A bioassay method for 237Np in fecal samples

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Abstract

Neptunium-237 (237Np), as a highly toxic radionuclide, poses a significant risk of internal exposure that cannot be underestimated. Once inadvertently inhaled by humans, it can pose an extremely serious threat to health. Therefore, to more effectively recover and accurately measure this radionuclide from various complex samples, this study comprehensively and systematically explored the specific impacts of multiple factors, including fecal sample calcination temperature, the addition of oxidizing and reducing agents, and the separation and purification process using TEVA resin, on the chemical recovery rate of 237Np. The experimental results clearly demonstrate that, under the condition of precisely controlling the fecal sample calcination temperature at 600°C, coupled with the rational addition of oxidizing and reducing agents during the pretreatment process, and employing TEVA resin for meticulous separation and purification (with strict control of column acidity and elution volume throughout the process), the chemical recovery rate of 237Np measured by ICP-MS technology reached an impressive 79.07%. This result convincingly proves that the method adopted in this study can significantly enhance the chemical recovery rate of 237Np. Consequently, this study not only provides solid and reliable technical support for accurately measuring the content of 237Np in fecal samples but also exhibits profound significance and value in safeguarding public health and maintaining environmental safety.

Full Text

A Bioassay Method for ^{237}Np in Fecal Samples

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Abstract: Neptunium-237 (^{237}Np), as a highly toxic radionuclide, poses a significant risk of internal exposure that cannot be underestimated. Once inadvertently inhaled by humans, it can pose an extremely serious threat to health. Therefore, to more effectively recover and accurately measure this radionuclide from various complex samples, this study comprehensively and systematically explored the specific impacts of multiple factors, including fecal sample calcination temperature, the addition of oxidizing and reducing agents, and the separation and purification process using TEVA resin, on the chemical recovery rate of ^{237}Np. The experimental results clearly demonstrate that, under the condition of precisely controlling the fecal sample calcination temperature at 600°C, coupled with the rational addition of oxidizing and reducing agents during the pretreatment process, and employing TEVA resin for meticulous separation and purification (with strict control of column acidity and elution volume throughout the process), the chemical recovery rate of ^{237}Np measured by ICP-MS technology reached an impressive 79.07%. This result convincingly proves that the method adopted in this study can significantly enhance the chemical recovery rate of ^{237}Np. Consequently, this study not only provides solid and reliable technical support for accurately measuring the content of ^{237}Np in fecal samples but also exhibits profound significance and value in safeguarding public health and maintaining environmental safety.

Keywords: fecal samples; ^{237}Np; chemical recovery

1. Introduction

^{237}Np is a radioactive isotope belonging to the actinide series, primarily originating from nuclear reactors, nuclear weapon tests, or the release of radioactive waste. It easily disseminates in air, soil, water bodies, and biological organisms, accumulating through the food chain and ultimately entering the human or animal body. It tends to deposit in bones and the liver, causing radiation damage to organs and potentially leading to cancer [1, 2]. However, a significant portion of ^{237}Np absorbed by the human body is excreted in urine and feces. By detecting ^{237}Np in excretions, exposure doses can be estimated, thereby assessing potential health risks.

Nevertheless, due to variations in dietary structures and changes in individuals' diets over time, the matrix composition of fecal samples is highly complex and variable [3, 4]. This characteristic poses a challenge for the effective separation of ^{237}Np from fecal samples, which explains the relatively low proportion of fecal samples in radionuclide research and analysis thus far. Furthermore, the low concentration of ^{237}Np in environmental and biological samples makes the sensitivity and accuracy of detection techniques crucial for research [5-7].

