

Scintillation light yield of Tb:Sr₂Gd₈(SiO₄)₆₀₂

Watanabe, Kenichi

Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University

Yanagida, Takayuki

Devision of Materials Science, Nara Institute of Science and Technology

Nakauchi, Daisuke

Devision of Materials Science, Nara Institute of Science and Technology

Kawaguchi, Noriaki

Devision of Materials Science, Nara Institute of Science and Technology

<https://hdl.handle.net/2324/7168661>

出版情報 : Japanese Journal of Applied Physics. 60 (10), pp.106002-1-106002-5, 2021-10-07. IOP Publishing

バージョン :

権利関係 : © 2021 The Japan Society of Applied Physics



Scintillation light yield of Tb: Sr₂Gd₈(SiO₄)₆O₂

Kenichi Watanabe^{1*}, Takayuki Yanagida², Daisuke Nakauchi², Noriaki Kawaguchi²

¹ Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University, 744 Motooka,
Nishi-ku, Fukuoka 819-0395, Japan

² Division of Materials Science, Nara Institute of Science and Technology, 8916-5 Takayama-cho, Ikoma,
Nara 630-0101, Japan

*Email of corresponding author: k-watanabe@nucl.kyushu-u.ac.jp

Abstract

We have constructed the scintillation light yield evaluation system for the scintillator with millisecond decay time, which is based on the digital signal processing and the analog integration circuit with the decay time of 0.1 ms. The digital pulse shape discrimination technique is introduced to check and/or remove the influence of the pile-up events. As the first trial of the absolute scintillation light yield evaluation for scintillators with millisecond decay time, we attempt to acquire the scintillation light yield spectrum of the Tb: Sr₂Gd₈(SiO₄)₆O₂ (Tb: SrGS). The pulse area spectrum obtained from the Tb: SrGS scintillator shows a clear peak corresponding to the full-energy absorption events of 662 keV gamma-rays from a Cs-137 source. The peak channels in the pulse area spectra are almost the same both for the Tb: SrGS and bismuthgermanate (BGO) scintillator. Considering the difference in the wavelength matching between scintillators and the photomultiplier tube (PMT), the Tb: SrGS scintillator has 2.8 times higher scintillation light yield compared with the BGO. We finally conclude that the scintillator light yield of the Tb: SrGS is evaluated to be 23,000 photons/MeV.

KEYWORDS: *ionizing radiation; scintillator; scintillation detector; scintillation light yield; millisecond decay; Tb: SrGS*

1. Introduction

Scintillators are one of luminescent materials, and have been used as ionizing radiation detectors in various applications, such as medical diagnostics,^{1)2),3)4)5)} security,⁶⁾⁷⁾⁸⁾ environmental monitoring⁹⁾¹⁰⁾, well-logging,¹¹⁾¹²⁾ and high energy physics.¹³⁾¹⁴⁾¹⁵⁾ Scintillation detectors generally consist of a scintillator and photodetector such as photomultiplier tube (PMT) or Si photodiode (PD). The functions of scintillators are to absorb ionizing radiation and convert the absorbed radiation energy to numerous number of photons immediately.¹⁶⁾⁻²⁰⁾ In the recent trend, scintillator materials consist of a host and an emission center, as same as the other phosphor materials. The main function of the host is to efficiently absorb the incident ionizing radiation, and that of the emission center is to emit longer-wavelength photons in the ultraviolet, visible or infrared region. The combination of the host and emission center is a key issue to design the scintillator material.

In order to search efficient scintillator materials, some kind of index is important to evaluate the scintillator performance. The required indexes depend on the applications. One of the fundamental and significant indexes is the light yield. To explain the scintillation process, the semi-empirical approach was done in 1980 by Robbins²¹⁾ based on semiconductor physic. According to the Robbin's model, the relative scintillation light yield per unit deposited energy L is described as following:

$$L = \frac{n_{e-h}}{E_i} SQ = \frac{\left(\frac{E_i}{\beta E_g} \right)}{E_i} SQ = \frac{SQ}{\beta E_g} \quad (1),$$

where n_{e-h} is the number of electron-hole pairs created by the incident ionizing radiation, E_i the incident radiation energy, E_g the bandgap energy, β is the constant parameter, S the energy migration efficiency from the host to emission centers and Q the quantum efficiency. The β and E_g depend on the host material. On the other hand, the S and Q are considered to depend on the combination of the host and emission center. Among them, the quantum efficiency or quantum yield Q can be determined by the photoluminescence measurement.^{22),23)} On the other hand, to determine the energy migration efficiency S , the scintillation light yield measurement is essential. To deeply understand the scintillation mechanism, we have to systematically

1 study various scintillator materials.

