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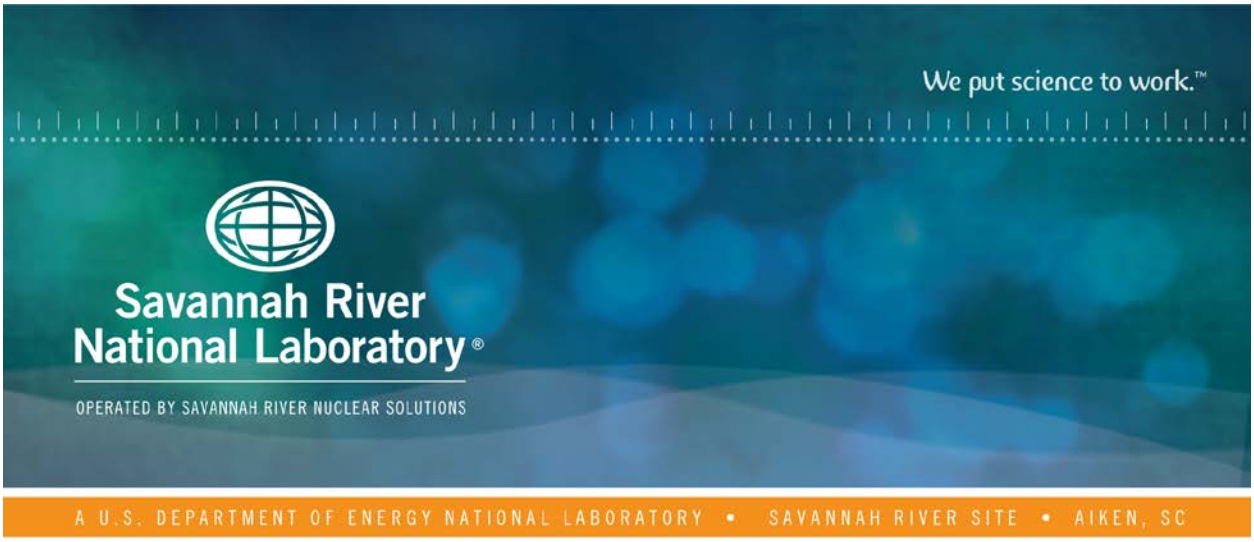

Analysis of H-Canyon 6.4D Dissolver Solids

T. E. Smith

T. S. Rudisill

D. H. Jones

T. B. Edwards

January 2020

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REVIEWS AND APPROVALS

AUTHORS:

_____
T. E. Smith, Separations & Actinide Science Programs Date

_____
T. S. Rudisill, Separations & Actinide Science Programs Date

_____
D. H. Jones, Environmental Stewardship Directorate Operations Date

_____
T. B. Edwards, Immobilization Technologies Date

TECHNICAL REVIEW:

_____
P. M. Almond, Nuclear Material Systems Date

_____
W. H. Clifton, Jr., H-Canyon Outside Facilities & Technical Support Date

APPROVAL:

_____
C. C. DiPrete, Group Leader Date
Separations & Actinide Science Programs

_____
T. L. Tice, Manager Date
Materials Disposition Engineering

_____
S. D. Fink, Director Date
Chemical Processing Technologies

EXECUTIVE SUMMARY

During reconfiguration of the 6.4D dissolver for the dissolution of High Flux Isotope Reactor (HFIR) fuel, solid material was observed on the bottom of the dissolver pot. Samples of the solids were taken from the north and south side of the 6.4D pot and were transported to the Savannah River National Laboratory (SRNL) for characterization. The samples were initially dried, and then a small portion of each sample set was removed, and rinsed with either 1 M HNO_3 or 6 M HNO_3. A portion of each unrinsed and rinsed sample was dissolved and analyzed for elemental and radiochemical concentrations. Results from the analyses indicated that the solids were primarily Si, Fe, Al, and Cr. The U content in the initial dried, unrinsed solids was 0.380 wt %, but was reduced to 0.057 wt % in the rinsed solids, indicating the majority of the U was mostly soluble in HNO_3. Small amounts of Pu were measured at 0.010 wt % in both the rinsed and unrinsed solids.

Undissolved solids (e.g., noble metal fission products and SiO_2 from U silicide (U_3Si or U_3Si_2) research reactor fuels) are a product of Spent Nuclear Fuel (SNF) dissolution and are regularly transferred from the dissolver and removed from the dissolving solution by centrifugation in Head End and transferred to waste. Based on the estimated volume of solids present in the 6.4D dissolver and the measured density, the bounding total masses of U and Pu present were 359 ± 45 g (126 ± 20 g U-235) and 8.6 ± 1.4 g, respectively, at 95% confidence limits for the unrinsed solids. The total masses of U and Pu present in the rinsed solids were 54 ± 6 g (12 ± 2 g of U-235) and 9.4 ± 1.1 g, respectively, at 95% confidence limits.

The calculated masses of U and Pu in the 6.4D dissolver solids were substantially reduced when more aggressive dissolving conditions were used. Contact with near boiling 7 M HNO_3 reduced the U and Pu masses to 39.4 ± 5.5 g (7.24 ± 1.17 g U-235) and 7.89 ± 1.13 g, respectively, at 95% confidence limits. When 0.1 M fluoride was added to the near boiling HNO_3, the remaining U and Pu masses were 16.9 ±2.9 g (4.25 ±0.91 g U-235) and 4.46 ± 1.12 g, respectively, at 95% confidence limits. The more aggressive dissolution conditions also reduced the mass of the solids by 50-70% leaving primarily Si as SiO_2. Continued dissolution of SNF in the 6.4D dissolver (using starting HNO_3 concentrations of approximately 7 M) should slowly dissolve the very small quantities of U and Pu remaining in the dissolver and address any significant accumulation of fissile material in the future. A dissolver clean-out may be performed using nominally 7 M HNO_3 containing 0.1 M fluoride which will reduce the total amounts of solids and fissionable materials. Based on this work, these solid are expected in either H-Canyon dissolver during SNF dissolution and H-Canyon SNF dissolution flowsheet is functioning as expected.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

APHA	alpha pulse height analysis
CVAAS	cold vapor atomic absorption spectroscopy
GPHA	gamma pulse height analysis
HFIR	High Flux Isotope Reactor
ICPES	inductively-coupled plasma emission spectrometry
ICPMS	inductively-coupled plasma mass spectroscopy
SEM	scanning electron microscopy
SNF	spent nuclear fuel
SRNL	Savannah River National Laboratory
TTA	thenoyltrifluoroacetone
XRD	X-ray diffraction

1.0 Introduction

During reconfiguration of the 6.4D dissolver for the dissolution of High Flux Isotope Reactor (HFIR) fuel, solid material was observed on the bottom of the vessel. Samples of the solids were taken by H-Canyon Operations from the north (HCA-North) and south side (HCA-South) of the 6.4D dissolver pot. The samples were transported to the Savannah River National Laboratory (SRNL) for characterization. H-Canyon Engineering requested that the SRNL determine the elemental content of the solids to verify that fissile materials (e.g., U and Pu) dissolved as expected during the dissolution of Spent Nuclear Fuel (SNF). Characterization of the solids included radiochemical analyses (e.g., actinides, fission products, and total alpha and beta emitters) and elemental analysis by inductively-coupled plasma mass spectrometry (ICPMS). Performing both radiochemical and ICPMS analyses provided a comparison of the measured amount of insoluble and soluble fissile materials by independent methods. Other characterization techniques performed included surface examination by scanning electron microscopy (SEM) and X-ray diffraction (XRD) analysis in an attempt to provide information on crystal structure and phase identification. The SRNL was also requested to measure the bulk density and settling times of the solids.[1]

Insoluble materials have been observed in equipment associated with H-Canyon dissolvers in the past. In 2007 during a periodic inspection of the physical spacers located within the 6.1D Dissolver MK-12 insert, a significant deposit of solid material was found in the spacer support structure (Figure 1-1).

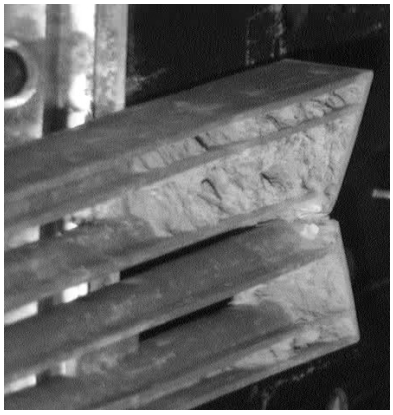

Figure 1-1. Solids Deposited in the MK-12 Insert Support Structure

A sample of the solids was collected and transported to SRNL for characterization. Data from the radiochemical and elemental analyses showed that the solids contained 0.15 wt % U (60% U-235 enrichment) with a relative standard deviation of 15%. The Pu and Np concentrations in the solids were 0.032 and 0.00066 wt % respectively, with relative standard deviations of 5 and 15%, respectively. The majority of the mass associated with the solids were fission product Zr and Mo oxides. X-ray diffraction analysis indicated that the only crystalline compound present was $Zr[Mo_2O_7(OH_2)(H_2O)_2]$.[2]

The precipitation of solids from completely dissolved fuels has been observed during intermediate storage of solutions. A number of hydrated oxides have been identified. Precipitated solids include compounds containing Mo, Tc, Ru, Rh, Pd, Te, Zr, U, and Pu.[3-4] The presence of Pu and U in these materials is generally associated with zirconium molybdate. The studies concerning the characterization and chemical behavior of the solids are limited; however, structures proposed for the zirconium molybdate include $Zr(MoO_4)_2 \cdot H_2O$ and $ZrMo_2O_7(OH)_2 \cdot H_2O$. Zirconium molybdate is known to act as a host lattice for Pu and can co-precipitate significant amounts of Pu from solution.[5-6]

The source of 6.4D dissolver solids was not known upon the receipt of the samples at SRNL. Post-dissolution precipitation of fission products with co-precipitation of U and Pu, like the process which deposited solids on the MK-12 insert support structure, was a possible mechanism. Incomplete dissolution of spent nuclear fuel processed in the 6.4D dissolver or subsequent precipitation of a component of the fuel were also possible mechanisms for generating solids. Uranium silicide fuels have been processed in the 6.4D dissolver in recent campaigns.[7] Following dissolution of silicide fuels during flowsheet development experiments using irradiated miniplates, precipitation of silica from the solution was observed.[8] During these experiments, there was no evidence that either U or Pu precipitated with the solids. During the characterization of the samples from the 6.4D dissolver, presented in this report, it was important to distinguish the fissile materials (i.e., U and Pu) which may be physically associated with the solids due to residual, entrained dissolver solution, from fissile material chemically associated with the solids. In anticipation that soluble U may be present in the solids, both unrinsed solids and solids rinsed with HNO_3 were dissolved and analyzed. A dissolution flowsheet was also tested which may be used to clean-out the dissolver pot by dissolving any residual solids.

2.0 Experimental Procedure

The HCA-North and HCA-South samples from the 6.4D dissolver were received into the SRNL Shielded Cells due to high beta/gamma dose rates. The samples were damp and dark gray in color. Each of the samples was weighed, transferred into 50-mL glass beakers, and dried in an oven overnight at 120 °C. The dry samples were then reweighed.

2.1 Phase 1: Initial Characterization of Solids

An initial characterization of the solids was performed to measure the elemental composition. A small portion (e.g., 0.25 g) of the unrinsed, dried HCA-North and HCA-South samples was removed from the beakers and dissolved using a sodium peroxide fusion method to prepare samples for elemental and radiochemical analyses.

A slightly larger portion (e.g., 0.7 g) of the HCA-North and HCA-South samples was rinsed to remove any soluble elements associated with the original liquid contained in the as received solids. The solids were placed in a beaker and rinsed with 50 mL of 1 M HNO_3. Each sample was initially combined with approximately 25 mL of HNO_3, shaken, and poured over a 0.45 µm vacuumed filter. Then the remaining approximately 25 mL aliquot of solution was transferred by slurry pipette to flush solids from the beaker and rinse the filtered solids. Samples of the filtrates were collected for elemental and radiochemical analyses. The rinsed solids were dried at ambient conditions on the filter for approximately 3 hours, so that they could be easily transferred into separate beakers and placed into an oven to dry overnight at 120 °C. Approximately 0.25 g each of the dry, rinsed solids were dissolved using the sodium peroxide fusion method to prepare samples for elemental and radiochemical analyses. The samples rinsed using 1 M HNO_3 were designated HCA-North 1 M and HCA-South 1 M.

