On residual ^{137}Cs on shallow rugged reefs lying inshore of Fukushima: Part 2: Methodological improvements

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Abstract

A newly improved underwater CsI(Tl) gamma-ray spectrometer was developed for in situ and stationary measurements of spectra of radiations on shallow rugged reefs lying inshore of Fukushima. Examination of the spectrometer using ^{40}K in a KCl test source and in slabs of granite justified the present method to confirm contact configuration between the detector and source. Furthermore, a spectrum of the Compton continuum due to coexisting ^{137}Cs and ^{134}Cs with the current ratio of ^{134}Cs/^{137}Cs was obtained to determine to a true energy spectrum of radiations on and near the interface between the contaminated surface of rock and seawater from a measured gamma-ray spectrum. The detector assembly was very useful in a situation where the depth was less than 30 m, the level of contamination was above 5×10^3 Bq/m^2 of ^{137}Cs, and an allocated time of measurement at a point was about ten minutes.

Keywords: underwater CsI(Tl) gamma-ray spectrometer, in situ and stationary measurement, ^{137}Cs, shallow rugged reefs.

Introduction

The Great East Japan Earthquake and subsequent tsunami hit the Fukushima Daiichi Nuclear Power Plant (FD1NPP) of the Tokyo Electric Power Company on March 11th, 2011 [1, 2]. As a consequence, heavily contaminated cooling water of the crippled nuclear reactors flowed toward the Pacific Ocean through shallow rugged reefs lying close to the coastline of Fukushima. A survey of contamination with anthropogenic radioactive nuclides on the reefs has required us to develop new methodological ideas because of the meter-scale ruggedness of the reefs.

Developments of underwater gamma-ray spectrometry and its application to in situ measurements of gamma rays from radioactive nuclides in the marine environment were comprehensibly reviewed by Jones [3], Osvath and Povinec [4], and Povinec et al. [5]. As early as 1996, gamma-ray energy spectra on the seafloor of the Irish Sea and that of the Kara Sea were measured with a high-purity Ge detector and a NaI(Tl) detector by Povinec et al. [6]. They identified natural and anthropogenic radioactive nuclides on the seafloor by the clearly observed photopeaks. Towed gamma-ray spectrometers were also developed to measure in situ and continuous distributions in a wide area, as described in the above-cited review articles. Right after the disaster of the FD1NPP, Thornton et al. [7, 8] developed a towed gamma-ray spectrometer. In their study, a standard combination of a NaI(Tl) scintillator and a photomultiplier tube with a signal processing module was accommodated into a flexible hose to be towed by a ship. They successfully carried out in situ and continuous measurements to survey the distribution of ^{137}Cs on the seafloor along the coastline of Fukushima. However, the meter-scale ruggedness of the reefs had denied traditional methodology developed for soft sea floor until a prototype of underwater CsI(Tl) spectrometer which aimed at the contact configuration between the detector and a flat part of rock surface was reported by Suzuki et al. [9] (hereafter called our previous study). In that study the coexisting photopeak of ^{40}K was used as an indicator to confirm the contact configuration.

In this report, we describe a newly improved underwater CsI(Tl) gamma-ray spectrometer for in situ and stationary measurements of radiations on the interface between rock and seawater in the rugged reefs lying inshore of Fukushima. The levels and distribution of ^{137}Cs measured with this spectrometer are found in our companion paper by Suzuki et al. [10] (hereafter called Part 1).

