Sister Rod Destructive Examinations (FY23)

Appendix D: Fission Gas, Fuel Burnup, and Fuel Isotopic Measurements

Spent Fuel and Waste Disposition

Prepared for
US Department of Energy

Spent Fuel and Waste Science and Technology

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SUMMARY

This report documents work performed under the Spent Fuel and Waste Disposition's Spent Fuel and Waste Science and Technology program for the US Department of Energy (DOE) Office of Nuclear Energy (NE). This work was performed to fulfill Level 2 Milestone M2SF-24OR010201024, "FY23 ORNL Testing on Sibling Pins," within work package SF-24OR01020102 and is an update to the work reported in M2SF-23OR010201024, M2SF-22OR010201047, M2SF-21OR010201032, M2SF-19ORO010201026, and M2SF-19OR010201028.

As a part of DOE NE High Burnup Spent Fuel Data Project, Oak Ridge National Laboratory (ORNL) is performing destructive examinations (DEs) of high burnup (HBU) (>45 GWd/MTU) spent nuclear fuel (SNF) rods from the North Anna Nuclear Power Station operated by Dominion Energy. The SNF rods, called *sister rods* or *sibling rods*, are all HBU and include four different kinds of fuel rod cladding: standard Zircaloy-4 (Zirc-4), low-tin (LT) Zirc-4, ZIRLO, and M5. The DEs are being conducted to obtain a baseline of the HBU rod's condition before dry storage and are focused on understanding overall SNF rod strength and durability. Composite fuel and defueled cladding will be tested to derive material properties. Although the data generated can be used for multiple purposes, one primary goal for obtaining the post-irradiation examination data and the associated measured mechanical properties is to support SNF dry storage licensing and relicensing activities by (1) addressing identified knowledge gaps and (2) enhancing the technical basis for post-storage transportation, handling, and subsequent disposition.

This appendix documents the status of the ORNL Phase 1 DE activities related to fission gas sample analysis, fuel burnup analysis, and fuel isotopic analysis of selected sister specimens in Phase 1 of the sister rod test program.

Table DS-1 provides a summary of the DE status.

Table DS-1. DE Status.

Planned DE		Status	Comments
DE.01	Collect fission gas samples and analyze	Complete	Fission gas samples were collected and analyzed. Results are consistent with publicly available database. Code-predicted fission gas production is not available; therefore, the fission gas release ratio is not available. ORNL and Pacific Northwest National Laboratory (PNNL) fission gas analyses are consistent with one another, and the data are as expected when differences in fission gas partial pressure are considered.
DE.03	Analyze fuel burnup to confirm predicted and extrapolated values	In progress	Eighteen specimens were sent to the ORNL Radiochemical Engineering Development Center for analysis. Twelve are complete. The work is co-funded by DOE-NE and the US Nuclear Regulatory Commission.

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ACKNOWLEDGMENTS

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Thanks go to Tracy Binger and Mark Walls for their quick support when radiation protection coverage is required.

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REVISION HISTORY

Date	Changes
3/29/2019	Initial release
9/27/2019	Revised to include additional data and incorporate comments from the previously released report.
10/29/2020	The detailed information supporting the sister rod testing was moved into appendices to retain the information but reduce the amount of detail in the main report.
11/30/2020	The document numbering was revised to reflect its M2 status and the date was changed.
3/31/2022	The document number was revised for inclusion in the FY21 report and the date was changed.
10/31/2023	Updated to include the measured isotopic results for 9 specimens.
1/31/2024	Updated the document ID to reflect the M2 status.

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ACRONYMS

CRM: certified reference material

DE	destructive examination
DOE	US Department of Energy
EPRI	Electric Power Research Institute
FIMA	fissions per initial metal atom
FHT	full-length fuel rod heat treatment
FY	fiscal year
GUM	Guide to the expression of uncertainty in measurements
HBU	high burnup
HP-RCA	High Precision Radiochemical analysis
ID	Isotope Dilution
IFEL	Irradiated Fuels Examination Laboratory
IHM	Initial Heavy Metal
IRMM	Institute for Reference Materials and Measurements (renamed as part of the European science hub to the joint research center at Geel (JRC-Geel))
LT	low tin
MC-ICPMS	Multi-collector Inductively Coupled Plasma Mass Spectrometer
MID	multiple ion detection
NBL	New Brunswick Laboratory
NE	Office of Nuclear Energy
NIST	National Institute of Standards and Technology
NDE	nondestructive examination
ORNL	Oak Ridge National Laboratory
PWR	pressurized water reactor
REDC	Radiochemical Engineering Development Center
RGA	residual gas analyzer
RPC	Research Project Cask
SEM	secondary electron multiplier
SFWD	Spent Fuel and Waste Disposition
SFWST	Spent Fuel and Waste Science and Technology
SNF	spent nuclear fuel
WRM	working reference material

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D-1. Fission Gas Sample Isotopic Composition and Calculated Fission Gas Release

Before the system evacuation for the void volume measurement, a fission gas sample was taken from each punctured sister rod. Gaseous fission products evolve in all uranium dioxide (UO_2) nuclear fuel pellets at all axial elevations during reactor operation [D-1]. They are located near the site of the fission, within the fuel grains, at a grain boundary, or at free surfaces on the pellet. The gaseous fission products form small bubbles within the pellet since the Xe and Kr gases produced are virtually insoluble in UO_2. Although much of the fission gas remains trapped within the fuel pellet microstructure as porosity, a fraction of the fission gas is released to the interior void volume of the fuel rod and contributes to an increase in the internal pressure of the fuel rod [D-2][D-3].

According to EPRI, less than 5% of the fission gas produced in the pellet stack during normal operation is released to the void volume of the rod [D-4]. The quantity of fission gas released from the pellet to the void volume of the rod during reactor operation has been the topic of much study because the gross rod pressure and localized rod pressure are important to rod performance during reactor transients such as loss-of-coolant accidents and reactivity-initiated accidents. The percentage of fission gas released is calculated as the moles of fission gas in the rod void volume divided by the total calculated fission gas produced during operation.

The eight sister rod samples were analyzed by the Oak Ridge National Laboratory (ORNL) Nuclear Analytical Chemistry and Isotopic Laboratories Group. Fission gas isotopic (in atom %) and concentration (in mole%) determinations were made using an OmniStar GSD 320 residual gas analyzer (RGA) analyzer coupled to a sample manifold located within a radiological fume hood (Figure D-1). The ion source and focus lens tune parameters are listed in Table D-1.

Figure D-1. The sample inlet manifold (left) coupled to an OmniStar GSD 320 residual gas analyzer (RGA) (right).

Table D-1. Ion source and focus lens tune parameters for the RGA analyzer.

Parameter	Setting
Filament	No 1
Active set	Set 1
Emission current	0.50 A
Protection current	3.50 A
R_f-polarity	positive
Ion reference	150.00 V
Cathode	-50.00 V
Focus	-3.00 V
Field axis	-5.00 V
Extraction	-45.00 V

The RGA analyzer is equipped with two detectors, a Faraday detector that is used for all fission gas concentration measurements and a secondary electron multiplier (SEM) that can be used for low parts per million gas concentration measurements. In general, the Faraday detector signal is more stable and offers slightly higher precision isotope ratio measurements. The SEM was not used for sister rod concentration measurements, but it was used to confirm the Faraday isotopic measurements. Gas samples are scanned from 1 to 150 atomic mass unit (amu) with either detector prior to starting the isotopic and mole percent determinations. These analog scans are performed to detect potential unknown gasses present in samples, and to have a record of detected peak shape and peak resolution. The natural (using a gas standard) or fission isotopes (unknown samples) are measured using a multiple ion detection (MID) data acquisition program. Gas concentrations are then measured by quantification of ^{84}Kr, ^{132}Xe, and ^{4}He isotopes using a multiple concentration determination program.

Quality control measures involved the analysis of two certified gas standard mixtures with independent production lot numbers. The primary standard is employed for calibration of the OmniStar GSD 320 RGA and the secondary standard is analyzed for calibration verifications and continuing instrument performance. The setup and calibration process of the OmniStar GSD 320 Gas Analysis (RGA) system involves an offset calibration with the evacuated sample manifold pressure controlled at 4–5 Torr. A mass scale adjustment and RGA ion source tuning are then performed with the certified mixed gas standard held at ~30 Torr in the sample manifold. Mass scale tuning, which is required for performance optimization, is performed when necessary. The final stage in the RGA setup is a second mass scale adjustment using a fine mass calibration setting and the same standard gas. The result of the calibration is verified with the secondary mixed gas standard held at the same pressure.

