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RAPID COMMUNICATION

Rapid quantification of radiocesium dissolved in water by using nonwoven fabric cartridge filters impregnated with potassium zinc ferrocyanide

Tetsuo Yasutaka^{a*}, Hideki Tsuji^b, Yoshihiko Kondo^c, Yasukazu Suzuki^d, Akira Takahashi^a and Tohru Kawamoto^a

^aNational Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan; ^bNational Institute for Environmental Studies (NIES), Tsukuba, Japan; ^cJapan Vilene Company, Ltd., Moriyama, Japan; ^dFukushima Agricultural Technology Centre, Koriyama, Japan

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This paper presents a cartridge filter incorporating a nonwoven fabric impregnated with potassium zinc ferrocyanide (Zn-C) to effectively concentrate and quantify cesium dissolved in water. Experiments conducted with ¹³⁷Cs in conditioned water showed that at a flow rate of 2.5 L/min the filter could absorb 97.9% of dissolved ¹³⁷Cs from 20 L of water; high recovery efficiency was achieved over a pH range of 3–10. Test measurements of ¹³⁷Cs concentrations using Zn-C in river water agreed with the results derived by using an evaporative concentration method (within the counting error of the detector). Using this method, the pre-concentration time of radiocesium in 20 L of fresh water can be reduced to just 8 minutes.

Keywords: dissolved radiocesium; potassium zinc ferrocyanide; fresh water; cartridge filter

1. Introduction

Since the 2011 nuclear accident at Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Station, measurement of radiocesium in natural waters has become important for determining the leak of radiocesium from the watershed, the influence on crops of radiocesium in irrigation water, and the movement of radiocesium from riverine environments toward coastal areas. Cesium in natural water exists mainly in particulate and dissolved forms; the behaviors of these two forms in the natural environment might influence the fate of radiocesium in ecosystems. Accordingly, it is necessary to monitor the concentrations in water of particulate and dissolved radiocesium separately.

The dissolved radiocesium concentration in river water one year after the Nuclear Power Station accident was in the range 10^{-3} – 10^{-1} Bq/L, as reported by the Japanese Ministry of Education, Sports, Science and Technology (MEXT) [1] and by Yasutaka et al. [2]. For effective detection of the radioactivity of dissolved radiocesium at such low activity, the dissolved radiocesium must be concentrated after filtering out the suspended particulate matter.

There are several methods of concentrating low-level dissolved radiocesium in waters. These include evaporative concentration [2,3] and co-precipitation by sorbents (e.g., ammonium phosphomolybdate [4,5] or fer-

rocyanide compounds [6,7]), which selectively absorb dissolved cesium. Moreover, Prussian blue (PB), a ferrocyanide compound widely used as a pigment, has been used to remove dissolved cesium from liquids [8,9]. PB has also been incorporated into cesium-recovery devices (e.g., columns filled with PB-impregnated nonwoven fabric [10] or U-8 containers packed with PB [11]). Cartridge filters impregnated with PB ("PB-C" hereinafter) have been shown to rapidly absorb dissolved radiocesium. Two such devices connected in series have recovered 92% of dissolved radiocesium at a flow rate of 2.5 L/min [12]. However, the absorption efficiency of the PB-C decreases in the case of water samples that are either somewhat acidic or somewhat basic [13,14]. To allow their use with water at a wide pH range, the efficiency of absorption of dissolved radiocesium by such cartridge filters must be improved.

Among the ferrocyanide compounds, potassium zinc ferrocyanide is a sorbent that can absorb alkali-metal ions, including the cesium ion, by ion-exchange with K^+ within its crystal structure [15–18]; it is also known to maintain a high absorption ability of dissolved radiocesium over a wide range of pH values [19]. Here, the authors attempted to improve the efficiency of absorption of radiocesium dissolved in water by developing and testing a cartridge filter incorporating a nonwoven fabric impregnated with potassium zinc ferrocyanide.

