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A compact and high efficiency GAGG well counter for radiocesium concentration measurements

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ABSTRACT

After the Fukushima nuclear disaster, social concern about radiocesium (^{137}Cs and ^{134}Cs) contamination in food increased. However, highly efficient instruments that can measure low level radioactivity are quite expensive and heavy. A compact, lightweight, and reliable radiation detector that can inexpensively monitor low level radiocesium is highly desired. We developed a compact and highly efficient radiocesium detector to detect ~32 keV X-rays from radiocesium instead of high energy gamma photons. A 1-mm thick GAGG scintillator was selected to effectively detect ~32 keV X-rays from ^{137}Cs to reduce the influence of ambient radiation. Four sets of 25 mm × 25 mm × 1 mm GAGG plates, each of which was optically coupled to a triangular-shaped light guide, were optically coupled to a photomultiplier tube (PMT) to form a square-shaped well counter. Another GAGG plate was directly optically coupled to the PMT to form its bottom detector. The energy resolution of the GAGG well counter was 22.3% FWHM for 122 keV gamma rays and 32% FWHM for ~32 keV X-rays. The counting efficiency for the X-rays from radiocesium (mixture of ^{137}Cs and ^{134}Cs) was 4.5%. In measurements of the low level radiocesium mixture, a photo-peak of ~32 keV X-rays can clearly be distinguished from the background. The minimum detectable activity (MDA) was estimated to be ~100 Bq/kg for 1000 s measurement. The results show that our developed GAGG well counter is promising for the detection of radiocesium in food.

1. Introduction

A giant earthquake and subsequent tsunami devastated the east coast of Japan's Tohoku region on March 11, 2011. In the ensuing nuclear accident, vast amounts of radioactive materials escaped from the Fukushima Daiichi Nuclear Power Plant. The Nuclear and Industrial Safety Agency estimated that 160 PBq of ^{131}I and 15 PBq of ^{137}Cs fell on Japan. Since the half-life of ^{131}I is as short as eight days, the effect is now minor. However, since the half-lives of radiocesium (^{134}Cs and ^{137}Cs) are long (2 years for ^{134}Cs and 30 years for ^{137}Cs), threats from them remain, especially in the food [1–4] of the people living around the Fukushima area.

Instruments to measure low level radioactivity are quite expensive and heavy, and they are unavailable for general civil and/or small establishments. Moreover, since the measurement needs 0.5 L–2 L of a chopped sample, most samples become waste after the measurements except for such granular grains as rice and beans [5]. A compact, less expensive, lightweight food monitor using a smaller sample may be of interest.

Most radiocesium detectors measure high energy gamma photons (662 keV for ^{137}Cs, 605 keV and 794 keV for ^{134}Cs). The detection of such high energy photons requires high density and large detectors to improve the efficiency. Radiation shielding for environmental radiation is also required to reduce the detection limit, which makes the system larger and heavier.

One possible method to reduce the size and improve the detection efficiency is to measure the X-rays from ^{137}Cs and ^{134}Cs, which emit X-rays after beta decay. Table 1 summarizes the X-rays from radiocesiums [6]. The sum of the probabilities of 32 keV–37 keV X-ray emission is 6.86% and 0.84% for ^{137}Cs and ^{134}Cs, respectively. Detection of low energy X-rays instead of high energy gamma rays reduces the size of the detector system because these can be detected with a much thinner detector, which reduces the probability of the detection of environmental radiation and the detection limit. Here we present the first results from our new radiocesium detector based on this concept.

2. Materials and methods

2.1. GAGG well detector

We selected well counter configuration to maximize the efficiency of the detector. Fig. 1 shows a cross-sectional schematic

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Table 1
X-rays emitted from radiocesiums. X-rays energy ranges are from 31.8 keV to 37.4 keV and highest probability is 32.2 keV X-rays.

