SCALE VALIDATION EXPERIENCE USING AN EXPANDED ISOTOPIC ASSAY DATABASE FOR SPENT NUCLEAR FUEL

I. C. Gauld, G. Radulescu, and G. Ilas
Oak Ridge National Laboratory
Nuclear Science and Technology Division
P.O. Box 2008
Oak Ridge, TN 37831-6370 USA
gauldi@ornl.gov

The availability of measured isotopic assay data to validate computer code predictions of spent fuel compositions applied in burnup-credit criticality calculations is an essential component for bias and uncertainty determination in safety and licensing analyses. In recent years, as many countries move closer to implementing or expanding the use of burnup credit in criticality safety for licensing, there has been growing interest in acquiring additional high-quality assay data. The well-known open sources of assay data are viewed as potentially limiting for validating depletion calculations for burnup credit due to the relatively small number of isotopes measured (primarily actinides with relatively few fission products), sometimes large measurement uncertainties, incomplete documentation, and the limited burnup and enrichment range of the fuel samples. Oak Ridge National Laboratory (ORNL) recently initiated an extensive isotopic validation study that includes most of the public data archived in the Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA) electronic database, SFCOMPO, and new datasets obtained through participation in commercial experimental programs. To date, ORNL has analyzed approximately 120 different spent fuel samples from pressurized-water reactors that span a wide enrichment and burnup range and represent a broad class of assembly designs. The validation studies, completed using SCALE 5.1, are being used to support a technical basis for expanded implementation of burnup credit for spent fuel storage facilities, and other spent fuel analyses including radiation source term, dose assessment, decay heat, and waste repository safety analyses. This paper summarizes the isotopic assay data selected for this study, presents validation results obtained with SCALE 5.1, and discusses some of the challenges and experience associated with evaluating the results. Preliminary results obtained using SCALE 6 and ENDF/B-VII cross sections libraries are also briefly summarized.

I. INTRODUCTION

Oak Ridge National Laboratory (ORNL) has been performing spent-fuel isotopic validation studies using the depletion analysis methods in the SCALE [1] code system for the past 20 years. These studies involve comparisons of calculated inventories against measured isotopic composition data obtained from destructive radiochemical analysis of commercial spent nuclear fuel samples. The results of these benchmark studies are used to quantify the bias and uncertainties associated with isotopic calculations and ultimately determine appropriate margins for uncertainty that can be applied in safety-related analyses such as burnup credit in criticality calculations, decay heat analysis, and source terms. Previous studies using several versions of SCALE and nuclear data libraries have been published in multiple validation reports [2–6] that evaluate selected experimental data obtained largely from public sources.

A study was recently initiated at ORNL with the objectives of updating and expanding the validation calculations using a comprehensive database of experimental isotopic assay data that includes isotopic composition data obtained from both publicly available sources and international commercial programs. As part of the study, an extensive isotopic database of nearly 120 measured spent fuel samples with an expanded range of initial enrichments and burnup values compared to previously analyzed data was reviewed and analyzed. The calculations were performed using two-dimensional (2-D) assembly models and a consistent set of modeling assumptions using the SCALE 5.1 code system and ENDF/B-V 44-group cross section library. As part of the current study, detailed benchmark modeling information and measurement data are being documented in a format that is readily usable for validating depletion and decay codes. The work is being extended to include analysis results using SCALE 6 and the ENDF/B-VII 238-group cross section library.

This paper describes the isotopic composition data evaluated in this study and highlights the preliminary findings to date.

II. EXPERIMENTAL ISOTOPIC VALIDATION DATA

The isotopic composition data evaluated for this study included 118 samples obtained from spent fuel irradiated in the following nine different pressurized-water reactors (PWRs):

- Trino Vercellese
- Obrigheim
- Turkey Point Unit 3
- H. B. Robinson Unit 2
- Calvert Cliffs Unit 1
- Three Mile Island (TMI) Unit 1
- Takahama Unit 3
- Gösgen
- Neckarwestheim (GKN) Unit 2

