Comparative study of transparent ceramic and single crystal Ce doped LuAG scintillators

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Abstract
Transparent ceramic Ce 0.5% doped Lu3Al5O12 (LuAG) scintillator grown by the sintering method and single crystalline Ce doped LuAG grown by the Czochralski method are prepared. They are cut to the physical dimensions 4 × 4 × 2 mm3. Their transmittance and radio luminescence spectra are evaluated. They are both transmissive in wavelength longer than 500 nm and intense Ce3+ 5d–4f emission appears around 520 nm. When 137Cs γ-ray is irradiated, 662 keV photo-absorption peaks are clearly observed in each sample. The transparent ceramic one shows higher light yield than that of the single crystalline one. The absolute light yield of the ceramic sample is turned out to be 14800 ± 1500 ph/MeV. The decay time constants are evaluated under pulse X-ray excitation. The main component of the decay time of ceramic and single crystalline one are determined as 37 and 46 ns, respectively.

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1. Introduction

Inorganic scintillators have been playing a major role as a radiation detector in the field of medical and security applications. The term of inorganic scintillator means generally single crystals. In spite of their usefulness, single crystals are subject to several difficulties. For example, they become progressively more difficult to produce, and more expensive, as the size increases. Transparent ceramics, made by sintering together a large number of single crystal grains, can potentially overcome these problems, and they suit to medical imaging applications which require bulk scintillators. In addition to above industrial applicability, it is still fresh in our minds that Nd-doped Y3Al5O12 (Nd:YAG) ceramics shows higher laser output than that of single crystalline Nd:YAG (Lu et al., 2004). Therefore, we can expect ceramics higher performance in scintillator applications.

In scintillator uses, transparent ceramic Ce:YAG was first developed in 1997 (Zych et al., 1997). At that time, scintillation performance of the ceramic one did not overcome the single crystal. After this invention, Konoshima Chemical has improved the fabrication processes. With a cooperation of Konoshima Chemical and our group, the scintillation light yield of the ceramic Ce:YAG exceeded that of the single crystalline counterpart (Yanagida et al., 2005). To improve the stopping power to γ-rays, we then developed ceramic Ce:(GdY)3Al5O12 (Ce:GYAG, Yanagida et al., 2007), and Pr:Lu3Al5O12 (Pr:LuAG, Yanagida et al., 2009). In recent years, some groups follow these developments, and especially, Ce:GYAG based scintillator (Ce:GYGAG) shows excellent scintillation performance (Cherepy et al., 2009). The present work describes the comparative study of transparent ceramic Ce:LuAG and its single crystalline counterpart in optical and radiation properties. Single crystalline Ce:LuAG showed the absolute light yield 12,500 ph/MeV (Mares et al., 2004). In single crystal growth, Ce concentration in LuAG host is limited to 0.2–0.3 mol% due to the segregation phenomenon. This low Ce concentration prevented us from getting high light yield from Ce:LuAG. Because ceramics are synthesized by the solid state reaction, higher doping of Ce is possible. Transparent ceramic Ce:LuAG was first examined in 2007 (Cherepy and Joshua, 2007). So far, some groups challenged to exceed the light yield of single crystal Ce:LuAG by the transparent ceramic one (Cherepy et al., 2009), but there is no one who realizes this purpose. As it is commonly known, physical properties of transparent ceramics strongly depend on the fabrication method.
of each group and this is the first study of transparent ceramic Ce:LuAG by our group.

2. Sample preparation and experimental procedures

Transparent ceramic Ce:LuAG was fabricated by Konoshima Chemical company. The Ce concentration was 0.5 mol% and the fabrication way was similar as that of Nd:YAG laser ceramics (Lu et al., 2004). On the other hand, single crystalline Ce:LuAG was grown by the conventional Czochralski method with an RF heating system. The rotation rate was 8–12 rpm and growth rate was 1.0 mm/h. An automatic diameter control system that uses crystal weighing was applied to control the crystal diameter. As a result, the crystal of 30 mm in diameter and 60 mm high were successfully grown. An Ar atmosphere was used to prevent oxidization of the crucible. Taking into account the segregation, the actual Ce concentration was 0.25 mol% which was almost the maximum doping level in single crystalline Ce:LuAG. Fig. 1 represents cut and polished ceramic and single crystalline Ce:LuAGs.