	Energy (keV)	Probability(%)	
		^{137}Cs	^{134}Cs
$K_{\alpha 1}$	31.8	1.95	0.24
$K_{\alpha 2}$	32.2	3.59	0.24
$K_{\beta 3}$	36.3		
$K_{\beta 1}$	36.4	1.06	0.13
$K_{\beta'' 5}$	36.7		
$K_{\beta 2}$	37.3		
$K_{\beta 5}$	37.3	0.26	0.03
$KO_{2,3}$	37.4		
	Sum	6.86	0.84

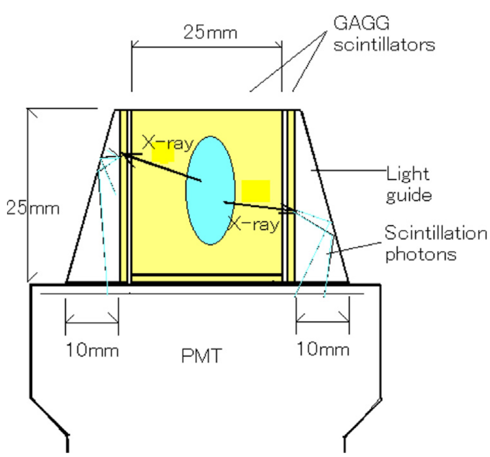

Fig. 1. Cross-sectional schematic of our developed GAGG well counter. X rays from subjects are detected by GAGG plates of sides and bottom.

of our developed well counter, which consists of Ce doped $Gd_3Al_2Ga_3O_{12}$ (GAGG) plates, light guides, and a photomultiplier tube (PMT).

We show performance comparison of possible scintillators [7–9] for proposed well counter in Table 2. CaF_2(Eu) is a possible candidate for our application. However, relatively long decay time (~900 ns) of CaF_2(Eu) limits count rate performance as well as this long decay time is not suitable for our digital data acquisition system. CsI(Tl) is slightly hygroscopic and has long decay time (1 μs). NaI(Tl) and $LaBr_3$ are suitable scintillators for our application because these have high light output. However the hygroscopic character of these scintillators will increase the fabrication cost for hermetic shield. In addition, the little natural activity in $LaBr_3$ is not suitable for our purpose of low background measurements. Because GAGG has high light output, no natural activity and is not hygroscopic, we selected GAGG for our well counter application [10].

The sample containing radiocesium is set inside the well counter and emits gamma photons and X-rays. The GAGG plates are thick enough to absorb the X-rays and produce scintillation light, which is transferred to the PMT through rectangular light guides.

Fig. 2(A) shows one set of a 1-mm-thick GAGG plate and a light guide used for the detector. The GAGG was 25 mm × 25 mm. The 1-mm thick GAGG can absorb most ~32 keV X-rays from the radiocesiums while minimizing the detection of environmental gamma photons, such as the scatter component of ^{40}K. Fig. 2 (B) shows one part of the assembled detector made from a GAGG plate and a light guide. All sides of the GAGG plate were polished. The GAGG was optically coupled to the rectangular side of the light

Table 2
Comparison of performance of possible scintillators for well counter.

Scintillator	GAGG	CaF_2:Eu	CsI:Tl	NaI:Tl	$LaBr_3$
Density (g/cm3)	6.63	3.19	4.5	3.67	5.08
Light yield (photons/Mev)	46,000	24,000	60,000	45,000	63,000
Decay time (ns)	90	900	1000	230	16
Peak Emission (nm)	520	425	545	415	380
Hygroscopic	no	no	little	yes	yes
Natural activity	no	no	no	no	little

guide, which effectively transfers the scintillation light to the photomultiplier tube (PMT).

Fig. 2(C) shows the detector part of our GAGG well counter. Four sets of assembled GAGG detectors were optically coupled to a 2-in. square PMT (Hamamatsu Photonics: R6236) to form the side wall part of a rectangular-shaped well counter. Another GAGG plate was optically coupled to the PMT directly to form its bottom part. All the surfaces except the bottom part were wrapped with Teflon tape as reflectors. The bottom part was covered with white paper to reduce the reflection because the light collection for the PMT of the bottom part was larger than the GAGG of the side parts. The GAGG plates, the light guides, and the PMT were optically shielded by black tape. The volume that can be measured by the well counter was ~10 cm^3.

