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The liquid waste treatment process generated sludge items at the plutonium conversion development facility. They are highly heterogeneous and contain large amounts of impurities (Na, Fe, Ni etc.). Therefore, the sludge items have very large sampling uncertainty and so the total measurement uncertainty is very large (approximately 24%). The plutonium scrap multiplicity counter (PSMC) measurement technique for sludge items was developed by joint research between the Japan Atomic Energy Agency (JAEA) and Los Alamos National Laboratory (LANL). The technical validity for sludge items using the PSMC was evaluated using various types of sample measurements and Monte Carlo N-Particle transport code calculations. The PSMC measurement parameters were found to be valid for use with sludge items and the validity of multiplicity analysis was confirmed and demonstrated through comparisons with standard MOX powder and a standard sludge. As a result, the PSMC measurement values were shown to be consistent and reasonable and the large amount of impurity (Fe, Ni etc.) did not impact the results. Therefore, the measurement uncertainty of the improved nuclear material accountancy (NMA) procedure by combined PSMC and high-resolution gamma spectrometry was shown to be 6.5%. In addition, an acceptance test was conducted using PSMC/HRGS and IAEA benchmark equipment. Measured Pu mass by both equipment agrees within the measurement uncertainty of each method, and so the validity for Pu mass measurement by PSMC/HRGS was confirmed.

The above results confirm the applicability of PSMC/HRGS as an additional NMA method for sludge and a newly designed NDA procedure based on this study is applied to sludge for NMA in PCDF.

Keywords: Sludge, Impurity, Neutron, Non-distractive Assay, Plutonium Scrap Multiplicity Counter

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This work has been conducted in the Japan Atomic Energy Agency as joint research with the United States Department of Energy.

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非破壊測定装置(PSMC)を用いたスラッジ測定に係る評価報告
(共同研究)

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(2024年7月10日受理)

プルトニウム転換技術開発施設の廃液処理工程で発生したスラッジ中の Pu 量を評価する上で、スラッジは生成上、多くの不純物(Na, Fe, Ni 等)を含み不均質であるため、従来の破壊分析ではサンプリングによる代表性が乏しく、Pu 量測定に係る測定不確かさが大きかった(約 24%)。この測定不確かさを低減させるために、日本原子力研究開発機構と米国ロスアラモス国立研究所は共同で中性子を利用した非破壊測定装置の Plutonium Scrap Multiplicity Counter(PSMC)を用いた測定技術の開発を進めた。MOX 粉末を用いた模擬スラッジやモンテカルロ法により検証等を行い PSMC 検出器パラメーター(predelay, doubles gate fraction 等)を最適化し、高分解能ガンマ線分光分析を組み合わせて測定した結果、含有不純物の影響はないことが確認でき、従来法と比べ新たに設定した非破壊測定方法における測定不確かさは約 6.5%まで低減できた。さらに、PSMC 測定値の妥当性を評価するため、IAEA 立ち合いの下、ガンマ線を測定し Pu 量を評価する In Situ Object Counting System (ISOCS)を用いた比較測定した結果、ISOCS と PSMC の測定値は両方の測定不確かさの範囲内で一致したため、PSMC による測定値の妥当性が確認された。

これらの結果より、本非破壊測定技術はスラッジのように不純物を多く含み、サンプリングの代表性が乏しいアイテムに有効であり、スラッジの計量管理に適用することが認められた。

本研究は、米国エネルギー省との共同研究として日本原子力研究開発機構により実施したものである。

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Contents

目次

Table list

Figure list

1. Introduction

1.1 Background and purpose

The Plutonium Conversion Development Facility (PCDF) in the Tokai Reprocessing Plant (TRP) was constructed in 1982 and operated for the conversion of plutonium (Pu) nitrate solution/uranium (U) nitrate solution into mixed oxide (MOX) powder after the recovery of Pu and U from spent fuel and the separation of the fission products. The PCDF has produced 6.8 t of MOX powder since 1983 and is now in a decommissioning phase since June 2018.

To reduce the radioactivity in the waste solution from the MOX conversion process, the PCDF conducts a coagulation sedimentation process using reagents, including iron (Fe), after a neutralization process using sodium hydroxide (NaOH), producing sediment as a solid waste, referred to as sludge, containing various chemical components, including Pu, U, Am, stainless steel components, halogens, $NaNO_3$, residual NaOH, and moisture.

The nuclear material accountancy (NMA) of sludge is conducted based on the results of a conventional method (weighing, sampling, and destructive analysis (DA)) for Pu and U obtained after filtration, dry-up, and calcination. However, due to the inhomogeneity and large amount of impurities, taking representative samples is difficult, triggering the start of this project to improve the NMA procedure of sludge items at the PCDF.

Therefore, the applicability of nondestructive assay (NDA) using the plutonium scrap multiplicity counter (PSMC) was investigated to improve the NMA of Pu and U for sludge. A newly designed NDA procedure based on this study will be applied to sludge for NMA in the PCDF after an acceptance test by the International Atomic Energy Agency (IAEA).

This study was conducted jointly by Japan (Japan Atomic Energy Agency (JAEA)) and the U.S. (United States Department of Energy (DOE)).

1.2 Organization

This technology development was conducted by the JAEA and Los Alamos National Laboratory (LANL) jointly in Project Arrangement (PA NP-09) under the “Implementing Arrangement between the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT) and Department of Energy of the United States of America (DOE) concerning Cooperation in the Field of Nuclear Energy-Related Research and Development.”

2. Task outline and schedule

This project comprised four tasks shown below and was conducted with the support of the DOE based on the NP-09 contract between the DOE and JAEA since February 2017.

Task 1

DOE-JAEA to develop and test an improved NMA procedure to reduce the uncertainty in NDA measurements of high alpha sludge items at PCDF.

Task 2※

DOE-JAEA to test the impact of replacing the Cd liner with Sn using normal and list mode acquisition and investigate gamma pile-up behavior based on dose rate using the sludge items measured in Task 1 and MOX powder.

※A recent field trial of the alternative ^{3}He coincidence counter (High Level Neutron coincidence counter - Boron, HLNB) at the PCDF showed the expectation of optimizing the low-energy gamma-ray shielding for high-mass solution samples simultaneously to improve neutron detection performance.

Task 3

JAEA to conduct acceptance tests for IAEA with DOE attendance and support.

Task 4

DOE-JAEA to prepare the final report.

Shortly after this project started based on the schedule shown in Table 2.1 (Before), a radioactive contamination incident in the Oarai Research and Development Center, JAEA, occurred on June 6, 2017, due to a lack of proper management of legacy waste storage containers, including nuclear material. One of the countermeasures of this incident was applied to the storage management of sludge in the PCDF, resulting in the implementation of sludge stabilization by water washing and changing of the container from polyethylene bottles to metal cans to prevent pressurized gas generation in storage containers and to enhance safety for sludge storage conditions.

The measurement object of this project had changed to be “stabilized sludge” based on the above circumstances. The schedule is revised to Table 2.1 (After). The agreement between the JAEA and DOE regarding this revision was done in August 2018.

Table 2.1 Project Schedule

Activity			CY2017				CY2018				CY2019			
			1Q	2Q	3Q	4Q	1Q	2Q	3Q	4Q	1Q	2Q	3Q	4Q
Task 1	DOE-JAEA to develop and test an improved NMA procedure to reduce the uncertainty in NDA measurements of high alpha sludge items at PCDF Item :stabilized sludge items	Before	■	■	■	■	■	■						
		After	■	■	■	■	■	■	■	■	■	■	■	■
Task 2	DOE-JAEA to test the impact of replacing the Cd liner with Sn using normal and list mode acquisition, and to investigate gamma pile-up behavior based on dose rate using the sludge items measured in Task 1.3 and MOX powder.	Before		■										
		After								■	■			
Task 3	JAEA to conduct acceptance tests for IAEA with DOE attendance and support (for Task 1).	Before						■	■					
		After										■	■	
Task 4	DOE-JAEA to prepare the final report.	Before							■	■				
		After											■	■

3. The characteristics of sludge generated in the PCDF

3.1 The waste treatment process

The PCDF adopts the microwave direct heating denitration method (MH method) for the U-Pu co-conversion process to produce MOX powder [1]. The MOX powder is stored in a storage area in the same facility after the calcination and blending processes (Figure 3.1).

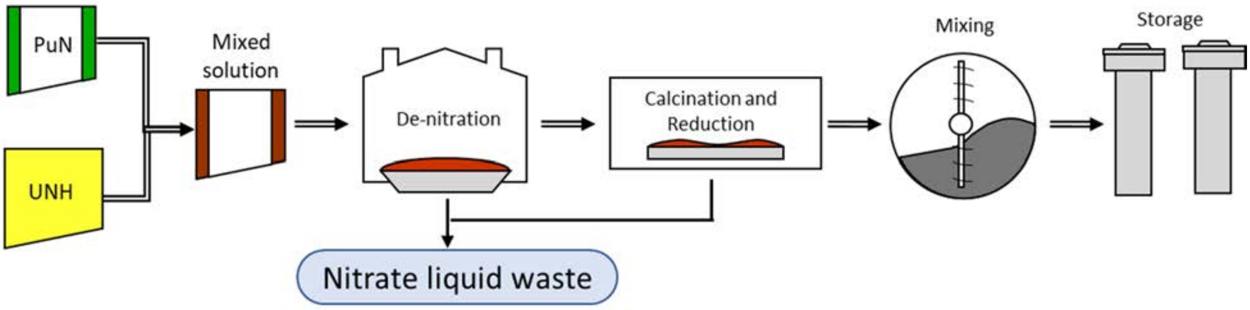


Figure 3.1 The MOX powder fabrication process in the PCDF

The waste solution is generated from the facility maintenance process (washing solution in the off-gas treatment process) and the steam condensate from the MH method in the PCDF. The evaporation and concentration of these waste solutions are performed in a waste evaporator to reduce the waste volume. To recover Pu and U from those waste solutions, two stages of the chemical process (neutralization sedimentation and coagulation sedimentation) are conducted, and the treated solution is transferred to TRP as a low-level radioactive waste solution (Figure 3.2).

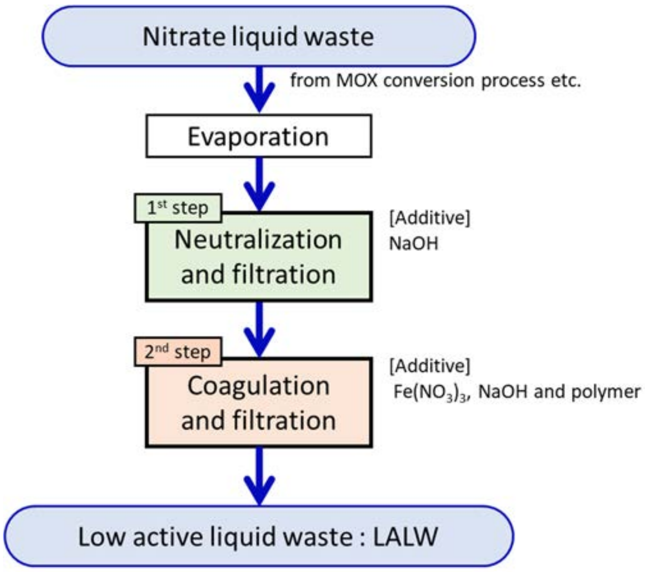


Figure 3.2 The complete process flow for the waste solution

3.1.1 The neutralization sedimentation process

The radioactive waste solution generated in the PCDF is transferred to the neutralizing sedimentation tank after the evaporation and concentration treatments. In this process, Pu and U in the waste solution are removed as sediment primarily comprising plutonium hydroxide (Pu $(OH)_4$) and sodium diuranate (SDU: $Na_2U_2O_7$) produced by the neutralization reaction.

$$\begin{aligned} &Pu\,(NO_3)_4 + 2UO_2(NO_3)_2 + HNO_3 + 6NaOH \\ &\quad \rightarrow \underline{Pu\,(OH)_4}\downarrow + \underline{Na_2U_2O_7}\downarrow + 7NaNO_3 + H_2O \end{aligned} \tag{3.1}$$

The sediment is filtered, recovered, and stored in 2-3 L polyethylene bottles as a calcinated product after drying (120°C, 5 h) and calcination (550°C, 5 h). The waste solution after filtration is transferred to the coagulation sedimentation process (Figure 3.3).

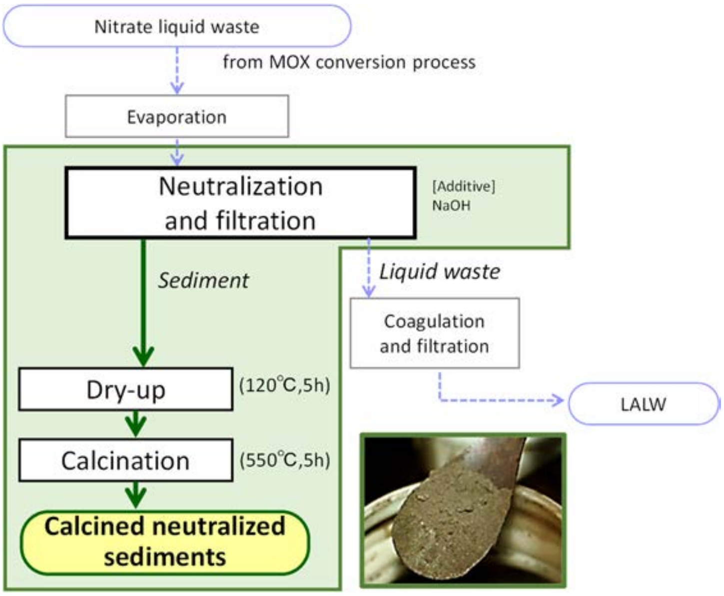


Figure 3.3 The neutralization sedimentation flow and product

3.1.2 The coagulation sedimentation process

The waste solution after the neutralization sedimentation process is adjusted in pH 9-11, then it is transferred to the coagulation sedimentation tank. The coagulation sedimentation treatment is conducted for these waste solutions by adding ferric nitrate and polymer flocculant. As a result, Pu and U accompanied with sediment are removed from the waste solution.

The produced sediments are filtered (natural drying, 120°C, 5 h) and calcinated (550°C, 5 h) and then stored in a polyethylene bottle (2-3 L) as calcined coagulation sediments (Figure 3.4). The radioactivity of the filtered waste solution is evaluated to confirm that it satisfies the standard value for transfer from the PCDF to the TRP waste treatment process.

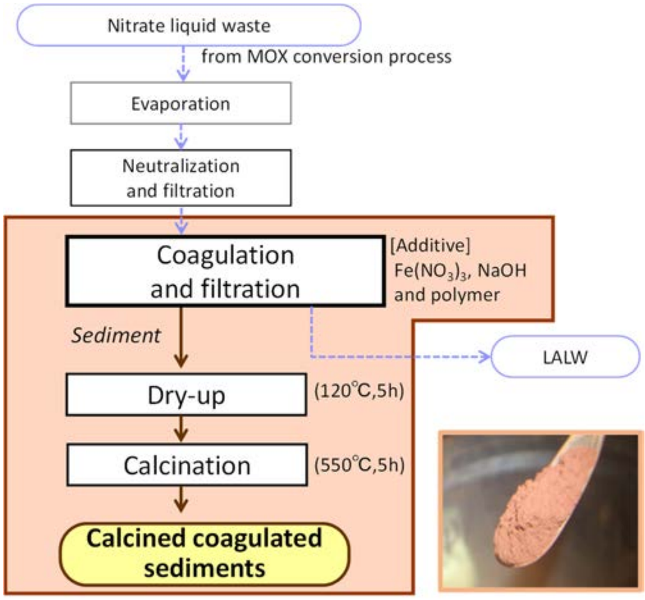


Figure 3.4 The process flow for calcined coagulated sediments

3.2 The determination method for Pu and U in sludge

The sludge generated from the neutralization and coagulation sedimentation processes is separated by type, and grinding treatment is performed. After taking a sample for DA, each sludge is stored in polyethylene bottles.

Because the sludge quantity for one batch is small, one container includes a plurality of batch sludges from multiple treatment processes, forming a layered structure (Figure 3.5). The DA results sampled each time of sludge generation (Figure 3.5) determine the amounts of Pu and U in the sludge of each batch (i.e., layer).

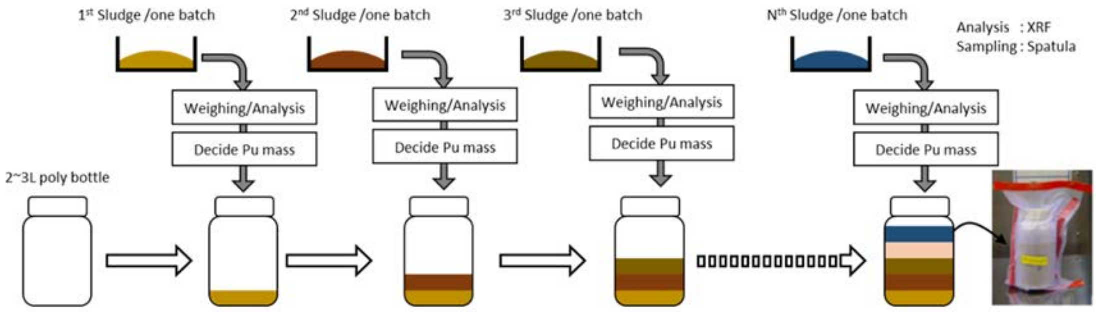


Figure 3.5 The quantitative determination method of nuclear material in sludge in polyethylene bottles using DA

3.3 Storage device and method

The sludge after the calcined neutralized sedimentation process and calcined coagulated sedimentation is stored in the facility. The sludge items are stored in a polyethylene bottle and packed in double plastic bags to prevent contamination.

Those sludges contain impurities with deliquescence and generate hydrogen gas caused by the radiolysis of water, swelling the plastic bag. Therefore, a visual inspection should ensure that the plastic bag containing the sludge does not have any swelling. When swelling occurs, that container is transferred to the glove box to change the plastic bag for degassing.

4. Sludge stabilization

4.1 The background of sludge stabilization

Radioactive contamination at the Pu fuel research facility in the Oarai Research and Development Center occurred on June 6, 2017. The resin bags containing solid Pu and U waste in the draft chamber were ruptured, and contamination occurred when the operators inspected a storage container for a safety check. This caused internal radiation exposure of some workers engaged in this work. The investigation revealed that the cause of the internal exposure was gas generation due to alpha-ray decomposition of mixed organic substances, including epoxy resin, which increased the internal pressure of the resin bags. The response after the incident was a review of all nuclear material-handling sites in the JAEA, and safety countermeasures were taken for similar storage cases, including the PCDF. The sludge storing in the PCDF was a similar storage case and was selected to stabilize the sludge by washing to prevent gas generation during storage.

4.2 The cause of gas generation from the PCDF sludge and countermeasures for the preventing a similar incident

The investigation results indicated that the cause of the gas generation from the PCDF sludge was the gas generated by the radiolytic decomposition of water absorbed in sodium nitrate ($NaNO_3$), which is included in sludge as a part of impurity. Therefore, washing the sludge with water was planned to remove $NaNO_3$ from sludge as a countermeasure to prevent gas generation.

4.3 The sludge stabilization procedure

Sludge stabilization was performed in a glove box using an existing liquid waste treatment process in the PCDF. Figure 4.1 shows the sludge stabilization procedure.

① Sludge transfer from the storage area
- ➢ Transfer the sludge in polyethylene bottles from the storage area (the storage cabinet or glove box in PCDF) to the glove box for water washing.
- ➢ Take a certain amount of sludge from the polyethylene bottle and grind it before washing. (Note that grinding is performed in small batches due to the mass tolerance of the grinding equipment. The full content of ground sludge is then combined and transferred to the washing step.)

② Water washing and filtration
- ➢ Place the ground sludge into a filter and mix with water for washing, stirring it for 1 min to dissolve $NaNO_3$ in the sludge in the water.
- ➢ Leave it for 10 min after stirring and divide it into precipitates and filtrates.
- ➢ Repeat the above process twice.

③ Drying and calcination
- ➢ The precipitate after filtration is dried at 120°C for 5 h, then calcined at 550°C for another 5 h for powderization.

④ Storage after stabilization
- ➢ The sludge after stabilization is stored in a metal storage bottle after sampling for DA analysis to determine the amount of nuclear material in stabilized sludge.

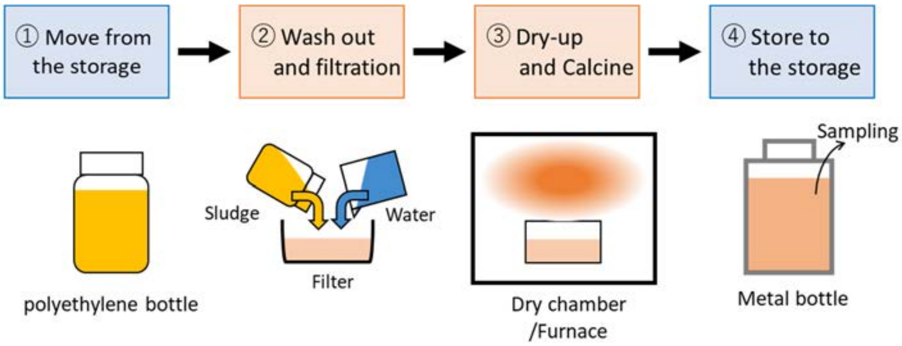


Figure 4.1 The sludge stabilization flow

The volume of sludge after stabilization is reduced due to rinsing of $NaNO_3$ from the sludge. Therefore, mixing a plurality of sludge batches after stabilization is performed for homogenization. Figure 4.2 shows the flow of the cross-blend method for sludge homogenization after stabilization. The cross-blend method is employed due to the limitation of the amount of sludge that can be handled in a glove box at one time in the PCDF.

a) Cross-blend method

- After uniformly grinding using a milling cutter for each batch, a fraction from each batch is collected and mixed to produce a new mixture batch of ~500 g (Figure 4.2 ①, ②).
- The mechanically mixed sludge is transferred to one tray (Figure 4.2 ③), then mixing and stirring are performed for those sludges on the tray using a spatula (Figure 4.2 ④).
- The sludge is divided into ~500 g of small fractions again for mechanically mixing using a milling cutter, and each of the divided sludge is mixed.
- Repeat the procedure above (Figure 4.2 ② to Figure 4.2 ⑤).

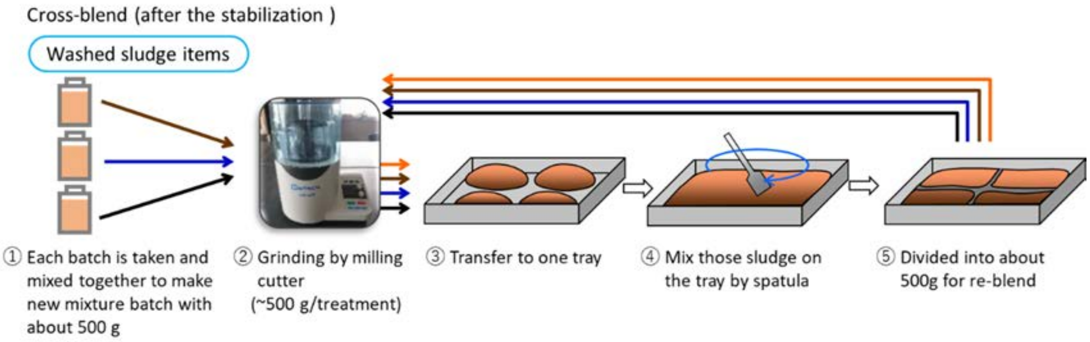


Figure 4.2 The cross-blend process

b) The confirmation method of sludge homogeneity using the PSMC measurement

The PSMC measurement test for the sludge after the cross-blending procedure is conducted to confirm sludge homogeneity at the NDA scale. A small sample is taken from some part of sludge for the PSMC measurements to compare the Pu content in sludge.

