Development of scintillator plates with high energy resolution for alpha particles made of GPS scintillator grains

Takehiro Shimaoka ^{a*}, Junichi H. Kaneko ^a, Kenji Izaki ^b, Youichi Tsubota ^a,
Mikio Higuchi ^a, Shusuke Nishiyama ^a

^a Hokkaido University, Kita 13, Nishi8, kita-ku, Sapporo, Hokkaido, 060-8628, Japan

^b Japan Atomic Energy Agency, Muramatsu 4-33, Tokai-mura, Ibaraki, 319-1194, Japan

Keywords: Alpha particle measurement, Scintillator plate, GPS:Ce, Energy resolution

*Corresponding author

ABSTRACT

A scintillator plate with high energy resolution was developed to produce an alpha particle monitor used in nuclear fuel reprocessing plants and mixed plutonium-uranium oxide (MOX) fuel plants. Grains of a Gd₂Si₂O₇ (GPS) scintillator of several 10 to 550 μm were fixed on a glass substrate and were then mechanically polished. By increasing the size of scintillator grains and removing fine powders, the collected light yield and energy resolution for alpha particles were drastically improved. Energy resolution of 9.3% was achieved using average grain size of 91 μm. Furthermore, the ratios between counts in a peak and total counts were improved by more than 60% by the further increase of grain size and adoption of mechanically polished surfaces on both sides. Beta and gamma ray influences were suppressed sufficiently by the thin 100 μm scintillator plates.

1. Introduction

In nuclear fuel reprocessing plants and mixed plutonium–uranium oxide (MOX) fuel plants, contamination control for plutonium and related radioactive nuclides is conducted according to law. Related laws prevent internal and external exposure of workers and also prevent the release of radioactive nuclides to the external environment. An alpha particle monitor, an important early warning instrument used for contamination control, measures alpha particles continuously from dust in workplace air that is collected using a paper filter [1, 2, 3]. For this measurement, alpha particles from plutonium, of which the energy is 5.5 or 5.1 MeV, must be identified in background alpha particles from radon progeny nuclides such as 7.7 MeV alpha particles from ²¹⁴Po.

In general, a silicon surface barrier semiconductor detector is chosen whenever high-energy resolution is necessary, e.g., contamination monitoring of workplace air. However, silicon surface barrier semiconductor detectors, which have poor endurance, are also easily affected by external noise. For example, a leakage current was increased by erosion on a surface contact by humidity and a weak acidic atmosphere. In addition, it had been affected by radiofrequency noise from a high-frequency sealing machine that is widely used in the workplace. They caused the issue of evacuation orders from the worksite.

To improve problems related to silicon surface barrier semiconductor detectors, Izaki et al. applied a ZnS:Ag

scintillator for this instrument. A ZnS:Ag scintillator is used in exhaust gas monitors for air from glove boxes in which plutonium is handled. For this instrument, the influence of alpha particles from radon progeny nuclides is negligible. Nevertheless, they measured alpha particles from plutonium in existence of radon progeny nuclide by optimizing the ZnS:Ag scintillator thickness. However, room for improvement remains in energy discrimination ability compared with a silicon surface barrier semiconductor detector [4].

Although a ZnS:Ag scintillator has large light yield, its energy resolution for alpha particles is several tens of percent because it consists of semi-transparent powders. This problem is solvable using a thin transparent single-crystal ZnS:Ag scintillator. However, crystal growth of a single-crystal ZnS:Ag requires a special technique. It is difficult to obtain a commercially available single-crystal ZnS:Ag scintillator.

As another approach for alpha particle measurement using a powder scintillator, Yasuda et al. reported a phoswich detector composed of powder YAlO₃ (YAP) and Y₃Al₅O₁₂ (YAG) single-crystal scintillators [5]. In their development, a YAP scintillator was adopted by its fast decay time of 27 ns. A single-crystal YAP scintillator had high-energy resolution of 5.7% for 662 keV gamma rays [6]. However, the powder YAP scintillator had similar poor energy resolution to that of a conventional ZnS:Ag scintillator.

As a conjecture from the YAP powder scintillator application, a key to improving the energy resolution for alpha particle is how to collect scintillation light effectively using a light detector. One method is the use of a large grain size scintillator compared with conventional powder scintillator. Janecek et al. reported correlation between increased grain size of scintillator and increased collected light yield for X-rays using several scintillators: Lu₂SiO₅, Bi₄Ge₃O₁₂, YAlO₃, and CsBa₂I₅ [7]. Fukabori et al. reported similar results with increased grain size of Y₂O₃ ceramic scintillator for alpha particles [8].