To measure the sister rod fission gas samples, the sample bottle used in the rod puncture sequence (Appendix C) is attached to the evacuated sample manifold for the measurement sequence. The known volume of the sample manifold is 18.4 mL, and the volume and pressure of each sister rod gas sample is known. The sample gas expands into the manifold until gas pressure reaches equilibrium. The sample manifold pressure is recorded and then reduced to ~30 Torr before injecting the sample into the RGA for analysis. Argon gas (99.999% purity) is used to flush the sample manifold between gas analyses and for verification testing. NIST tracible mixed gas standards from independent lots are used for calibration and verification testing, and these contain a gas mixture that is 5.00 mol% Kr, 5.00 mol% Xe, 1.05 mol% O, 3.96 mol% N, and the remaining 85.00 mol% is He. The individual gas concentrations in the mixed gas standards are certified with a blend tolerance and certified accuracy of a relative ±2%, and the certified test gas standards used are listed in Table D-2.

Table D-2. Matheson certified verification standards used for sister rod fission gas analysis.

Calibration-	Lot No:	1028601078B	Exp.	4/20/2021
Verification-	Lot No:	1027603245B	Exp.	12/8/2020

Measured gas concentrations for the eight sister rod gas samples collected during the rod puncture operation (see Section 7.1) are provided in Table D-3. Six of the sister rod samples were measured as many as three times on non-consecutive days and the determined fission gas concentrations were averaged for those samples. The concentrations measured were determined by linear regression monitoring ^{84}Kr and ^{132}Xe, which are naturally occurring isotopes present at 56.99 and 26.91 atom%, respectively. The isotopic concentration in the sister rod samples was determined by measuring the current responses corresponding to the ^{84}Kr and ^{132}Xe isotopes and comparing those with the current response of the known concentration calibration standards. The total uncertainty values reported are the combined uncertainties of the duplicate measurements at a 95% level of confidence. The number of digits in the reported mole% and their uncertainties are provided for information and are not intended to convey a significant degree of reliability for the long-term performance of precision for the measurement.

Table D-3. Sister rod gas sample measured elemental composition, mole%*

	Sample ID			
Detected gas**	30AK09 (M5) SR-Gr-02	30AD05 (M5) SR-Gr-05	30AE14 (M5, heat treated) SR-Gr-06	3A1F05 (LT Zirc-4) SR-Gr-04
Kr	1.60 ± 0.15	1.41 ± 0.19	1.45 ± 0.22	1.97 ± 0.25
Xe	15.31 ± 1.33	14.10 ± 0.70	14.11 ± 1.49	18.46 ± 1.72
He***	83.09 ± 1.10	84.49 ± 0.59	84.44 ± 1.62	79.57 ± 1.83

	Sample ID			
Detected gas**	6U3K09 (ZIRLO) SR-Gr-01	3D8E14 (ZIRLO) SR-Gr-03	3F9N05 (ZIRLO, heat treated) SR-Gr-07	F35P17 (Zirc-4, heat treated) SR-Gr-08
Kr	1.11 ± 0.10	2.36 ± 0.30	2.23 ± 0.30	1.93 ± 0.25
Xe	10.45 ± 1.47	22.44 ± 1.41	20.08 ± 2.01	19.87 ± 1.99
He***	88.44 ± 1.41	75.20 ± 1.41	77.69 ± 1.62	78.20 ± 1.62

* Reported uncertainties are the total combined uncertainties at the 95% level of confidence. Two decimal places are provided in the reported values for information only and are not intended to imply a significant degree of reliability. The precision contribution for samples 01-06 was the standard deviation of the values measured in August 2018 and September 2018. Because only a single data set was measured for samples 07 and 08, for conservatism, the precision contribution to the total uncertainty for those data was taken as the worst-case scenario observed for samples 01–06.

** Some residual air present in the sampling system was detected and the resulting O and N content has been neglected when determining the fission gas component percentages and fission gas release in the fuel rod.

***The measured He includes the pre-pressurization He and any He produced as fission/decay products.

Isotopic data, reported in Table D-4 and Table D-5 , were determined using the ion currents acquired from the MID run and includes both natural and fission product Kr and Xe isotopes. The isotopic composition of natural Kr and Xe in both primary and secondary standards were evaluated and compared with isotopic data sourced from the National Nuclear Data Center [D-5] to ensure analytical accuracy. The uncertainties reported in Table D-4 are a single standard deviation from the calculated mean, where $n = 2–4$, except for SR-Gr-07 and -08 in which only one measurement was completed. For those two samples, the averaged observed standard deviation for the other six samples was applied. The uncertainties reported in Table D-5

are 2σ standard deviations from the calculated mean, where n = 2–4, except for SR-Gr-07 and -08 in which only one measurement was completed. For those two samples, the largest observed uncertainty from the other six samples was applied. The number of digits in the reported atom% and their uncertainties are provided for information and are not intended to convey a significant degree of reliability. Some small bias in the measurement data was observed but was within the measurement uncertainty; no corrections were applied. Additionally, three independent measurements of selected sister rod samples were performed on three different dates between August 2018 and February 2019. The Faraday detector was used to generate most of the isotopic data; however, the SEM was also used for three of the fission gas samples in February 2019 to verify the precision of the Faraday measurements. The results of the three SEM measurements were incorporated into the uncertainty calculations for the measured samples.

Table D-4. Fission gas isotope ratios, atom% ratio

Sample	30AK09 (M5)			30AD05 (M5)			30AE14 (M5, heat treated)			3A1F05 (LT Zirc-4)		
Isotope Ratio	SP-Gr-02*			SP-Gr-05*			SP-Gr-06*			SP-Gr-04*		
$^{82}Kr/^{84}Kr$	0.021	+/-	0.009	0.018	+/-	0.007	0.019	+/-	0.008	0.023	+/-	0.003
$^{83}Kr/^{84}Kr$	0.322	+/-	0.009	0.307	+/-	0.008	0.318	+/-	0.010	0.310	+/-	0.002
$^{85}Kr/^{84}Kr$***	0.121	+/-	0.007	0.121	+/-	0.008	0.123	+/-	0.009	0.059	+/-	0.003
$^{86}Kr/^{84}Kr$	1.533	+/-	0.014	1.525	+/-	0.009	1.535	+/-	0.011	1.512	+/-	0.000
$^{128}Xe/^{132}Xe$	0.005	+/-	0.003	0.005	+/-	0.003	0.006	+/-	0.003	0.007	+/-	0.004
$^{130}Xe/^{132}Xe$	0.012	+/-	0.002	0.012	+/-	0.002	0.013	+/-	0.001	0.015	+/-	0.001
$^{131}Xe/^{132}Xe$	0.278	+/-	0.004	0.289	+/-	0.003	0.296	+/-	0.002	0.293	+/-	0.004
$^{134}Xe/^{132}Xe$	1.178	+/-	0.006	1.173	+/-	0.022	1.178	+/-	0.011	1.165	+/-	0.033
$^{136}Xe/^{132}Xe$	1.689	+/-	0.029	1.661	+/-	0.055	1.654	+/-	0.029	1.647	+/-	0.080
Sample	6U3K09 (ZIRLO)			3D8E14 (ZIRLO)			3F9N05 (ZIRLO, heat treated)			F35P17 (Zirc-4, heat treated)		
Isotope Ratio	SP-Gr-01*			SP-Gr-03*			SP-Gr-07**			SP-Gr-08**		
$^{82}Kr/^{84}Kr$	0.016	+/-	0.005	0.022	+/-	0.004	0.033	+/-	0.010	0.034	+/-	0.011
$^{83}Kr/^{84}Kr$	0.311	+/-	0.005	0.277	+/-	0.001	0.310	+/-	0.006	0.278	+/-	0.005
$^{85}Kr/^{84}Kr$***	0.100	+/-	0.004	0.073	+/-	0.004	0.088	+/-	0.005	0.050	+/-	0.003
$^{86}Kr/^{84}Kr$	1.537	+/-	0.008	1.474	+/-	0.007	1.530	+/-	0.008	1.469	+/-	0.008
$^{128}Xe/^{132}Xe$	0.005	+/-	0.003	0.007	+/-	0.004	0.011	+/-	0.006	0.011	+/-	0.006
$^{130}Xe/^{132}Xe$	0.012	+/-	0.002	0.015	+/-	0.002	0.019	+/-	0.002	0.018	+/-	0.002
$^{131}Xe/^{132}Xe$	0.278	+/-	0.004	0.254	+/-	0.005	0.290	+/-	0.004	0.249	+/-	0.003
$^{134}Xe/^{132}Xe$	1.178	+/-	0.006	1.126	+/-	0.033	1.156	+/-	0.018	1.094	+/-	0.017
$^{136}Xe/^{132}Xe$	1.689	+/-	0.029	1.582	+/-	0.083	1.600	+/-	0.048	1.545	+/-	0.046

* Uncertainty for samples defined as a 1σ external standard deviation of the replicate analyses (for 01, 03, and 04, n = 2; for 02, 05, and 06, n = 4).

