

A Rapid and Efficient Automated Method for the Sequential Separation of Plutonium and Radiostrontium in Seawater

Hyuncheol Kim^{1*}, Kun Ho Chung¹, Yoonhee Jung^{1,2}

Mi Jang¹, Mun ja Kang¹, Geun-Sik Choi¹

¹*Environmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute*

989-111 Deadeok-daero, Yuseong-gu, Daejeon, 305-353, Korea

²*Korea University of Science and Technology*

UST 217, Gajeong-ron, Yuseong-gu, Daejeon, 305-350, Korea

Keyword: plutonium, radiostrontium, emergency preparedness, automated radionuclide separator, seawater

* corresponding author: H. Kim (hckim3@kaeri.re.kr)

Abstract

In this work, a novel method for the simultaneous separation of Pu and ^{90}Sr in seawater is proposed that is based on precipitation and extraction chromatography with an automated radionuclide separator. In particular, Pu from seawater is co-precipitated with $\text{Fe}(\text{OH})_2$; ^{90}Sr is precipitated as SrCO_3 . The precipitates are then dissolved in HNO_3 and sequentially separated using TEVA and Sr-resin columns on an automated radionuclide separator (MARS, Modular Automated Radionuclide Separator). The yield of Pu and Sr from 1 L and 10 L of seawater ranged between 50 and 74 %, and between 77 and 95 %, respectively. The separation time for 1 L and 10 L seawater samples was estimated as 3.2 h and 9.4 h, respectively.

1 Introduction

2 Dangerous artificial radionuclides such as cesium, plutonium and $^{89/90}\text{Sr}$ are released into the
3 environment as a result of accidents in nuclear facilities or of nuclear weapon testing [1, 2]. To achieve a
4 full radiological protection of the environment, it is crucial to develop efficient methods that allow the
5 monitoring of these artificial radionuclides. After the Fukushima Daiichi Nuclear Power Plant (FDNPP)
6 accident, the dramatic increase in $^{134/137}\text{Cs}$ concentration in seawater has been well documented [3]. In
7 contrast, plutonium and $^{89/90}\text{Sr}$ concentrations in seawater have been rarely reported [4]. This may be due
8 to i) the difficulty of achieving an efficient separation of these radionuclides; ii) the tedious sample
9 treatment needed to determine the alpha/beta emitters as opposed to the analysis of gamma emitters in
10 seawater. Thus, the development of a rapid and valid analytical method is essential to assess the potential
11 hazards, and to allow early decision-making in case of a nuclear accident.

12 Sequential separation of Pu and Sr in environmental samples is a promising technique that can address
13 these issues. The great advantage is that chemical separation is performed only once for the two
14 radionuclides. To perform the sequential separation, several approaches have been proposed including
15 combined procedures of the anion-exchange resin and Sr-resin [5] or TRU and Sr resin [6]. Although the
16 above mentioned methods are normally reliable and accurate, most of them are operated manually and are
17 therefore labor intensive and time-consuming because of employing the gravity flow method. Maxell [7]
18 reported a sequential separation method for actinides and $^{89/90}\text{Sr}$ in environmental aqueous samples. The
19 method was based on the use of TEVA and Sr resins stacked on a vacuum box, which could reduce just
20 the analytical time as compared to the gravity-flow method. However, it still needs much manual
21 operations such as handling reagents.

22 In this study, we propose a sequential separation of Pu and ^{90}Sr from seawater using automated
23 separation system with minimal operational work. Pu and ^{90}Sr from seawater were concentrated by co-
24 precipitation/precipitation like $\text{Fe}(\text{OH})_2$ and SrCO_3 . Then they were purified by TEVA and Sr resin. The
25 behavior of Pu on TEVA and Sr on Sr-resin was investigated to optimize the operational procedure. The
26 performance of this method was tested for 1 L and 10 L seawater samples spiked with ^{238}Pu and ^{90}Sr . This
27 is the intermediate study to develop the fully automated sequential separation system for radionuclides.

