Article

Rapid and Simple Measurement Method of ^{90}Sr Concentration in Water by Measuring β-rays from ^{90}Y

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A rapid and simple method to measure the concentration of ^{90}Sr in water by measuring β-rays from ^{90}Y was presented. Under the situation that ^{90}Sr/^{90}Y, ^{134}Cs and ^{137}Cs are the main radionuclides included in the water sample, only β-rays from ^{90}Y can transmit through 1.5-mm-thick polyethylene. From this fact, it is possible to measure β-rays from ^{90}Y using a β-ray detector, such as the GM-counter, set beneath the 1.5-mm-thick bottom of the water bottle containing the sample with ^{90}Sr/^{90}Y. The acrylic resin collimator having 0 cm, 1.00 cm, 1.50 cm or 3.00 cm diameter was made to detect β-rays at the fixed region of the GM-counter used. Contributions from bremsstrahlung produced by β-rays and γ-rays from radionuclides such as ^{134}Cs and ^{137}Cs/^{137m}Ba are removed by subtracting the count rate measured with a 1.00 cm acrylic resin collimator without a hole as the background count rate. The developed method was studied using the bottle routinely used at the Fukushima Daiichi Nuclear Power Plant. It was confirmed that the developed method can be applied to measure the ^{90}Sr concentration in water to the order of several Bq/cm^3 if ^{134}Cs and ^{137}Cs concentrations are less than or equal to the ^{90}Sr/^{90}Y concentration.

KEYWORDS: Sr-90, Y-90, concentration in water, β-ray, GM-detector, range of β-ray, acrylic resin collimator

I. Introduction

At Fukushima Daiichi Nuclear Power Plant (hereinafter referred to as “1F”), measurement of the radioactive concentration of ^{90}Sr in numerous water samples containing ^{90}Sr in a wide range of radioactive concentrations is required. The radioactive concentrations of ^{90}Sr to be measured range widely from the effluent standard level (3×10^{-2} Bq/cm^3) to several tens of MBq/cm^3. The types of water to be measured vary, including water retained in the nuclear reactor building, RO concentrated water from which Cs is eliminated using a cesium removing system (SARRY) (hereinafter referred to as “RO concentrated water”), water treated with various

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nuclide removal equipment (ALPS), and stagnant water in the weir of the RO concentrated water tank. These water samples contain not only ^{90}Sr and ^{90}Y but also ^{134}Cs and ^{137}Cs at various radioactive concentrations. In addition, it is conceivable that the achievement of radioactive equilibrium between ^{90}Sr and ^{90}Y and the chemical dissolved state varies significantly among water samples. For water samples that are to be discharged, it is necessary to measure the radioactivity concentration of ^{90}Sr even for water samples containing ^{90}Sr at high radioactive concentrations that cannot be discharged. Tokyo Electric Power Co. measures ^{90}Sr in these water samples using a radioactive strontium analysis method requiring multistep chemical separation 1), a method of separating and concentrating ^{90}Sr through a chemical operation that allows ^{90}Sr to be measured using a β nuclide analysis device 2), a simplified measurement method for separating and measuring ^{90}Sr with cation paper 3), and a total beta method 4).

The former two methods not only require chemical separation and advanced measuring technologies but also take a substantial amount of time to obtain measurement results, thus they are not suitable for emergency response. The cation paper method has a drawback in that it is largely dependent on the amount of dissolved ions and the dissolved state. Given this background, the total beta measurement method has been used thus far to estimate the radioactive ^{90}Sr concentration in numerous water samples generated at 1F.

However, as described in the introduction of "Gross beta measurement method" 4), the total beta method is intended for the measurement of environmental samples and is used to check if the radioactive concentration is below the effluent standard. Its primary purpose is to determine whether another measurement is necessary or not. From the viewpoint of measuring the concentration of Sr-90/Y-90, which has a high radioactive concentration that cannot be drained, this method is not an appropriate one. Under such circumstances, a method that enables a wide range of radioactive ^{90}Sr concentrations to be measured rapidly and easily needs to be developed.

The method proposed in this study measures water samples containing ^{90}Sr/^{90}Y, ^{134}Cs, and ^{137}Cs as major radioactive nuclides in predetermined containers using a GM counter tube without performing any pretreatment. This is a new measurement method enables ^{90}Sr to be measured rapidly and easily at a concentration of more than several Bq/cm^3 by conducting the measurement twice at an interval of approximately 5 h even if the secular equilibrium of ^{90}Sr and ^{90}Y in the solution has not yet been confirmed. In addition to the measurement methods adopted by Tokyo Electric Power Co., the rapid analysis method 5) for discriminating radionuclides utilizing the difference in energy of β-rays using a liquid scintillation counter is also available. However, this method is intended to measure radionuclides in effluent and requires three or four days, despite its name. Because this method requires no chemical operation and can measure water samples placed in containers, it can be used to establish a system for measuring many samples in combination with a robotic operation and it is also possible to significantly shorten the time required to determine the radioactive ^{90}Sr concentration in numerous water samples generated at 1F.

The following section provides an outline of this method and the results of studying radioactive concentrations using known water samples containing ^{90}Sr/^{90}Y, ^{134}Cs, or ^{137}Cs.

II. Determining the ^{90}Sr Concentration in Water by Measuring β-rays from ^{90}Y

1. Outline of the Method

The following is a method to determine the radioactive ^{90}Sr concentration from the radioactive concentration of ^{90}Y obtained by measuring the number of β-rays from ^{90}Y using water samples containing ^{90}Sr/^{90}Y, ^{134}Cs, and ^{137}Cs as the main radionuclides. It is based on the fact that β-rays from ^{90}Sr, ^{134}Cs, and ^{137}Cs can be screened out by a 1.5-mm-thick (or more) polyethylene plate and that the maximum range for β-rays from ^{90}Y is 1 g/cm^2.

