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Original Article

Efficient design of a ∅2×2 inch NaI(Tl) scintillation detector coupled with a SiPM in an aquatic environment

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A B S T R A C T

After the Fukushima accident in 2011, there has been increased public concern about radioactive contamination of water resources through fallout in neighboring countries. However, there is still no available initial response system that can promptly detect radionuclides. The purpose of this research is to develop the most efficient gamma spectrometer to monitor radionuclides in an aquatic environment. We chose a thallium-doped sodium iodide (NaI(Tl)) scintillator readout with a silicon photo multiplier (SiPM) due to its compactness and low operating voltage. Three types of a scintillation detector were tested. One was composed of a scintillator and a photomultiplier tube (PMT) as a reference; another system consisted of a scintillator and an array of SiPMs with a light guide; and the other was a scintillator directly coupled with an array of SiPMs. Among the SiPM-based detectors, the direct coupling system showed the best energy resolution at all energy peaks. It achieved 9.76% energy resolution for a 662 keV gamma ray. Through additional experiments and a simulation, we proved that the light guide degraded energy resolution with increasing statistical uncertainty. The results indicated that the SiPM-based scintillation detector with no light guide is the most efficient design for monitoring radionuclides in an aquatic environment.
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1. Introduction

The Fukushima accident in March 2011 has stimulated increasing public concern about radioactive rain through fallout of radioactive nuclides in neighboring countries. When a radioactive disaster occurs, securing water resources is an important issue that directly affects the national security of every country. Hence, in order to quickly cope with the inflow of radioactive materials into the water resources, artificial radioisotopes (especially ^{134}Cs, ^{137}Cs, and ^{131}I[1]) need to be measured in real time under water. In Korea, the standard monitoring method prescribed by the Ministry of Environment is defined as sample sampling and analysis using a high purity germanium detector (HPGe) after pre-treatment. However, it takes a few days for the analysis, which makes it difficult to quickly respond and detect artificial radioisotopes. To protect water resources from fallout, real-time measurement is necessary to detect artificial radioisotopes in situ.

For in situ detection system, we chose an inorganic scintillation detector with portability instead of HPGe. The thallium-doped sodium iodide (NaI(Tl)) scintillator has been generally used for its high light yield (45,000 photons/MeV), high density, large linear response, reasonable price, and mature manufacturing technique [1]. The NaI(Tl) scintillation detector requires a photodetector to read out photons. A photomultiplier tube (PMT) is the most popular photodetector. The PMT, however, has disadvantages: bulky volume, sensitivity to a magnetic field, and high operating voltage (about 1500 V). Particularly, the high voltage for operation can cause safety problems and requires a large battery, which in turn requires a large space. Therefore, the scintillation detector readout

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with the exisiting PMT is not suitable in an aquatic environment. Recently, a silicon photomultiplier (SiPM) has been intensively studied and widely used as a photodetector in many fields. The SiPM is composed of several thousand microcells connected in parallel. Each microcell contains an avalanche photodiode (APD) that operates in Geiger-mode and generates photoelectrons. With the APD, the SiPM has a high intrinsic gain (10^5-10^6) that is comparable to the PMT. In addition, it has many advantages over the PMT, such as compactness, the ability to be mass produced, low bias operation (about 25–30 V) and low sensitivity to magnetic force [2–7].

In this study, we implemented a SiPM in a ⌀2 × 2 inch NaI(Tl) scintillator designed to be as efficient as possible. Previously, a small scintillator made with a rectangular crystal coupled with a SiPM showed a good energy resolution of about 5–8% at 662 keV [8–10]. However, it had a significantly low detection efficiency due to its small volume. A dedicated detector to monitor radioisotopes under water requires a high detection efficiency for gamma rays due to the shielding effect of water so we decided to use a ⌀2 × 2 inch NaI(Tl) scintillator that was based on the specifications of a commercial product [11]. Some studies have reported a SiPM in a ⌀2 × 2 inch NaI(Tl) scintillation detector that could maintain detection efficiency [12,13]. These studies estimated performance of the SiPM-based NaI(Tl) scintillation detector using a light guide or a tapered head scintillation crystal. These scintillation detectors demonstrated an energy resolution comparable to that of a PMT-based scintillation detector. However, it was somewhat tricky to maintain the structure or manufacture a specific shape of scintillation crystal. To avoid difficult maintenance and manufacturing processes, we propose a direct coupling system, which consists of only two components, a commercial ⌀2 × 2 inch NaI(Tl) scintillator and a SiPM. The energy resolution measured by the proposed detector was estimated by comparing it with that of other detectors and the results of previous research. Linearity was also taken into account for the performance assessment. Furthermore, the light collection efficiency of SiPM-based detectors was computed by Monte Carlo simulation to analyze the experimental results.

