SCALE 6 ANALYSIS OF ISOTOPIC ASSAY BENCHMARKS FOR PWR SPENT FUEL

Germina Ilas and Ian C. Gauld

Oak Ridge National Laboratory, Oak Ridge, TN, 37831-6170, ilasg@ornl.gov

Validation of the depletion capabilities and associated data libraries in the recently released SCALE 6 code system is ongoing at ORNL. This paper presents results of the analysis of isotopic assay measurement data for PWR spent fuel using SCALE 6 and ENDF/B-VII data libraries.

I. INTRODUCTION

A sustained effort is ongoing at Oak Ridge National Laboratory (ORNL) to compile and document high-quality radiochemical assay data for use in validating calculated isotopic compositions in spent fuel. The compiled database includes up-to-date measurement data for more than 120 samples from pressurized water reactors (PWR) spent fuel. This database is being applied to validate the depletion capabilities in the ORNL SCALE 6 code system [1]. Quantifying and evaluating the uncertainties in the calculated fuel composition are essential for understanding and reducing the uncertainties associated with spent fuel transportation and storage applications that involve decay heat, radiation sources, and criticality safety evaluations with burnup credit.

Depletion analysis can be performed in SCALE 6using the TRITON [2] depletion sequence. TRITON couples the ORIGEN-S point depletion and decay code with either the two-dimensional (2D) arbitrary-geometry discrete ordinates transport code NEWT or the three-dimensional Monte Carlo transport code KENO and computes isotopic concentrations for more than 1,400 nuclides. A major advancement in SCALE 6 is the addition of a multigroup transport library that is based on ENDF/B-VII data. The use of this library in a previous analysis of three spent fuel sample measurements from the MALIBU program showed [3] that several of the fission product isotopes of importance to spent fuel safety applications are calculated significantly better with the new library compared to computations carried out with previous SCALE libraries.

Additional validation results for SCALE 6 depletion codes and libraries are presented in this paper. The analysis includes isotopic measurement data for five uranium dioxide (UO_2) spent fuel samples from experiments performed in the ATM [4], ARIANE [5], and REBUS [6] experimental programs; benchmark specifications for two of these programs are included or soon to be included in the SFCOMPO database [7].

II. VALIDATION DATA

The experimental data used for validation in this study include measured isotopic compositions for five samples selected from fuel irradiated in the Calvert Cliffs, Gösgen, and GKN II PWRs that are operated in the United States, Switzerland, and Germany, respectively. The experimental measurements included a large number of actinides and fission products of importance to spent fuel safety applications.

The measurements on three spent fuel samples from the Calvert Cliffs reactor that are considered in this paper were carried out in 1986 at the Material Characterization Center at Pacific Northwest National Laboratory for the ATM Program designed to characterize medium-burnup spent fuel representative of reactors operating in the United States. Lanthanide measurements for the same three samples have been also performed at Khoplin Radium Institute in Russia. These three samples were selected from a rod of assembly D047 that was irradiated in the reactor for four consecutive cycles. The assembly, with a CE 15 x 15 design, had an initial fuel enrichment of 3.038 wt % ^{235}U, and the samples under consideration covered a burnup range from 27 to 44 GWd/MTU.

Three UO_2 samples selected from fuel rods irradiated in the Gösgen reactor were measured in the ARIANE program, an international program designed to improve the database of isotopic measurements for spent fuel source term and isotopic inventory validation, which was completed in 2001. The sample considered here, identified as GU3, was taken from a fuel rod in a rebuilt assembly, with 15 x 15 design and initial fuel enrichment of 4.1 wt% ^{235}U, which was irradiated for three reactor cycles. The sample had an estimated burnup of 52.5 GWd/MTU.

The REBUS international program was dedicated to the validation of computer codes for criticality calculations employing burnup credit. The program, completed in 2005, included measurements on a UO_2 sample from an 18×18 assembly irradiated in the

GKN II reactor. The measured sample had an initial enrichment of 3.8 wt % ^{235}U and a burnup of about 54 GWd/MTU.

III. COMPUTATIONAL MODELS

The computational analysis of the measurements was carried out using the 2D depletion option in TRITON, in which the 2D transport code NEWT is coupled with the depletion and decay code ORIGEN-S to perform the burnup simulation. The new SCALE 238-group cross section library based on ENDF/B-VII data and the CENTRM/PMC cross section processing methodology were used in the NEWT transport calculation. A set of 232 isotopes for which cross section data are available in the multigroup transport library used with NEWT were applied in updating cross sections for the ORIGEN-S fuel depletion calculation at each depletion step.

