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Review of Experimental Assay Data for PWR Spent Fuel

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OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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Reactor and Nuclear Systems Division

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ABSTRACT

The primary objective of this study is to review existing experimental assay data for pressurized water reactor (PWR) spent fuel, identify gaps in existing data, and asses the potential value of new isotopic measurements for nuclides relevant to burnup credit, shielding, and decay heat validation. The impact of adding new, high-quality experimental assay data on uncertainties associated with PWR depletion validation is discussed. In particular, the discussion focuses on potential benefits of additional isotopic measurements for spent fuel rods available at Oak Ridge National Laboratory. Selected measurements for these spent fuel rods are ongoing in Fiscal Year 2019 (FY19) as funded by the US Department of Energy (DOE) under the Spent Fuel and Waste Science and Technology (SFWST) Program.

ACRONYMS

ANL	Argonne National Laboratory
ATM	approved testing material
C/E	calculated-to-experimental
CEA	Commissariat à l'Énergie Atomique
DOE	US Department of Energy
FY	Fiscal year
GE-VNC	General Electric Vallecitos Nuclear Center
IAEA	International Atomic Energy Agency
ICPMS	inductively coupled plasma mass spectrometry
ID	isotope dilution
IRCh	Institute for Radiochemistry at Karlsruhe
ITU	European Institute for Transuranium Elements
JAERI	Japan Atomic Energy Research Institute (now Japan Atomic Energy Agency)
JRC	Joint Research Center, European Commission
KRI	Khlopin Radium Institute
LT	low tin
LWR	light water reactor
MC-ICPMS	multi-collector inductively coupled plasma mass spectrometry
NEA	Nuclear Energy Agency
NRC	US Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PNL	Pacific Northwest National Laboratory
PSI	Paul Scherrer Institute
PWR	pressurized water reactor
RCA	radiochemical assay
SCK·CEN	Studiecentrum voor Kernenergie – Centre d'étude de l'Energie Nucléaire
SFWST	Spent Fuel and Waste Science and Technology
TIMS	thermal ionization mass spectrometry
WAK	Karlsruhe Reprocessing Plant
YMP	Yucca Mountain Project.

1. INTRODUCTION

The primary objective of this report is to provide a review of existing experimental radiochemical assay (RCA) data for pressurized water reactor (PWR) spent fuel, identify gaps in existing RCA data, and assess the need of new isotopic measurements for nuclides relevant to burnup credit, shielding, and decay heat validation. Additionally, the impact of adding new, high-quality experimental assay data for potentially reducing uncertainties associated with PWR isotopic depletion validation is discussed. In particular, the discussion focuses on potential benefits of additional RCA measurements for spent fuel rods available at Oak Ridge National Laboratory (ORNL). Selected measurements for these rods are ongoing in FY19, as funded by the US Department of Energy (DOE) under the Spent Fuel and Waste Science and Technology (SFWST) Program.

The current study is the first step of a larger effort funded by the US Nuclear Regulatory Commission (NRC) to evaluate and update isotopic and criticality safety validation assessments documented in the NUREG/CR-7108 [1] and NUREG/CR-7109 [2] reports published in 2012. The goal of the study is to improve the isotopic data support for NRC criticality safety (burnup credit) confirmatory evaluations and licensing reviews, as well as NRC evaluations of industry's effort to move towards higher (e.g., more than 62.5 GWd/MTU) burnup fuels. Additionally, existing RCA data for nuclides relevant to future NRC guidance development for decay heat and shielding applications is summarized in this study.

The RCA data considered for isotopic depletion validation in NUREG/CR-7108 includes measurements of 100 PWR fuel samples for 28 actinides and fission products important for burnup credit. The bias and uncertainty in calculated concentrations for these nuclides were determined [2,3] by comparing the measured nuclide data with corresponding nuclide concentrations calculated using depletion capabilities [4,5] and ENDF/B-VII.0 nuclear data in the SCALE 6.1 code system [6]. Since the time at which the analyses documented in NUREG/CR-7108 were performed, additional PWR sample measurements became publicly available, as presented in Section 2 of this report.

A review of available RCA data for PWR spent fuel is provided in Section 2. The ORNL capabilities in performing high-quality RCA measurements, including measurement techniques and estimated accuracies, are summarized in Section 3, along with the characteristics for fuel samples planned to be measured under the SFWST Program. Section 4 discusses RCA data gaps and needs, and the impact of adding new, high-quality measurement data on reducing uncertainties associated with PWR isotopic depletion validation. The review focuses on nuclides important to burnup credit, shielding, and decay heat in PWR spent fuel. Concluding remarks are presented in Section 5.

2. SUMMARY OF EXISTING PWR EXPERIMENTAL ASSAY DATA

Quantifying and evaluating the bias and uncertainties in code predictions of spent nuclear fuel compositions is essential for validating the accuracy of the codes and nuclear data used for PWR safety and licensing calculations. Determination of the bias and uncertainties in code predictions of isotopic compositions is a continuous process. These values must be reassessed to keep pace with continuous changes in the characteristics of spent fuel currently discharged or planned for discharge from commercial reactors in the future. The modern fuels are characterized by higher burnups, higher enrichments, complex and heterogeneous assembly designs, and improved reactor operation. To cover the broad fuel characteristics of relevance to spent fuel applications, a comprehensive experimental database is needed.

2.1 BACKGROUND

Validation of depletion capabilities in the SCALE code system has been a continuous effort at ORNL since the time that these capabilities were first developed. Before the 2000s, analyses were performed with SCALE-4.2 to validate spent fuel isotopic predictions using experimental assay data for 38 PWR fuel samples irradiated in the Calvert Cliffs 1, H.B. Robinson 2, Obrigheim, Trino Vercellese, and Turkey Point 3 reactors [7,8]. The large majority of these samples had burnups lower than 35 GWd/MTU, and the measurements included mostly actinides.

Under a project sponsored by the NRC Office of Nuclear Regulatory Research that was initiated circa 2000s, new experimental RCA data for PWR spent fuel that is representative of modern fuel designs and higher burnups were acquired by ORNL through domestic and international experimental programs. These new data, along with other available measurements, are documented in a series of NUREG/CR reports [9,10,11,12] focused on high-burnup fuels. These data were also used to validate depletion capabilities and nuclear data in SCALE 5.1 [13]. The 51 documented samples were selected from fuel irradiated in the Calvert Cliffs 1, GKN II, Goesgen, Takahama 3, TMI-1 and Vandellos II reactors.

Since 2010 efforts focused on compiling new and old data sets for 118 samples [14] to cover both low and high burnup ranges. These samples included 45 high-burnup samples from the samples addressed in NUREG/CR-7012 [12], as well as samples representative of older assembly designs and lower burnup ranges [7,8]. These 118 samples originated from fuel irradiated in the Calvert Cliffs 1, GKN II, Goesgen, H.B. Robinson 2, Obrigheim, Takahama 3, TMI-1, Trino Vercellese, and Turkey Point 3 reactors. The measurements for these 118 samples were documented and used with SCALE 5.1 and ENDF/B-V cross section data to determine experimental-to-calculated isotopic concentration ratios for the measured nuclides [14].

These 118 PWR samples served as the basis for the PWR isotopic validation in NUREG/CR-7108 [1]. However, 18 of these samples were excluded from the validation data set because the information needed to adequately characterize the samples was incomplete or not permitted to be publicly released. Therefore, the validation data set in NUREG/CR-7108 consisted of measurements from 100 samples. The Vandellos II data (6 samples) were not included in the set of 118 samples, as they had not been fully analyzed at the time.

Since 2010, more PWR experimental RCA data with sufficient documentation to be considered for code benchmarking purposes have become available, including several data sets available through the international database of spent fuel compositions, SFCOMPO [15]. The new RCA data pertain to 6 samples from fuel irradiated in the Ohi-1 and Ohi-2 reactors [16] in Japan and 6 samples measured and analyzed by ORNL under support of DOE-Nuclear Energy (NE) and National Nuclear Energy Administration (NNSA) - 1 sample from Calvert Cliffs 1 fuel [17] and 5 samples from TMI-1 fuel [18].

Additionally, old measurements for 8 samples from 5 rods irradiated in Turkey Point 3 were recently added to SFCOMPO, after the primary reports describing the measurements data were retrieved. These latter samples, which were never analyzed by ORNL, have low burnups ranging between 19.89 and 27.54 GWd/MTU [15].

Under NRC support, ORNL participates in the ongoing REGAL international experimental program coordinated by SCK-CEN in Belgium. Four UO_2 and four UO_2-Gd_2O_3 samples from the Tihange PWR in Belgium are being measured through this program. The data for one of the UO_2 samples is already publicly available, with measurements for the other three UO_2 samples planned.

2.2 PWR RCA DATA

A summary of existing PWR RCA data for 147 samples, for which detailed information is available to develop analysis models, is presented in Table 1. This table lists the main characteristics of the fuel rods from which the samples were selected, including the fuel enrichment and burnup range. The data were obtained from fuel irradiated in 12 PWRs operated in six countries: Germany, Italy, Japan, Spain, Switzerland, and the United States. There were 139 samples selected from UO_2 fuel rods, while 8 samples were selected from UO_2-Gd_2O_3 fuel rods.

The number of PWR measured samples listed in the SFCOMPO 2.0 database (https://www.oecd-nea.org/sfcompo/) with isotopic measurements reported is greater than the data shown in Table 1. SFCOMPO shows RCA data for fuel irradiated in the Genkai 1 and Mihama 3 reactors operated in Japan and the Yankee 1 reactor operated in the United States. As previously noted [14], these data were omitted in the ORNL analyses due to incomplete documentation of the design and reactor operating information necessary to evaluate the measurements.

The number of samples shown in SFCOMPO may differ slightly in a few cases for the reactors listed in Table 1 because SFCOMPO lists all measured samples, including those for cross-check samples. Note that cross-check samples are samples selected from adjacent axial locations of the fuel rod and are characterized by similar operating history and burnup. RCA data for cross-check samples are sometimes combined into one set, as for example measurements for the Goesgen fuel measured under the ARIANE experimental program [10,14], the Vandellos II fuel [11], or the Calvert Cliffs 1 fuel measured by ORNL [17].

Additional RCA data for UO_2 samples would likely become publicly available in the near future through the REGAL international program in which ORNL participates with the support of the NRC. In addition to four UO_2-Gd_2O_3 samples measurements, which are proprietary, four UO_2 samples measurements are being performed. The UO_2 samples were selected from fuel irradiated in Tihange 15 × 15 fuel assemblies; these samples have 4.251 wt % ^{235}U enrichment and estimated burnups of 14, 30, 40, and 54 GWd/MTU.

The measurements of the three Gösgen UO_2 samples included in Table 1, measured under Phase I of the MALIBU program, are currently proprietary, even though the non-disclosure period has passed. These three Gösgen samples are not available in SFCOMPO, as well as one Ringhals PWR sample measured by Studsvik in Phase II of the MALIBU program. The Ringhals sample has not been previously analyzed.

The discussion below applies only to the data included in Table 1.

Table 1. Summary of existing RCA data.

Reactor	Country	Measurement laboratory [a]	Experimental program	Assembly lattice	Enrichment (wt % ^{235}U)	No. of samples [c] / fuel rods	Burnup (GWd/MTU)
Calvert Cliffs-1	US	PNL, KRI	ATM-104 [b]	14 × 14	3.038	3/1	27.4–44.3
		PNL	ATM-103	14 × 14	2.72	3/1	18.7–33.2
		PNL, KRI	ATM-106	14 × 14	2.453	3/1	31.4–46.5
		ORNL	ORNL	14 × 14	2.453	1/1	43.5
GKN II	Germany	SCK·CEN	REBUS [d]	18 × 18	3.8	1/1	54.1
Gösgen	Switzerland	SCK·CEN, ITU	ARIANE [d]	15 × 15	3.5, 4.1	3/2	29.1–59.7
		SCK·CEN, PSI, CEA	MALIBU [d]	15 × 15	4.3	3/1	47.2–70.4
H. B. Robinson-2	US	PNL, LANL	ATM-101	15 × 15	2.561	7/3	16.0–31.7
Obrigheim	Germany	JRC Ispra, Karlsruhe	EUR	14 × 14	2.83, 3.00	22/6	15.6–37.5
		ITU, IRCh, WAK, IAEA	ICE	14 × 14	3.13	5/5	27.0–29.4
Ohi-1	Japan	JAERI	JAERI	17 × 17	3.2	1/1	52.4
Ohi-2	Japan	JAERI	JAERI [e]	17 × 17	1.6874, 3.2	5/3	21.5-38.5
Takahama-3	Japan	JAERI	JAERI [f]	17 × 17	2.63, 4.11	16/3	7.8–47.3
TMI-1	US	ANL	DOE YMP [g]	15 × 15	4.013	11/1	44.8–55.7
		GE-VNC	DOE YMP	15 × 15	4.657	8/3	22.8–29.9
		ORNL	ORNL	15 × 15	4.013	5/2	45.9–55.0
Trino Vercellese	Italy	JRC Ispra, Karlsruhe	EUR	15 × 15	2.719, 3.13, 3.897	15/5	7.2–17.5
		JRC Ispra, Karlsruhe	EUR	15 × 15	3.13	16/5	12.8–25.3
Turkey Point-3	US	Battelle-Columbus	NWTS	15 × 15	2.556	13/7	19.9–31.6
Vandellos II	Spain	Studsvik	CSN/ENUSA	17 × 17	4.5	6/2	43.5–78.3

[a] ANL = Argonne National Laboratory; GE-VNC = General Electric Vallecitos Nuclear Center; PNL = Pacific Northwest National Laboratory; KRI = Khlopin Radium Institute; JAERI = Japan Atomic Energy Research Institute (now Japan Atomic Energy Agency); JRC = Joint Research Center, European Commission; ITU = European Institute for Transuranium Elements; IRCh = Institute for Radiochemistry at Karlsruhe; WAK = Karlsruhe Reprocessing Plant; IAEA = International Atomic Energy Agency; SCK·CEN = Studiecentrum voor Kernenergie – Centre d'étude de l'Energie Nucléaire; PSI = Paul Scherrer Institute; CEA = Commissariat à l'Énergie Atomique

[b] ATM = approved testing material

[c] Sister samples are not counted here; only the combined data of sister samples are counted, as listed in the reference reports

[d] International Experimental Programs coordinated by Belgonucleaire, Belgium, currently managed by SCK·CEN Laboratory

[e] One of the three measured rods had UO_2-Gd_2O_3 fuel with 5 measured samples

[f] Two of the three measured rods had UO_2-Gd_2O_3 fuel, for a total of three UO_2-Gd_2O_3 measured samples

[g] DOE YMP = US Department of Energy Yucca Mountain Project

2.3 FUEL CHARACTERISTICS

The samples listed in Table 1 span a large range of assembly designs—14 × 14, 15 × 15, 17 × 17 and 18 × 18 fuel rod lattices, and fuel enrichments—ranging from 1.6874 to 4.657 wt % ^{235}U. The sample burnups are from 7.8 to 78.2 GWd/MTU. The distribution of burnup as a function of enrichment for the 147 measured samples shown in Table 1 is illustrated in Figure 1; the colors indicate the fuels' origins by reactor name.