** For SP-Gr-07 and -08, only one replicate was performed; the assigned uncertainties are the averages of the other six samples.

*** ^{85}Kr was decay-corrected to February 2019 in each case.

Table D-5. Fission gas isotopic composition, atom %*

Sample	30AK09 (M5)	30AD05 (M5)	30AE14 (M5, heat treated)	3A1F05 (LT Zirc-4)
Isotope	SP-Gr-02*	SP-Gr-05*	SP-Gr-06*	SP-Gr-04*
^{82}Kr	0.69 ± 0.34	0.59 ± 0.30	0.64 ± 0.32	0.81 ± 0.40
^{83}Kr	10.73 ± 0.32	10.34 ± 0.31	10.62 ± 0.32	10.67 ± 0.32
^{84}Kr	33.37 ± 0.67	33.65 ± 0.67	33.39 ± 0.67	34.43 ± 0.69
^{85}Kr****	4.04 ± 0.20	4.08 ± 0.20	4.12 ± 0.21	2.03 ± 0.10
^{86}Kr	51.17 ± 0.51	51.33 ± 0.51	51.24 ± 0.51	52.06 ± 0.52
^{128}Xe	0.14 ± 0.07	0.13 ± 0.07	0.17 ± 0.09	0.16 ± 0.08
^{129}Xe	< 0.05	< 0.05	< 0.05	< 0.05
^{130}Xe	0.31 ± 0.16	0.30 ± 0.15	0.39 ± 0.19	0.36 ± 0.18
^{131}Xe	7.09 ± 0.35	6.98 ± 0.35	6.38 ± 0.32	7.11 ± 0.36
^{132}Xe	23.97 ± 0.48	24.16 ± 0.48	25.14 ± 0.50	24.24 ± 0.48
^{134}Xe	28.39 ± 0.57	28.33 ± 0.57	28.30 ± 0.57	28.23 ± 0.56
^{136}Xe	40.15 ± 0.40	40.10 ± 0.40	39.74 ± 0.40	39.90 ± 0.40
Sample	**6U3K09 (ZIRLO)**	**3D8E14 (ZIRLO)**	**3F9N05 (ZIRLO, heat treated)**	**F35P17 (Zirc-4, heat treated)**
Isotope	**SP-Gr-01***	**SP-Gr-03***	**SP-Gr-07****	**SP-Gr-08****
^{82}Kr	0.56 ± 0.28	0.77 ± 0.39	1.10 ± 0.55	1.20 ± 0.60
^{83}Kr	10.49 ± 0.31	9.75 ± 0.29	10.45 ± 0.31	9.83 ± 0.29
^{84}Kr	33.74 ± 0.67	35.14 ± 0.70	33.74 ± 0.67	35.30 ± 0.71
^{85}Kr****	3.36 ± 0.17	2.56 ± 0.13	3.06 ± 0.15	1.81 ± 0.09
^{86}Kr	51.85 ± 0.52	51.78 ± 0.52	51.63 ± 0.52	51.86 ± 0.52
^{128}Xe	0.12 ± 0.06	0.17 ± 0.09	0.27 ± 0.20	0.28 ± 0.21
^{129}Xe	< 0.05	< 0.05	< 0.05	< 0.05
^{130}Xe	0.29 ± 0.15	0.39 ± 0.19	0.46 ± 0.23	0.45 ± 0.23
^{131}Xe	6.68 ± 0.33	6.38 ± 0.32	7.12 ± 0.36	6.36 ± 0.32
^{132}Xe	24.03 ± 0.48	25.14 ± 0.50	24.54 ± 0.49	25.54 ± 0.51
^{134}Xe	28.31 ± 0.57	28.30 ± 0.57	28.36 ± 0.57	27.92 ± 0.56
^{136}Xe	40.57 ± 0.41	39.74 ± 0.40	39.25 ± 0.39	39.44 ± 0.39

* Reported numerical uncertainties are the 2σ external standard deviation of all duplicate analyses. The last digit in the measurements and uncertainties is provided for information and is not intended to convey a significant degree of reliability. The accuracy of the analysis was confirmed using a NIST tracible standard, and a bias correction did not measurably alter the data within the uncertainty of the 2σ standard deviation.

** SR-Gr-02, -05, and -06 also incorporate the uncertainty between two different modes of mass analysis, namely SEM and Faraday.

*** For SR-Gr-07 and -08, only one replicate was analyzed; for conservatism, the uncertainty attributed to those data was taken as the worst-case scenario observed for samples 01–06.

**** ^{85}Kr was decay-corrected to February 2019 in each case.

D-2. Fuel Burnup and Isotopic Measurements

Eighteen PWR samples were selected from the sister rods, as listed in Table D-6. Measurements of the samples are possible as a result of joint support from DOE-NE and the NRC under two different projects.

Three specimens are analyzed for chemical determination of burnup (called *burnup only*) as discussed in Section D2.1 and the remainder are analyzed for an expanded list of isotopes (Table D-7), called *full-isotopic* herein. The three samples selected for burnup-only measurements have operator-reported burnups of 41, 57, and 64 GWd/MTU. The samples planned for full-isotopic analysis cover a range of burnups from 53 to 66 GWd/MTU. They cover a cooling time from 12 to 33 years and an initial fuel enrichment ranging from 3.6 to 4.55 wt% ^{235}U. Six of the selected samples have operator-reported burnups greater than 60 GWd/t. Their addition to the current validation basis significantly increases the number of PWR samples available for this high-burnup range. For example, Figure D-2 presents the burnup range covered by the publicly available data provided in the SFCOMPO international database [D-16] maintained by OECD/NEA [D-17]]. This figure illustrates the scarcity of measurement data available for higher burnups. Approximately two-thirds of the illustrated samples have burnups lower than 40 GWd/t, and only 5 measured samples in the database are present for burnups above 60 GWd/t. Moreover, it was emphasized that limited high-quality measurement data are available for actinides and fission products relevant to spent fuel transportation, storage, and disposal [D-17]

Table D-6. List of specimens Selected for fuel burnup and isotopic measurements.

Rod	Originating segment elevation range (mm)		Estimated[a] burnup (GWd/MTU)	Analysis performed	Sample length (mm)	Status
30AD05	1280	1299	59	Full isotopic	11.7	To be dissolved
30AD05	0200	0215	44	Full isotopic	15.0	Complete
30AD05	2410	2429	59	Full isotopic	9.2	To be dissolved
30AE14	2675	2694	61[b]	Full isotopic	8.9	Complete
3A1F05	0150	0165	40	Full isotopic	9.8	Complete
3A1F05	0225	0240	41	Full isotopic	9.8	Complete
3A1F05	2383	2402	53[c]	Full isotopic	9.8	To be dissolved
3D8E14	0700	0719	61	Burnup only	14.9	Complete
3D8E14	0775	0796	64	Full isotopic	21.0	Complete
3D8E14	2303	2322	62	Full isotopic	12.4	To be dissolved
3D8E14	2655	2674	64	Full isotopic	8.3	To be dissolved
3D8E14	3206	3225	59	Burnup only	8.6	Complete
3D8E14	1375	1450	64	Full isotopic	12.2	Complete
3F9N05	2300	2329	57	Full isotopic	10.3	To be dissolved
3F9N05	2863	2882	58	Full isotopic	9.1	To be dissolved
3F9N05	3593	3615	30	Full isotopic	22.0	Complete
F35P17	2383	2402	66	Full isotopic	9.0	Complete
F35P17	3050	3069	61	Full isotopic	19.0	Complete
6U3K09	3506	3525	43	Burnup only	12.4	Complete
Total number of specimens					**19**	

[a] The burnup estimates provided in Table D-6 are based upon the reported rod-average burnup as calculated by the utility and adjusted for rod elevation using the gamma scanning results for the rod reported by Montgomery et al. [D-22]. They differ slightly from the estimates provided by Ilas et al. [D-17] where noted.

[b] Estimated by Ilas et al. as 57.8 GWd/MTU [D-17].

[c] Estimated by Ilas et al. as 55.5 GWd/MTU [D-17].

