Development of Alpha Particle Detectors for Detecting Radiological Contamination

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Abstract. Alpha contaminations in nuclear facilities should be detected instantaneously because alpha particle emitters such as plutonium isotopes are very harmful when a worker inhales them. We have developed new alpha particle detectors for detecting the alpha contaminations accurately and instantaneously: alpha imaging detectors and alpha dust monitors. An alpha detector which has a good energy resolution and spatial resolution is useful for distinguishing nuclear material such as plutonium from radon progeny. For the development of the alpha imaging detector, we used a thin cerium-doped $Gd_3(Ga,Al)_5O_{12}$ (Ce: GAGG) scintillator and silicon photomultipliers (SiPMs). The thin GAGG scintillator was optically coupled to a light-guide and the SiPMs for the detector fabrication. The detector showed a good energy resolution for 5.5 MeV alpha particles (~13 % at the full width at half maximum (FWHM)). The detector is capable of capturing two-dimensional alpha images. This detector also has the advantage of its compact size, which enables to measure the alpha contaminations in narrow spaces. We demonstrated the actual measurement of smear samples obtained from the Fukushima Daiichi Nuclear Power Station using the developed imaging detector. Commercial alpha dust monitors with a silicon surface barrier detector (SSBD) operating at some nuclear facilities frequently produced false alarms due to environmental conditions such as humidity. For the development of the alpha dust monitor, we used a cerium-doped $Gd_2Si_2O_7$ (GPS) scintillator plate and a photomultiplier tube (PMT). The energy resolution for 5.5-MeV alpha particles was ~12% FWHM. The count-rate of the radon progeny decreased by 77% with applying energy discrimination. The alpha dust monitor was capable of conducting alpha-particle spectroscopy even though the GPS scintillator got wet. The alpha dust monitor is an ideal choice for in places lacking temperature and humidity controls. In the presentation at the IRPA 15 conference, we will present the developments and measurement results of these alpha detectors.

KEYWORDS: Alpha particle detector; Fukushima Daiichi Nuclear Power Station; Continuous air monitoring; Plutonium; Radon;

1 INTRODUCTION

After the Great East Japan Earthquake hit the Fukushima Daiichi Nuclear Power Station (FDNPS), large quantities of radioactive materials were released inside and outside of the FDNPS [1]. Especially, a reactor building of the FDNPS was seriously contaminated by radioactive materials released from the Primary Containment Vessel. These radioactive materials were mostly beta and gamma emitters such as ^{137}Cs and ^{90}Sr, but there is a high possibility that alpha emitters such as ^{238}Pu and ^{239}Pu were released as well. As the decommissioning process continues, the frequency with which workers enter highly contaminated areas such as the reactor building will surely increase. Information about the activity and radionuclides of alpha emitters at a working site is extremely important for preventing workers from exposure to radiation. For detecting the alpha contaminations accurately and instantaneously, we have developed new alpha particle detectors: alpha imaging detectors and alpha dust monitors [2, 3].

So far using silver-doped zinc sulfide (ZnS(Ag)) survey meters used to be the only option for measuring alpha emitters at the FDNPS site. Such survey meters have been used for measuring alpha particle count rates. However, the energy resolutions obtained by the ZnS(Ag) scintillators are rather low, therefore, not showing where the alpha emitters are originating from: nuclear fuels or naturally occurring radionuclide such as Radon (^{222}Rn) and progeny [4]. To identify a radionuclide of an alpha emitter, an alpha particle detector with high energy resolution is required [5]. Another aim of measuring alpha emitters is obtaining information about the two-dimensional distribution of alpha emitters. In the previous study, alpha particles from Pu characteristically show up as spots in the two-dimensional distributions [4]. Information about the size and distribution of alpha emitters is very useful for radiation protection. For example, the particle size of alpha emitters is important for estimating internal exposure to a worker. Moreover, information about the two-dimensional distribution of alpha emitters is valuable for distinguishing Pu from ^{222}Rn progeny because the distributions of alpha particles between them are different [4]. An imaging plate (IP) has been widely used to obtain the two-dimensional distribution of alpha emitters [6], but this plate is sensitive to beta particles and gamma rays as well. There are large quantities of beta and gamma emitters at the FDNPS site. The IP cannot distinguish alpha particles from beta particles and gamma rays, and identify radionuclides of alpha emitters. Therefore, energy information is necessary to measure only alpha emitters in FDNPS. We have developed an alpha particle imaging detector that can measure the energy spectrum and the two-dimensional distribution of alpha particles by combining a cerium-doped $Gd_3(Ga,Al)_5O_{12}$ (Ce: GAGG) scintillator with a silicon photomultiplier (SiPM). This detector has good energy resolution (~13% full width at half maximum (FWHM)) and high spatial resolution (0.6 mm FWHM) for 5.5MeV alpha particles [7]. Furthermore, its beta and gamma sensitivity can be reduced by setting the thickness of the Ce: GAGG scintillator to 0.05 mm. Therefore, we measured alpha emitters in the reactor building of FDNPS for the first time by using the developed alpha particle imaging detector. The features of radionuclides of alpha emitters in the reactor building of FDNPS were clarified by these measurements.