*Corresponding author. Email: t.yasutaka@aist.go.jp

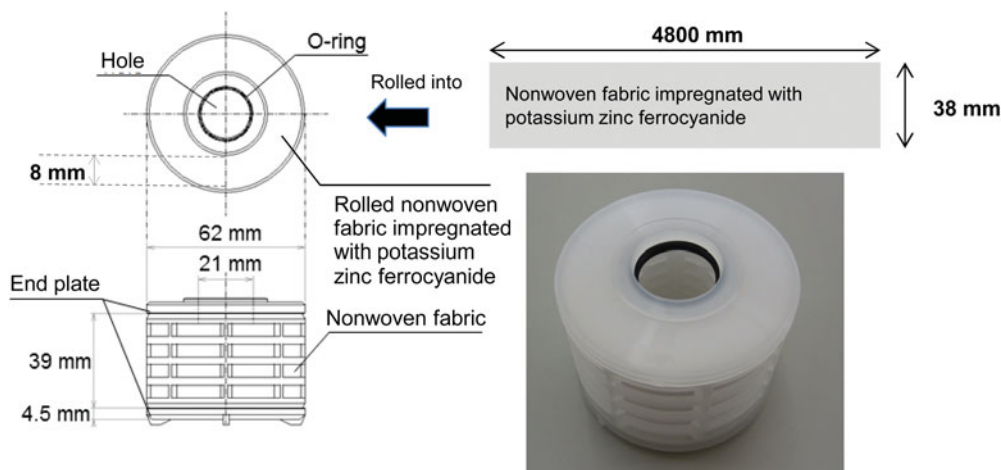


Figure 1. Schematic diagram of the cartridge filter (changing the nonwoven fabric type from Yasutaka et al. [12]).

2. Materials and methods

2.1. Experiment apparatus

Nonwoven fabric manufactured by using a wet papermaking process (Japan Vilene Co., Ltd., Japan) was impregnated with potassium zinc ferrocyanide hydrate ($K_2Zn_3[Fe(II)(CN)_6] \cdot xH_2O$, Kanto Chemical Co., Inc., Japan) by using the following process. First, the fabric (volumetric density 0.03 g cm^{-3} , thickness 0.01 cm) was infiltrated with potassium zinc ferrocyanide hydrate solution at a controlled concentration. The solution was prepared by mixing and stirring Zn solution and water to create a dispersion. The prepared dispersion was then placed in a tank and the untreated fabric dipped into it. In this operation, the areal density of impregnated potassium zinc ferrocyanide hydrate to nonwoven fabrics is 4 g/m^2 . The impregnated fabric was then dried at $130 \text{ }^\circ\text{C}$, cut into a rectangle measuring $3.8 \times 480 \text{ cm}$, and rolled up into a cartridge (Figure 1) to collect the dissolved radiocesium. This cartridge is hereafter called the nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide (“Zn-C”). A similar cartridge filter that employs nonwoven fabric impregnated with PB (i.e., a PB-C) has been developed; its performance in absorbing dissolved radiocesium in water has been reported by Yasutaka et al. [12] and Tsuji et al. [13].

The monitoring apparatus (Figure 2) comprised a feed-water tank, a peristaltic pump (Model-410, Solinst Canada, Ltd., Canada), a cartridge housing (polysulfonic housing PSF-500P, Advantec Co., Ltd. Japan), a Zn-C, a flow meter (DigiFlow 6710M, MRT Co., Ltd., or OF10ZZWN, Aichi Tokei Denki Co., Ltd. Japan), and a drainage tank. These components were connected in series by plastic tubes and the flow rate was controlled (or adjusted) by the pump. In most experiments only one cartridge housing was used. Before each experiment, residual sediment in the apparatus was washed out thoroughly with tap water. The pH value of the water was measured with a pH meter (D-50 series, Horiba Co., Ltd. Japan) and the electrical conductivity was mea-

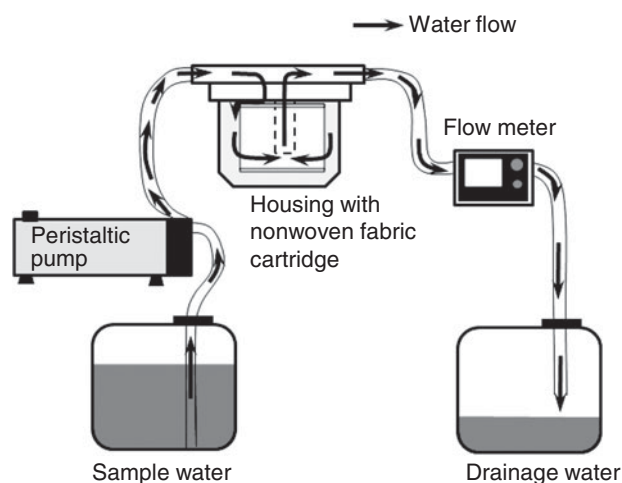


Figure 2. Conceptual drawing of the apparatus (modified from Yasutaka et al. [12]).

sured with a conductivity meter (ES-51, Horiba Co., Ltd.).

2.2. Preparation of conditioned water

For obtaining the recovery rate of dissolved ^{137}Cs by the Zn-C, sample water containing known dissolved ^{137}Cs concentrations (i.e., “conditioned” water) were prepared. The conditioned water was prepared by extracting ^{137}Cs from conifer leaves (picked in Fukushima Prefecture after this nuclear accident) by soaking the leaves in water and then filtering the resultant solution through a $0.45\text{-}\mu\text{m}$ -pore-size membrane filter (A045A047A Advantec Co., Ltd., Japan). The filtrate was then diluted with tap water (^{137}Cs concentration was 0.02 Bq/L) and the ^{137}Cs concentration measured with a Ge semiconductor detector in a 2-L Marinelli container to yield the conditioned water. The ^{137}Cs concentration in the water was conditioned to between 5 and 20 Bq/L (Table 1). The pH of the water was conditioned to

Table 1. Experimental conditions of water for applicability test of (a) potassium zinc ferrocyanide cartridges (Zn-Cs) and (b) Prussian blue cartridges (PB-Cs).