2 The light intensity of scintillators is frequently evaluated by the X-ray induced radioluminescence (RL)
3 measurement. However, in this method, the measured light intensity depends not only on the scintillation
4 efficiency but also on the X-ray absorption efficiency, sample size and afterglow component, which arises
5 from thermally stimulated luminescence at around room temperature. Therefore, to evaluate the absolute
6 scintillation light yield, the signal pulse height spectrum is acquired and compared with one obtained from the
7 reference scintillator, such as a bismuthgermanate (BGO), whose light yield has already been evaluated^(24),25).
8 Since the signal gain is affected by the wavelength matching between the scintillator and the photodetector,
9 the wavelength dependence of the photodetector sensitivity and the scintillation emission spectrum is
10 considered to correct the signal pulse height. The instrument for the pulse height spectrum measurements
11 consists of a photodetector, pre-amplifier, shaping amplifier and multichannel analyzer in addition to a
12 scintillator. The pre-amplifier and shaping amplifier are designed to focus only on the frequency band related
13 to the scintillation signals and improve a signal-to-noise ratio. Since the scintillation detectors used in the
14 pulse-mode operation have a relatively fast timing response shorter than a few tens of microsecond, these
15 amplifiers are designed for a microsecond-timing response. Therefore, the scintillator light yield has never
16 been evaluated for slow scintillators with a decay time longer than milliseconds.

17 However, there are a number of applications using scintillators with millisecond decay time. For
18 example, some of the flat-panel detectors (FPDs) for the X-ray radiography applies terbium doped
19 gadolinium oxysulfide (Tb:Gd₂O₂S, GOS), which has millisecond decay time, as a scintillator screen. In this
20 type of applications, since the brightness is more important than fast response, the relatively slow but bright
21 scintillators are frequently applied. Another example is use of red or infrared scintillators in an
22 optical-fiber-type dosimeter²⁶⁾. Since this type of detectors is operated in the current mode, scintillators with
23 the relatively long decay time can also be accepted. In addition, when they are used in an intense radiation
24 field, red or infrared scintillators are preferentially selected because the light attenuation in an optical fiber is
25 generally small in the red or infrared wavelength region. These red or infrared scintillators generally have
26 long decay time. As described above, the slow but bright scintillators have various applications. In addition, it

1 is important to systematically characterize these scintillators as fundamental information to deeply
2 understand the scintillation mechanism. As examples of the slow but bright scintillators, some recent works
3 focused on Tb-doped scintillators as an emission center, which is based on a Tb^{3+} 4f-4f transition with
4 relatively long decay time^{27),28)}. Among them, the $\text{Tb}:\text{Sr}_2\text{Gd}_8(\text{SiO}_4)_6\text{O}_2$ (Tb:SrGS) scintillator shows
5 relatively high light yield but millisecond decay time²⁸⁾.

6 As described above, for the scintillators with millisecond decay time, the scintillation light yield has
7 never been evaluated. In this paper, as the first trial of the absolute scintillation light yield evaluation for
8 scintillators with millisecond decay time, we attempt to acquire the scintillation light yield spectrum of the
9 Tb:SrGS. Since there is no standard system to evaluate the light yield spectrum of scintillators with
10 millisecond decay time, we first construct the evaluation system and then confirm its validity using the
11 reference BGO scintillator. We finally measure the light yield spectrum of the Tb:SrGS and compare with
12 one of the BGO scintillator.

15 2. Materials and methods

16 2.1. Tb:SrGS scintillator

17 3% Tb-doped SrGS crystal were synthesized by the floating zone (FZ) method. Here, we assumed the
18 substitution of Tb^{3+} ions to Gd^{3+} ions. First, raw material powders of Tb_4O_7 , Gd_2O_3 , SrCO_3 and SiO_2 were by
19 using mortar and pestle. The mixed powder was heated at 1100 °C for 10 h to remove CO_2 from SrCO_3 . Then
20 the obtained mixed powder was formed into a cylinder by a hydrostatic pressure. The cylinder rod of the
21 mixed powder was sintered at 1500 °C for 12 h to make a ceramic rod. Finally, a ceramic rod was loaded into
22 an FZ furnace (FZD0192, Canon Machinery Inc.) to synthesize a crystal under ambient atmosphere. The
23 pull-down and rotation rate were approximately 3 mm/h and 20 rpm, respectively. A relatively transparent
24 and less-crack part in the synthesized crystal was selected and cut out.

25 Properties of the synthesized crystal, such as the crystal structure, photoluminescence (PL) properties,
26 X-ray induced RL spectrum and decay time, were evaluated²⁸⁾. From the X-ray diffraction (XRD) pattern,

any impurity phases were not confirmed within the detection limit. In the PL measurement, some emission lines due to Tb^{3+} 4f-4f transition were observed from 350 to 650 nm. The excitation wavelength was mainly from 250 to 290 nm. The quantum efficiency Q was evaluated to be 31%. The PL decay time was determined to be 1.86 ms. In the X-ray induced RL measurement, some emission lines due to Tb^{3+} were observed as well as the PL spectrum. The most intense emission line was at 540 nm, corresponding to $^5\text{D}_4$ to $^7\text{F}_5$ transition. The scintillation decay time was a little faster than the PL decay and was determined to be 1.33 ms.

