

A simple and straightforward technique for analyzing radionuclides in seawater



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Technological innovation



Overview

^{90}Sr ^{137}Cs

~ 1 mBq/kg in seawater



Materials and Apparatus



AMP-PAN (or KNiFC-PAN)

DGA resin

2 mL column



SALT-100 (WITHTECH Ltd. ; South Korea)

Eight peristaltic pump
Flow rate: 10 – 100 mL min⁻¹
Applicable with 2 mL/ 5 mL column

https://www.withtech.co.kr/en/busi/new_busiList_5.php



Hidex Q-ARE

**Automated Radionuclide
Extraction System**

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A utomated
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E xtraction

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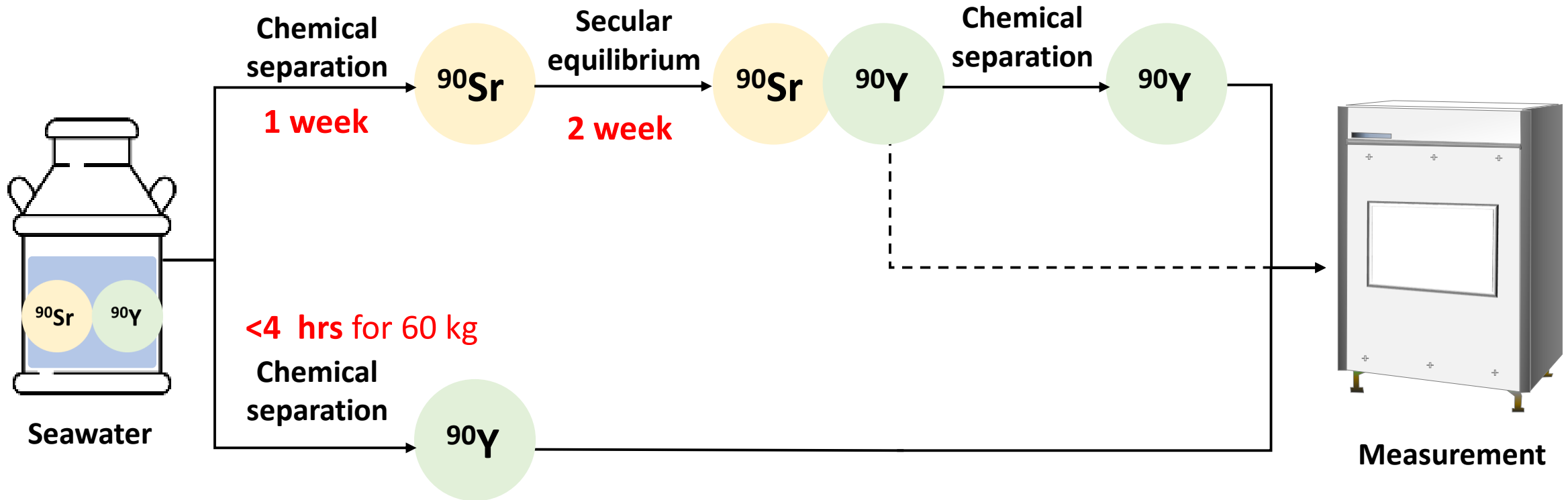
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^{90}Sr in seawater

This method focused on separation of ^{90}Sr from seawater sample.



This method focused on separation of ^{90}Y from seawater sample, which is equilibrium with ^{90}Sr

^{90}Sr in seawater; references

Talanta 152 (2016) 219–227

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Talanta

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Determination of strontium-90 from direct separation of yttrium-90 by solid phase extraction using DGA Resin for seawater monitoring

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^c College of Humanities and Sciences, Nihon University, 3-25-40, Sakurajosui, Setagaya-ku, Tokyo 156-8550, Japan

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ABSTRACT

It is important for public safety to monitor strontium-90 in aquatic environments in the vicinity of nuclear related facilities. Strontium-90 concentrations in seawater exceeding the background level have been observed in accidents of nuclear facilities. However, the analytical procedure for measuring strontium-90 in seawater is highly demanding. Here we show a simple and high throughput analytical technique for the determination of strontium-90 in seawater samples using a direct yttrium-90 separation. The DGA Resin is used to determine the abundance of strontium-90 by detecting yttrium-90 decay (beta-emission) in secular equilibrium. The DGA Resin can selectively collect yttrium-90 and remove naturally occurring radionuclides such as ^{40}K , ^{210}Pb , ^{214}Bi , ^{238}U , and ^{232}Th and anthropogenic radionuclides such as ^{140}Ba , and ^{140}La . Through a sample separation procedure, a high chemical yield of yttrium-90 was achieved at $95.5 \pm 2.3\%$. The result of IAEA-443 certified seawater analysis ($107.7 \pm 3.4 \text{ mBq kg}^{-1}$) was in good agreement with the certified value ($110 \pm 5 \text{ mBq kg}^{-1}$). By developed method, we can finish analyzing 8 samples per day after achieving secular equilibrium, which is a reasonably fast throughput in actual seawater monitoring. By processing 3 L of seawater sample and applying a counting time of 20 h, minimum detectable activity can be as low as 1.5 mBq kg^{-1} , which could be applied to monitoring for the contaminated marine environment. Reproducibility was found to be 3.4% according to 10 independent analyses of natural seawater samples from the vicinity of the Fukushima Daiichi Nuclear Power Plant in September 2013.