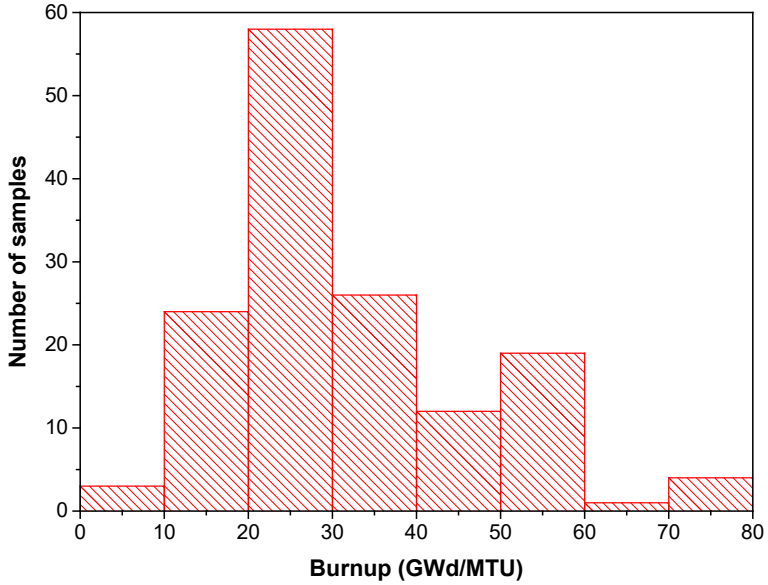


Figure D-2. Burnup distribution for existing PWR sample measurements [D-17].

The experiments performed and corresponding results are summarized in the following sections.

D2.1 Separation and Measurement Techniques for Burnup-Only Specimens

Chemical separation is achieved using ion chromatography using oxalic acid, diglycolic acid, and hydrochloric acid. A method called RAPID (Rapid Analysis of Post-Irradiation Debris) [D-6][D-5][D-7][D-8],[D-9][D-10], originally developed for the Defense Threat Reduction Agency (DTRA), has been modified for the successful separation of and isolation of all activation and fission isotopes, including ^{125}Sb, ^{95}Mo, ^{99}Tc, and ^{109}Ag. RAPID, when used in combination with isotope dilution mass spectrometry (IDMS) can yield direct isotopic compositions and concentrations with uncertainties as low as 2% at the 95% confidence level using a single detector inductively coupled plasma mass spectrometer (ICP-MS). When coupled with elemental isolation via fraction collection followed by analysis using a multi-collector ICP-MS isotopic the IDMS-RAPID protocol can yield isotopic concentrations with uncertainties as low as 0.5% at the 95% confidence level.

ORNL has developed and qualified a comprehensive analytical protocol for high precision radiochemical analysis (HP-RCA) of key fission product and actinide isotopes in select specimens from the HBU fuels. Table D-7 provides a summary of the improvements in uncertainty afforded by the high precision approach as compared with the previous analysis approach and lists the isotopes to be analyzed. A key to the HP-RCA is the state-of-the-art MC-ICPMS system installed for use with radioactive samples. This instrument is capable of measuring isotope ratios with relative uncertainties on the order of 0.1%. A comprehensive Sample Analysis Plan for the HP-RCA measurements has been developed [D-11] and a working reference material (WRM) has been prepared using NIST and NBL certified reference materials (CRM). For the HP-RCA, spikes were created using enriched isotopes procured from the DOE's Isotope Business Offices with traceability established to NIST standards. The WRM will be processed in parallel with the fuels and used as a laboratory control sample (LCS) to document performance of the analytical protocol. A full list of standards and controls can be found in Appendix D in ORNL/TM-202/1657 [D-11].

Measurement protocols with GUM-compliant[1] calculations to report total uncertainties for the results were established for all nuclides listed in Table D-7.

ORNL is unable to measure ^{14}C; ^{36}Cl, ^{221}Fr with current analytical capabilities.

Table D-7. Isotopes analyzed using RAPID protocols.

Isotope	Separation and detection method	Relative uncertainty single detector ICPMS, 2σ* (prior best practice)	Relative uncertainty HP-RCA test method, 2σ*
^{79}Se	RAPID-IDMS with a ^{77}Se spike	2%	0.5%
^{95}Mo	RAPID-IDMS with a ^{94}Mo spike	-	0.5%
^{90}Sr	RAPID-IDMS with a ^{86}Sr spike	5%	0.5%
^{99}Tc	Standard addition using ^{99}Tc	-	2%
^{101}Ru	RAPID-IDMS using a ^{99}Ru spike	2%	0.5%
^{106}Ru			0.5%
^{103}Rh	Standard Addition	5%	-
^{109}Ag	Rapid-IDMS using a ^{107}Ag spike	-	1%
^{125}Sb	RAPID-IDMS using a ^{123}Sb spike	2%	0.5%
^{133}Cs	RAPID-IDMS using a ^{133}Cs spike	2%	0.5%
^{134}Cs			0.5%
^{135}Cs			0.5%
^{137}Cs**			0.5%
^{143}Nd	RAPID-IDMS using a ^{150}Nd spike	2%	0.5%
^{145}Nd			0.5%
^{146}Nd			0.5%
^{148}Nd**			0.5%
^{144}Nd			0.5%

[1] ISO/IEC GUIDE 98-3:2008, Guide to the expression of uncertainty in measurement.

Isotope	Separation and detection method	Relative uncertainty single detector ICPMS, 2σ* (prior best practice)	Relative uncertainty HP-RCA test method, 2σ*
^{144}Ce	RAPID-IDMS using a ^{140}Ce spike	2%	1.0%
^{147}Pm	RAPID-IDMS using ^{149}Sm and a semi-quantitative approach	3%	-
^{147}Sm	RAPID-IDMS using a ^{149}Sm spike	2%	0.5%
^{149}Sm			0.5%
^{150}Sm			0.5%
^{151}Sm			0.5%
^{152}Sm			0.5%
^{151}Eu	RAPID-IDMS using a ^{151}Eu spike	2%	0.5%
^{153}Eu			0.5%
^{154}Eu			0.5%
^{155}Eu			0.5%
^{155}Gd	RAPID-IDMS using ^{155}Gd	2%	0.5%
^{227}Ac	Weighted dilution gamma	5%	-
^{234}U	Davies-Grey titration or IDMS with ^{233}U	0.5% or 2%	0.1%
^{235}U**			0.1%
^{236}U			0.1%
^{238}U**			0.1%
^{237}Np	RAPID using external calibration	5%	-
^{238}Pu	RAPID-IDMS using CRM-130, a^{242}Pu STD	2%	0.1%
^{239}Pu**			0.1%
^{240}Pu**			0.1%
^{241}Pu**			0.1%
^{242}Pu			0.1%
^{241}Am	RAPID-IDMS using an ^{243}Am , *verification of 241 using gamma*	2%	0.1%
^{242}mAm			0.5%
^{243}Am			0.1%
^{242}Cm	RAPID IDMS with in-house certified Cm-WRM, or using ^{243}Am and a semiquantitative approach	2%	1.0%
^{243}Cm			1.0%
^{244}Cm			1.0%
^{245}Cm			1.0%
^{246}Cm			1.0%
^{247}Cm			1.0%

*Based on the isotopic concentration of 1 µg/g fuel

**Isotopes required for the determination of chemical burnup

Burnup is estimated based on the measurement data using ASTM-321E standard methods [D-12], relying on ^{148}Nd as a burnup indicator. The sample's burnup, B in GWd/MTU, is determined as the product of the

estimated number of fissions per initial metal atom (FIMA) and a unit conversion factor C that relates FIMA% to GWd/MTU burnup units:

$$B = C \times FIMA\% \quad \text{(D-1)}$$

The recommended value for C is 9.6±0.3 GWd/MTU [D-12].

FIMA% was determined based on the measurement data for ^{148}Nd and uranium and plutonium isotopes:

$$FIMA\% = \frac{{}^{148}_{Nd}/Y_{eff}}{\left(U+Pu+{}^{148}_{Nd}/Y_{eff}\right)} \times 100 \quad \text{(D-2)}$$

where

^{148}Nd is number of ^{148}Nd atoms measured in the sample,

Y_{eff} is effective fission yield for ^{148}Nd,

U is number of uranium atoms measured in the sample, and

Pu is number of plutonium atoms measured in the sample.

The estimated burnup uncertainty is inferred by error propagation in (D-1), assuming all variables involved are independent:

$$\sigma_B^2 = \sum_{i=1}^{N} \left(\frac{\delta B}{\delta x_i}\right)^2 \sigma_i^2 \quad \text{(D-3)}$$

The uncertainty used for the number of ^{148}Nd atoms measured in the sample combined the measurement uncertainty for the isotope with an estimated uncertainty that corrects for radiative captures in ^{147}Nd and ^{148}Nd [D-12]. This correction is expected to be small (<0.5%).

D2.2 Measurement Techniques for Full Isotopics

The analytical protocols and analytical data sets for high-precision radiochemical assay (HP-RCA) performed for the full isotopics samples are described in detail in ORNL/TM-2022/2788 [D-15]. A summary is provided herein and is illustrated in Figure D-3.

