Scintillation properties of $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}^{3+}$ single crystal scintillators

Ongsa Sakthong^a, Weerapong Chewpraditkul^{a*}, Chalerm Wanarak^a, Kei Kamada^c, Akira Yoshikawa^{b,c}, Petr Prusa^{d,e} and Martin Nikl^e

^a Department of Physics, King Mongkut's University of Technology Thonburi, Bangkok 10140, Thailand

^b Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan

^c NICHe, Tohoku University 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai, Miyagi 980-8579, Japan

^d Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, 11519 Prague, Czech Republic

^e Institute of Physics, AS CR, Prague 16253, Czech Republic

*Corresponding author. Tel.: +662 470 8944; Fax: +662 470 8900; E-mail address: weerapong.che@kmutt.ac.th

Abstract

The scintillation properties of $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}^{3+}$ (GAGG:Ce) single crystals grown by Czochralski method with 1 at.% cerium in the melt were investigated and results compared with so far published results in literature. The light yield (LY) and energy resolution were measured using XP5200B photomultiplier. Despite about twice higher LY for GAGG:Ce, the energy resolution is only slightly better than that of LuAG:Ce due to its worse intrinsic resolution and non-proportionality of LY. The LY dependences on the sample thickness and amplifier shaping time were measured. The estimated photofraction in pulse height spectra of 320 and 662 keV γ -rays and the total mass attenuation coefficient at 662 keV γ -rays were also determined and compared with the theoretical ones calculated using the WinXCom program.

Keywords: Energy resolution; GAGG:Ce; Light yield; Photofraction; Scintillation detectors

1. Introduction

Single crystal scintillators are widely used for the detection of ionizing radiation in nuclear and high energy physics, modern medical imaging, space exploration, and industry. The fast and efficient $5d \rightarrow 4f$ luminescence of Ce^{3+} makes it a well-suited emission center in scintillator applications. As a result, new types of Ce-doped inorganic scintillators were intensively studied in the last two decades and some of them were successfully commercialized, for reviews see [1]–[4].

Oxide materials based on garnet structure are promising candidates for scintillator applications because of the well-mastered technology developed for laser hosts and easy doping by rare-earth elements. The Ce-doped $\text{Lu}_3\text{Al}_5\text{O}_{12}$ (LuAG:Ce) single crystal was shown to be a prospective scintillator material with a relatively high density of 6.7 g/cm^3 and a fast scintillation response of about 60–80 ns [5]. Due to technological improvements the reported light yield (LY) has gradually increased from about 14,000 up to 25,000 photons/MeV [6,7]. Scintillation performance of LuAG:Ce is strongly degraded by the presence of shallow electron traps which delay an energy delivery to the Ce^{3+} emission centers and give rise to intense slow components in the scintillation decay [8, 9]. These traps were ascribed to the antisite Lu_{Al} defects in the LuAG host [10] which are typical defects in the garnet crystals grown from high temperature

melt [11,12]. It has been reported that Ga admixture in LuAG host diminishes the energy trapping effects [13] and somewhat increased LY was obtained for Ga concentration up to 20 at. % in $\text{Lu}_3(\text{Al,Ga})_5\text{O}_{12}:\text{Ce}$ [14]. Recently, $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}$ single crystal grown by micro-pulling down method demonstrated high LY of 42,000 photons/MeV(ph/MeV) and energy resolution of 8.3% at 662 keV as measured with avalanche photodiode (APD) at room temperature [15]. An improvement of the scintillator performance was achieved in $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}$ single crystal grown by the Czochralski method which show high LY of 46,000-50,600 ph/MeV and good energy resolution of 4.9 -5.5 % at 662 keV [16,17]. However, lower LY of about 33,000 ph/MeV and energy resolution of 6.1% at 662 keV was reported at the samples of the same composition by another group [18]. The composition of multicomponent garnet scintillators influences strongly the energy transfer processes and interaction of Ce^{3+} and Pr^{3+} emission centers with the host. Fundamental aspects of these processes became intensively studied and of abroad interest [19-22] and the research results in the field of garnet scintillators in the last decade have been recently summarized in a review paper [23].

