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ANALYSIS OF SPENT NUCLEAR FUEL SAMPLES FROM THREE MILE ISLAND AND QUAD CITIES REACTORS: FINAL REPORT

by

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I. OBJECTIVES

This report summarizes results from a two-phase project whose objective was to determine the contents of 30 nuclides normalized to ^{238}U in 11 spent fuel samples from Three Mile Island reactor 1 (TMI-1) and 12 samples from Quad Cities reactor 1 (QC-1). The samples were prepared from known locations in fuel rod segments provided by General Electric Company, Vallecitos Nuclear Center (GE-VNC). The complete analysis of these samples includes the determination of the ^{238}U-normalized contents of the following nuclides: ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{137}Cs, ^{143}Nd, ^{145}Nd, ^{148}Nd, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{151}Eu, ^{153}Eu, ^{155}Eu, ^{155}Gd, ^{234}U, ^{235}U, ^{236}U, ^{237}Np ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, and ^{243}Am. This data provides the information necessary for the calculation of atom percent fission (fuel burnup) based on ^{148}Nd, Pu, and U content [1]. This data set may also serve as a basis for validation of isotopic models used for assessing repository criticality scenarios.

II. BACKGROUND AND APPROACH

An isotopic model is being used to calculate concentrations of fissionable and neutron-absorbing nuclides for post-closure criticality evaluation of the Yucca Mountain Repository. The model used for calculating initial isotopic concentrations in commercial spent nuclear fuel is the SAS2H sequence of the Scale-4.3 computer code with the 44-energy group cross-section library [2]. The validation of the isotopic model will be performed to support the license application to the Nuclear Regulatory Commission of the proposed repository. Radiochemical assay of spent fuel samples will be used, in part, to validate the isotopic model. The burnups of fuels used to validate the isotopic model to date range from 7 to 46.5 gigawatt-days per metric ton uranium (GWd/MTU). However, additional radiochemical assays of higher burnup spent nuclear fuels from both pressurized water and boiling water reactors are required to validate the isotopic model at higher burnup levels [2].

To provide the information required for this model validation, we have analyzed 23 segments from fuel rods with maximum burnups >45 GWd/MTU originating from TMI-1 and QC-1 reactors. The 12 samples analyzed for Phase 1 and the 11 samples analyzed for Phase 2 of this work are listed in Tables 1 and 2, respectively. The fuel sample segments were cut from fuel rods segments provided by GE-VNC. Tables 1 and 2 list the identification number designations for the GE-VNC fuel rod and fuel rod segment, the Argonne National Laboratory (ANL) Alpha Gamma Hot Cell Facility (AGHCF) fuel rod segment designation, the ANL Chemical Technology Division (CMT) sample designation, and the length and location in the fuel rod segment from which the individual samples were excised. In this report, samples will be identified by their ANL CMT designation so that samples prepared from different areas of the same fuel rod segment will have unique sample designations.

The 23 samples were cut from their respective fuel rod segments in the ANL AGHCF and shipped to the Senior Cave Facility located in the CMT Division, Building 205, at ANL for preparation and analysis. Preparation consisted of unloading the cut samples from the storage cask, identifying the fuel sample, and separating the fuel meat from the cladding. The fuel meat was separated from the cladding by use of a Plattner's diamond mortar modified with a 1-m long sleeve and a 2-kg tool steel pestle. In each case, the fuel fragments were recovered from the mortar and further crushed and homogenized with a Wig-L-Bug® shaker in hardened tool steel vials and ball pestle. A new vial and ball pestle was used on each sample to minimize cross-contamination. The powdered fuel was stored in glass vials and reserved for analysis.

An approximately 0.1- to 0.2-g aliquot of the homogenized fuel sample powder was taken for dissolution and analysis. The powdered samples were dissolved in Parr Bomb vessels with Teflon® liners. The fuel was added to a weighed Teflon® liner. A new liner was used for each sample. The liner was reweighed, and a mixture of HNO_3-HCl-HF was added to the liner. The Parr bomb was assembled and placed in a 150°C oven for 20 h for the TMI samples and 72 h for the QC samples. After heating for the required time, the vessel was removed from the oven and allowed to cool for 2 h. The vessel was opened, and the solution was examined. In all cases, the dissolution produced a yellow and translucent solution, indicating that the dissolution was complete. The solution was quantitatively transferred to a low-density polyethylene wash bottle that had been weighed. The Teflon® liner was triply rinsed with 2 M

HNO_3, and the rinse solutions were combined with the initial dissolution solution. The wash bottle was reweighed, and the total mass of solution was determined by a difference calculation. The final dissolved fuel solution weighed approximately 50 g.

