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Sequential separation of the actinides in environmental and radioactive waste samples

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Abstract

This paper presents a rapid and quantitative sequential radiochemical separation method for the Pu, Am, U and Np nuclides in radioactive waste samples as well as environmental samples in a hydrochloric acid medium with an anion exchange resin. From the recovery test for the actinides in acid solutions, the Pu, Am, Np and U isotopes were more rapidly purified from the actinides in a hydrochloric acid solution with the anion exchange resin than in a nitric acid solution with the anion exchange resin and TRU resin. Sequential separation method of the Pu, Np, Am and U isotopes was applied to IAEA reference samples, where the activity concentrations of the ^{239,240}Pu, ²⁴¹Am and ²³⁸U in the IAEA-375 and IAEA-326 soils were close to the reference values reported by the IAEA.

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1. Introduction

During the last 50 years, a great amount of attention has been given to the development of accurate and reliable methods for a determination of the actinides in various environmental and biological samples. A determination of low level radionuclides such as the Pu, Am, U and Np isotopes requires lengthy and tedious chemical processes, which include techniques such as an ion exchange, liquid–liquid extraction, and a precipitation for a separation and preconcentration of the nuclides. In some cases, the amount of a sample material available for an analysis is very small such that a radiochemical method is necessary by which the Pu, Am, Np and U isotopes can be separated from one sample and determined in sequence. Also, in an emergency situation, a rapid and reliable analytical method for radionuclides is necessary.

Recently, several study groups have reported on procedures for a determination of the radionuclides in environmental samples with extraction chromatographic materials such as TRU Spec, TEVA Spec and Diphonix resins [1–4]. These methods

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0925-8388/\$ - see front matter © 2007 Elsevier B.V. All rights reserved. doi:10.1016/j.jallcom.2007.06.100 for determining the amount of low level radionuclides in soil or sediment samples, however, are limited because it is somewhat difficult to purify the radionuclides due to the major salt ions in a large the soil sample. Also, with anion exchange chromatography, low level activity concentrations of radionuclides such as the Pu, Am and Sr isotopes were sequentially determined [5]. However, these sequential chromatographic methods have only focused on the Pu and Am isotopes, without including the Np and U isotopes. Therefore, it is desirable to develop a combined procedure for the radionuclides to obtain a reliable activity concentration and a correlation of the nuclides in the same materials sample. Also, in order to reduce the analysis cost and turnaround time of the conventional methods, a simultaneous method to determine the activity concentration of the Pu, Am, U and Np isotopes in a radioactive waste as well as environmental samples has become increasingly important for a waste management and environmental monitoring.

The objective of this work is to develop a rapid and quantitative sequential radiochemical separation method of the Pu, Am, U and Np nuclides in environmental samples with a standard anion exchange resin method, after comparing the chemical recoveries of the radionuclides in a hydrochloric acid medium with those in a nitric acid medium. Also, the sequential separation method developed in this study for the Pu, Np, Am and U

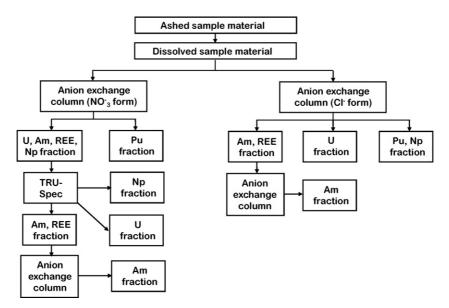


Fig. 1. Separation scheme of Pu, Np, Am and U in a nitric acid medium and hydrochloric acid medium.

isotopes was validated by an application to the IAEA-reference samples and radioactive waste samples.

2. Experimental

A flow chart of the conventional radiochemical procedure for the Pu, Np, Am and U isotopes in a nitric acid medium and hydrochloric acid medium is shown in Fig. 1.

