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Optimization of thickness of GAGG scintillator for detecting an alpha particle emitter in a field of high beta and gamma background

Yuki Morishita[a,*], Seiichi Yamamoto[b], Kenji Izaki[a], Junichi H. Kaneko[c], Katsuya Hoshi[a], Tatsuo Torii[a]

[a] Japan Atomic Energy Agency, 4-33, Muramatsu, Tokai-mura, Naka-gun, Ibaraki, Japan
[b] Nagoya University Graduate School of Medicine, 1-1-20 Daiko-Minami, Higashi-ku, Nagoya, Aichi, Japan
[c] Hokkaido University Graduate School of Engineering, Kita 13, Nishi 8, Kita-ku, Sapporo, Hokkaido, 060-8628, Japan

A B S T R A C T

At the site of the Fukushima Daiichi nuclear power plant (FDNPP), there is a large quantity of beta and gamma emitters such as ^{90}Sr and ^{137}Cs. Moreover, radon (Rn) progeny, which are naturally occurring radionuclides, exist and emit alpha and beta particles. To detect plutonium isotopes (^{238}Pu, ^{239}Pu, and ^{240}Pu) in a field of high beta and gamma background, an alpha particle detector with low beta- and gamma-ray sensitivity and good energy resolution for an alpha particle is required to distinguish plutonium isotopes from Rn progeny. Previously, we developed an alpha particle imaging detector by combining a 0.1-mm-thick gadolinium aluminum gallium garnet (GAGG) scintillator with a silicon photomultiplier (SiPM). However, this detector was sensitive to environmental gamma and beta rays. In this study, we optimized the thickness of the GAGG scintillator for alpha particle detection in a field of high beta and gamma background. We prepared three GAGG scintillators with thicknesses of 0.05 mm, 0.07 mm, and 0.1 mm. Each of the GAGG scintillators was coupled optically to the SiPM array, which was used as the photodetector. Alpha, beta, and gamma rays were irradiated onto the developed alpha particle detector, and their spectra were obtained. The energy resolution of the 0.05-mm-thick GAGG for 5.5-MeV alpha particles (~11.6% full width at half maximum [FWHM]) was the best among the three GAGG scintillators. All GAGG scintillators used in this study were not sensitive to gamma rays with a dose rate of 1 mSv/h. The beta particle count above the lower level discriminator (LLD) decreased because the scintillator was thinner, and the beta count of the 0.05-mm-thick GAGG was only 1/100 that of the 0.1-mm-thick GAGG. Because the alpha particle detector with the 0.05-mm-thick GAGG scintillator had low beta and gamma-ray sensitivity and good energy resolution for alpha particles, it is promising from the viewpoint of detecting plutonium contamination in a field with high beta and gamma background, such as the FDNPP site.

1. Introduction

At the site of the Fukushima Daiichi nuclear power plant (FDNPP), after the nuclear accident there exist large quantities of beta and gamma emitters, such as ^{90}Sr and ^{137}Cs. Not only beta and gamma emitters but also alpha particle emitters such as plutonium isotopes (^{238}Pu, ^{239}Pu, and ^{240}Pu) exist (Saito et al., 2015; Cao et al., 2017; Schneider et al., 2017). The activity of Pu isotopes is evaluated by measuring alpha particles, and this information is necessary to prevent internal exposure to workers (Dorrian, 1997; Foster, 1991). An alpha particle detector to be used at the FDNPP site must have two main features: low beta and gamma-ray sensitivity and good energy resolution for alpha particles. The background count of beta and gamma rays should be eliminated to measure the alpha activity accurately. Moreover, good energy resolution for alpha particles is required to distinguish radon (Rn) progeny, which are naturally occurring radionuclides that emit alpha particles.

