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Nuclear Analytical and Chemical Isotopics Laboratories Sample Analytical Plan for the Sister Rod Spent Nuclear Fuel Specimens

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Chemical Sciences Division
Nuclear Analytical Chemical and Isotopics Laboratories,

NUCLEAR ANALYTICAL AND CHEMICAL ISOTOPICS LABORATORIES SAMPLE ANALYTICAL PLAN FOR THE SISTER ROD SPENT NUCLEAR FUEL SPECIMENS

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ABSTRACT

This report documents the analytical protocols, data quality objectives, and reference materials that will be used by the Nuclear Analytical and Chemical Isotopics Laboratories (NACIL) in the Chemical Sciences Division at Oak Ridge National Laboratory (ORNL) for the high-precision characterization of pre-determined sections of spent nuclear fuel enclosed in zirconium-based cladding. This planning document was prepared to guide and direct NACIL staff in the sample preparation, analysis, and quality control processes. It includes the following activities:

- The quantitative, self-contained dissolution process in a radiological hot cell.

- Sample screening, using traditional radiochemical techniques, which will be performed at the Transuranium Analytical Laboratory based at the Radiochemical Engineering Development Center, a Hazard Category II nuclear facility, at ORNL.

- Aliquoting of analytical portions of the master dissolver solution, containing 25–30 mg portions of fuel. These will be prepared by mass and characterized for uranium assay by Davies-Gray titration. Cross-verification for the uranium will be made via isotope dilution mass spectrometry (IDMS) using a multi-collector mass spectrometer. Uranium isotopics will be determined using isotope ratio multi-collector mass spectrometry. These analyses will be used as the anchor measurements to achieve high-precision measurements of the fission and activation isotope analytes reported in micrograms/grams of uranium.

- Full mass screening of these samples using quadrupole-inductively coupled plasma mass spectrometry will be employed to enable accurate sample preparation for isotope dilution (ID) and standard addition analysis of the fission and activation analytes.

- Rapid Analysis of Post Irradiation Debris (RAPID), an ORNL-developed tool to elementally isolate and accurately measure the isotopic compositions of both radioactive and nonradioactive analytes found in irradiated materials, will also be employed for screening and quantifying measurements.

- ID-RAPID coupled with fraction collection capabilities will be employed to elementally isolate all analytes. These analytes will be analyzed using high-precision isotopic and IDMS assay determinations using multi-collector inductively coupled plasma mass spectrometry.

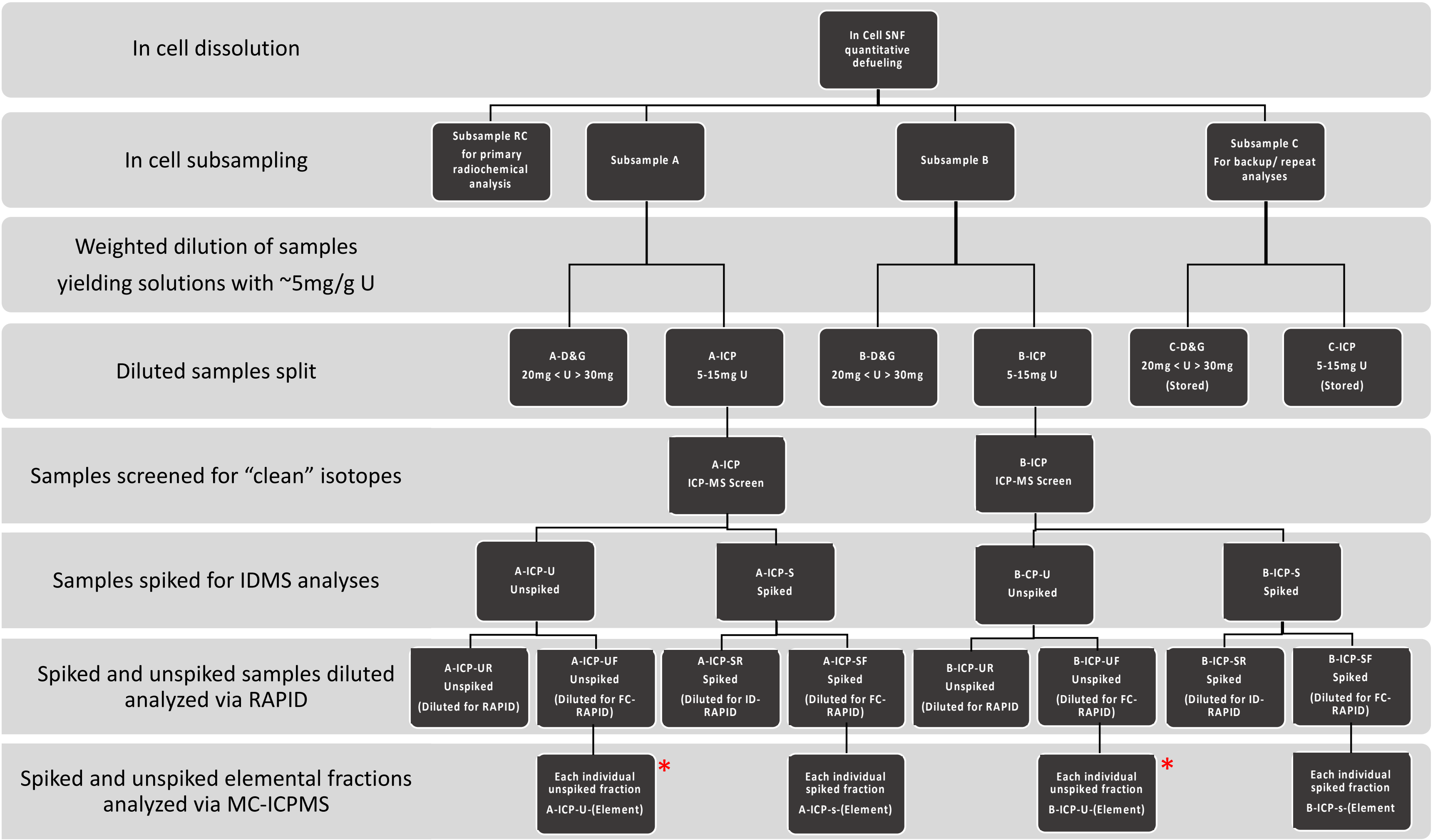
- Uncertainty calculations for reported results will be performed employing the principles of GUM at a 95% confidence level.

1. SAMPLE PREPARATION

1.1 SUMMARY

Approximately 2–3 in. segments of spent nuclear fuel (SNF) rods in zirconium-based cladding will be defueled via a quantitative, self-contained dissolution protocol in a radiological hot cell cubicle at the Radiochemical Engineering Development Center (REDC) at Oak Ridge National Laboratory (ORNL). Subsamples will be pulled from the master dissolver solution (MDS), undergo radiochemical analysis, and be shipped to a radiological facility for further analysis. The received samples will be diluted by weight and further subsampled for the assay determination of more than 50 analytes. Uranium (U) concentrations will be determined via Davies-Gray (D-G) titration in a radiological fume hood, with traceability of concentration measurements verified by bracketing with National Institute of Standards and Technology (NIST) -traceable standards. Isotope dilution (ID) mass spectrometry (IDMS) using certified enriched isotope spikes will be used as a concentration verification method for uranium and as a primary method of analysis for the remaining majority of analytes using a multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS). The majority of the analytes will require elemental isolation prior to analysis. Isolation will be accomplished using a hybrid high-pressure ion chromatography system coupled to a fraction collector (FC). Secondary methods of analysis will be used for verification purposes; the main method will be a modified separation scheme of the Rapid Analysis of Post Irradiation Debris (RAPID) method, whereby direct isotopic compositions of elements can be determined in real time. Figure 1 shows a sample flow chart outlining the primary and secondary modes of analysis, together with subsampling routines.

Further methods of analysis will include quadrupole inductively coupled plasma mass spectrometry (ICPMS), external calibration (EX), standard addition (SA) analysis, high-purity germanium (HPGe) gamma detection, liquid scintillation counting (LSC), and pulsed alpha analysis (PAA).



* Unspiked fractions for certain elements will also be subsampled and analyzed via radiochemical counting methods. the individual elements and methods are described within this document.

Figure 1. A sample flow diagram for the dissolution, sampling, separations, and analysis of spent nuclear fuel.

1.2 SOFTWARE AND DATA PROCESSING

The SCALE code system [1], used internationally in support of SNF transportation and storage, includes an isotope depletion and decay analysis module known as ORIGEN [2]. The model accurately reproduces the flux spectrum in the specific location of irradiation, enabling accurate prediction of total fissions in the target based on the mass of fissile material in the fuel. For RAPID, all measured isotopic data to be reported will reference the integrated isotopic peak areas in a transient signal as determined by the Thermo Fisher Scientific Qtegra software, using an *m/z* trace and a pre-determined elution time. Peak fitting and smoothing in Qtegra will be applied post-acquisition, statistically reducing the observed uncertainties and enabling peak area comparisons between isotope peaks from the same element, yielding isotopic ratios in atomic percent. The atomic percent ratios will be converted to mass percent before reporting. The chromatographic peak fitting and smoothing settings applied to determine peak areas, found within the "Peak Detection" settings of Qtegra, are as detailed previously [3-7].

1.3 DISSOLUTION

Dissolution of the SNF rod cladded segment will be carried out according to WA-TAL-2001: In Cell Dissolution of Spent Nuclear Fuel [8]. The total weight of the SNF rod cladded segment will be obtained on an in-cell, calibrated analytical balance (capable of measurements to 0.001 g). Triplicate weighings will be recorded with a balance tare done between each two weightings. The segment will then be transferred to a digestion bottle and 50 mL of 12 M nitric acid (Optima, Fisher Scientific) will be added. The bottle will then be mated to the condenser, and the chilled water flow will be established and maintained at 10°C for the entire heating period. A solid state ultra-compact thermoelectric recirculating chiller (Oasis 190, Solid State Cooling Systems, Wappingers Falls, NY) will be employed. The pressure vessel will be placed directly onto a laboratory hotplate, which will be heated to boiling. Boiling is typically observed at surface temperatures of 170°C to 180°C (solution temperature of ~150°C) (see Figure 2, left image). The heating period will be maintained for 40 to 50 hours. After completion of the nitric acid dissolution, the digest will be allowed to cool; the condenser will be removed; and a threaded Teflon spout will be attached. The digested fuel, intact clad segment, and metallic particulates will be rinsed through a 0.45 µm cellulose acetate filter into a pre-weighed dilution bottle using a vacuum draw (see Figure 2, right image).

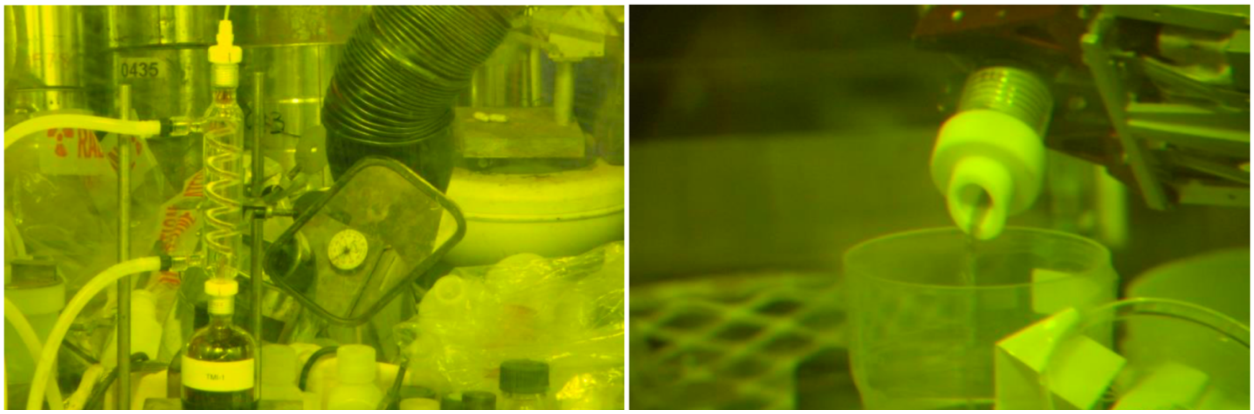


Figure 2. In-cell defueling apparatus (left) and filtration system (right).

After a thorough rinse, the clad segment will be removed, allowed to dry, and then weighed. Triplicate weighings will be recorded with a balance tare done between each. The weight of the fuel will then be calculated as the difference between the initial total "fuel + clad" weight and the post digestion "clad-only” weight. The filtered dissolver solution will then be diluted gravimetrically to a final weight using Optima 12 M nitric acid.

