OVERVIEW OF SPENT NUCLEAR FUEL INVENTORY RESULTS FOR THE ARIANE GU3 SAMPLE

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ABSTRACT

Accurate estimation of spent nuclear fuel (SNF) nuclide inventory is of great importance for safety aspects of the back-end of the nuclear fuel cycle. The Working Party on Nuclear Criticality Safety (WPNCS) of the OECD Nuclear Energy Agency (NEA) has agreed to support assessing the impact of nuclear data uncertainties on an SNF inventory. A PWR UO_2 post-irradiation-examination (PIE) fuel rod sample for which experimental data are available has been studied. Within this framework, Sub-Group 10 (SG10) focuses on the analysis of the code-to-code and nuclear data library-to-library impacts on the Calculation-over-Experiment (C/E-1) values for nuclides mainly important in burnup credit methodologies. This step will serve as a basis for the subsequent analysis of nuclear data uncertainties impact on C/E-1 biases and their uncertainties. Around 30 participants from 20 institutions and a large variety of calculation codes (15) and nuclear data libraries (8) are included. The exercise, the calculation methods, and the results are summarized and discussed in this paper.

KEYWORDS

OECD/WPNCS, spent nuclear fuel, depletion calculation, nuclear data uncertainty

1. INTRODUCTION

Estimation of an SNF nuclide inventory is of great importance in the context of fuel cycle safety assessment, for fuel transport, storage and disposal applications, including burnup credit (BUC) applications. Therefore, it is crucial to accurately estimate the SNF inventory resulting from code prediction. International efforts have been outlined for decades at the OECD NEA and beyond within dedicated Expert Groups, via meetings and benchmarks [1]-[7]. These efforts have contributed to increasing the confidence in code estimations (methods, assumptions, biases, and uncertainties). Some issues remain to be discussed for a better understanding of total biases and uncertainties, such as contributions from nuclear data uncertainties. The NEA WPNCS agreed to pursue the investigation of the impact of nuclear data uncertainties on an SNF inventory. This resulted in the endorsement of WPNCS dedicated subgroups in June 2018. To make the exercise valuable and innovative, it has been proposed to focus on an SNF realistic case where experimental data are available. These experimental data have a dual purpose: (1) to serve as a basis for the initial Calculation-over-Experiment comparison of the participants' fuel inventory calculations and (2) to analyse and assess the contribution of the nuclear data uncertainties to the total uncertainties. As a result, this work will provide new elements to improve biases and bias uncertainties accuracy of criticality calculations considering SNF.
The former subgroup SG7 focused on selecting an experimental dataset in the SFCOMPO database and on providing specifications of a benchmark model [8] (the measurand) to minimize the potential modelling discrepancies among the participants. The current subgroup SG10 focuses on the SNF calculation results and on analysis of the code-to-code and C/E-1 biases of mainly BUC nuclides for the selected fuel sample. These results will serve as a basis for a third subgroup, which will be proposed at a next WPNCS meeting in 2024.
This paper presents the current SG10 exercise and results. First, a brief description of the specified benchmark model is given. Secondly, the contributed results as well as the data and computer codes are detailed. Lastly, the code-to-code dispersion as well as the deviations of Calculation-over-Experiment (C/E-1) values are presented and discussed.

2. BENCHMARK SPECIFICATIONS

In the framework of SG7, a survey has been proposed to all the WPNCS members to select an SNF sample with fuel characteristics meeting the needs of the participants. The large majority of participants were interested in selecting a PWR UO_2 fuel with a specific burnup around 45 GWd/t (where the mass refers to initial actinides, i.e. unirradiated uranium). The ARIANE GU3 sample, selected from the SFCOMPO database [9], fulfilled these criteria. This sample has a ^{235}U initial mass enrichment of 4.1 % and an estimated burnup of 52.5 GWd/t. To minimize the potential modelling discrepancies among the participants, a detailed benchmark model has been proposed for this GU3 sample. The benchmark specifications were prepared by SG7 and are provided in a dedicated report [8]. A summary of these specifications is provided in this section.

2.1. General characteristics

Table I. General data of the benchmark model.

Lattice type	15 × 15
Fuel	UO_2
$^{235}U/U$	4.1%
Moderator	borated water

The general characteristics of the GU3 sample are recalled in Table I. The fuel rod containing the GU3 sample has been irradiated in the Gösgen PWR in Switzerland during three reactor cycles. The particularity of this sample is the changed position of the sample fuel rod between cycles. It was extracted from one fuel assembly FA 16-01 after two cycles (cycles 16 and 17) of irradiation and inserted

into the irradiated fuel assembly FA 17-01 for the third cycle (cycle 18). A cooling period of around three years before measurement is imposed. A detailed sample irradiation history was provided within SG7.

