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ARTICLE

Analysis of Uranium, Plutonium and Fission Product Nuclides in Process Solution during Flush-Out for Decommissioning of Reprocessing Plant

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Tokai Reprocessing Plant (TRP), Japan's first spent nuclear fuel reprocessing plant, is in decommissioning stage. Flush-out of TRP were performed and completed in February 2024 for preparation of post operation clean out. Since remaining nuclear materials were contained in main process of the facility, purpose of flush-out was to recover nuclear materials by transferring them to high level liquid waste storage tank and converting uranium (U) solution into uranium trioxide (UO_3). Then, all related processes were rinsed with nitric acid and deionized water. Analysis of U and plutonium (Pu) in process solution was performed by isotope dilution mass spectrometry (IDMS) and gravimetric method. It was found that U and Pu in process samples during flush-out were accurately and precisely determined with uncertainties below 0.13% (k=2). Rinsing solutions from related processes were determined either by IDMS, spectrophotometry and alpha-ray counting method depending on the concentrations in samples. Consequently, target values of flush-out, such as 1 g/L for U and 10 mg/L for Pu, were achieved.

KEYWORDS: Uranium, Plutonium, analysis, flush-out, reprocessing, decommissioning

I. Introduction

Tokai Reprocessing Plant (TRP), Japan's first spent nuclear fuel (SNF) reprocessing plant, began operations in 1977.[1)] It utilized the Plutonium Uranium Reduction EXtraction (PUREX) process, which employed solvent extraction with 30% tributyl phosphate in *n*-dodecane, to separate uranium (U) and plutonium (Pu) from fission products (FP). The TRP had a maximum reprocessing capacity of 0.7 tU per day and played a leading role in establishing reprocessing technologies in Japan. During the extraction process, U and Pu were recovered as U nitrate (UN) solution and Pu nitrate (PuN) solution, then converted to uranium trioxide (UO_3) and U-Pu mixed oxide (MOX), respectively.

In 2007, TRP suspended reprocessing operations to begin construction work aimed at improving earthquake resistance, in accordance with regulatory standards following the Niigata-ken Chuetsu-oki Earthquake. Although there were plans to resume operations, TRP shifted to decommissioning stage due to the post-Fukushima new safety regulations implemented after the Great East Japan Earthquake of 2011, considering cost effectiveness.[2)] Nuclear materials such as sheared powder, PuN solution and UN solution, were remained in the main process of TRP, because operations were halted abruptly before clean-up of nuclear materials. Consequently, the first stage of TRP decommissioning required flush-out involving the recovery of remaining nuclear materials by transferring them to the storage tank of high level liquid waste (HLLW) and rinsing all related processes with nitric acid (HNO_3) solution and deionized water (DWA).

Given the significant amounts of U and Pu in TRP, analysis work during flush-out was crucial for nuclear material control. Appropriate analytical techniques needed to be selected for samples, as concentrations of U and Pu would vary depending on status of flush-out and rinsing after each step. This report provides an overview of analysis work related to the flush-out and discusses radioactivity of FP nuclides in process solution after rinsing, which is essential for post operation clean out of TRP. As this is the first decommissioning of large-scale nuclear fuel facility in Japan, analytical data obtained is extremely valuable for future progress.

II. Outline of Flush-out

Flush-out at TRP involved transferring nuclear materials to the HLLW storage tank. **Figure 1** illustrates flow diagram and sampling points during flush-out. Three types of nuclear materials, such as sheared powder, PuN solution, and UN solution, remained in the process. Sheared powder, which was generated at loads of SNF into shearing machine and distributor during the past reprocessing operation, was dissolved in heated HNO_3 solution at the dissolver tank. Dissolved powder was sampled at the input accountability tank (IAT) for analysis, then transferred to HLLW. Hence TRP completed conversion of PuN solution into MOX powder, residual PuN liquid was diluted and stored in seven storage tanks. Those were collected, and analytical samples were taken at the rework process (RWP). Then, it was mixed with UN solution and transferred to HLLW by steam-jet. Considering a formation of Pu polymer by contact with water at the steam-jet transfer, U was mixed with Pu samples according to the procedure in past reprocessing operation. Remaining UN solution was de-nitrated and converted to UO_3