Many of these experiments are available in the public domain and are documented in the Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA) isotopic composition database, SFCOMPO [7]. The Trino Vercellese, Obrigheim, Turkey Point Unit 3, H. B. Robinson Unit 2, Calvert Cliffs Unit 1, and Takahama Unit 3 experiments have been previously analyzed by ORNL using earlier releases of SCALE and one-dimensional (1-D) assembly depletion models. The Obrigheim data analyzed previously included isotopic data for five fuel assemblies that were reprocessed at the Karlsruhe reprocessing plant. The current study includes additional Obrigheim measurements of fuel rod samples measured under an experimental program of the Joint European Commission (JRC). A common drawback of these data is the general lack of fission product measurements. Many of the early experiments focused mostly on measurements of major actinides for studies of uranium transmutation and plutonium production, and nuclides required for burnup determination of the fuel (e.g., ^{137}Cs and ^{148}Nd). Several later programs (Calvert Cliffs and Robinson) performed under the Office of Civilian Radioactive Waste Management (OCRWM) expanded the measurements to include radiological important isotopes such as ^{90}Sr, ^{137}Cs, and ^{154}Eu to support nuclear fuel safety analysis and ^{14}C, ^{135}Cs, ^{99}Tc, ^{126}Sn, ^{129}I and other long-lived fission products for waste repository analysis. However, very few experimental programs include many fission product isotopes considered important to burnup credit, with the exception of the Takahama measurements performed specifically to support the burnup-credit program in Japan by the Japan Atomic Energy Agency (JAEA), formerly the Japan Atomic Energy Research Institute (JAERI).

The current lack of sufficient experimental data for fission products is seen as one of the principal impediments to expanding the use of fission products in burnup-credit analyses. The data requirements for burnup credit have broadened the isotopes of interest to include many stable and long-lived fission products with large capture cross sections for which very few measurements were previously available, such as lanthanide isotopes of Sm, Eu, and Gd and several stable fission products including ^{95}Mo, ^{101}Ru, ^{103}Rh, ^{109}Ag, and ^{133}Cs. The largest six fission product contributors, ^{143}Nd, ^{149}Sm, ^{103}Rh, ^{151}Sm, ^{133}Cs, and ^{155}Gd, represent about 75% of the total fission product reactivity effect for typical fuel. Based on the experimental data widely available circa 2001, the isotopic validation studies performed to support development of the Nuclear Regulatory Commission regulatory guidance for burnup credit, ISG-8 revision 2 [8], included few samples with extensive fission product data. Only the Takahama measurements and Calvert Cliffs measurements, made under an extension program for burnup credit, provided fission product assay data relevant to validating depletion calculations for burnup-credit analyses. However, these experiments provided only one measurement of ^{103}Rh, three measurements of ^{133}Cs, and four measurements of ^{155}Gd.

Since 2001, ORNL has worked to acquire additional high-quality isotopic assay data to support the validation of the SCALE system. New data from commercial programs include measurements of Neckarwestheim-2 fuel from the international REBUS program and Gösgen fuel from the ARIANE and MALIBU international programs, currently managed by the Studiecentrum voor Kernenergie—Centre d'Etude de l'Energie Nucléaire (SCK•CEN) in Mol, Belgium. These programs provide high-quality data that involve multi-laboratory cross check measurements. Several programs have focused on high-burnup fuel to address current safety and licensing issues related to high-burnup fuel. Note however that for burnup credit, the range of interest is in lower-burnup fuel, particular burnup values typical of the ends of the fuel assembly, where reactivity is largest. Additional fuel data with extensive fission product measurements were obtained from TMI-1 fuel assay measurements, performed as part of the OCRWM Yucca Mountain Project (YMP). The TMI-1 data are open but are not widely known. The recently acquired program data include modern assembly designs and higher enrichment and burnup fuel samples, provide measured data for approximately 60 nuclides, and include isotopes relevant to burnup credit, decay heat (e.g., ^{134}Cs), and longer-term waste management safety (e.g., ^{79}Se, ^{135}Cs) analyses, as well as isotopes of radiological importance (neutron and gamma-ray emitters such as ^{244}Cm and ^{137}Cs).

An important attribute of this study is that virtually all samples from the selected experimental programs were included in the validation exercise. Only in the case of gross errors identified in the measurement, or in the case of Takahama samples located in very close proximity to the active end of the fuel rods where three-dimensional (3-D) neutron transport models are required to account for increased moderation near the ends, were samples or specific isotopic measurements rejected for this work. The intent of this study was, in part, to investigate the results obtained using a more comprehensive population of data compared to previous studies that typically used a selected subset of samples for validation.

The 118 fuel samples evaluated in this study, summarized in Table I, include initial enrichments from 2.453 to 4.657 wt % ^{235}U, and the sample burnup values vary from 7.2 to 70.4 GWd/MTU. The samples were obtained from both UO_2 and gadolinia fuel rods from typical UO_2 assemblies, some of which contained burnable poison rods. All evaluated samples provided measured data for the major actinide isotopes ^{235}U, ^{238}U, and ^{239}Pu through ^{242}Pu. The number of samples with measurement data for fission products relevant to burnup credit varied widely depending on the nuclide (e.g., from 13 samples for ^{133}Cs to 48 samples for ^{143}Nd and ^{145}Nd). The number of measurements available for the major fission products in burnup credit in this study is compared to prior work in 2002 in Table II.