29 **Experimental**

30 *Reagents*

31 Powders for TEVA (100~150 μm) and Sr resin (100~150 μm) were obtained from Eichrom
32 [Technologies](#), Inc. (Illinois, USA). The resins were swollen in water for 2 h prior to use. All solutions
33 were prepared using de-ionized water ([DIW](#), MilliQ-Plus, 18M Ω). ^{241}Pu , ^{242}Pu and ^{90}Sr were purchased
34 from the National Institute of Standards and Technology (NIST, Gaithersburg, MA, USA). ^{238}Pu was
35 obtained from [Korea Research Institute of Standards and Science](#) (KRISS, Daejeon, Korea). ^{241}Pu was
36 used to estimate the behavior of Pu on the TEVA. ^{242}Pu was used as a tracer in the whole procedure. ^{85}Sr
37 was obtained from Eckert & Ziegler Analytics Inc (GA, USA) and used as a tracer.

38

39 *Determination*

40 Cations such as Sr, Ca and Mg ions were measured by Inductively Coupled Plasma Optical Emission
41 Spectrometers (ICP-OES, Varian, USA). ^{85}Sr was analyzed by [gamma spectrometer \(\$\gamma\$ -spectrometer,](#)
42 [HPGe detector, Canberra, USA\)](#). Alpha spectrometer (α -spectrometer, Ortec, USA) was used for the
43 analysis of $^{238/242}\text{Pu}$. $^{238/242}\text{Pu}$ in 30 ml 1M HCl was taken in 100 ml Teflon beaker. It was evaporated to
44 dryness at 150 $^{\circ}\text{C}$ and dissolved in NaHSO₄ and Na₂SO₄ solution. Then it was electrodeposited on
45 stainless steel discs at a constant current of 0.9 A from (NH₄)₂C₂O₄ solution for 2 h. Liquid Scintillation
46 Counter (LSC, Quantulus 1220, Perkin Elmer, USA) was used for the analysis of ^{241}Pu , ^{90}Y ingrowth
47 from ^{90}Sr . ^{241}Pu in 30 ml 1M HCl was transferred in 20 ml glass vial and evaporated to dryness at 110 $^{\circ}\text{C}$.
48 8 mL of a 0.05 M HNO₃ solution was added to each glass vial and mixed with 12 mL of Ultima Gold
49 LLT. For analysis of ^{90}Sr by LSC, 0.5 ml of Sr carrier (10 mg ml⁻¹) was taken in 50 ml volume of conical
50 tube. The pH of the solution was adjusted to 10 with 25 % ammonia. Then, 5 mL of a saturated Na₂CO₃
51 solution was added to precipitate SrCO₃. The precipitate was transferred in a 20 mL glass vial and washed
52 with ethanol. It was dried at 80 $^{\circ}\text{C}$ and dissolved in 8 mL of 0.05 M HNO₃. Unfortunately SrCO₃
53 containing stable Sr 70 mg was not dissolved in 8 ml of 0.05 M HNO₃. Therefore, 8 ml of 0.5 M HNO₃
54 was used to dissolve SrCO₃. Then ^{90}Y fraction from ^{90}Sr was determined on LSC using Cerenkov
55 counting.

56

57 *Samples*

58 Seawater samples (salinity 3.2 %) were collected in the East Sea. All seawater samples were filtered
59 through a cellulose filter (0.45 µm). The samples were acidified to pH 2 with HCl and spiked with ⁹⁰Sr
60 (94 Bq) and ²³⁸Pu (0.0117 Bq). ²⁴²Pu (0.029 Bq) and ⁸⁵Sr (1 Bq) were added as tracers for Pu and Sr,
61 respectively. The concentrations of Ca, Mg and Sr were 350 mg L⁻¹, 1150 mg L⁻¹ and 6.5 mg L⁻¹,
62 respectively.

63

64 *Column preparation*

65 The powders of TEVA and Sr resin swollen in DIW were packed into a borosilicate glass column
66 (BenchMark, Omnifit, USA). The Sr resin was packed in a 10 mm × 100 mm column and in a 15 mm ×
67 400 mm column to achieve the desired resin volume. 4 ml (BV) of Sr resin (1.2 g) was used for the
68 purification of Sr in 1 L of seawater. 35 mL (BV) Sr resin (12 g) was used to separate Sr from 10 L of
69 seawater. TEVA resin was packed in a 10mm × 100mm column to obtain a BV of 4 mL (1.2 g) for 1 L
70 seawater or 6 mL (1.8 g) for 10L seawater, respectively.