Experimental

1. Justification of the use of the photopeak of ^{40}K for confirmation of contact configuration

As in our previous study, the contact between the detector and the flat part of the surface of rock is confirmed by measuring the count rate of the photopeak of coexisting ^{40}K ($T_{1/2}$ =1.25×10^9 y). The justification of the method given in our previous study, however, might need to be more convincing. In this paper, we have re-examined the rationale using ^{40}K in a KCl test source and in the slabs of granite obtained from a quarry in the northern part of the Abukuma Mountains located about 20 km inland along the coastline. The description of the details, such as the detector response to the KCl test source, determination of the activity of ^{40}K in the slabs of Abukuma granite, interpretation of near uniformity of the count rate of the photopeak of ^{40}K at all measurement points, and the overall uncertainty in the levels of ^{137}Cs, is rather lengthy for the main text. Therefore, it is contained in the Supplementary information. However, for the reader’s convenience, the most important conclusion is cited here; the overall uncertainty in the measured levels of ^{137}Cs is ± 10 % at most.

2. CsI(Tl) scintillator

A conceptual illustration of our in situ and stationary measurement of spectra of radiations on the rugged reefs is shown in Fig. 1. In order to attain the overall detection efficiency and stability better than those of our previous spectrometer described in Ref. [9], we have introduced a commercially available radiation detection module C12137-10 produced by Hamamatsu Photonics K.K. [11]. The centerpiece of the module is a scintillator crystal of CsI(Tl), the size of which is 25 mm in height and 110 mm in diameter coupled with a high-gain multi-pixel photon counter (MPPC). The volume of the scintillator is 19 times larger than that of our previous spectrometer.

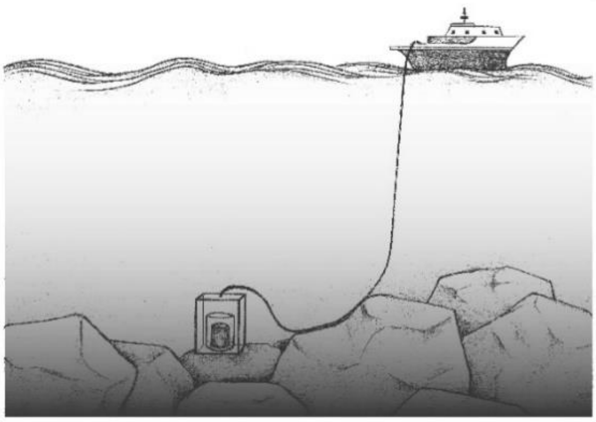

Fig. 1 Conceptual illustration of the in situ and stationary measurement

The detector assembly is lowered down manually and lifted up with a motor-drivable winch. The boat is not anchored, but the drift due to surface winds and currents is continuously corrected with engine and steering.

3. Pressure-tight vessel and cage

A pressure-tight vessel (PTV) to house the module is a hollow cylinder, 204 mm in diameter and 200 mm in height, made of aluminum, and the thickness of the wall is 15 mm. The bottom of the cylinder is a 30 mm thick aluminum plate. The central part of the bottom, which is 125 mm in diameter, is mechanically thinned to 1 mm thickness for a window to accept gamma rays. A plate of polycarbonate of 8 mm in thickness is attached to the bottom from the outside to protect the thin window. Figure 2 shows a cage to accommodate the PTV. It is rectangular in shape, 400×400×300 mm^3, and assembled with steel angles. Inside the cage is a vinyl chloride hollow-cylindrical guide of 210 mm in diameter and 200 mm in height for the PTV.

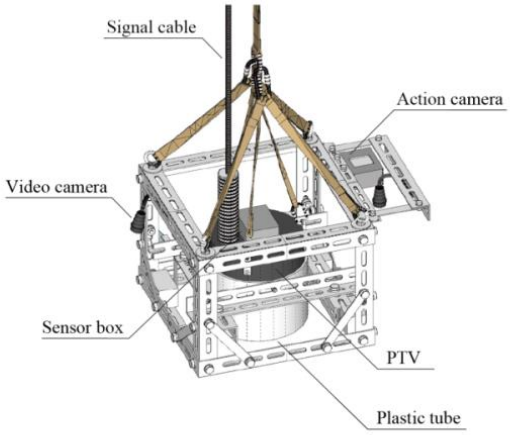

Fig. 2 Pressure-Tight Vessel (PTV) in the Cage
Although not shown fully in this illustration, two LED lights, two video cameras, and two action cameras are attached to the cage.