Currently, we utilize high-sensitivity instruments such as inductively coupled plasma mass spectrometry (ICP-MS) or alpha spectrometry [8]. These instruments can detect radioactive isotopes at low concentrations, ensuring accurate reflection of exposure levels. Among them, ICP-MS offers even higher sensitiv-

ity, enabling the detection of lower concentrations of ^{237}Np, which facilitates more convenient detection in fecal samples [9]. Additionally, based on the similar first ionization energies of Np (6.27 eV) and Pu (6.03 eV) and their comparable chemical behavior, there is precedence for using ^{242}Pu, an isotope of Pu, as a non-isotopic tracer for the chemical pretreatment and measurement of ^{237}Np in radioanalytical procedures [10, 11]. Therefore, in this paper, we employ ^{242}Pu as a tracer to measure the chemical recovery of ^{237}Np in fecal samples using ICP-MS. By adjusting the calcination temperature of fecal samples, adding oxidizing or reducing agents, and modifying the acidity of the sample solution during column loading and the volume of the eluent in the TEVA resin separation and purification process, we achieve a high chemical recovery rate for ^{237}Np in fecal samples. This provides reliable technical support for the monitoring of internal exposure to ^{237}Np and dose assessment.

2.1 Materials and Characterization

TEVA Resin: Particle size 100-150 μm, supplied by Triskem, France; Extraction Column: Homemade with an inner diameter of 5 mm and a height of 20 mm; ^{237}Np standard solution with an activity of 0.889 Bg/L and ^{242}Pu standard solution with an activity of 2.68 Bg/L, both sourced from the National Physical Laboratory, UK, and calibrated for activity before use; Chemical reagents and solvents including nitric acid (HNO_3), hydrochloric acid (HCl), hydrofluoric acid (HF), and sodium nitrite($NaNO_2$), all of analytical grade, supplied by Sinopharm Chemical Reagent Co., Ltd., China; Muffle Furnace, supplied by Tianjin Zhonghuan Electric Furnace Co., Ltd.; Centrifuge, supplied by Shandong Baiou Medical Technology Co., Ltd.; Electric Hotplate, supplied by Beijing Yongguangming Medical Instrument Co., Ltd.; ICP-MS, supplied by Thermo Fisher Scientific (China) Co., Ltd.

2.2 Pretreatment of Fecal Samples

The flowchart for the sample preparation process is shown in Figure 1 [Figure 1: see original paper]. To monitor any losses during the sample preparation process, ^{242}Pu was added as a tracer for chemical recovery correction. Firstly, the fecal samples were dried. Then, the samples were transferred to a muffle furnace and calcined at 600°C for 2 hours. Next, the calcined samples were thermally digested with aqua regia. Subsequently, the samples were diluted with deionized water, centrifuged, and the supernatants were collected after centrifugation. The precipitates after centrifugation were repeatedly washed with dilute HNO_3 solution to reduce the loss of ^{237}Np and ^{242}Pu. Then, the supernatants were evaporated to dryness, and the dried samples were dissolved in 20 mL of 3 mol/L HNO_3. Finally, before separation and purification, 100 μL of the redox agents $Fe(NH_2SO_3)_2$ and $NaNO_2$ were added respectively to adjust the valence states of Np and Pu in the fecal samples to Np(IV) and Pu(IV).

Figure 1. Schematic diagram of fecal sample pretreatment process.

2.3 Separation and Purification of Fecal Samples

Separation and purification is a crucial step in the analysis and monitoring of ^{237}Np in fecal samples. Horwitz et al. conducted research on the adsorption behavior of actinides such as Np, Pu, and Th on TEVA resin in HNO_3 medium and found that at an HNO_3 concentration of 3 mol/L, the distribution coefficients for Np(IV) and Pu(IV) on TEVA resin reach their maximum (K' ranging from 10^3 to 10^4), while the distribution coefficient for U is less than 10 (K' $<$ 10). Therefore, under these conditions, TEVA resin exhibits the strongest adsorption capacity for Np(IV) and Pu(IV) but a weaker adsorption capacity for U. This allows for the removal of interfering metal ions from the sample using a relatively high concentration of acid.

As the acidity decreases, the distribution coefficients for Np(IV) and Pu(IV) also rapidly decrease (K' $<$ 10), facilitating the desorption of Np and Pu from the TEVA resin [7].

Figure 2 [Figure 2: see original paper]. Schematic diagram of separation and purification using TEVA resin.

