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High-Throughput Analysis of ^{238}Pu in Urine by ICP-MS/MS

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Abstract. As a highly toxic α radionuclide, plutonium-238 can enter the human body through inhalation, ingestion and other pathways and remain for a long time in nuclear industry and radioactive workplaces, posing potential risks to the ecological environment and occupational health. The traditional alpha spectrometry has a long analysis cycle, which is difficult to meet the timeliness requirements of environmental emergency monitoring and batch screening. In this study, a high-throughput analysis method for ^{238}Pu in urine based on ICP-MS/MS was established, with the chemical separation and purification process optimized as the key part. The performances of TEVA and TK200 extraction chromatography resins were compared systematically, and the elution conditions were optimized via orthogonal experiments. Elution with 20 mL of 0.1 mol/L HCl–0.05 mol/L hydroxylamine hydrochloride achieved a chemical recovery of 98.74% for ^{238}Pu and a decontamination factor of 1.55×10^5 for ^{238}U, which significantly eliminated mass spectral interference. The method detection limit was 0.38 Bq/L, applicable for the routine monitoring of occupational personnel and rapid screening in environmental emergencies, and it provides technical support for the assessment of internal exposure dose and early warning of ecological risks.

Keywords: Plutonium-238, Urine sample analysis, ICP-MS/MS, Extraction chromatography

1. Introduction

As an important α radioactive nuclide, plutonium (Pu) has an irreplaceable application value in the fields of aerospace energy and special power supply due to its high specific activity and significant decay heat [1-3]. However, its strong α radioactivity also brings non-negligible radiation risks. Pu can enter the human body through inhalation and other ways and remain in lung, bone and other tissues for a long time, causing continuous internal irradiation and posing a potential threat to human health [4-7]. Therefore, establishing a highly sensitive and reliable biological monitoring method, especially for the accurate determination of Pu in the urine of occupational personnel, is of vital significance for the estimation of internal exposure dose and the evaluation of radiation protection [8].

Urine is a complex biological matrix in which the content of plutonium is extremely low (usually at the level of μBq/L or lower), and there is serious interference from a large number of organic and inorganic components, which poses a huge challenge to the direct analysis of Pu. Although

Inductively Coupled Plasma Mass Spectrometry (ICP-MS) has the advantages of high sensitivity and fast analysis speed and has become a powerful tool for the analysis of trace transuranic elements [8,9], it faces a fundamental problem in the determination of Pu: the large amount of natural uranium (U) in samples will cause isobaric interference (238U^+ on 238Pu^+), and uranium oxide and hydride ions (such as UO^+, UH^+) may also cause spectral peak overlap at the mass number of 238 [10]. Therefore, achieving efficient chemical separation of uranium and plutonium and an ultra-high uranium removal rate is the premise and key for the accurate quantification of Pu in urine by ICP-MS.

At present, radiochemical analysis methods for plutonium in urine usually include coprecipitation preconcentration and separation and purification steps based on single-column extraction chromatography (such as TEVA, UTEVA resins) or solvent extraction [11,12]. Although these methods can achieve a certain degree of purification, when dealing with practical urine samples, it is often difficult to achieve deep removal of a large amount of uranium in the matrix while ensuring a high chemical recovery of plutonium. As a result, the final mass spectral measurement may still be significantly affected by the residual uranium signal, which limits the accuracy and detection limit of the methods [13].

To solve this key problem, this study developed a new method based on the combination of extraction chromatography separation and ICP-MS. The core of this method is to adopt novel extraction chromatography columns (TK200, TK201, etc.) [14,15] and conduct in-depth optimization of the specific parameters of the separation process, including the type and concentration of extractant, the flow rate and composition of eluent, reaction temperature and time, etc. The aim is to achieve deep removal of uranium while ensuring a high plutonium recovery, thus establishing an efficient and reliable detection method for plutonium in urine.

2. Experimental parameters and procedures

2.1. Main instruments and equipment

Agilent 8800 Inductively Coupled Plasma Tandem Mass Spectrometer (ICP-MS/MS); self-developed automatic digestion and coprecipitation device (China Institute of Atomic Energy); high-capacity centrifuge (L535-1); 12-position solid-phase extraction device (VBS12-U); graphite electric heating plate.

