Performance comparison of scintillators for alpha particle detectors

Yuki Morishita [1) 2)], Seiichi Yamamoto [1)], Kenji Izaki [2)], Junichi H. Kaneko[3)], Kohei Toi[3)], Youichi Tsubota[3)], and Mikio Higuchi
[1)] Nagoya University Graduate School of Medicine, [2)]Japan Atomic Energy Agency, [3)]Hokkaido University Graduate School of Engineering

Abstract—Scintillation detectors for alpha particles are often used in nuclear fuel facilities. Alpha particle detectors have also become important in the research field of radionuclide therapy using alpha emitters. ZnS(Ag) is the most often used scintillator for alpha particle detectors because its light output is high. However, the energy resolution of ZnS(Ag)-based scintillation detectors is poor because they are not transparent. A new ceramic sample, namely the cerium doped $Gd_2Si_2O_7$ (GPS) scintillator, has been tested as alpha particle detector and its performances have been compared to that one of three different scintillating materials: ZnS(Ag), GAGG and a standard plastic scintillator. The different scintillating materials have been coupled to two different photodetectors, namely a photomultiplier tube (PMT) and a Silicon Photo-multiplier (Si-PM): the performances of each detection system have been compared. Promising results as far as the energy resolution performances (10% with PMT and 14% with Si-PM) have been obtained in the case of GPS and GAGG samples. Considering the quantum efficiencies of the photodetectors under test and their relation to the emission wavelength of the different scintillators, the best results were achieved coupling the GPS with the PMT and the GAGG with the Si-PM

I. INTRODUCTION

Scintillation detectors for alpha particles are often used in nuclear fuel facilities. In these facilities, ZnS(Ag) scintillation detectors are most often used because they have high light output [1] and they can sustain the highly corrosive and humid atmosphere. Because the scintillation efficiency of ZnS(Ag) is about the same as that of NaI(Tl) [2], the light yield of ZnS(Ag) would be 38000 photons/MeV. The scintillation detectors that employed ZnS(Ag) have high stability [3]. However ZnS(Ag)-based scintillators are not the best detectors to be used, because they are quite opaque and their energy resolution is very bad. Thus the energy information of ZnS(Ag) scintillation detectors cannot be used for energy spectroscopy [4-5]. For the energy spectroscopy of alpha emitters, silicon (Si) semiconductors are often used [6]. However they are much more expensive than scintillation detectors and sensitive to noise since their signal level is small.

Another growing research field of alpha particle detectors is radionuclide therapy. In radionuclide therapy, biological effects are obtained by the energy absorbed from the beta or alpha particles emitted from the radionuclides that are administered to cancer patients. For such therapy, alpha autoradiography is often used, to reconstruct the image of the relative concentration of radioactive material present within the biologic specimen or the human body part. Generally scintillators are used for this purpose. A ZnS(Ag) screen was used to produce the image of the distribution of the alpha emitter in animal slices [7]. A transparent scintillator would surely improve the spatial resolution of alpha autoradiography.

Currently, a thin plastic scintillator is used as a transparent scintillator for alpha spectroscopy and for imaging alpha particles [8]. However, the light output of plastic scintillators is relatively small (10000 photons/MeV) [2], limiting both the energy and spatial resolutions in imaging systems. A new transparent scintillator with high light output is desired for alpha particle detectors.

Recently, a cerium-doped $Gd_2Si_2O_7$ (GPS) has been developed that is suitable for alpha particle detectors because it has high light output to alpha particles [9-10] and improved performance [11]. Because light yield of GPS was reported to be 4.4 times higher than that of BGO scintillator [10] and the light yield of the BGO is 8200 photons/MeV [2], the light yield of the GPS would be about 36000 photons/MeV. We tested four kinds of scintillators for alpha particles including a GPS coupling them with photodetectors and evaluated the best combinations in term of response to alpha particles.