In this study, we utilized TEVA extraction chromatographic resin for separation and purification [12, 13]. As illustrated in Figure 2, we first activated the TEVA resin with 20 mL of 3 mol/L HNO_3. Subsequently, the treated fecal sample was transferred to the TEVA resin column, and the sample beaker was washed multiple times with 15 mL of 3 mol/L HNO_3 to minimize the loss of Np and Pu. After transferring the wash solution to the resin column, we adjusted the flow rate to 0.5 mL/min. At this flow rate, 20 mL of 8 mol/L HNO_3 was used to rinse and remove U, followed by 20 mL of 9 mol/L HCl to rinse and remove Th. These steps were taken to reduce the impurity content in the fecal sample. Next, 15 mL of a low-concentration mixed acid solution containing 0.1 mol/L HCl and 0.05 mol/L HF was used to desorb Np and Pu from the TEVA resin. The desorbed solution was then evaporated to dryness, concentrated, and diluted with 3 mL of 2% HNO_3. Finally, the solution was analyzed by ICP-MS.

3.1 The Effect of Calcination Temperature

We discuss the impact of calcination temperature on the chemical recovery rate of fecal samples as described in Section 2.2. Firstly, 1 mL of ^{237}Np standard solution (0.889 mBq) and 1 mL of ^{242}Pu tracer (2.68 mBq) were added to the dried fecal samples. The samples were then placed in a muffle furnace and calcined at temperatures of 400 °C, 500 °C, 600 °C, 700 °C, 800 °C, 900 °C, and 1000 °C, respectively, for 2 hours. As shown in Figure 3 [Figure 3: see original paper], with the increase in calcination temperature in the muffle furnace, the color of the resulting samples gradually changed from black to white. This indicates that the organic matter in the fecal samples, after carbonization, gradually transformed into gases such as carbon dioxide and escaped from the samples as the temperature rose [13, 14].

Figure 3. Fecal samples calcined at different temperatures: a) 400 ℃; b) 500 ℃; c) 600 ℃; d) 700 ℃; e) 800 ℃; f) 900 ℃; and g) 1000 ℃.

The calcined samples were subjected to hot digestion with aqua regia. Subsequently, the samples were diluted with deionized water and centrifuged, after which the supernatants were collected. The supernatants were then evaporated to dryness, and the dried samples were dissolved in 3 mol/L HNO_3. Oxidizing and reducing agents, $Fe(NH_2SO_3)_2$ and $NaNO_2$, were added to the solution. The samples were then loaded onto columns containing TEVA resin for separation and purification. Impurities such as U and Th were removed by washing with 8 mol/L HNO_3 and 9 mol/L HCl, and Np and Pu were eluted with 15 mL of a mixed acid solution containing 0.1 mol/L HCl and 0.05 mol/L HF. Finally, the samples were measured by ICP-MS. This experiment was repeated three times, and the average chemical recovery rate was calculated. The results are shown in Figure 4 [Figure 4: see original paper]: as the calcination temperature increased, the chemical recovery rate of ^{237}Np in the samples initially rose, then decreased, and finally stabilized. The maximum chemical recovery rate was achieved at a calcination temperature of 600 °C, indicating that calcining the fecal samples was beneficial for improving the chemical recovery rate. However, further increasing the temperature led to a decrease in the chemical recovery rate, possibly due to the formation of refractory particles at excessively high temperatures, which prevented effective extraction of the target elements [15, 16]. Therefore, we selected 600 °C as the optimal calcination temperature for the fecal samples.

Figure 4. Chemical recovery rate of fecal samples at different calcination temperatures.

3.2 The Effect of Oxidizing and Reducing Agents

We discuss the impact of adding oxidizing and reducing agents to fecal samples on the chemical recovery rate as described in Section 2.2. Firstly, 1 mL of ^{237}Np standard solution (0.889 mBq) and 1 mL of ^{242}Pu tracer (2.68 mBq) were added to the dried fecal samples. The samples were then transferred to a muffle furnace and calcined at 600 °C for 2 hours. Subsequently, the calcined samples were subjected to hot digestion with aqua regia, diluted with deionized water, and centrifuged, after which the supernatants were collected. The supernatants were evaporated to dryness, and the dried samples were dissolved in 3 mol/L HNO_3.

