analytical chemistry

Detection of Fuel Release in a Nuclear Accident: A Method for Preconcentration and Isolation of Reactor-Borne [239]Np Using Ion-Specific Extraction Chromatography

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ABSTRACT: Although actinides are the most informative elements with respect to the nature of a nuclear accident, plutonium analysis is complicated by the background created by fallout from atmospheric nuclear explosions. Therefore, we propose [239]Np, a short-lived actinide that emits several γ rays, as a preferred proxy. The aim of this study was to screen ion specific extraction chromatography resins (RE-, TEVA-, UTEVA-, TRU-, and Actinide-Resin) for the highest possible recovery and separation of trace amounts of [239]Np from samples with large activities of fission products such as radiocesium, radioiodine, and, most importantly, radiotellurium, the latter of which causes spectral interference in gamma spectrometry through overlapping peaks with [239]Np. The investigated environmental media for these separations were aqueous solutions simulating rainwater and soil. Spiked samples containing [239]Np and the aforementioned volatile radionuclides were separated through extraction chromatographic columns to ascertain the most effective means of separating [239]Np from other fission products for detection by gamma spectroscopy. We propose a method for nuclear accident preparedness based on the use of Eichrom's RE-Resin. The proposed method was found most effective for isolating [239]Np from interfering radionuclides in both aqueous solution and soil using 8 M HNO_3 as the loading solution and H_2O as the eluent. The RE-Resin outperforms the more commonly used TEVA-Resin because the TEVA-Resin showed a higher affinity for interfering radiotellurium and radioiodine.
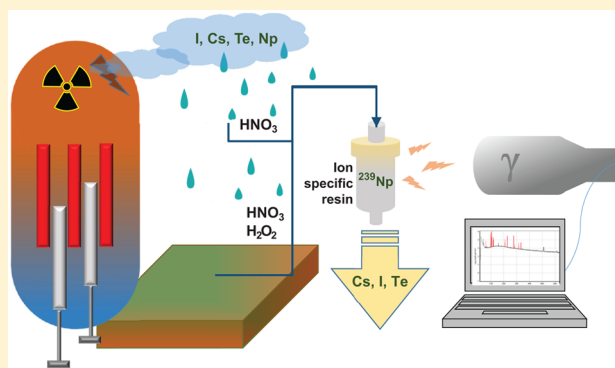
The nuclear accidents of Chernobyl and Fukushima caused the release of large amounts of radionuclides into the environment. Although both accidents are hardly comparable with respect to their cause and duration as well as reactor design, safety installations, physical state of the core during and after the accident, etc., they had in common that the main activities of their radioactive emissions were due to radionuclides of volatile elements such as Kr, Xe, I, Cs, and Te.[1] The presence in the environment of these radionuclides alone, therefore, does not provide much information on the nature of a nuclear accident, including the physical state of the core, reactor vessel, containment, or release mechanisms. Presence of nonvolatile elements such as actinides, however, provides much deeper insight into the nature of an accident, as for example, actinide emissions (primarily plutonium) from Chernobyl were significant,[2−5] thus indicating a massive thermal destruction of the core and the release of fuel particles. Emissions of actinides from Fukushima, in contrast, were relatively low,[6−14] thus indicating a very different type of accident.

After a nuclear accident, actinide analysis is therefore essential to understand the nature of the accident and the processes inside the reactor. Actinide analysis, however, is complicated by several facts. First, most actinides exhibit low specific activities due to their relatively long half-lives (e.g., [239]Pu, $T_{1/2}$ = 24 110 years; [240]Pu, $T_{1/2}$ = 6563 years), thus making radiometric detection of traces more difficult. Second, only a few relevant actinide nuclides are suitable γ-emitters. Detection by γ-spectrometry is the fastest and most straightforward radiation detection method, especially for nondestructive assays, as it requires only minimal sample preparation. For other analytical techniques such as mass spectrometry (MS) or α-spectrometry, the sample has to be treated chemically and thermally prior to separation and measurement, which sometimes introduces analytical pitfalls.[15] Third, there is a significant Pu background in the environment, caused by the releases of 8 tons of Pu (corresponding to an activity of 16 PBq)[16] in the course of the atmospheric nuclear explosions of the 20th century. Using the [240]Pu/[239]Pu ratio as an isotopic signature, it is possible to distinguish between the weapons fallout ([240]Pu/[239]Pu ∼0.18) and plutonium releases from a nuclear reactor ([240]Pu/[239]Pu ∼0.4−0.6).[17,18] When α-spectrometry is applied for the determination of Pu, the

