Scintillation properties of composite ceramic YAG and its capability on pulse shape descrimination

Takayuki YANAGIDA,¹ Kenichi WATANABE,² Yutaka FUJIMOTO, Akira URITANI,³ Hideki YAGI** and Takagimi YANAGITANI**

Kyushu Inst. Technol., 2–4 Hibikino, Wakamatsu, Kitakyushu 808–0196, Japan
¹Nagoya Univ., Furocho, Chikusa, Nagoya 464–8603, Japan
**Konoshima Chemical, 80 Kouda, Takuma, Mitoyo, Kagawa 769–1103, Japan

Transparent ceramic composite of non-doped and Ce-doped YAG was prepared by Konoshima Chemical with the vacuum sintering technique and investigated on pulse shape discrimination capability for functional collimator. Scintillation responses such as X-ray induced radioluminescence and decay time profiles of non-doped and Ce-doped YAG were evaluated. Pulse shape discrimination capability of the composite ceramic was evaluated by ¹³⁵Cs 662 keV γ-ray irradiation. As a result, clear separation of γ-ray events at non-doped or Ce-doped YAG layers was observed in two dimensional histogram by using signal processing with two different shaping times.

Key-words : Composite ceramic, YAG, Scintillator, Pulse shape discrimination, Gamma-ray

1. Introduction

Scintillators that convert an ionizing radiation to visible photons by an energy migration from host to emission centers¹ have been widely used for radiation detectors, including medical imaging,¹¹ security,¹¹ oil-logging,¹¹ astrophysics⁵ and particle physics.⁶ Especially, scintillation detectors for medical applications such as PET, X-ray CT, and SPECT have attracted much attentions due to the rise of the number of patients of cancer. In SPECT, one of the important techniques is a collimator that affects the spatial resolution of the detector in a large way.⁷–¹¹ Generally, collimators are passive materials like lead or tungsten block and they become a dead area of the detector. Recently, novel unique technique, a functional collimator,¹² was proposed. In this idea, they examined an active collimator also that had a sensitivity to γ-rays as same as the main scintillator and exhibited better performance from the viewpoint of spatial resolution and detection efficiency than the conventional detectors consisting of scintillators and passive collimators.¹²

In order to proceed this idea for actual applications, one of the important problem is a configuration of main scintillators and active collimator materials. For this configuration, it would be natural to use also scintillator materials as a collimator part to detect X- or γ-rays efficiently. If we use scintillator materials as active collimators, optical junctions of main scintillators and collimator scintillators are important because inferior optical junctions would make radiation imaging worse. In the present work, we investigated the transparent ceramic composite consisting of Ce:YAG and nondoped YAG aiming to apply functional collimator uses. Composite ceramic was developed in laser field for higher laser intensity.¹³–¹⁶ Unlike single crystals which possess a dominant position in scintillation detectors, ceramics are fabricated by the sintering method and the composite structure can be fabricated easily. The advantage of the composite ceramic is to combine different materials in nano-scale so that no optical adhesions are required. Figure 1 represents the sample for this study prepared by Konoshima Chemical using a vacuum sintering method. Yellowish and colorless parts were Ce 0.5% doped and nondoped YAG, respectively. Surface area of the sample was 10 × 6.5 mm² and thickness of each layer was 1 mm. All surfaces were polished for optical evaluations.

2. Experimental procedure

Backscattered electron images were observed by using scanning electron microscope (SEM, Hitachi S-3400). Basic scintillation properties of X-ray induced radioluminescence and scintillation decay time profiles were evaluated. In the radioluminescence, X-ray generator was used as an excitation source, and supplied bias voltage and tube current were 80 kV and 1 mA, respectively. Scintillation photons were detected monochromator (SR163, Andor) equipped CCD (DU420A-BU2, Andor) camera. The detailed description can be seen in past work.¹⁵ To avoid an observation of complicated spectrum, we evaluated separated Ce-doped and nondoped YAG specimens also prepared by Konoshima Chemical. Scintillation decay time profiles were evaluated by using pulse X-ray streak camera system which enabled us to observe wavelength resolved scintillation characteristics with 80 ps timing resolution.¹⁶,¹⁷ Monitoring wavelengths of Ce-doped and nondoped YAG pieces were 525 ± 15

¹ Corresponding author: T. Yanagida; E-mail: yanagida@lsse.kyutech.ac.jp
and 300 ± 15 nm, respectively. In decay time evaluation, we used the composite sample because our system can observe wavelength resolved scintillation decay time profiles. These basic evaluations were done at room temperature.

Then, pulse shape discrimination was examined to distinguish γ-ray events at Ce-doped YAG and non-doped YAG layers. The sample was optically attached with PMT (Hamamatsu, R7600U-200) with an optical grease (OKEN 6262A). The non-doped YAG side surface was faced to the PMT's photocathode window. The anode signal from the PMT was fed into a high-speed digitizer (Agilent, U1071A). In order to derive pulse height (PH) for fast and slow emission, signal waveforms were digitally processed with 50 and 500 ns shaping times, respectively.