Table I Summary of PWR Experimental Assay Data Used for Validation

Reactor	Measurement Laboratory[a]	Experimental Program[b]	Assembly Design	Enrichment (wt % ^{235}U)	No. of Samples/ Fuel Rods	Burnup (GWd/t)
Trino Vercellese	Ispra, Karlsruhe	JRC	15 × 15	2.72, 3.13, 3.897	15/5	7.2–17.5
	Ispra, Karlsruhe	JRC	15 × 15	3.13	16/5	12.8–25.3
Obrigheim	Ispra, Karlsruhe	JRC	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
H. B. Robinson-2	PNNL	ATM-101	15 × 15	2.561	4/1	16.0– 31.7
Turkey Point-3	Battelle-Columbus	NWTS	15 × 15	2.556	5/2	30.5–31.6
Calvert Cliffs-1	PNNL, KRI	ATM-104	14 × 14	3.038	3/1	27.4–44.3
	PNNL	ATM-103	14 × 14	2.72	3/1	18.7–33.2
	PNNL, KRI	ATM-106	14 × 14	2.453	3/1	31.4–46.5
Takahama-3	JAERI	JAERI	17 × 17	2.63, 4.11	16/3	7.8–47.3
TMI-1	ANL	DOE YMP	15 × 15	4.013	11/1	44.8–55.7
	GE-VNC	DOE YMP	15 × 15	4.657	8/3	22.8–29.9
Gösgen	SCK•CEN, ITU	ARIANE	15 × 15	3.5, 4.1	3/2	29.1–59.7
	SCK•CEN, PSI, CEA	MALIBU	15 × 15	4.3	3/1	47.2, 67.9, 70.4
GKN-2	SCK•CEN	REBUS	18 × 18	3.8	1/1	54.0

[a]ANL = Argonne National Laboratory; GE-VNC = General Electric Vallecitos Nuclear Center; PNNL = 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'Etude de l'Energie Nucléaire; PSI = Paul Scherrer Institute; CEA = Commissariat à l'Énergie Atomique.

[b]JRC = European Joint Nuclear Research Centre; ICE = European Isotopic Correlation Experiment; ATM = Approved Testing Material; NWTS = National Waste Terminal Storage Program; DOE YMP = U.S. Department of Energy Yucca Mountain Project; ARIANE, MALIBU, and REBUS are International Experimental Programs coordinated by Belgonucleaire—currently managed by SCK•CEN, Belgium.

Table II Summary of PWR Measurements for Major Burnup-Credit Fission Products

Fission product	Number of available measurements	
	ISG 8 – 2 (2002)	Expanded database (2009)
^{149}Sm	9	38
^{143}Nd	14	48
^{103}Rh	1	18
^{151}Sm	9	38
^{133}Cs	3	13
^{155}Gd	4	30

III. DATA EVALUATION

Prior to using the measured isotopic composition data for code validation, the data were first critically evaluated to identify potential errors associated with the actual measurements or documentation of the experiments. This evaluation was performed using several measures of experimental accuracy:

- Self-consistency of experimental data as a function of sample burnup and/or sample height, particular for multiple samples measured within the same fuel rod
- Self-consistency of measurement data for sample parent-daughter pairs, for example, ^{241}Pu–^{241}Am, ^{155}Eu–^{155}Gd, ^{144}Ce–^{144}Nd, ^{147}Pm–^{147}Sm
- Consistency of results from other experimental programs that have measured similar fuels

Isotopic composition data reported by laboratories were converted to a common mass unit of milligrams per gram of initial (unirradiated) uranium to facilitate comparison of measurement data from different experiments and provide a consistent basis for comparison of measured and calculated isotopic concentrations. Measurement uncertainties were assigned using values obtained either directly from primary references or, in cases where uncertainties were either not reported or deemed to be unreliable, by evaluating the variability of reported measurement results for different measurements made by the laboratory. This evaluation showed that laboratories used an inconsistent basis in determining measurement uncertainties. Some laboratories included a true estimate of the total uncertainty contributed by all phases of the measurement—from fuel cutting, weighing, dissolving, separations, standards, and mass spectrometry (MS), etc.—while other laboratories appear to have reported the precision associated with repeatability of the MS measurement.

