Analysis for the ARIANE GU1 sample: Nuclide inventory and decay heat

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ARTICLE INFO

Article history:
Received 16 February 2021
Received in revised form 9 April 2021
Accepted 21 April 2021

Keywords:
Post irradiation examination
Nuclear data
Expanded uncertainties
PIE sample
GU1
decay heat

ABSTRACT

The present study provides a detailed analysis of the calculations of 50 isotopic compositions for the Post Irradiation Examination sample GU1 from the ARIANE program. Two types of approach are performed: a lattice (2 dimensional) and a full core (3 dimensional) calculations. In the case of lattice calculations, the effect of location is also studied. Different code versions (for CASMO5 and SNF) are used in order to contribute to the validation work for specific calculation schemes and to assess the impact of nuclear data libraries as well. The effects of nuclear data are also quantified for cross sections, spectra and fission yields, together with their partial contributions. It is concluded that the consideration of the effect of the mass balance, nuclear data uncertainties, as well as uncertainties from operational conditions and manufacturing tolerances help in obtaining consistency between calculated and measured isotopic concentrations.

This study is then complemented with decay heat calculations for the assembly of interest, consistent with the PIE study. It indicates that, without considering geometrical deformation due to the long storage time, nuclear data are still the main source of uncertainties. Expanded uncertainties (including biases, experimental and calculation uncertainties) are finally proposed for both isotopic compositions and decay heat.

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1. Introduction

Post Irradiation Examination (PIE) are of significant importance for the validation of transport and depletion code systems. Such validation allows assessing performances for the predictions of isotopic contents in used nuclear fuels (UNF, also called Spent Nuclear Fuel, or SNF), which is mandatory for many types of studies (see for instance Refs. Kashima et al., 2015; Gauld et al., 2019; Santamarina et al., 2009; Rochman et al., 2020; Ebiwonjumi et al., 2020; Sanfelice et al., 2013; Rizzo et al., 2018). As a consequence, many institutes worldwide are developing various capabilities to compare calculated and measured isotopic concentrations, in the form of simulation codes, models, measurements, and analysis.

The UNF studies performed at PSI are no exception and the validation of the prediction codes is obtained against proprietary data (the LWR-PROTEUS Phase II and MALIBU programs Yamamoto et al., 2012; Boulanger et al., 2004; Grimm et al., 2014; Pecchia et al., 2016; Rochman et al., 2018), as well as data from the open literature, such as from the ARIANE programme (Michel-Sendis et al., 2017; ARIANE, 2000). They are mainly conducted for samples irradiated in Swiss reactors, but are also part of larger international efforts, allowing detailed comparisons, analysis or trend studies among various nuclear data libraries, codes, PIE samples and participants. To only cite four current international open projects, the readers are referred to the following work:

- the European Horizon 2020 project EURAD (European Joint Programme on Radioactive Waste Management EU H2020, xxxx). More specifically, the Work Package 8 is dedicated to the “spent fuel characterization and its evolution until disposal”. Concerning sample simulations, this project includes the calculation of isotopic concentrations from a number of PIE samples, as well as decay heat calculations, with an estimation of uncertainties and biases.
- the Coordinated Research Project (CRP) on “Spent Fuel Characterization” from International Atomic Energy Agency (CRP, xxxx). At the time of the writing of this paper, this CRP is starting for a duration of three years, and will include so-called “performance modeling” (i.e. validation through experimental data).
- the “SKB blind test in prediction of decay heat”, led by Vattenfall and the Swedish Nuclear Fuel and Waste Management Company (SKB), and hosted at the Nuclear Energy Agency. The goal of this exercise is to evaluate the accuracy of simulations for decay heat compared to measured values, as well as to assess the importance of specific simulation details.
and naturally to the SFcompo database for measured spent nuclear fuel isotopic assay data (Ilas et al., 2020), coordinated by the Organisation for Economic Co-operation and Development (OECD)/Nuclear Energy Agency (NEA).