71

72 *Apparatus – automated radionuclide separator*

73 The entire purification procedure was performed with MARS developed in our laboratory [8]. It is based
74 on fluidic systems using column and can control the flow rate and the types of the reagents in the system.
75 This allows a significant reduction of working labor. MARS was originally designed for the automated
76 single-radionuclide separation of four samples. In this study, it was modified to allow the sequential
77 separation of two radionuclides in a single sample with a minimum manual work. The sample was loaded
78 on TEVA/Sr resin (Figure 1-a). After all Sr in sample was eluted from TEVA column, Sr resin column
79 was manually disconnected from MARS (Figure 1-b). Washing solutions and eluent for Pu were passed
80 through TEVA column. Then, TEVA column was replaced by Sr resin column (figure 1-(c)). Washing
81 solutions and eluent for Sr were passed through Sr resin. Except connection/disconnection of columns, all
82 operational conditions (e.g., loading, washing and elution flow rates and the chemical composition of
83 reagents) were controlled via software developed in our laboratory [8]. The 6-inlet ports flow selection
84 valve (6I-FSV) is used to select different reagents, and the 2-inlet ports flow selection valve (2I-FSV) is

85 used to select a sample or a reagent. A 3-way flow distribution valve (3W-FDV) was used to obtain the
86 optimized volume of eluents in the elution step.

87

88 *Concentration of Pu and Sr in seawater*

89 The filtered seawater samples were acidified to pH 2 using HCl to avoid loss of actinides. This point is
90 crucial because hydrolyzed actinides have a great affinity to the surfaces of containers [9]. 1 g of $\text{Na}_2\text{S}_2\text{O}_5$
91 was added per 1 L seawater to adjust to oxidize all Pu to Pu(III). After that, 30 mg of Fe^{3+} as $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$
92 was added with stirring. It adjusted to pH 10 by 25 % ammonia to precipitate $\text{Fe}(\text{OH})_2$. 15g of Na_2CO_3
93 was added to precipitate SrCO_3 with intense mixing for 30 min. Concentrated HNO_3 was added to
94 dissolve the precipitate. Pu(III) can be oxidized to Pu(IV) with HNO_3 [10]. The concentration of HNO_3 in
95 the sample medium was adjusted to 6 M, before introducing it into the TEVA and the Sr-resin columns.

96 Concerning the recovery of Sr, 1 L seawater samples were tested using different amounts of Na_2CO_3
97 (from 5 g to 15 g), as described above. The precipitates were filtered by a glass fiber filter (GF/F,
98 Whatman). Concentration of Ca, Mg and Sr in the filtrate were analyzed by ICP-OES and compared with
99 the initial concentration. Recovery of Sr in 1 L seawater was increased with the amount of Na_2CO_3 and
100 was over 90 % with 15 g Na_2CO_3 (table 1). Most of divalent cations such as Ca and Mg were precipitated
101 as hydroxide and carbonate salt, also.

102

103 *Behavior of Sr and Pu on TEVA*

104 For efficient sequential separation of Pu and Sr, it is important to determine when Sr is eluted from
105 TEVA. TEVA and Sr resin column should be separated at that time. 25 mL of a 6 M HNO_3 solution,
106 containing ^{85}Sr and 10 mg of stable Sr, was passed through the 4 ml TEVA at 2 ml min^{-1} . After passing
107 the sample, 30 ml of 8 M HNO_3 was added to remove Sr at 2 ml min^{-1} . Every 3ml of solution coming
108 from TEVA was taken in 20 ml plastic vial and ^{85}Sr was determined (Fig 2-a). Sr was stripped from
109 TEVA resin with additional 15 ml of 8 M HNO_3 .

110 The elution behavior of Pu on TEVA 4 ml (BV) and 6 ml (BV) column was determined by LSC using
111 ^{241}Pu (Fig 2-b). 20 mL of a 6 M HNO_3 solution spiked with ^{241}Pu was passed through the TEVA at 2 ml
112 min^{-1} . ^{241}Pu was eluted with 2 ml of 1 M HCl using about 36 ml 1 M HCl at 1 ml min^{-1} .