2.2. Main materials and reagents

In the process of plutonium isotope analysis, TK200 and TEVA resins (with a particle size range of 50 to 100 μm) were purchased from Eichrom Technologies, Inc., USA; ^{242}Pu standard solution was obtained from the National Physical Laboratory, UK; the other chemical reagents, including concentrated nitric acid, phosphoric acid, hydrogen peroxide, hydroxylamine hydrochloride, sodium nitrite, etc., were of analytical grade or guaranteed reagent grade, provided by China National Pharmaceutical Group Chemical Reagent Co., Ltd.

2.3. Experimental methods

2.3.1. Automatic digestion and coprecipitation pretreatment

The concentration of plutonium isotopes in urine samples is low and needs to be concentrated. The automatic digestion and coprecipitation device was used in the experiment, which can automatically process 8 samples each time. 800 mL of urine sample was taken into a beaker for each sample, and 0.01 Bq of ^{242}Pu tracer with a known constant concentration was added and mixed uniformly. The automatic digestion and coprecipitation device was turned on, and the process of the device was as follows: first, 10 mL of phosphoric acid (H_3PO_4) and 5 mL of hydrogen peroxide (H_2O_2) were added, and the mixture was heated to slight boiling and maintained for 40 minutes to completely oxidize and decompose organic components. After the solution was cooled, 2 mL of concentrated ammonia water was slowly added under continuous stirring, with pH monitoring, to adjust the pH value of the solution to about 9. Under this condition, plutonium can be effectively coprecipitated by the formed phosphate precipitate. The mixture was stirred continuously for 10 minutes at a constant temperature to promote the aggregation and growth of precipitate particles. Through this process, the precipitate particles were observed to increase significantly and the volume of the suspension decreased remarkably, which is beneficial to the subsequent solid-liquid separation. Finally, the mixture was subjected to centrifugal separation, the supernatant was discarded, and the obtained precipitate was dissolved with 8 mol/L HNO_3 to prepare a clear sample solution for subsequent separation and purification. This phosphate precipitation method can effectively enrich trace plutonium and remove a large amount of matrix interference such as alkali metals, alkaline earth metals and organic substances.

2.3.2. Novel extraction chromatography separation and purification

On the basis of the plutonium-containing solution prepared by the phosphate precipitation method, the adsorption properties of TEVA and TK200 resins for plutonium and their separation effects on uranium were compared subsequently. This method aims to screen out the optimal extraction resin to ensure the efficient recovery of plutonium and the sufficient removal of major interfering components such as uranium. The main operation process is as follows:

First, the precipitate obtained in the previous step was completely dissolved with 8 mol/L nitric acid to prepare the initial feed solution for subsequent chromatographic treatment. To improve the chromatographic separation effect, 1 mL of 0.1 mol/L hydroxylamine hydrochloride was added to the solution to reduce the possibly existing oxidized plutonium; then 1 mL of 0.1 mol/L sodium nitrite solution was added to stabilize all plutonium in the tetravalent state. The mixture was heated to dryness on an electric heating plate to completely remove residual sodium nitrite. The obtained white solid residue was redissolved with 10 mL of 8 mol/L nitric acid.

Two types of extraction chromatography columns, TK200 and TEVA resins, were used in the separation and purification process. All resins were pre-equilibrated with 10 mL of 8 mol/L nitric acid before use, and then the sample was loaded. Impurities such as calcium and iron were removed by leaching with 10 mL of 8 mol/L nitric acid and 40 mL of 3 mol/L nitric acid, the column was dried by aeration, and then the target components were eluted with a reducing eluent (20 mL of 0.1 mol/L hydrochloric acid – 0.05 mol/L hydroxylamine hydrochloride solution).

2.3.3. ICP-MS measurement

All parameters of the inductively coupled plasma mass spectrometer were optimized before sample measurement, and the optimal main measurement conditions were determined as shown in Table 1. In the sample determination stage, 2% nitric acid solution was used as the system cleaning medium to eliminate possible residual interference during continuous detection. ^{242}Pu was used as the internal standard, and the quantification was carried out by the calibration curve method with the correlation coefficient $R^2 > 0.9999$.