The steel angles provide footholds for two video cameras, two action cameras, and two underwater LED lights. A water-tight sensor box which accommodates a 3-dimensional digital linear acceleration sensor and a 3-dimensional digital magnetic sensor (LSM303D, STMicroelectronics, Genève, Switzerland) is attached on the top of the PTV to detect the tilt of the PTV. To know an approximate depth, an underwater pressure sensor (MS5837-30BA, Measurement Specialties, CA, USA) is also attached. Computer programs are written for a live display of information sent from these sensors. The information from the sensors is indispensable to know how stable the cage stays on the surface of rock against currents. The signals from the C12137-10 module are forwarded to a laptop computer on a fishing boat through a LAN cable of 100 m in length with a USB2.0 extender, IC280A-R2 (Black Box Network Services, Lawrence, PA, USA). A gamma-ray energy spectrum in the energy range between 50 keV and 2 MeV can be readily seen on the laptop computer's screen with our programs which command acquisition, storage, and processing of the data.

The cage is tied by four belts to the lower end of the main rope; the other end is tied to a motor-drivable winch on board a fishing boat. Independently the PTV is tied to the same end of the rope by three belts. To protect the thin window, we adjust the length of the belts so that the bottom of the PTV is 50 mm above that of the cage. In this way, the PTV never lands on the rock's surface before the cage lands.

The total mass of the present PTV and the cage is about 40 kg. The cage is four times larger in the basal plane than our previous one. It stands more firmly against turbulent currents at the bottom of the sea during the measurement. Even when the cage tilts in a certain direction, as long as it keeps that tilt and the count rate of the photopeak of ^{40}K is above a preset value, we know that the contact between the PTV and the flat part of the surface of rock is stable on an inclined surface.

4. Property of the detector in the PTV

The residual ^{137}Cs on a flat surface of rock acts as a two-dimensionally extended source with practically no thickness, and our measurement is made in the contact configuration between the detector in the PTV and the flat surface of rock. Therefore, in carrying out measurements of the property of the detector in the PTV, we simulate the situation as close as possible to the actual situation at the bottom of the sea. As a test source of ^{137}Cs, we use a rectangular plate source of ^{137}Cs, the active area is 100×100 mm^2. The source was manufactured and calibrated by Japan Radioisotope Association, the source code was CS221, and the activity was 1.0×10^4 Bq on December 1st, 2011.

4. 1 Energy resolution

The detector in the PTV is placed directly on the plate source to secure the contact configuration. In this way, we take into account the effects of the aluminum case in which the CsI(Tl) scintillator crystal is sealed, the aluminum window, and the polycarbonate protection plate of the PTV. Figure 3 shows a raw gamma-ray spectrum of the plate source of ^{137}Cs measured for 600 s in our laboratory.