In this paper we have investigated the scintillation properties of the latest generation of $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}$ (GAGG:Ce) single crystals grown by the Czochralski method. The measurements of pulse height spectra were performed in order to evaluate LY, its non-proportionality and energy resolution. The LY dependences on the sample thickness and amplifier shaping time were measured. The estimated photofraction in pulse height spectra of 320 and 662 keV γ -rays and the total mass attenuation coefficient at 662 keV γ -rays were also determined and compared with the theoretical ones calculated using the WinXCom program.

2. Experimental

GAGG:Ce single crystals were produced in Materials Research Laboratory, Furukawa Co. Ltd. in Japan [16]. The crystals were grown by Czochralski method with cerium concentration of 1 at % ($\text{Ce}_{0.03}\text{Gd}_{2.97}\text{Ga}_3\text{Al}_2\text{O}_{12}$) from Ir crucible under Ar with adding 1.5% of O_2 atmosphere. A stoichiometric mixture of high purity 4N CeO_2 , Gd_2O_3 , $\beta\text{-Ga}_2\text{O}_3$, and $\alpha\text{-Al}_2\text{O}_3$ powders was used as starting materials. Polished plates of 1, 2 and 5 mm thickness cut from the parent sample were used for the measurements. The crystal density was 6.69 g/cm^3 determined by Archimedes method.

Light yield measurements were performed under the excitation of 662 keV γ -rays from a ^{137}Cs source using a photomultiplier (PMT) based-setup described in [24]: the signal from a Photonis XP5200B PMT anode was sent to a CANBERRA 2005 preamplifier and then to a Tennelec TC243 spectroscopy amplifier. The PC-based multichannel analyzer (Tukan 8k MCA) was used to record the pulse height spectra. The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) of energy deposited in the crystal, was determined by means of a single photoelectron method [24, 25]. In this method the number of photoelectrons is measured by comparing the position of a full energy peak of γ -rays detected in the crystals with that of the single photoelectron peak from the photocathode. The measurements of LY non-

proportionality and energy resolution were carried out for a series of X/ γ -rays emitted by different radioactive sources in the energy range from 32.1 to 1274.5 keV.

The total mass attenuation coefficient at 662 keV γ -rays was determined using the good geometry arrangement of source (15 mCi ^{137}Cs), absorber ($5 \times 5 \times 1 \text{ mm}^3$ GAGG:Ce sample) and NaI:Tl detector. A narrow beam of γ -rays is defined by circular apertures ($\varnothing 4 \text{ mm}$) in the Pb-collimators of source and detector, placed at a distance of 60 cm. All measurements were carried out at room temperature (RT).

3. Results and discussion

3.1. Light yield and energy resolution

Fig.1 presents the pulse height spectra of γ -rays from ^{137}Cs (662 keV) source as measured at **4 μs shaping time** for GAGG:Ce, LuAG:Ce and BGO crystals **with same size of $5 \times 5 \times 1 \text{ mm}^3$** , whereas the photoelectron yield and energy resolution values are summarized in Table 1. **The photoelectron yield (phe/MeV) was recalculated to the LY (ph/MeV) using the average quantum efficiency $\overline{QE}$ of 18.5 % for GAGG:Ce & LuAG:Ce and $\overline{QE}$ of 22 % for BGO, based on the typical quantum efficiency characteristic of PMT provided by manufacturer and the emission spectrum of the crystals.** Despite about twice higher LY for GAGG:Ce, its energy resolution is only slightly better than that of LuAG:Ce. It could be associated with its worse intrinsic resolution and non-proportionality of LY, see below.

Fig. 1. Pulse height spectra of 662 keV γ -rays (^{137}Cs source) measured with GAGG:Ce, LuAG:Ce and BGO crystals **with same size of $5 \times 5 \times 1 \text{ mm}^3$** .

