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研究課題名(和文)環境試料および廃炉関連廃棄物中重要Pu同位体同時測定のための先端質量分析法の開発

研究課題名(英文) Development of mass spectrometric method for simultaneous determination of hard-to-measure Pu isotopes in environmental and reactor decommissioning waste samples

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研究成果の概要(和文)：本研究では、トリプル四重極ICP-MS/MSを用いて、イオン-分子反応よりU-238とPu-238を分別し、これまで測定が困難であったPu同位体の同時測定を可能とする先端質量分析法の開発を行った。脱溶媒試料導入装置の導入によりプラズマへの導入効率の増加と水素化物生成の抑制によるPu同位体に関して10倍の感度向上を成功し、オンマスモードでfgレベルPu同位体の検出限界を達成した。環境試料と廃炉関連廃棄物試料分析応用ために、少量海水迅速分析法を確立及び新規低温溶融法を開発した。これを開発したICP-MS/MS測定法と組み合わせ、環境試料と廃炉関連廃棄物試料の迅速Pu同位体先端質量分析法を確立した。

研究成果の学術的意義や社会的意義

東電福島第一原発事故由来Pu同位体の環境動態解析、環境除染・廃炉作業に伴い発生する廃棄物の汚染状況の把握及び被ばく線量の評価には、重要同位体(Pu-238, Pu-239, Pu-240, Pu-241)の測定が必要である。本研究で開発した環境と廃炉関連試料中Pu迅速質量分析法は環境に優しい方法であり、環境科学研究と原子力緊急時の対応に幅広い応用を期待できる。

研究成果の概要(英文)：In this study, we developed an advanced mass spectrometry method for simultaneous analysis of important Pu isotopes (238,239,240,241Pu) using triple quadrupole ICP-MS/MS by employing the ion-molecule reaction separate U-238 and Pu-238, which was previously difficult to be measured. By introducing a desolvation sample introduction system, we succeeded in increasing 10 times the sensitivity of Pu isotopes and suppressing the production of uranium hydride, we achieved the detection limit of fg level in on-mass mode. We have established a rapid analytical method for small volume seawater and developed a new low-temperature fusion method for environmental and nuclear decommissioning samples. By combining these new sample preparation techniques with the developed ICP-MS/MS detection, a rapid Pu isotope mass spectrometry method for environmental samples and decommissioning related waste samples was established.

研究分野：環境学

キーワード：Pu同位体 トリプル四重極ICP-MS/MS 環境動態解析 廃炉関連廃棄物 環境試料 新規低温溶融法 先端質量分析法

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## 様式 C - 19、F - 19 - 1、Z - 19 (共通)

### 1 . 研究開始当初の背景 **Scientific background for the research**

Essentially all the Pu found in the environment is of anthropogenic origin, such as atmospheric nuclear weapons testing, reprocessing of nuclear fuel, and releases from nuclear accidents, of the past decades. Due to the high radiological toxicities and very long radioactive half-lives, Pu isotopes are regarded as highly hazardous contaminants in the environment, and the most frequently monitored Pu isotopes are  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  with half-lives of 87.7 y, 24110 y, 6561 y and 14.4 y, respectively. From the viewpoint of radiation protection,  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$  have been assigned as the most principle Pu isotopes contributing to the dose from external exposure from deposition for a reactor accident (IAEA, 1997).

Activities of  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$  varied significantly (more than one order of magnitude) in environmental mediums, for example, the typical global fallout  $^{239+240}\text{Pu}$  activity ranged from 0.15 to 4.31 mBq/g in Japanese soils before the Fukushima Daiichi nuclear power plant (FDNPP) accident (Muramatsu et al. 2003), thus, if only Pu activities were measured, it is hard to identify and assess any accident-derived new Pu input into the environment. It was well-known that Pu isotopic compositions (ratios of  $^{238}\text{Pu}/^{239+240}\text{Pu}$ ,  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{241}\text{Pu}/^{239}\text{Pu}$ ) vary significantly in the environment depending on the emission source (Zheng et al., 2013), the Pu isotopic ratio, therefore, has been used as a fingerprint for contamination source identification. After the FDNPP accident, the released Pu isotopes have been detected in various environmental mediums in our previous study (Zheng et al. 2012) and by other groups (Yamamoto et al. 2014; Sakaguchi et al. 2014; Shinonaga et al. 2014). Although the amounts of Pu isotopes released were small (almost 4 orders of magnitude lower than those released from the Chernobyl accident), knowledge on the distribution and long-term environmental behavior are important for strengthening radiological preparedness of future nuclear disasters. In addition, large amounts of contaminated materials have been accumulated during the environmental decontamination operations since 2011, information on the actinides, especially Pu contamination levels is essential for the strategy of their final disposal. Furthermore, for the disposal of radioactive wastes generated from nuclear reactor decommissioning, obtaining information on the activity and isotope compositions of Pu is inevitable for radiation dose assessment. All these studies have led to a rapidly increasing demand for reliable and rapid analysis of Pu isotopes ( $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$ ) in different environmental mediums.