1.4 SAMPLING

Before sampling, the MDS will be inverted no fewer than 20 times. Then 4 ×1–2 mL samples (25–35 mg U), labeled with the appropriate TAL sample code followed by -A, -B, -C, and -RAD, will be taken. The dissolver solution will be inverted no fewer than ten times between samplings. Sample-RAD will be employed for primary radiochemical analysis in triplicate, and Sample-A, Sample-B, and Sample -C will be shipped to a second facility for high-precision separation and analysis. If further cuts are required for specific analyses, those subsamples will be labeled Sample-D, Sample-E, etc., and their treatment and flow diagrams will be documented accordingly.

2. SAMPLE ANALYSIS

The MDS sample aliquots transferred to the analytical laboratory in the radiological facility will be diluted by weight to a concentration of ~5 mg U/g of solution, mixed by inverting no fewer than 20 times, and subsampled by weight for D-G analysis (20 mg < U > 30 mg) using a balance that reads to a minimum of 0.1 mg. The remaining sample (~5–15mg U) will be used for the remainder of the analyses outlined below. Sample preparations described in this sample analytical plan (SAP) will use analytical balances that are monitored through the ORNL Metrology Laboratory. The precision of the balance measurements will be a key contributor to the low uncertainties described in the data quality objectives for the analytes. The operation of the analytical balances will be conducted according to the ORNL Chemical Sciences Division document CSD-OP-RML-AD01, "Work Policies and Practices for the Radioactive Materials Analytical Laboratory" [9]. A complete list of all analytes and predicted concentrations (in ug/g fuel and ug/g U) is provided in Appendix A.

2.1 URANIUM ASSAY DAVIES- GRAY TITRATION.

The high-precision uranium assay will be measured by D-G titration based on "ASTM C1267-17 Standard Test Method for Uranium by Iron (II) Reduction in Phosphoric Acid Followed by Chromium (VI) Titration in the Presence of Vanadium." The procedures followed are found in references [10,11]. All uranium species in the solution are reduced to uranium (IV) using iron (II). The uranium (IV) is then titrated to a potentiometric end point using potassium dichromate.

2.2 SAMPLE SCREENING VIA MASS SPECTROMETRY

The samples ICP-A and ICP-B will be analyzed for fission product content to calculate the enriched isotope spike levels required for ID. "Clean" masses—i.e. masses that have been calculated to have a single isotope present—are listed in Table 1 for each element. They will be analyzed using a Thermo Scientific iCAP Q quadrupole ICPMS. External calibration using a multi-standard linear regression curve and NIST-traceable multi-element standards will be employed for isotopic quantitation. ORIGEN models will be employed to estimate both the elemental concentrations and the isotopic concentrations of all analytes.

Table 1. Isotopes to be analyzed for SNF screening via ICPMS

Actinide isotopes	Fission isotopes
^{238}U	^{82}Se
$^{239/240}$Pu	^{88}Sr
^{243}Am	$^{95/97/98}$Mo
^{227}Ac	^{99}Tc
^{237}Np	$^{101/102}$Ru
^{244}Cm	^{103}Rh
	^{109}Ag
	$^{121/123}$Sb
	^{129}I
	^{133}Cs
	$^{144/145}$Nd
	$^{147/152}$Sm
	^{153}Eu
	^{155}Gd

2.3 RAPID SEPARATION PROTOCOL

A Thermo Scientific Dionex ICS-5000+ HPIC system will be coupled to a Thermo Scientific iCAP Q quadrupole ICPMS. Chromatographic separation will be performed using a metal-free HPIC pump ICS 5000 (ThermoFisher Scientific), a 50 µL injection loop, an IonPac CG5A guard column, an IonPac CS5A column, and a thermal compartment set at 35°C for consistent elution times. The analytical column will be connected to a high-flow PTFE nebulizer (ESI, Omaha, NE) using PEEK tubing via a mixing tee-piece. Nitric acid (HNO_3, 1.25M, ~5%) will subsequently be pumped into the mixing tee-piece at 0.1 mL/min to acidify the eluent, post-column, before nebulization using the peri-pump as installed on the mass spectrometer. The nebulizer will be connected to a cyclonic quartz spray chamber, a nickel sampler cone, and a nickel skimmer cone with a high matrix skimmer cone insert (3.5 mm). The spray chamber will be cooled, using a Peltier-cooling jacket, to 2.5°C. The torch will use a 2.5 mm quartz injector. The coupled instruments will be housed within a radiological fume hood, facilitating the analyses of radioactive samples. The method development, isotopic detection limits, system stability, elution profiles, data analysis, method uncertainties, and various uranium-based applications have been reported previously [6-11]. RAPID will be used to directly measure the isotopic composition of the isolated elements in real time. For the majority of the elements, this method will be employed as the secondary analytical method for isotopic analysis. The separation scheme and eluent percentage contributions are given in Appendix B.

2.4 MC-ICPMS

Isotope/elemental fractions, isolated by the RAPID separation protocol and collected using an inline FC (AFC-3000 Automated Fraction Collector, Thermo Scientific Dionex), will be analyzed by an MC-IC-MS (Neptune, Thermo Scientific, Bremen, Germany). The instrument is a double-focusing MC-ICPMS equipped with nine Faraday collectors and secondary electron multipliers.

The uranium isotopic analysis, for example, will be performed in low-resolution mode ($m\,\Delta m^{-1} = 300$). The instrument will be fitted with a nickel sample cone and a nickel hydrogen skimmer cone. The samples will be aspirated using a 50 µL min^{-1} self-aspirating nebulizer with a standard introduction system consisting of a cyclonic coupled to a Scott double-pass spray chamber (ESI, Omaha, NE). During the U analysis, a 10^{-13} ohm amplifier resistor will be used for the 234 and 236 masses, and a 10^{-11} ohm will be used on the 235 and 238 mass. The analysis sequence will employ a standard sample bracketing method with quality control samples dispersed after every third sample. The mass fractionation corrections will be applied by direct comparison with an isotopic reference material. The sample fractions will also be corrected for instrument blank, potential isobaric interferences, and hydride contributions.

2.5 ID-MC-ICPMS

Isotope/elemental fractions, previously spiked with isotopically enriched standards, isolated by the RAPID separation protocol, and collected using an inline FC, will be analyzed by an MC-ICPMS. (see Section 2.4 for further analysis specifics)

2.6 SECONDARY ANALYTICAL METHODS

2.6.1 ID-RAPID

Samples previously spiked with isotopically enriched standards and isolated by the RAPID separation protocol will be measured in real time using the RAPID protocol. (see Section 2.3 for further analysis specifics)

2.6.2 EX-RAPID

Isotopes isolated by the RAPID separation protocol will be measured in real time using the RAPID protocol and NIST-traceable standards by EX using a linear regression curve. (see Section 2.3 for further analysis specifics)

2.6.3 SA-ICPMS

SA analysis will be performed by weight using a weighted dilution of the original aliquot, which will then be spiked by weight with NIST-traceable standards of known concentration. Linear regression analysis of the resultant slope will yield the concentration and uncertainty. An iCAP Q fitted with a cyclonic quartz spray chamber and a nickel skimmer cone with a 3.5 mm insert will be employed for these analyses.

2.7 TERTIARY ANALYTICAL METHODS

2.7.1 ICPMS

Samples will be diluted by weight, and predetermined “clean” isotopes will be analyzed via weighted EX by ICPMS using NIST-traceable standards and internal standards to correct for residual matrix effects.

2.7.2 RAPID-LSC

RAPID, in combination with fraction collection and subsequent measurement by LSC, will be employed for certain beta-emitting isotopes.

2.7.3 RAPID-PAA

RAPID, in combination with fraction collection and subsequent measurement by PAA, will be employed for certain alpha-emitting analytes—particularly those that have high specific activities but low mass within the sample.

2.7.4 RAPID-HPGe

RAPID, in combination with fraction collection and subsequent measurement by HPGe gamma analysis, will be employed for certain gamma-emitting analytes, particularly those that have high specific activities but low mass within the sample.

2.8 IDMS SPIKES

For each element, the primary method of analysis is ID-MC-ICPMS. The method relies on the use of isotopically enriched solutions as spike solutions directly added to the MDS. The certification of these enriched spikes will be made using NIST standard reference materials, naturally abundant elemental standards of known concentration and acceptably low uncertainty. An additional standard of each element with known isotopic ratios and acceptable low uncertainty is also required for use as a mass bias.

The spike certification process will proceed as follows:

- The isotopically enriched standard (SPIKE-STD) (oxide or salt) will be digested/dissolved by weight in an appropriate acid carrier. The solid oxide or salt will not be dried before digestion; however, the concentration of the resulting solution will be known to within ~5%

- The solution will be diluted by weight to an appropriate concentration for spiking the samples.

- The isotopically enriched solution will be spiked by weight using a NIST-traceable standard of natural isotopic abundance (Nat-STD).

- The isotopic composition of the spiked and unspiked solutions will be determined employing a third natural standard with known isotopic composition (or known isotope ratios) for mass bias corrections (MB-STD).

- The Nat-STD will then be also employed as an additional laboratory control for ID-MC-ICPMS and ID-RAPID to monitor effects of chemical separations.

3. ANALYTES

The isotopes to be analyzed are given in Table 2. They are categorized by abundance and mass into four groups: major actinides, minor actinides, high-mass fission products (HMFPs), and low-mass fission products (LMFPs). The primary method of quantification is given in Table 2. Full analysis details for each isotope/element are discussed in Sections 3.1–3.4, which cover these four main categories:

- 3.1 Major Actinides (U/Pu/Am)
- 3.2 Minor Actinides (Ac/Np/Cm)
- 3.3 High Mass Fission Products (mass 129–155)
- 3.4 Low Mass Fission Products (mass 79–125)

Table 2. Isotopes to be analyzed in spent nuclear fuel

Major actinides	Minor actinides	High-mass fission products	Low-mass fission products
░ ^{234}U	▓ ^{227}Ac	▒ ^{129}I	▒ ^{79}Se
░ ^{235}U	▓ ^{237}Np	▒ ^{133}Cs	▒ ^{95}Mo
░ ^{236}U	▒ ^{242}Cm	▒ ^{134}Cs	▒ ^{90}Sr
░ ^{238}U	▒ ^{243}Cm	▒ ^{135}Cs	▓ ^{99}Tc
▒ ^{238}Pu	▒ ^{244}Cm	▒ ^{137}Cs	▒ ^{101}Ru
▒ ^{239}Pu	▒ ^{245}Cm	▒ ^{137m}Ba	▒ ^{106}Ru
▒ ^{240}Pu	▒ ^{246}Cm	▒ ^{143}Nd	▓ ^{103}Rh
▒ ^{241}Pu	▒ ^{247}Cm	▒ ^{144}Nd	▒ ^{109}Ag
▒ ^{242}Pu		▒ ^{145}Nd	▒ ^{125}Sb
▒ ^{241}Am		▒ ^{146}Nd	
▒ ^{242m}Am		▒ ^{148}Nd	
▒ ^{243}Am		▒ ^{144}Ce	
		▒ ^{147}Pm	
		▒ ^{147}Sm	
		▒ ^{149}Sm	
		▒ ^{150}Sm	
		▒ ^{151}Sm	
		▒ ^{152}Sm	
		▒ ^{151}Eu	
		▒ ^{153}Eu	
		▒ ^{154}Eu	
		▒ ^{155}Eu	
		▒ ^{155}Gd	

Primary analytical method
░ Davies- Gray
▒ Isotope dilution
▓ Standard addition

3.1 MAJOR ACTINIDES

Throughout the SAP, the isotopes of uranium, plutonium, and americium will be referred to as the major actinide analytes. The analyses of the fissionable isotopes of these analytes, in particular, are vital for high-precision burn-up determinations. Figure 3 shows the m/z traces 134–242 with both logarithmic and linear y-axes shown. The americium, plutonium, and uranium peaks are highlighted to demonstrate the efficient separation of the major actinides from isobaric interferences. Table 3 shows each of the major actinide analytes together with the proposed analytical methods and appropriate traceable standards.