113

114 *Behavior of Sr on Sr resin*

115 Fig. 3 shows the elution behavior of Sr on Sr resin column. 10 mg and 70 mg of stable Sr in 8 M HNO₃
116 was passed through Sr resin 4 ml (BV) and 36 ml (BV) column at 2 ml min⁻¹, respectively. DIW was used
117 to elute Sr from Sr resin at 2 ml min⁻¹.

118

119 *Purification of Pu and Sr on MARS*

120 Processing standards were prepared to test purification steps of Pu and Sr on MARS. It contains ²⁴¹Pu,
121 ⁸⁵Sr and 7 mg stable Sr in 20 ml of 6 M HNO₃. It was loaded on 4 ml TEVA/4 ml Sr resin columns at 2
122 ml min⁻¹. Additional 15 ml 8 M HNO₃ was used to strip Sr from TEVA at 2 ml min⁻¹. As previously
123 described, Sr resin was removed from MARS. 20 ml of 1 M HNO₃ solution and 30 mL of 9 M HCl
124 solution were in order used to wash TEVA at 2 ml min⁻¹. 24 ml of 1 M HCl was used to elute Pu at 1 ml
125 min⁻¹. Sr resin column was then reconnected on MARS instead of TEVA column. 20 ml of 8 M HNO₃
126 was used to wash Sr resin at 2 ml min⁻¹. Sr was eluted with 20 ml DIW at 2 ml min⁻¹.

127

128 **Results and discussion**

129 *Recommend procedure for simultaneous Pu and Sr from seawater*

130 Based on above results, we propose the sequential separation of Pu and Sr from seawater as follows:

- 131 1. Add 1 g of Na₂S₂O₅ and 30 mg of Fe³⁺ with stirring
- 132 2. Adjust of pH to 10 by ammonia and then add 15 g of Na₂CO₃ with mixing for 30 min
- 133 3. Filter the sample by GF/F and dissolve the precipitates in concentrated HNO₃
- 134 4. Pre-condition TEVA (4 ml)/Sr resin (4ml) with 8M HNO₃ and load sample at 2 ml min⁻¹
- 135 5. Wash TEVA/Sr resin with 15 ml of 8 M HNO₃ at 2 ml min⁻¹ and then disconnect Sr resin column
136 from MARS
- 137 6. Wash TEVA with 15 ml 1 M HNO₃ and 30 ml 9 M HCl at 2 ml min⁻¹
- 138 7. Elute Pu from TEVA with 24 ml of 1 M HCl at 1 ml min⁻¹
- 139 8. Disconnect TEVA column and re-connect Sr resin column on MARS
- 140 9. Wash Sr resin with 20 ml 8 M HNO₃ and elute Sr with 20 ml DIW at 2 ml min⁻¹

141 The amount of reagents and bed volume of TEVA/Sr resin for 10 L seawater was written in Fig 4. The
142 separation time of Pu and Sr for 1 L and 10 L seawater was measured as 3.2 h and 9.4 h, respectively.

143

144 *Determination of ⁹⁰Sr and ²³⁸Pu in spiked seawater by proposed procedure*

145 Table 2 shows the analytical results for five samples of 1 L seawater and two samples of 10 L, spiked
146 with 94.2 Bq of ⁹⁰Sr and 0.0115 Bq of ²³⁸Pu. The average ⁹⁰Sr result was 91.6 ± 2.1 Bq L⁻¹ (1 SD,
147 standard deviation) with an average bias of -2.7 %. The average ²³⁸Pu was 0.0116 ± 0.0008 Bq L⁻¹ (1 SD)
148 with an average bias of -1.1 %. These excellent results confirmed that the proposed procedure is very
149 reliable. The recovery of Sr and Pu was 87.8 ± 6.5 % and 62.5 ± 10.4 % (1 SD) for sets of seawater,
150 respectively.