A silicon surface barrier detector (SSBD) is used to distinguish Pu isotopes from radon progeny because it has good energy resolution (Phillips and Lindeken, 1963). However, the thin entrance window of the SSBD can be broken easily owing to aging degradation, and thereafter it becomes sensitive to external light (Knoll, 2010). The SSBD might be used outdoors at the FDNPP site and be exposed to sunlight. Moreover, the SSBD was found to be easily affected by external noise. When the SSBD was broken, it had to be replaced with a new one, and this is very expensive. Therefore, the SSBD is unsuitable for use under harsh conditions such as those at the FDNPP site.

A silicon photomultiplier (SiPM), a novel photodetector, can resist

* Corresponding author.
E-mail address: morishita.yuki@jaea.go.jp (Y. Morishita).

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mechanical shocks (Yamamoto et al., 2013a). An alpha particle detector with SiPM would be suitable for use under harsh conditions. For alpha particle detection, silver-doped zinc sulfide (ZnS(Ag)) scintillators have typically been used. However, their poor energy resolution (approximately 50% FWHM) is unsuitable for distinguishing plutonium isotopes from radon progeny (Morishita et al., 2014a).

Recently, a cerium-doped $Gd_3(Ga,Al)_5O_{12}$ scintillator (GAGG scintillator) was developed. Because of its good energy resolution and high density, the GAGG scintillator has been applied to gamma cameras, a Compton camera, and positron emission tomography (PET) (Nakamori et al., 2012; Kataoka et al., 2013; Yamamoto et al., 2013b). The wavelength of the scintillation light of the GAGG scintillator (approximately 530 nm) is well suited for use in silicon photomultipliers (SiPM) (Morishita et al., 2014a; Iwanowska et al., 2013). We have developed an alpha particle imaging detector by combining a 0.1-mm-thick GAGG scintillator with a SiPM. Its energy resolution for 5.5 MeV alpha particles is approximately 13% FWHM, which is far superior to that of ZnS (Ag). However, in background measurement, this detector is sensitive to environmental gamma and beta rays. Therefore, it is necessary to reduce its sensitivity to beta and gamma rays for use in fields with high beta and gamma backgrounds.

There is plenty of room for reducing the influence of gamma rays and beta particles by optimizing the thickness of the GAGG scintillator because it is thicker than the range of 5.5 MeV alpha particles in the GAGG scintillator. In this study, we optimized the thickness of the GAGG scintillator to eliminate the influence of beta and gamma rays.

2. Materials and methods

2.1. Preparation of GAGG scintillator

Before determining the appropriate thickness of the GAGG scintillator, the range of 5.5 MeV alpha particles (^{238}Pu) was calculated using the Bragg–Kleeman rule as follows:

$$\frac{R_1}{R_0} \cong \frac{\rho_0\sqrt{A_1}}{\rho_1\sqrt{A_0}} \tag{1}$$

where ρ represents density, and A represents atomic weight. The density of the GAGG scintillator is 6.63 g/cm^3, and its effective molecular weight is 46. Here, A_l is the atomic weight of aluminum and R_1 is the range of 5.5 MeV alpha particles in aluminum (Williamson et al., 1966). The range in GAGG was calculated to be 0.02 mm (23 µm). However, it was challenging to fabricate a 0.02-mm-thick GAGG scintillator because of the risk of breakage. Therefore, a scintillator with a thickness of 0.05 mm was prepared.

We used three GAGG scintillators with thicknesses of 0.05 mm, 0.07 mm, and 0.1 mm made by Furukawa Co. Ltd. (Fig. 1). The length and the width of these scintillators were 26 mm and 26 mm, respectively. Each of these scintillators was coupled optically to a 1-mm-thick glass plate and a 1-mm-thick acrylic light guide. A SiPM array (Through Silicon Via (TSV) MPPC array S12642-0404PA-50, Hamamatsu Photonics K.K. Japan) was used as the photodetector. The SiPM was suitable for coupling to GAGG scintillator regarding wavelength matching and both combination achieved good energy resolution (Morishita et al., 2014a). A silicon oil compound (Shin-Etsu silicone, Shin-Etsu Chemical Co. Ltd. Japan) was used for optically coupling the glass plate, acrylic light guide, and the SiPM array. Fig. 2 shows a schematic drawing of the alpha particle detector. An incident alpha particle is converted to scintillation light. The SiPM detects the scintillation light and generates an electrical signal. The analog signals from the SiPM are transferred to a weight-summing amplifier that produces a summed signal of all channels of the SiPMs. An analog-to-digital (A/D) converter digitalizes the summed signal. Finally, the energy spectrum is displayed on a personal computer. The data acquisition system is similar to the one used previously for development of the alpha camera (Morishita et al., 2014b). The alpha detector, scintillator with the SiPM array, and source were placed in a black box in all measurements.