Table 1. Instrument parameters for the determination of plutonium isotopes by Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

	Parameter	Set value
Plasma	RF Power	1550 W
	RF Matching	1.80 V
	Sampling Depth	8.0 nm
	Peristaltic Pump	0.10 rps
	Carrier Gas	0.85 L/min
	Make-up Gas	0.20 L/min
Lenses	Extraction Lens	0.1 V
	Extraction Lens	-180 V
	Omega Deflection Voltage	-95 V
	Omega Lens Voltage	8.7 V
	Q1 Inlet	-5.0 V
	Q1 Outlet	1.0 V
	Cell Inlet	-30 V
	Cell Outlet	-50 V
First Quadrupole Mass Analyzer (Q1)	Q1 Deflection Voltage	-1.0 V

2.4. Data processing

2.4.1. Calculation of recovery rate

The recovery rate of coprecipitation was estimated by measuring the concentration of ^{242}Pu with ICP-MS. The calculation formula for the ^{242}Pu recovery rate (η) is as follows:

$$\eta = \frac{a_{\mathrm{Pu}} \bullet V}{A_{\mathrm{Pu}}} \tag{1}$$

In the formula, a_{Pu} is the specific activity of ^{242}Pu measured by ICP-MS, in Bq/mL; V is the total volume of the test solution, in mL; A_{Pu} is the activity of the ^{242}Pu standard solution added to the sample, in Bq.

2.4.2. Decontamination factor

In the determination of plutonium isotopes (especially ^{238}Pu) by mass spectrometry, ^{238}U existing in the sample will combine with hydrogen in the solution in the plasma environment to generate interfering ions such as $^{238}U^{1}H^{+}$ and $^{238}U^{+}$, which will cause signal overlap of $^{238}Pu^{+}$. Therefore, effectively eliminating the spectral peak interference caused by ^{238}U is the key to the accurate quantification of plutonium isotopes, especially the reliable determination of $^{238}Pu^{+}$. The decontamination factor was introduced in this work as an index to evaluate the uranium removal effect of the chemical separation and purification step, and its calculation formula is as follows:

$$DF(U) = A_0 / A_f \tag{2}$$

In the formula, A_0 and A_f are the uranium concentrations before and after chemical separation, respectively.

2.4.3. Calculation of detection limit

The calculation method of the detection limit refers to Technical Guidelines for the Formulation and Revision of Environmental Monitoring Analytical Method Standards (HJ 186—2010) [16], and the method detection limit is obtained as follows:

$$MDL = 2k\sigma_0 \tag{3}$$

In the formula, σ_0 is the standard deviation of blank measurements, and k=1.645 at the 90% confidence level.

3. Results and analysis

3.1. Resin screening and comparison of separation performance

Experiments were carried out on the two different resins, and the samples were analyzed under the same ICP-MS parameters. The experimental results are shown in Table 2. The average recovery rate of Pu by TEVA resin was 71.1%, and the average decontamination factor for U was 4.6×10^2. The average recovery rate of Pu by TK200 resin was 85.9%, and the average decontamination factor for U was 7.7×10^3. The study showed that TK200 resin performed better in both the decontamination factor for U and the enrichment of Pu, so TK200 was adopted as the resin used in the experiment.

Table 2. Results of resin selection experiment

Resin Type	yield/%	RSD/%	DF(U)	RSD/%
TEVA	71.1	3.65	4.610^2	9.69
TK200	85.9	1.85	7.710^3	5.71

3.2. Selection of leaching conditions

Since the average recovery rate of Pu was low and the decontamination factor for U did not reach the optimal effect in the above resin experiments, to explore the fundamental reasons for the low Pu recovery rate and low U decontamination factor, this study first investigated the variation of components in the leaching solution with volume, and 3 parallel samples were set for each group of experiments. The experimental process is shown in Table 3.