- A set of three sludge samples of the same weight after mixing by cross-blending was obtained. Weight adjustment is performed for these samples based on the amount of Pu in sludge (~ 1 g of Pu is required for the PSMC evaluation).
- PSMC measurements of 30 seconds and 10 cycles were performed, and sludge homogeneity was determined from the measurement results' consistency within the measurement errors of plus or minus 1 σ.

- If the Pu amount in the sampled sludge is unequal, another cross-blend and PSMC measurement are performed until the measurement results indicate equality within the limit of 1 σ.
- After the sampling for transferring the sludge to the analysis process, the sludge is stored in a metal bottle packed in double plastic bags.

The sludge after homogeneity using the cross-blend method followed by the stabilization procedure changes from brown to black (Figure 4.3). Those sludges are stored in a metal bottle (~2.4 L) and packed with double plastic bags, the same procedure for storage as for polyethylene bottles in the past.

Figure 4.3 The sludge after stabilization and storage conditions

4.4 The result of sludge stabilization

The sludge stabilization started on August 20, 2018, and 144 stabilized items were obtained. The items after mixing a plurality of sludge were 66. Table 4.1 and Table 4.2 shows the average composition of the sludge before and after stabilization.

Table 4.1 The average composition of neutralization sludge before and after stabilization

Item	Unit	Before	After
Weight	g	1,839.46	1,413.82
Pu	wt%	1.61	4.70
U	wt%	2.35	7.77
Am*	ppm/Pu	66,300	64,950
Moisture*	wt%	4.2	2.1
Na	wt%	39.0	2.3
Fe*	wt%	26.0	46.0
Cr*	wt%	5.5	0.1
Ni*	wt%	10.4	22.6
Dose rate(γ)	μSv/h	18,000	10,400
Dose rate(n)	μSv/h	90	50

*: Reference value

Stabilization washed out approximately 90% of the sodium. The sodium weight decreased from ~39 to ~2.3 wt%. It was considered that the sodium in the sludge was primarily sodium nitrate ($NaNO_3$), and the reduction in the sodium concentration was caused by the transfer of $NaNO_3$ from the sludge to the washing solution (water). The table also demonstrates that stabilization increased the concentrations of Fe and Ni.

The average Pu concentration value per one storage bottle changed from ~1.61 to ~4.70 wt%, and the U concentration changed from ~2.35 to ~7.77 wt%. The chemical element composition of sludge

is predominantly PuO_2, $Na_2U_2O_7$, Am, and metal impurities, such as Fe and Cr.
These compounds are insoluble in water; therefore, they tend to remain in the residue rather than in the washing solution.
As a result, consequently, the sludge volume per item decreased because more than 90% of sodium was removed from the calcined neutralized sediment before stabilization. The percentages of Pu, U, Fe, and Ni remaining in the sludge after stabilization increased due to sodium reduction, indicating that the stabilization procedure reduced the sodium content in the sludge.

Table 4.2 The average composition of coagulation sludge before and after stabilization

Item	Unit	Before	After
Weight	g	2,748.30	1,952.46
Pu	wt%	Not detected	Not detected
U	wt%	Not detected	Not detected
Am*	ppm/Pu	Not detected	Not detected
Moisture*	wt%	3.1	1.3
Na	wt%	6.2	3.0
Fe*	wt%	61.8	65.2
Cr*	wt%	0.1	0.1
Ni*	wt%	3.3	3.45
Dose rate(γ)	μSv/h	800	630
Dose rate(n)	μSv/h	2	0

*: Reference value

For coagulation sludge, the sodium content decreased from 6.2 to 3.0 wt%, the same trend as the neutralization sludge. However, the Pu and U concentrations of coagulation sludge did not increase because the Pu and U concentrations in these sludges before stabilization was originally low (under the detection level (0.02 g/L) by X-ray fluorescence analyzer (XRF) measurements under the analysis condition of PCDF analysis section).

The coagulation sludge has higher Fe and lower Na concentrations than the neutralization sludge because this process adds ferric nitrate as the precipitating reagent. Furthermore, the coagulation sludge is generated from the waste solution with Pu and U after the neutralization process. Therefore, the stabilization rate of the coagulation sludge is approximately half of the neutralization sludge.
From the results obtained, it is concluded that the sludge stabilization for both neutralization and coagulation sludge sediments was accomplished, and the safety of the storage form of sludge was improved.
The stabilization work by water washing prevented gas generation from the sludge, including some items that are stored over two years after stabilization.

5. The methodology for determining Pu in sludge

5.1 Conventional procedure by DA

Figure 5.1 shows the procedure for determining the Pu and U contents in sludge using the conventional method (weighing, sampling, and DA (i.e., XRF)) based on the design information questionnaire (DIQ) (Table 5.1).

Table 5.1 DIQ DA description for sludge

MBA	KMP	Chemical and physical form of material	Equipment or installed place	Sampling procedure and equipment used	Measurement and analytical method and equipment used	Source and level of random and systematic errors*	Method of converting source data to batch data
1	5	Sludge (1) and Sludge (2)	Mainly Liquid waste Treatment Room (A129)	Direct sampling	Weight; weighing apparatus Content; XRF or spectrophotometry	Weight measurement; R;2% S;3% Sampling; R;6% S;6% Analysis; XRF ; R;20% S;10% Spec.; R;20% S;10%	(Weight) × (Content)

*: The total measurement uncertainty: approximately 24.2%

The necessary amount of the powder sample is taken for the Pu and U determination using DA, and the rest of the powder is weighted to determine the net weight in Eq. 5.1. The amounts of Pu and U in sludge is determined from the following formula (Eq. 5.1), using the net weight (g) and Pu, U concentration (wt%) obtained by the XRF result.

$$\text{Pu, U mass [g]} = \text{Net weight [g]} \times \text{Pu, U concentration [wt\%]}/100 \qquad (5.1)$$

The Pu/U isotopic ratio and the content of ^{241}Am are determined from the estimated value based on the operational data.

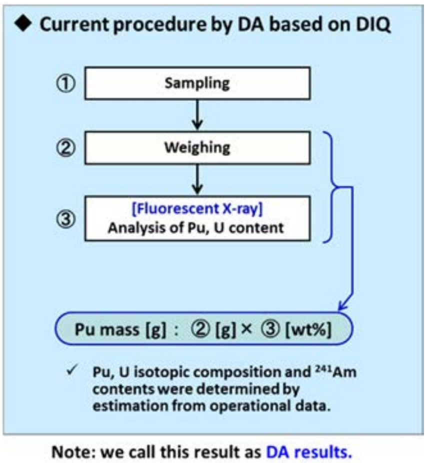


Figure 5.1 The flow of Pu determination using DA

The quantity of one batch of sludge from the waste treatment process is small, and the sludge contents in one polyethylene bottle will be formed as the laminations of different batches from the bottom. Therefore, the total nuclear material amount for Pu and U in one polyethylene bottle will be the sum of the Pu and U amounts in each sludge layer. This layered sludge structure in each storage bottle becomes problematic from the viewpoint of safeguards because one storage bottle comprising the plurality of different nuclear material quantities does not match the definition of a batch, resulting in an inability to ensure the representativeness of the sample for safeguards verification (Figures 3.5 and 4.2). Note that this problem also remains for the stabilized sludge stored in metal cans, obtained using the cross-blend method previously described; therefore, one metal can is consist of plural number of contents from several batches.

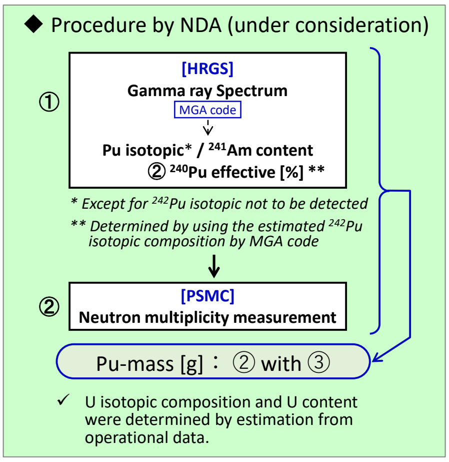
5.2 NDA

Figure 5.2 shows the procedure for determining Pu in sludge using NDA based on DIQ (Table 5.2). NDA can evaluate the entire storage bottle for Pu determination. The Pu isotopic ratio and ^{241}Am contents were obtained from the gamma-ray spectra using high-resolution gamma spectrometry (HRGS), followed by the pulse height analysis of those gamma energy spectra with the spectrum analysis method for the Pu gamma-ray, referred to as the multigroup analysis (MGA) code.

Table 5.2 DIQ NDA description for sludge

MBA	KMP	Chemical and physical form of material	Equipment or installed place	Sampling procedure and equipment used	Measurement and analytical method and equipment used	Source and level of random and systematic errors	Method of converting source data to batch data
1	5	Sludge (1) and Sludge (2)	R&D Area (A128)		Item counting Identification Pu amount; NDA (PSMC)	[Tentative] PSMC; 3 ~ 11%	Pu amount measured by NDA. U amount estimated from Pu amount measured by DA.

The Pu mass in the sludge is calculated by evaluating the neutron emission from the sludge measured using PSMC and the isotopic ratio from the MGA results. The U mass and isotopic ratio were obtained from the estimated value from the PCDF operation data (the detail is described in Section 9).

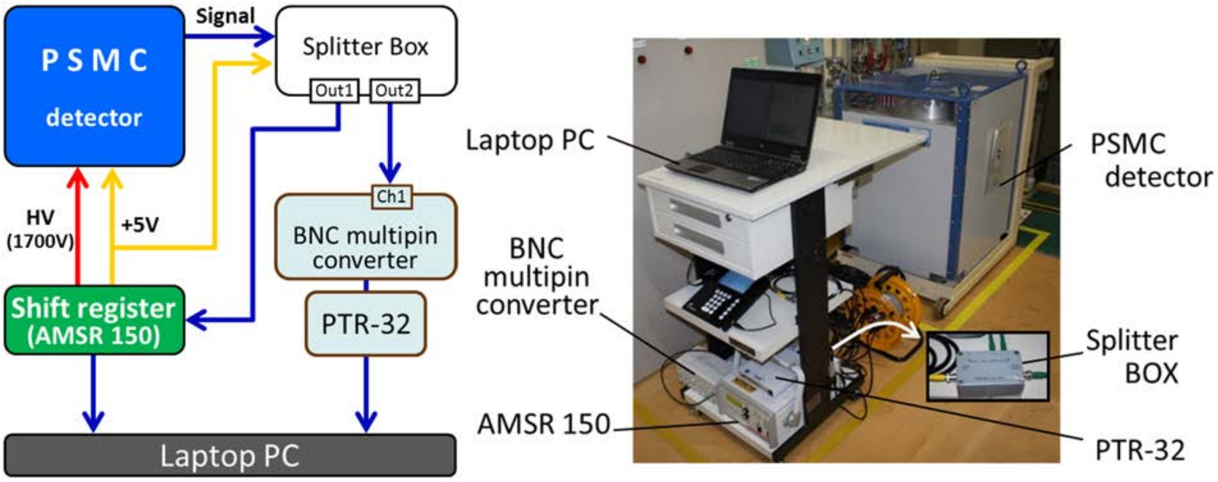


Note: we call this result as NDA results.

Figure 5.2 The flow of Pu determination using NDA

5.2.1 The PSMC measurement system

Figure 5.3 shows the PSMC measurement system.



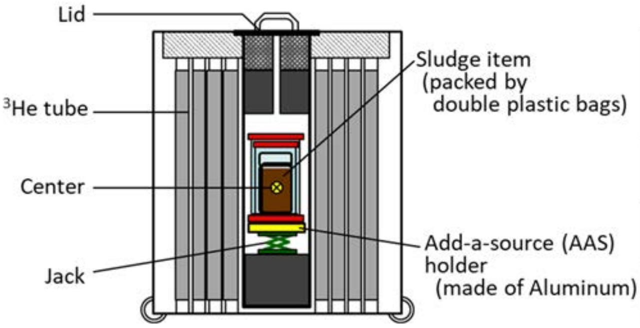
(a) The PSMC system configuration (b) The appearance of the PSMC system

Figure 5.3 The PSMC measurement system for sludge measurement

Figure 5.3(a) shows the PSMC measurement system comprising a PSMC detector, shift register (AMSR-150: ORTEC), list mode counter (PTR-32), splitter box, BNC-multipin converter, and laptop PC. A high voltage is supplied to a PSMC detector from the shift register. The signal from the detector is transmitted to the PC through the splitter box and shift register. When using the list mode, the signal from the detector is transmitted to the PC through the PTR-32 after the BNC-multipin converter.

a) The PSMC detector

The PSMC is a high-efficiency neutron counter designed for the measurement of the multiplicity of the neutron emission from both spontaneous fission and induced fission reactions in Pu and U. Figure 5.4 shows a diagram of the PSMC design with the 80 ^{3}He tubes surrounding the sample cavity.



Detector	^{3}He tube Number of tubes: 80 D: 25.4 mm Gas pressure: 0.4 MPa
Outside dimension	W × L: 660 mm × 660 mm H: 916 mm
Chamber	D: 200 mm H: 410 mm
Moderator	Polyethylene
Absorber	Cadmium

Figure 5.4 The design and dimensions of the PSMC detector

b) The PSMC operating parameters

Several operating parameters are required when using the PSMC in the multiplicity counting mode, including high voltage, neutron detection efficiency, Doubles and Triples gate fractions, predelay, and gate. As discussed in more detail in Section 7.1, these parameters were carefully evaluated and validated for use in sludge items assay. These evaluations included a high-voltage setting confirmation to ensure there is no gamma-ray interference, evaluation of neutron detection efficiency for the plastic bottle filled with sludge-like matrix and metal can used for sludge after stabilization, evaluation of Triples gate fraction, and confirmation of predelay and gate settings. The details of these calibration procedures are described in more detail in the LANL report [2] and [3]. The PSMC operating parameters were evaluated using a ^{252}Cf neutron source, MOX powder mixed with chemical reagents as mock-sludge, and a set of representative sludge items.

Table 5.3 shows the PSMC operating parameters for sludge measurements established in this comprehensive evaluation. Section 7 provides more details for those parameters.

Table 5.3 The PSMC operating parameters for sludge measurement - top for standard PSMC predelay and bottom for reduced predelay settings

Parameter	Unit	Setting value	Parameter	Unit	Setting value
Predelay	μs	4.5 (standard) 2.5 (reduced*)	Multiplicity deadtime	ns	112
Gate length	μs	64	Coefficient A	μs	0.379
2nd gate length	μs	64	Coefficient B	μs^2	0.123
High voltage	V	1700	Coefficient C	—	0.850
Die-away time	μs	49	Doubles gate fraction	—	0.615 (4.5 μs predelay) 0.643 (2.5 μs predelay)
Efficiency	—	0.543	Triples gate fraction	—	0.400 (4.5 μs predelay) 0.425 (2.5 μs predelay)

* For more details on this change, see Section 7.1 and [3].

The PSMC measurement conditions are as follows.

(1) Background measurement

- Confirm that the measurement system setup is appropriate and measure the background with no nuclear material present in the detector.
- The measurement time is 60 s, with 10 cycles.

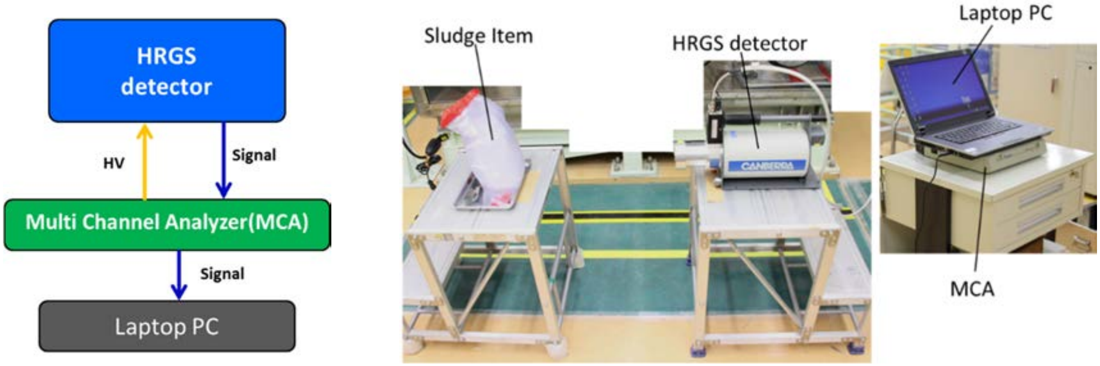
(2) Normalization measurement

- A series of measurements were obtained using a californium source placed at the center of the sample cavity to confirm that the expected response is obtained in the PSMC detector.
- The measurement time is 60 s, with 10 cycles.

(3) Sludge measurement

- Measure the amount of Pu in sludge positioned on the Add-A-Source (AAS) holder at the bottom of the sample cavity.
- The measurement is performed until the precision of the Triples signal is below 3%.
    - ✓ Before the stabilization sludge item: at least 60 s with 600 cycles (10 h) and the longest for 60 s with 1,440 cycles (24 h)
    - ✓ After the stabilization sludge item: at least 30 s with 120 cycles (1 h) and the longest for 30 s with 2,880 cycles (24 h).
- AAS measurement for 60 s with 60 cycles

5.2.2 The HRGS measurement system



(a) The HRGS system configuration (b) The appearance of the HRGS system

Figure 5.5 The HRGS measurement system for sludge measurement

Figure 5.5 shows the HRGS measurement system comprising a HRGS detector (model 2002CP: CANBERRA), multichannel analyzer (MCA), and laptop PC connected by cables.

a) HRGS detector

The HRGS detector contains a high-purity germanium detector (Canberra Industries, GL0510R), pulse height analyzer (MCA 1200 INSPECTOR), and control software (Genie 2000 and MGA (Ver.10.0)).

b) Pu isotopic composition

Pu isotopic composition is obtained by measuring the gamma-ray spectra using HRGS. However, it is difficult to directly measure the ^{242}Pu value because the intensity of emitted radiation rays is weak due to the longer ^{242}Pu half-life compared with other Pu isotopes. Therefore, the ^{242}Pu isotopic ratio is calculated using Equations 5.2 and 5.3.

$$^{242}Pu = 100 \cdot \frac{\left[\frac{242}{239}\right]}{\left[\frac{238}{239}\right]+\left[\frac{239}{239}\right]+\left[\frac{240}{239}\right]+\left[\frac{241}{239}\right]+\left[\frac{242}{239}\right]}, \tag{5.2}$$

where

$$\left[\frac{242}{239}\right] = C_1\left[\frac{238}{239}\right]^{C_2} \cdot \left[\frac{240}{239}\right]^{C_3} \cdot \left[\frac{241+Am}{239}\right]^{C_4} + C_5\left[\frac{238}{239}\right]^{C_8} + C_6\left[\frac{240}{239}\right]^{C_9} + C_7\left[\frac{241+Am}{239}\right]^{C_{10}}, \quad (5.3)$$

and $\left[\frac{238}{239}\right], \left[\frac{239}{239}\right], \left[\frac{240}{239}\right], \left[\frac{241}{239}\right], \left[\frac{241+Am}{239}\right]$ are isotopic ratios of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, respectively, to the existing against ^{239}Pu.

The factors from C_1 to C_{10} for Equation 5.3 are used for the sludge measurement using the MGA parameters shown in Table 5.4. For further details on determining these parameters, see Section 7.3.

Table 5.4 MGA parameters for sludge measurement

Coefficient	Setting value
C_1	4.40
C_2	1.016

*C_3-C_{10} are 0.0.

The HRGS measurement conditions for obtaining the Pu isotopic ratio and ^{241}Am content in sludge are as follows:

(1) Background measurement
- Confirm that the measurement system setup is appropriate and measure the background at the measurement location after supplying high voltage (HV) from the MCA.
- The measurement time is 300 s.

(2) Source measurement
- A series of measurements were taken using the ^{241}Am source placed at the tip of the detector to confirm the performance of the HRGS device.
- The measurement time is 300 s.

(3) The sludge measurement
- Measure the Pu isotopic ratio and ^{241}Am content in the sludge sample located in line with the detector.
- The measurement is performed with the distance between the detector and sample adjusted to set the deadtime to correspond to 20% or less. The number of 0 to 3 Cd plates is used for reducing the gamma-ray emitted from ^{241}Am (59.5 keV).

6 Comparing the measurement results between NDA and DA

6.1 NDA measurement results

6.1.1 Validating the PSMC/MGA parameters

A MOX powder sample with a Pu/U ratio of 1:1 and a sample called standard sludge (prepared from a mixed solution containing Pu and U) are measured using the PSMC and HRGS, respectively. The standard sludge is prepared using a mixed solution of Pu and U containing less impurities for reducing the influences of metal impurities in sludge. Table 6.1 shows the samples composition.

Table 6.1 The sample composition for validating the PSMC/MGA parameters

Item		MOX powder	Standard sludge	Unit
Weight		25.02	26.03	g
Pu mass		10.00	8.53	g
Pu contents		39.97	32.76	wt%
Pu isotopes	^{238}Pu	1.172	1.107	%
	^{239}Pu	59.381	61.127	%
	^{240}Pu	30.749	29.769	%
	^{241}Pu	3.103	2.719	%
	^{242}Pu	5.595	5.278	%
U mass		10.65	8.77	g
U contents		42.56	33.70	wt%
U isotopes	^{234}U	0.123	0.153	%
	^{235}U	0.624	0.607	%
	^{236}U	0.183	0.191	%
	^{238}U	99.070	99.049	%
Pu, U measurement day		May 9, 2017	July 2, 2020	-
^{241}Am		22500	36900	ppm
^{241}Am measurement day		May 8, 2017	July 2, 2020	-
Moisture		0.19	4.08	wt%
Na contents		0.03	5.60	wt%

Table 6.2 shows the sludge measurement results of MOX powder and standard sludge using the PSMC/MGA parameters. One σ represents the standard deviation in the DA results.

Table 6.2 The measurement result of MOX powder and standard sludge

Item	PSMC date	Method	Pu mass [g]		^{240}Pu effective[%]^{*1}		Alpha[-]
			Value	Difference[%]	Value	Difference[%]	
MOX powder	2020/2/26	NDA	10.08	-1.10	42.66	1.27	0.965
		DA^{*2}	9.97 ± 0.07	-	43.21 ± 0.58	-	-
Standard sludge	2020/6/25	NDA	8.48	0.64	42.09	-1.59	7.806
		DA^{*2}	8.53 ± 2.06	-	41.43 ± 10.02	-	-

*1: ^{240}Pu$_{\mathrm{eff}}$ is calculated as follows: ^{238}Pu, ^{240}Pu, and ^{242}Pu are isotopic compositions.

$$^{240}\mathrm{Pu}_{\mathrm{eff}} = 2.52\ ^{238}\mathrm{Pu} + {}^{240}\mathrm{Pu} + 1.68\ ^{242}\mathrm{Pu}$$

*2: The sigma of DA is referenced from PCDF DIQ

The Pu mass of MOX powder and standard sludge obtained using the NDA method agree with the DA within the uncertainty value (two sigma) of DA based on the DIQ. These measurements confirm the validity of the PSMC and MGA parameters determined for sludge NDA assay, as discussed earlier and in more detail in Section 7.

6.1.2 Confirmation results of the homogeneity of sludge

Figure 6.1 shows the sampling location of the sludge after cross-blending, and Table 6.3 shows the PSMC results for each sampling location.