2.1. Sample decomposition

A complete decomposition of a sample matrix such as the soil and sediment is important for a radiochemical analysis of environmental samples, because the destruction of a soil matrix makes it possible to enhance an isotopic exchange and convert the nuclides to an ionic form that can undergo chemical reactions. A total of 20 g of soil was weighed into a porcelain dish and ashed in a muffle furnace with a gradual heating program up to 600 °C to eliminate any organic matter. The liquid waste sample (10 mL) was evaporated to a dryness on a hotplate and the residue was dissolved in a hydrochloric acid solution. To compensate for the chemical recovery, 242 Pu (0.1 Bq), 243 Am (0.1–0.5 Bq) and 232 U (0.1 Bq), as yield tracers, were added into a sample.

In the dissolution method with nitric acid, the calcined soil samples were dissolved with 30 mL of 8 M HNO₃ with a stirring on a hot plate. In the HNO₃ + HF method, the samples were dissolved with 10 mL of concentrated HNO₃ and 10 mL of HF (48%) and evaporated to a dryness. Dissolution in HNO₃/HF was repeated and the sample was evaporated again to a dryness. The residue was dissolved with 30 mL of 8 M HNO₃ or 9 M HCl. Also, the fusion method with lithium metaborate has been applied to a soil for a decomposition of the soil matrix [6]. The sample solution was filtered with a 0.45 μ m membrane filter. In the 9 M HCl leaching solution, a small amount of concentrated HNO₃ was added to provide a matrix of 9 M HCl–0.1 M HNO₃.

2.2. Adjustment of oxidation states of Pu and Np

It is necessary to adjust the oxidation states of Pu and Np for a radiochemical analysis of Pu with an anion exchange column in a nitric or hydrochloric acid medium, because the distribution coefficients of Pu and Np on the anion exchange resin depend on the oxidation states of Pu and Np. In a solution, the oxidation states of Pu and Np are changeable due to slight differences in the oxidation–reduction potentials [7]. In the adjustment procedures of the oxidation states, for the leaching solution with an 8 M HNO₃ medium, the oxidation state of Pu was adjusted to a tetravalent state [8] and the oxidation state of Np was adjusted to a hexavalent state with 0.2 M NaNO₂ [9], after a heating at 80–90 °C for 0.5 h. Also, in the leaching solution with 9 M HCl–0.1 M HNO₃, the oxidation states of Pu and Np were adjusted to a tetravalent state with 0.2 M NaNO₂ [10].

2.3. Separation of Pu, Np and U in a nitric acid medium with an anion exchange resin and a TRU resin

The sample solution with an 8 M HNO₃ medium was passed through a pre-conditioned anion exchange resin (Bio-Rad, 100–200 Mesh) column (inner diameter, 10 mm; resin bead length, 120 mm) with 8 M HNO₃ at a rate of 0.5 mL/min. The column was then washed with 20 mL of 8 M HNO₃ to remove the Am, Np and U isotopes. The effluent was evaporated to a dryness and reserved for a subsequent separation of the Am, Np and U isotopes. Columns were washed with 20 mL of 9 M HCl to desorb the Th. Finally, Pu was eluted with 20 mL of 0.36 M HCl/0.01 M HF [11], which does not form a chloride complex on the anionic exchange resin.

The Am, Np and U residue that was separated from the Pu was dissolved in 30 mL of 2 M HNO₃. The content of Fe(III) was tested by adding one drop of 1 M NH₄SCN to the sample solution. If the test was positive (red color), about 200 mg of ascorbic acid was added to reduce the Fe(III) to Fe(II). TRU Spec columns (bed volume, 1.3 mL; length, 2.6 cm) were used to separate the Am, Np and U isotopes after a conditioning with 30 mL of 2 M HNO₃. Samples were loaded onto the column followed by a washing with 20 mL of 2 M HNO₃. The column was then washed with 4 mL of 9 M HCl and the Am fraction was eluted with 20 mL of 4 M HCl. The Np was eluted with 20 mL of 1 M HCl/0.03 M oxalic acid. Finally, the uranium was eluted with 20 mL of a 0.1 M ammonium oxalate solution.