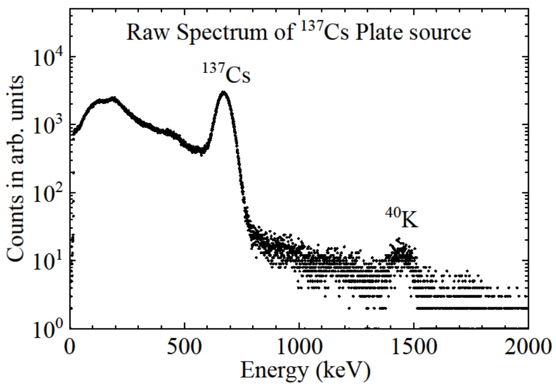

Fig. 3 Raw spectrum of the plate source of ^{137}Cs

To extract information about the photopeak from the raw spectrum, we use Origin Pro 2018 (Origin Lab Co., Northampton, MA, USA). After subtracting a B-spline curve fitted to a baseline from the raw spectrum, we fit a single Gaussian function to the photopeak of ^{137}Cs, as shown in Fig. 4 (a). The observed full width at half maximum (FWHM) of the photopeak gives an energy resolution of 10 % at 662 keV. The energy resolutions at other energies are measured using standard test sources of ^{241}Am and ^{60}Co, and ^{40}K in the KCl test source. The results are listed in Table 1. The photopeak of 59.5 keV of ^{241}Am is well represented by a single Gaussian function as shown in Fig. 4 (b). This fact and a fact that the background spectrum shown in Fig. 2 in Part 1 measured at 5 m above the bottom of the sea, the least noisy environment for the detector, has a sharp drop around 50 keV may indicate that the lowest end of the effective energy range is around 50 keV.

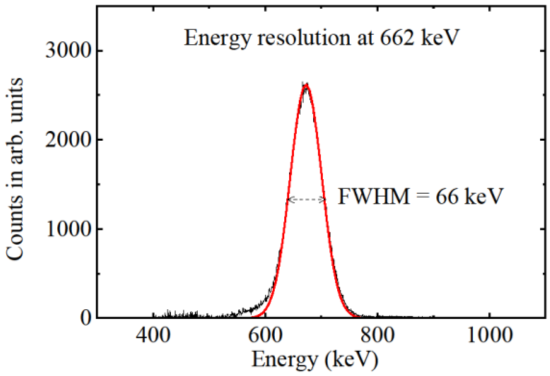

Fig. 4(a) Energy resolution at 662 keV

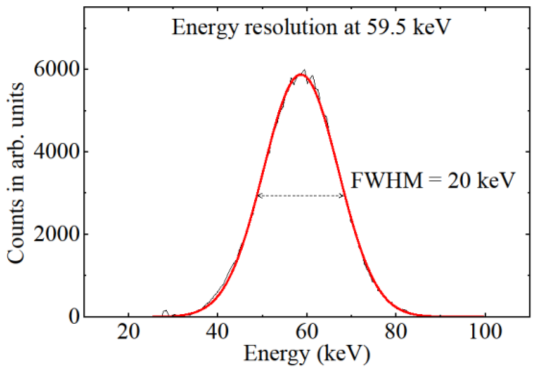

Fig. 4(b) Energy resolution at 59.5 keV

Table 1 Energy resolution

The energy resolution at 662 keV is measured using the plate source of ^{137}Cs mentioned in the text. At other energies standard test sources of ^{241}Am, ^{60}Co, and ^{40}K in KCl test source are used.

Energy in keV	Source	FWHM in keV (%)
59.5	^{241}Am	20 (34)
662	^{137}Cs	66 (10)
1173	^{60}Co	98 (8.4)
1332	^{60}Co	100 (7.5)
1461	^{40}K	109 (7.5)

4. 2 Conversion factor

In the present case of the contact configuration between the detector and source, a factor for conversion of the count rate of the ^{137}Cs photopeak in units of counts per second (cps) to the source strength is obtained from the number of counts under the photopeak of ^{137}Cs shown in Fig. 4 (a), the measurement time, and the amount of ^{137}Cs on the plate source under the detector. We have found that one count of the 662 keV gamma ray per second equals 3.29×10^3 Bq/m^2. Regarding overall detection efficiency, the present value is about 19 times higher than that reported in our previous study. The employment of the C12137-10 module, which has a wider area of 110 mm in diameter to accept gamma rays, has helped increase the overall detecting efficiency for a case of a two-dimensionally extended source.

4. 3 Spectrum of Compton continuum

As mentioned in Part 1, we are interested in an energy spectrum of gamma rays in an energy region below the photopeak of ^{137}Cs. Therefore, knowing the Compton continuum (a continuous spectrum caused by the Compton scattering of incoming gamma rays in the scintillator [12]) is essential.