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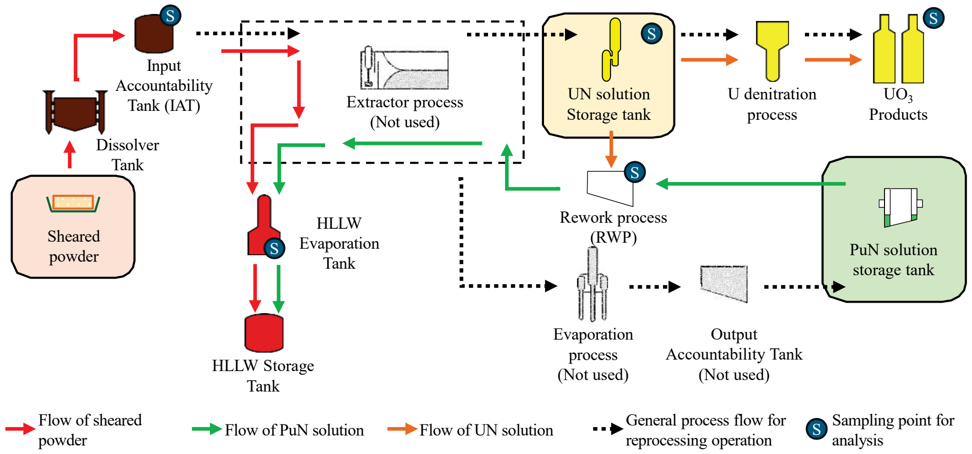

Fig. 1 Flow diagram of flush-out in TRP decommissioning. Sheared powder was dissolved in heated HNO_3 and then transferred to HLLW storage tank. The PuN solution in seven storage tanks was collected, mixed with UN solution, and then transferred to HLLW storage tank. The UN solution in storage tank was de-nitrated and converted to UO_3 powder.

powder at de-nitrification (DN) facility. Both samples from UN solution and UO_3 powder were taken at the storage tank and at DN facility during U conversion process. All related processes were rinsed with HNO_3 and DWA after each step until concentrations reached to the target values for flush-out, which were 1 g/L for U and 10 mg/L for Pu, from the past achievement of flush-out during normal TRP reprocessing operation.

III. Experimental

1. Analysis of Sheared Powder

Sheared powder was collected around shearing machine and distributor in TRP. Ten batches of approximately 30 kg sheared powder were dissolved in heated HNO_3 for about 8 hours. Amounts of sheared powder loaded into the dissolver tank were limited to prevent unexpected criticality occurrence of nuclear materials. Analytical sample solution of sheared powder was taken at IAT described in Fig.1. The solution in IAT was mixed by using airlift pump before sampling. Concentrations of U and Pu in sample were determined by isotope dilution mass spectrometry (IDMS) using thermal ionization mass spectrometer (TIMS) of TRITON™ from Thermo Fisher Scientific. A 1 mL of sample was mixed with dried tracer spike containing approximately 1 mg of ^{233}U (99% enriched) and 0.05 mg of ^{242}Pu (96% enriched) per vial. Prior to mass measurement, separation of FP was performed inside hot cell with manipulator operation to reduce radiation exposure. Valence of Pu in sheared powder sample was adjusted to Pu(IV) by adding aqueous mixture of 2.8% $FeSO_4\cdot 7H_2O$ and 5% sulfamic acid with aqueous solution of 7% $NaNO_2$. The FP nuclides in the sample were removed using anion-exchange resin, purchased from Bio-Rad Laboratories. Then, sample was transferred to glove-box and Pu was further purified by solid-phase extraction resin of TEVA®, purchased from Eichrom Technologies. The collected sample solution was measured by TIMS. Uncertainties of the measurements were evaluated according to the ISO-GUM standard.[3]

