Identification of ^{90}Sr/^{40}K based on Cherenkov detector for recovery from the Fukushima nuclear accident

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Although five years have passed since the Fukushima nuclear accident of 2011, the local fisheries have yet to recover from its effects. One reason for this situation is the difficulty of measuring the radioactivity owing to ^{90}Sr in seafood. After the accident, the radioactivity due to Cs isotopes in samples was measured with precision, which facilitated the enforcement of the maximum concentration of Cs radioisotopes in food at 100 Bq/kg, as defined by the Ministry of Health, Labour and Welfare in Japan. However, ^{90}Sr is more dangerous than Cs isotopes because it has an effective half-life of 18 years and accumulates in the bone. The radioactivity owing to ^{90}Sr in a sample is difficult to measure because the beta rays from ^{137}Cs or ^{40}K also contribute to the signal. When measured based on the endpoint pulse height as determined by a conventional survey meter, the beta ray signal from ^{90}Y (daughter of ^{90}Sr) cannot be differentiated from the beta rays from other sources. To overcome this difficulty, in this study, we develop a Cherenkov detector based on a silica aerogel with a refractive index of 1.034 that can identify beta rays from ^{90}Y within a background of beta rays from ^{137}Cs and ^{40}K. This instrument involves a detector that is sensitive to beta rays from ^{90}Sr but less sensitive to radiation from other sources. This detector comprises a trigger counter that uses scintillating fibers, an aerogel Cherenkov counter with wavelength-shifting fibers, and a veto counter to suppress cosmic rays. We characterize the detector using a ^{90}Sr source, ^{137}Cs source, and pure potassium chloride reagent of 16.6 Bq/g, where the radioactivity of natural ^{40}K is estimated to be 31.7 Bq/g. The following results are obtained: the absolute detection efficiency for ^{90}Sr, ^{137}Cs, and ^{40}K is $[2.24 \pm 0.01\ (\text{stat}) \pm 0.44\ (\text{sys})] \times 10^{-3}$ Bq^{-1}s^{-1}, $[1.27 \pm 0.08\ (\text{stat}) \pm 0.25\ (\text{sys})] \times 10^{-6}$ Bq^{-1}s^{-1}, and $[5.05 \pm 2.40\ (\text{stat}) \pm 0.15\ (\text{sys})] \times 10^{-5}$ Bq^{-1}s^{-1}, respectively. To aid in the recovery efforts for the Fukushima accident, this study reports the current development status of this Cherenkov-radiation-based detector.

KEYWORDS: Strontium-90, Cherenekov detector, Fukushima

1. Introduction

Five years have passed since the Fukushima nuclear accident in 2011. As part of the recovery effort, a recent study focused on detecting soluble, longer-lived radioisotopes such as ^{90}Sr and ^{137}Cs. The Fukushima accident caused greater damage to local fisheries than to local livestock or agriculture [1]; the fisheries have yet to recover. This situation is mainly attributed to the fact that measuring radioactivity due to ^{90}Sr remains difficult, so foodstuffs may contain ^{90}Sr without it being detected.

After the accident, methods were developed to measure the radioactive concentration of cesium isotopes (radio-cesium) in samples with precision [1–3], which facilitates the enforcement of the maximum radioactive concentration of radio-cesium in food at 100 Bq/kg, as defined by the Ministry of Health, Labour and Welfare in Japan. This concentration leads to a dose that is less than that due

to internal exposure from natural ^{40}K (0.18 mSv/year). However, the maximum radioactive concentration of ^{90}Sr is not set: if it is set at the same level as radio-cesium, the resulting estimated dose is more dangerous than that of radio-cesium, because ^{137}Cs and ^{90}Sr have effective half-lives time of 70 days and 18 years and a maximum energy deposition of 1.17 and 2.28 MeV, respectively. In addition, Cs is distributed throughout the body whereas Sr is focused in the bone. Based on chemical surveillance of food, flatfish were reported to have a maximum ^{90}Sr contamination concentration of 0.46 ± 0.05 Bq/kg with the relative concentration ^{90}Sr/(^{134}Cs +^{137} Cs) = 0.027 [4]. Here the sample must be rapidly inspected a few percent of ^{90}Sr concentration compared with the limit of radio-cesium. Thus, detecting the ^{90}Sr concentration in food with a quantitative lower limit of detection of a few Bq/kg requires a detection limit of 1 Bq/kg.