The present PIE study falls into the category of code system validation, more specifically in the case of the simulation codes used for the core reload licensing of Swiss reactors (Leray et al., 2017; Rochman et al., 2020), as well as UNF studies for the final repository as planned in Switzerland (Rochman et al., 2018; Ferroukhi et al., 2008; Rochman et al., 2018; Rochman et al., 2020; Rochman et al., 2020). The codes of interest consist in CASMO5, SIMULATE and SNF (Rhodes et al., 2006; DiGiovine et al., 1995; Rochman et al., 2020). The codes of interest consist in CASMO5, SIMULATE and SNF (Rhodes et al., 2006; DiGiovine et al., 1995; Simeonov and Wemple, 2017) from the Studsvik company and are applied to a validation set of 27 PIE samples from PROTEUS, MALIBU and ARIANE. In this paper, the specific case of the GU1 sample from the ARIANE programme is presented and the study includes a comparison between calculated and measured isotopic compositions, as well as the calculation of uncertainties for such concentrations, due to nuclear data and a number of technological parameters. As the decay heat mainly depends on the sample or assembly isotopic composition, decay heat calculations are also performed, using the same nominal parameters and variations as for the PIE analysis. These quantities, isotopic concentrations and decay heat (as a function of cooling time), are of prime importance for the specification of UNF and their safe handling regarding criticality and efficient cooling & storage.

In the following, the simulation tools are first presented (Section 2), a description of the GU1 sample is given in Section 3, followed by the results of this study. The main conclusions of this study are that (1) for this sample, both lattice (2D) and full core calculation (3D) provide equivalent results for the isotopic compositions, (2) the effect of adjacent rod relocation is not of primary importance, (3) the nuclear data and technological parameter uncertainties help in explaining apparent inconsistency between calculations and measurements, (4) that a limited number of cross sections and fission yields are of importance for the estimation of uncertainties due to nuclear data, and finally (5) that the calculated decay heat uncertainty mainly depends on nuclear data, themselves showing strong variations from one library to the other. The quantified results of this study are presented in Tables A.3, A.4, A.5, A.6, A.7.

2. Simulation tools and calculation schemes

For the present calculation of the GU1 isotopic contents, two different approaches are followed, as described below: two- and three-dimensional calculations (also referred to as 2D and 3D).

2.1. Definition of uncertainty

In the following, the term “uncertainty” is used to indicate the spread of a calculated quantity. This term has often different meanings in publications, which can lead to confusion. For the present study, the uncertainty represents one standard deviation of the probability density function of a specific calculated quantity. There is no assumption that such distribution is Normal, therefore the uncertainty can cover more (or less) than 68% of the total probability.

Additionally, the calculated uncertainties are obtained for specific input variations. Taking different assumptions for the relevant inputs would necessarily lead to different uncertainties for calculated outputs. As presented in the following, variations of nuclear data are relatively well framed by covariance matrices from nuclear data libraries. For other quantities, such as the engineering parameters, specific assumptions are taken, undeniably affected the outcome. Thus, we would like to indicate here that the proposed uncertainties do not correspond to “true” ones. They are bounded by our assumptions on the input variations, the propagation method, and potentially the simulation codes.

2.2. Two dimensional calculations

The first type of calculations is based on two-dimensional calculations using the deterministic transport code CASMO5. The characteristics of the assembly containing GU1 are modeled for this code (geometry, initial concentration, irradiation history, cooling periods), using consistent description with the 3D analysis. CASMO5 also includes a simplified depletion module, called SNFlight, used here for the calculations of the isotopic concentrations after cooling times corresponding to the measurement dates (provided in the ARIANE report). The resonance treatment of CASMO-5 relies on the standard equivalence theory (Weinberg and Wigner, 1958). The self-shielding of the resonance absorption is corrected for each pin depending on its position in the lattice through the use of Dancoff factors. When a fuel rod is moved through the lattice, its Dancoff factor is re-calculated ensuring that no calculation bias is introduced (in the limit of the inherent biases associated with the deterministic treatment of resonance self-shielding). For the calculation of the concentrations of fission products, CASMO5 is using either cumulative of independent fission yields. This is of importance for nuclear data evaluators, in case of inconsistent evaluated cumulative and independent fission yields and their uncertainties. For the following fission products, cumulative yields are used: 90Sr, 106Ru, 144Ce, 145Nd and 152, 154Sm.

For other ones, the independent yields are calculated.

As part of the validation study, three different versions of CASMO5 are tested, potentially leading to different isotopic concentration values. The three versions are as follows:

1. CASMO5 version 2.03.00, with the nuclear data library based on the US ENDF/B-VII.1 original library (Chadwick, 2011), called version “e7r1.201.586.bin”.
2. CASMO5 version 2.13.00, also with the same nuclear data library version (“e7r1.201.586.bin”), and
3. CASMO5 version 2.13.00, with an updated processed library version, called “e7r1.202.586.bin”, and also based on ENDF/B-VII.1.