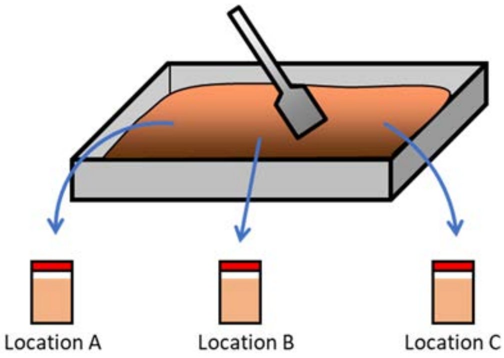


Figure 6.1 Sampling point after the cross-blend

The PSMC results of Pu mass for three sampling locations for each sludge item after cross-blending coincided with the range of 1 σ of uncertainty. This result confirmed the homogeneity of sludge for the PSMC measurement for Pu mass determination, despite the inhomogeneity for the DA method (see Section 6.2 for more details on DA sampling uncertainty). This homogeneity confirmation sampling procedure was performed for 15 of 31 sludge items assayed using the PSMC.

Table 6.3 PSMC measurement results for cross-blending

No.	Item ID	Net weight[g]	Pu mass [g]				Pu contents[wt%] from PSMC
			Location A	Location B	Location C	Average	
1	T109A	17.00	0.932 ± 0.032	0.949 ± 0.033	0.930 ± 0.031	0.937 ± 0.032	5.512
2	T086A	15.00	0.706 ± 0.027	0.748 ± 0.026	0.717 ± 0.027	0.724 ± 0.027	4.824
3	T080A	21.00	0.905 ± 0.027	0.929 ± 0.027	0.919 ± 0.027	0.918 ± 0.027	4.370
4	T089A	28.00	0.916 ± 0.028	0.914 ± 0.029	0.936 ± 0.029	0.922 ± 0.029	3.293
5	T087A	55.00	1.082 ± 0.061	1.140 ± 0.059	1.133 ± 0.059	1.118 ± 0.060	2.033
6	T060A	21.00	1.028 ± 0.037	1.064 ± 0.037	1.028 ± 0.037	1.040 ± 0.037	4.952
7	T004A	17.00	1.066 ± 0.040	1.048 ± 0.040	1.077 ± 0.040	1.064 ± 0.040	6.257
8	T028A	15.00	1.188 ± 0.044	1.232 ± 0.044	1.213 ± 0.044	1.211 ± 0.044	8.073
9	T142A	35.00	1.293 ± 0.040	1.226 ± 0.041	1.308 ± 0.041	1.276 ± 0.041	3.645
10	T139A	15.00	0.677 ± 0.030	0.657 ± 0.029	0.660 ± 0.030	0.665 ± 0.030	4.431
11	T066A	19.00	1.027 ± 0.032	1.023 ± 0.032	1.024 ± 0.032	1.025 ± 0.032	5.393
12	T065A	23.00	1.531 ± 0.045	1.512 ± 0.045	1.519 ± 0.046	1.521 ± 0.045	6.612
13	T074A	11.00	0.963 ± 0.042	0.945 ± 0.042	0.958 ± 0.041	0.955 ± 0.042	8.685
14	T126A	17.00	0.974 ± 0.029	0.952 ± 0.030	0.969 ± 0.029	0.965 ± 0.029	5.676
15	T053A	23.00	1.414 ± 0.043	1.406 ± 0.043	1.407 ± 0.043	1.409 ± 0.043	6.126

Other sludge items are a small amount of net weight item or very low Pu concentrations and were not needed for the cross-blend operation.

6.1.3 NDA measurement

The NDA measurements using the PSMC/MGA parameters validated in Section 6.1.1 were performed.

50 before stabilization sludge items were selected and measured form the storage items (approximately 200) in the facility. On the other hand, 31 after stabilization sludge items were selected and measured.

6.2 Measurement uncertainty by DA

Samples were taken for DA analysis from each sludge storage bottle after stabilization to determine the amounts of Pu and U in each sludge. Sampling uncertainty evaluation was performed by confirming the level of sludge homogeneity in one storage bottle.

Test 1: The sampling uncertainty per sludge storage bottle
Test 2: Validating the XRF measurement results by comparison with another analysis methodology

In Test 1, numerous samplings from an arbitrary location for sludge (ID T086A) were collected, and the Pu content (wt%) measured using XRF was compared. In Test 2, the Pu content results between XRF and isotope dilution mass spectrometry (IDMS) were compared. Figure 6.2 shows the results of Test 1 obtained from the eight sampling results measured using XRF.

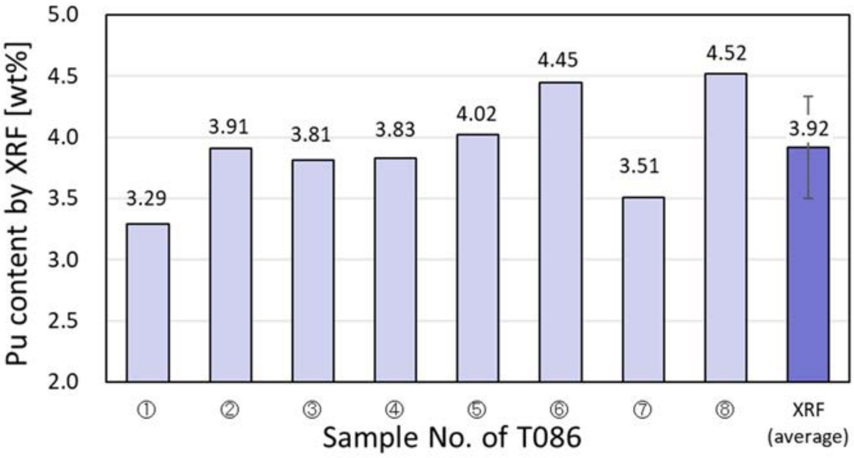


Figure 6.2 The sampling uncertainty of sample ID T086 (Test 1)

As a result, the average Pu content was 3.92 wt%, with an uncertainty of 1 σ of 10.7% (0.42 wt%). All the Pu content results are within the range of 2 σ from the average value, and the sampling uncertainty value exceeds the XRF equipment uncertainty of ±5%. These results indicate that the variation between samples is much greater than the expected variation from the XRF equipment uncertainty; therefore, sampling uncertainty is significant. The total uncertainty value is the sum of the sampling and DA (XRF) procedure uncertainties measured using the following equation:

$$U_t = \sqrt{U_s^2 + U_a^2} \tag{6.1}$$

where U_t, denotes the total uncertainty of this measurement (10.7%)
U_s, the sampling uncertainty.
U_a, the DA (XRF) procedure uncertainty (5%).

The sampling uncertainty after sludge stabilization is 9.5% using the following equation:

$$U_s = \sqrt{U_t^2 - U_a^2} \tag{6.2}$$

Figure 6.3 shows the Test 2 results, the Pu content measured using XRF and IDMS. The sample used in Test 2 is the same as ID T086⑧ in Test 1.

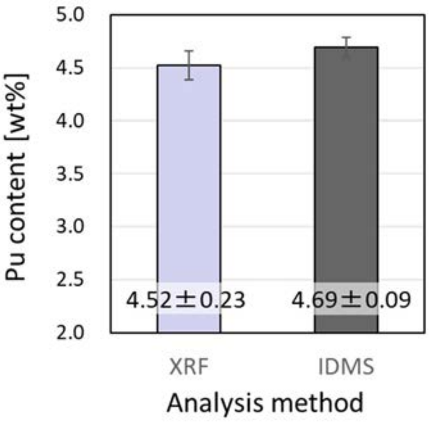


Figure 6.3 The comparison results between XRF and IDMS (Test 2)

The XRF and IDMS results are 4.52 ± 0.23 wt% and 4.69 ± 0.09 wt%, respectively. The difference between XRF and IDMS is ~3.8%, within the range of XRF uncertainty of ± 5%; therefore, the XRF measurement results are valid for the sludge items. The results of Tests 1 and 2 indicate that the sludge analysis using DA (XRF) has ~9.5% of sampling uncertainty and no bias with measurement equipment. Further XRF evaluation is provided in Section 7.2 in the context of the applicability assessment of PSMC to sludge NMA.

Table 6.4 shows the result of total measurement uncertainty (TMU) of DA for the Pu mass in sludge items based on the results of Tests 1 and 2. The TMU is calculated from the sum of the weighing, sampling, and measurement equipment uncertainties.

Table 6.4 Reevaluation results of current nuclear material accountancy (NMA) procedure by destructive analysis (DA)

		Weight[*1]	Sampling	Analysis	TMU[*3]
Practical value (evaluation result)	R	2%	9.5%	5%[*2]	11.3%[*4]
	S	3%	-		
DIQ (current)	R	2%	6%	20%	24.2%
	S	3%	6%	10%	

*1: The DIQ value for weighing

*2: The control value in analytical section

*3: The TMU was calculated as $\sigma = \sqrt{\sigma^2_{wei(R,S)} + \sigma^2_{sam(R,S)} + \sigma^2_{ana(R,S)}}$

*4: This data (11.3%) was calculated from DA(XRF) results using same sample for NDA evaluation.

From the above results in the Table 6.4, the total XRF measurement uncertainty is 11.3% for the sludge after stabilization. The sampling uncertainty of sludge after cross-blending for homogenization is ~10%. This reevaluation result of the current NMA procedure using DA would be reflected in the revision of DIQ and the NDA procedure in consultation with IAEA.

7. Applicability of PSMC and HRGS measurement results for sludge NMA

7.1 Applicability of PSMC

The LANL evaluated two datasets for the potential of the PSMC to be used as an NDA technique for sludge measurements. An initial set of 50 items was provided before stabilization, and an additional set of 31 items was provided to evaluate the PSMC performance after stabilization. Furthermore, a set of five items was provided with the same Pu content before and after stabilization (i.e., unmixed items). As discussed in Section 4.1, the stabilization procedure typically involves mixing of multiple items, complicating the evaluation of the effects of the procedure on the PSMC performance. The five items, down selected from the initial 50 sludge items, were individually stabilized without mixing with other items for the direct evaluation of the stabilization process on the PSMC performance. The following sections summarize the evaluation and PSMC performance results for each group.

7.1.1 Sludge items before stabilization

Table 7.1 shows the overview of 50 sludge items measured in the PSMC before stabilization. The items represent a complex mixture of Pu in a matrix comprising low-Z elements (O, Na, N) and stainless components (Fe, Cr, Ni). The low-Z elements cause high and variable α, n reactions, since the concentration and type of low-Z elements in the sludge changes from item to item. Therefore, the NDA assay using a passive calibration curve method cannot be used, and a multiplicity assay represents the most reliable and robust method recommended for this type of material [4]. In addition, the moisture content of the sludge before stabilization was not well controlled. The passive multiplicity calibration method and an AAS method were employed for the evaluation of moisture content. A dedicated AAS holder was designed in collaboration with the LANL and machined by the JAEA. The 50 sludge items provided to the LANL were measured twice, once in a passive mode for passive multiplicity assay and once with a ^{252}Cf source for the AAS technique.

Table 7.1 Overview of 50 sludge items measured before stabilization and evaluated by the LANL

Item No.	Item ID	Item No.	Item ID
1	A123T161	26	A123T095
2	A123T175	27	A123T142
3	A123T110	28	A123T054
4	A123T076	29	A123T156
5	A123T002	30	A123T162
6	A123T143	31	A123T150
7	A123T043	32	A123T130
8	A123T065	33	A123T152
9	A123T053	34	A123T123
10	A123T125	35	A123T132
11	A123T085	36	A123T159
12	A123T074	37	A123T067
13	A123T120	38	A123T154
14	A123T124	39	A123T024
15	A123T068	40	A123T147
16	A123T106	41	A123T096
17	A123T131	42	A123T039
18	A123T028	43	A123T077
19	A123T045	44	A123T157
20	A123T135	45	A123T118
21	A123T122	46	A123T145
22	A123T137	47	A123T140
23	A123T035	48	A123T081
24	A123T151	49	A123T033
25	A123T098	50	A123T034

To use the PSMC in the passive multiplicity calibration mode, its operating parameters had to be established specifically for the configuration of sludge items to account for the matrix and packaging effects on the PSMC detection performance. All sludge items before stabilization were contained in

21 standard plastic bottles (Figure 7.1), and the LANL provided a dedicated operating parameter calibration procedure to the JAEA to establish or confirm the key parameters. The calibration included confirmation of high-voltage settings to ensure no gamma interference, evaluation of neutron detection efficiency for plastic bottle filled with sludge-like matrix, evaluation of Triples gate fractions, and confirmation of predelay and gate settings. The LANL report [2] describes details of the calibration, and Table 7.2 summarizes the results.

Table 7.2 Overview of the PSMC calibration parameters used for the measurement of sludge items before stabilization

Parameter	Unit	Setting value	Parameter	Unit	Setting value
Predelay	μs	4.5	Multiplicity deadtime	ns	112
Gate length	μs	64	Coefficient A	μs	0.379
2nd gate length	μs	64	Coefficient B	μs^2	0.123
High voltage	V	1700	Coefficient C	—	0.850
Die-away time	μs	49	Doubles gate fraction	—	0.615
Efficiency	—	0.543	Triples gate fraction	—	0.400

Figure 7.1 Plastic bottle used for sludge items before stabilization

The 50 sludge items (and all remaining items before stabilization) were measured using the operating parameters from Table 7.2. The first evaluation step focused on the AAS measurements to establish the moisture content. The AAS technique uses a ^{252}Cf source to establish the moisture content of an item through perturbation measurements of ^{252}Cf Doubles count rate [5]. The californium perturbation (Eq. 7.1) is calculated using the unperturbed Doubles directly from ^{252}Cf with no item in the PSMC (D_{empty}) and the difference in Doubles measured with ^{252}Cf and the item present (D_{AAS}) as well as the Doubles from the item itself (D_{noAAS}). Figure 7.2 summarizes the results of this evaluation. The californium perturbation for all 50 items is <3%, demonstrating low moisture and confirming no need for additional moisture correction in the passive multiplicity analysis.

$$Cf_{perturbation} = \frac{D_{empty}}{(D_{AAS} - D_{noAAS})} - 1 \tag{7.1}$$

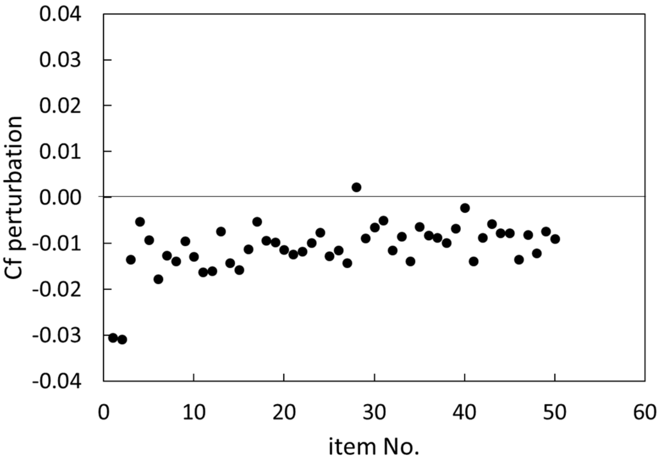


Figure 7.2 Californium perturbation for 50 sludge items measured before stabilization
(The error bars are smaller than the size of the symbols)

PSMC measurements using the passive multiplicity mode were used as the primary analysis method for all 50 sludge items. The passive multiplicity analysis in a properly characterized system provides independent information on the Pu mass, α-value (i.e., (α, n) neutrons contribution), and multiplication. Figures 7.3 and 7.4 show the results for the α-value and multiplication.

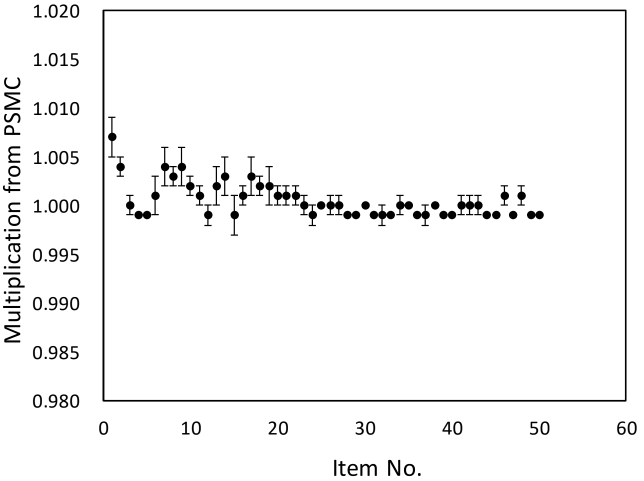


Figure 7.3 Multiplication extracted from the PSMC assay for 50 sludge items before stabilization

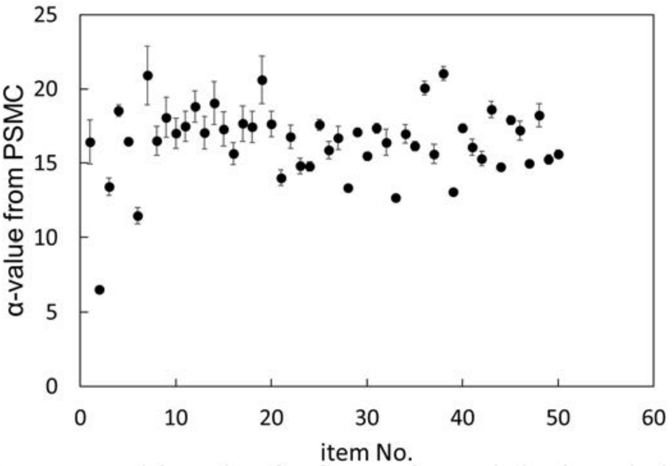


Figure 7.4 α-values extracted from the PSMC assay for 50 sludge items before stabilization

Figure 7.3 demonstrates negligible multiplication for all sludge items, expected for small quantities of fissile material dispersed in a large-volume (~2 L) matrix. Figure 7.4 shows a high α-value, corresponding to 16 on average. Figure 7.5 shows the comparison of the PSMC mass with the DA results. Note that the DA for these sludge items is especially challenging due to their nonhomogeneous composition and could, therefore, bias the results. A similar overall trend is observed in the PSMC as in the DA measurements; however, some variations can be observed. Note that the minimum Pu concentration limit for the DA (XRF) analysis is 0.02 g/L under the condition of the PCDF analysis section, and for concentrations below this value, the DA defaults to 0.3 wt%. This explains the discrepancy observed between the mass measured in the PSMC and DA for groupings of items between 25 and 50. For a better overview of these differences, Figure 7.6 summarizes the PSMC/DA ratio, where the items with the most significant discrepancies are highlighted in different colors. Note that the grouping of items with low PSMC/DA ratios (circled in blue) correspond to those with default DA results of 0.02 g/L Pu concentration.

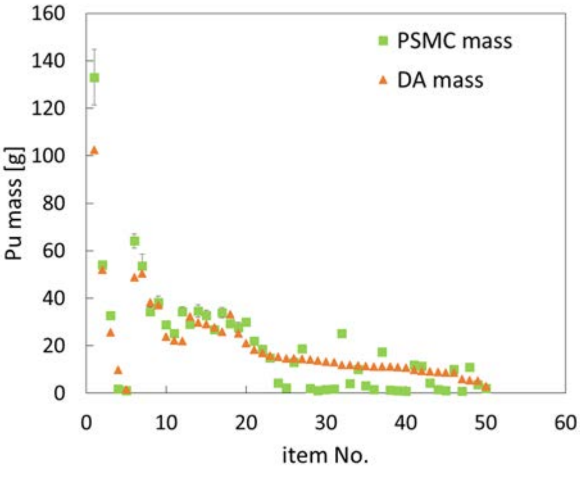


Figure 7.5 Pu mass versus item number measured using the PSMC and extracted from DA

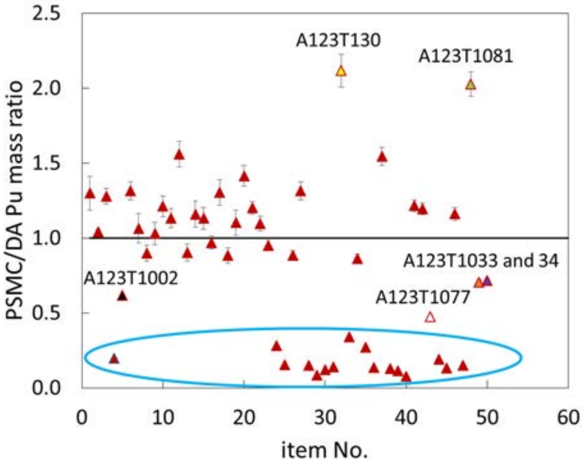


Figure 7.6 Ratio of the PSMC versus DA-determined Pu mass
Note: The grouping of items with low PSMC/DA ratios (circled in blue) correspond to those with default DA results of 0.02 g/L Pu concentration.

For assessing the PSMC assay validity relative to the DA results, it is useful to evaluate a calibration curve of the PSMC-measured Doubles count rate as a function of Pu mass. Ideally, such a calibration curve should result in a unique relationship between the measured Doubles rate and Pu mass. Because of the low multiplication of sludge items (Figure 7.3), the calibration is expected to exhibit a linear trend. Figure 7.7 compares the PSMC calibration curve with the Doubles plotted as a function of the DA Pu mass. Note that the linear fit of the data excludes the highest mass item. Items with the highest PSMC/DA discrepancy are color-coded, as shown in Figure 7.6. From Figure 7.7, the PSMC measurement results are in an improved calibration compared with DA. Note the clustering of items with low Doubles along the *x*-axis for the DA calibration curve due to 0.3 wt% (NMA value for under-the-detection-limit items), removed in the PSMC calibration. The color-coded items from Figure 7.6 exhibit significant scatter in the DA calibration but follow a near-linear trend in the PSMC calibration. These observations support the improved performance in sludge item characterization using PSMC compared with DA for all but the highest mass item.

The significant deviation of the highest mass item (A123T161) from the expected linear calibration trend can be attributed to high accidentals due to the combination of its high Pu mass and high α-value. These accidentals significantly increase the uncertainty of correlated count rates (Doubles and Triples) and can overestimate them. Item T161A shows the most challenging scenario from the evaluated sludge items. The high Doubles rate for this sample would be expected due to the induced fission reactions from the random (α, n) source neutrons.

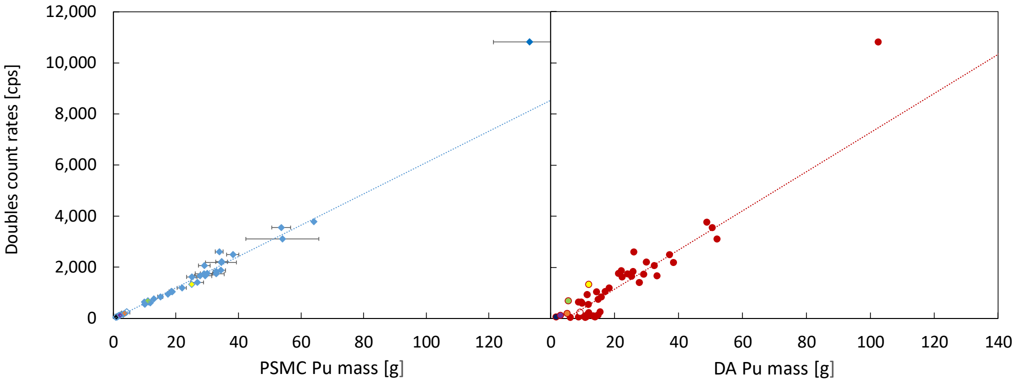


Figure 7.7 Comparison between PSMC calibration and DA calibration for the DA Pu mass for sludge before stabilization

7.1.2 Effects of sludge stabilization on the PSMC performance

Based on the evaluation of the 50 items, 5 items were selected to evaluate the effects of the stabilization process. To fully evaluate these effects of stabilization process, these items underwent the process without mixing with other sludge items (Figure 7.8), contrary to the routine stabilization procedure (Section 4.1). This provided a unique opportunity to observe the direct impact of stabilization on item properties (α-value, multiplication, and mass). Table 7.3 shows an overview of the selected items. These items were selected, because they provide a representative set of items with range of Pu mass and concentration that resulted in very good as well as very poor agreement with DA.