2.2. Light yield evaluation system

Figure 1 shows the experimental setup for scintillation light yield evaluation. The evaluation system consists of a scintillator, PMT (Hamamatsu, R7600U-200), high voltage supply, digitizer (Pico Technology, picoScope 6402D) and control computer. The scintillator was attached with the PMT by a transparent resin and wrapped by a polytetrafluorethylene reflector sheet. The scintillator and PMT were covered by a black sheet to shield an ambient light. The applied voltage to the PMT was -700 V for all measurements. The PMT signal was directly fed into the digitizer to convert analog scintillation pulse signals to digital data. The digitized pulse shape signals were transferred to the control computer and analyzed. The input impedance of the digitizer was set to 1 M Ω to integrate the PMT current pulse signal with an adequate time constant. As a gamma-ray source, a Cs-137 source, which emits mono-energy gamma-rays with an energy of 662 keV, was used. For the purpose of validation of the constructed evaluation system and determination of the reference light yield, the light yield spectrum of the BGO scintillator was also acquired.

2.3. Signal processing procedure

Figure 2 shows an example of the digitized signal pulse shape obtained from the BGO scintillator. The pulse signal has a decay time of approximately 0.1 ms. This decay time is attributed mainly to the time constant of the RC integration circuit accidentally formed in this signal processing system. To determine the light yield for each event, the pulse area is integrated in the digitized signal pulse. The integration time should

be adequately set depending on the scintillator decay time. For the Tb:SrGS scintillator having the scintillation decay time of 1.33 ms, the integration time was set to be 3.75 ms, which is roughly three times longer than the decay time. The integration system with a long open window frequently suffers from the pile-up events, in which multiple-event pulses are integrated as a single event in the light yield determination process. This effect might distort the light yield spectrum. To check and/or remove the pile-up events, the pulse shape discrimination (PSD) technique can be applied. Here, the two integration windows were applied to extract the information of the pulse shape.²⁹⁾³⁰⁾ The First and second windows were set for 0.5 ms and 3.5 ms from the trigger time point, respectively. The first window corresponds to the fast component of the scintillation pulse. The second window is for the entire component.

3. Results and Discussion

3.1 Validation of the constructed system using BGO

Figure 3 a) and b) show the two-dimensional plot between the fast and entire components and the pulse area spectrum obtained from the BGO scintillator, respectively. Here, the pulse area is the entire component. **Figure 3 c), d) and e)** show examples of typical pulse shapes obtained in the characteristic regions indicated in Fig. 3 a). In this case, the full energy peak corresponding to 662 keV gamma-ray from Cs-137 source can be seen in the pulse area spectrum. However, the pile-up events significantly occur and the pulse area spectrum has some distortion. In this case, the counting rate was more than 200 cps. From the two-dimensional plot, we can confirm an influence of the pile-up events.

We can adjust the counting rate by changing the source-to-detector distance. **Figure 4 a) and b)** also show the two-dimensional plot between the fast and entire components and the pulse area spectrum. In this case, the counting rate was adjusted to be approximately 50 cps or less in order to suppress the pile-up events within an acceptable level. From the two-dimensional plot, no significant pile-up component is confirmed. The clear full-energy peak can be seen in the pulse area spectrum. The energy resolution is evaluated to be 18% FWHM. This resolution is a little worse compared with one obtained by the conventional pulse height

spectrum measurement system, in which the resolution is evaluated to be 12% FWHM for the same BGO scintillator. This is because that the wide integration window system is sensitive to the noise. However, this noise level is acceptable to evaluate the scintillation light yield. We confirm that the constructed evaluation system of the scintillation light yield works as expected.

3.2 Light yield evaluation of Tb:SrGS

Figure 5 a) shows the two-dimensional plot between the fast and entire components obtained from the Tb:SrGS scintillator. Since the source-to-detector distance was adjusted to suppress the pile-up events, the two-dimensional plot shows no pile-up component. **Figure 5 b)** shows the signal pulse shape averaged over the region indicated as red plots in Fig. 5 a), corresponding to the full-energy absorption events. The time constant of this decay profile is evaluated to be 1.2 ms. This well agrees with one obtained by the X-ray induced RL decay profile because this is much larger than the instrumental time constant, which comes from the RC integration circuit formed in the system, that is 0.1 ms.

Figure 6 shows the pulse area spectrum obtained from the Tb:SrGS scintillator. The spectrum obtained from the BGO scintillation by the same system is also plotted as a reference. The both spectra are confirmed to have a clear full-energy absorption peak. This is the first example of the pulse area or light yield spectrum obtained from the scintillator with millisecond decay time. The spectrum of the Tb:SrGS scintillator has a little smaller peak-to-Compton ratio than one of the BGO. This is considered to be valid taking into account the difference in the effective atomic number. The peak channels of the both scintillations are almost the same. However, since they have different emission spectra, we have to consider the difference in the wavelength matching between the scintillators and PMT. The PMT used in this study has the quantum efficiency of 25% for a wavelength of 480 nm, which is the peak emission wavelength of the BGO³¹⁾. On the other hand, this has the quantum efficiency of 9% for a wavelength of 540 nm, where the Tb:SrGS has the most intense emission line. The PMT has 2.8 times lower sensitivity for the Tb:SrGS scintillator compared with the BGO. This means that the Tb:SrGS has a 2.8 times higher scintillation light yield than the BGO. Therefore, we conclude that the Tb:SrGS has the scintillation light yield of 23,000 photons/MeV, assuming the light yield of

8,200 photons/MeV for the BGO²⁴⁾.