No.	Dissolved ^{137}Cs concentration (Bq/L)	Dissolved ^{133}Cs concentration ($\times 10^{-9}$ g/L)	Sample volume (L)	pH	Flow rate (L/min)	Number of repetition
(a) Zn-C						
Z-1	20	50	20	6–7	0.5	2
Z-2	5	50	20	6–7	2.5	6
Z-3	5	50	20	6–7	5	2
Z-4	5	50	20	6–7	10	2
Z-5	5	50	100	6–7	2.5	2
Z-6 ^{*c}	5	50	20	3	2.5	2
Z-7	20	50	20	5.8	2.5	2
Z-8	20	50	20	8.6	2.5	2
Z-9 ^c	5	50	20	10	2.5	2
Z-10	10	500	20	6–7	2.5	1
Z-11	10	5000	20	6–7	2.5	1
(b) PB-C						
P-1	20	50	20	6–7	0.5	2
P-2 ^{*d}	5	50	20	6–7	2.5	9
P-3	5	50	20	6–7	5	2
P-4	5	50	20	6–7	10	2
P-5 ^{*d}	5	50	95	6–7	2.5	2
P-6 ^{*d}	5	50	20	3	2.5	2
P-7 ^{*d}	5	50	20	10	2.5	2
P-8	10	500	20	6–7	0.5	1
P-9	10	5000	20	6–7	0.5	1

^{*c}The ZnCs used for the experiment no. Z-6 and no. Z-7 consist of nonwoven fabric impregnated with 5 g/m² of potassium zinc ferrocyanide.

^{*d}The result of the no. P-5 and the no. P-6 was described in Yasutaka et al. [12], and the result of the no. P-7 and the no. P-8 was described in Tsuji et al. [13].

between 6.0 and 7.0; the electrical conductivity ranged from 467 to 1482 $\mu\text{S}/\text{cm}$.

2.3. Gamma spectroscopy methodology

Activity of ^{137}Cs was measured by an SEG-EMS GEM 35-70a Ge semiconductor detector (detection efficiency 22.7%; resolution 1.76 keV; Seiko EG&G Co. Ltd., Japan) and an MCA 7600 multichannel analyzer (hereinafter “MCA”; Seiko EG&G Co., Ltd.). The MCA was setup with 4000 channels (at 0.5-keV intervals) from about 0 to 2000 keV. The spectra were analyzed with Gamma Studio software (Seiko EG&G Co., Ltd.). Detector efficiency was calibrated by using a source with nine nuclides in a 2-L Marinelli container (MX033MR) (Japan Radioisotope Association, Japan) and a source with nine nuclides in a U-8 container (MX033U8PP) (Japan Radioisotope Association, Japan) during installation of the semiconductor detector.

The accuracy of the analytical equipment was calibrated by taking weekly background measurements over 24 h; gamma ray energy was also calibrated weekly. The semiconductor detector was calibrated weekly by using standard volume sources, and decay compensation was applied. The minimum detection count for each sample was set to three times the standard deviation from the counting statistics, as calculated by using Cooper’s method [20].

2.4. Experimental conditions

One series of experiments with various pH values, flow rates, and ^{133}Cs (stable isotope) concentrations were conducted with the Zn-C and another with the PB-C (Table 1). The pH of the conditioned water was adjusted to between 3 and 10 for experiments Z-6 to Z-9 by adding HCl or NaOH.

To estimate the capacity of the Zn-C to absorb dissolved cesium, the ^{133}Cs concentration in the conditioned water was adjusted for experiments Z-10 and Z-11. A standard solution of $^{133}\text{CsCl}$ (Wako pure chemical industries, Ltd., Japan) was added in water with a known ^{137}Cs activity to adjust the ^{133}Cs concentration from its conditioned value from 5×10^{-8} (Z-2) to 5×10^{-7} g L⁻¹ for experiment Z-10 and to 5×10^{-6} g L⁻¹ for experiment Z-11. The concentration of ^{133}Cs was analyzed by inductively coupled plasma mass spectrometry (7500cx; Agilent Technologies, US). The Z-series experiments (Table 1(a)) used one Zn-C in each experiment. The number of repetitions was six for experiment Z-2, one for experiments Z-10 and Z-11, and two for all other Z-series experiments.