As described in this paper, to resolve these issues on an alpha particles monitor for contamination of workplace air, energy resolution of a scintillator plate was improved using the following method: scintillator grains of several tens to several hundred micrometers were fixed on a glass substrate by adhesive. Then they were polished mechanically. In addition, gadolinium pyrosilicate (Gd₂Si₂O₇:Ce, GPS:Ce) scintillators with light yield two times greater than that of the YAP scintillator were adopted.

2. Experimental

2.1. Benefits of the GPS scintillator and growth using TSSG

A GPS scintillator has several benefits including high-energy resolution of 4.6% for 662 keV gamma rays, high density of 5.5 g/cm³, fast decay time of 50–100 ns, and non-deliquescent and non-self-radioactivity [9, 10, 11]. Actually, GPS with low cerium concentration has light yield that is 4.4 times higher than that of a conventional BGO scintillator [12, 13]. Moreover, gadolinium, a main composite element of GPS, has the largest capture cross-section among thermal neutrons. An internal conversion electron is emitted from the ¹⁵⁵Gd or ¹⁵⁷Gd nuclei after neutron capture [14, 15]. Therefore, the GPS scintillator is expected to become an excellent neutron scintillator with high detection efficiency [16, 17, 18, 19].

In this study, a GPS scintillator was obtained using top seeded solution growth (TSSG). As starting materials, Gd₂O₃, SiO₂, and CeO₂ powders were used. The former two powders were pure grade of 99.999%. The latter

powder was pure grade of 99.99%. These materials were mixed at compositions of $(\text{Gd}_{0.975}\text{Ce}_{0.025})_2\text{Si}_2\text{O}_7$ (2.5% of cerium concentration) and then sintered at 1650°C for 6 hr in air. The sintered material was set into an iridium crucible in a radio frequency Czocharlski furnace (TCH-05; Techno Search Corp.).

Typically, a 20-mm-diameter, 30-mm-long GPS polycrystalline is grown through 100 hr synthesis. The GPS polycrystalline was composed of single-crystal grains with size of several millimeters. For this growth, a typical growth rate of 0.3–0.5 mm/h and rotation rate of 10–20 rpm were adopted [12].

2.2. Scintillator plate fabrication

The GPS polycrystalline was ground and screened using sieves. GPS scintillator grains were classified in eight sizes presented in Table 1. The classified GPS scintillator grains were fixed on glass substrates with sizes of $25 \times 25 \times 1.25$ mm using adhesive (Blanchard Wax; J.H. Young Co. Inc.). Then they were polished mechanically using wet abrasive papers. Eight samples were fabricated to evaluate the influence of grain size on energy resolution.

Previous to the fabrication, ranges of alpha particles in a GPS scintillator were calculated based on ranges in silicon and the Bragg–Kleeman rule. The calculated ranges were 16 μm and 26 μm , respectively, for 5.5 and 7.7 MeV alpha particles. Consequently, the required minimum thickness for a GPS scintillator plate was ca. 30 μm . Although the thin scintillator gave the benefit of reduction of gamma rays and beta rays, 100 μm thickness was adopted for this study with the objective of fabrication accuracy. When the scintillator thickness became thinner than 100 μm , non-uniform thickness occurred gradually.

Figure 1 presents an optical photograph of an example of GPS scintillator plates and a conventional ZnS:Ag scintillator of 10 mg/cm² in mass thickness. In this image, the scintillator surfaces were settled on school seals of Hokkaido University printed on paper. Although the school seal is visible through the GPS scintillator plate, it was not visible through the ZnS scintillator.

2.3. Measurement system

To evaluate the response for alpha particles, a photomultiplier tube (H1161; Hamamatsu Photonics K.K.), a delay line amplifier (460; Ortec), a pulse stretcher (542; Ortec), and a multi-channel analyzer (WE7562; Yokogawa Electric Corp.) were used. The integration time constant of the delay line amplifier was set at 250 ns, considering the decay time of scintillators. A scintillator plate was fixed on the photomultiplier tube using optical grease. An ²⁴¹Am alpha source was set on a copper collimator. Alpha particles were injected into the scintillator plate through a hole ($\phi 1.5 \times 0.1$ mm thick). The irradiation area and the distance between the alpha source and the scintillator were fixed.

In addition, the influences of gamma rays and beta rays on the scintillator plates were evaluated using the same measurement system. A ¹³⁷Cs and a ⁹⁰Sr radioactive source were used, respectively, in response function measurements for gamma rays and beta rays. Distances between the radioactive source and a scintillator plates were 15 cm for gamma rays and 2 mm for beta rays. For beta-ray measurements, a lead collimator of $\phi 1.5 \times 2.0$ mm was used. All measurements for radiation sources were conducted in air at room temperature.