II. MATERIALS AND METHODS

Table 1 summarizes the scintillators used for our performance evaluation. We selected ZnS(Ag) and a plastic scintillator (NE-102, Ohyo Koken, Japan) as conventional scintillators for alpha particles and compared their performances with GPS. We also selected a new scintillator, Ce-doped $Gd_3Al_2Ga_3O_{12}$ (GAGG) that has high light output and high energy resolution for gamma photons [12-13]. CsI(Tl) is a possible scintillator for alpha particles. Although CsI(Tl) is much less hygroscopic than NaI(Tl), it is slightly hygroscopic and degrades after exposure to humid air. Sometimes alpha particle scintillation detector is continuously used in harsh conditions such as dust and exhaust monitoring for a nuclear facility. Therefore, CsI(Tl) was not considered as a candidate of the scintillators for performance evaluation. All the scintillators were 18 mm x 16 mm. The GPS and GAGG were 0.1 mm thick because it is the current, technically minimum thickness. To increase the mechanical strength, these scintillators were optically coupled to 1-mm thick transparent acrylic resin.

Table 1 Scintillators for performance evaluation

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Size (mm)	18 x 16	18 x 16	18 x 16	18 x 16
Thickness (mm)	0.1	0.05	0.1	0.05

Figure 1 shows a photograph of these scintillators. The plastic scintillator and GAGG (Furukawa Company, Japan) were transparent. ZnS(Ag) (Ohyo Koken Japan) was not transparent because it was made of white powder. The GPS was a ceramic scintillator made by Hokkaido University that was partly transparent with some irregular white patterns on it.

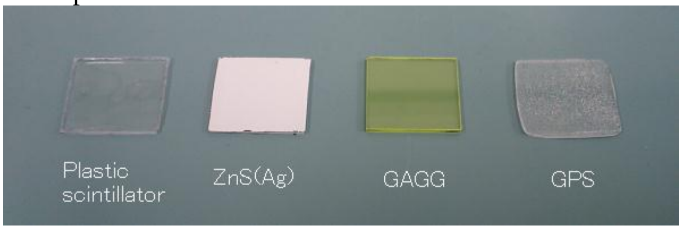


Fig. 1 Scintillators for performance evaluation

Figure 2(a) shows a schematic drawing of the coupling configuration of the thin scintillators with a PMT, which is 3-inch round high quantum efficiency (HQE) photomultiplier tube (Hamamatsu Photonics R6233-100 HA). A HQE PMT was chosen because besides the better light yield, a better energy resolution was observed for gamma-rays as shown in [14]. Each sample among the four scintillators listed in Table 1 was coupled to the PMT. The scintillator part was on the upper side, and the aluminized Mylar was covered on the scintillator as a reflector. A 5.5 MeV alpha source (Am-241, 500 Bq) was positioned on the aluminized Mylar to irradiate the 5.5-MeV alpha particles. High voltage for PMT was fixed to -750 V and the acquisition time was 600 s. The PMT signals were fed to a standard NIM module and a multichannel analyzer (ADC Model 1125P, Clear-Pulse Co., Tokyo). The energy resolution was evaluated by Gaussian fitting software in the MCA. The measurement was conducted five times to evaluate the standard deviation. We evaluated the relative light output of the scintillators by a ratio of the peak channel to that of the plastic scintillator (plastic scintillator=1).

Figure 2(b) shows a schematic drawing of the coupling to a Si-PM: a multi-pixel photon counter (MPPC) array from Hamamatsu Photonics S11064-025P. Since Si-PMs have advantages as far as magnetic field operation with respect to PMTs, they are commonly used in medical applications [15-16]. Si-PM is also suitable for alpha-particle detectors [17]. One of the scintillators in Table 1 was optically coupled to a 3-mm thick light guide and to Si-PM. The light guide distributed the scintillation light. The aluminized Mylar was placed on the scintillator as a reflector. The Am-241 alpha source, which is the same as in the PMT evaluation, was positioned on the aluminized Mylar. High voltage for Si-PM was fixed to -71 V, and the acquisition time was 1800 s. The signals from the 4x4 channels of the Si-PM array were read out by 16 coaxial cables and fed to high-speed amplifiers whose outputs were fed to the weighted summing amplifiers and digitized by 100-MHz free running A-D converters in the data acquisition system. The digitally calculated position and energy data were accumulated in the memory and transferred to a personal computer (PC). The data acquisition system is identical as a previously reported one [17]. Finally, 2-dimensional distribution and energy spectrum were simultaneously acquired. As with PMT, we conducted five measurements and evaluated the standard deviation. Energy resolution and relative light output have been obtained with the same procedure as in the previous case.

