Nonstochiometry of $Lu_3Al_5O_{12}$ single crystal and its effects of on luminescence and scintillation properties

[1]**Kei Kamada**[1,2], **Shunsuke Kurosawa**[3] , **Yuui Yokota**[1], **Jan Pejchal**[1,] **Martin Nikl**[4], **Yuji Ohashi**[3] **and Akira Yoshikawa**[1,2,3]
[1]New Industry Creation Hatchery Center, Tohoku University, 6-6-10 Aoba, Sendai 980-8579
[2]C&A corporation, 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
[3]Institute for Material Reseach, Tohoku University, 2-1-1, Sendai,980-8577 Japan
[4]Institute of Physics AS CR,16253 Prague, Czech Republic
E-mail: kamada@imr.tohoku.ac.jp

Abstract. Nonstoichiometric $Lu_3Al_{5+x}O_{12}$ (x= 0.05, 0.15, 0.35, 0, -0.05, -0.15, -0.35) crystals were grown by the μ-PD method. Luminescence and scintillation properties such as absorption, excitation and emission spectra, light yield and decay time were evaluated. Expected anti-site defect related host emission have been observed in 250-420nm. Emission intensity was increased by increasing the nonstochiometry. The x=-0.35 sample showed the highest light yield of around 12000 photons/MeV and slowest scintillation decay time of 1.96μs.

1. Introduction

Scintillator materials combined with photodetectors are used to detect high energy photons and accelerated particles in medical imaging techniques, high energy and nuclear physics detectors, high-tech industrial applications and most recently also in the advanced homeland security related techniques. Oxide materials based on garnet structure single crystals are promising candidates for scintillator applications because of well mastered technology developed for laser hosts and other applications, optical transparency and easy doping by rare-earth elements. For example, Pr:$Lu_3Al_5O_{12}$ (Pr:LuAG) has attracted attention because of its favorable combination of fast Pr^{3+} 5d-4f emission peaking at 311 nm, non-hygroscopic nature, high density (6.7 g/cm^3), high light output (around 20000 photon/MeV), very short decay time (20 ns), and good energy resolution (4.8% at 662 keV for a 10 × 10 × 10 mm LuAG:Pr sample and a Hamamatsu R1791 PMT) [1-3] Isostructural cubic garnet structures of YAG and LuAG form a solid solution and any intermediate mixed composition (i.e. $Y_xLu_{3-x}Al_5O_{12}$, for 0>x>3) can be grown. The solidification points of mixed compositions range between 2010 ^{0}C (for LuAG) and 1930 ^{0}C (for YAG) [4]. The garnet structure exhibits very flexible cation substitution and such substitution was used to prepare materials with tailored lattice constants [5] and can, in principle, be used for the preparation of highly substituted crystals with a homogeneous composition. However, at the same time, this readiness of solid solution formation in the garnet structure hints at the relative ease of intrinsic anti-site defect formation. Indeed, theoretical studies have shown that the anti-site defect, Y_{Al}, was found as the lowest energy intrinsic defect in both YAG [6-8] and all other REAGs, (RE ranging from Lu to Gd) [9]. Anti-site defect related host emission is

[1] Kei Kamada, New Industry Creation Hatchery Center, Tohoku University, 6-6-10 Aoba, Sendai 980-8579

observed around 280-360nm with decay time of few hundreds nano second. Anti-site defect can be enhanced by Sc doping in Al-site and Sc doped LuAG shows high light yield of 22500 photon/MeV with decay time of 610ns[10]
In this report, nonstochiometry of LuAG single crystal and its effects of on luminescence and scintillation properties were investigated. Nonstoichiometric $Lu_3Al_{5-x}O_{12}$ single crystals were grown by the micro-pulling down (μ-PD) method. Luminescence and scintillation properties were evaluated.

2. Experimental

2.1. Crystal growth procedure

A stoichiometric mixture of 4N Lu_2O_3 and α-Al_2O_3 powders (High Purity Chemicals Co.) was used as starting material. Nonstoichiometric $Lu_3Al_{5+x}O_{12}$ were grown by the micro-pulling-down (μ-PD) method with an RF heating system. A schematic of the μ-PD growth apparatus is given in Refs. [11,12]. Typical pulling rates were 0.05–0.12 mm/min and the diameter was around 3 mm. Crystals were grown from an Ir crucible under N_2 atmosphere. The seed crystals were <100> oriented LuAG crystals. Plates of 3mmϕ x 1mm were cut and polished for the absorption and luminescence spectra measurement.