2.4. Separation of Pu, Np and U in a hydrochloric acid medium with an anion exchange resin

The sample solution with a 9 M HCl–0.1 M HNO₃ medium was passed through a pre-conditioned anion exchange resin (Bio-Rad, 100–200 Mesh) column (inner diameter, 10 mm; resin bead length, 120 mm) with 9 M HCl–0.1 M HNO₃ at a rate of 0.5 mL/min. The column was then washed with 20 mL of 9 M HCl–0.1 M HNO₃. The effluent was evaporated to a dryness and reserved for a purification of the Am from the rare earth elements (REEs) with an anion exchange resin. Uranium was eluted on to the anion exchange column with 20 mL of 8 M HNO₃ and 20 mL of 4 M HNO₃. The eluted U fraction was evaporated to a dryness and held for an electrodeposition. The column was then washed with 10 mL of 9 M HCl to remove the Th, which was discarded. Finally,

the Pu and Np isotopes were eluted with 20 mL of 0.36 M HCl/0.01 M HF and held for an electrodeposition.

2.5. Separation of Am from the REEs

The Am fraction was evaporated to a dryness and converted to a nitrate with 65% HNO₃. The residue was dissolved in 10 mL of 1 M HNO₃–93% CH₃OH, and then, passed over a 12 cm³ column of an anion exchange resin (Dowex 1 × 4), which had previously been conditioned with 10 mL of 1 M HNO₃–93% CH₃OH. The column was washed with 20 mL of 1 M HNO₃–93% CH₃OH and 50 mL of 0.1 M HCl–0.5 M NH₄SCN–80% methanol to remove the REEs [5]. Finally, the Am isotopes were stripped with 40 mL of a 1.5 M HCl–86% methanol solution after a washing with 10 mL of 1 M HNO₃–93% CH₃OH.

2.6. Recovery test for Pu, Np, Am and U in a simulated solution

The Pu separation with an anion exchange resin was mainly carried out in a nitric acid medium because most of the handling processes of a nuclear fuel are in a nitric acid medium. However, a separation of the Pu isotopes in the environmental samples can be performed in a hydrochloric medium, since the distribution coefficients of Pu on the anion exchange resin in high hydrochloric acid concentrations are very high [12]. Therefore, it is necessary to compare the chemical recovery of the Pu isotopes with the anion exchange resin in the nitric acid and hydrochloric acid solutions. To check on the chemical recovery for the Pu, Np, Am and U isotopes at each separation step, ²⁴²Pu, ²³⁷Np, ²⁴¹Am and ²³²U (about 0.5 Bq) were added into the simulated acid (8 M HNO₃ and 9 M HCl–0.1 M HNO₃) solutions. The elution profiles for ²⁴²Pu, ²³⁷Np, ²⁴¹Am and ²³²U in the anion exchange resin are shown in Figs. 2 and 3. Eluting fractions were collected at 5 mL intervals. The chemical recoveries of ²³⁹Pu, ²³⁷Np, ²⁴¹Am and ²³²U were calculated with an alpha spectrometer.

2.7. Measurement of the Pu, Np, Am and U isotopes with an alpha spectrometer

The separated fractions of the Pu (Np), Am and U isotopes were evaporated to a dryness. Each residue was dissolved in 1 mL of concentrated HCl and evaporated to a dryness. The Pu (Np), Am and U isotopes were electroplated on to stainless steel platelets and measured by an alpha spectrometer [13]. The measured activities for ^{239,240}Pu, ²⁴¹ Am and ²³⁸U were corrected for a chemical yield by using the observed activities of ²⁴²Pu, ²⁴³ Am and ²³²U. Also, the chemical yield for ²³⁷Np eluted from the TRU resin was obtained by measuring the gamma activity of ²³⁹Np ($t_{1/2}$: 2.35 days; E_γ : 228 keV, 278 keV) on the electroplated disc. Typical chemical yields were in the range of 60–90%. At a counting time of 1000 min, the detection limits were 0.0009 Bq/sample, 0.0012 Bq/sample and 0.0010 Bq/sample for ^{239,240}Pu, ²⁴¹Am and ²³⁸U, respectively.