2.2. Major assumptions and simplifications

As underlined in the previous section, a benchmark model was defined to reduce arbitrary user effects when comparing code predictions. A detailed description of the experimental data [10] is available, but the benchmark model is simplified. The specifications therefore converged to consider:

- Cycle averaged boron concentration.
- Simplified and constant temperatures (the cladding and coolant temperature is 600 K while the average fuel temperatures are 1200 K, 1000 K and 900 K for cycle 16, 17 and 18 respectively).
- Discretization of specific power history by steps of 2 GWd/t.
- Reflective boundary condition approximation (the surrounding assemblies are not included in the model).
- The change of the sample rod position from FA 16-01 to FA 17-01 at the beginning of cycle 18.
- For FA 17-01 at beginning of cycle 18, an average fuel assembly inventory for 157 nuclides, at a burnup of 20 GWd/t, is provided for the fuel rods, excluding the sample fuel rod. This inventory does not account for the burnup gradient in the assembly.
- Burnup re-normalization to one or more measured nuclide concentrations (participants were free to use the method of their choice).

2.3. Experimental results

Measurements of the GU3 sample were performed by two of the laboratories involved in the ARIANE program, ITU in Germany and SCK CEN in Belgium. A set of recommended values was established by consensus of experts participating in the program, based on a detailed cross-check analysis of the two sets of measurements. The cross-check was based on a comparison of the 95 % confidence intervals associated with the measured values. In the framework of SG10, only the set of recommended values [8] is considered.

2.4. Requested results

Among the measured nuclides in the GU3 sample, SG10 has decided to focus on a restricted list of nuclides concerning originally only some BUC nuclides, i.e. within the scope of criticality safety assessment. Next to BUC nuclides, neodymium isotopes (as burnup tracers) were reported to ease the interpretation of individual participant results, as well as some other nuclides of common interest for participants (mainly related to decay heat and radiation emission calculations).The list of requested nuclides is summarized in Table II.

Table II. **Requested list of nuclide inventory results.**

BUC nuclides	^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{237}Np, ^{241}Am ^{155}Gd, ^{103}Rh, ^{149}Sm, ^{143}Nd, ^{133}Cs, ^{151}Sm, ^{152}Sm, ^{99}Tc, ^{153}Eu, ^{95}Mo, ^{150}Sm, ^{109}Ag, ^{147}Sm, ^{101}Ru
Burnup tracers	^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd
Other nuclides of interest	^{234}U, ^{236}U, ^{243}Am, ^{137}Cs, ^{242}Cm, ^{244}Cm, ^{147}Pm, ^{90}Sr

The usual depletion calculation results for physical parameters have been requested from the participants such as: k_∞ for assembly and nuclide mass as a function of irradiation and decay time. Calculation-over-Experiment values were also requested.

3. PARTICIPANTS, DATA AND COMPUTER CODES

3.1. Overview of participation, codes and nuclear data libraries

Table III summarizes the participants' information, including the computer codes and the nuclear data libraries used. SG10 accounts for 29 participations from 20 institutions and 12 countries, highlighting the high level of interest in this exercise. The exceptional level of contributions provides a large display of involved calculation codes and nuclear data libraries. Almost all the latest nuclear data evaluations releases are represented. A balance is found between the use of Monte Carlo (grey boxes) and deterministic codes (white boxes), illustrating the rising trend of the use of Monte Carlo codes for depletion calculations.

Table III. List of participants, computer codes and nuclear data libraries.