discrimination of the reactor plutonium and weapons fallout plutonium via their characteristic isotopic ratio $^{240}Pu/^{239}Pu$ is not possible, because the α-particle energies of both nuclides cannot be resolved from each other. Plutonium analysis, therefore, usually relies on the application of MS, such as accelerator MS (AMS) or sector field inductively coupled plasma MS (SF-ICP-MS) and requires a rather laborious and time-consuming sample preparation, when compared with rapid γ-spectrometry.

To overcome this problem, ^{239}Np may be a suitable proxy for Pu in the initial assessment of the nature of a nuclear accident and may indicate fuel release into the environment. Neptunium-239 is produced by neutron capture of ^{238}U in the nuclear fuel and subsequent β^--decay of the resulting ^{239}U. Neptunium-239 is a short-lived ($T_{1/2}$ = 2.356 days) γ-emitting actinide, which makes it an ideal nuclide for rapid and γ-spectrometric analysis. Because of the short half-life, there is no ^{239}Np background from global fallout in the environment, which is another important advantage. Finally, both elements Np and Pu are characterized by a similar volatility and thus ^{239}Np may act as an indicator of fuel particle releases.

Analysis of ^{239}Np, however, faces one significant drawback in such a scenario: Its main γ-peak (intensity 26.3% at 106.1 keV) coincides with the γ-peak of the fission product ^{129m}Te (intensity 0.14% at 105.5 keV, $T_{1/2}$ = 33.6 days), which is usually emitted from major nuclear accident scenarios in great excess. This spectral interference has also been addressed in environmental samples after the Fukushima accident.[19] Although not as pronounced, there is also some spectral interference of the neptunium's second-most intense γ-peak at 277.6 keV (intensity 14.4%) with a γ-photon (284.3 keV with an intensity of 6.1%) emitted by the prominent fission product ^{131}I ($T_{1/2}$ = 8.03 days).

This spectral interference calls for the separation of Np from fission products in environmental samples taken after a nuclear accident. In this study, we have screened several Np specific extraction chromatographic resins for the applicability of the attempted Np/fission product separation. Although much work has been done on the separation, isolation, and analysis of elements within the group of the actinides,[20−28] separation of Np from fission products such as I, Te, and Cs by using ion specific extraction chromatography resins has not been investigated nearly as thoroughly, as they are not typically regarded as relevant interferences for actinide analysis with mass spectrometric methods and α-spectrometry.

The objective of this work is to establish a robust protocol for the rapid detection of ^{239}Np within aqueous and soil environmental matrices. The key of this study, however, is not only recovery of Np but also simultaneous suppression/removal of fission products. This scenario was accomplished through addition of commonly released volatile radionuclides to aqueous samples (simulating contaminated rainwater) and homogenized soil samples containing relatively minute, but detectable, quantities of ^{239}Np.

