Luminescence and scintillation properties of $(Lu_xGd_{3-x})(Al_{2.4}Ga_{2.6})O_{12}$:Ce multicomponent garnet crystals

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This report summarizes my research work during five-week visit as a Visiting Associate Professor at Yoshikawa's Laboratory, Institute of Materials Research (IMR), Tohoku University, from June 5 to July 8, 2017.

The purpose of the visit was to investigate the scintillation properties of new multicomponent garnet crystals (LuxGd3-x)(Al2.4Ga2.6)O12; doped with Ce and codoped with Ce + Mg. These crystals were grown by micro-pulling down method.

1. Crystal growth and characterization

LuGdAGG:Ce and LuGdAGG:Ce,Mg crystals were grown by the micro-pulling-down method using an RF heating system (Fig.1). Starting materials with the composition $(Lu_xGd_{3-0.015-x})(Al_{2.4}Ga_{2.6})O_{12}:Ce_{0.015}$ and $(Lu_xGd_{3-0.0165-x})(Al_{2.4}Ga_{2.6})O_{12}:Ce_{0.015}Mg_{0.0015}$, x = 0.2, 0.4, 0.6, 0.8 were prepared by mixing of 99.99% pure oxide materials at a stoichiometric composition. An Ir crucible was used in the atmosphere of Ar + 2%O_2 to prevent evaporation of gallium oxide. The seed was undoped GAGG crystal rod attached to the alumina seed holder (Fig. 2). The pulling rate was 0.05 mm/min and the crystal diameter was around 4 mm. The photograph of as grown $(Lu_{0.6}Gd)$AGG:Ce0.5% crystals is shown in Fig. 3. The polished plates of about 3.8 x 3.8 x 2 mm cut from the parent rods were used for all the measurements, i.e. X-ray induced RL spectra, scintillation decay time and light yield value.

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

The RL spectra measurements were performed using CCD- coupled monochromator under excitation with X-ray tube (20 kV, 0.15 mA). Fig.4 shows the RL spectra of the μ-PD grown $(Lu_{0.8}Gd)$AGG:Ce and $(Lu_{0.8}Gd)$AGG:Ce,Mg crystal samples compared to the CZ-grown GAGG:Ce crystal. The blue-shifted of RL spectra for the studied crystals with respect to the GAGG:Ce crystal was clearly observed. It can be attributed to the decrease in the crystal field strength around Ce^{3+} ion at the dodecahedral site from a partial substitution of Gd^{3+} by the smaller Lu^{3+} ions, which causes a high-energy shift of the 5d1 level of Ce^{3+} center.

Fig.2. An Ir crucible attached to the after-heater and GAGG seed mounted to the alumina rod.

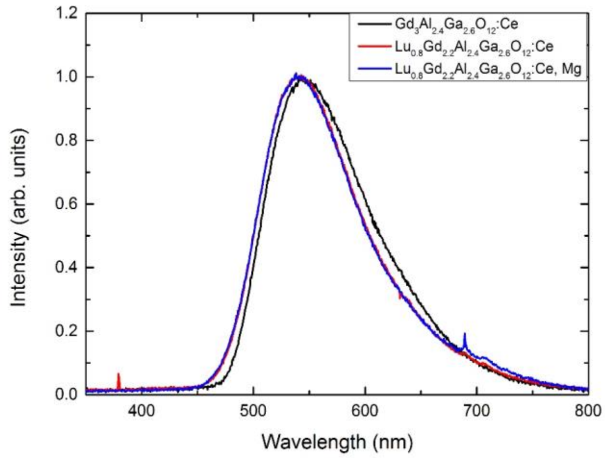

Fig.4. RL spectra of μ-PD grown ($Lu_{0.8}$Gd)AGG:Ce and ($Lu_{0.8}$Gd)AGG:Ce,Mg samples compared to the CZ-grown GAGG:Ce crystal.