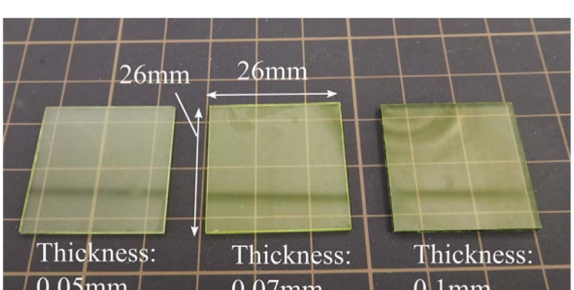

Fig. 1. GAGG scintillators of different thicknesses. From the left, the thicknesses of the GAGG scintillator are 0.05 mm, 0.07 mm, and 0.1 mm. The length and the width of these scintillators of all scintillators are the same (26 mm × 26 mm).

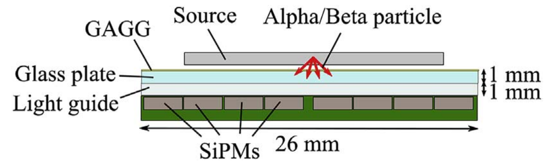

Fig. 2. Schematic drawing of alpha particle detector.

2.2. Irradiation of alpha particles

A 3.7-kBq alpha source (^{241}Am) was placed on the scintillator to irradiate 5.5 MeV alpha particles, and the pulse height spectra of the three GAGG scintillators were compared. A 0.1-mm-thick plastic scintillator was also measured as a reference. The three GAGG scintillators with different thicknesses and the plastic scintillator were replaced one-by-one. High voltage (HV) of the SiPM was fixed at −67.0 V. The measurement was conducted for 5 min and repeated three times.

2.3. Irradiation of beta particles

A 74-kBq ^{90}Sr/^{90}Y source was placed on the scintillator to irradiate beta particles, and the pulse height spectra of the three GAGG scintillators were compared. The measurement was conducted for 5 min and repeated three times. HV for the SiPM was the same as that in the case of alpha particle measurement.

2.4. Irradiation of gamma rays

The influence of 59.5 keV gamma rays from ^{241}Am was evaluated. A 296-GBq ^{241}Am gamma-ray source was placed 78.5 cm away from the center of the detector and the dose rate at the center of the detector was 1 mSv/h (Fig. 3). HV of the SiPM was the same as that in the alpha particle measurement.

2.5. Verification using Monte Carlo simulation

To verify the experimental results of the influence of beta particles and gamma rays, a Monte Carlo simulation using Electron Gamma Shower code version 5 was also conducted (Hirayama et al., 2005). In the simulation, the thicknesses of the GAGG scintillators were reproduced, that is, 0.05 mm, 0.07 mm, and 0.1 mm with 1mm-thick glass plate (Fig. 4). Gamma-ray photons of 59.5 keV (^{241}Am) and 662 keV (^{137}Cs), and beta particles from ^{90}Sr and ^{90}Y were irradiated from 0mm above the scintillator. RADAR beta spectrum data was used as the beta spectrum input (DOSEINFO-RADAR database). The Monte Carlo simulation scored the energy deposited in the scintillator for one radiation (energy deposition per source). The standard error of the mean of the

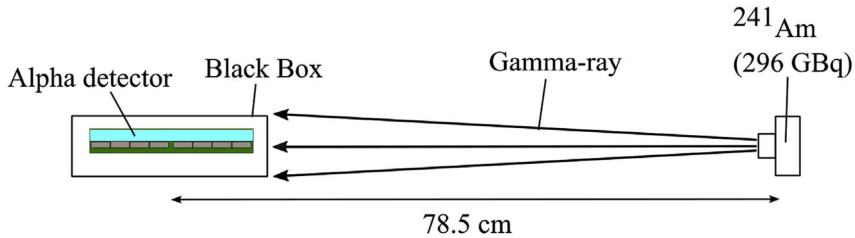

Fig. 3. Schematic drawing of gamma-ray irradiation.

energy deposition in all calculations was below 1%.