Conventionally, alpha spectrometry has usually been used for Pu ( $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ) measurement in various environmental samples. This method, however, normally requires a tedious chemical separation procedure and long counting times (days to weeks). Moreover, alpha spectrometry cannot distinguish  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  due to the closeness of the alpha radiation energies (5.16 MeV and 5.17 MeV for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , respectively), so it cannot provide isotopic composition information. In contrast, mass spectrometry, especially ICP-MS, as an atom-counting approach, is gradually replacing alpha spectrometry as a main-stream technique for Pu isotopic analysis. However, as Pu is often concomitant with a large (5-10 orders of magnitude) excess of U in environmental matrix, which clearly obscures the determination of  $^{238}\text{Pu}$  using any mass spectrometer due to the isobaric interference from  $^{238}\text{U}$ . The required resolution power to overcome U/Pu isobaric interference is around 190000, which is beyond the actual possibilities of any mass spectrometer. So far, there is no established mass spectrometric method for simultaneous determination of these hard-to-measure  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  in environmental mediums.

To overcome the U/Pu isobaric interference for  $^{238}\text{Pu}$  analysis, a promising approach is the *in-situ* interference resolution using ion-gas molecule reaction in collision/reaction cell devices. The choice of an appropriate reactant gas is based on the different behavior between the analyte and the interfering species with respect to the considered gas. Recent studies have shown the different behaviors of U and Pu towards  $\text{CO}_2$ ,  $\text{NH}_3$  and  $\text{C}_2\text{H}_4$  reaction gases in collision/reaction cell (Tanner et al. 2004; Epov et al. 2005; Gourgiotis et al. 2010). In this study, these novel *in-situ* isobar separation processes combined with advanced sample preparation techniques for removing sample matrix and interference elements will be applied to the development of a mass spectrometric method for simultaneous determination of the hard-to-measure Pu isotopes in various environmental mediums.

### 2 . 研究の目的 **Aims of the study**

This study aims to development of mass spectrometric method for simultaneous determination of the hard-to-measure Pu isotopes ( $^{238-241}\text{Pu}$ ) in various environmental mediums, such as contaminated soil, sediment, seawater, vegetation, and radioactive wastes generated from the nuclear reactor decommissioning, for instance steel and concrete samples. For this purpose, effective chemical separation procedures prior to mass spectrometric measurement and the *in-situ* isobar interference resolution techniques need to be developed.

### 3 . 研究の方法 **Research methods**

Mass spectrometric determination of  $^{238}\text{Pu}$  has long been a great challenge. In this project, an ICP-MS/MS method will be explored to realize simultaneous determination of  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$

in environmental mediums. To accomplish this purpose, in-situ ion-gas molecule reaction technique will be tested to resolve U/Pu in ICP-MS/MS, and several new approaches will be adopted to improve ICP-MS/MS sensitivity to realize ultra-trace Pu analysis comparable to that of SF-ICP-MS. Therefore, the following aspects will be elucidated.