Specifically, a water sample containing ^{90}Sr/^{90}Y is placed in a container with an approximately 1.5-mm-thick bottom, and the number of β-rays is measured in the range limited by a GM counter tube using a β-ray detector equipped with a 1-cm-thick acrylic collimator. In addition, the count rate of β-rays from ^{90}Y is determined by deducting the count rate based on the bremsstrahlung X-rays generated because the γ- and β-rays from ^{134}Cs, ^{137}Cs/^{137m}Ba, etc. are present in the water sample as background while blocking the β-rays from ^{90}Y with a 1-cm-thick acrylic plate with no opening. The count rates of the β-rays from ^{90}Y are converted into radioactive concentrations using the conversion coefficient obtained from water samples with known radioactive concentrations. If it has not yet been confirmed that ^{90}Sr and ^{90}Y have achieved secular equilibrium, the measurement is conducted twice under the same conditions at an interval of approximately 5 h to determine the ^{90}Sr concentration from the ^{90}Y concentration obtained using the parent nuclide and daughter nuclide relation between ^{90}Sr and ^{90}Y. In this case, the information on the achievement of radioactive equilibrium between ^{90}Sr and ^{90}Y during the first measurement can be obtained together from the ^{90}Sr and ^{90}Y concentrations acquired during the first measurement.

2. β-Emitting Radionuclides to be Measured

Water containing ^{90}Sr/^{90}Y that cannot be discharged because of high radioactive concentrations, generated at 1F, includes retained water such as RO concentrated water, revetment groundwater, and water related to leakage of reserved water, etc. The radionuclide in RO concentrated water obtained by removing cesium from stagnant water at turbine building consist mainly ^{90}Sr/^{90}Y, except tritium, and ^{134}Cs, ^{137}Cs, ^{54}Mn, ^{60}Co, ^{106}Ru/^{106}Rh and ^{125}Sb at a concentration of one-thousandth or less of the ^{90}Sr/^{90}Y concentration[6)]. In the case of revetment groundwater, ^{90}Sr/^{90}Y occur at the highest radioactive concentrations except for tritium, and there are cases in which ^{134}Cs and ^{137}Cs also occur at the same concentration in some areas. In addition, there are cases in which ^{54}Mn, ^{60}Co, ^{106}Ru/^{106}Rh, and ^{125}Sb are also present at a concentration of one-thousandth of ^{90}Sr/^{90}Y[7)]. Immediately after the accident, ^{89}Sr emitting β-rays with a maximum energy of 1.495 MeV were at a level equal to ^{90}Sr. However, this nuclide can be ignored because its half-life period of 50.53 d and 28 half-life periods have already passed. Concerning these nuclides as detected, **Table 1** shows the maximum energy and emission rate of β-rays as well as the energy and emission rate of γ-rays[8)]. The nuclides that emit β-rays with a maximum energy of 1 MeV or higher in addition to ^{90}Y are ^{60}Co, ^{106}Rh as a progeny nuclide of ^{106}Ru, and ^{137}Cs. The radioactive concentrations of ^{60}Co and ^{106}Ru/^{106}Rh occurring in water samples to be measured are not greater than one-thousandth of the radioactive concentrations of ^{90}Sr/^{90}Y, thus their contributions can be ignored in the measurement of β-rays. In addition, because water samples containing ^{90}Sr/^{90}Y that cannot be discharged because of high radioactive concentrations are also included in the measuring objects, the contribution of β-rays from ^{40}K,

Table 1 β-rays and γ-rays from main radionuclides identified in water samples at the Fukushima Daiichi Nuclear Power Station

Nuclide	Half-life	Decay mode	Maximum β-ray energy (MeV)	Emission rate	γ-ray energy (MeV)	Emission rate
^{54}Mn	312.03 d	EC		1.0	0.836	1.0
^{60}Co	5.2713 y	β^-	0.318	0.999	1.173	0.999
			1.491	0.0012	1.333	1.0
^{90}Sr	28.79 y	β^-	0.546	1.0		
^{90}Y	64.00 h	β^-	2.28	1.0		
^{106}Ru	373.59 d	β^-	0.0394	1.0		
^{106}Rh	29.80 s	β^-	2.407	0.100	0.512	0.204
			3.029	0.081	0.662	0.099
			3.541	0.786	1.05	0.016
			Others		Others	
^{125}Sb	2.75856 y	β^-	0.0953	0.134	0.176	0.069
			0.125	0.058	0.38	0.015
			0.131	0.179	0.428	0.298
			0.241	0.016	0.463	0.106
			0.303	0.404	0.601	0.178
			0.446	0.072	0.607	0.05
			0.622	0.136	0.636	0.113
			Others		0.671	0.018
					Others	
^{134}Cs	2.065 y	β^-	0.0888	0.273	0.563	0.084
			0.415	0.025	0.569	0.154
			0.658	0.702	0.605	0.976
			Others		0.796	0.866
					0.802	0.087
					1.365	0.03
					Others	
^{137}Cs	30.1671 y	β^-	0.514	0.944		
			1.176	0.056		
^{137m}Ba	2.532 m	IT			0.662	0.851

which is a form of natural radioactivity, can also be ignored.

Based on such circumstances, a method for measuring the β-rays in water samples containing ^{90}Sr/^{90}Y, ^{134}Cs, and ^{137}Cs as the main nuclides was studied.

3. How to Separate β-rays from ^{90}Sr, ^{134}Cs, and ^{137}Cs in the Water Samples as well as β-rays from ^{90}Y

Except for the β-rays with a maximum energy of 1.176 MeV from ^{137}Cs with an emission rate of 5.6%, the maximum energy of the β-rays from ^{90}Sr, ^{134}Cs, and ^{137}Cs is 0.6 MeV or lower. The maximum range of the β-rays of 0.6 MeV was calculated to be 0.2 g/cm^2 using the following equation 9):

$$R(g/cm^2) = 0.407\,E^{1.38} \tag{1}$$

where E is the maximum energy of the β-rays expressed in MeV. However, the maximum range of the β-rays from ^{90}Y with a maximum energy of 2.25 MeV is 1.05 g/cm^2 as determined using the following equation 9):