In addition, in order to instantaneously check for leakage of alpha-particle emitters, an airborne monitoring system is installed in the facility [8]. The airborne monitoring system consists of an alpha-particle detector, an air sampler, and an air filter to capture radioactive dust. If the alpha count measured using the monitoring system exceeds a given alarm threshold, the monitoring system sounds an alarm to notify workers so that they can escape from the working room. A silicon surface barrier detector (SSBD) is often used as the alpha-particle detector because it has good energy resolution [9]. However, dust monitors with SSBD frequently produce a false alarm, especially if in a room of high humidity. If a false alarm from the dust monitor sounds, workers in this room should escape to the outside to avoid inhaling Pu particles, which interrupts the operations in this room. Therefore, the dust monitor is required to have high energy resolution and to be strong against external noise to reduce false alarms. Since dust monitors using an SSBD have produced false alarms, replacing the SSBD with a scintillation detector is one potential solution to reduce the rate of false alarms. The gadolinium pyrosilicate (GPS, $Gd_2Si_2O_7$:Ce) scintillator developed by Hokkaido University has a high light output and good energy resolution for alpha particles [10]. Energy resolution up to 13% of the FWHM was achieved. Also, its thin thickness is suitable for reducing beta and gamma background noise. The light emission spectrum of the GPS has two peaks, namely, 375 and 390 nm [11], and they are matched with the quantum efficiency of a photomultiplier tube (PMT). It is not suitable for continuous monitoring in a work room where environmental conditions change because a gain of SiPM is sensitive to temperature variations [12]. Thus, we developed an alpha dust monitor using a GPS scintillator plate with the PMT for a highly reliable dust monitor.