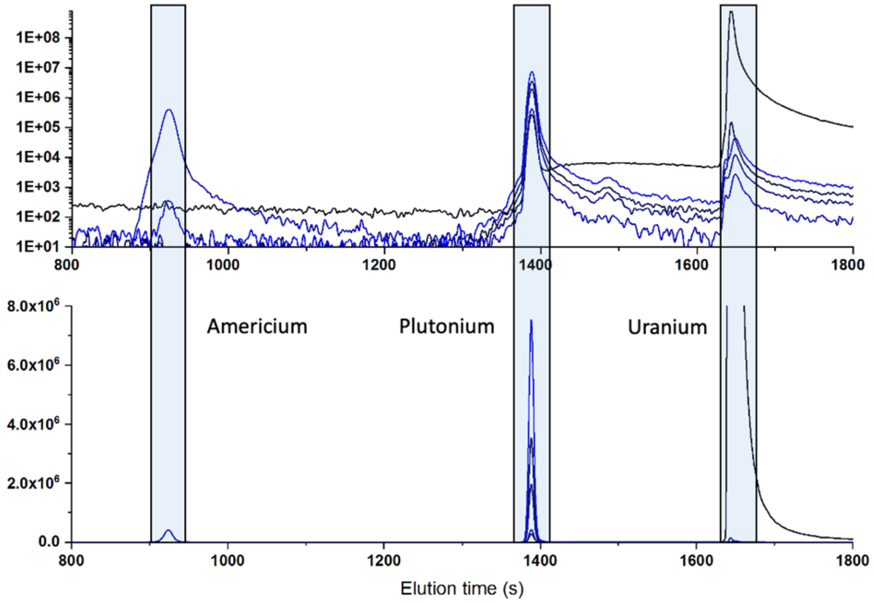


Figure 3. Graph showing the monitored m/z traces for uranium, plutonium and americium. The baseline resolution between ^{241}Am and ^{241}Pu is clearly visible; and, although ^{238}Pu does not achieve baseline post-elution, there are ~5 orders of magnitude difference between ^{238}Pu and ^{238}U.

Table 3. Each of the major actinide analytes, the ORIGEN-calculated content in fuel, and the proposed analytical methods and appropriate traceable standards.

Analyte	Half-life	ORIGEN content* (µg/g fuel)	Primary analytical method /standard	Secondary/tertiary analytical method/s	Control standards (See APPENDIX D)	Mass bias standards
Uranium	NA	7.5-8.5E^5	Davies-Gray	ID-MC-ICPMS DHS UTHX 233 ONRL-WEWA (93% 235)	NBLPO-CRM-129, NBLPO-CRM-112a	NBLPO-CRM-117
^{234}U	2.45E5 Y		MC-ICPMS	RAPID	NBLPO-CRM-010	IRMM-73 series
^{235}U	7.04E8 Y					
^{236}U	2.34E4 Y					
^{238}U	4.47E9 Y					
Plutonium	NA	9.0-9.9E^3	ID–MC-ICPMS NBLPO-CRM-130	ID-RAPID,	NBLPO-CRM-136 NBLPO-CRM-137 ORNL-WRM-003 NBLPO-CRM-138	NBLPO-CRM-137
^{238}Pu	87.7 Y	180–198 (2%)	MC-ICPMS	RAPID, RAPID-PAA Pu238/Pu239/240		NBLPO-CRM-137 ORNL-WRM-003
^{239}Pu	24110 Y	5310–5841 (59%)				
^{240}Pu	6561 Y	2250–2475 (25%)				

Table 3. Each of the major actinide analytes, the ORIGEN-calculated content in fuel, and the proposed analytical methods and appropriate traceable standards (continued).

Analyte	Half-life	ORIGEN content* (µg/g fuel)	Primary analytical method /standard	Secondary/tertiary analytical method/s	Control standards (See APPENDIX D)	Mass bias standards
^{241}Pu	14.329 Y	540–594 (6%)				
^{242}Pu	3.75E5 Y	630–693 (7%)				
Americium	NA	1.0–1.2 E^{3}	ID-MC-ICPMS CETAMA ^{243}Am STD	ID-RAPID HPGe	SRM-4332E	CETAMA ^{241}Am/^{243}Am STD
^{241}Am	432.6 Y	870–1044 (87%)	MC-ICPMS	RAPID	CETAMA ^{241}Am/^{243}Am STD	CETAMA ^{241}Am/^{243}Am STD
^{242m}Am	141 Y	0.7—0.84 (0.07%)				
^{243}Am	7364 Y	130–156 (13%)				

*ORIGEN-calculated analyte content in µg/g fuel—using a standard 23.9 MW power reactor, three cycles of 416 days irradiation, each irradiation followed by 30-day decay, employing 4% enriched UO_2 green fuel. To be used for estimates only. Full ORIGEN models for each of the spent nuclear fuel segments are available, with partial analyte tables in Appendix C.

3.1.1 Uranium

- Assay
    - D-G analysis employing NBLPO-CRM-129 and NBLPO-CRM-112a as control standards
    - ID-RAPID using ORNL-WEWA (93% enriched ^{235}U spike) and NBLPO-CRM-117 as a mass bias standard
    - EX using NIST-traceable natural or depleted U standards and ICPMS.

- Isotopic
    - FC-MC-ICPMS using NBLPO-CRM-010 and NBLPO-CRM-138 as controls
    - RAPID using a traceable natural U standard and NBLPO-CRM-117 as a mass bias standard
    - ICPMS without separation using a traceable natural U standard and NBLPO-CRM-117 as a mass bias standard

3.1.2 Plutonium

- Assay
    - ID-FC-MC-ICPMS using NBLPO-CRM-130 (Pu-242) as a spike, NBLPO-CRM-137 as a mass bias standard and control, and NBLPO CRM 136 and ORNL WRM-003 as controls
    - ID-RAPID using NBLPO-CRM-130 (Pu-242) as a spike

- Isotopic
    - FC-RAPID and MC-ICPMS using NBLPO-CRM-136, NBLPO-CRM-137, and ORNL WRM-003 as controls
    - RAPID using NBLPO-CRM-137 as a control
    - ICPMS without separation NBLPO-CRM-137 as a control
    - FC-RAPID-PAA to determine the ^{238}Pu (5.50 KeV) ratio compared with ^{239}Pu+^{240}Pu (5.15 KeV) according to ASTM C1415-18, “Standard Test Method for ^{238}Pu Isotopic Abundance by Alpha Spectrometry”

3.1.3 Americium

- Assay
  - ID-FC-MC-ICPMS using SRM-4332E as a spike and CETAMA 241/243 as a mass bias standard
  - ID-RAPID using SRM-4332E as a spike

- Isotopic
  - FC-RAPID and MC-ICPMS using CETAMA 241/243 as a mass bias standard.
  - RAPID using CETAMA 241/243 as a mass bias standard.
  - FC-RAPID-HPGe for ^{241}Am/^{243}Am ratio. Note: For gamma analysis, traditionally, the emissions from the ^{239}Np daughter (106.1, 277.6, and 228.2 KeV) of ^{243}Am are used for counting and secular equilibrium is assumed for most solutions. If the ^{239}Np emissions are to be used the solutions will need to be "aged" for ~2 weeks prior to analysis, though the ^{243}Am direct emission* (74.7 KeV) is also measurable.
  - FC-RAPID-PAA* for the ^{241}Am/^{243}Am ratio is also possible as long as the sample deposition is clean and the PAA baseline allows for resolution between the ^{243}Am emissions (5.18–5.28 KeV) and those from ^{241}Am (5.39–5.49 KeV).

The ^{243}Am half-life (7364[22] years) is an order of magnitude longer than that of ^{241}Am (432.6[6] years), so direct measurement of ^{243}Am is less favorable.

3.2 MINOR ACTINIDES

The minor actinides are designated as actinides that cannot be directly measured by any traditional form of high-precision IDMS. The analyses of neptunium and actinium will be performed using SA in combination with EX using NIST-traceable standards. Curium content will be determined using an in-house–produced Cm-244/245/246 working reference material (WRM) with known isotopic composition and activity. ID-RAPID and ID-MC-ICPMS will be employed for minor actinide analyses. Figure 4 shows the monitored m/z traces: 241 (red), 242 (gray), 243 (blue), 244 (green), 245 (purple), 246 (ochre), and 247 (cyan) showing the fractions to be collected to isolate curium. Figure 4 shows that americium can be cleanly isolated. However, because of the low intensity of the ^{243}Cm relative to that of the preceding ^{243}Am, the curium fraction must be monitored for ^{241}Am. Any observed ^{241}Am in the curium fraction will be used to correct the ^{243}Cm. Table 4 shows each of the minor actinide analytes, the ORIGEN-calculated content in fuel, and the proposed analytical methods and appropriate traceable standards.

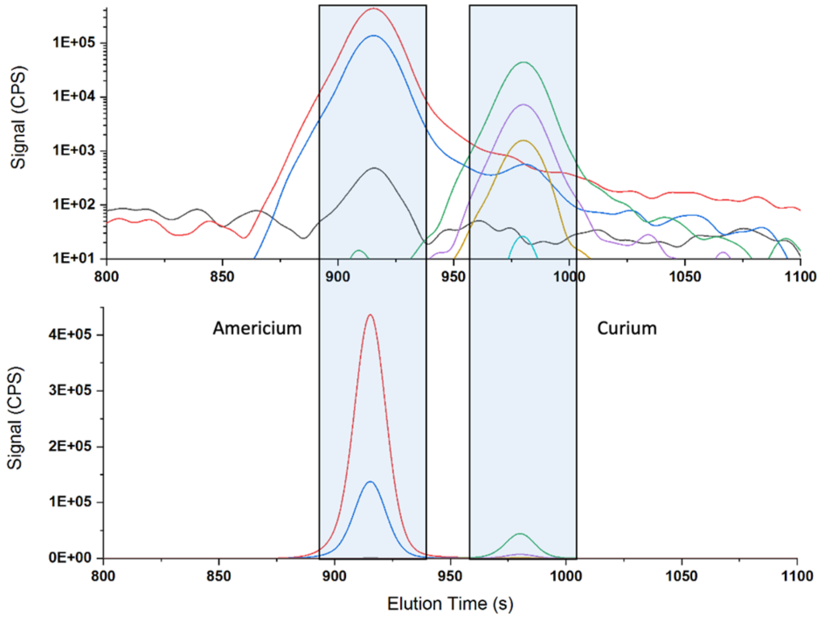


Figure 4. Graph showing the monitored m/z traces: 241 (red), 242 (gray), 243 (blue), 244 (green), 245 (purple), 246 (ochre), and 247 (cyan). The peaks for americium and curium are highlighted, the y-axis is shown on both a linear (bottom) and logarithmic (top) scale.

Table 4. The minor actinide analytes together with the ORIGEN-calculated content in fuel, the proposed analytical methods, and appropriate traceable standards to be employed.

Analyte	Half-life	ORIGEN content* (µg/g fuel)	Primary analytical method /standard	Separation method	Secondary/ tertiary analytical method/s	Control standards (See APPENDIX D)	Mass bias standards
^{227}Ac	21.772 Y		SA-ICPMS	NA	EX-ICPMS	EZIP Ac-227	NA
^{237}Np	2.144E^{6} Y		SA-RAPID	RAPID	EX-RAPID	NIST4341a	NA
Curium	NA	30–35	ID-RAPID, Gross alpha/PAA (direct)	RAPID	PAA HPGe EX-ICPMS	ORNL-Cm-WRM (^{246}Cm) **SRM 4320b**	CETAMA ^{241}Am/^{243}Am STD
^{242}Cm	162 D	0.00	MC-ICPMS	RAPID	RAPID RAPID-HPGe,	ORNL-Cm-WRM **SRM 4320b**	CETAMA ^{241}Am/^{243}Am STD
^{243}Cm	29.1 Y	0.39 (1%)					
^{244}Cm	18.1 Y	27.41 (85%)					
^{245}Cm	8423 Y	3.82 (12%)					
^{246}Cm	4706 Y	0.46 (1%)					
^{247}Cm	1.57E^{7} Y	?					

*ORIGEN-calculated analyte content in µg/g fuel—using a standard 23.9 MW power reactor, three cycles of 416 days irradiation, each irradiation followed by 30-day decay, employing 4% enriched UO_2 green fuel. To be used for estimates only.

3.2.1 Actinium

- Assay
    - SA using NIST-traceable EZIP-Ac-227.
    - EX using NIST-traceable EZIP-Ac-227.
    - FC-RAPID-HPGe has potential to be used for ^{227}Ac assay measurement. However, if the ^{227}Th/^{223}Ra daughter emissions (235.96 KeV among others) are to be used, the solutions will need to be aged for ~120 days prior to analysis to allow for secular equilibrium to occur.

3.2.2 Neptunium

- Assay
    - SA using NIST4341a
    - EX-RAPID using NIST4341a

3.2.3 Curium

- Assay
    - ID-RAPID using an in-house–produced WRM enriched in ^{246}Cm, certified via IDMS using NIST SRM 4320b and verified via PAA and HPGe. [12]
    - Direct alpha spectrometry and PAA without separation for ^{243}Cm + ^{244}Cm (5.80 KeV). Weighted dilutions if necessary, and sample self-attenuation must be taken into consideration (20–40% of alpha activity). Dilutions by weight and duplicate samples should be plated by volume.
    - Direct HPGe without separation for ^{243}Cm (277.6 KeV). Weighted dilutions and, if necessary, potential signal suppression must be taken into consideration.