2. Materials & methods

2.1. Configurations of the three types of detectors

Three systems for the gamma spectrometer were tested in an aluminum dark box at room temperature, as shown in Fig. 1. One was composed of a scintillator (Epic Crystal, ⌀2 × 2 inch NaI(Tl)) and a PMT (Hamamatsu, H7195); another consisted of a scintillator, an array of SiPMs (SensL, ArrayJ-60035-4P-EVB, 12 × 12 mm^2) and a light guide (PMMA, 10 mm and 30 mm); and the other system was a scintillator directly coupled with an array of SiPMs. The main parameters of the PMT and SiPM are presented in Tables 1 and 2, respectively. The parts where the components contacted each other were coated with optical grease (Saint Gobain, BC-630) to minimize loss of photons due to the difference in the reflective index. Light guides were made with heights of 10 mm and 30 mm. We fabricated two different heights for the light guide so we could estimate the effect of the reflective angle on detection. Polytetrafluoroethylene (PTFE) was used as a diffuse reflector on the surface of the light guide and a window of the scintillation crystal except for the part of coupling the SiPM to transmit photons generated in the scintillator to the photo sensor. A circuit for processing pulses obtained through the cells of the SiPM was designed at the Korea Advanced Institute of Science and Technology (KAIST, South Korea). Finally, the signals from the circuit were expressed as a spectrum through a shaping amplifier (ORETEC, model 673) and a multichannel analysis (ORTEC, TRUMP-PCI – 2K).

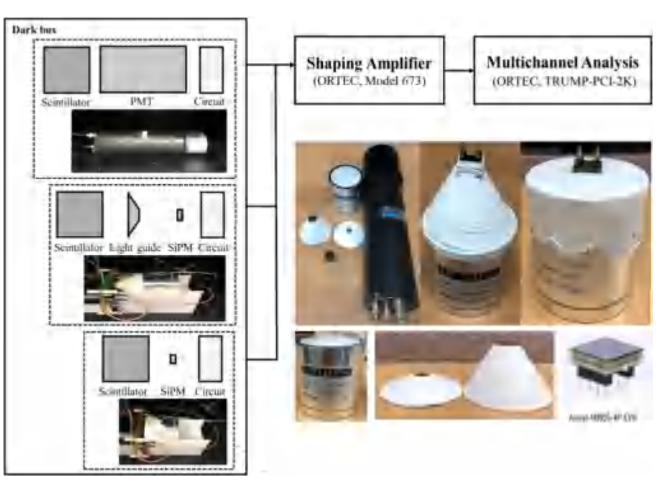

Fig. 1. Configurations of the gamma spectrometers used for the experiments.

2.2. Experimental setup

First, the optimum operational voltage of a SiPM needs to be determined because critical characteristics such as Photo Detection Efficiency (PDE), Dark Current Rate (DCR), Excess Noise Factor (ENF), Cross-talks, and after-pulses strongly depend on the voltage [4,5]. Using ^{137}Cs to measure the standard reference of energy resolution, the applied voltage with the best resolution was 28.5 V as shown in Fig. 2. After optimization, several sources were measured to estimate the energy resolution at various energy peaks and the linearity of the spectrometers. The distance between radionuclides and the scintillator was 15 cm. Details of the sources used in this work are described in Table 3. The operational voltage of PMT was set to 1450 V.

In fact, when a detector is set up under water, gamma-rays are incident at a scintillator in various directions. Energy resolution measured by the SiPM-based detector without a light guide may change according to where photons are generated in the scintillator because the active area of the SiPM is smaller than that of the ⌀2 × 2 inch scintillator. In contrast, a detector using a light guide may alleviate the dependence of energy resolution on the generating location of photons because the photons can eventually be transmitted to the photo sensor through the light guide regardless of where it occurred. To confirm these predictions, we measured ^{137}Cs at different locations at 30° intervals from 0° to 180°, as presented in Fig. 3.

Table 1
Main parameters of the PMT.