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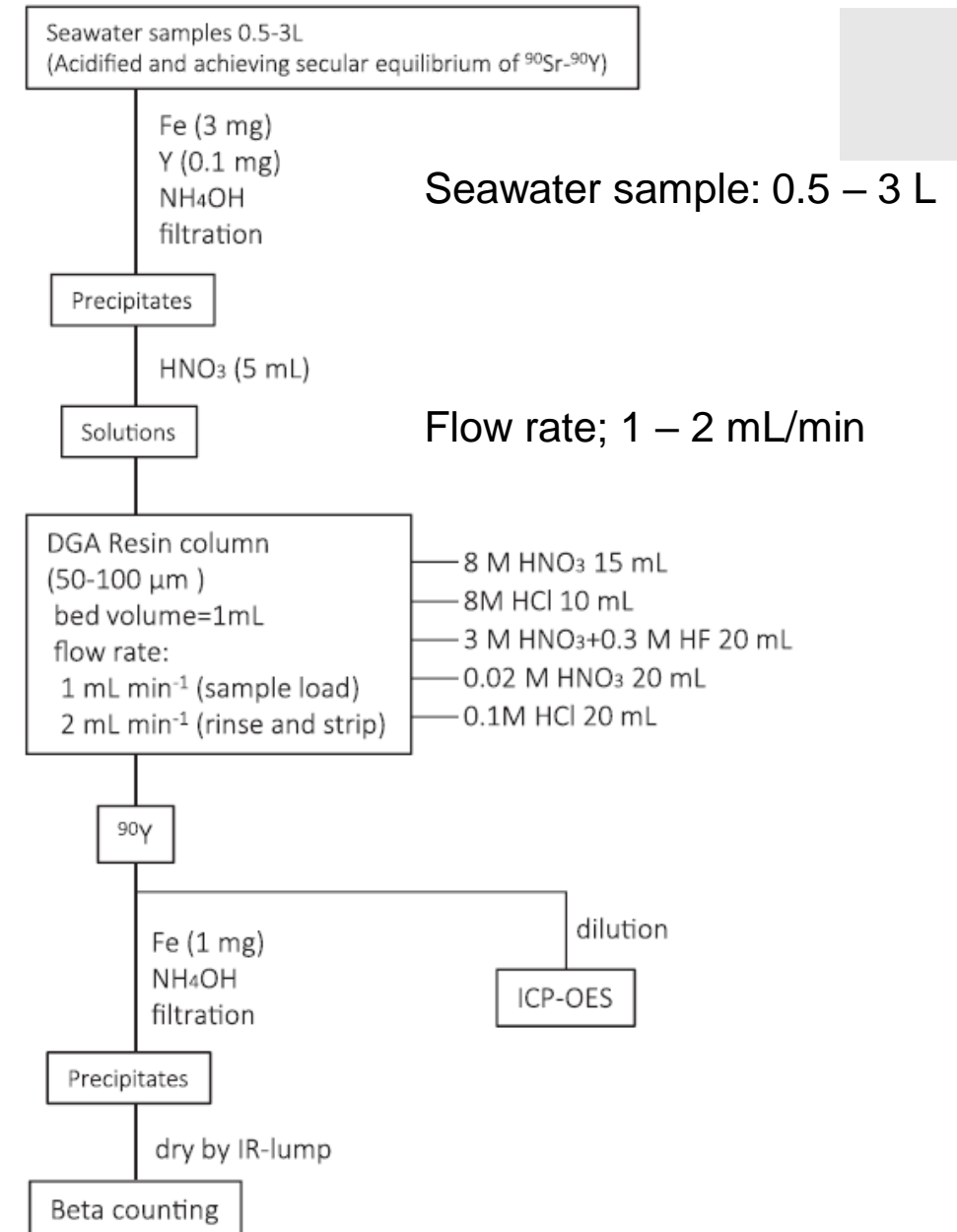


Fig. 2. Schematic chart of the analytical procedure for the determination of ^{90}Sr concentration in seawater.

^{90}Sr in seawater; references

Talanta 217 (2020) 121055

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Automated extraction chromatographic radionuclide separation system for analysis of ^{90}Sr in seawater

Hyuncheol Kim^{a,*}, Yoo Gyum Kang^{a,b}, Yong-Jin Lee^{a,b}, Sang-Do Choi^a, Jong-Myoung Lim^a, Jin-Hong Lee^b

^a Nuclear Emergency and Environmental Protection Division, Korea Atomic Energy Research Institute (KAERI), 989-111 Daedeok-daero, Yuseong-gu, Daejeon, 34057, Republic of Korea

^b Environmental Engineering, Chungnam National University, 99 Daehak-ro, Yuseong-gu, Daejeon, 34134, Republic of Korea

ARTICLE INFO

Keywords:
Automated separation system
Environmental monitoring
 ^{90}Sr
Seawater
Emergency preparedness

ABSTRACT

After the Fukushima Dai-ichi nuclear power plant disaster, the demand for a rapid method for the detection of environmental radioactivity increased drastically. Since the development of extraction chromatography using resins, analytical methods have advanced significantly in terms of simplicity and required labor. Herein, a home-made automated separation system that is applicable radio-extraction chromatographic separation techniques is reported. A simple, rapid, and high-throughput method was developed using this home-made automated separation system to analyze radiostromium in seawater in emergency and routine situations. For emergency situations, radiostromium in seawater is pre-concentrated on a cation exchange resin and consecutively purified using the Sr-resin. Fifty minutes are required for the purification of ^{90}Sr in four samples (100 ml). The minimum detectable activity (MDA) for ^{90}Sr is 0.2 Bq kg^{-1} at 100 min counting, with a recovery of 70% and counting efficiency of 95% in the scintillation mode. For routine monitoring, ^{90}Y that is in equilibrium with ^{90}Sr is first separated from the sample matrix using DGA. Treatment of 30 L of each seawater sample requires ~2 h. The MDA for this method is 0.3 mBq kg^{-1} at 400 min counting with a recovery of 70% and counting efficiency of 67% in the Cerenkov mode. By employing the developed method, the measured ^{90}Sr in seawater collected from the coastal area of Korea is $0.92 \pm 0.18 \text{ mBq kg}^{-1}$, which is comparable to that reported previously. The measurements were obtained using a liquid scintillation counter, and the entire separation process was performed by employing the home-made separation system.

Seawater sample: 10 ~ 30 L

Flow rate: 50 mL/min

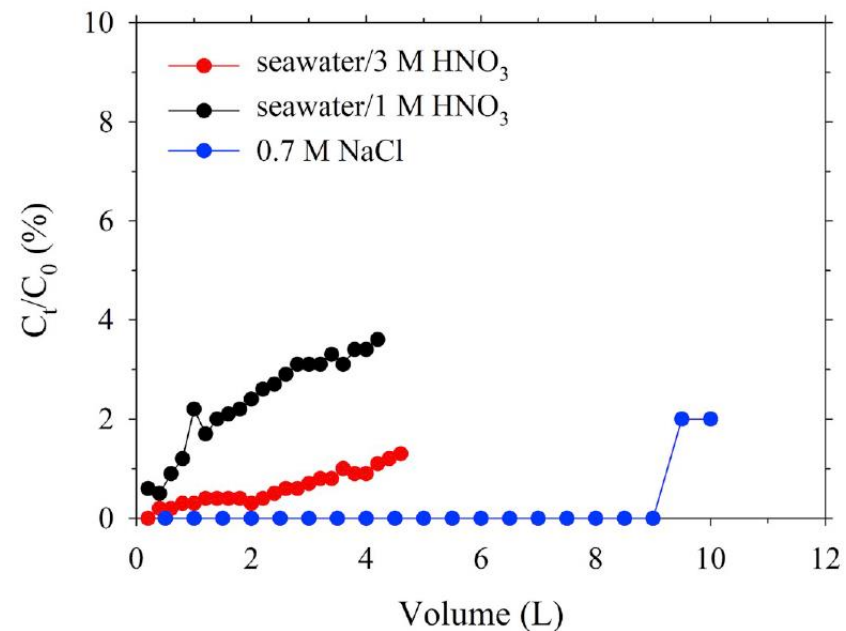


Fig. 4. Breakthrough of Y in different matrices of the sample.