Such a detector would require the following capabilities to ensure the recovery of fisheries in Fukushima: it must measure an individual sample individually within an hour, it must function properly anywhere outside of the radiation-controlled area, and it must be as simple to handle as a survey meter. A conventional chemical method exists to estimate the radioactive concentration of ^{90}Sr [5]. However, this method cannot be directly applied to the Fukushima recovery because it is a destructive method and requires long measuring time (on the order of a month). More rapid conventional methods involve measuring the surface contamination of a sample using a Geiger-Müller chamber and measuring a range of beta rays using a collimator; the detection limits of such instruments for ^{90}Sr are a few Bq/g [6, 7]. Measuring the radioactivity by a conventional spectroscopic method is difficult because ^{90}Sr and its daughter ^{90}Y only emit beta rays, which have a continuous energy spectrum and cannot be differentiated from beta rays from other radionuclides. In addition, estimating the radioactivity using the pulse-height endpoint (with a maximum energy of 2.28 MeV) as determined by a conventional survey meter is difficult because higher-energy cosmic muons may contaminate the signal. In the present study, therefore, beta rays from ^{137}Cs and ^{40}K (with maximum energy of 1.17 and 1.31 MeV) and cosmic muons are the main background when measuring the activity of ^{90}Sr. To measure the radioactive concentration of ^{90}Sr, we are thus developing a real-time ^{90}Sr counter that is sensitive only to ^{90}Sr. This detector, based on Cherenkov radiation, suppresses background radiation from other sources. This study discusses the development status of this detector.

2. Materials and Detector Design

We developed a real-time ^{90}Sr counter that consists of a trigger counter based on scintillating fibers, an aerogel Cherenkov counter with wavelength-shifting fibers, and a veto counter to suppress the background events due to cosmic rays [8, 9]. The prototype counter has an effective footprint of 300 mm $\times$ 100 mm. In this section, we describe in detail the detector components and its design.

2.1 Scintillating fibers for trigger

A sheet made of scintillating fibers was adopted to ensure that minimal energy is deposited in the trigger. The fibers (type: SCSF-78MJ) are made by Kuraray Co., Ltd. and are 0.2 mm in diameter with a double-cladding structure and a trapping efficiency of 5.4% [12]. The trapping efficiency is the percent of photons that undergo total internal reflection from one side of the fiber. Both fiber ends are bundled and optically connected to photomultiplier tubes (PMTs). The PMTs are manufactured by Hamamatsu Photonics K.K. (R9880U-210) and have an 8-mm-diameter photocathode [13]. All PMTs used in this study were of this type. The detection efficiency of the trigger counter was determined to be 54.4% using a ^{90}Sr source.

2.2 Aerogel Cherenkov counter with wavelength shifting fibers

Cherenkov radiation is a kind of shock wave. In a material with index of refraction n, Cherenkov photons are emitted when the velocity v of a charged particle exceeds that of a photon in that material.

The Cherenkov condition is $v > c/n$, where c is the speed of light in a vacuum. From the relation between the kinetic energy K of an electron and the velocity ratio of electrons to photons ($\beta = v/c$), we determined that an index of 1.018-1.042 for electrons with kinetic energy of 1.31-2.28 MeV would satisfy the Cherenkov condition. Silica aerogel tiles with an index of 1.034 [10] were used for the test; these are 30 mm thick and have an optical transmission length of 53 mm at 400 nm. A Cherenkov counter that uses silica aerogel as radiator is called an aerogel Cherenkov counter (AC); such counters can identify beta rays from ^{90}Y and measure the radioactivity of ^{90}Sr [11].