(1) Study on the ion-molecule reactions between U/Pu ions and reaction gas to identify the best collision/reaction cell parameters for *in-situ* separation of U and UH<sup>+</sup> from Pu in triple quadrupole ICP-MS/MS. In-situ interference resolution using ion-gas molecule reactions has been shown as a promising approach to separate isobaric and polyatomic interferences in mass spectrometric analysis (Bandura *et al.* 2006). In this research project, we will investigate the ion-gas reactions between U<sup>+</sup>, UH<sup>+</sup> and reaction gas, CO<sub>2</sub>, in an octopole reaction cell installed in a triple-quadrupole ICP-MS/MS. A schematic diagram of U<sup>+</sup>, UH<sup>+</sup> separation using CO<sub>2</sub> reaction gas in ICP-MS/MS is shown in Figure 1. The residue U in the Pu sample solution is separated from Pu with the reaction to form UO<sup>+</sup> species, thus being rejected by the second quadrupole mass filter to realize the mass spectrometric determination of <sup>238</sup>Pu, which has long been a great challenge in the field of radioanalytical chemistry. The flow rate of reaction gas is crucially important to remove U<sup>+</sup> by the formation of UO<sup>+</sup>, and to control the signal loss of Pu<sup>+</sup> by decreasing the PuO<sup>+</sup> formation. Thus, gas flowrate and reaction cell parameters will be optimized in this research.

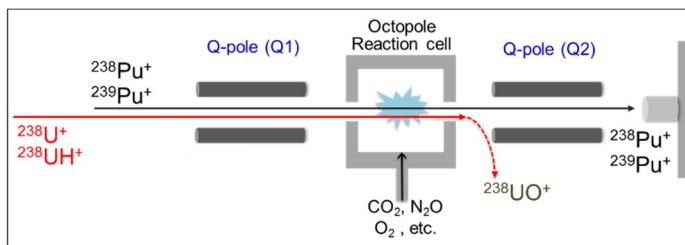


Fig. 1 Diagram of U<sup>+</sup> and UH<sup>+</sup> separation using CO<sub>2</sub> reaction gas in ICP-MS/MS.

(2) Study on the sensitivity enhancement of ICP-MS/MS for ultra-trace level analysis by combine high efficiency desolvation sample introduction system. In our previous study on fission products <sup>135</sup>Cs and <sup>137</sup>Cs analysis using ICP-MS/MS, we found that the hyphenation of a desolvation sample introduction system (APEX-Q) resulted in a 10-fold sensitivity improvement (Zheng *et al.* 2016). In this research, we will test the APEX-Q desolvation sample introduction system for the potential sensitivity improvement for actinides, Pu isotope analysis. The goal is to achieve a similar sensitivity of the SF-ICP-MS, thus to realize Pu analysis at ppq (fg/ml) level.

(3) Development of new sample digestion method for environmental samples (soil/sediment) and for nuclear decommissioning wastes, such as steel and concrete, and establish effective chemical separation/pre-concentration method to separate sample matrix and interfering elements, especially U. Nuclear decommissioning waste samples consist of different matrix from the conventional environmental samples, such as soil, sediment etc. The standard digestion method for Pu release from those sample matrix was not available in Japanese radioactivity measurement guideline (文科省放射能測定法シリーズ), therefore, besides the conventional acidic digestion, new methods such as novel fusion techniques will be investigated.

(4) Validation of the developed methods by analysis of reference materials (soil, sediment, seawater etc.), and application analysis for environmental samples (seawater and marine sediment in Japanese coast) and simulated nuclear decommissioning samples.

#### 4 . 研究成果 Results

During the three years project study, we achieved the following important results.

(1) in-situ ion-gas molecule reaction technique to resolve U/Pu in ICP-MS/MS

We combined the APEX-Q membrane desolvation sample introduction to the ICP-MS/MS instrument (Agilent 8800) to optimize the in-situ ion-gas molecule reaction technique to resolve U/Pu. The outlet of the APEX-Q system was connected directly to the torch of the ICP-MS/MS, and CO<sub>2</sub> gas was introduced into the octupole collision/reaction cell via the fourth cell gas line. Figure 2 shows effects of CO<sub>2</sub> flow rate on <sup>238</sup>U intensity, <sup>239</sup>Pu intensity, and the formation of <sup>238</sup>UO<sup>+</sup> and <sup>239</sup>PuO<sup>+</sup> evaluated using a <sup>239</sup>Pu standard solution (4.5 ppt) and an ICP-MS tune solution containing <sup>238</sup>U at different concentrations (0.02, 0.05 and 0.1 ppb). With the increase of CO<sub>2</sub> flow rate, the formation of <sup>238</sup>UO<sup>+</sup> and <sup>239</sup>PuO<sup>+</sup> was immediately observed. Under the optimized cell condition, the different reactivity observed for U and Pu towards CO<sub>2</sub> gas led it possible to measure <sup>238</sup>Pu because the signal intensity of <sup>238</sup>U decreased quickly with the increase of CO<sub>2</sub> gas flow rate. As

