Growth and characterization of $(Lu,Y,Gd)_3(Al,Ga)_5O_{12}$:Ce;Mg crystals for timing application

---

Assoc. Prof. Weerapong Chewpraditkul, Dr. Ongsa Sakthong, Mr. Warut Chewpraditkul

King Mongkut's University of Technology Thonburi, Thailand

This report summarizes our research work during short research visit at Yoshikawa's Laboratory, Institute of Materials Research (IMR), Tohoku University, from June 13 to July 4, 2018.

The purpose of this visit was to grow $(Lu,Y,Gd)_3(Al,Ga)_5O_{12}$:Ce;Mg multicomponent garnet crystals and characterize their scintillation properties. These crystals were grown by micro-pulling-down method.

1. Crystal growth and characterization

The garnet crystals of $Y_{0.8}Gd_{2.2}(Al_{5-x}Ga_x)O_{12}$:Ce;Mg (YGdAGG:Ce;Mg) & $Lu_{0.8}Gd_{2.2}(Al_{5-x}Ga_x)O_{12}$:Ce;Mg (LuGdAGG:Ce;Mg) with x = 2.6 and 3 were grown by the micro-pulling-down method using an RF heating system (Fig.1). The dopant concentration in the melts was 1 at.% Ce and 0.15 at.% Mg. An Ir crucible was used in the atmosphere of Ar + 2%O_2 to prevent evaporation of gallium oxide. The seed was GAGG:Ce crystal rod attached to the alumina seed holder. The pulling rate was 0.05 mm/min and the crystal diameter was around 4 mm. The photograph of as grown LuGdAGG:Ce;Mg crystal is shown in Fig. 2. The polished plates with thickness of 1 mm cut from the parent rods were used for all the measurements, i.e. X-ray induced radioluminescence (RL) spectra, scintillation decay time, light yield (LY) and thermoluminescence characteristics.

The RL spectra measurements were performed using CCD -coupled monochromator under excitation with X-ray tube (20 kV, 0.15 mA). Fig.3 shows the RL spectra of the µ-PD grown LuGdAGG:Ce;Mg crystal samples. The RL spectrum of Ga = 3 sample was slightly blue-shifted with respect to the sample with Ga = 2.6 was observed. It can be attributed to a decrease in a crystal field strength around Ce^{3+} ion at the dodecahedral site from partial substitution of Gd^{3+} by the smaller Lu^{3+} ions, which causes a high-energy shift of the $5d_1$ level of Ce^{3+} center.

Fig. 1. Photograph of the µ-PD growth machine.

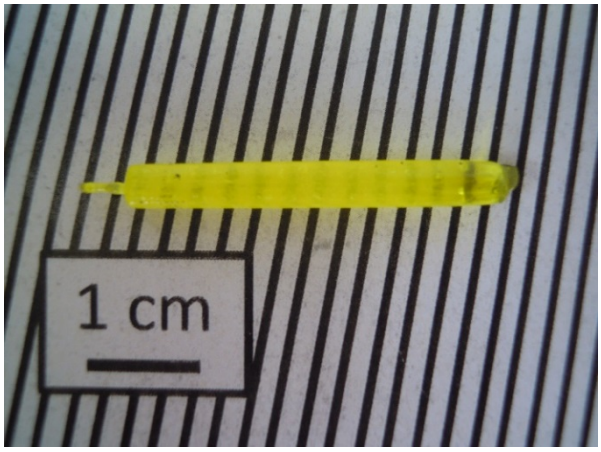

Fig. 2. Photograph of as-grown LuGdAGG:Ce;Mg crystal grown by the μ-PD method.

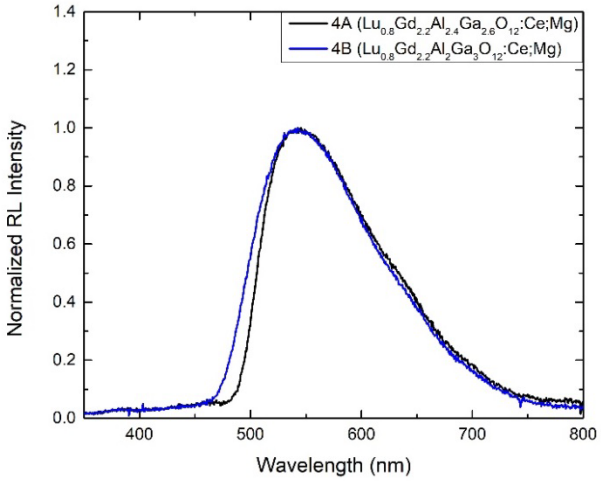

Fig. 3. RL spectra of LuGdAGG:Ce;Mg samples.