Since the quantum efficiency (QE) distributions of PMT and Si-PM are different, we evaluated the relation between the luminescence spectra of the scintillators and the QE distribution of the photodetectors. The spectral response range of PMT was from 300 to 650 nm, and the peak sensitivity wavelength was 350 nm [14]. The spectral response range of Si-PM was 320 to 900 nm, and the peak sensitivity wavelength was 440 nm. The luminescence spectra of the scintillators listed in Table 1 were measured with a spectrofluorometer (JASCO FP-6500 Series, Tokyo). The excitation light wavelengths of these scintillators were selected as 290 nm, on the basis of a photoluminescence excitation/emission (PLE) data map to obtain luminescence emission spectra. To evaluate the theoretical light outputs for our experiments, first, integral values were calculated by multiplying each QEs of Si-PM and PMT by emission intensity of each scintillators at every wavelength. Then, a ratio was calculated by dividing integral value of Si-PM by that of PMT. Then the measured ratio of Si-PM/PMT and the calculated ratio of Si-PM/PMT were compared.

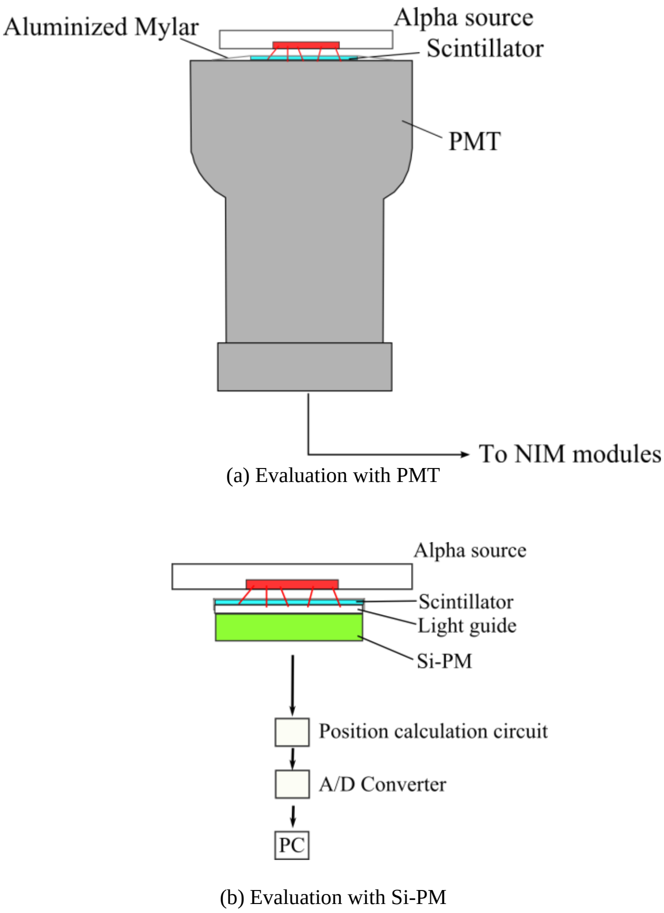


Fig. 2 Schematic diagram of performance evaluation system: PMT (a) and Si-PM (b)

III. RESULTS

3.1 Energy resolution

The energy spectra of the scintillators for the 5.5-MeV alpha particles using PMT and Si-PM are shown in Figs. 3(a) and (b). The energy resolutions of the four scintillators and standard deviations are shown in Table 2.