Some problems observed in the data evaluation were attributed to the method of back calculating isotopic compositions to a reference date, often to the time of discharge. For example, the Takahama measurement data for ^{241}Am were found to be highly erratic due to the procedure of adjusting the measured compositions to the time of discharge. The problem arises because the majority of ^{241}Am at the time of measurement is from the decay of ^{241}Pu. Subtracting the large ^{241}Pu contribution results in a small ^{241}Am value with a very large uncertainty. Other errors in data adjustment have been found for isotopes that have decay precursors that were not measured, leading to large errors when the isotope concentration is adjusted for decay only, and neglecting the contribution from the precursor. Other errors were found in the reporting, likely due to typographical errors that produced measurement results that were not physically possible.

Only a very small number of data points were identified as being likely outliers based on the evaluation of measurement data.

Other data were evaluated with respect to the ability of the computational models to accurately represent the fuel assembly neutronics. Three samples from Takahama, one from each of the three measured rods SF95, SF96, and SF97, were removed from the considered dataset due to their proximity to the active ends of the fuel rod. These samples require 3-D models to accurately account for the spectral change near the ends of the fuel [9]. The use of 2-D analysis models used in this study resulted in anomalous results for isotopes that are very sensitive to the neutron spectrum.

IV. COMPUTATIONAL MODELING

Computational analysis of the measurements was carried out using the TRITON depletion analysis sequence in SCALE 5.1, which couples the 2-D discrete-ordinates transport code NEWT with the

depletion and decay code ORIGEN-S to perform the isotopic depletion and decay simulation. Cross sections for the ORIGEN-S analysis are generated by NEWT using the 44GROUPNDF5 cross-section library, which is based on ENDF/B-V data. The ENDF/B-V multigroup cross sections, available for 232 isotopes, are collapsed by NEWT using the flux solution for the fuel at each time interval and applied in the ORIGEN-S library. Cross sections for all other isotopes in the ORIGEN-S library are also updated to account for the flux changes in the fuel during irradiation. Cross section resonance processing was performed using the established NITAWL module of SCALE. New cross section libraries based on ENDF/B-VII data are available in SCALE. However, the present validation calculations are based on the ENDF/B-V data libraries that have been used extensively by ORNL for spent-fuel isotopic predictions for more than a decade and are widely used by other organizations, in order to provide a validation baseline against which the performance of newer data can be assessed.

Two-dimensional TRITON depletion calculation models were developed for each of the samples considered using assembly design and operating data available in primary reference documents. A consistent modeling methodology was developed to derive input values for parameters with no documented data. Input parameters frequently not available in documentation, particularly in some of the older experimental reports, include

- moderator density at the sample location (axial height) of the fuel rod,
- effective temperature of fuel sample,
- soluble boron concentration in the moderator,
- information related to absorber rod exposure,
- assembly pitch (gap between adjacent assemblies), and
- information on surrounding neighbor assemblies that can be important to modeling samples from fuel rods located at the periphery of the assembly.

The depletion histories of the fuel rods from which the samples were selected, including samples selected from rebuilt Gösgen fuel assemblies, were explicitly simulated with TRITON. The geometry of the assemblies hosting these fuel rods was modeled in full detail. All details on material and irradiation history data, as available, were included in the TRITON model. The time-dependent variation of the soluble boron content in the coolant, temperature in coolant or fuel, and coolant density during irradiation was also included in the model when data were provided by the utility or fuel vendor.

An example of the modeling detail is provided in Fig. 1 for the H. B. Robinson assembly B-05 showing the location of the measured fuel rod (test pin) and neighboring fuel rods. The model is developed using a one-quarter assembly representation, taking advantage of the assembly symmetry. Separate models were developed to represent the borosilicate glass absorber rods, inserted in the guide tubes during the first irradiation cycle, and removal of the rods in the second cycle. The fuel rod isotopic compositions after cycle 1 are transferred to the initial fuel rod compositions in the cycle 2 model. A reflective boundary condition was applied to the outer boundary of this configuration. Individual depleting mixtures were specified for the measured rod and its nearest neighbor fuel rods, as illustrated in Fig. 1; all other fuel rods in the assembly were treated as a single depletion material with uniform composition, as their impact on the neutron flux spectrum in the fuel rod from which samples were measured has been found to be a second-order effect.