Another small portion (e.g., 0.7 g) of the unrinsed, dried HCA-North and HCA-South samples was subsequently transferred to small beakers for rinsing with 50 mL of 6 M HNO_3 using the same rinse and dry procedure prior to the preparation of samples by the sodium peroxide fusion method for analysis. The samples rinsed using 6 M HNO_3 were designated HCA-North 6 M and HCA-South 6 M.

The filtrate and solutions generated from the sodium peroxide fusion dissolution method were analyzed using liquid scintillation counting (for total alpha and beta emitters), gamma pulse height analysis (GPHA) (for gamma emitters), GPHA with Cs removal (for lower activity gamma emitters), thenoyltrifluoroacetone (TTA) extraction/alpha pulse height analysis (APHA) (for Pu-238 and Pu-239/240), cold vapor atomic absorption spectroscopy (CVAAS) (for Hg), ICPMS (for fission products, actinides, and U/Pu isotopic information), and inductively-coupled plasma emission spectroscopy (ICPES) (for U and low molecular

weight fission and corrosion products). A portion of the rinsed and unrinsed solids was prepared for surface analysis by SEM and XRD.

The bulk density of each of the prepared samples: HCA-North, HCA-South, HCA-North 1 M, HCA-South 1 M, HCA-North 6 M, and HCA-South 6 M, was measured by placing approximately 1 mL of material in a graduated cylinder and recording the weight and volume.

2.2 Phase 2: Secondary Rinse

A small portion of the HCA-North, HCA-South, and HCA-South 1 M rinsed solids was transferred to a glovebox for additional density measurements. A secondary rinse was also performed on the HCA-South 1 M sample. The bulk density was measured by placing approximately 1 mL of material in a small graduated cylinder. Once the solids were added, the weight of the solids was obtained. This process was repeated twice per sample.

The HCA-North and HCA-South samples were prepared for an additional density/settling test. Approximately 5 mL of pre-weighed 1 M HNO_3 was added to 1 mL of pre-weighed HCA-North solids. The mixture was then swirled in a beaker, poured back into the graduated cylinder, and timed to measure how long the solids took to settle. Once the solids settled, the volume was measured, and density calculated from the mass balance. This procedure was repeated for the HCA-South sample.

The South 1M rinsed solids (from Phase 1) were rinsed a second time and prepared for additional analysis. Approximately 0.20 g of the sample was rinsed with a total of 50 mL of 1 M HNO_3. It was first mixed with 25 mL of HNO_3 and filtered using a 0.45 µm vacuumed filter. The filtered solids were then rinsed with the remaining 25 mL of HNO_3 using a pipette. Both the (dissolved) solids and a sample of the filtrate were analyzed by GPHA (for gamma emitters), GPHA with Cs removal (for lower activity gamma emitters), TTA extraction/APHA (for Pu-238 and Pu-239/240), ICPMS (for fission products, actinides, and U/Pu isotopic information), and ICPES (for U and low molecular weight fission and corrosion products).

2.3 Phase 3: Dissolution in 7 M HNO_3

A portion of the HCA-North, HCA-South, and the rinsed solids samples (both the 1 M and 6 M rinse for each sample) were prepared for solubility testing in the SRNL Shielded Cells. A starting weight was obtained for each of the six solid samples. The HCA-North and HCA-South samples were combined into one sample of non-rinsed solids, while the 1 M and 6 M rinsed solids were combined into one rinsed sample. A total weight was obtained for both combined samples. A target mass of 0.4 g each of both samples was added to 50 mL of pre-weighed 7 M HNO_3. The solutions were placed on a hot plate, covered with a watch glass that contained a small amount of water to reduce evaporation losses, and heated to near boiling while stirring. The solution was held at near boiling for approximately 3 hours and then observed for visible solids. Afterwards, the solutions were left to cool and again observed for changes in the solution and solids. Once cooled, the solutions were poured through a 0.45 µm vacuumed filter. A sample of each filtrate was collected for analysis. The solids were dried in ambient air for approximately 1 hour, then transferred to a pre-weighed crucible and dried in an oven at 120 ˚C before being weighed. Samples of the (dissolved) solids and the filtrate solutions were analyzed by GPHA (for gamma emitters), GPHA with Cs removal (for lower activity gamma emitters), TTA extraction/APHA (for Pu-238 and Pu-239/240), ICPMS (for fission products, actinides, and U/Pu isotopic information), and ICPES (for U and low molecular weight fission and corrosion products).

2.4 Phase 4: Dissolution in 7 M HNO_3 containing 0.1 M Fluoride

To demonstrate a chemical cleanout flowsheet that could be utilized in the H-canyon 6.4D dissolver, approximately 0.5 g of rinsed solids (from both the HCA-North and HCA-South samples) were placed in 100 mL of pre-weighed 7 M HNO_3 containing 0.1 M fluoride. The solution was placed on a hot plate,

covered with a watch glass that contained a small amount of water to reduce the evaporation rate, and heated to near boiling while stirring. The solution was held at near boiling for approximately 12 hours over the course of two shifts. The solution was periodically checked for evaporation and when more than 20 mL of solution evaporated an additional aliquot was added. After completing the dissolution, the solution was cooled and poured through a 0.45 µm vacuumed filter. The remaining solids were dried in ambient air for approximately one hour and transferred to a pre-weighed crucible which was dried in an oven at 120 ˚C before being weighed. A sample of the (dissolved) solids was analyzed by GPHA (for gamma emitters), GPHA with Cs removal (for lower activity gamma emitters), TTA extraction/APHA (for Pu-238 and Pu-239/240), ICPMS (for fission products, actinides, and U/Pu isotopic information), and ICPES (for U and low molecular weight fission and corrosion products).

2.5 Quality Assurance

A Functional Classification of Safety Significant was applied to this work. Analytical measurement systems with a General Service functional classification were used to collect data during the characterization of solids from the 6.4D Dissolver. Standards used to calibrate these systems were purchased at level 2 with a certificate of analysis. Chemical reagents used in experiments and sample preparation were purchased at levels 2 or 3. Standards used for analytical measurements were traceable to NIST or equivalent per manual 1Q, 2-7 section 5.2.3.

To match the requested functional classification, this report received technical review by design verification. Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

3.0 Results and Discussion

3.1 Phase 1 & 2: Characterization of Solids

Elemental concentrations were measured by ICPES, ICPMS (reported by m/z numbers), and TTA/GPHA for the rinsed and unrinsed solids. Unrinsed solids are designated “unrinsed” and the rinsed solids are identified using the concentration of HNO_3 used for the rinse solution. A second rinse was performed on the South-1M material, which is designated as South-1M-B. The ICPES results for selected elements provided in Table 3-1 indicate that solids are mostly comprised of Si, Fe, Al, and Cr and would include a corresponding anion (e.g., oxide or nitrate). The analytical results for all elements are provided in Appendix A (Table A-1).

Table 3-1. ICPES Analysis of Solids Before and After Rinse

Element	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Al	4.88	5.15	0.337	0.357	0.388	0.314	0.386
Ca	0.380	0.323	0.311	0.251	0.481	0.357	0.262
Cr	1.340	0.347	0.508	0.164	0.176	0.475	0.205
Fe	4.95	1.31	1.90	0.642	0.717	1.79	0.831
Mg	0.023	0.026	0.002	< 0.001	< 0.014	< 0.002	0.003
Ni	0.733	0.193	0.261	0.083	< 0.169	0.260	0.117
Si	18.8	26.2	32.2	27.6	33.2	31.2	28.6

The ICPES filtrate results for selected elements are provided in Table 3-2. The analytical results for all elements are provided in Appendix A (Table A-3). The data for the solids (Table 3-1) and filtrate analyses

show that much of the Al, Fe, Cr and Ni was removed from the solids by both the 1 and 6 M HNO_3 rinses. There does not appear to be a significant impact of the second rinse with 1 M HNO_3 (Table 3-1). The filtrate results also show that U was rinsed from the solids; however, the U concentration in the solids was below the ICPES detection limit. The filtrate results indicate that Si was not dissolved by either of the acid rinses. Rinses with 1 M HNO_3 removed more Al, Fe, Cr, and Ni than rinses using 6 M HNO_3.

Table 3-2. ICPES Analysis of Rinse Filtrate

Element	North-1M	South-1M	North-6M	South-6M
	wt %	wt %	wt %	wt %
Al	12.6	15.0	10.2	10.5
Ba	0.012	0.015	0.010	0.011
Ca	0.055	0.048	0.045	0.039
Cr	0.321	0.377	0.257	0.258
Fe	1.135	1.348	0.894	0.854
La	< 0.008	0.010	< 0.009	0.008
Mn	0.033	0.040	0.026	0.028
Ni	0.173	0.214	0.142	0.149
Si	< 0.065	< 0.071	< 0.076	< 0.061
Sr	0.006	0.008	0.005	0.005
U	0.858	1.156	0.657	0.811
Zr	0.061	0.089	0.091	0.091

The most prevalent elements measured by the ICPMS analysis for the solids are shown in Table 3-3. The filtrate results are provided in Table 3-4. The analytical results for all elements are provided in Appendix A (Table A-4 and Table A-6, respectively). The ICPMS method reports the mass of isotopes with the same mass number divided by charge number. Therefore, using the decay chain of U-235 and half-lives of the isotopes, the most likely species were provided in the tables. Fissionable species were further resolved with radiochemical analytical techniques.

Table 3-3. ICPMS Analysis of Solids Before and After Rinse

Most Likely Isotope	Other Possible Isotopes	m/z	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %	wt %	wt %
Nb-93	Zr-93 Mo-93	93	0.164	0.168	0.212	0.197	0.184	0.207	0.212
Mo-95		95	0.024	0.013	0.018	0.011	0.011	0.016	0.013
Mo-96	Zr-96 Ru-96	96	0.444	0.464	0.382	0.401	0.369	0.45	0.397
Mo-97	Tc-97	97	0.021	0.011	0.017	0.01	0.01	0.014	0.012
Mo-98	Ru-98 Tc-98	98	0.025	0.013	0.018	0.011	0.011	0.016	0.013
Ru-100 Mo-100		100	0.022	0.012	0.018	0.011	0.01	0.015	0.013
Ru-101	Rh-101	101	0.005	0.009	0.007	0.009	0.009	0.01	0.01
Te-126	Sn-126	126	0.021	0.025	0.030	0.026	0.033	0.030	0.027
Gd-156		156	0.009	0.011	0.001	0.001	0.001	0.001	0.001
Gd-158		158	0.011	0.013	0.001	0.001	0.001	0.001	0.001
Gd-160		160	0.009	0.012	0.001	0.001	0.001	0.001	0.001
Hf-177		177	0.024	0.026	0.017	0.018	0.019	0.022	0.017
Hf-178		178	0.035	0.038	0.025	0.027	0.028	0.032	0.025
Hf-179	Ta-179	179	0.018	0.02	0.013	0.014	0.014	0.017	0.013
Hf-180	Ta-180 W-180	180	0.045	0.049	0.032	0.034	0.035	0.042	0.033
Ta-181		181	0.001	0.001	0.001	0.001	0.003	0.001	0.001
W-182		182	0.013	0.008	0.012	0.008	0.011	0.011	0.009
W-183		183	0.008	0.005	0.008	0.005	0.007	0.007	0.006
W-184		184	0.016	0.010	0.016	0.010	0.014	0.014	0.012
W-186		186	0.014	0.008	0.014	0.009	0.012	0.012	0.01
Pt-196		196	0.001	0.001	0.000	0.000	0.000	0.001	0.000
U-232	Th-232	232	0.01	0.01	0.02	0.016	0.019	0.02	0.02
U-235	Np-235	235	0.12	0.147	0.012	0.013	0.007	0.01	0.014
U-236	Np-236 Pu-236	236	0.015	0.018	0.002	0.002	0.001	0.001	0.002
Np-237		237	0.001	0.001	0.000	0.000	0.000	0.000	0.000
U-238	Pu-238	238	0.205	0.251	0.042	0.041	0.026	0.041	0.046
Pu-239		239	0.007	0.008	0.008	0.008	1.682	0.008	0.009
Pu-240		240	0.002	0.002	0.002	0.002	0.104	0.002	0.002