2.2. GAGG well counter system

A schematic diagram of the entire GAGG well counter system is shown in Fig. 3. The system consists of a detector, a data acquisition system, and a personal computer (PC). The detector part of the GAGG well counter was contained in a 10-mm-thick lead shield to reduce the influence of the ambient radiation. The PMT signal was amplified and fed to a 100-MHz analog to digital (A–D) converter of the data acquisition system. A–D converted signals higher than the threshold level (40 mV) are digitally integrated for 320 ns. The counts within the energy window (±50%) are transferred to a PC and stored. Also, the data acquisition system can acquire and display the energy spectrum and set the energy window from the PC [11]. Fig. 4 show a photo of the developed GAGG well counter system. The GAGG well counter system weights ~30 kg. Most of the weight depends on the lead shield around the detector.

2.3. Performance evaluation

2.3.1. Energy resolution

Energy resolution reduces the background counts from environments. With higher energy resolution, we can reduce the energy window width and minimize the background counts from environmental gamma photons [9]. For this purpose, we measured the energy distribution for low energy gamma photons for 122 keV gamma photons from ^{57}Co as well as ~32 keV X-rays from ^{137}Cs and estimated the energy resolution.

2.3.2. Stability

The system stability is important because the quantitative measurements of the radiocesium activity are the goal of this system. We measured the count rate of the X-rays from ^{137}Cs (half-life: 30 year) for 4 h and evaluated the count rate change.

2.3.3. Background counts

The background counts are important for this type of detector because they determine the minimum detectable counts or activity of the system. We measured the energy spectrum for the

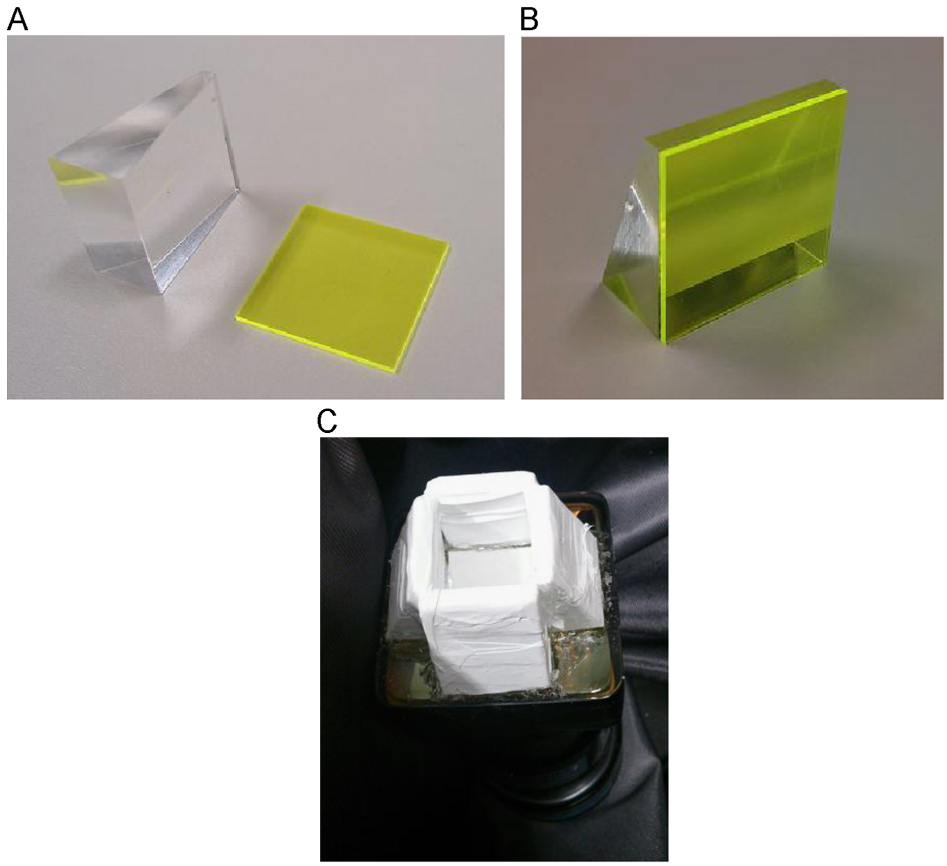

Fig. 2. Photographs of materials. GAGG of 1-mm-thick plate and light guides (A), assembled GAGG plate and light guide (B), and developed detector part of GAGG well counter (C).