Cherenkov photons are observed via a light guide consisting of a wavelength-shifting fiber (WLSF). After the WLSFs absorb photons, they reemit photons redshifted to a longer wavelength. These emitted photons propagate to both ends of the fibers by undergoing total internal reflection at the fiber walls. Therefore, the fiber wall plays a role in determining the photosensitive area. The fibers are made of two types of WLSFs: B-3 and Y-11, manufactured by Kuraray Co. Ltd. They are 0.2 mm in diameter, have an attenuation length of approximately 1 m and a double cladding structure, and offer a trapping efficiency of 5.4%. B-3 and Y-11 have a purple-to-blueshift and a blue-to-greenshift, respectively [12]. Using the WLSFs as light guides, the optical collection efficiency reaches about 10%, and a photosensitive area of 300 cm^2 can be read out using smaller PMTs, where the collection efficiency contains the trapping efficiency, absorption efficiency, and emission efficiency for Cherenkov photons in WLSFs. The absorption and the emission efficiency depend on wavelength. Because the range of absorption covered the wavelength of Cherenkov photons when using two kinds of fibers, the collection efficiency recovered to approximately twice the trapping efficiency. As shown in Fig. 1, the fiber light guide was set downstream of the silica aerogel.

In addition, noise in the system can be suppressed. Such noise is caused by two types of gamma rays. When the gamma rays interact with the material upstream of the photocathode, electrons are knocked out of the material. One source of noise is caused by these electrons entering the PMT photocathode. Another source of noise is Cherenkov emission from these electrons. The probability of the noise is proportional to the area of the photocathode and inversely proportional to the square of the distance between the samples and the photocathode. Therefore, designing WLSFs with smaller PMTs can suppress this noise in comparison with the noise obtained when using a bigger PMT.

2.3 Veto counter to suppress cosmic rays

A veto counter was set over the AC to suppress the background events due to cosmic muons. The shielding block (made of aluminum) set between the veto counter and the AC, clearly prevents all electrons emitted from the sample from entering the veto counter. The veto counter itself consists of a 400 mm × 200 mm × 5 mm plastic scintillator bar, WLSF, and a PMT. The WLSFs are connected to the four sides of the scintillator, bundling both ends of the fibers into one. The PMT is connected to the bundled fibers, which allows scintillation light to be detected. In a bench test using cosmic muons, and average of 6.6 photoelectrons were detected by the PMT.

2.4 Detector design

Figure 2 shows the detector design. The sample was ground to an approximately 1-mm-thick paste and set under the trigger counter. The 10-mm-thick silica aerogel tiles were stacked into three layers to stop the 1.31 MeV beta ray. Electrons ejected via Compton scattering with gamma rays of energy less than 1.72 MeV also do not satisfy the condition for Cherenkov radiation in the silica aerogel with an index of 1.034. Thus, the detector is less sensitive to gamma rays originating from ^{137}Cs (0.66 MeV), ^{134}Cs (maximum energy of 1.37 MeV), and ^{40}K (1.46 MeV). A beta ray from ^{90}Y in the detector is counted when the trigger counter and the AC are triggered and the veto counter is not triggered.