The HP-RCA methodology consisted of the following analytical protocols:

- Dissolution: High-precision gravimetric hot cell dissolutions
- Separation via the rapid analysis of post-irradiation debris (RAPID) protocol: high-resolution chemical separations using high-pressure ion chromatography (HPIC). RAPID was used to prescreen and independently quantify fission and activation isotopes and isotopic distributions using a hyphenated HPIC and inductively coupled plasma–mass spectroscopy (ICP-MS) instrument throughout the fuel sample analysis
- Measurement protocols
    - Uranium titrations using the Davies–Gray (D-G) uranium titration method
    - High-precision isotope ratio measurements using multi-collector ICP-MS (MC-ICPMS) or single-detector ICP-MS
    - Internal ratios using isotope dilution (ID)-MC-ICPMS high-precision methodologies to quantify isotopes in the sample with equivalent detector responses (within the uncertainty of the analysis)
    - External calibration using gravimetric procedures to prepare the calibration standards when isotope ratio measurements were not possible

- Gamma spectroscopy for measuring low-mass high-activity isotopes (^{134}Cs, ^{144}Ce, and ^{106}Ru)

Each of the full isotopics specimens, which were ~8 to 25 mm long, were defueled via a gravimetric, self-contained dissolution protocol in a heavily shielded radiological hot cell cubicle. Subsamples of each digested fuel solution were analyzed for radionuclide activities and were then shipped to a separate radiological facility for preparations for analyses. After removal of an aliquot for uranium analysis, the received samples were further diluted by mass and subsampled for the assay and isotopic determinations. The weights of the specimens measured during the dissolution process are presented in Table D-8.

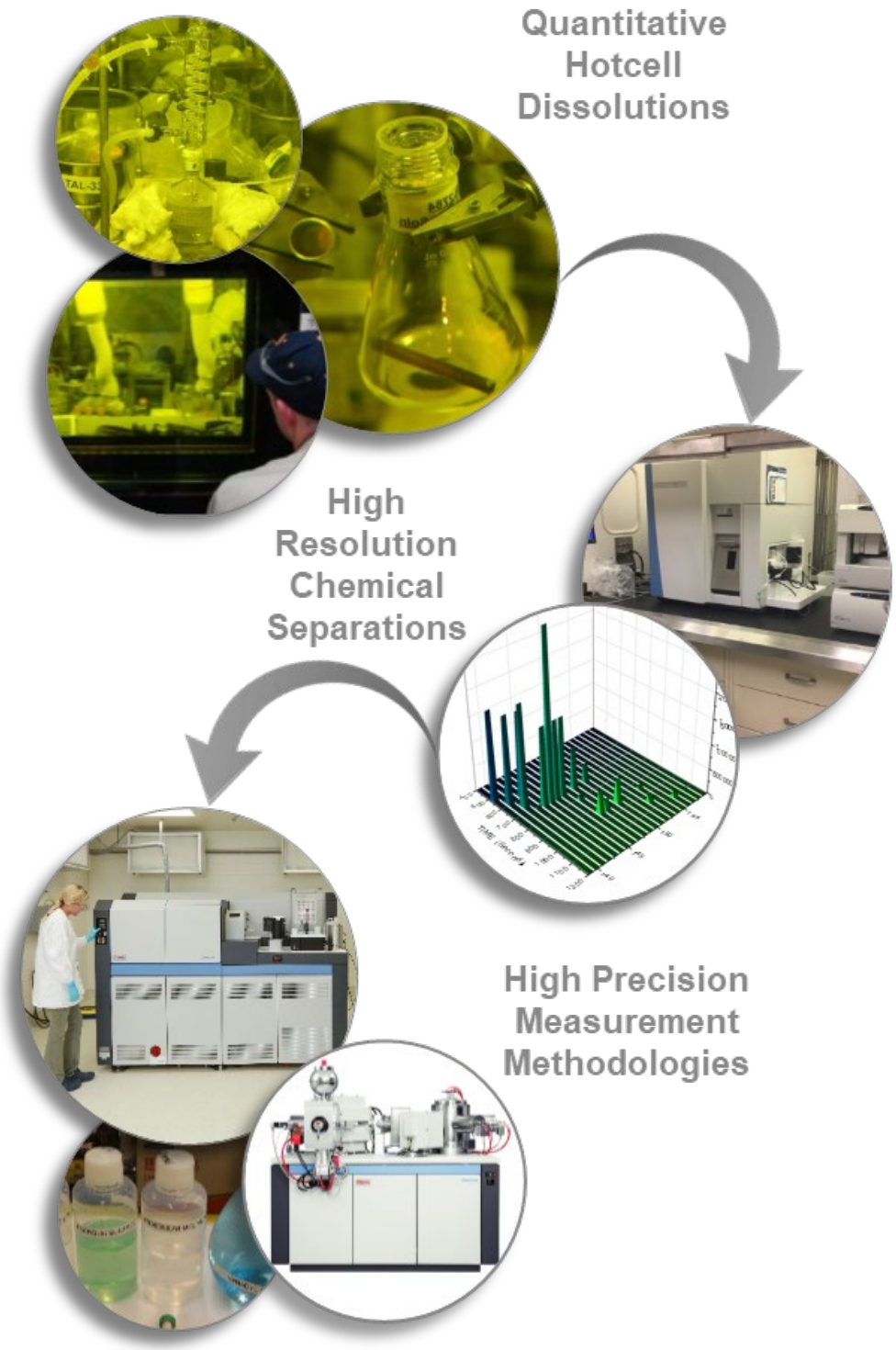


Figure D-3. Illustration of the Analytical Methodology used for HP-RCA [D-15].

Table D-8. Cladding and fuel weights [D-15].

Sample Parent Segment ID	Fuel + cladding weight (g)	Cladding weight (g)	Fuel weight (g)
3D8E14-1375-1450	7.952	1.308	6.664
F35P17-2383-2402	7.597	1.289	6.308
30AE14-2675-2694	5.715	0.934	4.781
3D8E14-0775-0796	12.297	2.649	9.648
F35P17-3050-3069	5.585	0.951	4.634
3F9N05-3593-3615	13.795	2.282	11.513
3A1F05-0150-0165	9.334	1.490	7.844
30AD05-0200-0215	9.324	1.458	7.866
3A1F05-0225-0240	9.309	1.524	7.785

The analyzed isotopes are listed in Table D-9 [D-15] for four categories: major actinides, minor actinides, high-mass fission products, and low-mass fission products.

The first sample listed in Table D-8 was used as a pilot sample to test the developed analytical protocols, therefore allowing the chemistry analysts to optimize the protocols and gain experience before analyzing the other samples.

Table D-9. Isotopes analyzed for full isotopics [D-15].

Major actinides	Minor actinides	High-mass fission products	Low-mass fission products
^{234}U	^{237}Np	^{129}I (not reported)	^{79}Se (not reported)
^{235}U	^{241}Am	^{133}Cs	^{95}Mo
^{236}U	^{242m}Am	^{134}Cs	^{90}Sr
^{238}U	^{243}Am	^{135}Cs	^{99}Tc
^{238}Pu	^{242}Cm	^{137}Cs	^{101}Ru
^{239}Pu	^{243}Cm	^{137m}Ba (calculated)	^{106}Ru
^{240}Pu	^{244}Cm	^{143}Nd	^{103}Rh
^{241}Pu	^{245}Cm	^{144}Nd	^{109}Ag (not reported)
^{242}Pu	^{246}Cm	^{145}Nd	^{125}Sb (not reported)
	^{247}Cm	^{146}Nd	
		^{148}Nd	
Primary analytical method		^{144}Ce	
		^{147}Pm	
Davies–Gray		^{147}Sm	
IDMS (MC-ICPMS or RAPID)		^{149}Sm	
		^{150}Sm	
Calibrated internal ratio (ICP-MS or RAPID)		^{151}Sm	
		^{152}Sm	
		^{151}Eu	
Weighted external calibration (ICP-MS)		^{153}Eu	
		^{154}Eu	
RAPID-HPGe		^{155}Eu	
		^{155}Gd	

Note: MC-ICPMS is multicollector inductively coupled plasma–mass spectroscopy, RAPID is rapid analysis of post-irradiation debris, ICP-MS is inductively coupled plasma–mass spectroscopy, and RAPID-HPGe is RAPID separated fuel fractions analyzed via gamma spectroscopy using high-purity germanium detection.

D-3. Results

D3.1 Burnup-Only Specimens

Chemical determination of burnup per the established ASTM method [D-13] has been completed for the three selected fuel specimens. A drop of sample 3D8E14-3206-3225 was lost during filtering of the final digested solution in the hot cell. At the time of loss, the solution was homogeneous and therefore the ratio of Nd-148 burnup indicator to uranium and plutonium was not compromised. For this sample burnup was calculated using total atom ratios in the final solution vs. the pellet as is convention. Performing the calculation in this manner does not affect the results and they are considered accurate.