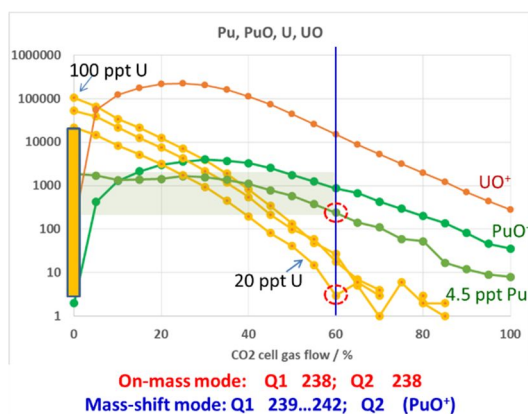


Fig. 2 Effects of CO<sub>2</sub> flow rate on intensities of U and Pu, and the formation of UO<sup>+</sup> and PuO<sup>+</sup>.

shown in Fig. 2, the signal intensity of 20 ppt  $^{238}\text{U}$  in Pu solution dropped more than 4 orders of magnitude to ca. 2 cps, when  $\text{CO}_2$  gas flow rate was increased to 60 % (0.6 mL/min), while the signal intensity of Pu just decreased ca. 60 %. This ion-gas molecule reaction almost eliminated the interference of  $^{238}\text{U}$  for the detection of  $^{238}\text{Pu}$  using on-mass mode, that is setting both Q1 and Q2 to  $m/z = 238$ , when  $^{238}\text{U}$  concentration is less than 20 ppt. Since the signal intensity of  $\text{PuO}^+$  is higher than  $\text{Pu}^+$ , mass-shift mode could be employed for the detection of  $^{239,240,241,242}\text{Pu}$  isotopes. For environmental samples, we have developed effective chemical separation methods to separate U from Pu, and the residue  $^{238}\text{U}$  concentration is normally less than 10 ppt in the final Pu solution prior to ICP-MS/MS measurement (see details in section (3)), therefore, by combining effective chemical separation techniques, the developed APEX-ICP-MS/MS system demonstrated great capacity to realize simultaneous determination of important Pu isotopes, including  $^{238}\text{Pu}$ .

## (2) Sensitivity enhancement and detection limits of Pu isotopes

Due to the improvement of sample transport efficiency and minimizing  $\text{PuO}^+$  formation in the plasma, a significant sensitivity enhancement could be expected by using desolvation sample introduce device. Using a  $^{238}\text{U}$  standard solution, we compared the sensitivity of Pu and U isotopes achievable with ICP-MS/MS and APEX-ICP-MS/MS under no-reaction gas condition. We found that a 10-fold sensitivity enhancement was achieved with APEX-ICP-MS/MS. For 1 ppb  $^{238}\text{U}$  (or Pu) standard solution, the highest sensitivity up to 3M cps was obtained, which is comparable to that of SF-ICP-MS. When  $\text{CO}_2$  gas was introduced, at a low rate of 0.6 mL/min, similar to the no-gas mode, a 10-fold sensitivity improvement still remained with APEX-ICP-MS/MS. On the basis of the definition of 3 $\sigma$  of the blank solution, for  $^{238}\text{Pu}$ , a detection limit of 3.7 fg/ml was obtained under on-mass mode detection; for  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$ , detection limits of 1.4 fg/ml, 2.9 fg/ml and 1.1 fg/ml, respectively, were achieved under mass-shift mode detection. And for the purpose of just analysis of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  in samples, APEX-ICP-MS/MS without using reaction gas achieved much lower detection limits of 0.29 fg/ml, 0.09 fg/ml, and 0.05 fg/ml, respectively. These detection limits are *ca.* one order of magnitude lower than those obtained with using  $\text{CO}_2$  reaction gas, and comparable to the SF-ICP-MS detection limit.