151 The recovery of Pu in this method is relatively lower than obtained by Maxwell et al. (87.7 ± 6.4 %)
152 [11]. When the processing standards were followed just the purification steps on MARS, the recovery of
153 Pu was higher than 90 %. The discrepancy mentioned above may result from incomplete valence
154 adjustments of Pu in concentration step before introducing to TEVA resin. The step for the valence
155 adjustments of Pu should be improved for better recovery of Pu. For 10 L seawater, the recovery of Sr
156 was lower than that in 1 L seawater. Although Sr recovery on Sr resin was quantitative for Ca level up to
157 320 mg per Sr resin 2 ml (BV), chemical yields declined above that level [12]. Most of Ca in seawater
158 was precipitated with Sr in the concentration step (table 1). It is shown that high level of Ca in sample
159 loaded on Sr resin results in a small deterioration of Sr recovery for 10 L seawater.

160 The minimum detectable activity (MDA) for radionuclides using this method with alpha spectrometry
161 and LSC was calculated with the equations from Currie [13]:

$$MDA = \frac{(2.71 + 4.65\sqrt{BKG \cdot C_T})}{60 \times C_T \times R \times E_C \times V_{SMP}}$$

162 where BKG is the background count rate; C_T is the counting time; R is the chemical recovery; V_{SMP} is
163 the sample volume (L); E_C is the detector counting efficiency; 60 is the conversion factor from dpm to Bq.
164 The MDA for ⁹⁰Sr is 5.2 mBq L⁻¹ with 100 min counting, V_{SMP} 10 L, BKG 10 cpm, E_C 60% and R 80.0 %.
165 MDA for ²³⁸Pu is 39 μBq L⁻¹ with 2 days counting, V_{SMP} 10 L, BKG 1 count, E_C 20% and R 54.2 %.

166

167 A large-volume sampling (typically 50 ~ 200 L) is necessary for a reliable analysis of Pu and Sr in
168 seawater because of their low concentration [14, 15]. However, in an emergency scenario, such as the
169 FDNPP accident, smaller amounts of seawater can be used to estimate the radiological activity. This is
170 because the concentration of a radionuclide increases drastically after an accident. Casauberta et al. [16]
171 reported that the level of ^{90}Sr ranged between 8 and 85 mBq L^{-1} in surface water which was 10 times
172 larger than the pre-Fukushima level (1.2 mBq L^{-1}). In this study, the target sample aliquot is about 10 L,
173 which is 1/10 of a typical sample volume for background-level analysis. In emergency situation, ^{89}Sr level
174 is high, also. The analysis of ^{89}Sr was not shown in this work. However, if $^{89/90}\text{Sr}$ results are required,
175 Cerenkov counting is useful technique for determination of $^{89/90}\text{Sr}$ [17].
176

176

177 **Conclusion**

178 A procedure based on an automated radionuclide separator was proposed to simultaneously separate
179 ^{238}Pu and ^{90}Sr from seawater with a reliable performance. The separation time was measured to be 3.2 h
180 and 9.4 h, for the 1 L and 10 L seawater samples, respectively. Although MDA in this approach is higher
181 than that of the background levels of ^{238}Pu and ^{90}Sr in the ocean, our method is suitable in the event of
182 emergencies, in which concentration of plutonium and $^{89/90}\text{Sr}$ drastically increases. Our results show that
183 MARS has a small limit such as column replacements. The development of a fully automated procedure
184 is currently being investigated in our laboratory for a safe and rapid sequential separation of several
185 radionuclides.
186

186

187 **Acknowledgements**

188 This work was performed under the auspices of the Ministry of Science, ICT and Future Planning (MSIP)
189 of Korea, NRF contract No. 2012M2A8A4025915.