Manufacturer	Hamamatsu
Model	H7195
Built-in PMT	R329-02[a]
PMT diameter	51 mm (2 inch)
Photocathode Materials	Bialkali
Window Materials	Borosillicate
Typical Gain	3.0 x 10^6
Spectral range	300–650 nm (maximum sensitivity 420 nm)
Quantum efficiency	27%

[a] It is composed of the R329-02 type of PMT, which is used to continuously measure changes in environmental radiation due to its long term stability, low background noise, and good plateau characteristic [14].

Table 2
Main parameters of the SiPM.

Manufacturer	SensL
Model	ArrayJ-60035-4P-EVB
Number of cell	4 (2 x 2 array)
Active area/cell	6.07 x 6.07 mm^2
Microcell size	35 µm
Number of microcells	22,292
Microcell fill factor	75%
Typical gain	5.3 x 10^6
Sepctral range	200–900 nm (maximum sensitivity 420 nm)
Capacitance	4140 pF

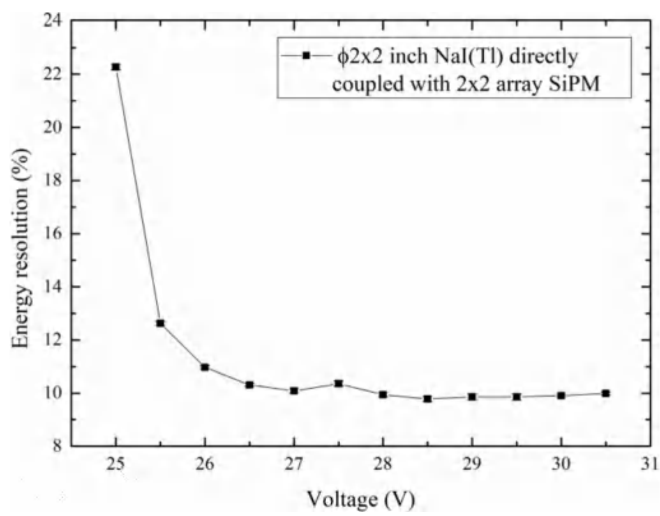

Fig. 2. Energy resolution of 662 keV gamma rays versus bias voltage for a ⌀2 × 2 inch NaI(Tl) scintillation detector with no light guide and without PTFE.

Table 3
Information on the radioisotope sources.

5. Radioisotope	^{57}Co	^{133}Ba	^{22}Na	^{137}Cs	^{54}Mn	^{60}Co
Activity (µCi)	0.02	0.62	7.31	43.26	1.82	0.38
Energy 1 (keV)	122	356	511	662	834.8	1173.2
Energy 2 (keV)	-	-	1274.5	-	-	1332.5

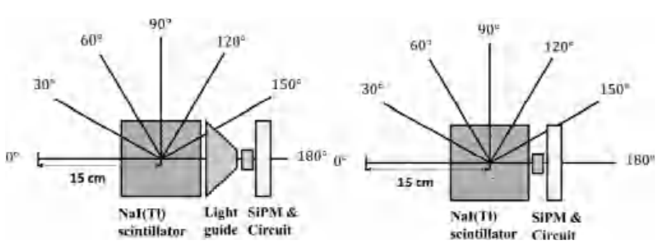

Fig. 3. Schematics of the ^{137}Cs position to vary the incident direction of photons to the scintillator.

2.3. Optical simulation of scintillation light

The role of a light guide is to transmit photons generated from a scintillator to the active area of a SiPM so that it has an influence on light collection efficiency in a detection system. To determine whether or not a light guide is helpful in transferring photons, we performed an optical simulation by a Geant4 application for tomographic emission (GATE) Monte-Carlo simulation. GATE is a recently developed simulation platform based on Geant4 physical models, specifically designed for PET and SPECT studies [15,16].

We tested the probability of collecting photons generated at various points in the scintillator. Fig. 4 shows a flow chart of the optical simulation. Geometries and materials were set to components used in the experiment. Fig. 5 shows the indicated positions of the optical source in the x-y and x-z cross section of the ⌀2 × 2 inch scintillator. Source files with different positions were generated by Python code. There were a total of 800 positions, which was composed of 80 positions in the x-y coordinate and 10 positions along the z-axis for each position of the x-y coordinate. The energy of a photon was set at 2.988 eV which corresponded to 415 nm, the most frequently emitted wavelength in the NaI(Tl) scintillator. After running the GATE, Python processed the GATE output files and computed the light collection efficiency (LCE) at each position.

