Radiation Measurements 115 (2018) 13–19

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Development of a high-resolution alpha-particle imaging system for detection of plutonium particles from the Fukushima Daiichi nuclear power plant

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ARTICLE INFO

Keywords:
Position-sensitive photomultiplier tube
ZnS(Ag)
High resolution
Alpha particles
Plutonium
Particles

ABSTRACT

For the detection of plutonium particles released from the Fukushima Daiichi nuclear power plant, we developed a high-resolution alpha-particle imaging system. The detector of the alpha-particle imaging system consists of a thin ZnS(Ag) sheet, a light guide, and a high quantum efficiency 1-inch square position-sensitive photomultiplier tube (PSPMT). The ZnS(Ag) sheet was optically coupled to the PSPMT with a 1.5 mm thick light guide between them. The Anger principle was used for the position calculation of the alpha particles. The spatial resolution of the alpha-particle imaging detector was 0.45 mm full width at half maximum for 5.5 MeV alpha particles. The uniformity of the imaging detector in the central part of the field of view was ± 7%. The detection efficiency was 76% for 5.5 MeV alpha particles. Although the background count rate was 8.1 counts per minute because of the collection and detection of alpha particles from radon daughters in the air, it could be decreased to 0.1 counts per minute if the detector surface was covered with paper. There was no increase in the background count rates of ^{137}Cs gamma photons and ^{45}Ca beta particles. We obtained high-resolution variable phantom images and particles of alpha emitters with the system. We conclude that the alpha-particle imaging system developed is promising for the detection of plutonium particles in samples collected near the Fukushima Daiichi nuclear power plant.

1. Introduction

The accident at the Fukushima Daiichi nuclear power plant (FDNPP) in 2011 caused massive contamination of the Japanese land surface. Large areas near Fukushima were contaminated with ^{137}Cs, ^{134}Cs, or other radioisotopes (Kinoshita et al., 2011; Shozugawa et al., 2012; Steinhauser, 2014). Among these FDNPP-released radioisotopes, alpha emitters, plutonium (Pu) and uranium, were recently detected and reported (Zheng et al, 2012, 2013; Shinonaga et al., 2014). Among these alpha emitters, ^{239}Pu has a long half-life (24,110 years) and is a most remarkable radioisotope in terms of both the exposure dose and chemical toxicity (Gillies et al., 2017). ^{239}Pu was released from FDNPP in the form of PuO_2 particles, and these are thought to be distributed around FDNPP. PuO_2 particles can accumulate in the lungs, emit alpha particles, and cause lung cancer (Gillies et al., 2017). Although the radioactivity of the detected Pu particles from FDNPP is low (Zheng et al, 2012, 2013; Shinonaga et al., 2014), the survey of Pu released from FDNPP was not conducted extensively, and thus the whole aspect of Pu contamination in Japan from FDNPP is not clear. One reason is that the detection of low-activity Pu particles is difficult because natural radionuclides such as radon and its daughter radionuclides also emit alpha particles (Iida et al., 1990).

For Pu detection and quantitation, accelerator mass spectrometry is usually used (Zheng et al, 2012, 2013; Shinonaga et al., 2014). Before the accelerator mass spectrometry measurements, detection of the position of the PuO_2 particles in samples is a promising method to increase the sensitivity. It reduces the time for the preprocessing and improves the quality of the data. A high-resolution and low-background real-time alpha-particle imaging system is suitable to detect PuO_2 particles. Because PuO_2 particles emit alpha particles in the same position in the imaging detector, a spot is formed in the image of the alpha-particle imaging system. The method is also used to distinguish PuO_2 particles from natural alpha emitters such as radon and its daughters in air (Iida et al., 1990; Koarashi et al., 2007).

In alpha-particle imaging detectors, Anger camera–based imaging systems using a scintillator combined with a position-sensitive photomultiplier tube (PSPMT) detector are suitable for Pu detection because they can measure the position and energy of alpha particles

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https://doi.org/10.1016/j.radmeas.2018.05.009
Received 2 March 2018; Received in revised form 21 May 2018; Accepted 21 May 2018

simultaneously. Because the pulse heights of the alpha particles detected by scintillation detectors are commonly much higher than those of the environmental gamma photons or beta particles, a low background can be achieved if a thin scintillator is used. However, the spatial resolution of these previously developed systems was limited (Yamamoto and Iida, 1998; Yamamoto et al., 1997). A silicon photomultiplier (Si-PM) array is a possible new photodetector to be used for radiation imaging detectors using the Anger principle (Yamamoto et al., 2010; Yoon et al., 2012), including an alpha-particle imaging detector (Morishita et al., 2014, 2017). However, the temperature dependency of the gain of the Si-PM is high (Yamamoto et al., 2011), so the stability of Si-PM-based imaging systems is generally lower than that of PSPMT-based imaging systems. A means of temperature control, such as water cooling, is usually required to maintain the stability of an imaging system such as an Si-PM-based positron emission tomography system (Grant et al., 2016).