Table 1 Photoelectron yield, LY and energy resolution for GAGG:Ce, LuAG:Ce and BGO crystals **with same size of 5 x 5 x 1 mm³** measured at 4 μ s shaping time.

Crystal	Photoelectron yield (phe/MeV)	LY (ph/MeV)	$\Delta E/E$ (%)
GAGG:Ce	8,860 $\pm$ 440	47,900 $\pm$ 4800	6.8 $\pm$ 0.2
LuAG:Ce	4,300 $\pm$ 220	23,200 $\pm$ 2300	7.3 $\pm$ 0.2
BGO	1,860 $\pm$ 100	8,450 $\pm$ 800	9.1 $\pm$ 0.3

We also studied the dependence of LY on the **sample thickness (h)** for GAGG:Ce. The pulse height spectra of 662 keV γ -rays measured for the studied samples are shown in Fig. 2, whereas the values of LY and energy resolution are summarized in Table 2. **The LY value of 47,900 ph/MeV (energy resolution of 6.8%) obtained in this work for a 5 x 5 x 1 mm³ GAGG:Ce sample is comparable to that of 46,000 - 50,600 ph/MeV (energy resolution of 4.9 - 7.3%) recently measured for the same size GAGG:Ce samples [16,17]. We note that LY value of the studied GAGG:Ce decreases with thickness down to 42,100 ph/MeV (88 % LY of 1 mm thick sample) for 5 mm thick sample, which is better than the value (85 % LY of 1 mm thick sample) measured for 4.5 mm thick sample in Ref. [17]. It indicates that scintillation light loss due to self-absorption and photon scattering in the studied GAGG:Ce is smaller than that of the samples in Ref. [17]. This feature is of importance for maintaining high LY with increasing sample size. Despite a comparable photoelectron yield, the energy resolution of 6.8% obtained for a 5 x 5 x 1 mm³ sample is worse than that of 6.1% observed for a 10 x 10 x 5 mm³ GAGG:Ce sample in Ref.[18], which is due to a higher contribution of the intrinsic resolution (6.0 % vs. 5.2%) for the studied sample, see below.**

Fig. 2. Pulse height spectra of ¹³⁷Cs γ -rays measured for the GAGG:Ce samples with thickness **(h)** of 1, 2 and 5 mm.

Table 2 LY and energy resolution (at 662 keV) measured at 4 μ s shaping time for studied GAGG:Ce samples with **thickness of 1, 2 and 5 mm**.

Sample size (mm ³)	LY (ph/MeV)	Relative LY (% of 1 mm)	$\Delta E/E$ (%)
5 x 5 x 1	47,900 $\pm$ 4800	100	6.8 $\pm$ 0.2
5 x 5 x 2	45,400 $\pm$ 4500	95	7.0 $\pm$ 0.2
5 x 5 x 5	42,100 $\pm$ 4200	88	7.3 $\pm$ 0.3

The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a PMT can be written as [26]

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2, \quad (1)$$

where δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution and δ_{st} is the statistical contribution of PMT to the resolution.

The statistical uncertainty of the signal from the PMT can be described as

$$\delta_{st} = 2.355 \times 1/N^{1/2} \times (1 + \epsilon)^{1/2}, \quad (2)$$

where **N is the number of the photoelectrons** and ϵ is the variance of the electron multiplier gain, equal to 0.1 for an XP5200B PMT.

Overall energy resolution and PMT resolution can be determined experimentally. If δ_p is negligible, intrinsic resolution δ_{sc} of a crystal can be written as follows