- Isotopic
- FC-RAPID and MC-ICPMS using CETAMA 241/243 as a mass bias standard. The ^{241}Am/^{242m}Am/^{243}Am ratios should be determined and employed to correct for isobaric interferences on ^{242}Cm (if detectable) and ^{243}Cm.
- FC-RAPID-PAA for ^{242}Cm/^{243}Cm+^{244}Cm/^{245}Cm+^{246}Cm ratio at 6.11 KeV/5.8 KeV/ 5.3KeV.
    - RAPID for full isotopic composition (ISO).

3.3 HIGH MASS FISSION PRODUCTS

With regard to this SAP, the high-mass fission isotopes are defined as those with a mass of 129–155 amu. This encompasses cesium together with the lower-mass lanthanides; cerium, neodymium, promethium, samarium, europium, and gadolinium. As a rule, the HMFPs have very good resolution from both isobaric and potential polyatomic interferences. However, to achieve the analytical precision required, several potential analytes will be monitored using various clean masses with no possible isobaric interferences. Table 5 shows the HMFP analytes together with the ORIGEN-calculated content in fuel, the proposed analytical methods, and appropriate traceable standards to be employed.

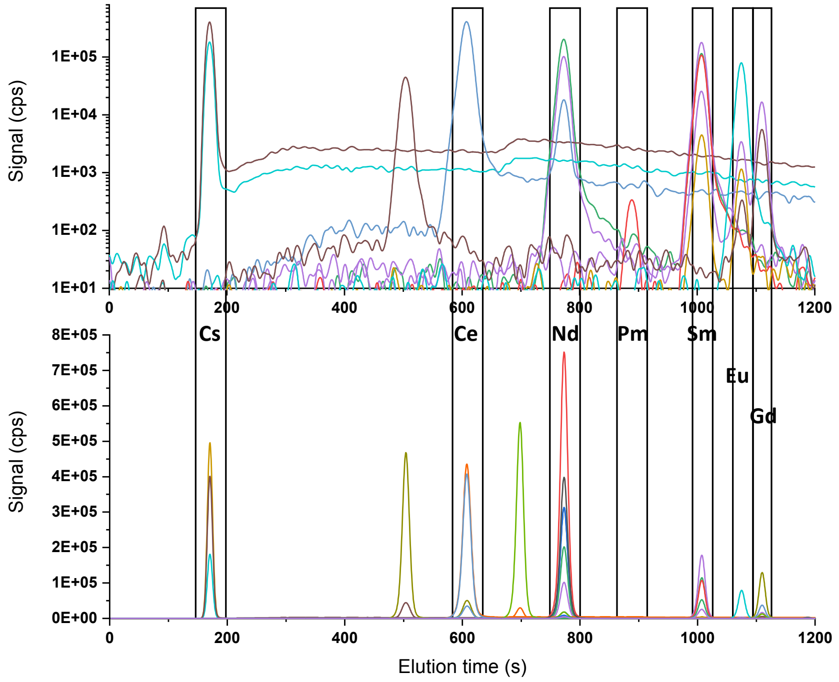


Figure 5. Graph showing the separated spectra of a spent nuclear fuel sample monitored m/z traces: 133–155 for the elution period of 0–1200s. The peaks for cesium, cerium, neodymium, promethium, samarium, europium, and gadolinium are highlighted. The remaining peaks, lanthanum and praseodymium, are shown to further illustrate peak resolution. The top figure has a y-axis in *l*og scale and depicts all of the traces with potential isobaric or oxide interferences.

Table 5. The high-mass fission product analytes together with the ORIGEN-calculated content in fuel, the proposed analytical methods, and appropriate traceable standards to be employed.

Analyte	Half-life	ORIGEN content* (µg/g fuel)	Primary analytical method /standard	Secondary/ tertiary analytical method/s	SPIKE-STD (enriched isotope)	Nat-STD (See APPENDIX D)	MB-STD (See APPENDIX D)
^{129}I	1.6E7 Y	~140	ID-ICPMS	ICPMS	^{127}I	NA	NIST 3104a barium
Cesium	NA	~2600	ID-MC-ICPMS	ID-RAPID	Inorganic ventures CGCS1 (^{133}Cs)	NA	NIST 3104a barium
^{133}Cs	Stable	1304 (49%)	MC-ICPMS	RAPID, RAPID-HPGe	NA	NA	NIST 3104a barium
^{134}Cs	2.06 Y	0.2 (0.007%)					
^{135}Cs	2.3E6 Y	475 (18%)					
^{137}Cs	30.08 Y	900 (34%)					
^{137m}Ba	2.55 m	–	MC-ICPMS (Cs)	HPGe	NIST 3111a (^{133}Cs)	Inorganic ventures Cs	NA
Neodymium	NA	5000-8500	ID-MC-ICPMS	ID-RAPID	Nd-150-STD (^{150}Nd)	Legolas (La Jolla) NIST 3135a	JNDI (jNNDi-1)

Table 5. The high-mass fission product analytes together with the ORIGEN-calculated content in fuel, the proposed analytical methods, and appropriate traceable standards to be employed (continued).

Analyte	Half-life	ORIGEN content* (µg/g fuel)	Primary analytical method /standard	Secondary/ tertiary analytical method/s	SPIKE-STD (enriched isotope)	Nat-STD (See APPENDIX D)	MB-STD (See APPENDIX D)
^{143}Nd	Stable	912 (19%)	MC-ICPMS	RAPID	NA	Legolas (La Jolla) NIST 3135a	JNDI (jNNDi-1)
^{144}Nd	Stable	1584 (33%)					
^{145}Nd	Stable	778 (16%)					
^{146}Nd	Stable	832 (17%)					
^{148}Nd	Stable	437 (9%)					
^{144}Ce	284 D	0	ID-MC-ICPMS, HPGe	ID-RAPID	Ce-140-STD (^{140}Ce)	HPS-Ce	NIST 3110
^{147}Pm	2.62 Y	1 (100%)	ID-RAPID (semi-quant)	ICPMS RAPID-LSC	Sm-149-STD (^{149}Sm)	NIST 3147a	NA
Samarium	NA	~1100	IDMS Sm-149	ID-RAPID	Sm-149-STD (^{149}Sm)	HPS-Sm	NIST 3147a ^{150}SM/^{154}Sm
^{147}Sm	Stable	284 (29%)	ID-MC-ICPMS	RAPID	NA	HPS-Sm	NIST 3147a ^{150}SM/^{154}Sm
^{149}Sm	Stable	3 (0.35%)					
^{150}Sm	Stable	348 (35%)					
^{151}Sm	90 Y	10 (1%)					
^{152}Sm	Stable	113 (11%)					
Europium	NA	5000-8500	ID-MC-ICPMS	ID-RAPID	Eu-141-STD (^{151}Eu)	NIST 3117a	NIST 3147a ^{150}SM/^{154}Sm
^{151}Eu	Stable	2 (1%)	MC-ICPMS	RAPID RAPID-HPGe	NA	NIST 3117a	NIST 3147a ^{150}SM/^{154}Sm
^{153}Eu	Stable	135 (94%)					
^{154}Eu	8.60 Y	7 (4%)					
^{155}Eu	4.75 Y	0.5 (0.4%)					
^{155}Gd	Stable	10 (5%)	ID-MC-ICPMS	ID-RAPID	Gd-155-STD (^{155}Gd)	HPS-Gd	NIST 3118a

*ORIGEN calculated analyte content in µg/g fuel – using a standard 23.9 MW power reactor, three cycles of 416 days irradiation, each irradiation followed by 30-day decay, employing 4% enriched UO_2 green fuel. To be used for estimates only.

3.3.1 Iodine-129

- Assay
  - ID-ICPMS using Inorganic Ventures iodide (^{127}I) as SPIKE-STD and using a NIST 3104a barium standard as an MB-STD
  - ICPMS using Inorganic Ventures iodide (^{127}I) as a semi-quant EX STD

Iodine (I_2) is known to form in heated nitric acid. It is volatile at high temperatures, and thus there is potential for iodine loss during the dissolution of the fuel. Although the reflux condenser being used has been previously shown to limit loss via volatilization, there is no guarantee that it will completely stop it. It is suggested for both ID-ICPMS and ICPMS that numerous channels be monitored, as there is a known partial interference of $^{238}UO^{++}$ at m/z127.5 which has the potential to lead to a negative bias on the determination of ^{129}I using ^{127}I. A minimum of five channels should be monitored spaced at 0.1AMU from the centroid mass for both methods of analysis.

3.3.2 Cesium

- Assay
  - ID-MC-ICPMS using NIST 3111a (^{133}Cs) as SPIKE-STD and using an NIST 3104a barium standard as an MB-STD
  - ID-RAPID using NIST 3111a (^{133}Cs) as SPIKE-STD and using an offline NIST 3104a barium standard as an MB-STD.

Natural barium is used as it covers the masses of the non-natural cesium isotopes and although elementally the responses will be differently, the machine bias will be the same for the same masses.

- Isotopic
  - MC-ICPMS NIST 3104a barium standard as an MB-STD
  - RAPID NIST 3104a barium standard as an MB-STD
  - RAPID-HPGe for ^{134}Cs/^{137}Cs ratio, measuring on three independent HPGe detectors

Natural barium is used as the Nat-STD and MB-STD, as natural cesium is monoisotopic and barium covers the masses of the non-natural cesium isotopes produced in fission. Although it could be argued that elementally the responses of cesium and barium will be different, the machine bias will be the same for the same masses within the element.

3.3.3 Barium-137m

- Assay
  - Cesium-137 mass will be determined via ID-MC-ICPMS using NIST 3111a (^{133}Cs) as SPIKE-STD and using an NIST 3104a barium standard as an MB-STD.
  - Cesium-137 mass will be determined via ID-RAPID using NIST 3111a (^{133}Cs) as SPIKE-STD and using an offline NIST 3104a barium standard as an MB-STD.
  - HPGe will be used for direct analysis of ^{137m}Ba from a weighted dilution, measuring on three independent HPGe detectors.

The determined ^{137}Cs mass in ug/g fuel will be converted to activity in Bq/g fuel. Since ^{137m}Ba is in secular equilibrium with ^{137}Cs after ~18 minutes in the fuel sample, the activity and thus the mass of ^{137m}Ba remains constant and is directly related to the ^{137}Cs concentration. The uncertainty assigned will be equal to that determined for ^{137}Cs

Natural barium is used as it covers the masses of the non-natural cesium isotopes and although elementally the responses will be differently, the machine bias will be the same for the same masses.

3.3.4 Neodymium

- Assay
  - ID-MC-ICPMS using Nd-150-STD (^{150}Nd) as SPIKE-STD, using La Jolla and NIST 3135a as the Nat-STD, and using JNDI (JNdi-1) as an MB-STD
  - ID-RAPID using Nd-150-STD (^{150}Nd) as SPIKE-STD, using NIST 3135a as the Nat-STD, and using HPS-Nd as an MB-STD

- Isotopic
  - MC-ICPMS using JNDI (JNdi-1) as lanthanum as an MB-STD
  - RAPID using La Jolla neodymium standard as an MB-STD

Neodymium-142, ^{144}Nd, ^{148}Nd, and ^{150}Nd have potential ^{142}Ce and ^{144}Ce, ^{148}Sm, and ^{150}Sm interferences. Cesium-140, ^{147}Sm, and ^{152}Sm should also be monitored (monitoring mass 152 will avoid overcorrection using mass 147 because of potential contribution being from ^{147}Pm.

3.3.5 Cerium-144

- Assay
  - ID-MC-ICPMS using Ce-140-STD (^{140}Ce) as a SPIKE-STD, using HPS-Ce as the Nat-STD, and using NIST 3110 standard as an MB-STD. Cerium-142 will be used for a higher-precision cerium concentration.
  - ID-RAPID using Ce-140-STD (^{140}Ce) as SPIKE-STD, using HPS-Ce as the Nat-STD, and using NIST 3110 standard as an MB-STD. Cerium-142 will be used for a higher-precision cerium concentration.
  - HPGe will be employed without separation to directly measure ^{144}Ce by aliquoting a weighted fraction of the fuel sample and measuring on three independent HPGe detectors.

- Isotopic
  - MC-ICPMS using NIST 3110 standard as an MB-STD
  - RAPID using NIST 3110 standard as an MB-STD

Cerium-144 could potentially have a ^{144}Nd interference by mass; ^{146}Nd should also be monitored for potential neodymium content.

3.3.6 Promethium-147

- Assay
  - ID-RAPID using Sm-149-STD (^{149}Sm) as SPIKE-STD, using NIST 3147a as the Nat-STD, and using HPS-Sm standard as an MB-STD. The approach is a semi-quantitative approach based on the mass, the charge density, and first ionization enthalpy being almost identical to the ^{147}Pm isobar ^{147}Sm.
  - ICPMS using EX and using NIST 3147a with the ^{147}Pm/^{147}Sm ratio obtained from RAPID to yield a percentage contribution of ^{147}Pm to the quantified ^{147}Sm.
  - RAPID-LSC will be used as a tertiary analysis. Although lower precision is likely, the direct traceability of this method will validate the other two analytical methods.