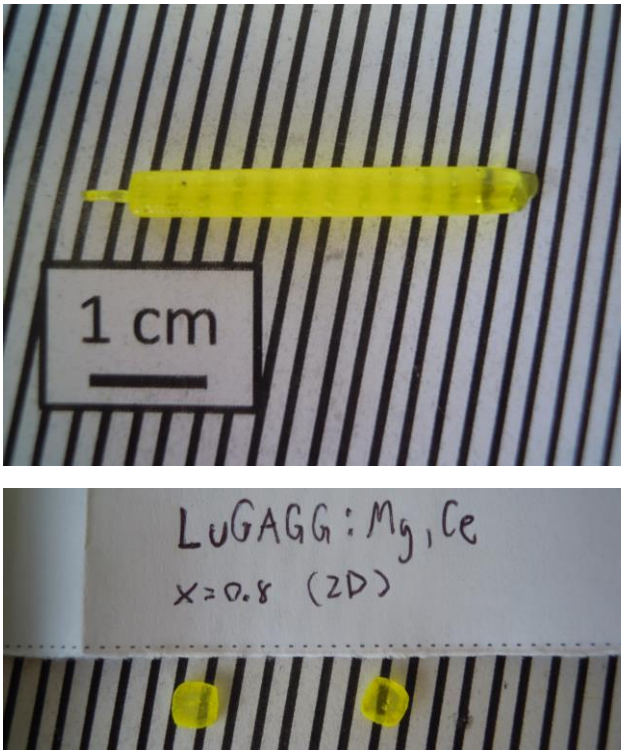

Fig.3. Photograph of ($Lu_{0.6}$Gd)AGG:Ce crystals grown by the μ-PD method.

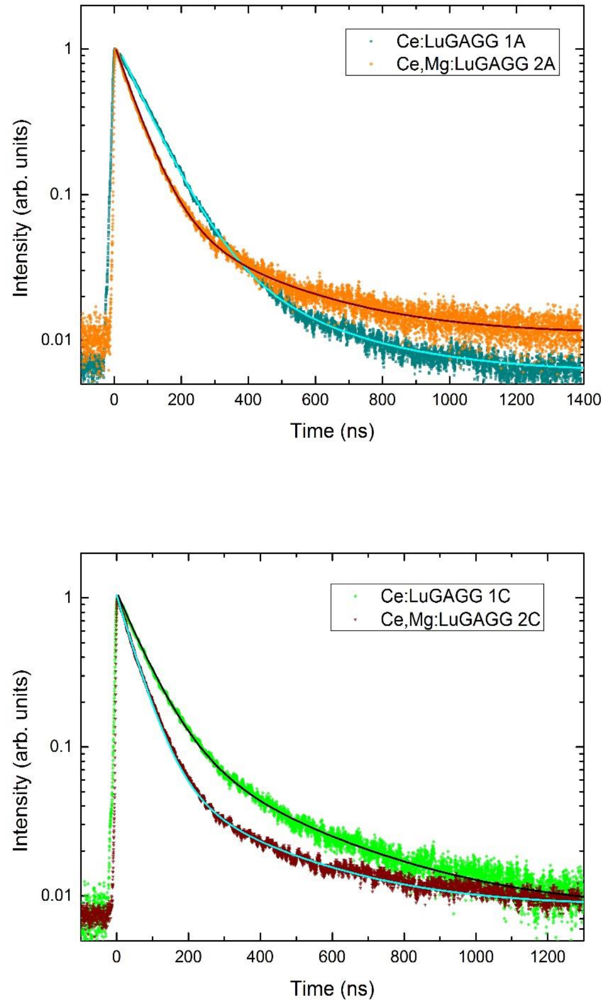

Fig.5. Scintillation decays of some μ-PD grown (Lu_xGd)AGG:Ce and (Lu_xGd)AGG:Ce,Mg crystals.

The scintillation decay time measurements were performed using a Hamamatsu R7600U-200 PMT and Tektronix TDS3052 digital storage oscilloscope under excitation with γ - rays from a ^{137}Cs source. Fig.5 presents examples of the scintillation decay spectra of (Lu_xGd)AGG:Ce crystals in comparison with Mg^{2+}-codoped (Lu_xGd)AGG:Ce,Mg crystals.

The decay time components and relative intensities were estimated using a double-exponential fit to the scintillation decay spectra, and the results are collected in Table 1. The Mg co-doped crystals show a faster decay time with respect to LuGdAGG:Ce crystals.

Table.1. Scintillation decay time and relative intensity of the $(Lu_xGd)AGG$:Ce and $(Lu_xGd)AGG$:Ce,Mg crystals.