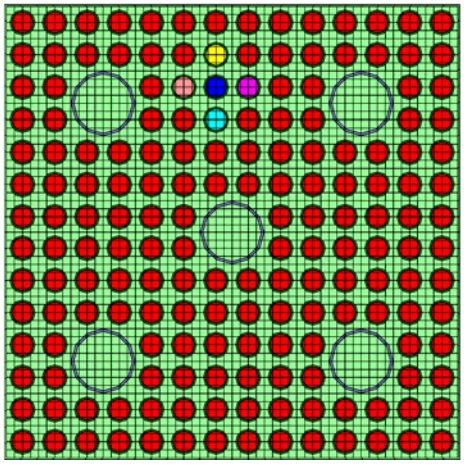
Individual TRITON models were developed for each of the samples considered. The depletion history of the fuel rods from which the samples were selected, including the reconstitution of the fuel assembly in the case of sample GU3, was explicitly simulated with TRITON. The geometry of the assembly hosting these fuel rods was modeled in full detail. All the details on material and irradiation history data, as available, were included in the TRITON model. The variation during irradiation of the soluble boron content in the coolant, temperature in coolant or fuel, and coolant density was also included in the model when data were available.

The TRITON assembly models are illustrated in Fig. 1(a) for the Calvert Cliffs samples and Fig. 1(b) for sample GU3. Taking advantage of the fuel assembly symmetry, a quarter assembly model was used for the analysis. Individual depleting mixtures were specified for the measured rod and its nearest neighbor fuel rods, as indicated by the use of different colors in Fig. 1; all other fuel rods in the assembly were treated as a single depletion material with uniform composition, given that their impact on the neutron flux spectrum in the measured fuel rod is a second-order effect.

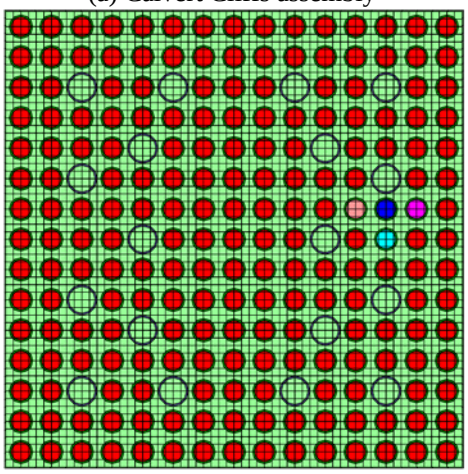
IV. RESULTS

A comparison of the calculated and measured isotopic content in the samples, using the calculated-to-experimental ratio (C/E), is presented in Fig. 2 for major actinides and selected fission products – cesium, neodymium, and samarium isotopes. The results are shown as obtained with SCALE 5.1 and ENDF/B-V libraries, as well as with the recently released SCALE 6 and ENDF/B-VII libraries.

As seen in Fig. 2, the overall agreement calculation-experiment for most major actinides is similar for both libraries used; the average C/E over the five samples for ^{235}U and ^{239}Pu is about 0.5 and 5%, respectively, in both cases.



(a) Calvert Cliffs assembly



(b) Gösgen assembly

Fig. 1. Illustration of TRITON models

Cesium and neodymium isotopes, of importance to reactivity of spent fuel (burnup credit), source terms, or used as burnup indicators, are well estimated with both libraries. The exception is ^{134}Cs, important to decay heat and gamma sources at short cooling times, for which the C/E is improved by about 10% if using ENDF/B-VII data.

Large improvement in C/E is seen for samarium nuclides, when using the ENDF/B-VII libraries, specifically for those with large absorption cross section and of importance to burnup credit criticality calculations. The average C/E ratios for ^{151}Sm and ^{152}Sm decrease from the 30% range when using ENDF/B-V data to less than 5% when using the new ENDF/B-VII libraries.

More samples would need to be added to the set analyzed in the current paper to be able to draw conclusions about systematic behavior and trends in the C/E results as a function of burnup or other parameters.