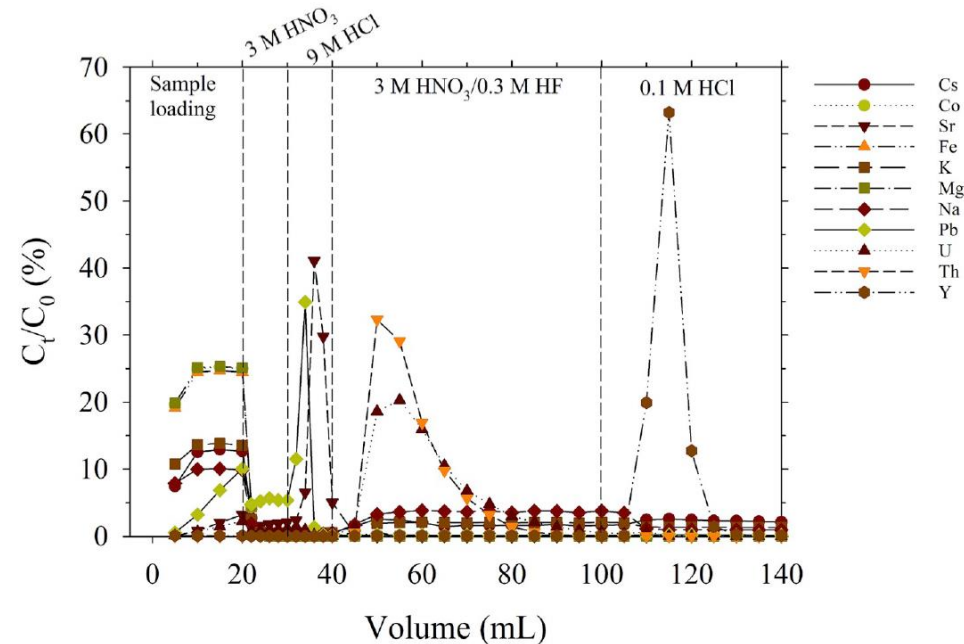


Fig. 5. Separation behavior of the interferences and Y on DGA.

^{90}Sr in seawater; references

Marine Pollution Bulletin 193 (2023) 115258

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul



Strontium-90 levels in seawater southeast of Jeju Island during 2021–2023

Gahyun Kim^{a,b}, Sang-Do Choi^a, Jong-Myoung Lim^a, Hyuncheol Kim^{a,b,*}

^a Environmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute, 111 Daedeokdae-ro 989 beongil, Yuseong-gu, Daejeon 34057, Republic of Korea

^b Nuclear Science and Technology, University of Science and Technology, 217, Gajeong-ro, Yuseong-gu, Daejeon 34113, Republic of Korea

ARTICLE INFO

Keywords:

Strontium-90
 ^{90}Sr analysis method
 Seawater
 Fukushima accident impact
 Marine radioactivity

ABSTRACT

This study introduces an efficient method for determining ^{90}Sr activity levels in seawater, reducing the processing time to <3 h for 50 L of seawater. The key feature of the proposed method is the chemical separation of ^{90}Y when it is in equilibrium with ^{90}Sr , which is achieved by utilizing custom-made sample-loading equipment and an automated radionuclide separation instrument. As a result, the procedure consistently yields a recovery rate > 90 % for ^{90}Y . Investigations of ^{90}Sr levels were conducted in the ocean southeast of Jeju Island from November 2021 to January 2023. Owing to the regional ocean circulation, this region was among the first within the Korean Peninsula to experience the impact of the Fukushima-accident-derived radionuclides. Throughout the investigation period, the observed ^{90}Sr activity concentration ranged from 0.57 to 1.0 Bq m⁻³. No distinct temporal variation of ^{90}Sr was observed in the selected area during the investigation.

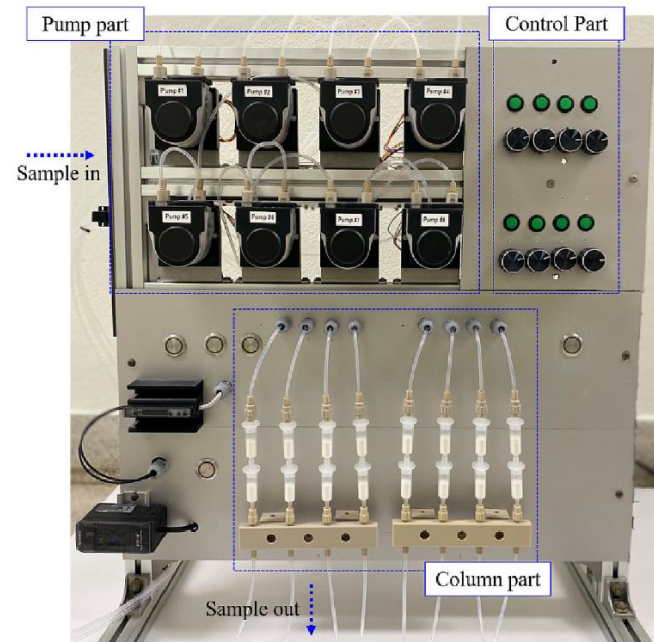
1. Introduction

Strontium-90 (half-life: 28.91 y) and ^{137}Cs (half-life: 30.08 y) have been recognized as two of the most hazardous artificial radionuclides owing to their high radiotoxicity and long half-lives (IAEA, 2023). As ^{90}Sr exhibits a chemical behavior similar to that of calcium and emits high-energy β particles along with its daughter ^{90}Y , it is accumulated in mammalian bones, causing bone cancer and leukemia (Vajda and Kim,

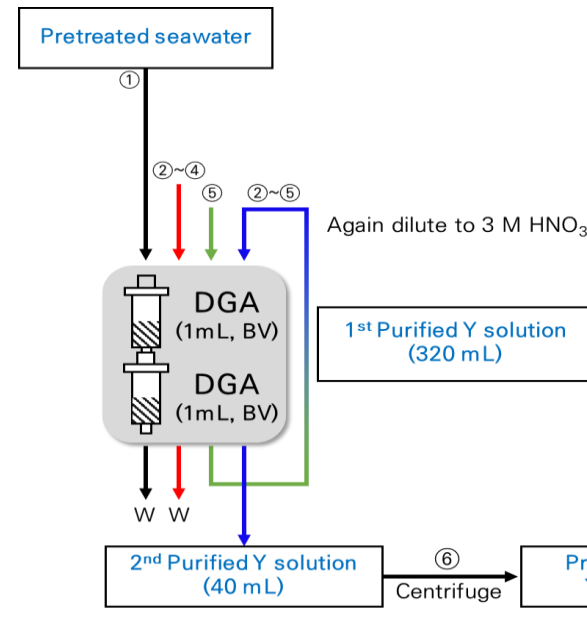
been used as an oceanic water mass tracer (Hirose and Povinec, 2020). As ^{90}Sr and ^{137}Cs are soluble in seawater and have similar half-lives, they have specific global fallout ratios (~0.63) (UNSCEAR, 2000). The Fukushima accident presented different oceanic inputs of ^{90}Sr compared to ^{137}Cs , thus causing deviations from the global fallout ratio between ^{90}Sr and ^{137}Cs (Povinec et al., 2013). For instance, seawater samples collected from the vicinity of the Fukushima Daiichi Nuclear Power Plant (FDNPP) in September 2013 yielded a $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratio of