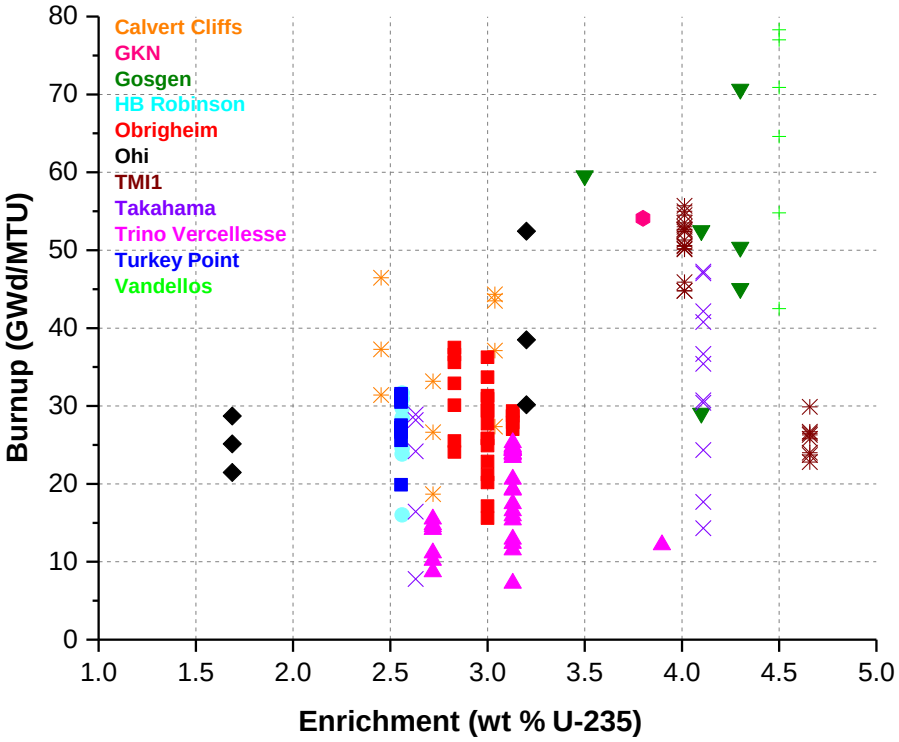

Figure 1. Enrichment distribution vs. burnup for measured samples.

The histogram of the enrichments for the measured samples is provided in Figure 2, and the plotted data are listed in Table 2. As can be seen, almost two thirds of the samples have enrichments lower than 4.0 wt % ^{235}U, and they originate from fuel of older assembly designs. There are 46 samples (~30%) with enrichments ranging between 4.0–5.0 wt % ^{235}U: with 5 from Gösgen, 6 from Vandellos II, 11 from Takahama, and 24 from TMI-1 fuel.

The histogram of the burnups for all measured samples is presented in Figure 3, and the corresponding data are listed in Table 3. Approximately two thirds of the considered samples have burnups lower than 40 GWd/MTU. There are 28 high burnup samples for this set with burnups greater than 45 GWd/MTU, 14 of which originate from TMI-1 fuel. The median of the burnup distribution in Figure 3 is 28.2 GWd/MTU. For comparison, the distribution as a function of burnup of the number of PWR assemblies discharged in the United States prior to 2013 is provided in Fig. 4 of NUREG/CR-7227 [19] and is

reproduced here as Figure 4. The median of this distribution is 42 GWd/MTU, and it is skewed toward higher burnups.

The sample distribution according to fuel origin is shown in Figure 5. The majority of the samples considered originate from three reactors: 31 from Trino Vercellese, 27 from Obrigheim, and 24 from TMI-1.

The number of samples originating from US reactors is 55, with 7 from H. B. Robinson 2, 13 from Turkey Point 3, 11 from Calvert Cliffs-1, and 24 from TMI-1. Only the TMI-1 samples have enrichments greater than 4 wt % ^{235}U. Fourteen of the US fuel samples have burnups greater than 40 GWd/MTU, with 11 of these samples originating from one fuel rod that was irradiated in TMI-1.

A summary of assembly lattice type, fuel rod diameter, and fuel rod pitch for all fuel samples included in Table 1 is provided in Table 4. The sample distribution by assembly lattice type is shown in Figure 6.

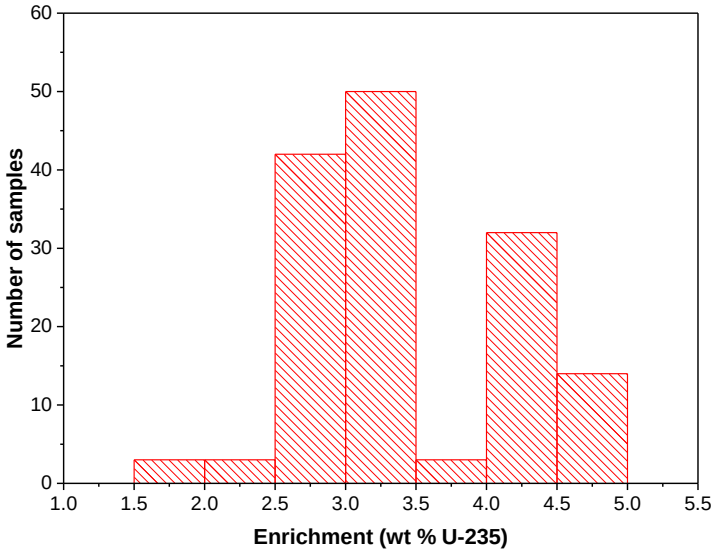

Figure 2. Enrichment distribution histogram for measured samples.

Table 2. Enrichment distribution data for measured samples.

Enrichment range (wt % ^{235}U)	Number of samples
1.5–2.0	3
2.0–2.5	3
2.5–3.0	42
3.0–3.5	50
3.5–4.0	3
4.0–4.5	32
4.5–5.0	14

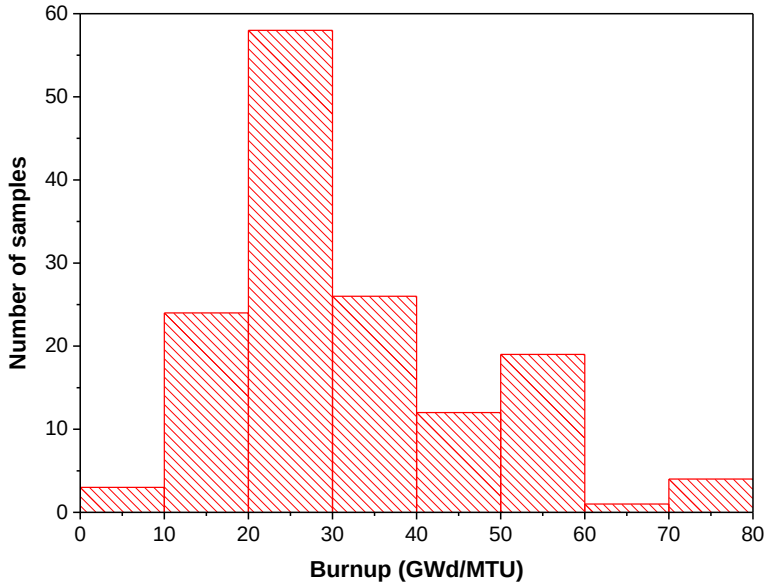

Figure 3. Burnup distribution histogram for measured samples.

Table 3. Burnup distribution data for measured samples.

Burnup range (GWd/MTU)	Number of samples
0–10	3
10–20	24
20–30	58
30–40	26
40–50	12
50–60	19
60–70	1
70–80	4

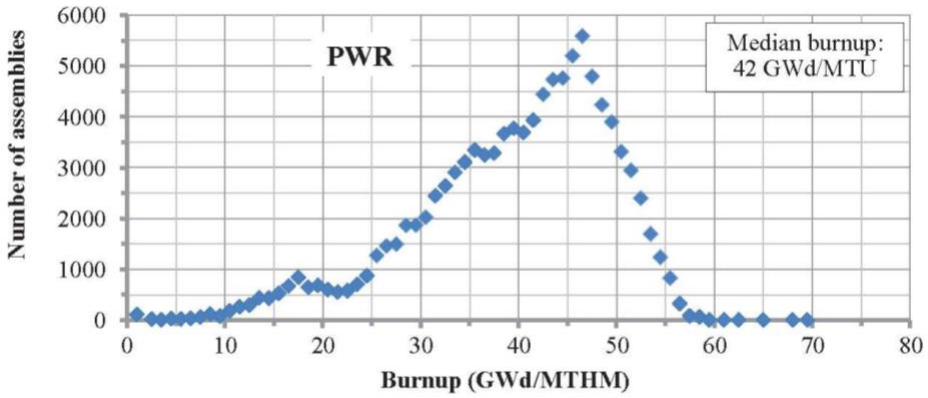

Figure 4. Burnup distribution for US PWR assemblies discharged by 2013 [19].

Table 4. Summary of assembly and fuel rod data.

Reactor	Country	Assembly lattice	Fuel rod diameter (mm)	Fuel rod pitch (mm)	H/X Ratio [a]
Calvert Cliffs-1	US	14 × 14	14.73	11.18	0.42
Obrigheim	Germany	14 × 14	14.3	10.76	0.38
H. B. Robinson-2	US	15 × 15	14.3	10.72	0.40
Turkey Point-3	US	15 × 15	14.3	10.72	0.40
TMI-1	US	15 × 15	14.43	10.92	0.39
Trino Vercellese	Italy	15 × 15	13.03	9.79	0.46
Gösgen	Switzerland	15 × 15	14.3	10.75	0.39
Ohi-1	Japan	17 × 17	12.6	9.5	0.39
Ohi-2	Japan	17 × 17	12.6	9.5	0.39
Takahama-3	Japan	17 × 17	12.6	9.5	0.38
Vandellos II	Spain	17 × 17	12.6	9.5	0.39
GKN II	Germany	18 × 18	12.70	9.5	0.43

[a] Water-to-metal ratio assuming guide tubes are empty.

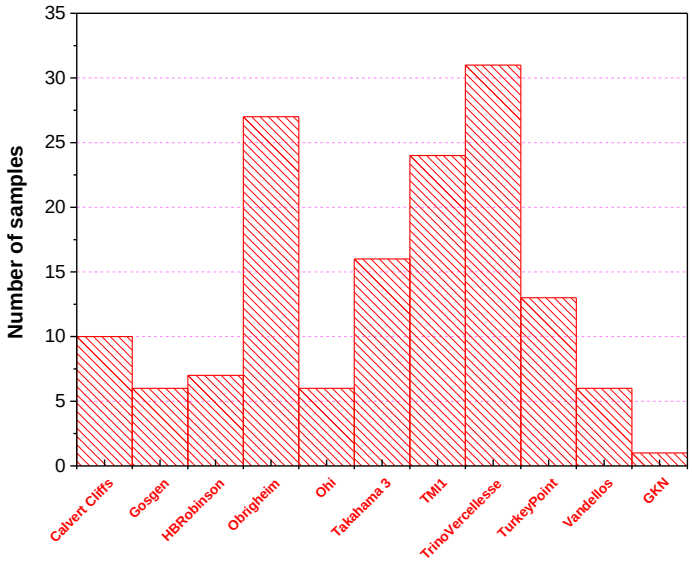

Figure 5. Distribution by reactor for measured samples.

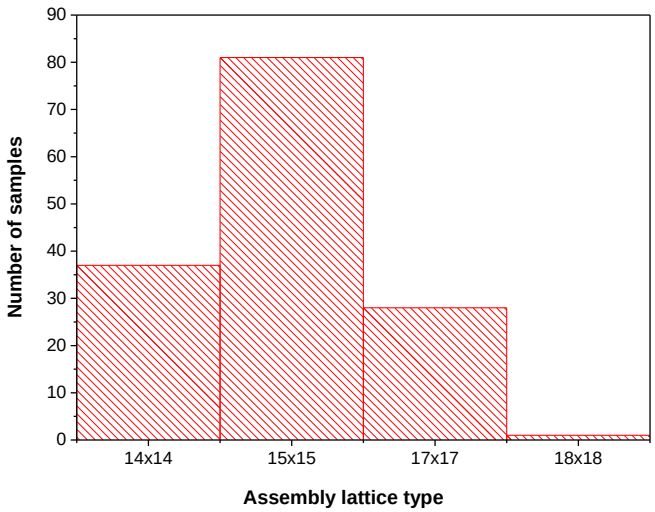

Figure 6. Distribution by assembly lattice type for measured samples.

2.4 NUCLIDE IMPORTANCE TO SAFETY APPLICATIONS

High-quality RCA data are important for evaluating uncertainties in spent nuclear fuel safety analyses, including burnup credit, decay heat, neutron and gamma sources, or waste management applications. In particular, they provide one means for determining uncertainties in integral quantities important to safety, such as decay heat or spent fuel reactivity. Direct measurements of such integral quantities can be expensive or impractical for covering the multitude of existing fuel designs, operating conditions, and specific application purposes. However, as these integral quantities are mainly driven by the nuclide composition in spent fuel at the end of irradiation and the decay time after discharge, measured nuclide compositions can serve as an indirect way to determine uncertainties associated with code predictions of these quantities.

Previous studies have investigated nuclides of high relevance to various safety applications and the relative importance of these nuclides to the metrics characterizing spent nuclear fuel [12, 20, 21, 22]. Findings of these previous investigations are summarized herein.