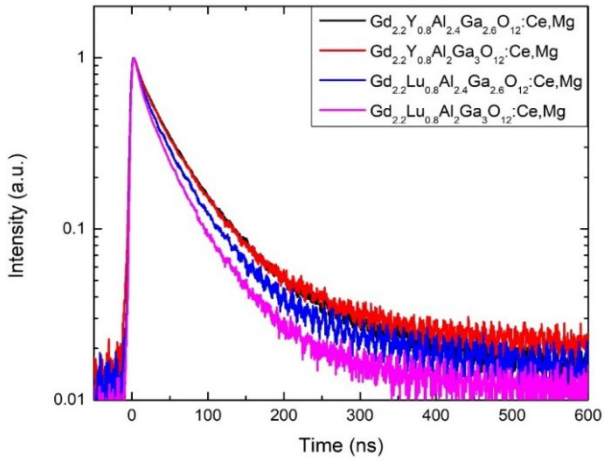

Fig. 4. Scintillation decays of LuGdAGG:Ce;Mg and YGdAGG:Ce;Mg crystals.

The scintillation decay time measurements were performed using a Hamamatsu R7600U-200 PMT and a Tektronix TDS3052 digital storage oscilloscope under excitation with γ rays from a ^{137}Cs source. Fig.4 presents the scintillation decay curves of LuGdAGG:Ce;Mg and YGdAGG:Ce;Mg crystals. The decay components and relative intensities were estimated by fitting the scintillation decay curves with a double - exponential function: $I(t) = \Sigma A_i \exp(-t/\tau_i) +$ background, and the results are collected in Table 1. The LuGdAGG:Ce;Mg crystals show a faster decay time with respect to YGdAGG:Ce;Mg ones. The scintillation decay time was accelerated with increasing Ga content. It can be due to a blue-shifted emission spectra caused by a high-energy shift of the $5d_1$ level upon partial substitution of Al^{3+} by a larger Ga^{3+} ions, which reduces the crystal field strength around Ce^{3+} ions at the dodecahedral site. Scintillation decay time for $Gd_3Al_2Ga_3O_{12}$:Ce;Mg is also included in Table 1 for a comparison.

Table 1 Scintillation decay time and relative intensity of YGdAGG:Ce;Mg and LuGdAGG:Ce;Mg crystals.

Crystals	τ_1(I%) ns	τ_2(I%) ns
$Y_{0.8}Gd_{2.2}Al_{2.4}Ga_{2.6}O_{12}$:Ce;Mg	25 (28%)	81 (72%)
$Y_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg	20 (22%)	69 (78%)
$Lu_{0.8}Gd_{2.2}Al_{2.4}Ga_{2.6}O_{12}$:Ce;Mg	16 (18%)	61 (82%)
$Lu_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg	12 (18%)	52 (82%)
$Gd_3Al_2Ga_3O_{12}$:Ce;Mg [1]	45 (41%)	108 (59%)

Light yield (LY) measurements were performed using a Hamamatsu R6231 PMT under excitation with a ^{137}Cs γ rays. To improve the light collection efficiency each sample was coupled to the PMT window with silicone grease and covered with several layers of Teflon tape. The signal from the PMT anode was processed by a CANBERRA 2005 preamplifier and a Tennelec TC243 spectroscopy amplifier set at 4 μs shaping time constant. Tukan 8 k MCA was used to record the pulse height spectra. The photoelectron yield (phe/MeV) was determined by relating the full-energy peak position with that of the single photoelectron peak from the PMT photocathode. Fig. 5 shows the pulse height spectra of γ rays from a ^{137}Cs source measured with the studied crystals. The LY(ph/MeV) values, calculated from the photoelectron yield using the quantum efficiency of PMT weighted over the RL spectrum, and energy resolution (ΔE/E) are collected in Table 2. An increase of LY value with increasing Ga content (from 2.6 to 3) was obtained for both crystal hosts. It can be attributed to a decrease of bandgap energy (E_g), caused by more down-shift of the bottom of the conduction band with more substitution of Al^{3+} by a larger Ga^{3+} ions.