Seawater sample: 50 - 100 L

Flow rate: 80 mL/min



Custom-made sample loading equipment



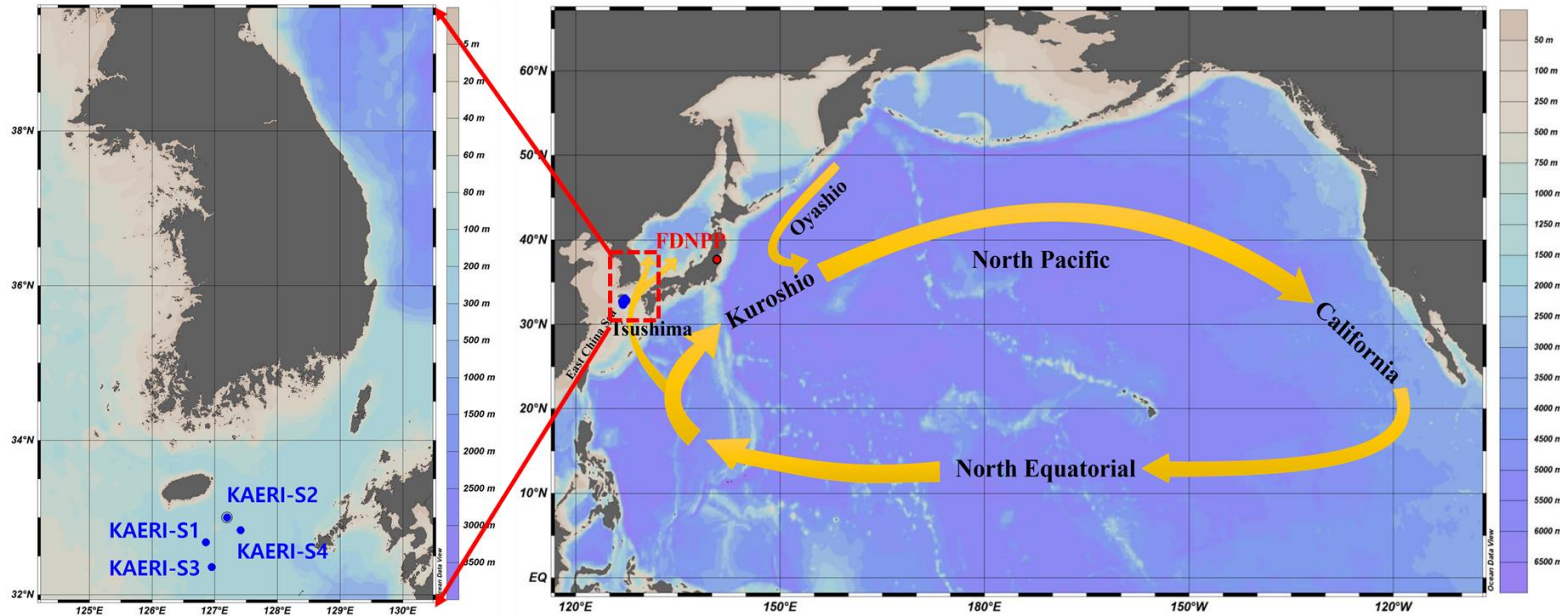
[Pretreatment]
 Filtration(GF/F)
 Dilute to 3 M HNO₃ using Conc. HNO₃
 Add Y carrier

[Chemical separation]
 ① Sample
 ② 10 mL of 3 M HNO₃
 ③ 10 mL of 9 M HCl
 ④ 60 mL of 3 M HNO₃/0.3 M HF
 ⑤ 40 mL of 0.1 M HCl
 ⑥ Adjust to pH 10 using NaOH
 ⑦ Dissolve with 10 mL of 0.1 M HNO₃

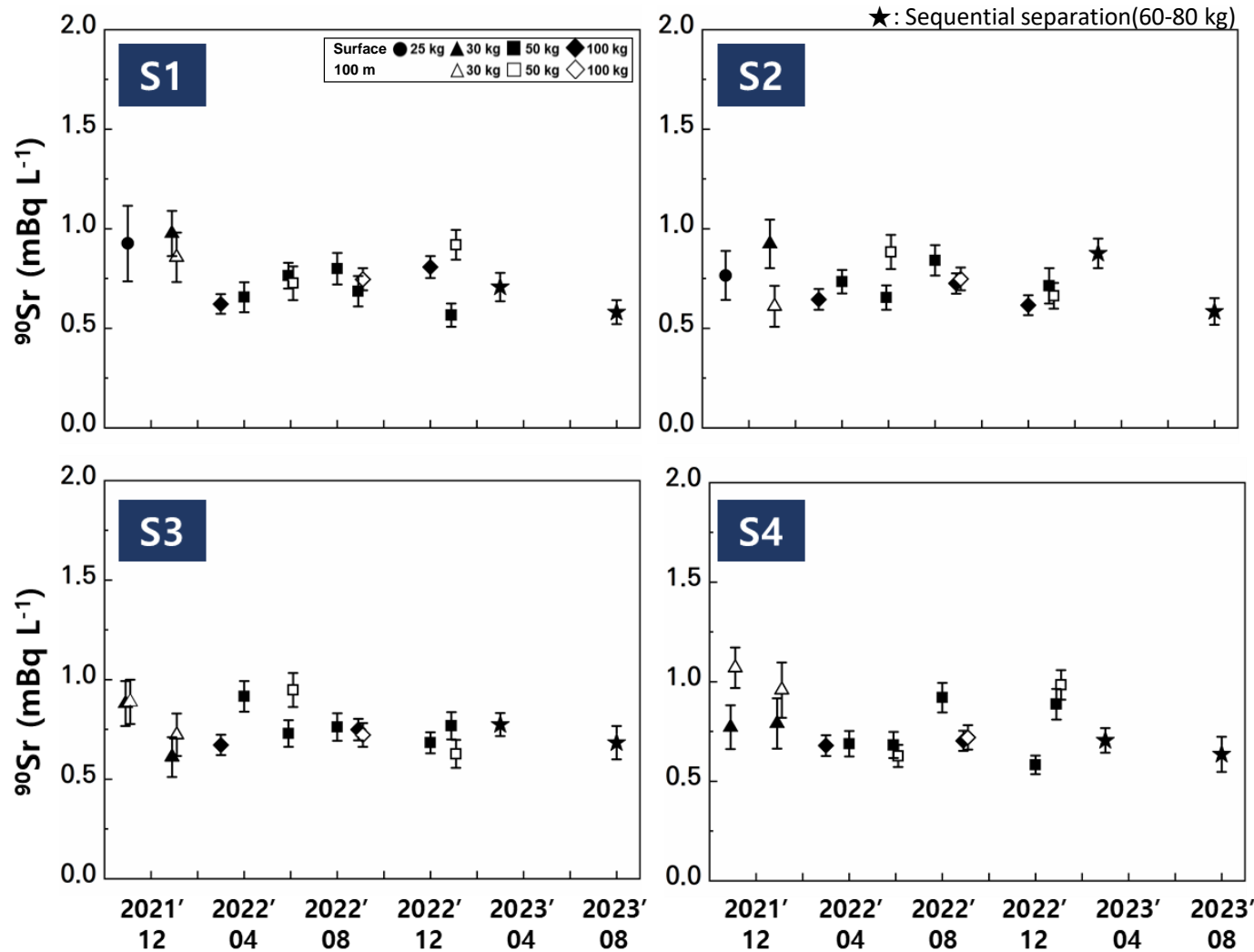
[Measurement]
 40 minutes each 10 times by LSC