Table 5 presents a list of nuclides [12, 22] that are highly important to burnup credit, radiological safety, and waste management applications for which measurement data are available. The focus in the current report is on nuclides important to burnup credit and radiological safety (decay heat, source terms).

2.4.1 Nuclides important to burnup credit

There are 28 nuclides of high importance to burnup credit for storage and transportation (see Table 6) that were used as the basis for isotopic validation in NUREG/CR-7108. They include 12 actinides and 16 fission products, which have large neutron fission cross sections and/or large neutron absorption cross sections. Their relative importance to spent fuel reactivity varies with burnup, cooling time, enrichment, and assembly design. Nine of the 12 actinides listed in Table 5 (^{234}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am) account for ~ 95% of the reactivity's worth of the actinides and ~ 70% of the total reactivity's worth of all nuclides in typical spent fuel, whereas 6 of the listed 16 fission products (^{143}Nd, ^{149}Sm, ^{103}Rh, ^{151}Sm, ^{133}Cs, and ^{155}Gd) account for ~75% of the fission product reactivity's worth and ~20% of the total reactivity's worth in typical spent fuel [21].

The importance of these 28 nuclides to fuel reactivity for the GBC-32 PWR cask analyzed in NUREG/CR-7108 is presented in Figure 7 [1], for 10 and 40 GWd/MTU assembly burnups and a 5-year cooling time. This figure shows sensitivity coefficients estimated with a 3D TSUNAMI [6] model of the cask. At the 5-year cooling time, the major contributing actinides are ^{235}U, ^{239}Pu, and ^{238}U, whereas the top three fission product contributors are ^{143}Nd, ^{103}Rh, and ^{151}Sm.

A ranking of the individual actinides and fission product nuclides based on their contribution to the total neutron absorption is provided as a function of burnup, enrichment, and cooling time in NUREG/CR-6700 [21]. For fuel with 4.0 wt% ^{235}U enrichment and 40 GWd/MTU fuel at 5- and 100-year cooling times, some of these data are illustrated in Figure 8 for the nuclides with contributions greater than 0.5% to the total absorption. In this figure, the top three contributors to total absorption in the actinide and fission product categories include the same nuclides identified as important for the sensitivities shown in Figure 7.

Table 5. Nuclides important to spent fuel safety applications [12,22].

Nuclide	Half life	Burnup credit	Radiological safety	Waste management
^{79}Se	2.95×10^5 years			■
^{95}Mo	Stable	■		
^{90}Sr/^{90}Y	28.9 years		■	■
^{99}Tc	2.11×10^5	■		■
^{101}Ru	Stable	■		
^{106}Ru	371.6 days		■	
^{103}Rh	Stable	■		
^{109}Ag	Stable	■		
^{125}Sb	2.76 years		■	
^{129}I	1.6×10^7 years			■
^{133}Cs	Stable	■		
^{134}Cs	2.06 years		■	
^{135}Cs	2.3×10^6 years			■
^{137}Cs/^{137}Ba	30.0 years		■	■
^{139}La[a]	Stable			
^{143}Nd	Stable	■		
^{145}Nd	Stable	■		
^{148}Nd[a]	Stable			
^{144}Ce/^{144}Pr[a]	284.9 days		■	
^{155}Gd	Stable	■		
^{147}Sm	1.06×10^{11} years	■		
^{149}Sm	Stable	■		
^{150}Sm	Stable	■		
^{151}Sm	90 years	■		
^{152}Sm	Stable	■		
^{151}Eu	Stable	■		
^{153}Eu	Stable	■		
^{154}Eu	8.59 years		■	
^{155}Eu[b]	4.75 years	■	■	
^{234}U	2.45×10^5 years	■		■
^{235}U	7.04×10^8 years	■		■
^{236}U	2.34×10^7 years	■		■
^{238}U	4.47×10^9 years	■		■
^{237}Np	2.14×10^6 years	■		■
^{238}Pu	87.71 years	■	■	■
^{239}Pu	2.41×10^4 years	■	■	■
^{240}Pu	6.56×10^3 years	■	■	■
^{241}Pu	14.29 years	■		■
^{242}Pu	3.75×10^5 years	■		■
^{241}Am	433 years	■	■	■
^{243}Am	7,370 years	■		■
^{242}Cm	162.8 days		■	
^{243}Cm[c]	29.1 years	■		
^{244}Cm	18.1 years		■	
^{245}Cm	8.5×10^3 years	■		■
^{246}Cm [c]	18.1 years		■	

[a] Nuclides used as burnup indicators; [b] Important not directly, but as parent nuclide of ^{155}Gd; [c] Important for very high burnup.

Table 6. Nuclides important to burnup credit for storage and transportation [1].

^{234}U	^{235}U	^{236}U	^{238}U	^{237}Np	^{238}Pu
^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{243}Am
^{95}Mo	^{99}Tc	^{101}Ru	^{103}Rh	^{109}Ag	^{133}Cs
^{143}Nd	^{145}Nd	^{147}Sm	^{149}Sm	^{150}Sm	^{151}Sm
^{152}Sm	^{151}Eu	^{153}Eu	^{155}Gd	–	–

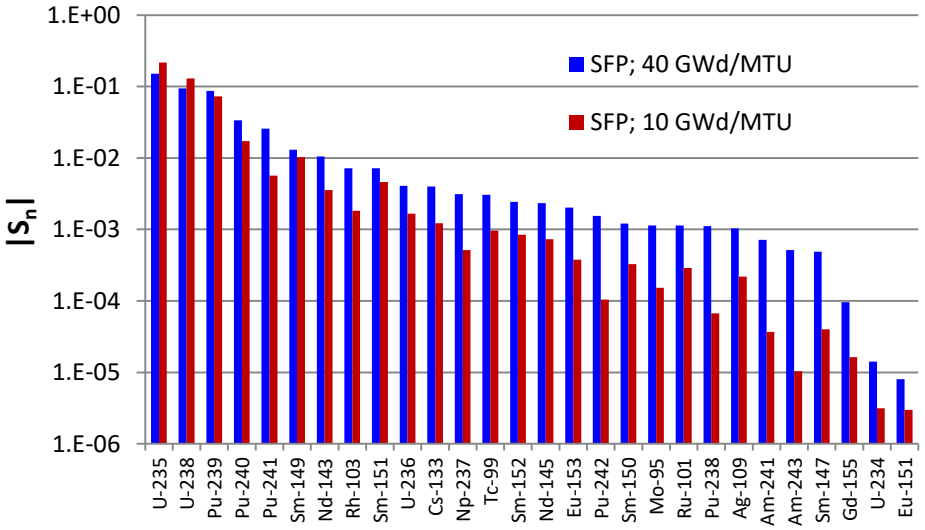

Figure 7. Sensitivity coefficients for GBC-32 PWR cask at 5-year cooling time [1].

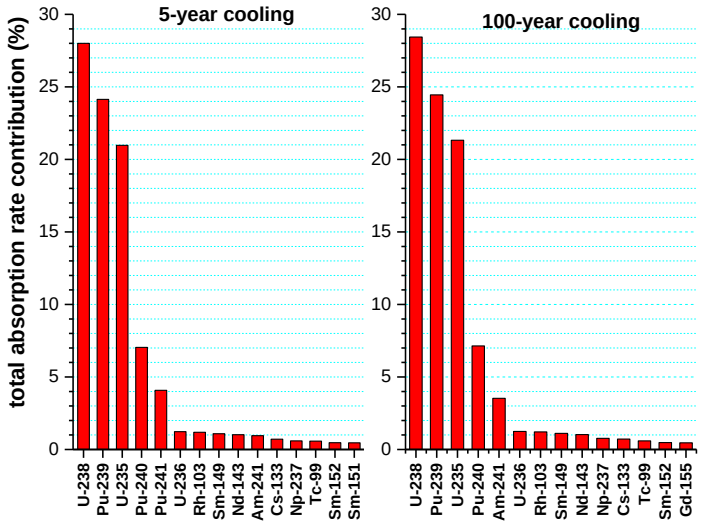

Figure 8. Nuclide contribution to total absorption for 40 GWd/MTU burnup and 5 and 100 years cooling time.

2.4.2 Nuclides important to decay heat

The most important nuclides for decay heat in typical high burnup spent fuel as a function of cooling time are illustrated in Figure 9 [12,22] for fuel with 4.5 wt% ^{235}U initial enrichment and 50 GWd/MTU burnup. This figure shows the fraction of the total decay heat produced by specific nuclides as a function of decay time for up to 1,000 years. Figure 10 provides another example of decay heat contributors as a function of decay time for up to 100 years for spent fuel with 2.9 wt% ^{235}U initial enrichment and 37 GWd/MTU burnup [23].

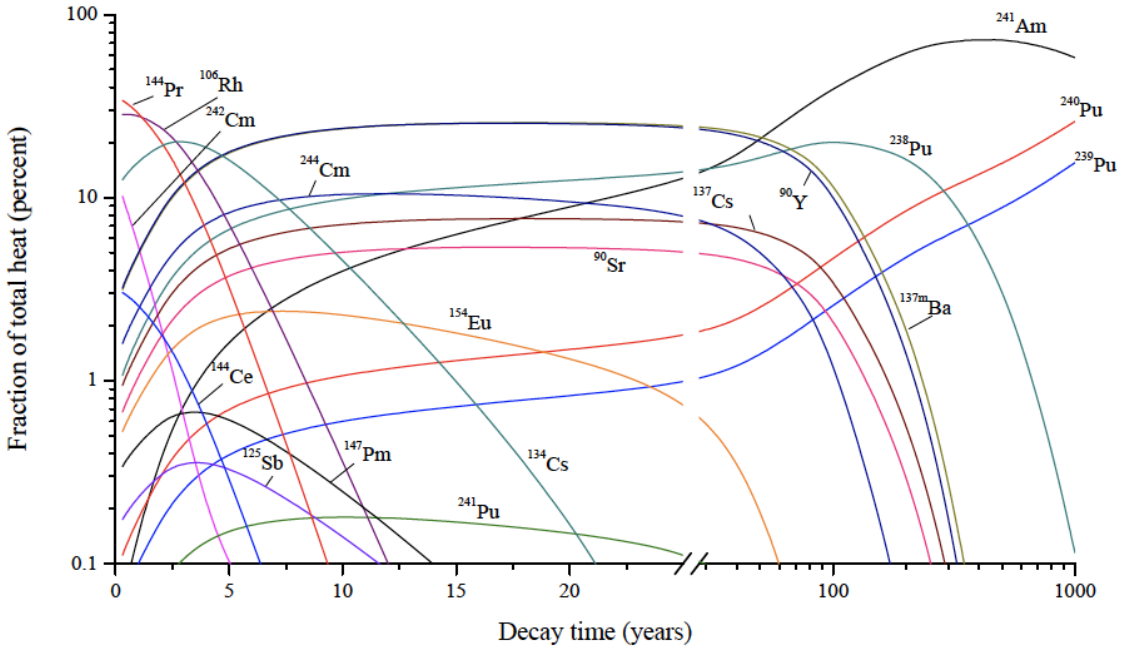

Figure 9. Important nuclides to decay heat for 50 GWd/MTU burnup and 1–1,000 years cooling time [12,22].

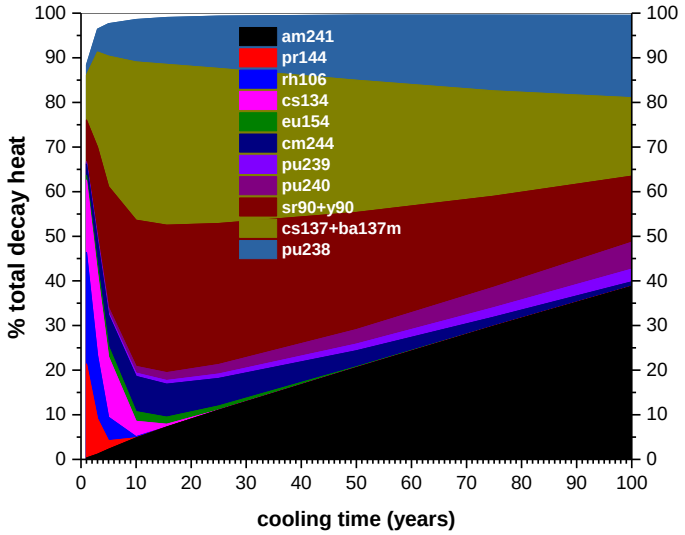

Figure 10. Important nuclides to decay heat for 37 GWd/MTU burnup and 1–100 years cooling time [23].

At less than 30-day cooling time, most of major contributors are fission products with short half lives of minutes or days. The total decay heat decreases in the 30 days after discharge by two orders of magnitude, from ~10^6 W/MTU to ~10^4 W/MTU. At 1 to 50-year cooling times, fission products are main contributors to decay heat, with the top contributors being ^{137}Cs (and its metastable isomer ^{137m}Ba) and ^{90}Sr (and its decay progeny ^{90}Y). The relative contribution to decay heat of fission products decreases with increasing burnup and increasing cooling time. At cooling times greater than ~50 years, fission products are outranked by ^{241}Am, the concentration of which increases with increasing cooling time due to β-decay of ^{241}Pu (14.4-year half life). ^{244}Cm is also a top contributor for approximate 10–100-year cooling times. For example, for 60 GWd/MTU fuel, the contribution of ^{244}Cm to decay heat at 10-year cooling time is ~ 20%, and at 30-year cooling time is ~15%, after which it decreases, as this nuclide decays out (18.1-year half life). At long cooling times over 100 years, ^{241}Am, ^{238}Pu, and ^{239}Pu are the top contributors to decay heat.

2.4.3 Nuclides important to shielding

Nuclides of importance to shielding applications are strong neutron and gamma emitters that contribute to dose rates. Many of these nuclides are similar to those important to decay heat. As the spent fuel is generally shielded, the charged particles and low-energy gamma emitters are less important in shielding applications, while the nuclides emitting high-energy gammas are major contributors to the gamma dose rates outside the shielded fuel; the nuclide importance varies with the type of shielding material and the shield thickness.