$$R(g/cm^2) = 0.542\,E - 0.133 \tag{2}$$

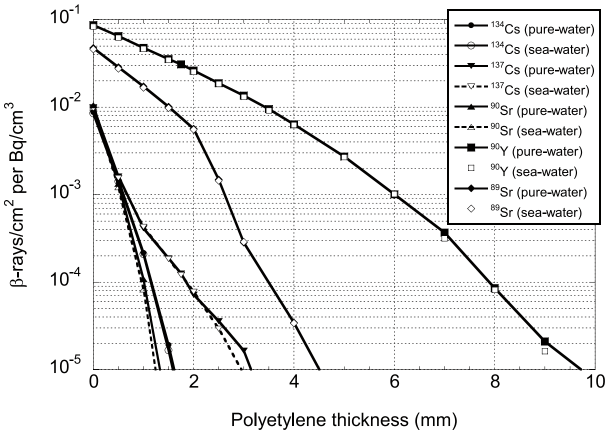

Figure 1 Calculated β-ray flux with egs5 from 1.5 cm pure-water or sea-water including ^{90}Sr, ^{90}Y, ^{89}Sr, ^{134}Cs, or ^{137}Cs with concentration of 1 Bq/cm^3 as a function of polyethylene thickness inserted

These figures are different from the effective ranges because they are just relative to the maximum energy of the β-rays and the energy decreases in water if these nuclides occur in water. To more closely examine the attenuation of the actual situation, we calculated the amount of β-rays originating from a water sample that can transmit a polyethylene plate 1.5 cm thick containing ^{90}Sr, ^{90}Y, ^{134}Cs, or ^{137}Cs at a concentration of 1 Bq/cm^3, using egs5 10) the electromagnetic cascade Monte Carlo code. For reference, we also calculated the β-rays from ^{89}Sr although they do not need to be considered in the case of water samples collected at 1F. In addition, we also performed a calculation for the case where these nuclides occur in seawater having a salt content of 3.4% and a density of 1.035 g/cm^3. Because the number of electrons that reached the outside of the polyethylene plate was determined in the calculation, the secondary electrons generated because of scattering with β-rays and those generated by bremsstrahlung X-rays were also included. **Figure 1** shows the results. As is clear from the figure, even when ^{90}Sr, ^{134}Cs, or ^{137}Cs with the same radioactive concentration as ^{90}Y is present, the number of β-rays from the respective nuclide after transmitting the 1.5-mm-thick (0.136 g/cm^2) polyethylene plate is less than one-hundredth of the number of β-rays from ^{90}Y. There was little difference found between fresh water and seawater. Therefore, assuming that the radioactive concentrations of ^{134}Cs and 137Cs measured are lower than the concentration of ^{90}Y, the number of β-rays from ^{90}Y can be obtained by measuring the β-rays after penetrating the polyethylene plate [a].

4. Elimination of the Influence of Bremsstrahlung X-Rays and γ-Rays

β-ray measuring instruments can also sense γ-rays and X-rays, although the sensitivity

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[a] Although different from the environment covered by this method, if ^{90}Sr and ^{90}Y are present at the same concentration, the result of the measurement conducted according to this method overestimates the concentration of ^{90}Y. According to Figure 1, the number of β-rays from ^{89}Sr after penetrating a 1.5-mm-thick polyethylene plate is approximately 30% of the number of the β-rays of ^{90}Y. Therefore, the concentration of ^{90}Y which is in secular equilibrium with ^{90}Sr is overestimated by approximately 30%. Even if the concentration of ^{89}Sr was the same as that of ^{90}Sr when the accident occurred, the concentration of ^{89}Sr decreases to less than 0.7% of that of ^{90}Sr over one year. Therefore, the overestimation of the concentration of ^{90}Y will be approximately 0.2% in the secular equilibrium state or approximately 2% even in an extreme case where only 10% equilibrium is achieved.

varies. Because bremsstrahlung X-rays are generated in water or a container by the β-rays from ^{90}Sr and ^{90}Y in water and γ-rays are emitted when various concentrations of γ-emitting nuclides such as ^{134}Cs and ^{137}Cs/^{137m}Ba occur in a water sample, it is necessary to deduct the count rates based on the γ-rays and bremsstrahlung X-rays (hereinafter referred to as "photons") as background to measure the number of β-rays.

The high-energy β-rays from ^{90}Y can also be screened out using a 1-cm-thick acrylic plate, which is thicker than the maximum range. Although photons are slightly attenuated by an acrylic plate 1 cm thick, the attenuation rate is much lower than that of β-rays. Thus, the results of the measurement after penetration through the acrylic plate can be used as the background count rates based on photons.

5. Relation between the Radioactive Concentration of ^{90}Sr and that of ^{90}Y in Water Samples

If it has been confirmed that ^{90}Sr and ^{90}Y in a water sample have achieved secular equilibrium, the radioactive ^{90}Sr concentration can be determined from the radioactive ^{90}Y concentration obtained during the first measurement. Even if they have not yet achieved secular equilibrium, conducting the second measurement after a certain period (approximately 5 h) enables the radioactive ^{90}Sr concentration to be obtained.

Because the ^{90}Sr and ^{90}Y half-life periods are 28.79 y and 64.00 h, respectively, their decay constants are 2.75×10^{-6} (h^{-1}) and 1.08×10^{-2} (h^{-1}), respectively.

Assuming the number of atoms of ^{90}Sr and ^{90}Y per cm^3 at time $t = 0$ is $N_{Sr\text{-}90}(0)$ and $N_{Y\text{-}90}(0)$, respectively, the number of atoms of ^{90}Y per cm^3 after t hours $N_{Y\text{-}90}(t)$ is calculated as follows:

$$N_{Y\text{-}90}(t) = \frac{\lambda_{Sr\text{-}90}}{\lambda_{Y\text{-}90} - \lambda_{Sr\text{-}90}} N_{Sr\text{-}90}(0)\,[\exp(-\lambda_{Sr\text{-}90}t) - \exp(-\lambda_{Y\text{-}90}t)] + N_{Y\text{-}90}(0)\exp(-\lambda_{Y\text{-}90}t) \tag{3}$$