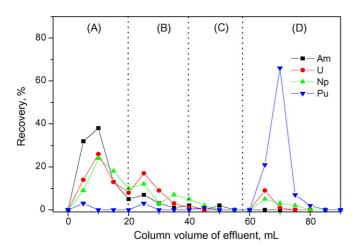


Fig. 2. Elution profile for Pu, Np, Am and U for an ion exchange resin in a HNO_3 medium: (A) 8.0 M HNO_3 , (B) 8.0 M HNO_3 , (C) 9.0 M HC1 and (D) 0.36 M HC1/0.01 M HF.

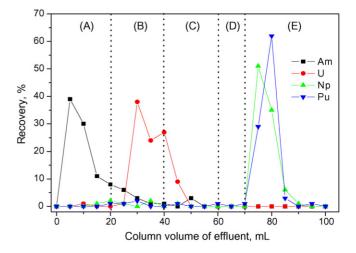


Fig. 3. Elution profile for Pu, Np, Am and U for an ion exchange resin in a HCl medium: (A) 9.0 M HCl, (B) 8.0 M HNO₃, (C) 4.0 M HNO₃, (D) 9.0 M HCl, and (E) 0.36 M HCl/0.01 M HF.

3. Results and discussion

3.1. Radiochemical separation of the Pu, Np, Am and U isotopes in a nitric acid medium

It is interesting to compare the separation of the Pu isotopes in a nitric acid solution with that in a hydrochloric acid solution. As shown in Fig. 2, a trace level of U was often detected in the final Pu fraction with the anion exchange method in a 8 M HNO₃ medium. This means that a small quantity of the uranium adsorbed on to the anion exchange column in the 8 M HNO₃ medium is eluted easily into the final Pu solution when a lower concentration of hydrochloric acid (0.36 M HCl/0.01 M HF) is used. In the loading solution (8 M HNO₃), the oxidation states of Pu and Np were adjusted to IV and VI with NaNO₂, respectively. It has been reported that Np(V) in a high nitric acid concentration (>3 M HNO₃) is oxidized to Np(VI) due to the NO_2^{-} [9,14], after a heating at 80–90 °C for 0.5 h. Under the same conditions Pu exists as Pu(IV). Because the distribution coefficient of Np(VI) on the anion exchange resin in a nitric acid medium is similar to U(VI) [15], most of the Np and U were passed into the eluting solution, while the Pu was strongly adsorbed on to the anion exchange resin.

With a TRU resin, the U and Np fractions were purified from the effluent solution passed through the anion exchange column, as shown in Fig. 1. For the determination of the Np and Am fractions, it is not necessary to add a yield tracer (²³⁹Np), because ²³⁹Np is generated by milking it from its precursor, ²⁴³Am. Therefore, if a sufficient amount of ²⁴³Am to generate ²³⁹Np, as a yield tracer, is used for determining ²⁴¹Am, it is possible to sequentially determine the activity concentrations of ²⁴¹Am and ²³⁷Np without adding a ²³⁹Np tracer. The Am fraction passed through the TRU resin was contained the REEs so that they were removed by anion exchange resin. A radiochemical separation of Pu, Np, Am and U isotopes in a nitric acid medium requires lengthy and tedious steps such as a preconcentration and coprecipitation due to using an anion exchange resin and TRU resin. Therefore, in an emergency situation, it is necessary to develop a rapid and reliable analytical method for determining the amount of Pu, Np, Am and U isotopes.