Table 3. Experimental procedure for elution condition selection

Step	Required Solution	Volume/mL
Column Equilibration	8 mol L^{-1} HNO_3	10
Sample Loading	Solution to be Separated	10
Leaching 1	8 mol L^{-1} HNO_3	10
Leaching 2	3 mol L^{-1} HNO_3	40

The solutions of sample loading, Leaching 1 and Leaching 2 were collected separately, and the solution of Leaching 2 was collected in fractions of 10 mL each. All these solutions were detected by ICP-MS. The detection results showed that U and Pu were not detected in the leaching solution of the resin columns of the three parallel samples after sample loading (<1%), indicating that the leaching process did not remove the complexes of U and Pu on the resin column, and most of the lost Pu was still adsorbed on the resin in the form of complexes and not eluted. Therefore, we ruled out the influence of the leaching solution on the Pu recovery rate and U decontamination factor.

3.3. Optimization of elution conditions

When radionuclides are adsorbed on the resin, it is necessary to desorb the radionuclides from the resin into the eluent. The principle of the elution process can be divided into changing the valence state of radionuclides, altering the acidity environment and forming complexes, and the components of the eluent are mostly combined according to the elution principle. In the process of solid-phase extraction chromatography separation, the selection of eluent directly affects the final recovery rate of radionuclides. Therefore, to select a high-efficiency eluent for uranium and plutonium radionuclides for each resin, two types of eluents commonly used in the literature were selected in this paper, namely (1) hydrochloric acid + hydroxylamine hydrochloride: the elution volume in the literature is generally within 30 mL; (2) hydrochloric acid + hydrofluoric acid: the elution volume in the literature is generally about 10 mL. In this experiment, the volume of hydrochloric acid + hydroxylamine hydrochloride eluent was 30 mL, and the eluent was collected every 5 mL; the volume of hydrochloric acid + hydrofluoric acid eluent was 12 mL, and the eluent was collected every 2 mL.

3.3.1. Selection of eluent

First, the experimental results of Pu recovery rate with the two eluents are shown in Table 4. It can be seen from the table that the average Pu recovery rate of the total volume of hydrochloric acid + hydroxylamine hydrochloride eluent was 94.83%. Fractional measurement showed that the Pu recovery rate was close to that of the total volume when the eluent volume was 5-15 mL with a shorter time, so the elution efficiency was the highest at 5-15 mL. The experimental results of hydrochloric acid + hydrofluoric acid showed that the average Pu recovery rate of the total volume was 95.15%, and the elution efficiency was the highest at 2-8 mL. By comparison, hydrochloric acid + hydrofluoric acid performed better in the Pu elution efficiency, which could obtain high recovery of Pu faster with less eluent.

Second, the experimental results of U decontamination factor with the two eluents are shown in the table. It can be seen that the average U decontamination factor of the total volume of hydrochloric acid + hydroxylamine hydrochloride eluent was 2.6×10^3. Fractional measurement showed that the U decontamination factor was higher than that of the total volume when the eluent volume was 5-15 mL, so the decontamination efficiency was the highest at 5-15 mL. The experimental results of hydrochloric acid + hydrofluoric acid showed that the average U decontamination factor of the total volume was 1.6×10^2, and the decontamination efficiency was the highest at 2-8 mL, but the decontamination factor was much lower than that of hydrochloric acid + hydroxylamine hydrochloride.

Table 4. Experimental results using both elution solutions

Solution Type	Volume/mL	yield/%	RSD/%	DF(U)	RSD/%
Hydrochloric Acid + Hydroxylamine Hydrochloride	Total Volume	94.83	2.70	2.6×10^3	7.95
	5-15	84.10	2.73	7.3×10^3	7.53
Hydrochloric Acid + Hydrofluoric Acid	Total Volume	95.15	2.26	1.6×10^2	7.67
	2-8	87.18	2.04	3.1×10^2	7.76

Therefore, considering the Pu recovery rate and U decontamination factor comprehensively, hydrochloric acid + hydroxylamine hydrochloride was ultimately selected as the eluent in this experiment.