3. Results and discussion

3.1. Energy resolution at normal incidence

The energy resolution measured by three detectors is shown in Fig. 6. We distinguished it as “bare” and “PTFE” depending on whether or not the surface of the light guide or the scintillator, except for the part coupling to the photo sensor, was coated with PTFE. “PTFE″ showed better energy resolution than “bare” for all cases of detectors. In particular, at above the 1200 keV energy peak, the energy resolution measured by “10 mm_bare” and “30 mm_bare” increased, which was not physical behavior. Because signals from a detector using a light guide without reflective material is very small, photopeaks were shown on a low range of channel. Overall spectrum including the photopeaks was concentrated in the lower channels so that the channel-to-channel fluctuation of the peaks was not noticeable [1]. This resulted in overlapping between 1173 keV and 1332 keV peaks and an increment of energy resolution at the 1332 keV peak for both “bare” detectors.

Among SiPM-based detectors, the “Direct_PTFE” had the best energy resolution at all energy peaks. It achieved a 9.76% energy resolution at 662 keV, while the detector composed of a scintillator, a SiPM and a 10 mm light guide coated with PTFE had a 11.32% energy resolution. Even the resolution of the “Direct_bare” was close to that of the “10 mm_PTFE”. Based on the simulation study in Ref. [12], there was no remarkable improvement in light collection efficiency at the optimal height of the light guide. In other words, even an SiPM-based detector that uses an optimized light guide does not perform better than one with a direct coupling system.

Fig. 7 shows the spectra of the ^{137}Cs source measured by each detection system. To keep the photoelectric peak at the same position in the channel, we adjusted the shaping amplifier gain. A backscattering effect was observed because of the aluminum dark

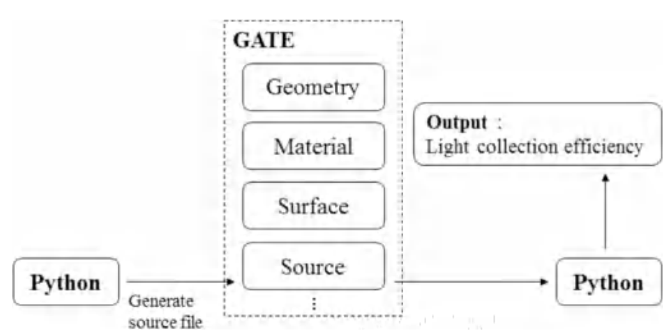

Fig. 4. A flowchart of the optical transport simulation.

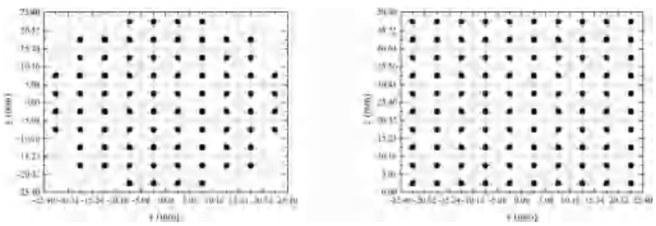

Fig. 5. Positions of optical sources in the x-y and x-z coordinates of the scintillator.

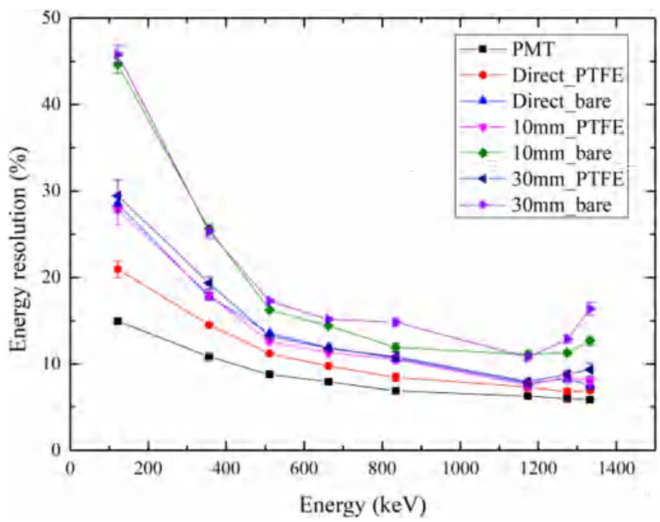

Fig. 6. Plot of the energy resolution versus the energy peaks for each detector.