Crystals	τ_1(I%) ns	τ_2(I%) ns
(1A) Lu0.2GAGG:Ce	85(83%)	290(17%)
(1B) Lu0.4GAGG:Ce	72(76%)	340(23%)
(1C) Lu0.6GAGG:Ce	72(65%)	318(35%)
(1D) Lu0.8GAGG:Ce	68(73%)	290(27%)
(2A) Lu0.2GAGG:Ce, Mg	62(72%)	296(28%)
(2B) Lu0.4GAGG:Ce, Mg	56(71%)	280(29%)
(2C) Lu0.6GAGG:Ce, Mg	52(74%)	280(26%)
(2D) Lu0.8GAGG:Ce, Mg	52(73%)	266(27%)

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. 6 shows the pulse height spectra of ^{137}Cs γ - rays measured with the μ-PD grown $(Lu_{0.2}Gd)AGG$:Ce and $(Lu_{0.2}Gd)AGG$:Ce,Mg crystals compared to the old $Gd_3Al_2Ga_3O_{12}$:Ce (GAGG:Ce) CZ-grown crystal. The LY (ph/MeV) was calculated using the PMT average quantum efficiency of 13% for the emission spectrum of both crystals. The best crystal sample (Lu0.2GAGG:Ce, Mg) shows very good pulse height spectrum of 662 keV γ -rays from a ^{137}Cs source due to its good crystal quality. It exhibits LY value of 43,300 ph/MeV and energy resolution of 9.3% in comparison - to the values of 56,100 ph/MeV and 6.9% for the old CZ- grown GAGG:Ce crystal.

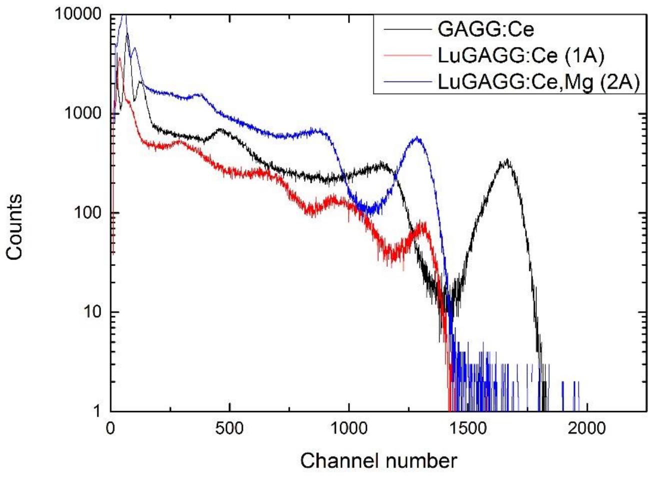

Fig.6. Pulse height spectra of ^{137}Cs gamma rays measured with the studied $(Lu_{0.2}Gd)AGG$:Ce and $(Lu_{0.2}Gd)AGG$:Ce,Mg crystals compared to a CZ-grown $Gd_3Al_2Ga_3O_{12}$:Ce crystal.

2. Topics of Other Activities (Assistance to IMR Staff, Students, etc.)

Characterizing the scintillation properties of Mg^{2+}- and Li^{1+}- codoped GAGG:Ce crystals

$Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Mg and $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce, Li crystals (with dimension 5x5x5 mm^3) were grown by the Czochralski method and the scintillation characteristics were investigated and compared to the old CZ-grown GAGG:Ce crystal. Fig. 7 shows pulse height spectra of 662 keV γ -rays from a ^{137}Cs source measured for the studied crystals with same size of 5x5x5 mm^3. LY and energy resolution are collected in Table 2. The LY value of 46,326 ph/MeV measured for the $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Li samples is larger than that of 44,768 ph/MeV for the old GAGG:Ce sample, whereas the $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Mg samples show a lower LY value of 22,069 MeV and 27,027 MeV. Superior energy resolution of $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Li crystals was also confirmed in the better LY -proportionality (see Fig. 8) in addition to a larger LY value.

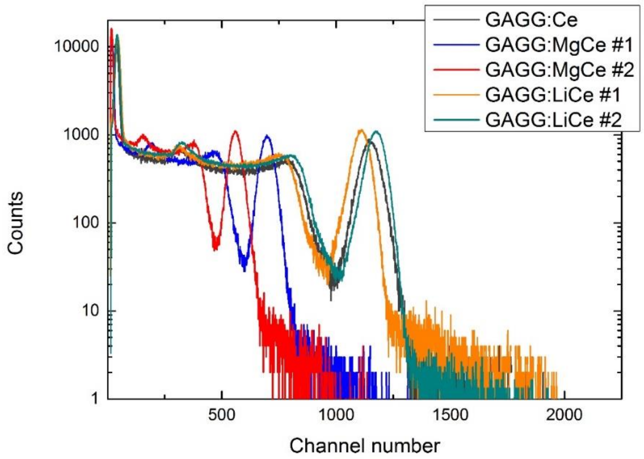

Fig.7. Pulse height spectra of 662 keV γ - rays from a [137]Cs source measured with $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Mg $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Li and GAGG:Ce crystals.