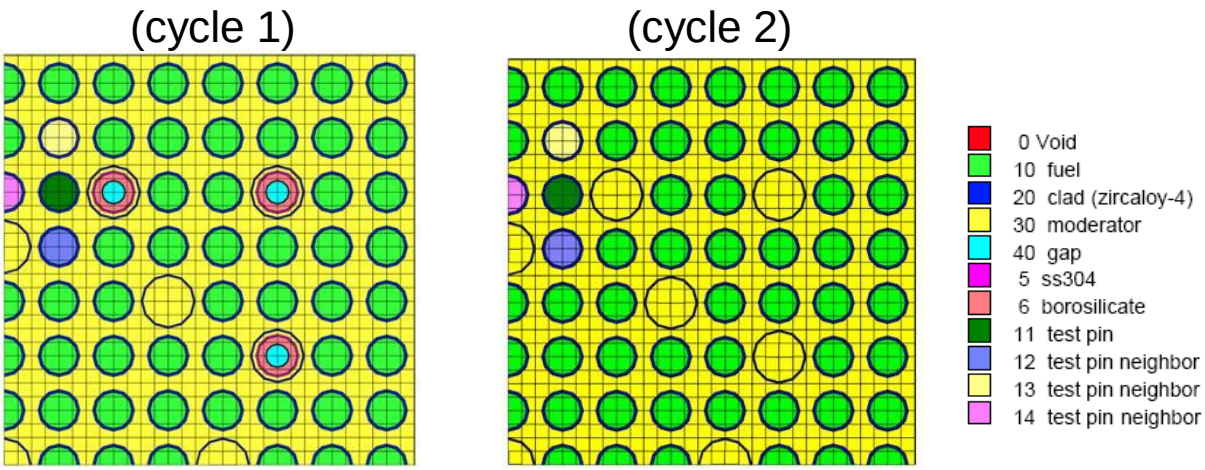

Fig. 1. TRITON/NEWT ¼ assembly models for H. B. Robinson assembly B0-5.

V. ENDF/B-V RESULTS

The isotopic results were analyzed by comparing the calculated isotopic contents with the measured values for each of the 118 fuel samples, as the percent difference between calculated (C) and experimental (E) mass values, that is, (C/E — 1)%. The results were reviewed to identify samples with significant deviations compared to results of other similar samples that would warrant further investigation. The characteristics of the (C/E — 1)% frequency distributions for the isotopes relevant to safety-related analyses were examined further to establish the requirements for a data analysis methodology for use in isotopic bias and bias uncertainty determination.

The results for ^{235}U and ^{239}Pu isotopes, which are important indicators of measurement data and modeling accuracy, and two important burnup-credit fission products, ^{149}Sm and ^{103}Rh, are plotted in Fig. 2. These results have characteristics common to many other nuclides. The error bars ($\pm 1\ \sigma$) reflect the experimental uncertainties as reported by the measurement laboratory but do not include added uncertainty due to burnup error, also measured experimentally. The additional nuclide uncertainty arising from burnup uncertainty is dependent on the nuclide. For example, ^{235}U is very sensitive to changes in burnup, whereas the ^{239}Pu concentration is less sensitive to changes in burnup over the range of typical discharged fuel. In the case of the TMI-1 data from assembly NJ05YU (2-cycle fuel), the uncertainty in the burnup value typically exceeds 7%. Consequently the error bars shown in Fig. 2 for these TMI-1 measurements are in some cases underestimated.

Large ^{235}U deviations are seen for the Obrigheim samples from fuel rods located at the edge of assembly BE210 (5 to 11%) and for the set of TMI-1 samples from assembly NJ05YU (-18 to +21%). The calculated ^{239}Pu concentrations for these Obrigheim and TMI-1 samples were also overestimated on average by up to 11 and 20%, respectively. The difference between measured and calculated ^{235}U and ^{239}Pu concentrations may be due, in the case of the Obrigheim BE210 samples, to incomplete documentation of the operating/design data. Obrigheim is known to have operated with mixed oxide (MOX) fuel assemblies at the time that assembly BE210 was irradiated; however, the core locations of the MOX assemblies were not provided in the primary reference documents. In the case of the TMI-1 NJ05YU samples, the large uncertainties associated with the burnup have been established as the major source of the variability observed in the ^{235}U results shown in Fig. 2.