■ MATERIALS AND METHODS

Materials and Radionuclides. All chemicals used in this study were of analytical grade or higher purity. Nitric acid and hydrochloric acid were provided by Fisher Scientific, and hydrogen peroxide was purchased from Sigma-Aldrich. Radionuclides mimicking the typical mix of fission and activation products were generated by sealing into separate Suprasil quartz glass vials $CsNO_3$ (50 mg of Cs; Strem Chemicals, Newburyport, MA), $UO_2(NO_3)_2 \cdot 6H_2O$ (20 mg of depleted U; J.T. Baker Chemical Co., Phillipsburg, NJ), and $Te(OH)_6 \cdot 2H_2O$ (30 mg of Te; Strem Chemicals, Newburyport, MA) and irradiating the vials with neutrons at the USGS TRIGA Reactor in Denver, Colorado. Peak thermal neutron flux densities were 4.3×10^{12} cm^{-2} s^{-1} for $Te(OH)_6 \cdot 2H_2O$ (irradiated for 12 h and cooled for 5 days) and 1.4×10^{11} cm^{-2} s^{-1} for $CsNO_3$ and $UO_2(NO_3)_2 \cdot 6H_2O$ (irradiated for 2 h, cooled for 48 h). Neutron activation yielded the radionuclides of interest for this study, in particular ^{131}I, ^{123m}Te ($T_{1/2}$ = 119 days), ^{134}Cs ($T_{1/2}$ = 2.1 years), and ^{239}Np. Following irradiation, the $CsNO_3$ sample was washed from its quartz vial using 0.5 M HNO_3 to create a 50 mL stock solution with a concentration of 1 mg Cs mL^{-1} (7.5 mM Cs; with an initial activity concentration (A_0) of 1.32 ± 0.10 kBq mL^{-1} ^{134}Cs); the $Te(OH)_6 \cdot 2H_2O$ and $UO_2(NO_3)_2 \cdot 6H_2O$ were washed from their vials with deionized H_2O to create 30 and 20 mL stock solutions with concentrations of 1 mg Te mL^{-1} (7.8 mM Te; A_0 = 0.93 ± 0.12 kBq mL^{-1} ^{123m}Te) and 1 mg U mL^{-1} (4.2 mM U; A_0 = 12.44 ± 0.62 kBq mL^{-1} ^{239}Np), respectively. The activity concentration of ^{131}I in the Te solution, after decay of the primary activation product ^{131}Te, was 25.76 ± 0.94 kBq mL^{-1}. The radionuclides were applied in quantities that would not only mimic the characteristic releases in the course of a nuclear accident (activities of $^{131}I > ^{129m}Te \approx ^{134+137}Cs > ^{239}Np$) but also make the small amounts of ^{239}Np undetectable in the high background by direct γ-spectrometric measurement of the mixture, hence making the need for separation and isolation of ^{239}Np self-evident.

The chemistry of Np is highly influenced by its valency;[29] it is typically present in the tetravalent or pentavalent form. The resin columns used in this study (Actinide-, RE-, TEVA-, TRU-, and RE-Resins) are more selective for Np(IV) than Np(V), thereby requiring a reducing agent to reduce any Np(V) present in the uranium solution to Np(IV).[30] The presence of Fe(II) and ascorbic acid effectively reduce Np(V). A 5 mg Fe(II) mL^{-1} (89.5 mM) solution was freshly prepared for every experimental series with 0.025 g of $FeSO_4 \cdot 7H_2O$ (Mallinckrodt, MO) in 10 mL of H_2O. A 1.5 M ascorbic acid (Macron Chemicals, PA) solution was prepared by carefully heating and dissolving 13.2 g of ascorbic acid in 50 mL of H_2O.

Ion Specific Resins and Technology. Five ion specific resins were purchased from Eichrom Technologies, LLC, and evaluated for their effectiveness in separating neptunium from volatile radionuclides released during nuclear events. The Actinide-Resin (ACT), RE-Resin, TEVA-Resin, TRU-Resin, and UTEVA-Resin have been screened for their applicability of the purpose of this study. All resins were purchased in cartridge form (2 mL) with 50−100 μm particle size. A vacuum box system was used to enhance the flow of samples through the Eichrom extraction chromatography columns. The system was set up in accordance with section 7.2.2 of ref 31. Pressures within the vacuum box were maintained below 17 kPa when vacuum was applied. Prior to loading the sample solution, resins were conditioned with 5 × 1 mL (1 mL 5 times) of each respective conditioning solution to ensure homogeneity of solutions within the columns. After loading, the columns were rinsed to remove foreign substances and radionuclides. Finally the analytes were eluted from the resin columns (see below).

Rainwater Analogue Preparation. Eichrom extraction chromatography columns have affinities for actinides that vary by acid type and concentration. Preconditioning, loading,

rinsing, and eluting conditions for the rainwater analogues and soil are summarized in Table 1.