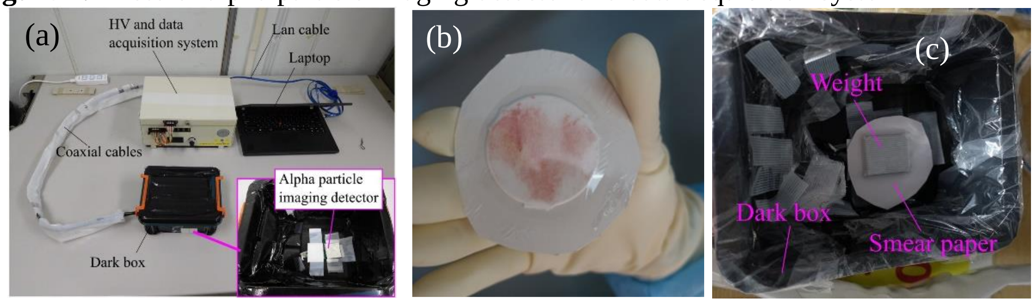
2 MATERIALS AND METHODS

2.1 Measurement of FDNPS smear papers by using alpha particle imaging detector

An alpha particle imaging detector must have good energy resolution for identifying radionuclides of alpha emitters. Moreover, because there are large quantities of beta and gamma emitters at the FDNPS site, the influences of beta and gamma rays must be distinguished from those of alpha particles. Figure 1(a) shows a schematic drawing of the alpha particle imaging detector developed in this study. A very thin (0.05-mm-thick) GAGG scintillator was used employed to limit beta sensitivity [13]. The length and the width of the scintillator size 26 mm × 26 mm. The 0.05-mm-thick GAGG scintillator was coupled to a 1-mm-thick glass plate and a 1-mm-thick acrylic light guide. The bottom of the acrylic light guide was optically coupled to silicon photomultiplier (SiPM) arrays having 8 × 8 channels (Through Silicon Via (TSV) MPPC array S12642-0404PA-50, Hamamatsu Photonics K.K., Japan) by using an optical compound (Shin-Etsu silicone, Shin-Etsu Chemical Co., Ltd., Japan). The detector was covered with aluminized mylar to shield it from external light. The GAGG scintillator produces scintillation light when an alpha particle deposits energy. The SiPMs convert this scintillation light into electric signals. The analog signals produced by the 8 × 8 SiPMs were fed to preamplifiers and a weight-summing circuit, which creates X+, X-, Y+, Y- signals for position calculation. These signals are converted to digital signals by using analog-to-digital converters and subsequently fed to a field programmable gate array. Finally, the data are transferred to a laptop via a lan cable, and the two-dimensional position and energy spectrum of the alpha particles are displayed on the laptop. High voltage (HV) was adjusted to -67 V, and the temperature of the SiPM circuit was monitored to compensate for temperature-dependent changes in gain. During measurement, the detector and sample were enclosed in a dark box to shield them from external light.

Surface contamination on the floor inside the FDNPS reactor building was wiped off by using smear papers for the measurement. Figure 1(b) shows photograph of one of the smear papers. The diameter of the smear papers was 5 cm. The smear papers were covered with a thin polyethylene film to prevent cross-contamination to the detector. Figure 1(c) shows a photograph of the measurement of the smear papers by using the developed alpha particle imaging detector. Each of the smear papers was placed on the detector and close contact between the paper and the detector was achieved by placing a weight on top. The smear paper was placed in a black box, and the door of the container was closed to prevent penetration of external light. The measurement was repeated twice during a 5 min period. Also, the measurement was started at around noon to maintain the room temperature. The dose-rate of gamma rays in the measurement environment was approximately 10 μSv/h. Moreover, we measured two Pu samples obtained from a MOX fuel fabrication facility for comparison with the FDNPS smear papers. The measurement was conducted under the same conditions as those used in FDNPS (measurement time, HV).

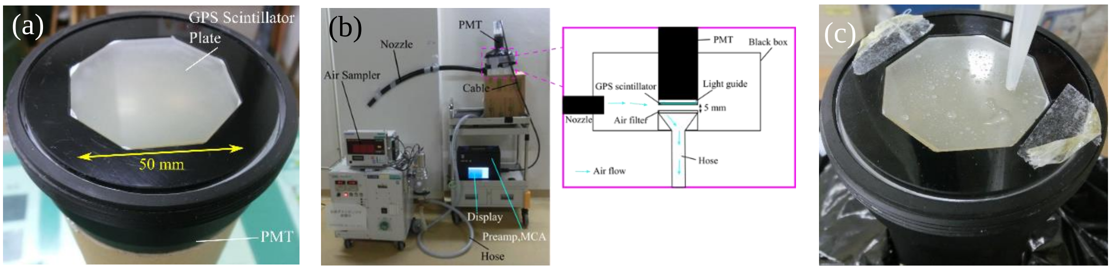
Figure 1: Photo of alpha particle imaging detector and data acquisition system