To compare the performance of the Zn-C with that of the PB-C, experiments with the PB-C (Table 1(b)) under the same conditions as those used for the Z-series experiments were performed.

First, conditioned water with a known dissolved ^{137}Cs concentration was passed through the Zn-C or



Photo 1. PBC and plastic case for direct measurement of ^{137}Cs concentrations by Ge semiconductor detector (left) and measurement situation of PBC with plastic case by Ge semiconductor detector (right).

PB-C to concentrate the dissolved ^{137}Cs . This “treated” water was collected in a 20-L bottle and shaken well; a 2-L water sample was then extracted and delivered to the Ge semiconductor detector in a 2-L Marinelli container. The rates of recovery of dissolved ^{137}Cs from the Zn-C and the PB-C were calculated as the difference between the amount of ^{137}Cs in the conditioned water and that in the treated water.

2.5. Field experiment

To evaluate the effectiveness of the Zn-C in measuring dissolved ^{137}Cs concentrations in river waters, the concentrations of ^{137}Cs measured by using the Zn-C method compared with those measured after conventional evaporative concentration [2,3]. River water samples were taken from two locations in the Abukuma upland area (about 50 km from the Fukushima Dai-ichi Nuclear Power Station): 17.3 L from location A and 20.7 L from location B.

With the Zn-C method, suspended particles in the sampled river water were immediately filtered on-site with the 1- μm pore size nonwoven fabric cartridge filters (1- μm nominal pore size [13]; capable of collecting more than 81% of particles of pore size 0.4–0.5 μm [21]). Two Zn-Cs were used in series along the direction of flow. The flow rate was set at 2.5 L/min. To calculate the actual ^{137}Cs radioactivity (Bq), the detected ^{137}Cs radioactivity was divided by a non-destructive detection efficiency (geometric efficiency) of 0.67 (see Section 3.4); the resulting ^{137}Cs radioactivity was divided by the flow volume for conversion to the dissolved ^{137}Cs concentration in the water (Bq/L).

With the evaporative concentration method, sampled water was passed through a 0.45- μm -pore-size membrane filter; the filtrate was concentrated to 2 L by evaporation and the ^{137}Cs concentration in this water

was detected with an SEG-EMS GEM 35-70a Ge semiconductor detector in a 2-L Marinelli container.

The pH and the electrical conductivity of the water were measured immediately after the sampling. The pH of the water from location A was 7.42 and that for location B was 7.10. The electrical conductivity of the water from location A was 83.3 $\mu\text{S}/\text{cm}$ and that for location B was 65.0 $\mu\text{S}/\text{cm}$.

2.6. Non-destructive detection of ^{137}Cs collected by the Zn-C, and distribution of Cs in the Zn-C

Yasutaka et al. [12] and Tsuji et al. [13] recommend that, for non-destructive measurement of ^{137}Cs concentrations, a Ge semiconductor detector covered in a plastic case be used to test samples collected by PB-Cs (Photo 1). Using non-destructive detection measurement, to calculate the actual ^{137}Cs radioactivity (Bq) or concentration (Bq L^{-1}), the detected ^{137}Cs radioactivity or concentration was divided by a non-destructive detection efficiency (geometric efficiency).

The authors previously found the non-destructive detection efficiency of the PB-C to be 0.77 by calculating the ratio of ^{137}Cs radioactivity in the cartridge as measured by non-destructive detection to the actual radioactivity of ^{137}Cs [12]. Here, the non-destructive detection efficiency of the Zn-C was calculated by using the same method, compared actual radioactivity of the ^{137}Cs in the Zn-C and radioactivity of the ^{137}Cs measured by non-destructive detection method. The actual radioactivity of the ^{137}Cs collected by the Zn-C was determined as the difference between the ^{137}Cs radioactivity of the conditioned water and that of the treated water.

For obtaining more information about non-destructive detection efficiency, the relationship between the non-destructive detection efficiency and the distribution of ^{137}Cs on the nonwoven fabric were