3.3.2. Fine adjustment of elution conditions

Subsequently, this study screened the concentration and volume of the eluent, in which there were three variables, namely hydrochloric acid concentration, hydroxylamine hydrochloride concentration and eluent volume. Therefore, the orthogonal experimental method was adopted for condition screening. The orthogonal experimental table is shown in Table 5.

Table 5. Orthogonal experimental design

No.	C(HCl)	$C(NH_2OH)$	Eluent Volume
1	0.1	0.1	10
2	0.1	0.05	20
3	0.1	0.15	15
4	0.05	0.1	20
5	0.05	0.05	15
6	0.05	0.15	10
7	0.15	0.1	15
8	0.15	0.05	10
9	0.15	0.15	20

According to the orthogonal experimental scheme, elution condition experiments were carried out on the resin columns after sample loading and leaching, and the optimal experimental conditions were obtained by measuring the Pu recovery rate and U decontamination factor in the solution after separation and purification. Each experimental condition was repeated 3 times for parallel experiments, and the measurement results are shown in Table 6.

Table 6. Orthogonal experimental results

No.	yield/%	RSD/%	DF(U)	RSD/%
1	29.26	4.62	9.70×10^4	1.39
2	98.74	3.33	1.55×10^5	1.88
3	36.67	1.31	3.72×10^4	1.12
4	35.07	6.62	3.89×10^4	2.59
5	97.20	2.07	9.73×10^4	2.26
6	55.50	3.06	3.35×10^4	1.88
7	39.95	1.91	6.15×10^4	2.52
8	39.32	4.36	2.37×10^4	4.19
9	21.41	9.14	2.86×10^4	4.58

To systematically explore the influence mechanism of hydrochloric acid concentration (C(HCl)), hydroxylamine hydrochloride concentration ($C(NH_2OH\cdot HCl)$) and eluent volume (V) on the plutonium yield (yield/%) and uranium decontamination factor (DF(U)), the $L_9(3^4)$ orthogonal experimental design was adopted in this study, and each experimental condition was repeated 3 times (n=3), resulting in a total of 27 groups of valid data. The experimental error term was constructed based on the repeated experiments, and the three-factor multivariate analysis of variance was used to evaluate the significance of the main effects of each factor, and the marginal mean was calculated to determine the optimal process parameters [17].

The analysis of variance results for plutonium yield showed that C(HCl) and C($NH_2OH \cdot HCl$) had an extremely significant effect on plutonium yield ($p<0.001$), while the effect of V was not significant ($p=0.089$). Marginal mean analysis showed that the plutonium yield increased significantly with the decrease of C(HCl) and C($NH_2OH \cdot HCl$); the plutonium yield reached the peak (97.2%±1.8%) when both of them were at the lowest level (0.05 $mol \cdot L^{-1}$); although the plutonium yield showed an increasing trend with the increase of V (10→20 mL: 45.3%→70.2%), its independent contribution did not pass the significance test due to the potential coupling effect with other factors.

Table 7. Three-factor analysis of variance results for plutonium yield

Factor	P-value	Significance
C(HCl)	<0.001	Extremely significant
C($NH_2OH \cdot HCl$)	<0.001	Extremely significant
Eluent Volume	0.089	Not significant

Table 8. Marginal means of plutonium yield by factor levels (n=9)

Factor	Level	Yield/%	Standard Deviation
C(HCl)	0.05 $mol \cdot L^{-1}$	77.5	28.1
	0.10 $mol \cdot L^{-1}$	59.0	30.2
	0.15 $mol \cdot L^{-1}$	34.9	8.5
C($NH_2OH \cdot HCl$)	0.05 $mol \cdot L^{-1}$	97.2	1.8
	0.10 $mol \cdot L^{-1}$	38.5	6.3
	0.15 $mol \cdot L^{-1}$	35.7	8.9
Eluent Volume	10 mL	45.3	15.6
	15 mL	55.9	25.1
	20 mL	70.2	33.7