4. Conclusion

We construct the scintillation light yield evaluation system for the scintillator with millisecond decay time, which is based on the digital signal processing and the analog integration circuit with the decay time of 0.1 ms. The digital PSD technique is introduced to check and/or remove the influence of the pile-up events, because the signal integration system with relatively long-time window frequently suffers from the pile-up events. The PSD technique is quite useful to judge the system to be operated under an adequate counting rate. As the first trial of the absolute scintillation light yield evaluation for scintillators with millisecond decay time, we attempt to acquire the scintillation light yield spectrum of the Tb:SrGS. The pulse area spectrum obtained from the Tb:SrGS scintillator shows a clear peak corresponding to the full-energy absorption events of 662 keV gamma-rays from a Cs-137 source. In this experiment, the peak channels in the pulse area spectra are almost the same both for the Tb:SrGS and BGO scintillator. Considering the difference in the wavelength matching between scintillators and the PMT, the Tb:SrGS scintillator has 2.8 times higher scintillation light yield compared with the BGO. We finally conclude that the scintillator light yield of the Tb:SrGS is evaluated to be 23,000 photons/MeV. This is the first example of the light yield evaluation for scintillators with millisecond decay time.

As future works, we will measure the scintillation light yield of other scintillators with millisecond decay time by using the constructed system. Through systematic studies of the light yield evaluation, we will attempt to deeply understand the scintillation process, especially the energy migration from the host to emission centers.

Acknowledgements

This work was partially supported by Grant-in-Aid for Scientific Research (A) (17H01375) from the

Ministry of Education, Culture, Sports, Science and Technology of the Japanese government (MEXT).

References

- 1) S. Yamamoto, K. Kuroda and M. Senda, IEEE Trans. Nucl. Sci. **50** [5 II], 1683 (2003).
- 2) H. O. Anger, Nature **170**, 200 (1952).
- 3) C. L. Melcher, J. Nucl. Med. **41** [6], 1051 (2000).
- 4) S. Duclos, Interface-Electrochemical Soc. **7** [2], 34 (1998).
- 5) J. Krimmer, J. L. Ley, C. Abellan, J. P. Cachemiche, L. Caponetto, X. Chen, M. Dahoumane, D. Dauvergne, N. Freud, B. Joly, D. Lambert, L. Lestand, J. M. Létang, M. Magne, H. Mathez, V. Maxim, G. Montarou, C. Morel, M. Pinto, C. Ray, V. Reithinger, E. Testa and Y. Zoccarato, Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **787**, 98 (2015).
- 6) J. Glodo, Y. Wang, R. Shawgo, C. Brecher, R. H. Hawrami, J. Tower and K. S. Shah, Phys. Procedia **90** [November 2016], 285 (2017).
- 7) K. Watanabe, T. Yamazaki, D. Sugimoto, A. Yamazaki, A. Uritani, T. Iguchi, K. Fukuda, S. Ishidu, T. Yanagida and Y. Fujimoto, Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **784**, 260 (2015).
- 8) M. E. Ellis, K. Duroe and P. A. Kendall, Int. J. Mod. Phys. Conf. Ser. **44** [2010], 1660214 (2016).
- 9) K. Watanabe, T. Yanagida, K. Fukuda, A. Koike, T. Aoki and A. Uritani, Sensors Mater. **27** [3], 269 (2015).
- 10) M. Lowdon, P. G. Martin, M. W. J. Hubbard, M. P. Taggart, D. T. Connor, Y. Verbelen, P. J. Sellin and T. B. Scott, Sensors **19** [18], 3828 (2019).
- 11) T. Yanagida, Y. Fujimoto, S. Kurosawa, K. Kamada, H. Takahashi, Y. Fukazawa, M. Nikl and V. Chani, Jpn. J. Appl. Phys. **52** [7 PART 1], 3 (2013).
- 12) L. G. Howell and A. Froscht, GEOPHYSICS **4**, 81 (1939).
- 13) P. Lecoql and M. Korzhik, IEEE Trans. Nucl. Sci. **47** [4], 1311 (2000).