3.3.7 Samarium

- Assay
  - ID-MC-ICPMS using Sm-149-STD (^{149}Sm) as SPIKE-STD, using HPS-Sm as the Nat-STD, and using NIST 3147a standard as an MB-STD
  - ID-RAPID using Sm-149-STD (^{149}Sm) as SPIKE-STD, using HPS-Sm as the Nat-STD, and using NIST 3147a standard as an MB-STD

- Isotopic
  - MC-ICPMS using NIST 3147a standard as an MB-STD
  - RAPID using NIST 3147a standard as an MB-STD

Samarium-142, ^{144}Sm, ^{145}Sm, ^{148}Sm, and ^{150}Sm, have potential ^{142}Nd, ^{144}Nd, ^{145}Nd, ^{148}Nd, and ^{150}Nd interferences; and ^{151}Sm, and ^{154}Sm have ^{151}Eu, and ^{154}Eu interferences. Neodymium-146 and ^{153}Eu should also be monitored. [13]

3.3.8 Europium

- Assay
  - ID-MC-ICPMS using Eu-151-STD (^{151}Eu) as SPIKE-STD, using NIST 3117a as the Nat-STD, and using NIST 3147a standard as an MB-STD
  - ID-RAPID using Eu-151-STD (^{151}Eu) as SPIKE-STD, using NIST 3117a as the Nat-STD, and using NIST 3147a standard as an MB-STD

- Isotopic
- MC-ICPMS using NIST 3147a standard as a MB-STD.
- RAPID using NIST 3147a standard as a MB-STD.
- RAPID-HPGe for Eu-154/155 ratio confirmation measurement

Europium-151 and ^{154}Eu have potential ^{151}Sm and ^{154}Sm/^{154}Gd interferences. Europium-153 and ^{155}Eu have potential ^{153}Gd and ^{155}Gd interferences Gadolinium-156 and ^{147}Sm should also be monitored. NIST 3147a is a natural samarium standard that has a known 150/154 ratio to high precision (0.3244) that has been used to determine ^{151}Eu/^{153}Eu isotope ratios [14].

3.3.9 Gadolinium-155

- Assay
  - ID-MC-ICPMS using Gd-155-STD (^{155}Gd) as SPIKE-STD, using NIST 3118a as the Nat-STD, and using NIST 3118a as an MB-STD.
  - ID-RAPID using Gd-155-STD (^{155}Gd) as SPIKE-STD, using NIST 3118a as the Nat-STD, and using NIST 3118a as an MB-STD.

This will be determined using ^{156}Gd and reverse IDMS.

3.4 LOW-MASS FISSION PRODUCTS

In this SAP, the low-mass fission isotopes are defined as those with a mass of 79-125 amu. This encompasses selenium and antimony as well as the second-row transition elements: strontium, molybdenum, technetium, ruthenium, rhodium, and silver. Analyses of the LMFPs are more complicated than analysis of their high-mass equivalents, as elemental isolation is not facile. The observed baseline resolutions from both isobaric and potential polyatomic interferences are more complicated. As with the HMFPs, to achieve the analytical precision required, several potential analytes will be monitored using various clean masses with no possible isobaric interferences. Table 6 shows the LMFP analytes together with the ORIGEN-calculated content in fuel, the proposed analytical methods, and appropriate traceable standards to be employed.

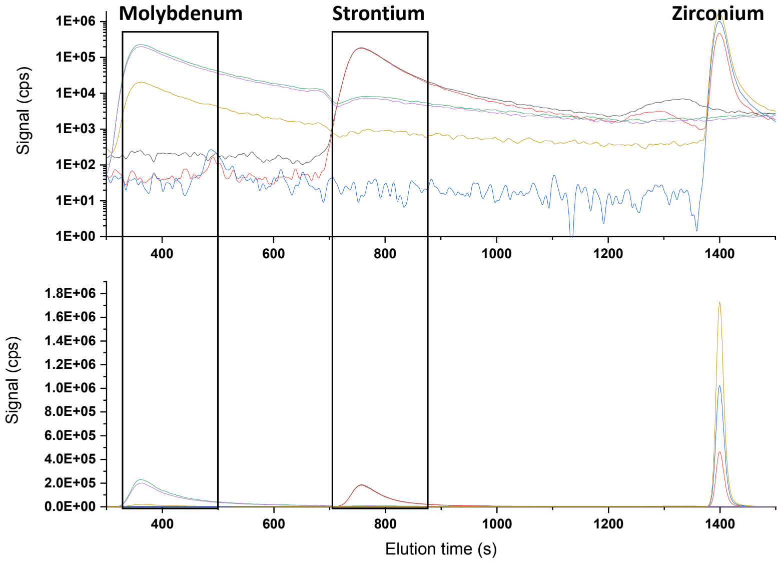


Figure 6. Graph showing the separated spectra of a spent nuclear fuel sample, the monitored m/z traces: 88 (gray), 90 (red), 91 (blue), 95 (purple), 96 (ochre), and 97 (green) for the elution period 250–1500 s. The highlighted peaks are molybdenum and strontium; zirconium also is monitored because of its interfering isobars. The bottom figure has a y-axis in linear scale and the top figure has a y-axis in log scale.

Table 6. The high-mass fission product analytes, together with the ORIGEN-calculated content in fuel, the proposed analytical methods, and appropriate traceable standards to be employed.

Analyte	Half-life	ORIGEN content* (µg/g fuel)	Primary analytical method /standard	Secondary/ tertiary analytical method/s	SPIKE-STD (enriched isotope)	Nat-STD (See APPENDIX D)	MB-STD (See APPENDIX D)
^{79}Se	$3.27E^5$ Y	6 (9%)	ID-MC-ICPMS	ID-RAPID	Se-77-STD (^{77}Se)	HPS-Se	NIST 3149
^{95}Mo	Stable	883 (22%)	ID-MC-ICPMS	ID-RAPID	Mo-94-STD (^{94}Mo)	HPS-Mo	NIST 3134
^{90}Sr	28.79 Y	384 (49%)	ID-MC-ICPMS	ID-RAPID	Sr-86-STD (^{86}Sr)	HPS-Sr	NIST 3153a
^{99}Tc	$2.11E^5$ Y	923 (100%)	SA-RAPID	SA-ICPMS RAPID-LSC	NIST 4288B	N/A	N/A
^{101}Ru	Stable	910 (34%)	ID-MC-ICPMS	ID-RAPID	Ru-99-STD (^{99}Ru)	Inorganic Ventures CGRU10	HPS-Ru
^{106}Ru	371.8 D	0.2ng	ID-MC-ICPMS HPGe	HPGe	Ru-99-STD (^{99}Ru)	NA	NA
^{103}Rh	Stable	545 (100%)	SA-ICPMS	ICPMS	N/A	SRM3144	NA
^{109}Ag	Stable	70 (100%)	ID-MC-ICPMS	ID-RAPID	Ag-107-STD (^{107}Ag)	HPS-Ag	SRM3151
^{125}Sb	2.758 Y	0.01(0.6%)	ID-MC-ICPMS HPGe	N/A	Sb-123-STD (^{123}Sb)	HPS-Se	NIST 3102a

*ORIGEN-calculated analyte content in µg/g fuel—using a standard 23.9 MW power reactor, three cycles of 416 days irradiation, each irradiation followed by 30-day decay, employing 4% enriched UO2 green fuel. To be used for estimates only.

3.4.1 Selenium-79

Selenium readily dissolves in nitric acid to form selenic acid, which has been shown to be a volatile substance at high temperatures. In high uranium solutions, selenium also has the potential to form U-based complexes. To mitigate loss of selenium during analyses and processing, any heat applied to either standards or samples should be limited to 200°C.

- Assay
  - ID-MC-ICPMS using Se-77-STD (^{77}Se) as SPIKE-STD, using HPS-Se as the Nat-STD, and using NIST 3149 standard as an MB-STD
  - ID-RAPID using Se-77-STD (^{77}Se) as SPIKE-STD, using HPS-Se as the Nat-STD, and using NIST 3149 standard as an MB-STD

Mass 81 should be monitored for residual bromine. However, the collected fraction will be fumed in 16N HNO_3 to remove any HBr in the sample before analysis by ID-MC-ICPMS. Mass 77 has the potential to have Ar-^{37}Cl interferences. Mass 75 should be monitored for the analogous Ar-^{35}Cl species

3.4.2 Molybdenum-95

- Assay
  - ID-MC-ICPMS using Mo-94-STD (^{94}Mo) as SPIKE-STD, using HPS-Mo as the Nat-STD, and using the NIST 3134 standard as an MB-STD
  - ID-RAPID using Mo-94-STD (^{94}Mo) as SPIKE-STD, using HPS-Mo as the Nat-STD, and using NIST 3134 standard as an MB-STD

Mass 91 should be monitored for potential zirconium interference on ^{94}Mo.

3.4.3 Strontium-90

- Assay
  - ID-MC-ICPMS using Sr-86-STD (^{86}Sr) as SPIKE-STD, using HPS-Sr as the Nat-STD, and using NIST 3153a standard as an MB-STD
  - ID-RAPID using Sr-86-STD (^{86}Sr) as SPIKE-STD, using HPS-Sr as the Nat-STD, and using NIST 3153a standard as an MB-STD

Krypton mass 91 should be monitored for potential krypton interference on ^{86}Sr and zirconium interference on ^{90}Sr.

3.4.4 Technetium-99

- Assay
  - SA-RAPID using SRM3144 SRM4288B
  - SA-ICPMS using SRM4288B
  - RAPID-LSC using SRM4288B

Mass 101 should be monitored for potential ruthenium interference.

3.4.5 Ruthenium-101

- Assay
  - ID-MC-ICPMS using Ru-99-STD (^{99}Ru) as SPIKE-STD, Inorganic Ventures CGRU10 as the Nat-STD, and HPS-Ru as an MB-STD
  - ID-RAPID using Ru-99-STD (^{99}Ru) as SPIKE-STD, Inorganic Ventures CGRU10 as the Nat-STD, and HPS-Ru as an MB-STD

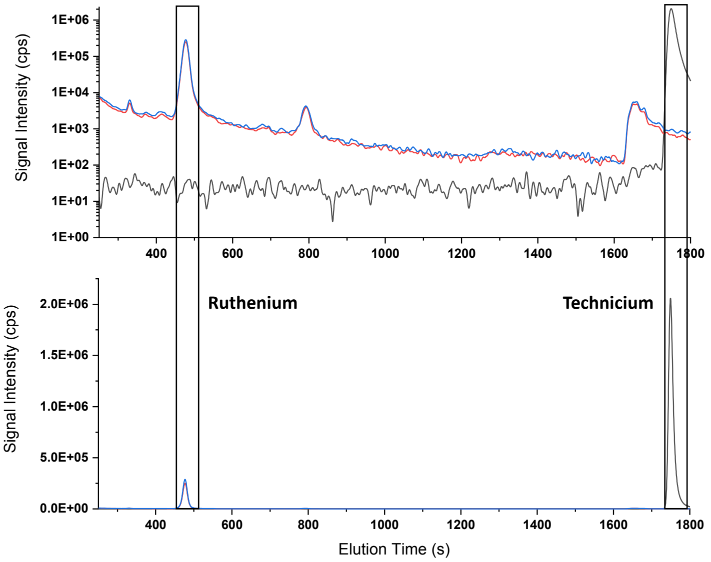


Figure 7. Graph showing the monitored m/z traces: 99 (gray), 101 (red), and 102 (blue) for the elution period 250–1850s. The highlighted peaks are ruthenium and technetium, showing no risk of ^{99}Tc interfering with the suggested ^{99}Ru spike. The bottom figure has a y-axis in linear scale and top figure has a y-axis in *l*og scale.

3.4.6 Ruthenium-106

- Assay
  - Indirect ID-MC-ICPMS using Ru-99-STD (^{99}Ru) as SPIKE-STD, using Inorganic Ventures CGRU10 as the Nat-STD, and using HPS-Ru as a MB-STD to determine ^{101}Ru.
  - RAPID-HPGe using ID-MC-ICPMS using Ru-99-STD (^{99}Ru) as SPIKE-STD to determine recovery of ^{101}Ru. This will yield a ^{106}Ru/^{101}Ru ratio to be used to determine the ^{106}Ru assay.