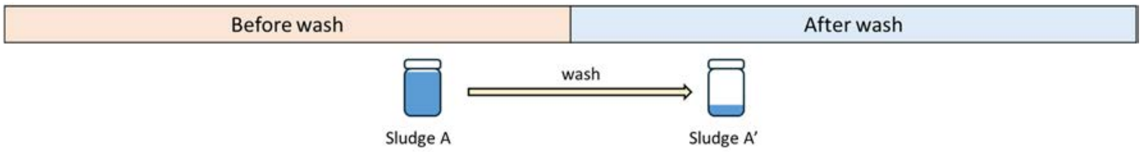


Figure 7.8 Schematic material flow in the sludge stabilization process for the subset of items selected to be stabilized without mixing with other items

Table 7.3 Overview of sludge items selected for the evaluation of the stabilization process

Sludge ID (Before stabilization)	DA Pu mass [g] (Before stabilization)	PSMC Pu mass [g] (Before stabilization)	Net weight [g]	Pu [wt%]	Sludge ID (After stabilization)
A123T145	8.6	10.0	1871.6	0.5	T145A
A123T035	15.8	15.0	2132.3	0.7	T035A
A123T175	52.0	54.1	1061.9	5.1	T175A
A123T131	26.0	33.9	2240.7	1.5	T131A
A123T161	102.4	133.1	2251.2	5.9	T161A

During the stabilization process, the $NaNO_3$ is removed, and items are washed, dried, homogenized, and rebottled in 2 or 2.4 L metal cans.

Figure 7.9 shows the specifications and dimensions of the metal cans. The removal of $NaNO_3$ is expected to significantly reduce the α-value, resulting in an improved Pu mass uncertainty due to the reduced contribution of accidentals.

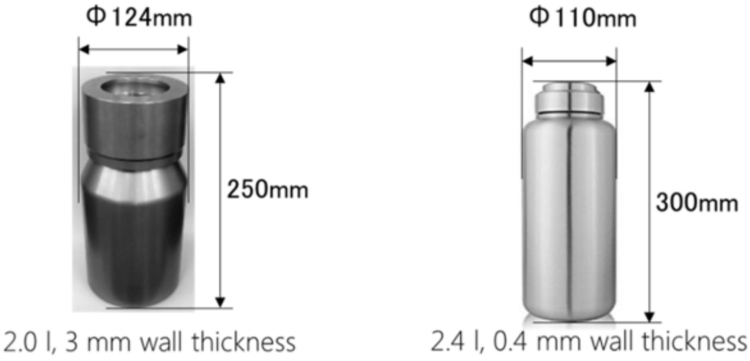


Figure 7.9 Metal cans used for stabilized sludge items

Following the joint LANL/JAEA measurements during the LANL visit at the JAEA in December 2018, the LANL recommended additional optimization of PSMC measurement parameters to further improve measurement uncertainty by reducing the PSMC predelay setting. Reduced predelay allows for higher number of genuine coincidences to be registered within the gate due to the close proximity of the detected pulses to the trigger pulse. Such a technique is well known for reducing measurement uncertainty and was demonstrated for the specific case of PSMC and sludge items in [3].

Since the original sludge items' measurements (before stabilization) were well underway during the LANL visit, they recommended changing the predelay setting, starting with the sludge items after stabilization, to prevent unnecessary remeasurements. The sludges after stabilization were measured using updated PSMC predelay settings. Note that predelay reduction impacts the gate fraction values, as discussed in detail in [3]. Updated gate fractions for the new predelay setting were established, and Table 7.4 summarizes them with the other PSMC parameters. Other PSMC parameters were unchanged for the sludge after stabilization. Note that independent verification of the PSMC operating parameters (efficiency and gate fractions) for sludge after stabilization contained in metal bottles was performed to verify that modification of the PSMC parameter for sludge after stabilization is not needed.

Table 7.4 Overview of the PSMC parameters recommended for stabilized sludge measurements

Parameter	Unit	Setting value	Parameter	Unit	Setting value
Predelay	**μs**	**2.5**	Multiplicity deadtime	ns	112
Gate length	μs	64	Coefficient A	μs	0.379
2nd gate length	μs	64	Coefficient B	ps	0.123
High voltage	V	1700	Coefficient C	—	0.850
Die-away time	μs	49	**Doubles gate fraction**	—	**0.643**
Efficiency	—	0.543	**Triples gate fraction**	—	**0.425**

Table 7.5 summarizes the PSMC measurement results of the selected sludge items before and after stabilization. Figure 7.10 shows the effects of the stabilization process through the Singles count rate and α-value ratios. Figure 7.10 (top) demonstrates that stabilization significantly reduces the Singles count rate on average by a factor of ~4, indicating the reduced contribution of accidental neutrons from the (α, n) reaction due to the $NaNO_3$ removal. Figure 7.10 (bottom) shows the evidence of the reduced α-value. As discussed earlier, a reduced α-value should positively affect the improved Pu mass uncertainty (column 5 of Table 7.5).

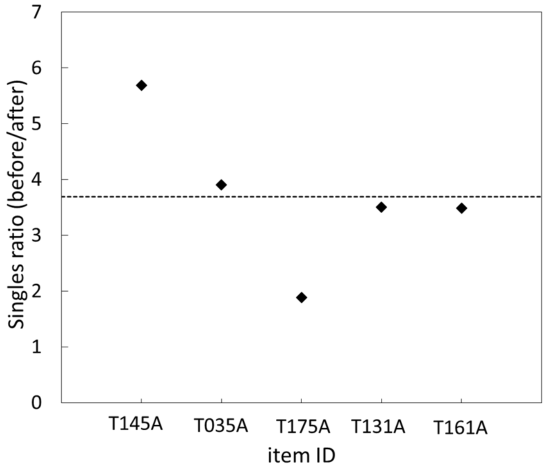
Table 7.5 PSMC results for the subset of sludge items before and after stabilization

(a) Before stabilization sludge items results

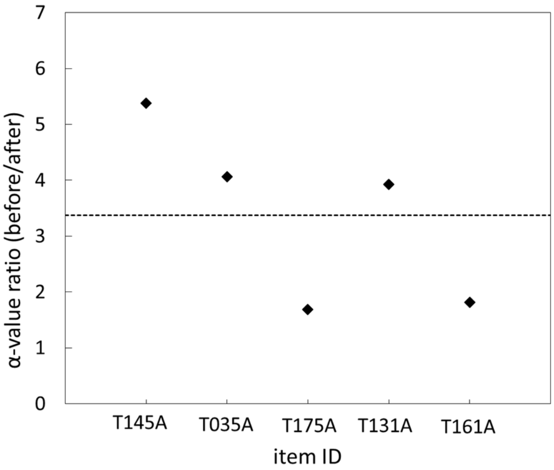
ID before stabilization	S [cps]	σ[cps]	Pu mass [g]	σ [%]	α [-]	σ [%]	M [-]	σ [%]
A123T145	38307.9	1.0	10.0	3.7	17.2	3.8	1.001	0.1
A123T035	45570.1	1.3	15.0	3.4	14.8	3.5	1.000	0.1
A123T175	74629.4	1.6	54.1	1.6	6.5	1.7	1.004	0.1
A123T131	145085.9	1.9	33.9	6.5	17.7	6.7	1.003	0.2
A123T161	496916.0	3.1	133.1	8.8	16.4	9.1	1.007	0.2

(b) After stabilization sludge items results

ID after stabilization	S [cps]	σ[cps]	Pu mass [g]	σ [%]	α [-]	σ [%]	M [-]	σ [%]
T145A	6731.6	1.5	7.6	0.8	3.2	1.0	1.002	0.1
T035A	11667.1	1.9	13.1	1.2	3.6	1.4	0.999	0.1
T175A	39568.7	3.6	44.2	1.9	3.9	2.2	1.007	0.1
T131A	41387.5	3.3	35.4	2.3	4.5	2.6	1.006	0.2
T161A	142695.6	3.4	66.2	5.0	9.1	5.3	1.007	0.2



(a) Ratio of Singles count rates



(b) Ratio of PSMC measured α-values

Figure 7.10 Ratio of Singles count rates and PSMC measured α-values before and after stabilization

Item T161A shows the most significant change in Pu mass, with a reduction of a factor of two. This can be attributed primarily to the very high-Singles count rate due to the combination of high α-value and Pu mass, responsible for the significant accidental contribution to measured coincidences. As discussed earlier, very high rates of accidental coincidences significantly impact the uncertainty of measured correlations (Doubles and Triples). Even after stabilization, the α-value for item T161A is considerably higher than the other smaller items. The average Doubles-over-Singles (D/S) ratio for the four smaller items after stabilization corresponds to 0.07 but is a factor of two lower for T161A, indicating a very high-Singles count rate compared with the Doubles count rate for this item, resulting in increased accidentals compared with other items.

Based on the above results, the key effects of the stabilization process on the PSMC results can be summarized as follows:

- Significant reduction in the α-value, on average by a factor of 3
- Minimal change in multiplication (note that multiplication might change when multiple sludge items are mixed)
- Improving Pu mass uncertainty

7.1.3 Sludge items after stabilization

A dataset of 31 stabilized sludge items was provided to the LANL for a broader evaluation of the PSMC performance for this type of material. The previous section discussed the impact of the stabilization process in more detail, and this section focuses on the overall performance and applicability of the PSMC to assay this type of material. The standard stabilization procedure involved mixing of multiple items into a single product (Figure 7.11). As concluded in the previous section, improved performance is anticipated compared with items before stabilization due to the reduction in the α-value.

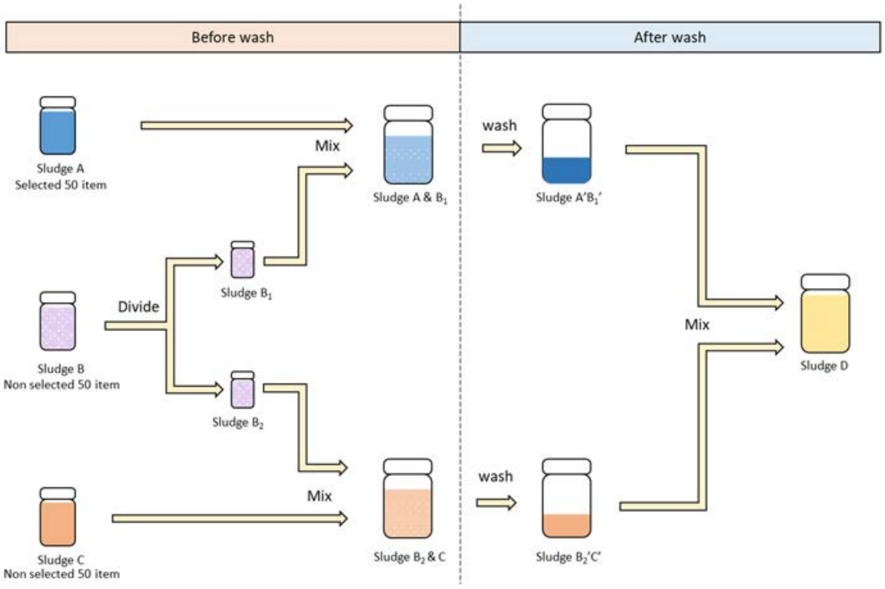


Figure 7.11 Schematic material flow in the routine sludge stabilization process

Table 7.6 shows an overview of 31 items evaluated by the LANL. The measurements were performed using the PSMC in multiplicity mode. All items were stored in metal cans (Figure 7.9), and the JAEA independently calibrated the PSMC with the new metal can, confirming the validity of existing PSMC parameters. As described in the previous section, the LANL recommended the use of a reduced predelay setting (2.5 µs) for the measurement of sludge items after stabilization. However, based on the provided International Neutron Coincidence Counting software (INCC) results, approximately 1/2 of the 31 sludge items used the original PSMC operating parameters listed in Table 7.2. (i.e., 4.5 µs predelay). The LANL reevaluated the data to ensure consistency in the PSMC parameters, as outlined in Tables 7.2 and 7.4, and removed the normalization correction factor, as detailed in [6]. The plots below include data from both PSMC settings. As explained in [3], the choice of predelay has no impact on the assay mass; instead, it improves the overall measurement uncertainty.

Table 7.6 Overview of the 31 sludge items after stabilization provided to the LANL for evaluation

Item No.	Item ID	Item No.	Item ID
1	T078A	17	T183A
2	T056A2	18	T060A
3	T181A	19	T004A
4	T109A	20	T028A
5	T086A	21	T142A
6	T080A	22	T139A
7	T089A	23	T066A
8	T014A2A	24	T065A
9	T014AB	25	T074A
10	T071A	26	T126A
11	T083A	27	T053A
12	T063A	28	09SS001
13	T111A	29	SNS001A
14	T087A	30	17B-05A
15	T042A	31	PNS004A
16	T018A	—	—

The first evaluation of sludge items after stabilization focused on residual moisture content. Although low moisture is expected due to stabilization, it is recommended to confirm with a dedicated AAS measurement. Figure 7.12 shows these measurement results. The ^{252}Cf perturbation corresponds to <1% on average, confirming a low moisture content. Note that the AAS results are shown for only a subset of items measured using a 4.5 μs predelay setting to correspond to the original ^{252}Cf AAS measurement, also performed using this predelay value.

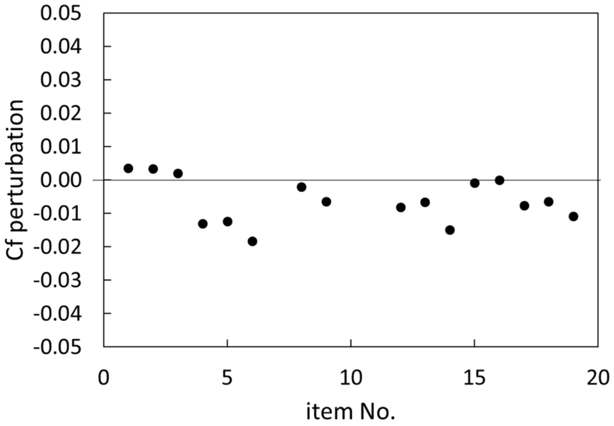


Figure 7.12 Californium perturbation for sludge items measured after stabilization (the error bars are smaller than the symbol sizes)

Figures 7.13 and 7.14 show the α-value and multiplication results for 31 stabilized items. An increase in multiplication compared with the 50 items before stabilization is observed. This is a consequence of mixing multiple items into a single final product, increasing the total Pu mass in the stabilized sludge. As expected, a significant reduction in the α-value compared with the 50 items before stabilization is observed for all stabilized items, corresponding to a factor of more than three.

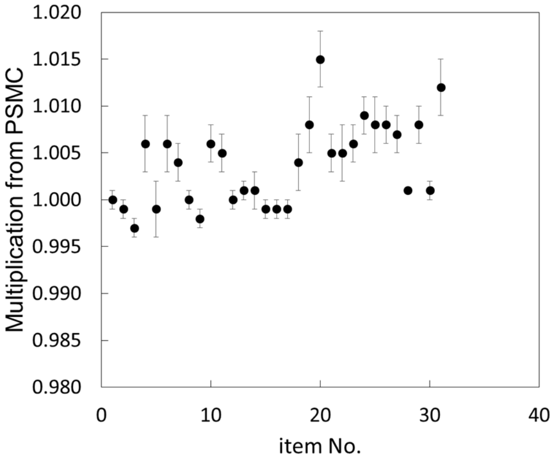


Figure 7.13 Multiplication extracted from the PSMC assay for 31 sludge items after stabilization

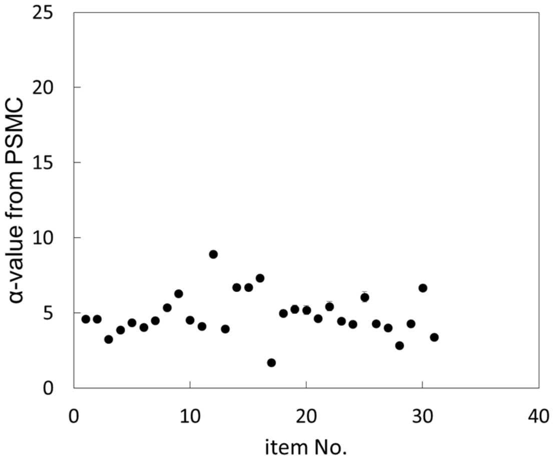


Figure 7.14 α-value extracted from the PSMC assay for 31 sludge items after stabilization

Figure 7.15 shows a comparison of the PSMC mass with the DA results. Note that the minimum Pu concentration limit for the DA (XRF) analysis is 0.02 g/L under the condition of PCDF analysis section. For concentrations below this value, the DA values were set to 0. For a better overview of the differences between the PSMC and DA, Figure 7.16 summarizes the PSMC/DA ratio. The grouping of items with a low PSMC/DA ratio (circled in blue) corresponds to the items with DA set to 0. Item numbers below ~20 were measured using original PSMC settings (Table 7.2), whereas item numbers >20 used reduced predelay settings and parameters from Table 7.4.

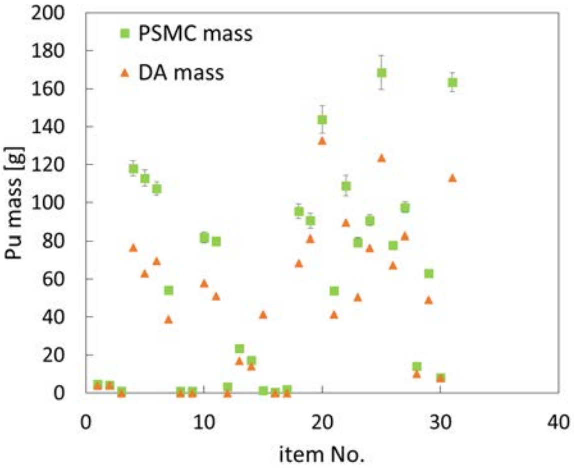


Figure 7.15 Pu mass versus item number measured using the PSMC and extracted from DA for sludge after stabilization

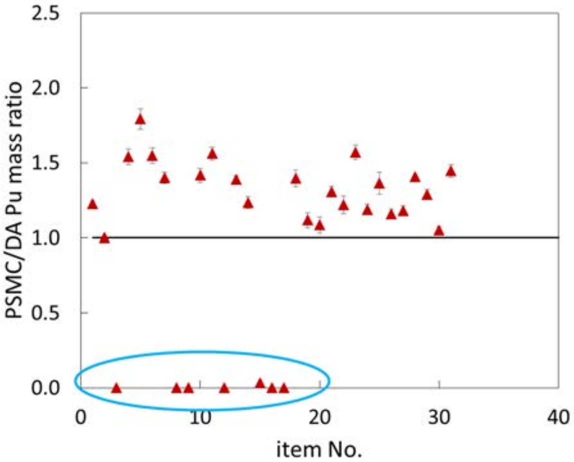


Figure 7.16 Ratio of PSMC versus DA determined Pu mass for sludge after stabilization
Note: The grouping of items with circled in blue corresponds to the items with DA set to zero.

The results in Figure 7.16 indicate a bias in the PSMC/DA results of ~30%. This is a significant discrepancy that merits further evaluation. The following sections detail the additional efforts by the JAEA and LANL to identify the reasons for such a discrepancy. The DA and NDA methodologies were thoroughly evaluated. The evaluation focused on (a) the DA methodology and use of XRF, (b) isotopic composition impact on the NDA results, and (c) validity of the PSMC operating parameters and multiplicity techniques for sludge. These three areas are further discussed.

7.2 Evaluating DA methodology

The JAEA uses XRF to obtain Pu concentration in sludge items in combination with weighing to extract total Pu mass. This technique is labeled DA in this document. The LANL evaluation focused on several aspects of XRF on the sludge matrix that could potentially contribute to the discrepancies in the DA results. Based on a review of XRF results provided by the JAEA presented during a joint technical meeting in March 2021, the provided results exhibited potentially diverging trends, prompting the assessment of XRF performance in more detail to eliminate plausible yet improbable scenarios of how the bias between the PSMC and XRF might be caused.

The JAEA conducted a detailed evaluation of the XRF methodology using a standard sludge prepared by them and one realistic sludge item (T086A). In both cases, the evaluation focused on comparing XRF with IDMS to validate the XRF approach. The standard sludge results confirmed a good agreement between the XRF and IDMS results, with an overall difference of less than 3% and good reproducibility of XRF results. However, the analysis of T086A, a subsample, revealed excellent agreement with IDMS (less than 4%). The other subsample extracted from the same sludge item exhibited XRF 25% lower than IDMS. This result could be explained by the heterogeneity of the T086A sludge item. However, six more subsamples extracted from item T086A and measured using XRF only (no IDMS data are available for these subsamples) exhibited a trend in XRF with time. The XRF results monotonically increased the mass prediction with the measurement date. The difference between the two measurements with a maximum separation in time (approximately 1 year) results in a discrepancy of ~26%. Such a discrepancy could point to potentially drifting XRF calibration. Overall, two trends are observed in the XRF evaluation results, one that could be potentially explained by the heterogeneity of items and one that suggests systematic drift in the XRF results. Both effects are of similar order as the observed NDA/DA discrepancy; therefore, further evaluation focused on several key aspects that could potentially contribute and help confirm or rule out these trends:

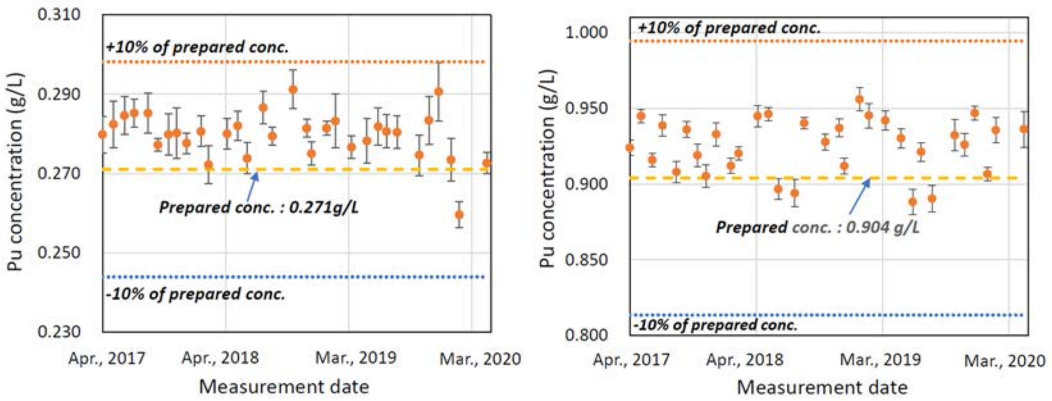
a) drifting XRF efficiency calibration
b) biased XRF efficiency calibration
c) potential heterogeneity

To further evaluate these aspects, the JAEA provided additional information:

- XRF calibration over an extended period with images of raw calibration spectra for primary and secondary standards.
- Images of raw XRF spectra of T086A measurements.
- Description and photographs of physical properties of the containers used to hold the calibration standards and/or sludge sample solutions during the XRF assay.

The additional information concluded the following:

- No evidence of drifting XRF efficiency was observed. Figure 7.17 shows the Pu concentration measurements of two Pu standards over approximately 3 years. No clear drift is evident, and the XRF efficiency seems constant over that period. However, a small (2-3%) calibration seems to be apparent in both instances, for the 0.271 and 0.904 g/L standards. This bias is well within the control range (±10%), declared by the JAEA; hence, it did not trigger corrective action. However, note that this bias is in the opposite direction than between DA and PSMC.