Table 1. Preconditioning/Loading/Rinsing and Eluting Conditions of the Ion Specific Resins Used for Recovery of ^{239}Np from Aqueous Solution (Rainwater Analogue) and Soil

resin	replicates	conditioning/loading/rinsing	eluting
Rainwater Analogues			
ACT	4	0.1 M HCl	10 M HCl
RE	4	8 M HNO$_3$	0.01 M HNO$_3$
RE	2	8 M HNO$_3$	H$_2$O
TRU	4	3 M HNO$_3$	0.1 M HNO$_3$
TRU	2	3 M HNO$_3$	H$_2$O
UTEVA	4	10 M HCl	0.1 M HCl
UTEVA	2	8 M HNO$_3$	0.1 M HNO$_3$
UTEVA	2	10 M HCl	H$_2$O
TEVA	3	2 M HNO$_3$	0.01 M HNO$_3$
TEVA	3	10 M HCl	0.1 M HCl
TEVA	3	8 M HNO$_3$	H$_2$O
Soil Matrix			
RE	2	8 M HNO$_3$	H$_2$O
UTEVA	2	8 M HNO$_3$	H$_2$O
TEVA	2	8 M HNO$_3$	H$_2$O

Natural rainwater exhibits considerable variability with respect to chemical composition and pH. It is influenced by weather phenomena,[32] urbanization,[33] air mass sources,[34] and other factors. For the preparation of rainwater analogues, in-house deionized water (0.014 mg L^{-1} Ca^{2+}, 0.001 mg L^{-1} Cl$^-$, and each 0.0001 mg L^{-1} Mg^{2+}, Na$^+$, and K$^+$) was used, which is within 1 or 2 orders of magnitude of the composition of rainwater reported in literature.[32] The pH of the deionized water (5.0) is within the typical range of rainwater.[32] Rainwater analogues were contaminated and made ready for loading onto the resin columns by adding, in the following order: 1.25 mL of 1.5 M ascorbic acid, 0.1 mL of 89.5 mM Fe(II) solution, 20 μL of 4.2 mM neutron-activated U, 20 mL of conditioning solution, 1 mL of 7.8 mM neutron-activated Te, and 2 mL of 7.5 mM neutron-activated Cs. Conditioning solutions were prepared from deionized water and concentrated HCl and HNO$_3$ stock solutions. The final concentrations of the solutes in 24.37 mL of solution were 76.9 mM ascorbic acid, 0.4 mM Fe(II), 3.4 μM U, 0.3 mM Te, and 0.6 mM Cs, the latter three being radionuclide carriers. These solutions were loaded onto their respective resin columns. The columns were then rinsed with 10 × 1 mL of conditioning solution. Additional conditioning solution was added to bring the final volume up to 35 mL, which corresponds to the γ-spectrometry calibration geometry used. Eluents were pulled through the resin columns by vacuum into separate centrifuge tubes for collection. Eluents were applied by 10 × 1 mL. Final collected volumes were brought up to 35 mL with additional eluent (see Table 1) to ensure identical filling levels of the vial, which is important for γ-spectrometry.

Soil Sample Preparation. Soil from the Colorado State University campus was homogenized in a ceramic mortar to a particle size of 3 μm. The soil has a clay loam texture (28% sand, 33% silt, and 39% clay), a very high lime estimate, and a rather low organic fraction (1.5%). The 6-fold replicates of soil samples (10.0 g aliquots from the homogenized stock) were spiked with the above-mentioned stock solutions (20 μL of 4.2

mM U (^{239}Np), 2 mL of 7.5 mM Cs, 1 mL of 7.8 mM Te) and then promptly leached by boiling under reflux with a mixture of 1 mL of 30% H$_2$O$_2$, 4 mL of concentrated HNO$_3$ (16 M), and 4 mL of 8 M HNO$_3$ for 30 min. The samples were filtered through a Carl Schleicher & Schuell Co. paper filter (no. 576) using a vacuum flask and Büchner funnel. Ascorbic acid (1.25 mL, 1.5 M) and Fe(II) (0.1 mL, 89.5 mM) were added to the filtrates before loading onto the RE-, TEVA-, and UTEVA-Resin columns in duplicate (see Table 1). The columns were conditioned with 8 M HNO$_3$. After loading the soil extracts, the columns were rinsed with 10 × 1 mL of 8 M HNO$_3$. The eluent used was 10 × 1 mL of H$_2$O. Final eluted volumes were brought up to 35 mL for analysis by γ-spectrometry.

