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Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation—ARIANE and REBUS Programs (UO_2 Fuel)

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Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation—ARIANE and REBUS Programs (UO_2 Fuel)

Manuscript Completed: June 2009
Date Published: February 2010

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ABSTRACT

This report is part of a report series designed to document benchmark-quality radiochemical assay data against which computer code predictions of isotopic composition for spent nuclear fuel can be validated to establish the uncertainty and bias associated with the code predictions. The experimental data analyzed in the present report were acquired from two international programs: (1) ARIANE and (2) REBUS, both coordinated by Belgonucleaire. All measurements include extensive actinide and fission product data of importance to spent fuel safety applications including burnup credit, decay heat, and radiation source terms. The analyzed four spent fuel samples were selected from fuel rods with initial enrichments of 3.5, 3.8, and 4.1 wt % ^{235}U, which were irradiated in two pressurized water reactors operated in Germany and Switzerland to reach burnups in the 30 to 60 GWd/MTU range. Analysis of the measurements was performed by using the two-dimensional depletion sequence of the TRITON module in the SCALE computer code system.

TABLE OF CONTENTS

LIST OF FIGURES

Page

LIST OF TABLES

ACKNOWLEDGMENTS

This work was performed under contract with the U.S. Nuclear Regulatory Commission Office of Nuclear Regulatory Research, under Project JCN Y6685, *Experimental Data for High Burnup Spent Fuel Validation*. The authors acknowledge the review and helpful comments of R. Y. Lee and D. E. Carlson of the Office of Nuclear Regulatory Research and C. J. Withee, formerly of the Spent Fuel Storage and Transportation Office. Review of the manuscript by our colleagues at Oak Ridge National Laboratory, M. DeHart and G. Radulescu, and the careful formatting of this document by D. J. Weaver is very much appreciated and acknowledged.

ACRONYMS

ANL	Argonne National Laboratory
ARIANE	Actinides Research In A Nuclear Element
BOC	beginning of cycle
C/E	calculated-to-experimental
CEA	Commisariat à l'Énergie Atomique
DOE	U.S. Department of Energy
EOC	end of cycle
GE-VNC	General Electric – Vallecitos Nuclear Center
GKN II	Gemeinschaftskernkraftwerk Unit II
ICP-MS	inductively coupled plasma mass spectrometry
IDA	isotope dilution analysis
ID-MS	isotope dilution mass spectrometry
ITU	Institute for Transuranium Elements
JAERI	Japanese Atomic Energy Research Institute
KRI	Khoplin Radium Institute
LA	luminescent analysis
LWR	light water reactor
MALIBU	MOX and UOX LWR Fuels Irradiated to High Burnup
MOX	mixed oxide
MS	mass spectrometry
MTU	metric ton uranium (10^6 grams)
NRC	U.S. Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PNNL	Pacific Northwest National Laboratory
PSI	Paul Scherrer Institute
PWR	pressurized water reactor
REBUS	Reactivity Tests for a Direct Evaluation of the Burnup Credit on Selected Irradiated LWR Fuel Bundles
SCALE	Standardized Computer Analyses for Licensing Evaluations
SCK-CEN	Studiecentrum voor Kernenergie - Centre d'Étude de l'Energie Nucléaire
TIMS	thermal ionization mass spectrometry
TMI	Three Mile Island
UO_2	uranium dioxide
YMP	Yucca Mountain Project
WABA	wet annular burnable absorber

1. INTRODUCTION

The current trend toward extended irradiation cycles and higher fuel enrichments of up to 5 wt % ^{235}U has led to an increase of the burnup range for discharged nuclear fuel assemblies in the United States expected to exceed 60 GWd/MTU. An accurate analysis and evaluation of the uncertainties in the predicted isotopic composition for spent nuclear fuel in the high burnup regime requires rigorous computational tools and experimental data against which these tools can be benchmarked. However, the majority of isotopic assay measurements available to date involve spent fuel with burnups of less than 40 GWd/MTU and initial enrichments below 4 wt % ^{235}U, limiting the ability to directly validate computer code predictions and accurately quantify the uncertainties of isotopic analyses for modern, high burnup fuel.

This report is part of a report series that documents high-quality radiochemical assay data against which computer code predictions of the isotopic composition in high burnup fuel can be validated. Quantifying and evaluating these uncertainties is fundamental for understanding and reducing the uncertainties associated with predicting the high burnup fuel characteristics for spent fuel transportation and storage applications involving decay heat, radiation sources, and criticality safety evaluations with burnup credit, as well as for reactor safety studies and accident consequence analysis. The report series presents a compilation of recently available isotopic measurements involving high burnup pressurized water reactor (PWR) fuel as well as older isotopic measurements for low- and medium-range burnup fuel that can be used for code validation purposes. Previous experiments were selected primarily on the basis of having extensive fission product measurements.

The experimental data included in the report series prepared for high burnup fuel isotopic validation were compiled from domestic and international programs. The isotopic assay measurements include data for a total of 45 spent fuel samples selected from fuel rods enriched from 2.6 to 4.7 wt % ^{235}U and irradiated in five different PWRs operated in Germany, Japan, Switzerland, and the United States. The samples cover a large burnup range, from 14 to 70 GWd/MTU. A summary of the experimental programs and measured fuel characteristics is listed in Table 1.1.

The current report documents the analysis of experimental data acquired by Oak Ridge National Laboratory (ORNL) through participation in two international programs: (1) ARIANE (Actinides Research In A Nuclear Element) and (2) REBUS (Reactivity Tests for a Direct Evaluation of the Burnup Credit on Selected Irradiated LWR Fuel Bundles), both coordinated by the Belgian company Belgonucleaire. The assay measurements documented in this report include four spent fuel samples selected from fuel rods with 3.5, 3.8, and 4.1 wt % ^{235}U initial enrichments that were irradiated in two PWRs operated in Germany and Switzerland. The four samples cover the burnup range 30 to 60 GWd/MTU.

A brief description of the experimental programs is given in Section 2 of the report. The radiochemical methods employed, the measurement results, and the associated experimental uncertainties are provided in Section 3. Information on the assembly design data and irradiation history is presented in Section 4, and details on the computational models developed and simulation methodology used are given in Section 5. A comparison of the experimental data to the results obtained from code simulations are presented in Section 6.

Table 1.1 Summary of spent fuel measurements

Reactor (country)	Measurement facility	Experimental program name	Assembly design	Enrichment (wt % ^{235}U)	No. of samples	Measurement methods	Burnup(s)[a] (GWd/MTU)
TMI-1[b] (USA)	ANL (USA)	YMP	15 × 15	4.013	11	ICP-MS, α-spec, γ-spec	44.8 – 55.7
TMI-1[b] (USA)	GE-VNC (USA)	YMP	15 × 15	4.657	8	TIMS, α-spec, γ-spec	22.8 – 29.9
Calvert Cliffs[b] (USA)	PNNL, KRI (USA, Russia)	ATM	14 × 14 CE	3.038	3	ID-MS, LA, α-spec, γ-spec	27.4 – 44.3
Takahama 3[b] (Japan)	JAERI (Japan)	JAERI	17 × 17	2.63, 4.11	16	ID-MS, α-spec, γ-spec	14.3 – 47.3
Gösgen[c] (Switzerland)	SCK-CEN, ITU (Belgium, Germany)	ARIANE	15 × 15	3.5, 4.1	3	TIMS, ICP-MS, α-spec, β-spec, γ-spec	29.1, 52.5, 59.7
GKN II[c] (Germany)	SCK-CEN (Belgium)	REBUS	18 × 18	3.8	1	TIMS, ICP-MS α-spec, γ-spec	54.0
Gösgen[d] (Switzerland)	CEA, PSI, SCK-CEN (France, Switzerland, Belgium)	MALIBU	15 × 15	4.3	3	TIMS, ICP-MS, α-spec, γ-spec	46.0, 50.8, 70.4

[a] Correspond to operator-based values, as reported, except for data for MALIBU program samples, which correspond to measured data for burnup indicators.

[b] Documented in G. Ilas, I. C. Gauld, F. C. Difilippo, and M. B. Emmett, *Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation—Calvert Cliffs, Takahama, and Three Mile island Reactors*, NUREG/CR-6968 (ORNL/TM-2008/071), Oak Ridge National Laboratory, Oak Ridge, Tennessee (May 2008).

[c] Documented in current report.

[d] Documented in G. Ilas and I. C. Gauld, *Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation—MALIBU Program (UO_2 Fuel)*, NUREG/CR-6970 (ORNL/TM-2008/13), Oak Ridge National Laboratory, Oak Ridge, Tennessee (May 2008).

2. EXPERIMENTAL PROGRAMS

This section provides a brief overview of the experimental isotopic assay data compiled in this report for code validation and of the international programs through which these data were acquired. A detailed description of the measurement results, techniques, and accuracies is provided in Section 3.

2.1 ARIANE

ARIANE, an international program designed to improve the database of isotopic measurements for spent fuel source term and isotopic inventory validation, was coordinated by Belgonucleaire and completed in March 2001.[1] This collaborative project involved participants from laboratories and utilities from seven countries: Belgium, Germany, Japan, Netherlands, Switzerland, the United Kingdom, and the United States. ORNL participated in this program through support of the U.S. Department of Energy (DOE) Fissile Materials Disposition Program.

A key feature of the ARIANE program was that three cross-checking laboratories participated in radiochemical assay measurements to reduce the experimental uncertainties and improve confidence in the measured data: Studiecentrum voor Kernenergie - Centre d'Étude de l'Énergie Nucléaire (SCK-CEN) in Belgium, Paul Scherrer Institute (PSI) in Switzerland, and Institute for Transuranium Elements (ITU) in Germany. Measurements were carried out on both uranium dioxide (UO_2) and mixed oxide (MOX) fuels between 1996 and 1999. Only the UO_2 samples are discussed in this report.

The three UO_2 samples considered were selected from fuel rods irradiated in the Gösgen reactor operated in Switzerland. One of these samples was obtained from an assembly with an initial enrichment of 3.5 wt % ^{235}U that was irradiated for four consecutive cycles. The other two samples, irradiated for three cycles, were taken from a rebuilt assembly with initial fuel enrichment of 4.1 wt% ^{235}U. The three samples analyzed span the burnup range 30–60 GWd/MTU.

2.2 REBUS

The REBUS International Program[2] coordinated by Belgonucleaire was dedicated to the validation of computer codes for criticality calculations that take into account the reduction of reactivity of spent fuel as a result of burnup credit. Participants in REBUS included institutes from Belgium, France, Germany, Japan, and the United States. ORNL was a participant in the early stages of the program under support from the U.S. Nuclear Regulatory Commission (NRC) and negotiated access to the data from this program. The REBUS program was completed in December 2005.

REBUS involved critical measurements in the VENUS critical facility at SCK-CEN using spent fuel rod segments. One of the segments was assayed to experimentally determine the isotopic content of the fuel. The results for this sample, measured by the SCK-CEN laboratory in Belgium, were reported. The sample was obtained from a fuel rod of an 18 × 18 PWR assembly operated in the German reactor Gemeinschaftskernkraftwerk Unit II (GKN II) in Neckarwestheim/Neckar. Although this reactor currently operates with a MOX core, the assembly was obtained from the reactor during a period when it operated with only UO_2 fuel. The measured sample had an initial enrichment of 3.8 wt % ^{235}U and a burnup of about 54 GWd/MTU.

3. ISOTOPIC MEASUREMENTS

3.1 Gösgen (ARIANE) Samples

Three UO_2 samples, identified as GU1, GU3, and GU4, were measured in the ARIANE program. Duplicate measurements for sample GU3 were carried out at two different facilities, SCK-CEN in Belgium and ITU in Germany. Measurements for sample GU1 were performed at SCK-CEN, and measurements for sample GU4 were carried out at ITU.

The following main experimental techniques have been applied for measurements performed at SCK-CEN:

- Thermal ionization mass spectrometry (TIMS)
  - major (uranium, plutonium) and minor (americium and ^{245,246}Cm) actinides
  - lanthanides: neodymium, samarium, ^{144}Ce, ^{155}Gd, ^{151}Eu, ^{153}Eu
  - cesium nuclides: $^{133\text{-}135}$Cs
- Inductively coupled plasma mass spectrometry (ICP-MS) with external calibration
  - metallics: ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{125}Sb
  - ^{237}Np
- γ-spectrometry
  - ^{106}Ru, ^{137}Cs, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{243}Cm
- α-spectrometry
  - ^{242}Cm, ^{244}Cm
- β-spectrometry
  - ^{90}Sr

The following two main experimental techniques have been used for measurements performed at ITU:

- TIMS
  - major actinides (uranium, plutonium)
- ICP-MS with IDA (isotope dilution analysis)
  - all other measured nuclides

Because of the variety of the analysis techniques, the varying properties of the nuclides being analyzed, and their differing concentrations, uncertainties in the measured concentrations can vary considerably. Table 3.1 lists the measurement method used and the experimental uncertainty, expressed both as uncertainty at 95% confidence level, as reported,[1] and as relative standard deviation, calculated as half of the 95% confidence level uncertainty reported. Only the maximum uncertainty corresponding to the measurements at each laboratory is shown in Table 3.1. The nuclide concentrations were reported both in mg/g fuel and mg/g U in the measured sample for most of the measured isotopes. For metallic fission products, however, the values reported in the final set of data were in mg/g fuel only; these values represent a combination of the separate measurements done on the main solution and undissolved residue.

The experimental isotope concentrations in mg/g fuel are presented in Table 3.2. For samples GU1 and GU4, the data shown in the table (as reported) correspond to measurement date, except for ^{106}Ru, ^{125}Sb, and ^{147}Pm, for which they correspond to discharge. For sample GU3, most of the isotopes considered by the program were measured at both SCK-CEN and ITU. For the isotopes with two independent measurements, the recommended values were established by consensus of experts participating in the program, based on a detailed cross-check analysis of the measurements. The cross-check was based on a comparison of the 95% confidence intervals associated to the measured values. If there was an intersection zone between the two 95% confidence intervals, the concentration results were combined in a weighted average. If the two concentration values were outside this intersection zone, either only one of the two values was recommended based on a detailed analysis of the measurement process or both values were maintained without recommendation.

The cross-checked values were reported either at measurement date or discharge. The isotope concentration data shown in Table 3.2 for sample GU3, as reported, corresponds to the discharge date for the following isotopes: ^{241}Pu, ^{242m}Am, ^{242,243,244}Cm, ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{134,137}Cs, ^{144}Ce, ^{147}Pm, ^{151}Sm, ^{154,155}Eu; for the other considered isotopes, the data correspond to the most recent (longer cooling time) of the two dates at which measurements were performed at the two laboratories. There were four nuclides (^{244,245}Cm, ^{133}Cs, and ^{155}Gd) measured in sample GU3 for which no recommendations were provided. For these four nuclides, the data shown in Table 3.2 were calculated as weighted averages of the two results provided by the program as:

$$c_{avg} = \left(\frac{c_1}{\sigma_1^2} + \frac{c_2}{\sigma_2^2} \right) / \left(\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} \right) , \tag{3-1}$$

where c_1 and c_2 are the reported concentration values and σ_1 and σ_2 the corresponding relative experimental errors. Note that the two reported concentrations for these four nuclides differed by about 6% for ^{133}Cs, 14% for ^{155}Gd, and 20% for ^{244,245}Cm. The measured isotopic data presented in Table 3.2 are also shown in Table 3.3 in units of g/g $U_{initial}$, using as a basis the uranium mass in the sample before the irradiation. The unit conversion from mg/g fuel to g/g $U_{initial}$ was done as[1]

$$m(g / gU_{initial}) = 1.1345 \times 10^{-3} m(mg / g_{fuel}) . \tag{3-2}$$

The measurement dates and the time duration from discharge to the measurement date for each of the analyzed nuclides and samples are provided in Table 3.4.

The material balance for the ARIANE Gösgen samples was confirmed[1] using two independent measures to verify the consistency of the experimental data. The material balance ratio was calculated as

$$MB = \frac{1.1345(W_U + W_{Pu} + W_{MA} + \Delta W)}{W_{sample}^{total}} , \tag{3-3}$$

where W_U, W_{Pu}, and W_{MA} are the weights of the uranium, plutonium, and minor actinides (americium and curium) measured in the dissolved solution, ΔW is the loss on the initial uranium mass due to fission, and W_{sample}^{total} is the actual mass of the fuel sample as measured on the mass balance. The coefficient 1.1345 represents the approximate ratio of the fuel weight to uranium weight. The loss due to fission, ΔW, was determined using the measured concentrations of the burnup indicator fission product ^{148}Nd. The fuel mass ratio obtained for samples GU3 and GU4 (all laboratories) was 1.00; however, the ratio obtained for sample GU1 was 1.12, indicating that the mass derived from the sum of measured actinides was about 12% greater than the actual measured fuel sample mass. The experimental data was therefore adjusted to the initial fuel mass as derived from the heavy metal isotopic measurements. The only plausible source of such significant error in the isotopic data would be the absolute measured mass of uranium in the solution.

