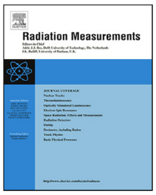
Radiation Measurements 82 (2015) 26–30

Contents lists available at ScienceDirect

Radiation Measurements

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



Characteristic X-ray detector: In-situ imaging of radioactive contaminant distributions

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HIGHLIGHTS

- We tested a characteristic X-ray detector (CXRD) to image Cs-137 distribution.
- The CXRD can make an image of Cs-137 contamination by detecting characteristic X-rays.
- The correlation between X-ray image and Cs137 contamination in-situ was tested.
- The CXRD has good performance to image hotspots in a contaminated area.

ARTICLE INFO

Article history:
Received 12 July 2014
Received in revised form
27 June 2015
Accepted 6 August 2015
Available online 12 August 2015

Keywords:
Characteristic X-ray detector
Gamma camera
Fukushima Dai-ichi nuclear power station
Radiation safety
Decontamination

ABSTRACT

The characteristic X-ray detector (CXRD), a CsI(Tl) scintillator with a 50-mm diameter, is a directional X-ray sensor that measures characteristic X-rays from radioactive material, such as ^{137}Cs, and identifies the direction of radioactive contamination. We evaluated a CXRD and visualized the distribution of radioactivity in the contaminated area near the Fukushima Dai-ichi nuclear power station, where the ambient dose equivalent rate was 2.1 μSv/h at 1 m above ground level. We found a good correlation between the characteristic X-ray fluxes and the distribution of radioactive contaminants with a 0.823 Pearson product–moment correlation coefficient.

1. Introduction

A significant amount of radioactive material was released into the environment by the Fukushima Dai-ichi nuclear power station (FDNPS) accident in March 2011, around which the Japanese government designated an evacuation zone (Baba, 2013). As of May 2014, efforts to decontaminate radioactive material from houses, roads, and agricultural fields were still ongoing in an effort to restore the environment and let residents return to their homes.

A portable gamma camera (GC) may help Fukushima decontamination efforts; it can be used to look for partially contaminated areas (hotspots) and confirm cleanliness after decontamination. Several groups have developed portable GCs since the 1990s and used them to assess radiation during uranium enrichment in nuclear power plants. They have also been used successfully in visualizing radioactive sources and waste (Guru et al., 1996; Gal et al., 1997; Woodring et al., 1999). Recently, new GCs using a semiconductor sensor have been developed (e.g. Ueno et al., 2013). However, the weight (~10–45 kg) and price (~10 million yen; ~100,000 US dollars) make them less accessible to public institutions, individuals, and companies who work on Fukushima decontamination. To reduce the weight of GCs, Compton cameras (CC) have also been explored as alternatives (Amman et al., 2009; Takeda et al., 2012; Kataoka et al., 2013; Shimazoe et al., 2013; Wahl et al., 2015). CCs that weigh about 2–13 kg are available for purchase but the price still exceeds 10 million yen (Matsuura et al., 2014). It would be a significant advancement to reduce the weight and price of GCs and CCs while maintaining sufficient sensitivity.

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http://dx.doi.org/10.1016/j.radmeas.2015.08.001
1350-4487/

Kobayashi et al. (2014) proposed a light-weight device to image radioactive contaminants from nuclear accidents, in particular the FDNPS accident, by detecting characteristic X-rays from ^{137}Cs (32 and 36 keV) instead of gamma rays (662 keV), which is the major FDNPS contaminants. By detecting characteristic X-rays instead of gamma-rays, the weight of a camera shield can be reduced because the mean free path of 32 keV X-rays in a shield material is ~0.01 of that of 662 keV gamma-rays. The probability of 32 and 36 keV characteristic X-ray emission from ^{137}Cs is 6.9% and is lower than that of 662 keV gamma-rays (85.1%). However, this disadvantage is compensated by the higher photo-peak efficiency for X-rays in comparison with that for gamma-rays. Kobayashi et al. (2014, 2015) developed a characteristic X-ray detector (CXRD), which is an X-ray sensor with a sensitivity for a specific direction to detect environmental ^{134}Cs and ^{137}Cs. Furthermore, a proto-type characteristic X-ray camera (CXRC) that is a pin-hole camera with a two-dimensional scintillator array (effective sensitive area ~200 cm^2) has been also developed and it successfully images environmental radioactivity (Kobayashi et al., 2015). The weight of the CXRC is 6.9 kg, which includes a sensor head (6.28 kg), an external 10,000 mAh Li-ion battery (0.26 kg) and a tablet PC (0.34 kg), and the reference price is <5 million yen. The battery provides ~3 h operation of the CXRC. However, evaluation of the sensitivity of these devices in-situ, where gamma-ray background is high, has not been sufficient. Sensitivity generally depends on gamma-ray background flux; CXRDs and CXRCs will be used in a high-background environment, which differs from that in a laboratory. In addition, the contamination around FDNPS is not a point source such as a check source, but is diffuse. In this paper, we evaluated the detector based on a characteristic X-ray detection method using the CXRD in situ, in the evacuation zone around FDNPS. The design and basic performance of the CXRD are described in Section 2 and the results of imaging performance tests around FDNPS are given in Section 3.