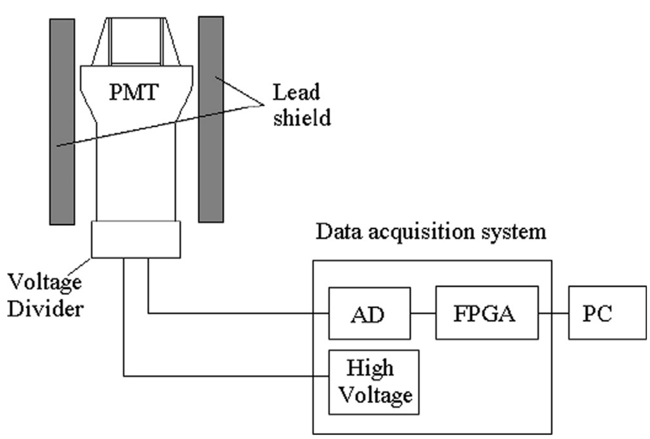

Fig. 3. Block diagram of developed GAGG well counter system. Signal from PMT is fed to analog-to-digital (A-D) converter and field-programmable gate array (FPGA) for processing signals.

background counts without any activity inside the detector for 3600 s. The background counts within the energy window were also used to calculate the minimum detectable counts and the activity of the system.

2.3.4. Measurement of radiocesium sampled in Fukushima

To estimate the counting efficiency for ~32 keV X-rays from radiocesium, we prepared solution including ^{137}Cs (9.2 Bq) and ^{134}Cs (7.3 Bq) whose ^{134}Cs/^{137}Cs ratios resembled the Fukushima samples and measured its energy spectrum for 3600 s. The counts within the energy window were also used to calculate the detection efficiency (DE) of our GAGG well counter system with following equation:

Fig. 4. Developed GAGG well counter system. Sample is set inside hole of upper side of counter. High voltage supply is included in system.

$$DE = CR/TRA \tag{1}$$

where CR: count rate (cps) of the GAGG well counter TRA: total radioactivity of the ^{137}Cs and ^{134}Cs solution

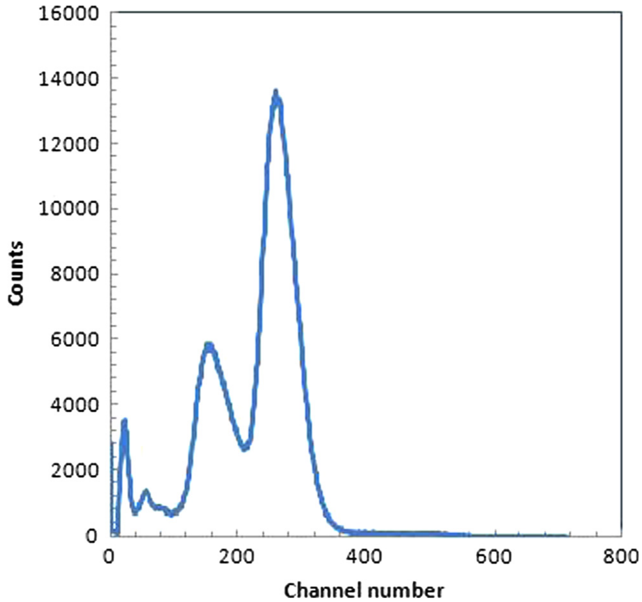

Fig. 5. Energy spectrum of GAGG well counter for 122 keV gamma photons. Clear photo-peak was observed with energy resolution of 22.3% FWHM.

2.3.5. *Detection limit calculation*

The detection limit (DL) of the system with 1000 s acquisition time was evaluated by the following equation [12]:

$$DL = 3\sigma_B = 3\sqrt{N_{B1000s}} \quad (2)$$

where σ_B: standard deviation of blank counts and N_{B1000s}: blank counts for 1000 s acquisition.

We used the background count rate to estimate the DL of the GAGG well counter.

3. Results

3.1. *Energy resolution*

Fig. 5 shows the energy spectrum for ^{57}Co gamma photons measured by the GAGG well counter. The energy resolution was 22.3% FWHM for 122 keV gamma photons.