An assessment of radionuclide importance to the radioactivity of spent fuel and radiation dose rates for three spent fuel casks with different shielding materials (concrete, steel, and lead) was documented in the ORNL/TM-12724 [20] report published in 1995; results were presented in this report for two burnup values—20 and 50 Wd/MTU—and cooling times ranging between 2–10,000 years. The nuclides listed as the top six contributors to the total radioactivity of unshielded fuel for the two burnups under consideration at 5-year cooling time are ^{241}Pu, ^{137}Cs (and progeny ^{137}Ba), ^{90}Sr (and progeny ^{90}Y), and

^{147}Pm. These nuclides contribute more than 10% to the total activity. At the 10,000-year cooling time, a handful of nuclides contribute ~95% to total radioactivity: ^{239}Pu, ^{240}Pu, ^{243}Am, ^{239}Np, and ^{99}Tc.

Another assessment was documented in NUREG/CR-6700 [21], which was published in 2003. This study focused on high burnup fuel and showed nuclide rankings for burnups of 20 and 70 Wd/MTU and 5- and 100-year cooling times. The nuclides contributing more than 1% to the total dose for a steel cask are illustrated in Figure 11. While ^{60}Co is produced from steel cask activation, the other nuclides shown are present in the spent fuel. At the 5-year cooling time, the following fission products are dominant contributors: ^{90}Y (progeny of ^{90}Sr), ^{106}Rh, ^{144}Pr (progeny of ^{144}Ce), ^{154}Eu, ^{134}Cs, and ^{137m}Ba (progeny of ^{137}Cs). The nuclide ^{244}Cm is the only actinide contributing more than 1% to the total dose at this cooling time. At the 100-year cooling time, actinides are becoming major contributors, including ^{244}Cm, ^{246}Cm, ^{241}Am, $^{238\text{-}242}$Pu, and (^{137}Cs-^{137m}Ba) is a top contributor at both low and high burnups.

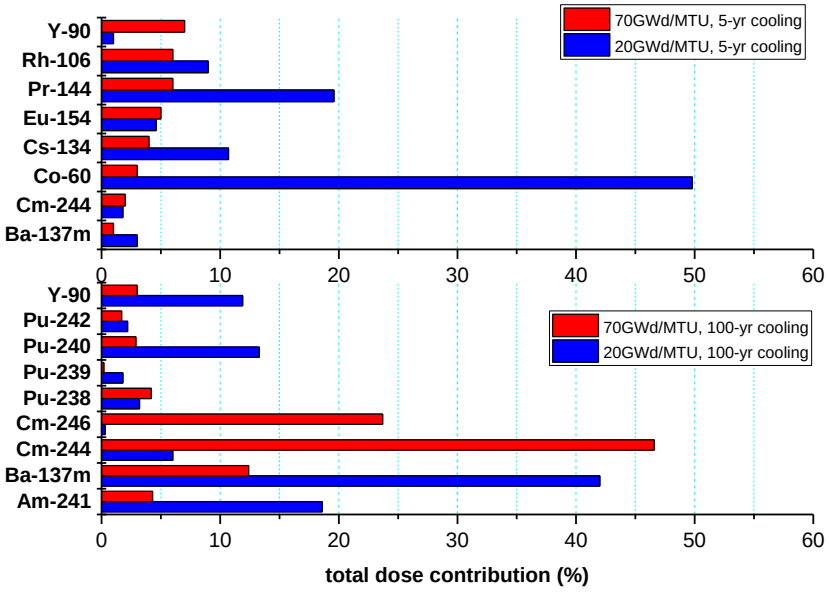

Figure 11. Nuclide contribution to total dose for steel cask (based on data from [21]).

2.5 ISOTOPIC MEASUREMENTS

2.5.1 Isotopic measurements and uncertainties

Isotopic measurements are summarized in Tables 7–15 for the PWR spent fuel datasets presented in Table 1, and include experimental program measurements, the main measurement method, and the typical reported uncertainties (maximum values shown). The acronyms used to list the isotopic methods are identified in Table 16.

A large variety of measurement procedures are used at different laboratories and for different experimental programs when performing sampling and dissolution of fuel, isotope separation, mass spectrometry, and radiometric techniques. These procedures are described in detail in the State-of-the-Art Report [22] published by the Nuclear Energy Agency (NEA) in 2011. Radiometric measurements include α-, β-, and γ-spectrometry, or a combination of these. The minimum uncertainties associated with this type of measurements are 2–3% at a 95% confidence level [22]. Thermal ionization mass spectrometry (TIMS) and multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS) are among the most accurate mass spectrometry techniques; they can reach accuracies of 0.1–0.4% when used in tandem with isotope dilution (ID) as a calibration technique after separation [22]. The main calibration techniques used are (1) *isotope dilution*, which is based on addition of an element with known isotopic composition, or the "spike," which is added after chemical separation to overcome isobaric interferences, and (2) *external calibration*, which is based on the use of standards of different concentrations for measurements with no chemical separation. The measurement uncertainties associated with external calibration are typically a few percent [22].

The reported measurement uncertainties, including their value and significance, differ greatly among measurement laboratories. There is a general lack of consistency in reporting uncertainties—overall uncertainty vs. spectrometry-only uncertainty—across laboratories and different experimental programs [9,10,12,14,17,18,22]. This inconsistency is more prevalent in the experiments performed before modern instruments were available (old vs. new programs). In some cases, the reported experimental uncertainties refer only to instrument precision and are based on multiple measurements of standard solutions. In other cases, the reported uncertainties reflect general laboratory experience in analyzing fuel samples. Recent experimental programs such as ARIANE and MALIBU have reported overall measurement uncertainties for each measured nuclide and sample, including uncertainties resulting from each of the steps involved in the measurement process, beginning with sample cutting and dissolution all the way through the analysis of the mass spectrometry results. In addition, these programs included cross-check measurements that were used to confirm estimated measurement uncertainties.

Table 7. Methods and typical reported uncertainties for actinide measurements (U, Np).

Reactor	Measurement Laboratory [a]	Experimental program	^{234}U		^{235}U		^{236}U		^{238}U-		^{237}Np	
			Method	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*
Calvert Cliffs-1	PNL, KRI	ATM-104 [b]	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	α-sp	1.9
	PNL	ATM-103	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	α-sp	1.9
	PNL, KRI	ATM-106	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	α-sp	1.9
	ORNL	ORNL	IDMS	5.0	IDMS	1.4	IDMS	1.4	IDMS	1.0	ICPMS	6.1
GKN II	SCK·CEN	REBUS	TIMS	2.5	TIMS	0.4	TIMS	0.4	TIMS	0.3	ICPMS	10.0
Gösgen	SCK·CEN, ITU	ARIANE	TIMS	2.5	TIMS	1.2	TIMS	0.8	TIMS	0.2	ICPMS	10.0
	SCK·CEN, PSI, CEA	MALIBU	TIMS	2.5	TIMS	0.4	TIMS	0.4	TIMS	0.3	ICPMS	10.0
H. B. Robinson-2	PNL, LANL	ATM-101	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.9
Obrigheim	JRC Ispra, Karlsruhe	EUR	na	na	IDMS	3.8	IDMS	0.1	IDMS	0.1	na	na
	ITU, IRCh, WAK, IAEA	ICE	na	na	IDMS	0.7	IDMS	0.9	IDMS	0.2	na	na
Ohi-1	JAERI	JAERI	IDMS	1.3	IDMS	0.5	IDMS	0.5	IDMS	0.5	IDMS	5.4
Ohi-2	JAERI	JAERI	IDMS	13	IDMS	0.5	IDMS	0.5	IDMS	0.5	IDMS	5.4
Takahama-3	JAERI	JAERI	IDMS	1.0	IDMS	0.1	IDMS	2.0	IDMS	0.1	α-sp	10.0
TMI-1	ANL	DOE YMP	ICPMS	4.4	ICPMS	3.7	ICPMS	5.8	ICPMS	4.2	ICPMS	5.6
	GE-VNC	DOE YMP	TIMS	0.5	TIMS	0.5	TIMS	0.5	TIMS	0.5	α-sp	2.9
	ORNL	ORNL	ID-ICPMS	12.5	ID-ICPMS	1.2	ID-ICPMS	1.2	ID-ICPMS	0.8	ICPMS	5.0
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	na	na	IDMS	1.6	IDMS	2.4	IDMS	4.3	na	na
Turkey Point-3	Battelle-Columbus	NWTS	IDMS	5.2	IDMS	2.1	IDMS	0.9	IDMS	0.1	na	na
Vandellos II	Studsvik	CSN/ENUSA	ICPMS	20.1	ICPMS	6.8	ICPMS	6.7	ICPMS	na	ICPMS	8.0

Table 8. Methods and typical reported uncertainties for actinide measurements (Pu).

Reactor	Measurement Laboratory	Experimental program	^{238}Pu		^{239}Pu		^{240}Pu		^{241}Pu		^{242}Pu	
			Method	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*
Calvert Cliffs-1	PNL, KRI	ATM-104	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6
	PNL	ATM-103	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6
	PNL, KRI	ATM-106	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6
	ORNL	ORNL	IDMS	1.0	IDMS	1.0	IDMS	1.0	IDMS	1.0	IDMS	1.4
GKN II	SCK·CEN	REBUS	TIMS	1.6	TIMS	0.3	TIMS	0.3	TIMS	0.3	TIMS	0.3
Gösgen	SCK·CEN, ITU	ARIANE	TIMS	1.5	TIMS	0.3	TIMS	0.3	TIMS	1.7	TIMS	0.3
	SCK·CEN, PSI, CEA	MALIBU	TIMS	1.5	TIMS	0.3	TIMS	0.3	TIMS	1.7	TIMS	0.3
H. B. Robinson-2	PNL, LANL	ATM-101	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6	IDMS	1.6
Obrigheim	JRC Ispra, Karlsruhe	EUR	α-sp	14.3	IDMS	0.3	IDMS	0.2	IDMS	1.3	IDMS	5.3
	ITU, IRCh, WAK, IAEA	ICE	IDMS	6.3	IDMS	2.4	IDMS	2.7	IDMS	2.5	IDMS	3.6
Ohi-1	JAERI	JAERI	IDMS	1.6	IDMS	0.5	IDMS	0.5	IDMS	0.5	IDMS	0.5
Ohi-2	JAERI	JAERI	IDMS	6.6	IDMS	0.5	IDMS	0.5	IDMS	0.5	IDMS	0.5
Takahama-3	JAERI	JAERI	IDMS	0.5	IDMS	0.3	IDMS	0.3	IDMS	0.3	IDMS	0.3
TMI-1	ANL	DOE YMP	α-sp	7.9	ICPMS	5.7	ICPMS	6.2	ICPMS	4.6	ICPMS	6.7
	GE-VNC	DOE YMP	α-sp	2.5	TIMS	0.6	TIMS	0.6	TIMS	0.6	TIMS	0.6
	ORNL	ORNL	ID-ICPMS	1.5	ID-ICPMS	0.8	ID-ICPMS	0.9	ID-ICPMS	1.3	ID-ICPMS	1.0
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	IDMS	2.7	IDMS	2.0	IDMS	2.3	IDMS	2.2	IDMS	2.8
Turkey Point-3	Battelle-Columbus	NWTS	IDMS	1.0	IDMS	0.9	IDMS	0.8	IDMS	1.6	IDMS	2.1
Vandellos II	Studsvik	CSN/ENUSA	ICPMS	6.7	ICPMS	3.5	ICPMS	3.5	ICPMS	3.6	ICPMS	13.8

Table 9. Methods and typical reported uncertainties for actinide measurements (Cm).

Reactor	Measurement Laboratory	Experimental program	^{242}Cm		^{243}Cm		^{244}Cm		^{245}Cm		^{246}Cm	
			Method	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*
Calvert Cliffs-1	PNL, KRI	ATM-104	na	na	na	na	na	na	na	na	na	na
	PNL	ATM-103	na	na	na	na	na	na	na	na	na	na
	PNL, KRI	ATM-106	na	na	na	na	na	na	na	na	na	na
	ORNL	ORNL	na	na	na	na	ICPMS	5.0	na	na	na	na
GKN II	SCK·CEN	REBUS	α-sp	16.0	γ-spec	10.0	α-sp	1.3	TIMS	2.8	na	na
Gösgen	SCK·CEN, ITU	ARIANE	α-sp	3.6	γ-spec	36.8	ICPMS	6.4	ICPMS	10.1	TIMS	10.1
	SCK·CEN, PSI, CEA	MALIBU	α-sp	16.0	γ-spec	10.0	α-sp	1.3	TIMS	2.8	TIMS	10.1
H. B. Robinson-2	PNL, LANL	ATM-101	IDMS	5	na	na	IDMS	20	na	na	na	na
Obrigheim	JRC Ispra, Karlsruhe	EUR	α-sp	72	na	na	α-sp	28	na	na	na	na
	ITU, IRCh, WAK, IAEA	ICE	α-sp	100	na	na	α-sp	20	na	na	na	na
Ohi-1	JAERI	JAERI	MS, α-sp	0.5	MS, α-sp	19.1	MS, α-sp	0.9	MS, α-sp	1.1	MS, α-sp	1.4
Ohi-2	JAERI	JAERI	MS, α-sp	0.5	MS, α-sp	19.0	MS, α-sp	8.9	MS, α-sp	1.4	MS, α-sp	1.6
Takahama-3	JAERI	JAERI	MS, α-sp	10.0	MS, α-sp	2.0	MS, α-sp	2.0	MS, α-sp	2.0	MS, α-sp	5.0
TMI-1	ANL	DOE YMP	na	na	na	na	na	na	na	na	na	na
	GE-VNC	DOE YMP	TIMS, α-sp	10.1	TIMS, α-sp	2.8	TIMS, α-sp	2.8	TIMS, α-sp	2.8	na	na
	ORNL	ORNL	na	na	na	na	ICPMS	5.0	na	na	na	na
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	α-sp	2.0	na	na	α-sp	7.0	na	na	na	na
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	na	na	na	na	ICPMS	10.0	na	na	ICPMS	15.0

Table 10. Methods and typical reported uncertainties for actinide measurements (Am).