Table 3-4. ICPMS Analysis for Rinse Filtrate

Most Likely Isotope	Other Possible Isotopes	m/z	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %
Zr-90	Sr-90	90	0.006	0.009	0.000	0.008	0.008
Zr-91	Nb-91 Y-91	91	0.012	0.017	0.000	0.017	0.017
Zr-92	Nb-92 Mo-92	92	0.011	0.016	0.000	0.015	0.016
Nb-93	Zr-93 Mo-93	93	0.014	0.020	0.005	0.019	0.019
Zr-94	Mo-94 Nb-94	94	0.012	0.018	0.000	0.017	0.018
Mo-95		95	0.004	0.006	0.000	0.004	0.004
Mo-96	Zr-96 Ru-96	96	0.011	0.016	0.004	0.015	0.016
Mo-97	Tc-97	97	0.004	0.006	0.000	0.003	0.004
Mo-98	Ru-98 Tc-98	98	0.004	0.006	0.000	0.004	0.004
Ru-99	Tc-99 Mo-99	99	0.004	0.005	0.000	0.003	0.003
Ru-100 Mo-100		100	0.004	0.006	0.000	0.004	0.004
Ru-101	Rh-101	101	0.004	0.005	0.000	0.003	0.004
Ru-102	Pd-102	102	0.003	0.005	0.000	0.003	0.003
Sn-119		119	0.009	0.013	0.000	0.007	0.009
Cs-133	Ba-133	133	0.006	0.008	0.000	0.004	0.005
Cs-137	Ba-137	137	0.006	0.008	0.000	0.004	0.005
Ba-138	La-138 Ce-138	138	0.008	0.010	0.000	0.007	0.007
La-139		139	0.007	0.010	0.000	0.006	0.007
Ce-140		140	0.007	0.009	0.000	0.006	0.007
Pr-141		141	0.006	0.009	0.000	0.005	0.007
Ce-142	Nd-142	142	0.007	0.010	0.000	0.006	0.007
Nd-143		143	0.006	0.008	0.000	0.004	0.005
Nd-144	Ce-144	144	0.008	0.010	0.000	0.006	0.007
Nd-145	Pm-145	145	0.004	0.006	0.000	0.003	0.004
Nd-146	Sm-146 Pm-146	146	0.004	0.005	0.000	0.003	0.004
Eu-154 Sm-154		154	0.003	0.004	0.000	0.002	0.003
Eu-155 Gd-155		155	0.017	0.023	0.000	0.013	0.016
Gd-156		156	0.024	0.031	0.000	0.018	0.022
Gd-157		157	0.018	0.024	0.000	0.014	0.017
Gd-158		158	0.029	0.038	0.000	0.022	0.026
Gd-160		160	0.026	0.034	0.000	0.020	0.024
Pt-198 Hg-198		198	0.030	0.045	0.000	0.024	0.031
Hg-204	Ti-204 Pb-204	204	0.017	0.025	0.000	0.014	0.018
U-234		234	0.005	0.006	0.000	0.004	0.004
U-235	Np-235	235	0.318	0.433	0.005	0.256	0.299
U-236	Np-236 Pu-236	236	0.040	0.055	0.001	0.032	0.038
U-238	Pu-238	238	0.494	0.671	0.007	0.392	0.453
Pu-239		239	0.003	0.004	0.008	0.002	0.003

The most prevalent species in the solids identified by ICPMS are Mo, Nb, and U. Solid and filtrate results indicate U is readily rinsed out. Niobium and Mo mostly remain as a solid. Uranium, Hg, and Gd are the most abundant species in the filtrate. Uranium and Hg appear slightly more soluble in 1 M HNO_3; whereas Gd appears to be equally soluble in both acid concentrations. The slight increase in solubility may be due to the dynamic equilibrium of the nitrate salts.

Radiochemical analyses were performed to better resolve the concentrations of the fissionable species in the solids. The analytical results for selected isotopes are provided in Table 3-5. Complete results for the solids and filtrates are provided in Appendix A (Table A-8 and Table A-10 respectively). Very low concentrations of the radionuclides were measured in the filtrates causing most of the calculated mass percentages to be less than 0.000 wt % (to three significant digits). The solids contained low amounts of Pu-238, which also resulted in calculated mass percentages less than 0.000 wt %. Some Pu-239 and Pu-240 were measured that does not appear to rinse out. However, the total Pu content of the solids was very low, averaging 0.010 wt %. The significantly higher concentrations of Pu-239 and Pu-240 measured in sample South-1MB is due to cross-contamination from performing the work in a glovebox which was highly contaminated with weapons grade Pu.

Table 3-5. Radiochemical Analysis of Solids Before and After Rinse

	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Sn-126	0.002	0.003	0.003	0.002	0.005	0.003	0.002
Cs-137	0.002	0.000	0.000	0.000	0.000	0.000	0.000
Pu-238	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Pu-239	0.007	0.008	0.007	0.008	1.752	0.008	0.008
Pu-240	0.002	0.002	0.002	0.002	0.108	0.002	0.002
Am-241	0.000	0.000	0.000	0.000	0.000	0.000	0.000

The primary contents of the solids are summarized in Table 3-6. Because the solids were wet when received at SRNL, then dried as necessary for proper analysis, it was difficult to determine which species were present in the 6.4D dissolver as solids verses solids generated by evaporation (i.e., drying) after receipt at SRNL. The analysis of the rinsed solids indicate that the primary components are Si, Fe, Mo and Al. Uranium is present in the unrinsed samples at 0.342 wt % (0.120 wt % U-235) (HCA-North) and 0.418 wt % (0.147 wt % U-235) (HCA-South). The U content of the rinsed solids (0.052 to 0.062 wt % total U) is much lower than the unrinsed solids indicating that a majority of the U present in the solids is soluble in HNO_3. It is likely that the soluble U was present in the dried, unrinsed solids either interstitially or as nitrate crystals that formed during evaporation. The second rinse, South-1M-B, removed only slightly more Nb, and U.

Table 3-6. Dissolved Solids Summary

Unrinsed Solids				*Rinsed Solids*						
		North-Unrinsed	South-Unrinsed			North-1M	South-1M	South-1M-B	North-6M	South-6M
Species	**Method**	**wt %**	**wt %**	**Species**	**Method**	**wt %**	**wt %**	**wt %**	**wt %**	**wt %**
Si	ICPES	18.8	26.2	Si	ICPES	32.2	27.6	33.2	31.2	28.6
Al	ICPES	4.88	5.15	Fe	ICPES	1.90	0.642	0.717	1.79	0.831
Fe	ICPES	4.95	1.31	Mo-96	ICPMS	0.401	0.382	0.369	0.450	0.397
Mo-96	ICPMS	0.464	0.444	Al	ICPES	0.337	0.357	0.388	0.314	0.386
Cr	ICPES	1.34	0.347	Ca	ICPES	0.311	0.251	0.481	0.357	0.262
U-238	ICPMS	0.205	0.251	Cr	ICPES	0.508	0.164	0.176	0.475	0.205
Ni	ICPES	0.733	0.193	Ni	ICPES	0.261	0.083	< 0.169	0.26	0.117
Nb-93	ICPMS	0.168	0.164	U-238	ICPMS	0.042	0.041	0.026	0.041	0.046
U-235	ICPMS	0.120	0.147	U-235	ICPMS	0.012	0.013	0.007	0.010	0.014
Pu-239	ICPMS	0.007	0.008	Pu-239	ICPMS	0.008	0.008	-	0.008	0.009
Pu-240	Pu TTA	0.002	0.002	Pu-240	Pu TTA	0.002	0.002	-	0.002	0.002

The bulk density was measured by placing dry solids into a graduated cylinder to determine the volume and to weigh the material. In addition, 5 mL of pre-weighed, 1 M HNO_3 was mixed with the dried solids. The solids were allowed to settle over time in the graduated cylinder. The settling times were less than one minute. The volume of the settled solids in the solution and the total mass were measured to calculate an alternative ('settling') density. Results of the density tests are summarized in Table 3-7. Test Set 1 was performed in the SRNL Shielded Cells using manipulators. Test Sets 2-5 were performed manually in a glovebox.

The average density of the dry and settled solids was 0.6 ± 0.1 g/cm^3. H-Canyon Engineering personnel estimated that 157,000 cm^3 of solids were present on the bottom of the 6.4D dissolve pot.[9] Based on this volume and the measured density, the total mass of solids was 94.4 kg. Undissolved solids (e.g., noble metal fission products and SiO_2 from U silicide (U_3Si or U_3Si_2) research reactor fuels) are always a product of SNF dissolution and are regularly transferred from the dissolver to be removed from the solution by centrifugation in Head End and then transferred to waste. Visual inspection of the 6.4D dissolver pot indicated the solids were primarily located on the high side of the vessel away from the jet, which may be a result of the solids settling during the transfer of solution.[9]

Table 3-7. Calculated Density of Dry Solids

Test Set	*Sample ID: Location- Rinse*	Solids Mass	Solid Volume	Calculated Density	Method
		g	cm^3	g/cm^3	
1	North-Unrinsed	0.72	1.1	0.7	Bulk
	North-1M	0.45	0.8	0.6	Bulk
	North-6M	0.33	0.6	0.5	Bulk
	South- Unrinsed	0.57	1.1	0.5	Bulk
	South-1M	0.60	1.0	0.6	Bulk
	South-6M	0.38	0.7	0.5	Bulk
2	North- Unrinsed	0.72	1.0	0.7	Bulk
	South	0.58	1.0	0.6	Bulk
	South-1M	0.20	0.3	0.7	Bulk
3	North- Unrinsed	0.68	1.0	0.7	Bulk
	South- Unrinsed	0.55	1.0	0.6	Bulk
	South-1M	0.21	0.3	0.7	Bulk
4	North- Unrinsed	0.74	1.0	0.7	Bulk
	South- Unrinsed	0.59	1.0	0.6	Bulk
	South-1M	0.21	0.3	0.7	Bulk
5	North- Unrinsed	0.40	0.8	0.5	Settling
	South- Unrinsed	0.30	0.8	0.4	Settling
			Avg.	*0.6 ± 0.1*	

Using the analytical results for the unrinsed material (Table 3-6), the bounding total masses of U and Pu present were 359 ± 45 g (126 ± 20 g U-235) and 8.6 ± 1.4 g, respectively, at 95% confidence limits. The total masses of U and Pu present in the rinsed solids were 54 ± 6 g (12 ± 2 g U-235) and 9.4 ± 1.1 g, respectively, at 95% confidence limits. The confidence limits in the masses of the fissionable materials were calculated by GUM Workbench[10] using propagation of error techniques based on the uncertainty in the ICPMS method (a one sigma uncertainty of 10%), the individually measured uncertainties of the Pu isotopes from the TTA/APHA method (a one sigma uncertainty of 5-10%), and the uncertainty in the average density of the solids. The volume of the solids in the 6.4D dissolver was a bounding value; therefore, an uncertainty was not used in the calculations. A summary of the estimated masses and 95% confidence limits of the fissionable material in the 6.4D dissolver solids is provided in Table 3-8.

Table 3-8. Estimated Mass of Fissionable Species

Isotope	Unrinsed		1 M		6 M	
	Mass	Uncertainty	Mass	Uncertainty	Mass	Uncertainty
	(g)	(g)	(g)	(g)	(g)	(g)
U-234	1.78	0.29	0.16	0.03	0.15	0.03
U-235	126	20	11.8	1.9	11.7	1.9
U-236	15.7	2.5	1.56	0.25	1.57	0.26
U-238	215	35	39.4	6.3	41.1	6.6
*Total U**	*359*	*45*	*52.9*	*7.1*	*54.5*	*7.4*
Pu-238	0.16	0.03	0.13	0.02	0.13	0.02
Pu-239	6.86	1.28	7.34	1.23	7.71	1.32
Pu-240	1.53	0.30	1.67	0.30	1.75	0.32
*Total Pu**	*8.55*	*1.45*	*9.14*	*1.36*	*9.59*	*1.47*

***** Isotopic masses may not sum to the elemental total due to rounding.

3.2 Phase 3 & 4: Dissolution Strategy

To evaluate the dissolution behavior of the solids using more aggressive conditions (similar to conditions in the 6.4D dissolver during SNF dissolution), samples of the unrinsed and rinsed solids were contacted with near boiling 7 M HNO_3. North and South samples were combined to create two distinct composites (unrinsed and rinsed). A subsample of each composite, designated as Unrinsed-7M and Rinsed-7M, was then contacted with the HNO_3 solution for about three hours. To demonstrate a chemical cleanout method that could be utilized in the 6.4D dissolver, a sample of the rinsed composite was also contacted with near boiling 7 M HNO_3 containing 0.1 M KF (Rinsed-7M-F) for approximately 12 hours. The resulting mass balances for the dissolution experiment are shown in Table 3-9.