190 **References**

- 191 1. Perelygin, V. P. and Y. T. Chuburkov (1997). Man-made plutonium in environment - Possible serious
192 hazard for living species. *Radiat Meas* 28 (1-6):385-392
193
- 194 2. Vajda, N. and C. K. Kim (2010). Determination of radiostrontium isotopes: A review of analytical
195 methodology. *Appl Radiat Isotopes* 68 (12):2306-2326
196
- 197 3. Chino, M., H. Nakayama, H. Nagai, H. Terada, G. Katata and H. Yamazawa (2011). Preliminary
198 estimation of release amounts of ¹³¹I and ¹³⁷Cs accidentally discharged from the Fukushima Daiichi
199 Nuclear power plant into the atmosphere. *J Nucl Sci Technol* 48 (7):1129-1134
200
- 201 4. Povinec, P. P., K. Hirose and M. Aoyama (2012). Radiostrontium in the western North Pacific:
202 Characteristics, behavior, and the Fukushima impact. *Environ Sci Technol* 46 (18):10356-10363
203
- 204 5. Moreno, J., N. Vajda, P. R. Danesi, J. J. Larosa, E. Zeiller and M. Sinojmeri (1997). Combined
205 procedure for the determination of ⁹⁰Sr, ²⁴¹Am and Pu radionuclides in soil samples. *J Radioanal Nucl*
206 *Chem* 226 (1-2):279-284
207
- 208 6. Mellado, J., M. Llauradó and G. Rauret (2001). Determination of Pu, Am, U, Th and Sr in marine
209 sediment by extraction chromatography. *Anal Chim Acta* 443 (1):81-90
210
- 211 7. Maxwell Iii, S. L. (2006). Rapid column extraction method for actinides and ^{89/90}Sr in water samples.
212 *J Radioanal Nucl Chem* 267 (3):537-543
213
- 214 8. Chung, K. H., S. D. Choi, G. S. Choi and M. J. Kang (2013). Design and performance of an automated
215 radionuclide separator: Its application on the determination of ⁹⁹Tc in groundwater. *Appl Radiat Isotopes*
216 81:57-61
217
- 218 9. Salonen, L. and H. Hukkanen (1997). Advantages of low-background liquid scintillation alpha-
219 spectrometry and pulse shape analysis in measuring ²²²Rn, uranium and ²²⁶Ra in groundwater samples.
220 *J Radioanal Nucl Chem* 226 (1-2):67-74
221
- 222 10. Chen, Q., H. Dahlgaard, S. P. Nielsen and A. Aarkrog (2002). ²⁴²Pu as tracer for simultaneous
223 determination of ²³⁷Np and ^{239,240}Pu in environmental samples. *J Radioanal Nucl Chem* 253 (3):451-
224 458
225
- 226 11. Maxwell, S. L., B. K. Culligan, J. B. Hutchison, R. C. Utsey and D. R. McAlister (2014). Rapid
227 determination of actinides in seawater samples. *J Radioanal Nucl Chem*:1-15
228
- 229 12. Sr resin Eichrom Technologies, http://www.eichrom.com/eichrom/products/info/sr_resin.aspx.
230 Accessed 5 Aug 2014
231
- 232 13. Currie, L. A. (1968). Limits for Qualitative Detection and Quantitative Determination: application to
233 radiochemistry. *Anal Chem* 40 (3):586-593
234
- 235 14. Lee, S. H., J. Gastaud, J. J. La Rosa, L. L. W. Kwong, P. P. Povinec, E. Wyse, L. K. Fifield, P. A.
236 Hausladen, L. M. Di Tada and G. M. Santos (2001). Analysis of plutonium isotopes in marine samples by
237 radiometric, ICP-MS and AMS techniques. *J Radioanal Nucl Chem* 248 (3):757-764
238
- 239 15. Nagaya, Y. and K. Nakamura (1981). Artificial radionuclides in the western Northwest Pacific (¹³⁷Cs and
240 ⁹⁰Sr) in the deep waters. *Journal of the Oceanographical Society of Japan* 37 (3):135-144
241
- 242 16. Casacuberta, N., P. Masqué, J. Garcia-Orellana, R. Garcia-Tenorio and K. O. Buesseler (2013). ⁹⁰Sr

243 and ^{89}Sr in seawater off Japan as a consequence of the Fukushima Dai-ichi nuclear accident.
244 *Biogeosciences* 10 (6):3649-3659
245
246 17. Günther, K., S. Lange and M. Veit (2009). A rapid method for determining ^{89}Sr and ^{90}Sr by
247 Cerenkov counting. *Appl Radiat Isotopes* 67 (5):781-785
248
249