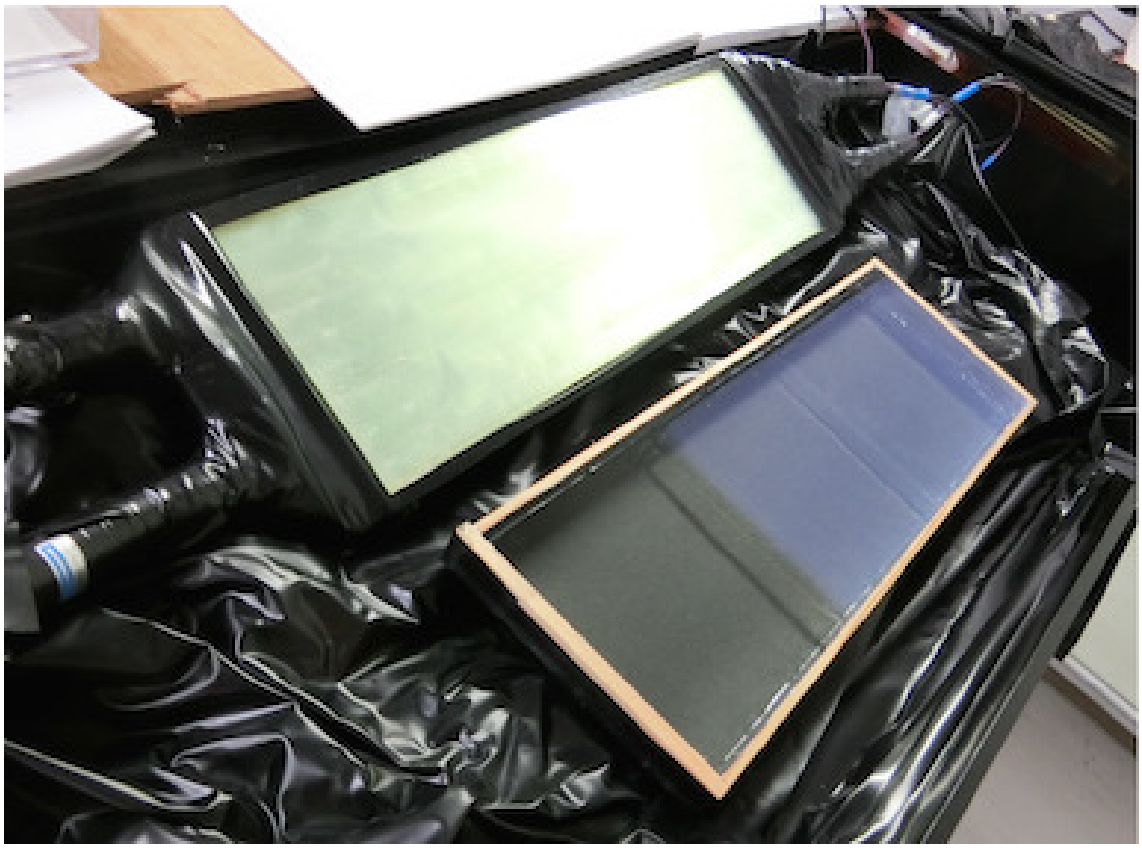

Fig. 1. Installed silica aerogel tiles and WLSF light guide coupled with PMTs.

3. Results and Method for Identifying ^{90}Sr

The radioactivity of sources is estimated using

$$A(t) = A_0 \, \mathrm{e}^{-\mathrm{t} \ln 2/\tau}, \tag{1}$$

where A_0 = 37 kBq (±20%) as per the Japan Radioisotope Association, t is measured in years since the last calibration, and τ is the half-life of the substance in question. The activity of ^{90}Sr (A_{Sr}) and ^{137}Cs (A_{Cs}) was calculated to be 23.8 ± 4.7 (sys) and 26.0 ± 5.2 (sys) kBq, respectively.

Pure potassium chloride (KCl) was used as a ^{40}K source. KCl has stable mass and an activity of 16.6 Bq/g (from ^{40}K). KCl with a purity exceeding 99.5% was obtained from Hayashi Pure Chemical Ind. Ltd. [14]. A KCl source with a mass of 4.0 ± 0.1 g (and an activity of 66 ± 2 (sys) Bq) was used for the test.

Then, 1 h measurements of the background counts gave N_{BG} = 190 ± 5. For these measurements, each source was placed under the center of the detector. The counts N_{Sr}, N_{Cs} and N_{K} were 191752 ± 535, 309 ± 5 and 202 ± 4, respectively (N_x is the counts per hour from source x, where x = ^{90}Sr, ^{137}Cs, or ^{40}K). The errors were estimated from standard errors ($\sigma/\sqrt{n}$, where σ is the standard deviation).