3. Experimental results and discussion

3.1. Influence of grain sizes, removal of fine powder, and surface polishing on alpha particle spectra

Figure 2 presents an example of energy spectra for 5.5 MeV alpha particles obtained using a YAP single-crystal scintillator ($10 \times 10 \times 1$ mm; Oken Co. Ltd.) and a GPS single-crystal ($10 \times 12 \times 1.2$ mm) grown using the same conditions as those used for this study. The GPS single-crystal showed a light yield that was two times higher than that of the YAP single crystal.

Figure 3 presents a change of spectra for the alpha particles obtained using GPS scintillator plates made of several different average grain sizes up to $91 \mu\text{m}$. With increased grain sizes, the collected light yield increased gradually. For sample #4, with average grain size of $91 \mu\text{m}$, an obvious peak appeared near the channel of a peak obtained using the GPS single crystal.

Moreover, great improvement in spectra for alpha particles was achieved by removal of fine powders. Figure 4a) presents a transmission image of sample #4 obtained using an optical microscope. Fine powders produced the granular figure. However, as shown in Figure 4b), no fine powder was observed in sample #5. These were removed by rinsing using alcohol after grinding of the scintillator. Spectra obtained using samples #4 and #5, which had the same average grain size, are presented in Figures 3 and 5, respectively. Integrated counts distributed under 600 channels were decreased drastically for sample #5. Energy resolution of 9.3% was achieved using the sample. It was close to energy resolution of the GPS single-crystal scintillator: 6.3%. In addition, a spectrum obtained using the ZnS:Ag scintillator [4] is shown in Figure 5. The energy resolution of the ZnS:Ag was ca. 40%. The sample #5 had larger collected light yield and higher energy resolution than that of the ZnS:Ag scintillator.

Figure 6 presents an alpha spectrum obtained using sample #8, of which the average grain size was $550 \mu\text{m}$. Only for this sample were both sides of the scintillator layer polished. Although sample #5 had slightly better energy resolution, integrated counts distributed in lower channels than the peak were decreased drastically. High-energy resolution near the single-crystal was achieved by the combination of large and mechanically polished scintillator grains.

Figure 4c) shows sample #8 obtained using the optical microscope. The filing ratio of GPS grains on the surface was ca. 73%. This value was consistent with detection efficiency obtained using the alpha particle measurement.

In alpha particle measurements for dust in workplace air, beta rays and gamma rays are background radiation aside from alpha particles from radon progeny nuclides. Influences of gamma rays and beta rays from the ^{137}Cs and ^{90}Sr are shown in Figure 6. It is readily apparent that the influence of this background radiation can be removed easily by setting of an energy threshold.

3.2. Evaluation of the discrimination capability of fabricated GPS scintillator plates

To evaluate the discrimination ability of the GPS scintillator plates quantitatively, an evaluation index of $S_{\text{peak}}/S_{\text{total}}$ was defined, where S_{peak} is a double value of integrated counts distributed at a higher channel from a peak and S_{total} is the integrated count in a spectrum higher than 50 channels. When $S_{\text{peak}}/S_{\text{total}}$ becomes larger, its spectrum closes to the spectrum obtained using the GPS single-crystal. In addition, the energy resolution simply adopted full width at half maximum of the peak.

Figure 7 portrays a scatter plot of $S_{\text{peak}}/S_{\text{total}}$ and reciprocal quantities of energy resolution for the samples. For samples #2 to #4, the discrimination ability was improved gradually with increased grain size. Huge improvement caused by removal of fine powders was observed between samples #4 and #5. However, for samples of #5 to #8, $S_{\text{peak}}/S_{\text{total}}$ values were improved with increased grain size, although the energy resolution remained at similar values.

3.3. Discussion of scintillator grain size effects on spectra for alpha particles

Figure 8 presents sectional views of GPS scintillator plates estimated from experimentally obtained results. The GPS scintillator plates were composed of the classified scintillator grains and adhesive. The scintillator plate thickness was fixed at 100 μm .

Figures 8a) and 8b) show estimated cross-sections of the scintillator plate with average grain sizes of 60 μm and 96 μm . In such cases, the scintillator grains were smaller than the scintillator plate thickness. Consequently, several parts of alpha particles lost their energy in the adhesive because the adhesives partially covered the surface of the scintillator grains. In this region, $S_{\text{peak}}/S_{\text{total}}$ and energy resolutions were improved with increased grain size.