ID.	First name	Family name	Institution	Country	Depletion code	Nuclear data library
1	Coralie	Carmouze	CEA	France	APOLLO2.8/ DARWIN2.3	JEFF-3.1.1 281G; ENDFB-VII.1 281G
2	Raphaëlle	Ichou	IRSN	France	VESTA2.2	ENDF/B-VII.1
3	Yohannes Lydie	Molla Giot	SUBATECH	France	SERPENT2.1.32; OPENMC	JEFF-3.2, -3.3; ENDFB-VII.1, -VIII.0
4	Volker / Fabian	Hannstein / Sommer	GRS GmbH	Germany	MOTIVE	ENDFB-VII.1, -VIII.0
5	Gašper	Žerovnik	JSI	Slovenia	SERPENT2.1.31	ENDF/B-VII.1
6	Dimitri	Rochman	PSI	Switzerland	CASMO5	ENDF/B-VII.1 586G
7	Kevin	Govers	FANC	Belgium	SCALE-6.2.3 (TRITON-KENO-Va)	ENDFB-VII.1 252G
8	Nicolas	Slosse	Tractebel	Belgium	WIMS	JEFF-3.1.2 172G
9	Pablo Federico	Romojaro Grimaldi	SCK-CEN	Belgium	ALEPH2, SERPENT2.2.0	ENDF/B-VII.1
10	Dennis	Mennerdahl	EMS	Sweden	SCALE-6.2.4 (POLARIS)	ENDF/B-VII.1 252G
11	Pauli	Juutilainen	VTT	Finland	SERPENT2.1.32	JEFF-3.2; ENDF/B-VII.1; JENDL-4.0
12	James	Lam	Rolls-Royce	UK	MONK DV1	JEFF-3.1 CE, 172G
13	Dirk	Schulze Grachtrup	BASE	Germany	SCALE 6.2.4 (TRITON-NEWT)**;** (TRITON-KENO-VI)	ENDF/B-VII.1 252G **;** ENDF/B-VII.1; JEFF-3.1.1,-3.3
14	Tomoaki	Watanabe	JAEA	Japan	SWAT4.0	JENDL-4.0
15	Germina Ugur	Ilas Mertyurek	ORNL	USA	SCALE-6.2.4, -6.3.0 (TRITON-NEWT)**;** SCALE-6.3.0 (POLARIS)	ENDF/B-VII.1 252G ; ENDF/B-VIII.0 252G
16	Ahmed	Shama	Nagra	Switzerland	SCALE-6.3.0 (POLARIS)	ENDF/B-VII.1 56G
17	Simon Paul	Richards Smith	Jacobs	UK	MONK-11A	JEFF-3.3
18	Axel	Hoefer	Framatome Gmbh	Germany	SCALE-6.2.3 (TRITON-NEWT)	ENDF/B-VII.1 252G
19	Roberto Pedro Francisco	García-Baonza Martínez-Moreno Álvarez-Velarde	CIEMAT	Spain	EVOLCODE	JEFF-3.3,-4T1; ENDF/B-VIII.0
20	Sven Maksym	Tittelbach Chernykh	WTI GmbH	Germany	SCALE-6.2.3 (TRITON-NEWT)	ENDF/B-VII.0 238G; ENDF/B-VII.1 252G

3.2. Deviation from the benchmark specifications, including burnup calibration

As some participants may have faced constraints related to their codes with respect to using the exact specifications, some deviations from the specifications were reported. This information is crucial for analyzing the results and will be highlighted in the SG10 report planned for the end-of-year 2023. Most of the noticed deviations appear to have only a slight impact on the results except for the significant change on the given sample power history. The participants were invited, but not requested, to calibrate the burnup using burnup tracers. Eleven participants performed sample burnup calibration, but nine did not. It is to note that such re-normalization makes a consistent comparison between participant results more complicated.

4. SIMULATION RESULTS AND INTERPRETATION

4.1. Assembly infinite neutron multiplication factor (k_∞) as a function of time

The evolution of k_∞ and the related reactivity effects are important and directly used in baseline safety studies. The k_∞ is defined at the assembly level. The computed values are displayed in Figure 1. The first step of the analysis consisted in studying the k_∞ results as a function of time to first highlight possible issues in the modelling and to provide some feedback to the participants.
Given the large number of participants, codes and nuclear data libraries involved, a general agreement on the time dependence of the k_∞ is observed. Nevertheless, a large dispersion is noticed between the participants' k_∞ calculation results. The mean value is 1.31076 at beginning of life (BoL), with a standard deviation of around 0.3 % and a maximal deviation of 1.6 %. These deviations cannot be imputed only to the involved variety of codes and nuclear data. Strong discrepancies were initially observed for results of different participants that used the same code and the same nuclear data library. This is the case of participants #13, #15, #18 and #20, that all used SCALE-6.2.*i* (*i*=3 or 4)/ TRITON-NEWT and the ENDF/B-VII.1 library. A deviation up to 0.6 % is observed at BoL for these cases, indicating potential user effects. It was later identified that most of this k_∞ deviation is due to using material or geometry input data that were different than the provided benchmark specifications. For example, participant #18 reported the use of a moderator density greater than 3% and boron concentration greater than 5 % compared to the benchmark data. Another similar and interesting example can be given comparing participants #3, #5, #9 and #11 results, obtained using SERPENT2 and the ENDF/B-VII.1 library. For these cases, a deviation up to 0.2 % is observed at BoL due to the use of the Doppler Broadening Rejection Correction Method by one of the participants.