3.4.7 Rhodium-103

- Assay
  - SA-RAPID using NIST 4288B
  - SA-ICPMS using NIST 4288B

3.4.8 Silver-109

- Assay
  - ID-MC-ICPMS using Ag-107-STD (^{107}Ag) as SPIKE-STD, using HPS-Ag as the Nat-STD, and using SRM3151as an MB-STD
  - ID-RAPID using Ag-107-STD (^{107}Ag) as SPIKE-STD, using HPS-Ag as the Nat-STD, and using SRM3151as an MB-STD.

Mass 105 and 108 should be monitored for potential palladium interference on ^{107}Ag.

3.4.9 Antimony-125

- Assay
  - Indirect ID-MC-ICPMS using Sb-123-STD (^{123}Sb) as SPIKE-STD, using NIST 3102a as the Nat-STD, and using ??? standard as an MB-STD to determine ^{121}Sb.
  - RAPID-HPGe to determine ^{125}Sb in the fraction, and ID-MC-ICPMS using Sb-123-STD (^{123}Sb) as SPIKE-STD to determine recovery of ^{121}Sb. This will yield a ^{125}Sb/^{121}Sb ratio to be used to determine the ^{125}Sb assay.

3.5 CREATING A MULTI-ELEMENT SPIKE WITH ENRICHED ISOTOPES FOR IDMS

To achieve the highest-precision measurement by IDMS, the spike isotope that is added to the sample should result in an analyte-isotope (*i*) to spiked-isotope (*k*) (e.g., ^{144}Nd:^{150}Nd) ratio that is as close to 1 as possible. Ideally, each individual enriched spike would be added to the sample by weight to achieve *i:k* ratios of 1 for all analytes. Adding 16 different *k* isotopes, however, would significantly dilute the sample and would require sample concentration post-spiking. The larger the total solution volume, the longer the heat treatment that will be required, potentially leading to loss/contamination of the sample. To avoid this, the samples will be spiked with only two multi-element spikes. The first will be a multi-element spike to measure each of the fission elements whose concentration is to be determined by IDMS. The second will be an actinide spike plus ruthenium and iodine for the same purpose.

The approximate spike concentrations used in the multi-element spikes are given in Table 7 in ug/g solution and were calculated as the mean of 16 independent ORIGEN calculations (see Appendix C) for SNF segments to be analyzed in ug/g U and the approximate U concentration of the 5 mg U/g solution, as detailed in Section 2.

Table 7. The standard, enriched isotope and recommended approximate concentration for multi-spike-A and multi-spike-B.

Multi-spike-A			Multi-spike-B		
Enriched isotope	**Standard**	**Multi-spike concentration (ug/g soln.)**	**Enriched isotope**	**Standard**	**Multi-spike concentration (ug/g soln.)**
^{107}Ag	Ag-107-STD	0.85	^{242}Pu	CRM-130	44.79
^{140}Ce	Ce-140-STD	13.95	^{243}Am	SRM-4332E	7.67
^{133}Cs	CGCS1	12.3	^{246}Cm	WRM-Cm-246	0.254
^{151}Eu	Eu-151-STD	1.261	^{99}Ru	Ru-99-STD	8.585
^{155}Gd	Gd-155-STD	1.044	^{127}I	I-127-STD	1.228
^{94}Mo	Mo-94-STD	8.334			
^{150}Nd	Nd-150-STD	12.96			
^{123}Sb	Sb-123-STD	0.064			
^{77}Se	Se-77-STD	0.045			
^{149}Sm	Sm-149-STD	3.244			
^{86}Sr	Sr-86-STD	3.621			

4. ANALYTICAL PROTOCOLS

4.1 LABWARE CLEANING

The low-density polyethylene bottles and PFA labware/vials used for digestion, dilutions, and analysis will be purchased new for this project from Fisher Scientific or Savillex. Initial washing will be done with Micro-90. Labware will be leached in 6 M HCl followed by 8 M HNO_3 and finally 18.2 MΩ•cm water each for a 24-hour time period. After rinsing. the labware will be dried in a Class 10 laminar flow hood and stored in new, clean plastic containers until needed for project activities.

4.2 DAVIES-GRAY TITRATION

The D-G titration scheme will be carried out according to Table 8.

Table 8. Davies-Gray titration scheme.

Titration day 1
Blank
CRM 112-A
CRM 129-A
Process blank-1
LCS
Sample-A
Sample-B
CRM 112-A
CRM 129-A

4.3 ID-MC-ICPMS

The ID-MC-ICPMS measurements will be carried out according to Table 9 for each individual element.

Table 9. ID-MC-ICPMS measurement scheme.

ID-MC-ICPMS Day 1
Blank
Control
Control
Process blank-1
LCS
Sample A
Sample B
Blank
Control
Blank
Spiked sample A
Spiked sample B
Control
Control

4.4 ID-RAPID

ID-RAPID measurements will be carried out according to Table 10. The analytes will be split into two independent runs varying only by dilution and m/z traces monitored. The first will be for actinides and HMFPs (analyzed at a higher dilution), and the second will be for the LMFPs (analyzed at a lower dilution).

Table 10. ID-RAPID measurement scheme.

ID-RAPID run 1 (HMFP + actinides)	ID-RAPID run 2 (LMFP)
Blank × 2	Blank × 2
MB-STD × 3	MB-STD × 3
Blank	Blank
Process blank-1	Process blank-1
LCS × 3	LCS × 3
Blank × 2	Blank × 2
Sample A × 3	Sample A × 3
Blank	Blank
Sample B × 3	Sample B × 3
Blank	Blank
Sample A × 3	Sample A × 3
Blank	Blank
Spiked sample A × 3	Spiked sample A × 3
Blank	Blank
Spiked sample B × 3	Spiked sample B × 3
Blank × 2	Blank × 2
MB-STD × 3	MB-STD × 3

4.5 EX-RAPID

Isotope measurements via the EX-RAPID separation protocol will be conducted using the measurement protocol in Table 11 and NIST-traceable standards by EX using a linear regression curve.

Table 11. EX-RAPID measurement scheme.

EX-RAPID
Blank
STD-1
STD-2
STD-3
ICV
ICB
LCS
Blank
Sample A
Blank
Sample B
Blank
FCV
FCB

4.6 SA-ICPMS

Isotopes measured using SA analysis will be performed by weight using a weighted dilution of the original aliquot, which will then be spiked by weight with NIST-traceable standards of known concentration. The measurement scheme is outlined in Table 12 and linear regression analysis of the resultant slope will yield the concentration and uncertainty.

Table 12. SA-ICPMS measurement scheme.

SA-ICPMS
Blank
Sample A
Sample A + Spike-1
Sample A + Spike-2
Sample A + Spike-3
Blank
Sample B
Sample B + Spike-1
Sample B + Spike-2
Sample B + Spike-3
Blank
LCS
LCS + Spike-1
LCS + Spike-2
LCS + Spike-3
Blank

4.7 ICPMS

Isotopes measured via ICPMS will be conducted using the measurement protocol in

Table 12 and NIST-traceable standards by EX using a linear regression curve. Additional isotopes that have isobaric interferences from the EX standards will be semi-quantified to a reference response.

Table 12. ICPMS measurement scheme.

ICPMS
Blank
STD-1
STD-2
STD-3
ICV
ICB
LCS
Blank
Sample A
Blank
Sample B
Blank
CCV
CCB

5. QUALITY ASSURANCE AND QUALITY CONTROL

All analytical operations performed will be compliant with the ORNL Chemical Sciences Division Quality Assurance Plan [15].

5.1 BALANCE CALIBRATION

Analytical balances readable to 0.01 mg that are within calibration will be used for all spike preparations and D-G analyses. Analytical balances readable to 0.10 mg will be used for the rest of the analytical mass measurements. The balance will be checked before and after use, and sample weights will be recorded into a logbook and log report that will be provided. The balances and calibration weight sets will have been calibrated by the ORNL Metrology Laboratory, which maintains accreditation to ISO/IEC 17025 through the National Voluntary Laboratory Accreditation Program.

5.2 PRODUCTION OF A LABORATORY CONTROL STANDARD

The laboratory control standard (LCS) will be produced by heat treating a portion of the WRM, which will be produced and certified according to “WA-LCS-01: Production of an Irradiated Nuclear Fuel Surrogate Working Reference Material” [16]. The WRM will be produced in accordance with ASTM C1128-18, “Standard Guide for Preparation of Working Reference Materials for Use in Analysis of Nuclear Fuel Cycle Materials” [17]. The composition of the WRM will be representative of approximately 2.25 g of dissolved irradiated nuclear fuel in 100 ml 12M HNO3.

In a radiological fume hood, the WRM will be quantitatively transferred by weight into a dissolution pressure vessel and reflux apparatus setup as described in Section 1.3. In short, a digestion bottle will be mated to a recirculating water-chilled condenser and maintained at 10°C for the entire heating period. The pressure vessel will be placed directly onto a laboratory hotplate, which will be heated to surface temperatures of 170°C to 180°C (solution temperature of ~150°C). The heating period will then be maintained for 40 to 50 hours under constant reflux.

The solution will be cooled to room temperature with the recirculating condenser in place. When cool, the solution will be rinsed through a 0.45 µm cellulose acetate filter into a pre-weighed Teflon dilution bottle using a vacuum draw. The cellulose acetate filter will be rinsed three times with HNO_3 (10 mL, 12M, Optima) into the dilution bottle. The solution will be brought to a total volume of ~100 mL with HNO_3 (12M, Optima), and a final solution weight will be taken. The LCS will be inverted no fewer than 30 times to homogenize the solution, and then it will be stored in a sealed Mylar bag.

5.3 ANALYTICAL BLANKS

Analytical blanks and process blanks will be included with each sample set to identify and, if appropriate, correct for any contamination that may occur during processing.

5.4 RECORD KEEPING

Careful documentation will be kept for the sample preparation, analysis procedure, and data reduction. For ease of data reduction and eventual reporting, Microsoft Excel spreadsheets will be submitted after each group of elements are measured by the specific technique. Quantitative results for blanks and quality control samples will be included in the final report. Logbooks will be kept for all mass measurements, environmental parameter recording (temperature, humidity, atmospheric pressure), and associated dates, and submitted in the form of a digital logbook that is a transcription of the physical logbook into an Excel spreadsheet.

All laboratory work should be documented in a project-numbered notebook, according to the Records Management standard in the Standards-Based Management System. The following information should also be documented.

- This document in its most current version
- Subsampling aliquot weights together with times and dates
- Any ORIGEN model output files with details about the irradiations
- The NIST and NIST-traceable certificate of analyses (COAs) for all standards and reference materials
- The GUM Workbench budget tables for the assigned uncertainty
- All measurement methods used for each analyte identified
- The names and badge numbers of all analysts who performed the work
- All reagent vendor supplier and lot numbers.
- All balances, balance check weight sets, and pipettes used and their calibrations
- The environmental conditions measured for buoyancy correction, as needed.

All electronic documentation will be locked and stored, including but not limited to spreadsheets used for planning, uncertainty calculations, raw analytical data on instrument computers, and data reduction spreadsheets. All required calculations will be recorded.

5.5 UNITS, UNCERTAINTY, AND TRACEABILITY

Uranium concentration results will be reported on a mass basis as g U/g solutions. Uranium isotopic compositions will be reported as isotope mass ratios. The remainder of the analytes will be reported as isotope concentrations and will be reported as mass fractions relative to the determined U content in µg/gU. Calculations will be made to determine the µg/g sample to calculate the mass balance. Measurement uncertainties will be calculated for all measurement results reported by ORNL using GUM Workbench software. International Union of Pure and Applied Chemistry values for atomic weights will be used in all calculations. All environmental readings will be taken using SI-traceable devices. [15, 18]

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APPENDIX A. ORIGEN Calculated Fission Isotope Content in SNF

Table A.1. Complete analyte isotope list and ORIGEN calculated amount in ug/g Fuel and ug/g U together with elemental concentration in ug/g U.