(a) Pu concentration: 0.271 g/L (b) Pu concentration: 0.904 g/L

Figure 7.17 The results of the Pu concentration measurements for two standards items by XRF

- An image where energy calibrated the XRF spectra of measurements of four different Pu standards are superimposed over one another together with one of the measurements of the sludge sample T086A (Figure 7.18). This image indicates a linear relationship between the Pu concentrations and the Pu-Lα peak size but also raises questions about the potential interference with other elemental components in the sludge sample but not in the calibration standards. Those unique peaks were identified as the components of stainless steel (i.e., Fe, Cr, Ni) in sludge. It does seem that the ROI around the Pu peak at 14.3 keV is clear of any interference from unidentified elements within the T086A case. Moreover, if any unidentified element contributed to the Pu-Lα peak's area at 14.3 keV, it would falsely increase the Pu concentration, i.e., contribute to the increased disagreement between the DA and PSMC measurements. Thus, we conclude that the direct yet unidentified contribution of elements unique to the sludge samples but missing in the calibration standards is unlikely to cause the discrepancy between the DA and PSMC results. However, complete analysis of the XRF spectra of the Pu sludge is required to fully understand the potential interference (if any) of any unidentified element unique to the sludge and absent in the calibration standards.

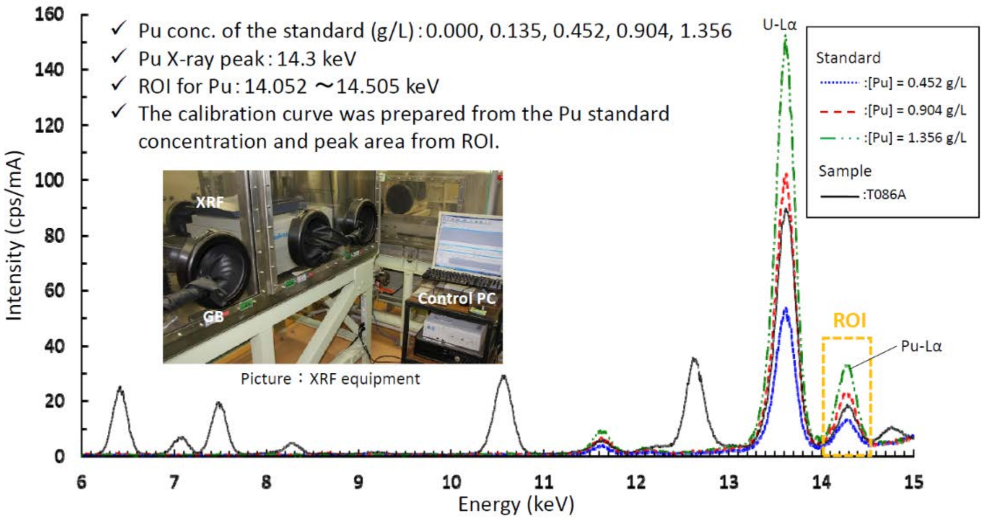


Figure 7.18 XRF spectra of four calibration standards and one of the T086A samples

- Because XRF calibration is performed to the state-of-the-art standard and in a sufficient quality regarding the associated task, an independent review of the sludge measurements themselves is proposed to help rule out systematic errors associated with the difference between the sludge sample and calibration standard.

Overall, it does not seem that XRF calibration by itself could cause any major discrepancy between the DA and PSMC measurements. The measurements and XRF calibration seem to be stable over the relevant time (~3 years), with the apparent bias being too small and of the opposite magnitude than between the DA and PSMC measurements. However, as the XRF measurements available for independent evaluation by the LANL reveal elements unique to the sludge and absent in the calibration standards, an independent review of sludge measurements by providing raw XRF spectra is required for further investigation.

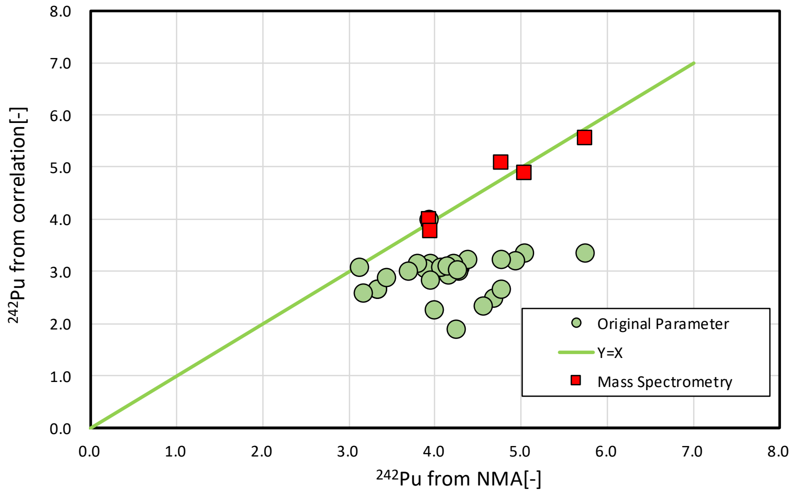
Since no direct evidence of the observed NDA/DA bias was identified in the DA methodology evaluation, the following sections focus in more detail on the evaluation of key components affecting quantitative NDA results.

7.3 Influence of isotopic composition

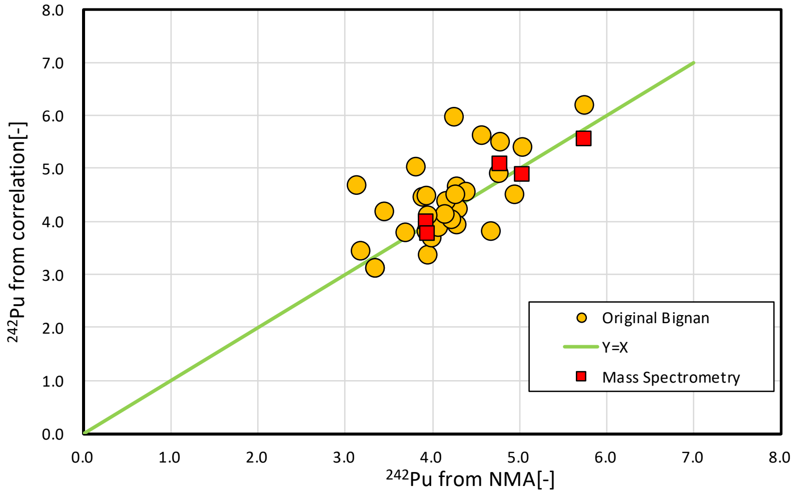
The PSMC NDA measurements rely on two key components provided by the user—PSMC operating parameters and isotopic composition. This section focuses on the impact of isotopic composition. The validity of the PSMC operating parameters was evaluated in earlier reports [2] and [3] and will be further assessed in greater detail in Section 7.4 of this document.

The JAEA conducted an additional evaluation using HRGS to provide isotopic composition input for the PSMC measurements. This evaluation identified underestimating ^{240}Pu effects using MGA analysis as a potential contributor to the observed bias. The LANL performed a detailed evaluation of the MGA parameters and, based on isotopic composition information obtained from the JAEA, established an improved correlation using DA, stream averages, and HRGS data. Figure 7.19 shows the analysis.

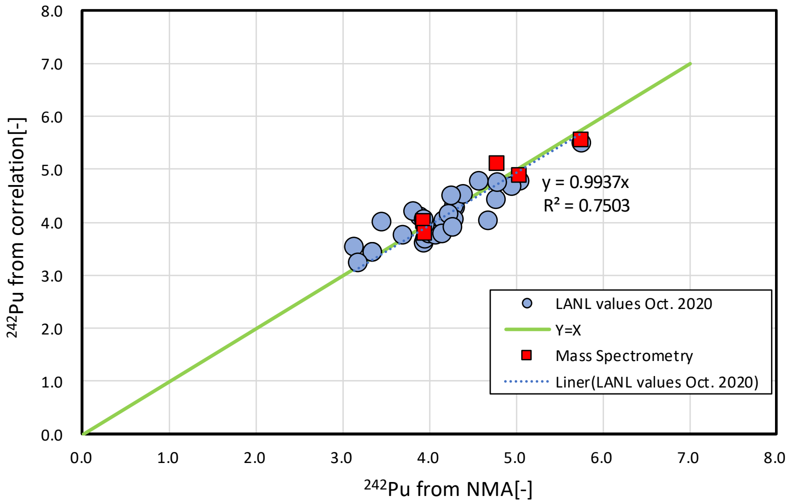
Figure 7.19 (a) shows the correlation between ^{242}Pu extracted using the original MGA analysis and stream average compared with the DA (Mass spec.) versus stream average correlation. Whereas the ^{242}Pu concentration extracted from the DA corresponds excellently to the stream average, the MGA correlation underestimates the stream average and DA values. Consequently, the LANL recommended the use of Bignan coefficients for LWR and PWR assemblies corresponding to C_1 = ~1.3, C_2 = 0.33, and C_3 = 1.7 [7]. Figure 7.19 (b) shows the resulting ^{242}Pu concentration compared with the stream average and DA. This new correlation resulted in a significant improvement, however, with a broad spread of extracted ^{242}Pu values. The LANL, therefore, further refined the C_1, C_2, and C_3 coefficients using the known correlation between DA and stream average ^{242}Pu concentration. The final coefficients (C_1 = 4.40, C_2 = 1.016, and C_3 = 0) resulted in a significant improvement (Figure 7.19 (c)) and were adopted in subsequent MGA analysis by the JAEA.



(a) Original MGA correlation used by the JAEA



(b) initial LANL recommendation using standard Bignan coefficients



(c) LANL updated MGA correlation based on the DA and stream average values provided by the JAEA

Figure 7.19 Overview of ^{242}Pu content extracted using MGA correlation with different correlation parameters

Overall, this modification of the MGA parameters based on the first 31 datasets and, in total, 54, including additional datasets, reduced the NDA/DA bias from ~30% to ~20% (Figure 7.20). To further evaluate the remaining bias, a detailed review of the PSMC operating parameters and validity of multiplicity counting assumptions was performed, as described in the following section.

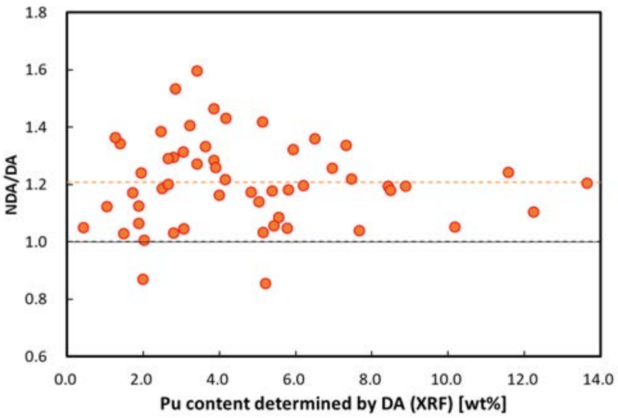


Figure 7.20 JAEA reanalysis of 54 stabilized sludge items using the updated MGA parameters

7.4 Evaluating PSMC operating parameters

This section reviews the PSMC operating parameters and the range of aspects underlining the assumptions of multiplicity counting to evaluate whether any remaining biases can be explained. As mentioned in Section 7.2, the second key inputs affecting the quantitative PSMC multiplicity assay results are the PSMC operating parameters. To obtain correct results from multiplicity assay, the instrument must be correctly calibrated to the nature and form of material to be assayed. The details of the multiplicity calibration procedure are outlined in [4] and include the determination of key instrument operating parameters, such as efficiency, gate fractions, deadtime coefficients, and HV setting. All these parameters were evaluated before starting the sludge measurement campaign, and the details are outlined in [2] and [3]. The evaluation confirmed that no change in the PSMC operating parameters was required for use with sludge items. Tables 7.2 and 7.4 show the currently used operating parameters. These evaluations involved the measurement of a small MOX standard inside a sludge-like matrix to resemble realistic sludge material composition.

However, the observed NDA/DA bias prompted an additional thorough assessment of PSMC calibration to ensure that the current operating parameters represent the correct settings for high-alpha sludge items and confirm the validity of point-model assumptions for sludge measurements.

To validate the operating parameters and overall applicability of the multiplicity counting (point-model) concept, we conducted a step-by-step assessment of material characteristics unique to sludge items fundamentally different from standard materials, such as pure Pu materials, for which the multiplicity assay works well. Contrary to pure Pu material, sludge items present several key characteristics, making them fundamentally different, including the following:

- High-level impurities, resulting in a high α-value
- Large volume, occupying a large fraction of sample cavity
- High gamma dose

The first aspect is the most important and requires careful evaluation of the PSMC operating parameters and overall approach. It can affect the sludge assay in three ways: (1) *via* neutron scattering through the sludge matrix, (2) *via* additional neutrons created in (α, n) reactions, and (3) *via* higher deadtime corrections. The first effect can impact the neutron energy spectrum emitted from the sludge, affecting the efficiency of neutron detection. The second effect contributes to increased neutron production, which is correctly accounted for in point-model equations [4] but can result in a shift in the neutron energy spectrum due to different energies of neutrons emitted from (α, n) reactions than spontaneous fission. Finally, the impact of deadtime on the measurement results was evaluated for completeness.

7.4.1 Impact of neutron scattering

To evaluate the impact of neutron scattering, the JAEA conducted a dedicated calibration with a 10 gPu MOX standard surrounded by a representative (nonnuclear) sludge-like matrix. Figure 7.21 shows the MOX configuration inside the standard sludge metal bottle. Two measurements were performed: the first one only involved bare MOX inside the empty metal bottle, and the second one involved MOX inside a sludge-like matrix. The metal bottle was placed inside the PSMC to measure the Pu mass for these two configurations. These measurements confirmed that the response of the PSMC was unaffected by the sludge matrix, confirming that neutrons emitted from Pu are not significantly affected by interactions of neutrons with the sludge matrix, confirming the standard PSMC detection efficiency. Table 7.7 summarizes the results of this evaluation.

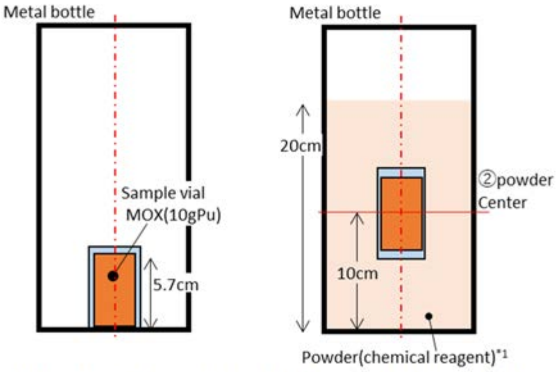


Figure 7.21 A drawing of the location of the MOX standard inside the sludge metal bottle for the PSMC calibration measurements
(The left figure shows the MOX surrounded by a sludge-like matrix.)

Table 7.7 PSMC response results for bare MOX and MOX surrounded by a sludge-like matrix

Material	Singles [cps]	In sludge/bare
Bare MOX (10 gPu)	4702.394	—
MOX in sludge	4693.662	**0.998**

7.4.2 Impact of sample volume

Additional aspect that might affect the PSMC performance for sludge measurements is that sludge is stored in 2 L bottles (Figure 7.9), occupying a large volume inside the PSMC sample cavity. Additionally, Pu is distributed throughout the bottle volume, contrary to the MOX calibration, where the material is localized inside the small MOX can volume. The distribution of Pu could modify efficiency due to neutrons coming from different regions within PSMC and PSMC efficiency variation over the sample cavity.

To investigate the first aspect, LANL performed a series of dedicated Monte Carlo N-Particle transport code (MCNP) simulations with the following:

a) a bare MOX can (10 gPu, ~6 cm tall cylinder, ~2 cm diameter, Figure 7.8)
b) MOX (10 gPu) inside a sludge matrix (Figure 7.8 left)

c) 10 gPu distributed throughout the sludge matrix

The definition of the sludge composition was derived from the information provided by the JAEA, shown in Table 7.8 for the MCNP model.

Table 7.8 Definition of the sludge composition used in the MCNP model

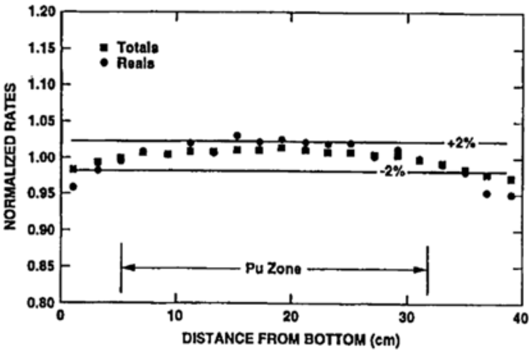
Element	wt [%]	Element	wt [%]
Pu	5.26	Fe	53.31
U	6.98	O	31.16
Na	2.04	N	1.24

These simulations confirmed that the distributed Pu source negligibly affects the PSMC performance, and no adjustment of the PSMC operating parameters is necessary. Table 7.9 shows the results of this MCNP evaluation.

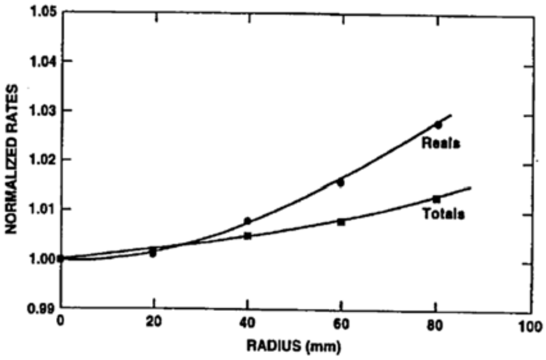
Table 7.9 Results of the MCNP evaluation of the PSMC response to various distributions of Pu inside the PSMC cavity

Material	**MCNP source**	**Efficiency**
Bare MOX (10 gPu)	SF inside MOX	54.52%
MOX in sludge (10 gPu)	SF inside MOX	55.00%
U/Pu sludge (10 gPu)	SF distributed	54.23%

The second aspect related to sludge volume that could affect the PSMC performance is changing the neutron detection efficiency as a function of the neutron emission position within the sample cavity. This aspect is typically considered in the design stages of each instrument, and effort is made to minimize any position dependence of instrument response to support the measurement of large-scale items. This aspect was evaluated for the PSMC, and the results illustrated in Figure 7.22 confirm minimal variation (<2%) of PSMC efficiency across the volume corresponding to the sludge material (~11 cm diameter and 30 cm height) [8].



(a) axial direction profile



(b) radial direction profile

Figure 7.22 Variation of the PSMC measurement response

7.4.3 Contribution from (α, n) reactions

The contribution from (α, n) reactions from interactions within the sludge is absent in the MOX calibration measurements because α particles do not penetrate into the MOX container. The (α, n) reactions will have two main effects:

a) Increase in Singles and correlated (Doubles and Triples) count rates
b) Generation of neutrons with a different energy than spontaneous fission (i.e., two-energy source term)

Both effects will increase the count rates and could affect the PSMC operating parameters. Dedicated MCNP simulations were performed to study the effect of spontaneous fission (~2 MeV) and random neutron source terms. The main contribution to (α, n) reactions in the sludge is expected from O and Na. The average neutron energy from the (α, n) reactions on O corresponds to ~1.9 MeV [9]. The average neutron energy from Na is expected to be <1 MeV [9]. Therefore, to illustrate the effect of different neutron energy spectra, monoenergetic random neutron source with 1 MeV energy and PuO_2 (α, n) reactions source were assumed in the MCNP evaluation. The MCNP calculations were performed, assuming 10 and 100 g of Pu to represent the range of Pu masses observed in the sludge items. Table 7.10 shows the results of this evaluation. Table 7.10 confirms that the neutron energy spectrum and the contribution of (α, n) reactions neutrons have a minimal impact on the expected PSMC performance.

Table 7.10 Comparison of the MCNP predicted ^{240}Pu effective mass for various neutron energy spectra with declared ^{240}Pu effective masses

Sample configuration	Source	Singles [cps]	Doubles [cps]	Triples [cps]	MCNP ^{240}Pu$_{eff}$ [g]	Declared ^{240}Pu$_{eff}$ [g]	Mass ratio MCNP/ declared [−]
U/Pu sludge (10 gPu) only SF	SF distributed	2232	740	132	4.38	4.32	1.01
U/Pu sludge (10 gPu) SF + alpha	1 MeV random neutrons	14473	743	132	4.17		0.96
U/Pu sludge (100 gPu) only SF	SF distributed	22363	7493	1385	43.67	43.24	1.01
U/Pu sludge (100 gPu) SF + alpha	1 MeV random neutrons	144926	7778	1423	42.40		0.98
U/Pu sludge (100 gPu) SF + alpha	PuO_2 (α, n) spectrum	129421	7819	1435	42.77		0.99

7.4.4 Effect of gamma doses

Some sludge items measured in the PSMC exhibited extremely high gamma doses. Although the original PSMC evaluation included measurements of HV plateaus [2] and confirmed no gamma sensitivity of PSMC for standard operating HV even for the highest dose item measured, the subsequent evaluation revealed a potential trend of NDA/DA discrepancy with increasing gamma doses (Figure 7.23).

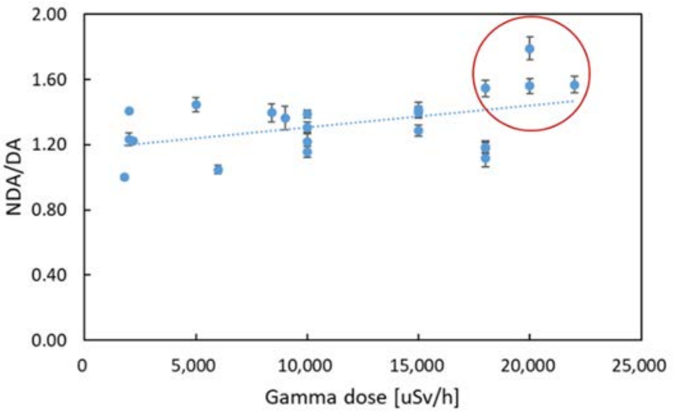


Figure 7.23 NDA/DA ratio as a function of gamma dose for ranges of stabilized sludge items
Note: The grouping of items with red circle were selected for this detail evaluation of the influence.

To fully evaluate this aspect, the JAEA performed a series of dedicated measurements for the set of the highest dose items (red circle in Figure 7.23). During this evaluation, the items were remeasured in the PSMC with an additional 1-mm Pb shielding. Consequently, the dose reduced from ~20,000 to ~100 µSv/h (i.e., well within the ^{3}He tube operating range). These measurements confirmed no impact of gamma dose on the PSMC measurements (Figure 7.24).

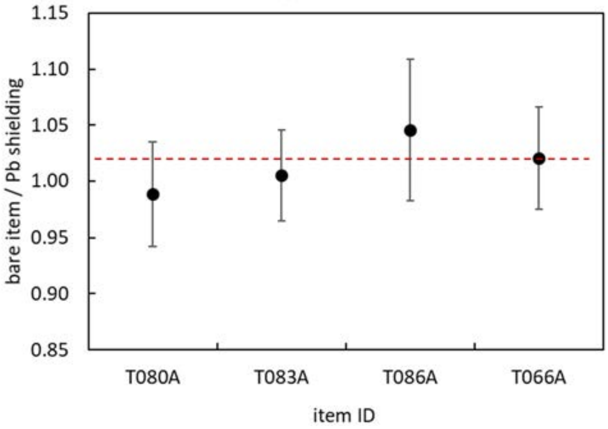


Figure 7.24 Ratio of Pu mass measured in the PSMC for bare sludge items and items shielded with 1-mm Pb

7.4.5 Impact of PSMC deadtime

The JAEA standard PSMC deadtime parameters listed in Table 7.4 were used in this analysis. However, these parameters are different from the recommended PSMC operating parameters based on [8], which are generally adopted deadtime coefficients for the PSMC instruments. Since sludge items represent a broad range of count rates, including count rates of more than 100,000 counts per second, the contribution of deadtime correction is expected to be sizable. Therefore, an independent evaluation was conducted to compare the JAEA standard PSMC deadtime coefficients with those recommended by the PSMC manual. This evaluation included a subset of 12 stabilized sludge items that cover the full range of count rates encountered in the sludge measurements, providing a complete overview of the expected deadtime trends. Table 7.11 summarizes the JAEA standard and PSMC manual deadtime coefficients for reference. Figure 7.25 shows the evaluation results in the form of the ratio of Pu mass obtained assuming PSMC manual deadtime coefficients versus Pu mass obtained assuming JAEA standard deadtime coefficients. Figure 7.25 demonstrates that changing deadtime coefficients (although the individual coefficients differ by ~8%) negligibly affect the PSMC assay results for the entire range of count rates expected from stabilized sludge. This provides additional validation of the JAEA standard deadtime coefficients for sludge item assay.