- 1 14) T. Itoh, T. Yanagida, M. Kokubun, M. Sato, R. Miyawaki, K. Makishima, T. Takashima, T. Tanaka,
2 K. Nakazawa, T. Takahashi, N. Shimura and H. Ishibashi, Nucl. Instruments Methods Phys. Res. Sect.
3 A Accel. Spectrometers, Detect. Assoc. Equip. **579** [1], 239 (2007).
- 4 15) K. Yu, J. Lasers, Opt. Photonics **04** [01], 1 (2017).
- 5 16) P. A. Rodnyi, *Physical Processes in Inorganic Scintillators* (CRC Press, London, 1997).
- 6 17) C. Ronda, H. Wiczorek, V. Khanin and P. Rodnyi, ECS J. Solid State Sci. Technol. **5** [1], R3121
7 (2016).
- 8 18) S. E. Derenzo, M. J. Weber, E. Bourret-Courchesne and M. K. Klintenberg, Nucl. Instruments
9 Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **505** [1–2], 111 (2003).
- 10 19) C. W. E. Van Eijk, **460**, 1 (2001).
- 11 20) T. Yanagida, J. Lumin. **169**, 544 (2016).
- 12 21) D. J. Robbins, J. Electrochem. Soc. **127** [12], 2694 (1980).
- 13 22) A. Endo, K. Suzuki, T. Yoshihara, S. Tobita, M. Yahiro and C. Adachi, Chem. Phys. Lett. **460** [1–3],
14 155 (2008).
- 15 23) T. Yanagida, Y. Fujimoto, K. Fukuda and V. Chani, Nucl. Instruments Methods Phys. Res. Sect. A
16 Accel. Spectrometers, Detect. Assoc. Equip. **729**, 58 (2013).
- 17 24) I. Holl, E. Lorenz and G. Mageras, IEEE Trans. Nucl. Sci. **35** [1], 105 (1988).
- 18 25) M. Moszynski, M. Kapusta, M. Mayhugh, D. Wolski and S. O. Flyckt, IEEE Trans. Nucl. Sci. **44** [3],
19 1052 (1997).
- 20 26) K. Ueno, T. Tadokoro, T. Fukumoto and S. Okada, J. Nucl. Sci. Technol. **56** [9–10], 822 (2019).
- 21 27) I. Kandarakis and D. Cavouras, Appl. Radiat. Isot. **54** [5], 821 (2001).
- 22 28) T. Yanagida and N. Kawaguchi, J. Ceram. Process. Res. **20** [6], 577 (2019).
- 23 29) K. Watanabe, Y. Kondo, A. Yamazaki, A. Uritani, T. Iguchi, N. Kawaguchi, K. Fukuda, S. Ishidu, T.
24 Yanagida, Y. Fujimoto and A. Yoshikawa, IEEE Trans. Nucl. Sci. **60** [2], 959 (2013).
- 25 30) T. Yanagida, K. Watanabe and Y. Fujimoto, Nucl. Instruments Methods Phys. Res. Sect. A Accel.
26 Spectrometers, Detect. Assoc. Equip. **784**, 111 (2015).

1 31) O. H. Nestor and C. Y. Huang, IEEE Trans. Nucl. Sci. **22** [1], 68 (1975).

2

3

Figure Captions

- Fig. 1** Experimental setup for the scintillation light yield evaluation.
- Fig. 2** An example of the digitized signal pulse shape obtained from the BGO scintillator.
- Fig. 3** a) Two-dimensional plot between areas of the fast and entire components and b) the pulse area spectrum obtained from the BGO scintillator. Examples of pulse shapes, which are extracted from the regions c), d) and e) indicated in Fig. 3 a), are also plotted. In this case, the pile-up events significantly occur.
- Fig. 4** a) Two-dimensional plot between areas of the fast and entire components, and b) the pulse area spectrum obtained from the BGO scintillator. In this case, the source-to-detector distance was adjusted to suppress the pile-up events within an acceptable level.
- Fig. 5** a) Two-dimensional plot between the fast and entire components and b) the averaged signal pulse shape obtained from the Tb:SrGS scintillator. The signal pulses are averaged over the region indicated as red plots in Fig. 5 a).
- Fig. 6** Pulse area spectrum obtained from the Tb:SrGS scintillator. The spectrum obtained from the BGO scintillator is also plotted as a reference.