A working solution for analysis was prepared by diluting approximately 1 g of the dissolved fuel solution to 100 mL. Solution analyses were performed with inductively coupled plasma-mass spectrometry (ICPMS), γ-spectrometry (γ-S), and α-spectrometry (α-S). Table 3 lists the nuclides determined along with the analytical and calibration method used. Uranium-238 was determined for a solution diluted 100-fold from the initial working solution. This solution was analyzed by two independent calibration methods: (1) internal standardization with ^{209}Bi and external calibration via linear regression using a ^{238}U standard (SPEX Industries) and (2) isotope dilution with a ^{233}U spike (New Brunswick Laboratory CRM 111A). The concentrations of ^{241}Am, ^{137}Cs, and ^{155}Eu were determined by direct γ-spectrometric analysis of a 5-mL aliquot of the 100-fold diluted sample.

Fission product nuclides free of isobaric interferences were determined directly in the working solution with ICPMS. Aliquots of the working solution were spiked with ^{115}In and ^{209}Bi internal standards and quantitation was performed via external calibration with multi-element standards (SPEX Industries) with known natural abundance isotopic content followed by linear regression. Ruthenium-99 was used as a surrogate calibration element for ^{99}Tc in the standard solutions. Several fission product nuclides could not be determined directly with ICPMS due to significant isobaric interferences. As a result, analysis of these nuclides required chemical separation of interfering species. These nuclides are identified in Table 3. Chemical separations of Nd, Pm, Sm, Eu, and Gd were performed using high-performance liquid chromatography (HPLC) with ICPMS functioning as the detector (see Table 3). The relative isotopic abundance of individual isotopes of each element was measured by isotope ratio (IR) determination with ICPMS. The absolute concentration of each isotope could then be calculated relative to the absolute concentration of an isobar-free nuclide, as determined from direct ICPMS analysis. Samarium-152 was the benchmark nuclide for Sm isotopes. Europium-153 was the benchmark nuclide for Eu isotopes. Gadolinium-156 was the benchmark for Gd isotopes. Neodymium isotopes were determined by isotope dilution (ID) with a ^{150}Nd isotopic spike (Oak Ridge National Laboratory) for definitive results.

Minor uranium isotopes, ^{237}Np, ^{239}Pu, ^{240}Pu, and ^{243}Am were determined by direct ICPMS analysis of the working solution using the ID method with a ^{233}U isotopic spike. Other actinides were determined in chemically separated aliquots of the working solution. Actinide elements were chemically separated by ion exchange chromatography, providing Pu fractions and Am fractions for analyses of individual isotopes of each element. All plutonium isotopes except ^{238}Pu were determined by the ID method with a ^{244}Pu isotopic spike (National Bureau of Standards SRM 996), including a reanalysis of ^{239}Pu and ^{240}Pu. Plutonium-239 and -240 were determined by two independent methods. Plutonium-238 was determined by α-spectrometry using the mean ^{239}Pu/^{240}Pu mass ratio that had been determined by direct analysis of the dissolved fuel with ICPMS in the chemically separated Pu fraction. This ratio was converted to total ^{239}Pu+^{240}Pu activity, and the ^{239}Pu+^{240}Pu composite peak was used as a benchmark for the determination of ^{238}Pu by α-pulse height analysis. The isotopic composition of Am was determined from the isotopic ratio measured by ICPMS of the chemically separated Am fraction. These ratios were corrected for the presence of small amounts of ^{242}Pu in the Am fraction due to incomplete radiochemical separation of the two elements. The concentration of ^{242m}Am was calculated using ^{241}Am, a nuclide determined with γ-spectrometry, as a benchmark.

III. RESULTS AND DISCUSSION

A. Nuclide Content and Fuel Burnup

Results of analyses performed at ANL and calculated fuel burnups for the 11 TMI samples from Phase 1 and Phase 2 are given in Tables 4 and 5, respectively. Results of analyses performed at ANL and calculated fuel burnups for the 12 QC samples from Phase 1 and Phase 2 are given in Tables 6 and 7, respectively. Phase 1 TMI samples were analyzed in September 1998. Phase 1 QC samples were analyzed in December 1998. All Phase 2 samples were analyzed in May 2000.