3.2. Radiochemical separation of the Pu, Np, Am and U isotopes in a hydrochloric acid medium

To overcome the demerits of the conventional Pu separation method in a nitric acid medium, Pu can be separated from other elements in a hydrochloric acid medium. A chloride-based anion exchange system has advantages in that the concentration of HCl and the temperature of the operation are not critical, and a complete loading can be obtained either with Pu in a tetravalent or the hexavalent state when compared with a nitrate-based system, even though a decontamination from certain impurities and fission products is lower and the solution is more corrosive to most metal equipment [12]. In this study, for purifying the Pu and Np isotopes, 9 M HCl-0.1 M HNO₃ medium was used to maintain the oxidation states of the Pu and Np isotopes at a tetravalent state instead of using only a 9 M HCl medium, because the Pu adsorbed on to the resin from the 9M HCl solution may be reduced to a trivalent state in the resin phase, and loosened and removed from the resin column as the elution with HCl proceeds [16].

In the 9 M HCl-0.1 M HNO₃ medium, Np(IV) was so strongly adsorbed on to the anion resin due to a high distribution coefficient on the anion resin, where most of the Np was detected in the eluting solution (0.36 M HCl/0.01 M HF). Therefore, as shown in Fig. 3, the total chemical recovery (96%) of the Np measured in the final eluting solution was similar to that of the Pu (98%), though it is a little difficult to distinguish between the energy spectrum of ²⁴²Pu and that of ²³⁷Np on the alpha spectrometer due to their similar emitting alpha energy $(^{242}Pu,$ 4.901 MeV; ²³⁷Np, 4.790 MeV). These similar chemical recoveries imply that ²⁴²Pu could be used as a tracer for compensating for a chemical recovery of Pu as well as Np during a simultaneous determination of Np and Pu in environmental samples [17]. Therefore, the chloride system can be used for analytical procedures to separate the various valence states of Pu, as well as to separate Np from transplutonium elements.

In the 9 M HCl-0.1 M HNO3 medium, the U fraction was separated easily from the other nuclides through the anion exchange resin with 8.0 M HNO₃ and 4.0 M HNO₃ without a step with the TRU resin, as shown in Fig. 3. Also, the Am isotopes were purified from other REEs with an anion-exchange procedure in acid-methanol as shown in Fig. 1. To save on the analytical time for the Am isotopes in the routine liquid samples, an anion-exchange procedure in an acid-methanol to remove the REEs can be omitted because the concentration of the REEs is very low in the liquid samples.

3.3. Application to environmental and radioactive waste samples

As presented in Table 1, the concentrations of ^{239,240}Pu in the IAEA-375 and IAEA-326 samples with different decomposition methods and loading solutions were consistent with the reference values reported by the IAEA. The activity concentrations of ^{239,240}Pu in the radioactive waste solution adjusted with 9 M HCl-0.1 M HNO3 were lower than those of ²³⁸Pu. Also, the chemical recoveries of the Pu isotopes for the IAEA reference samples and the radioactive waste solution were measured with high values.

It is interesting to compare the chemical recovery of the Pu isotopes with various decomposition methods in a soil sample. As shown in Table 1, there was no significant difference in the chemical recoveries for the Pu isotopes between the nitric acid medium (8 M HNO₃) and the hydrochloric acid medium (9 M HCl-0.1 M HNO₃). Also, the chemical recoveries of the Pu isotopes with the acid leaching methods (HNO₃, HNO₃ + HF) were similar to those with the fusion method. However, the fusion method is time consuming, requires expensive platinum dishes and is relatively dangerous, especially for an inexperienced analyst. Therefore, the acid leaching method with nitric or hydrochloric acid is an excellent decomposition method for routine environmental samples. With only a strong mineral acid

Table 1

Activity concentrations and chemical recoveries of the Pu and Np isotopes in the IAEA reference soils and liquid waste samples