2.6. Measurement of Rn progeny

Rn progeny were collected electrically on a polyethylene film placed on a photomultiplier tube (PMT) by using an electrostatic collection method (Yamamoto and Iida, 1998; Hopke, 1989). High voltage of −2000V was applied to the PMT, and positively charged ^{218}Po and ^{214}Po of Rn progeny were collected in a nuclear fuel facility. The collection time was over 3 h. After the collection of Rn progeny, the polyethylene film (approximately 1000 dpm measured by an alpha counter) was placed on the developed alpha particle detector and measured. The 0.05-mm-thick GAGG scintillator was used for the measurement because its energy resolution was the best. High voltage of the SiPM was reduced slightly so that the output signal was not saturated by 7.7 MeV alpha particles from ^{214}Po. The measurement time was 1 h.

3. Results

3.1. Pulse height spectra of alpha particles

Table 1 summarizes the energy resolutions and the relative pulse heights of the three GAGG scintillators and the plastic scintillator, and Fig. 5 shows the pulse height spectra of the three GAGG scintillators and the plastic scintillator. The peak channel of the plastic scintillator was the lowest among the four scintillators. As the thickness of the GAGG scintillator decreased, the pulse height increased. The pulse height of the 0.05-mm-thick GAGG scintillator was 1.6 times higher than that of the 0.1-mm-thick GAGG scintillator and 3.7 times higher than that of the plastic scintillator. The energy resolutions of the 0.1-mm-, 0.07-mm-, and 0.05-mm-thick scintillators were 13.4% FWHM, 12.1% FWHM, and 11.6% FWHM, respectively. As the thickness of the GAGG scintillator decreased, its energy resolution increased.

3.2. Influence of beta particles and gamma rays

To eliminate the counts of beta particles and gamma rays, the lower level discriminator (LLD) was set to the lower channel of 1% of the peak channel counts of the ^{241}Am spectrum. The counts above the LLD were usable for alpha particle detection. The LLDs were 42-channel for 0.1 mm, 53-channel for 0.07 mm, and 65-channel for 0.05 mm.

Fig. 6 shows the pulse height spectra of the beta particles from the ^{90}Sr/^{90}Y source with the three GAGG scintillators. The counts below 18-channel were discriminated to remove electrical noise. This confirms that the counts above the LLD decreased as the scintillator thickness decreased. The counts above the LLDs of each scintillator were compared and normalized against 0.1-mm-thick GAGG = 1. The relative counts were 0.64 for the 0.07-mm-thick GAGG and 0.01 for the 0.05-mm-thick GAGG (Fig. 7).

Fig. 8 shows pulse height spectra of the 59.5 keV gamma rays with the three GAGG scintillators. The gamma-ray counts were considerably lower compared to those of the beta particles. Although the dose rate of the gamma rays was high (1 mSv/h), the count above the LLD was zero. Therefore, the influence of gamma rays was negligible.

Table 1
Energy resolutions and beta count rates of the three GAGG scintillators.