In preliminary trials for extraction of Np from soil, two ion specific resins (RE- and UTEVA-Resin) were used. In this case, soils were spiked with ^{239}Np (20 μL, 4.2 mM U) only and were boiled under reflux with 8 M HNO$_3$ in the absence of 30% H$_2$O$_2$.

Sample Analysis. Final activities were determined by placing the samples on top of an ORTEC 364 cm^3 HPGe detector with a 0.76 mm Be window (2.32 keV resolution at the 1332 keV ^{60}Co peak; 87.4% relative efficiency). Activity yields were decay-corrected to a reference time for accurate determination of the true radionuclide recovery. Samples were measured for 600 s live time in identical geometries. The Peak Locate algorithm used the Unidentified Second Differential along channels 1−4096 with a significance threshold of 3.00 and a tolerance of 1.00 keV. The Peak Area algorithm used the Sum/Nonlinear LSQ Fit with a fixed tail parameter and 4 channel continuum. For ROI (region of interest) Limits Determination, the maximum number of FWHMs (full width at half-maximum) was 5.00 between peaks, 2.00 for the left limit, and 2.00 for the right limit.

RESULTS AND DISCUSSION

Separation from Aqueous Matrixes. The recovery of ^{239}Np from aqueous solution and simultaneous suppression of fission products are shown in Figure 1 (standard deviation shown to 1σ). The most prominent finding that will affect the selection of conditioning/loading/rinsing and eluting solutions is not only recovery of Np but also the behavior of radiotellurium in the resin columns because ^{129m}Te will exhibit
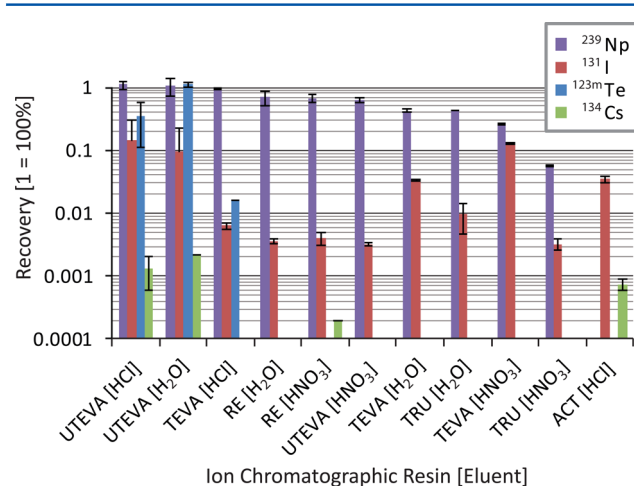
Figure 1. Recovery of ^{239}Np and associated volatile radionuclides from aqueous matrixes; uncertainties are expressed to 1σ.

the leading spectral interference with ^{239}Np in an accident scenario. Evaluation of ^{123m}Te (intensity of the 159 keV γ-photon is 84%) content in the eluates, resins, and loading solutions was conducted approximately 90 days following ^{239}Np isolation, after complete decay of ^{131}I and ^{239}Np.

There was no detectable activity of ^{239}Np eluted from the four replicates of the Actinide-Resin columns. According to the manufacturer, the affinity for Np on the Actinide-Resin decreases with increasing acid concentration. However, using 10 M HCl was not effective at eluting detectable quantities of ^{239}Np, resulting in effectively all of the Np being retained on the resin column (as confirmed by γ-spectrometry).

Neptunium could not be eluted in detectable amounts from two of the four replicates of the TRU-Resin columns with 0.1 M HNO$_3$. The other two replicates of the TRU-Resins using 0.1 M HNO$_3$ as eluent yielded 5.7 ± 0.2% of the original ^{239}Np activity in the eluate (calculated using the 278 keV peak yield). Similarly, only one of the two replicates of the TRU-Resins using H$_2$O as eluent yielded detectable amounts of ^{239}Np (4.2% of the initially spiked amount). In those samples where ^{239}Np was not detected, visual analysis of the spectra suggested the presence of traces of ^{239}Np both on the column and in the eluate. However, there was too much spectral interference from prominent amounts of residual ^{131}I on the column as well as in the eluate for the ^{239}Np peaks to be discerned by the peak-locating algorithm we used.