20 y decay	ug/g fuel	ug/gU	Total ug/g U
Ag-107	**NA**	**NA**	**9.48E+01**
ag-109	**7.96E+01**	**9.48E+01**	
ag-110m	9.51E-10	1.13E-09	
am-241	**8.37E+02**	**9.97E+02**	**1.14E+03**
am-242m	**6.26E-01**	**7.46E-01**	
am-243	**1.22E+02**	**1.45E+02**	
ba-134	*2.02E+02*	*2.41E+02*	***2.49E+03***
ba-135	*5.02E-01*	*5.98E-01*	
ba-136	*2.25E+01*	*2.68E+01*	
ba-137	*5.16E+02*	*6.15E+02*	
ba-137m	**NA**	**NA**	
ba-138	*1.35E+03*	*1.61E+03*	
ba-140	*0.00E+00*	*0.00E+00*	
cd-110	*4.03E+01*	*4.80E+01*	***1.19E+02***
cd-111	*2.53E+01*	*3.01E+01*	
cd-112	*1.31E+01*	*1.56E+01*	
cd-113	*1.13E-01*	*1.34E-01*	
cd-114	*1.53E+01*	*1.82E+01*	
cd-116	*5.66E+00*	*6.74E+00*	
ce-140	**1.30E+03**	**1.55E+03**	**2.93E+03**
ce-141	0.00E+00	0.00E+00	
ce-142*	**1.16E+03**	**1.39E+03**	
ce-144	**5.71E-06**	**6.81E-06**	
cm-242	**1.63E-03**	**1.94E-03**	**3.36E+01**
cm-243	**3.43E-01**	**4.09E-01**	
cm-244	**2.41E+01**	**2.87E+01**	
cm-245	**3.36E+00**	**4.00E+00**	
cm-246	**4.03E-01**	**4.80E-01**	
cs-133	**1.15E+03**	**1.37E+03**	**2.81E+03**
cs-134	**1.72E-01**	**2.05E-01**	
cs-135	**4.18E+02**	**4.98E+02**	
cs-137	**7.92E+02**	**9.43E+02**	
eu-151	**1.62E+00**	**1.93E+00**	**1.51E+02**
eu-153	**1.19E+02**	**1.42E+02**	
eu-154	**5.69E+00**	**6.78E+00**	
eu-155	**4.78E-01**	**5.69E-01**	
gd-152	2.84E-02	3.38E-02	**1.92E+02**
gd-154	2.56E+01	3.05E+01	

Table A.1. Complete analyte isotope list and ORIGEN calculated amount in ug/g Fuel and ug/g U together with elemental concentration in ug/g U (continued).

20 y decay	ug/g fuel	ug/gU	Total ug/g U
gd-155	**8.43E+00**	**1.00E+01**	
gd-156*	**1.06E+02**	**1.26E+02**	
gd-157	1.21E-01	1.45E-01	
gd-158	1.97E+01	2.35E+01	
gd-160	1.13E+00	1.35E+00	
i-127	**4.58E+01**	**5.46E+01**	**2.46E+02**
i-129	**1.61E+02**	**1.91E+02**	
i-131	0.00E+00	0.00E+00	
mo-100	9.72E+02	1.16E+03	**4.15E+03**
mo-94	**1.13E-02**	**1.35E-02**	
mo-95	**7.77E+02**	**9.26E+02**	
mo-96	4.54E+01	5.41E+01	
mo-97	8.29E+02	9.87E+02	
mo-98	8.60E+02	1.02E+03	
nd-142	2.36E+01	2.81E+01	**5.01E+03**
nd-143	**8.03E+02**	**9.56E+02**	
nd-144	**1.39E+03**	**1.66E+03**	
nd-145	**6.85E+02**	**8.16E+02**	
nd-146	**7.32E+02**	**8.72E+02**	
nd-147	**0.00E+00**	**0.00E+00**	
nd-148	**3.84E+02**	**4.58E+02**	
nd-150	**1.85E+02**	**2.20E+02**	
np-237	**4.76E+02**	**5.67E+02**	**5.67E+02**
pd-104	*2.51E+02*	*2.99E+02*	**1.77E+03**
pd-105	*4.09E+02*	*4.87E+02*	
pd-106	*3.92E+02*	*4.67E+02*	
pd-107	*2.30E+02*	*2.74E+02*	
pd-108	*1.53E+02*	*1.82E+02*	
pd-110	*5.08E+01*	*6.05E+01*	
pu-238	**1.91E+02**	**2.27E+02**	**9.62E+03**
pu-239	**4.74E+03**	**5.65E+03**	
pu-240	**2.08E+03**	**2.47E+03**	
pu-241	**4.96E+02**	**5.91E+02**	
pu-242	**5.65E+02**	**6.73E+02**	
pu-244	3.63E-02	4.33E-02	
rh-103	**4.80E+02**	**5.71E+02**	**5.71E+02**
ru-100	1.14E+02	1.35E+02	**2.78E+03**
ru-101	**8.01E+02**	**9.54E+02**	
ru-102	8.38E+02	9.99E+02	
ru-103	**3.54E-55**	**4.22E-55**	

Table A.1. Complete analyte isotope list and ORIGEN calculated amount in ug/g Fuel and ug/g U together with elemental concentration in ug/g U (continued).

20 y decay	ug/g fuel	ug/gU	Total ug/g U
ru-104	5.81E+02	6.92E+02	
ru-106	**1.86E-04**	**2.21E-04**	
ru-99	**8.56E-02**	**1.02E-01**	
sb-121	4.62E+00	5.51E+00	**1.27E+01**
sb-123	**5.97E+00**	**7.11E+00**	
sb-125	**5.86E-02**	**6.98E-02**	
se-77	**8.10E-01**	**9.65E-01**	**6.64E+01**
se-78	2.36E+00	2.82E+00	
se-79	**4.98E+00**	**5.93E+00**	
se-80	1.31E+01	1.57E+01	
se-82	3.44E+01	4.10E+01	
sm-147	**2.50E+02**	**2.98E+02**	**1.03E+03**
sm-148	1.55E+02	1.85E+02	
sm-149	**3.01E+00**	**3.59E+00**	
sm-150	**3.06E+02**	**3.64E+02**	
sm-151	**9.66E+00**	**1.15E+01**	
sm-152	**9.93E+01**	**1.18E+02**	
sm-154	3.87E+01	4.62E+01	
sn-115	*2.47E-01*	*2.94E-01*	**7.22E+01**
sn-116	*3.62E+00*	*4.31E+00*	
sn-117	*5.15E+00*	*6.14E+00*	
sn-118	*4.81E+00*	*5.73E+00*	
sn-119	*4.75E+00*	*5.66E+00*	
sn-120	*4.80E+00*	*5.72E+00*	
sn-121m	*2.71E-01*	*3.23E-01*	
sn-122	*6.11E+00*	*7.28E+00*	
sn-124	*9.70E+00*	*1.16E+01*	
sn-126	*2.11E+01*	*2.52E+01*	
sr-86	**5.49E-01**	**6.54E-01**	**8.19E+02**
sr-88	3.49E+02	4.16E+02	
sr-89	6.15E-43	7.33E-43	
sr-90	**3.38E+02**	**4.03E+02**	
tc-99	**8.12E+02**	**9.67E+02**	**9.67E+02**
te-122	*3.95E-01*	*4.71E-01*	**5.99E+02**
te-124	*3.29E-01*	*3.92E-01*	
te-125	*1.29E+01*	*1.53E+01*	
te-125m	*8.18E-04*	*9.74E-04*	
te-126	*5.71E-01*	*6.80E-01*	
te-127m	*4.16E-21*	*4.96E-21*	
te-128	*9.69E+01*	*1.15E+02*	

Table A.1. Complete analyte isotope list and ORIGEN calculated amount in ug/g Fuel and ug/g U together with elemental concentration in ug/g U (continued).

20 y decay	**ug/g fuel**	**ug/gU**	**Total ug/g U**
te-129m	*4.23E-66*	*5.04E-66*	
te-130	*3.91E+02*	*4.66E+02*	
u-233	**NA**	**NA**	
u-234	**3.49E+01**	**4.16E+01**	**1.00E+06**
u-235	**6.71E+03**	**7.99E+03**	
u-236	**4.13E+03**	**4.92E+03**	
u-237	**1.55E-05**	**1.84E-05**	
u-238	**8.28E+05**	**9.87E+05**	
zr-90	*2.36E+02*	*2.82E+02*	**4.60E+03**
zr-91	*6.08E+02*	*7.24E+02*	
zr-92	*6.63E+02*	*7.89E+02*	
zr-93	*7.31E+02*	*8.71E+02*	
zr-94	*7.88E+02*	*9.39E+02*	
zr-95	*2.43E-33*	*2.90E-33*	
zr-96	*8.33E+02*	*9.93E+02*	

*Pseudo-analyte isotopes where the intended analyte to be measured by IDMS is also the isotope in the enriched spike.

All isotopes highlighted in **red** are predetermined analytes. All isotopes highlighted in **blue** are the defined enriched isotope to be used in IDMS. All isotopes highlighted in **green** are to be determined using SA. All isotopes in *italics* are potential impurities to be considered. All remaining isotopes (non-bold/colored/italicized) should preferentially be measured also.

APPENDIX B. RAPID Separation Scheme and Elution Profile

Table B.1. Separation scheme for the elemental isolation of neodymium, plutonium, and uranium.

Segment type	Segment duration	Segment end time	Deionized water	2 M hydrochloric acid	0.1 M diglycolic acid	0.15 M oxalic acid
Start	0 s	0 s	100%	0%	0%	0%
Ramp	300 s	300 s	40%	0%	0%	60%
Hold	240 s	540 s	40%	0%	0%	60%
Ramp	0 s	540 s	20%	0%	0%	80%
Ramp	540 s	1080 s	51%	0%	26%	23%
Ramp	120 s	1200 s	0%	0%	100%	0%
Hold	300 s	1500 s	0%	0%	100%	0%
Ramp	0 s	1500 s	0%	100%	0%	0%
Hold	120 s	1620 s	0%	100%	0%	0%
Ramp	120 s	1740 s	100%	0%	0%	0%
Hold	60 s	1800 s	100%	0%	0%	0%

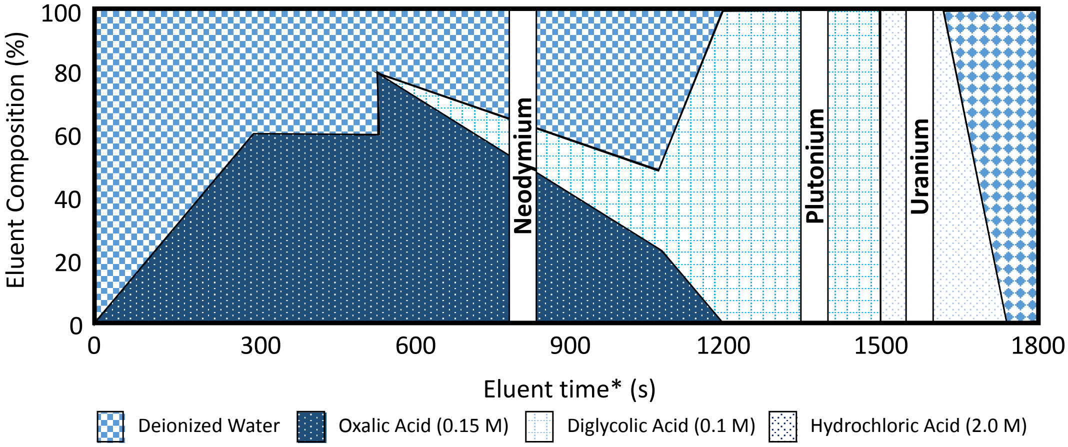

Figure B.1. Mixed-gradient elution profile illustrating the eluent composition at the time of neodymium, plutonium, and uranium detection.

APPENDIX C. ORIGEN-calculated Fission Isotope Masses for Individual SNF Samples

Tables C.1–C.4 are ORIGEN-calculated fission isotope masses (in ug/g U) for 16 specific SNF rod segments to be analyzed.

Table C.1. ORIGEN-calculated fission isotope masses (in ug/g U) for 3A1F05_150_165, 3F9N05_3593_3615, 30AD05_200_215, and 3A1F05_225_240.

Analyte	3A1F05_150_165 ug/gU	3F9N05_3593_3615 ug/gU	30AD05_200_215 ug/gU	3A1F05_225_240 ug/gU
se79	5	6	7	7
sr90	334	441	588	401
ru101	858	890	1032	1087
rh103	539	553	624	654
i129	165	171	200	218
cs133	1268	1317	1504	1553
cs135	413	448	543	519
cs137	743	964	1296	943
nd143	949	993	1114	1085
nd144	1429	1487	1762	1889
nd145	765	796	911	930
nd146	761	793	937	994
nd148	411	427	496	523
nd150	192	199	233	251
sm147	305	313	329	337
sm150	320	332	388	414
sm151	11	13	15	12
sm152	115	118	130	135
eu153	121	125	148	160
gd155	8	8	8	12

Table C.2. ORIGEN-calculated fission isotope masses (in ug/g U) for F35P17_3050_3069, F35P17_2383_2402, 3F9N05_2863_2882, and 3F9N05_2300_2329.

Analyte	F35P17_3050_3069 ug/gU	F35P17_2383_2402 ug/gU	3F9N05_2863_2882 ug/gU	3F9N05_2300_2329 ug/gU
se79	10	10	9	9
sr90	445	450	630	626
ru101	1604	1630	1428	1417
rh103	853	861	797	793
i129	350	357	297	294
cs133	2077	2102	1947	1934
cs135	729	743	710	703
cs137	1231	1253	1554	1541
nd143	1174	1177	1251	1248
nd144	3069	3134	2633	2605
nd145	1224	1238	1164	1157
nd146	1586	1619	1372	1357
nd148	785	799	695	688
nd150	400	408	341	338
sm147	354	355	371	370
sm150	617	627	551	546
sm151	13	14	15	15

sm152	171	173	160	159
eu153	235	238	211	209
gd155	20	20	16	15

Table C.3. ORIGEN-calculated fission isotope masses (in ug/g U) for 3D8E14_2655_2674, 3D8E14_2303_2322, 3D8E14_1375_1450, and 30AE14_2675_2694.