2. Analysis of PuN Solution

Describing in Fig. 1, PuN solutions in seven storage tanks were collected in tank at RWP, where the sample solution was taken. The solution in tank was mixed by using airlift pump. For IDMS of PuN solution, two types of liquid spikes with ^{239}Pu (98% enriched), naming liquid large-size (LS) spikes, were used. A liquid LS spike containing Pu with 2 mg per vial was used to analyze above 1 g/L of PuN solution. The other liquid LS spike containing Pu with 0.2 mg per vial was used to analyze the lower concentration of PuN solution. The Pu in PuN sample solution was also purified by TEVA® before mass measurement. Uncertainties of measurements were evaluated according to the ISO-GUM standard.[3]

3. Analysis of UN Solution and UO_3 Powder

Analytical samples were taken from UN solution storage tank and UO_3 products. The U in UN solution and UO_3 powders were measured by gravimetric method. Sample weights were measured by electric mass balance of XP204 from Mettler Toledo and samples were ignited by electric muffle furnace of Y-2025-P, Yamada Denki with crucibles made from Inconel®. Crucibles including their lids were treated to heat several times in an electric muffle furnace before the usage for being no weight change during the ignition. Ignition was set at 900°C in air for 60 minutes. The U contents were evaluated from the difference between masses before and after ignition according to literatures.[4,5] Uncertainties of measurements were evaluated according to the ISO-GUM standard.[3]

4. Analysis of Rinsing Solution

The U and Pu in several rinsing solution, which were below quantification limit of IDMS and gravimetric method, were measured by spectrophotometry with spectrophotometers of UV-2450 from Shimadzu and V-550 from JASCO. Dibenzoyl methane (DBM) solution was prepared by mixing DBM (4.5×10^{-3} mol/L), $Mg(NO_3)\cdot 6H_2O$ (4.3×10^{-2} mol/L), ethylenediamine-N,N,N',N'-tetraacetic acid tetrasodium salt tetrahydrate (3.4×10^{-2} mol/L) in 500 mL of pyridine with 500 mL of pure water. The prepared solution was used as chromogenic reagents of U determination with absorption peak at 410 nm. Prior to measurement, U was extracted by ethyl acetate to separate interfering species. The Pu in rinsing solution was directly determined by spectrophotometry at 830 nm with following Pu(VI) valence adjustment by $(NH_4)_2[Ce(NO_3)_6]$. The HNO_3 concentration in sample was adjusted to 3 mol/L. The Pu concentrations below 10 mg/L were determined by an alpha-ray counting method after Pu extraction using 2-thenoyltrifluoroacetone (TTA) in xylene (0.5 mol/L). Extracted Pu was dried onto stainless-steel planchet and alpha-ray counts were detected by ZnS(Ag) scintillation counter with scaler (TDC-451F and TDC-511, Aloka). Errors in analytical results of rinsing solution were estimated from propagation of system and random error. System error was calculated from difference between the measured and prepared values during standard measurements. Random error was calculated from the standard deviation during repeated sample measurements. For gamma-ray spectroscopy of FP nuclides measurements, highly purified germanium detector (HPGe) detector and multi-channel analyzer of GC2020 from CANBERRA were used.

IV. Results and Discussion

1. Analysis of Sheared Powder Dissolution

It was evident that chemical composition in sheared powder was very similar to those of SNF because both were originated from the spent nuclear fuel. However, U and Pu concentration in sheared powder was estimated to be 20-30 g/L and 200-300 mg/L, respectively, lower than those in SNF due to the smaller quantities of loads into the dissolver.

The IDMS is known as one of the most reliable analytical techniques and often applied for determination of dissolved solution of SNF. Since chemical components of sheared powder were similar to those of SNF, IDMS was applied for determining dissolved solution of sheared powder. It is important that an optimal spike-sample ratio is required for IDMS to achieve the best accuracy. As concentrations of U and Pu in sheared powder sample were lower than those in SNF, spike-sample ratio was evaluated and optimized for analysis of sheared powder by minimizing the error multiplication factor (EMF) following in Eq.(1), according to the reported literature.[6)]

$$EMF = \frac{(R_S - R_T)\cdot R_{Mix}}{(R_{Mix} - R_S)\cdot(R_T - R_{Mix})} \tag{1}$$