2.2. Measurements of scintillation properties

Radio-luminescence spectrum at room temperature was measured with the above spectrometer (EI FLS920) excited by 5.5MeV alpha rays from an 241Am source. To determine the light output, we obtained the pulse height spectrum of this crystal under excitation with 662 keV gamma rays from a ^{137}Cs source. The scintillation photons were detected using by a photomultiplier (PMT; Hamamatsu R7600U), and then the signals from the PMT were amplified (ORTEC 113), shaped (ORTEC 572A), and read out with a multi-channel analyzer (AMPTEK 8000A). The pulse height of $Bi_4Ge_3O_{12}$ (BGO) crystal, which has a light output of 8,500 photons/MeV [13] was also measured as a reference. Additionally, its scintillation decay time excited by the gamma rays (^{137}Cs) was measured with the PMT and an oscilloscope (Tektronix TDS 3052B). The scintillation decay time was calculated from exponential approximation.

3. Results

3.1. Crystal growth procedure

Nonstoichiometric $Lu_3Al_{5+x}O_{12}$ (x= 0.05, 0.15, 0.35, 0, -0.05, -0.15, -0.35) crystals were grown by the μ-PD method. Example photos are shown in Figure 1. The grown crystals were colorless transparent with 2-3 mm in diameter and 15-23 mm in length.

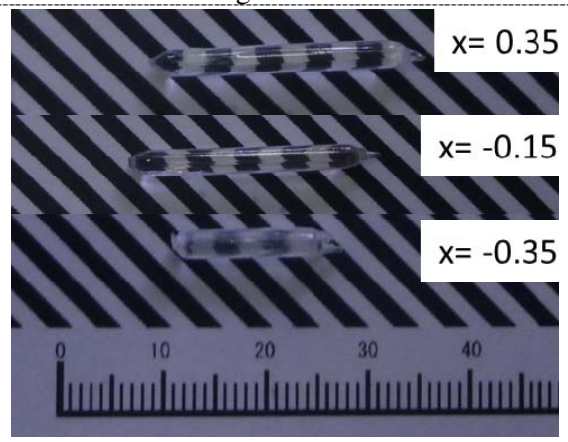

Figure 1. Example photographs of the grown $Lu_3Al_{5+x}O_{12}$ single crystals.

3.2. Luminescence and gamma-ray response measurement procedurembering

Figure 2 shows radioluminescence spectra of the $Lu_3Al_{5+x}O_{12}$ samples. Expected anti-site defect related host emission have been observed in 250-420nm. Emission intensity was increased by increasing the nonstochiometry. The x=+0.35 sample showed the highest emission intensity among the grown $Lu_3Al_{5+x}O_{12}$ samples.

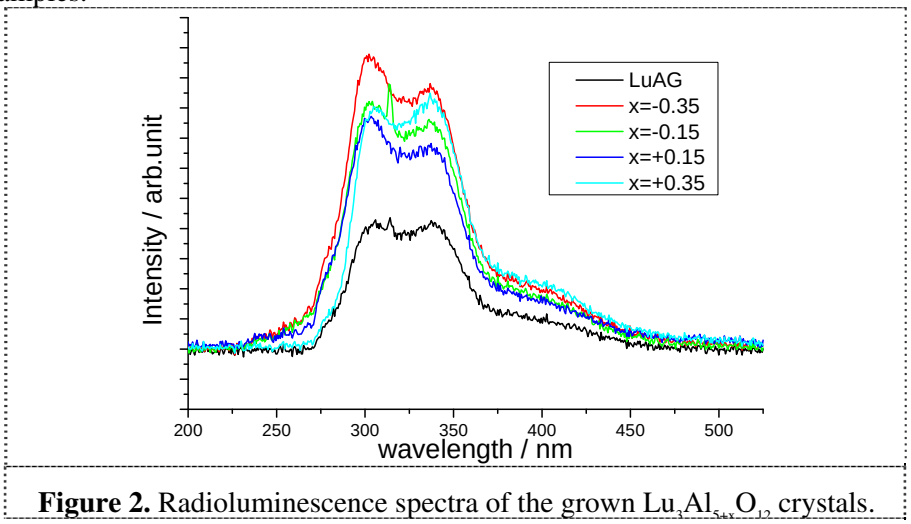

Figure 2. Radioluminescence spectra of the grown $Lu_3Al_{5+x}O_{12}$ crystals.

Figure 3 exemplifies energy spectra of the grown $Lu_3Al_{5+x}O_{12}$ samples irradiated by ^{137}Cs, where 662 keV. Light yield was increased with increasing the nonstochiometry. This result is good agreement with the result of radioluminescence spectra measurement. The x=-0.35 sample showed the highest light yield of around 12000 photons/MeV considering with the emission wavelength of the samples and quantum efficiency of the PMT (44%@330nm, 30%@480nm). Light yields of x= 0, 0.05, 0.15, 0.35, -0.05, -0.15, -0.35 were 8000, 8600, 9200, 12000, 8200, 9000 and 11000 photons/MeV, respectively.

Scintillation decay curves were obtained by using the PMT and digital oscilloscope and the sample was irradiated by ^{137}Cs (Figure 4). Scintillation decay time became slower with increasing the nonstochiometry. Scintillation decay time of x= 0, 0.05, 0.15, 0.35, -0.05, -0.15, -0.35 were 1.10, 1.32, 1.43, 1.96, 1.59, 1.62 and 1.88μs, respectively.