The RE-, TEVA-, and UTEVA-Resins exhibited the best and most consistent yield for neptunium recovery. The RE-Resins performed well in recovering ^{239}Np (yields 69.2 ± 19% for H$_2$O as eluent; 67.3 ± 11% for 0.01 M HNO$_3$ as eluent). At the same time, only 0.4 ± 0.02% ^{131}I and 0.4 ± 0.1% ^{131}I have been coeluted with the ^{239}Np when H$_2$O and 0.01 M HNO$_3$ were used as eluents, respectively. Suppression of ^{123m}Te (not detectable) and ^{134}Cs worked well with RE-Resin (only minute traces of ^{134}Cs were detected in the 0.01 M HNO$_3$ eluate of one trial: 0.02%).

The TEVA-Resin is the most commonly used resin for Np separation and isolation in the literature.[21,22] Indeed the use of TEVA-Resin with 0.1 M HCl as eluent recovered 95.6 ± 2.7% of the initial ^{239}Np activity. However, in this experiment, the TEVA-Resin also yielded considerable amounts of ^{123m}Te (1.6 ± 0.01%) and also some ^{131}I (0.63 ± 0.07%). The use of H$_2$O or 0.01 M HNO$_3$ as eluents decreased not only the Np yield (42.1 ± 2.7% and 25.9 ± 0.001%, respectively) considerably but also diminished the separation efficiency from ^{131}I (3.3 ± 0.1% and 13.0 ± 0.4% of ^{131}I have been recovered in these trials, respectively). Both series, however, did not yield detectable ^{123m}Te in the eluate.

The manufacturer's UTEVA-Resin protocol requires elution with 0.1 M HCl. However, different eluents were also tested to seek higher yields of Np and better suppression of interfering fission products. The UTEVA-Resin seemed to have recovered all of the spiked ^{239}Np when 0.1 M HCl and H$_2$O are used for elution. Unfortunately, these trials also recovered 34 ± 24% and 100% of the ^{123m}Te, respectively, and also showed poor performance while suppressing ^{131}I (14.2 ± 15% and 9.6 ± 13%, respectively, have been recovered). Also traces of ^{134}Cs have been eluted when 0.1 M HCl and H$_2$O are used as eluates. Only little ^{131}I (0.33 ± 0.02%) and no detectable ^{134}Cs and ^{123m}Te have been found in the eluates when 8 M HNO$_3$ is used for conditioning/loading/rinsing and 0.1 M HNO$_3$ is used for elution. Unfortunately, however, the recovery of ^{239}Np was significantly diminished in these tests (62.8 ± 0.05%).

Separation and recovery of Np has been well investigated for these resins; however, retention of possible interfering fission products, especially radioiodine or radiotellurium, has not been studied or reported nearly as systematically. Radiocesium generally showed the lowest affinity to the resins scrutinized in this study. Neptunium could be separated from more than 99.9% of the radiocesium present in the samples. In the light of the very purpose of this study, utmost exclusion of fission products (especially radiotellurium) together with a good recovery of Np, RE-Resin performed best for the rainwater analogue scenario, as it recovers a worthwhile amount of ^{239}Np (69%) but mostly excluded ^{131}I and ^{123m}Te from the eluate.

Separation from Soil Matrixes. The RE-, TEVA-, and UTEVA-Resins were evaluated for their ability to isolate neptunium from the volatile fission products in a soil matrix. Figure 2 shows that RE- (64.8 ± 4.2%) and TEVA-Resins (53.6
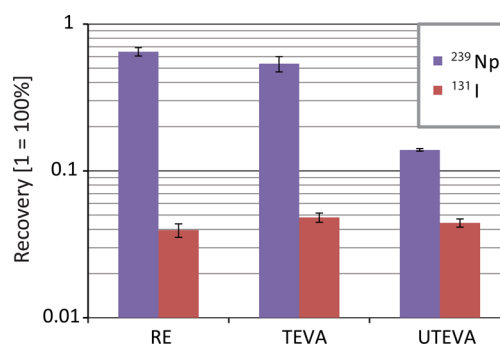
Figure 2. Recovery of ^{239}Np and associated volatile radionuclides from soil matrixes; uncertainties are expressed to 1σ.