$$(\delta_{sc})^2 = (\Delta E/E)^2 - (\delta_{st})^2. \quad (3)$$

Fig. 3 presents the measured energy resolution versus energy of γ -rays for a **5 x 5 x 1 mm³** GAGG:Ce crystal. Other curves shown in Fig. 3 represent the PMT resolution (δ_{st}) and intrinsic resolution (δ_{sc}). Apparently, the energy resolution for the GAGG:Ce crystal is mainly contributed by the intrinsic resolution over whole energy range from 32 to 1274.5 keV, reflecting to its high LY. An analysis of the 662 keV energy resolution for GAGG:Ce compared to LuAG:Ce is presented in Table 3. Despite about twice higher LY for GAGG:Ce, its energy resolution is slightly better than that of LuAG:Ce, which is due to its worse intrinsic resolution δ_{sc} . Fig. 4 presents the non-proportionality characteristics of **5 x 5 x 1 mm³** GAGG:Ce and LuAG:Ce crystals. Over the energy range from 662 keV down to 32 keV, the LY non-proportionality for GAGG:Ce is ~20% which is worse than that of ~14% for LuAG:Ce, reflecting somewhat higher intrinsic resolution (see Table 3) as the non-proportionality of LY is a fundamental limitation to intrinsic resolution [27]. We note that the LY non-proportionality (20 % at 32 keV) obtained for a studied 5 x 5 x 1 mm³ GAGG:Ce is comparable to those measured for 5 x 5 x 5 mm³ and 10 x 10 x 5 mm³ GAGG:Ce samples [18], and there is no sample size dependence.

Fig. 3. Energy resolution and contributed factors versus energy of γ -rays for a $5 \times 5 \times 1 \text{ mm}^3$ GAGG:Ce crystal.

Table 3 Analysis of the 662 keV energy resolution for GAGG:Ce and LuAG:Ce crystals with same size of $5 \times 5 \times 1 \text{ mm}^3$. N is the number of photoelectrons measured at 662 keV.

Crystal	N	Energy resolution (%)		
		$\Delta E/E$	δ_{st}	δ_{sc}
GGAG:Ce	5,860	6.8	3.1	6.0
LuAG:Ce	2,850	7.3	4.4	5.8

Fig. 4. Non-proportionality characteristics of $5 \times 5 \times 1 \text{ mm}^3$ GAGG:Ce and LuAG:Ce crystals.

In order to investigate the contribution of slow components in scintillation response for GAGG:Ce, the LY dependence on amplifier shaping time was measured under 662 keV γ -ray excitation and compared with LuAG:Ce. The result in Fig. 5 shows a lower contribution of slow component in the scintillation response of GAGG:Ce with respect to LuAG:Ce. In the former material it confirms the diminished trapping effect due to intrinsic shallow electron traps and at the same time yet sufficient separation between the conduction band edge and $5d_1$ excited state of Ce^{3+} achieved due to the balanced admixture of Gd and Ga into aluminum garnet matrix [20, 21, 28].

Shaping time dependences of LY in Fig. 5 were approximated by the model of Prusa et al. in Ref. [29]. Based on a two-exponential approximation of scintillation decay the normalized LY(t) time dependence is expressed as

$$LY(t) = m_1 - m_2 \exp[-m_3 t] \quad (4)$$

where $m_{1,2,3}$ are parameters. Their physical meaning can be described as follows: m_1 is relative increase of LY for an infinite shaping time, relative fraction of the fast response part $K_{f/e}$ ($t = 0.5 \mu s$) in the entire LY value ($t = \infty$) is obtained as $K_{f/e} = 1 - m_2/m_1$, and m_3 is proportional to the inverse decay time of the slow component responsible for the LY(t) increase for increasing shaping time values. The values of $m_1 = 123\%$, $K_{f/e} = 80\%$ and $1/m_3 = 4.2 \mu s$ for GAGG:Ce and $m_1 = 182\%$, $K_{f/e} = 50\%$ and $1/m_3 = 3.7 \mu s$ for LuAG:Ce were obtained from Eq. (4) fitted to the data in Fig. 5, solid and dashed lines, respectively, for further evaluation details see Ref. [29]. This measurement supports higher content of fast component with contributed intensity of 85% and 47%, respectively, in the scintillation decay of GAGG:Ce [18] and LuAG:Ce [8, 9]. $K_{f/e}$ value of 80% obtained for a studied GAGG:Ce is lower than those measured earlier for GAGG:1%Ce [17].

Fig. 5. Light yield dependence on amplifier shaping time normalized at $0.5 \mu s$ for GAGG:Ce and LuAG:Ce crystals. Solid lines come from the model described in the text.