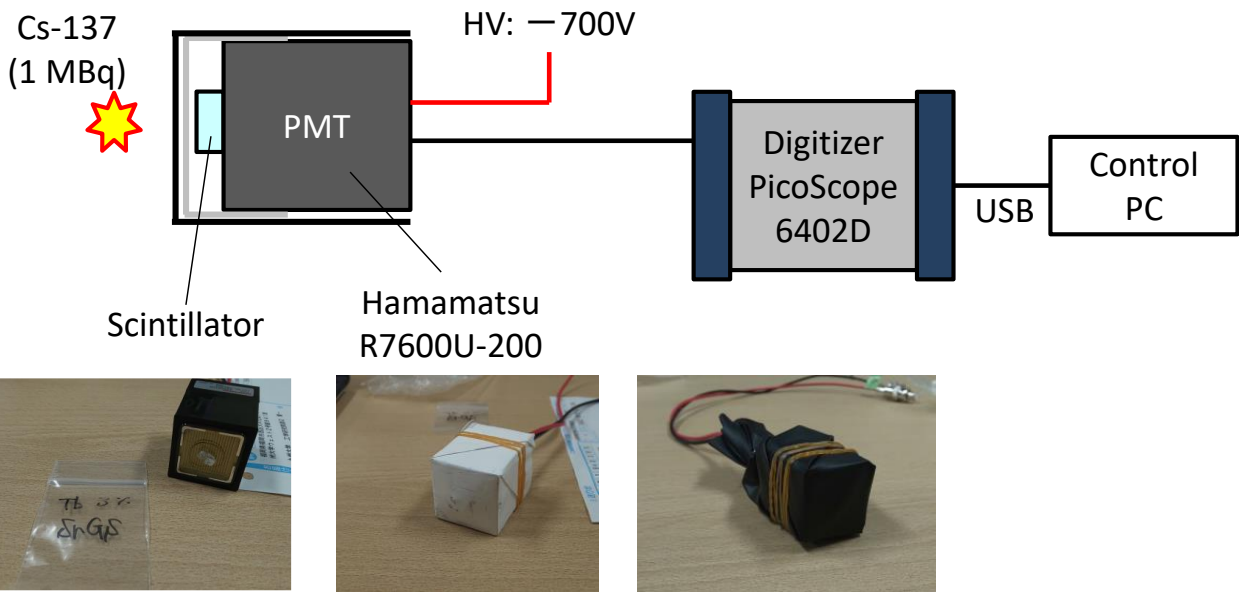


Fig. 1 Experimental setup for the scintillation light yield evaluation.

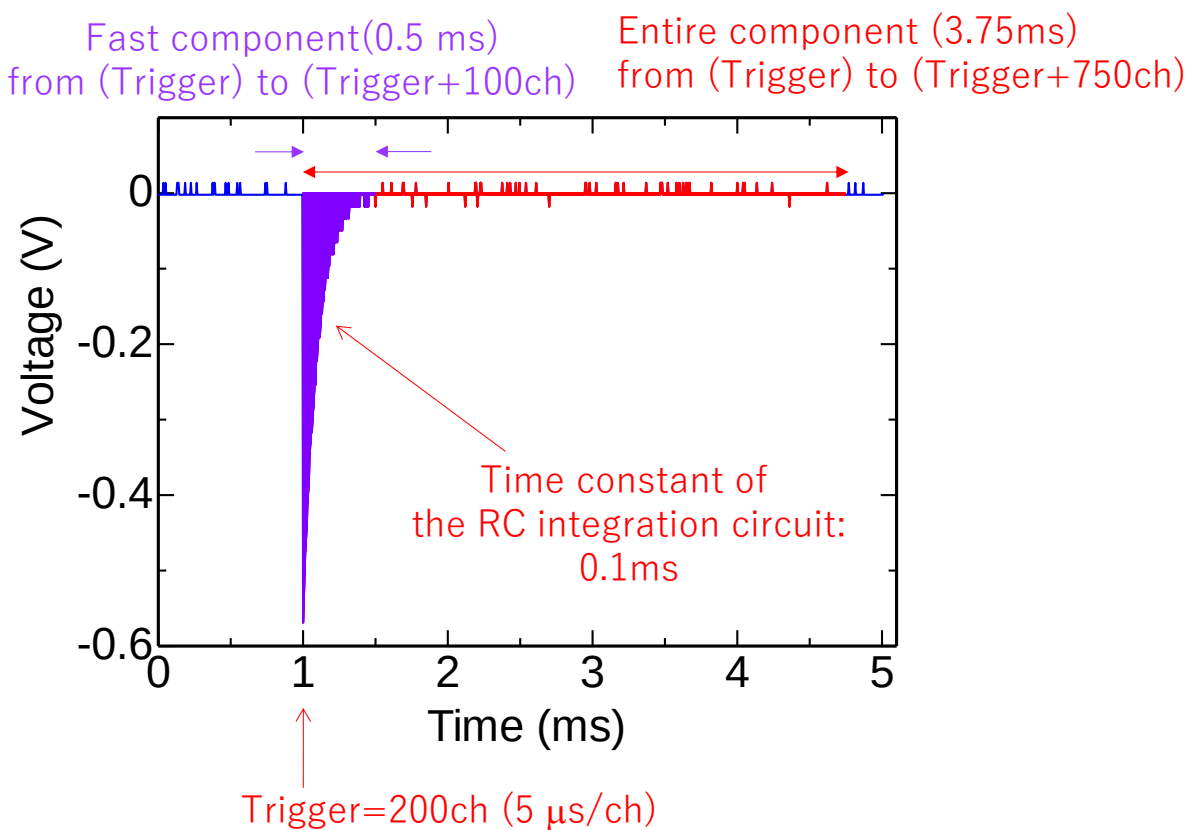


Fig. 2 An example of the digitized signal pulse shape obtained from the BGO scintillator.