Table 7.11 Overview of the JAEA standard and PSMC manual deadtime coefficients

Deadtime coefficient	JAEA standard	PSMC manual
A	0.379	0.409
B	0.123	0.132
C	0.850	0.000
Multiplicity	112	121

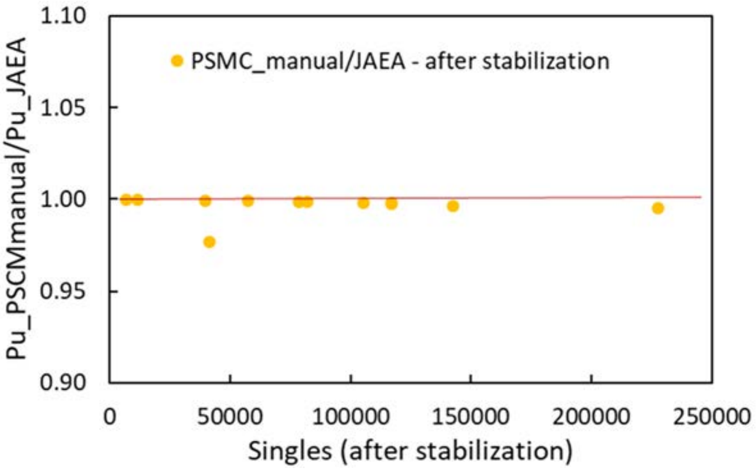


Figure 7.25 Ratio of Pu mass using PSMC manual based deadtime coefficients versus standard JAEA deadtime coefficients

7.4.6 PSMC parameter conclusions

Based on the results shown in previous sections, there was no impact on various aspects of sludge materials fundamentally different from the MOX standard used for the PSMC calibration. The evaluation focused on key aspects that could impact the PSMC response and, as such, its operating parameters. Since no significant impact was observed, the previous sections confirm the validity and provide justification for use of original PSMC operating parameters for sludge item measurements.

7.5 Applicability of the PSMC to sludge NDA

Previous sections demonstrated the applicability of routine PSMC parameters to sludge materials and did not reveal any significant effects contributing to the observed NDA/DA bias. Therefore, this PSMC NDA provides sludge assay results that are consistently ~20% higher than expected from DA. Since neither detailed analysis of DA nor NDA methodology revealed any direct evidence of such bias, this section will focus on demonstrating PSMC applicability and suitability for sludge NDA as a standalone method based on a series of additional tests and evaluations.

Here, we focus on two aspects:

- PSMC performance for MOX, standard sludge, and all other sludge items
- Validity of point-model assumptions and the multiplicity counting approach

As outlined in previous sections, the PSMC operating parameters were calibrated using a 10 gPu MOX standard surrounded by a sludge-like matrix. However, contrary to sludge items, the PSMC performance for the MOX standard is in very good agreement with the DA value. To provide additional confidence in the PSMC performance for sludge material, the JAEA prepared a small standard sludge (26 g total mass, 8 gPu). The sludge standard was assayed in the PSMC using its standard operating parameters and evaluated using DA. The PSMC results for this standard sludge are in a very good agreement with the DA result (Table 7.12), providing additional confidence in the PSMC performance for a material closer to the realistic sludge. However, some aspects are unique to realistic sludge items warranting additional consideration.

Table 7.12 Comparison of PSMC-measured (NDA) and DA mass for MOX standard in sludge-like matrix and standard sludge

Item	Declared Pu mass [g]	PSMC Pu mass for regular fd, ft [g]	NDA/DA
MOX in sludge	10.00	9.90	0.99
Standard sludge	8.54	8.40	0.98

The extent to which the point-model assumptions are valid for such an unusual material with low Pu concentrations in a light element matrix with a significant contribution of the (α, n) reactions is critical. Therefore, we focused on the key element that drives such items away from the point-model assumptions, which is the contribution from the (α, n) reactions. Note that all sludge items have low multiplication due to a low Pu concentration in the sludge matrix; therefore, multiplication will play a minor role in the point-model assumptions. The topic of deviation from the point-source approximation was addressed in Section 7.4.2, and evidence of uniform PSMC responses, independent of the source location, was provided. In addition, large-volume materials are routinely assayed in the PSMC during the LANL training activities, and the assay results are in good agreement with the declared values.

The contribution of the (α, n) reactions and their impact on point-model prediction is the focus of this evaluation. Some aspects of the contribution of the (α, n) reactions were addressed in Section 7.4.3 and focused on the differences in the energy spectrum of neutrons originating from the (α, n) reactions. An alternative approach to directly evaluate the emitted neutron energy spectrum is the use of a ring ratio [4]. This method uses information from the inner and outer rings of a neutron multiplicity counter. The ratio of the inner-to-outer ring depends on the neutron energy and typically increases with the decrease in neutron energy. Unfortunately, the PSMC does not provide independent outputs from individual rings. Therefore, to assess the neutron energy spectrum from sludge items compared with MOX standard and standard sludge, dedicated measurements were performed by the JAEA using the Fast Carton Assay System (FCAS) counter, including this capability [10]. Figure 7.26 shows the results and illustrates a significant difference between the ring ratio of MOX standard (green data point) and sludge items (blue data points). The red data point represents a standard sludge. Figure 7.26 clearly demonstrates that sludge items have a different neutron energy spectrum than the MOX standard. Simultaneously, it shows that the ring ratio of standard sludge is similar to the realistic sludge items. This is additional evidence that the standard sludge provides a good representation of a key feature unique to the sludge material, namely, the neutron energy spectrum. For completeness, Figure 7.27 shows a comparison of the FCAS Pu mass results with the PSMC. Note that standard FCAS operating parameters were used in this measurement. From Figure 7.27, FCAS confirms the NDA results obtained from the PSMC.

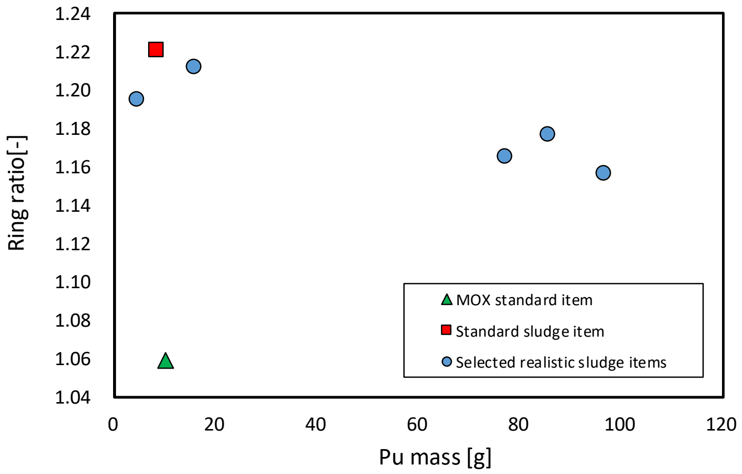


Figure 7.26 Ring ratios for the MOX standard, standard sludge, and selected realistic sludge items measured in FCAS

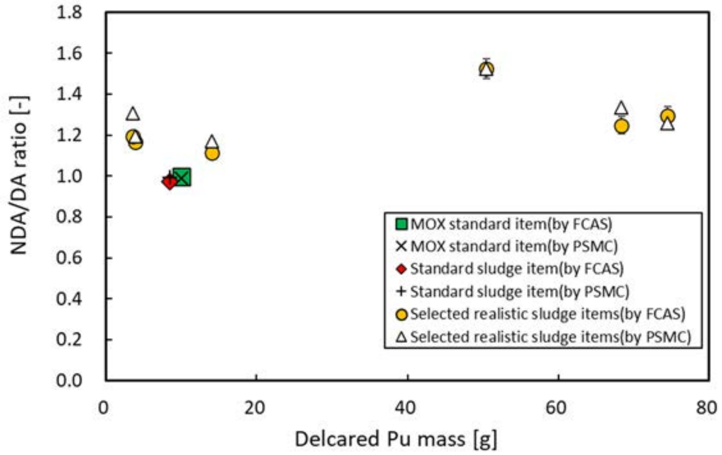


Figure 7.27 Comparison of the NDA/DA ratio for the PSMC and FCAS for the MOX standard, sludge standard, and selected realistic sludge items

Despite this agreement, it is important to realize that the sludge, including standard sludge, ring ratio is significantly different from the MOX standard, demonstrating different energy spectra for neutrons originating from the MOX standards and sludge materials. This is an important observation because changes in the neutron energy spectra are typically linked to changes in the neutron detection efficiency, a key PSMC operating parameter. Figure 7.28 shows the typical PSMC energy dependence on neutron detection efficiency. As discussed in Section 7.4.4, the light elements contained in the sludge material are expected to produce neutrons with energies lower than spontaneous fission. The corresponding expected PSMC response is increased neutron detection efficiency.

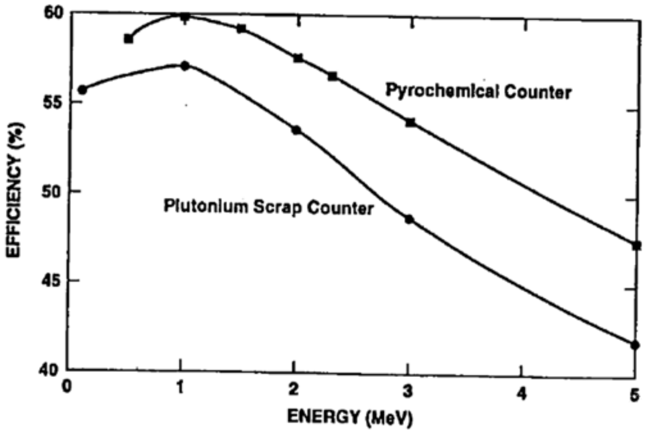


Figure 7.28 PSMC efficiency profile as a function of energy

Section 7.4.1 and 7.4.3 indicated that change in neutron energy spectra due to the sludge matrix is not expected to affect the PSMC performance and efficiency. However, to underscore this point, we conducted an additional parametric study. This study uses a dual-energy point model, which was developed specifically to address the issue with point-model assumptions failing for high-alpha items due to the differences in neutron energy spectra between spontaneous fission and (α, n) reactions neutron [11]. The dual-energy point model essentially breaks the point-model equations into two subcomponents, depending on the neutron energy. The contributions to measured Singles, Doubles, and Triples from spontaneous fissions and (α, n) reaction neutrons are separated to appropriately account for increased S, D, and T due to the (α, n) reactions neutron detection efficiency and associated induced fissions. Ref. [11] illustrates this effect for 1 kgPu oxide items with moderate impurities (α up to 3) and shows a comparison of the bias caused using standard point-model assumptions for such items for several neutron multiplicity counters, including the PSMC. For the PSMC, the bias is <8% for the 0-2 MeV energy range.

To address this aspect, the LANL conducted a bias study for sludge-like materials encountered in the present measurements. The study involved MCNP modeling to evaluate the neutron detection efficiency to neutrons originating from the (α, n) reactions in a sludge matrix, and using this input, bias calculations were performed, where the S, D, and T rates were calculated for various masses, assuming typical sludge α-values and multiplication based on dual-energy point-model equations. These S, D, and T rates were analyzed to calculate the Pu mass (*m*) using standard point-model equations to evaluate the bias introduced by imposing point-model assumptions on a problem with two different neutron energy spectra. The bias is defined as $(m - m_{ref})/m_{ref}$, where m_{ref} denotes the assumed input mass (Figure 7.29). The dual-energy point model S, D, and T rates were calculated assuming the JAEA sludge item characteristics (typical multiplication of 1.004 and an α-value of 5). The calculation also used PSMC detection efficiency of 58.7%, calculated using the MCNP for 1 MeV neutrons and a probability of 1 MeV (α, n) reactions neutrons to induce fission relative to fission neutrons of 1.05 extracted from [11]. Note that the variation in the neutron detection efficiency within ±5% and neutron energy-dependent-induced fission probability by ~10% introduced a minimal variation in the calculated bias. Figure 7.29 demonstrates that the bias due to additional (α, n) reaction neutrons from JAEA sludge materials is low. This can be largely attributed to the low multiplication of these items, minimizing induced fission contributions of these additional (α, n) reaction neutrons to the measured correlated count rates.

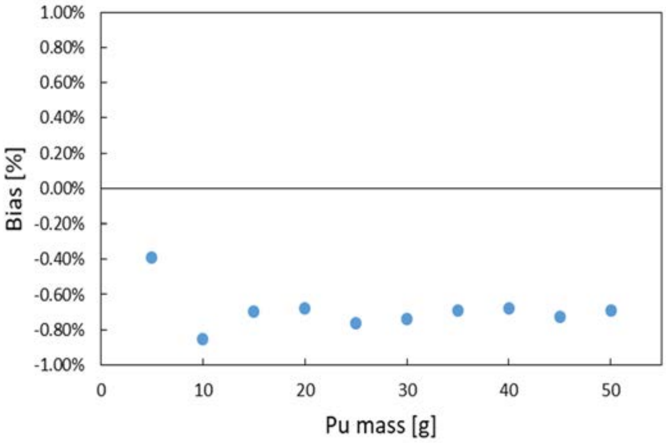


Figure 7.29 Bias in the Pu mass calculated using energy-independent point-model equations on the S, D, and T rates from dual-energy problem representative of sludge items measured in the PSMC

Finally, to complete this evaluation and point-model validation, the dual-energy point-model equations were directly used to solve the measured S, D, and T count rates for the Pu mass. The dual-energy point model is implemented in the INCC and relies on the measured ring ratios to extract the average neutron energy [12]. Since PSMC does not provide ring-ratio information, the FCAS measurements were used to conduct this study. As shown earlier, the FCAS measurements provide NDA assay results in close agreement with the PSMC.

In this evaluation, the FCAS ring input was used with the MCNP calculated efficiency for 1 MeV neutrons in the INCC. The INCC used the FCAS measured ring ratio to extract the corresponding efficiency and calculate the S, D, and T rates based on the dual-energy point model outlined in [12]. These S, D, and T rates were used in the INCC to calculate the Pu mass. Table 7.13 shows the results of this test. In addition, these results demonstrate that the impact of the additional (α, n) reactions contributions and different neutron energy spectra on the standard multiplicity analysis is negligible for the JAEA sludge items.

Table 7.13 Comparison of the Pu mass measured in the FCAS and extracted using the standard point-model and dual-energy point-model equations in the INCC

Item ID	Inner ring Singles [cps]	Outer ring Singles [cps]	Ring ratio [—]	Standard point-model Pu mass [g]	Pu mass from dual-energy point model [g]	Standard point model / dual-energy point model
T078A	1887.5	1577.5	1.20	4.50	4.521	1.004
T087A	10089.5	8319.8	1.21	15.63	15.604	0.998
T0562A	1783.0	1491.4	1.20	4.24	4.241	1.000
T066A	32491.8	27867.2	1.17	76.93	76.903	1.000
T060A	42282.2	35899.1	1.18	85.41	85.311	0.999

In conclusion, the above analysis and evidence demonstrate that the PSMC and standard point-model assumptions are fully applicable to the sludge materials encountered at the JAEA. In addition, because the thorough evaluation of the PSMC parameters presented in Section 7.4 did not reveal any of their modifications due to the sludge materials, this evaluation concludes that the PSMC could provide a reliable, independent NDA assay of sludge materials.

7.6 Measurement uncertainty by the PSMC/HRGS

This section summarizes individual contributions to the TMU for NDA(PSMC). The key contributions include the uncertainty of calibration parameters, deadtime correction, isotopic composition from HRGS, item positioning, and statistical uncertainty. The details of the individual contributions are summarized below. Table 7.14 show the summary of the TMU for PSMC sludge measurements.

Calibration

This component includes uncertainty in the PSMC operating parameters required to calculate Pu mass from the multiplicity assay. The relevant operating parameters include efficiency and Triples gate fraction. These parameters were carefully evaluated and confirmed for the Pu energy spectra using a MOX standard, as discussed in [2] and [3]. The variation in neutron detection efficiency due to the contribution of the sludge matrix was evaluated in Section 7.5 and demonstrated a <1% effect on the assayed mass. The Triples gate fraction calibration measurements demonstrated that varying Triples gate fraction of less than 2% was needed to obtain a <1% agreement with the expected mass. Therefore, an overall uncertainty of 1% is assigned to the impacts of the PSMC calibration.

Deadtime correction

The uncertainty of deadtime correction depends on the used deadtime coefficients. Deadtime coefficients were unmodified during the evaluation of the PSMC operating parameters due to the low mass (i.e., low-count rates) of the MOX standard [2]. Conservative uncertainty on deadtime coefficients of 5% was assumed to assess its impact on the overall assay uncertainty of stabilized sludge items. Impact evaluations of the modification of the original deadtime coefficients (Table 7.2) by ±5% revealed its very small impact on the assay mass, except for the highest mass items (Pu mass > 130 gPu). Overall uncertainty of 0.5% was estimated based on this evaluation.

Isotopics (HRGS)

Isotopic composition uncertainty stems primarily from the ^{242}Pu content uncertainty using HRGS. As discussed in Section 7.3, the MGA parameterization used to extract ^{242}Pu represents the lead contribution to uncertainty and can bias the results. Section 7.3 describes the removal of such bias by appropriately updating the MGA analysis parameters. Following this updated MGA analysis, the JAEA reanalyzed isotopic composition for all stabilized sludge items. The updated isotopic composition values were provided to the LANL along with the DA (Mass spec.) results for several stabilized sludge items. These sludge items, for which the Mass spec. and HRGS results were available, were used to evaluate the overall HRGS uncertainty. Based on this evaluation, an overall uncertainty of 4.5% was established for the HRGS.

Positioning

As discussed in Section 7.4.2, the PSMC count rate exhibits a small variation horizontally and vertically. For the dimensions of the sludge cans, such variation is <2% (Figures 7.9 and 7.22). An uncertainty of 2% in the measured count rates was assumed, and its impact on assay mass was evaluated. The 2% value represents a conservative estimate because item positioning in the PSMC cavity is expected to be well reproducible due to its large volume. Furthermore, any changes in position could result in mutually canceling effects due to an increase in the count rate from some parts of the item, simultaneously compensated by a decrease in the count rate from other item regions (see the trend in Figure 7.22 (a)). Nevertheless, the 2% uncertainty in the measured count rates was used as a conservative estimate, contributing to an overall uncertainty of assay mass of 3.5%.

Statistical

The overall statistical uncertainty of the PSMC assay was evaluated based on the PSMC performance across 31 stabilized sludge items measured in the PSMC. The overall performance for this broad range of items with Pu masses between 1 and 170 g was less than 3%. In cases of 170 gPu in actual PSMC measurements, the statistical part of uncertainty is separately calculated for the TMU.

Table 7.14 Evaluation of the TMU contributions for the PSMC sludge measurements

TMU contribution	Uncertainty [%]
Calibration	1.0
Deadtime correction	0.5
Isotopics (HRGS)	4.5
Positioning	3.5
Statistical	3.0
TMU*	6.5

* The TMU was calculated as $\sigma = \sqrt{\sigma_{calib}^2 + \sigma_{Deadt}^2 + \sigma_{isot}^2 + \sigma_{pos}^2 + \sigma_{stat}^2}$

7.7 Applicability of the Known-M method

Figures 7.3, 7.4 and 7.13, 7.14 show the multiplication and alpha value of the PSMC measurement results by passive multiplicity method of the before and after the stabilization sludge items, respectively. Those results show that the alpha values are high and the multiplication of the sludge items is close to one. Therefore, the applicability of the Known-multiplication(Known-M) method was studied for the sludge items.

LANL carried out a feasibility study of applying the "Known-M" method to the analysis of sludge items, both before and after washing. The items investigated are shown in Table 7.15, which also gives the existing results for the classic passive multiplicity analysis method. (The detection efficiency was taken as 0.543 for all items.)

Table 7.15 Items selected for Known-M feasibility study

Item ID	Date	Time	fD [-]	fT [-]	M [-]	α [-]	Pu mass [g]
T109A	February15, 2019	10:36	0.615	0.400	1.006	3.896	117.9 ± 4.0
T083A	September 3, 2019	13:30	0.643	0.425	1.005	4.110	79.7 ± 2.3
T066A	August 6, 2019	16:48	0.643	0.425	1.006	4.461	79.2 ± 2.5
T028A	July 17, 2019	11:44	0.643	0.425	1.013	5.397	139.6 ± 7.1
T074A	August 26, 2019	16:37	0.643	0.425	1.008	6.071	168.6 ± 9.0
A123T161	June 26, 2018	16:27	0.615	0.400	1.007	16.341	138.2 ± 12.0
A123T175	June 27, 2018	16:47	0.615	0.400	1.004	6.542	54.1 ± 0.9
A123T110	March 15, 2018	14:44	0.615	0.400	1.000	13.428	32.9 ± 1.4
A123T143	March 26, 2018	15:31	0.615	0.400	1.001	11.463	64.1 ± 3.0

7.7.1 Known-M relationship

The Known-M method is based on a known relationship between the mass of $^{239}\mathrm{Pu}_{\mathrm{eff}}$ of the item and the neutron multiplication. The $^{239}\mathrm{Pu}_{\mathrm{eff}}$ of an item is given by:

$$\begin{aligned} {}^{239}Pu_{eff} = m_{239} = 0.786\,{}^{238}Pu + {}^{239}Pu + 0.515\,{}^{240}Pu + 1.414\,{}^{241}Pu \\ +0.422\,{}^{242}Pu + 0.545\,{}^{241}Am + 0.671\,{}^{235}U + 0.082\,{}^{238}U \end{aligned} \tag{7.2}$$

(Note that in INCC, the calculation does not include the two U isotopes). The multiplication of a representative sludge sample was calculated using an MCNP simulation. The sample was 1,873.12 cm^3 with bulk density 1.0 g cm^{-3}, with a $^{239}\mathrm{Pu}_{\mathrm{eff}}$ mass of 97.99 g. The multiplication was 1.0051 (with a statistical uncertainty of less than 1 in the least significant figure). The shape of the curve of multiplication versus $^{239}\text{Pu}_{\text{eff}}$ mass is not known from this single calculation. However, INCC can only use a polynomial relationship:

$$M = b_{kM} m_{239} + c_{kM} m_{239}^{2} \tag{7.3}$$

and so, as a first estimate, a simple x^2-only relationship was used, with $b_{KM} = 0$ and $c_{KM} = 5.31 \times 10^{-7}$, as shown in Figure 7.30. Note that Y-axis is shown the multiplication minus one.