$\boldsymbol{EMF}$	Error multiplication factor
$\boldsymbol{R_S}$	Ratio of isotope in sample
$\boldsymbol{R_T}$	Ratio of isotope in spike
$\boldsymbol{R_{Mix}}$	Ratio of isotope in mixture

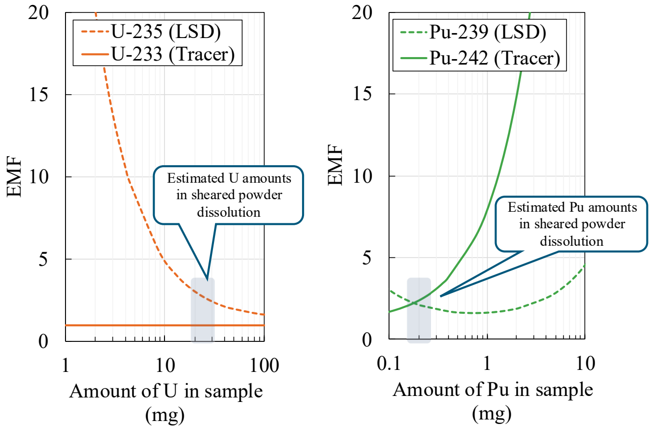

Fig. 2 Calculated EMF of dissolved solution of sheared powder for LSD spike and dried tracer spike. The filled area in graph indicated the estimated U and Pu amounts in sheared powder sample with solution volume of 1 mL.

For selecting appropriate spikes, EMF of sheared powder for two types of spikes were calculated. One was the large-size dried (LSD) spike, containing approximately 40 mg of U with 20% enrichment of ^{235}U and approximately 2 mg of Pu with 98% abundance of ^{239}Pu per vial.[7,8)] This spike was often used for analyzing dissolved solution of SNF. The other was dried tracer spike, containing approximately 1 mg of U with 99% of ^{233}U and approximately 0.05 mg of Pu with 96% of ^{242}Pu per vial. This was often used for analyzing samples from HLLW.[9)] **Figure 2** indicated the calculated EMF of dissolved solution of sheared powder for LSD spike and dried tracer spike. In U measurement, EMF values of dried tracer spike were stable and lower than those of the LSD spike for ranges in estimated U sample amount. On the other hand, EMF values in LSD spike and dried tracer spike were almost same for Pu sample amount. The EMF theoretically described by what factor the deviation of isotope ratio affected quantification results and minimizing EMF value minimized errors in quantification by IDMS. It was found that the dried tracer spike could provide less analytical errors for both U and

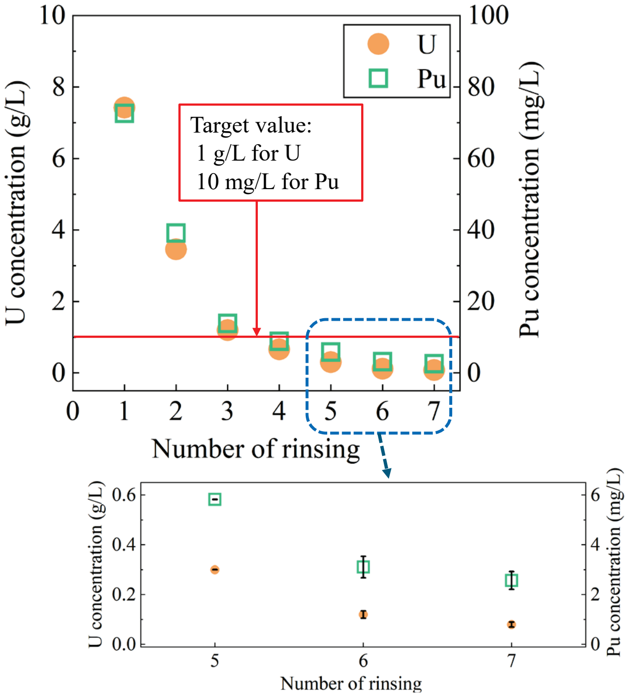

Fig. 3 Analytical results of rinsing solution in sheared powder process. The red line indicated the target values of flush-out. The last 2 sample batches were analyzed by spectrophotometry for U and alpha-ray counting method for Pu. The others were analyzed by IDMS using dried tracer spikes.