Table 3.1 Experimental techniques and uncertainties for Gösgen (ARIANE) samples

Nuclide ID	Measurements at SCK/CEN			Measurements at ITU		
	Method[a]	Uncertainty[b] 95% confidence (%)	RSD[c] (%)	Method	Uncertainty[b] 95% confidence (%)	RSD (%)
U-234	TIMS	5.02	2.51	TIMS	0.02	0.01
U-235	TIMS	2.05	1.03	TIMS	2.40	1.20
U-236	TIMS	0.67	0.34	TIMS	1.57	0.79
U-238	TIMS	0.45	0.23	TIMS	0.02	0.01
Pu-238	TIMS	3.05	1.53	TIMS	2.15	1.08
Pu-239	TIMS	0.57	0.29	TIMS	0.51	0.26
Pu-240	TIMS	0.57	0.29	TIMS	0.51	0.26
Pu-241	TIMS	0.57	0.29	TIMS	3.40	1.70
Pu-242	TIMS	0.59	0.29	TIMS	0.55	0.28
Np-237	ICP-MS	20.60	10.30	ICP-MS	9.61	4.81
Am-241	TIMS	3.56	1.78	ICP-MS	11.87	5.94
Am-242m	TIMS	10.60	5.30			
Am-243	TIMS	3.56	1.78	ICP-MS	13.29	6.65
Cm-242	α-spec	7.22	3.61			
Cm-243	γ-spec	73.49	36.75			
Cm-244	α-spec	3.24	1.62	ICP-MS	12.85	6.43
Cm-245	TIMS	5.89	2.95	ICP-MS	20.29	10.15
Cm-246	TIMS	20.24	10.12			
Cs-133	TIMS	4.91	2.46	ICP-MS	3.27	1.64
Cs-134	TIMS	4.91	2.46	ICP-MS	8.20	4.10
Cs-135	TIMS	4.91	2.46	ICP-MS	3.29	1.65
Cs-137	γ-spec	4.90	2.45	ICP-MS	3.00	1.50
Ce-144	γ-spec	7.84	3.92	ICP-MS	7.49	3.75
Nd-142	TIMS	10.01	5.01	ICP-MS	10.18	5.09
Nd-143	TIMS	0.57	0.29	ICP-MS	12.32	6.16
Nd-144	TIMS	0.57	0.29	ICP-MS	11.89	5.95
Nd-145	TIMS	0.57	0.29	ICP-MS	11.78	5.89
Nd-146	TIMS	0.57	0.29	ICP-MS	14.73	7.37
Nd-148	TIMS	0.59	0.30	ICP-MS	13.4	6.70
Nd-150	TIMS	0.59	0.30	ICP-MS	13.55	6.78
Pm-147	β-spec	18.01	9.00	ICP-MS	13.51	6,76
Sm-147	TIMS	0.64	0.32	ICP-MS	21.14	10.57
Sm-148	TIMS	0.64	0.32	ICP-MS	8.01	4.01
Sm-149	TIMS	2.09	1.05	ICP-MS	42.83	21.42
Sm-150	TIMS	0.64	0.32	ICP-MS	6.87	3.44
Sm-151	TIMS	0.79	0.40	ICP-MS	67.63	33.82
Sm-152	TIMS	0.64	0.32	ICP-MS	6.41	3.21
Sm-154	TIMS	0.66	0.33	ICP-MS	11.3	5.65
Eu-151	TIMS	2.10	1.05			
Eu-153	TIMS	0.67	0.34	ICP-MS	10.97	5.49
Eu-154	γ-spec	5.29	2.65	ICP-MS	23.73	11.87
Eu-155	γ-spec	9.83	4.92	ICP-MS	32.13	16.07
Gd-155	TIMS	5.00	2.50	ICP-MS	13.72	6.86
Sr-90	β-spec	16.01	8.01	ICP-MS	0.77	0.39
Mo-95	ICP-MS	9.14	4.57	ICP-MS	2.20	1.10
Tc-99	ICP-MS	17.7	8.85	ICP-MS	1.78	0.89
Ru-101	ICP-MS	24.42	12.21	ICP-MS	1.88	0.94
Ru-106	γ-spec	28.41	14.21	ICP-MS	8.18	4.09
Rh-103	ICP-MS	9.77	4.89	ICP-MS	6.53	3.27
Ag-109	ICP-MS	18.12	9.06			
Sb-125	ICP-MS	18.85	9.43			

[a] Main technique is mentioned; some nuclides required multiple techniques to eliminate interferences.
[b] The maximum of the values for the two UO_2 samples measured at this facility is shown.
[c] Relative standard deviation.

Table 3.2 Experimental results (mg/g fuel) for Gösgen (ARIANE) samples

Sample ID	GU1		GU3		GU4	
Burnup[a]	59.7		52.5		29.1	
Enrichment (wt% ^{235}U)	3.5		4.1		4.1	
Measuring lab	SCK/CEN		SCK/CN & ITU		ITU	
Nuclide ID	mg/g fuel	% error[b]	mg/g fuel	% error	mg/g fuel	% error
U-234	1.06E-01	5.02	1.26E-01	0.02	1.72E-01	0.02
U-235	1.86E+00	2.05	5.33E+00	0.64	1.28E+01	0.89
U-236	4.26E+00	0.67	4.98E+00	0.61	4.05E+00	0.89
U-238	8.11E+02	0.45	8.17E+02	0.02	8.32E+02	0.02
Pu-238	4.00E-01	3.05	3.28E-01	0.55	9.80E-02	2.15
Pu-239	4.31E+00	0.57	5.12E+00	0.38	4.55E+00	0.47
Pu-240	2.80E+00	0.57	2.50E+00	0.30	1.62E+00	0.51
Pu-241	1.27E+00	0.57	1.60E+00	0.56	8.70E-01	3.40
Pu-242	1.37E+00	0.57	8.95E-01	0.04	2.73E-01	0.55
Np-237			7.15E-01	6.00	4.63E-01	4.81
Am-241	2.19E-01	3.56	2.01E-01	1.58	1.30E-01	11.87
Am-242m	6.04E-04	10.60	8.20E-04	10.58		
Am-243	3.55E-01	3.56	2.10E-01	3.48	3.86E-02	13.29
Cm-242	2.72E-04	7.22	2.43E-02	4.03		
Cm-243	2.98E-04	73.49	5.50E-04	19.21		
Cm-244	2.15E-01	3.02	1.24E-01	3.14	1.09E-02	3.13
Cm-245	1.54E-02	5.89	9.69E-03	2.86	5.06E-04	20.29
Cm-246	4.66E-03	20.24	1.27E-03	10.52		
Cs-133	1.52E+00	4.91	1.44E+00	1.87	9.54E-01	3.27
Cs-134	9.56E-02	4.94	2.21E-01	2.87	3.65E-02	2.54
Cs-135	4.55E-01	4.91	4.13E-01	2.24	3.29E-01	2.41
Cs-137	1.79E+00	4.90	1.65E+00	1.04	8.77E-01	3.00
Ce-144	2.97E-02	7.84	3.89E-01	2.01	3.20E-02	7.49
Nd-142	5.97E-02	10.01	3.73E-02	10.01	1.06E-02	10.18
Nd-143	8.22E-01	0.57	9.45E-01	0.56	7.60E-01	12.32
Nd-144	2.32E+00	0.57	1.89E+00	0.56	1.08E+00	11.89
Nd-145	9.18E-01	0.57	8.72E-01	0.56	5.96E-01	11.78
Nd-146	1.17E+00	0.57	1.01E+00	0.56	5.73E-01	14.73
Nd-148	5.87E-01	0.59	5.17E-01	0.58	3.06E-01	13.40
Nd-150	2.99E-01	0.59	2.52E-01	0.58	1.39E-01	13.55
Pm-147	1.21E-01	10.25	1.70E-01	18.01	1.78E-01	13.51
Sm-147	1.96E-01	0.64	1.73E-01	0.64	1.42E-01	21.14
Sm-148	2.86E-01	0.64	2.24E-01	0.64	9.74E-02	8.01
Sm-149	2.89E-03	2.09	2.96E-03	2.09	2.66E-03	11.76
Sm-150	4.48E-01	0.64	3.93E-01	0.64	2.14E-01	6.87
Sm-151	1.15E-02	0.79	1.30E-02	0.81	9.94E-03	4.41
Sm-152	1.46E-01	0.64	1.18E-01	0.64	8.34E-02	6.41
Sm-154	7.09E-02	0.66	5.05E-02	0.66	2.33E-02	11.30
Eu-151	6.33E-04	2.10	3.70E-04	2.10		
Eu-153	1.85E-01	0.67	1.62E-01	0.66	8.28E-02	10.97
Eu-154	2.84E-02	3.89	3.79E-02	1.53	1.22E-02	23.73
Eu-155	9.95E-03	5.28	1.35E-02	9.43	3.88E-03	7.28
Gd-155	4.96E-03	5.00	3.46E-03	1.99	2.33E-03	13.72
Sr-90	8.57E-01	15.00	6.83E-01	0.63	4.45E-01	0.77
Mo-95	1.08E+00	7.74	1.04E+00	2.94	6.68E-01	3.11
Tc-99	1.10E+00	12.60	9.83E-01	3.87	5.28E-01	2.35
Ru-101	1.14E+00	9.15	1.07E+00	3.49	6.60E-01	4.00
Ru-106	2.26E-01	5.64	2.56E-01	28.41	1.14E-01	5.37
Rh-103	5.40E-01	8.98	4.76E-01	4.88	4.00E-01	4.71
Ag-109	6.62E-02	10.35	1.05E-01	18.12		
Sb-125	8.19E-03	10.14	6.61E-03	18.85		

[a] In GWd/MTU; as reported in *ARIANE International Programme–Final Report*, ORNL/SUB/97-XSV750-1, Oak Ridge National Laboratory, Oak Ridge, Tennessee (May 1, 2003).
[b] Reported uncertainty at 95% confidence level.

Table 3.3 Experimental results (g/g $U_{initial}$) for Gösgen (ARIANE) samples

Sample ID	GU1		GU3		GU4	
Burnup[a]	59.7		52.5		29.1	
Enrichment (wt% ^{235}U)	3.5		4.1		4.1	
Measuring lab	SCK/CEN		SCK/CEN & ITU		ITU	
Nuclide ID	g/g $U_{initial}$	RSD[b] (%)	g/g $U_{initial}$	RSD (%)	g/g $U_{initial}$	RSD (%)
U-234	1.20E-04	2.51	1.43E-04	0.01	1.95E-04	0.01
U-235	2.11E-03	1.03	6.05E-03	0.32	1.45E-02	0.45
U-236	4.83E-03	0.34	5.65E-03	0.31	4.59E-03	0.45
U-238	9.20E-01	0.23	9.27E-01	0.01	9.44E-01	0.01
Pu-238	4.54E-04	1.53	3.72E-04	0.28	1.11E-04	1.08
Pu-239	4.89E-03	0.29	5.81E-03	0.19	5.16E-03	0.24
Pu-240	3.18E-03	0.29	2.84E-03	0.15	1.84E-03	0.26
Pu-241	1.44E-03	0.29	1.82E-03	0.28	9.87E-04	1.70
Pu-242	1.55E-03	0.29	1.02E-03	0.02	3.10E-04	0.28
Np-237			8.11E-04	3.00	5.25E-04	2.41
Am-241	2.48E-04	1.78	2.28E-04	0.79	1.47E-04	5.94
Am-242m	6.85E-07	5.30	9.30E-07	5.29		
Am-243	4.03E-04	1.78	2.38E-04	1.74	4.38E-05	6.65
Cm-242	3.09E-07	3.61	2.76E-05	2.02		
Cm-243	3.38E-07	36.75	6.24E-07	9.61		
Cm-244	2.44E-04	1.51	1.41E-04	1.57	1.24E-05	1.57
Cm-245	1.75E-05	2.95	1.10E-05	1.43	5.74E-07	10.15
Cm-246	5.29E-06	10.12	1.44E-06	5.26		
Cs-133	1.72E-03	2.46	1.63E-03	0.94	1.08E-03	1.64
Cs-134	1.08E-04	2.47	2.51E-04	1.44	4.14E-05	1.27
Cs-135	5.16E-04	2.46	4.69E-04	1.12	3.73E-04	1.21
Cs-137	2.03E-03	2.45	1.87E-03	0.52	9.95E-04	1.50
Ce-144	3.37E-05	3.92	4.41E-04	1.01	3.63E-05	2.75
Nd-142	6.77E-05	5.01	4.23E-05	5.01	1.20E-05	5.09
Nd-143	9.33E-04	0.29	1.07E-03	0.28	8.62E-04	6.16
Nd-144	2.63E-03	0.29	2.14E-03	0.28	1.23E-03	5.95
Nd-145	1.04E-03	0.29	9.89E-04	0.28	6.76E-04	6.89
Nd-146	1.33E-03	0.29	1.15E-03	0.28	6.50E-04	7.37
Nd-148	6.66E-04	0.30	5.87E-04	0.29	3.47E-04	6.70
Nd-150	3.39E-04	0.30	2.86E-04	0.29	1.58E-04	6.78
Pm-147	1.37E-04	5.13	1.93E-04	9.01	2.02E-04	
Sm-147	2.22E-04	0.32	1.96E-04	0.32	1.61E-04	10.57
Sm-148	3.24E-04	0.32	2.54E-04	0.32	1.11E-04	4.01
Sm-149	3.28E-06	1.05	3.36E-06	1.05	3.02E-06	5.88
Sm-150	5.08E-04	0.32	4.46E-04	0.32	2.43E-04	3.44
Sm-151	1.30E-05	0.40	1.47E-05	0.41	1.13E-05	2.21
Sm-152	1.66E-04	0.32	1.34E-04	0.32	9.46E-05	3.21
Sm-154	8.04E-05	0.33	5.73E-05	0.33	2.64E-05	5.65
Eu-151	7.18E-07	1.05	4.20E-07	1.05		
Eu-153	2.10E-04	0.34	1.84E-04	0.33	9.39E-05	5.49
Eu-154	3.22E-05	1.95	4.30E-05	0.77	1.38E-05	11.87
Eu-155	1.13E-05	2.64	1.53E-05	4.72	4.40E-06	4.64
Gd-155	5.63E-06	2.50	3.93E-06	1.00	2.64E-06	6.81
Sr-90	9.72E-04	7.50	7.75E-04	0.32	5.05E-04	
Mo-95	1.23E-03	3.87	1.18E-03	1.47	7.58E-04	1.56
Tc-99	1.25E-03	6.30	1.12E-03	1.94	5.99E-04	1.18
Ru-101	1.29E-03	4.58	1.21E-03	1.75	7.49E-04	2.00
Ru-106	2.56E-04	2.82	2.90E-04	14.21	1.29E-04	2.69
Rh-103	6.13E-04	4.49	5.40E-04	2.44	4.54E-04	2.36
Ag-109	7.51E-05	5.18	1.19E-04	9.06		
Sb-125	9.29E-06	5.07	7.50E-06	9.43		

[a] In GWd/MTU; as reported in *ARIANE International Programme–Final Report*, ORNL/SUB/97-XSV750-1, Oak Ridge National Laboratory, Oak Ridge, Tennessee (May 1, 2003).
[b] Relative standard deviation.

Table 3.4 Decay time data for Gösgen (ARIANE) samples

Sample ID	*GU1*		*GU3*				*GU4*	
Experimental facility	*SCK/CEN*		*SCK/CEN*		*ITU*		*ITU*	
Nuclides	**Measurement date (month/day/year)**	**Decay time (days)**	**Measurement date (month/day/year)**	**Decay time (days)**	**Measurement date (month/day/year)**	**Decay time (days)**	**Measurement date (month/day/year)**	**Decay time (days)**
Uranium	4/9/97	1040	10/12/99	857	1/28/99	600	5/20/99	712
Plutonium	4/22/97	1053	10/11/99	856	1/28/99	600	8/17/99	801
Neptunium			12/22/99	928	6/16/99	739	6/16/99	739
Americium	4/9/97	1040	12/21/99	927	6/16/99	739	6/16/99	739
Curium	6/4/97	1096	7/1/99	754	6/16/99	739	6/16/99	739
Neodymium	4/11/97	1042	11/24/99	900	3/26/99	657	9/30/99	845
Cesium	5/30/97	1091	7/1/99	754	5/4/99	696	10/6/99	851
Cerium	2/28/97	1000	7/1/99	754	5/3/99	695	9/30/99	845
Samarium	4/23/97	1054	12/13/99	919	5/4/99	696	10/1/99	846
Europium	4/23/97	1054	7/1/99	754	5/4/99	696	10/4/99	849
Gadolinium			12/1/99	907	10/6/99	851	10/4/99	849
Strontium	6/24/97	1116	4/28/00	1056	10/7/99	852	11/15/99	891
^{95}Mo,^{99}Tc,^{101}Ru	4/10/00	2137	4/10/00	1038	10/7/99	852	11/15/99	891
^{103}Rh,^{109}Ag	4/10/00	2137	4/10/00	1038	10/7/99	852	11/15/99	891
^{106}Ru,^{125}Sb[a]	2/28/97	1000	10/7/99	852	10/7/99	852	11/15/99	891

[a] This date correspond to measurements in the main solution. Measurements were also done on the residue. The reported measurement data at discharge time (0 days decay) was a combination of the data measured in both main solution and residue.