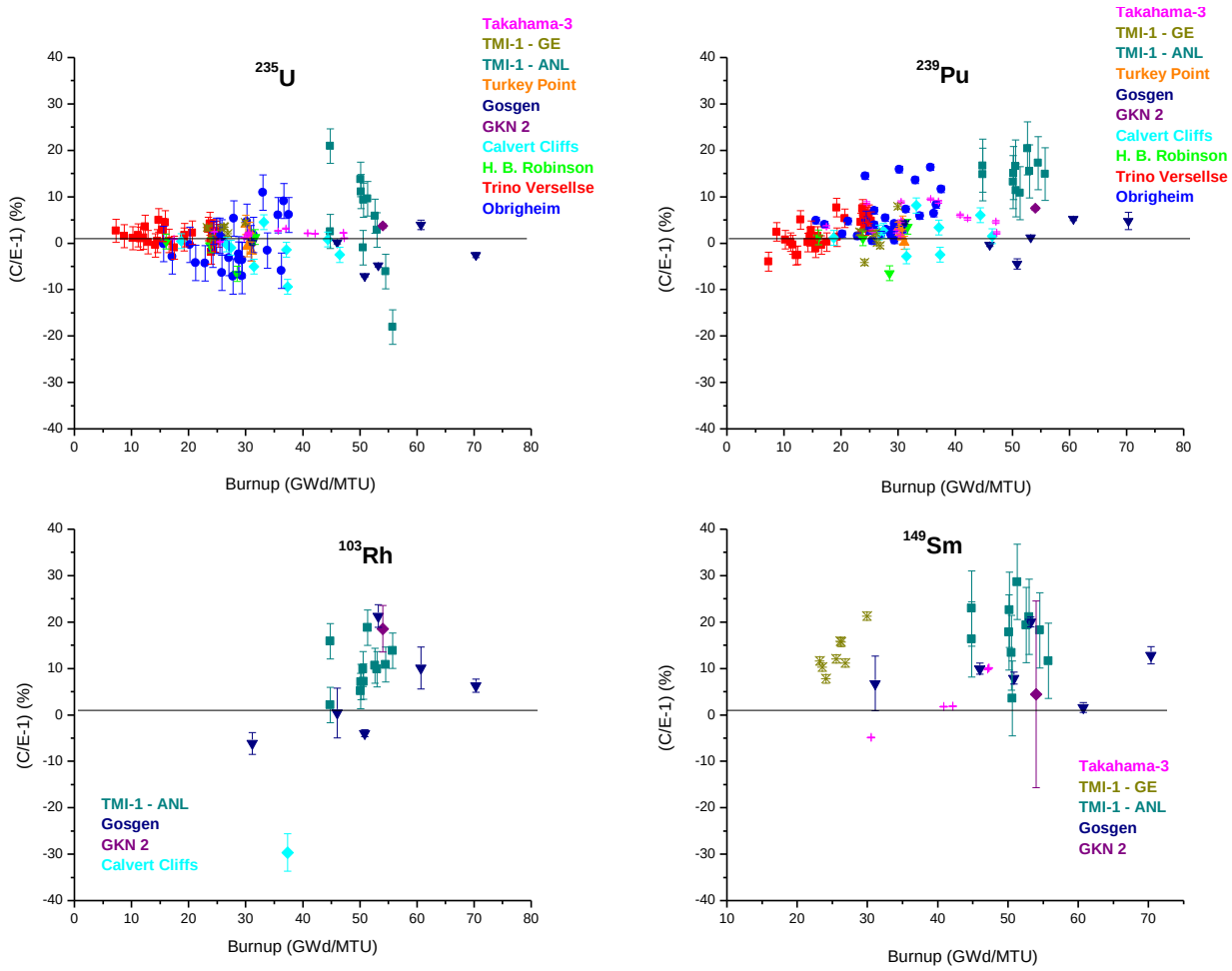

Fig. 2. Percent differences between calculated and measured concentrations vs. burnup for ^{235}U, ^{239}Pu, ^{149}Sm, and ^{103}Rh. Error bars included reported measurement uncertainties (1 standard deviation).

The reported uncertainties for sample burnup varied greatly among the evaluated samples, from less than 1% for many samples to more than 7% for the TMI-1 NJ05YU assembly samples. The TMI-1 NJ05YU samples are of high interest because they provide important measurement data for fission products relevant to burnup credit. There are currently plans to perform additional isotopic cross check measurements of samples from the same TMI-1 rod to confirm some of the earlier results. Another observation is that several laboratories report uncertainties that appear unrealistically small. If measurement uncertainties are used to weight different data sets, it will be critical to ensure that a consistent basis for the uncertainty values is used in order to avoid over or under weighting certain data sets.

The bias and bias uncertainty associated with the calculation of each burnup-credit isotope may be determined based on the assumption that the (C/E — 1)% results for all evaluated spent fuel samples are independent variables that follow a normal distribution. Shapiro-Wilk normality tests performed on the data however show that the results for the majority of the nuclides do not pass the normality test at the 0.05 significance level. The data distributions for many of the burnup-credit nuclides are unimodal and peaked (positive kurtosis) or skewed. Several nuclides with smaller sample sizes, such as ^{133}Cs, ^{151}Eu, and ^{153}Eu, appear to have bi-modal distributions.

The characteristics of the data distributions are attributed in some cases to the following.