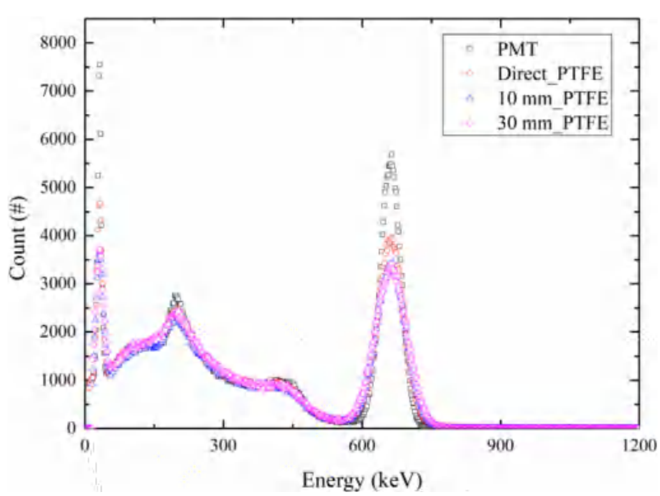

Fig. 7. Spectra of the ^{137}Cs source measured by each detection system.

box where the experiment was performed. Corresponding to the previous results, "Direct_PTFE" showed the best energy resolution among the SiPM detection systems, which had a less broadening Gaussian distribution.

Table 4 shows the energy resolution measured at 662 keV in this study and other studies [12,13]. The NaI(Tl) scintillator directly coupled with a SiPM had better results than all the other systems. Thus, the proposed detection system is the most efficient in energy resolution.

3.2. Linearity at normal incidence

Figs. 8–10 show the linearity of the SiPM-based scintillation detecotor. Linearity was tested according to the method described in Ref. [17]. Under the same conditions, several gamma-ray emitting nuclides were measured to find a channel of energy peaks with both an SiPM and a PMT. The excellent linearity of the PMT made it possible to relatively evaluate the linearity of the SiPM-based scintillation detector. The X-ray (32 keV) from ^{137}Cs was used for normalization of the PMT response to the SiPM response. The dashed line is the ideal line, where the SiPM response is fully proportional to the PMT response. A response is considered to be linear if the response of the SiPM does not deviate by more than 2% from the diagonal line.

In conclusion, all detectors had a nonlinear response in a range up to 1330 keV. For the detector using a light guide, the error was 6–8%. The SiPM-based detector with no light guide had only a 2–5% error. Although the direct coupling system had nonlinearity, it showed the possibility that its linearity can be easily improved by increasing the number of SiPMs. Based on these results, it was clear that the light guide was not helplful to transport photons to the SiPM.

3.3. Energy resolution according to the incident direction

Placing the ^{137}Cs in the positions shown in Fig. 3, the energy resolution was measured by each gamma spectrometer to check the effect of a light guide and the stability of the direct coupling system when the gamma ray was from various directions. Fig. 11 shows the energy resolution as a function of the position of the ^{137}Cs source. When the position of ^{137}Cs was near a circuit (when the angle was greater), the energy resolution was relatively degraded compared to that measured at 0°. This tendency was pronounced in the scintillation detector directly coupled with an SiPM, while the detector using the light guide showed less variance of energy resolution. However, there was no significant difference between the worst energy resolution of the directly coupled system and the best energy resolution of the detector with the light guide. As a result, the main factor inducing the degradation of the energy resolution was not the fact that the active area of the SiPM was relatively smaller than that of the scintillator, but the main factor was the implementation of the light guide in the detector.

3.4. Simulation of light collection efficiencies according to scintillation positions

Energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled with a photomultiplier or APD is described as

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2 + (\delta_n)^2 \qquad (1)$$

where δ_{sc} is the intrinsic resolution of a scintillation crystal, δ_p is the transfer resolution, δ_{st} is the statistical contribution of a PMT or photo diode and δ_n is the dark noise contribution connected with the detector's current and the noise of electronics [17,18].

Because two detectors with and without a light guide were composed of the same NaI(Tl) scintillator, SiPM, and signal processing circuit, we assumed that both δ_{sc} and δ_n were equal, and we could consider them negligible when comparing the energy resolution of each detection system. Then, we considered only δ_p and δ_{st} to reveal the effect of the light guide.

The transfer factor δ_p is described by the variation of probability that a photon generated in the scintillator results in the arrival of a photoelectron. The statistical uncertainty of the signal from the SiPM can be described as

Table 4
Energy resolution at 662 keV for each NaI(Tl) scintillation detector.