Fig. 6 shows the energy spectrum for ~32 keV X-rays from ^{137}Cs. We observed a clear peak from ~32 keV X-rays from ^{137}Cs. The photo-peak channel was higher than the noise level of the signal, indicating the detection of this lower energy signals is possible without electrical noise of the system. The energy resolution was 32% FWHM.

3.2. *Stability*

The stability of the GAGG well counter is shown in Fig. 7. The raw count rate data was averaged for 100 s to reduce the statistical deviation. The stability was within ±6.1%, including statistical deviation.

3.3. *Background counts*

Fig. 8(A) shows the energy spectra for the background counts. We did not observe any obvious radiocesium peaks in the low energy level of the spectrum. The background counts within the energy window were 860 counts per 3600 s, so the background count rate was calculated to be 0.24 cps.

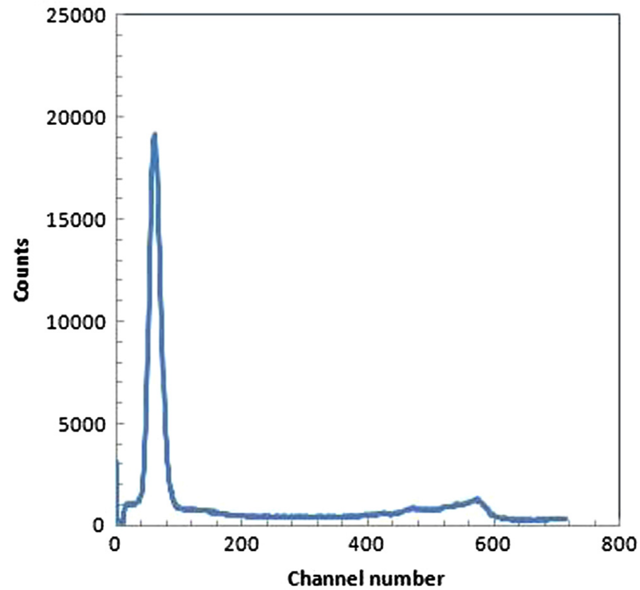

Fig. 6. Energy spectrum for ~32 keV X-rays from ^{137}Cs. The energy resolution was 32% FWHM.

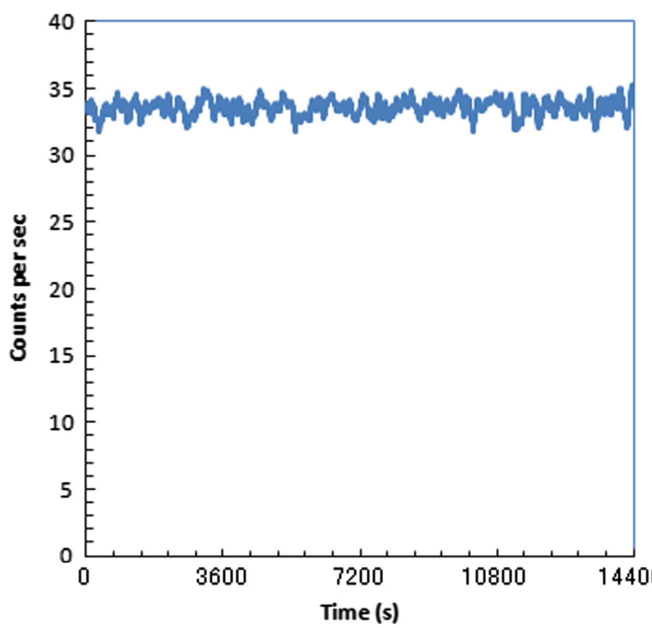

Fig. 7. Stability of the GAGG well counter. No systematic change was observed.

3.4. *Measurement of radiocesium sampled in Fukushima*

Fig. 8(B) shows the energy spectra for the radiocesium solution. In the measurements, the photo-peak of ~32 keV X-rays is clearly distinguished from the background (Fig. 8(A)). The counting efficiency for the radiocesium mixture was estimated to be 4.5% for ~32 keV X-rays.

3.5. *Detection limit calculation*

The detection limit of our system with 1000 s acquisition time was evaluated by Eq. (1). The detection limit with 1000 s measurement time was calculated to be 46.3 counts.