B. Uncertainty Analysis

Two measures of analytical uncertainty are given in Table 8: the estimated precision and the estimated bias uncertainty of analysis. The precision of analyses was calculated for each nuclide and is expressed in terms of 1 times the sample standard deviation percent relative to concentration of the nuclide in the fuel. All analyses were performed in duplicate or triplicate to establish the within-sample precision of the methodologies employed. Each replicate analysis was performed on aliquots from the same initial dissolution solution in non-consecutive analysis procedures and, in most cases, on different days. This was done so that the estimated precision took into account the procedure-to-procedure and day-to-day variability of analysis. The ^{238}U, ^{239}Pu, and ^{240}Pu contents were determined using at least two independent calibration methods and were calculated from at least three replicate analyses.

The estimated bias uncertainty of analysis was estimated from deviations of Quality Control standard solutions that were analyzed before and after fuel samples. The reported values are the root of the mean sum of squares of these deviations, includes the propagation of error for normalization to ^{238}U content of each sample, and is expressed in terms of percent relative to the ^{238}U normalized concentration of the nuclide in the fuel. Both the estimated precision and the bias uncertainty values reflect the uncertainties in the analytical measurement techniques employed in this work.

C. Evaluation of GE-VNC Data

One of the authors of this report (SFW) has been asked to provide a technical evaluation of GE-VNC data as a basis to support data qualification. The technical evaluation is based on several actions: examination of two documents summarizing the methodologies employed in this work [3,4], visited the personnel at the facilities where work was performed, and compared results of analyses of samples possessing near-identical irradiation histories [3,4]. The available information supports data qualification.

The available technical documentation and laboratory visits both indicate that, while the analytical procedures utilized by GE-VNC differ slightly from those employed by ANL, the procedures are fundamentally identical in most cases. In the two cases where different techniques were used, the alternative approach was technically sound.

General Electric-VNC employed an open-vessel digestion procedure to dissolve the samples. The mineral acids and temperatures employed would be expected to dissolve all fuel matrix elements including actinides, lanthanides, and soluble fission products such as Cs. The only elements that this technique would not be expected to dissolve are those present in ε-particles such as Tc, Ru, Rh, Pd, and Mo. These elements were not included in GE-VNC results so that the dissolution would be adequate for their purposes.

General Electric-VNC used the high-precision technique of isotope dilution thermal ionization mass spectrometry for the determination of most U, Pu, Am, Cm, Nd, Sm, Eu, and Gd isotopes. Thermal ionization mass spectrometry has traditionally been the technique for determining isotopes of these elements in dissolved fuel [1] and provided the necessary chemical separations from interfering isobars are achieved, this methodology should produce quality data. Argonne employed ICPMS as the method for mass spectrometric determination.

Two nuclides were determined by essentially identical methods in both laboratories. Plutonium-238 activity determination was performed by α-spectrometry by benchmarking to the $^{239}Pu+^{240}Pu$ α-activity in a Pu-separated sample. The ^{238}Pu concentration was then calculated based on the ^{239}Pu and ^{240}Pu concentrations as determined by ICPMS. Cesium-137 was determined by γ-spectrometry.

Two nuclides were determined by fundamentally different methods in both laboratories. General Electric-VNC determined ^{241}Am by α-spectrometry after chemical separation. This method differs from ANL's direct determination by gamma spectrometry. General Electric-VNC determined ^{237}Np by α-spectrometry after chemical separation with yield determination by γ-spectrometric determination of a ^{239}Np tracer. This method differs from ANL's direct determination by ICPMS. While the methods ^{241}Am and ^{237}Np differ from those employed by ANL, the procedures are fundamentally sound and should produce quality data. In both cases the isotopic spikes, standards, and analytical instrumentation employed by GE-VNC are all technically appropriate for the analyses that were performed.

IV. CONCLUSION

Our analyses of fissionable and neutron-absorbing nuclides in high-burnup spent nuclear fuels will allow testing of isotopic models for predicting criticality scenarios in a nuclear waste repository. We have generated a database of concentrations for 30 nuclides in samples of high-burnup fuels (>45 GWd/MTU) from the Three Mile Island and Quad Cities Reactors. These measurements were obtained with ICPMS, as well as α- and γ-spectrometry. For those nuclides with significant isobaric interferences in the ICPMS analysis, chemical separation of the interfering species was performed by either ion-exchange or HPLC followed by detection by ICPMS.

V. REFERENCES

[1] "Standard Test Method for Atom Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method)," ASTM E 321-79, American Society for Testing and Materials, Philadelphia, PA, (1995).

[2] "Disposal Criticality Analysis Methodology Topical Report," YMP-TR-004Q.