Table 3-9. Dissolution Mass Balance

	Starting Solids Mass	Ending Solids Mass	Mass Loss	Mass Loss
	(g)	(g)	(g)	(%)
Unrinsed-7M	0.427	0.166	0.261	61.1%
Rinsed-7M	0.465	0.133	0.332	71.4%
Rinsed-7M-F	0.515	0.257	0.258	50.1%

The analytical results for the elements and isotopes of interest from the ICPES and radiochemical analyses for the solids dissolution experiments are provided in Table 3-10 and the ICPMS results for the three samples are provided in Table 3-11. The complete analysis of the samples are provided in Appendix A (Table A-2, Table A-3, Table A-5, Table A-7, Table A-9, and Table A-11). The initial element and isotope concentrations for the unrinsed and rinsed composite samples are also provided in the two tables. The concentrations were calculated from data in Table 3-1, Table 3-3, and Table 3-5 using a weighted average based on the masses of the combined solids. There is a significant reduction in the amount of Al, Fe, and Cr, when compared to the unrinsed solids. The dissolved solids still contain very high amounts of Si due to the low solubility of SiO_2 in HNO_3 solutions.[11] The undissolved solids remaining from contact with the 7 M HNO_3 solution containing 0.1 M fluoride appear to be primarily Si, consistent with the low solubility of SiO_2 in HNO_3. The Pu concentration in the solids remained nearly constant and was only reduced in concentration when contacted with the HNO_3 containing fluoride. During dissolution of the composite solids with 7M HNO_3, which is similar to the conditions for dissolution of SNF in H-Canyon,[12,13] the U concentration appears to have reached a relatively constant value in the solids. The U concentrations following the HNO_3 dissolution for both the rinsed and unrinsed solids are nearly the same. The U isotopic concentrations are 0.007-0.008 wt % U-235, 0.001 wt % U-236, and 0.029-0.033 wt % U-238 (Table 3-11). The addition of 0.1 M fluoride to the HNO_3 solution reduced the total U concentration to 0.018 wt %.

Table 3-10. ICPES and Radiochemical Analyses for Solids Dissolutions

Element	Initial Unrinsed	Unrinsed-7M		Initial Rinsed	Rinsed-7M		Rinsed-7M-F	
	Solids	Solids	Filtrate	Solids	Solids	Filtrate	Solids	Filtrate
	wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Al	5.069	0.460	9.133	< 0.012	0.448	0.265	0.329	-
Ba	< 0.007	0.020	0.010	0.307	0.025	< 0.001	< 0.004	-
Be	0.000	< 0.003	0.001	< 0.006	< 0.004	< 0.001	0.000	-
Cr	0.646	0.080	0.283	0.000	0.073	0.059	< 0.018	-
Fe	2.405	0.337	1.063	0.286	0.351	0.300	< 0.086	-
Gd	< 0.030	< 0.046	0.093	1.092	< 0.052	< 0.014	< 0.011	-
Li	0.014	< 0.147	< 0.043	0.012	< 0.167	< 0.044	0.015	-

Element	Initial Unrinsed	Unrinsed-7M		Initial Rinsed	Rinsed-7M		Rinsed-7M-F	
	Solids	Solids	Filtrate	Solids	Solids	Filtrate	Solids	Filtrate
	wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Mg	0.025	< 0.010	0.093	< 0.024	< 0.011	0.033	0.030	-
Mn	< 0.007	< 0.008	0.027	0.002	< 0.010	0.003	< 0.002	-
Na	-	-	0.115	-	-	0.226	-	-
Ni	0.355	< 0.421	0.145	0.152	< 0.478	< 0.124	< 0.059	-
Si	23.975	20.0	0.119	26.01	40.0	0.168	28.5	-
Sr	< 0.083	0.000	0.005	< 0.066	< 0.005	< 0.001	< 0.009	-
U	< 0.183	1.76	0.696	< 0.144	< 2.00	< 0.520	< 0.426	-
Zn	< 0.010	< 0.027	0.013	< 0.007	< 0.037	0.008	< 0.006	-
Zr	-	-	0.187	-	-	0.173	-	-
Pu-239	0.008	0.007	0.000	0.008	0.007	0.000	0.004	0.000
Pu-240	0.002	0.002	0.000	0.002	0.002	0.000	0.001	0.000

Table 3-11. ICPMS Analyses for Solids Dissolutions

Most likely	Possible	m/z	Initial unrinsed	Unrinsed-7M		Initial unrinsed	Rinsed-7M		Rinsed-7M-F	
			solids	solids	filtrate	solids	solids	filtrate	solids	filtrate
			wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %
		90	-	-	0.007	-	-	0.007	-	0.061
		91	-	-	0.017	-	-	0.014	-	0.161
		92	-	-	0.017	-	-	0.014	-	0.148
Nb-93	Zr-93 Mo-93	93	0.167	0.218	0.019	0.206	0.211	0.218	0.140	0.219
		94	-	-	0.018	-	-	0.015	-	0.171
Mo-95		95	0.016	0.010	0.007	0.014	0.007	0.005	0.004	0.019
Mo-96	Zr-96 Ru-96	96	0.458	-	0.017	0.404	-	0.014	-	0.150
Mo-97	Tc-97	97	0.014	0.009	0.006	0.013	0.006	0.005	0.004	0.018
Mo-98	Ru-98 Tc-98	98	0.016	0.010	0.006	0.014	0.007	0.005	0.004	0.020
Ru-100 Mo-100		100	0.015	0.001	0.006	0.014	0.001	0.005	0.004	0.019
Ru-101	Rh-101	101	0.008	0.010	0.002	0.009	0.006	0.001	0.006	0.007
Ru-102	Pd-102	102	0.007	0.009	0.002	0.008	0.009	0.000	0.005	0.006
Rh-103		103	0.004	0.008	0.001	0.004	0.008	0.000	0.003	0.004
Ru-104	Pd-104	104	0.004	0.005	0.001	0.005	0.004	0.000	0.003	0.003
		106	0.009	0.014	0.000	0.007	0.019	0.000	0.010	0.000
Pd-107	Ag-107	107	0.003	0.005	0.000	0.003	0.007	0.000	0.010	0.000
		108	0.003	0.005	0.000	0.002	0.007	0.000	0.010	0.000
Sn-118		118	0.005	0.006	0.001	0.004	0.008	0.000	0.005	0.002
Te-126	Sn-126	126	0.024	0.026	0.002	0.028	0.032	0.002	0.027	0.027
Pr-141		141	0.003	0.000	0.005	0.000	0.000	0.000	0.000	0.000
Gd-156		156	0.010	0.001	0.009	0.001	0.000	0.000	0.000	0.001
Gd-157		157	0.008	0.000	0.007	0.001	0.000	0.000	0.000	0.001
Gd-158		158	0.012	0.001	0.011	0.001	0.001	0.000	0.001	0.000
Lu-176	Hf-176 Yb-176	176	0.007	0.010	0.000	0.005	0.014	0.000	0.006	0.000
Hf-177		177	0.026	0.038	0.000	0.018	0.053	0.000	0.023	0.000
Hf-178		178	0.037	0.057	0.000	0.027	0.079	0.000	0.033	0.000
Hf-179	Ta-179	179	0.019	0.027	0.000	0.014	0.038	0.000	0.017	0.000
Hf-180	Ta-180 W-180	180	0.048	0.074	0.000	0.035	0.100	0.000	0.043	0.000
W-182		182	0.009	0.012	0.001	0.010	0.009	0.001	0.004	0.014
W-183		183	0.006	0.007	0.000	0.006	0.005	0.001	0.003	0.008
W-184		184	0.012	0.015	0.001	0.012	0.011	0.001	0.006	0.018

Most likely	Possible	m/z	Initial unrinsed	Unrinsed-7M		Initial unrinsed	Rinsed-7M		Rinsed-7M-F	
			solids	solids	filtrate	solids	solids	filtrate	solids	filtrate
			wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %
W-186		186	0.010	< 0.013	0.001	0.011	0.009	0.001	0.005	0.016
Pt-198 Hg-198		198	0.005	0.001	0.012	0.001	0.000	0.000	0.000	0.001
U-232 Th-232		232	0.011	0.018	0.001	0.016	0.021	0.000	0.021	0.005
U-235	Np-235	235	0.139	0.008	0.131	0.013	0.007	0.005	0.005	0.018
U-236	Np-236 Pu-236	236	0.017	0.001	0.016	0.002	0.001	0.001	0.000	0.003
U-238	Pu-238	238	0.237	0.037	0.201	0.042	0.029	0.009	0.013	0.060
Pu-239		239	0.007	0.007	0.002	0.008	0.007	0.000	0.004	0.009

3.3 U Isotopic Concentrations of Solids

The U isotopic concentrations measured in the solids are given in Table 3-12. The average U-235 enrichment in the unrinsed solids is slightly greater than 35 wt %. The U-235 enrichment of the U in the rinsed solids was significantly less, ranging from about 19 to 23 wt %. The solids contacted with 7 M HNO_3 acid show further evidence of the preferential dissolution of U-235 and U-236; however, the addition of fluoride to the 7 M HNO_3 solution results in a reduction in the U-238 concentration. Based on these data, it is apparent that solids containing different enrichment levels exist. The solids containing U at approximately 35 wt % U-235 enrichment are more soluble than the other solids in the dissolver; however, the addition of fluoride aids in dissolution of U in all solids.

Table 3-12. U Isotopic Concentrations in Dissolved Solids

	Isotope	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
		wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Solids	U-234	0.5	0.5	0.3	0.3	0.4	0.2	0.3	0.2	0.2	0.3
	U-235	35.1	35.2	21.8	22.7	21.2	19.5	23.1	18.0	18.9	25.1
	U-236	4.4	4.4	2.9	3.0	2.8	2.6	3.1	2.6	2.5	2.5
	U-238	60.1	59.9	75.0	74.0	75.7	77.7	73.5	79.1	78.4	72.1
Filtrate	U-234	-	-	0.5	0.5	0.6	0.6	0.6	0.5	0.5	0.3
	U-235	-	-	37.1	37.2	37.9	37.4	37.6	37.4	35.1	22.0
	U-236	-	-	4.7	4.7	4.3	4.7	4.8	4.7	4.4	3.2
	U-238	-	-	57.6	57.6	57.3	57.3	57.0	57.4	60.0	74.5

A review of the recent 6.4D dissolver SNF batch charging history shows the average U-235 content was approximately 35-36 wt %, which is consistent with the U concentrations measured in the unrinsed solids samples. Various MTR type fuels have been previously dissolved that initially contained approximately 19-20 wt % U-235 before irradiation which is consistent with the concentrations measured in the rinsed solids.

3.4 Fissionable Mass in Dissolved Solids

The analytical data in Table 3-10 and Table 3-11 show that contacting the 6.4D dissolver solids with near boiling HNO_3 (with and without fluoride) reduces the concentrations of the U and Pu isotopes. The concentrations of the fissionable isotopes following dissolution in 7 M HNO_3 with and without using 0.1 M fluoride are summarized in Table 3-13. The measured concentrations following the dissolution of the rinsed and unrinsed solids were averaged.

Table 3-13. Fissionable Concentration in Dissolved Solids

Isotope	Dissolution-7M	Dissolution-7M-F
	wt %	wt %
U-234	0.000	0.000
U-235	0.008	0.005
U-236	0.001	0.000
U-238	0.033	0.013
Total U	*0.042*	*0.018*
Pu-238	0.000	0.000
Pu-239	0.007	0.004
Pu-240	0.002	0.001
Total Pu	*0.009*	*0.005*

The concentrations measured in the 6.4D dissolver solids were used to calculate the masses of the U and Pu isotopes remaining in the solids following dissolution in the HNO_3 solutions. The average density of the dry and settled solids (Table 3-7) and the estimated volume of solids present on the bottom of the 6.4D dissolver pot[9] were used for the calculations. The 95% confidence limits in the masses of the fissionable isotopes were calculated by GUM Workbench[10] using propagation of error techniques based on the uncertainty of the ICPMS method (a one sigma uncertainty of 10%), the individually measured uncertainties of the Pu isotopes from the TTA/APHA method (a one sigma uncertainty of 5-10%), and the uncertainty in the average density of the solids. The volume of the solids in the 6.4D dissolver was a bounding value; therefore, an uncertainty was not used in the calculations. A summary of the estimated masses of fissionable isotopes remaining in the 6.4D dissolver following dissolution with 7 M HNO_3 and 7 M HNO_3 containing 0.1 M fluoride is given in Table 3-14.