- Unbalanced number of samples among the experimental datasets
  - Measurements for GKN II involved one sample, whereas Trino Vercellese included 31 samples all having fairly similar properties.
  - The combined number of Trino Vercellese and Obrigheim spent-fuel samples represents about half of the total number of the evaluated samples.
  - Thus, the selection of samples gives more weight to some of the experimental datasets in the distribution of results.
  - Ideally, the sample sizes will be uniform and representative of a broader range for fuel characteristics.
- Significantly different measurement uncertainties among the experimental datasets
  - Experimental isotopic concentrations characterized by large measurement uncertainties have greater potential to generate anomalous values in the data that tend to increase the estimate of sample variance.
  - Reported uncertainties may not be reliable for weighting different measurements.
- Systematic measurement bias (i.e., correlation within an experimental dataset) characterizing some of the experimental datasets, observed as significantly different sample means and/or variances between the different datasets.
- Systematic bias in the calculated isotopic concentration values (i.e., correlation within an experimental dataset) due to assumptions or uncertainties in the modeling data for some of the spent fuel samples.

A statistical test for trending was also applied to the data, with most data sets showing no significant trending with burnup. For data that did show a positive trend, it is not clear, given the variability in the results and biases observed between the different experimental programs, if the trend is the result of an actual dependence on the code bias with burnup or an artifact of the biases between measurements performed at different laboratories. Trending is more reliably determined using measurements performed at one laboratory using similar fuel rod samples that span a large burnup range.

The relative standard deviation (RSD) of the (C/E — 1)% isotopic results, based on results for 118 evaluated samples, varied from 0.37% (^{238}U) to 23% (^{242m}Am) for actinides and from 2.18% (^{133}Cs) to 36.03% (^{109}Ag) for fission products important to burnup credit. The RSD values for ^{235}U and ^{239}Pu were 4.6 and 5.9%, respectively, including all 18 TMI-1 and 7 Obrigheim BE210 samples characterized by relatively large uncertainties in the measured data and input modeling parameters.

VI. PRELIMINARY ENDF/B-VII RESULTS

Preliminary results have been obtained using the SCALE 6 238-group ENDF/B-VII cross section library. Because of the increased number of resonances and the format representation of the resonance cross sections in ENDF/B-VII, the CENTRM/PMC resonance self-shielding methods are required. A problem-dependent pointwise energy flux (50,000–70,000 energy points) solution from a CENTRM 1-D transport calculation is determined for the system's components (i.e, fuel pin cells in an assembly). The CENTRM flux solution is then used by PMC to collapse pointwise cross section data over the resonance region to obtain problem-dependent multigroup cross sections (238-group) that are used in the transport calculations, and collapsed for use in the ORIGEN-S burnup simulations.

Samples analyzed for these studies were selected from the MALIBU program data because of the high quality and confidence in the measurement data and the large number of isotopes measured. Three

samples, all obtained from the same fuel rod, were evaluated, having a burnup range from 46.0 to 70.4 GWd/t. Results for isotopes of Am, Np, Cm, Sm, Gd, Eu, and Cs are shown in Fig. 3. Results for the major uranium and plutonium isotopes show no significant differences compared to results obtained using ENDF/B-V cross sections. Significant improvements in the calculated isotopic compositions are observed for the ^{245}Cm and ^{246}Cm, ^{155}Eu and ^{155}Gd, ^{133}Cs and ^{134}Cs, and all Sm isotopes. Initial studies indicate that improvements in the evaluated cross section data, more than the CENTRM processing methods, are responsible for most of the change. The new ENDF/B-VII data have the potential to significantly reduce the bias associated with calculations of fission products important to burnup credit, because ^{133}Cs, ^{149}Sm, ^{151}Sm, and ^{155}Gd represent four of the six most important fission product isotopes.

VII. CONCLUSIONS

Isotopic composition data for nearly 120 PWR spent fuel samples obtained from publicly available sources and from commercial programs have been evaluated for use in validating SCALE depletion calculations. The increased number of available fission product measurements provides an improved experimental basis to evaluate the bias and uncertainty in the calculated isotopic content of spent fuel as compared to previous studies. These data have been analyzed by ORNL using the 2-D depletion analysis methods in SCALE 5.1 and the 44-group ENDF/B-V cross section library. These results provide a reference basis for ongoing calculations being performed using SCALE 6 with ENDF/B-VII cross section data.

Evaluation of experimental measurements was performed to vet the data for errors associated with measurements and/or reporting. Although some datasets were subject to large measurement uncertainties or uncertainties related to the actual irradiation of the fuel in the reactor, effectively all samples were retained in this study to provide a comprehensive study of all current data. The only samples not included were the Takahama samples located in close proximity to the active end of the fuel rod that could not be accurately modeled using 2-D methods.