[3] TRW Yucca Mountain Project Test Report Phase 1," TRW Purchase Order No. AO9112CC8A

[4] TRW Yucca Mountain Project Test Report Phase 2," TRW Purchase Order No. AO9112CC8A

[5] "Standard Practice for Conducting an Interlaboratory Test Program to Determine the Precision of Test Methods." ASTM E691-79, American Society for Testing and Materials, Philadelphia, PA, (1979).

Table 1. Spent nuclear fuel samples analyzed in Phase 1 of this work

Fuel Rod Number	GE-VNC Sample ID	AGHCF Sample ID	Location, in.	Sample Length, in.	ANLCMT Sample ID
TMI/I05243	TMI-1 H6-1	AG536A	29.4[a]	0.56	TMI A2
TMI/I05243	TMI-1 H6-2	AG536B	45.3[a]	0.41	TMI B2
TMI/I05243	TMI-1 H6-3	AG536C	92.7[a]	0.50	TMI C1
TMI/I05243	TMI-1 H6-3	AG536C	61.5[a]	0.50	TMI C3
TMI/I05243	TMI-1 H6-4	AG536D	126.8[a]	0.50	TMI D2
VW00101	QC-1 C7-2	AG539A	57.2[b]	0.44	QC A2
VW00101	QC-1 C7-2	AG539B	93.3[b]	0.41	QC B1A
VW00101	QC-1 C7-2	AG539C	115.2[b]	0.44	QC C2
ZS00102	QC-1 G5-3A	AG539D	85.2[b]	0.41	QC D4A
ZS00102	QC-1 G5-4	AG539E	113.8[b]	0.44	QC E2
ZB00113	QC-1 B1-4	AG539F	95.1[b]	1.09	QC F1
ZB00113	QC-1 B1-4	AG539F	128.0[b]	0.41	QC F8

[a] Length measured from the bottom of the end plug.

Table 2. Spent nuclear fuel samples analyzed in Phase 2 of this work

Fuel Rod Number	GE-VNC Fuel Rod ID	AGHCF Fuel Rod ID	Location, in.	Sample Length, in.	ANLCMT Sample ID
TMI/I05243	TMI-1 H6-1	AG536A	15.25	0.50	TMI A1B
TMI/I05243	TMI-1 H6-2	AG536B	61.40	0.93	TMI B1B
TMI/I05243	TMI-1 H6-2	AG536B	30.32	0.63	TMI B3J
TMI/I05243	TMI-1 H6-3	AG536C	76.62	0.50	TMI C2B
TMI/I05243	TMI-1 H6-4	AG536D	103.11	0.50	TMI D1A2
TMI/I05243	TMI-1 H6-4	AG536D	115.11	0.50	TMI D1A4
VW00101	QC-1 C7-2	AG539A	31.25	0.50	QC A3A
VW00101	QC-1 C7-3	AG539B	84.12	0.50	QC B4B2
VW00101	QC-1 C7-4	AG539C	128.07	0.50	QC C1B
ZS00102	QC-1 G5-3A	AG539D	76.94	0.50	QC D4B1
ZS00102	QC-1 G5-3A	AG539E	62.44	0.50	QC D6C

LEP: Length measured from the bottom endplug.

Table 3. Analytical techniques and calibration methods for nuclides determined in this work.

Nuclide	Technique(s)	Calibration Method(s)
^{95}Mo	ICPMS	LR
^{99}Tc	ICPMS	LR
^{101}Ru	ICPMS	LR
^{103}Rh	ICPMS	LR
^{109}Ag	ICPMS	LR
^{137}Cs	γ-s	ES
^{143}Nd	ICPMS	LR, ID
^{145}Nd	ICPMS	LR, ID
^{148}Nd	HPLC-ICPMS	ID
^{147}Sm	HPLC-ICPMS	RD
^{149}Sm	ICPMS	LR
^{150}Sm	ICPMS	RD
^{151}Sm	HPLC-ICPMS	RD
^{152}Sm	ICPMS	LR
^{151}Eu	ICPMS	RD
^{153}Eu	ICPMS	LR
^{155}Eu	γ-s	ES
^{155}Gd	ICPMS	RD
^{234}U	ICPMS	ID
^{235}U	ICPMS	ID
^{236}U	ICPMS	ID
^{238}U	ICPMS	ID, LR
^{237}Np	ICPMS	ID
^{238}Pu	α-s	RD, ES
^{239}Pu	ICPMS	ID
^{240}Pu	ICPMS	ID
^{241}Pu	ICPMS	ID
^{242}Pu	ICPMS	ID
^{241}Am	γ-s	ES
^{242m}Am	ICPMS	RD
^{243}Am	ICPMS	ID

LR: linear regression calibration.
ID: isotope dilution analysis.
ES: external calibration of instrument response.
RD: determination of isotopic abundance on chemically separated sample with calculation of isotopic concentration

Table 4. Results of analyses of five samples of TMI spent nuclear fuel rod TMI/I05243 analyzed for Phase 1 of this work. Results are expressed as g/g ^{238}U by nuclide.