The isotope measurement data used for burnup determination for the 3 samples are provided in terms of g/g initial heavy metal (IHM) in Table D-10; the uncertainty is 1σ.

The estimated sample burnups and the FIMA% values based on the measurement data are presented in Table D-10. A value of 1.706% [D-13] was used for the effective fission yield of ^{148}Nd. The quantified 1σ burnup uncertainties for U and Pu are consistent with the measurement data in Table D-10. The uncertainty used for these calculations was conservatively assumed as 0.5% [D-14].

Table D-11 also compares the measurement-based burnup estimate with the operator-reported burnup. There is good agreement, with a relative difference on the order of 5%. 3D8E14-3206-3225 and 6U3K09-3506-3525 are both from the steep burnup gradient region at the top of the fuel rod, whereas sample 3D8E14-700-719 was cut from the flat burnup region of the rod. The samples from the steep gradient region are likely to have a higher uncertainty associated with the operator-reported burnup that is related to the fidelity of the simulations, and this is consistent with the differential from the measured burnup, with the steep gradient samples having a slightly higher differential from predictions.

Additionally, as a proof of concept, the Nd fractions from the three burnup samples were analyzed using HP-RCA. A comparison of the two datasets shows an improvement in relative precision for elemental Nd from +/- 3 % to +/- 0.5 % and its isotopic abundances from +/- 0.08 % to +/- 0.005 % (2-σ).

Table D-10. Chemical Isotopic analysis (burnup only) of sister rod specimens.

Project ID	**3D8E14-0700-0719**		**3D8E14-3206-3225**		**6U3K09-3506-3525**	
Lab ID	**TAL SR-719**		**TAL SR-3225**		**TAL SR-3525**	
Specimen weight (g)	6.648		4.532		6.707	
Units	**g/gIHM**	**1σ uncertainty**	**g/gU**	**1σ uncertainty**	**g/gIHM**	**1σ uncertainty**
Nd (isotopics over six runs)	7.526E-03	3.919E-05	7.724E-03	3.987E-05	5.237E-03	2.755E-05
^{124}Nd wt %	0.8687%	0.0080%	0.7634%	0.0034%	0.4903%	0.0039%
^{143}Nd wt %	15.1330%	0.0017%	16.3629%	0.0011%	20.1298%	0.0016%
^{144}Nd wt %	36.4023%	0.0034%	35.2676%	0.0019%	32.3057%	0.0021%
^{145}Nd wt %	14.9920%	0.0016%	15.2970%	0.0010%	16.5945%	0.0014%
^{146}Nd wt %	18.7087%	0.0025%	18.4323%	0.0014%	17.1489%	0.0016%
^{148}Nd wt %	9.2956%	0.0019%	9.3012%	0.0016%	9.0248%	0.0016%
^{150}Nd wt %	4.5996%	0.0017%	4.5756%	0.0016%	4.3059%	0.0015%
Units	**g/gIHM**	**1σ uncertainty**	**g/gSoln**	**1σ uncertainty**	**g/gIHM**	**1σ uncertainty**
U	9.150E-01	3.235E-03	3.424E-03	0.342E-05	9.418E-01	1.561E-03
^{233}U wt %	0.0010%	NA	0.0010%	NA	0.0010%	NA
^{234}U wt %	0.0248%	0.0002%	0.0254%	0.0002%	0.0272%	0.0003%
^{235}U wt %	0.4368%	0.0005%	0.5822%	0.0007%	1.3359%	0.0016%
^{236}U wt %	0.6425%	0.0066%	0.6370%	0.0066%	0.5944%	0.0061%
^{238}U wt %	98.8958%	0.0066%	98.7554%	0.0066%	98.0425%	0.0063%
Units	**g/gIHM**	**1σ uncertainty**	**g/gU**	**1σ uncertainty**	**g/gU**	**1σ uncertainty**
Pu (isotopics over six runs)	1.119E-02	1.976E-04	1.276E-02	4.890E-04	1.028E-02	7.288E-05
^{238}Pu wt %	3.4261%	0.0598%	3.6434%	0.2286%	2.1232%	0.0597%
^{239}Pu wt %	50.9118%	0.1726%	51.7021%	0.9671%	59.1770%	0.4432%
^{240}Pu wt %	27.8080%	0.1372%	27.3843%	0.8421%	24.6114%	0.4178%
^{241}Pu wt %	6.3293%	0.1139%	6.5196%	0.2642%	8.3739%	0.1013%
^{242}Pu wt %	11.5248%	0.1091%	10.7505%	0.3482%	5.7145%	0.0265%

Table D-11. Estimated Burnup for Sister Rod Specimens.

Project ID	**3D8E14-0700-0719**	**3D8E14-3206-3225**	**6U3K09-3506-3525**
Lab ID	**TAL SR-719**	**TAL SR-3225**	**TAL SR-3525**
Measured FIMA (%)	6.651	6.239	4.473
Measured burnup (GWd/MTU)	63.849	59.895	42.940
Measured burnup uncertainty, 1σ (%)	0.9	0.8	0.7
Operator-estimated burnup (GWd/MTU)	63.564	56.779	40.658
Measured/operator burnup ratio	1.004	1.055	1.056

D3.2 Full Isotopic Specimens

Measurement results as a function of sample burnup are illustrated in Figure D-4 to Figure D-6 for selected actinides and fission products. The error bars in the figures are reported experimental errors at 95% confidence level (2σ). Detailed measurement results are provided in [D-15] for all isotopes shown in Table D-10.

Measured concentrations of selected uranium and plutonium isotopes in g/gU are illustrated in Figure D-4 and Figure D-5. Measured concentrations of selected isotopes of the Cs and Nd fission products are illustrated in Figure D-6 and Figure D-7.

Experimental determination of the sample burnup for the nine measured samples was documented in [D-18]. The estimation was based on the measured value of the burnup indicator fission product ^{148}Nd and the ASTM-321E standard method [D-20] for burnup determination. The reported burnup estimates [D-18] are shown in Table D-11, along with the measured value and relative measurement uncertainty for ^{148}Nd and the determined FIMA% values. The reported measurement uncertainty for ^{148}Nd is less than 0.3% for all nine samples. The burnups based on experimental data varied from 34.5 67.2 GWd/t.

Table D-12. Burnup estimates based on measurement data [D-18].

Sample ID	^{148}Nd		FIMA%	Burnup
	g/gU	2σ (%)		(GWd/MTU)
3D8E14-1375-1450	7.85E-04	0.24	6.81	65.4
F35P17-2383-2402	8.08E-04	0.25	7.00	67.2
30AE14-2675-2694	7.04E-04	0.24	6.15	59.1
3D8E14-0775-0796	7.77E-04	0.24	6.75	64.8
F35P17-3050-3069	7.88E-04	0.25	6.84	65.6
3F9N05-3593-3615	3.99E-04	0.35	3.59	34.5
3A1F05-0150-0165	4.51E-04	0.27	4.04	38.8
30AD05-0200-0215	5.50E-04	0.27	4.88	46.9
3A1F05-0225-0240	5.23E-04	0.27	4.66	44.7