Thickness of GAGG	0.1 mm	0.07 mm	0.05 mm
Energy resolution [%]	13.4	12.1	11.6
Relative pulse height (plastic scintillator = 1)	2.4	2.9	3.7

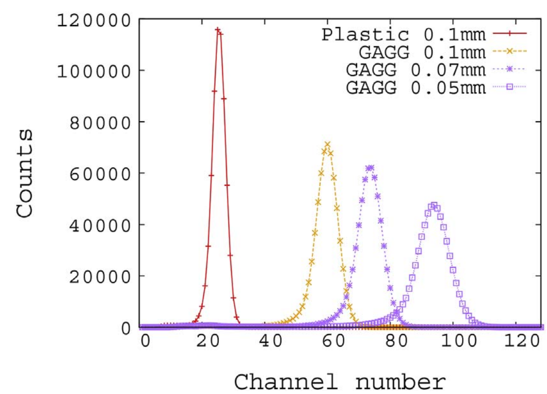

Fig. 5. Pulse height spectra of the three GAGG scintillators and the plastic scintillator for 5.5-MeV alpha particles. The gain and HV were adjusted to −67.0 V.

3.3. Monte Carlo simulation

Fig. 9 shows the calculated energy deposition per source for different thicknesses of the GAGG scintillators. 59.5-keV (^{241}Am) and 662-keV (^{137}Cs) gamma rays deposited very low amounts of energy compared to the beta particles. In case of the 0.5-mm-thick GAGG scintillator, the energy depositions per source were 6.3 keV for the 59.5-keV photons and 0.25 keV for the 662-keV photons. The energy depositions

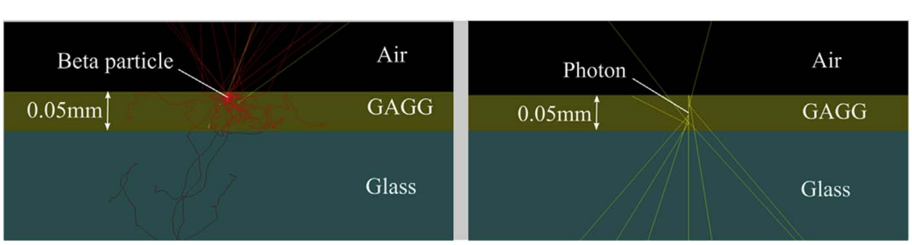

Fig. 4. Geometry of Monte Carlo simulation; irradiation of beta particle (a) and gamma photon (b) onto GAGG scintillator.

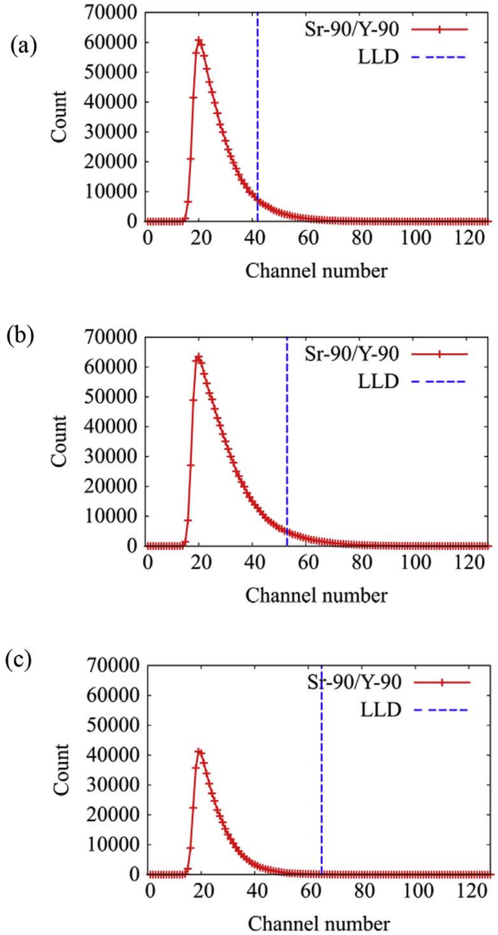

Fig. 6. Pulse height spectra of beta particles from ^{90}Sr/^{90}Y with 74 kBq; thicknesses of the GAGG scintillators are 0.1 mm (a), 0.07 mm (b), and 0.05 mm (c).