## (3) Development of novel chemical separation methods

### Rapid chemical separation method for small-volume seawater Pu isotope analysis

In order to assess the impact of the Fukushima accident derived Pu isotopes on seawater, a new analytical method to rapidly determine Pu isotopes in seawater including  $\text{Fe}(\text{OH})_2$  primary co-precipitation,  $\text{CaF}_2/\text{LaF}_3$  secondary co-precipitation and TEVA+UTEVA+DGA extraction chromatographic separation and ICP-MS detection was established (Fig. 3). High concentration efficiency ( $\sim 100\%$ ) and high U decontamination factor ( $\sim 10^7$ ) were achieved. The plutonium chemical recoveries were 74-88% with the mean of  $83\pm 5\%$ . The precisions for both  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios and  $^{239+240}\text{Pu}$  activity concentrations were less than 5% when 15 L of seawater samples with the typical  $^{239+240}\text{Pu}$  activity of the Northwest Pacific were measured. It just needs 12 hours to determine plutonium using this new method. This method has been successfully applied to the study of Pu contamination impact in coastal seawaters collected within two months after the FDNPP accident.

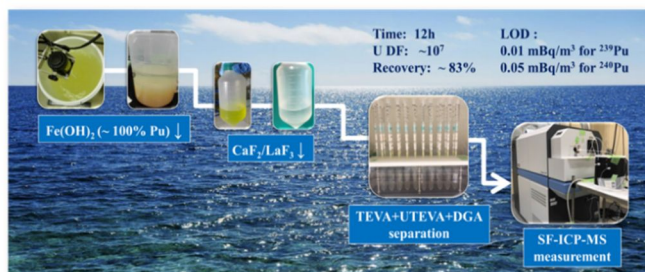


Fig. 3 Rapid chemical separation for small volume seawater Pu analysis.

### Low-temperature fusion for rapid determination of Pu isotopes in environmental samples

Two fusion methods were established for rapid determination of Pu in soil and sediment samples. The methods consisted of  $\text{NH}_4\text{HSO}_4$  or  $\text{NH}_4\text{HF}_2$  fusion procedures incorporated with procedures for  $\text{CaF}_2/\text{LaF}_3$  co-precipitation, extraction chromatography and ICP-MS measurement (Fig. 4). The fusion procedures were done on a portable hot plate instead of in a cumbersome muffle furnace and took only 15 min heating-up time from room temperature to  $250^\circ\text{C}$  and 15 min fusion time at  $250^\circ\text{C}$ . Chemical recoveries of Pu after completing the  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{HF}_2$  fusion methods for 0.5-1 g sample were approximately 70% and more than 90%, respectively. Sediment samples were pre-ignited at different temperatures ranging from  $450^\circ\text{C}$  to  $1000^\circ\text{C}$  to form refractory fractions of Pu, with which the dissolution rates of Pu by the  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{HF}_2$  fusion were investigated. With the increase of pre-ignition temperature of the sediment samples, the dissolution rates of Pu from the samples prepared by  $\text{NH}_4\text{HSO}_4$  fusion declined dramatically from near 100% for  $450^\circ\text{C}$  to 8% for  $1000^\circ\text{C}$ . In



contrast, the  $\text{NH}_4\text{HF}_2$  fusion was capable of completely releasing Pu from samples that were pre-ignited at temperatures over  $450\text{ }^\circ\text{C}$  to  $1000\text{ }^\circ\text{C}$ , which was comparable to releases obtained by the hazardous and time-consuming  $\text{HNO}_3$ -HF digestion, and was superior to the conventional  $\text{HNO}_3$  digestion. Additionally, because HF is not used in any procedure of the  $\text{NH}_4\text{HF}_2$  fusion, a safer and greener alternative to  $\text{NH}_4\text{HSO}_4$  fusion and  $\text{HNO}_3$ -HF digestion is realized for rapid Pu determination in environmental samples for nuclear emergency response and application in environmental studies.