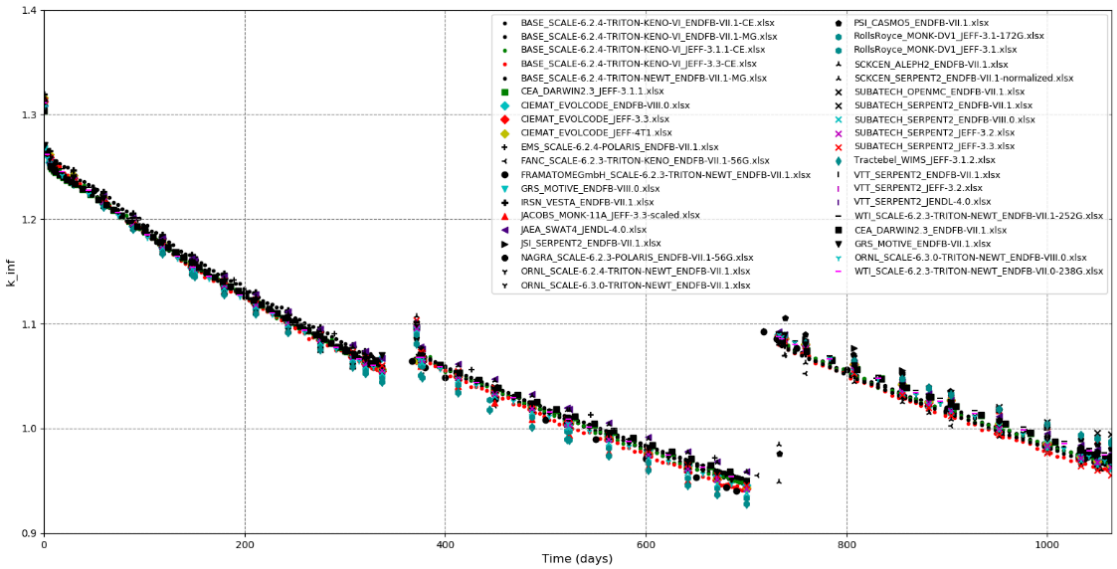

Figure 1. Assembly k_∞ as a function of time.

4.2. Nuclide inventory

4.2.1 Nuclide concentration evolution with burnup

The second step of the analysis consisted in studying the evolution of concentrations of the selected nuclides as a function of the sample burnup to investigate possible issues in the modelling. The ^{148}Nd concentration evolution with burnup is displayed in Figure 2.

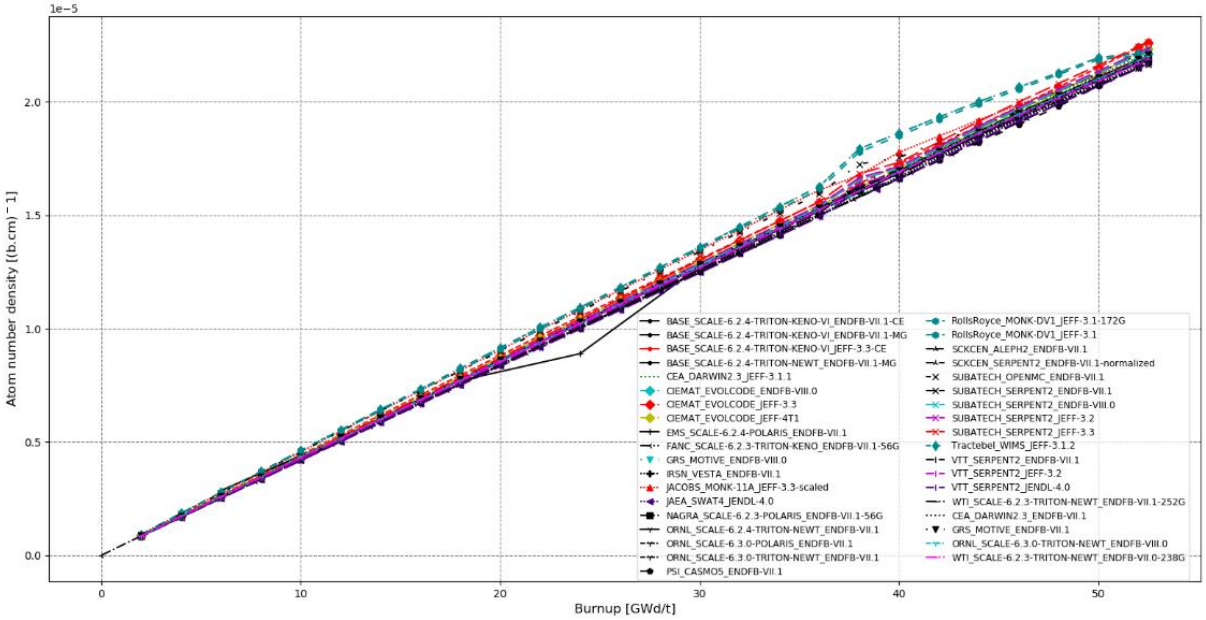

Figure 2. ^{148}Nd atom number density as a function of burnup.