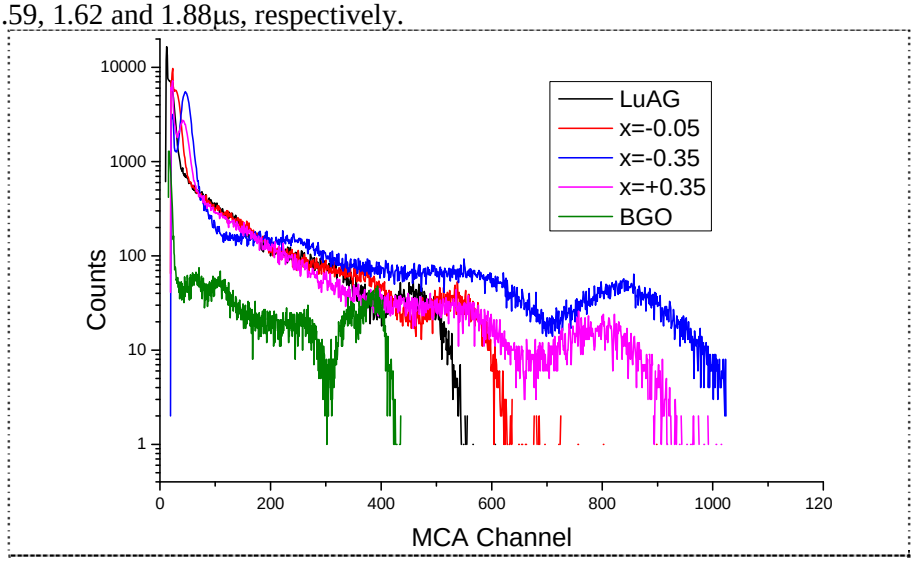

Figure 3. Energy spectra of the grown $Lu_3Al_{5+x}O_{12}$ crystals and BGO standard excited by a 662 keV gamma ray.

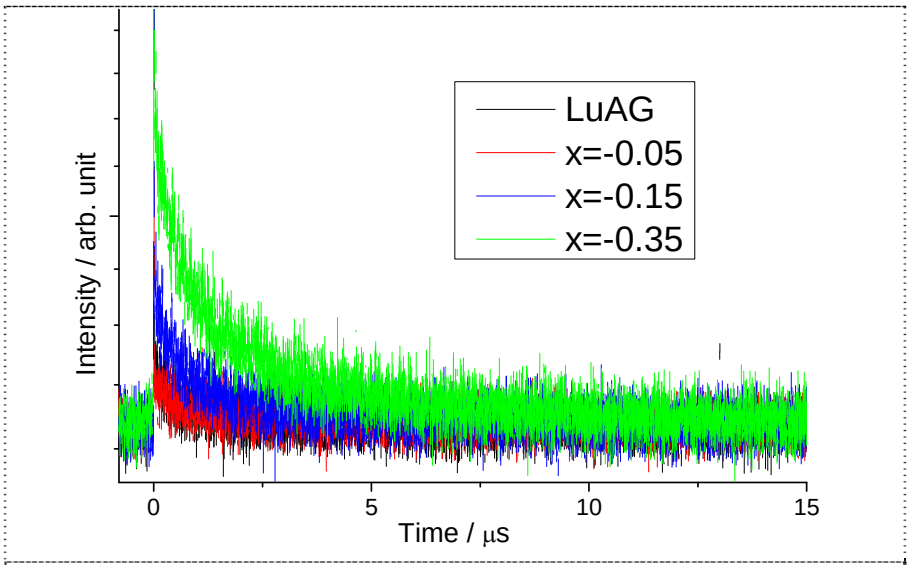

Figure 4. Decay curves of Nd1%:$Y_3Al_5O_{12}$ and Nd1%:$Y_3Ga_3Al_2O_{12}$ excited by a 662 keV gamma-ray using the PMT

4. Conclusion

Nonstoichiometric $Lu_3Al_{5+x}O_{12}$ (x= 0.05, 0.15, 0.35, 0, -0.05, -0.15, -0.35) crystals were grown by the μ-PD method. The grown crystals were colorless transparent with 2-3 mm in diameter and 15-23 mm in length. Expected anti-site defect related host emission have been observed in 250-420nm. Emission intensity was increased by increasing the nonstochiometry. Light yield was increased with increasing the nonstochiometry. The x=-0.35 sample showed the highest light yield of around 12000 photons/MeV. Scintillation decay curves were obtained by using the PMT and digital oscilloscope and the sample was irradiated by ^{137}Cs. Scintillation decay time became slower with increasing the nonstochiometry. Scintillation decay time of x= 0, 0.05, 0.15, 0.35, -0.05, -0.15, -0.35 samples were 1.10, 1.32, 1.43, 1.96, 1.59, 1.62 and 1.88μs, respectively.

5. References

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