^{90}Sr in seawater; procedure

Campaign for ^{90}Sr in the ocean (2021-2023)



^{90}Sr in seawater; procedure



Marine Pollution Bulletin 193 (2023) 115258

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Gahyun Kim ^{a,b}

^a Environmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute, 111 Daedeokdae-ro 989 beongil, Yuseong-gu, Daejeon 34057, Republic of Korea

ARTICLE INFO

Keywords: Strontium-90, ^{90}Sr analysis method, Seawater, Fukushima accident impact, Marine radioactivity

ABSTRACT

This study introduces an efficient method for determining ^{90}Sr activity levels in seawater, reducing the processing time to <3 h for 50 L of seawater. The key feature of the proposed method is the chemical separation of ^{90}Sr when it is in equilibrium with ^{90}Y , which is achieved by utilizing custom-made sample-loading equipment and an automated radionuclide separation instrument. As a result, the procedure consistently yields a recovery rate > 90 % for ^{90}Y . Investigations of ^{90}Sr levels were conducted in the ocean southeast of Jeju Island from November 2021 to January 2023. Owing to the regional ocean circulation, this region was among the first within the Korean Peninsula to experience the impact of the Fukushima-accident-derived radionuclides. Throughout the investigation period, the observed ^{90}Sr activity concentration ranged from 0.57 to 1.0 Bq m⁻³. No distinct temporal variation of ^{90}Sr was observed in the selected area during the investigation.

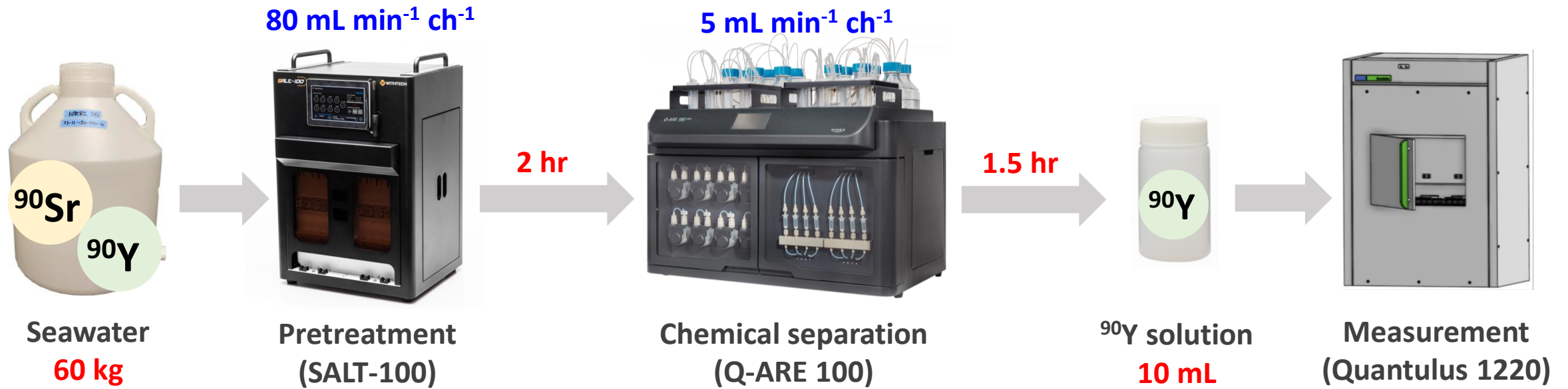
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Strontium-90, a highly soluble radionuclide with a long half-life, behaves conservatively in seawater and is transported via physical processes, such as advection, diffusion, and mixing. Therefore, ^{90}Sr has been used as an oceanic water mass tracer (Hirose and Povinec, 2020). As ^{90}Sr and ^{137}Cs are soluble in seawater and have similar half-lives, they have specific global fallout ratios (~0.63) (UNSCEAR, 2000). The Fukushima accident presented different oceanic inputs of ^{90}Sr compared to ^{137}Cs , thus causing deviations from the global fallout ratio between ^{90}Sr and ^{137}Cs (Povinec et al., 2013). For instance, seawater samples collected from the vicinity of the Fukushima Daiichi Nuclear Power Plant (FDNPP) in September 2013 yielded a $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratio of 0.28, which is lower than the global ratio (~0.63) (Castrillejo et al., 2016). The comparison of ^{90}Sr and ^{137}Cs activity in seawater can therefore aid in the estimation of the long-term impact of the Fukushima disaster on the marine environment (Hirose and Povinec, 2019). However, data regarding ^{90}Sr activity in the ocean are limited owing to the intricate, time-intensive, and laborious methods required for the determination of ^{90}Sr , in contrast to that of ^{137}Cs . As ^{90}Sr plays a significant role in tracing complex ocean circulation and understanding the behavior of the Fukushima-accident-derived radionuclides (Hirose and Povinec, 2019), an analytically friendly method for ^{90}Sr determination provides a better understanding of the marine environment.

During an investigation of the potential impact of the Fukushima accident, ^{134}Cs was observed in the South China Sea and the East China Sea from 2013 to 2014 (Wang et al., 2022). Despite its short half-life of 2.06 y (which makes it difficult to detect after 10 y), ^{134}Cs serves as a