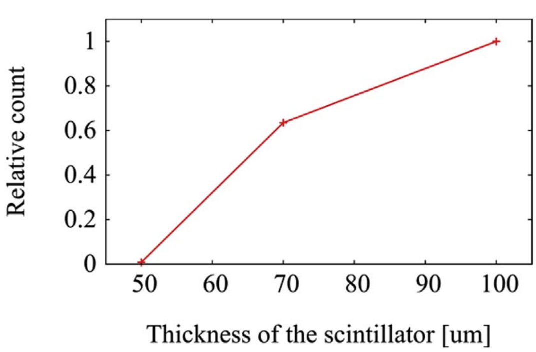

Fig. 7. Relative counts of beta particles from ^{90}Sr/^{90}Y for different thicknesses of the GAGG scintillators.

of the beta particles of ^{90}Y were 220–320 times higher than those of the 662-keV gamma rays. In all calculations, the energy deposition decreased with decreasing scintillator thickness. In the calculation of beta particles of ^{90}Y, the energy deposition of the 0.05-mm-thick GAGG was 0.47 times less than that of the 0.1-mm-thick GAGG.

3.4. Measurement of Rn progeny

Fig. 10 shows the alpha spectra of ^{241}Am and Rn progeny. In the spectrum of Rn progeny, a peak at the 92nd channel that is higher than the peak of 5.5 MeV (at 59th channel) was confirmed. This peak was ascribed to 7.7-MeV ^{214}Po alpha particles. Also, the peak at the 20th was ascribed to beta particles mainly from ^{214}Pb and ^{214}Bi.

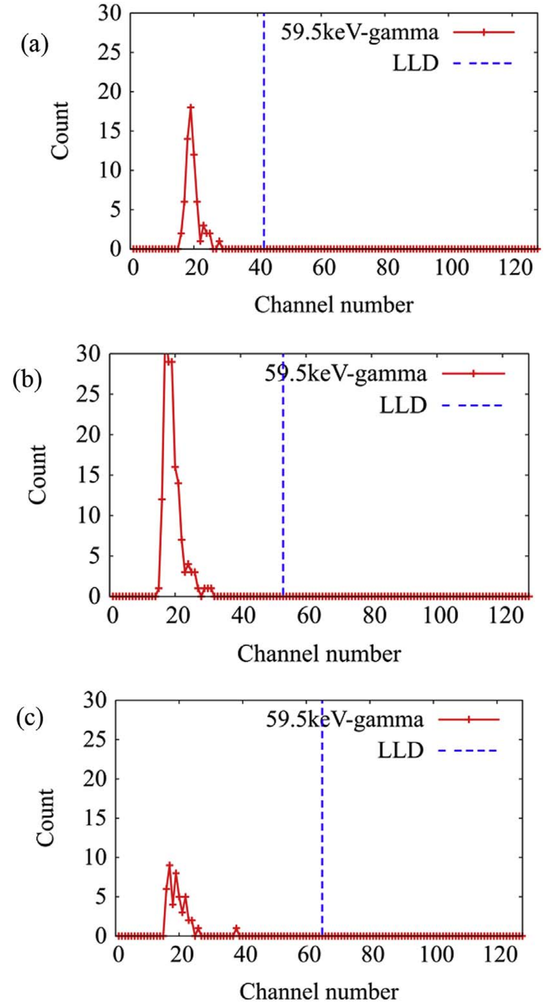

Fig. 8. Pulse height spectra of 59.5 keV gamma rays; thicknesses of GAGG scintillators are 0.1 mm (a), 0.07 mm (b), and 0.05 mm (c). Counts were confirmed to be below the LLDs in all GAGG scintillators.

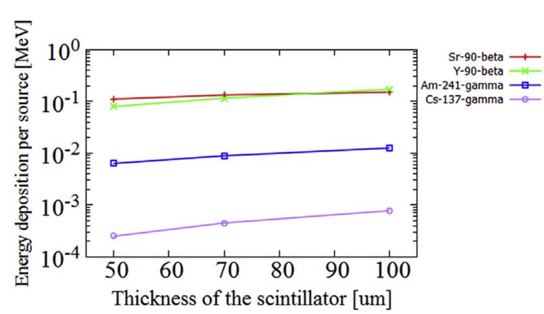

Fig. 9. Energy deposition per source for different thicknesses of GAGG scintillators irradiated with various radiation beams (beta particles from ^{90}Sr and ^{90}Y, and 59.5-keV and 662-keV gamma rays) calculated by Monte Carlo simulation.