Sample	Decomposition method	Loading solution	Activity concentration (Bq/kg or Bq/mL)			Chemical recovery (%)
			^{239,240} Pu	²³⁸ Pu	²³⁷ Np	
IAEA-375 ^a	Acid (HNO ₃)	8 M HNO ₃	0.26 ± 0.04	<mda<sup>b</mda<sup>	_	67 ± 4
	Acid (HNO ₃ + HF)	8 M HNO ₃	0.33 ± 0.04	<mda< td=""><td>_</td><td>70 ± 6</td></mda<>	_	70 ± 6
	Acid (HNO ₃ + HF)	9 M HCl/0.1 M HNO3	0.30 ± 0.04	0.061 ± 0.007	_	73 ± 5
	Fusion	9 M HCl/0.1 M HNO3	0.29 ± 0.04	<mda< td=""><td>_</td><td>75 ± 4</td></mda<>	_	75 ± 4
IAEA-326 ^c	Acid (HNO ₃)	8 M HNO3	0.47 ± 0.05	<mda< td=""><td>_</td><td>72 ± 5</td></mda<>	_	72 ± 5
	Acid (HNO ₃ + HF)	8 M HNO3	0.50 ± 0.06	<mda< td=""><td>-</td><td>76 ± 8</td></mda<>	-	76 ± 8
	Acid (HNO ₃ + HF)	9 M HCl/0.1 M HNO3	0.52 ± 0.05	<mda< td=""><td>_</td><td>73 ± 5</td></mda<>	_	73 ± 5
	Fusion	9 M HCl/0.1 M HNO3	0.50 ± 0.04	<mda< td=""><td>_</td><td>75 ± 6</td></mda<>	_	75 ± 6
RWS1	Acid (HCl)	9 M HCl/0.1 M HNO3	0.44 ± 0.04	1.02 ± 0.08	_	76 ± 5
RWS2	Acid (HCl)	9 M HCl/0.1 M HNO3	0.05 ± 0.01	0.15 ± 0.01	-	88 ± 5
RWS3	Acid (HCl)	9 M HCl/0.1 M HNO3	0.19 ± 0.02	0.28 ± 0.03	-	83 ± 7

^a Reference value: ^{239,240}Pu, 0.30 Bq/kg; ²³⁸Pu, 0.071 Bq/kg.

^b MDA: 0.0043 Bq/kg.
^c Reference value: ^{239,240}Pu, 0.50 Bq/kg; ²³⁸Pu, 0.019 Bq/kg.

Table 2

RWS1

RWS2

RWS3

Activity concentrations and chemical recoveries of the Am and U isotopes in the IAEA reference soils and liquid waste samples									
Sample	Decomposition method	Loading solution	²⁴¹ Am		²³⁸ U	²³⁸ U			
			Concentration (Bq/kg, Bq/mL)	Chemical recovery (%)	Concentration (Bq/kg, Bq/mL)	Chemical recovery (%)			
IAEA-375 ^a	Acid (HNO ₃)	8 M HNO3	0.15 ± 0.03	72 ± 6	18.02 ± 1.94	72 ± 4			
	Acid (HNO ₃ + HF)	8 M HNO3	0.14 ± 0.04	68 ± 7	27.53 ± 2.30	70 ± 6			
	Acid (HNO ₃ + HF)	9 M HCl/0.1 M HNO3	0.10 ± 0.04	75 ± 7	23.87 ± 4.07	74 ± 5			
	Fusion	9 M HCl/0.1 M HNO ₃	0.11 ± 0.05	78 ± 5	28.06 ± 3.56	76 ± 7			
IAEA-326 ^b	Acid (HNO ₃)	8 M HNO3	0.20 ± 0.04	69 ± 6	25.22 ± 2.14	71 ± 6			
	Acid (HNO ₃ +HF)	8 M HNO3	0.15 ± 0.02	72 ± 8	29.85 ± 3.17	68 ± 4			
	Acid (HNO ₃ + HF)	9 M HCl/0.1 M HNO3	0.19 ± 0.03	72 ± 5	27.78 ± 2.59	74 ± 6			
	Fusion	9 M HCl/0.1 M HNO3	0.21 ± 0.03	75 ± 6	30.71 ± 3.24	72 ± 5			

 0.67 ± 0.07

 0.08 ± 0.02

 0.29 ± 0.05

^a Reference value: ²⁴¹Am, 0.13 Bq/kg; ²³⁸U, 24.4 Bq/kg.