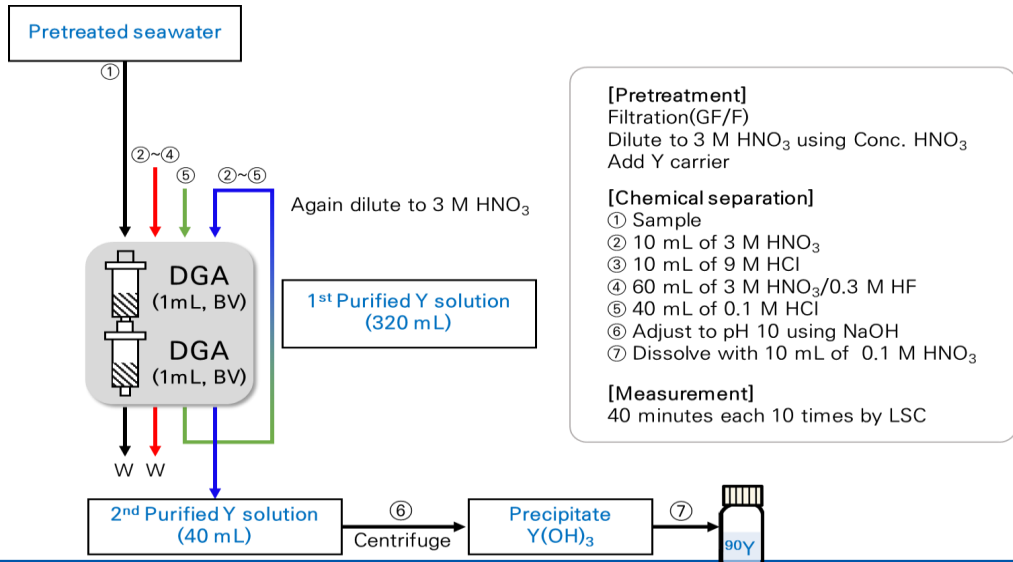
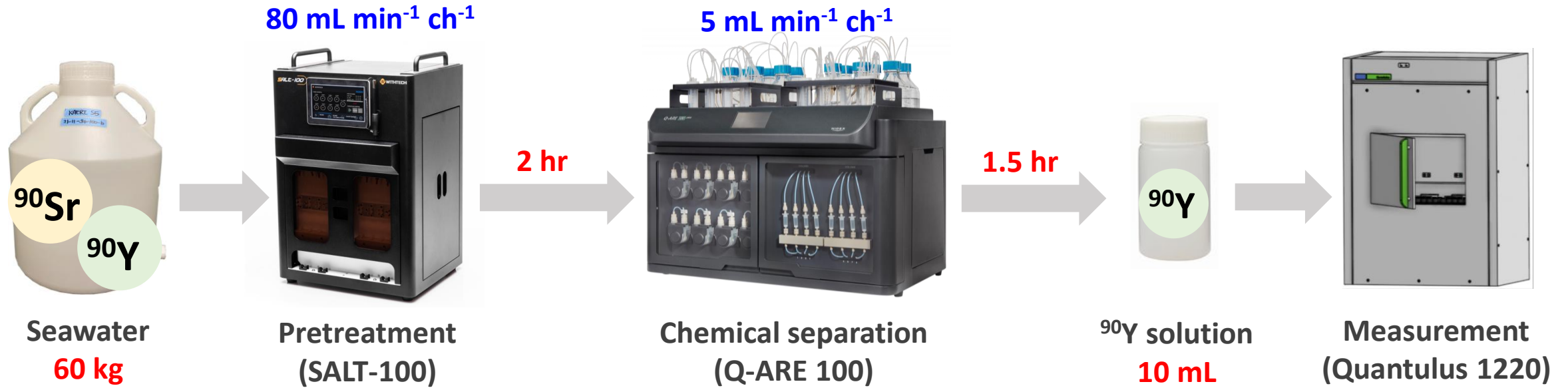
^{90}Sr in seawater; procedure



Elapsed time for analyzing the 60 kg of seawater?

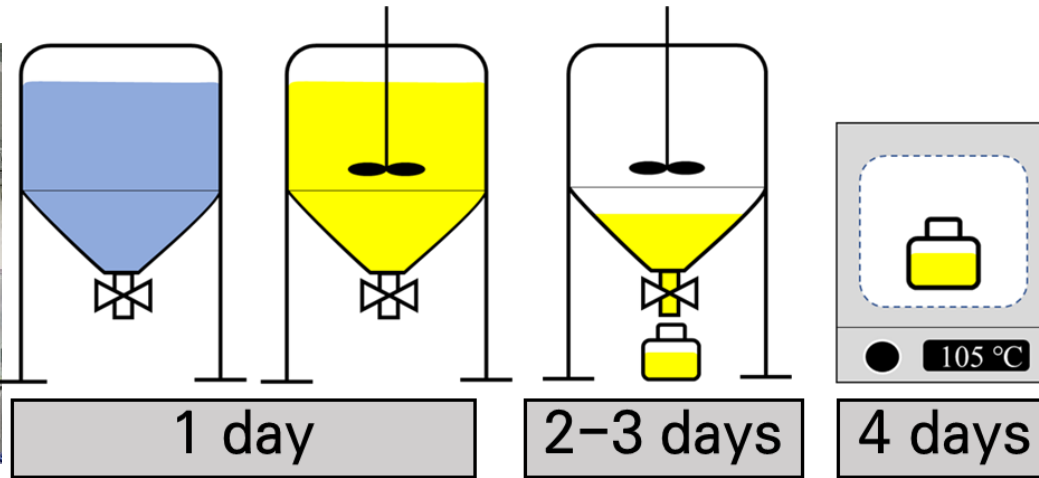
4 hr excepting the measurement time

^{90}Sr in seawater; procedure



Video clip~

^{137}Cs in seawater

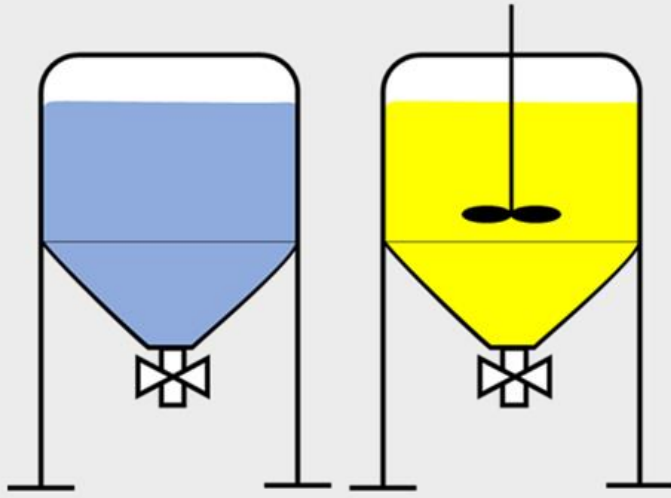


(From Sanghan Lee)

Ref. Korea Institute of Nuclear Safety(KINS)

^{137}Cs in seawater

Batch (using AMP powder)



- ✓ It is labor-intensive work.
- ✓ Elapsed time for analyze: **3-4 days**
(excepted measurement time)

Column chromatography (using AMP-PAN resin)



- ✓ User friendly and table-top equipment
(size: 0.4 m x 0.3 m x 0.6 m)
- ✓ Elapsed time for analyze: **within 1 day**
(excepted measurement time)

¹³⁷Cs in seawater; references

J Radioanal Nucl Chem (2013) 296:369–374
DOI 10.1007/s10967-012-2014-5

Extraction of cesium in seawater off Japan using AMP-PAN resin and quantification via gamma spectroscopy and inductively coupled mass spectrometry

S. M. Pike · K. O. Buesseler · C. F. Breier ·
H. Dulaiova · K. Stastna · F. Sebesta

Received: 13 July 2012 / Published online: 8 August 2012
© Akadémiai Kiadó, Budapest, Hungary 2012

Abstract The March 2011 earthquake off the Japanese coast and subsequent tsunami that devastated the Fukushima Dai-Ichi nuclear power plant resulted in the largest accidental release of cesium 137 and 134 to the oceans. Seawater samples were collected in June 2011 from 30 to 600 km off the coast of Japan as part of initial mapping of the spread of contamination in the ocean. Cesium was extracted from unfiltered and filtered (<1.0 μm) seawater using an absorber based upon an organic polymer polyacrylonitrile (PAN) containing ammonium molybdophosphate (AMP) Sebesta and Stefula (J Radioanal Nucl Chem 140:15–21, 1990). The AMP-PAN resin can be counted directly using gamma spectroscopy for ¹³⁴Cs and ¹³⁷Cs. Stable ¹³³Cs was added to evaluate extraction efficiency and quantified by ICP-MS. Our 5 mL AMP-PAN resin column was on average 95 % efficient in the removal of

cesium from 20 L samples at an average flow rate of 35 mL min⁻¹. Measured activities of ¹³⁷Cs ranged from a few Bq m⁻³ to >300 Bq m⁻³. The method can be adapted to different sample sizes and is easily used in the field.