Because $\lambda_{Y\text{-}90} \gg \lambda_{Sr\text{-}90}$, assuming $\lambda_{Y\text{-}90} - \lambda_{Sr\text{-}90} \approx \lambda_{Y\text{-}90}$, the equation becomes as follows:

$$\begin{aligned} A_{Y\text{-}90}(t) &= \lambda_{Y\text{-}90} N_{Y\text{-}90}(t) \\ &= \lambda_{Sr\text{-}90} N_{Sr\text{-}90}(0)\,[\exp(-\lambda_{Sr\text{-}90}t) - \exp(\lambda_{Y\text{-}90}t)] + \lambda_{Y\text{-}90} N_{Y\text{-}90}(0)\exp(-\lambda_{Y\text{-}90}t) \\ &= A_{Sr\text{-}90}(0)\,[\exp(-\lambda_{Sr\text{-}90}t) - \exp(-\lambda_{Y\text{-}90}t)] + A_{Y\text{-}90}(0)\exp(-\lambda_{Y\text{-}90}t) \end{aligned} \tag{4}$$

where, $A_{Y\text{-}90}(0)$ and $A_{Y\text{-}90}(t)$ are the radioactive concentrations of ^{90}Y at time $t = 0$ and t, respectively, and $A_{Sr\text{-}90}(0)$ is the radioactive ^{90}Sr concentration at time $t = 0$. Based on equation (4), the radioactive concentration is calculated as follows:

$$A_{Sr\text{-}90}(0) = \frac{A_{Y\text{-}90}(t) - A_{Y\text{-}90}(0)\exp(-\lambda_{Y\text{-}90}t)}{\exp(-\lambda_{Sr\text{-}90}t) - \exp(-\lambda_{Y\text{-}90}t)} \tag{5}$$

If the measurement is conducted at an interval of 5 h, it is calculated as follows:

$$A_{Sr\text{-}90}(0) = \frac{A_{Y\text{-}90}(t) - 0.054 \times A_{Y\text{-}90}(0)}{0.946} \tag{6}$$

Actually measuring $A_{Y\text{-}90}(0)$ and $A_{Y\text{-}90}(t)$ enables the radioactive ^{90}Sr concentration to be determined using equation (6).

6. Achievement of Radioactive Equilibrium between ^{90}Sr and ^{90}Y in a Solution

The achievement of radioactive equilibrium between ^{90}Sr and ^{90}Y in a sample can possibly be used as information for determining the history of the contaminated water. In the measurement conducted according to this method, not only can the ^{90}Sr concentration in a sample be measured but also the achievement of radioactive equilibrium between ^{90}Sr and ^{90}Y at the time of the first measurement as follows:

The radioactivity ratio of ^{90}Y to ^{90}Sr at $t = 0$ is determined by equation (5) as follows:

$$f=\frac{A_{Y\text{-}90}(0)}{A_{Sr\text{-}90}(0)}=\frac{A_{Y\text{-}90}(0)\,[\exp(-\lambda_{Sr\text{-}90}t)-\exp(-\lambda_{Y\text{-}90}t)]}{A_{Y\text{-}90}(t)-A_{Y\text{-}90}(0)\exp(-\lambda_{Y\text{-}90}t)} \tag{7}$$

Although f can be determined as a value using equation (7) regardless of the value of t, it is possible that it may not be because of the influence of the statistical error of the counted value, if the measurement interval is short, when these nuclides are near radioactive equilibrium. The ratio of $A_{Y\text{-}90}$ (t) to $A_{Y\text{-}90}$ (0) is determined using equations (4) and (5) with f as a parameter as follows:

$$\begin{aligned}\frac{A_{Y\text{-}90}(t)}{A_{Y\text{-}90}(0)}&=\frac{A_{Y\text{-}90}(0)/f\,[\exp(-\lambda_{Sr\text{-}90}t)-\exp(-\lambda_{Y\text{-}90}t)]}{A_{Y\text{-}90}(0)}+\frac{A_{Y\text{-}90}(0)\exp(-\lambda_{Y\text{-}90}t)}{A_{Y\text{-}90}(0)}\\&=\frac{\exp(-\lambda_{Sr\text{-}90}t)-\exp(-\lambda_{Y\text{-}90}t)}{f}+\exp(-\lambda_{Y\text{-}90}t)\end{aligned} \tag{8}$$

Figure 2 show the changes in $A_{Y\text{-}90}$ $(t)/A_{Y\text{-}90}$ (0) because of f when t is 5, 10, and 24 h. As shown in the figure, it is necessary to conduct the second measurement after at least 10 h and preferably 24 h to precisely determine the value of f when f is 0.5 or greater.

When ^{90}Sr and ^{90}Y are present in water, a bias in concentration may be caused by the formation of colloids and adsorption to suspended solids mainly because of a difference in chemical properties. In addition, immediately after the water is treated with a separation device utilizing the difference in chemical properties of both of the nuclides, there are cases in which the value of f becomes greater than 1 and there are also cases in which it is less than 1. Although this does not affect the result of the radioactive ^{90}Sr concentration, if the value of f is greater than 1, attention needs to be paid to the achievement of radioactive equilibrium between ^{90}Sr and ^{90}Y in the solution.

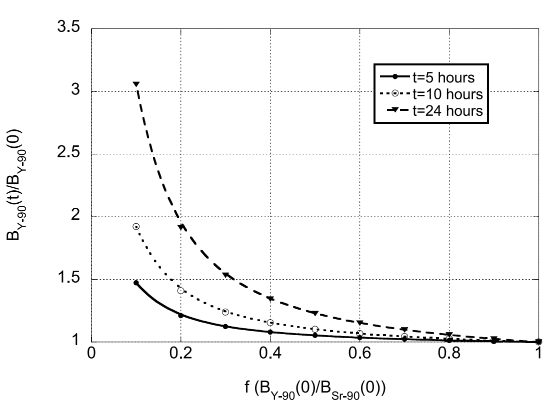


Figure 2 Increase in ^{90}Y concentration after t hours as a function of the equivalent ratio between ^{90}Y and ^{90}Sr

III. Measurement System

1. β-Ray Detector

Although there are various types of β-ray detectors, it is desirable if they can be handled easily because they are used for routine measurements. In the demonstration experiment described below, we used a TGS-146B [11)] (Hitachi-Aloka Medical) equipped with a large-area end-window organic GM tube utilized in contamination inspections as a β-ray detector, considering its sensitivity to β-rays. For precise measurement of the β-rays from ^{90}Y in a solution, the positional relation between the solution, which is a radiation source, and the detector is important. To detect the β-rays in the fixed range of the detector, we fabricated a 1.00-cm-thick acrylic collimator and used it during the experiment. We designed a jig to make the collimator center coincide with the centers of the GM detector as well as the water sample container at all times. We used four types of collimators; those with diameters of 1.00 cm, 1.50 cm, and 3.0 cm and one with no opening, for BG measurement. **Figure 3** shows the collimator and jig used. The aluminum holder shown in the figure is the jig used to place the container in a predetermined position.

