectrum measured using the 3 inch PMT. Since a ratio of the peak channels is a ratio of the light yields, the absolute light yield of the GAGG (TPS) is approximately 74,000 ± 7400 ph/MeV. The TPS GAGG shows 65% higher light yield than that of the Russian GAGG.

An α/β ratio show the ratio of emitted light under the α and γ-ray. In the case of the α-ray interaction with a crystal, lots of the energy is lost in other ways, not the light. Fig. 6 shows the pulse height spectra of the GAGG under the radiation from ^{137}Cs (662 keV, γ-ray) and ^{241}Am (5486 keV, α-ray). Let us suppose that the pulse height for the α-ray divided the energy, 5486 keV

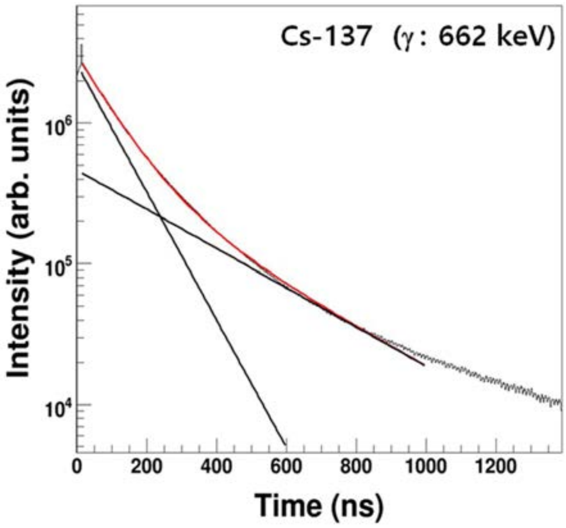

Fig. 7. The decay time spectra of the GAGG crystal with ^{137}Cs radio-active source.

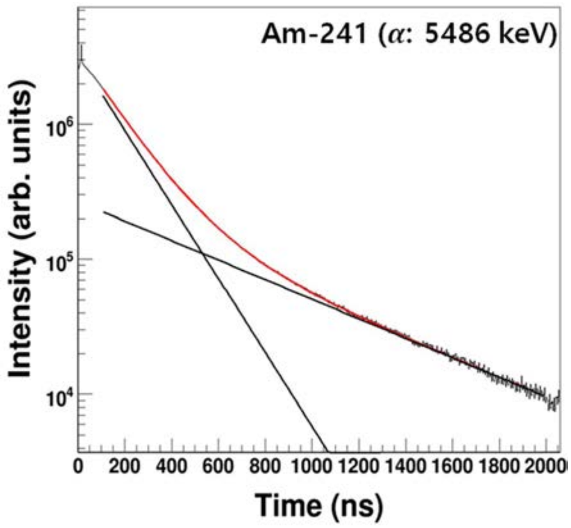

Fig. 8. The decay time spectra of the GAGG crystal with ^{241}Am radio-active source.

is an α and the pulse height for the γ-ray divided the 662 keV is a β. Then, the α/β is 0.2, which means that if the amount of emitted light for the 1 keV γ-ray is 1, the amount of emitted light for the 1 keV α-ray is 0.2 [12].

A decay time of the GAGG crystal under the radiation of α and γ-ray was determined. The decay time has 2 components. Under the 662 keV γ-ray, the main component is 92 ns (60%) and the slow component is 278 ns (40%). Under the 5486 keV α-ray, the main component is 158 ns (76%) and the long component is 601 ns (24%). The decay time for the γ-ray is much shorter than the that of the α-ray. From this, we can expect that the signals of γ-ray and α-ray discriminate with the pulse shape.

We show the two dimensional histogram of mean time vs. pulse height as shown in Fig. 9. The mean time is the time average weighting the pulse height,

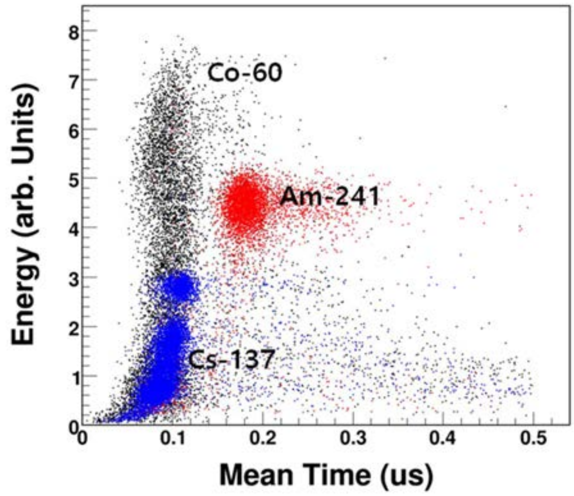

Fig. 9. Energy vs. mean time distribution of the GAGG (TPS) crystal with ^{137}Cs, ^{60}Co and ^{241}Am radio-active source.

defined as

$$\langle t \rangle = \frac{\sum t_i \times E_i}{\sum E_i}$$

, and where, E_i is the amplitude of the pulse at the time t_i up to 24 μs. It is practically the same as the decay time. Two clear bands in the Fig. 9 indicate that we can make good separation between the α particles and γ-rays. Two peaks are well separated by more than 3 sigma at the same energy region of α particles [13].

Conclusions

We performed the luminescence and scintillation properties measurement of the GAGG crystal grown by the Czhocralski method. The emission wavelength under the X-ray is 480-780 nm peaking at 550 nm. In the pulse height spectra with a bi-alkali PMT the energy resolution under the 662 keV γ-ray (^{137}Cs) obtained with FWHM is 7.8%, which is better than that of the crystal made in the Russia (9.9%). The absolute light yield under the same excitation, ^{137}Cs, is 74,000 ± 7400 ph/MeV, which is 65% higher than previously reported one. The α/β ratio obtained under the α-ray from the ^{241}Am and the γ-ray from the ^{137}Cs obtained to be 0.2. The decay time for the γ-ray is 92 ns (60%) and 278 ns (40%) and for the α-ray is 158 ns (76%) and 601 ns (24%). We demonstrated that α and γ-ray are separated more than 3 sigma using mean-time pulse shape discrimination method since the GAGG crystal have the different decay times for α and γ-ray.

Acknowledgments

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