$Y_{0.8}Gd_{2.2}Al_{2.4}Ga_{2.6}O_{12}$:Ce;Mg shows highest LY value and best ΔE/E while $Lu_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg exhibits lowest LY value but fastest scintillation decay time. The LY and ΔE/E for $Gd_3Al_2Ga_3O_{12}$:Ce;Mg [1] and $Gd_3Al_2Ga_3O_{12}$:Ce [2] crystals measured under the

same experimental conditions are also included in Table 2 for a comparison.

Time resolution spectra of $Y_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg (YGAGG:Ce;Mg) and $Lu_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg (LGAGG:Ce;Mg) detectors measured in coincidence with a BaF_2 detector are shown in Fig. 6. An analysis of

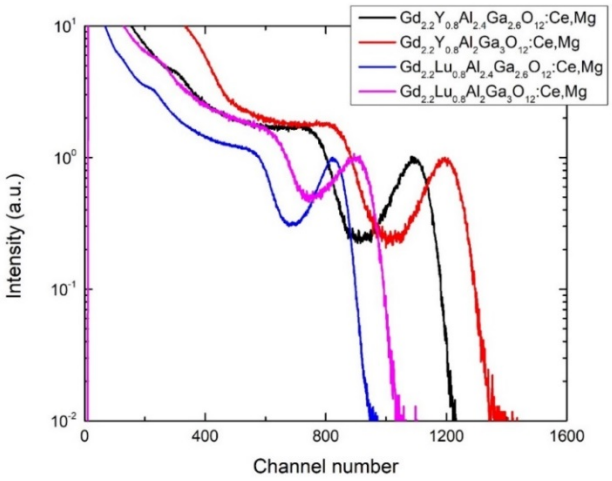

Fig.5. Pulse height spectra of ^{137}Cs γ rays measured for LuGdAGG:Ce;Mg and YGdAGG:Ce;Mg crystals.

Table 2 LY and ΔE/E of YGdAGG:Ce;Mg and LuGdAGG:Ce;Mg crystals.

Sample	LY (ph/MeV)	ΔE/E (%)
$Y_{0.8}Gd_{2.2}Al_{2.4}Ga_{2.6}O_{12}$:Ce;Mg	33,700	7.7
$Y_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg	36,800	7.9
$Lu_{0.8}Gd_{2.2}Al_{2.4}Ga_{2.6}O_{12}$:Ce;Mg	25,400	8.3
$Lu_{0.8}Gd_{2.2}Al_2Ga_3O_{12}$:Ce;Mg	27,600	11
$Gd_3Al_2Ga_3O_{12}$:Ce;Mg [1]	36,400	7.1
$Gd_3Al_2Ga_3O_{12}$:Ce [2]	51,900	5.6

the measured time resolution is presented in Table 3. The time resolution (δ_t) in a second column is obtained after correction for a contribution of 128 ps time resolution of a BaF_2 detector. The third column shows the mean decay time (τ_m) defined by equation: $\tau_m = \Sigma(A_i\tau_i^2) / \Sigma(A_i\tau_i)$. The number of photoelectrons (N_f) measured at 0.5 μs time gate, is listed in the fourth column. The last column shows initial photoelectron rate (N/τ_m), a figure of merit in timing applications. Despite about 24% lower LY value, time resolution of LGAGG:Ce;Mg is slightly worse than that of YGAGG:Ce;Mg. This is mainly due to its faster scintillation decay time, which leads to a comparable N/τ_m value.