Nuclide	TMI A2	TMI B1A	TMI C1	TMI C3	TMI D2
^{95}Mo	1.21E-03	1.22E-03	1.19E-03	1.09E-03	9.90E-04
^{99}Tc	1.17E-03	1.18E-03	1.17E-03	1.12E-03	1.05E-03
^{101}Ru	1.25E-03	1.30E-03	1.26E-03	1.11E-03	1.02E-03
^{103}Rh	6.70E-04	6.80E-04	6.69E-04	5.93E-04	5.55E-04
^{109}Ag	6.46E-05	5.71E-05	5.80E-05	1.00E-04	5.01E-05
^{137}Cs	1.91E-03	1.89E-03	1.96E-03	1.84E-03	1.74E-03
^{143}Nd	1.03E-03	1.08E-03	1.06E-03	1.03E-03	9.83E-04
^{145}Nd	9.50E-04	9.80E-04	9.71E-04	9.71E-04	8.92E-04
^{148}Nd	5.96E-04	5.89E-04	5.90E-04	6.04E-04	5.24E-04
^{147}Sm	2.13E-04	2.01E-04	2.02E-04	1.97E-04	1.96E-04
^{149}Sm	4.13E-06	3.53E-06	3.45E-06	3.14E-06	3.33E-06
^{150}Sm	4.05E-04	4.06E-04	4.15E-04	3.92E-04	3.75E-04
^{151}Sm	1.36E-05	1.45E-05	1.35E-05	1.36E-05	1.36E-05
^{152}Sm	1.43E-04	1.40E-04	1.37E-04	1.36E-04	1.30E-04
^{151}Eu	9.56E-07	8.58E-07	7.42E-07	9.18E-07	7.57E-07
^{153}Eu	1.85E-04	1.81E-04	1.81E-04	1.74E-04	1.68E-04
^{155}Eu	1.39E-05	1.42E-05	1.55E-05	1.38E-05	1.32E-05
^{155}Gd	5.65E-06	7.08E-06	6.88E-06	7.22E-06	6.02E-06
^{234}U	2.07E-04	2.02E-04	2.14E-04	2.00E-04	2.07E-04
^{235}U	6.84E-03	6.71E-03	7.13E-03	6.77E-03	7.94E-03
^{236}U	5.95E-03	5.84E-03	5.92E-03	5.77E-03	5.74E-03
^{237}Np	7.51E-04	7.48E-04	7.62E-04	7.39E-04	7.27E-04
^{238}Pu	3.83E-04	3.40E-04	3.57E-04	2.72E-04	3.50E-04
^{239}Pu	5.78E-03	5.72E-03	5.85E-03	5.97E-03	5.84E-03
^{240}Pu	3.01E-03	2.95E-03	2.98E-03	3.08E-03	2.87E-03
^{241}Pu	1.47E-03	1.50E-03	1.54E-03	1.52E-03	1.47E-03
^{242}Pu	9.99E-04	9.89E-04	9.74E-04	1.00E-03	8.55E-04
^{241}Am	3.27E-04	3.69E-04	4.08E-04	3.28E-04	3.72E-04
^{242m}Am	<1E-05	<1E-05	<1E-05	<1E-05	<1E-05
^{243}Am	2.75E-04	2.76E-04	2.66E-04	2.67E-04	2.07E-04
Burnup*	50.6	50.1	50.2	51.3	44.8

* Burnup calculation is based on ^{148}Nd and expressed in units of GWd/MTU.

Table 5. Results of analyses of six samples of TMI spent nuclear fuel rod TMI/I05243 analyzed for Phase 2 of this work. Results are expressed as g/g ^{238}U by nuclide.