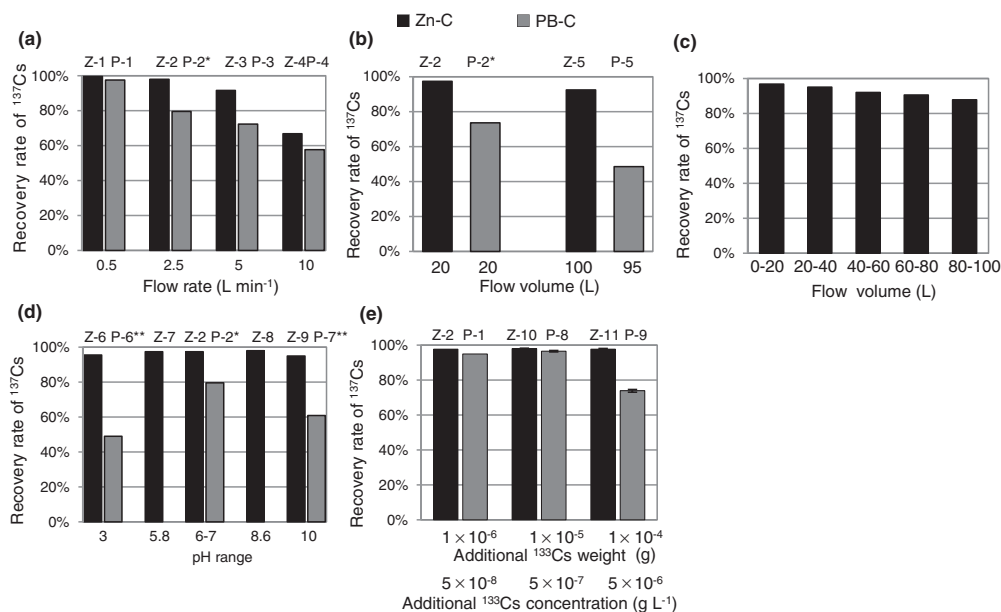


Figure 3. Relationships between rate of recovery of dissolved ^{137}Cs and (a) flow rate (0.5–10 L/min), (b) and (c) flow volume (20–100 L), (d) pH range (3–10), and (e) weight and concentration of ^{137}Cs (5×10^{-8} – 5×10^{-6} g L⁻¹). *Yasutaka et al. [12], **Tsuji et al. [13]. Zn-C, nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide; PB-C, cartridge filter using nonwoven fabric impregnated with Prussian blue

investigated. All of the Zn-Cs used in experiments from Z-1 to Z-5 were disassembled; the fabric was then unrolled and stretched. Each piece of fabric was cut into three pieces, each 160 cm long, representing the outer, middle, and inner sections of the rolled fabric. For experiments from Z-1 to Z-4, these three sections were then cut into smaller pieces (2×2 mm) and each section was packed into a 100-mL U-8 container for analysis by the Ge semiconductor detector. The three 160-cm pieces of nonwoven fabric from experiment Z-5 were each cut into only two pieces, representing the upper and lower parts of the filter material. Two Zn-Cs were disassembled for each of the experimental conditions (Z-1 to Z-5).

3. Results

3.1. Efficiencies of absorption of dissolved ^{137}Cs by the Zn-C and PB-C

Figure 3 indicates the effects of flow rate, flow volume, pH, and ^{137}Cs concentration in the water on the rates of recovery of dissolved ^{137}Cs by the Zn-C and the PB-C.

^{137}Cs recovery by the Zn-C was more than 99.5% at a flow rate of 0.5 L/min and 97.9% at 2.5 L/min (Figure 3(a)). Recovery decreased with increasing flow rate (91.5% at 5 L/min, 66.8% at 10 L/min). For all experimental flow rates, the rate of recovery of dissolved ^{137}Cs was higher with the Zn-C than with the PB-C.

With increasing flow volume, the recovery of dissolved ^{137}Cs by the Zn-C decreased from 97.9% (flow volume 20 L) to 92.4% (flow volume 100 L)

(Figure 3(b)). However, recovery by the Zn-C at flow volumes of both 20 and 100 L was higher than that by the PB-C at flow volumes of 20 and 95 L. Investigations of the Zn-C at other flow volumes (Figure 3(c)) showed that recovery decreased continuously with increasing flow volume (>96.7% for 20 L, 87.8% for 80–100 L).

Recovery rates with the Zn-C were consistently high (Figure 3(d)) for waters of pH 3 (>95.5%) to 10 (>94.8%); furthermore, for waters of pH 5.8 to 8.6, the dissolved ^{137}Cs recoveries were from 97.4% to 97.9%. In contrast, the rate of recovery of dissolved ^{137}Cs by the PB-C decreased when the pH of the water deviated from neutral (Figure 3(d) and Tsuji et al. [13]).

The concentration of ^{137}Cs in the water had no marked effect on the recovery of dissolved ^{137}Cs by the Zn-C (Figure 3(e)): the recovery rates stayed at 97.6% to 97.7% when the ^{137}Cs content of water increased from 1×10^{-6} g (concentration 5×10^{-8} g L⁻¹, sample volume 20 L) to 1×10^{-4} g (concentration 5×10^{-6} g L⁻¹, sample volume 20 L) (Figure 3(e)). In contrast, the recovery of dissolved ^{137}Cs by the PB-C decreased from 94.8% to 73.9% when the ^{137}Cs concentration of the conditioned water increased to 1×10^{-4} g.