An assumption at the onset of this analysis was that by including many more samples in the analysis compared to any previous study, the statistical behavior of the data would improve. In fact, the distribution of results for many nuclides in this study failed the normality test. It is clear that correlations in the data due to laboratory measurement methods and/or modeling have a significant influence on the results. Experimental results involving multiple measurements for a single fuel rod or assembly, or measured at a single laboratory, have a very strong influence on the frequency distributions of data. Any bias associated with these measurements or the modeling data will be propagated in a similar way to all samples. These correlations tend to result in observed clustering of results for different programs. The statistical characteristics of the validation results are currently being evaluated to ensure that appropriate statistical analysis methods for isotopic composition bias determination are applied in determining isotopic bias and uncertainty values for safety-related applications.

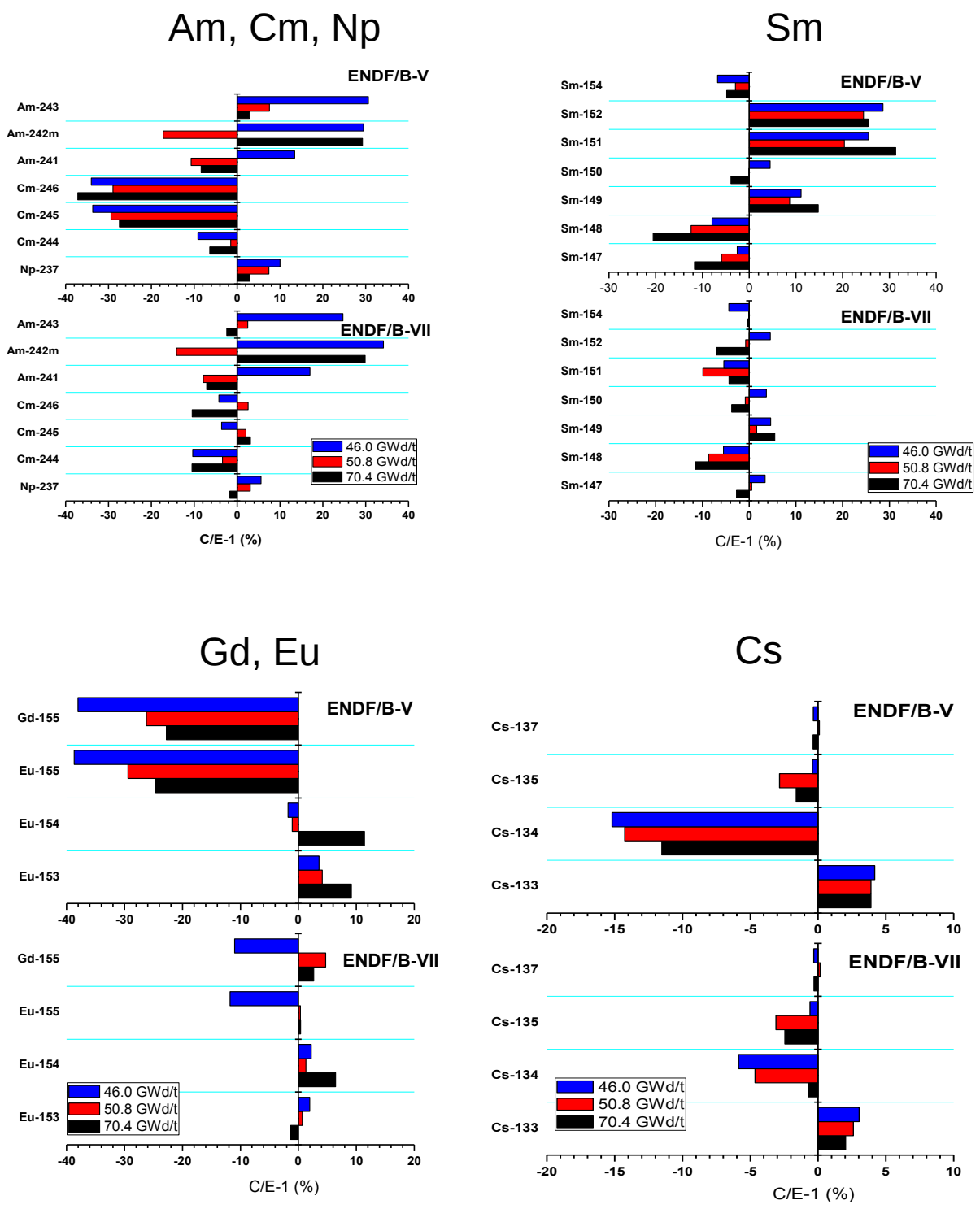

Fig. 3. Comparison of isotopic results obtained using ENDF/B-V and -VII SCALE Libraries.

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