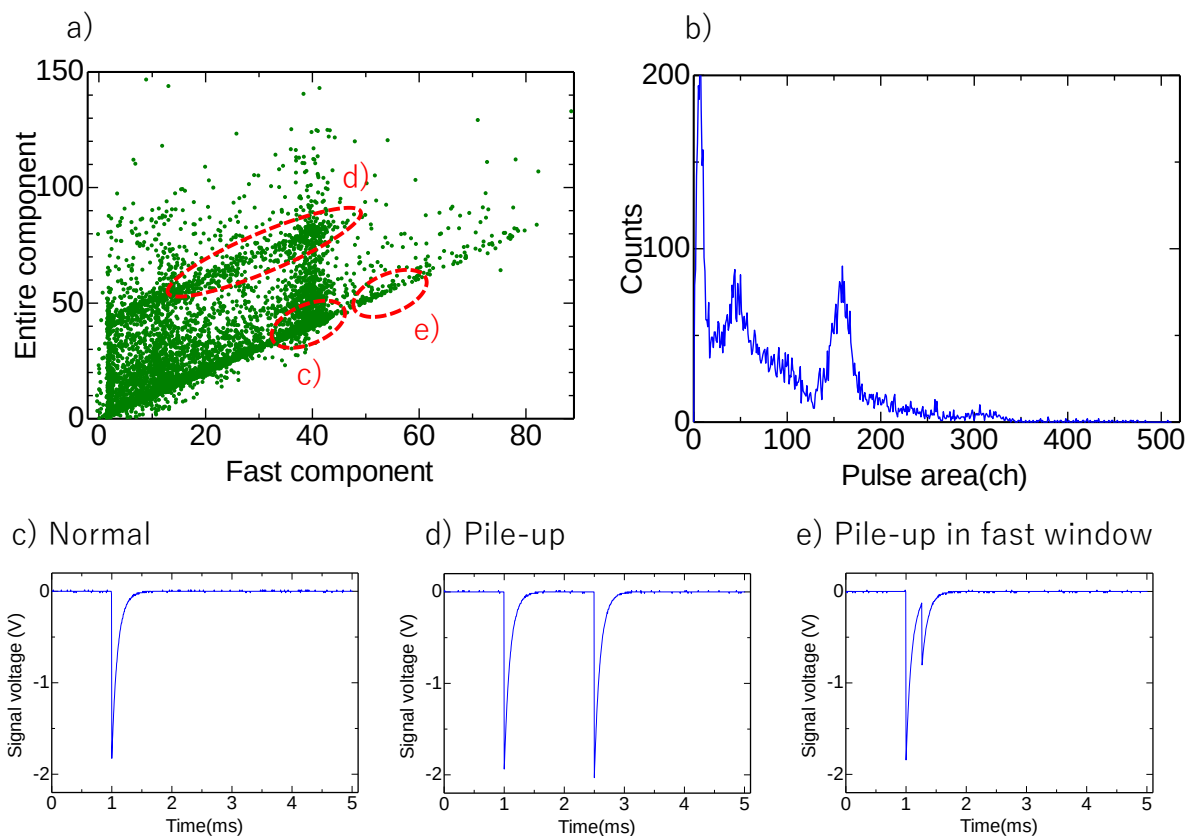


Fig. 3 a) Two-dimensional plot between areas of the fast and entire components and b) the pulse area spectrum obtained from the BGO scintillator. Examples of pulse shapes, which are extracted from the regions c), d) and e) indicated in Fig. 3 a), are also plotted. In this case, the pile-up events significantly occur.

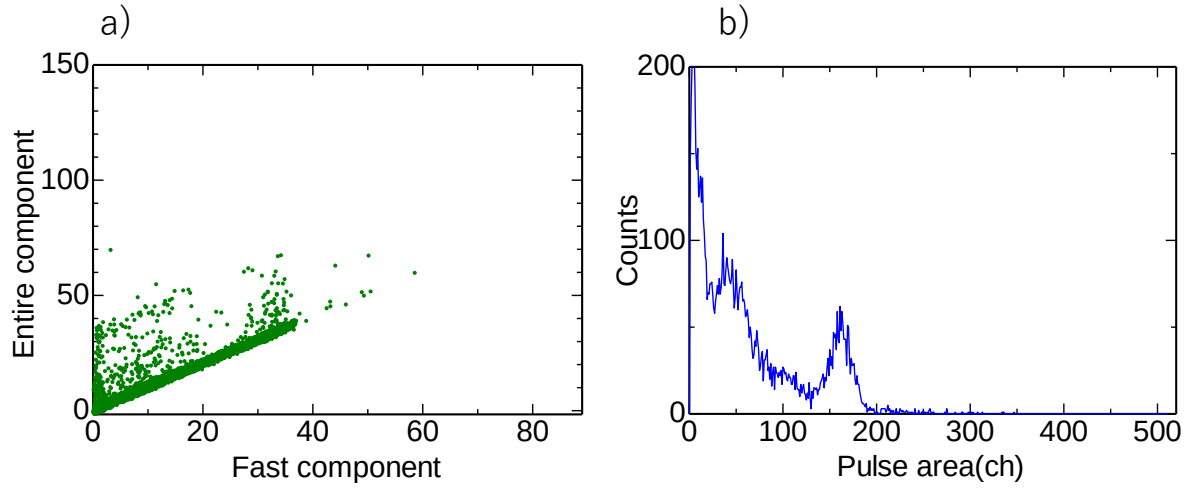


Fig. 4 a) Two-dimensional plot between areas of the fast and entire components, and b) the pulse area spectrum obtained from the BGO scintillator. In this case, the source-to-detector distance was adjusted to suppress the pile-up events within an acceptable level.