2. Design and performance of the CXRD used in the in-situ measurement

2.1. Design

The CXRD sensor was a disk (thickness = 1 mm, dia. = 50 mm) of CsI(Tl) (Hilger Crystals, Margate, UK) coupled with a 51-mm diameter photomultiplier tube (PMT R10131, Hamamatsu Photonics K.K., Shizuoka, Japan, effective sensitive area = 16.6 cm^2). It was installed in a 1.5-mm-thick cylindrical stainless steel vessel (all stainless steel used here is SUS304 as specified in Japanese industrial standards) (Fig. 1). A stainless steel multi-hole collimator (Fig. 1c) with nineteen 10-mm-diameter apertures and a length of 25 mm was prepared. The collimation coefficient is defined as: ρ = the length/the diameter = 2.5. A single-hole collimator (Fig. 1b) with a cylindrical stainless steel tube with an inner diameter of 62.5 mm and a thickness of 1 mm was also prepared ($\rho = 1.5$). Here the intrinsic efficiency is defined as the ratio of the number of X-ray photons which detected by the CsI(Tl) scintillator to the number of X-ray photons which perpendicularly enter the collimator hole(s). The intrinsic efficiencies are 0.53 ± 0.02 and 0.76 ± 0.02 for the single-hole collimator and the multi-hole collimator, respectively. The areas of the collimator hole(s) are 30.7 cm^2 and 14.9 cm^2, respectively. The collimators are removable and can be exchanged. Because the CsI(Tl) is shielded in all directions by stainless steel except for the collimator aperture(s), only X-rays that come through the collimator are detected. Thus, the CXRD has a directional sensitivity to characteristic X-rays in the energy range 10–60 keV. The CXRD direction can be adjusted precisely by a θ stage and a α gonio stage (SIGMA Koki Co.,Ltd., Tokyo, Japan) attached at the bottom of CXRD.

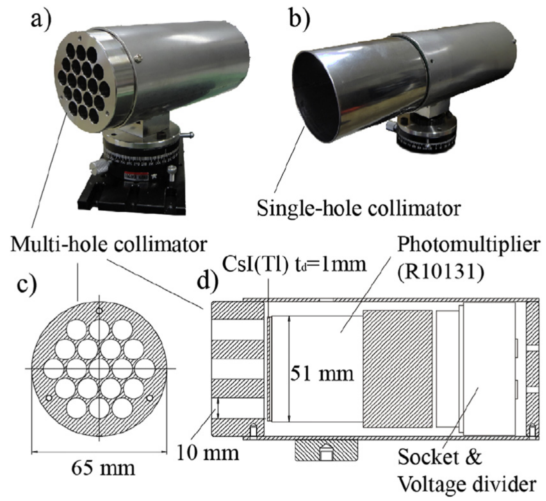

Fig. 1. A characteristic X-ray detector (CXRD). a) CXRD with a multi-hole collimator; b) CXRD with a single-hole collimator; c) Schematic drawing of a multi-hole collimator; d) Cross section of CXRD with a multi-hole collimator.

We used CsI(Tl) because of the low acquisition and handling costs compared with semiconductors. This feature is crucial when a CXRD is actually used for the decontamination work. The CsI(Tl) thickness is determined based on Monte Carlo simulations and optimized to increase the efficiency of detecting 32 and 36 keV, and to decrease gamma ray background according to Kobayashi et al. (2014). Consequently, we use 1-mm-thick CsI(Tl) with an average 92% photopeak efficiency for the characteristic X-rays from ^{137}Cs (32 and 36 keV). We used stainless steel for the vessel and collimator material, using a thickness determined to shield an average of 99% of 32 and 36 keV characteristic X-rays of ^{134}Cs and ^{137}Cs from outside the field of view (FOV). Achieving such a high shielding efficiency is difficult for a GC: 99% shielding efficiency of 662 keV gamma-rays from ^{137}Cs requires ~37-mm-thick lead shielding, which is very heavy to carry it. Lead (Pb) and tungsten (W) are commonly used to shield gamma rays, but are not suitable for a CXRD because they affect measurement of the 32 and 36 keV peaks of ^{134}Cs and ^{137}Cs. Pb and W also emit KX-rays with an energy around 70–80 keV when they are irradiated by gamma rays, which is typical around FDNPS. The escape peak energy that results from interacting Pb and W KX-rays with CsI(Tl) is about 35–50 keV, and partly overlaps with the ^{134}Cs and ^{137}Cs characteristic X-ray peaks.

The CXRD signal processing method is conventional. The PMT current signal is fed to a charge-sensitive preamplifier (595H, Clear Pulse Co., Ltd., Tokyo, Japan) with a decay time constant of 50 μs and a shaping amplifier (4417, Clear Pulse Co., Ltd.), with a time constant of 0.5 μs (in-situ measurement), or 2 μs (measurement in a laboratory) to decrease noise. Finally, the shaping amplifier signal pulse height was analyzed by a multi-channel analyzer (MCA-8000, AMPTEK Inc., Bedford, MA, USA) and the energy spectrum obtained. The PMT was operated at −582 V.

2.2. Basic performance

The experimental setup of the CXRD is shown in Fig. 2. First, to evaluate energy resolution, the CXRD was irradiated with a ^{137}Cs source (1.09 MBq) at a distance of 1 m and the energy spectrum obtained (Fig. 3). The peaks due to 32 and 36 keV X-rays were obtained with an energy resolution of 10.2 keV (full width at half maximum (FWHM) at 32 and 36 keV), where it was assumed to be a single Gaussian peak.