NaI(Tl) Scintillator	Light guide	Photodetector	Energy resolution at 662 keV (%)
∅2 × 2inch	-	PMT (∅2 × 2 inch)	7.94 ± 0.03
∅2 × 2inch	30 mm, PTFE	Array SiPMs (12 × 12 mm^2)	11.81 ± 0.03
∅2 × 2inch	10 mm, PTFE	Array SiPMs (12 × 12 mm^2)	11.32 ± 0.07
∅2 × 2inch	Direct, bare	Array SiPMs (12 × 12 mm^2)	11.91 ± 0.06
∅2 × 2inch	Direct, PTFE	Array SiPMs (12 × 12 mm^2)	9.76 ± 0.03
Experimental result in Ref. [12]			
∅2 × 2inch	17 mm, PTFE	Array SiPMs (12 × 12 mm^2)	14.11
Experimental result in Ref. [13]			
∅2 × 2inch	20 mm, PTFE	Array SiPM (12 × 12 mm^2)	11.7 ± 0.7
∅2 × 2inch	-	Array SiPM (12 × 12 mm^2)	10.2 ± 0.3
70. (Taperd head)			

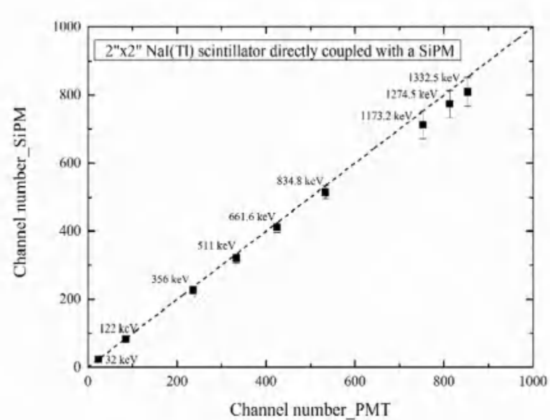

Fig. 8. Linearity of the 2 × 2 array SiPM-based ∅2 × 2 inch NaI(Tl) scintillation detector with no light guide.

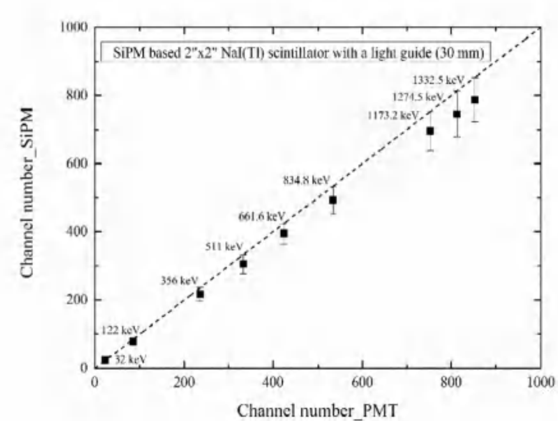

Fig. 10. Linearity of the 2 × 2 array SiPM-based ∅2 × 2 inch NaI(Tl) scintillation detector with a light guide (30 mm).

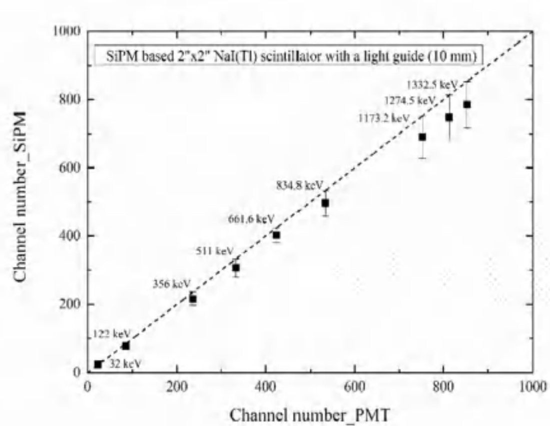

Fig. 9. Linearity of the 2 × 2 array SiPM-based ∅2 × 2 inch NaI(Tl) scintillation detector with a light guide (10 mm).

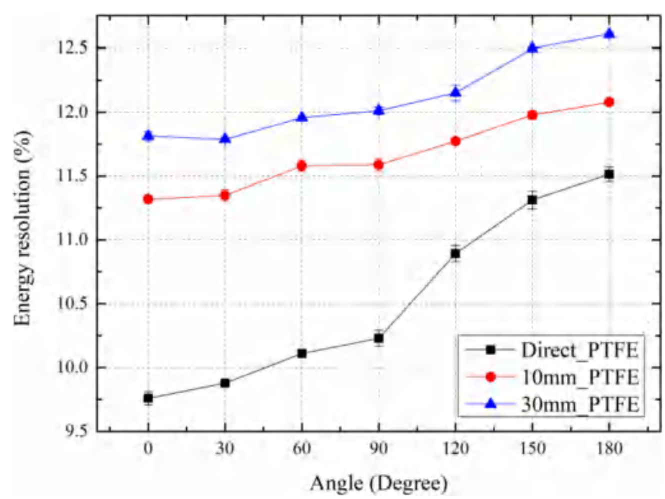

Fig. 11. Plot of energy resolution as a function of location of a 662 keV gamma-ray source expressed as the angle.