2.2 Development of a GPS scintillator plate-based alpha dust monitor

Figure 2(a) shows photos of the developed GPS scintillator plate and the alpha-particle detector. The GPS scintillator plate was composed of a GPS scintillator and a glass substrate. The GPS scintillator was fabricated from a sintered compact prepared by liquid-phase sintering, using SiO_2 as a self-flux. The thickness of the layer of the GPS scintillator was approximately 40 μm, which is able to reduce the beta and gamma background. The GPS scintillator was optically coupled with a 3-mm-thick high-transmission glass. The shape of the GPS scintillator plate was hexagonal, and its diameter is 50 mm. This diameter is the same as that of an air filter deployed for monitoring airborne contamination (Fig. 1). The bottom face of the GPS scintillator plate was optically coupled to a 3-in-diameter PMT (R6233, Hamamatsu Photonics K.K., Hamamatsu, Japan) using an optical grease. The PMT was connected to a case that contains electronics via a signal and HV cable. An output signal from the PMT was transferred to a preamplifier (5625, CLEAR PULSE CO., LTD., Tokyo, Japan) in the case. The preamplifier's output signal was transferred to a Multi-Chanel Analyzer (MCA; A2731, CLEAR PULSE CO., LTD., Tokyo, Japan). A display on the case showed an energy spectrum in real time. The high voltage for the PMT was adjusted to −490 V. Figure 2(b) shows the developed dust monitor. The alpha-particle detector was positioned facing downward, and the detection end was placed in a light-tight box. A nozzle and an air sampler were also connected to the same light-tight box. When the air sampler operates, the nozzle sucks up the air around the monitor, and radioactive dust in the air are captured on the filter. The distance between the GPS scintillator plate and the filter was approximately 5 mm, so that air can pass through. The alpha detector continually measures the alpha particles from the radioactive dust on the filter, thus enabling real time monitoring. The alpha detector and the filter were placed in the light-tight box in order to shield them from external light. The air was discharged from the air sampler, after passing through the air filter. The entire system was transportable. Measurements were conducted in a place with limited air ventilation and a high ^{222}Rn concentration. The average ^{222}Rn concentration, measured using an AlphaGUARD monitor (Genitron Instruments, Germany), was 57 ± 3 Bq/m^3. The air sampler collected the air on the filter. To ensure that the activity of Rn progeny was saturated, the air sampling commenced 3 h before the start of the measurement. The measurement time was 1700 s, with 30 s intervals for continual monitoring. Additionally, a Pu sample (single PuO2 particle, 19.2 Bq) was measured, with 30 s intervals, by placing the sample on the air filter's position. In the time variation graph, Pu sample counts were added to ^{222}Rn progeny counts with a 240 s delay. In order to simulate high humidity conditions, water was dropped directly on the surface of the GPS plate using a dropper. Figure 2(c) shows a schematic of this measurement and a photo of the water-dropping process. A 2-in-diameter ^{241}Am source (same as in the air filter) was used for the measurement, and the alpha spectra with and without water drops were compared.

Figure 2: Photo of the GPS scintillator plate (a) and a developed alpha dust monitor (b), example of dropping water (c).



3 RESULTS

3.1 Measurement of smear papers by using alpha particle imaging detector

Figure 3(a)(b) shows comparisons of the energy spectra of the smear papers and the Pu sample measured at the nuclear fuel facility. The energy spectrum of smear papers no. 2 or 4 and that of the Pu sample confirmed up to 6.5 MeV. The shape of the spectra of smear papers no. 2 or 4 and the Pu samples were different because both smear paper and the Pu sample were affected by self-absorption and their peaks were shifted to lower energy. In the results of all smear papers, the alpha spectrum was not confirmed at energies exceeding 6 MeV, which means that this spectrum was not created by naturally occurring radionuclides such as Radon progeny (mainly 7.7 MeV alpha particle from ^{214}Po). Therefore, this alpha spectrum must have been created by artificial radioactivity originating from a nuclear fuel such as ^{238}Pu (5.5 MeV alpha particle), ^{239}Pu (5.15 MeV alpha particle), and ^{241}Am (5.5 MeV alpha particle).

Figure 3: Comparisons of alpha spectra between smear papers and Pu samples measured at nuclear fuel facility: smear paper no.2(a) and no.4(b). The error bars indicate the standard deviation by taking the square root of the number of counts of each bin in the spectrum.