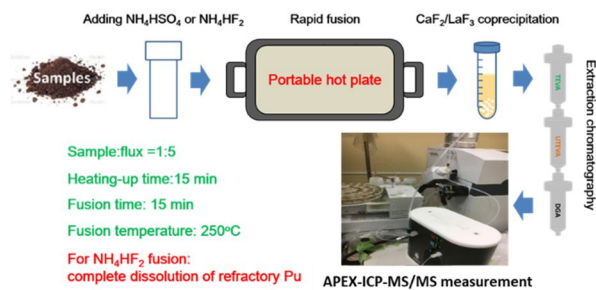


Fig. 4 Low-temperature fusion methods for rapid Pu analysis.

#### (4) Validation of the development mass spectrometric method

To assess the accuracy and applicability of the developed low-temperature fusion using  $\text{NH}_4\text{HF}_2$  combined to APEX-ICP-MS analytical method for ultra-trace level Pu in environmental samples, two sediment reference materials, namely IAEA-384 (Fangataufa Lagoon) and IAEA-385 (Irish Sea) were analyzed. The measured  $^{239+240}\text{Pu}$  activities were  $103.1 \pm 8.8\text{ mBq/g}$  and  $3.07 \pm 0.16\text{ mBq/g}$  for IAEA-384 and IAEA-385, respectively, and agreed well with the certified values of 103-110 mBq/g and 2.89-3.00 mBq/g. Isotope ratios of  $^{240}\text{Pu}/^{239}\text{Pu}$  measured were  $0.050 \pm 0.002$  and  $0.180 \pm 0.019$  for IAEA-384 and IAEA-385, respectively, also agreed well with values reported in the literature. These results demonstrated that the APEX-ICP-MS/MS analytical method combined with rapid low-temperature fusion sample preparation provided sufficient sensitivity and accuracy for ultra-trace Pu isotopes in environmental samples.

#### (5) Application analysis of environmental samples and simulated nuclear decommissioning samples

##### Determination of Pu isotopes in marine sediments

The developed  $\text{NH}_4\text{HF}_2$  low-temperature fusion method combined with the APEX-ICP-MS/MS detection was applied to the determination of Pu isotopes in Japanese coast sediment samples collected off Hokkaido, Aomori, Kagoshima, Niigata, Shimane, Ishikawa, Fukushima and Shizuoka after the FDNPP accident. Fig. 5 shows the comparison of  $^{239+240}\text{Pu}$  activity concentrations measured by APEX-ICP-MS/MS and APEX-SF-ICP-MS. These measurement results agreed very well, demonstrating the reliability of the developed APEX-ICP-MS/MS analytical method. A range of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio (0.205-0.269) found in surface sediments off Japanese coast after the accident was comparable to those ranges of 0.201-0.249 observed in Japanese estuaries and 0.150-0.281 in the western North Pacific and its marginal seas before the FDNPP accident, indicating that Pu contamination from the accident are negligible in the coast sediments.

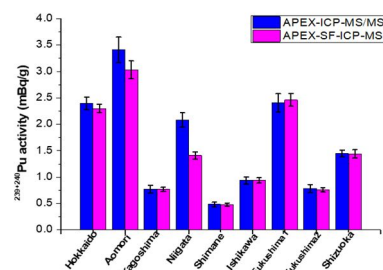


Fig. 5 Comparison of  $^{239+240}\text{Pu}$  activities measured by ICP-MS/MS and SF-ICP-MS.

##### Determination of Pu isotopes in simulated nuclear decommissioning samples: concrete and steel

The applicability of the developed APEX-ICP-MS/MS analytical system for the determination of Pu isotopes in nuclear decommissioning samples was demonstrated by the analysis of concrete and steel samples. For concrete, samples were pulverized to about 80 mesh, then a sample of about 1 g was weighed and put into a Teflon vessel (120 mL). After adding 28 fg  $^{239}\text{Pu}$ , the concrete sample was ashed at  $1000\text{ }^\circ\text{C}$  for 4 hours in a muffle furnace for 4 h. After cooling, 0.57 pg  $^{242}\text{Pu}$  tracer was added. Then, the developed low-temperature fusion was employed to digest the sample and followed by extraction chromatographic separation to prepare sample for APEX-ICP-MS/MS measurement. High recovery of 83-87% of  $^{242}\text{Pu}$  and high dissolution rate of added  $^{239}\text{Pu}$  (97-98%) were obtained. For steel samples, we developed a rapid acidic digestion method using  $\text{HCl-HNO}_3\text{-NH}_4\text{HF}_2$ . Steel sample (1g) spiked with 0.57 pg  $^{242}\text{Pu}$  tracer was digested for 40 min in a hot plate, then followed by  $\text{LaF}_3/\text{CaF}_2$  co-precipitation-extraction chromatographic separation prior to APEX-ICP-MS/MS measurement. A satisfactory Pu recovery of  $78 \pm 3\%$  was obtained. These results demonstrated the reliability of the developed APEX-ICP-MS/MS based analytical method for rapid analysis of trace level Pu isotopes in nuclear decommissioning samples.