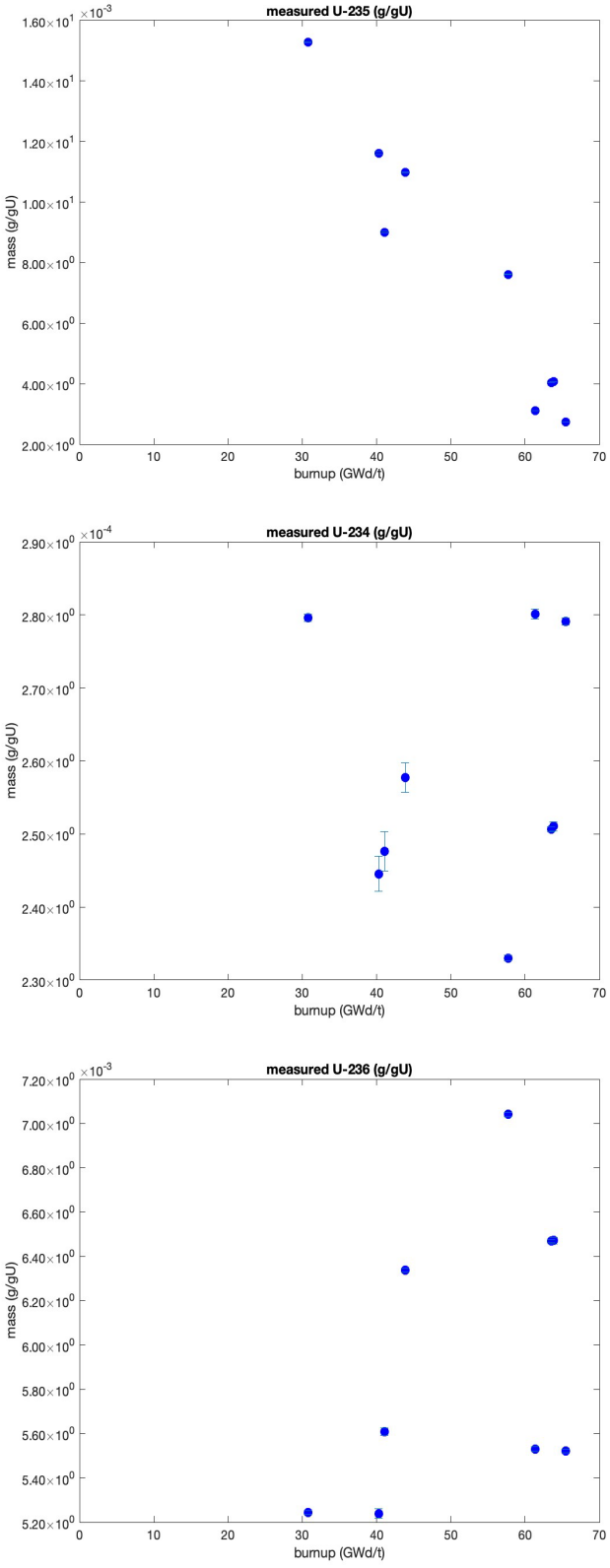
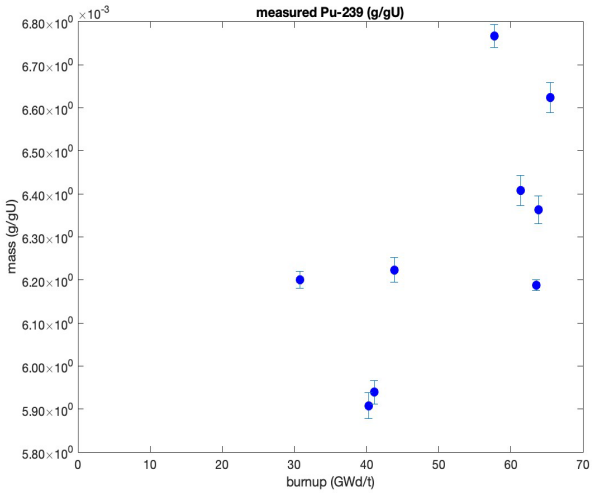
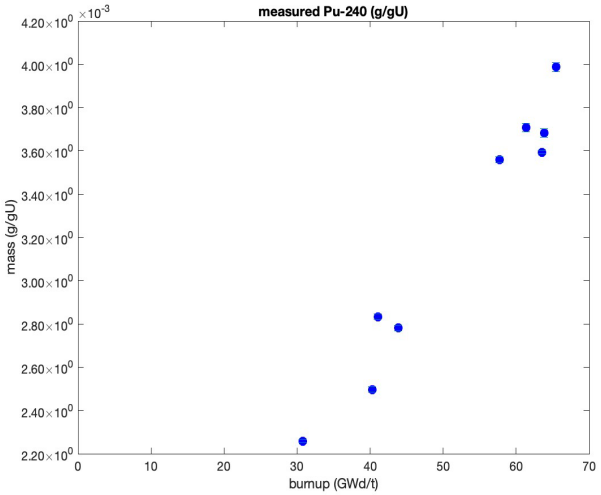


Figure D-4. Measured mass of U isotopes (^{235}U,^{234}U,^{236}U) vs. burnup.





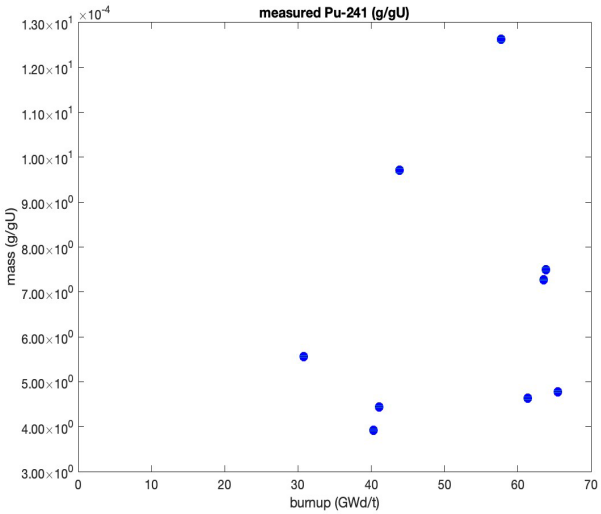
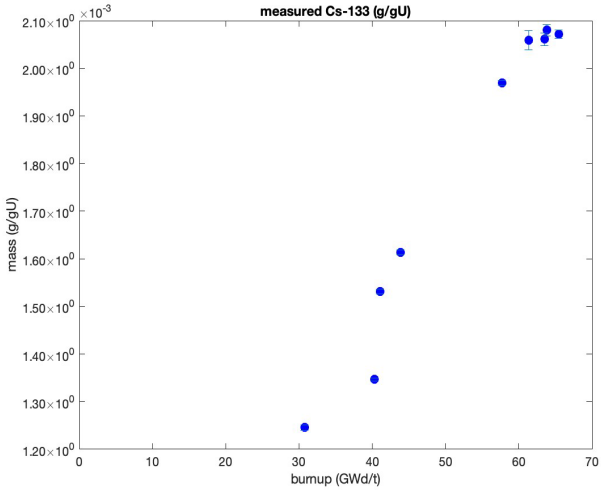
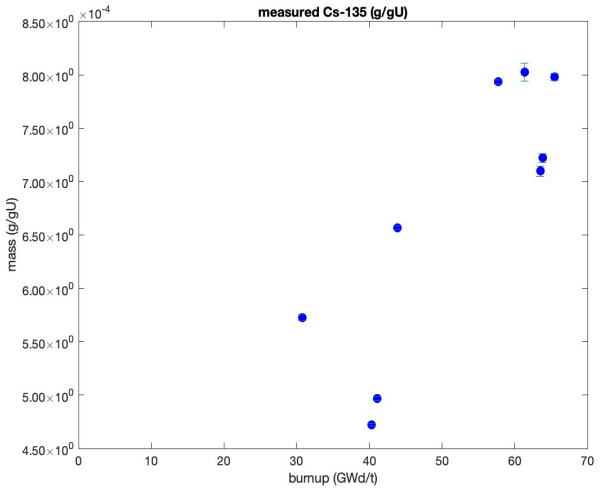


Figure D-5. Measured mass of Pu isotopes (^{239}Pu,^{240}Pu,^{241}Pu) vs. burnup.





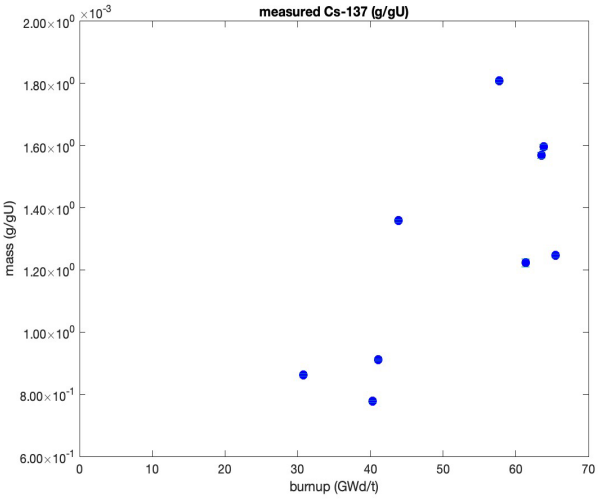


Figure D-6. Measured mass of Cs isotopes (^{133}Cs,^{135}Cs,^{137}Cs) vs. burnup.