Table 3-14. Mass of Fissionable Solids in the 6.4D Dissolver Following HNO_3 Dissolutions

Isotope	7 M HNO_3		7 M HNO_3/0.1 M Fluoride	
	Total Remaining Mass	Uncertainty	Total Remaining Mass	Uncertainty
	(g)	(g)	(g)	(g)
U-234	0.089	0.014	0.046	0.010
U-235	7.24	1.17	4.25	0.91
U-236	1.00	0.16	0.43	0.09
U-238	31.0	5.0	12.2	2.6
*Total U**	*39.4*	*5.5*	*16.9*	*2.9*
Pu-238	0.10	0.01	0.05	0.01
Pu-239	6.34	1.03	3.59	0.98
Pu-240	1.45	0.25	0.83	0.24
*Total Pu**	*7.89*	*1.13*	*4.46*	*1.12*

* Isotopic masses may not sum to the elemental total due to rounding.

The calculated masses of U and Pu in the 6.4D dissolver solids were substantially reduced when more aggressive dissolving conditions were used. The mass of U was reduced from 53-54 g (Table 3-8) in the samples rinsed with HNO_3 to 39 g in the solids when contacted with near boiling 7 M HNO_3. When the solids were contacted with near boiling 7 M HNO_3 containing 0.1 M fluoride, the total mass of U was further reduced to 17 g. Likewise the Pu mass in the solids was reduced from 9-10 g when rinsed with HNO_3 to 7.9 g and 4.5 g when contacted with near boiling 7 M HNO_3 and near boiling 7 M HNO_3 containing 0.1 M fluoride, respectively. Therefore, continued dissolution of SNF in the 6.4D dissolver (using starting HNO_3 concentrations of approximately 7 M) should slowly dissolve the very small quantities of U and Pu remaining in the dissolver and address any significant accumulation of fissile material in the future. A dissolver clean-out may be performed using nominally 7 M HNO_3 containing 0.1 M fluoride. The more

aggressive conditions would facilitate the removal of a higher percentage of the fissionable materials and reduce the amount of Si-containing solids in the 6.4D vessel.

4.0 Conclusions

Two solid samples taken from the 6.4D dissolver pot were transferred to SRNL for characterization. The samples were dried. The density of the dried solids was 0.6 ± 0.1 g/cm^3. A small portion of each sample set was removed, and rinsed with 1 or 6 M HNO_3. A portion of the unrinsed and rinsed solids was dissolved and analyzed for elemental and radiochemical concentrations. Results from the analyses indicated that the solids were primarily Si, Fe, Al, and Cr. Comparison of the analysis of the rinsed solids and filtrate to the analysis of unrinsed solids indicated that U was mostly soluble. Based on the estimated volume of solids present in the 6.4D dissolver (157,000 cm^3),[9] the bounding total masses of U and Pu present in the unrinsed solids were 359 ± 45 g (126 ± 20 g U-235) and 8.6 ± 1.4 g, respectively, at 95% confidence limits. The masses of U and Pu in the rinsed solids were 54 ± 6 g (12 ± 2 g of U-235) and 9.4 ± 1.1 g, respectively, at 95% confidence limits.

The calculated masses of U and Pu in the 6.4D dissolver solids were substantially reduced when more aggressive dissolving conditions were used. Contact with near boiling 7 M HNO_3 reduced the U and Pu masses to 39.4 ± 5.5 g (7.24 ± 1.17 g U-235) and 7.89 ± 1.13 g, respectively, at 95% confidence limits. When 0.1 M fluoride was added to the near boiling HNO_3, the remaining U and Pu masses were 16.9 ± 2.9 g (4.25 ± 0.91 g U-235) and 4.46 ± 1.12 g, respectively, at 95% confidence limits. The dissolution experiments also reduced the mass of the solids by 50-70% leaving primarily Si as SiO_2. Continued dissolution of SNF in the 6.4D dissolver (using starting HNO_3 concentrations of approximately 7 M) should slowly dissolve the very small quantities of U and Pu remaining in the dissolver and address any significant accumulation of fissile material in the future. As an alternative, a dissolver clean-out could be performed using nominally 7 M HNO_3 containing 0.1 M fluoride to reduce the total amounts of solids and fissionable materials. A periodic clean-out of the dissolver could also be considered. Based on this work, the H-Canyon SNF dissolution flowsheet is functioning as expected.

5.0 References

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Appendix A. Analytical Results

The ICPES analytical results from the analysis of the solutions following dissolution of unrinsed and rinsed solids are provided in Table A-1.

Table A-1. ICPES Analyses of HCA-North and HCA-South Solids

Element	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Ag	< 0.017	< 0.015	< 0.015	< 0.012	< 0.101	< 0.017	< 0.013
Al	4.88	5.15	0.337	0.357	0.388	0.314	0.386
B	< 0.018	< 0.016	< 0.016	< 0.013	< 0.129	< 0.018	< 0.014
Ba	< 0.008	< 0.007	< 0.007	< 0.006	< 0.025	< 0.008	< 0.006
Be	0.000	0.000	0.000	0.000	0.003	0.000	0.000
Ca	< 0.380	< 0.323	< 0.311	< 0.251	< 0.481	< 0.357	< 0.262
Cd	< 0.015	< 0.013	< 0.013	< 0.011	< 0.099	< 0.015	< 0.011
Ce	< 0.039	< 0.035	< 0.035	< 0.028	< 0.307	< 0.039	< 0.029
Co	< 0.015	< 0.013	< 0.014	< 0.011	< 0.107	< 0.015	< 0.011
Cr	1.340	0.347	0.508	0.164	0.176	0.475	0.205
Cu	< 0.050	< 0.045	< 0.045	< 0.037	< 0.360	< 0.051	< 0.038
Fe	4.95	1.31	1.90	0.642	0.717	1.79	0.831
Gd	< 0.032	< 0.029	< 0.029	< 0.024	< 0.076	< 0.033	< 0.024
K	< 1.200	< 1.070	< 1.080%	< 0.876	< 1.940	< 1.210	< 0.906
La	< 0.011	< 0.010	< 0.010	< 0.008	< 0.060	< 0.011	< 0.008
Li	< 0.014	< 0.013	< 0.013	< 0.011	< 0.103	< 0.015	< 0.011
Mg	0.023	0.026	0.002	< 0.001	< 0.014	< 0.002	0.003
Mn	< 0.007	< 0.006	< 0.006	< 0.005	< 0.014	< 0.007	< 0.005
Mo	0.087	< 0.071	< 0.071	< 0.058	< 0.307	< 0.080	< 0.060
Ni	0.733	0.193	0.261	0.083	< 0.169	0.260	0.117
P	< 0.192	< 0.171	< 0.172	< 0.140	< 1.370	< 0.194	< 0.145
Pb	< 0.191	< 0.171	< 0.171	< 0.139	-	< 0.193	< 0.144
S	< 10.0	< 10.0	< 10.0	< 8.73	-	< 10.0	< 9.04
Sb	< 0.201	< 0.179	< 0.180	< 0.146	< 1.440	< 0.203	< 0.151
Si	18.8	26.2	32.2	27.6	33.2	31.2	28.6
Sn	< 0.121	< 0.108	< 0.108	< 0.088	< 0.863	< 0.122	< 0.091
Sr	< 0.090	< 0.081	< 0.081	< 0.066	< 0.007	< 0.091	< 0.068
Th	< 0.005	< 0.004	< 0.004	< 0.003	< 0.033	< 0.005	< 0.003
Ti	< 0.043	< 0.038	< 0.038	< 0.031	< 0.306	< 0.043	< 0.032
U	< 0.198	< 0.176	< 0.177	< 0.144	< 1.410	< 0.199	< 0.149
V	< 0.006	< 0.006	< 0.006	< 0.005	< 0.040	< 0.007	< 0.005
Zn	< 0.010	< 0.009	< 0.009	< 0.007	< 0.039	< 0.010	< 0.008

The ICPES analytical results from the analysis of the solutions generated from the dissolution of unrinsed and rinsed solids remaining from contact with near boiling 7 M HNO_3 solutions are provided in Table A-2.

Table A-2. ICPES Analysis for Dissolution of Solids

Element	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
	wt %	wt %	wt %
Ag	< 0.074	< 0.084	< 0.018
Al	0.460	0.448	0.329
B	< 0.078	< 0.089	-
Ba	0.020	0.025	< 0.004
Be	< 0.003	< 0.004	0.000
Ca	0.432	0.509	< 0.351
Cd	< 0.060	< 0.068	< 0.015
Ce	< 0.169	< 0.192	< 0.045
Co	< 0.090	< 0.103	< 0.016
Cu	< 0.004	< 0.248	< 0.053
Cr	0.080	0.073	< 0.018
Fe	0.337	0.354	< 0.086
Gd	< 0.046	< 0.052	< 0.011
K	1.17	< 1.34	< 0.500
La	< 0.047	< 0.053	0.011
Li	< 0.147	< 0.167	0.015
Mg	< 0.010	< 0.011	0.030
Mn	< 0.008	< 0.010	< 0.002
Na	-	-	-
Ni	< 0.421	< 0.478	< 0.059
Mo	< 0.186	< 0.211	< 0.045
P	< 0.832	< 0.946	< 0.202
Pb	< 0.832	< 0.946	< 0.201
S	< 52.2	< 59.4	< 10.0
Sb	< 1.70	< 1.94	< 0.211
Si	20.0	40.0	28.5
Sn	< 0.524	< 0.596	< 0.127
Sr	< 0.004	< 0.005	< 0.009
Th	< 0.357	< 0.451	< 0.055
Ti	< 0.358	< 0.407	< 0.049
U	1.76	< 2.00	< 0.426
V	< 0.028	< 0.032	< 0.007
Zn	< 0.027	< 0.037	< 0.006
Zr	-	-	-
Pu-239	0.007	0.007	0.004
Pu-240	0.002	0.002	0.001

The ICPES analytical results from the analysis of the filtrate solutions generated from rinsing the solids in 1 and 6 M HNO_3 and dissolution of unrinsed and rinsed solids in near boiling 7 M HNO_3 are provided in Table A-3.

Table A-3. ICPES Analysis of Rinse Filtrate

Element	North-1M	South-1M	North-6M	South-6M	Unrinsed-7M	Rinsed-7M
	wt %	wt %	wt %	wt %	wt %	wt %
Ag	< 0.010	< 0.011	< 0.012	< 0.009	< 0.022	< 0.022
Al	12.6	15.0	10.2	10.5	9.133	0.265
B	< 0.013	< 0.014	< 0.015	< 0.012	< 0.023	< 0.023
Ba	0.012	0.015	0.010	0.011	0.010	< 0.001
Be	0.000	0.000	0.000	0.000	0.001	< 0.001
Ca	0.055	0.048	0.045	0.039	0.157	0.132
Cd	< 0.010	< 0.011	< 0.011	< 0.009	< 0.018	< 0.018
Ce	< 0.026	< 0.028	< 0.031	< 0.024	< 0.049	< 0.050
Co	< 0.011	< 0.012	< 0.012	< 0.010	< 0.026	< 0.027
Cr	0.321	0.377	0.257	0.258	0.283	0.059
Cu	< 0.035	< 0.039	< 0.042	< 0.033	< 0.064	< 0.065
Fe	1.135	1.348	0.894	0.854	1.063	0.300
Gd	< 0.023	< 0.025	< 0.027	< 0.021	0.093	< 0.014
K	< 0.190	< 0.209	< 0.225	< 0.179	< 0.344	< 0.348
La	< 0.008	0.010	< 0.009	0.008	< 0.014	< 0.014
Li	< 0.010	< 0.011	< 0.012	< 0.010	< 0.043	< 0.044
Mg	< 0.079	< 0.087	< 0.094	< 0.075	0.093	0.033
Mn	0.033	0.040	0.026	0.028	0.027	0.003
Mo	< 0.030	< 0.033	< 0.036	< 0.028	< 0.055	< 0.055
Na	< 0.157	< 0.172	< 0.186	< 0.148	0.115	0.226
Ni	0.173	0.214	0.142	0.149	0.145	< 0.124
P	< 0.134	< 0.148	< 0.159	< 0.127	< 0.244	< 0.246
Pb	< 0.134	< 0.147	< 0.158	< 0.126	< 0.244	< 0.246
S	< 8.39	< 9.22	< 9.94	< 7.93	< 15.29	< 15.45
Sb	< 0.141	< 0.155	< 0.167	< 0.133	< 0.499	< 0.504
Si	< 0.065	< 0.071	< 0.076	< 0.061	0.119	0.168
Sn	< 0.085	< 0.093	< 0.100	< 0.080	< 0.153	< 0.155
Sr	0.006	0.008	0.005	0.005	0.005	< 0.001
Th	< 0.008	< 0.008	< 0.009	< 0.007	< 0.116	< 0.117
Ti	< 0.030	< 0.033	< 0.036	< 0.028	< 0.105	< 0.106
U	0.858	1.156	0.657	0.811	0.696	< 0.520
V	< 0.004	< 0.004	< 0.005	< 0.004	< 0.008	< 0.008
Zn	< 0.015	< 0.016	< 0.017	< 0.014	0.013	0.008
Zr	0.061	0.089	0.091	0.091	0.187	0.173

The ICPMS analytical results from the analysis of the solutions following dissolution of unrinsed and rinsed solids are provided in Table A-4.