## 5. 主な発表論文等

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3. 雑誌名 Microchemical Journal	6. 最初と最後の頁 597 ~ 604
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2. 論文標題 First determination of Pu isotopes (239Pu, 240Pu and 241Pu) in radioactive particles derived from Fukushima Daiichi Nuclear Power Plant accident	5. 発行年 2019年
3. 雑誌名 Scientific Reports	6. 最初と最後の頁 9:11807
掲載論文のDOI (デジタルオブジェクト識別子) 10.1038/s41598-019-48210-4	査読の有無 有
オープンアクセス オープンアクセスとしている (また、その予定である)	国際共著 該当する

1. 著者名 Ni Youyi, Guo Qiuju, Huang Zhaoya, Zheng Jian, Li Sixuan, Huang Wenna, Bu Wenting	4. 巻 253
2. 論文標題 First study of <sup>237</sup> Np in Chinese soils: Source, distribution and mobility in comparison with plutonium isotopes	5. 発行年 2020年
3. 雑誌名 Chemosphere	6. 最初と最後の頁 126683 ~ 126683
掲載論文のDOI (デジタルオブジェクト識別子) 10.1016/j.chemosphere.2020.126683	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1. 著者名 Junwen Wu, Jian Zheng	4. 巻 324
2. 論文標題 Reference materials fro quality assurance of environmental plutonium analysis	5. 発行年 2020年
3. 雑誌名 Journal of Radioanalytical and Nuclear Chemistry	6. 最初と最後の頁 169-188
掲載論文のDOI (デジタルオブジェクト識別子) 10.1007/s10967-020-07053-4	査読の有無 有
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1. 著者名 Jinlong Wang, Jinzhou Du, Jian Zheng	4. 巻 20
2. 論文標題 Plutonium isotopes research in the marine environment: A synthesis	5. 発行年 2020年
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掲載論文のDOI (デジタルオブジェクト識別子) なし	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1. 著者名 Ni Youyi, Wang Zhongtang, Guo Qiuju, Zheng Jian, Li Sixuan, Lin Jinxian, Tan Zhaoyi, Huang Wenna	4. 巻 212
2. 論文標題 Distinctive distributions and migrations of <sup>239+240</sup> Pu and <sup>241</sup> Am in Chinese forest, grassland and desert soils	5. 発行年 2018年
3. 雑誌名 Chemosphere	6. 最初と最後の頁 1002 ~ 1009
掲載論文のDOI (デジタルオブジェクト識別子) 10.1016/j.chemosphere.2018.09.021	査読の有無 有
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1. 著者名 Men Wu, Zheng Jian, Wang Hai, Ni Youyi, Kumamoto Yuichiro, Yamada Masatoshi, Uchida Shigeo	4. 巻 246
2. 論文標題 Pu isotopes in the seawater off Fukushima Daiichi Nuclear Power Plant site within two months after the severe nuclear accident	5. 発行年 2019年
3. 雑誌名 Environmental Pollution	6. 最初と最後の頁 303 ~ 310
掲載論文のDOI (デジタルオブジェクト識別子) 10.1016/j.envpol.2018.12.007	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1. 著者名 Wang Hai, Ni Youyi, Zheng Jian, Huang Zhaoya, Xiao Detao, Aono Tatsuo	4. 巻 1050
2. 論文標題 Low-temperature fusion using NH4HSO4 and NH4HF2 for rapid determination of Pu in soil and sediment samples	5. 発行年 2019年
3. 雑誌名 Analytica Chimica Acta	6. 最初と最後の頁 71 ~ 79
掲載論文のDOI (デジタルオブジェクト識別子) 10.1016/j.aca.2018.10.065	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1. 著者名 Wu Junwen, Dai Minhan, Xu Yi, Zheng Jian	4. 巻 511
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3. 雑誌名 Chemical Geology	6. 最初と最後の頁 256 ~ 264
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オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1. 著者名 Bu Wenting, Tang Lei, Liu Xuemei, Wang Zhongtang, Fukuda Miho, Zheng Jian, Aono Tatsuo, Hu Sheng, Wang Xiaolin	4. 巻 34
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1. 著者名 Ni Youyi, Wang Zhongtang, Zheng Jian, Tagami Keiko, Guo Qiuju, Uchida Shigeo, Tsukada Hirofumi	4. 巻 196