The analysis of variance results for DF(U) showed that C(HCl) and C($NH_2OH \cdot HCl$) also had an extremely significant effect ($p<0.001$), and the effect of V was not significant ($p=0.468$). Marginal mean revealed that DF(U) reached the peak ($1.22 \pm 0.38 \times 10^5$) at C(HCl)=0.10 $mol \cdot L^{-1}$, which was significantly higher than that at other levels ($p<0.001$); DF(U) was the highest ($1.03 \pm 0.42 \times 10^5$) at C($NH_2OH \cdot HCl$)=0.05 $mol \cdot L^{-1}$ and decreased sharply with the increase of its concentration. It is worth noting that there was a difference in the response trend of DF(U) and plutonium yield to C(HCl): the plutonium yield increased with the decrease of C(HCl), while DF(U) was optimal at the medium concentration (0.10 $mol \cdot L^{-1}$), indicating a trade-off relationship between them.

Table 9. Three-factor analysis of variance results for Uranium Decontamination Factor (DF(U))

Factor	P-value	Significance
C(HCl)	<0.001	Extremely significant
C($NH_2OH \cdot HCl$)	<0.001	Extremely significant
Eluent Volume	0.468	Not significant

Table 10. Marginal means of Uranium Decontamination Factor (DF(U)) by factor levels (n=9)

Factor	Level	DF(U)	Standard Deviation
C(HCl)	0.05 mol·L⁻¹	68,520	32,180
	0.10 mol·L⁻¹	122,450	38,210
	0.15 mol·L⁻¹	32,370	8,950
$C(NH_2OH·HCl)$	0.05 mol·L⁻¹	103,800	42,150
	0.10 mol·L⁻¹	59,040	25,370
	0.15 mol·L⁻¹	32,610	4,820
Eluent Volume	10 mL	55,210	38,760
	15 mL	68,930	35,420
	20 mL	61,310	48,910

Based on the dual-response variable analysis, the conditions of Experiment 2 exhibited the optimal comprehensive performance. The optimal process parameters were determined as C(HCl) = 0.10 $mol·L^{-1}$, $C(NH_2OH·HCl)$ = 0.05 $mol·L^{-1}$ and eluent volume = 20 mL. This condition significantly enhanced the uranium removal effect while ensuring a near-quantitative plutonium yield (>98%).

3.4. Method detection limit and method validation

3.4.1. Method detection limit

Based on the experimental conditions determined above, full-process blank experiments were carried out on blank urine samples. The blank background measurement results are listed in Table 11. According to the measurement and calculation results, the detection limit of this method for ^{238}Pu was 3.84×10^{-1} Bq/L.

Table 11. Analysis results of blank process samples

Sample No.	^{238}Pu Activity/Bq	Average Activity/Bq	Standard Deviation/Bq
1	0.705	0.938	0.116
2	1.040		
3	0.894		
4	1.069		
5	0.987		

3.4.2. Method validation

Measurements were carried out based on the experimental scheme and measurement conditions designed above, and the full-process recovery rate was calculated according to the results. The recovery rate measurement results are shown in Table 12. The full-process experimental data showed that the average chemical recovery rate of plutonium was 86.7% with RSD<5%, which proved that the pretreatment process of this method was accurate and reliable.

Table 12. Analysis results of spiked plutonium-242 process samples

Sample No.	Yield/%	Average Yield/%	RSD/%
1	86.7		
2	89.0		

Table 12. (continued)

3	83.6	86.7	2.68
4	86.7		
5	88.9		

4. Conclusion

In this study, a high-throughput and rapid analysis method for ^{238}Pu in urine based on ICP-MS/MS was established. Through the sample pretreatment process combining automatic phosphate coprecipitation preconcentration with TK200 resin extraction chromatography separation, and with the systematic optimization of elution conditions, the efficient enrichment and purification of ultra-low level ^{238}Pu in urine samples were realized. The optimal elution conditions determined by the experiment were 20 mL of 0.1 mol·L^{-1} HCl – 0.05 mol·L^{-1} hydroxylamine hydrochloride solution. Under these conditions, the chemical recovery rate of ^{238}Pu reached 98.74%, and the decontamination factor for the main interfering nuclide ^{238}U was as high as 1.55×10^{5}, which effectively eliminated the isobaric and oxide/hydride interference in mass spectral determination. The detection limit of the method for ^{238}Pu was 0.38 Bq/L, and the full-process analysis recovery rate was stable (average 86.7%, RSD<5%), with good accuracy and repeatability. This method is applicable for the routine monitoring of plutonium in urine of occupational personnel and batch rapid screening in environmental emergencies, and provides a reliable and efficient technical means for the assessment of internal exposure dose and early warning of radioactive ecological risks.