Keywords Cesium · Fukushima · AMP-PAN · Gamma spectroscopy

Introduction

The Earthquake and following Tsunami off the coast of Japan on March 11, 2011 severely damaged several nuclear reactors at the Fukushima Dai-Ichi nuclear power plant (NPP). The total direct release of ¹³⁷Cs into the environment by the damaged reactors was

J Radioanal Nucl Chem (2013) 296:841–846
DOI 10.1007/s10967-012-2007-4

Fast concentration of dissolved forms of cesium radioisotopes from large seawater samples

Jan Kameník · Henrieta Dulaiova ·
Ferdinand Šebesta · Kamila Šťastná

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Abstract The method developed for cesium concentration from large freshwater samples was tested and adapted for analysis of cesium radionuclides in seawater. Concentration of dissolved forms of cesium in large seawater samples (about 100 L) was performed using composite absorbers AMP-PAN and KNiFC-PAN with ammonium molybdophosphate and potassium–nickel hexacyanoferrate(II) as active components, respectively, and polyacrylonitrile as a binding polymer. A specially designed chromatography column with bed volume (BV) 25 mL allowed fast flow rates of seawater (up to 1,200 BV h⁻¹). The recovery yields were determined by ICP-MS analysis of stable cesium added to seawater sample. Both absorbers proved usability for cesium concentration from large seawater samples. KNiFC-PAN material was slightly more

Keywords Cesium · ¹³⁴Cs · ¹³⁷Cs · Seawater · Composite ion-exchangers

Introduction

Cesium concentration from larger sample volumes is usually required for radioanalytical determination of ¹³⁴Cs ($T_{1/2} = 2.07$ years) and ¹³⁷Cs ($T_{1/2} = 30.08$ years) in seawater. Some of the well-established methods include cesium adsorption on ammonium molybdophosphate (AMP) (e.g., [1–3]) and co-precipitation with various insoluble hexacyanoferrates(II) (e.g., [4–6]). Applications of these materials in chromatography columns for seawater analysis were reviewed by Gaur [7]. Insoluble hexacy-

^{137}Cs in seawater; references



Fig. 1 Column used for concentration of cesium from 100 L of seawater (internal diameter 42 mm, 25 mL of AMP-PAN)

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Fast concentration of dissolved forms of cesium radioisotopes in large seawater samples

Table 2 Cesium recovery yields from seawater determined by ICP-MS

	Volume (L)	Average flow-rate (mL min ⁻¹)	Cs added (mg)	Recovery yield (%)
AMP-PAN (acidified seawater)				
A	106	280	25	88.7 ± 0.3
B	93	285	4	88.8 ± 0.4
C	101	319	4	84.9 ± 0.5
D	95	243	4	94.1 ± 0.3
E	98	326	4	86.3 ± 0.5
F	96	298	4	85.7 ± 0.6
KNiFC-PAN (acidified seawater)				
A	97	287	4	92.4 ± 0.4
B	98	288	4	94.4 ± 0.3
C	101	293	4	93.0 ± 0.4
D	102	260	4	91.5 ± 0.5
E	100	293	4	93.2 ± 0.5
KNiFC-PAN (natural seawater)				
A	102	279	4	91.9 ± 0.5
B	100	250	4	92.7 ± 0.5
C	98	276	4	88.3 ± 0.5
D	101	291	4	91.6 ± 0.5
E	100	315	4	90.9 ± 0.5
F	102	470	4	85.6 ± 0.6

August 2012

cesium concentra-
tested and adapted
seawater. Concen-
n large seawater
using composite
with ammonium
el hexacyanoferrate
ly, and polyacry-
specially designed
me (BV) 25 mL
o 1,200 BV h⁻¹).
ICP-MS analysis
le. Both absorbers
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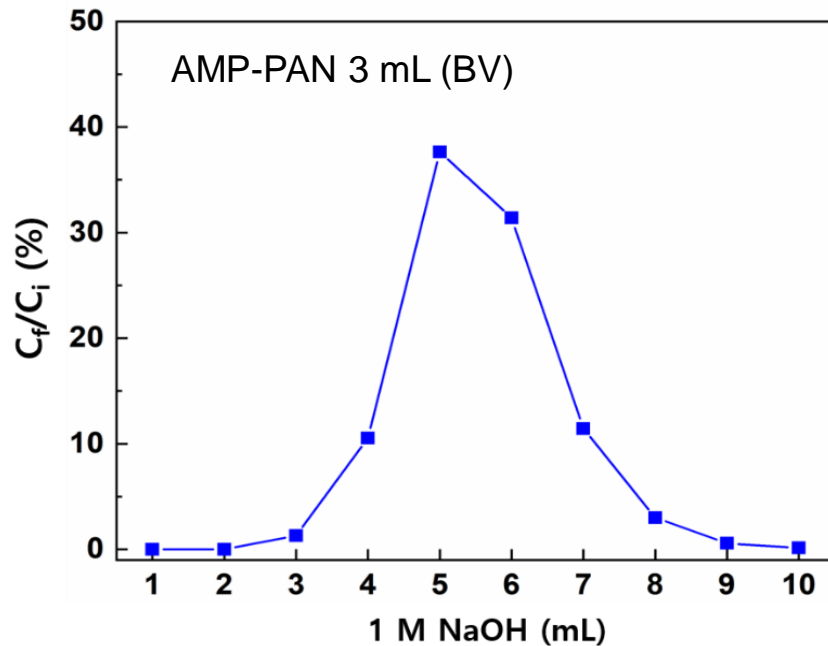
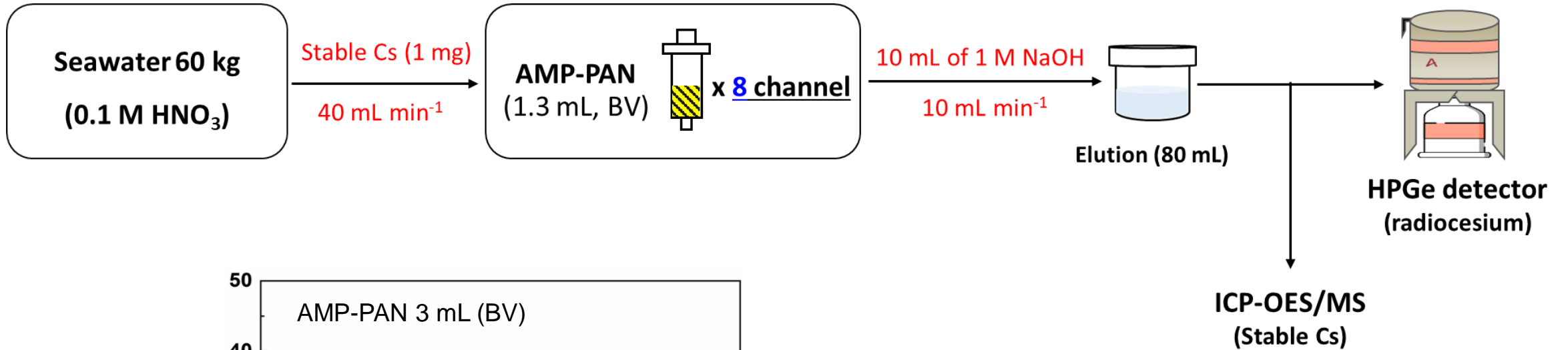
water samples. KNiFC-PAN material was slightly more