Table A-4. ICPMS Analysis of Unrinsed and Rinsed Solids

Most Likely Isotope	Possible Other Isotopes	m/z	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %	wt %	wt %
Ni-59	Co-59	59	0.004	0.001	0.002	0.001	0.001	0.002	0.001
		82	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		84	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Rb-85		85	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		86	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Rb-87		87	0.001	0.001	0.000	0.000	0.000	0.001	0.001
Sr-88		88	0.003	0.002	0.002	0.001	0.002	0.002	0.001
Y-89		89	0.001	0.002	0.000	0.000	0.000	0.000	0.000
Nb-93	Zr-93 Mo-93	93	0.164	0.168	0.212	0.197	0.184	0.207	0.212
Mo-95		95	0.024	0.013	0.018	0.011	0.011	0.016	0.013
Mo-96	Zr-96 Ru-96	96	0.444	0.464	0.382	0.401	0.369	0.45	0.397
Mo-97	Tc-97	97	0.021	0.011	0.017	0.01	0.01	0.014	0.012
Mo-98	Ru-98 Tc-98	98	0.025	0.013	0.018	0.011	0.011	0.016	0.013
Ru-100 Mo-100		100	0.022	0.012	0.018	0.011	0.01	0.015	0.013
Ru-101	Rh-101	101	0.005	0.009	0.007	0.009	0.009	0.01	0.01
Ru-99	Tc-99 Mo-99	99	0.164	0.168	0.212	0.197	0.184	0.207	0.212
Ru-102	Pd-102	102	0.004	0.008	0.007	0.008	0.008	0.009	0.009
Rh-103		103	0.444	0.464	0.382	0.401	0.369	0.450	0.397
Ru- 104	Pd-104	104	0.021	0.012	0.017	0.010	0.010	0.014	0.012
		105	0.025	0.013	0.018	0.011	0.011	0.016	0.013
		106	0.002	0.003	0.001	0.001	0.001	0.001	0.001
Pd-107	Ag-107	107	0.022	0.013	0.018	0.011	0.010	0.015	0.013
		108	0.005	0.009	0.007	0.009	0.009	0.010	0.010
Ag-109	Cd-109	109	0.004	0.008	0.007	0.008	0.008	0.009	0.009
Pd-110	Cd-110 Ag-110	110	0.004	0.004	0.004	0.004	0.005	0.004	0.005
Cd-111		111	0.003	0.005	0.004	0.005	0.005	0.005	0.005
Cd-112	Sn-112	112	0.001	0.001	0.001	0.001	0.001	0.001	0.001
		113	0.009	0.009	0.006	0.007	0.007	0.008	0.007
Cd-114	Sn-114	114	0.003	0.003	0.002	0.003	0.010	0.003	0.003
Cd-116	Sn-116	116	0.003	0.003	0.002	0.002	0.003	0.003	0.002
Sn-117		117	0.000	0.000	0.000	0.000	0.007	0.000	0.000
Sn-118		118	0.004	0.004	0.003	0.003	0.003	0.004	0.003
Sn-119		119	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sn-120		120	0.002	0.002	0.001	0.001	0.001	0.001	0.001
Sb-121		121	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sn-122	Te-122	122	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sb-123	Te-123 Sn-123	123	0.002	0.003	0.003	0.002	0.003	0.003	0.002
Sn-124	Te-124	124	0.002	0.002	0.002	0.001	0.002	0.002	0.001
Te-125	Sb-125	125	0.004	0.005	0.004	0.003	0.005	0.005	0.003
Te-126	Sn-126	126	0.021	0.025	0.03	0.026	0.033	0.03	0.027
Te-128		128	0.005	0.006	0.002	0.002	0.002	0.002	0.002
Te-130	Ba-130	130	0.005	0.006	0.006	0.005	0.008	0.006	0.004
Cs-133	Ba-133	133	0.002	0.002	0.002	0.002	0.003	0.002	0.002
Cs-134	Ba-134	134	0.002	0.002	0.002	0.002	0.003	0.002	0.002
Ba-135		135	0.002	0.003	0.003	0.002	0.004	0.003	0.002
Ba-136		136	0.003	0.004	0.004	0.003	0.005	0.004	0.003
Cs-137	Ba-137	137	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ba-138	La-138 Ce-138	138	0.021	0.025	0.030	0.026	0.033	0.030	0.027

Most Likely Isotope	Possible Other Isotopes	m/z	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %	wt %	wt %
La-139		139	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ce-140		140	0.002	0.002	0.002	0.001	0.001	0.001	0.002
Pr-141		141	0.003	0.003	0.001	0.001	0.001	0.001	0.001
Ce-142	Nd-142	142	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Nd-143		143	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Nd-144	Ce-144	144	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Nd-145	Pm-145	145	0.002	0.003	0.001	0.000	0.001	0.001	0.001
Nd-146	Sm-146 Pm-146	146	0.003	0.004	0.001	0.000	0.002	0.001	0.000
Sm-147	Pm-147	147	0.003	0.003	0.000	0.000	0.000	0.000	0.000
Nd-148	Sm-148	148	0.003	0.003	0.000	0.000	0.000	0.000	0.000
Sm-149		149	0.002	0.003	0.000	0.000	0.000	0.000	0.000
Sm-150 Nd-150		150	0.000	0.003	0.003	0.000	0.000	0.000	0.000
Eu-151		151	0.002	0.002	0.000	0.000	0.000	0.000	0.000
Sm-152	Gd-152	152	0.003	0.003	0.000	0.000	0.000	0.000	0.000
Eu-153		153	0.002	0.002	0.000	0.000	0.000	0.000	0.000
Eu-154 Sm-154	Gd-154	154	0.001	0.002	0.000	0.000	0.000	0.000	0.000
Eu-155 Gd-155		155	0.010	0.001	0.001	0.000	0.000	0.000	0.000
Gd-156		156	0.009	0.011	0.001	0.001	0.001	0.001	0.001
Gd-157		157	0.001	0.001	0.000	0.000	0.000	0.000	0.000
Gd-158		158	0.011	0.013	0.001	0.001	0.001	0.001	0.001
Tb-159		159	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Gd-160		160	0.009	0.012	0.001	0.001	0.001	0.001	0.001
		161	0.001	0.001	0.000	0.000	0.000	0.000	0.000
		162	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		163	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		164	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		165	0.001	0.001	0.000	0.000	0.000	0.000	0.000
		166	0.006	0.008	0.001	0.001	0.000	0.001	0.001
		167	0.009	0.011	0.001	0.001	0.001	0.001	0.001
		168	0.007	0.008	0.001	0.001	0.000	0.001	0.001
		169	0.011	0.013	0.001	0.001	0.001	0.001	0.001
		170	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		171	0.009	0.012	0.001	0.001	0.001	0.001	0.001
		172	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		173	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		174	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		175	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Lu-176	Hf-176 Yb-176	176	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Hf-177		177	0.024	0.026	0.017	0.018	0.019	0.022	0.017
Hf-178		178	0.035	0.038	0.025	0.027	0.028	0.032	0.025
Hf-179	Ta-179	179	0.018	0.02	0.013	0.014	0.014	0.017	0.013
Hf-180	Ta-180 W-180	180	0.045	0.049	0.032	0.034	0.035	0.042	0.033
Ta-181		181	0.001	0.001	0.001	0.001	0.003	0.001	0.001
W-182		182	0.013	0.008	0.012	0.008	0.011	0.011	0.009
W-183		183	0.008	0.005	0.008	0.005	0.007	0.007	0.006
W-184		184	0.016	0.01	0.016	0.01	0.014	0.014	0.012
Re-185		185	0.000	0.000	0.000	0.000	0.000	0.000	0.000
W-186		186	0.014	0.008	0.014	0.009	0.012	0.012	0.01
Re-187		187	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ir-191		191	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ir-193		193	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Most Likely Isotope	Possible Other Isotopes	m/z	North-Unrinsed	South-Unrinsed	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %	wt %	wt %
		194	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		195	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Pt-196		196	0.001	0.001	0.000	0.000	0.000	0.001	0.000
Pt-198 Hg-198		198	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		203	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Hg-204	Ti-204 Pb204	204	0.000	0.000	0.000	0.000	0.000	0.000	0.000
		205	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Pb-206		206	0.007	0.007	0.005	0.005	0.005	0.006	0.005
Pb-207		207	0.024	0.026	0.017	0.018	0.019	0.022	0.017
Pb-208	Bi-208 Po-208	208	0.035	0.038	0.025	0.027	0.028	0.032	0.026
		230	0.018	0.020	0.013	0.014	0.014	0.017	0.013
		233	0.045	0.049	0.032	0.034	0.035	0.042	0.033
U-232	Th-232	232	0.01	0.01	0.02	0.016	0.019	0.02	0.02
U-234		234	0.001	0.001	0.001	0.001	0.003	0.001	0.001
U-235	Np-235	235	0.12	0.147	0.012	0.013	0.007	0.01	0.014
U-236	Np-236 Pu-236	236	0.015	0.018	0.002	0.002	0.001	0.001	0.002
Np-237		237	0.001	0.001	0.000	0.000	0.000	0.000	0.000
U-238	Pu-238	238	0.205	0.251	0.042	0.041	0.026	0.041	0.046
Pu-239		239	0.007	0.008	0.008	0.008	1.682	0.008	0.009
Pu-240		240	0.002	0.002	0.002	0.002	0.104	0.002	0.002
Pu-241	Am-241	241	0.013	0.008	0.012	0.008	0.011	0.011	0.009
Pu-242	Am-242	242	0.008	0.005	0.008	0.005	0.007	0.007	0.006
		243	0.016	0.010	0.016	0.010	0.014	0.014	0.012
		244	0.000	0.000	0.000	0.000	0.000	0.000	0.000

The ICPMS analytical results from the analysis of the solutions generated from the dissolution of unrinsed and rinsed solids remaining from contact with near boiling 7 M HNO_3 solutions are provided in Table A-5.