2. 論文標題 The transfer of fallout plutonium from paddy soil to rice: A field study in Japan	5. 発行年 2019年
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1. 著者名 Wang Z. T., Zheng J., Imanaka T., Uchida S.	4. 巻 32
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オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1. 著者名 Bu Wenting, Zheng Jian, Ketterer Michael E., Hu Sheng, Uchida Shigeo, Wang Xiaolin	4. 巻 995
2. 論文標題 Development and application of mass spectrometric techniques for ultra-trace determination of <sup>236</sup> U in environmental samples-A review	5. 発行年 2017年
3. 雑誌名 Analytica Chimica Acta	6. 最初と最後の頁 1 ~ 20
掲載論文のDOI (デジタルオブジェクト識別子) 10.1016/j.aca.2017.09.029	査読の有無 有
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1. 著者名 Bu Wenting, Zheng Jian, Guo Qiuju, Xiao Detao, Aono Tatsuo	4. 巻 314
2. 論文標題 Determination of Pu isotopes in sediment and soil samples by SF-ICP-MS: an improved anion-exchange procedure for Pu separation	5. 発行年 2017年
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掲載論文のDOI (デジタルオブジェクト識別子) 10.1007/s10967-017-5618-y	査読の有無 有
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1. 著者名 Ni Youyi, Zheng Jian, Guo Qiuju, Men Wu, Tagami Keiko, Uchida Shigeo	4. 巻 1000
2. 論文標題 Rapid determination of ultra-trace plutonium isotopes ( $^{239}\text{Pu}$ , $^{240}\text{Pu}$ and $^{241}\text{Pu}$ ) in small-volume human urine bioassay using sector-field inductively coupled plasma mass spectrometry	5. 発行年 2018年
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1. 発表者名 Jian Zheng
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2. 発表標題 Simultaneous determination of ultra-trace level <sup>237</sup> Np and Pu isotopes in soil/sediment samples by sector-field ICP-MS with a single-column chromatographic separation
3. 学会等名 日本分析化学会第67年会, 日本分析化学会
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1. 発表者名 福田 美保, 鄭 建, 山崎 慎之介, 青野 辰雄
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1. 発表者名 Jian Zheng
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3. 学会等名 Fukushima University, 4th Annual Report Symposium (招待講演)
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1. 発表者名 Jian Zheng, Zhongtang Wang, Youyi Ni, Keiko Tagami, Shigeo Uchida
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4. 発表年 2017年

1. 発表者名 Jian Zheng
2. 発表標題 Tracing the Fukushima nuclear accident released radiocesium and Pu isotopes in the environment: the role of isotope ratio analysis for nuclear forensics.
3. 学会等名 Nuclear Emergency Expert Meeting (NEXT 2017), Hannover, Germany (招待講演) (国際学会)
4. 発表年 2017年

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3. 学会等名 Actinides 2017, Sendai, Japan (国際学会)
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1. 著者名 Youyi Ni, Jian Zheng, Qiuju Guo, Hai Wan	4. 発行年 2019年
2. 出版社 Nature Springer	5. 総ページ数 236
3. 書名 Chapter, 1.The Key Role of Isotopic Analysis in Tracing the Fukushima Nuclear Accident-Released Pu and Radiocesium Isotopes in the Environment. In Nuclear Emergencies. Current Topics in Environmental Health and Preventive Medicine	

〔産業財産権〕

〔その他〕

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6. 研究組織

	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考
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