2. Container Used

Because numerous samples were measured, we adopted the container that is routinely used at 1F for water samples as shown in **Figure 4** (white JK wide mouth bottle manufactured by Kokugo [12)], hereinafter referred to as a "T-container"). The T-container is made of polyethylene and has a bottom face the center of which is 1.5 mm thick and with a periphery that is 1.0 mm thick. The button face functions as a shield that eliminates the β-rays from the nuclides other than ^{90}Y. We conducted the measurement on a 25-cm^3 water sample with the aforementioned concentration in the T-container. The water surface height was approximately 1.6 cm. Because the maximum range of ^{90}Y, which has the widest range, is 1 cm in water, it is believed that the water surface height does not influence the measurement result as long as it is 1 cm or higher.

During the measurement conducted to examine the influence of the uneven bottom face, we also used a U8 container, which is commonly used in radioactive sample measurement, having a bottom surface of constant thickness,.

Figure 3 Picture of acrylic resin collimators and aluminum holder used for measurement

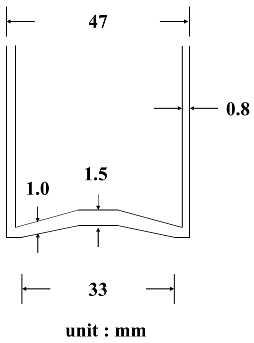


Figure 4 Schematic diagram of T-bottle used

3. Points to Note when Preparing Samples

The method is based on the premise that ^{90}Y generated by the decay of ^{90}Sr is uniformly distributed in a water sample. The radioactive isotopes occurring in a water sample undergoing measurement, such as ^{134}Cs, ^{137}Cs, ^{90}Sr, etc., are considered to be present in the carrier-free state and neutral in most cases, but there are also neutral or slightly alkaline solutions such as RO concentrated saltwater 13). It has been reported that carrier-free elements do not display their original physicochemical behavior and cause uneven solutions because of the formation of colloids and adsorption to suspended solids in some cases 14,15). In particular, it is well known that carrier-free ^{90}Y causes uneven solutions because of the formation of colloids and adsorption to suspended solids when the solutions are alkaline 14,16).

In RO concentrated saltwater, it has been shown that not only ^{90}Y but also other nuclides are present in the colloidal state or they are adsorbed to fine particles at a high rate 13).

To secure the uniformity of ^{90}Y, which is important for this measurement method, in a water sample for measurement, it is essential to:

(1) acidify the solution to inhibit the generation of colloids and

(2) add a small amount of carrier to prevent adsorption to fine particles.

In the demonstration experiment described in the following section, we acidified the water samples containing ^{90}Sr/^{90}Y, ^{134}Cs, or ^{137}Cs with hydrochloric acid (approximately pH = 1) and added the carriers of Cs, Sr, and Y.

IV. Demonstration Experiment and Results

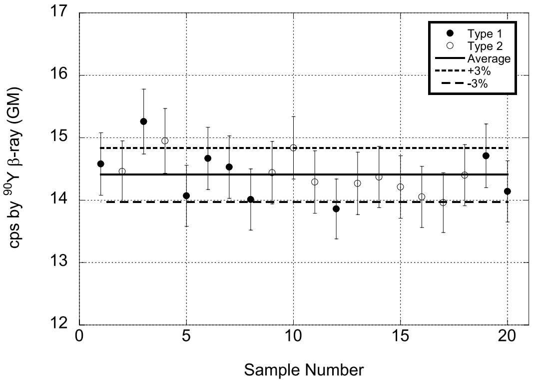
1. Radiation Source Samples and Measurement Method Used

To validate this method, we conducted an experiment using ^{90}Sr/^{90}Y solutions in the radioactive equilibrium state, which were adjusted to the radioactive concentrations of 138.0, 523.9, 5,244, and 11,660 Bq/cm^3 as well as ^{134}Cs and ^{137}Cs solutions adjusted to the radioactive concentrations of 7,405 and 10,800 Bq/cm^3, respectively. These were all hydrochloric acid solutions of 0.1 M, and the carrier concentrations of the strontium chloride and yttrium chloride were adjusted to be 0.05 mg/g in the ^{90}Sr/^{90}Y solutions and 0.1 mg/g in the ^{134} Cs and ^{137}Cs solutions. The radioactive ^{90}Sr/^{90}Y concentrations in the solutions were measured according to the efficiency tracer method using a liquid scintillation counter. However, the radioactive concentrations of ^{134}Cs and ^{137}Cs were measured using the γ-ray spectrometry method with a high-purity germanium semiconductor detector whose counting efficiency was calibrated in advance. The expanded uncertainties of the radioactive concentrations (coverage factor of 2) were 1.4% in the case of ^{90}Sr/^{90}Y and 11,660 Bq/cm^3 and 1.5% in the case of the other nuclides.

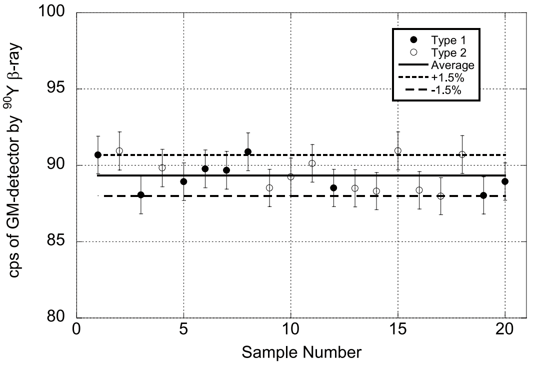
The measurement was conducted three times for each combination of container and collimator, and the dead times were corrected using the dead time (225 μs) measured in advance according to the two-source method.