^{137}Cs in seawater; procedure

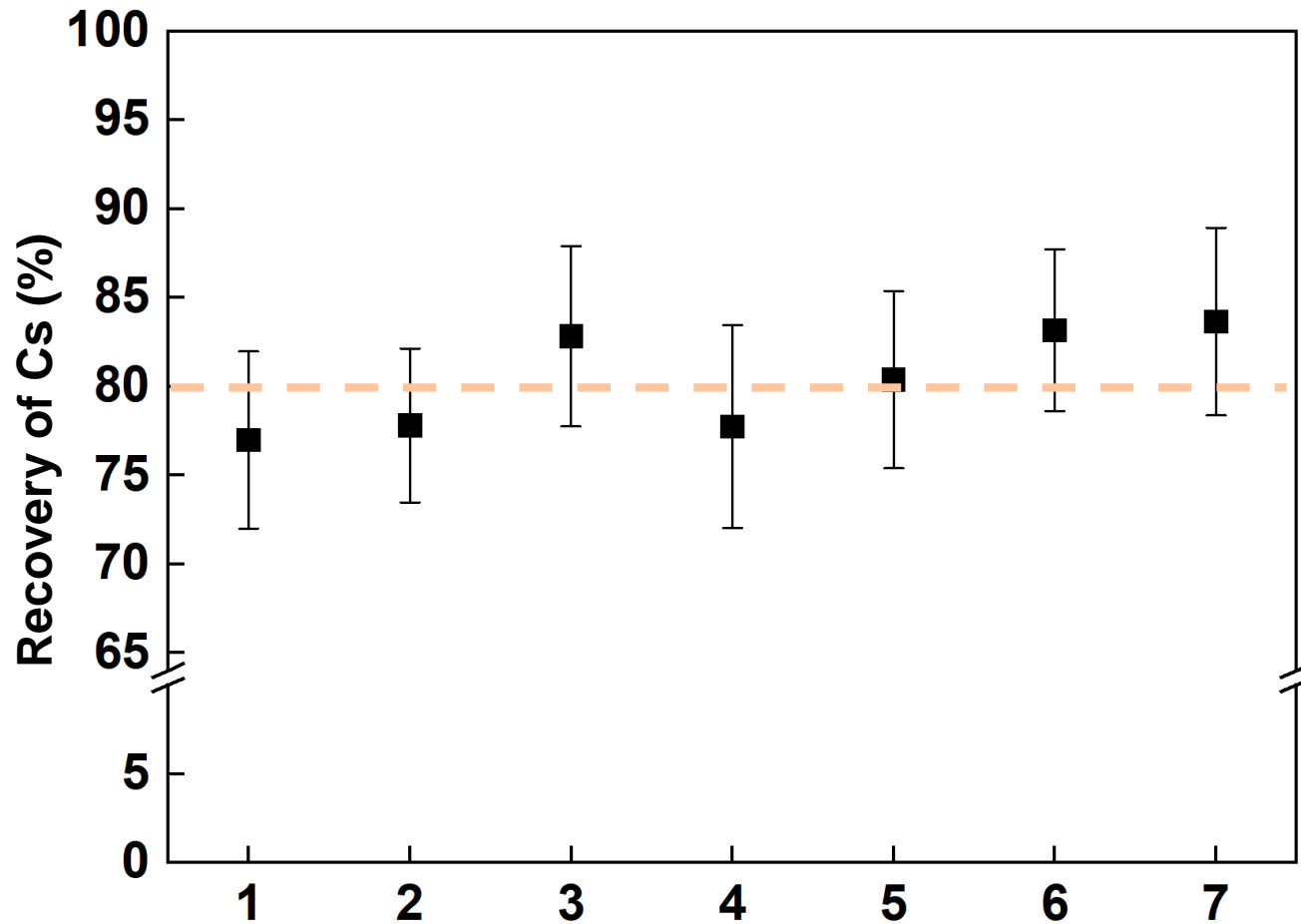


Elapsed time for analyzing the 60 kg of seawater?

4 hr excepting the measurement time



^{137}Cs in seawater; procedure

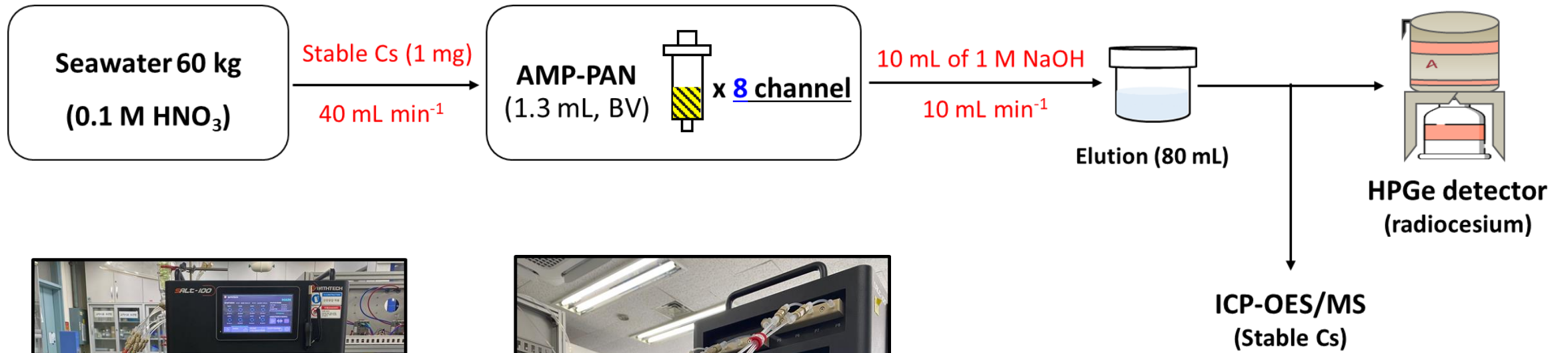


- Seawater : 60 kg
- Cs carrier: 1 mg
- AMP-PAN resin: 1.3 mL (BV) * 8 channel
- Loading flow rate: 40 mL min⁻¹



Average \pm SD: **80.4 \pm 2.9** (n=7)

^{137}Cs in seawater; procedure



Sample loading



Cs elution

1. AMP-PAN column: 1.3 mL (BV)

Video clip~

Thank you