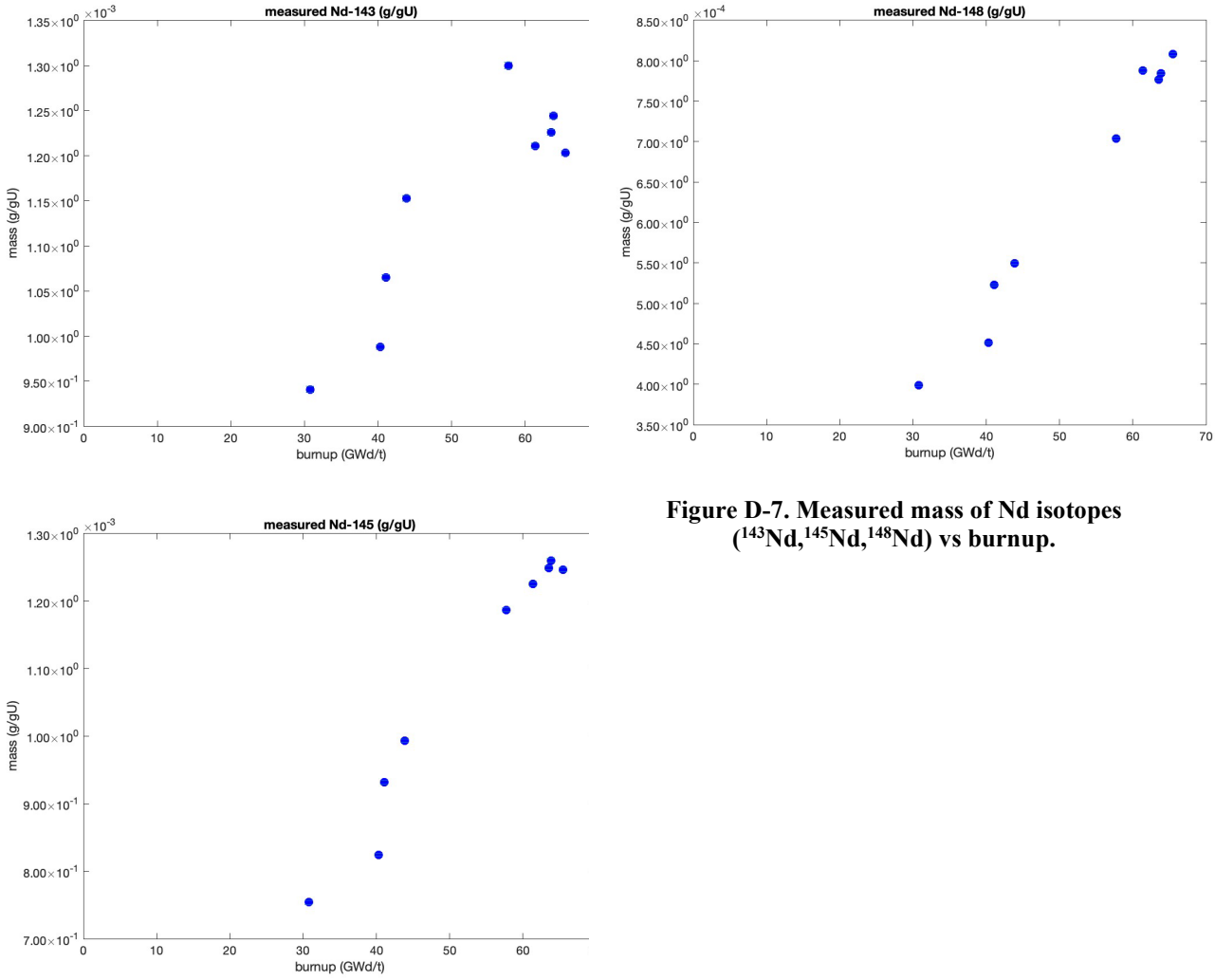


Figure D-7. Measured mass of Nd isotopes (^{143}Nd,^{145}Nd,^{148}Nd) vs burnup.

D3.3 Impact of New Measurement Data on SNF Applications

The new measurements significantly expanded the technical basis for validating actinide and fission product material compositions for SNF storage, transportation, and disposal applications, as well as for burnup credit criticality safety analyses. Calculated-to-measured ratios for all nine samples were presented in Ilas et al. [D-18] for selected uranium, plutonium, cesium, neodymium, and samarium isotopes. They indicated an overall excellent agreement between calculation and experiment.

The new measurements significantly expanded the validation basis for burnup credit applications as documented in detail in the recently published NUREG/CR-7303 [D-18]. The variations with burnup of the measured-to-calculated nuclide concentration ratios that were reported for the major actinides ^{235}U and ^{239}Pu are presented in Figure D-8. The figure is based on measurement data and simulation results for 129 PWR spent fuel samples. These measurements were obtained from various experimental programs for different fuels, and they include the new measurements for North Anna fuel. The different colors indicate the origins of the measured fuel by reactor name, and the error bars account for the reported measurement uncertainty and are shown only for qualitative purposes.

The cited report [D-11] discusses the bias and bias uncertainty in the calculated neutron multiplication factor (k_{eff}) that is associated to calculated spent nuclear fuel nuclide compositions using radiochemical assay measurement data as validation basis for nuclide concentrations. It was shown that the significant improvement of the validation basis, along with the increase in the number of measurements, helped fill data gaps for burnup credit nuclides, especially at high burnups, and resulted in an overall decrease in the k_{eff} bias uncertainty for the considered cask criticality model.

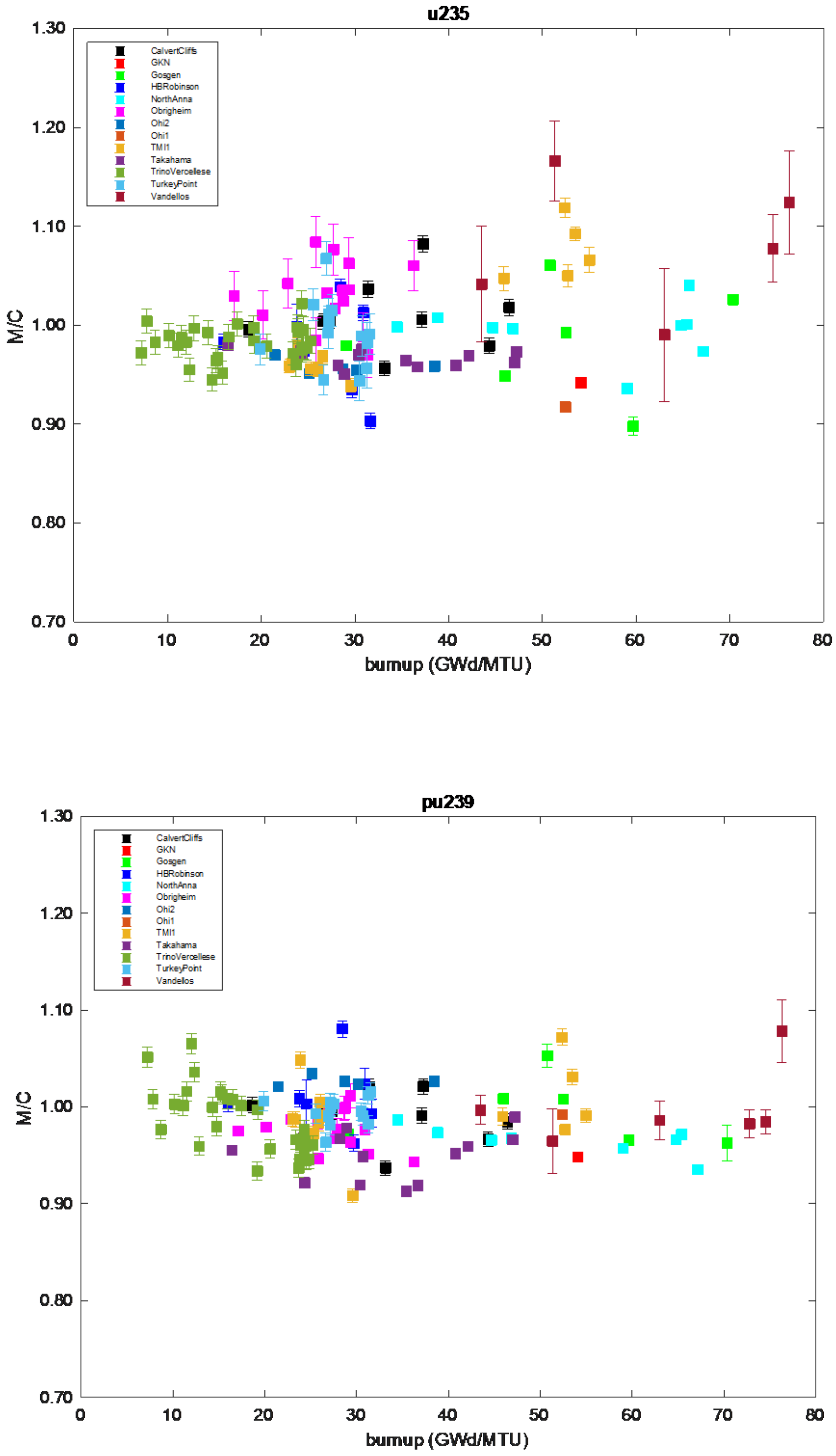


Figure D-8. Measured-to-calcuated concentration ratio for ^{235}U and ^{239}Pu vs. burnup [D-18]].

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