3.2. Photofraction

The photofraction is defined as the ratio of counts under the full-energy peak to the total counts of the spectrum as measured at a specific γ -ray energy. Fig. 6 presents the pulse height spectra of γ -rays from ^{137}Cs (662 keV) and ^{51}Cr (320 keV) sources as measured with a $5 \times 5 \times 5 \text{ mm}^3$ GAGG:Ce sample while the photofraction for all GAGG:Ce samples is presented in Table 4. The ratio of the cross-section for the photoelectric absorption to the total one calculated using the WinXCom program [30] is also given for a comparison. Note a higher photofraction as measured for the larger samples as expected due to a higher contribution from (multiple) Compton scattering and terminated by photoelectric absorption, leading to a full-energy peak in the pulse height spectrum, for the larger crystals. The higher photofraction for 320 keV γ -rays was also observed in a same trend with the cross-section ratio (σ -ratio) obtained from the WinXCom program. It is due to the increase of photoelectric absorption cross section with decreasing energy of γ -rays.

Fig. 6. Pulse height spectra of γ -rays from ^{137}Cs (662 keV) and ^{51}Cr (320 keV) sources as measured with a $5 \times 5 \times 5 \text{ mm}^3$ GAGG:Ce crystal.

Table 4 Photofraction at 320 and 662 keV γ -rays for studied GAGG:Ce crystals.

Energy (keV)	Sample size (mm ³)	Photofraction (%)	σ -ratio (%)
320	5x5x1	33.4	40.8
	5x5x2	37.2	
	5x5x5	43.4	
662	5x5x1	10.74	12.97
	5x5x2	14.24	
	5x5x5	16.35	

3.3. Total mass attenuation coefficient

A parallel beam of monoenergetic γ -rays is attenuated in an absorber according to the Lambert–Beer law:

$$I(x) = I_0 \exp (-\mu_m \rho x) \quad (5)$$

where I_0 and I are incident and transmitted intensities of γ -rays, respectively, μ_m is the mass attenuation coefficient, ρ is the density of the absorber, and x is the thickness of the absorber. The product $\mu_m \rho$ is called the linear attenuation coefficient. Theoretical values of the mass attenuation coefficient of mixture have been calculated using the WinXCom program [30]. Table 5 shows the experimental value $(\mu_m)_{ex}$ of the mass attenuation coefficient at 662 keV γ -rays for GAGG:Ce together with the theoretical ones $(\mu_m)_{th}$ calculated at 320 and 662 keV. The higher value $(\mu_m)_{th}$ for 320 keV was clearly seen in a same trend with the photofraction value (Table 4). It is mainly due to the increase of photoelectric absorption cross section with decreasing energy of γ -rays.

Table 5 Mass attenuation coefficient at 320 and 662 keV γ -rays for GAGG:Ce crystal.

γ - energy (keV)	$(\mu_m)_{ex}$ (cm ² /g)	$(\mu_m)_{th}$ (cm ² /g)
320	n.m.*	0.151
662	0.078	0.077

*not measured

4. Conclusion

The scintillation properties of the latest generation of $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}$ crystal grown by the Czochralski method were investigated. It shows a high LY value over 47,000 ph/MeV and a good energy resolution of 6.8 % at 662 keV γ -rays, comparable to that of NaI(Tl). Dependence of LY value on amplifier shaping time shows distinctly lower content of slower component (in the range of microseconds) in scintillation response compared to LuAG:Ce, which confirms the diminished trapping effect of shallow electron traps due to the balanced admixture of Gd and Ga into aluminum garnet matrix. These properties together with a high density of 6.69 g/cm^3 , non-hygroscopic and intrinsic radioactivity-free host assure the capability of GAGG:Ce crystal for high energy γ -ray measurements and medical imaging applications. Further improvement of scintillation performance can be obtained by fine composition tuning in the range of $\text{Gd}_3\text{Al}_{2-x}\text{Ga}_{3+x}\text{O}_{12}$.

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