The counts for 1000 s are 45 with the 4.5% detection efficiency system for 10 cm^3 volume of 100 Bq/kg radiocesium sample. Thus, the minimum detection activity of this GAGG well counter with

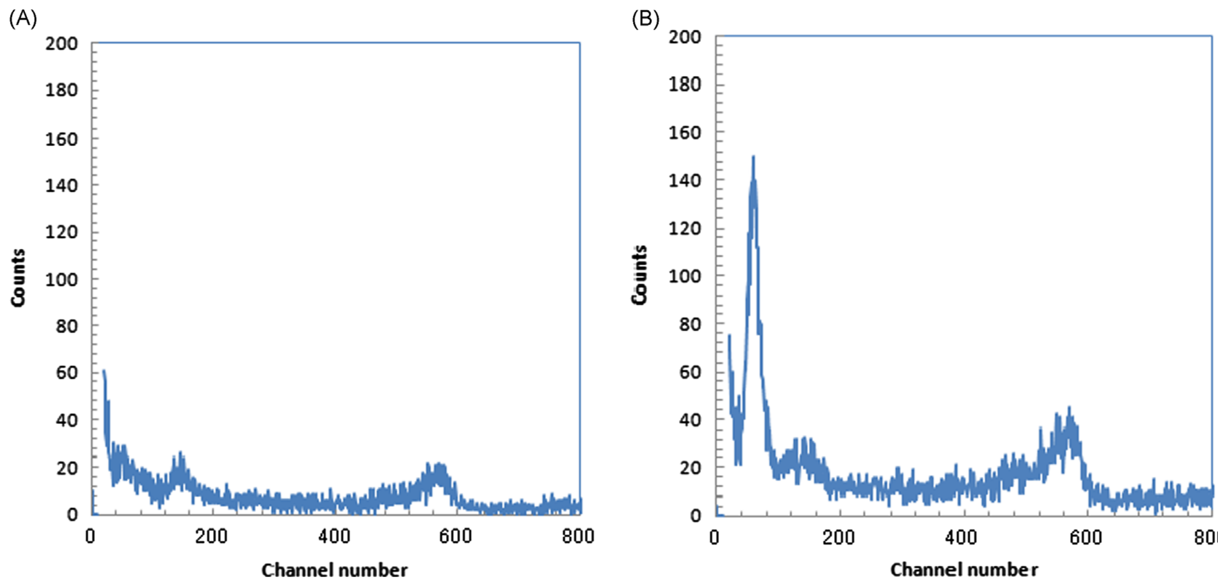

Fig. 8. Energy spectra for background (A) and radiocesium solution (B). We observe photo-peak of ~32 keV gamma photons in left side of Fig. 8(B).

10 cm^3 sample was approximately 100 Bq/kg with 1000 s acquisition time.

4. Discussion

We successfully developed a GAGG well counter for radiocesium detection. With this system, we detected ~32 keV X-rays from radiocesium with a low background count rate of 0.24 cps. With this background count rate, the minimum detection concentration for a 10-gr sample was ~100 Bq/kg, which is larger than the minimum requirement for radiocesium food monitors in Japan (25 Bq/kg) [13]. However, the minimum detection concentration depends on the total measured volume by the system. Thus, by increasing the total volume for the GAGG well counter, the minimum detection concentration will be reduced. For example, if we increase the total volume of the GAGG well counter for 4 times (40 cm^3), the minimum detection concentration will be 1/4 or ~25 Bq/kg, which is smaller than the minimum requirement for food monitors in Japan.

Another possible method to reduce the detection limit of our GAGG well counter is by increasing the shield to reduce the detection of gamma photons from outside the well counter. However, increasing the shield also increases its weight and negates one advantage of our system.

Although our GAGG well counter system was developed for radiocesium detection, it can be applied to other radioisotopes, such as single photon emitters ^{99m}Tc or ^{125}I. The high energy resolution of our developed system is also useful to set a narrower energy window for other radioisotopes.

5. Conclusion

We successfully developed a small, lightweight GAGG well counter system that showed considerable performance for detecting low level radiocesium contaminating samples. The results show that our developed GAGG well counter is promising for the detection of radiocesium in food.

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