Our process to empirically determine the spectrum of the Compton continuum in the present detector is as follows. A test source is made of dead leaves collected at a site near the FD1NPP to record the radioactivity of ^{134}Cs and ^{137}Cs at the time of the accident and to follow the subsequent change in the ratio of ^{134}Cs/^{137}Cs. It is widely accepted that the ratio of ^{134}Cs/^{137}Cs was very close to unity when spewed. At present, it is calculated to be 0.054. The activity of ^{137}Cs in the test source was found to be 8.2×10^5 Bq/m^2 as of December 2017 by a cross-calibration with the plate source mentioned above. We use the dead leaves as our test source for an important reason. As shown in Part 1, in all the spectra measured at the bottom of the reefs, the photopeak of 796 keV (^{134}Cs) is still distinguishable and the photopeak of ^{134}Cs at 605 keV overlapped with that of ^{214}Bi at 609 keV was also observable as a little hump on the tail of the 662 keV peak of ^{137}Cs. Therefore, the spectrum of the Compton continuum created by the coexisting ^{134}Cs with the current ratio of ^{134}Cs/^{137}Cs should be taken into account. The measurements are made in the low-background underground facility of the Institute of Cosmic Ray Research (ICRR), the University of Tokyo. First, the background spectrum is measured with the window of the PTV facing down on the concrete floor. An energy spectrum of the test source of ^{137}Cs with ^{134}Cs is then measured in a way that the test source is inserted between the bottom of the PTV and the concrete floor. Thus, gamma rays scattered back from the concrete floor are recorded on the spectrum. The raw spectra of the test source and the background are shown in Fig. 5. Subtraction of the spectrum of the backgrounds from that of the test source gives the spectrum of the photopeaks of ^{134}Cs and ^{137}Cs, and the Compton continuum in the detector due to the test source, as shown in Fig. 6. At the same time, the subtraction nulls the effects of noises in the electronic circuits of the detector and contributions from gamma rays from ^{40}K in the surrounding walls and floor of the laboratory.

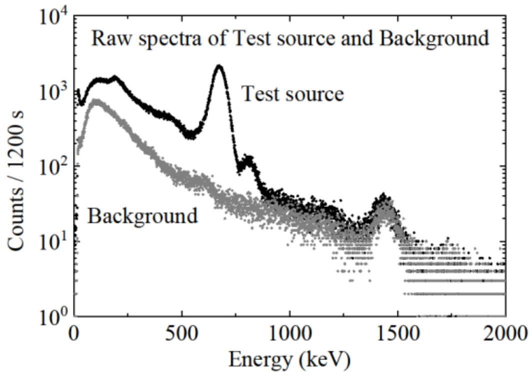

Fig. 5 Raw spectrum of the test source

The spectrum of the test source made of the dead leaves is shown by the black dots and that of the background is show by the gray dots. Both are measured in the low-background underground facility of the ICRR. Subtraction of the gray spectrum from the black one gives the Compton continuum and the photopeaks of ^{134}Cs and ^{137}Cs with the current ratio.

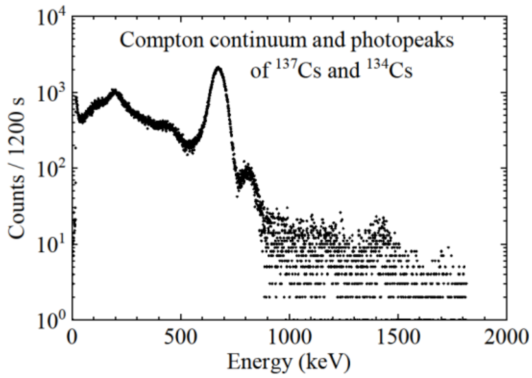

Fig. 6 Compton continuum and photopeaks of ^{137}Cs and ^{134}Cs

The empirically determined spectrum of the Compton continuum and the photopeaks of coexisting ^{134}Cs and ^{137}Cs, the ratio $^{134}Cs/^{137}Cs$ is 0.0543, with those backscattered from the concrete floor are shown.