Next, we proceeded with the addition of oxidizing and reducing agents. The first set of samples (A1) did not receive any oxidizing or reducing agents. The second set (A2) was treated with 100 µL of $Fe(NH_2SO_3)_2$ solution only. The third set (A3) was treated with both 100 µL of $Fe(NH_2SO_3)_2$ and $NaNO_2$ solutions. For TEVA resin separation and purification, the samples were loaded onto columns, washed with 8 mol/L HNO_3 and 9 mol/L HCl to remove impurities such as U and Th, and then eluted with 15 mL of a mixed acid solution containing 0.1

mol/L HCl and 0.05 mol/L HF to obtain ^{237}Np and ^{242}Pu. The samples were then measured by ICP-MS. This experiment was repeated three times, and the average chemical recovery rate was calculated.

Figure 5 [Figure 5: see original paper]. Relationship between redox agents and chemical recovery rate: A1 represents no addition of both $Fe(NH_2SO_3)_2$ and $NaNO_2$; A2 represents addition of only $Fe(NH_2SO_3)_2$; A3 represents addition of both $Fe(NH_2SO_3)_2$ and $NaNO_2$.

The results, shown in Figure 5, indicate that the chemical recovery rates of ^{237}Np and ^{242}Pu were lowest when no oxidizing or reducing agents ($Fe(NH_2SO_3)_2$ and $NaNO_2$) were added to the fecal samples. The highest chemical recovery rate was achieved when both $Fe(NH_2SO_3)_2$ and $NaNO_2$ were added to the samples. Adding only $Fe(NH_2SO_3)_2$ resulted in a moderate chemical recovery rate compared to the other two conditions. This is because $Fe(NH_2SO_3)_2$ can oxidize Np(V) present in the solution to Np(IV), and the subsequent addition of $NaNO_2$ ensures that all Pu in the solution is converted to Pu(IV). This facilitates better adsorption of these elements onto the TEVA resin, reducing sample loss and thereby improving the chemical recovery rate [7]. Therefore, the oxidizing and reducing agents $Fe(NH_2SO_3)_2$ and $NaNO_2$ are crucial for improving the chemical recovery rates of ^{237}Np and ^{242}Pu in fecal samples. In this study, we chose to add both $Fe(NH_2SO_3)_2$ and $NaNO_2$ as oxidizing and reducing agents.

3.3 The Effect of Column Loading Acidity

We discuss the impact of different column loading acidities on the chemical recovery rate during TEVA resin separation and purification as described in Section 2.3. Firstly, 1 mL of ^{237}Np standard solution (0.889 mBq) and 1 mL of ^{242}Pu tracer (2.68 mBq) were added to the dried fecal samples. The samples were then transferred to a muffle furnace and calcined at 600 °C for 2 hours. Subsequently, the calcined samples were subjected to hot digestion with aqua regia, diluted with deionized water, and centrifuged, after which the supernatants were collected. The supernatants were evaporated to dryness, and the dried samples were dissolved in HNO_3 solutions of 1, 2, 3, 4, and 5 mol/L, respectively. Oxidizing and reducing agents, $Fe(NH_2SO_3)_2$ and $NaNO_2$, were then added to each solution.

For TEVA resin separation and purification, the samples were loaded onto columns, washed with 8 mol/L HNO_3 and 9 mol/L HCl to remove impurities such as U and Th, and then eluted with 15 mL of a mixed acid solution containing 0.1 mol/L HCl and 0.05 mol/L HF to obtain ^{237}Np and ^{242}Pu. The samples were then measured by ICP-MS. This experiment was repeated three times, and the average chemical recovery rate was calculated.

Figure 6 [Figure 6: see original paper]. Chemical recovery rates at different HNO_3 concentrations during column loading of the pre-processed samples.

The results, shown in Figure 6, indicate that the chemical recovery rates of

^{237}Np and ^{242}Pu increased with increasing HNO_3 concentration from 1 to 3 mol/L. However, when the HNO_3 concentration was between 3 and 5 mol/L, the chemical recovery rates of ^{237}Np and ^{242}Pu tended to stabilize. Additionally, the experiment demonstrated that using ^{242}Pu as a tracer for ^{237}Np resulted in small errors and relatively accurate chemical recovery rates. Given that the elution solution, a mixed acid of 0.1 mol/L HCl and 0.05 mol/L HF, has a relatively low concentration, and to ensure efficient desorption of ^{237}Np and ^{242}Pu under low acidity conditions, we chose a column loading acidity of 3 mol/L HNO_3 for this work.