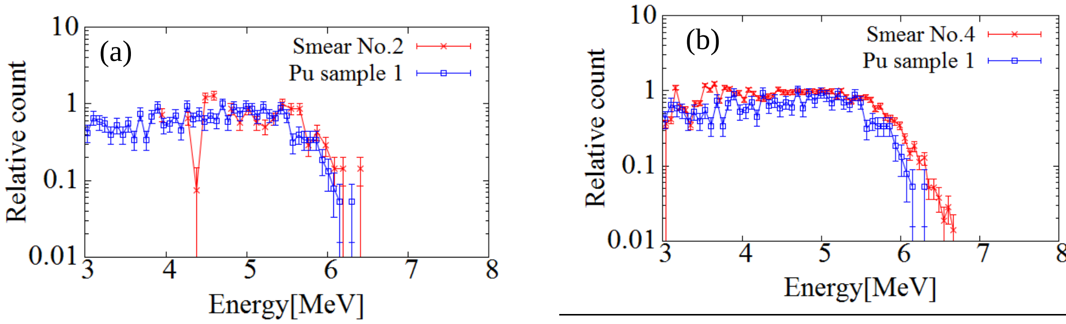
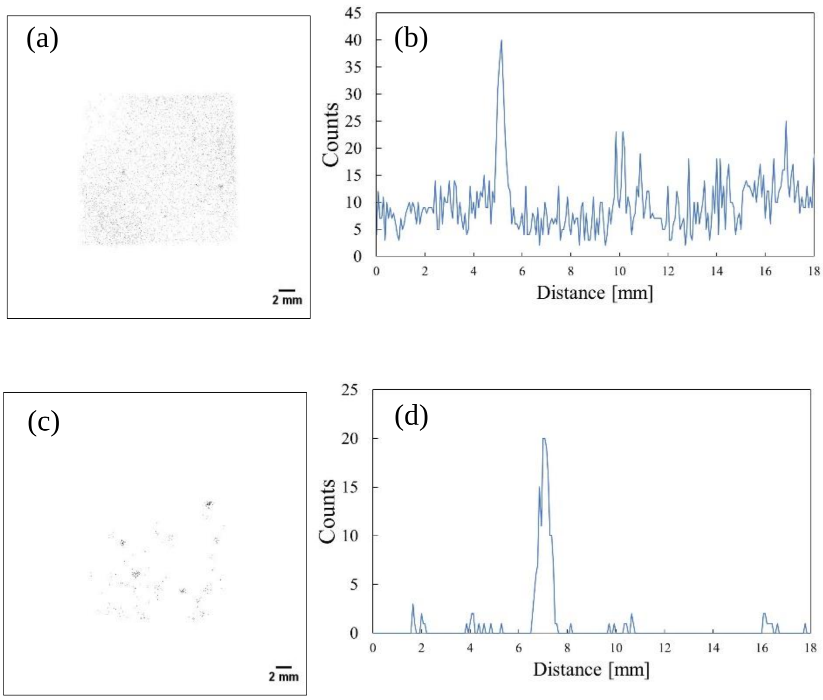


Figure 4(a) shows the two-dimensional distribution of the smear papers in the energy range of 5–6.5 MeV (alpha particles) measured for 30 min. Alpha particle emitters were widely distributed on the smear paper, as opposed to forming spots. In addition, Fig. 4(c) shows the two-dimensional distribution of the Pu sample obtained from a fabrication process of mixed oxide (MOX) fuel in the nuclear fuel facility. These two-dimensional distribution images were created in the same energy range of 5–6.5 MeV. The spots were confirmed in both images of the Pu samples. Even though the energy ranges of the FDNPS smear paper and the Pu sample are identical (5–6.5 MeV), their two-dimensional distributions are different. In the intensity profile of smear paper no. 4 shown in Fig.4(b), the maximum count of 40 can be confirmed at the distance of 5.1 mm, whereas counts of other area fluctuated between 4 to 25. In contrast, in the intensity profile of Pu sample 1 shown in Fig.4(d), the maximum count of 20 can be confirmed at the distance of 7.0 mm. It is possible that minute particles and large particles were mixed in one smear paper.

Figure 4: Two-dimensional distribution of Pu samples measured at nuclear fuel facility in energy range of 5–6.5 MeV (alpha particles): smear paper no. 4 (a), its intensity profile (b), Pu sample 1 (c), and its intensity profile (d)



3.2 Development of a GPS scintillator plate-based alpha dust monitor

Figure 5(a) shows the alpha energy spectrum of the ^{241}Am source placed on the GPS scintillator plate, measured over 600 s. Energy resolution was calculated using Gaussian fitting. The energy resolution of this spectrum was 11.9% ± 0.2% of the FWHM. The mean peak channel of the three measurement times was 602.7 ± 0.8. This energy resolution largely improved compared to that of the ZnS(Ag) scintillator (~50% of the FWHM). Figure 5(b) shows the energy spectra of the Pu sample placed at the same position as the air filter, measured for 1500 s. Figure 5(c) also shows the energy spectra of ^{222}Rn progeny, measured for 1500 s. The major isotope of the Pu sample is ^{238}Pu, which emits a 5.5-MeV alpha-particle. The ^{218}Po and ^{214}Po decayed from ^{222}Rn existed as a radioactive aerosol and were collected on the filter. Therefore, the Rn progeny spectrum counts were created by the ^{218}Po-emitted 6.0-MeV alpha-particle and ^{214}Po-emitted 7.7-MeV alpha particles. In the Rn progeny spectrum, a peak below 20 channels was identified and was likely created by beta particles from the Rn progeny (^{214}Pb and ^{214}Bi). In order to eliminate Rn progeny counts, an energy window was set with the upper (ULD) and lower (LLD) channels of the Pu sample spectrum. The 15-channel LLD was the best setting for achieving the highest Pu/Rn ratio. The Pu/Rn ratio becomes lower when using an LLD below 15 channels because of the section of beta particles. By applying the optimized energy window, Rn counts were reduced by 77%.