Table A-5. ICPMS Analysis for Dissolution of Solids

Most Likely Isotopes	Other Possible Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
Ni-59	Co-59	59	0.000	0.000	0.000
		82	0.000	0.000	0.000
		84	0.000	0.000	0.000
Rb-85		85	0.000	0.000	0.000
		86	0.000	0.000	0.000
Rb-87		87	0.000	0.000	0.000
Sr-88		88	0.002	0.003	0.001
Y-89		89	0.000	0.000	0.000
Nb-93	Zr-93 Mo-93	93	0.218	0.211	0.140
		94	-	-	-
Mo-95		95	0.010	0.007	0.004
Mo-96	Zr-96 Ru-96	96	-	-	-
Mo-97	Tc-97	97	0.009	0.006	0.004
Mo-98	Ru-98 Tc-98	98	0.010	0.007	0.004
Ru-99	Tc-99 Mo-99	99	0.218	0.211	0.140
Ru-100 Mo-100		100	0.001	0.001	0.004

Most Likely Isotopes	Other Possible Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
Ru-101	Rh-101	101	0.010	0.006	0.006
Ru-102	Pd-102	102	0.009	0.009	0.005
Rh-103		103	0.008	0.008	0.003
Ru- 104	Pd-104	104	0.005	0.004	0.003
		105	0.010	0.007	0.004
		106	0.014	0.019	0.010
Pd-107	Ag-107	107	0.010	0.007	0.004
		108	0.005	0.007	0.010
Ag-109	Cd-109	109	0.008	0.008	0.005
Pd-110	Cd-110 Ag-110	110	0.005	0.004	0.003
Cd-111		111	0.005	0.004	0.003
Cd-112	Sn-112	112	0.001	0.001	0.000
		113	0.014	0.019	0.010
Cd-114	Sn-114	114	0.005	0.007	0.010
Cd-116	Sn-116	116	0.005	0.007	0.010
Sn-117		117	0.001	0.001	0.002
Sn-118		118	0.006	0.008	0.005
Sn-119		119	0.001	0.001	0.003
Sn-120		120	0.002	0.003	0.002
Sb-121		121	0.000	0.000	0.000
Sn-122	Te-122	122	0.000	0.000	0.000
Sb-123	Te-123 Sn-123	123	0.004	0.005	0.003
Sn-124	Te-124	124	0.002	0.003	0.002
Te-125	Sb-125	125	0.006	0.008	0.005
Te-126	Sn-126	126	0.026	0.032	0.027
Te-128		128	0.003	0.003	0.002
Te-130	Ba-130	130	0.008	0.011	0.006
Cs-133	Ba-133	133	0.002	0.003	0.003
Cs-134	Ba-134	134	0.003	0.004	0.002
Ba-135		135	0.002	0.004	0.003
Ba-136		136	0.005	0.006	0.004
Cs-137	Ba-137	137	0.000	0.000	0.000
Ba-138	La-138 Ce-138	138	0.026	0.032	0.027
La-139		139	0.000	0.000	0.000
Ce-140		140	0.001	0.001	0.000
Pr-141		141	0.000	0.000	0.000
Ce-142	Nd-142	142	0.000	0.000	0.000
Nd-143		143	0.000	0.000	0.000
Nd-144	Ce-144	144	0.000	0.000	0.000
Nd-145	Pm-145	145	0.000	0.000	0.000
Nd-146	Sm-146 Pm-146	146	0.001	0.001	0.001
Sm-147	Pm-147	147	0.000	0.000	0.000
Nd-148	Sm-148	148	0.000	0.000	0.000
Sm-149		149	0.000	0.000	0.000
Sm-150 Nd-150		150	0.000	0.000	0.000
Eu-151		151	0.000	0.000	0.000
Sm-152	Gd-152	152	0.000	0.000	0.000
Eu-153		153	0.000	0.000	0.000
Eu-154 Sm-154	Gd-154	154	0.000	0.000	0.000
Eu-155 Gd-155		155	0.000	0.000	0.000
Gd-156			0.009	0.000	0.000

Most Likely Isotopes	Other Possible Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
Gd-157		157	0.000	0.000	0.000
Gd-158		158	0.000	0.000	0.000
Tb-159		159	0.000	0.000	0.000
		161	0.000	0.000	0.000
		162	0.000	0.000	0.000
		163	0.000	0.000	0.000
		164	0.000	0.000	0.000
		165	0.000	0.000	0.000
		166	0.000	0.000	0.000
		167	0.001	0.000	0.000
		168	0.000	0.000	0.000
		169	0.001	0.001	0.001
		170	0.000	0.000	0.000
		171	0.001	0.001	0.001
		172	0.000	0.000	0.000
		173	0.000	0.000	0.000
		174	0.000	0.000	0.000
		175	0.000	0.000	0.000
Lu-176	Hf-176 Yb-176	176	0.010	0.014	0.006
Hf-177		177	0.038	0.053	0.023
Hf-178		178	0.057	0.079	0.033
Hf-179	Ta-179	179	0.027	0.038	0.017
Hf-180	Ta-180 W-180	180	0.074	0.100	0.043
W-182		182	0.012	0.009	0.004
W-183		183	0.007	0.005	0.003
W-184		184	0.015	0.011	0.006
Re-185		185	0.000	0.000	0.000
W-186		186	< 0.013	0.009	0.005
Re-187		187	0.000	0.000	0.000
Ir-191		191	0.000	0.000	0.000
Ir-193		193	0.000	0.000	0.000
		194	0.000	0.000	0.000
		195	0.000	0.000	0.000
Pt-198 Hg-198		198	0.001	0.000	0.000
		203	0.000	0.000	0.000
Hg-204	Ti-204 Pb204	204	0.000	0.000	0.000
		205	0.000	0.000	0.000
Pb-206		206	0.010	0.014	0.006
Pb-207		207	0.038	0.053	0.023
Pb-208	Bi-208 Po-208	208	0.057	0.079	0.033
		230	0.027	0.038	0.017
U-232 Th-232		232	0.018	0.021	0.021
		233	0.074	0.100	0.043
U-234		234	0.001	0.001	0.003
U-235	Np-235	235	0.008	0.007	0.005
U-236	Np-236 Pu-236	236	0.001	0.001	0.000
U-238	Pu-238	238	0.0037	0.029	0.013
Pu-239		239	0.007	0.007	0.004
Pu-241	Am-241	241	0.012	0.009	0.004
Pu-242	Am-242	242	0.007	0.005	0.003
		243	0.015	0.011	0.006

Most Likely Isotopes	Other Possible Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
		244	0.000	0.000	0.000

The ICPMS analytical results from the analysis of the filtrate solutions generated from rinsing the solids in 1 and 6 M HNO_3 ae provided in Table A-6.

Table A-6. ICPMS Analysis of Filtrate Solution from HNO_3 Rinses

Most Likely Isotope	Possible Other Isotopes	m/z	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %
Ni-59	Co-59	59	0.001	0.002	0.000	0.001	0.001
		82	0.000	0.000	0.000	0.000	0.000
		84	0.000	0.000	0.000	0.000	0.000
Rb-85		85	0.001	0.001	0.000	0.000	0.000
		86	0.000	0.000	0.000	0.000	0.000
Rb-87		87	0.001	0.002	0.000	0.001	0.001
Sr-88		88	0.003	0.004	0.000	0.002	0.002
Y-89		89	0.003	0.004	0.000	0.003	0.003
Zr-90	Sr-90	90	0.006	0.009	0.000	0.008	0.008
Zr-91	Nb-91 Y-91	91	0.012	0.017	0.000	0.017	0.017
Zr-92	Nb-92 Mo-92	92	0.011	0.016	0.000	0.015	0.016
Nb-93	Zr-93 Mo-93	93	0.014	0.020	0.005	0.019	0.019
Zr-94	Mo-94 Nb-94	94	0.012	0.018	0.000	0.017	0.018
Mo-95		95	0.004	0.006	0.000	0.004	0.004
Mo-96	Zr-96 Ru-96	96	0.011	0.016	0.004	0.015	0.016
Mo-97	Tc-97	97	0.004	0.006	0.000	0.003	0.004
Mo-98	Ru-98 Tc-98	98	0.004	0.006	0.000	0.004	0.004
Ru-99	Tc-99 Mo-99	99	0.004	0.005	0.000	0.003	0.003
Ru-100 Mo-100		100	0.004	0.006	0.000	0.004	0.004
Ru-101	Rh-101	101	0.004	0.005	0.000	0.003	0.004
Ru-102	Pd-102	102	0.003	0.005	0.000	0.003	0.003
Rh-103		103	0.002	0.003	0.000	0.002	0.002
Ru-104		104	0.002	0.002	0.000	0.001	0.002
		105	0.000	0.000	0.000	0.000	0.000
		106	0.000	0.000	0.000	0.000	0.000
Pd-107	Ag-107	107	0.000	0.000	0.000	0.000	0.000
		108	0.000	0.000	0.000	0.000	0.000
Ag-109	Cd-109	109	0.000	BDL	0.000	0.000	0.000
Pd-110	Cd-110 Ag-110	110	0.000	0.000	0.000	0.000	0.000
Cd-111		111	0.000	0.000	0.000	0.000	0.000

Most Likely Isotope	Possible Other Isotopes	m/z	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %
Cd-112	Sn-112	112	0.000	0.000	0.000	0.000	0.000
Cd-114	Sn-114	114	0.000	0.000	0.000	0.000	0.000
Cd-116	Sn-116	116	0.000	0.000	0.000	0.000	0.000
Sn-117		117	0.000	0.000	0.000	0.000	0.000
Sn-118		118	0.001	0.001	0.000	0.001	0.001
Sn-119		119	0.009	0.013	0.000	0.007	0.009
Sn-120		120	0.000	0.000	0.000	0.000	0.000
Sb-121		121	0.000	0.000	0.000	0.000	0.000
Sn-122	Te-122	122	0.000	0.000	0.000	0.000	0.000
Sb-123	Te-123 Sn-123	123	0.000	0.000	0.000	0.000	0.000
Sn-124	Te-124	124	0.000	0.000	0.000	0.000	0.000
Te-125	Sb-125	125	0.000	0.000	0.000	0.000	0.000
Te-126	Sb-126	126	0.001	0.001	0.000	0.001	0.001
Te-128		128	0.000	0.000	0.000	0.000	0.000
Te-130	Ba-130	130	0.001	0.001	0.000	0.001	0.001
Cs-133	Ba-133	133	0.006	0.008	0.000	0.004	0.005
Cs-134	Ba-134	134	0.000	0.001	0.000	0.000	0.000
Ba-135		135	0.001	0.001	0.000	0.001	0.001
Ba-136		136	0.000	0.000	0.000	0.000	0.000
Cs-137	Ba-137	137	0.006%	0.008%	0.000%	0.004%	0.005%
Ba-138	La-138 Ce-138	138	0.008	0.010	0.000	0.007	0.007
La-139		139	0.007	0.010	0.000	0.006	0.007
Ce-140		140	0.007	0.009	0.000	0.006	0.007
Pr-141		141	0.006	0.009	0.000	0.005	0.007
Ce-142	Nd-142	142	0.007	0.010	0.000	0.006	0.007
Nd-143		143	0.006	0.008	0.000	0.004	0.005
Nd-144	Ce-144	144	0.008	0.010	0.000	0.006	0.007
Nd-145	Pm-145	145	0.004	0.006	0.000	0.003	0.004
Nd-146	Sm-146, Pm-146	146	0.004	0.005	0.000	0.003	0.004
Sm-147	Pm-147	147	0.002	0.003	0.000	0.002	0.002
Nd-148	Sm-148	148	0.002	0.003	0.000	0.002	0.002
Sm-149		149	0.000	0.000	0.000	0.000	0.000
Sm-150 Nd-150		150	0.002	0.003	0.000	0.002	0.002
Eu-151		151	0.000	0.000	0.000	0.000	0.000
Sm-152	Gd-152	152	0.001	0.001	0.000	0.001	0.001
Eu-153		153	0.000	0.000	0.000	0.000	0.000
Eu-154 Sm-154	Gd-154	154	0.003	0.004	0.000	0.002	0.003
Eu-155 Gd-155		155	0.017	0.023	0.000	0.013	0.016
Gd-156		156	0.024	0.031	0.000	0.018	0.022
Gd-157		157	0.018	0.024	0.000	0.014	0.017