$$\delta_{st} = 2.355 \times (ENF/PHE)^{1/2} \quad (2)$$

where PHE is the number of photoelectrons and ENF is the excess noise factor.

Fig. 12 shows the relative pulse height obtained from SiPM-based scintillation detectors in the experiment and the optical simulation. The measured pulse height (i.e., the center channel of a photopeak) was obtained from the 662 keV photopeak. The light collection efficiency (LCE) can be converted to a pulse height by multiplying the gain because the pulse height is proportional to the number of photons collected in the sensor. The exact gain of the SiPM and PMT was unknown, but that of the SiPM was constant in all the SiPM-based detectors. Using the constant gain of the SiPM, we performed a comparison between the LCE of each SiPM-based

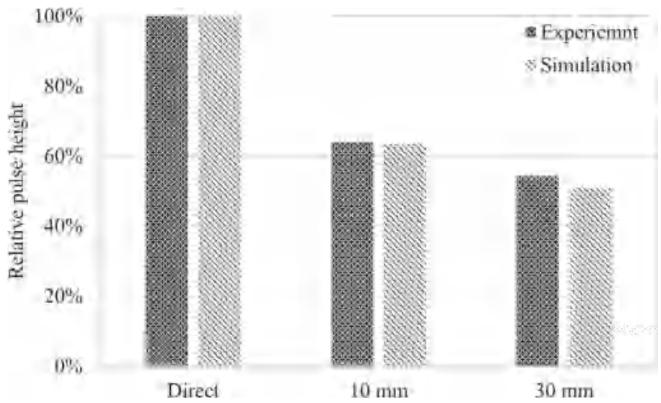

Fig. 12. Relative pulse height estimated from the measured 662 keV peak and the optical simulation.

detector and the measured pulse height. In the simulation the relative LCE obtained from the SiPM-based detector with a 10 mm light guide and the detector with a 30 mm light guide were respectively 63.4% and 50.9% of the LCE in the directly coupled system. The error between the experiment and simulation results was within about 6%.

The LCE at each location in different detectors is shown in Fig. 13. The mean value of each system was about 11.9%, 14.9%, and 23.5%. Considering the fact that approximately 30% of the light emitted by the crystal reaches the cathode of the PMT [19], these value were acceptable. Although the mean of the LCE at each point increasd as the length of the light guide shortened, the LCE achieved by the SiPM-based detector with no light guide was significantly higher. This result corresponded to the result of the experiment where the detector without the light guide achieved the best energy resolution at all photo peaks. It demonstrated that the light guide prevented the photons from transmitting to the sensor, which is not its intended role. On the other hand, the LCE of the SiPM-based scintillation detectors deviated depending on where the scintillation photons were generated in the NaI(Tl) crystal as described in Fig. 13. The deviation of the direct coupling system was higher than that of the detector using a light guide.

These findings revealed that the light guide improved the energy resolution by reducing $(\delta_p)^2$, the variance of efficiency at each location, but it finally degraded the resolution with an increasing $(\delta_{st})^2$, the statistical uncertainty, as described in Eq. (1). Table 5 shows the quantitative comparison of energy resolution between the direct coupling system and the detector using a light guide.

The transfer factor, δ_p was defined as the variance of LCE depending on where the photons were generated. It can be calculated based on the simulation results, the LCE of each 800 point within the scintillator. Looking back to Eq. (2), δ_{st} is composed of PHE (the number of photoelectrons) and ENF (excess noise factor). We considered the LCE as PHE because PHE is proportional to the LCE, and the ENF was omitted because the same SiPM was used in all SiPM-based detectors. Hence, the relative $(\delta_{st})^2$ is expressed as

$$\delta_{st} = 2.355 \times (1/\mathrm{LCE})^{1/2} \tag{3}$$

As with the qualitative discussion above, this quantitative comparison also indicated a trade-off effect of the light guide with respect to energy resolution. The SiPM-based detector with a light guide had a lower variance of LCE, δ_p than the direct coupling system. The low δ_p clearly improved the energy resolution according to Eq. (1). However, positive effect of the light guide with reducing δ_p was insignificant, given the statistical uncertainty. Implementation of the light guide caused the loss of photons, which negatively influenced on energy resolution with increasing the statistical error. This corresponded to the fact that the transfer factor δ_p is negligible compared to the other components of the energy resolution in the modern scintillation detectors [20].