Table 1. Removal of Sr in 1L of seawater with amount of Na₂CO₃

Na ₂ CO ₃	Removal of Sr ^a (%)	Removal of Ca ^b (%)	Removal of Mg ^b (%)
5g	55.0 ± 3.9	99.1	97.9
10g	84.9 ± 1.2	99.5	96.9
15g	92.3 ± 4.5	98.7	96.3

^a sample number = 3, "average ± 1 standard deviation"; ^b sample number = 1

Table 2. ⁹⁰Sr and ²³⁸Pu results in a set of seawater samples

Seawater	Recovery (%)	⁹⁰ Sr reference value (Bq) ^a	⁹⁰ Sr measured value (Bq)	Difference (%)	Recovery (%)	²³⁸ Pu reference value (Bq) ^b	²³⁸ Pu measured value (Bq)	Difference (%)
1L	94.9	94.3	90.3	-4.2	73.7	0.0117	0.0128	9.4
	85.4	94.3	89.2	-5.4	51.5	0.0117	0.0107	-8.5
	95.0	93.9	95.2	1.4	58.0	0.0117	0.0117	-0.2
	90.1	94.3	92.0	-2.4	72.8	0.0117	0.0125	6.8
	89.5	94.3	91.2	-3.3	73.5	0.0117	0.0107	-8.5
10L	82.5	94.1	93.2	-1.0	56.5	0.0117	0.0114	-2.6
	77.5	94.2	90.2	-4.2	51.8	0.0117	0.0112	-4.3
Avr.	87.8		91.6	-2.7	62.5		0.0116	-1.1
SD	6.5		2.1		10.4		0.0008	
Processing standard ^c (n=3)	92.1 ± 1.2				91.9 ± 0.6			

^a uncertainty 3.0 % (1 SD)

^b uncertainty 5.0 % (1 SD)

^c Processing standards are used to test the purification step (1 SD)

Figure 1. Schematic flow diagram of MARS (R1: 8 M HNO₃, R2: 1 M HNO₃, R3: 9 M HCl, R4: DIW, In the column section: solid line (1), connection of TEVA and Sr-resin column; dash line (2), connection only with TEVA column; dot line (3), connection only with Sr-resin column.

Figure 2. Behavior of (a) Sr and (b) Pu on the TEVA column (open triangle: Sr on TEVA 4 ml (BV); open circle: Pu on TEVA 6 ml (BV); closed circle: Pu on TEVA 4 ml (BV)).

Figure 3. Elution behavior of Sr on Sr resin (open circle: Sr resin 35 ml (BV); closed circle: Sr resin 4 ml (BV)).

Figure 4. Flow chart of simultaneous separation of Pu and Sr in seawater using TEVA and Sr resin columns.