Recently, the quantum efficiency (QE) of the PSPMT photocathode was increased, and high spatial resolution may be achieved with the newly developed PSPMT for the imaging system. Because high QE of the photocathode increases the number of photoelectrons for the scintillation photons, the energy resolution as well as the spatial resolution of the imaging system increases (Knoll, 2000). Consequently, we developed an alpha-particle imaging system using a high QE PSPMT and evaluated the performance for alpha particles to realize high resolution and low background as well as high stability that is suitable for the future detection of Pu particles from FDNPP.

2. Methods

2.1. Principle of operation of the alpha-particle imaging detector

The structure of the alpha-particle imaging detector we developed is shown in Fig. 1. In this detector, a thin ZnS(Ag) sheet was used as a scintillator. This was because the light output of ZnS(Ag) is large, so spatial resolution can be increased by the Anger principle. Thin ZnS(Ag) can be penetrated by environmental gamma photons and beta particles, while it absorbs alpha particles of alpha emitters such as Pu. A light guide is used between the scintillator and the PSPMT to distribute the scintillation light among several anodes of the PSPMT to calculate the position by the Anger principle. The PSPMT detects the scintillation light produced by the interaction between alpha particles and the ZnS(Ag) scintillator, and the photocathode of the PSPMT converts light photons to photoelectrons. The photoelectrons are multiplied by the dynodes of the PSPMT, and the electrons are detected by the anodes of the PSPMT. High voltage is supplied to the voltage divider of the PSPMT. The anode signals are fed to the amplifiers of the data acquisition electronics for position and energy calculation.

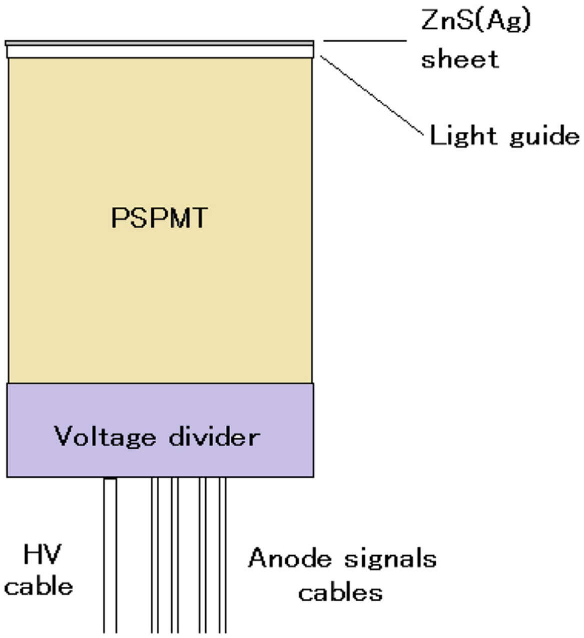

Fig. 1. Structure of the alpha-particle imaging detector. HV, high voltage; PSPMT, position-sensitive photomultiplier tube.

Fig. 2. Alpha-particle imaging detector supported by a flexible arm stand.

2.2. Alpha-particle imaging detector

A photograph of the alpha-particle imaging detector we developed is shown in Fig. 2. The ZnS(Ag) sheet used for the detector had a thickness of 3.25 mg/cm^2 and was painted on a 0.25 mm thick peristyle sheet (EJ-420, G-Tech, Japan). The ZnS(Ag) sheet was optically coupled to a 1.5 mm thick light guide made of acrylic plate with silicone rubber (KE420, Shin-etsu Silicone, Japan). The ZnS(Ag) sheet and the acrylic plate were optically coupled to a PSPMT (R8900-100-C12, Hamamatsu Photonics, Japan) with silicone rubber. The PSPMT was a 1 inch square metal package type with 6 × 6 crosswire anodes. The photocathode of the PSPMT was a super bialkali photocathode, which had approximately 10% higher QE than a normal PSPMT. An E7514 voltage divider (Hamamatsu Photonics, Japan) was used. A high voltage of -650 V was supplied to the voltage divider. For the light shield of the detector, aluminized Mylar sheet was used on the ZnS(Ag) scintillator. The sides of the PSPMT were covered with black tape for the light shield and insulation of the high voltage that is supplied to the metal surfaces of the PSPMT. The alpha-particle imaging detector was contained in a case and attached to a flexible arm stand that can freely change the position of the detector as shown in Fig. 2.

The six analog signals from the crosswire anodes in the X-direction and the six analog signals in the Y-direction were fed to weight summing boards with approximately 50 cm long coaxial cables. The analog signals were individually amplified and were weight summed by amplifiers (AD8056, Analog Devices) to form four weight-summed signals ($X+$, X-, $Y+$, and Y-). The weight-summed signals were fed to 100-MHz, 10 bit analog-to-digital converters (AD9218, Analog Devices) of the data acquisition system and digitally integrated for 320 ns; the positions were digitally calculated with use of the Anger principle by a field programmable gate array. The calculated results were stored in a personal computer in list mode. The list mode data were sorted to form a 512 × 512 image with 128 channel energy information for each pixel by a computer. When the alpha-particle imaging detector is used for a portable imaging system, the weight-summed signals are fed to a small data acquisition system that provides a 45 × 45 matrix image with a fixed energy window.

2.3. Performance evaluation

For the performance evaluation of the imaging system, we measured