Table. 2 Light yield and energy resolution of studied crystals.

Sample	LY (ph/MeV)	ΔE/E(%)
GAGG:MgCe #1	27,070	9.4
GAGG:MgCe #2	22,070	10.8
GAGG:LiCe #1	43,810	6.9
GAGG:LiCe #2	46,330	7.0
GAGG:Ce (old)	44,770	8.6

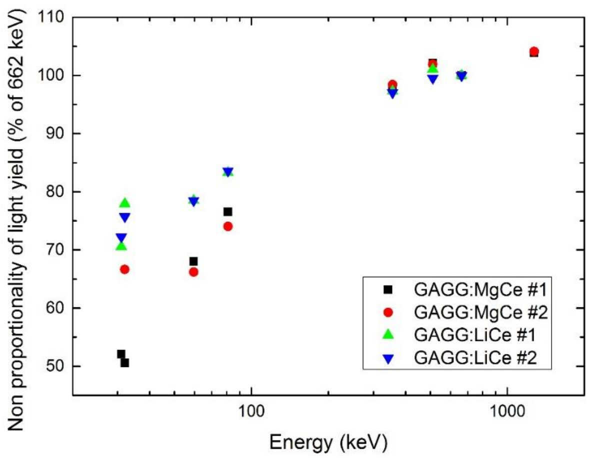

Fig. 8 LY non-proportionality of the studied crystals.

In order to investigate the scintillation timing characteristics, their scintillation decay spectra were measured as shown in Fig. 9. The decay time components (τ_i) were determined by performing a double exponential fit: $I(t) = \Sigma A_i \exp(-t/\tau_i) + B$. The results are collected in Table 5.

The $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Mg crystals show faster scintillation decay time with respect to $Gd_3(Al_{2.4}Ga_{2.6})O_{12}$:Ce,Li and $Gd_3Al_2Ga_3O_{12}$:Ce. The coincidence time resolution for these crystals will be measured and discussed.

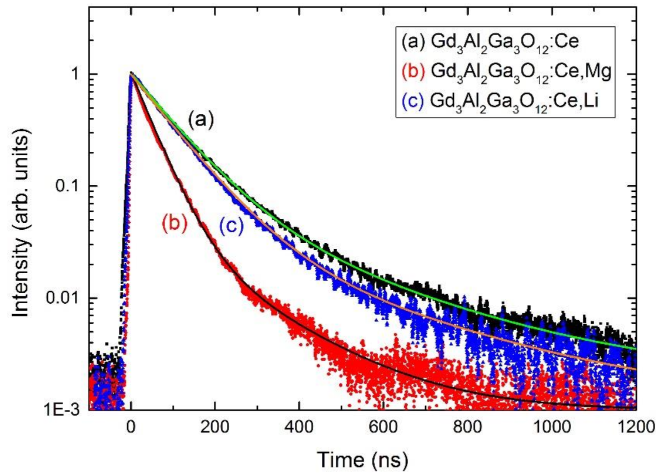

Fig. 9. Scintillation decay spectra of studied crystals

Table. 3. Scintillation decay of the studied crystals

Sample	τ_1(ns) (I%)	τ_2(ns) (I%)	τ_{ave} (ns)
GAGG:Ce	92 (76%)	300 (24%)	141
GAGG:Mg,Ce	46 (84%)	174 (16%)	66
GAGG:Li,Ce	90 (86%)	335 (14%)	124

3. Co-Authored Manuscript Preparation for Publication.

1. 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.
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.
3. O. Sakthong, W.R. Chewpraditkul, W. Chewpraditkul, T. Szczesniak, L. Swiderski, M. Moszynski, K. Kamada, A. Yoshikawa, M. Nikl, Comparative study of $GdLu_2Al_2Ga_3O_{12}$:Ce and $GdY_2Al_2Ga_3O_{12}$:Ce scintillation crystals for γ - ray detection.

4. Acknowledgments

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

July 2017

Keywords: Micro-pulling-down method; Scintillator; Multicomponent garnet; Scintillation decays
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