Checking the evolution of the ^{148}Nd, commonly used as a burnup tracer since it evolves in an almost linear way with burnup, could highlight issues in modelling or use of code options. Specific issues were due to user's approach in modelling the specific power for codes that do not have shuffling capabilities to easily account for the transfer of the GU3 sample from one assembly to another in cycle 18. This study supported work of coherence on the code options to be used. These were discussed within the SG10 and will be detailed in the final report. Overall, a generally good agreement is observed, given the large number of codes, nuclear data libraries, and users involved. A quasi-linear behaviour with burnup is obtained for ^{148}Nd for all codes, even if some non-linearity exceptions remain to be investigated within SG10.

4.2.2 Comparison with experimental results

In the following subsections, the calculated nuclide mass concentrations were compared against the benchmark values (experimental measurement) using a C/E-1 (%) format, where C is the calculated value (per code) and E is the experimental value, for the list of selected nuclides. A correct decay time is considered when evaluating the nuclides, as stated in the benchmark specifications.
As previously explained, learning from the previous SNF exercises, the experimental values serve as a basis for the comparison of the participants results and not necessarily for best estimate calculations. They help to identify and investigate modelling issues by highlighting results obtained and revealing significant deviations from the mean results. First, the mean, max., and min. C/E-1 values are given for all the studied nuclides and then, the focus is on the ^{235}U results as good candidates to highlight the major encountered issues. A comprehensive evaluation for all nuclides will be detailed in the SG10 report. It is to note that the experimental uncertainties are not displayed in the following plots as they are not significant compared to the C/E-1 bias between participants' results.

- All nuclide results

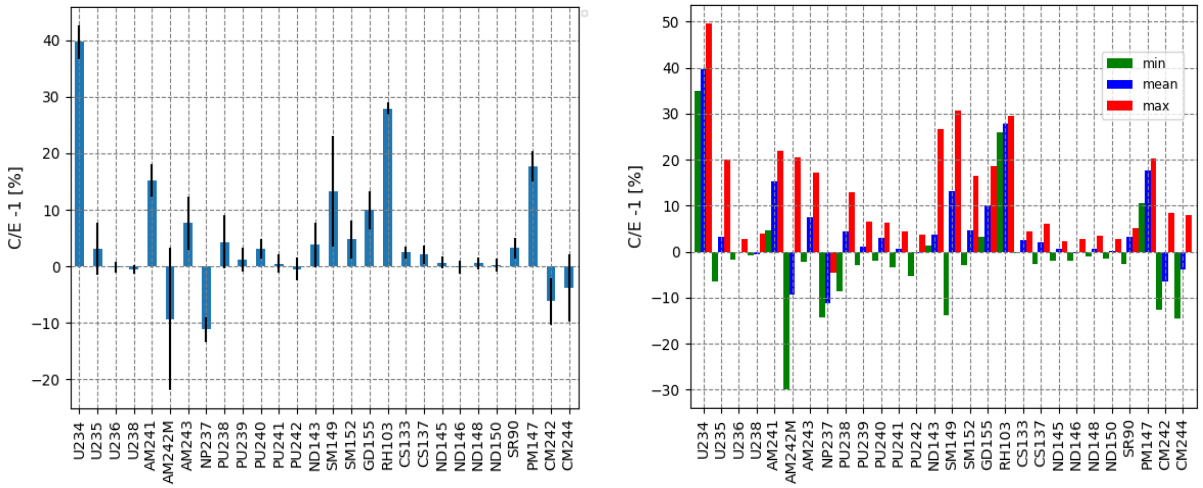

Figure 3. Mean C/E-1 (%) values for all cases and all nuclides, with the standard deviation (left) and with max. and min. divergences (right).

The mean C/E-1 values for each nuclide, among all results, are displayed in Figure 3 together with the standard deviation between results and the maximal positive and negative divergences (max. and min. values). If one focuses on the Calculation-over-Experiment comparison, an overall acceptable agreement is observed, except for a few nuclides. An exception is ^{234}U which is overestimated by ~40%, likely linked with the initial isotopic fraction which appears to be much higher than usual for uranium that is not reprocessed. The C/E-1 values are larger than 10 % for ^{241}Am, ^{237}Np, ^{149}Sm, ^{103}Rh and ^{147}Pm and not covered by the experimental uncertainty nor by the standard deviation between the participants' results. Focusing on the code-to-code comparison, it can be first noticed that large deviations impact the results of the participants for a significant number of nuclides, when comparing the extreme results. Nevertheless, these deviations are not reflected in the mean values, which highlight the fact that the extreme values remain exceptions. The obtained mean values for most nuclides are smaller than 4 %. Small differences are expected for nuclides used as burnup tracers, even though a max-to-min deviation about 6 % is noticed for ^{148}Nd for example. This remains to be investigated. Moreover, a standard deviation larger than 4 % is observed for ^{235}U, ^{242m}Am, ^{243}Am, ^{238}Pu, ^{149}Sm, and Cm isotopes. As mentioned before, the results for these nuclides appear to be directly connected to the observed user effect. Moreover, some of those nuclides are known to be very sensitive to the burnup or to the power history. The example of the use of the SCALE-6.2.*i* TRITON-NEWT code associated with the ENDF/B-VII.1 library, pointed out in subsection 4.1, helps here again to point out the significant impact of the user effect.