Nuclide	TMI A1B	TMI B1B	TMI B3J	TMI C2B	TMI D1A2	TMI D1A4
^{95}Mo	1.12E-03	1.25E-03	1.22E-03	1.19E-03	1.21E-03	1.18E-03
^{99}Tc	1.53E-03	1.43E-03	1.35E-03	1.47E-03	1.24E-03	1.29E-03
^{101}Ru	1.20E-03	1.29E-03	1.27E-03	1.27E-03	1.23E-03	1.19E-03
^{103}Rh	6.41E-04	6.81E-04	6.73E-04	6.66E-04	6.72E-04	6.53E-04
^{109}Ag	5.50E-05	4.78E-05	8.45E-05	7.08E-05	5.02E-05	9.17E-05
^{137}Cs	1.81E-03	1.91E-03	1.88E-03	1.91E-03	1.67E-03	1.79E-03
^{143}Nd	1.06E-03	1.18E-03	1.15E-03	1.12E-03	1.21E-03	1.17E-03
^{145}Nd	9.17E-04	1.07E-03	1.06E-03	1.02E-03	1.09E-03	1.04E-03
^{148}Nd	5.24E-04	6.44E-04	6.25E-04	6.20E-04	6.60E-04	5.94E-04
^{147}Sm	2.43E-04	2.77E-04	2.69E-04	2.48E-04	2.74E-04	2.55E-04
^{149}Sm	3.35E-06	3.72E-06	3.46E-06	3.64E-06	4.20E-06	3.90E-06
^{150}Sm	3.85E-04	5.08E-04	4.91E-04	4.54E-04	4.93E-04	4.47E-04
^{151}Sm	1.39E-05	1.63E-05	1.60E-05	1.44E-05	1.69E-05	1.53E-05
^{152}Sm	1.31E-04	1.56E-04	1.54E-04	1.41E-04	1.55E-04	1.45E-04
^{151}Eu	7.08E-07	6.19E-07	8.11E-07	7.62E-07	7.21E-07	7.23E-07
^{153}Eu	1.58E-04	2.02E-04	1.99E-04	1.87E-04	2.06E-04	1.89E-04
^{155}Eu	1.08E-05	1.68E-05	1.12E-05	1.08E-05	1.07E-05	1.37E-05
^{155}Gd	8.85E-06	1.09E-05	1.13E-05	1.02E-05	1.11E-05	1.51E-05
^{234}U	2.21E-04	2.04E-04	1.99E-04	1.96E-04	2.10E-04	2.14E-04
^{235}U	9.26E-03	6.94E-03	6.63E-03	6.75E-03	7.59E-03	8.11E-03
^{236}U	5.50E-03	5.87E-03	5.92E-03	5.62E-03	5.94E-03	5.81E-03
^{237}Np	6.50E-04	7.62E-04	7.66E-04	7.44E-04	7.69E-04	7.42E-04
^{238}Pu	4.34E-04	4.69E-04	4.32E-04	4.97E-04	4.15E-04	4.06E-04
^{239}Pu	5.45E-03	5.55E-03	5.52E-03	5.41E-03	5.94E-03	5.85E-03
^{240}Pu	2.52E-03	2.86E-03	2.88E-03	2.76E-03	2.95E-03	2.84E-03
^{241}Pu	1.30E-03	1.48E-03	1.48E-03	1.44E-03	1.60E-03	1.55E-03
^{242}Pu	7.31E-04	1.04E-03	1.20E-03	1.01E-03	1.05E-03	1.02E-03
^{241}Am	3.73E-04	3.13E-04	5.49E-04	5.50E-04	3.65E-04	5.70E-04
^{242m}Am	<1E-05	1.12E-06	1.35E-06	1.82E-06	6.63E-07	9.09E-07
^{243}Am	1.34E-04	2.22E-04	2.29E-04	2.12E-04	2.24E-04	2.00E-04
Burnup*	44.8	54.5	53.0	52.6	55.7	50.5

* Burnup calculation is based on ^{148}Nd and expressed in units of GWd/MTU.

Table 6. Results of analyses of seven samples of QC spent nuclear fuel rods VW00101, ZB00113, and ZS00102 analyzed for Phase 1 of this work. Results are expressed as g/g ^{238}U by nuclide.