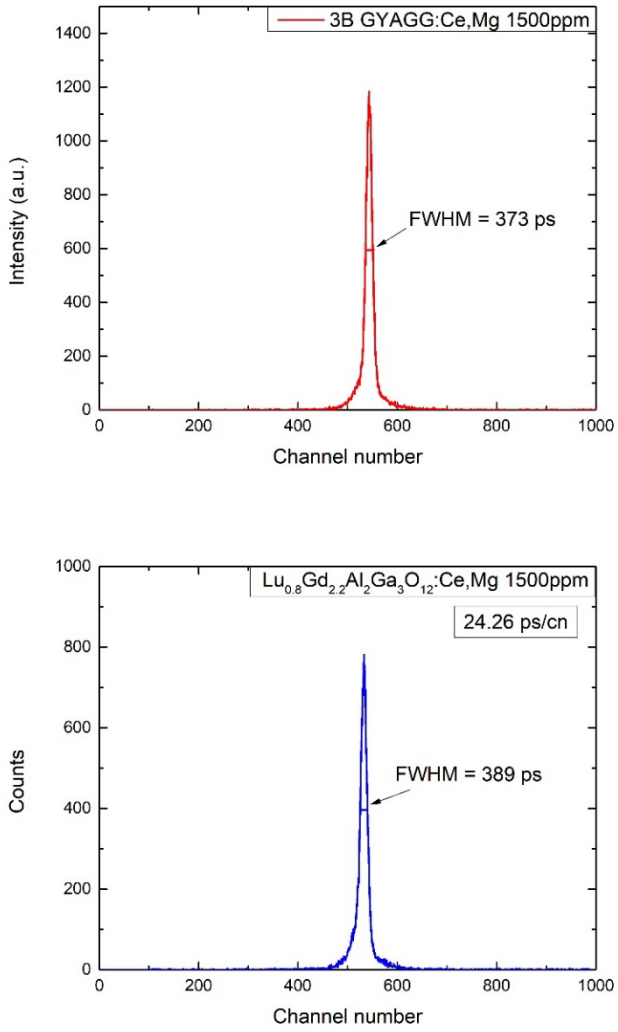

Fig. 6. Coincidence time resolution spectra of YGAGG:Ce;Mg-BaF_2 and LGAGG:Ce;Mg-BaF_2 detectors.

Table 3 Analysis of coincidence time resolution for $YGAl_2Ga_3G$:Ce;Mg and $LGAl_2Ga_3G$:Ce;Mg crystals coupled to an XP20D0 PMT.

Crystal	δ_t (ps)	τ_m (ns)	N (phe)	N/τ_m (phe/ns)
YGAGG:Ce;Mg	373	65	2120	33
LGAGG:Ce;Mg	389	50	1620	32

2. Co-Authored Publications

[1] W.R. Chewpraditkul, N. Pattanaboonmee, O. Sakthong, K. Wantong, W. Chewpraditkul, A. Yoshikawa, K. Kamada, S. Kurosawa, T. Szczesniak, M. Moszynski, V. Babin, M. Nikl, Scintillation

properties of GAGG:Ce;Li and GAGG:Ce;Mg single crystal scintillators: A comparative study, Optical Materials 92 (2019) 181–186.
[2] W.R. Chewpraditkul, N. Pattanaboonmee, O. Sakthong, W. Chewpraditkul, K. Kamada, A. Yoshikawa, M. Nikl, Scintillation properties of $Gd_3(Al_{5-x}Ga_x)O_{12}$:Ce; x = 2.3, 2.6, 3.0 single crystals, Optical Materials 81 (2018) 23–29.
[3] W.R. Chewpraditkul, N. Pattanaboonmee, W. Chewpraditkul, O. Sakthong, T. Szczesniak, M. Moszynski, K. Kamada, A. Yoshikawa, M. Nikl, Luminescence and scintillation characteristics of $(Gd_xY_{3-x})Al_2Ga_3O_{12}$:Ce; x = 1, 2, 3 single crystals, Optical Materials 76 (2018) 162-168.

3. Acknowledgments

I would like to thank all the members of Yoshikawa Laboratory and other Departments of IMR for their kind assistance and support.

June 2019

Keywords: Micro-pulling-down method; Scintillator;
Multicomponent garnet; Scintillation decays, Coincidence time resolution
E-mail: weerapong.che@kmutt.ac.th; ongsa.sak@kmutt.ac.th; raychew.rc@gmail.com