Figure 4 shows the C/E-1 results, obtained for all nuclides, with SCALE 6.3.0 and SCALE-6.2.*i* / TRITON-NEWT code system and 252G ENDF/B-VII.1 libraries. While results obtained by one participant varying only the SCALE code version (green and red results) show no significant discrepancy, not the same conclusion would apply to the othe participants. When one compares the results reported by four participants (in blue, orange, green and purple), significant discrepancies are observed for many nuclides. This is the case for ^{235}U C/E-1 which varies by a factor 2.7 between participants #18 and #20, ^{242m}Am which varies by a factor 4 between participants #18 and #20, and ^{155}Gd which varies by a factor 2.3 between participants #13 and #18. These results indicate very important user effects. The effect of deviations between what the users applied as input values in their models and the provided benchmark specifications was identified as impactful. Moreover, the code usage, especially for depletion calculations options, is clearly impactful here. It was identified that the users assumptions for modelling the provided irradiation history and the differences in the depletion calculation options can lead to huge deviations between users' results. As it has been the object of working groups in the

past for deterministic calculations, this issue remains to be discussed regarding Monte Carlo depletion calculations. Similar conclusions, whereas not detailed here, are obtained from the SERPENT2 Monte Carlo depletion calculation results with the same library.

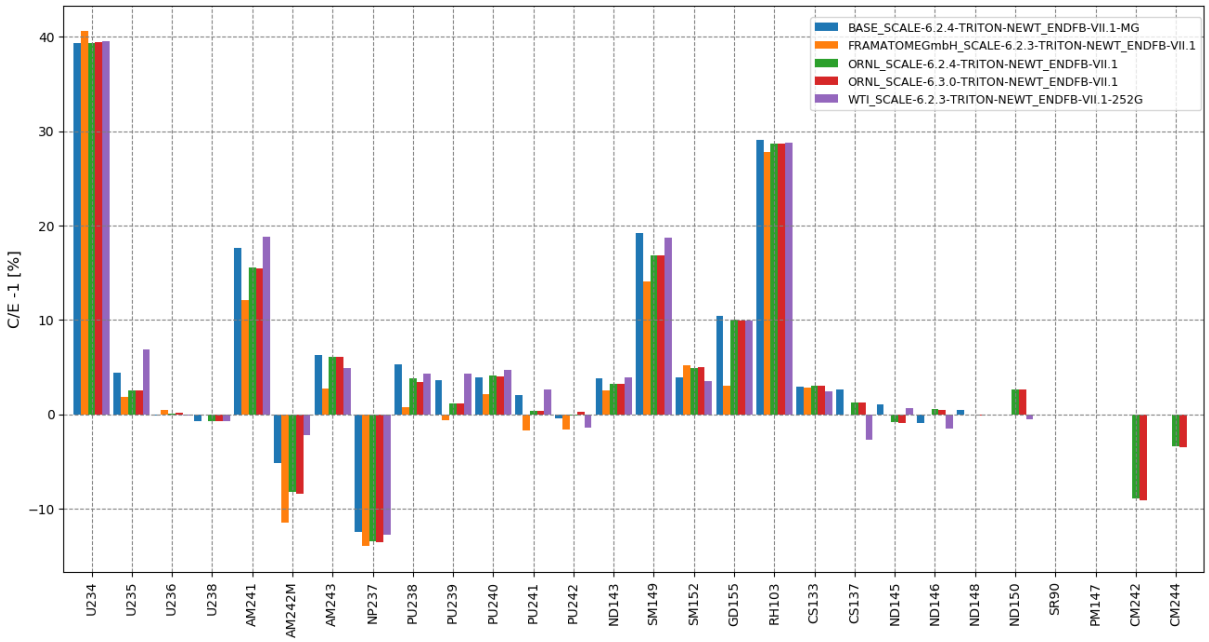

Figure 4. C/E-1(%) values for TRITON/NEWT code with ENDF/B-VII.1 libraries.

- ^{235}U results

C/E-1 trends on SNF ^{235}U estimation are well studied and documented by the scientific community. It is also a good candidate to present the major issues on this exercise.