As described in Part 1, the spectrum of the Compton continuum thus obtained makes it possible to obtain a true energy spectrum of the scalar flux of gamma rays near the interface between the surface of rock and seawater.

4. 4 Gain stability

A long-term (from May 2018 to November 2019, 16 months) drift of the gain of the C12137-10 module was 0.7 percent at 662 keV in our laboratory environment. Regarding a short-term drift, the temperature on the deck of our fishing boat was around 28 ºC and that at the bottom of the sea was around 20 ºC in summer, while in winter the temperature on the deck was around 5 ºC and at the bottom was around 15 ºC. We did not observe any shift of the photopeak of ^{137}Cs in the repetition of a cycle of lowering to the bottom of the sea, keeping at the bottom for 600 s for measurement of the gamma-ray spectrum, and lifting up on the deck every half an hour. The short-term gain drift due to a temperature change of about 10 ºC was negligible. The built-in temperature-compensation mechanism seemed to be working well.

5. Operation

With the aid of the live views sent from the video cameras, the cage is manually lowered from the fishing boat toward a place that looks like a part of a flat surface of a rock. After placing the cage there, we monitor the count rate of the photopeak of ^{40}K for the first few minutes. When the count rate is less than 2.4 counts/s, we judge that the contact is not properly established and abandon the measurement. Then, we replace the cage on a nearby rock. During the measurement, the tilt and movement of the cage are monitored with the linear acceleration sensor. The video cameras also send a live view of the cage and its surroundings. Any movement of the cage, if any, due to turbulent currents can be detected. The depth of reefs varies from 7 m to 15 m. We use sonar installed on the fishing boat and the pressure sensor attached to the cage for depth measurements. However, the depth may not be accurate because no correction of the height of waves and the tidal effect is made. Therefore, the value of depth is not listed in Table 1S of Part 1. The fishing boat is not anchored during the measurements, but the captain continuously corrects the drift due to surface winds and currents. It has been found in surveying the levels of residual ^{137}Cs on rugged reefs that the spectrometer is very useful in a situation where the depth is within 30 m, a level of ^{137}Cs contamination is above 5×10^3 Bq/m^2, and an allocated time of measurement at a point is about ten minutes.

The effects of unidentifiable noises in the sea should be removed from the observed spectrum. For this purpose, the detector is kept about 5 m above the bottom, and a spectrum is measured. This position is so chosen that the gamma rays emitted from ^{134}Cs and ^{137}Cs on the bottom would not reach the detector; note that the mean free path of the 662 keV gamma rays is only about 0.08 m in water. According to JAEA Seawater Registered Data Catalogue (2017) [13], the amount of ^{137}Cs in seawater is less than 10^{-3} Bq/L, which is too low to be detected by our detector for a measuring time of 600 s. Therefore, the spectrum thus obtained is that of background consisting of electronic and unidentifiable noises. The measured background spectrum is shown in Fig. 2 in Part 1.

Conclusions

A handy underwater CsI(Tl) gamma-ray spectrometer has been successfully improved for the in situ and stationary measurements of gamma-ray spectra on shallow rugged reefs. The justification for the use of ^{40}K to confirm the contact configuration between the detector and a flat part of the surface of rock at the bottom of the sea has been examined in greater detail. The overall uncertainty in the measured levels of ^{137}Cs is ± 10 % at most. The spectrum of the Compton continuum in the detector which is in the contact configuration with a given amount of coexisting ^{134}Cs and ^{137}Cs with the current ratio of ^{134}Cs/^{137}Cs has been empirically determined. Thus, the spectrum obtained makes it possible to obtain an energy spectrum of the scalar flux on and near the interface between the surface of rock and seawater.

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Supplementary information

Justification of our method to confirm contact configuration between the detector and source--- examination with the use of ^{40}K in a KCl test source and in slabs of granite---

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