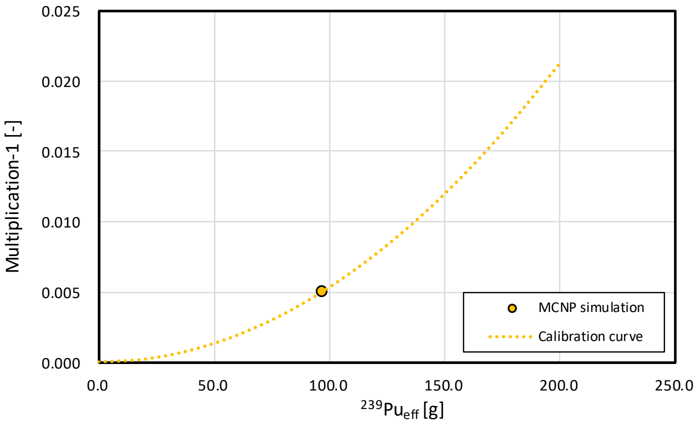


Figure 7.30 Multiplication minus one from single MCNP simulation

7.7.2 Comparison with multiplicity multiplication

It is interesting to compare the MCNP calculated multiplication with that determined by the classic passive multiplicity analysis. The measured multiplication values (from Table 7.15) have been plotted in Figure 7.31 together with the curve fitted from Figure 7.30. The $^{239}\text{Pu}_{\text{eff}}$ values for the measured items were obtained by using the isotopic composition and the measured Pu masses determined by the passive multiplicity method. Figure 7.31 shows that the behavior of the multiplication from the MCNP simulation is very similar to that determined by the passive multiplicity analysis and gives additional confidence in both analysis methods.

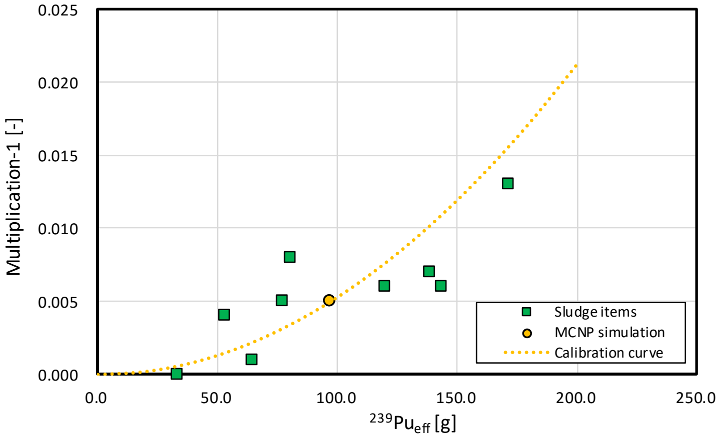


Figure 7.31 Comparison of multiplication from passive multiplicity analysis of individual items and the Known-M relationship from the MCNP evaluation point

7.7.3 Results from first Known-M analysis

The results from the Known-M analysis with $c_{KM} = 5.31 \times 10^{-7}$ are shown in Table 7.16, together with the original multiplicity results that were given in Table 7.15. The differences between the results are given in the final column. They range from -5% to 5%. These results were calculated with Excel and include the effect of the U isotopes.

Table 7.16 Results from Known-M analysis with c_{KM} = 5.31 x 10^{-7} compared to original multiplicity analysis (Table 7.15)

Item ID	Original (Passive multiplicity)			Known-M			Mass change from original [%]
	M [-]	α [-]	Pu mass [g]	M [-]	α [-]	Pu mass [g]	
T109A	1.006	3.896	117.9	1.008	3.92	117.3	-0.5
T083A	1.005	4.110	79.7	1.003	4.00	81.5	2.2
T066A	1.006	4.461	79.2	1.004	4.26	82.4	4.0
T028A	1.013	5.397	139.6	1.012	5.18	144.7	3.7
T074A	1.008	6.071	168.6	1.011	6.36	161.4	-4.3
A123T161	1.007	16.341	138.2	1.009	17.3	130.7	-5.4
A123T175	1.004	6.542	54.1	1.002	6.23	56.4	4.3
A123T110	1.000	13.428	32.9	1.001	13.57	32.6	-1.0
A123T143	1.001	11.463	64.1	1.002	11.79	61.8	-3.6

The Known-M results are very similar to the passive multiplicity results and are distributed both above and below the passive multiplicity values. This gives additional confidence that there is not a systematic bias in either method.

7.7.4. Impact of neglecting U contribution

The Known-M method assumes that the neutron multiplication, M, is a function of the $^{239}\mathrm{Pu_{eff}}$ mass only, with the form listed in Equation (7.3)

Combining the expression in Equation (7.3) with Singles and Doubles in point model equations, we can write:

$$f_M(m_{239eff}) = \frac{\varepsilon f_D M}{2}\left[k_M m_{239} F_0 \varepsilon M F \nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right)\nu_{i2} S\right] - D = 0 \qquad (7.4)$$

Where:

m_{239eff} = mass of $^{239}\mathrm{Pu_{eff}}$
m_{240eff} = mass of $^{240}\mathrm{Pu_{eff}}$
ε = detection efficiency (0.543)
f_D =Doubles gate fraction (0.615)
k_M =$^{239}\mathrm{Pu_{eff}}$/ $^{240}\mathrm{Pu_{eff}}$ (0.345)
F_0 = fission / s / g ^{240}Pu (473.5)
ν_{s2} =2nd moment of spontaneous fission (3.789)
S = item Singles rate (113,607)
D = item Doubles rate (7,329.8)

We call this the Known-M equation. Additionally:

$$\alpha = \frac{S}{m_{240} F_0 \varepsilon M \nu_{s1}} - 1 \qquad (7.5)$$

The Known-M equation can be solved by Newton's method. Table 7.17 shows the results of five samples analyzed by INCC (without U contribution) and with Excel (with and without the U contribution). For the values in the table c_{kM} was taken as an earlier estimate of 5.663 × 10^{-7}. The results from INCC analysis and Excel analysis (without U) differ by 1% or less. The effect of ignoring the U contribution increases rapidly with Pu mass (and multiplication) and the INCC results are systematically higher than the true values.

Table 7.17 Comparison of Known-M method estimated mass with and without U contribution

Item ID	INCC Pu mass [g]	EXCEL Pu mass no U [g]	Excel Pu mass with U [g]	delta [%]
T109A	118.0	118.7	116.7	1.1
T083A	81.8	82.0	81.3	0.6
T066A	82.7	83.0	82.2	0.6
T028A	146.4	147.7	143.5	2.0
T074A	158.3	159.9	154.2	2.7

7.7.5. Known-M feasibility summary

The first point is that the deduced multiplication from passive multiplicity analysis is very similar to that from the MCNP simulation. This gives additional confidence in the passive multiplicity analysis. Secondly, the Known-M analysis approach gives plutonium mass results which are similar to those from the original passive multiplicity analysis. This gives added confidence in the results of both methods.

Thirdly, as the Known-M method uses only Singles and Doubles (and not Triples), the statistical uncertainty on the Pu mass is likely to be less than that of the passive multiplicity analysis. Preliminary numerical estimates suggest a statistical uncertainty of 0.25% for item T109A, compared to a statistical uncertainty of over 3% for the passive multiplicity analysis for the same measurement time. It is important to note, however, that the total measurement uncertainty would also include potential systematic errors from the shape of the multiplication versus $^{239}Pu_{eff}$ mass curve.

However, the INCC Known-M analysis currently does not include an uncertainty analysis on the final Pu mass. Additionally, INCC Known-M implementation does not include U isotopes and as shown in Section 7.7.4, the Pu mass results obtained using INCC in the Known-M mode are systematically higher than those of the complete analysis including U isotopes (done here with spreadsheet software). The passive multiplicity method is therefore confirmed as the more appropriate for Pu mass in those sludge items at the moment.

8. The acceptance test

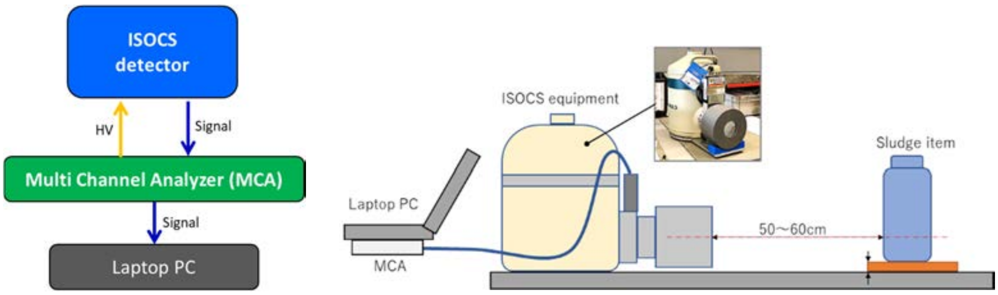
JAEA explained the above re-evaluation results by the Known-M method to IAEA and got an understanding of the results. After that, acceptance test preparation was started. Regarding the confirmation of the PSMC/HRGS capability to determine Pu mass in the sludge items, IAEA suggested to benchmark the PSMC/HRGS measurements against a different technique, namely quantitative gamma assay-with ISOCS and MCNP calibration. JAEA accepted the proposal.

The acceptance test was conducted using ISOCS and PSMC/HRGS equipment and six sludge items from the 23rd to 25th of May 2023 in PCDF.

8.1 The ISOCS measurement system

For the measurements IAEA used ISOCS characterized high purity germanium detector with BEGe 3830 crystal type (: CANBERRA). Figure 8.1 shows the ISOCS measurement system comprising a ISOCS detector, multichannel analyzer (model INSPECTOR 2000: CANBERRA), and laptop PC connected by cables. The control software is used for gamma acquisition and analysis. The detector had tungsten collimator for side shielding and 2mm tin filter at front.

The item-to-detector distance was adjusted for each item to maintain a reasonable dead-time below 30%. Each item was measured from two sides with 180 degrees turn to account for possible inhomogeneities.



(a)The ISOCS system configuration (b) The appearance of the ISOCS system

Figure 8.1 The ISOCS measurement system configuration for sludge measurement

8.2 The ISOCS measurement condition

The ISOCS measurement condition are as follows.

(1) Background measurement

- ISOCS detector is filled the liquid nitrogen before the measurement day by operator.
- Confirm that the measurement system setup is appropriate and measure the background with no nuclear material present near the detector.
- The measurement time is 600 minutes.

(2) Sludge measurement

- Measure the spectrum from the sludge item. Each item is measured twice, each for 20 minutes of real time.
- Before the spectrum acquisition a short 10-20 sec measurement is made to check that the dead-time is below 20%. If higher, adjust the distance to reduce the deadtime.

8.3 Contents of implementation for the test

8.3.1 The measurement sludge item list

Six measurement sludge items were selected by IAEA form 134 items at PCDF storage area.

Those sludge items are shown Table 8.1. Items both before and after stabilization were selected.

Table 8.1 Sludge measurement item list for acceptance test

No.	Batch name	Pu mass[g] (Declared value by XRF)	Net weight[g] (declared value)	Stabilization	Container type	Gamma Dose rate [μSv/h] (18th May 2023)
1	03SS006A	92.47	2,712.42	After (washed)	MOX can	13,500
2	03SS006B	94.30	2,662.68	After (washed)	MOX can	10,000
3	T069A	76.86	3,086.83	After (washed)	SUS304 bottle	10,000
4	T010A2B	66.03	2,492.80	After (washed)	SUS304 bottle	11,000
5	A123T155	11.48	3,827.90	Before (un-washed)	SUS304 bottle	650
6	19G-01-A	0.00	2,252.36	After (washed)	SUS304 bottle	1,500

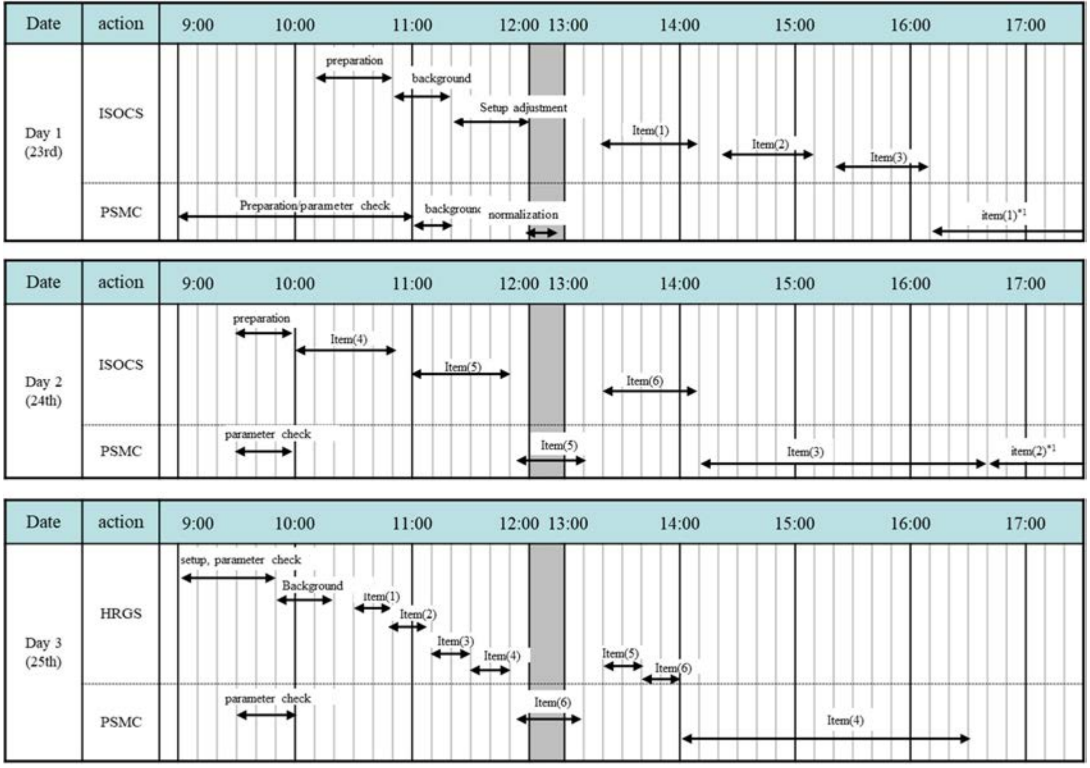
8.3.2 The equipment condition

ISOCS measurement was conducted in the measurement configuration described in section 8.1.
On the other hand, measurement parameters of the HRGS(MGA) were used as Table 5.4, and measurement parameters of the PSMC were used as Table 5.3. Pu mass is evaluated by passive multiplicity method for the sludge items.

8.3.3 The acceptance test schedule

The acceptance test schedule is shown the Table 8.2.
ISOCS, PSMC and HRGS measurement were conducted with the selected six sludge items.

Table 8.2 The time schedule of the acceptance test



*1: The measurements time of item (1) and item (2) were approximately 3.5 hours. Therefore, the measurement order was changed and they were measured all night on first and second day.

8.4 The evaluation results for the acceptance test

8.4.1 Analysis of the ISOCS measurements

Table 8.3 shows the item to detector cap distances for both measurement sides.

Table 8.3 Item-to-detector cap distances and live times for each side item measurement

Item ID	Side A		Side B	
	Distance [mm]	Live time[sec]	Distance [mm]	Live time[sec]
03SS006A	950	924.01	955	928.06
03SS006B	805	869.67	805	895.39
T069A	787	921.07	790	920.30
T010A2B	825	946.09	825	942.11
19G-01-A	280	927.64	280	911.64
A123T155	83	935.14	86	930.02

It is also useful for the analysis to estimate the fill-height of the matrix material inside the container. Based on dose rate readings of the fill height scans, the matrix density for the items with high Pu content was determined as approximately 1.9-2.0 g/cm^3.

The ISOCS technique allows to calculate detection efficiency for given modelled geometry and composition for multiple gamma line energies at once. Benchmark of Pu mass for different energies against each other enables model optimization and refinement of parameters, such as matrix density and heavy metal weight fraction. For this task we employed three gamma lines which provide excellent counting statistics and energy span: 722keV (^{241}Am), 208keV(^{241}Pu), 148.5keV(^{241}Pu).

a) Isotopics composition and U/Pu fraction analysis

For each measurement we ran low-and high-energy Fixed-Energy, Response Function Analysis with Multiple Efficiency (FRAM) analysis with the setup parameters for MOX items; the high energy parameter set was modified to account for specific properties of the item spectra: presence of ^{137}Cs peak, ^{232}U decay chain and other spectral artefacts.

The ^{242}Pu fraction is calculated using the isotopic correlation defined in Equations 5.3 and Table 5.4 as,

$$\frac{f_{242}}{f_{239}} = 4.4 \cdot \left(\frac{f_{240}}{f_{239}}\right)^{1.016} \tag{8.1}$$

The results of the analysis are Pu isotopic composition, 148keV, 208keV and 722keV peak area and (U+Pu), or Metal/Pu ratio. For this IAEA used high-energy result as its analysis based on 1001keV line provides substantially better statistical uncertainty than low-energy analysis based on fluorescence peaks, as well as lower potential bias due to inhomogeneities.

b) Modelling of ISOCS efficiency

This is done in iterative manner with the step c) below. The modelled Pu wt% is calculated based on Pu mass obtained at previous iteration or starting value as:

$$Pu\ wt\% = m_{pu} \cdot 100\% / Net\ weight \tag{8.2}$$

Uranium wt% is calculated based on FRAM analysis result, and its modelled value was allowed to vary within 3 RSD of the obtained U/Pu ratio.

The variation of matrix density was allowed with the incremental/decremental step 0.05 g/cm^3. The measurement modelling and efficiency calculations were done with the geometry composer software.

c) Pu mass calculation

The Pu mass is calculated individually for the three gamma lines of interest as:

$$m_{Pu} = \frac{A_x}{LT \cdot ef_x \cdot Y_x \cdot fr_x} \quad (8.3)$$

Where “x” denotes the three gamma peaks: 148, 208 and 722 keV. A-peak area, LT: live time, ef: ISOCS calculated efficiency, Y: specific yield, fr: isotopic fraction (^{241}Pu for 148 and 208keV lines and ^{241}Am for 722keV line).

The iteration of matrix density (or fill-height) and heavy metal wt% in steps b and c are performed until Pu mass calculated based on 722 keV and 208 keV match within statistical error. Pu mass for 148 keV line is an additional indicator of the model consistency, though certain deviation is allowed, since for this low energy line the matrix heterogeneity and ISOCS modelled efficiency have higher biases.

8.4.2 FRAM and ISOCS results

The following table summarize the FRAM and ISOCS for the low energy (Table 8.4) , for high energy (Table 8.5), for Calculated figures in support of ISOCS analysis (Table 8.6) , and analysis results(Table 8.7).

Table 8.4 Results of low energy FRAM analysis for Pu isotopic composition and ^{240}Pu effective fraction

Item ID	Isotopic fraction*						^{240}Pu effective		Difference
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	FRAM	JAEA	[%]
03SS006A	1.028	65.94	26.35	2.439	4.232	4.644	36.05	35.81	-0.67
03SS006B	1.042	65.46	26.78	2.422	4.290	5.203	36.61	37.52	2.42
T069A	0.949	67.17	26.18	1.801	3.899	7.836	35.12	35.62	1.41
T010A2B	0.955	67.38	25.90	1.834	3.925	8.106	34.90	36.35	4.00
19G-01-A	0.954	66.98	26.23	1.911	3.919	5.736	35.21	34.92	-0.83
A123T155	1.077	64.46	27.72	2.298	4.435	7.484	37.88	37.47	-1.10

*: Each item was measured from two sides with 180 degrees turn. The average values are shown.

Table 8.5 Results of high energy FRAM analysis for heavy metal (U+Pu) to Pu ratio

Item ID	Side A		Side B		Average	
	Metal/Pu	s[%]	Metal/Pu	s[%]	Metal/Pu	s[%]
03SS006A	1.67	27.2	2.23	22.9	1.95	17.8
03SS006B	2.24	18.2	2.20	21.1	2.22	13.9
T069A	3.37	17.3	3.90	15.1	3.64	11.5
T010A2B	3.41	13.7	3.92	12.8	3.67	9.3
19G-01-A	9.86	8.4	8.20	11.7	9.03	7.2
A123T155	12.10	10.6	12.10	7.6	12.10	6.5

Table 8.6 Calculated figures in support of ISOCS analysis: ^{241}Am and ^{241}Pu isotopic, count rates in the peaks of interest and optimized ISOCS efficiencies for the peaks

Item ID	side	LE FRAM		Count rate			Final ISOCS efficiency		
		^{241}Am	^{241}Pu	722keV	208keV	148keV	722keV	208keV	148keV
03SS006A	A	4.663	2.444	32.53	1743.88	290.16	2.399×10^{-5}	2.837×10^{-5}	1.426×10^{-5}
	B	4.625	2.434	31.86	1697.20	281.14	2.375×10^{-5}	2.809×10^{-5}	1.412×10^{-5}
03SS006B	A	5.172	2.424	37.40	1904.48	323.18	3.267×10^{-5}	4.101×10^{-5}	2.140×10^{-5}
	B	5.234	2.419	38.06	1895.70	322.29	3.267×10^{-5}	4.101×10^{-5}	2.140×10^{-5}
T069A	A	7.796	1.786	65.91	1384.06	205.18	3.776×10^{-5}	4.055×10^{-5}	1.864×10^{-5}
	B	7.876	1.815	65.96	1396.56	207.66	3.701×10^{-5}	3.936×10^{-5}	1.806×10^{-5}
T010A2B	A	8.110	1.831	57.48	1251.08	187.56	3.428×10^{-5}	3.791×10^{-5}	1.758×10^{-5}
	B	8.102	1.836	58.12	1281.54	190.96	3.428×10^{-5}	3.791×10^{-5}	1.758×10^{-5}
19G-01-A	A	5.717	1.909	25.40	1288.36	274.07	2.518×10^{-5}	4.426×10^{-5}	2.742×10^{-4}
	B	5.755	1.913	26.88	1378.23	295.47	2.518×10^{-5}	4.426×10^{-5}	2.742×10^{-4}
A123T155	A	7.519	2.305	26.57	1276.43	291.59	9.734×10^{-5}	1.769×10^{-5}	1.192×10^{-3}
	B	7.448	2.291	27.03	1304.41	300.72	9.539×10^{-5}	1.736×10^{-5}	1.171×10^{-3}

Table 8.7 Optimized ISOCS geometry data and analysis results

Item ID	side	Density	FH	Pu+U	Pu mass			
		g/cm^3	cm	wt%	148keV	208keV	722keV	Weight[g]*
03SS006A	A	1.90	14.2	9.0	121.15	123.25	116.39	120.98 ± 3.44
	B				120.81	121.63	114.36	
03SS006B	A	1.90	14.0	7.6	92.19	93.86	87.09	92.46 ± 3.17
	B				92.73	93.62	87.04	
T069A	A	1.90	17.7	12.8	93.26	93.63	86.17	93.75 ± 3.72
	B				94.24	95.79	88.59	
T010A2B	A	1.80	15.1	13.3	86.11	88.31	81.48	86.64 ± 3.33
	B				87.16	90.21	82.73	
19G-01-A	A	1.60	15.4	2.4	7.35	7.47	7.32	7.54 ± 0.28
	B				7.72	7.98	7.88	
A123T155	A	1.70	24.6	0.5	1.51	1.53	1.48	1.55 ± 0.05
	B				1.58	1.61	1.57	

*: the Pu mass uncertainty is quoted at 1 sigma level and includes only random part calculated based on results' scattering for different gamma energies for both measured sides of the items.

8.4.3 PSMC/HRGS analysis

Table 8.8 PSMC passive multiplicity method analysis results

No.	Item ID	Cycles	Singles [cps]	Doubles [cps]	Triples [cps]	Multiplication[-]	Alpha [-]	Pu mass[g]
1	03SS006A	383	245,066.928 ± 4.722	9,475.333 ± 19.246	1,903.812 ± 61.605	1.012 ± 0.003	9.117 ± 0.653	120.688 ± 8.115
2	03SS006B	494	204,150.893 ± 4.010	7,023.233 ± 15.084	1,326.351 ± 37.607	1.007 ± 0.002	9.528 ± 0.595	92.606 ± 5.452
3	T069A	213	146,898.251 ± 5.198	6,279.851 ± 14.323	1,171.716 ± 35.900	1.008 ± 0.003	7.211 ± 0.423	90.000 ± 4.883
4	T010A2B	238	136,748.062 ± 4.729	5,672.215 ± 13.903	1,050.440 ± 30.827	1.007 ± 0.003	7.404 ± 0.410	80.261 ± 4.113
5	19G-01-A	120	22,144.072 ± 2.663	511.489 ± 3.214	90.963 ± 2.418	1.003 ± 0.002	13.735 ± 0.911	7.745 ± 0.493
6	A123T155	188	8,012.383 ± 1.229	111.889 ± 0.870	19.305 ± 0.542	1.001 ± 0.001	22.605 ± 1.401	1.633 ± 0.099

The Table 8.8 shows the PSMC passive multiplicity method analysis results. The dominant source of the statistical uncertainty for the passive multiplicity measurement with the PSMC is Triples count rate, which for the sludge items due to high (alpha, n) neutrons contribution, is at the level of 5-10 % for one hour measurement.