3.2 GKN II (REBUS) Sample

The GKN II sample was obtained from one of the inner rods of 18 × 18 PWR assembly 419, which was irradiated in the GKN II German reactor. The sample consisted of about three fuel pellets cut from the fuel rod identified as M11. The reported sample burnup was about 54 GWd/MTU. Radiochemical analyses of this sample were performed at SCK-CEN.

The selected sample was subjected to a two-step dissolution process followed by sample preparation for the various analytical techniques employed. The radiochemical analysis techniques included α- and γ-spectrometry, ICP-MS, and TIMS. For the actinides, the analysis was performed for isotopes of uranium, neptunium, plutonium, americium, and curium. The fission products that were analyzed were of two types: there were burnup indicators consisting of neodymium isotopes, as well as ^{137}Cs and ^{144}Ce; and there were absorbing fission products consisting of metallic species (^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{105}Pd, ^{108}Pd, and ^{109}Ag), ^{133}Cs, plus samarium, europium, and gadolinium isotopes. The metallic species were difficult to dissolve completely, and, as a result, the dissolution residue had to be analyzed separately.

Because of the variety of the analysis techniques, the varying properties of the nuclides being analyzed, and their differing concentrations, uncertainties in the measured concentrations vary greatly. Table 3.5 lists the measurement method and, for each of the measured nuclides, the reported experimental uncertainty at 95% confidence level, corresponding to the experimental results reported in mg/g ^{238}U (Ref. 3). Also shown in Table 3.5 is the relative standard deviation calculated as half of the reported 95% confidence level uncertainty.

Nuclide concentrations were reported both in mg/g fuel and mg/g ^{238}U in the sample at the measurement date. However, the REBUS report[3] on isotopic measurements recommends use of values reported in mg/g ^{238}U for further calculations because these values do not include uncertainties resulting from manipulations or spills during dissolution or dilution of the sample. The measured data reported in mg/g ^{238}U are presented in Table 3.6. For the purpose of comparison to measured data from other programs, the experimental data for the GKN II sample are also presented in g/g $U_{initial}$ units in Table 3.6. The unit conversion was done as[1]

$$\frac{m_i}{\sum_k m_{U_k} + \sum_l m_{Pu_l} + \sum_m m_{Am_m} + \sum_n m_{Cm_n} + 238\frac{m_{^{148}Nd}}{148\bar{Y}}}, \qquad (3\text{-}4)$$

where m_i is the mass of nuclide i, as reported in mg/g ^{238}U. The denominator in Eq. (3-4) is the initial uranium content derived as a sum of the actinide (U, Pu, Am, Cm) concentrations in the measured sample and the weight loss in initial uranium due to burnup. The weight loss due to burnup is approximated by $238\frac{m_{^{148}Nd}}{148\bar{Y}}$, where $\bar{Y}$ is the average fission yield of ^{148}Nd. A value $\bar{Y} = 0.0176$ is recommended[1] for PWR UO_2 fuel. Note that $m_{U_{238}} = 1000$ in Eq. (3-4).

The measurement date and the time duration from discharge to the measurement date for each of the analyzed nuclides is provided in Table 3.7.

Table 3.5 Experimental techniques and uncertainties for GKN II (REBUS) sample

Nuclide ID	Method [a]	Uncertainty [b] at 95% confidence level (%)	RSD [c] (%)
U-234	TIMS	5.0	2.5
U-235	TIMS	0.73	0.37
U-236	TIMS	0.73	0.37
U-238	TIMS	0.57	0.29
Total U		0.53	0.22
Np-237	ICP-MS	20.0	10.0
Pu-238	TIMS, α-spec	3.1	1.6
Pu-239	TIMS	0.59	0.30
Pu-240	TIMS	0.59	0.30
Pu-241	TIMS	0.59	0.30
Pu-242	TIMS	0.61	0.31
Am-241	TIMS	3.5	1.8
Am-242m	TIMS	11.0	5.5
Am-243	TIMS	3.5	1.8
Cm-242	α-spec	32.0	16.0
Cm-243	γ-spec	20.0	10.0
Cm-244	α-spec	2.5	1.3
Cm-245	TIMS	5.6	2.8
Mo-95	ICP-MS	9.9	5.0
Tc-99	ICP-MS	10.0	5.0
Ru-101	ICP-MS	9.9	5.0
Rh-103	ICP-MS	10.0	5.0
Pd-105	ICP-MS	9.8	4.9
Pd-108	ICP-MS	9.8	4.9
Ag-109	ICP-MS	10.0	5.0
Cs-133	TIMS	2.6	1.3
Cs-134	γ-spec	2.6	1.3
Cs-137	γ-spec	2.6	1.3
Nd-142	TIMS	0.78	0.39
Nd-143	TIMS	0.64	0.32
Nd-144	TIMS	0.64	0.32
Nd-145	TIMS	0.64	0.32
Nd-146	TIMS	0.64	0.32
Nd-148	TIMS	0.64	0.32
Nd-150	TIMS	0.65	0.33
Ce-144	γ-spec	10.0	5.0
Sm-147	TIMS	0.75	0.38
Sm-148	TIMS	0.75	0.38
Sm-149	TIMS	2.13	1.07
Sm-150	TIMS	0.75	0.38
Sm-151	TIMS	0.88	0.44
Sm-152	TIMS	0.75	0.38
Sm-154	TIMS	0.76	0.38
Eu-153	TIMS	0.9	0.5
Eu-154	γ-spec	3.4	1.7
Eu-155	γ-spec	6.0	3.0
Gd-155	TIMS	5.0	2.5

[a] Main technique is listed; some nuclides may require multiple techniques to eliminate interferences.
[b] As reported for the measured data expressed in mg/g ^{238}U in *REBUS International Program—Reactivity Tests for a Direct Evaluation of the Burnup Credit on Selected Irradiated LWR Fuel Bundles, Destructive Radiochemical Spent Fuel Characterization of a PWR UO_2 Fuel Sample*, SCK-CEN, Belgonucleaire (May 2006).
[c] Relative standard deviation.

Table 3.6 Experimental results for GKN II (REBUS) sample

Nuclide	Concentration[a] (mg/g ^{238}U)	RSD[b] (%)	Concentration[c] (g/g $U_{initial}$)	RSD[d] (%)
U-234	0.162	2.5	1.49E-04	2.52
U-235	5.56	0.37	5.13E-03	0.46
U-236	5.81	0.37	5.36E-03	0.46
U-238	1000	0.29	9.22E-01	2.52
Np-237	0.66	10.0	6.09E-04	10.0
Pu-238	0.465	1.6	4.29E-04	1.58
Pu-239	6.26	0.30	5.77E-03	0.41
Pu-240	3.49	0.30	3.22E-03	0.41
Pu-241	1.407	0.30	1.30E-03	0.41
Pu-242	1.271	0.31	1.17E-03	0.42
Am-241	0.57	1.8	5.26E-04	1.77
Am-242m	0.00170	5.5	1.57E-06	5.51
Am-243	0.270	1.8	2.49E-04	1.77
Cm-242	4.7E-06	16.0	4.33E-09	16.00
Cm-243	8.4E-04	10.0	7.75E-07	10.00
Cm-244	0.144	1.3	1.33E-04	1.28
Cm-245	0.0144	2.8	1.33E-05	2.81
Mo-95	1.13	5.0	1.04E-03	5.01
Tc-99	1.36	5.0	1.25E-03	5.01
Ru-101	1.05	5.0	9.68E-04	5.01
Rh-103	0.63	5.0	5.81E-04	5.01
Pd-105	0.49	4.9	4.52E-04	5.01
Pd-108	0.192	4.9	1.77E-04	5.01
Ag-109	0.116	5.0	1.07E-04	5.01
Cs-133	1.74	1.3	1.60E-03	1.33
Cs-135	0.625	1.3	5.76E-04	1.33
Cs-137	1.82	1.3	1.68E-03	1.33
Ce-144	5.3E-04	5.0	4.89E-07	5.01
Nd-142	0.0566	0.39	5.22E-05	0.48
Nd-143	1.162	0.32	1.07E-03	0.43
Nd-144	2.449	0.32	2.26E-03	0.43
Nd-145	1.081	0.32	9.97E-04	0.43
Nd-146	1.276	0.32	1.18E-03	0.43
Nd-148	0.647	0.33	5.97E-04	0.43
Nd-150	0.320	0.33	2.95E-04	0.43

Table 3.6 Experimental results for the GKN II (REBUS) sample (continued)

Nuclide	Concentration[a] (mg/g ^{238}U)	RSD[b] (%)	Concentration[c] (g/g $U_{initial}$)	RSD[d] (%)
Sm-147	0.324	0.38	2.99E-04	0.47
Sm-148	0.313	0.38	2.89E-04	0.47
Sm-149	0.00259	1.07	2.39E-06	1.10
Sm-150	0.518	0.38	4.78E-04	0.47
Sm-151	0.01551	0.44	1.43E-05	0.52
Sm-152	0.1598	0.38	1.47E-04	0.47
Sm-154	0.0727	0.38	6.70E-05	0.48
Eu-153	0.2086	0.5	1.92E-04	0.53
Eu-154	0.0250	1.7	2.31E-05	1.72
Eu-155	0.0067	3.0	6.18E-06	3.01
Gd-155	0.0110	2.5	1.01E-05	2.52

[a] As reported in *REBUS International Program—Reactivity Tests for a Direct Evaluation of the Burnup Credit on Selected Irradiated LWR Fuel Bundles, Destructive Radiochemical Spent Fuel Characterization of a PWR UO_2 Fuel Sample*, SCK-CEN, Belgonucleaire (May 2006).
[b] Relative standard deviation.
[c] Calculated using Eq. (3-4).
[d] Accounts for reported error in measured ^{238}U.

Table 3.7 Decay time data for GKN II (REBUS) sample

Measurement date (month/day/year)	Decay time (days)	Measured nuclides
9/28/2004	2600	^{144}Ce, ^{154}Eu, ^{155}Eu, ^{137}Cs
9/29/2004	2601	^{242}Cm, ^{244}Cm
11/02/2004	2635	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu
11/15/2004	2648	^{133}Cs, ^{135}Cs
12/09/2004	2672	^{234}U, ^{235}U, ^{236}U, ^{238}U
2/10/2005	2735	^{147}Sm, ^{148}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{154}Sm, ^{153}Eu, ^{155}Gd
2/28/2005	2753	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd
3/07/2005	2760	^{243}Cm, ^{241}Am, ^{242m}Am, ^{243}Am
4/29/2005	2813	^{237}Np, ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{105}Pd, ^{108}Pd, ^{109}Ag
6/01/2005	2846	^{245}Cm

4. ASSEMBLY AND IRRADIATION HISTORY DATA

This section presents information on the fuel assembly geometry, irradiation history, and sample burnup that is necessary for developing a computational model to calculate the isotopic composition of the samples under consideration. For the cases in which insufficient information was available, assumptions are stated.

4.1 Gösgen (ARIANE) Samples

Three UO_2 samples were measured for the ARIANE program, identified as GU1, GU3, and GU4. Samples GU3 and GU4 were from the same fuel rod. The layout of the assembly, showing the location of the measured rod at the beginning of cycles 12 and 16 for samples GU1 and GU3 (GU4), respectively, is illustrated in Figure 4.1. Assembly geometry and fuel data are presented in Table 4.1. Table 4.2 shows the operating history data for sample GU1 as provided[1]: irradiation cycle start and end dates, actual cycle duration and down days, effective full power days and down days, core load factor, concentration of soluble boron in the moderator, operator estimated sample burnup, and sample fuel temperature. The same type of information is presented in Table 4.3 for samples GU3 and GU4.

Sample GU1 was selected from a fuel rod with 3.5 wt % ^{235}U initial enrichment of assembly 1240, which was irradiated in the reactor for four consecutive cycles, from cycle 12 to cycle 15. The sample was cut from an axial location at about 97.7 cm from the bottom of the active region of the fuel rod. There were several changes in the fuel rod configuration of assembly 1240 during cycles 14 and 15: in each of these cycles, three fuel rods were replaced by irradiated fuel rods from other assemblies, as specified in Ref. 1. At the start of cycle 14, three fuel rods corresponding to assembly 1240 positions L12, M12, and N12, which were adjacent to the GU1 rod position M13 (see Figure 4.1), were replaced. After cycle 14, the rods at positions N12, K14, and L14 were also replaced. The reconfiguration of the rods is potentially of consequence to the analysis because of the close proximity of the replacement rods to the measured rod, and the potential influence on the local neutronic environment of the measured sample. Further review found that the replacement rods, in general, had a burnup similar to that of the original rods for the nearest neighbors (rods located at M12 and M14) of the M13 rod. Based on diagrams provided in Ref. 1, the burnup of these above mentioned neighboring rods did not differ by more than 3–4% from the burnup of rods placed in symmetric locations, with respect to the location of the rod from which sample GU1 was cut. Because additional details were not available (e.g., location of replacement rods from the donor assemblies), reconfiguration of the rods was not simulated in the computational analysis. Although the potential impact on the analysis results is believed to be minor, additional uncertainties introduced during the irradiation of the GU1 sample need to be considered when evaluating the data.

Samples GU3 and GU4 were selected from different axial locations of a single fuel rod irradiated in the Gösgen reactor for three consecutive cycles: cycle 16 to cycle 18. During cycles 16 and 17, this rod belonged to assembly 1601 with an initial fuel enrichment of 4.1 wt % ^{235}U, whereas during last cycle 18, it was part of a different assembly identified as 1701 with an initial fuel enrichment of 4.3 wt % ^{235}U. The assemblies had a 15 × 15 configuration, with 205 fuel rods and 20 guide tubes. The estimated axial locations for samples GU3 and GU4 are 127.42 cm and 7.42 cm, respectively, from the bottom of the active fuel region.

Four rods from assembly 1601, including the rod from which samples were selected, were taken out of the assembly after cycle 17 and inserted into assembly 1701. The rod from which samples GU3 and GU4 were selected at the end of cycle 18 changed location, with respect to the layout shown in Figure 4.1, from P7 in assembly 1601 to R11 in assembly 1701. The other three replacement rods in assembly 1701 that were transferred from assembly 1601 into assembly 1701 at the end of cycle 17 were located at N9, N12, and S13 in assembly 1701. Assembly 1701 is known to have had an average burnup at the beginning of cycle 18 of about 20.0 GWd/MTU at the axial level of sample GU3 and about 9.7 GWd/MTU at axial level of sample GU4. [1]

The temperature T of the moderator at the sample axial location z with respect to the bottom of the active fuel region was calculated as[4]

$$T(z) = T_{in} + \frac{T_{out} - T_{in}}{2}\left(1 - \cos\pi\frac{z}{L}\right), \tag{4-1}$$

where T_{in} and T_{out} are the inlet and outlet coolant temperatures, and L is the active fuel rod length. Based on the moderator temperature value for each sample, the corresponding moderator density was calculated by using tabulated temperature vs. pressure data[5] corresponding to a 154×10^5 Pa operating system pressure.