3.2. Use of Zn-C to measure radiocesium in river waters

The measured ^{137}Cs concentration in the water from location A was 0.038 Bq/L as collected by the Zn-C and 0.042 Bq/L according to the evaporative concentration method (Figure 4). At location B the value was

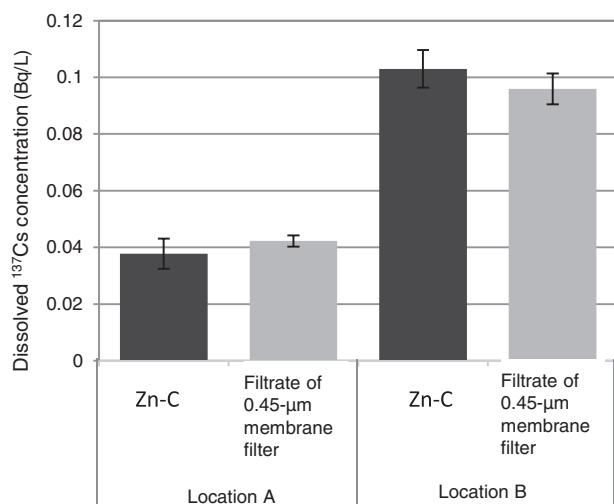


Figure 4. Comparison of concentration of ^{137}Cs determined by using the nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide (Zn-C) with that determined by using a conventional evaporative concentration method (filtrate from a 0.45- μm -pore-size membrane filter) in samples from two locations in the Abukuma upland. Error bars indicate counting error of the Ge semiconductor detector.

0.103 Bq/L with the Zn-C and 0.096 Bq/L with the evaporative concentration method. The dissolved ^{137}Cs concentrations determined by using the two methods at both locations agreed within the range of the counting error.

3.3. Distribution of ^{137}Cs in the rolled nonwoven fabric

During experiment Z-1 (flow rate 0.5 L/min), 93% of the available ^{137}Cs was collected on the outer section of the rolled nonwoven fabric, but the proportion of ^{137}Cs collected there decreased gradually with increasing flow rate (Figure 5(a)). In contrast, the proportion of ^{137}Cs collected on the inner section of fabric increased with increasing flow rate from 0.3% (0.5 L/min) to 18% (10 L/min). Thus dissolved ^{137}Cs was absorbed mainly by the outer section of the fabric and the amount absorbed decreased as the water progressed through the apparatus. Comparison of the amounts of dissolved ^{137}Cs absorbed by the upper and lower halves of the filter medium in Z-5 (Figure 5(b)) showed no notable difference.

3.4. Non-destructive detection efficiency

Non-destructive detection efficiency increased with increasing flow rate (Figure 6). Therefore, the actual ^{137}Cs radioactivity in the cartridge at a flow volume of 20 L and a flow rate of 0.5, 2.5, and 5.0 L min^{-1} could be determined without cartridge disassembly, by calculation: the detected ^{137}Cs radioactivity was divided by the non-destructive detection efficiency value of

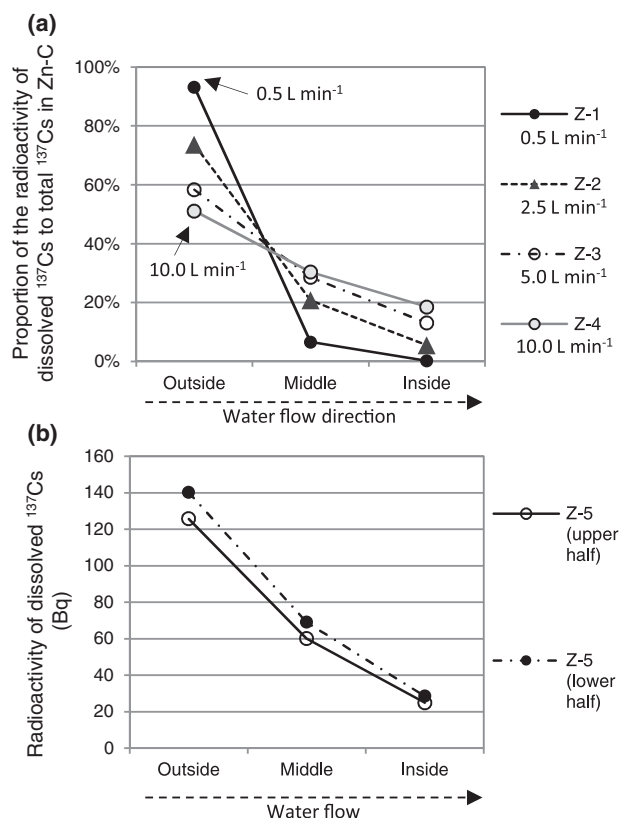


Figure 5. Distribution of recovered ^{137}Cs on nonwoven fabric after the experiments. (a) Proportion of dissolved ^{137}Cs recovered in the outer, middle, and inner sections of the nonwoven fabric in the nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide (Zn-C) relative to the total amount of ^{137}Cs in the water sample at different flow rates. (b) Radioactivity of dissolved ^{137}Cs on the upper and lower halves of the three sections of the nonwoven fabric.