The approximate matrix density (full height) and (U+Pu)/Pu mass ratio obtained with the ISOCS and FRAM analysis enables utilization of Known-M analysis method for the PSMC measurement. The advantage of this is substantially better counting statistics, since it uses only Singles and Doubles count rate.

For the method implementation, one needs to establish relation between neutron multiplication M and ^{239}Pu effective mass. To use the standard Known-M method implemented in INCC software, in the acceptance evaluation, we use simplistic linear dependence:

$$M = 1 + {}^{239}m_{eff} \cdot C_{avg} \cdot 1.21 \cdot 10^{-5} \qquad (8.4)$$

Where C_{avg} is average chord of the item matrix material (neutron average escape length), which can be found as:

$$C_{avg} = \frac{2rh}{r+h} \qquad (8.5)$$

r: container internal effective radius, h: Pu sludge fill height, 1.21×10^{-5} is constant determined based on neutron transport modeling for multiplication in sludge, given the container dimensions.

For generalization an average value of the average chord of 8.2 can be used for the analysis, however during this exercise, an individual C_{avg} values were calculated for each measured container, since the fill-height was individually determined during the ISOCS analysis.

The Table 8.9 summarized the calculations of M(^{239}m$_{eff}$) dependence and Known-M analysis results for the items measured during the acceptance test.

Table 8.9 PSMC Known-M method analysis results

Item ID	Singles [cps]	Doubles [cps]	Density [g/cm^3]	Effective radius [cm]	Height [cm]	Cavg [-]	Multiplication [-]	Alpha [-]	Pu mass [g]
03SS006A	245,066	9,475.3	1.90	5.65	14.24	8.09	1.0118	9.12	120.68
03SS006B	204,151	7,023.2	1.90	5.65	13.97	8.05	1.0086	9.81	90.13
T069A	146,898	6,279.9	1.55	5.40	21.74	8.65	1.0068	7.14	90.81
T010A2B	136,748	5,672.2	1.90	5.40	14.32	7.84	1.0085	7.63	78.05
19G-01-A	22,144	511.5	1.70	5.40	14.46	7.86	1.0011	12.88	8.24
A123T155	8,012	111.9	1.70	5.40	24.58	8.85	1.0002	21.32	1.73

In the evaluation of the acceptance test by Known-M method, in order to calculate the relationship between multiplication and $^{239}m_{\mathrm{eff}}$, we adjusted coefficients and calibration curve based on the selected sludge items in this test. Therefore, the evaluation conditions and the results values are slightly different between section 7.7 and this test. However, calculated Pu mass values from each evaluation are generally consistent within their respective uncertainty.

Table 8.10 comparison of the results of PSMC passive multiplicity (JAEA operator results), Known-M and ISOCS measurement and evaluation of uncertainty components.

Table 8.10 The evaluation results for the acceptance test

No.	Item ID	PSMC/HRGS [Passive multiplicity]	ISOCS [IAEA reference]		PSMC/HRGS [Known-M]	
		Pu mass[g]	Pu mass[g]	Difference*[%]	Pu mass[g]	Difference* [%]
1	03SS006A	120.69 ± 8.11	120.98 ± 3.44	-0.24	120.68	0.01
2	03SS006B	92.61 ± 5.45	92.46 ± 3.17	0.16	90.13	2.68
3	T069A	90.00 ± 4.88	93.75 ± 3.72	-4.17	90.81	-0.90
4	T010A2B	80.26 ± 4.11	86.64 ± 3.33	-7.95	78.05	2.75
5	19G-01-A	7.75 ± 0.49	7.54 ± 0.28	2.71	8.24	-6.32
6	A123T155	1.63 ± 0.10	1.55 ± 0.05	4.91	1.73	-6.13

*: The value is difference from the PSMC (passive multiplicity method) results.

8.4.4 Conclusion of the acceptance test

Both validating measurement methods (ISOCS and PSMC – Known-M) show that the JAEA reference values (i.e. PSMC/HRGS passive multiplicity results) have statistically insignificant bias and only minorly contribute to the total RSD between the methods. Therefore, PSMC/HRGS passive multiplicity measurement for sludge items is confirmed to reduce the measurement uncertainty relative to previous destructive analysis method and it is confirmed that this measurement is applicable to NMA for sludge items based on the acceptance test.

These figures are well within the uncertainty requirements for the standard method per DIQ: DA sampling with XRF measurements (total measurement uncertainty: approximately: 24.2%) for sludge characterization.

Then, JAEA received an IAEA evaluation result that “It was good decision to use the NDA methodology for the Pu sludge instead of the standard (DA) methodology)” from IAEA.

9. Nuclear material control and accounting for sludge items using PSMC/HRGS

9.1 Nuclear material control and accounting for sludge items using PSMC/HRGS

The elements for the nuclear material control and accounting for sludge are Pu, U, and Am and the calculation method is shown below.

9.1.1 The declaration flow of nuclear material in sludge using NDA measurement results

Figure 9.1 shows the evaluation procedure for determining the Pu, U, and Am mass in sludge in a storage bottle.

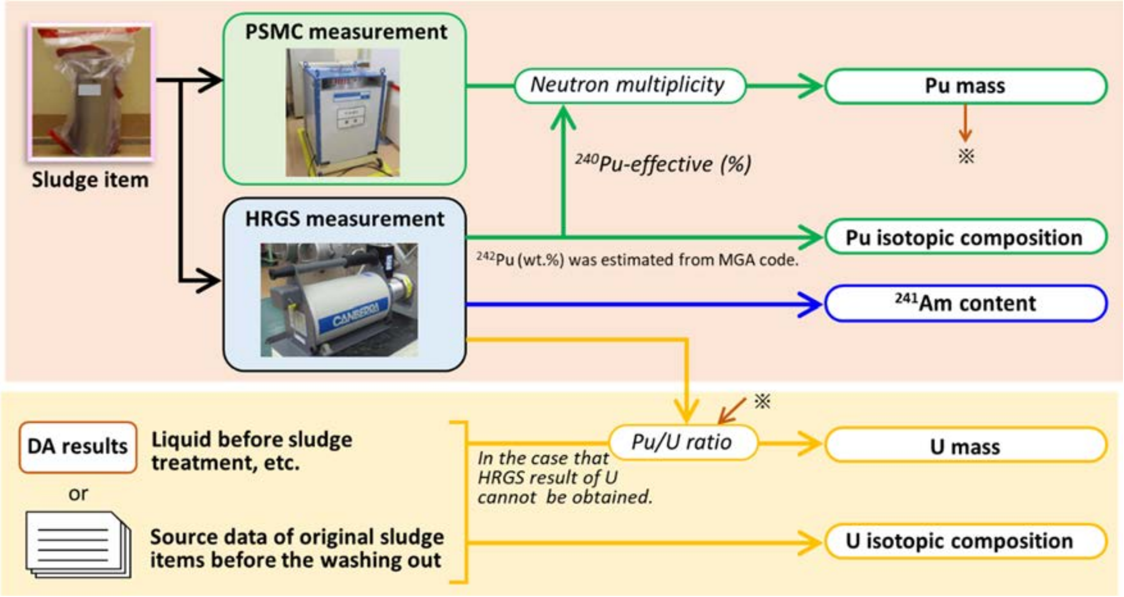


Figure 9.1 The evaluation procedure for determining the Pu, U, and Am mass in sludge in a storage bottle

Pu mass: The Pu mass is obtained from the PSMC measurement results. The Pu isotopic composition is obtained from the HRGS (MGA) results. If HRGS is unavailable when the amount of Pu is small, the estimated value from the process operation data (sludge source data before stabilization, the original solution data composing the sludge) is used for the Pu mass value estimation.

U mass: The U mass is obtained from the U/Pu ratio and the Pu mass from the NDA results (PSMC and HRGS), calculated using Equation 9.1.

$$U\ mass[g]\ = Pu\ mass[g]\ \times\ {}^{U}/_{Pu} \tag{9.1}$$

The U/Pu ratio is obtained using the HRGS measurement results or the estimated value based on the process operation data. The U/Pu ratio is the weight proportion. Table 9.1 shows the U/Pu ratio using HRGS for sludge after stabilization. The 1 σ of the U/Pu ratio is ~9.7% from the PSMC measurements results for stabilized sludge. The TMU of U mass is 11.4% by including the Pu mass TMU of 6.5% (Table 7.14). When the U/Pu ratio is unavailable, it is estimated from the process operation data and used for the U mass evaluation. The U isotopic ratio is obtained from the estimated value based on the process operation data as before.

Table 9.1 The results of the U/Pu ratio by HRGS

No.	Item ID	U/Pu		No.	Item ID	U/Pu	
		ratio	%			ratio	%
1	T078A	0.74	17.40	17	T183A	-*	-*
2	T056A2	2.96	14.77	18	T060A	1.25	3.84
3	T181A	-*	-*	19	T004A	1.19	5.48
4	T109A	1.21	6.18	20	T028A	1.18	7.05
5	T086A	1.60	9.06	21	T142A	1.71	5.07
6	T080A	1.29	4.49	22	T139A	7.27	6.49
7	T089A	1.37	5.55	23	T066A	1.58	8.50
8	T014A2A	12.89	13.22	24	T065A	1.16	3.61
9	T014AB	12.89	13.22	25	T074A	1.23	9.06
10	T071A	1.64	5.60	26	T126A	1.19	10.57
11	T083A	1.18	4.06	27	T053A	1.14	8.63
12	T063A	0.56	23.80	28	09SS001	0.82	17.58
13	T111A	1.43	12.07	29	SNS001A	1.04	4.40
14	T087A	14.41	22.96	30	17B-05A	1.79	6.32
15	T042A	2.60	17.90	31	PNS004A	1.05	5.67
16	T018A	-*	-*	average		2.87	9.73

*: Unmeasurable U/Pu for low-count rate by HRGS

Am: The Am content is obtained from the ^{241}Am content value measured using HRGS. If HRGS is unavailable when the amount of Pu is small, the estimated value from the process operation data (sludge source data before stabilization, the original solution data composing the sludge) is used for the Am value estimation.

9.1.2 The advantages of the NDA method for NMA for sludge

The following are the expected advantages of applying the NDA method for nuclear material control and accountancy for sludge.

1) Technological advantages of the NDA method dedicated for sludge in PCDF

Even though mixing of the sludge after stabilization by water washing is thoroughly performed, the sludge does not have sufficient homogeneity for sample-taking for the DA method. The NDA method has the advantage of measuring the entire sludge item storage bottle without sample-taking to avoid this issue.

2) Reducing the number of procedures for determining the Pu mass (Figure 9.2)

The procedure for determining the Pu mass in sludge using the DA method comprises five steps: (1-1) sampling, (1-2) transfer for analysis, (1-3) analysis operation (DA: XRF analysis), (1-4) weighting, and (1-5) Pu calculation. During the (1-3) XRF analysis, preparation such as sludge dissolution, filtration, and dilution is necessary. However, the NDA requires only two steps: (2-1) transfer for measurement and (2-1) NDA measurement (HRGS and PSMC). The reduced number could reduce the accumulation of errors.

Also, the DA process requires glove box work during the entire analysis, a plurality of bag-in/bag-out procedures for sample transferring with a risk of radiation contamination and exposure. However, the advantages of NDA reduce the probability of radiation contamination and low-radiation exposure due to the few steps for the Pu mass determination. Furthermore, the NDA method evaluates the Pu mass without losing Pu by sample-taking.

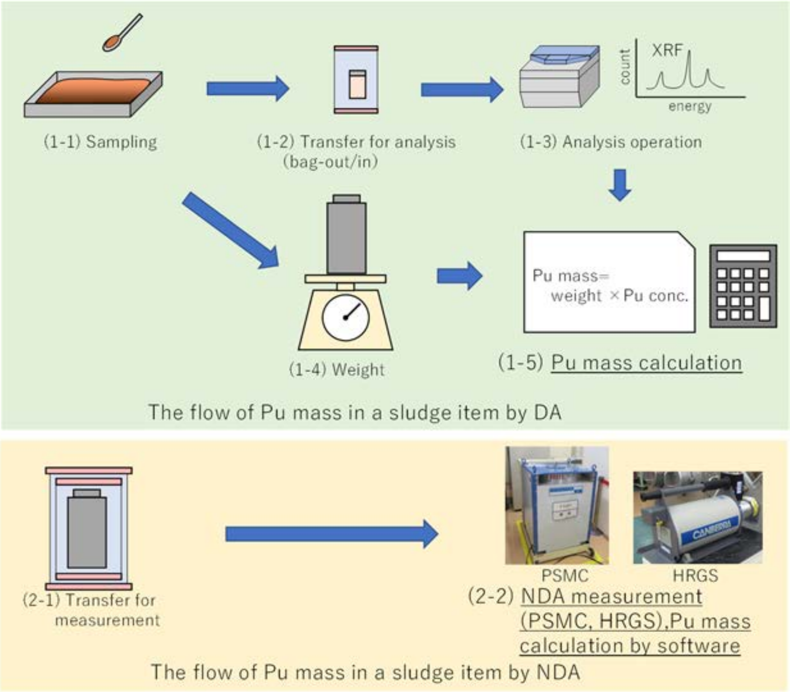


Figure 9.2 The Pu mass calculation procedure for DA and NDA

3) The reduction of time of Pu determination

Contrary to the DA method that typically takes 3 to 4 days for Pu determination, the NDA method takes approximately 1 h for HRGS measurement and another hour for PSMC measurement. The NDA method takes 1 day, including the preparation processes, such as sample setting and measurement equipment setup.

4) Reducing radioactive waste

The DA method generates radioactive solid wastes, such as sample bottles and plastic bags. Also, radioactive liquid waste is generated after the DA method. The application of the NDA method for Pu determination allows one to determine the Pu mass in sludge without producing radioactive waste.

9.2 Revising DIQ for the application of the NDA method for sludge

Table 9.2 shows the current DIQ contents of PSMC for sludge items of 3% to 11% (tentative). Table 9.3 shows the proposed draft of the DIQ contents of PSMC for sludge items that is 6.5% based on the evaluation results in Section 7.

Table 9.2 Current DIQ description

MBA	KMP	Chemical and physical form of material	Equipment or installed place	Sampling procedure and equipment used	Measurement and analytical method and equipment used	Source and level of random and systematic errors	Method of converting source data to batch data
1	5	Sludge (1) and Sludge (2)	Mainly Liquid waste Treatment Toom(A129)	Direct sampling	Weight; Weighing Apparatus Content; XRF or spectrophotometry	Weight measurement; R ; 2% S ; 3% Sampling; R ; 6% S ; 6% Analysis; XRF R ; 20% S ; 10% Spec.; R ; 20% S ; 10%	(Weight) × (Content)
			R&D Area (A128)		Item counting Identification Pu amount; NDA (PSMC)	[Tentative] PSMC; 3 ~ 11%	Pu amount measured by NDA. U amount estimated from Pu amount measured by DA

Table 9.3 Proposed draft DIQ description

MBA	KMP	Chemical and physical form of material	Equipment or installed place	Sampling procedure and equipment used	Measurement and analytical method and equipment used	Source and level of random and systematic errors	Method of converting source data to batch data
1	5	Sludge (1) and Sludge (2)	Mainly Liquid waste Treatment Toom(A129)	Direct sampling	Weight; Weighing Apparatus Content; XRF or spectrophotometry	Weight measurement; R ; 2% S ; 3% Sampling; R ; 6% S ; 6% Analysis; XRF R ; 20% S ; 10% Spec.; R ; 20% S ; 10%	(Weight) x (Content)
			R&D Area (A128)		Item counting Identification Pu and U amount by NDA (PSMC/HRGS)	PSMC/HRGS ; 6.5 % for Pu	Pu amount is directly obtained by NDA. U amount is obtained from the U/Pu ratio measured using HRGS or other estimation method

9.3 The PSMC measurement procedure for routine use

Based on the NDA evaluation results for the Pu mass in sludge items at PCDF described in earlier sections, the proposed NDA (PSMC/HRGS) measurement procedures for routine use are shown below.

9.3.1 The measurement system

Figures 9.3 and 9.4 show the PSMC and HRGS measurement systems, respectively.

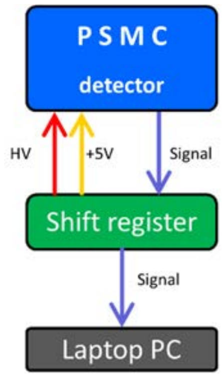


Figure 9.3 The PSMC measurement system

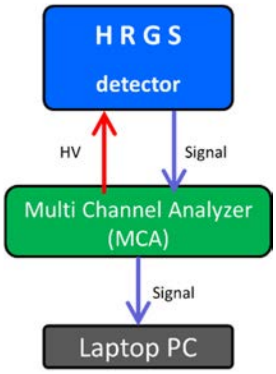


Figure 9.4 The HRGS measurement system

9.3.2 The measurement parameters

Tables 9.4 and 9.5 show the PSMC operating and HRGS (MGA) parameters, respectively.

Table 9.4 The PSMC operating parameters for sludge measurement

Parameter	Unit	Setting value	Parameter	Unit	Setting value
Predelay	μs	2.5	Multiplicity deadtime	ns	112
Gate length	μs	64	Coefficient A	μs	0.379
2nd gate length	μs	64	Coefficient B	μs^2	0.123
High voltage	V	1700	Coefficient C	—	0.850
Die-away time	μs	49	Doubles gate fraction	—	0.643
Efficiency	—	0.543	Triples gate fraction	—	0.425

Table 9.5 The HRGS (MGA) parameters for the sludge measurement

Coefficient	Setting value
C_1	4.40
C_2	1.016

*C_3-C_{10} are 0.0.

9.3.3 The measurement conditions

As shown in the Figure 8.1, for the Pu mass in sludge items, the Pu isotopic composition of sludge items is obtained from the HRGS (MGA) results. The Pu mass is calculated from the PSMC measurement results of ^{240}Pu effective mass combined with the HRGS isotopic composition. The measurement steps for the equipment are listed below.

The HRGS measurement steps for obtaining the Pu isotopic composition and ^{241}Am content in sludge are as follows:

(1) Background measurement

- Confirm that the measurement system setup is appropriate and measure the background at the measurement location after supplying HV from MCA.
- The measurement time is 300 s.

(2) Source measurement

- A series of measurements are taken using the ^{241}Am source placed at the tip of the detector to confirm the performance of the HRGS device.
- The measurement time is 300 s.

(3) The sludge measurement

- Measure the Pu isotopic ratio and ^{241}Am content in the sludge sample located in line with the detector.
- The measurement is conducted with the distance between the detector and sample adjusted to set the deadtime to correspond to 20% or less. The number of 0-3 Cd plates (t: 1 mm) is used for reducing the gamma-rays emitted from ^{241}Am (59.5 keV).

The PSMC measurement steps are as follows:

(1) Background measurement

- Confirm that the measurement system setup is appropriate and measure the background with no nuclear material present.
- The measurement time is 30 s with 20 cycles.

(2) Normalization measurement

- A series of measurements are taken using a californium source placed at the center of the sample cavity to confirm that the expected response is obtained in the PSMC detector.
- The measurement time is 30 s with 20 cycles.

(3) Background measurement (exchange: PSMC parameter for sludge items)

- Measure the background with no nuclear material present.
- The measurement time is 30 s with 20 cycles.

(4) The sludge measurement

- Measure the amount of Pu in the sludge positioned on the AAS holder at the bottom of the sample cavity.
- The measurement is conducted until the precision of the Triples signal becomes below 3%.
  - ✓ At least 30 s with 120 cycles (1 h), the longest 30 s with 2880 cycles (24 h).
- A passive multiplicity method is used in analysis method for the sludge items.

As described in Sections 7.1.1 and 7.1.3, from the ^{252}Cf perturbation results, the influence of residual moisture content in stabilized sludge is negligible. Therefore, the AAS measurement is not performed for routine use but will be used as a quality control tool for selected fractions of sludge items.

The lower limit of detection (LLD) of Pu by the PSMC for sludge samples is evaluated based on the background measurement values using newly designed PSMC parameters (Table 9.4). As a result, the 0.46 mgPu of LLD is confirmed, and the LLD value of 0.01 g is applied as the NMA value of PSMC for practical use in PCDF.

10. Conclusion

The applicability of the plutonium scrap multiplicity counter (PSMC) as the Pu determination method for the nuclear material accountancy (NMA) of sludge with various chemical components generated in the Pu conversion process at PCDF was evaluated jointly by the Japan (JAEA) and the U.S. (United States Department of Energy (DOE)).

The problem of the current NMA for the sludge at PCDF using the DA method, including sample-taking, is the inhomogeneity of the sludge in each item's storage bottle due to the layered structure of multiple sludge batches from multiple treatment processes. Therefore, the amounts of plutonium (Pu) and uranium (U) in sludge in each polyethylene bottle are determined using DA results sampled at each time of sludge generation. However, a large amount of impurity makes the representativeness of each sample difficult.

The inhomogeneity of the sludge was slightly improved, especially in solving the layered structure in each storage bottle, by the implementation of the stabilization of sludge by water washing due to the occurrence of a radioactive contamination incident at another JAEA site. However, the homogeneity of sludge for the DA method was insufficient to ensure the representativeness of the sample to safeguards verification. The measurement object of this project was changed to be stabilized sludge based on the above circumstances.

The PSMC detector parameters were evaluated and optimized appropriately for the stabilized sludge washed with water. The reduced predelay value changing from 4.5 to 2.5 µs, with the modification of Doubles and Triples gate fractions, improved the uncertainty of multiplicity count rates for all sludge items, that means the shorter modified predelay value improved the measurement times for various sludge compositions, independent of the Pu mass and total sludge mass. Furthermore, we evaluated and confirmed that no gate-width change was required.

By using the above detector parameters, we conducted PSMC measurements using actual sludge items (54 items). It was confirmed that the PSMC measurement results were technically consistent and reasonable compared with the mixed oxide powder (α: 0.965) and standard sludge (α: 7.806), and a large amount of impurity (Fe, Ni) did not impact the measurement result. Finally, we evaluated the PSMC/HRGS measurement uncertainty as 6.5% (Pu) and 11.4% (U), equivalent to the current method, and within the range of current DIQ values (tentative) for an NDA method (3-11% for Pu).

A NDA technique, such as PSMC, has a big advantage for sludge items containing impurities, high α-value, and inhomogeneity that could cause the difficulty for reliable DA results.

Based on the above results, an acceptance test was conducted using PSMC/HRGS and IAEA benchmark equipment. Measured Pu mass by both equipment agrees within the measurement uncertainty of each method and the validity for Pu mass measurement of PSMC/HRGS was confirmed.

By comparing the current mass determination procedure (DA based), application of PSMC/HRGS decreases the measurement uncertainty from ~24% to 6.5% (Pu). The advantage of using PSMC as the NMA method for sludge is that it reduces the number of sampling and analytical processes that could increase the total measurement uncertainty and work hours.

Therefore, the applicability of PSMC as an additional NMA method for sludge is technically confirmed, and a newly designed NDA procedure based on this study will be applied to sludge for NMA in PCDF.

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