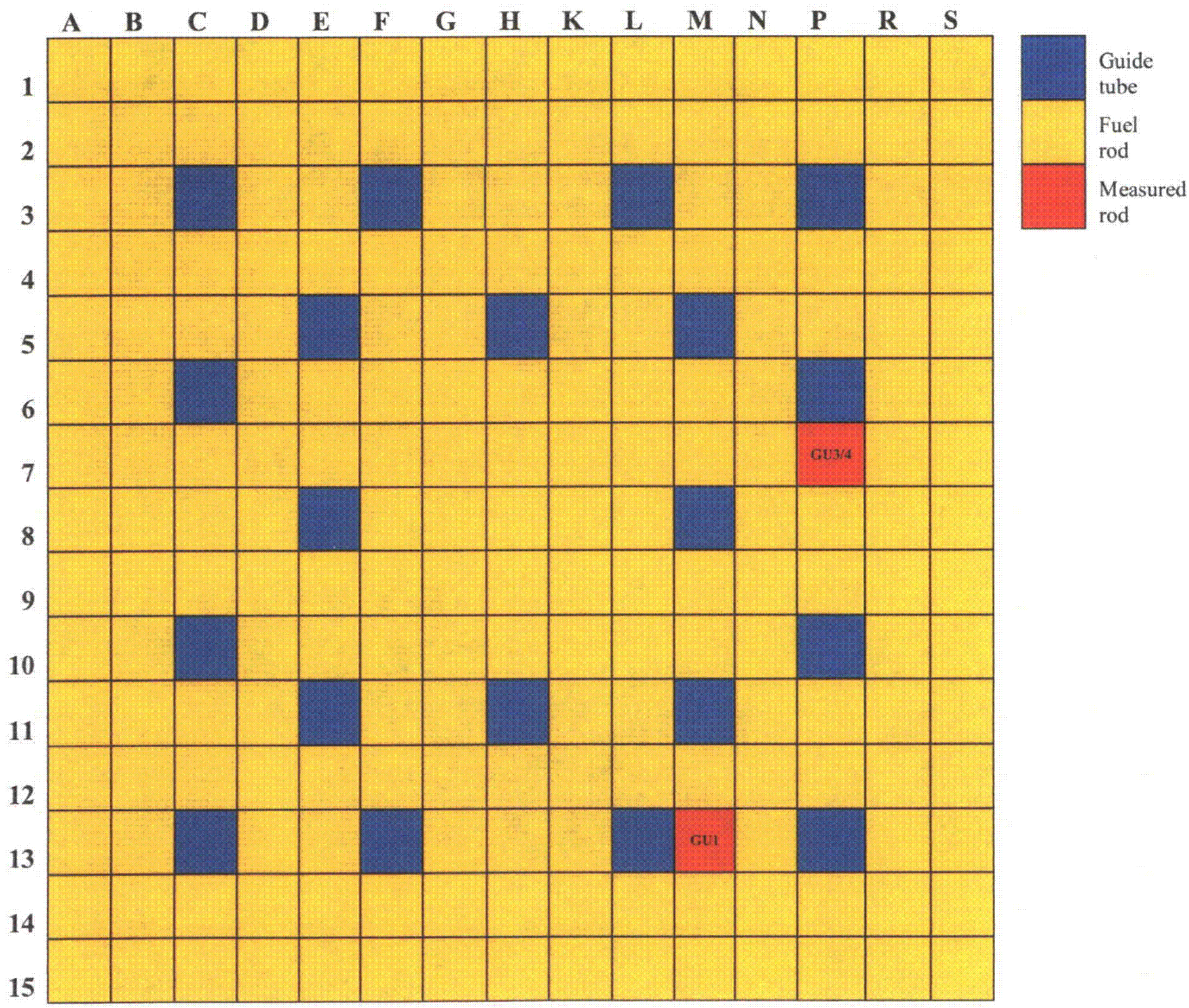

Figure 4.1 Assembly layout for Gösgen (ARIANE) samples

Table 4.1 Assembly design data for Gösgen (ARIANE) samples

Parameter	Data for GU1	Data for GU3/4
Assembly and reactor data		
Reactor	Gösgen	Gösgen
Operating pressure (Pa)	154×10^5	154×10^5
Lattice geometry	15×15	15×15
Rod pitch (cm)	1.43	1.43
Number of fuel rods	205	205
Number of guide tubes	20	20
Active fuel rod length (cm)	340	355
Assembly pitch (cm)	21.56	21.56
Fuel rod data		
Fuel material type	UO_2	UO_2
Fuel pellet density (g/cm^3)	10.4	10.4
Fuel pellet diameter (cm)	0.913	0.911
Sample axial location [a] (cm)	97.7	127.42/7.42
Fuel temperature (K)	see Table 4.2	see Table 4.3
U isotopic composition (wt %)		
^{234}U	0.036	0.042
^{235}U	3.5	4.1
^{236}U	0.0	0.0
^{238}U	96.464	95.858
Clad material	Zircaloy-4	Zircaloy-4
Clad inner diameter (cm)	0.93	0.93
Clad outer diameter (cm)	1.075	1.075
Average clad temperature [b](K)	619	619
Moderator data		
Inlet temperature (K)	565	565
Outlet temperature (K)	599	599
Moderator density [c] (g/cm^3)	0.730	0.723/0.743
Moderator temperature [c] (K)	572	575/565
Soluble boron content (ppm)	see Table 4.2	see Table 4.3
Guide tube data		
Guide tube material	Zircaloy-4	Zircaloy-4
Inner diameter (cm)	1.24	1.24
Outer diameter (cm)	1.38	1.38

[a] With respect to the bottom of the active fuel region.
[b] Assumed value; maximum clad temperature as given in *1998 World Nuclear Industry Handbook.*[6]
[c] Corresponding to sample axial location.

Table 4.2 Operating history data for Gösgen (ARIANE) sample GU1

Cycle no.	Start date	End date	Duration (days)	Down (days)	Effective full power days	Effective[a] down days	Load factor (%)	Soluble boron in coolant (ppm)	Sample GU1 Nominal burnup (GWd/MTU)	Sample GU1 Fuel temperature (K)
					0			1511		1151.3
					6		100	1179		1171.5
12	07/06/90	06/01/91	330	32	150	45	100	565		1136.0
					294.9		100	8		1078.3
					317		90.4	8	18.649	1046.7
					0			1477		919.3
					6		100	1145		967.7
13	07/03/91	05/30/92	332	16	150	27	100	542		957.9
					292.3		100	7		943.1
					321.3		87.3	7	33.594	842.0
					0			1517		888.9
					6		100	1178		894.4
14	06/15/92	06/05/93	355	26	150	50	100	549		854.8
					290.1		100	5		841.4
					331.3		72.0	5	47.911	709.8
					0			1594		806.6
					6		100	1243		829.8
15	07/01/93	06/04/94	338		150	11	100	605		810.6
					301.9		100	5		804.0
					326.7		87.0	5	59.656	738.9

[a] Sum of the actual down days and the difference between the actual cycle duration and effective full-power days.

Table 4.3 Operating history data for Gösgen (ARIANE) samples GU3 and GU4

Cycle no.	Start date	End date	Duration (days)	Down (days)	Effective full power days	Effective[a] down days	Load factor (%)	Soluble boron in coolant (ppm)	Sample GU3 Nominal burnup (GWd/MTU)	Sample GU3 Fuel temperature (K)	Sample GU4 Nominal burnup (GWd/MTU)	Sample GU4 Fuel Temperature (K)
					0			1705		1203.1		731.1
					6		100	1347		1244.1		782.0
16	06/29/94	06/10/95	346	25	150	34	100	690		1194.6		901.1
					320		100	5		1154.1		1008.5
					336.8		92.0	5	21.771	1065.2	11.248	919.9
					0			1601		1052.5		744.8
					6		100	1247		1068.5		786.9
17	07/05/95	06/08/96	339	22	150	32	100	602		1005.0		865.5
					299.5		100	9		978.7		949.8
					328.7		89.6	9	38.866	865.4	21.762	851.2
					0			1675		944.7		687.0
					6		100	1300		933.6		709.1
18	06/30/96	06/07/97	342		150	10	100	631		866.6		756.8
					301.2		100	17		858.0		805.6
					331.6		89.3	17	52.504	794.9	29.067	744.6

[a] Sum of the actual down days and the difference between the actual cycle duration and effective full-power days.

4.2 GKN II (REBUS) Sample

The radiochemical analysis was performed on a sample taken from a fuel rod identified as M11 of assembly 419 irradiated in the GKN II PWR reactor between August 1993 and August 1996. The sample was cut from an axial location on the fuel rod between 105.5 cm and 108.5 cm from the top end of the rod, which is approximately 300 cm from the bottom of the active fuel region. The estimated burnup[7] based on the measured ^{137}Cs gamma scan data was 54.1 GWd/MTU.

The assembly had an 18 × 18 configuration, as illustrated in Figure 4.2, with 300 fuel rods and 24 guide tubes. Twelve of the fuel rods contained Gd_2O_3 at 7.0 wt %. The rods with Gd_2O_3 had an initial fuel enrichment of 2.6 wt % ^{235}U; the regular fuel rods had an enrichment of 3.8 wt % ^{235}U. The composition of uranium in the fresh fuel was obtained from Ref. 8. The content of ^{234}U and ^{236}U in the fresh fuel for the gadolinia-bearing fuel rods was not available.

Assembly design data are listed in Table 4.4. The content of soluble boron in moderator as a function of the irradiation time is listed in Table 4.5, along with the sample cumulative burnup at the end of each cycle as reported by the utility.[2] The cycle duration and the sample cumulative burnup and average power values used in the calculations are shown in Table 4.6. The value for the burnup at the end of each cycle shown in Table 4.6 was obtained by normalizing the operator-based burnup data in Table 4.5 such that the sample final cumulative burnup corresponds to the reported value of 54.1 GWd/MTU based on the gamma scan. The cycle average fuel and moderator temperatures presented in Table 4.7 were calculated based on a more detailed time-dependent data[3] supplied by the utility for an axial location corresponding to the measured sample. Also shown in Table 4.7 are the moderator density data; they were calculated based on the moderator temperature by using temperature vs. pressure tabulated data[5] corresponding to the operating system pressure of 158×10^5 Pa.

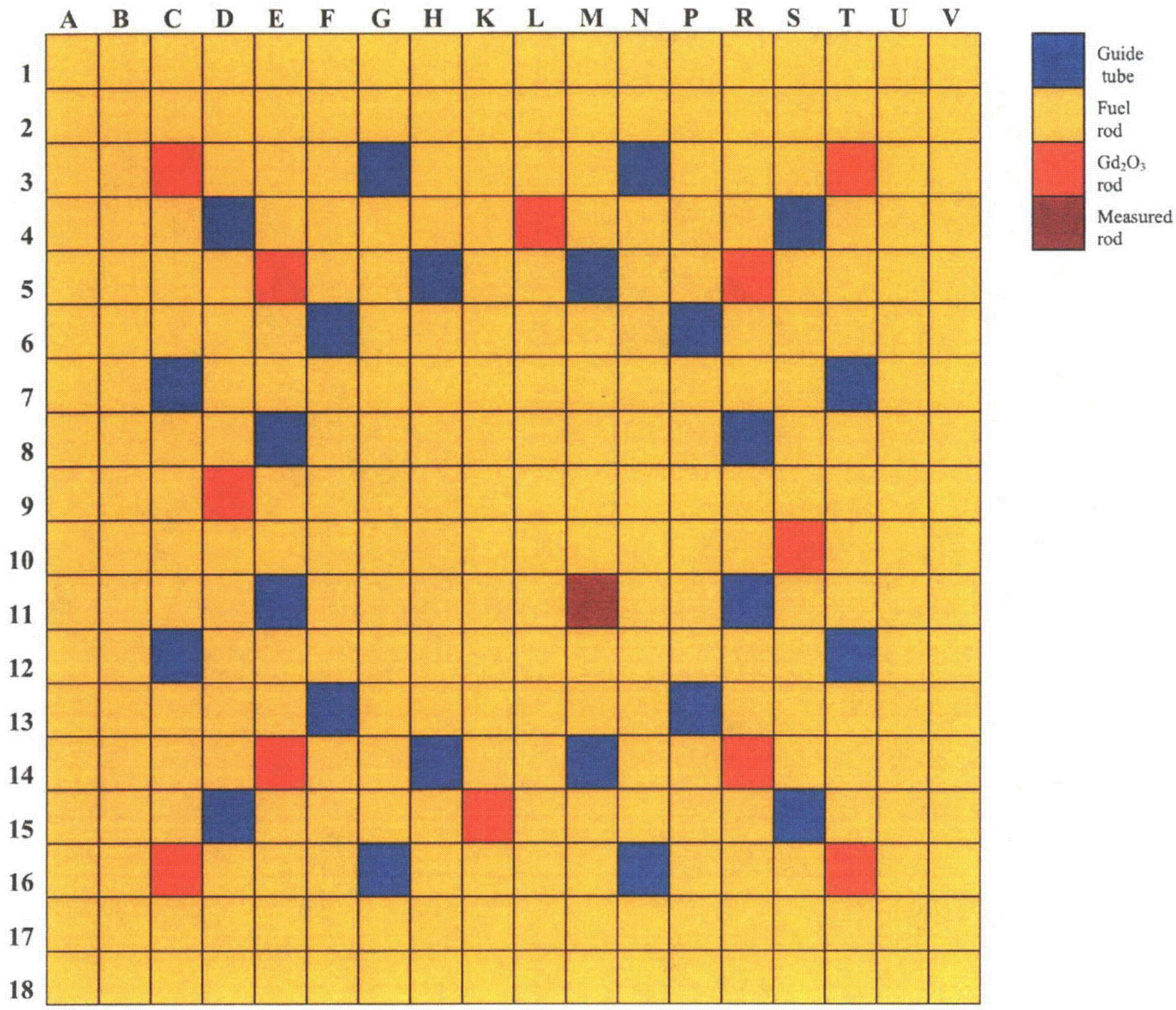

Figure 4.2 Assembly layout for GKN II (REBUS) sample

Table 4.4 Assembly design data for GKN II (REBUS) sample

Parameter	Data
Assembly and reactor data	
Reactor	GKN II
Lattice geometry	18 × 18
Rod pitch (cm)	1.27
Number of fuel rods	300
Number of guide tubes	24
Active fuel rod length (cm)	390
Assembly pitch (cm)	23.116
Fuel rod data	
Fuel material type	UO_2
Fuel pellet density (g/cm^3)	10.4
Enrichment (wt % ^{235}U)	3.8 (2.6)[b]
Sample location[a] (cm)	303
Fuel pellet diameter (cm)	0.805
Fuel temperature (K)	see Table 4.7
Clad material	Zircaloy-4
Clad inner diameter (cm)	0.822
Clad outer diameter (cm)	0.95
Average clad temperature[c] (K)	619
Number of rods with Gd_2O_3	12
Gd_2O_3 content (wt %)	7.0
U isotopic composition[d] (wt %)	
^{234}U	0.036 (0.0)[b]
^{235}U	3.798 (2.6)[b]
^{236}U	0.0 (0.0)[b]
^{238}U	96.166 (97.4)[b]
Moderator data	
Moderator temperature (K)	see Table 4.7
Moderator density (g/cm^3)	see Table 4.7
Soluble boron content (ppm)	see Table 4.7
Guide tube data	
Guide tube material	Zircaloy-4
Inner diameter (cm)	1.11
Outer diameter (cm)	1.232

[a] Relative to the bottom of the active fuel region.
[b] Values in parentheses correspond to gadolinia-bearing fuel.
[c] Maximum clad temperature as given in *1998 World Nuclear Industry Handbook*.[6]
[d] Initial (fresh fuel) values.

Table 4.5 Operating history data for GKN II (REBUS) sample

Cycle	Cumulative[a] time (days)	Burn time (days)	Soluble[b] boron in moderator (ppm)	Cumulative[a] burnup (GWd/MTU)
	6.0	6.0	965.6	
	30.0	30.0	876.6	
	60.0	60.0	783.2	
	90.0	90.0	681.8	
	120.0	120.0	583.2	
5	150.0	150.0	489.4	
	180.0	180.0	400.9	
	210.0	210.0	308.3	
	240.0	240.0	206.9	
	270.0	270.0	99.4	
	295.4	295.4	10.0	
	310.0	310.0	10.0	17.196
Down	332.0			
	338.0	316.0	1175.9	
	362.0	340.0	1088.9	
	392.0	370.0	998.8	
	422.0	400.0	898.8	
	452.0	430.0	800.2	
	482.0	460.0	706.1	
6	512.0	490.0	617.3	
	542.0	520.0	529.3	
	572.0	580.0	432.0	
	602.0	580.0	323.7	
	632.0	610.0	212.4	
	652.0	640.0	101.8	
	687.0	665.0	10.0	
	718.7	696.7	10.0	35.356
Down	735.7			
	741.7	702.7	1016.0	
	765.7	726.7	926.5	
	795.7	756.7	533.8	
	825.7	766.7	732.3	
	855.7	816.7	632.7	
	885.7	846.7	537.4	
7	915.7	876.7	447.5	
	945.7	906.7	355.7	
	975.7	936.7	255.0	
	1005.7	966.7	148.6	
	1044.6	1005.6	7.8	
	1083.6	1044.6	7.8	49.356
Down	1098.6			
	1104.0	1050.6	1228.9	
	1128.6	1074.6	1119.9	
	1158.6	1104.6	1001.3	
	1188.6	1134.6	874.3	
	1218.6	1164.6	749.2	
	1248.6	1194.6	627.3	
8	1278.6	1224.6	509.1	
	1308.6	1254.6	395.4	
	1338.6	1284.6	282.6	
	1368.6	1314.6	169.4	
	1411.0	1357.0	11.9	
	1445.4	1391.4	11.9	53.331

[a] From beginning of cycle 5 based on operating data.