When we optically coupled the scintillators to the PMT, the energy resolution of GAGG (~8.4%) was the best among the four scintillators. The energy resolution of ZnS(Ag) (~42.8%) was the worst among the four scintillators. When we optically coupled the scintillators to the Si-PM array, the energy resolution of GAGG (~13.0%) was still the best among the four scintillators. The relative standard deviations of five times were less than 10%. The measurement error was small enough.

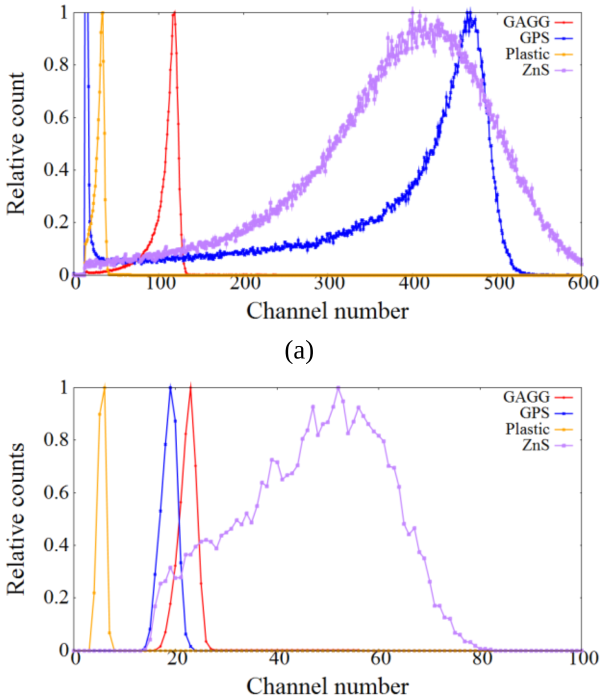
3.2 Relative light output

The relative light outputs with respect to the standard plastic sample are shown in Table 3. In the test performed coupling the samples to the PMT, the GPS relative light output resulted to be the highest: up to 14 times the light yield of the plastic scintillator. GAGG's light output was worse than GPS and ZnS(Ag), although its energy resolution was the best.

When we optically coupled the scintillators to the Si-PM, the light output of GPS was 3.5 times better than that of the plastic scintillator but worse than ZnS(Ag) and GAGG. The GAGG relative light output was, on the contrary, slightly higher when using the PMT coupling.

3.3 Relation between luminescence spectra of scintillators and QE distribution of photodetectors (wavelength matching)

The QE distributions of PMT and Si-PM are shown in Fig. 4, which also shows the luminescence emission spectra of the scintillators listed in Table 1. The emission wavelength of GPS is relatively shorter than that one of ZnS(Ag) and of the NE102. On the contrary, the emission wavelength of GAGG is relatively longer than that one of ZnS(Ag) and the plastic scintillator. The emission wavelengths of ZnS(Ag) and the plastic scintillator are closer. Table 4 shows the ratio of the integral value (Si-PM to PMT). The ratio of the calculated areas below 1 means that the GPS matches better the PMT QE spectrum than that one of the Si-PM, while the contrary is true for the GAGG, for which the calculated ratio is 1.88.



(a)

(b)

Fig. 3 Energy spectra of scintillators for 5.5-MeV alpha particles: PMT (a) and Si-PM (b)

Table 2 Energy resolution of four scintillators with PMT and Si-PM

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Energy resolution with PMT (%FWHM)	9.7±0.6	42.8±1.8	8.4±0.5	16.1±1.6

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Energy resolution with Si-PM (%FWHM)	13.8±0.3	43.6±1.0	13.0±0.3	24.2±0.5

Table 3 Relative light output of four scintillators with PMT and Si-PM

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Relative light output with PMT	14.0	12.7	3.6	1

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Relative light output with Si-PM	3.5	10.4	4.6	1