The absolute detection efficiency is defined as the ratio of count rate and the activity of the given sample. The efficiency η of the radionuclides is

$$\eta_{\mathrm{Sr}} \equiv \frac{N_{\mathrm{Sr}} - N_{BG}}{A_{\mathrm{Sr}} T}, \tag{2}$$

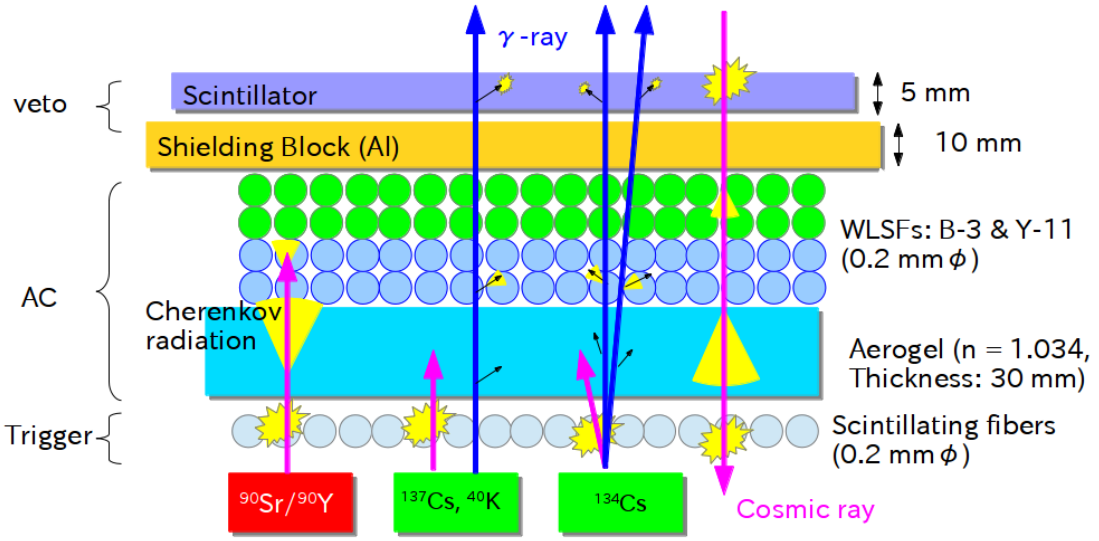

Fig. 2. The sketch of the structure of the real-time ^{90}Sr counter comprising the trigger counter (trigger) using scintillating fibers, the aerogel Cherenkov counter (AC) with wavelength-shifting fibers (WLSFs), and the veto counter (veto) for suppression of cosmic rays. The magenta, blue, and black arrows represent the charged particle, gamma ray and knocked-out electron, respectively. The yellow cone shape and spreading shape denote Cherenkov radiation and scintillation emission, respectively.

where $N_{\rm Sr}$ is the number of counts on the ^{90}Sr source, N_{BG} is background counts, $A_{\rm Sr}$ is the activity of the ^{90}Sr source, and $T = 3600$ s. Table I shows the efficiencies for ^{90}Sr, ^{137}Cs and ^{40}K.

Table I. Absolute efficiencies

Parameter	Value (Bq^{-1}s^{-1})
$\eta_{\rm Sr}$	$[2.24 \pm 0.01\ (\text{stat}) \pm 0.44\ (\text{sys})] \times 10^{-3}$
$\eta_{\rm Cs}$	$[1.27 \pm 0.08\ (\text{stat}) \pm 0.25\ (\text{sys})] \times 10^{-6}$
$\eta_{\rm K}$	$[5.05 \pm 2.40\ (\text{stat}) \pm 0.15\ (\text{sys})] \times 10^{-5}$

4. Discussion

The sample under inspection should basically be heated and compressed to a thickness of 1 mm. The detector has an effective area of 300 cm^2. For a sample density of 1 g/cm^3, 30 g is the maximum mass of heated sample that can be measured. Seafood and seawater are approximately 70% and 99% or more pure water, respectively, and the compressibility is 0.3 for seafood and 0.01 for seawater. As per the Japanese Government, the sample should have an activity of 100 Bq/kg due to ^{137}Cs. In nature, seafood and the seawater have an activity due to ^{40}K of approximately 150 and 12.1 Bq/kg, respectively. In this section, we discuss the detection limit of ^{90}Sr activity.