Reactor	Measurement Laboratory	Experimental program	^{241}Am		^{242m}Am		^{243}Am	
			Method	***RSD (%)***	***Method***	***RSD (%)***	***Method***	***RSD (%)***
Calvert Cliffs-1	PNL, KRI	ATM-104	α-sp	4.9	na	na	na	na
	PNL	ATM-103	α-sp	4.9	na	na	na	na
	PNL, KRI	ATM-106	α-sp	4.9	na	na	na	na
	ORNL	ORNL	α-sp	5.0	na	na	ICPMS	5.0
GKN II	SCK·CEN	REBUS	TIMS	1.8	TIMS	5.5	TIMS	1.8
Gösgen	SCK·CEN, ITU	ARIANE	ICPMS	5.9	TIMS	5.3	ICPMS	6.7
	SCK·CEN, PSI, CEA	MALIBU	TIMS	1.8	TIMS	5.5	TIMS	1.8
H. B. Robinson-2	PNL, LANL	ATM-101	IDMS	5	na	na	IDMS	20
Obrigheim	JRC Ispra, Karlsruhe	EUR	IDMS, α-sp	20.0	IDMS, α-sp	na	IDMS	na
	ITU, IRCh, WAK, IAEA	ICE	α-sp	100	na	na	α-sp	100
Ohi-1	JAERI	JAERI	MS, α-sp	8.3	MS, α-sp	1.5	MS, α-sp	10.9
Ohi-2	JAERI	JAERI	MS, α-sp	8.6	MS, α-sp	1.6	MS, α-sp	11.0
Takahama-3	JAERI	JAERI	MS, α-sp	2.0	MS, α-sp	10.0	MS, α-sp	5.0
TMI-1	ANL	DOE YMP	γ-spec	7.1	ICPMS	3.1	ICPMS	5.9
	GE-VNC	DOE YMP	TIMS, α-sp	3.5	TIMS, α-sp	3.5	TIMS, α-sp	3.5
	ORNL	ORNL	γ-spec	5.0	na	na	ICPMS	5.0
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	α-sp	20.0	na	na	α-sp	8.0
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	ICPMS	3.7	na	na	ICPMS	6.0

Table 11. Methods and typical reported uncertainties for fission products measurements (Nd, Cs).

Reactor	Measurement Laboratory	Experimental program	^{143}Nd		^{145}Nd		^{148}Nd		^{133}Cs		^{134}Cs		^{137}Cs	
			Method	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*
Calvert Cliffs-1	PNL, KRI	ATM-104	IDMS	1.0	IDMS	1.0	IDMS	na	IDMS	1.0	na	na	γ-spec	3.5
	PNL	ATM-103	na	na	na	na	na	na	na	na	na	na	γ-spec	3.5
	PNL, KRI	ATM-106	IDMS	1.0	IDMS	1.0	IDMS	na	IDMS	1.0	na	na	γ-spec	3.5
	ORNL	ORNL	IDMS	1.0	IDMS	1.0	IDMS	1.0	IDMS	1.0	na	na	IDMS	1.0
GKN II	SCK·CEN	REBUS	TIMS	0.3	TIMS	0.3	TIMS	0.3	TIMS	1.3	na	na	γ-spec	1.3
Gösgen	SCK·CEN, ITU	ARIANE	ICPMS	5.1	ICPMS	5.9	ICPMS	6.7	ICPMS	1.6	ICPMS	4.1	ICPMS	1.5
	SCK·CEN, PSI, CEA	MALIBU	TIMS	0.3	TIMS	0.3	TIMS	0.3	TIMS	1.3	ICPMS	4.1	γ-spec	1.3
H. B. Robinson-2	PNL, LANL	ATM-101	IDMS	1.0	IDMS	1.0	IDMS	1.0	na	na	IDMS	5.0	γ-spec	3.5
Obrigheim	JRC Ispra, Karlsruhe	EUR	na	na	na	na	IDMS	1.4	na	na	γ-spec	1.5	γ-spec	1.5
	ITU, IRCh, WAK, IAEA	ICE	na	na	na	na	na	na	na	na	na	na	na	na
Ohi-1	JAERI	JAERI	IDMS	0.1	IDMS	0.1	IDMS	0.1	na	na	γ-spec	3.7	γ-spec	1.9
Ohi-2	JAERI	JAERI	IDMS	0.1	IDMS	0.3	IDMS	03	na	na	γ-spec	3.7	γ-spec	2.4
Takahama-3	JAERI	JAERI	IDMS	0.1	IDMS	0.1	IDMS	0.1	na	na	γ-spec	3.0	γ-spec	3.0
TMI-1	ANL	DOE YMP	CPMS	5.5	CPMS	6.2	CPMS	7.1	na	na	na	na	γ-spec	4.8
	GE-VNC	DOE YMP	TIMS	0.75	TIMS	0.75	TIMS	0.75	na	na	γ-spec	1.8	γ-spec	1.8
	ORNL	ORNL	ID-ICPMS	1.0	ID-ICPMS	1.0	ID-ICPMS	1.1	ID-ICPMS	1.0	γ-spec	5.0	ID-ICPMS	1.1
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	na	na	na	na	IDMS	1.0	na	na	γ-spec	2.5	γ-spec	1.5
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	IDMS	na	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	ICPMS	2.7	ICPMS	2.7	ICPMS	4.1	ICPMS	8.0	γ-spec	5.9	ICPMS	4.0

Table 12. Methods and typical reported uncertainties for fission products measurements (Sm, Ce).

Reactor	Measurement Laboratory	Experimental program	^{147}Sm		^{149}Sm-		^{150}Sm		^{151}Sm		^{152}Sm		^{144}Ce	
			Method	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*
Calvert Cliffs-1	PNL, KRI	ATM-104	MS,LA	3.3	MS,LA	20.01	MS,LA	4.2	MS,LA	38.5	MS,LA	3.2	na	na
	PNL	ATM-103	na	na	na	na	na	na	na	na	na	na	na	na
	PNL, KRI	ATM-106	MS,LA	2.5	MS,LA	13.3	MS,LA	1.5	MS,LA	4.6	MS,LA	2.4	na	na
	ORNL	ORNL	IDMS	1.0	IDMS	2.7	IDMS	1.0	IDMS	2.5	IDMS	1.0	na	na
GKN II	SCK·CEN	REBUS	TIMS	0.4	TIMS	1.1	TIMS	0.4	TIMS	0.4	TIMS	0.4	γ-spec	5.0
Gösgen	SCK·CEN, ITU	ARIANE	ICPMS	10.6	ICPMS	21.4	ICPMS	3.4	ICPMS	33.8	ICPMS	3.2	ICPMS	3.8
	SCK·CEN, PSI, CEA	MALIBU	TIMS	0.4	TIMS	1.1	TIMS	0.4	TIMS	0.4	TIMS	0.4	γ-spec	5.0
H. B. Robinson-2	PNL, LANL	ATM-101	na	na	na	na	na	na	na	na	na	na	IDMS	5.0
Obrigheim	JRC Ispra, Karlsruhe	EUR	na	na	na	na	na	na	na	na	na	na	na	na
	ITU, IRCh, WAK, IAEA	ICE	na	na	na	na	na	na	na	na	na	na	na	na
Ohi-1	JAERI	JAERI	na	na	na	na	na	na	na	na	na	na	γ-spec	2.1
Ohi-2	JAERI	JAERI	na	na	na	na	na	na	na	na	na	na	γ-spec	2.1
Takahama-3	JAERI	JAERI	IDMS	0.1	IDMS	0.1	IDMS	0.1	IDMS	0.1	IDMS	0.1	γ-spec	10.0
TMI-1	ANL	DOE YMP	ICPMS	10.1	ICPMS	8.1	ICPMS	5.0	ICPMS	7.1	ICPMS	4.5	na	na
	GE-VNC	DOE YMP	TIMS	0.9	TIMS	0.9	TIMS	0.9	TIMS	0.9	TIMS	0.9	na	na
	ORNL	ORNL	ID-ICPMS	0.9	ID-ICPMS	1.2	ID-ICPMS	0.9	ID-ICPMS	1.3	ID-ICPMS	1.1	na	na
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	na	na	na	na	na	na	na	na	na	na	γ-spec	1.7
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	na	na	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	ICPMS	4.5	ICPMS	23.8	ICPMS	4.1	ICPMS	5.4	ICPMS	3.7	γ-spec	19.5

Table 13. Methods and typical reported uncertainties for fission products measurements (Eu, Gd).

Reactor	Measurement Laboratory	Experimental program	^{151}Eu		^{153}Eu		^{154}Eu		^{155}Eu		^{155}Gd	
			Method	***RSD (%)***	***Method***	***RSD (%)***	***Method***	***RSD (%)***	***Method***	***RSD (%)***	***Method***	***RSD (%)***
Calvert Cliffs-1	PNL, KRI	ATM-104	MS,LA	9.79	MS,LA	1.8	MS, γ-sp	8.6	MS, γ-sp	16.7	IDMS	6.6
	PNL	ATM-103	na	na	na	na	na	na	na	na	na	na
	PNL, KRI	ATM-106	MS,LA	9.7	MS,LA	1.8	MS, γ-sp	8.6	MS, γ-sp	3.2	IDMS	4.1
	ORNL	ORNL	IDMS	2.7	IDMS	1.0	IDMS	2.7	IDMS	15.0	IDMS	1.0
GKN II	SCK·CEN	REBUS	na	na	TIMS	0.5	γ-spec	1.7	γ-spec	3.0	TIMS	2.5
Gösgen	SCK·CEN, ITU	ARIANE	TIMS	1.1	ICPMS	5.6	ICPMS	11.9	ICPMS	16.1	ICPMS	6.7
	SCK·CEN, PSI, CEA	MALIBU	na	na	TIMS	0.5	γ-spec	1.7	γ-spec	3.0	TIMS	2.5
H. B. Robinson-2	PNL, LANL	ATM-101	na	na	na	na	IDMS	5.0	IDMS	5.0	na	na
Obrigheim	JRC Ispra, Karlsruhe	EUR	na	na	na	na	γ-spec	5.0	na	na	na	na
	ITU, IRCh, WAK, IAEA	ICE	na	na	na	na	na	na	na	na	na	na
Ohi-1	JAERI	JAERI	na	na	na	na	γ-spec	4.3	na	na	na	na
Ohi-2	JAERI	JAERI	na	na	na	na	γ-spec	4.3	na	na	na	na
Takahama-3	JAERI	JAERI	na	na	na	na	γ-spec	3.0	na	na	na	na
TMI-1	ANL	DOE YMP	ICPMS	12.5	ICPMS	5.2	na	na	γ-spec	7.2	ICPMS	8.0
	GE-VNC	DOE YMP	TIMS	0.9	TIMS	0.9	na	na	na	na	TIMS	1.4
	ORNL	ORNL	ID-ICPMS	1.3	ID-ICPMS	1.0	ID-ICPMS	1.1	ID-ICPMS	1.5	ID-ICPMS	1.0
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	na	na	na	na	γ-spec	5.0	na	na	na	na
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	na	na	ICPMS	4.4	ICPMS	6.6	ICPMS	5.8	ICPMS	8.4

Table 14. Methods and typical reported uncertainties for fission products measurements (Ru, Rh, Ag, Sb).

Reactor	Measurement Laboratory	Experimental program	Ru-101		Ru-106		Rh-103		Ag-109		Sb-125	
			Method	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*	*Method*	*RSD (%)*
Calvert Cliffs-1	PNL, KRI	ATM-104	na	na	na	na	na	na	na	na	na	na
	PNL	ATM-103	na	na	na	na	na	na	na	na	na	na
	PNL, KRI	ATM-106	na	na	na	na	ICPMS	4.0	na	na	na	na
	ORNL	ORNL	ICPMS	5.0	na	na	ICPMS	5.0	na	na	na	na
GKN II	SCK·CEN	REBUS	ICPMS	5.0	na	na	ICPMS	5.0	ICPMS	5.0	na	na
Gösgen	SCK·CEN, ITU	ARIANE	ICPMS	12.2	ICPMS	12.2	ICPMS	14.2	ICPMS	9.1	ICPMS	9.4
	SCK·CEN, PSI, CEA	MALIBU	ICPMS	5.0	ICPMS	5.0	ICPMS	5.0	ICPMS	5.0	na	na
H. B. Robinson-2	PNL, LANL	ATM-101	na	na	na	na	na	na	na	na	IDMS	5.0
Obrigheim	JRC Ispra, Karlsruhe	EUR	na	na	na	na	na	na	na	na	na	na
	ITU, IRCh, WAK, IAEA	ICE	na	na	na	na	na	na	na	na	na	na
Ohi-1	JAERI	JAERI	na	na	γ-spec	4.3	na	na	na	na	γ-spec	1.4
Ohi-2	JAERI	JAERI	na	na	γ-spec	4.3	na	na	na	na	γ-spec	6.5
Takahama-3	JAERI	JAERI	na	na	γ-spec	5.0	na	na	na	na	γ-spec	10.0
TMI-1	ANL	DOE YMP	ICPMS	5.8	na	na	ICPMS	3.8	ICPMS	5.9	na	na
	GE-VNC	DOE YMP	na	na	na	na	na	na	na	na	na	na
	ORNL	ORNL	na	na	na	na	ICPMS	5.0	ICPMS	5.0	na	na
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	na	na	γ-spec	3.0	na	na	na	na	na	na
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	na	na	γ-spec	5.2	ICPMS	7.5	na	na	na	na

Table 15. Methods and typical reported uncertainties for fission products measurements (Mo, Tc, Sr).

Reactor	Measurement Laboratory	Experimental program	^{95}Mo		^{99}Tc		^{90}Sr	
			Method	***RSD (%)***	***Method***	***RSD (%)***	***Method***	***RSD (%)***
Calvert Cliffs-1	PNL, KRI	ATM-104	na	na	β-sp	3.5	β-sp	5.7
	PNL	ATM-103	na	na	β-sp	3.5	β-sp	5.7
	PNL, KRI	ATM-106	na	na	β-sp	3.5	β-sp	5.7
	ORNL	ORNL	ICPMS	5.0	na	na	β-sp	5.0
GKN II	SCK·CEN	REBUS	ICPMS	5.0	ICPMS	5.0	na	na
Gösgen	SCK·CEN, ITU	ARIANE	ICPMS	4.60	ICPMS	8.9	β-sp	8.0
	SCK·CEN, PSI, CEA	MALIBU	ICPMS	5.0	ICPMS	5.0	ICPMS	1.5
H. B. Robinson-2	PNL	ATM-101	na	na	β-spec	3.5	IDMS	5.0
Obrigheim	JRC Ispra, Karlsruhe	EUR	na	na	na	na	na	na
	ITU, IRCh, WAK, IAEA	ICE	na	na	na	na	na	na
Ohi-1	JAERI	JAERI	na	na	na	na	na	na
Ohi-2	JAERI	JAERI	na	na	na	na	na	na
Takahama-3	JAERI	JAERI	na	na	na	na	na	na
TMI-1	ANL	DOE YMP	ICPMS	4.2	ICPMS	8.0	na	na
	GE-VNC	DOE YMP	na	na	na	na	na	na
	ORNL	ORNL	ICPMS	5.0	ICPMS	5.0	na	na
Trino Vercellese	JRC Ispra, Karlsruhe	EUR	na	na	na	na	na	na
Turkey Point-3	Battelle-Columbus	NWTS	na	na	na	na	na	na
Vandellos II	Studsvik	CSN/ENUSA	na	na	ICPMS	8.0	na	na

Table 16. Acronyms used for isotopic measurement methods.