[b] As provided in *REBUS International Program—Reactivity Tests for a Direct Evaluation of the Burnup Credit on Selected Irradiated LWR Fuel Bundles, Fuel Irradiation History*, SCK-CEN, Belgonucleaire (June 2005).

Table 4.6 Cycle average power data for GKN II (REBUS) sample

Cycle #	Duration (effective power days)	Down (days)	Cumulative burnup (GWd/MTU)	Power (MW/MTU)
5	310.0	22	17.442	56.264
6	386.7	17	35.862	47.634
7	347.9	15	50.063	40.820
8	346.8	–	54.095	11.626

Table 4.7 Cycle average moderator and fuel data for GKN II (REBUS) sample

Cycle #	Moderator density (g/cm^3)	Moderator temperature (K)	Fuel temperature (K)
5	0.646	605.0	1018.0
6	0.665	599.0	904.3
7	0.681	593.3	819.7
8	0.725	574.2	646.1

5. COMPUTATIONAL MODELS

5.1 Computational Tools

The computational analysis of the measurements was carried out using the two-dimensional (2-D) depletion sequence of the TRITON module in the SCALE computer code system.[9] The T-DEPL sequence in TRITON couples the 2-D arbitrary polygonal mesh, discrete ordinates transport code NEWT with the depletion and decay code ORIGEN-S in order to perform the burnup simulation. At each depletion step, the transport flux solution from NEWT is used to generate cross sections and assembly power distributions for the ORIGEN-S calculations; the isotopic composition data resulting from ORIGEN-S is employed in the subsequent transport calculation to obtain cross sections and power distributions for the next depletion step in an iterative manner throughout the irradiation history.

TRITON has the capability of simulating the depletion of multiple mixtures in a fuel assembly model. This is a very useful and powerful feature in a nuclide inventory analysis, as it allows a more appropriate representation of the local flux distribution and neutronic environment for a specific measured fuel rod in the assembly. The flux normalization in a TRITON calculation can be performed using as a basis the power in a specified mixture, the total power corresponding to multiple mixtures, or the assembly power. The first of the above-mentioned options permits specification of the burnup (power) in the measured sample, usually inferred from experimental measurements of burnup indicators (such as ^{148}Nd).

Individual TRITON models were developed for each of the sample measurements discussed in the previous sections. The models will be presented in this section. In all cases, the calculations were carried out by normalizing the power to reproduce the measured concentration of ^{148}Nd in the sample within the experimental uncertainty.

All TRITON calculations employed the SCALE 44-group cross-section library based on ENDF/B-V data and NITAWL as processor for the pin-cell cross section treatment. Default values were used for the convergence parameters in the NEWT transport calculation. Selected TRITON input files are provided in Appendix A.

5.2 Gösgen (ARIANE) Samples

The analysis of sample GU1 was carried out by using a quarter assembly model of assembly 1240, as shown in Figure 5.1. The geometry, material, and burnup data used in the TRITON model were as given in Tables 4.1 to 4.3. Replacement of some of the fuel rods during cycles 14 and 15 was not modeled because insufficient information on the configurations was available. However, the replacement rods were indicated to have burnup similar to that of the original rods and not modeling the fuel rods reconfiguration was deemed to be of minor importance.

The depletion history of the fuel rod from which samples GU3 and GU4 were selected, including the reconstitution of the fuel assembly, was explicitly simulated with TRITON. One TRITON model, as illustrated in Figure 5.2, was used to model the depletion of assembly 1601 during cycles 16 and 17; individual depleting mixtures were used for the measured rod and its nearest neighbor fuel rods, whereas all other fuel rods in the assembly were treated as a single depletion material with uniform composition. The nuclide compositions for the measured rod and the average composition for the regular fuel rods in assembly 1601 were saved at the end of the simulation for cycle 17 and used in the input file for simulating assembly 1701 during cycle 18. The average composition for the regular fuel rods from assembly 1601 was used as composition data for the three replacement rods that were, in addition to the measured rod, inserted in the rebuilt assembly 1701 at the beginning of cycle (BOC) 18.

The TRITON model for assembly 1701 is illustrated in Figure 5.3. As mentioned in Section 4.1, it is known that the average burnup of assembly 1701 at BOC-18 was about 20 GWd/MTU. To determine the composition of the spent fuel for the 201 fuel rods in this assembly from the total of 205 rods, once the composition for the four replacement rods was calculated, an additional TRITON model was used to simulate the depletion of assembly 1701 prior to the reconstitution. This model is similar to that illustrated in Figure 5.2 but considered a single depletion mixture for all the fuel rods in the assembly; this mixture was depleted to a burnup of 20 GWd/MTU for sample GU3 and 9.7 GWd/MTU for sample GU4 and the composition of the depletion mixture was saved to be used in the depletion model of assembly 1701 during cycle 18.

The sample burnups used in the code simulations were normalized to the measured ^{148}Nd concentration. The sample burnup values based on measured ^{148}Nd for samples GU1, GU3, and GU4 were 60.7, 52.5, and 31.1 GWd/MTU, respectively. These burnups based on experimental data are in good agreement with the burnup values 59.7, 52.5, and 29.1 GWd/MTU from operator data. The burnup history data presented in Tables 4.2 and 4.3 were adjusted by a constant factor to correspond to the measurement-based burnup.

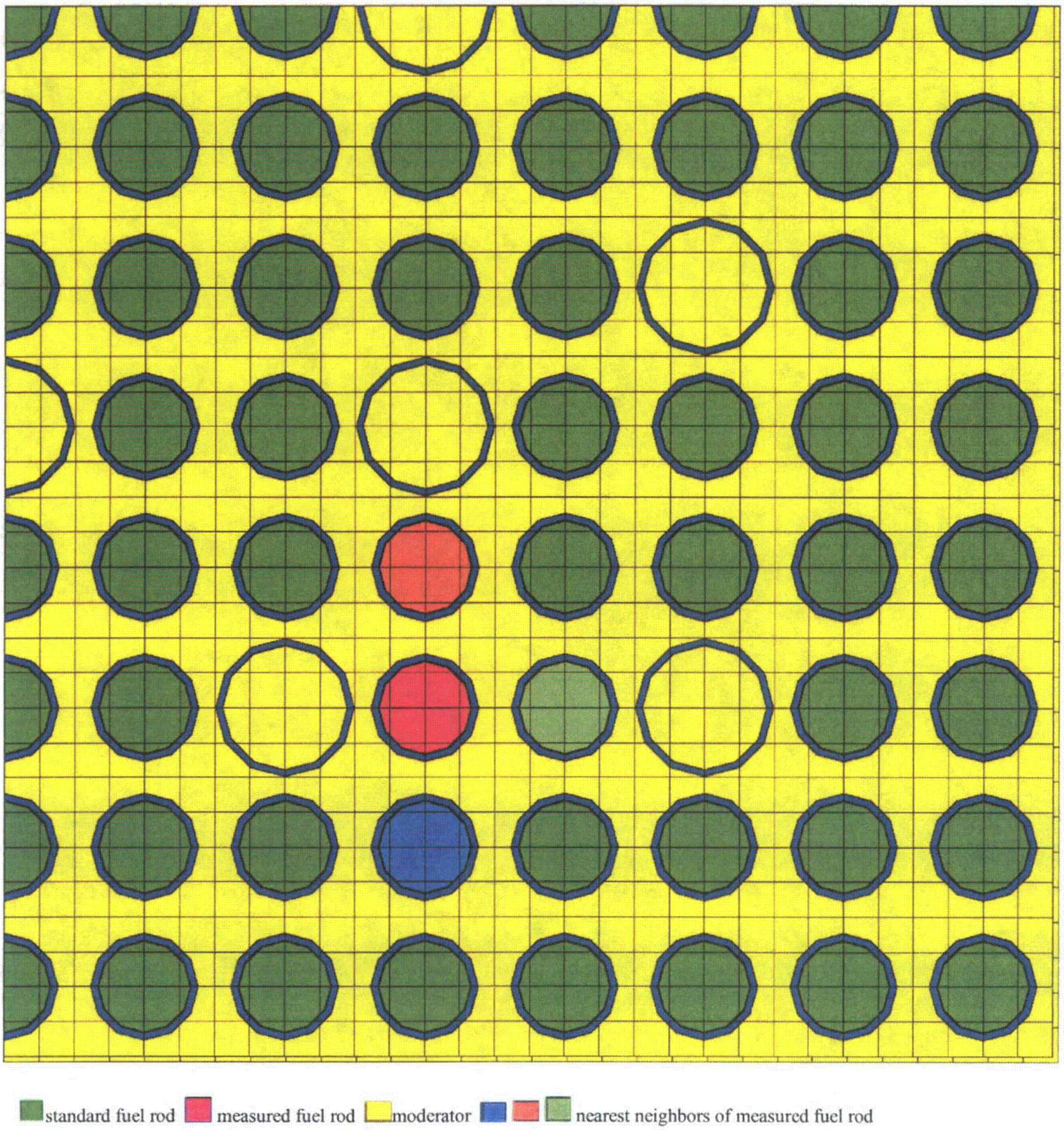

Figure 5.1 TRITON assembly model for Gösgen (ARIANE)—sample GU1

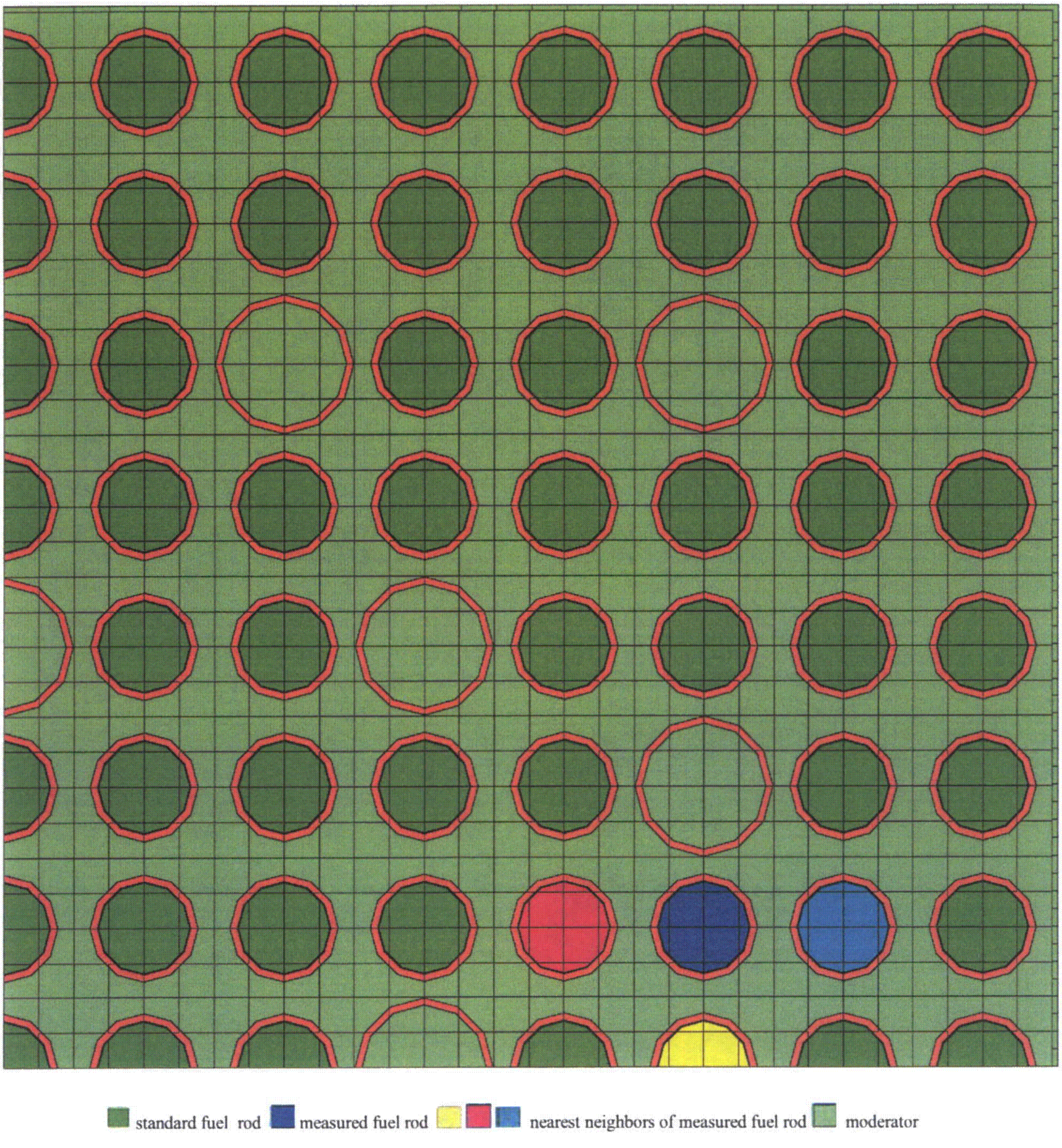

Figure 5.2 TRITON assembly model for Gösgen (ARIANE)—sample GU3/4, cycles 16–17

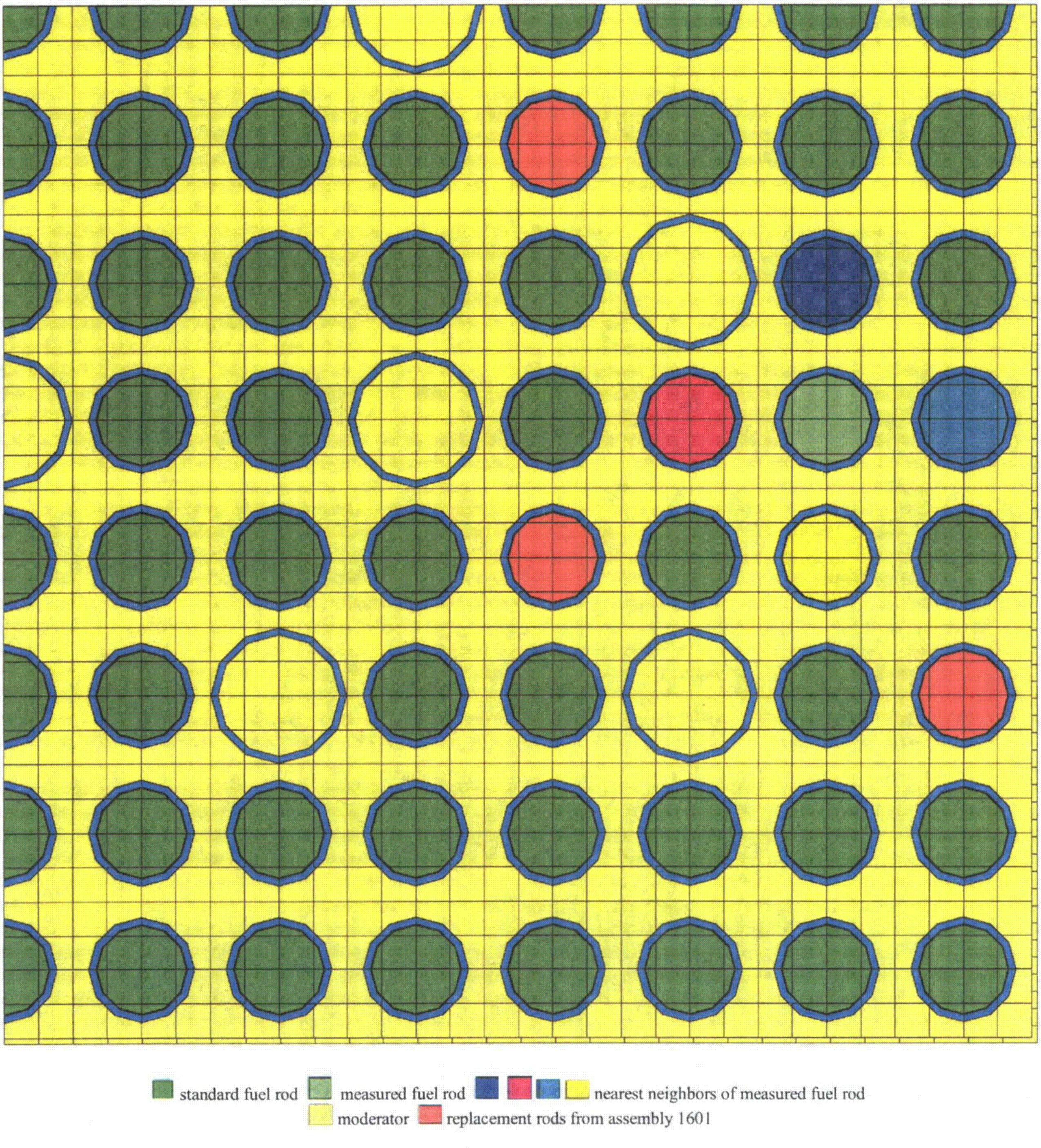

Figure 5.3 TRITON assembly model for Gösgen (ARIANE)—sample GU3/4, cycle 18

5.3 GKN II (REBUS) Sample

The geometry of the 18 × 18 GKN II assembly 419 was modeled in full detail, as illustrated in Figure 5.4. White boundary conditions were used for the assembly bounding surfaces. As observed, there is a slight asymmetry in the assembly with respect to the placement of the gadolinia-bearing rods. The average power used in the simulations for each of the four irradiation cycles was taken from Table 4.7. The time-dependent variation of the boron concentration in the moderator, as well as of the moderator density and fuel and moderator temperatures, as given in Tables 4.6 and 4.7, were simulated through the TIMETABLE input block in the TRITON input. The use of the provided sample burnup, 54.1 GWd/MTU, yielded a calculated ^{148}Nd consistent with the measured value.