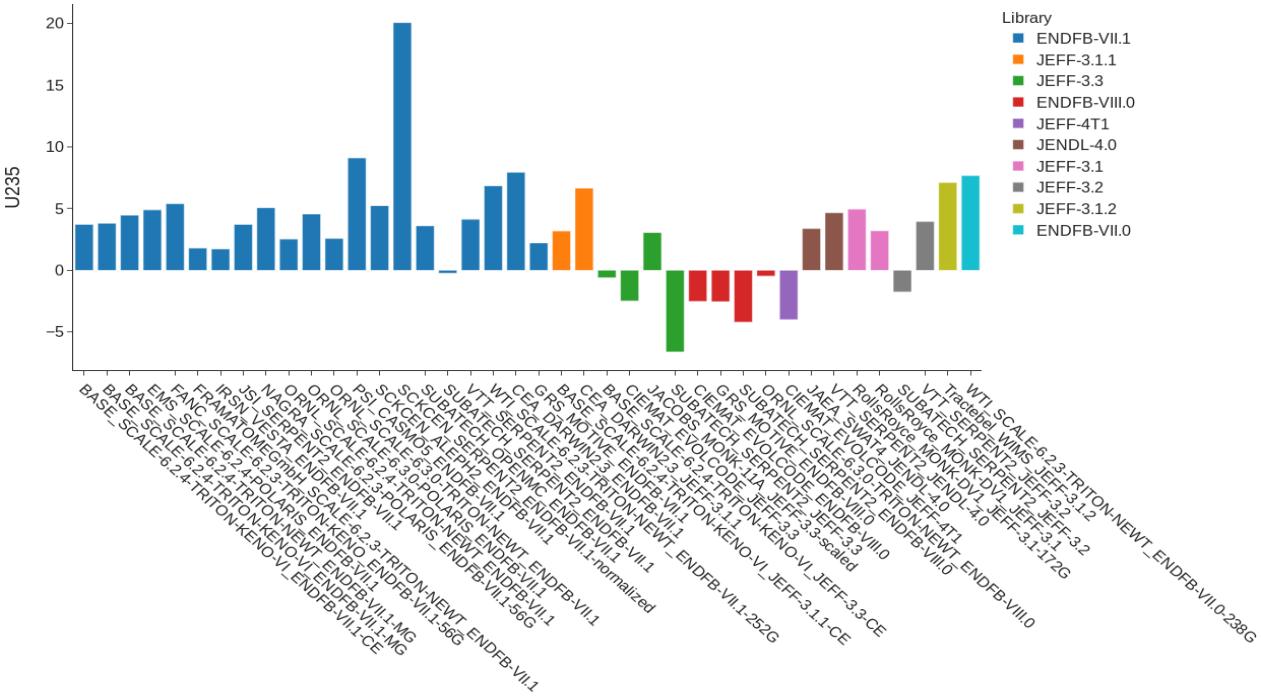

Figure 5. ^{235}U C/E-1(%) values for all cases.

Figure 5 shows a comparison of code predictions to experimental measurements for ^{235}U, considering all the participants results, ordered as a function of the nuclear data library. Depending on the result (obtained for a given code and a given nuclear data library), ^{235}U is predicted generally within 5 % of the experimental value, with a few exceptions. Some variation is nonetheless observed among the codes. An average standard deviation of 3.1 % is obtained for ^{235}U among all results. Because the ENDF/B-VII.1 library (in blue) is the most used library for this exercise, an inter-code comparison at identical nuclear data is possible[1]. Nevertheless, a rather high dispersion among the calculations using ENDF/B-VII.1 is observed, although some common trends can be noted. A common overestimation of the experimental values is obtained for all the results with ENDF/B-VII.1. An opposite behaviour is obtained for the results with ENDF/B-VIII.0 (in red), where all the four calculation results underestimate the experimental value. This is consistent with the modifications of ^{235}U(n,f) [≈ +3 %], (n,γ) [≈ -7 %] cross sections and fission neutron multiplicity (nu-bar) [≈ -2.4 %] between the two ENDF/B- versions, known to lead to a slight underestimation of the k_{eff} in small critical systems. Similar trends are observed among the JEFF-3.1.1 versus JEFF-3.3 and -4T0 calculation results. Here again these results are consistent with the ^{235}U cross section revisions done between the 3 JEFF versions.