Figure 5: Energy spectra: ^{241}Am source (a), Pu sample (b), and Rn progeny (c).

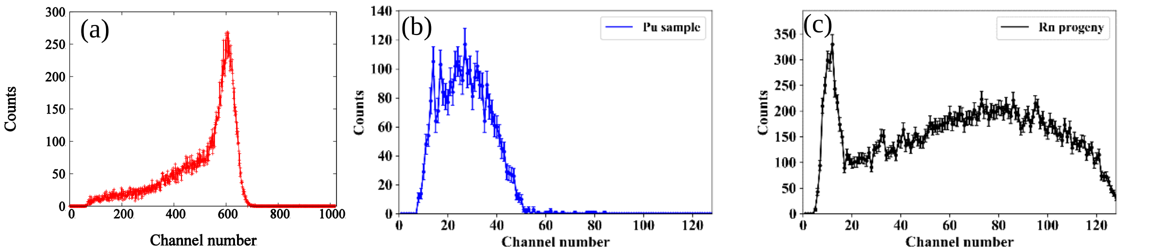


Figure 6 shows the alpha spectra with and without dropping water; the measurement time was 5 min. The GPS scintillator plate was able to measure the alpha spectrum even though the GPS scintillator got wet. The ratio of the total counts with/without water was 0.84. The peak channel was calculated using the Gaussian fitting. The peak numbers of channels without water and with water were 47.1 and 47.3 channels, respectively.

Figure 6: Alpha energy spectra with and without dropping water. The distance between the source and the GPS scintillator was 5 mm, which is the same as in the alpha dust monitor.

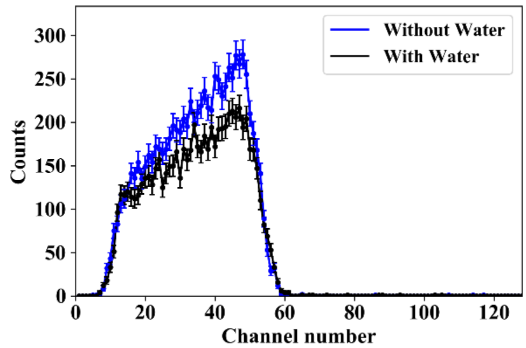
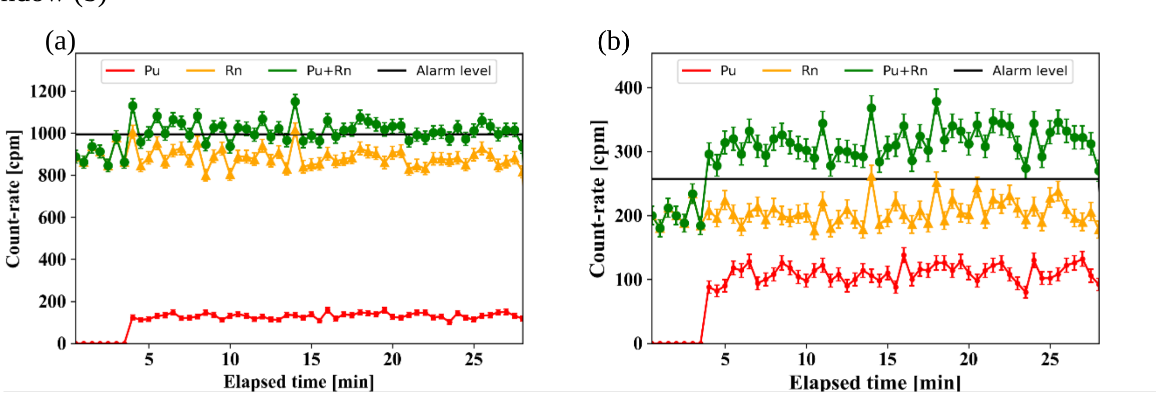


Figure 7 shows the time variation of count-rate (in count per minutes [cpm]), with 30 s intervals. The energy window can effectively trigger the PuO_2 particle alarm instantaneously and with certainty. The count-rate of the Rn progeny was decreased by 77%.