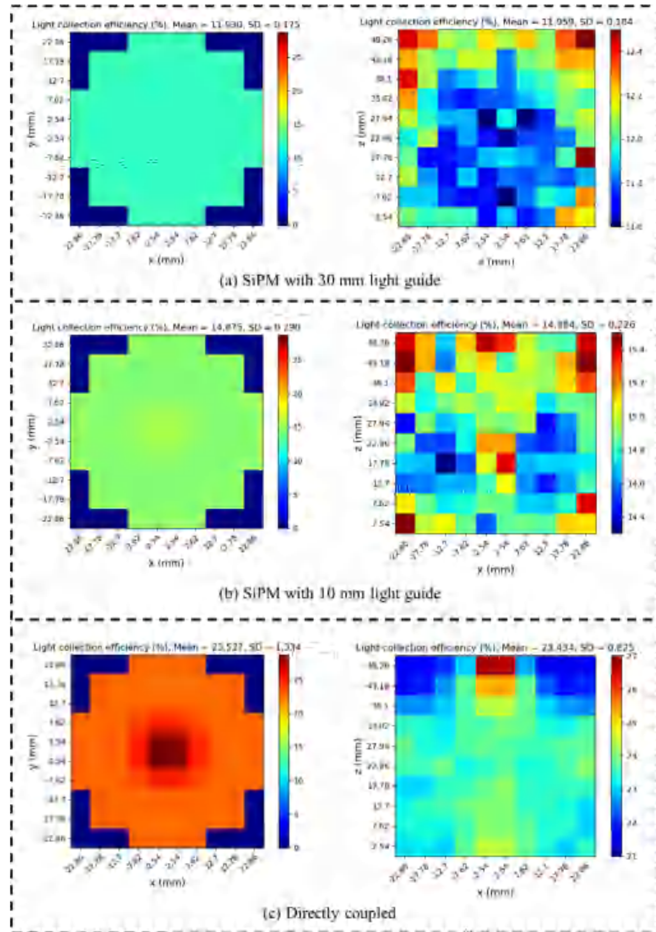

Fig. 13. Simulation results of light collection efficiency in both the x-y and x-z coordinates for each detector: (a) SiPM-based detector using a 30 mm light guide. (b) SiPM-based detector using a 10 mm light guide. (c) SiPM-based detector without a light guide.

4. Conclusions

This paper reported our study on the design of a gamma spectrometer composed of a ⌀2 × 2 inch NaI(Tl) scintillator and a SiPM to be applied in an environment with limited weight and power supply where it is difficult to use a traditional detector using a PMT. Among the SiPM-based scintillation detectors, the one with direct coupling to the SiPM had the best energy resolution (~9.76% for a 662 keV gamma-ray). The response of the ⌀2 × 2 inch NaI(Tl) scintillation detector without a light guide was less nonlinear than the detector with the light guide. This study confirmed that the light guide had a negative effect on photon transmission. To thoroughly understand the effect of the light guide and the behavior of photons in the scintillator and light guide, we performed an optical simulation with the GATE. The simulation result indicated that the light guide reduced the variation in light collection efficiency at each point, which improved the energy resolution. However, it prevented photons from arriving at the SiPM. Loss of photons

Table 5
Relative energy resolution of SiPM-based scintillation detectors based on Eq. (2).

Type of detector	Mean of LCE	Deviation of LCE	Variance of LCE	Transfer factor, $(\delta_p)^2$	Statistical factor, $(\delta_{st})^2$	Relative energy resolution, $(\Delta E/E)^2$
Direct	0.235	1.84E-02	3.38E-04	1.14E-07	23.60	23.60
10 mm	0.149	4.19E-03	1.75E-05	3.08E-10	37.22	37.22
30 mm	0.119	2.27E-03	5.16E-06	2.66E-11	46.61	46.61

eventually degraded the energy resolution by increasing the statistical uncertainty. Hence, the ∅2 × 2 inch NaI(Tl) scintillation detector directly coupled with the SiPM was the most efficient for use in an aquatic environment.

Acknowledgments

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