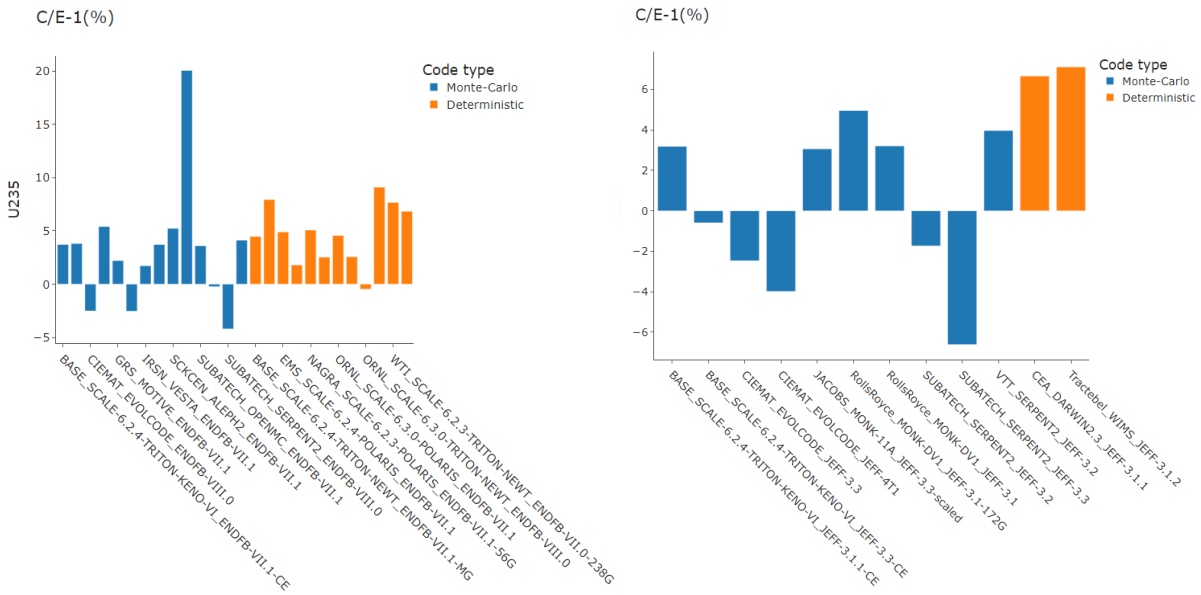

Figure 6. ^{235}U C/E-1 values (%) for ENDF/B (left) and JEFF (right) libraries.

Figure **6** shows the ^{235}U C/E-1 results, obtained for all the ENDF/B libraries (ENDF/B-VII.0, ENDF/B-VII.1 and ENDF/B-VIII.0) (left) and obtained for all the JEFF libraries (JEFF-3.1, JEFF-3.1.1, JEFF-3.1.2, JEFF-3.2 and JEFF-3.3) (right) by displaying them as a function of their code type, that is to say either Monte-Carlo (in blue) or deterministic (in orange). Except for the participant #9 – SERPENT2 results which show an ~20% overestimation of ^{235}U compared to the experimental value (due to an error in the post-processing that will be corrected), the results tend to agree within 5 % of the measurements or slightly more. However, the significant deviations observed cannot be imputed solely to the type of code (Monte Carlo vs deterministic) or to the nuclear data library. As previously explained, participants have calibrated the burnup based on various methods (see §3.2). Given that ^{235}U prediction at high burnup is significantly affected by burnup, part of the observed deviations come from the difference in the final burnup value used by the participants. This point will be further investigated within SG10.

[1] Some caution should be applied to this comparison as 1/ some codes use modified nuclear data libraries for their applications (case of CASMO5), and 2/ nuclear data processing can impact the library as well as the energy group structure.

5. CONCLUSIONS

To assess the impact of nuclear data uncertainties on an SNF inventory, a PWR UO_2 PIE sample with experimental data available, has been selected and detailed benchmark modelling specifications have been provided within the framework of the WPNCS SG7. The present study is performed under the WPNCS SG10, focusing on the analysis of the code-to-code dispersions and the Calculation-over-Experiment bias (C/E-1) values for nuclides mainly important in burnup credit methodologies. This step will serve as a basis for the subsequent analysis of nuclear data uncertainties impacts on the C/E-1 bias and uncertainties in calculated individual nuclide concentrations.
A wide participation of WPNCS members is achieved, with ~ 30 participants from 20 institutions and a large panel of calculation codes (15) and nuclear data libraries (8). It allows for a large scale and valuable comparison of the results. It has been possible to draw conclusions and provide a comparison between the participant results based on the calculation code, the calculation method, and the nuclear data library. An overall good agreement with the experimental values is observed for most participants and most nuclides. However, some dispersion between the results is also observed. Despite detailed specifications being provided to enable common implementation among codes rather than developing a best-estimate model, it was noted that sometimes large code-to-code dispersions in results remains. It was identified that these discrepancies are not exclusively related to differences in the nuclear data library or the code used. Unexpected discrepancies have been observed for k_∞ results at BoL, mainly due to code user effect or to differences between the input data applied and the benchmark specifications. This valuable work emphasizes the important code user effect at each stage of the depletion calculations. Valuable feedback is provided to the participants and will be expressly detailed within the SG10 report planned for the end-of-year 2023. Lastly, efforts remain to be made within the SG10 to outline code user and/or modelling issues for results that significantly deviated from the mean values. This point is crucial before moving to the second phase of the nuclear data uncertainties quantification, planned for the beginning of the year 2024.

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