Acronym	Method
IDMS	isotope dilution mass spectrometry
ICPMS	inductive coupled plasma mass spectrometry
ID-ICPMS	isotope dilution - inductive coupled plasma mass spectrometry
LA	luminescent analysis
MS	mass spectrometry
TIMS	thermal ionization mass spectrometry
α-sp	α-spectrometry
β-sp	β-spectrometry
γ-sp	γ-spectrometry

2.5.2 Number of available measurements per nuclide

Table 17 summarizes the number of measurements available for each of the nuclides listed in Table 5 that are important for burnup credit, decay heat, and shielding applications. Table 17 also identifies which data set has the largest number of measurements for each nuclide, and for this dataset, it lists the burnup range for the measured samples and the percentage of the measurements relative to the total number of measurements for the nuclide. Uranium and plutonium measurements are available for most samples.

Table 17. Number of measurements per nuclide.

Nuclide	Number of measurements	Set with most measurements	Data for set with most measurements		
			Number of meas.	*Contribution to total number (%)*	*Burnup range (GWd/MTU)*
^{234}U	98	TMI-1	24	24	7.2–25.3
^{235}U	146	Trino Vercellese	31	21	15.6–37.5
^{236}U	131	Obrigheim	27	20	7.2–25.3
^{238}U	122	Trino Vercellese	31	25	22.8–55.7
^{237}Np	61	TMI-1	24	39	15.6–37.5
^{238}Pu	132	Obrigheim	27	20	7.2–25.3
^{239}Pu	146	Trino Vercellese	31	21	7.2–25.3
^{240}Pu	146	Trino Vercellese	31	21	7.2–25.3
^{241}Pu	146	Trino Vercellese	31	21	7.2–25.3
^{242}Pu	145	Trino Vercellese	30	21	22.8–55.7
^{241}Am	95	TMI-1	24	25	22.8–55.7
^{243}Am	82	TMI-1	24	29	15.6–37.5
^{242}Cm	73	Obrigheim	21	29	7.8–47.3
^{243}Cm	30	Takahama	11	37	7.8–47.3
^{244}Cm	91	Takahama	16	18	14.3–47.3
^{245}Cm	32	Takahama	11	34	14.3–47.3
^{246}Cm	28	Takahama	11	39	44.8–55.7
^{95}Mo	24	TMI-1	11	46	18.7–44.3
^{90}Sr	17	Calvert Cliffs	10	59	45.9–55.7
^{99}Tc	38	TMI-1	16	42	44.8–55.7
^{101}Ru	19	TMI-1	11	58	7.8–47.3
^{106}Ru	45	Takahama	16	36	45.9–55.7
^{103}Rh	26	TMI-1	16	62	45.9–55.7
^{109}Ag	22	TMI-1	16	73	7.8–47.3
^{125}Sb	28	Takahama	16	57	18.7–44.3
^{133}Cs	24	Calvert Cliffs	7	29	7.2–25.3
^{134}Cs	88	Trino Vercellese	24	27	7.2–25.3
^{137}Cs	114	Trino Vercellese	24	21	7.8–47.3
^{143}Nd	70	Takahama	16	23	7.8–47.3
^{134}Nd	70	Takahama	16	23	7.2–25.3
^{148}Nd	132	Trino Vercellese	27	20	7.8–47.3
^{144}Ce	47	Takahama	16	34	22.8–55.7
^{147}Sm	49	TMI-1	24	49	22.8–55.7
^{149}Sm	50	TMI-1	24	48	22.8–55.7
^{150}Sm	50	TMI-1	24	48	22.8– 55.7
^{151}Sm	50	TMI-1	24	48	22.8–55.7
^{152}Sm	50	TMI-1	24	48	22.8–55.0
^{151}Eu-	36	TMI-1	13	36	22.8–55.7
^{153}Eu	43	TMI-1	24	56	15.6–37.5
^{154}Eu	75	Obrigheim	16	21	45.9–55.7
^{155}Eu	36	TMI-1	16	44	22.8–55.7
^{155}Gd	40	TMI-1	24	60	7.2–25.3

2.6 EVALUATION STATUS

Most of the available PWR RCA data have been analyzed by ORNL using the TRITON [4] depletion sequence in SCALE, as summarized in Table 18. The list includes the number of samples measured, the number of analyzed samples, the code and nuclear data used for analysis, and the main references where the analyses are documented.

Three of the 7 samples for H. B. Robinson and 8 of the 13 Turkey Point samples included in Table 1 were never analyzed by ORNL because important measurement or modeling data reports were missing until recently. New measurement reports were identified for these samples and were contributed to the SFCOMPO database. The 6 samples from the Ohi reactors were not analyzed by ORNL, as the corresponding data were recently contributed to SFCOMPO by Japan. The total number of samples not analyzed previously by ORNL is 18.

The measurements for the 6 Vandellos II samples were analyzed with SCALE 5.1 and ENDF/B/V data [11]. The analysis of the three Gösgen samples measured in the MALIBU experimental program were documented in a NUREG/CR report that was not published because the data were proprietary at the time. Relative comparisons were documented in a conference paper [24] that showed ratios of measured and calculated nuclide concentrations obtained with SCALE 5.1 and ENDF/B/V data. The MALIBU data have not been publicly released to date, and therefore not included in SFCOMPO.

There is very limited validation for SCALE 6.2/TRITON with ENDF/B-VII.1 cross section data for the 147 samples listed in Table 1. The only documented analyses include one Calvert Cliffs sample measured at ORNL [17] and one Calvert Cliffs sample measured under the ATM-104 program [25].

Table 18. Evaluation status.

Reactor	Measurement laboratory	# of measured samples	# of analyzed samples	SCALE code/data	Main Refs.
Calvert Cliffs-1	PNL, KRI	9	9	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	9,14 1
	ORNL	1	1	6.1/ENDF/B-VII.0 6.2/ENDF/B-VII.1	17 17
Obrigheim	EUR	27	27	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	14 1
H. B. Robinson-2	PNL	4	4	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	14 1
	LANL	3	na	na	na
Turkey Point-3	Battelle	13	5	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	14 1
TMI-1	ANL	24	24	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	9,14 1
	GE-VNC			5.1/ENDF/B-V 6.1/ENDF/B-VII.0	9,14 1
	ORNL			6.1/ENDF/B-VII.0	18
Trino Vercellese	EUR	31	31	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	14 1
Gösgen	SCK, ITU	3	3	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	10,14 1
	SCK, PSI, CEA	3	3	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	24 1
Ohi-1	JAERI	1	na	na	na
Ohi-2	JAERI	5	na	na	na
Takahama-3	JAERI	16	16	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	9,14 1
Vandellos II	Studsvik	6	6	5.1/ENDF/B-V	11
GKN II	SCK	1	1	5.1/ENDF/B-V 6.1/ENDF/B-VII.0	10,14 1

3. ASSAY DATA MEASUREMENTS UNDER THE SISTER RODS PROGRAM

In an ongoing ORNL program funded by DOE-NE, the effects of long-term storage and transportation on light water reactor (LWR) high burnup fuel are being investigated. As part of this program, 25 spent fuel rods selected from seven assemblies irradiated in the North Anna PWR were transported to ORNL in 2016 for further nondestructive and destructive analyses.

These 25 fuel rods, collectively known as *sister rods* due to their selection from symmetric positions in the core/assembly as those used in the long-term storage tests, were selected from 17 × 17 fuel assemblies and had different types of cladding: M5, ZIRLO, Zircaloy-4 (Zirc-4), and Zircaloy-4 with low tin content (LT Zirc-4). The 25 rods had initial enrichments between 3.59 and 4.55 wt% ^{235}U, rod-average burnups between 48 and 59 GWd/MTU, and cooling times in the approximate range of 10–32 years [26]. Complete gamma scans of these rods were performed in the ORNL hot cells. Details about these gamma measurements and more information on the sister rods are provided elsewhere [26].

ORNL's radiochemistry capabilities have been significantly enhanced by acquisition of a state-of-the-art MC-ICPMS system capable of providing relative measurement uncertainties on the order of 0.1%. This new, high-performing measurement instrument which was used last year for measurements on a safeguards-related project, will be used to performed the RCA measurements for the sister rod samples. Measurement protocols have been established for all but three of the important nuclides listed in Section 2.4. A further study of the optimal separation technique is needed for the three remaining nuclides—^{95}Mo, ^{99}Tc, and ^{109}Ag—metallics that are very difficult to measure [22].

At the time of this writing, 15 of the 25 sister rods are available at ORNL, as 10 rods were transported to PNL for additional mechanical testing. These 15 rods originate from different assemblies that were irradiated for three cycles. Full isotopic RCA measurements are scheduled to start at ORNL in the summer of 2019 for 8 samples selected from 5 of the sister rods. These full isotopic measurements are funded by DOE-NE under the SFWST Program. The list of nuclides planned to be measured and the estimated measurement uncertainties corresponding to two different spectrometry techniques are shown in Table 19. The shown measurement uncertainties are provided at 2σ level, and they characterize the spectrometry measurements. These uncertainties were estimated based on an isotopic concentration of 1μg/g fuel. They do not include the uncertainty contribution from the sample dissolution or other steps in the measurement process.

The characteristics of the 8 samples scheduled to be measured are listed in Table 20. Fuel enrichments for the samples are between 4.0 and 4.55 wt% ^{235}U, and the assembly average burnups varies between 50 and 55 GWd/MTU [27]. A plot of the gamma scan for fuel rods in assembly 30A [26] is reproduced herein to illustrate the burnup profile as a function of the axial location along the length of the fuel rod. As seen from this plot, all sample locations selected under the SFWST program (Table 20) would correspond to the plateau region of the burnup profile and therefore would have burnups greater than the reported assembly average burnup. Therefore, these samples would likely have burnups between 50 to 55 GWd/MTU or higher, depending on the peak burnup in the rod. Assuming a rod peak burnup of 110% of the average rod burnup, the burnups for the considered samples could be as high as 60 GWd/MTU.

Table 19. Nuclides to be measured at ORNL for sister rods samples.

Nuclide	Relative uncertainty single-detector ICPMS (2-sigma)	Relative uncertainty multi-detector ICPMS (2-sigma)
^{79}Se	2%	0.1%
^{90}Sr	5%	0.1%
^{101}Ru	5%	0.1%
^{106}Ru		0.1%
^{103}Rh		0.1%
^{125}Sb	2%	0.1%
^{129}I	2%	0.1%
^{133}Cs	2%	0.1%
^{134}Cs		0.1%
^{135}Cs		0.1%
^{137}Cs		0.1%
^{143}Nd	2%	0.1%
^{145}Nd		0.1%
^{146}Nd		0.1%
^{148}Nd		0.1%
^{144}Nd		0.1%
^{144}Ce	2%	0.1%
^{147}Pm	5%	-
^{147}Sm	2%	0.1%
^{149}Sm		0.1%
^{150}Sm		0.1%
^{151}Sm		0.1%
^{152}Sm		0.1%
^{151}Eu	2%	0.1%
^{153}Eu		0.1%
^{154}Eu		0.1%
^{155}Eu		0.1%
^{155}Gd	2%	0.1%
^{234}U	0.5% or 2%	0.1%
^{235}U		0.1%
^{236}U		0.1%
^{238}U		0.1%
^{237}Np	5%	-
^{238}Pu	2%	0.1%
^{239}Pu		0.1%
^{240}Pu		0.1%
^{241}Pu		0.1%
^{242}Pu		0.1%
^{241}Am	2%	0.1%
^{242m}Am		0.1%
^{243}Am		0.1%
^{242}Cm	2%	0.1%
^{243}Cm		0.1%
^{244}Cm		0.1%
^{245}Cm		0.1%
^{246}Cm		0.1%
^{247}Cm		0.1%

Table 20. Characteristics of the 8 samples scheduled for measurements under the SWST Program.

Sample #	Assembly ID	Fuel rod ID	Clad type [a]	Enrichment [b] (wt% ^{235}U)	Assembly [c] average burnup (GWd/MTU)	Sample axial location (mm)
1	30A	D05	M5	4.55	52.0	1280–1299
2	30A	D05	M5	4.55	52.0	2410–2429
3	30A	E14	LT Zirc-4	4.55	52.0	2675–2694
4	3A1	F05	LT Zirc-4	4.0	50.0	2383–2402
5	3D8	E14	ZIRLO	4.2	55.0	2303–2322
6	3D8	E14	ZIRLO	4.2	55.0	2655–2674
7	3F9	N05	Zirc-4	4.25	52.3	2300–2329
8	3F9	N05	Zirc-4	4.25	52.3	2863–2882

[a] provided in Montgomery et al. [26]; [b] provided in Scaglione et al. [27]; [b] provided in Saltzstein et al. [28].