3.4 The Effect of Eluent Volume

We discuss the impact of eluent volume on the chemical recovery rate during TEVA resin separation and purification as described in Section 2.3. The primary objective is to investigate the optimal eluent volume required to desorb ^{237}Np from the TEVA resin, ensuring maximum chemical recovery. Initially, 1 mL of ^{237}Np standard solution (0.889 mBq) and 1 mL of ^{242}Pu tracer (2.68 mBq) were added to the dried fecal samples. The samples were then transferred to a muffle furnace and calcined at 600 °C for 2 hours. Subsequently, the calcined samples were subjected to hot digestion with aqua regia, diluted with deionized water, centrifuged, and the supernatants were collected. The supernatants were evaporated to dryness, and the dried samples were dissolved in 3 mol/L HNO_3. Oxidizing and reducing agents, $Fe(NH_2SO_3)_2$ and $NaNO_2$, were then added.

Figure 7 [Figure 7: see original paper]. Relationship between the volume of eluent and chemical recovery rate.

For TEVA resin separation and purification, the samples were loaded onto columns, washed with 8 mol/L HNO_3 and 9 mol/L HCl to remove impurities such as U and Th, and then eluted using a mixed acid solution of 0.1 mol/L HCl and 0.05 mol/L HF at a flow rate of 0.5 mL/min. Samples were collected every 2 mL for ICP-MS testing. The results, shown in Figure 7, indicate that the chemical recovery rate of ^{237}Np initially increased with increasing eluent volume and then gradually decreased. When the eluent volume reached 12 mL, the chemical recovery rate of ^{237}Np tended to maximize and stabilize. By calculating the standard deviation of the chemical recovery rates from six repeated experiments, it was found that the standard deviation s was less than 5%. This indicates that when the eluent volume reaches 12 mL, ^{237}Np can be completely desorbed. In this work, we chose a 15 mL eluent volume to ensure optimal desorption efficiency.

3.5 The Validation of Analytical Method

We conducted experimental validation to determine the reliability of experimental conditions, following the preliminary established experimental procedures (as shown in Figures 1 and 2) and based on the results obtained during the pretreatment and separation purification processes of the fecal samples.

Figure 8 [Figure 8: see original paper]. Chemical recovery rates for different groups of fecal samples, with 0 designating the blank sample.

As shown in Figure 8, we performed 11 sets of full-process experiments, where Sample 0 was a blank sample without fecal material added. Each set included 1 mL of ^{237}Np standard solution (0.889 mBq) and 1 mL of ^{242}Pu tracer (2.68 mBq). To ensure the accuracy of the experiments, each set was repeated three times, and the chemical recovery rates were averaged. Figure 8 demonstrates that the chemical recovery rates for the 10 sets of full-process experiments with fecal samples were relatively stable, with the chemical recovery rate of ^{237}Np consistently ranging from 70.85% to 79.07%. This confirms the feasibility of the experimental method and procedure.

4. Conclusion

In this study, a full-process experiment was conducted on fecal samples to establish an analytical monitoring procedure for ^{237}Np in feces, achieving a chemical recovery rate of up to 79.07%. This fully demonstrates the significance of conditions such as a calcination temperature of 600°C for fecal samples, oxidizing and reducing agents $Fe(NH_2SO_3)_2$ and $NaNO_2$, and adjustments to the column acidity and eluent volume during TEVA resin separation and purification for improving the chemical recovery rate of ^{237}Np. To some extent, this study fills the gap in research on measurement methods for ^{237}Np in fecal samples. It provides technical support for internal exposure monitoring and dose assessment of workers in subsequent practical applications.

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Data Availability Statement

The data that support the findings of this study are available from the author upon reasonable request.

Ethics Statement

The authors declare no ethical issue in relation to this study.

Conflicts of Interest

The authors declare no conflicts of interest.

Author Contributions

All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Xiao-Rui Wang, Chuan-Gao Wang, Ying Wang, and Ai-Yun Li. The first draft of the manuscript was written by Xiao-Rui Wang, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Note: Figure translations are in progress. See original paper for figures.