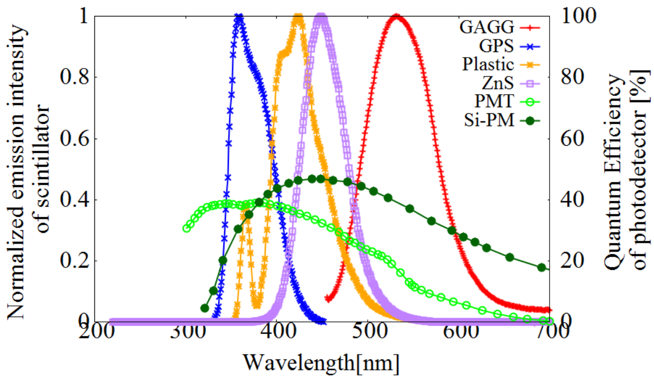


Fig. 4 QE distributions of PMT and Si-PM and distributions of scintillation wavelengths of four scintillators

Table 4 Ratio of integral value (Si-PM/PMT)

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Calculated ratio (Si-PM/PMT)	0.72	1.15	1.88	1.0

	GPS	ZnS(Ag)	GAGG	Plastic scintillator
Measured ratio (Si-PM/PMT)	0.25	0.82	1.28	1.0

IV. DISCUSSION

The energy resolutions of GAGG with both PMT and Si-PM were the best among the four scintillators. This is because it is transparent with a single-crystal structure so its crystal uniformity is good and the loss of scintillation light in the scintillator is small. The energy resolution of GPS was the second best among the four scintillators: slightly inferior to GAGG. The GAGG and GPS structures are different. GAGG is a single-crystal structure. In contrast, since GPS has scintillator grains fixed on a glass plate [10], the scintillation light may be inflected or refracted on the border of the scintillator grains, causing a loss of scintillation light in the scintillator. Even though GPS suffered a loss of scintillation light in the scintillator, the energy resolution of GAGG and GPS showed no significant difference. This result may induce to think that a GPS with a single-crystal structure will improve its performance in terms of energy resolution.

The energy resolution of ZnS(Ag) was poor (with PMT: ~42.8%, with Si-PM: ~43.6%) because it is not transparent and the scintillation light is partly absorbed in the scintillator. ZnS(Ag) is therefore not suitable for alpha spectroscopy measurement. The energy resolution of the plastic scintillator (with PMT: ~16.1%, with Si-PM: ~24.2%) confirmed standard results, which are better than the ZnS(Ag), because it is transparent, but its performance was anyhow much worse than that one of GPS and GAGG.

Concerning the relative light output, GPS was 14 times better than the plastic scintillator with PMT, while only 3.5 times higher than the plastic response, when coupled with Si-PM. The discrepancy is due to the difference between the emission spectrum of GPS and the QE distributions of the photodetectors. Since the GPS wavelength was relatively short, the wavelength matching to the tested Si-PM is not optimized. On the contrary, the wavelength matching to used PMT was good. When the GPS scintillator is used as an alpha particle detector, a combination of GPS and PMT is better because the PMT has QE distributions more suitable for relatively short wavelength (about 300 to 400 nm). Moreover, it is likely that using a GPS with a single-crystal structure a larger light output could be also obtained together with an improved energy resolution.

V. CONCLUSIONS

We evaluated the performances for alpha particles of four scintillators: GPS, GAGG, ZnS(Ag), and a standard plastic scintillator. GPS and GAGG resulted to be the most promising scintillators as alpha particle detectors among the four scintillators because their energy resolutions were good: ~10% with PMT, ~14% with Si-PM. For the relationship among the QE distributions of the photodetectors and the distribution of the scintillation wavelengths, the best combinations among these four scintillators and two types of photodetectors are GPS coupled to PMT and GAGG to Si-PM. The alpha particle detectors with the best combinations will be suitable for energy spectroscopy for alpha emitters in nuclear fuel facilities and for autoradiography for radionuclide therapy.

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