Pu, compared to LSD spike. Therefore, dried tracer spike was selected for IDMS analysis of sheared powder.

Measured U in 10 batches of dissolved solution of sheared powder were from 6.9051 ± 0.0090 g/L to 29.989 ± 0.039 g/L (*k*=2), and Pu in those were from 172.50 ± 0.19 to 327.60 ± 0.36 mg/L (*k*=2), respectively. Uncertainties of measurements were below 0.13% (*k*=2). It was confirmed that U and Pu in sheared powder solution was about 1/10 of those in SNF and successfully recovered by transferring to the HLLW storage tank.

After that, the process was rinsed with HNO_3 and DWA. Analytical results of rinsing solution were indicated in **Fig. 3**. The IDMS with dried tracer spike was applied except for last 2 bathes of rinsing solution. Hence U and Pu in these samples were expected to be lower than quantification limit of IDMS, the last 2 batches were determined by spectrophotometry for U and alpha-ray counting method for Pu. As a result, all samples from rinsing solution were successfully measured. It was found that nuclear materials in related processes of sheared powder decreased as increased rinsing number. Consequently, it reached the target value of flush-out.

2. Analysis of PuN Solution

Highly concentrated PuN solution, about 200-300 g/L, was normally treated during TRP reprocessing operation and IDMS was applied for Pu determination. The PuN solution during reprocessing operation was diluted to 2-3 g/L, then mixed with LS spikes, which contained 2 mg of Pu with 98% of ^{239}Pu.[10] Since Pu concentration in PuN solution during flush-out was estimated to be about 3 g/L, similar to that of diluted Pu solution in normal TRP reprocessing operation, liquid type of LS spike was directly mixed.

Analytical results of PuN solution in flush-out by IDMS using liquid type LS spikes were shown in **Table 1**. The Pu concentrations in PuN solutions were about 3-4 g/L, as expected. Uncertainties of measured values were below 0.11% with *k*=2, which satisfied international target value (ITV), that can be achieved in routine analysis of nuclear materials.[11] It was found that Pu in PuN solution were accurately and precisely determined by IDMS, and successfully transferred to the HLLW storage tank.

Table 1 Analytical results of PuN solution in flush-out by IDMS using liquid typed LS spikes. Uncertainties of measurements were evaluated according to ISO-GUM standard.[3]

Sample	Element	Measured value (g/L, *k*=2)
No.1	Pu	2.9857±0.0032
No.2	Pu	3.5626±0.0038
No.3	Pu	3.5500±0.0038

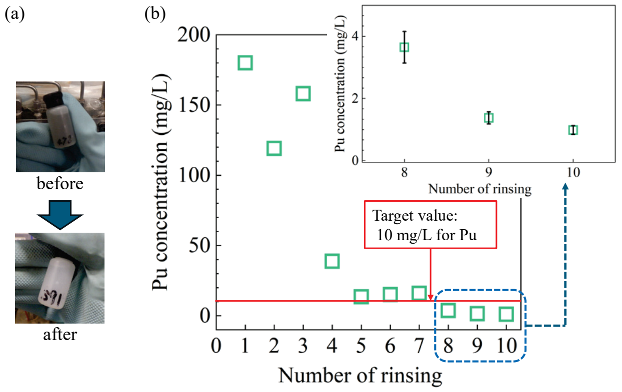

Fig. 4 Pictures of samples and analytical results of rinsing solution. (a) Pictures of samples taken from PuN solution process with white colored polyethylene vial before and after rinsing. (b) The Pu concentration trend of rinsing solution in PuN solution process. Red line in the graph indicated the target values (10 mg/L for Pu) of flush-out. The first 2 batches were determined by IDMS using LS spikes, next 5 batches were determined by spectrophotometry and last 3 batches were determined by alpha-ray counting method.