Most Likely Isotope	Possible Other Isotopes	m/z	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %
Gd-158		158	0.029	0.038	0.000	0.022	0.026
Tb-159		159	0.000	0.000	0.000	0.000	0.000
Gd-160		160	0.026	0.034	0.000	0.020	0.024
Lu-176	Hf-176 Yb-176	176	0.000	0.000	0.000	0.000	0.000
Hf-177		177	0.000	0.000	0.000	0.000	0.000
Hf-178		178	0.000	0.000	0.000	0.000	0.000
Hf-179	Ta-179	179	0.000	0.000	0.000	0.000	0.000
Hf-180	Ta-180 W-180	180	0.000	0.000	0.000	0.000	0.000
Ta-181		181	0.000	0.000	0.000	0.000	0.000
W-182		182	0.000	0.000	0.000	0.000	0.000
W-183		183	0.000	0.000	0.000	0.000	0.000
W-184		184	0.000	0.000	0.000	0.000	0.000
Re-185		185	0.000	0.000	0.000	0.000	0.000
W-186		186	0.000	0.000	0.000	0.000	0.000
Re-187		187	0.000	0.000	0.000	0.000	0.000
Ir-191		191	0.000	0.000	0.000	0.000	0.000
Ir-193		193	0.000	0.000	0.000	0.000	0.000
		194	0.000	0.000	0.000	0.000	0.000
		195	0.000	0.000	0.000	0.000	0.000
Pt-196		196	0.001	0.001	0.000	0.001	0.001
Au-197		197	-	-	0.000	-	-
Pt-198 Hg-198		198	0.030	0.045	0.000	0.024	0.031
		203	0.000	0.000	0.000	0.000	0.000
Hg-204	Ti-204 Pb-201	204	0.017	0.025	0.000	0.014	0.018
		205	0.000	0.000	0.000	0.000	0.000
Pb-206		206	0.000	0.000	0.000	0.000	0.000
Pb-207		207	0.000	0.000	0.000	0.000	0.000
Pb-208	Bi-208 Po-208	208	0.000	0.001	0.000	0.000	0.000
		230	0.000	0.000	0.000	0.000	0.000
U-232	Th-232	232	0.000	0.000	0.000	0.000	0.000
		233	0.000	0.000	0.000	0.000	0.000
U-234		234	0.005	0.006	0.000	0.004	0.004
U-235		235	0.318	0.433	0.005	0.256	0.299
U-236	Np-236 Pu-236	236	0.040	0.055	0.001	0.032	0.038
Np-237		237	0.001	0.002	0.000	0.001	0.001
U-238	Pu-238	238	0.494	0.671	0.007	0.392	0.453
Pu-239		239	0.003	0.004	0.008	0.002	0.003
Pu-240		240	0.001	0.001	0.000	0.001	0.001
Pu-241	Am-241	241	0.000	0.001	0.000	0.000	0.000
Pu-242	Am-242	242	0.000	0.000	0.000	0.000	0.000

Most Likely Isotope	Possible Other Isotopes	m/z	North-1M	South-1M	South-1M-B	North-6M	South-6M
			wt %	wt %	wt %	wt %	wt %
		243	0.000	0.000	0.000	0.000	0.000
		244	0.000	0.000	0.000	0.000	0.000

The ICPMS analytical results from the analysis of the filtrate solutions generated from dissolution of unrinsed and rinsed solids remaining from contact with near boiling 7 M HNO_3 solutions in Table A-7.

Table A-7. ICPMS Analysis of Filtrate Solution from 7 M HNO_3 Dissolutions

Most Likely Isotope	Possible Other Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
Ni-59	Co-59	59	0.001	0.000	0.002
		82	0.000	0.000	0.000
		84	0.000	0.000	0.000
Rb-85		85	0.000	0.000	0.020
		86	0.000	0.000	0.000
Rb-87		87	0.001	0.000	0.008
Sr-88		88	0.001	0.000	0.001
Y-89		89	0.001	0.000	0.000
Rh-103		103	0.001	0.000	0.004
Ru-104		104	0.001	0.000	0.003
		105	0.000	0.000	0.000
		106	0.000	0.000	0.000
Pd-107	Ag-107	107	0.000	0.000	0.000
		108	0.000	0.000	0.000
Ag-109	Cd-109	109	0.000	0.000	0.000
Pd-110	Cd-110 Ag-110	110	0.000	0.000	0.000
Cd-111		111	0.000	BDL	0.000
Cd-112	Sn-112	112	0.000	0.000	0.001
Cd-114	Sn-114	114	0.000	0.000	0.000
Cd-116	Sn-116	116	0.000	0.000	0.001
Sn-117		117	0.000	0.000	0.001
Sn-118		118	0.001	0.000	0.002
Sn-119		119	0.000	0.000	0.002
Sn-120		120	0.000	0.000	0.003
Sb-121		121	0.000	0.000	0.002
Sn-122	Te-122	122	0.000	0.000	0.003
Sb-123	Te-123 Sn-123	123	0.000	0.000	0.003
Sn-124	Te-124	124	0.000	0.000	0.003
Te-125	Sb-125	125	0.000	0.000	0.020
Te-126	Sb-126	126	0.002	0.002	0.027
Te-128		128	0.000	0.000	0.000

Most Likely Isotope	Possible Other Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
Te-130	Ba-130	130	0.001	0.001	0.002
Cs-134	Ba-134	134	0.000	0.000	0.000
Ba-135		135	0.000	0.000	0.001
Ba-136		136	0.000	0.000	0.000
Sm-147	Pm-147	147	0.001	0.000	0.000
Nd-148	Sm-148	148	0.001	0.000	0.000
Sm-149		149	0.000	0.000	0.000
Sm-150 Nd-150		150	0.001	0.000	0.000
Eu-151		151	0.000	0.000	0.000
Sm-152	Gd-152	152	0.001	0.000	0.000
Eu-153		153	0.000	0.000	0.000
Tb-159		159	0.000	0.000	0.000
Lu-176	Hf-176 Yb-176	176	0.000	0.000	0.000
Hf-177		177	0.000	0.000	0.000
Hf-178		178	0.000	0.000	0.000
Hf-179	Ta-179	179	0.000	0.000	0.000
Hf-180	Ta-180 W-180	180	0.000	0.000	0.000
Ta-181		181	0.000	0.000	0.001
W-182		182	0.001	0.001	0.014
W-183		183	0.000	0.000	0.000
W-184		184	0.001	0.001	0.008
Re-185		185	0.000	0.000	0.000
W-186		186	0.001	0.001	0.016
Re-187		187	0.000	0.000	0.000
Ir-191		191	0.000	0.000	0.000
Ir-193		193	0.000	0.000	0.000
		194	0.000	0.000	0.000
		195	0.000	0.000	0.000
Pt-196		196	0.000	0.000	0.000
Au-197		197	0.000	0.000	0.000
		203	0.000	0.000	0.000
		205	0.000	0.000	0.000
Pb-206		206	0.000	0.000	0.000
Pb-207		207	0.000	0.000	0.000
Pb-208	Bi-208 Po-208	208	0.000	0.000	0.000
		230	0.000	0.000	0.000
U-232	Th-232	232	0.001	0.000	0.005
		233	0.000	0.000	0.002
Np-237		237	0.001	0.000	0.000
Pu-240		240	0.000	0.000	0.002
Pu-241	Am-241	241	0.000	0.000	0.000

Most Likely Isotope	Possible Other Isotopes	m/z	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
			wt %	wt %	wt %
Pu-242	Am-242	242	0.000	0.000	0.000
		243	0.000	0.000	0.000
		244	0.000	0.000	0.000

The radiochemical analysis for unrinsed solids and solids rinsed with 1 and 6 M HNO_3 are provided in Table A-8. The Pu isotopes were measured by initially performing a TTA extraction followed by APHA. All other isotopes were quantified by GPHA.

Table A-8. Radiochemical Analysis of Unrinsed and Rinsed Solids

Isotope	North	South	North-1M	South-1M	South-1M-B	North-6M	South-6M
	wt %	wt %	wt %	wt %	wt %	wt %	wt %
Co-60	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ru-103	0.000	-	-	0.000	-	-	0.000
Ru-106	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sb-125	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sb-126	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sn-126	0.002	0.003	0.003	0.002	0.005	0.003	0.002
Cs-134	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Cs-137	0.002	0.000	0.000	0.000	0.000	0.000	0.000
Ce-144	0.000	0.000	0.000	0.000	-	0.000	0.000
Eu-154	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Eu-155	0.000	0.000	0.000	0.000	-	0.000	0.000
Pu-238	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Pu-239	0.007	0.008	0.007	0.008	1.752	0.008	0.008
Pu-240	0.002	0.002	0.002	0.002	0.108	0.002	0.002
Am-241	0.000	0.000	0.000	0.000	0.000	0.000	0.000

The Pu isotopic concentrations in the solids contacted by near boiling 7 M HNO_3 are provided in Table A-9. The concentrations were measured by initially performing a TTA extraction followed by APHA.

Table A-9. Radiochemical Analysis of Solids Contacted with Near Boiling 7 M HNO_3

Isotope	Unrinsed-7M	Rinsed-7M	Rinsed-7M-F
	wt %	wt %	wt %
Pu-239	0.007	0.007	0.004
Pu-240	0.002	0.002	0.001

The radiochemical analysis for the filtrate solutions generated from rinsing the solids with 1 and 6 M HNO_3 are provided in Table A-10. The Pu isotopes were measured by initially performing a TTA extraction followed by APHA. All other isotopes were quantified by GPHA.

Table A-10. Radiochemical Analysis of Filtrates from Rinsing Solids in HNO_3

Isotope	North-1M	South-1M	South-1M-B	North-6M	South-6M
	wt %	wt %	wt %	wt %	wt %
Cs-134	0.000	0.000	BDL	0.000	0.000
Cs-137	0.000	0.000	0.000	0.000	0.000
Co-60	0.000	0.000	0.000	0.000	0.000
Ru-106	0.000	0.000	0.000	0.000	0.000
Sb-125	0.000	0.000	0.000	0.000	0.000
Sb-126	BDL	BDL	0.000	BDL	BDL
Sn-126	BDL	BDL	0.000	BDL	BDL
Ce-144	0.000	0.000	0.000	0.000	0.000
Eu-154	0.000	0.000	0.000	0.000	0.000
Eu-155	0.000	0.000	0.000	0.000	0.000
Pu-238	0.000	0.000	0.000	0.000	0.000
Pu-239	0.000	0.000	0.000	0.000	0.000
Pu-240	0.000	0.000	0.000	0.000	0.000
Am-241	0.000	0.000	0.000	0.000	0.000

BDL – below detection limit

The Pu isotopic concentrations in the filtrate solutions from contacting solids with near boiling 7 $MHNO_3$ are provided in Table A-11. The concentrations were measured by initially performing a TTA extraction followed by APHA.

Table A-11. Radiochemical Analysis of Filtrates from Contacting Solids with Near Boiling HNO_3

Isotope	Unrinsed -7M	Rinsed -7M	Rinsed -7M-F
	wt %	wt %	wt %
Pu-238	0.000	0.000	0.000
Pu-239	0.000	0.000	0.000
Pu-240	0.000	0.000	0.000

Distribution

alex.cozzi@srnl.doe.gov
david.crowley@srnl.doe.gov
c.diprete@srnl.doe.gov
a.fellinger@srnl.doe.gov
samuel.fink@srnl.doe.gov
connie.herman@srnl.doe.gov
dennis.jackson@srnl.doe.gov
patricia.lee@srnl.doe.gov
Joseph.Manna@srnl.doe.gov
john.mayer@srnl.doe.gov
daniel.mccabe@srnl.doe.gov
Gregg.Morgan@srnl.doe.gov
frank.pennebaker@srnl.doe.gov
Amy.Ramsey@srnl.doe.gov
William.Ramsey@SRNL.DOE.gov
michael.stone@srnl.doe.gov
Boyd.Wiedenman@srnl.doe.gov
philip.almond@srnl.doe.gov
daniel.jones@srnl.doe.gov
tommy.edwards@srnl.doe.gov
kim.crapse@srnl.doe.gov
gene.daniel@srs.doe.gov
jonathan.duffey@srnl.doe.gov
harris.eldridge@srnl.doe.gov
eddie.kyser@srnl.doe.gov
robert.pierce@srnl.doe.gov
tracy.rudisill@srnl.doe.gov
john.scogin@srnl.doe.gov
thomas.shehee@srnl.doe.gov
nicholas.karay@srnl.doe.gov
Records Administration (EDWS)
timothy.tice@srs.gov
steven.brown@srs.gov
brett.clinton@srs.gov
tara.smith@srnl.doe.gov
bill.clifton@srs.gov
perry.levin@srs.gov
hayley.williams@srs.gov
james.therrell@srs.gov
kenneth.burrows@srs.gov
kevin.usher@srs.gov
anthony.carraway@srs.gov
sara.shirley@srs.gov
jaclyn.fitzpatrick@srs.gov
stephen.garrison@srs.gov
anthony.carraway@srs.gov
tracy.stover@srs.gov
joshua.butler@srs.gov
Richard.Burns@srs.gov