Figure 8c) shows a corresponding drawing of the average grain size of 196 μm . In this case, the adhesive did not cover the surfaces of scintillator grains anymore. The grains were sufficiently larger than the scintillator plate thickness. Consequently, a further increase in grain sizes, i.e., sample #7, resulted in no improvement in $S_{\text{peak}}/S_{\text{total}}$ and no energy resolution.

In contrast, sample #8 showed improvement in $S_{\text{peak}}/S_{\text{total}}$. In this case, as shown in Figure 8d), both sides of the scintillator layers were polished mechanically as a chance outcome of the fabrication process. Absorption and/or scattering of scintillation lights at boundaries of lower part of scintillator grains were suppressed and $S_{\text{peak}}/S_{\text{total}}$ was improved.

Combinational usage of a scintillator with large grain size and mechanical polishing for scintillator plates improved the energy resolution for alpha particles. Thickness of 100 μm was sufficient to reduce the influence of beta rays and gamma rays. This technique has no limitation in the scintillator plate area size. In addition, adoption of GPS scintillator brought high energy resolution of around 10% for 5.5 MeV alpha particles. This technique will be applied in alpha particle monitors and other devices in the near future.

Acknowledgments

This study was conducted under the framework of the JAEA Cooperative Research Scheme (A) on the

Nuclear Fuel Cycle during 2010–2012. In addition, a part of this study was conducted by the “Development of Systems and Technologies for Advanced Measurement and Analysis” Program of JST.

References

- [1] T.J. Yule, IEEE Transactions on Nuclear Science, 25 (1978) 762
- [2] J.E. Sayers, F.E. Monroe JR., D.D. Smith, S.A. Wallance, IEEE Transactions on Nuclear Science, 37 (1990) 2165.
- [3] W.O. Gentry, G.B. Seaborn, IRE Transactions on Nuclear Science, 8 (1961) 104.
- [4] K. Izaki, Study on Pulse Height Discrimination of Alpha-ray Using ZnS(Ag) Scintillation Detector, JAEA Research 2008-107.
- [5] K. Yasuda, S. Usuda, H. Gunji, Applied Radiation and Isotopes, 52 (2000) 365.
- [6] M. Moszyhski, M. Kapusta, D. Wolski, W. Klamra, B. Cederwall, Nuclear Instruments and Methods in Physics Section A, 404 (1998) 157.
- [7] M. Janecek, R. Borade, E. Bouret-Courchesne, Stephen E. Derenzo, Nuclear Instruments and Methods in Physics Section A, 659 (2012) 252.
- [8] A. Fukabori, L. An, A. Ito, A. Chani, V. Kamada, K. Yoshikawa, Ceramics International, 38 (2012) 2119.
- [9] S. Saeki, J.H. Kaneko, M. Higuchi, S. Kawamura, J. Haruna, F. Fujita, A. Homma, S. Nishiyama, S. Ueda, K. Kurashige, H. Ishibashi, M. Furusaka, IEEE Transactions on Nuclear Science, 54 (2007) 1426.
- [10] S. kawamura, J.H. Kaneko, M. Higuchi, T. Yamaguchi, J. Haruna, Y. Yagi, F. Fujita, A. Homma, S. Nishiyama, H. Ishibashi, K. Kurashige, M. Furusaka, IEEE Transactions on Nuclear Science, 54 (2007) 1383.
- [11] S. kawamura, J.H. Kaneko, M. Higuchi, F. Fujita, A. Homma, J. Haruna, S. Saeki, K. Kurashige, H. Ishibashi, M. Furusaka, Nuclear Instruments and Methods in Physics Section A, 583 (2007) 356.
- [12] Y. Tsubota, J.H. Kaneko, M. Higuchi, T. Shimaoka, S. Nishiyama, H. Ishibashi, F. Fujita, IEEE Transactions on Nuclear Science, 58 (2011) 1923.
- [13] S. Kawamura, J.H. Kaneko, M. Higuchi, J. Haruna, S. Saeki, F. Fujita, A. Homma, S. Nishiyama, S. Ueda, K. Kurashige, H. Ishibashi, M. Furusaka, IEEE Transactions on Nuclear Science, 56 (2009) 328.
- [14] P. L. Reeder, Nuclear Instruments and Methods in Physics Section A, 353 (1994) 134.
- [15] P. L. Reeder, Nuclear Instruments and Methods in Physics Section A, 340 (1994) 371.
- [16] J. Haruna, J.H. Kaneko, M. Higuchi, S. Kawamura, S. Saeki, Y. Yagi, H. Ishibashi, F. Fujita, A. Homma, M. Furusaka, Y. Kiyanagi IEEE Transactions on Nuclear Science, 54 (2007) 1421.
- [17] O. Sidletskiy, V. Baumer, I. Gerasymov, B. Grinyov, K. Katrunov, N. Strazhinsky, O. Tarasenko, V. Tarasov, S. Tkachenko, Radiation Measurements, 45 (2010) 365.