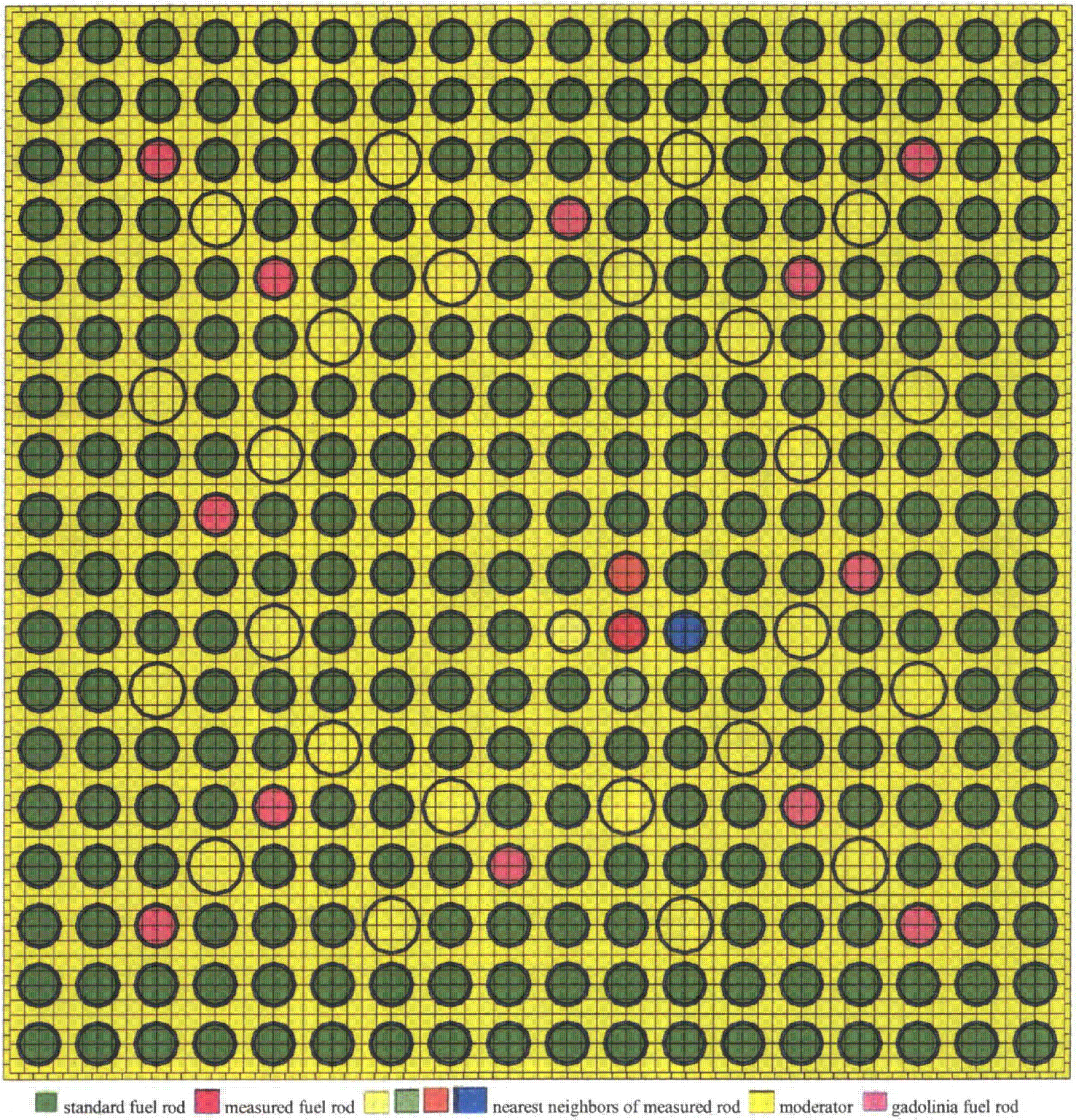

Figure 5.4 TRITON assembly model for GKN II (REBUS) sample

6. RESULTS

6.1 Gösgen (ARIANE) Samples

The results of the TRITON simulations, given as percentage difference between calculated and measured nuclide concentrations, are illustrated in Figures 6.1 to 6.5 and listed in Table 6.1. The sample burnups shown in the figures are the values based on the measured ^{148}Nd concentration. The comparison experiment-calculation for sample GU3, which was measured at two laboratories, was done by using the recommended measured isotopic concentrations presented in Table 3.3.

The uranium and plutonium nuclides, except for ^{234}U, are predicted within 6% of the measurement for all three samples (see Figure 6.1). The most important fissionable actinides, ^{235}U and ^{239}Pu, are on average overpredicted by about 1 and 4%, respectively. There is a large variation in the prediction of the minor actinides, depending on the nuclide considered, as seen in Figure 6.2. The ^{241}Am and ^{244}Cm nuclides, which are important contributors to decay heat in spent fuel, are estimated on average within 6% of the measured data. In general, the results of the comparison in the case of samples GU3 and GU4 are consistent with the results of a previous analysis using the HELIOS code.[10]

As illustrated in Figure 6.3, the cesium isotopes ^{133}Cs, ^{135}Cs, and ^{137}Cs are overestimated in all three samples on average by less than 5% of measured data, whereas ^{134}Cs, important to decay heat and gamma sources at short cooling times, is underpredicted by 9% on average. The neodymium nuclides, except for ^{142}Nd, are on average predicted within about 2% of the measurement. The ^{149}Sm isotope, an important fission product for burnup credit criticality calculations, is overestimated on average by 11%. The ^{147}Sm and ^{148}Sm nuclides are on average predicted within about 1 and 9% of the measurement, whereas ^{151}Sm and ^{152}Sm are consistently overestimated in the 30% range; ^{150}Sm and ^{154}Sm are overpredicted, on average, by 8 and 5% of the measurement. The nuclides ^{153}Eu, important for burnup credit criticality calculations, and ^{154}Eu, an important gamma emitter, are overpredicted on average by 7 and 8%. The ^{155}Eu nuclide and its decay daughter ^{155}Gd are both underestimated in the 30% range.

When assessing the level of agreement between calculation and experiment, one needs to consider the experimental errors as well as other problems or limitations related to measurement or data required for simulations. Also, the user of the ARIANE experimental data needs to be aware of the fact that calculated data for samples selected from fuel rods from rebuilt assemblies may have additional uncertainties related to modeling and simulation as compared to typical commercial fuel. However, these data are valuable for code validation purposes, as they enlarge the burnup and enrichment ranges of the limited set of available measurement data and may be used for testing different code capabilities, such as changes in assembly geometry and composition during a depletion simulation.

To establish any conclusion as to whether or not the calculated-to-experimental (C/E) ratios exhibit a systematic behavior versus burnup, the samples considered in this report would need to be evaluated in the framework of a larger set of data covering an extensive burnup range.

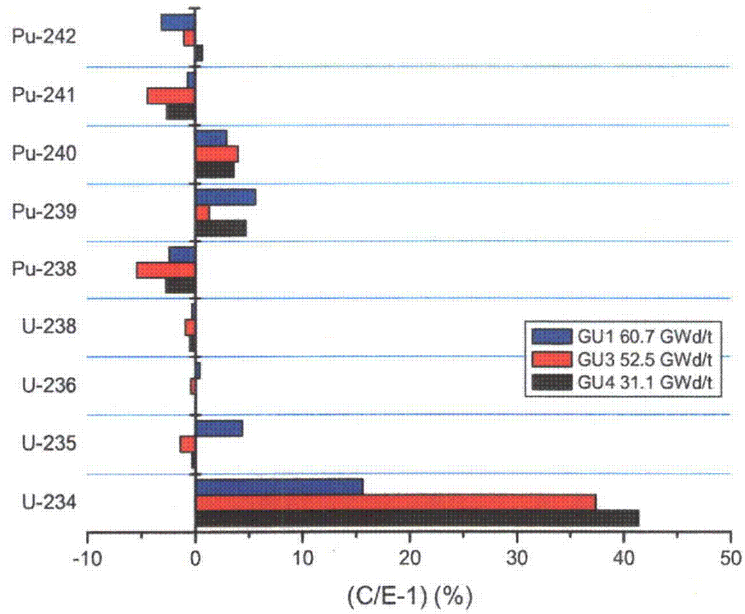

Figure 6.1 Gösgen (ARIANE) samples—major actinides

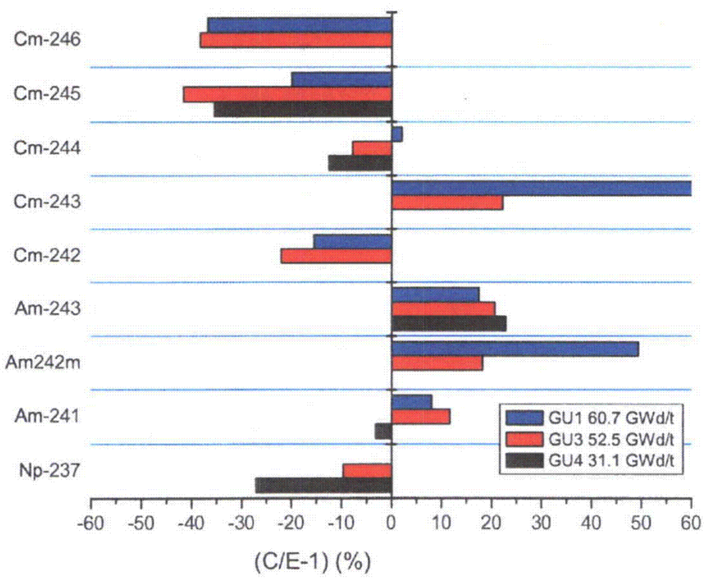

Figure 6.2 Gösgen (ARIANE) samples—minor actinides

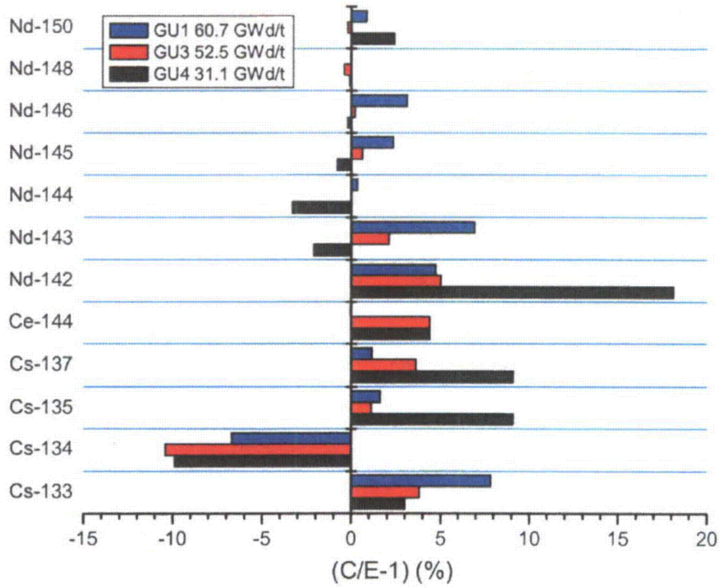

Figure 6.3 Gösgen (ARIANE) samples—fission products (Cs, Ce, Nd)

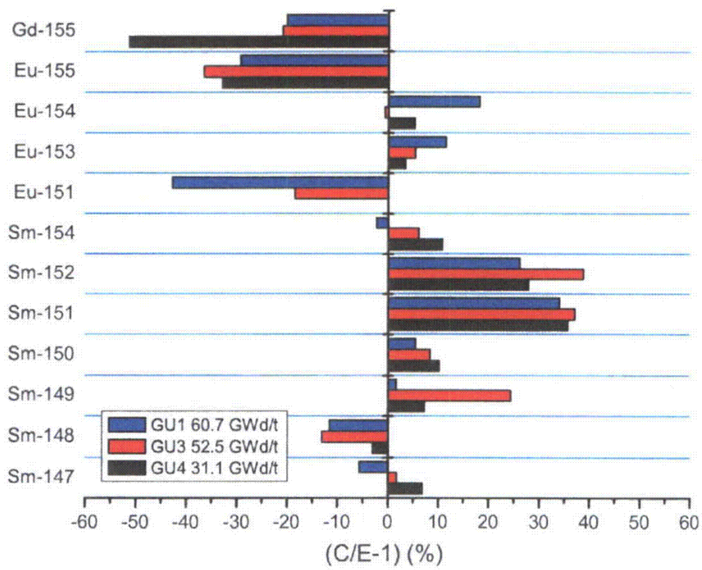

Figure 6.4 Gösgen (ARIANE) samples—fission products (Sm, Eu, Gd)

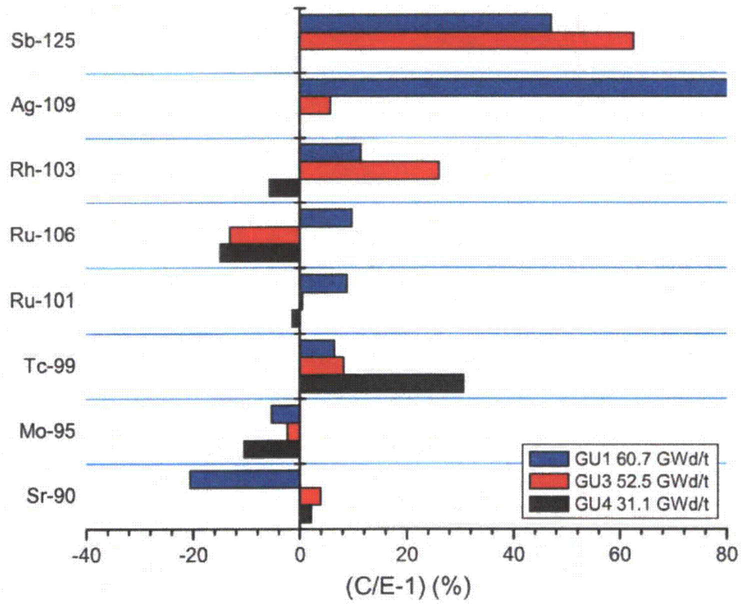

Figure 6.5 Gösgen (ARIANE) samples—fission products (metallics)

Table 6.1 C/E-1 (%) for Gösgen (ARIANE) samples

Sample ID	GU4	GU3	GU1			
Burnup[a] (GWd/MTU)	31.1	52.5	60.7			
				Avg	Min	Max
U-234	41.4	37.4	15.6	31.5	15.6	41.4
U-235	-0.3	-1.4	4.4	0.9	-1.4	4.4
U-236	0.1	-0.4	0.4	0.1	-0.4	0.4
U-238	-0.5	-0.9	-0.3	-0.6	-0.9	-0.3
Pu-238	-2.7	-5.4	-2.4	-3.5	-5.4	-2.4
Pu-239	4.7	1.3	5.6	3.9	1.3	5.6
Pu-240	3.6	4.0	2.9	3.5	2.9	4.0
Pu-241	-2.6	-4.4	-0.6	-2.6	-4.4	-0.6
Pu-242	0.7	-1.0	-3.1	-1.1	-3.1	0.7
Np-237	-27.1	-9.6		-18.3	-27.1	-9.6
Am-241	-3.2	11.7	8.0	5.5	-3.2	11.7
Am242m		18.2	49.4	33.8	18.2	49.4
Am-243	22.8	20.6	17.5	20.3	17.5	22.8
Cm-242		-22.0	-15.5	-18.8	-22.0	-15.5
Cm-243		22.3	202.3	112.3	22.3	202.3
Cm-244	-12.5	-7.8	2.1	-6.1	-12.5	2.1
Cm-245	-35.4	-41.6	-19.9	-32.3	-41.6	-19.9
Cm-246		-38.2	-36.7	-37.5	-38.2	-36.7
Sr-90	2.1	3.9	-20.6	-4.9	-20.6	3.9
Mo-95	-10.5	-2.4	-5.4	-6.1	-10.5	-2.4
Tc-99	30.6	8.1	6.4	15.0	6.4	30.6
Ru-101	-1.4	0.5	8.8	2.6	-1.4	8.8
Ru-106	-14.9	-13.1	9.7	-6.1	-14.9	9.7
Rh-103	-5.7	26.0	11.3	10.5	-5.7	26.0
Ag-109		5.7	118.6	62.1	5.7	118.6
Sb-125		62.5	47.1	54.8	47.1	62.5
Cs-133	3.0	3.8	7.8	4.9	3.0	7.8
Cs-134	-9.9	-10.4	-6.7	-9.0	-10.4	-6.7
Cs-135	9.1	1.1	1.6	3.9	1.1	9.1
Cs-137	9.1	3.6	1.1	4.6	1.1	9.1
Ce-144	4.4	4.4	-0.1	2.9	-0.1	4.4
Nd-142	18.1	5.0	4.7	9.3	4.7	18.1
Nd-143	-2.1	2.1	6.9	2.3	-2.1	6.9
Nd-144	-3.3	0.0	0.3	-1.0	-3.3	0.3
Nd-145	-0.8	0.6	2.3	0.7	-0.8	2.3
Nd-146	-0.2	0.2	3.1	1.0	-0.2	3.1
Nd-148	-0.1	-0.4	0.0	-0.2	-0.4	0.0
Nd-150	2.4	-0.2	0.9	1.0	-0.2	2.4