Figure 1

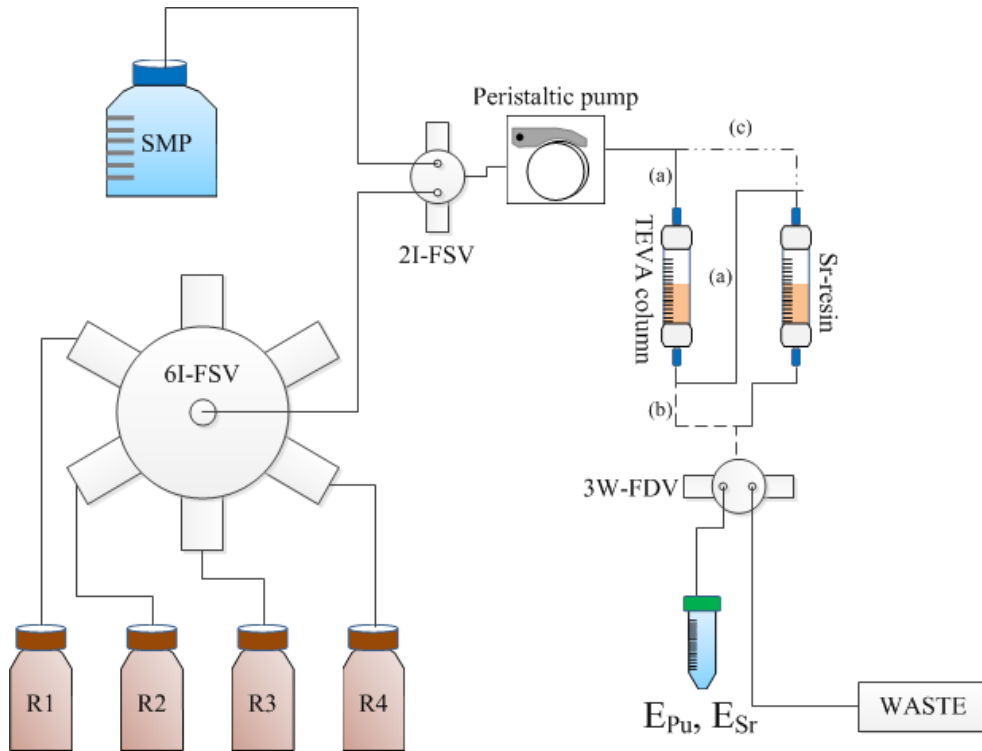


Figure 2

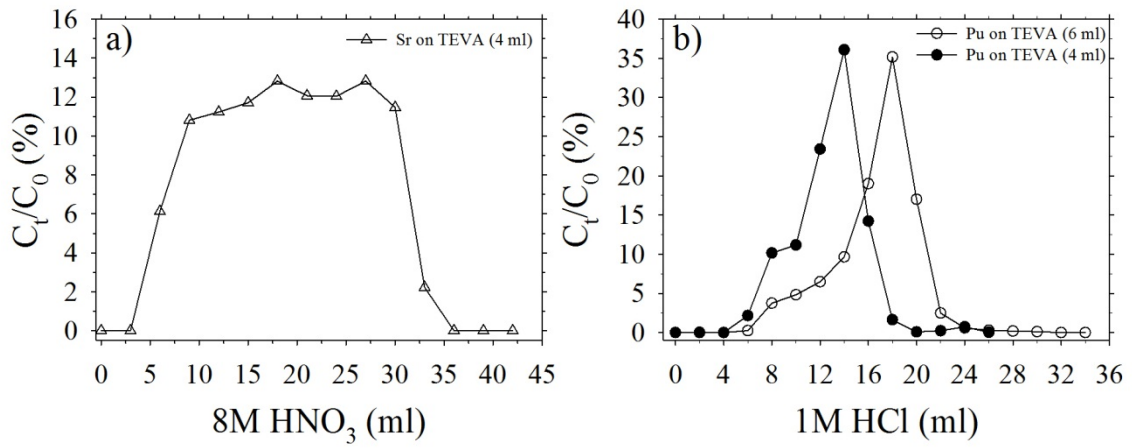


Figure 3

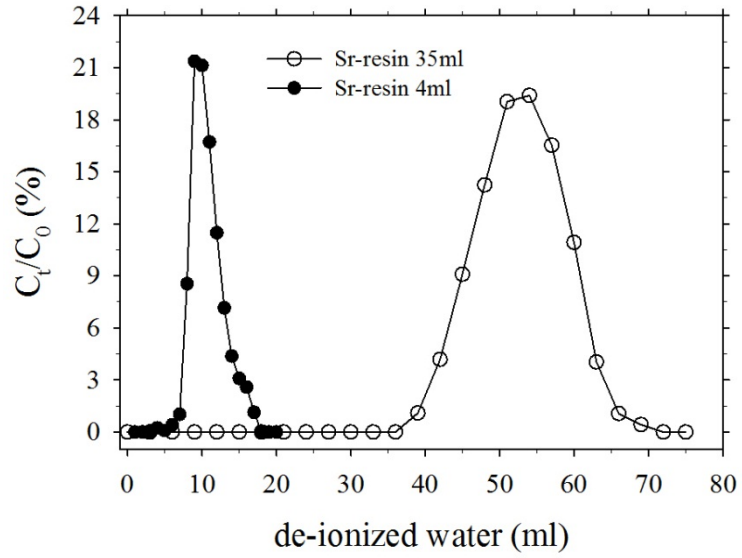


Figure 4