	3D8E14_2655_2674 ug/gU	3D8E14_2303_2322 ug/gU	3D8E14_1375_1450 ug/gU	30AE14_2675_2694 ug/gU
se79	10	9	10	9
sr90	606	597	611	777
ru101	1569	1537	1589	1498
rh103	846	836	853	824
i129	332	324	337	310
cs133	2086	2055	2106	2037
cs135	775	759	785	778
cs137	1555	1522	1575	1890
nd143	1275	1269	1279	1320
nd144	2966	2889	3012	2782
nd145	1244	1225	1256	1223
nd146	1539	1500	1563	1452
nd148	767	750	777	730
nd150	382	373	387	358
sm147	377	376	377	371
sm150	605	593	613	577
sm151	15	15	15	17
sm152	169	167	170	164
eu153	230	226	233	220
gd155	18	18	18	14

Table C.4. ORIGEN-calculated fission isotope masses (in ug/g U) for 30AD05_2410_2439, 30AD05_1280_1299, 3D8E14_775_796, 3A1F05_2383_2402.

Analyte	30AD05_2410_2439 ug/gU	30AD05_1280_1299 ug/gU	3D8E14_775_796 ug/gU	3A1F05_2383_2402 ug/gU
se79	9	9	10	8
sr90	761	764	610	461
ru101	1454	1464	1583	1317
rh103	807	811	851	754
i129	299	302	336	273
cs133	1990	2001	2101	1814
cs135	755	761	783	628
cs137	1832	1845	1569	1145
nd143	1307	1310	1278	1179
nd144	2679	2702	3001	2389
nd145	1196	1202	1252	1081
nd146	1400	1412	1557	1245
nd148	708	713	774	638
nd150	345	348	386	313
sm147	369	369	377	357
sm150	559	563	611	507
sm151	17	17	15	13
sm152	161	162	170	152
eu153	214	215	232	196
gd155	13	13	18	15

Tables C.5–C.8 show the calculated deviation from the mean of the ORIGEN-calculated fission isotope masses (in ug/g U) for 16 specific SNF rod segments. The average of the percentage deviation from the mean is calculated, as is the standard deviation of each data set. If a standard deviation is lower than 20%, then a single multi-spike standard, comprising isotopically enriched standards at varying levels, can be employed for all samples.

Table C.5. The calculated deviation from the mean of the ORIGEN-calculated fission isotope masses (in ug/g U) for 3A1F05_150_165, 3F9N05_3593_3615, 30AD05_200_215, and 3A1F05_225_240.

Analyte	3A1F05_150_165	3F9N05_3593_3615	30AD05_200_215	3A1F05_225_240
se79	35%	32%	21%	18%
sr90	41%	21%	-5%	29%
ru101	36%	34%	23%	19%
rh103	29%	27%	17%	13%
i129	41%	39%	28%	22%
cs133	31%	28%	18%	16%
cs135	38%	33%	18%	22%
cs137	46%	30%	6%	32%
nd143	20%	17%	6%	9%
nd144	42%	40%	29%	24%
nd145	31%	28%	17%	16%
nd146	41%	39%	27%	23%
nd148	37%	35%	24%	20%
nd150	40%	38%	28%	22%
sm147	14%	12%	7%	5%
sm150	38%	36%	25%	20%
sm151	21%	10%	-3%	14%
sm152	25%	23%	15%	12%
eu153	39%	37%	25%	19%
gd155	42%	45%	43%	17%
Average Deviation	**34%**	**30%**	**19%**	**19%**
STD	**9%**	**9%**	**12%**	**6%**

Table C.6. The calculated deviation from the mean of the ORIGEN-calculated fission isotope masses (in ug/g U) for F35P17_3050_3069, F35P17_2383_2402, 3F9N05_2863_2882, and 3F9N05_2300_2329.

Analyte	F35P17_3050_3069	F35P17_2383_2402	3F9N05_2863_2882	3F9N05_2300_2329
se79	-15%	-17%	-6%	-5%
sr90	21%	20%	-12%	-11%
ru101	-19%	-21%	-6%	-5%
rh103	-13%	-14%	-5%	-5%
i129	-25%	-28%	-6%	-5%
cs133	-13%	-14%	-6%	-5%
cs135	-10%	-12%	-7%	-6%
cs137	11%	9%	-13%	-12%
nd143	1%	1%	-5%	-5%
nd144	-24%	-27%	-6%	-5%
nd145	-11%	-13%	-6%	-5%
nd146	-23%	-25%	-6%	-5%
nd148	-20%	-22%	-6%	-5%
nd150	-24%	-27%	-6%	-5%
sm147	0%	0%	-5%	-4%

sm150	-19%	-21%	-7%	-6%
sm151	7%	6%	-5%	-4%
sm152	-12%	-13%	-4%	-4%
eu153	-19%	-21%	-7%	-6%
gd155	-37%	-39%	-8%	-7%
Average Deviation	**-12%**	**-14%**	**-7%**	**-6%**
STD	**14%**	**14%**	**2%**	**2%**

Table C.7. The calculated deviation from the mean of the ORIGEN-calculated fission isotope masses (in ug/g U) for 3D8E14_2655_2674, 3D8E14_2303_2322, 3D8E14_1375_1450, and 30AE14_2675_2694.

Analyte	3D8E14_2655_2674	3D8E14_2303_2322	3D8E14_1375_1450	30AE14_2675_2694
se79	-15%	-13%	-17%	-12%
sr90	-8%	-6%	-9%	-38%
ru101	-16%	-14%	-18%	-11%
rh103	-12%	-11%	-13%	-9%
i129	-19%	-16%	-21%	-11%
cs133	-13%	-12%	-14%	-11%
cs135	-16%	-14%	-18%	-17%
cs137	-13%	-10%	-14%	-37%
nd143	-7%	-7%	-7%	-11%
nd144	-20%	-17%	-22%	-12%
nd145	-13%	-11%	-14%	-11%
nd146	-19%	-16%	-21%	-12%
nd148	-17%	-14%	-18%	-11%
nd150	-19%	-16%	-20%	-11%
sm147	-6%	-6%	-6%	-5%
sm150	-17%	-15%	-19%	-12%
sm151	-5%	-4%	-5%	-16%
sm152	-11%	-9%	-11%	-7%
eu153	-17%	-15%	-18%	-11%
gd155	-26%	-24%	-28%	4%
Average Deviation	**-14%**	**-12%**	**-16%**	**-13%**
STD	**5%**	**5%**	**6%**	**9%**

Table C.8. The calculated deviation from the mean of the ORIGEN-calculated fission isotope masses (in ug/g U) for

Analyte	30AD05_2410_2439	30AD05_1280_1299	3D8E14_775_796	3A1F05_2383_2402
se79	-9%	-9%	-16%	2%
sr90	-36%	-36%	-9%	18%
ru101	-8%	-9%	-17%	2%
rh103	-7%	-7%	-13%	0%
i129	-7%	-8%	-20%	2%
cs133	-8%	-9%	-14%	1%
cs135	-14%	-14%	-18%	6%
cs137	-33%	-34%	-14%	17%
nd143	-10%	-10%	-7%	1%
nd144	-8%	-9%	-21%	3%
nd145	-9%	-9%	-14%	2%
nd146	-8%	-9%	-21%	4%
nd148	-8%	-9%	-18%	3%

nd150	-7%	-8%	-20%	3%
sm147	-4%	-4%	-6%	-1%
sm150	-8%	-9%	-18%	2%
sm151	-14%	-15%	-5%	8%
sm152	-5%	-6%	-11%	1%
eu153	-8%	-9%	-18%	1%
gd155	8%	7%	-28%	-7%
Average Deviation	**-10%**	**-11%**	**-15%**	**3%**
STD	**9%**	**9%**	**6%**	**6%**

APPENDIX D. NIST and NIST-Traceable Reference Material

Table D.1. NIST and NIST-traceable reference materials.

Reference Material Code	Element	STD Description	STD Producer
EZIP Ac-227	Actinium	Assay/Isotopic STD	Isotope Products
CETAMA 241Am/243Am STD	Americium	Assay/Isotopic STD	CETAMA
NIST SRM-4332E	Americium	Assay/Isotopic STD	NIST
HPS-Se	Antimony	Assay/Isotopic STD	High Purity Standards
NIST 3102a	Antimony	Assay/Isotopic STD	NIST
Sb-123-STD	Antimony	Assay/Isotopic STD	ORNL Isotope Program
NIST 3104a	Barium	Assay/Isotopic STD	NIST
Ce-140-STD	Cerium	Assay/Isotopic STD	ORNL Isotope Program
HPS-Ce	Cerium	Assay/Isotopic STD	High Purity Standards
NIST 3110	Cerium	Assay/Isotopic STD	NIST
Inorganic ventures Cs	Cesium	Assay STD	Inorganic Ventures
NIST 3111a	Cesium	Assay STD	NIST
ORNL-Cm-WRM (^{246}Cm)	Curium	Assay/Isotopic STD	NACIL
NIST SRM 4320b	Curium	Assay STD	NIST
Eu-151-STD	Europium	Assay/Isotopic STD	ORNL Isotope Program
NIST 3117a	Europium	Assay/Isotopic STD	NIST
Gd-155-STD	Gadolinium	Assay/Isotopic STD	ORNL Isotope Program
HPS-Gd	Gadolinium	Assay/Isotopic STD	High Purity Standards
NIST 3118a	Gadolinium	Assay/Isotopic STD	NIST
HPS-Mo	Molybdenum	Assay/Isotopic STD	High Purity Standards
Mo-94-STD	Molybdenum	Assay/Isotopic STD	ORNL Isotope Program
NIST 3134	Molybdenum	Assay/Isotopic STD	NIST
JNDI (jNNDi-1)	Neodymium	Isotopic STD	NA
Legolas (La Jolla)	Neodymium	Isotopic STD	NA
Nd-150-STD	Neodymium	Assay/Isotopic STD	ORNL Isotope Program
NIST 3135a	Neodymium	Assay/Isotopic STD	NIST
NIST4341a	Neptunium	Assay STD	NIST
IRMM-73 series	Plutonium	Assay/Isotopic STD	IRMM
NBLPO-CRM-136	Plutonium	Assay/Isotopic STD	NBL Program Office
NBLPO-CRM-137	Plutonium	Assay/Isotopic STD	NBL Program Office
NBLPO-CRM-138	Plutonium	Assay/Isotopic STD	NBL Program Office
ORNL-WRM-003	Plutonium	Assay/Isotopic STD	NACIL
NIST SRM3144	Rhodium	Assay STD	NIST
HPS-Ru	Ruthenium	Assay/Isotopic STD	High Purity Standards

Inorganic Ventures CGRU10	Ruthenium	Assay/Isotopic STD	Inorganic Ventures
Ru-99-STD	Ruthenium	Assay/Isotopic STD	ORNL Isotope Program
HPS-Sm	Samarium	Assay/Isotopic STD	High Purity Standards
NIST 3147a	Samarium	Assay/Isotopic STD	NIST
Sm-149-STD	Samarium	Assay/Isotopic STD	ORNL Isotope Program
HPS-Se	Selenium	Assay/Isotopic STD	High Purity Standards
NIST 3149	Selenium	Assay/Isotopic STD	NIST
Se-77-STD	Selenium	Assay/Isotopic STD	ORNL Isotope Program
Ag-107-STD	Silver	Assay/Isotopic STD	ORNL Isotope Program
HPS-Ag	Silver	Assay/Isotopic STD	High Purity Standards
NIST SRM3151	Silver	Assay/Isotopic STD	NIST
HPS-Sr	Strontium	Assay/Isotopic STD	High Purity Standards
NIST 3153a	Strontium	Assay/Isotopic STD	NIST
Sr-86-STD	Strontium	Assay/Isotopic STD	ORNL Isotope Program
NIST 4288B	Technetium	Assay STD	NIST
NBLPO-CRM-129	Uranium	Assay/Isotopic STD	NBL Program Office
NBLPO-CRM-010	Uranium	Isotopic STD	NBL Program Office
NBLPO-CRM-112a	Uranium	Assay/Isotopic STD	NBL Program Office
NBLPO-CRM-117	Uranium	Assay/Isotopic STD	NBL Program Office
ONRL-WEWA	Uranium	Assay/Isotopic STD	NACIL