Nuclide	QC A2	QC B1A	QC C2	QC D4A	QC E2	QC F1	QC F8
^{95}Mo	1.32E-03	1.41E-03	1.28E-03	1.33E-03	1.19E-03	1.55E-03	1.27E-03
^{99}Tc	1.52E-03	1.53E-03	1.48E-03	1.42E-03	1.38E-03	1.62E-03	1.47E-03
^{101}Ru	1.54E-03	1.65E-03	1.50E-03	1.49E-03	1.36E-03	1.72E-03	1.39E-03
^{103}Rh	6.18E-04	6.84E-04	6.38E-04	6.86E-04	6.28E-04	7.49E-04	6.77E-04
^{109}Ag	4.68E-05	1.02E-04	8.18E-05	5.96E-05	8.09E-05	1.38E-04	5.23E-05
^{137}Cs	2.40E-03	2.04E-03	2.44E-03	1.84E-03	2.07E-03	2.13E-03	2.19E-03
^{143}Nd	6.42E-04	7.24E-04	7.44E-04	8.67E-04	9.34E-04	8.98E-04	1.09E-03
^{145}Nd	1.18E-03	1.22E-03	1.13E-03	1.13E-03	1.08E-03	1.24E-03	1.18E-03
^{148}Nd	8.38E-04	8.56E-04	8.14E-04	7.65E-04	7.05E-04	8.49E-04	7.36E-04
^{147}Sm	3.15E-04	3.24E-04	2.92E-04	3.13E-04	2.94E-04	3.64E-04	3.42E-04
^{149}Sm	1.33E-06	1.61E-06	1.84E-06	1.79E-06	2.36E-06	1.48E-06	2.26E-06
^{150}Sm	4.25E-04	4.77E-04	4.08E-04	4.40E-04	3.98E-04	5.29E-04	4.09E-04
^{151}Sm	8.06E-06	8.94E-06	8.28E-06	1.15E-05	1.18E-05	1.07E-05	1.05E-05
^{152}Sm	1.82E-04	1.95E-04	1.67E-04	1.59E-04	1.39E-04	1.96E-04	1.59E-04
^{151}Eu	7.73E-07	7.44E-07	8.52E-07	1.02E-06	1.16E-06	8.33E-07	1.15E-06
^{153}Eu	2.14E-04	2.35E-04	2.12E-04	2.26E-04	2.61E-04	2.45E-04	2.03E-04
^{155}Eu	1.06E-05	1.02E-05	1.11E-05	1.08E-05	1.08E-05	8.63E-06	9.42E-06
^{155}Gd	1.24E-05	1.41E-05	1.57E-05	1.35E-05	1.49E-05	1.60E-05	1.40E-05
^{234}U	1.11E-04	1.10E-04	1.19E-04	1.44E-04	1.52E-04	1.56E-04	1.86E-04
^{235}U	2.99E-04	3.46E-04	5.71E-04	1.00E-03	1.55E-03	1.20E-03	3.31E-03
^{236}U	4.61E-03	4.43E-03	4.58E-03	3.86E-03	4.69E-03	4.54E-03	6.08E-03
^{237}Np	5.98E-04	6.45E-04	6.58E-04	7.16E-04	7.20E-04	8.32E-04	6.72E-04
^{238}Pu	4.32E-04	5.78E-04	4.72E-04	5.26E-04	5.27E-04	6.11E-04	5.54E-04
^{239}Pu	3.19E-03	3.75E-03	3.78E-03	5.65E-03	5.72E-03	4.01E-03	4.73E-03
^{240}Pu	3.15E-03	3.59E-03	3.34E-03	3.61E-03	3.31E-03	3.44E-03	3.38E-03
^{241}Pu	7.33E-04	9.83E-04	8.71E-04	1.32E-03	1.17E-03	1.01E-03	1.09E-03
^{242}Pu	2.16E-03	2.36E-03	2.05E-03	1.84E-03	1.52E-03	1.82E-03	1.44E-03
^{241}Am	4.44E-04	3.77E-04	5.27E-04	5.87E-04	7.82E-04	6.43E-04	6.30E-04
^{242m}Am	<1E-05	<1E-05	<1E-05	<1E-05	<1E-05	2.76E-06	<1E-05
^{243}Am	6.65E-04	5.98E-04	6.16E-04	5.33E-04	5.72E-04	7.43E-04	4.18E-04
Burnup*	71.2	72.6	69.6	62.0	61.1	77.6	63.6

* Burnup calculation is based on ^{148}Nd and expressed in units of GWd/MTU.

Table 7. Results of analyses of five samples of QC spent nuclear fuel rods VW00101 and ZS00102 analyzed for Phase 2 of this work. Results are expressed as g/g ^{238}U by nuclide.