Figure 7: Time variation of counts with 30 s intervals; without the energy window (a) and with energy window (b)



4 DISCUSSION AND CONCLUSIONS

We measured the existing alpha emitter contamination in smear papers at the FDNPS site by using the alpha particle imaging detector, and revealed that the alpha emitters in the reactor buildings at the FDNPS site originated from the nuclear fuels. This result has been impossible to obtain using ZnS(Ag) detectors. The Pu sample was obtained from the MOX fuel facility. Although ^{239}Pu is the dominant Pu isotope in the MOX fuel (^{238}Pu: 3.5%, ^{239}Pu: 47.4 %) [14], ^{238}Pu has the highest specific activity (^{238}Pu: 600 GBq/g, ^{239}Pu is 2 GBq/g) among the plutonium isotopes. Considering the specific activity and abundance ratio of Pu isotopes, 5.5 MeV alpha particles from ^{238}Pu majorly contribute to the alpha spectra. The beta activity of the FDNPS smear papers was approximately 10–1000 times higher than their alpha activity. Therefore, the influence of beta particles must be considered in alpha spectrum measurement. In this study, we used a very thin (0.05-mm-thick) scintillator to reduce the influence of beta particles. However, in measured spectra of the smear papers, beta spectrum and alpha spectrum overlapped partly. By subtraction with/without paper, the beta spectrum could be removed. In the future, a technique for discrimination of beta particles is necessary for accurate alpha spectrometry. The two-dimensional distribution of alpha particles on the FDNPS smear papers was different from that of the Pu sample; In the Pu sample, spots were observed, but spots and wide distribution of minute particles were simultaneously observed in the FDNPS smear papers. However, the alpha emitters in FDNPS would have different diameters and compositions compared to the Pu particle in the MOX fuel fabrication facility. Further studies are required to clarify the diameter and composition of the Pu in the FDNPS. Even though our measurements were conducted on-site at the FDNPS without destroying the samples, the results showed the radioactive concentration of alpha particles, which was of the same order of magnitude as measured using a radiochemical analysis. Therefore, our measurement method proved to be valuable for detecting alpha emitter contamination at the FDNPS site.

We also developed an alpha dust monitor using a GPS scintillator plate. The GPS scintillator plate exhibited a uniform sensitivity in the lateral direction (between −2 cm and +2 cm) with respect to the 5.5-MeV alpha particles. Additionally, this detector has good efficiency with respect to the 5.5-MeV alpha particles (92%). A PuO_2 particle is generally tiny (on average, 5 μm diameter in a MOX fuel facility), so it attaches onto anywhere on the air filter. The GPS scintillator plate has uniform sensitivities and is able to detect PuO_2 particles if they attach anywhere on the filter. The energy resolution of the spectrometer for 5.5-MeV alpha particles was 11.9% ± 0.2% of the FWHM, which is superior to a ZnS(Ag) scintillation detector (~50% of the FWHM). This resolution is comparable to an alpha detector using a GAGG scintillator with the SiPM. However, the SiPM is temperature-dependent and is thus not suitable for continuous monitoring, especially outside of buildings. The PMT is generally stable against temperature changes, relative to the SiPM. We set the energy window to the energy range of ^{238}Pu (~5.5 MeV); this approach was effective in reducing counts created by ^{214}Po alpha particles with 7.7 MeV. We used the 15-channel LLD to eliminate the effect of beta particles from the Rn progeny, although 17% Pu counts were also reduced by applying the energy window. Therefore, reducing beta-particle sensitivity is important in improving the Pu/Rn ratio. In this study, in order for air to pass, the distance between the air filter and the GPS scintillator plate was set to 5 mm, which is the same distance as in a commercial one. Optimizing this distance will further improve the Pu/Rn ratio. In this study, the average count-rate ratio of Rn was 6.8 times higher than that of the Pu sample. Applying the energy window allowed for the instantaneous triggering from a PuO_2 particle with 19.0 Bq activity. The GPS scintillator plate was able to measure the alpha spectrum and to identify the peak location of a 5.5-MeV alpha-particle, even though water was attached to the surface of the GPS scintillator plate. The alpha dust monitor using the GPS scintillator plate is an ideal choice for detecting alpha-particle emitters in places, which lack the ability to control temperature and humidity, such as the outside of building

5 ACKNOWLEDGEMENTS

This work was partly supported by JSPS KAKENHI Grant Number JP15K21618. This work was done in collaboration with from Tokyo Electric Power Company Holdings, Inc.

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