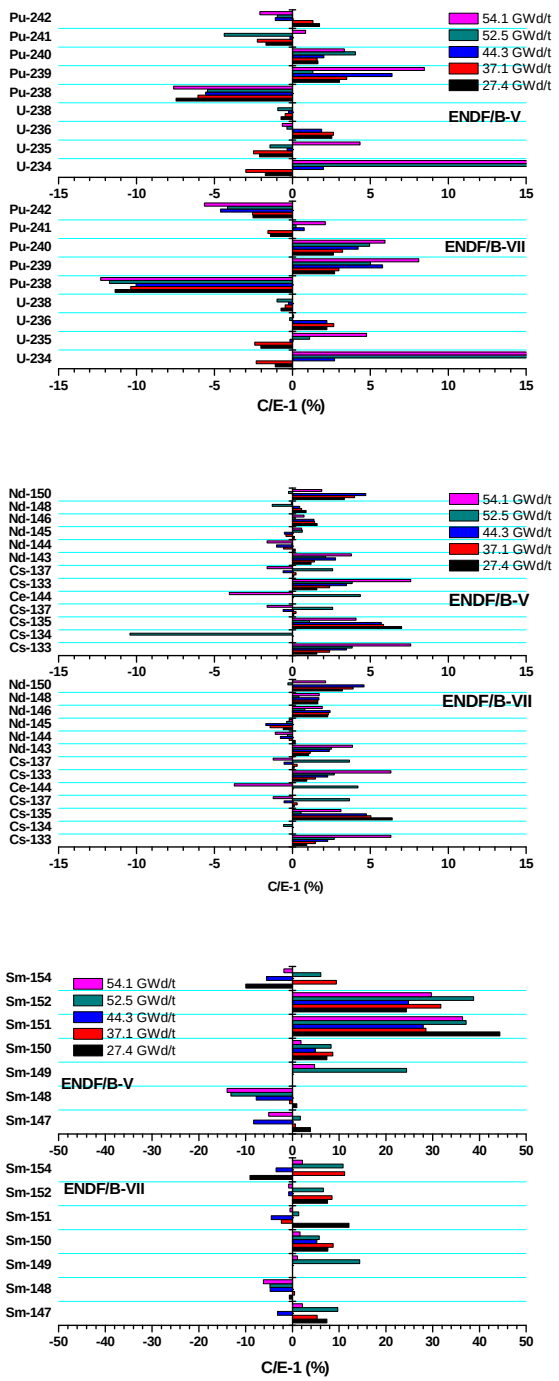


Fig. 2 Relative C/E comparison

The evaluation of the data is a work in progress and the comparison shows here the C/E ratios only, not including uncertainties that may be due to uncertainties in experimental data or computational models parameters. However, when evaluating the level of agreement between calculation and experiment, one should also consider the experimental errors as well as other problems or limitations related to measurement or data required for simulations. Both the uncertainty in the data used to build the computational model and the measurement uncertainty are manifested in an observed code bias, and there is no prescription on how to isolate and quantify the sources of uncertainties. The isotopic radiochemical measurement is a very complex process with a highly elaborate methodology that includes not only the mass spectrometric analysis, but also many other steps, such as dissolution of the sample to obtain a solution to be used for analyses, chemical separations of elements from the solution before measuring the isotopic content of those elements through a spectrometric method, spiking, isotopic dilution, etc.

IV. CONCLUSION

The depletion capabilities in SCALE 6 and the newly available ENDF/B-VII libraries are validated against isotopic measurement data for five PWR spent fuel samples with burnup between 27 and 54 GWd/MTU. The results of the study show that, while the major actinides are calculated with similar accuracy with the old ENDF/B-V libraries and new ENDF/B-VII libraries, the new libraries provide significantly better estimates, compared to measurement data, for several fission product nuclides.

REFERENCES

1. *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, ORNL/TM-2005/39, Version 6, Vols. I–III, January 2009. (Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-750).
2. M. D. DeHart, "Lattice Physics Capabilities of the SCALE Code System Using TRITON", CD Proceedings, ANS Topical Meeting on Reactor Physics PHYSOR 2006 (2006).
3. M. L. Williams and G. Ilas**,** "ENDF/B-VII Nuclear Data Libraries for SCALE 6.0", CD Proceedings, Advances in Nuclear Fuel Management IV (2009).
4. R. J. Guenther et. al, “Characterization of Spent Fuel Approved Testing Material ATM-104”, PNL-5109-104, Pacific Northwest Laboratory (1991).
5. M. Lippens et. al., "Source Term Assessment: The ARIANE Programme", 8th International Conference on Environment Management (2001).
6. P. Baeten, et. al. “The REBUS Experimental Programme for Burn-Up Credit”, Proceedings of the 7th International Conference on Nuclear Criticality Safety (2003).
7. SFCOMPO - Spent Fuel Isotopic Composition Database, www.nea.fr/sfcompo/, Nuclear Energy Agency Data Bank.