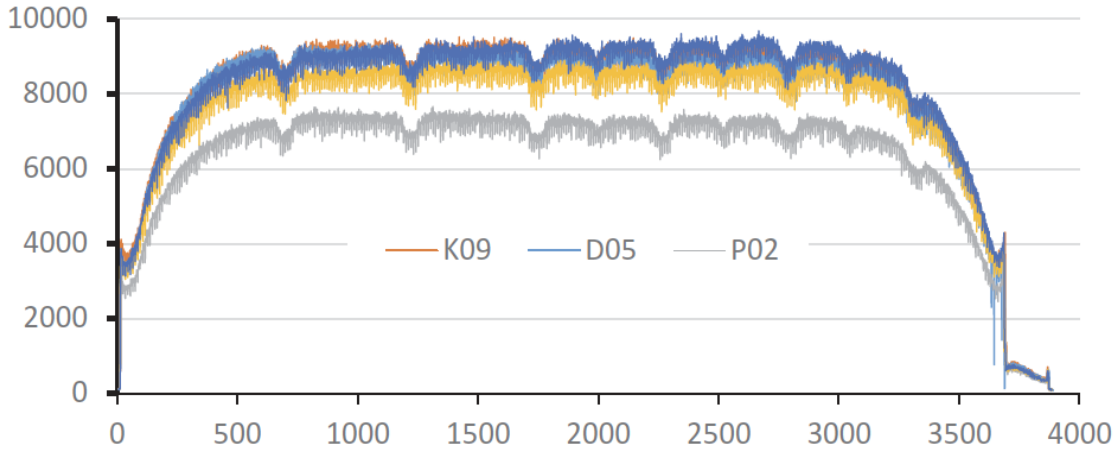

Figure 12. Gamma scan for rods in assembly 30A (axial location in mm on x-axis and number of gamma counts on y-axis) [26, Fig. 12].

4. DATA GAPS AND NEEDS

Validation of computer codes and associated nuclear data against measurement data relevant for the application of interest is important for determining and understanding biases and uncertainties associated with code predictions for that application. The accuracy and precision of predicted isotopic compositions in spent nuclear fuel is generally assessed via calculated-to-experimental (C/E) nuclide concentration ratios for the nuclides of interest. Uncertainties are inherent to computations and measurements, and disentangling the contribution of each of the uncertainty sources is difficult. Understanding the performance, limitations, reliability, and uncertainty in both measurements and computations is highly desirable. The discussion in this section is based on analyses of C/E values reported in previous studies for the important nuclides summarized in Section 2.4 of this report.

Previous results are included here to enhance the discussion. Table 21 reproduces Table 6.1 from NUREG/CR-7108 [1], which shows average E/C values and relative standard deviations for selected burnup ranges. These results were obtained with SCALE 6.1 and ENDF/B-VII.0 cross sections. Table 22 shows C/E results [29] obtained with SCALE 6.1 and ENDF/B-VII.0 data for the same samples that are included in NUREG/CR-7108, less the TMI-1/ANL data and with the addition of the Gösgen /MALIBU samples. Neither of these two tables includes the 6 Vandellos II samples or the one Calvert Cliffs sample and 5 TMI-1 samples measured at ORNL.

Table 21. E/C values (average and standard deviations) in NUREG/CR-7108 [1].

Burnup range	**5 < Burnup ≤ 15 GWd/MTU**			**15 < Burnup ≤ 40 GWd/MTU**			**40 < Burnup ≤ 60 GWd/MTU**		
Nuclide	***No. of samples***	***E/C***	***1-sigma[a]***	***No. of samples***	***E/C***	***1-sigma***	***No. of samples***	***E/C***	***1-sigma***
^{235}U	11	0.9814	0.0284	69	0.9907	0.0416	20	0.9459	0.1096
^{238}U	11	0.9990	0.0063	69	1.0017	0.0042	20	1.0020	0.0021
^{239}Pu	11	0.9906	0.0453	69	0.9587	0.0375	20	0.8984	0.0727
^{240}Pu	11	1.0155	0.0700	69	0.9801	0.0317	20	0.8981	0.0810
^{241}Pu	11	1.0648	0.1103	69	1.0108	0.0514	20	0.9833	0.0839
^{242}Pu	10	1.1029	0.1905	69	1.0647	0.0783	20	1.0636	0.0852

Burnup range	**5 < Burnup ≤ 40 GWd/MTU**			**40 < Burnup ≤ 60 GWd/MTU**		
Nuclide	***No. of samples***	***E/C***	***1-sigma***	***No. of samples***	***E/C***	***1-sigma***
^{234}U	43	0.9119	0.1749	20	0.9114	0.1077
^{236}U	65	1.0249	0.0445	20	0.9862	0.0303
^{237}Np	25	0.9905	0.2429	19	1.0011	0.1072
^{238}Pu	65	1.1500	0.0923	20	1.1375	0.2331
^{241}Am	27	0.9312	0.2077	20	0.9947	0.3224
^{243}Am	30	0.9998	0.2269	18	0.9216	0.2124

Burnup range	**5 < Burnup ≤ 60 GWd/MTU**		
Nuclide	***No. of samples***	***E/C***	***1-sigma***
^{95}Mo	15	1.0002	0.0745
^{99}Tc	25	0.9400	0.2030
^{101}Ru	15	0.9726	0.1152
^{103}Rh	16	0.9021	0.0894
^{109}Ag	14	0.5546	0.2694
^{133}Cs [c]	7	0.9810	0.0680
^{143}Nd	44	0.9779	0.0526
^{145}Nd	44	0.9978	0.0291
^{147}Sm	32	0.9379	0.0967
^{149}Sm	28	0.9634	0.0995
^{150}Sm	32	0.9656	0.0663
^{151}Sm	32	0.9961	0.0782
^{152}Sm	32	0.9736	0.0427
^{151}Eu	21	1.4721	0.7644
^{153}Eu	27	0.9967	0.0480
^{155}Gd	27	1.2556	0.3391

[a] Standard deviation for the distribution of C/E values around the mean

Table 22. C/E values for important nuclides in burnup credit, decay heat, and radiation shielding [29].

Isotope	Number of fuel samples	SCALE 6.1 ENDF/B-VII[a]	
		$(C/E)_{avg}$	σ
^{234}U	55	1.124	0.176
^{235}U	92	1.012	0.035
^{236}U	77	0.981	0.035
^{238}U	92	0.999	0.004
^{238}Pu	77	0.883	0.059
^{239}Pu	92	1.041	0.035
^{240}Pu	92	1.022	0.034
^{241}Pu	92	0.986	0.045
^{242}Pu	91	0.941	0.061
^{237}Np	36	1.039	0.195
^{241}Am	39	1.102	0.207
^{243}Am	38	1.029	0.140
^{244}Cm	57	0.956	0.111
^{245}Cm	24	0.985	0.156
^{246}Cm	14	0.956	0.255
^{90}Sr	15	0.991	0.069
^{99}Tc	20	1.152	0.154
^{101}Ru	7	1.058	0.123
^{106}Ru	31	1.079	0.227
^{103}Rh	8	1.091	0.109
^{109}Ag	6	1.773	0.746
^{125}Sb	18	1.996	0.466
^{133}Cs	10	1.019	0.017
^{134}Cs	59	0.930	0.071
^{135}Cs	16	1.027	0.037
^{137}Cs	73	0.993	0.031
^{143}Nd	36	1.008	0.032
^{145}Nd	36	0.995	0.022
^{148}Nd	77	1.006	0.014
^{144}Ce	32	0.979	0.081
^{147}Sm	24	1.016	0.034
^{149}Sm	20	1.019	0.062
^{150}Sm	24	1.008	0.032
^{151}Sm	24	0.979	0.044
^{152}Sm	24	1.016	0.037
^{151}Eu	12	0.893	0.198
^{153}Eu	19	0.991	0.031
^{154}Eu	44	1.042	0.104
^{155}Eu	11	0.956	0.077
^{155}Gd	19	0.916	0.144

[a] σ is the standard deviation of the C/E values around the mean.

4.1 MAJOR ACTINIDES

Accurate predictions for the two major actinides ^{235}U and ^{239}Pu are critical, as they are the primary drivers for higher actinides and fission products. The samples considered in this report had measurements for these two nuclides that cover a wide burnup range of 7–78 GWd/MTU. Relative C/E ratios for these two nuclides are shown in Figure 13 for the samples considered in NUREG/CR-7108, along with 15 more samples (6 Vandellos II, 3 Gösgen/MALIBU, 1 Calvert Cliffs and 5 TMI-1 measured at ORNL) for which analysis results are available. The error bars account for the reported measurement uncertainties for nuclide concentrations and do not reflect computational uncertainties or uncertainties in the measured burnup, which are generally assessed based on the concentration for the ^{148}Nd burnup indicator fission product. Note that the calculated results included in NUREG/CR-7108 and the calculated results for the TMI-1 samples measured at ORNL were obtained with SCALE 6.1 and ENDF/B-VII.0 cross sections, while the Vandellos and Gösgen /MALIBU calculations were performed with SCALE 5.1 and ENDF/B-V cross sections. Therefore, there is no complete consistency with respect to the code version and nuclear data used to obtain these results. However, as shown in previous work [29], predictions obtained with these SCALE and nuclear data versions are on average similar for the two major actinides.

Most of the samples correspond to lower burnups, as illustrated in Figure 13. Twenty-four of the samples have burnups greater than 50GWd/MTU, with most samples being grouped in the 20–40 GWd/MTU burnup range. The TMI-1 samples measured at ANL show the largest C/E deviations compared to other samples. This is likely due to the large uncertainty in the measured sample burnup; the 1σ measurement uncertainty of 7% in the ^{148}Nd burnup indicator for the ANL measurements is extremely large compared to reported uncertainties for this nuclide from other experimental programs, which are generally below 2%. The sample burnup uncertainty is not included in the error bars in Figure 13. Note that the concentration of ^{235}U is extremely sensitive to burnup [12], especially for high burnup fuel.

The average C/E corresponding to all data over all burnup ranges included in Figure 13 is 1.2 ± 5.6% for ^{235}U and 5.0 ± 5.2% for ^{239}Pu. The mean and standard deviation are significantly reduced if the TMI-1/ANL data set is removed, leading to 0.5 ± 4.1% for ^{235}U and 3.8 ± 3.6% for ^{239}Pu.

The analysis documented in NUREG/CR-7108 concluded that the k_{eff} bias and k_{eff} bias uncertainty values for the considered applications are fairly constant for the burnup range of 5 to 40 GWd/MTU, and they gradually increase with increasing assembly average burnups above 40 GWd/MTU. The analysis for burnups higher than 40 GWd/MTU was based on data for 20 samples, 11 of which are from the TMI-1/ANL data set.

The plot of the ^{239}Pu C/E values for burnups greater than 40 GWd/MTU is provided in Figure 14 for two cases. The first case, illustrated on the right side of Figure 14, shows data for all samples with available C/E data; the dataset is larger than in NUREG/CR-7108 due to addition of 14 samples (Vandellos II, Gösgen /MALIBU, and TMI-1/ORNL). The average C/E of ^{239}Pu is 7.4 ± 7.3% in this case. The average C/E of ^{239}Pu only for the ANL data in this plot is 16.7 ± 3.7%. For the second case, illustrated on the left side of Figure 14, the TMI-1/ANL data were replaced with data likely to be obtained from the 8 North Anna samples scheduled for measurements in the summer of 2019. It is assumed that these samples have burnups between 48 and 60 GWd/MTU (consistent with the discussion in Section 3) and C/E values of 0.5%± 1.0% for each of the eight samples. This assumption is based on the expected measurement uncertainties (see Table 19) and average data for the three Goesgen/MALIBU samples; the average C/E for these three samples is 0.2%, and the average uncertainty in C/E is 0.8%. The average C/E of ^{239}Pu for the data shown in left plot in Figure 14 is 2.5 ± 3.4%, which is significantly improved as compared to the first case.

If the TMI-1/ANL data are removed for burnups greater than 40 GWd/MTU, the number of samples with ^{239}Pu measurements is 24, and the corresponding C/E is 3.1 ± 3.7%. Addition to this set of 8 samples with C/E values of 0.5%± 1.0% for ^{239}Pu would lead to an increase of the data set size to 32 samples and an average C/E of 2.5 ± 3.4%.

All plutonium nuclides are important to the safety applications discussed in Section 2.4. Because the TMI-1/ANL data showed large C/E deviations for all these nuclides, replacing this data with new, better quality data from the ORNL sister rods program would improve the statistics.

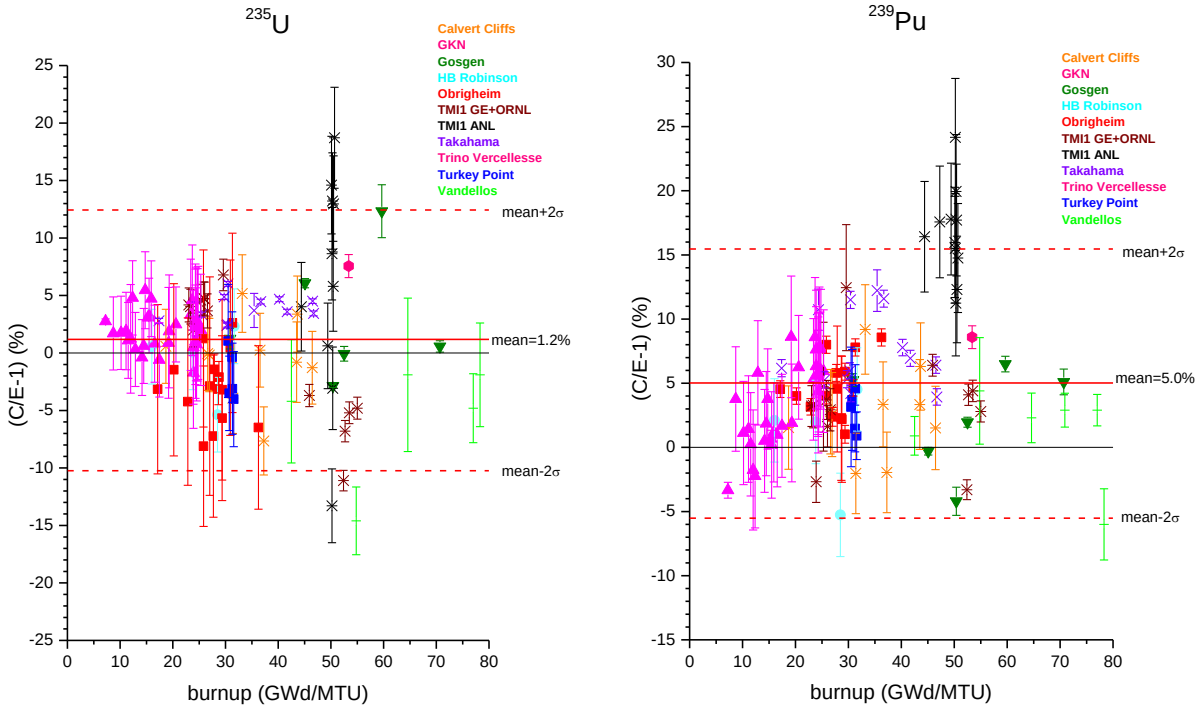

Figure 13. Comparison C/E for major actinides ^{235}U and ^{239}Pu.