2. Variation in Results Depending on Container Type

Because the measurement is based on the premise of the use of a T-container but not a special dedicated container as described in Section III-2, it was feared that the variation in the bottom thicknesses of T-containers used might have possibly caused a variation in measurement results. To examine the degree of variation, we conducted a measurement using 20 samples containing ^{90}Sr/^{90}Y at a concentration of 523.9 Bq/cm^3. The containers used were designated, "No. 1 Type" and "No. 2 Type", and there was a slight difference in weight between the two types. We also examined if a difference in type influenced the result. **Figure 5** shows the results obtained for the collimators with diameters of 1.50 cm and 3.00 cm. The lengths of the error bars in the figure represent statistical uncertainties of the count rates for 1 min. Of course, because the variation in measurement results is affected by statistical uncertainty, the variation increases when a collimator having a smaller diameter is used. This measurement showed that



(a) 1.50 cm diameter collimator



(b) 3.00 cm diameter collimator

Figure 5 Count rate unevenness of ^{90}Y β-rays from water including ^{90}Sr/^{90}Y with concentration of 523.9 Bq/cm^3 due to T-bottle used (a) 1.50 cm diameter collimator, (b) 3.00 cm diameter collimator the variations when a 3.00-cm-diameter collimator and 1.50-cm-diameter collimator were used were 1.5% and 3.0%, respectively, which proved to be the same as the statistical uncertainty. No systematic difference was observed between the No. 1 and No. 2 types. From the aforementioned result, it was concluded that the influence of the variation among T-containers could be ignored.

3. Influence of the Bottom Face Structure of the T-Container

As shown in Figure 4, the T-container used in the measurement has a bottom face that is not flat but raised and thicker in the center. To examine the influence of this structure on count rates, we compared the results of the measurement conducted using a T-container containing ^{90}Sr/^{90}Y to a concentration of 523.9 Bq/cm^3 and a U8 container with a flat bottom face with the number of β-rays at the collimator exit calculated using egs5. The calculation using egs5 was made to determine the number of β-rays per second at the collimator exit when a 1.00-cm-thick acrylic collimator is placed behind 1.5-mm-thick polyethylene plate, assuming the amounts of ^{90}Sr and ^{90}Y in the water sample are uniform. The T-container is composed of polyethylene and its bottom face is 1.5 mm thick at its center and 1.0 mm thick along its periphery.

The U8 container is composed of polystyrene and has a bottom surface with a uniform thickness of 1.5 mm. **Table 2** shows a comparison between the measured and calculated values for various collimator diameters. Although the difference from the calculation result was less than 10% for both types of containers, the measured values of the U8 container are generally in better agreement with the calculated values because the calculation was conducted assuming the bottom face of the U8 container. The count values of the U8 container were higher for the collimators with diameters of 1.00 cm and 1.50 cm, apparently because of the influence of the center part of the T-container, which was slightly raised. However, the count values of the T-container were higher for the collimator with a diameter of 3.00 cm, apparently because of the contribution of the thin polyethylene along the periphery of the bottom face of the T-container. From the aforementioned results, it is believed that the shape of the bottom face of the T-container, which is not flat, has a certain influence on the results but does not become a major factor of uncertainty as long as the conversion coefficient is determined for the container to be used.

4. Influence of Radioactive Cesium

To examine the influence of γ-rays when ^{134}Cs and ^{137}Cs/^{137m}Ba occur in water samples, we conducted a measurement using water samples containing 7,405 Bq/cm^3 of ^{134}Cs or 10,800 Bq/cm^3 of ^{137}Cs/^{137m}Ba. **Table 3** shows the comparison between the count rates based on the β-rays from ^{90}Y per 10 kBq/cm^3 and the count rates of ^{134}Cs or ^{137}Cs/^{137m}Ba. Regardless

Table 2 Comparison between measured count rate and number of β-rays calculated with egs5

Diameter of collimator (cm)	Type of bottole	Count rate (A) cps for 524.6 Bq/cm^3	egs5 calculation (B) β-rays for 524.6 Bq/cm^3	A/B
1	T-bottole	4.5 ± 0.3	5.0 ± 0.2	0.91
	U8	4.9 ± 0.2		0.97
1.5	T-bottole	14.4 ± 0.9	15.8 ± 0.3	0.91
	U8	15.4 ± 0.4		0.98
3	T-bottole	89 ± 2	90 ± 1	0.99
	U8	82.8 ± 0.6		0.92

Table 3 Comparison of count rate between ^{134}Cs, ^{137}Cs and ^{90}Sr/^{90}Y

Colimator diameter	cps for 10 kBq/cm^3				
	^{90}Y (A)	^{134}Cs (B)	^{137}Cs/^{137m}Ba (C)	(B)/(A)	(C)/(A)
1.00 cm	86.8 ± 1.1	−4.92 ± 2.29	−4.80 ± 0.57	—	—
1.50 cm	276 ± 0.3	7.22 ± 1.76	2.25 ± 1.20	2.6%	0.8%
3.00 cm	1,824 ± 13	61.6 ± 3.1	38.2 ± 2.6	3.4%	2.1%

of which collimator was used, the count rates of ^{134}Cs and ^{137}Cs/^{137m}Ba were less than 4% of the count rates of the β-rays from ^{90}Y [b]. For the case in which a collimator with a diameter of 3 cm was used, we calculated the attenuation of the γ-rays from ^{134}Cs and ^{137}Cs caused by a 1-cm-thick acrylic plate using egs5. The ratio of the energy of γ-rays with and without the 1-cm-thick acrylic plate behind the 1.5-mm-thick polyethylene plate was 0.96 for both ^{134}Cs and ^{137}Cs/^{137m}Ba. However, the ratio of the count rate when a collimator with a 3-cm diameter and 1-cm-thick acrylic plate with no opening were used was 0.85 for ^{134}Cs and 0.78 for ^{137}Cs/^{137m}Ba, larger than the attenuation of the penetrating energy calculated. As is clear from Figure 1, this was mainly due to the influence of β-rays originating from the difference in the sensitivity of the GM detector to β-rays and γ-rays because a small number of penetrating β-rays was present behind the 1.5-mm-thick polyethylene plate. This can also be confirmed as the attenuation of ^{137}Cs/^{137m}Ba is greater. Therefore, the attenuation of γ-rays caused by the 1-cm-thick acrylic plate is approximately 5% of the value calculated using egs5.