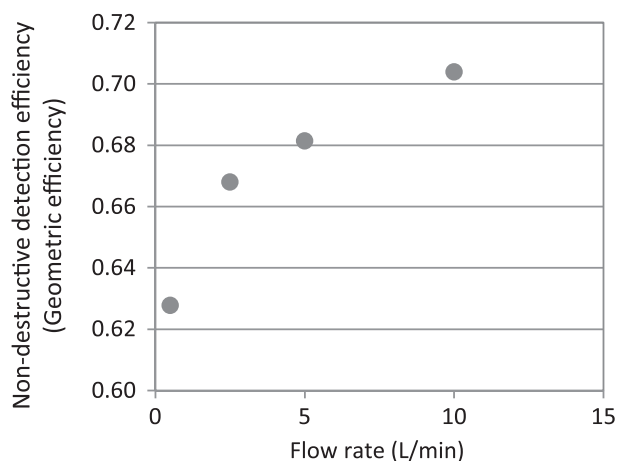


Figure 6. Non-destructive detection efficiency versus flow rate. Values are averages of two tests. Non-destructive detection efficiency was the ratio of ^{137}Cs radioactivity (Bq) in the cartridge, as measured by non-destructive detection, to the actual radioactivity of ^{137}Cs , as calculated from the difference in ^{137}Cs concentration between the conditioned water and the cartridge-treated water.

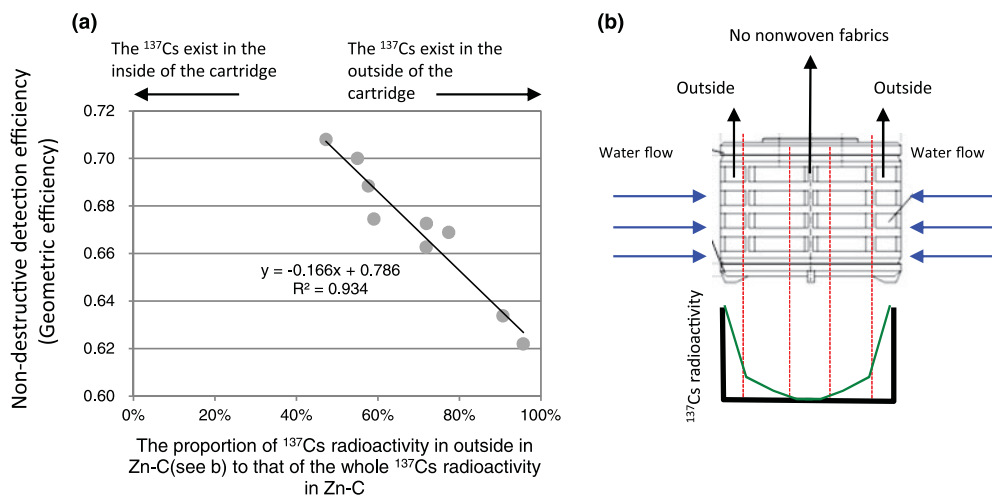


Figure 7. (a) Relationship between the proportion of ¹³⁷Cs in the outside (outer one-third see (b)) of the nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide (Zn-C) and non-destructive detection efficiency. As the proportion of ¹³⁷Cs in the outside one-third of the Zn-C increased, the non-destructive detection efficiency decreased because of the increasing distance of the ¹³⁷Cs from the Ge semiconductor detector. (b) Indicate the outside of the nonwoven fabric cartridge.

0.63 (0.5 L min⁻¹), 0.67 (2.5 L min⁻¹), 0.68 (5.0 L min⁻¹), and 0.70 (10.0 L min⁻¹).

4. Discussion

4.1. Efficiency of absorption of dissolved ¹³⁷Cs under different experimental conditions

The efficiency of absorption of dissolved ¹³⁷Cs by the Zn-C decreased with increasing flow rate (Figure 3(a)), because faster water flows shortened the amount of contact time between the dissolved ¹³⁷Cs and the nonwoven fabric.