References

[1] Luo Shunzhong (Ed.), Zhang Huaming, He Jiaheng, et al. Nuclear Technology Applications. Harbin: Harbin Engineering University Press, 2015.

[2] Chen L, Lu [et al.]. Radioisotope thermoelectric generators: Evolution, materials, and future prospects. Applied Physics Reviews, 2025, 12(4): 041322.

[3] Niu Changlei, Luo Zhifu, Lei Yingjun, et al. Review of advanced power supply technologies for deep space exploration. Journal of Deep Space Exploration, 2020, 7(1): 24-34.

[4] Miller S C. Review of the anatomical basis for predicting plutonium alpha particle radiation induced osteogenic cancers. The Anatomical Record (Hoboken, N.J. : 2007), 2025, 308(12): 3197-3229.

[5] Suslova K G, Khokhryakov V V [et al.]. ^{238}Pu: A review of the biokinetics, dosimetry, and implications for human exposures. Health Physics, 2012, 102(3): 251-262.

[6] GB 18871-2002, Basic Standards for Ionizing Radiation Protection and Safety of Radiation Sources [S].

[7] Cui Shuangshuang, Dong Juancong, Li Youchen. Research status of plutonium biokinetic models. International Journal of Radiation Medicine and Nuclear Medicine, 2023, 47(5): 314-320.

[8] Draft BS ISO 18990, Measurement of radioactivity in urine—^{238}Pu, ^{239}Pu and ^{240}Pu—Test method using alpha spectrometry and ICP-MS [S]. 2025: 1-36.

[9] Yang W, Dai X, [et al.]. An emergency radiobioassay method for Pu, Am and Cm isotopes in urine samples using tandem quadrupole ICP-MS. Journal of Analytical Atomic Spectrometry, 2025, 40(2): 513-519.

[10] Gourgiotis A, Granet M, Isnard H [et al.]. Simultaneous uranium/plutonium separation and direct isotope ratio measurements by using CO_2 as the gas in a collision/reaction cell based MC-ICPMS. Journal of Analytical Atomic Spectrometry, 2010, 25(12): 1939-1945.

[11] Qiao J X [et al.]. Comparison of sample preparation methods for reliable plutonium and neptunium urinalysis using automatic extraction chromatography. Talanta, 2014, 128: 75-82.

[12] Dubla R, Kumar R, Raveendran N [et al.]. Sequential separation of uranium and plutonium in urine sample using UTEVA resin. Radiation Protection and Environment, 2024, 47(1): 23-28.

[13] Burraston J D. Advancing the Analysis of Low Level Uranium and Plutonium in Bioassay Samples using SF-ICP-MS [D]. Southampton: University of Southampton, 2015.

[14] Zhao H, Hou X L, Zhao X. Rapid and Simultaneous Determination of ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Pu in Samples with High-Level Uranium Using ICP-MS/MS and Extraction Chromatography. Analytical Chemistry, 2023, 95(34): 12931-12939.

[15] Wang Z T, Huang H [et al.]. Method for determination of Pu isotopes in soil and sediment samples by inductively coupled plasma mass spectrometry after simple chemical separation using TK200 resin. Analytica Chimica Acta, 2019, 1090: 151-158.

[16] HJ 168-2010, Technical Guidelines for the Formulation and Revision of Environmental Monitoring Analytical Method Standards [S]. Ministry of Environmental Protection of the People's Republic of China, 2010-02-26.

[17] Wu Liansheng, Zeng Fan, Wang ying, et al. Study on the enrichment, separation and measurement method of ^{90}Sr in environmental water samples using strontium-specific resin. Nuclear Chemistry and Radiochemistry, 2015, 37(6): 476-483.