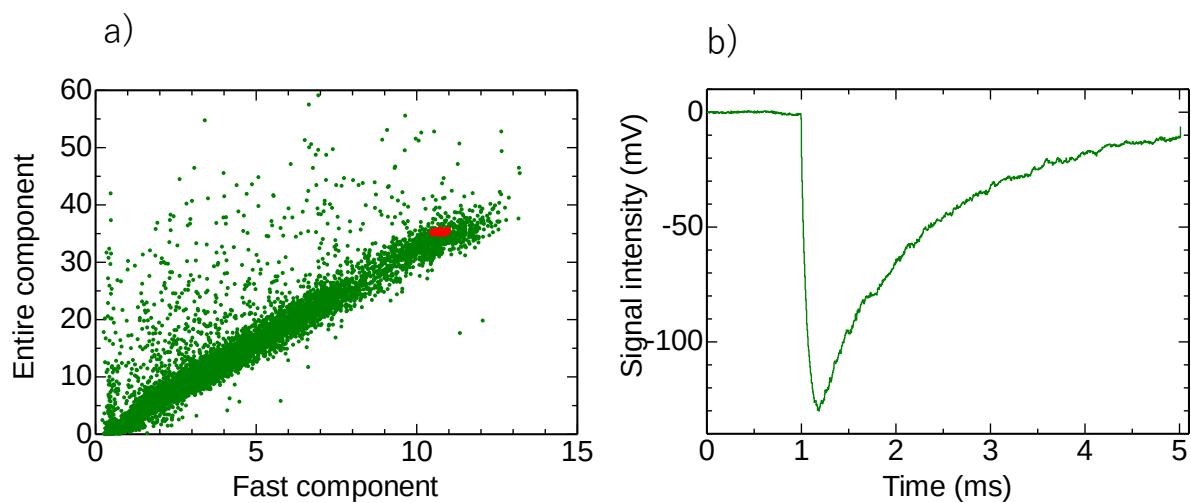


Fig. 5 a) Two-dimensional plot between the fast and entire components and b) the averaged signal pulse shape obtained from the Tb:SrGS scintillator. The signal pulses are averaged over the region indicated as red plots in Fig. 5 a).

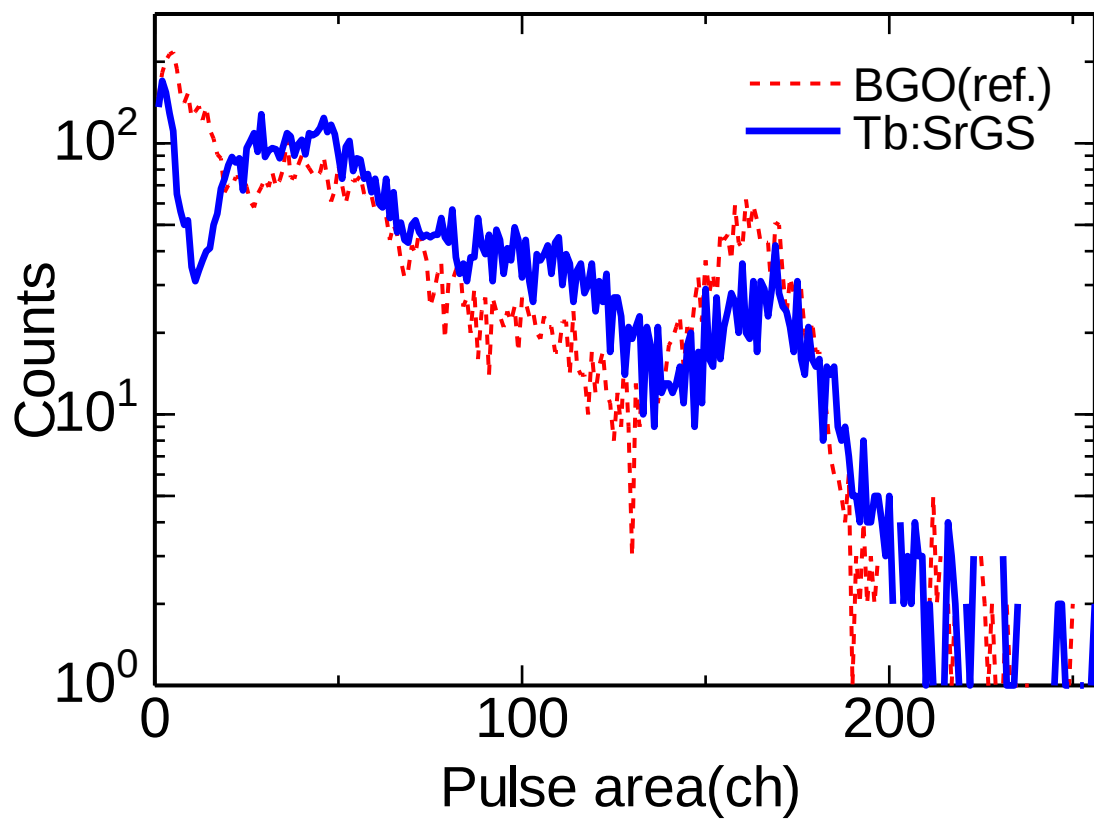


Fig. 6 Pulse area spectrum obtained from the Tb:SrGS scintillator. The spectrum obtained from the BGO scintillator is also plotted as a reference.