The efficiency of the Zn-C also decreased with increasing volume of the water sample (Figure 3(b) and 3(c)). We assumed there were two possible reasons for this decrease, namely saturation of the Zn-C by stable Cs ions, or generation of water pathways within the Zn-C. Because the concentration of stable Cs ions in the water used in Z-5 experiments was 5×10^{-8} g L⁻¹, it is possible that the absorption sites of Cs ions in the outer section of the cartridge filter gradually became saturated with stable Cs from the water as the experiment progressed. However, the results of experiments Zn-9 to Zn-11 (Figure 3(e)) show that the Zn-C had a high capacity to absorb Cs ions (over 1×10^{-4} g) and still maintain high rates of recovery of ¹³⁷Cs with water ¹³³Cs concentrations of up to 5×10^{-6} g L⁻¹. Therefore, we concluded that the decrease in efficiency of the Zn-C with increasing sample volume was caused by the development of preferential water flow paths in the filter; these paths likely formed in response to increasing water pressure in the cartridge as the flow rate increased.

The Zn-C, unlike the PB-C, showed minimal changes in efficiency with changes in pH (Figure 3(d)). Under the same experimental conditions, the Zn-C absorbed dis-

solved ¹³⁷Cs more efficiently than did the PB-C, and with greater tolerance for acidic or basic water.

The high efficiency of the Zn-C was maintained at flow rates of up to 2.5 L/min for 20-L water samples. This combination of flow rate and sample volume provided a shorter processing time than has been achieved by conventional methods of ¹³⁷Cs extraction (about 8–40 h) [10]. However, the authors did not test the performance of the Zn-C for waters such as sea water, which contains high concentrations of other ions. Sea water contains several coexisting ions, such as K⁺ and NH₄⁺ that might influence the efficiency of the Zn-C. We intend to investigate the effects of coexisting ions in future experiments by using the Zn-C to filter sea water.

4.2. Relationship between non-destructive detection efficiency and ¹³⁷Cs distribution in the Zn-C

In this paper, we recommended the non-destructive detection efficiency value of the Zn-C at a flow rate of 2.5 L min⁻¹ and flow volume of 20 L was 0.67 with the Zn-C. The non-destructive detection efficiency of the Zn-C increased as the flow rate of water through it increased, whereas the proportion of ¹³⁷Cs absorbed onto the outer section of the fabric decreased (Figures 5 and 6). These observations suggest that faster flow rates led to shorter contact times between the Cs ions and the filter fabric and resulted in a more homogeneous distribution of radioactivity on the fabric. Moreover, the higher proportion of radiocesium absorbed onto the outer section of the filter medium at slower flow rates likely resulted in the detection of less radioactivity because more of the radiation source was then located farther from the sensing element of the Ge semiconductor detector. Accordingly, the distribution of radiocesium ions in the Zn-

C determined the non-destructive detection efficiency of the non-destructive detection method used, as indicated by the strong negative correlation between the proportion of ^{137}Cs in the outer section of the fabric and the non-destructive detection correction coefficient (**Figure 7(a)**). The distribution of radiocesium ions in the filter medium is determined by its absorption capability, which in turn depends on the experimental conditions, including the flow rate and the concentration of coexisting ions in the conditioned water.

The non-destructive detection efficiency value of the PB-C at a flow rate of 2.5 L min^{-1} was 0.77 [12] – higher than that of the Zn-C (0.67). The reason of this difference could be explained from the difference of adsorption ability of cartridge type. The fact that ^{137}Cs adsorption ability of the Zn-C was greater than PB-C at 2.5 L min^{-1} (**Figure 3(a)**) suggested the more ^{137}Cs absorbed onto the outer section of the fabric in Zn-C compared with PB-C. As a result, the non-destructive detection efficiency of Zn-C is lower than that of PB-C.

5. Conclusions

From our laboratory experiments, we reached the following conclusions.

- (1) The Zn-C was capable of absorbing 97.9% of dissolved ^{137}Cs from 20 L of water at a flow rate of 2.5 L/min; it was therefore more effective than conventional methods of concentrating dissolved ^{137}Cs from fresh water.
- (2) As was the case with the PB-C, the efficiency of the Zn-C decreased with increasing flow rate and with increasing flow volume.
- (3) High recovery efficiency of ^{137}Cs was achieved for the Zn-C over a pH range of 3–10; the Zn-C can therefore be used to extract ^{137}Cs from fresh waters with a wide range of pH values unlike with the PB-C.
- (4) The Zn-C could absorb at least $1 \times 10^{-6} \text{ g}$ of ionic cesium at a flow rate of 2.5 L/min – a much more efficient result than those reported for the PB-C.
- (5) The non-destructive detection efficiency of the Zn-C was possibly determined by the location of the cesium absorption sites on the filter medium. The localization of absorption sites is determined by flow rate, flow volume, and the concentration of coexisting ions. The non-destructive detection efficiency obtained for treatment of 20 L at water at a flow rate of 2.5 L/min was 0.67.
- (6) By comparing dissolved ^{137}Cs concentrations determined by using the Zn-C in river water samples from each of the two locations with those determined by using a conventional evaporative concentration method, we demonstrated clearly that the Zn-C could be used effectively to measure radiocesium concentrations in fresh waters.

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