± 6.4%) had the highest yield of ^{239}Np, compared with the 13.9 ± 0.3% Np yield achieved by UTEVA-Resin (standard deviation shown to 1σ). The RE-Resin not only showed the highest recovery of ^{239}Np but also the lowest contamination with ^{131}I (3.9 ± 0.4%) in the eluate, compared with TEVA-Resin (4.8 ± 0.3%) and UTEVA-Resin (4.4 ± 0.3%). Again, RE-Resin outperformed the more commonly used TEVA-Resin for the very purpose of this study. There was no detectable ^{123m}Te and ^{134}Cs activity observed in any of the eluates of this series (hence these were not listed in Figure 2). Furthermore, we have observed that the elution of samples through TEVA-Resin columns requires longer intervals of time and/or greater pressure differentials applied through the vacuum box, thereby being not as rapid as elution through the RE-Resin and potentially not as complete.

Separation of ^{239}Np from soil matrixes was conducted by boiling samples under reflux in the presence and, in preliminary trials, absence of 30% H$_2$O$_2$. It may be an interesting side-note that elution of Np from the RE- and UTEVA-Resin columns with H$_2$O was possible only with soil samples that were refluxed with a mixture containing 30% H$_2$O$_2$. For samples refluxed in the absence of 30% H$_2$O$_2$, elution of Np from these resin columns could not be accomplished with various eluents that were supposed to either shift the pH to neutral, oxidize the Np(IV) to Np(V), or chelate the Np and thus aid its elution. The eluents tried in the scenario without 30% H$_2$O$_2$ included H$_2$O, 0.01 M HNO$_3$, 30% H$_2$O$_2$, and 0.05 M oxalic acid. Removal of Np from the resin columns required dissolution of the entire stationary phase from the resin beads using acetone. However, the use of acetone cannot be recommended as a standardized protocol as it may elute any volatile radionuclides

(primarily ^{131}I) remaining on the column after loading and rinsing. Apparently the presence of H_2O_2 changes the chemistry in the soil extract during refluxing in a favorable way. We conclude that hydrogen peroxide must be used in the leaching process of soil to later effectively elute Np from the resin columns.

CONCLUSIONS

In the event of a nuclear accident, [239]Np detected in the environment signifies that fuel has leaked from the core elements of a reactor. Continuous monitoring of environmental samples for [239]Np would allow for faster and more accurate evaluations of whether or not a reactor core has emitted actinides. We present a rapid method for the isolation and detection of reactor-borne [239]Np using Eichrom's RE resin. For 25 mL of acidified rainwater, 45 min should be allotted for sample preparation, extraction chromatographic separation, and measuring each sample. Soil samples, however, would require an additional 60 min for boiling, cooling, and filtering. In our study, 600 s of γ-spectrometric measurement were sufficient to detect and quantify the radionuclides. Depending on detector efficiencies of the utilized γ-spectrometer, measurement geometry, levels of contamination, and the desired level of sensitivity, measurement time may be longer (or possibly even shorter). In addition, also chemical pretreatment for the removal of radioiodine may be required.[35] From the point of view of rapidness, the RE-Resin is also preferred as the preparation of chemicals requires the least efforts and chemical lab skills (conditioning/loading/rinsing with half-concentrated HNO_3 and elution with H_2O).

Although the TEVA- and UTEVA-Resins have also shown to be reliable in separating neptunium from volatile fission products, RE-Resin outperforms TEVA- and UTEVA-Resins when using 8 M HNO_3 as a conditioning/loading solution and H_2O as an eluent. The RE-Resin recovers a worthwhile amount of [239]Np in the rainwater scenario (69 ± 19%) and shows the highest yield when Np is extracted from soil (65 ± 4%). Moreover, the RE-Resin excluded ^{131}I and [123m]Te from the eluate to the highest degree of all resins. Therefore, the RE-Resin is favored over the TEVA-Resin (the most commonly used resin for Np separation in the literature) for the very purpose of the study, establishing a rapid method for emergency response.

Although a difference in performance is not expected, one should note that the presence of carriers (Cs, Te, and U) in this study may have a slight impact on the performance of the proposed methods when applied to real samples without any carriers. The proposed methods, of course, are only intended to act as a rapid response after a nuclear accident to answer the urgent yes-or-no question for major releases of fuel particles. Further analyses would have to be conducted in an accident scenario to establish the true quantity of actinides released.

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Notes

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