^b Reference value: ²⁴¹Am, 0.19 Bq/kg; ²³⁸U, 29.4 Bq/kg.

Acid (HCl)

Acid (HCl)

Acid (HCl)

(HCl, HNO₃), the actinides are effectively extracted from a soil component into an acidic solution for the global fallout environmental samples, because they may be associated with the soil via a surface phenomena such a surface complexation or surface precipitation [18]. Also, the acid leaching methods have a merit in that the leaching solutions could be loaded directly onto the anion exchange column with no further adjustment to their acid strength or additional preconcentration stages. But, for the refractory substances, so called hot particles, it is necessary to use LiBO₄, HNO₃/HF or HCl/HF for a complete extraction of the highly refractory Pu oxides from a sample matrix, since it is not enough to extract the refractory Pu oxides from a sample matrix with only the acid leaching method.

9 M HCl/0.1 M HNO3

9 M HCl/0.1 M HNO3

9 M HCl/0.1 M HNO3

As presented in Table 1, there was no ²³⁷Np detected in the IAEA reference soil and radioactive waste samples. For detecting the level of ²³⁷Np in environmental or radioactive samples, mass analytical techniques with TIMS or ICP-MS are preferred due to its long half-life $(2.14 \times 10^6 \text{ years})$ rather than a radiation measurement.

The concentrations of ²⁴¹Am and ²³⁸U in the IAEA-375 and IAEA-326 samples were within the confidence interval, though in the acid leaching method, the concentrations of the ²³⁸U were a little lower than the other decomposition methods due to an incomplete destruction of the soil matrix, as presented in Table 2. Also, the chemical recoveries for the Am and U isotopes with the anion exchange resin in the 9.0 M HCl-0.1 M HNO3 medium were higher than those in the 8.0 M HNO₃ medium, because in the hydrochloric medium, it is not necessary to use TRU resin to purify the Am and U isotopes from the hindrance nuclides or elements. In the nitric acid medium, a lot of Fe which leached from the soil hindered the adsorption of the Am and U isotopes onto the TRU resin, even when an ascorbic acid was added to the solution to reduce the amount of Fe(III). Therefore, it is necessary to remove the Fe with an oxalic coprecipitation before loading the solution containing the Am and U isotopes onto the TRU resin [5]. The extra separation steps such as an oxalic coprecipitation and TRU resin method in the nitric acid medium may cause a lower chemical recovery of Am and U than in the hydrochloric acid medium.

4. Conclusions

 73 ± 5

 80 ± 2

 78 ± 6

From the recovery tests for the Pu, Np, Am and U isotopes in a simulated solution, the Am, Np and U isotopes were sequentially separated from the simulated solution in the nitric acid medium with expensive extraction chromatography materials (TRU resin), after separating the Pu isotopes with an anion exchange resin. While, with the anion exchange resin, the Pu (Np), U and Am isotopes were simply separated from the simulated solution in the hydrochloric medium.

From the results of an application of the sequential separation method for the Pu, Np, Am and U isotopes to IAEA reference samples, the activity concentrations of the ^{239,240}Pu, ²⁴¹Am and ²³⁸U in the IAEA-375 and IAEA-326 soils were close to the reference values reported by the IAEA. Also, the activity concentrations of the ^{239,240}Pu and ²⁴¹Am in the liquid waste samples adjusted with the hydrochloric acid medium were measured with high chemical yield. There was no significant difference in the chemical recoveries for the Pu isotopes between the nitric acid medium and the hydrochloric acid medium, while the chemical recoveries for the Am and U isotopes with the anion exchange resin in the hydrochloric acid medium were higher than those with the TRU resin in the nitric acid medium. The activity concentration of ²³⁷Np was not detected in the IAEA reference soil and radioactive waste samples.

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