The activity of ^{90}Sr, ^{137}Cs, and ^{90}K in a unit-mass sample are labeled $A'_{\rm Sr}$, $A'_{\rm Cs}$, and $A'_{\rm K}$, respectively. We assume $A'_{\rm Cs} = 100$ Bq/kg and $A'_{\rm K} = 150$ (12.1) Bq/kg for seafood (seawater). When the sample is placed in the detector, the count is given by

$$N = \left(\eta_{\rm Sr}A'_{\rm Sr} + \eta_{\rm Cs}A'_{\rm Cs} + \eta_{\rm K}A'_{\rm K}\right)m\varepsilon^{-1}T + N_{BG}, \tag{3}$$

where m denotes the sample mass of 30 g, ε is compressibility of 0.3 (seafood) or 0.01 (seawater), and T is inspection time of 3600 s. The background count is given as by

$$N'_{BG} = \left(\eta_{\mathrm{Cs}} A'_{\mathrm{Cs}} + \eta_{\mathrm{K}} A'_{\mathrm{K}}\right) m\varepsilon^{-1} T + N_{BG}. \tag{4}$$

The detection limit of the activity of ^{90}Sr (A^{min}_{Sr}) is calculated to be

$$A^{min}_{\mathrm{Sr}} = \frac{3\sqrt{N_{BG} + (\eta_{\mathrm{Cs}} A'_{\mathrm{Cs}} + \eta_{\mathrm{K}} A'_{\mathrm{K}}) m\varepsilon^{-1} T}}{\eta_{\mathrm{Sr}} m\varepsilon^{-1} T}, \tag{5}$$

Kaiser's theorem [15]: $N > N'_{BG} + 3\sqrt{N'_{BG}}$. As the result, the detection limit is estimated to 52 ± 8 (stat) ± 10 (sys) Bq/kg (seafood) and 1.7 ± 0.3 (stat) ± 0.3 (sys) Bq/kg (seawater).

Therefore, the detector can detect a radioactive concentration of ^{90}Sr in seawater above 1.7 ± 0.3 (stat) ± 0.3 (sys) Bq/kg, which has nearly sufficient performance for inspecting seawater. However, this performance is insufficient for inspecting seafood. Equation (5) shows that the detection limit depends on the sample mass, so extending the effective area should improve the detector performance. Furthermore, the veto counter was placed on the AC, which is problematic because it cannot suppress muons that enter from the sides. Thus, the detector performance should be improved by adding scintillator bars to side faces.

5. Conclusion

This study presents the status of the development of a real-time ^{90}Sr counter based on Cherenkov radiation to aid in the recovery of fisheries in Fukushima. The prototype detector has an effective area of 300 mm × 100 mm and is sensitive to ^{90}Sr but less sensitive to ^{40}K, ^{137}Cs, or cosmic muons. The following results are obtained: the absolute detection efficiency for ^{90}Sr, ^{137}Cs, and ^{40}K is [2.24 ± 0.01 (stat) ± 0.44 (sys)] × 10^{-3} Bq^{-1}s^{-1}, [1.27 ± 0.08 (stat) ± 0.25 (sys)] × 10^{-6} Bq^{-1}s^{-1}, and [5.05 ± 2.40 (stat) ± 0.15 (sys)] × 10^{-5} Bq^{-1}s^{-1}, respectively. The detector performance was estimated by testing it on radioactive sources of ^{90}Sr (23.8 ± 4.7 kBq), ^{137}Cs (26.0 ± 5.2 kBq), and ^{40}K (66 ± 2 Bq). In future work, we plan to fabricate the next-generation detector with an effective area of 500 mm × 200 mm or with veto counters added to the sides of the AC to suppress cosmic muons.

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