Nuclide	QC A3A	QCB4B2	QC C1B	QC D4B1	QC D6C
^{95}Mo	1.36E-03	1.48E-03	1.31E-03	1.29E-03	1.31E-03
^{99}Tc	1.54E-03	1.83E-03	2.13E-03	1.59E-03	1.60E-03
^{101}Ru	1.56E-03	1.78E-03	1.60E-03	1.49E-03	1.51E-03
^{103}Rh	6.24E-04	6.88E-04	6.76E-04	6.31E-04	6.39E-04
^{109}Ag	2.97E-05	5.47E-05	7.10E-05	6.36E-05	6.00E-05
^{137}Cs	1.73E-03	2.01E-03	2.17E-03	1.74E-03	1.75E-03
^{143}Nd	6.08E-04	6.66E-04	7.48E-04	7.85E-04	7.76E-04
^{145}Nd	1.07E-03	1.14E-03	9.78E-04	1.05E-03	1.06E-03
^{148}Nd	7.54E-04	8.17E-04	6.74E-04	7.33E-04	7.35E-04
^{147}Sm	3.11E-04	3.18E-04	2.85E-04	2.88E-04	3.00E-04
^{149}Sm	8.65E-07	1.41E-06	1.36E-06	1.46E-06	1.29E-06
^{150}Sm	4.04E-04	4.78E-04	3.85E-04	4.33E-04	4.37E-04
^{151}Sm	7.29E-06	8.62E-06	8.27E-06	1.08E-05	1.05E-05
^{152}Sm	1.84E-04	1.92E-04	1.47E-04	1.47E-04	1.54E-04
^{151}Eu	5.02E-07	7.52E-07	6.42E-07	8.33E-07	9.51E-07
^{153}Eu	2.05E-04	2.26E-04	1.83E-04	2.08E-04	2.10E-04
^{155}Eu	8.47E-06	8.88E-06	1.30E-05	1.28E-05	8.97E-06
^{155}Gd	1.26E-05	1.62E-05	1.29E-05	1.52E-05	2.72E-05
^{234}U	1.21E-04	1.18E-04	1.30E-04	1.48E-04	1.52E-04
^{235}U	3.50E-04	3.33E-04	1.09E-03	8.82E-04	8.93E-04
^{236}U	4.33E-03	4.21E-03	4.29E-03	4.26E-03	4.36E-03
^{237}Np	5.23E-04	5.94E-04	6.04E-04	6.43E-04	6.37E-04
^{238}Pu	3.50E-04	6.11E-04	6.07E-04	5.98E-04	5.77E-04
^{239}Pu	2.83E-03	3.47E-03	3.92E-03	4.38E-03	4.28E-03
^{240}Pu	2.72E-03	3.27E-03	3.21E-03	2.83E-03	2.85E-03
^{241}Pu	5.73E-04	8.20E-04	8.96E-04	8.28E-04	8.09E-04
^{242}Pu	1.71E-03	2.15E-03	1.55E-03	1.42E-03	1.45E-03
^{241}Am	4.63E-04	5.69E-04	5.77E-04	6.63E-04	7.00E-04
^{242m}Am	9.62E-07	3.48E-06	6.47E-06	3.81E-06	3.56E-06
^{243}Am	3.88E-04	4.86E-04	3.28E-04	4.00E-04	3.95E-04
Burnup*	63.9	68.7	57.4	62.1	62.3

* Burnup calculation is based on ^{148}Nd and expressed in units of GWd/MTU.

Table 8. The estimated precision and estimated bias uncertainty of analysis for each nuclide.

Nuclide	Precision* (1s rel. %)	Bias uncertainty** (rel. %)
^{95}Mo	1.7	3.4
^{99}Tc	2.7	7.3
^{101}Ru	1.6	5.3
^{103}Rh	1.5	3.1
^{109}Ag	4.7	3.1
^{137}Cs	3.6	2.7
^{143}Nd	3.5	3.9
^{145}Nd	4.8	3.5
^{148}Nd	4.2	5.5
^{147}Sm	3.3	9.4
^{149}Sm	7.1	3.5
^{150}Sm	3.5	3.2
^{151}Sm	6.1	3.2
^{152}Sm	2.7	3.2
^{151}Eu	12	2.9
^{153}Eu	3.9	3.0
^{155}Eu	6.4	2.7
^{155}Gd	6.8	3.8
^{234}U	3.0	2.7
^{235}U	1.5	2.9
^{236}U	4.6	3.1
^{238}U	1.7	3.8
^{237}Np	4.1	3.4
^{238}Pu	6.8	3.6
^{239}Pu	4.3	3.3
^{240}Pu	5.1	3.1
^{241}Pu	3.2	2.9
^{242}Pu	5.9	2.8
^{241}Am	6.1	3.1
^{242m}Am	-	3.1
^{243}Am	4.2	3.8

*ANL within-sample standard deviation (rel. %)
** Calculated as the square root of the average of the squares of the deviations from multiple QC samples analyzed prior to and subsequent to fuel samples. The calculated values are for ^{238}U normalized concentrations.