4. Discussion

The energy resolution of 0.05-mm-thick GAGG for 5.5-MeV alpha particles (~11.6%FWHM) was the best among the three GAGG scintillators. This energy resolution was adequate to distinguish alpha particles of Pu isotopes (^{238}Pu with 5.5-MeV alpha particles and ^{239}Pu with 5.2-MeV alpha particle) from those of Rn progeny (mainly ^{214}Po with 7.7-MeV alpha particles). By reducing the thickness of the GAGG scintillator to 0.03 mm or 0.04 mm, more light would be transmitted from the scintillator, and the energy resolution might be improved

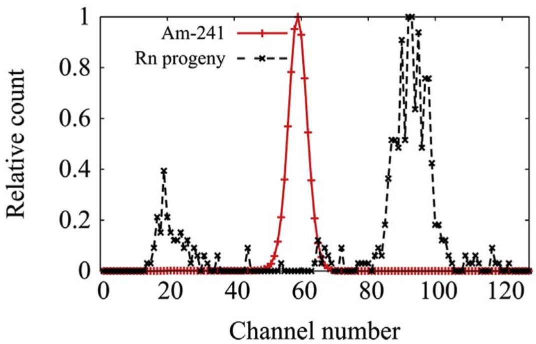

Fig. 10. The alpha spectra of ^{241}Am and Rn progeny measured with 0.05-mm-thick GAGG. In this measurement, HV for SiPM was reduced slightly from −67.0V so that the output signal was not saturated by the 7.7-MeV alpha particles from ^{214}Po.

further.

All GAGG scintillators used in this study were not sensitive to gamma rays with a dose rate of 1 mSv/h. At the site of the FDNPP, except in a reactor building, the maximum dose rate of gamma rays is ~1.4 mSv/h (Fukushima Daiichi NPS Survey Map), and at this rate, gamma rays would not impede alpha particle measurement.

The results of our experiment and the Monte Carlo simulation suggest that beta particles deposited higher energy than gamma rays. At the FDNPP site, there will be many beta particle emitters on the floor surface of buildings and in rubble. The detector must approach floor surfaces and rubble for alpha particle measurement, which is when it will be exposed to beta particles. Given that the beta count of the 0.05-mm-thick GAGG was only 1/100th that of the 0.1-mm-thick GAGG, it will be valuable for use in this field. By reducing the thickness of the GAGG scintillator to 0.03 mm or 0.04 mm, the beta count can be decreased further.

It is known that gamma rays and beta particles with high dose rate interact with the glass of the PMT and induce a scintillation, or Cherenkov light (Knoll, 2010). This would become a background count. We used SiPM (MPPC) as the photodetector, and it had no sensitivity to gamma rays and beta particles, even under high dose rates of beta and gamma rays. Therefore, SiPM is a promising photodetector in fields of high beta and gamma background. The 0.05-mm-thick GAGG scintillator with SiPM can be adapted to various devices to detect surface contamination of alpha emitters, such as an alpha survey meter, a hand and foot monitor, a dust monitor, and so on.

5. Conclusions

We evaluated the beta and gamma sensitivities of three GAGG scintillators with thicknesses of 0.05 mm, 0.07 mm, and 0.1 mm for alpha particle detection in a field of high beta and gamma background. The 0.05-mm-thick GAGG scintillator had the best energy resolution of 11.6% FWHM for 5.5 MeV alpha particles, and the lowest beta particle sensitivity (1/100th of the sensitivity of the 0.1-mm-thick GAGG). The alpha particle detector with the 0.05-mm-thick GAGG scintillator is promising for detecting plutonium contamination in a field with high beta and gamma background, such as the site of the FDNPP.

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