After the Pu recovery, related processes of PuN solution were rinsed with HNO_3 and DWA. Since Pu concentration was expected to decrease drastically by rinsing, spikes including less Pu amounts were used for IDMS of several rinsing solutions. According to the Eq.(1), Pu amounts in this spike were set as 0.2 mg and optimized to samples containing 100 mg/L to 1 g/L of Pu. **Figure 4** indicated pictures and analytical results of Pu in rinsing solution. As shown in Fig. 4 (a), colors of PuN solution in analytical samples disappeared by rinsing. It was found in Fig. 4 (b) that Pu in rinsing solution decreased by increasing number of rinsing. The first 2 batches of rinsing solution in Fig. 4 (b) were analyzed by IDMS. Samples with Pu lower than 100 mg/L was determined by Pu(VI) spectrophotometry. Although Pu in TRP contained Am due to the decay from ^{241}Pu and its peak was observed at 811 nm, spectral interference was not observed for Pu(VI) determination at 830 nm in PuN rinsing solution. The Pu in last 3 batches in Fig. 4 (b) was lower than 10 mg/L and difficult to determine by spectrophotometry, thus they were determined by alpha-ray counting method. It was shown that Pu in final rinsing bath decreased to be about 1 mg/L and reached the target value of flush-out. The increase of Pu concentration in 3rd batch of rinsing solution was due to the rinsing procedure at flush-out. The other process solution was simultaneously added into the RWP tank and caused the Pu increase in 3rd batch of rinsing sample.

The Pu decontamination factor (DF) was also calculated from analytical results of final rinsing batch and initial PuN solution. The DF value of flush-out in PuN solution process was about 4×10^3. It was found that high DF value for PuN solution process was obtained by rinsing only HNO_3 and DWA.

3. Analysis of UN Solution and UO_3 Powder

In the normal reprocessing operation of TRP, U was recovered as highly concentrated UN solution, about 300-400 g/L, and converted to UO_3 powder at DN facility. During flush-out, the same conversion procedure from UN solution to UO_3 powder was performed. The U products in TRP such as UN solution and UO_3 were highly purified chemical compounds. Thus, gravimetric method, which was used extensively throughout the nuclear industry to measure nearly pure U samples, was applied to determine U.[4,5,12] This method utilizes the chemical reaction of UN solution and UO_3 to U_3O_8 with ignition as following equation (2) to (4). The method provides excellent trueness for U analysis because it relies on fundamental quantity of mass.

$$UO_2(NO_3)_2{\cdot}6H_2O \rightarrow UO_2(NO_3)_2 + 6H_2O \tag{2}$$

$$UO_2(NO_3)_2 \rightarrow UO_3 + 2NO_2 + 1/2O_2 \tag{3}$$

$$3UO_3 \rightarrow U_3O_8 + 1/2O_2 \tag{4}$$

The UN solution in 9 storage tanks of TRP was de-nitrated and converted to 14 batches of UO_3 powder during flush-out. Analytical results of U in UN solution were from 155.54 ± 0.10 g/L to 377.39 ± 0.13 g/L (k=2), and U in UO_3 were from 0.82197 ± 0.00021 g/g to 0.82742 ± 0.00020 g/g (k=2). It was found that U in UN solution and UO_3 powder were accurately and precisely determined by gravimetric method within uncertainties below 0.10% (k=2), which satisfied ITV value.[11] Purification of UO_3, calculated from analytical results, were above 99%, indicating that UN solution in the process was successfully converted to highly purified UO_3. After the de-nitration and conversion, related processes were also rinsed with DWA. The rinsing solution of U process was transferred to low radioactive liquid waste treatment facility in TRP. Since U contents in rinsing solution of the process were drastically decreased, U was determined by spectrophotometry. It was found that U contents in all related process reached the U target value of 1 g/L by only 2-3 times of DWA rinsing.