- [18] N.Z. Galunov, B.V. Grinyov, N.L. Karavaeva, Y.V. Gerasymov, O.T. Sidletskit, O.A. Tarasenko, IEEE Transactions on Nuclear Science, 58 (2011) 339.
- [19] I. Gerasymov, O. Sidletskiy, S. Neicheva, B. Grinyov, V. Baumer, E. Galenin, K. Katrunov, S. Tkachenko, O. Voloshina, A. Zhukov, Journal of Crystal Growth, 318 (2011) 805.

Table and Figure captions

Table 1
GPS scintillator plates fabricated for this study

Figure 1

Optical photograph of a conventional ZnS:Ag scintillator of 10 mg/cm² mass thickness (left) and an example of GPS scintillator plates fabricated on a glass substrate of 25 × 25 × 1.25 mm (right). Scintillator parts are surrounded by dashed lines. Although the school seals printed on the paper are visible through the GPS scintillator plate, they are not visible through the ZnS:Ag scintillator.

Figure 2

Examples of energy spectra for 5.5 MeV alpha particles obtained from YAP and GPS single-crystal scintillators. The GPS scintillator showed a light yield that was two times higher than that of the YAP scintillator.

Figure 3

Examples of spectra for 5.5 MeV alpha particles obtained by GPS scintillator plates with several grain sizes: a) GPS single crystal, b) sample #1 (< 5 μm), c) sample #2 (35 μm), d) sample #3 (60 μm), and e) sample #4 (91 μm). These samples, except for the GPS single crystal, included fine powders.

Figure 4

Transmission images of a) sample #4 (91 μm with fine powders), b) sample #5 (91 μm without fine powders), and c) sample #8 (550 μm without fine powders). These images were obtained using an optical microscope. Fine powders make a) appear to be granulated. The filling ratio of sample #8, 73%, was calculated from this picture.

Figure 5

Examples of spectra for 5.5 MeV alpha particles obtained using sample #5, the ZnS:Ag and the GPS single crystal. Sample #5 showed light yield higher than that of the ZnS:Ag. The energy resolution of a peak obtained from sample #5 was 9.3%.

Figure 6

Examples of spectra for 5.5 MeV alpha particles, 662 keV gamma rays from the ^{137}Cs and beta rays from the ^{90}Sr obtained from sample #8 (550 μm). Both surfaces of the scintillator layer were polished mechanically. Influences of gamma and beta-rays are easily removed by setting an energy threshold.

Figure 7

A scattered plot of $S_{\text{peak}}/S_{\text{total}}$ and reciprocal quantities of energy resolutions of the samples. The discrimination ability was improved with increased grain sizes for samples #2 to #4. Great improvement attributable to the removal of fine powders was apparent between samples #4 and #5. After sample #5, the energy resolutions were almost steady with increased grain sizes. Further improvement in $S_{\text{peak}}/S_{\text{total}}$ was achieved from sample #8, the surfaces of which were polished on both sides.

Figure 8

Sectional views of the GPS scintillator plates estimated from experimentally obtained results. Average grain sizes in the figures were the following: a) 60 μm , b) 96 μm , c) 196 μm , and d) 550 μm . The scintillator layer thickness was fixed at 100 μm .

Figure and Table

Table 1

Thickness of scintillator layers, 100 μm ; Glass substrates, $25 \times 25 \times 1.25$ mm

ID	Average grain size (μm)	Removal of fine powder	Energy resolution (%)
#1	< 5	-	68.9
#2	35	-	70.9
#3	60	-	70.9
#4	91	-	20.0
#5	91	○	9.3
#6	196	○	12.8
#7	275	○	12.3
#8	550	○	10.3

Figure 1

Figure 2

369
370
371
372
373
374
375
376

377
378
379
380

381
382
383
384
385
386
387
388
389

Figure 3

Figure 4

390
391
392
393
394
395
396
397
398
399
400
401
402
403
404
405
406
407
408

Figure 5

409
410
411
412
413
414
415
416
417
418
419
420
421
422
423
424
425
426
427

Figure 6

428
429
430
431
432
433
434
435
436
437
438
439
440
441
442
443
444

Figure 7

446

447

448

449

450

451

452

453

454

455

456

457

458

459

460

461

462

463

464

465

Figure 8

467

468

469

470

471

472

473

474

475

476

477

478

479

480

481