Table 6.1 C/E-1 (%) for Gösgen (ARIANE) samples (continued)

Sample ID	GU4	GU3	GU1			
Burnup[a] (GWd/MTU)	**31.1**	**52.5**	**60.7**			
				Avg	Min	Max
Pm-147	-61.4	124.8	39.0	34.1	-61.4	124.8
Sm-147	6.8	1.7	-5.7	0.9	-5.7	6.8
Sm-148	-3.0	-13.1	-11.6	-9.2	-13.1	-3.0
Sm-149	7.2	24.4	1.6	11.1	1.6	24.4
Sm-150	10.2	8.3	5.4	8.0	5.4	10.2
Sm-151	35.7	37.2	34.0	35.6	34.0	37.2
Sm-152	27.9	38.8	26.2	31.0	26.2	38.8
Sm-154	10.8	6.1	-2.3	4.9	-2.3	10.8
Eu-151		-18.5	-42.7	-30.6	-42.7	-18.5
Eu-153	3.4	5.4	11.5	6.8	3.4	11.5
Eu-154	5.2	-0.6	18.1	7.6	-0.6	18.1
Eu-155	-32.9	-36.5	-29.3	-32.9	-36.5	-29.3
Gd-155	-51.3	-20.9	-22.1	-31.5	-51.3	-20.9

[a] Based on measured ^{148}Nd.

6.2 GKN II (REBUS) Sample

The total sample burnup of 54.1 GWd/MTU was used in the simulations for the GKN II sample. The results of the calculation are illustrated in Figures 6.6 to 6.9 and listed in Table 6.2. The calculated concentration of ^{148}Nd is, within the experimental error, consistent with the measured values. The uranium nuclides, except for ^{234}U, are predicted within about 4% of the measured value. The large overprediction of ^{234}U, about 20%, may be indicative of uncertainty in the ^{234}U concentration in the fresh fuel. The plutonium isotopes ^{240}Pu, ^{241}Pu, and ^{242}Pu are well predicted, within about 3% of the measurement, whereas ^{238}Pu and ^{239}Pu are predicted within about 8%. The americium isotopes are overpredicted by about 30% on average. The ^{244}Cm nuclide, an important contributor to decay heat and the neutron source terms, is well predicted, within about 6% of the measurement.

The comparison for cesium, cerium, and neodymium isotopes is presented in Figure 6.7. Concentrations for this group of nuclides tend to be well predicted: all calculated concentrations for neodymium nuclides except for ^{142}Nd and ^{143}Nd are within 2% of the measurement; both ^{148}Nd and ^{137}Cs, which can be used as burnup indicators are well predicted, within 0.3 and 1.3% of the measured values, respectively. The results for the fission product group consisting of samarium, europium, and gadolinium isotopes are illustrated in Figure 6.8. With the exception of ^{151}Sm and ^{152}Sm, the measured samarium nuclides are predicted within 14% of the measurement. The results for the metallic elements consisting of isotopes of molybdenum, ruthenium, rhodium, technetium, silver, and paladium are shown in Figure 6.9. With the exception of ^{99}Tc, the metallic isotopes are overpredicted, with larger overpredictions seen for palladium isotopes. It is possible that this is caused by the experimental problems in recovering and measuring all of the material in the undissolved residues, as these species are difficult to dissolve and must be measured in both the main solution and the residue.

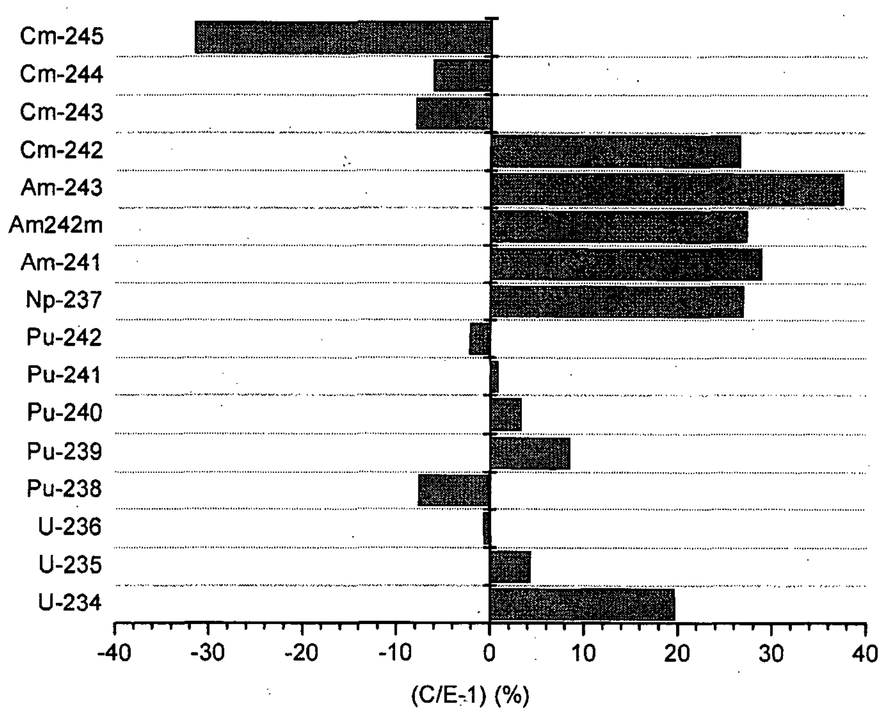

Figure 6.6 GKN II (REBUS) sample—actinides

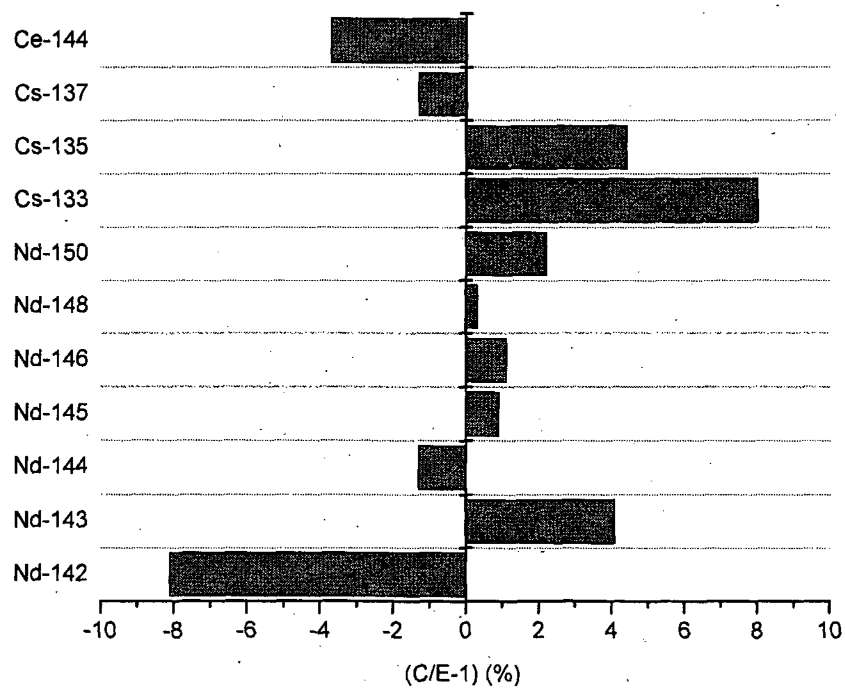

Figure 6.7 GKN II (REBUS) sample—fission products (Cs, Ce, Nd)

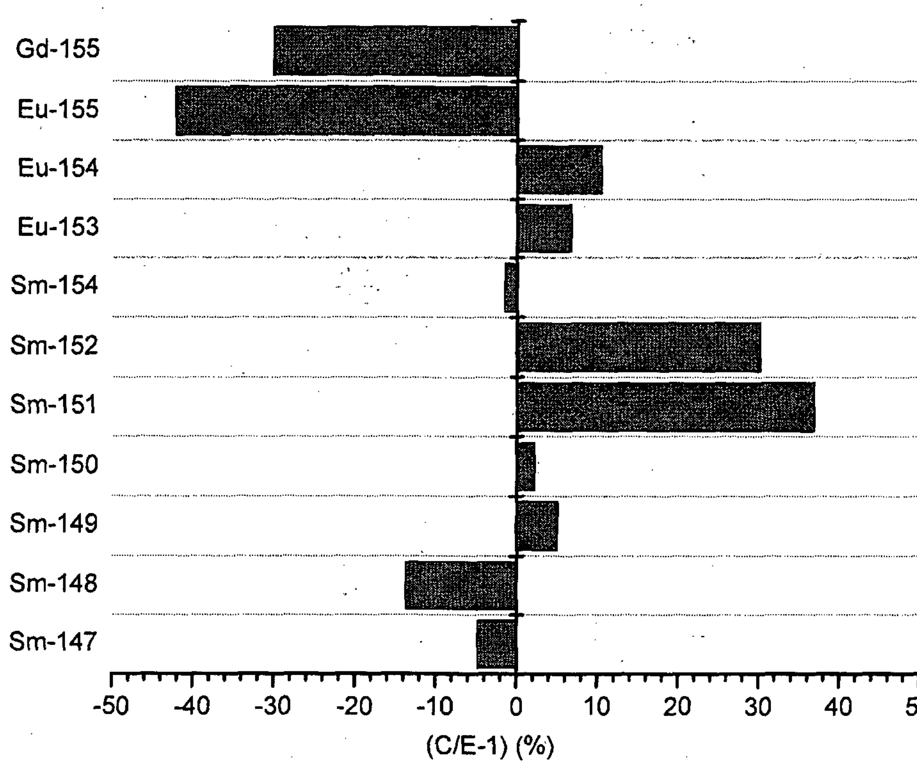

Figure 6.8 GKN II (REBUS) sample—fission products (Sm, Eu, Gd)

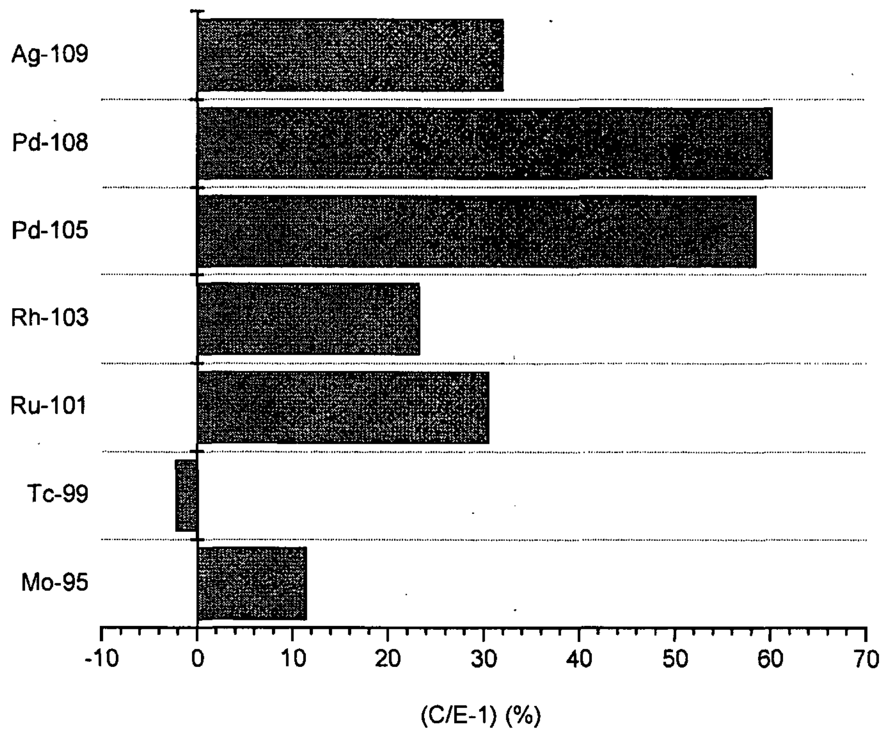

Figure 6.9 GKN II (REBUS) sample—fission products (Mo, Tc, Ru, Rh, Pd, Ag)

Table 6.2 C/E-1 (%) for GKN II (REBUS) sample

Nuclide ID	C/E-1 (%)
U-234	19.7
U-235	4.3
U-236	-0.6
Pu-238	-7.6
Pu-239	8.5
Pu-240	3.3
Pu-241	0.8
Pu-242	-2.1
Np-237	27.0
Am-241	28.9
Am-242m	27.3
Am-243	37.5
Cm-242	26.6
Cm-243	-7.9
Cm-244	-6.1
Cm-245	-31.6
Nd-142	-8.1
Nd-143	4.1
Nd-144	-1.3
Nd-145	0.9
Nd-146	1.1
Nd-148	0.3
Nd-150	2.2
Cs-133	8.0
Cs-135	4.4
Cs-137	-1.3
Ce-144	-3.7
Sm-147	-4.8
Sm-148	-13.7
Sm-149	5.1
Sm-150	2.2
Sm-151	36.9
Sm-152	30.2
Sm-154	-1.5
Eu-153	6.8
Eu-154	10.5
Eu-155	-42.2
Gd-155	-30.2
Mo-95	11.4
Tc-99	-2.2
Ru-101	30.5
Rh-103	23.3
Pd-105	58.5
Pd-108	60.1
Ag-109	32.0

7. SUMMARY

This report is part of a series of reports documenting high-quality radiochemical assay data against which computer code predictions of the isotopic composition in high-burnup spent nuclear fuel can be validated to quantify the uncertainty and bias associated with the code predictions. The experimental data documented and analyzed in this report were acquired by ORNL through participation in two international programs designed to provide benchmark-quality radiochemical assay data: (1) ARIANE and (2) REBUS, both coordinated by Belgonucleaire.

The measurements analyzed include four spent fuel samples from fuel irradiated in two PWRs: GKN II and Gösgen, operated in Germany and Switzerland, respectively. The samples cover a large burnup range, from 30 to 60 GWd/MTU, and have initial fuel enrichments between 3.5 and 4.1 wt % ^{235}U. An analysis of the experimental data was carried out using the two-dimensional depletion module TRITON in the SCALE code system. Individual TRITON models were developed for each of the samples considered. In the case of the Gösgen GU3 and GU4 samples, the reconstitution of the assembly was simulated explicitly. Information on the radiochemical analysis methods and uncertainties, assembly design description and irradiation history, and computational models and results obtained using the SCALE code system are included. The data are presented in sufficient detail to allow an independent analysis to be performed.