3. Results and discussion

Figure 2 exhibits SEM image around the junction of the sample. Clear and smooth nano-scale junction can be observed. X-ray induced radioluminescence spectra of Ce-doped and non-doped YAG are demonstrated in Fig. 3. Emission peak wavelengths of Ce-doped and non-doped YAG were 525 and 300 nm, respectively. These results were consistent with previous results.20,21 The origin of the former one was Ce\(^{3+}\) 5d-4f transition\(^{20}\) and the latter one was some kinds of intrinsic luminescence of garnet materials.\(^{21}\) The wavelength sensitivity of the PMT was from 250 to 650 nm so that both scintillation photons could be detected in the pulse shape discrimination test. Of course, wavelength discrimination is potentially possible for this composite. In actual radiation detectors for medical applications, scintillator array and position sensitive photodetectors are used and it would be difficult to use optical filters due to a spatial restriction. Figure 4 represents X-ray induced decay time profiles of Ce-doped and non-doped YAG using X-ray streak camera system.\(^{18,19}\) As clearly seen, main decay times of Ce:YAG and non-doped YAG were ~100 ns and ~1 μs, respectively. Therefore pulse shape discrimination is possible in this material system because there was an enough difference in decay time. Except Fig. 3, we used the composite ceramic material to whole experiments.

Figure 5 represents a simple two-dimensional histogram of fast and slow emission components for the ceramic composite under \(^{137}\)Cs γ-ray irradiation. The vertical and horizontal axes represent pulse heights processed with fast (50 ns) and slow (500 ns) shaping times, respectively. γ-ray events detected at Ce:YAG should show relatively similar pulse height in fast and slow shaping times because its scintillation decay time was fast. On the other hand, γ-ray events detected at non-doped YAG should exhibit higher pulse height in the slow shaping time than that in the fast slow shaping time because its scintillation signal cannot be enough accumulated in the fast shaping time due to the ballistic deficit. In this figure, γ-ray signals were clearly separated in the composite ceramic. Then, Fig. 6 shows a similar histogram but the vertical axis is changed to the ratio of fast and slow pulse height (fast/slow PH). In this figure, we could distinguish Ce-doped YAG and non-doped YAG events clearly by using only one line.

Scintillation decay time profiles under \(^{137}\)Cs γ-ray excitation extracted by two dimensional event selection is demonstrated in Fig. 7. These profiles were separately averaged for extracted events of each component. Extracted decay time profiles resembled to Fig. 4. The hump of Ce-doped YAG around 800 ns was not significant and it would be corrected by using other event selection criteria. By the same event selection, we reconstructed pulse height spectra of Ce-doped and non-doped YAG layers, as shown in Fig. 8. Photoabsorption peak of \(^{137}\)Cs 662 keV γ-ray was clearly observed in both layers. In previous report in transparent ceramic Ce 0.5% doped YAG, scintillation light yield
was around 20000 ph/MeV\(^2\)) and that of nondoped one was around 2700 ph/MeV\(^2\)) so that presented results were consistent with previous works by using ceramics of Konoshima Chemical.

One of the problems of this composite is that intrinsic luminescence of the garnet also arises in Ce-doped YAG layer and such a signal cannot be distinguished with the signal from the nondoped YAG layer. In the composite ceramic of this work, the intrinsic luminescence in Ce-doped YAG layer is negligible (see Fig. 3) so that it would not cause a problem. In addition, if we also use PH information, such events can be fully cut due to a discrepancy of the light yield as well the decay curve.

Throughout this work, it was experimentally confirmed that transparent ceramic composite materials could be applied for functional collimators. The advantage of the composite ceramic is that we can easily fabricate several layers with various shapes in one composite materials. Though material systems of transparent ceramics are restricted to cubic structure materials, this new technology has a large potential for radiation detection applications.

4. Conclusion

Ce 0.5% doped and nondoped YAG composite ceramic was examined in scintillation responses and pulse shape discrimination. Ce-doped YAG exhibited 525 nm scintillation with ~100 ns decay time while nondoped one showed 300 nm with ~1 µs under X-ray excitation. Under \(^{137}\text{Cs}\) \(\gamma\)-ray irradiation, pulse shape discrimination by using two different shaping times was examined. As a result, \(\gamma\)-ray events at Ce-doped and nondoped YAG layers were clearly separated in two dimensional histogram between different shaping pulse heights. By conducting event selection, scintillation decay time profiles and pulse height spectra under \(^{137}\text{Cs}\) excitation were successfully reconstructed. The composite ceramic can be useful for functional collimator in various ionizing radiation detectors.

Acknowledgement  This work was mainly supported by a Grant in Aid for Scientific Research (A)-26249147 from the Ministry of Education, Culture, Sports, Science and Technology of the Japanese government (MEXT) and partially by JST A-step. Partial assistance from Nippon Sheet Glass Foundation for Materials Science and Engineering, Tokuyama Science foundation, Iketani Science and Technology Foundation, Hitachi Metals Materials Science Foundation, Mazda Foundation, JFE 21st century Foundation, and The Asahi Glass Foundation, the Cooperative Research Project of Research Institute of Electronics, Shizuoka University and Collaborative Research Program of Institute for Chemical Research, Kyoto University (2014-31) are also gratefully acknowledged.

References