Although the influence of γ-rays increases when the concentration of ^{134}Cs or ^{137}Cs is higher than that of ^{90}Sr/^{90}Y, overestimation can be avoided using the following method in this case. However, in the case where the concentration is 100 times or higher, the correction using this method cannot be applied because the contribution of the β-rays from ^{134}Cs and ^{137}Cs cannot be ignored.

(1) Measure the ^{134}Cs and ^{137}Cs concentrations in an aqueous solution using a Ge detector. (This concentration measurement is performed in many cases.).

(2) Determine the count rate of the γ-rays from ^{134}Cs and ^{137}Cs/^{137m}Ba using the concentrations of respective nuclides and the coefficients for converting from concentration to count rate. (According to Table 3, ^{134}Cs: 0.0062, ^{137}Cs/^{137m}Ba: 0.0038 cps per Bq/cm^3, using a 3.00-cm-diameter collimator)

(3) Determine the count rate of the β-rays originating from ^{90}Y by deducting the count value previously determined from the count value obtained by deducting the measurement rate of the collimator with no opening from the count rate of the collimator.

5. Confirmation of Linearity with Respect to Count Rate and ^{90}Y Concentration as well as Determination of the Coefficient for Converting from the Count Rate of β-Rays from ^{90}Y to Concentration

To examine the relation between the radioactive concentration of ^{90}Y obtained according to this method and the count rate of the GM detector, we conducted a measurement using samples containing ^{90}Sr/^{90}Y at a concentration of 138.0, 523.9, 5,244, and 11,660 Bq/cm^3.

Figure 6 shows the results. Superior linearity was observed for all collimator diameters. The count rates per Bq/cm^3 for the collimators with a diameter of 1.0 cm, 1.50 cm, and 3.00 cm

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[b] The reason why the value is negative in the case of the collimator with a diameter of 1.00 cm is because the count value obtained when a 1.00-cm-thick acrylic plate with no opening was used was deducted from the count value obtained when the collimator was used, and it is considered that both are practically the same.

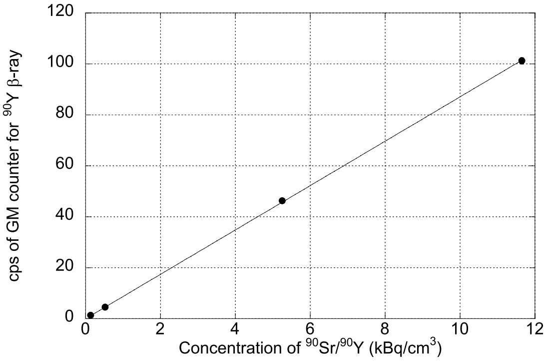
were 0.00871, 0.0277, and 0.181 cps, respectively.

From the measurement results, coefficients for converting from count rate to concentration using the least squares method were obtained as follows:

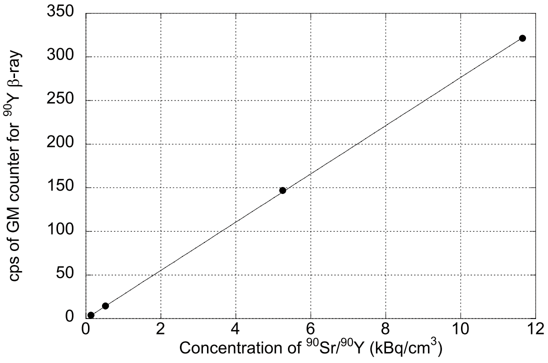
A collimator with a diameter of 1.00 cm: 115 ± 0.4 Bq/cm^3 per cps

A collimator with a diameter of 1.50 cm: 36.2 ± 0.1 Bq/cm^3 per cps

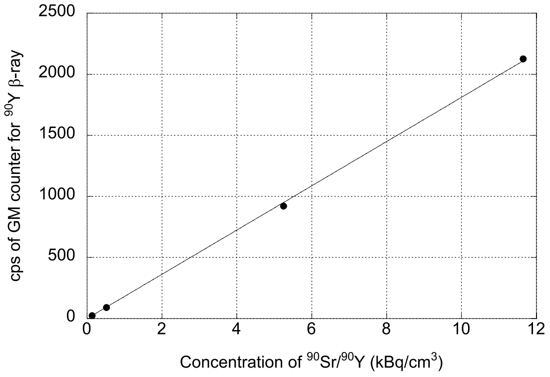
A collimator with a diameter of 3.00 cm: 5.52 ± 0.04 Bq/cm^3 per cps



(a) 1.00 cm diameter collimator



(b) 1.50 cm diameter collimator



(c) 3.00 cm diameter collimator

Figure 6 Relation between ^{90}Sr/^{90}Y concentration and count rate of ^{90}Y β-rays
(a) 1.00 cm diameter collimator, (b) 1.50 cm diameter collimator, (c) 3.00 cm diameter collimator

6. Detection Limit of This Method

The detection limit is expressed as follows:

$$n = (k/2)\left(k/t + \sqrt{(k/t)^2 + 4n_b(1/t + 1/t_b)}\right) \tag{9}$$

Where n is the count rate of the detection limit (cps), k is the confidence level, t is the sample measurement time, t_b is the background measurement time, and n_b is the background count rate (cps). The measuring environment background level of the location in which the demonstration experiment was conducted when no sample was placed was 1.3 cps. The background level of the measurement site at 1F may possibly be several times higher. In this method, the count rates of the photons through a collimator with no opening needs to be included in the background count rate. Because the background count rate of the bremsstrahlung X-rays based on the β-rays from ^{90}Sr/^{90}Y occurring in a water sample is approximately 0.0007 (cps per Bq/cm^3), the background of the measuring environment becomes dominant when the concentration of ^{90}Sr/^{90}Y is 1,800 Bq/cm^3 or less. In addition, because the background count rates based on γ-rays for ^{134}Cs and ^{137}Cs/^{137m}Ba are 0.036 and 0.014 (cps per Bq/cm^3), respectively, the background of the measuring environment dominates when the concentration of ^{134}Cs is 35 Bq/cm^3 or less and that of ^{137}Cs/^{137m}Ba is 91 Bq/cm^3 or less. Assuming that k is 3 and the water sample and background measurement time is 10 min when the background of the measuring environment is dominant, the detection limit count rate is 0.21 cps when the background level of the measuring environment is 1.3 cps and it is 0.31 cps when the background level is 3 cps. Based on the conversion coefficient of the 3-cm-diameter collimator with the highest sensitivity of 5.52 (Bq/cm^3 per cps), the respective detection limit radioactive concentrations correspond to 1.1 and 1.7 (Bq/cm^3). Of course, although the detection limit is dependent on the ^{134}Cs and ^{137}Cs concentrations in the water sample as well as the background level of the measuring environment, it can be stated that the detection limit based on this method is only several Bq/cm^3 if the contribution of the γ-rays from ^{134}Cs and ^{137}Cs is less than the background of the measuring environment.

V. Conclusions

Under the condition where the main nuclides occurring in a water sample are ^{90}Sr/^{90}Y, ^{134}Cs, and ^{137}Cs, we proposed a new method that allows a wide range of radioactive ^{90}Sr concentrations to measured quickly and easily without the need for any special operations by conducting a measurement using a water sample containing ^{90}Sr/^{90}Y placed in a container. Based on the results of a demonstration experiment conducted using water samples, the radioactive concentrations of which were precisely evaluated, and the T-containers used routinely at 1F, we drew the following conclusions:

(1) The variation among T-containers used at 1F is within the range of statistical uncertainty and does not affect the result.

(2) The bottom face structure of the T-container, which is not flat, caused a slight difference from the case in which the U8 container with a flat bottom face was used. However, because the influence is small, the uncertainty of the result is not affected as long as the "conversion coefficient" is determined in advance for the container to be used.

(3) The contribution of bremsstrahlung X-rays generated by β-rays in the water samples and containers can be eliminated by deducting the count rate based on the collimator with no

opening as the background.

(4) The influence of the γ-rays from ^{134}Cs or ^{137}Cs/^{137m}Ba occurring in the water samples is 4% or less of the count value based on the β-rays from ^{90}Y having the same radioactive concentration. If the radioactive concentration of ^{134}Cs or ^{137}Cs is 100 times or less than that of ^{90}Sr/^{90}Y, the overestimation of ^{90}Y can be avoided by deducting the count rate of γ-rays based on the radioactive concentration of ^{134}Cs or ^{137}Cs from the count value of the β-rays from ^{90}Y.

(5) The relation between the count rate of the GM detector and the radioactive ^{90}Y concentration achieves superior linearity in a wide range of radioactive ^{90}Y concentrations regardless of the diameter of the collimator.

(6) Even if it has not yet been confirmed that ^{90}Sr and ^{90}Y in a water sample have achieved secular equilibrium, conducting the measurement twice at an interval of approximately 5 h enables the radioactive ^{90}Sr concentration to be obtained.

(7) If the contribution of the γ-rays from ^{134}Cs and ^{137}Cs/^{137m}Ba is less than the background level of the measuring environment when the collimator with a diameter of 3.00 cm with the highest sensitivity is used, the minimum detectable radioactive concentration is only several Bq/cm^3, although it depends on the background level of the measuring environment.

From these results, it would appear that this method could be used to measure the radioactive ^{90}Sr concentration occurring in many water samples over a wide range of concentrations, which need to be measured at 1F, but the following needs to be noted when applying this method in the field.

(1) This method is based on the premise that ^{90}Y generated by the decay of ^{90}Sr is uniformly distributed in a water sample. Therefore, it is required to "acidify the solution to inhibit the generation of colloids and to add a small number of carriers to prevent adsorption to fine particles."

(2) It is necessary to determine the dead time of the GM detector to be used in advance according to the two-source method as well as to correct the dead time in the measurement results.

(3) Because the sensitivity may vary among GM detectors even if they are made by the same manufacturer, the conversion coefficient of the GM detector needs to be determined using a sample with a known radioactive concentration in the same manner as the demonstration experiment or determined in a similar manner with the TGS-146B used in this demonstration experiment.

(4) If the radioactive concentration of ^{90}Sr/^{90}Y in a solution is very high, it is not appropriate to use the GM detector for the measurement of the β-rays from ^{90}Y because the correction of dead time becomes significant. In such a case, a collimator with a smaller diameter[c] and a measuring instrument that can measure β-rays without being influenced by the dead time (such as an ionization chamber type survey meter that can be easily handled) needs to be used. It is necessary to determine the conversion coefficient using a sample with a known concentration in the same manner as in this study.

(5) To obtain precise information regarding the achievement of radioactive equilibrium between ^{90}Sr and ^{90}Y in a water sample, it is necessary to conduct measurements for at least 10 h, preferably at an interval of 1 day.

(6) This method can be used under the condition where ^{90}Sr/^{90}Y, ^{134}Cs and ^{137}Cs occur as the main nuclides, such as in the water samples currently obtained at 1F. If a water sample containing nuclear fuel materials is obtained in the course of future decommissioning work, it will be necessary to examine the relative ratio of the nuclides occurring to determine whether

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[c] In the case of a 1 cm collimator with the smallest diameter used in this study, the upper limit of measurement is 10^5 Bq/cm^3 if setting an upper limit of 1,000 cps. In the calculation of egs 5, it is possible to measure up to 2×10^6 Bq/cm^3 in the case of a collimator with a diameter of 0.4 cm.

this method can be applied or not.

For 1F, it is very important to control ^{90}Sr, and it is necessary to quickly and precisely measure the radioactive ^{90}Sr concentration in water samples. Although the water samples with a radioactive concentration of several Bq/cm^3 or higher can be the measuring objects, because no chemical operation is required and the measurement can be conducted with water samples placed in containers, it is possible to establish a system for many samples in combination with robotic operation. It is also expected that applying this method will significantly shorten the time needed to obtain a result and improve precision as well.

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