8. REFERENCES

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APPENDIX A

TRITON INPUT FILES

A.1 TRITON INPUT FILE FOR THE GU1 (ARIANE) SAMPLE

```
=t-depl parm=(nitawl,addnux=3)
   Gosgen 15x15 PWR.  Sample GU1. Nd-148 burnup 60.75 GWd/MTU.
44groupndf5
=============================================================================
read alias
 $fuel 10 11 12 13 14 end
 $clad 20 21 22 23 24 end
 $mod  30 31 32 33 34 end
 $gap  40 41 42 43 44 end
end alias
=============================================================================
read comp
' fuel
uo2 $fuel den=10.4 1 1151.3 92234  0.036
                            92235  3.500
                            92238 96.464 end
' clad
zirc4 $clad 1 619 end
' moderator
h2o $mod den=0.7299 1 571.6 end
arbmb 0.7299 1 1 0 0 5000 100 $mod 1511e-06 571.6 end
' gap
n $gap den=0.00125 1 619 end
end comp
=============================================================================
read celldata
 latticecell squarepitch pitch=1.43   $mod
                         fueld=0.913  $fuel
                         cladd=1.075  $clad
                         gapd=0.93    $gap  end
end celldata
=============================================================================
read depletion
 10 -11 12 13 14
end depletion
=============================================================================
read burndata
power= 60.31  burn=  6      down=  0  nlib=1 end
power= 60.31  burn=  144    down=  0  nlib=3 end
power= 60.31  burn=  144.9  down=  0  nlib=3 end
power= 54.52  burn=  22.1   down=  45 nlib=1 end
power= 47.92  burn=  6      down=  0  nlib=1 end
power= 47.92  burn=  144    down=  0  nlib=3 end
power= 47.92  burn=  142.3  down=  0  nlib=3 end
power= 41.83  burn=  29     down=  27 nlib=1 end
power= 45.59  burn=  6      down=  0  nlib=1 end
power= 45.59  burn=  144    down=  0  nlib=3 end
power= 45.59  burn=  140.1  down=  0  nlib=3 end
power= 32.83  burn=  41.2   down=  50 nlib=1 end
power= 36.97  burn=  6      down=  0  nlib=1 end
power= 36.97  burn=  144    down=  0  nlib=3 end
power= 36.97  burn=  151.9  down=  0  nlib=3 end
power= 32.17  burn=  24.8   down=  11 nlib=1 end
end burndata
=============================================================================
```

```
read opus
units=grams
symnuc=u-234  u-235   u-236  u-238  np-237 pu-238 pu-239 pu-240 pu-241 pu-242
       am-241 am-242m am-243 cm-242 cm-243 cm-244 cm-245 cm-246 ce-144 nd-142
       nd-143 nd-144  nd-145 nd-146 nd-148 nd-150 pm-147 sm-147 sm-148 sm-149
       sm-150 sm-151  sm-152 sm-154 eu-151 eu-153 eu-154 eu-155 gd-155 sr-90
       cs-133 cs-134  cs-135 cs-137 mo-95  tc-99  ru-101 ru-106 rh-103 ag-109
       sb-125 end
matl=11 end
end opus
=============================================================================
read timetable
' soluble boron in moderator
densmult $mod 2 5010 5011
 0         1.000
 6         0.780
 150       0.374
 295       0.005
 330       0.005
 361.99    0.005
 362       0.977
 368       0.758
 512       0.359
 654       0.005
 694       0.005
 709.99    0.005
 710       1.004
 716       0.780
 860       0.363
 1000      0.003
 1065      0.003
 1090.99   0.003
 1091      1.055
 1097      0.823
 1241      0.400
 1393      0.003
 1429      0.003 end
' fuel temperature
temperature $fuel
0   1151.3
6   1171.5
150 1136.0
295 1078.3
330 1046.7
362 919.3
368 967.7
512 957.9
654 943.1
694 842.0
710 888.9
716 894.4
860 854.8
1000    841.4
1065    709.8
1091    806.6
1097    829.8
1241    810.6
```

```
1393   804.0
1429   738.9 end
end timetable
============================================================================
read model
 Gosgen 15x15 PWR. Sample GU1.
read parm
 run=yes drawit=yes echo=yes fillmix=30
end parm
read materials
 10  1 ! regular pin          ! end
 20  1 ! clad                 ! end
 30  2 ! water moderator      ! end
 40  1 ! gap                  ! end
 11  1 ! test pin             ! end
 12  1 ! N test pin           ! end
 13  1 ! E test pin           ! end
 14  1 ! S test pin           ! end
end materials
read geom
unit 1
com='fuel pin cell'
 cylinder  1 0.4565
 cylinder  2 0.465
 cylinder  3 0.5375
 cuboid    4 4p0.715
 media 10 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 11
com='bottom half fuel pin cell'
 cylinder  1 0.4565   chord -y=0
 cylinder  2 0.465    chord -y=0
 cylinder  3 0.5375   chord -y=0
 cuboid    4 2p0.715  0.0 -0.715
 media 10 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 2
unit 12
com='right half fuel pin cell'
 cylinder  1 0.4565   chord +x=0
 cylinder  2 0.465    chord +x=0
 cylinder  3 0.5375   chord +x=0
 cuboid    4 0.715 0.0 2p0.715
 media 10 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 2 4
unit 13
com='bottom right quarter fuel pin cell'
 cylinder  1 0.4565   chord +x=0 chord -y=0
 cylinder  2 0.465    chord +x=0 chord -y=0
```

```
 cylinder  3 0.5375   chord +x=0 chord -y=0
 cuboid    4 0.715 0.0 0.0 -0.715
 media 10 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 2 2
unit 2
com='test pin cell'
 cylinder  1 0.4565
 cylinder  2 0.465
 cylinder  3 0.5375
 cuboid    4 4p0.715
 media 11 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 3
com='N neigbor of test pin'
 cylinder  1 0.4565
 cylinder  2 0.465
 cylinder  3 0.5375
 cuboid    4 4p0.715
 media 12 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 4
com='E neigbor of test pin'
 cylinder  1 0.4565
 cylinder  2 0.465
 cylinder  3 0.5375
 cuboid    4 4p0.715
 media 13 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 5
com='S neigbor of test pin'
 cylinder  1 0.4565
 cylinder  2 0.465
 cylinder  3 0.5375
 cuboid    4 4p0.715
 media 14 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 6
com='guide tube'
 cylinder  1 0.62
 cylinder  2 0.69
 cuboid    3 4p0.715
 media 30 1 1
```

```
 media 20 1 2 -1
 media 30 1 3 -2
 boundary 3 4 4
unit 61
com='bottom half guide tube'
 cylinder  1 0.62     chord -y=0
 cylinder  2 0.69     chord -y=0
 cuboid    3 2p0.715  0.0 -0.715
 media 30 1 1
 media 20 1 2 -1
 media 30 1 3 -2
 boundary 3 4 2
unit 62
com='right half guide tube'
 cylinder  1 0.62     chord +x=0
 cylinder  2 0.69     chord +x=0
 cuboid    3 0.715 0.0 2p0.715
 media 30 1 1
 media 20 1 2 -1
 media 30 1 3 -2
 boundary 3 2 4
global unit 10
 cuboid 10 10.78 0.0 10.78 0.0
 array 1 10 place 1 1 0.0 0.77
 media 30 1 10
 boundary 10 30 30
end geom
read array
 ara=1 nux=8 nuy=8 typ=cuboidal
 fill
 12  1  1  1  1  1  1  1
 12  1  1  5  1  1  1  1
 12  1  6  2  4  6  1  1
 12  1  1  3  1  1  1  1
 62  1  1  6  1  1  1  1
 12  1  1  1  1  6  1  1
 12  1  1  1  1  1  1  1
 13 11 11 61 11 11 11 11 end fill
end array
read bounds
 all=refl
end bounds
end model
=================================================================================
end
=shell
 cp ft71f001 $RTNDIR/GU1.den
end
```

A.2 TRITON INPUT FILE FOR THE GKN II (REBUS) SAMPLE

```
=t-depl parm=(nitawl,addnux=3)
GKN II 18x18 PWR  Assembly FA 419 Pin M11 REBUS program
44groupndf5
=============================================================================
read alias
$fuel1 10 11 12 13 14 15 end
$clad1 20 21 22 23 24 25 end
$mod1  30 31 32 33 34 35 end
$gap1  40 41 42 43 44 45 end
$fuel2 50 end
$clad2 60 end
$mod2  70 end
$gap2  80 end
end alias
=============================================================================
read comp
uo2 $fuel1 den=10.4 1 1018.04 92234  0.036
                              92235  3.798
                              92238 96.166  end
zirc4 $clad1 1 619 end
h2o   $mod1 den=0.646 1  605.01 end
n     $gap1 den=0.00125 1 619 end
arbm-bormod  0.646  1 1 0 0 5000 100 $mod1 974.4e-6 605.01 end
uo2   $fuel2 den=10.13 0.93 1018.04 92235  2.6
                                    92238 97.4 end
arbmgd 10.13 2 0 1 0 64000 2 8016 3 $fuel2 0.07 1018.04 end
zirc4 $clad2 1 619 end
h2o   $mod2 den=0.646 1  605.01 end
arbm-bormod  0.646  1 1 0 0 5000 100 $mod2 974.4e-6 605.01 end
n     $gap2 den=0.00125 1 619 end
end comp
=============================================================================
read celldata
 latticecell squarepitch pitch=1.27   $mod1
                         fuelr=0.4025 $fuel1
                         cladr=0.475  $clad1
                         gapr=0.411   $gap1  end
 latticecell squarepitch pitch=1.27   $mod2
                         fuelr=0.4025 $fuel2
                         cladr=0.475  $clad2
                         gapr=0.411   $gap2 end
end celldata
=============================================================================
read timetable
 density $mod1 2 5010 5011
   0       1.000
 310       0.010
 331.99    0.010
 332       1.222
 718.7     0.010
 735.69    0.010
 735.7     0.974
1083.6     0.008
1098.59    0.008
1098.6     1.246
```

```
1445.4      0.012 end
 density $mod2 2 5010 5011
   0        1.000
 310        0.010
 331.99     0.010
 332        1.222
 718.7      0.010
 735.69     0.010
 735.7      0.974
1083.6      0.008
1098.59     0.008
1098.6      1.246
1445.4      0.012 end
dens $mod1 2 1001 8016
   0        1.000
 331.99     1.000
 332        1.029
 735.69     1.029
 735.7      1.053
1098.59     1.053
1098.6      1.122
1445.4      1.122 end
dens $mod2 2 1001 8016
   0        1.000
 331.99     1.000
 332        1.029
 735.69     1.029
 735.7      1.053
1098.59     1.053
1098.6      1.122
1445.4      1.122 end
temperature $fuel1
   0        1018.04
 331.99     1018.04
 332        904.25
 735.69     904.25
 735.7      819.69
1098.59     819.69
1098.6      646.13
1445.4      646.13 end
temperature $fuel2
   0        1018.04
 331.99     1018.04
 332        904.25
 735.69     904.25
 735.7      819.69
1098.59     819.69
1098.6      646.13
1445.4      646.13 end
temperature $mod1
0           605.01
331.99      605.01
332         598.98
735.69      598.98
735.7       593.34
1098.59     593.34
1098.6      574.23
```

```
1445.4     574.23 end
temperature $mod2
0          605.01
331.99     605.01
332        598.98
735.69     598.98
735.7      593.34
1098.59    593.34
1098.6     574.23
1445.4     574.23 end
end timetable
=============================================================================
 read depletion
  10 -11 12 13 14 15 50
 end depletion
=============================================================================
read burndata
  power=56.264  burn=310   down=22 nlib=6 end
  power=47.634  burn=386.7 down=17 nlib=6 end
  power=40.820  burn=347.9 down=15 nlib=5 end
  power=11.626  burn=346.8 down=0  nlib=2 end
 end burndata
=============================================================================
read model
GKN II 18x18 PWR  Assembly FA 419
read parm
  run=yes drawit=yes fillmix=30 echo=yes cmfd=yes xycmfd=4
end parm
read materials
 10  1 ! fuel               ! end
 20  1 ! clad               ! end
 30  2 ! moderator          ! end
 40  0 ! gap                ! end
 11  1 ! test rod           ! end
 12  1 ! N test rod         ! end
 13  1 ! S test rod         ! end
 14  1 ! E test rod         ! end
 15  1 ! W test rod         ! end
 50  1 ! fuel-gd            ! end
end materials
read geom
unit 1
com='fuel pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 10 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 3
com='guide tube'
 cylinder  1 0.555
 cylinder  2 0.616
 cuboid    3 4p0.635
```

```
 media  30 1 1
 media  20 1 2 -1
 media  30 1 3 -2
 boundary 3 4 4
unit 4
com='test pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 11 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 41
com='N test pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 12 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 42
com='S test pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 13 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 43
com='E test pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 14 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
unit 44
com='W test pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 15 1 1
 media 40 1 2 -1
 media 20 1 3 -2
```

```
 media 30 1 4 -3
 boundary 4 4 4
unit 5
com='UO2-Gd pin cell'
 cylinder  1 0.4025
 cylinder  2 0.411
 cylinder  3 0.475
 cuboid    4 4p0.635
 media 50 1 1
 media 40 1 2 -1
 media 20 1 3 -2
 media 30 1 4 -3
 boundary 4 4 4
global unit 10
 cuboid 10 23.116 0.0 23.116 0.0
 array 1 10 place 1 1 0.763 0.763
 media 30 1 10
 boundary 10 72 72
end geom
read array
 ara=1 nux= 18  nuy=18 typ=cuboidal
 fill
 1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1
 1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1
 1  1  5  1  1  1  3  1  1  1  1  3  1  1  1  5  1  1
 1  1  1  3  1  1  1  1  5  1  1  1  1  1  3  1  1  1
 1  1  1  1  5  1  1  3  1  1  3  1  1  5  1  1  1  1
 1  1  1  1  1  3  1  1  1  1  1  1  3  1  1  1  1  1
 1  1  3  1  1  1  1  1  1  1 42  1  1  1  1  3  1  1
 1  1  1  1  3  1  1  1  1 44  4 43  1  3  1  1  1  1
 1  1  1  1  1  1  1  1  1  1 41  1  1  1  5  1  1  1
 1  1  1  5  1  1  1  1  1  1  1  1  1  1  1  1  1  1
 1  1  1  1  3  1  1  1  1  1  1  1  1  3  1  1  1  1
 1  1  3  1  1  1  1  1  1  1  1  1  1  1  1  3  1  1
 1  1  1  1  1  3  1  1  1  1  1  1  3  1  1  1  1  1
 1  1  1  1  5  1  1  3  1  1  3  1  1  5  1  1  1  1
 1  1  1  3  1  1  1  1  1  5  1  1  1  1  3  1  1  1
 1  1  5  1  1  1  3  1  1  1  1  3  1  1  1  5  1  1
 1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1
 1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  1  end fill
 end array
 read bounds
  all=refl
 end bounds
 end model
end
=shell
  cp ft71f001 $RTNDIR/GKN.ft71
end
```

NRC FORM 335 (9-2004) NRCMD 3.7 — U.S. NUCLEAR REGULATORY COMMISSION	1. REPORT NUMBER (Assigned by NRC, Add Vol., Supp., Rev., and Addendum Numbers, if any.)
BIBLIOGRAPHIC DATA SHEET *(See instructions on the reverse)*	NUREG/CR-6969 (ORNL/TM-2008/072)
2. TITLE AND SUBTITLE Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation—ARIANE and REBUS Programs (UO2 Fuel)	3. DATE REPORT PUBLISHED MONTH: February YEAR: 2010
	4. FIN OR GRANT NUMBER Y6685
5. AUTHOR(S) G. Ilas, I. C. Gauld, and B. D. Murphy	6. TYPE OF REPORT Technical
	7. PERIOD COVERED *(Inclusive Dates)*

8. PERFORMING ORGANIZATION - NAME AND ADDRESS *(If NRC, provide Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address; if contractor, provide name and mailing address.)*

Oak Ridge National Laboratory
Managed by UT-Battelle, LLC
Oak Ridge, TN 37831-6170

9. SPONSORING ORGANIZATION - NAME AND ADDRESS *(If NRC, type "Same as above"; if contractor, provide NRC Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address.)*

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Office of Nuclear Regulatory Research
U. S. Nuclear Regulatory Commission
Washington, DC 20555–0001

10. SUPPLEMENTARY NOTES
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11. ABSTRACT *(200 words or less)*

This report is part of a report series designed to document benchmark-quality radiochemical assay data against which computer code predictions of isotopic composition for spent nuclear fuel can be validated to establish the uncertainty and bias associated with the code predictions. The experimental data analyzed in the present report were acquired from two international programs: (1) ARIANE and (2) REBUS, both coordinated by Belgonucleaire. All measurements include extensive actinide and fission product data of importance to spent fuel safety applications including burnup credit, decay heat, and radiation source terms. The analyzed four spent fuel samples were selected from fuel rods with 3.5, 3.8 and 4.1 wt % 235U initial enrichments that were irradiated in two pressurized water reactors operated in Germany and Switzerland to reach burnups in the 30 to 60 GWd/MTU range. Analysis of the measurements was performed by using the two-dimensional depletion module TRITON in the SCALE computer code system.

12. KEY WORDS/DESCRIPTORS *(List words or phrases that will assist researchers in locating the report.)*	13. AVAILABILITY STATEMENT unlimited
SCALE, isotopic measurements, radiochemical analysis, ARIANE, REBUS, experimental programs, TRITON, ORIGEN-S, spent nuclear fuel, isotopic validation, burnup credit	14. SECURITY CLASSIFICATION *(This Page)* unclassified *(This Report)* unclassified
	15. NUMBER OF PAGES
	16. PRICE

NRC FORM 335 (9-2004) PRINTED ON RECYCLED PAPER

Printed on recycled paper

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