Transmittance was measured by using JASCO V550 spectrometer in the 190–900 nm range. The wavelength resolution was set to 1 nm. Then, to grasp emission wavelength of the sample, photoluminescence and excitation spectra were investigated by the Spectrofluorometer FLS920 (Edinburgh Instruments) equipped with the hydrogen steady-state, nanosecond and Xe microsecond pulsed flashlamps (IBH Scotland).

In pulse height analysis, Si avalanche photodiode (Si-APD) was selected as a photodetector because Ce:LuAG showed green-yellow emission. The samples were optically coupled with reverse type APD (Hamamatsu, S8664-55) and wrapped by Teflon seat to collect scintillation photons. Once the radiation was detected, signals were fed into preamplifier (CP 581K), the shaping amplifier (CP4417) with 2 µs shaping time, multichannel analyzer (Amptek 8000A), and finally to the computer. The decay time was also evaluated using our original apparatus which has a timing resolution as several tens of ps (Yanagida et al., 2010).

3. Experimental results

Transmittance and emission spectra of these samples are represented in Fig. 2. In both Ce:LuAGs, absorption lines appeared at 230, 340, and 460 nm. In wavelengths shorter than 400 nm, absorption depth was stronger in the ceramic one due to higher Ce concentration. On the other hand, they both showed ~80% transparency at wavelength longer than 500 nm. Thus, it is confirmed that the Ce concentration of ceramics is much denser than that of the single crystalline counterpart. This is the first result in Ce:LuAG that the light yield of the ceramic exceeds that of the single crystalline counterpart. Then in emission spectra, the emission wavelengths of the ceramic and the single crystal were 626 ch and 771 ch, respectively. The light yield of the ceramic one was 23% larger than that of the single crystalline counterpart. As a calibration, we irradiated 55Fe 5.9 keV X-ray directly to Si-APD to know the relation between MCA channel and number of electron-hole pairs in advance. Compared with 5.9 keV (=1640 electron-hole pairs) peak, absolute light yields of the ceramic and the single crystal were calculated as 14,800 ± 1500 and 12,000 ± 1200 ph/MeV.

In X-ray excited decay time measurements, main decay time of the ceramic and the single crystal were 37.3 ± 2.6 and 46.1 ± 2.0 ns, respectively. The observed value of the ceramic is slightly faster than reported value in the previous work (43 ns, Cherepy and Joshua, 2007).

137Cs pulse height spectra are exhibited in Fig. 3. 662 keV photo-absorption peak and Compton edge were clearly observed. Taking into account Lu escape peak, we used a double Gaussian function to fit. As a result, the photo-absorption peaks of the ceramic and the single crystal were 626 ch and 771 ch, respectively. The light yield of the ceramic one was 23% larger than that of the single crystalline counterpart. This is the first result in Ce:LuAG that the light yield of the ceramic exceeds that of the single crystal. As a calibration, we irradiated 55Fe 5.9 keV X-ray directly to Si-APD to know the relation between MCA channel and number of electron-hole pairs in advance. Compared with 5.9 keV (=1640 electron-hole pairs) peak, absolute light yields of the ceramic and the single crystal were calculated as 14,800 ± 1500 and 12,000 ± 1200 ph/MeV.
4. Conclusion

We fabricated ceramic and single crystalline Ce:LuAG and compared the optical and radiation properties of both scintillators. They were both transparent in wavelength longer than 500 nm and showed strong emission due to Ce$^{3+}$ 5d→4f transition around 500 nm. The light yield of the ceramic one exceeded that of the single crystalline one. As a result, the absolute light of the ceramic was evaluated as 14,800 ± 1500 ph/MeV and overcame the single crystal. The main component of the decay time was around 37 ns which was the typical value of Ce$^{3+}$ doped scintillators.

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