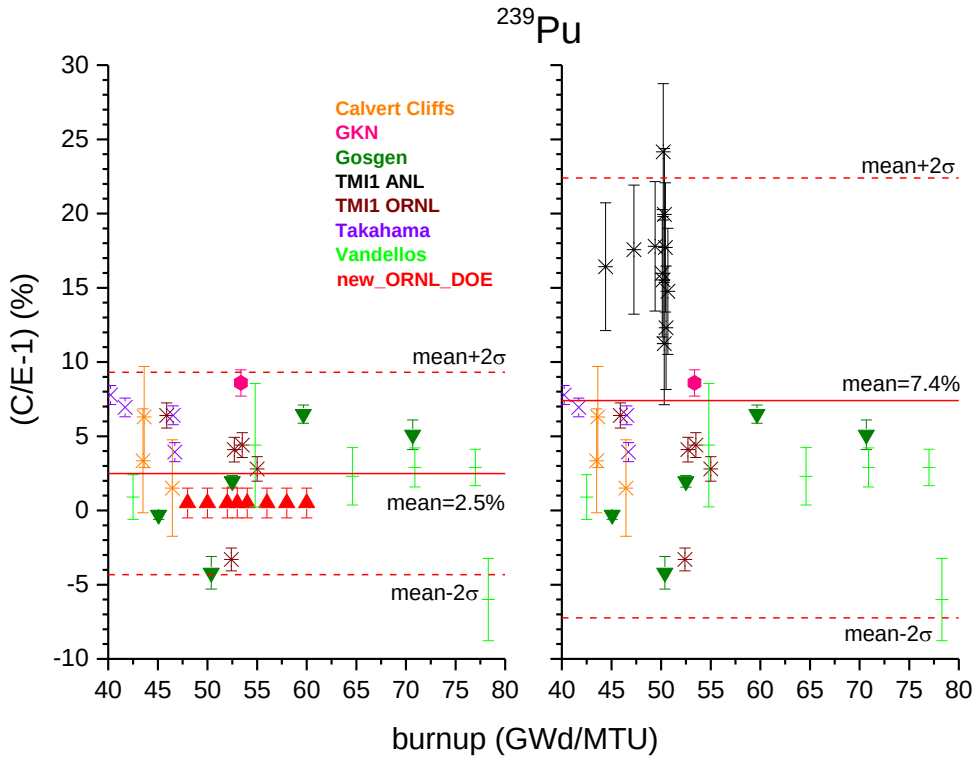

Figure 14. Comparison C/E for ^{239}Pu for burnup > 40 GWd/MTU.

4.2 MINOR ACTINIDES

^{241}Am is a major nuclide for decay heat (Figure 10) and radiation shielding (Figure 11), and it is also included on the list of 12 actinides important to burnup credit (Table 6). The uncertainty for predicting this nuclide as shown in NUREG/CR-7108 is quite large, at 32% for burnups greater than 40 GWd/MTU and 20% for burnups smaller than 40 GWd/MTU. Availability of new, high-quality measurement data over all burnup ranges would improve the aggregate uncertainty and would allow for removal of data sets known to have large uncertainty in burnup, or for which the reported measured data were actually back-calculated to the time of discharge, a process which can introduce large errors in the reported measured data.

Among the Cm nuclides, ^{244}Cm is a major nuclide for both decay heat and radiation shielding applications, whereas ^{246}Cm is important for shielding applications at high burnups (Figure 11). While there are 91 measurements available for ^{244}Cm, there are 26 samples of data for ^{246}Cm, including the Ohi samples that were not analyzed by ORNL. Some existing ^{246}Cm data, such as those for Vandellos [12], may be of little value for code validation due to the large uncertainties associated with α-spectrometry, which are likely underestimated. There is a large variability in the C/E for the samples analyzed to date for both ^{244}Cm and ^{246}Cm. The average relative standard deviation in C/E, which was previously estimated [29], was ~12% for ^{244}Cm and ~27% for ^{246}Cm, though the mean calculated value over all considered samples at the time was within ~5% of the measurement on average. Given the low number of available data for ^{246}Cm and the variability observed, addition of new measurement data over a large burnup range would be beneficial for improving the uncertainty in C/E for this nuclide. The ^{244}Cm data would also benefit from this addition.

4.3 FISSION PRODUCTS

Fission products important to burnup credit that have C/E uncertainties greater than 10% include ^{99}Tc, ^{101}Ru, ^{109}Ag, ^{149}Sm, ^{151}Eu, and ^{155}Gd, with ^{155}Gd having the largest variability. Some of these nuclides have relatively small numbers of measurements available, such as ^{101}Ru, with 19 samples available (Table 17). For ^{151}Eu, for which 36 samples are now available, the large variability is attributed to the 11 TMI-1/ANL samples that show large deviations in C/E compared to other samples. Note that large measurement uncertainties of 12.5% for this nuclide were reported for the ANL set. Among the six nuclides mentioned above, the ^{155}Gd nuclide has the largest uncertainty. The relative C/E data currently available, including the new TMI-1 sample measurements at ORNL [18], are illustrated in Figure 15. Addition of new high quality measurements would reduce the uncertainty and would support the removal of the measurements exhibiting an erratic behavior.

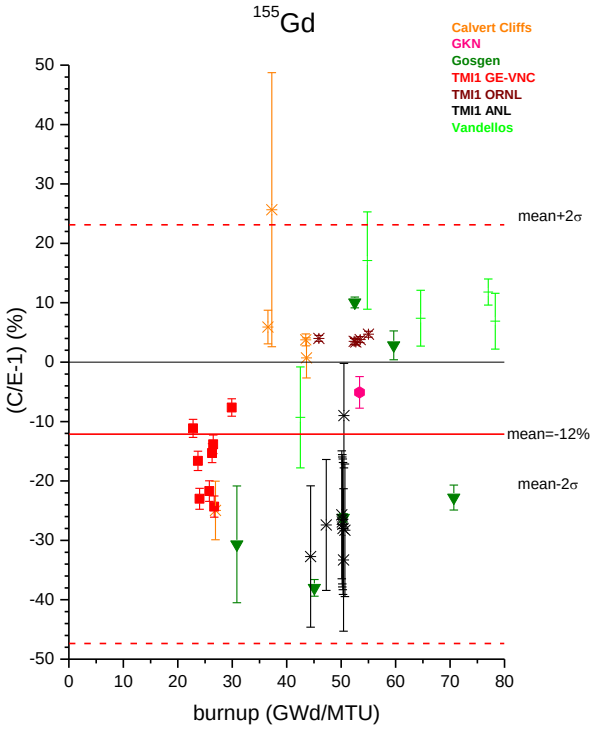

Figure 15. Comparison C/E for ^{155}Gd.

Although this nuclide is on average well predicted, any reduction in the uncertainty of calculated ^{90}Sr would have a sizable impact for shielding and decay heat applications, as its progeny ^{90}Y is a top contributor (see Figures 9–11). Reduction of uncertainty would be possible if new data could be added to the currently available 17 sample measurements.

Among the fission products important for decay heat that have large uncertainties, ^{106}Rh (determined via its parent nuclide ^{106}Ru) shows the largest uncertainty (Table 21). ^{106}Rh is ranked as the fourth most important nuclide for decay heat at the 5-year cooling time [21]; its importance is also illustrated in Figure 10 herein. There are 46 sample measurements available for ^{106}Ru, with 16 of them being measured by JAERI for the Takahama samples. Previous studies [11] indicated potential problems with the JAERI measurements for this nuclide, likely associated with a potential loss of the metallic species in the residue. Addition of new high-quality measurements could lead to reduction of the uncertainty in the code

prediction for this nuclide. Note that ^{106}Rh is also ranked as one of the most important nuclides for cask shielding calculations [21].

4.4 RECCOMENDATIONS

Radiochemical assay measurements are well recognized by the international community as the most reliable approach for validating calculated nuclide concentrations in irradiated fuel [22]. However, this type of measurements are very complex, requiring specialized instruments, experimental techniques and procedures, knowledge, and infrastructure (e.g., hot cells). RCA measurements can be performed only at specialized laboratories. There is a long process from the discharge of a fuel from a reactor to the nuclide concentrations measured in this fuel, including—transportation of spent fuel from the reactor to the laboratory, selection of representative samples and cutting, sample weighting, dissolution, separation, performing radiometric measurements, conducting mass spectrometry, and interpreting measurement steps and results—to provide measured isotopic concentrations and measurement uncertainties.

Given the complexity and cost associated with RCA measurements, as well as the relative scarcity of data for some important nuclides, analysts prefer to use all available sample measurements to support their analyses. At the same time, it is recognized that inclusion in the analysis of some of the problematic data with unreconciled large deviations will yield relatively large estimates of nuclide concentration uncertainty [12]. As previously mentioned, one of the challenges in selecting adequate measurement data to support a specific application is the large inconsistency in the manner that the measurement uncertainties are reported, as some account for all steps in the measurement process, while others show spectrometry uncertainty only, or they are not sample-specific and are based on the laboratory experience for that type of nuclide measurement. Therefore, it is not possible to use the reported measurement uncertainty as a weight function to account for differences in the experiments. Careful observation of the trends in data for different nuclides, samples, and measurement laboratories can serve as a means to identify problematic data with unphysical behavior. Available measurement data should not be removed from a data set without clear evidence of erratic behavior [12].

Reducing isotopic uncertainties requires that additional measurements be performed with high-precision measurement techniques and instruments with low overall measurement uncertainties. Availability of new, high-quality measurements can add new data for nuclides where data are scarce, thus making it possible to replace old data that are clearly known as problematic. Addition of the 8 sample measurements scheduled under the DOE SFWST Program would be of value for improving the quality of existing experimental data. Onsite availability of the sister rods, combined with the specialized experimental capabilities at ORNL, provide a unique opportunity to further expand the RCA measurements by adding even more samples from these rods to the existing measurement plan. This could be accomplished at a significantly reduced cost compared to experiments starting from scratch.

Six additional sample measurements are proposed to supplement the 8 DOE SFWST samples.

- **2 samples from the peak burnup region of rod F35P17 available at ORNL.** Assembly F35 has the largest assembly average burnup at 57.9 GWd/MTU [28] among the seven assemblies from which the sister rods were selected. This rod has a Zirc-4 clad [26]. These two samples will likely have burnups higher than those of the 8 DOE SFWST samples, maybe as high as 65 GWd/MTU (assuming a peak burnup of 110% compared to the assembly average burnup). Note that the number of sample measurements currently available for burnups greater than 62.5 GWd/MTU is 5.
- **2 samples from the low-burnup region of rod 3A1B16 available at ORNL.** Assembly 3A1 has the smallest assembly average burnup at 50 GWd/MTU among the considered assemblies [28].

The cladding for this rod was LT Zirc-4. Sample locations will be selected based on the available gamma scans that correspond to burnups between 30 and 40 GWd/MTU.

- **2 samples from the low-burnup region of rod 30AK09 available at ORNL.** Assembly 30A has an assembly average burnup of 52 GWd/MTU. The cladding for this rod was M5. Sample locations will be selected based on the available gamma scans to correspond to burnups between 40 and 50 GWd/MTU.

Benefits of adding new measurements to the current existing measurement database include:

- The majority of available data are for fuel from 15 × 15 assemblies (Figure 6). The new data to be measured at ORNL is for fuel from 17 × 17 assemblies. Most of the PWR assemblies in the United States have a 17 × 17 lattice [19]. The samples in the SFCOMPO database for 17 × 17 lattices originate from fuel irradiated in Ohi (6 samples), Vandellos (6 samples), and Takahama (16 samples). Addition of new data will improve the coverage for this assembly lattice type.
- Addition of new samples for high-burnup fuel will increase the number of samples in the burnup range > 40 GWd/MTU, for which a large uncertainty in the major actinide ^{239}Pu and other Pu nuclides, as well as in fission products, were noted in NUREG/CR-7108.
- Addition of new samples in the 30–40 and >40 GWd/MTU ranges will increase the number of measurements for nuclides for which data are relatively scarce, including ^{246}Cm, ^{101}Ru, ^{106}Ru, ^{151}Eu, ^{155}Gd. This would also improve statistics.
- Addition of new, high-quality data with low measurement uncertainty will improve overall isotopic uncertainty, including for nuclides that are well predicted on average, but for which uncertainty reduction would have great impact (e.g., ^{90}Y, ^{99}Tc) on applications.
- Addition of new, high-quality measurements for all actinides and fission products over all burnup ranges would improve the aggregate uncertainty for each of these nuclides and would allow for removal of those data sets that exhibit an erratic, unphysical behavior.

5. CONCLUSIONS

A review of existing experimental assay data for PWR spent fuel is provided, with the purpose of identifying gaps in existing data and assessing the potential value of adding new isotopic measurements. The discussion is focused on nuclides important to burnup credit and the impact that new data would have for improving the isotopic uncertainties reported in NUREG/CR-7108. Nuclides important to decay heat and radiation shielding applications are also addressed.

A summary of existing PWR RCA data is provided. This summary is mainly based on experiments included in the international SFCOMPO database or data that were previous analyzed by ORNL. Analysis of the coverage provided by these fuel samples characteristics data, such as burnup, enrichment, and assembly lattice type, is presented. The measurement techniques and their typical measurements uncertainties are presented, as well as the number of available measurements for nuclides that are important to burnup credit, shielding, and decay heat analyses.

The type, quality, and significance of existing data for selected actinides and fission products are discussed, and the value of adding new, high-quality measurement data is emphasized. This discussion is not exhaustive, and it continues from previous similar assessments performed by ORNL [11, 21,1]. In particular, the benefits of additional, high-quality isotopic measurements for spent fuel rods that are available at ORNL through the DOE SFWST Program is addressed, and the addition of six new samples to those already planned for analysis under the SFWST Program is recommended. The proposed six samples include two samples in each of these three burnup ranges: 30–40, 40–50, and >60GWd/MTU. The planned eight SFWST samples cover an estimated burnup range of 50–60 GWd/MTU. There are 44 nuclides important to burnup credit, decay heat, and radiation shielding, and these are planned to be measured by ORNL using state-of-the art instruments and techniques with low measurement uncertainties.

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