4. Gamma-Ray Nuclides before and after Rinsing

Analytical samples were also taken from the HLLW evaporation tank, where rinsing solutions from sheared powder dissolution process and PuN solution process were transferred. Therefore, FP nuclides in sample before and after rinsing were measured for post operation clean out. A 1 mL from each sample was collected after appropriate dilution to ensure that dead-time of detector was less than 10%, then measured by HPGe for 10000 s. Figure 5 shows gamma-ray spectra of sample before and after rinsing during flush-out, and contributions of nuclides to total gamma radioactivity in final rinsing solution.

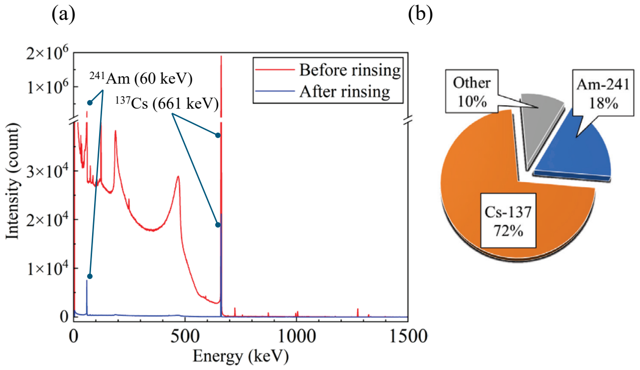

Fig. 5 Gamma-ray spectra of sample at HLLW evaporation tank and contribution of nuclides to total gamma radioactivity. (a) Gamma-ray spectra of sample before and after rinsing. (b) The contribution of nuclides to total gamma radioactivity in final rinsing batch sample

It was found from **Fig. 5 (a)** that significant gamma-ray peaks from ^{137}Cs and ^{241}Am were detected in sample taken from HLLW evaporation tank. Gamma-ray activities were $3{\times}10^7$ Bq/mL for ^{137}Cs and $3{\times}10^6$ Bq/mL for ^{241}Am. Even after rising at flush-out, ^{137}Cs and ^{241}Am were detected and those were $4{\times}10^4$ Bq/mL and $9{\times}10^3$ Bq/mL, respectively. Although TRP operation was stopped about 15 years ago, ^{137}Cs was still main gamma-ray source as shown in **Fig. 5 (b)**, and ^{241}Am radioactivity was dominant because ^{241}Pu decayed in 14-year half-life. The DF values before and after rinsing were $8{\times}10^2$ for ^{137}Cs and $3{\times}10^2$ for ^{241}Am, respectively. These were similar to DF of Pu shown in section “IV. 2”. It was considered from gamma-ray measurement results that decommissioning of TRP would require careful management of these FP nuclides.

V. Conclusion

Although more than 15 years had passed since previous TRP operations, flush-out, which were to recover nuclear materials from main process by transferring dissolved solution of sheared powder and PuN solution to the HLLW storage tank was performed. The UN solutions in storage tanks were also converted to UO_3, then all related processes were rinsed with HNO_3 and DWA. It was successfully completed in February 2024. The U and Pu in sheared powder and PuN solution were determined by IDMS with uncertainties below 0.13% (k=2). The U in UN solution and UO_3 powder during de-nitration were determined by gravimetric method with uncertainties below 0.10% (k=2). It was concluded that U and Pu in process samples during flush-out were accurately and precisely determined by appropriate analytical techniques. Rinsing solutions from related processes were determined either by IDMS, spectrophotometry, and alpha-ray counting method depending on concentrations of samples. Consequently, target values of flush-out, such as 1 g/L for U and 10 mg/L for Pu, were achieved. As a result, U and Pu concentration in all related process solution of flush-out could be promptly provided and feedbacked as requested.

In addition to those, gamma-ray emitted FP nuclides in rinsing solution were measured by HPGe detector. The ^{137}Cs and ^{241}Am were detected and found as dominant even after the flush-out. Also, DF values of Pu, ^{137}Cs, ^{241}Am by rinsing were found to be 10^2-10^3, indicating those were rinsed only by HNO_3 and DWA.

The TRP decommissioning project is planned to continue for approximately 70 years until the release from radioactive controlled area. The data obtained in flush-out will be reflected as post operation clean out of TRP.

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