This numbering (1) to (3) will be used in the rest of the study. The difference between case (2) and (3) is solely the change of library, from the “201” to the “202” version. Some details of these processed libraries are presented in Hykes et al. (2014) and are succinctly repeated here: the 202 library contains a larger number of isotopes based on the ENDF/B-VII.1 and TENDL-2012 (Koning et al., 2019) libraries, 1095 isotopes with full or absorption-only cross section models (526 more than its predecessor 201 library), 119 actinides and 491 fission products in the depletion calculation (the 201 library used 60 and 259, respectively). Such changes can potentially modify the calculated isotopic concentrations, as indicated in Section 4.1.

As presented in the following, the input description is the same for the three types of calculation: it is directly derived from the PSI CMSYS database, which includes CASMO5/SIMULATE cycle calculation for the segment of the assembly of interest and validated full core models (in the following, the term CMSYS will be equivalent to CASMO5/SIMULATE). By using an in-house tool called BOHR, information as a function of the irradiation and inter-cycle time is extracted (e.g. moderator and fuel temperatures, boron concen-
tration, power density, segment burnup, see Rochman et al., 2018; Ferroukhi et al., 2008 for more details) and converted into a CASMO5 input file. This way, similar input quantities (and cross sections) are used in the lattice and core calculation, with the limitation of neglecting the effect of the surrounding assemblies in the lattice case (see Rochman et al., 2018 for another application of BOHR). Lattice calculations can require an adjustment of the sample burnup, as the absolute sample power is often approximately guessed. Such adjustment is often applied based on the $^{148}$Nd concentration, considered as a burnup indicator. In the present case, as the sample characteristics are derived from the full core calculations, no burnup adjustment was necessary (calculated and measured $^{148}$Nd concentrations were judged to be in good agreement). This is not automatically the case, even when using the BOHR approach, as the core simulations are performed within a number of approximations (for instance the number of vertical segments), usually necessitating small burnup corrections.

As presented later, the relocation of specific rods surrounding the GU1 sample will also be performed, based on the code version (1).

2.3. Three dimensional calculations

The second type of calculations considers the assembly irradiation within the different reactor cycles and the full core characteristics. A similar approach was already followed in Rochman et al. (2020) for the U1 sample from the PROTEUS program, as well as in Zuwat et al. (2012). In this case, as the GU1 sample was irradiated in the Gösgen reactor in Switzerland, all the cycle information is available at PSI in the framework of the reload licensing procedure. Therefore, the full three-dimensional information is available (core loading, cycle length, core power) and the SIMULATE core simulator can be used to estimate various power profiles and their variations over time. The validated models for CASMO5 and SIMULATE are stored in the CMSYS database. SIMULATE does not by itself calculate isotopic compositions for a specific rod segment, but it can be used in combination with the SNF program. In such a case, SNF takes advantage of the various macroscopic cross sections and other factors from CASMO5, the irradiation conditions from SIMULATE and provides a number of isotopic concentrations as a function of cooling time for a specific rod segment. As in the case of the two-dimensional simulations, a few code versions are used, grouped in three different calculation schemes:

1. [4]: CASMO5 version 2.03.00, with e7r1.201.586.bin, SIMULATE3 version 6.07.17_MOX_4 and SNF version 1.6.4.
2. [5]: CASMO5 version 2.13.00, with e7r1.201.586.bin, SIMULATE3 version 6.07.17_MOX_4 and SNF version 1.6.4, and
3. [6]: CASMO5 version 2.13.00, with e7r1.202.586.bin, SIMULATE-5 version 1.16.00 and SNF version 1.07.

One of the main advantages of SNF 1.07 compared to version 1.6.4 (which is an anterior version) is that it provides a larger number of isotopic compositions, covering the measured isotopes in the case of GU1. Therefore in the following, a limited number of measured compositions will be compared with SNF 1.6.4, whereas comparisons for all isotopes will be obtained with version 1.07.

2.4. Uncertainty propagation: effect of input variations

In addition to the nominal calculations, the effect of the variation of various input parameters is also considered in this study. It includes nuclear data, as well as operating and engineering parameters. The method to propagate uncertainties from various inputs is based on the Monte Carlo randomization of values, followed by CASMO5 calculations. It is a straightforward method, given specific assumptions for the variations of input variables. As a single CASMO5 segment calculation takes less than one hour on a single CPU, such uncertainty propagation is feasible on a small-size cluster.

2.4.1. Nuclear data

The study of the effect of nuclear data is performed in both the two- and three-dimensional simulations. In the 2D case, the CASMO5 version 2.03.00 is used with the in–house tool SHARK-X (Wieselquist et al., 2013; Leray et al., 2016). Similarly in the 3D case, only the versions presented in item (4) of the previous section are used. The uncertainty propagation method is similar to previous PSI studies, and is based on the perturbation of group cross sections and the repetition of CASMO5 calculations, each time with perturbed nuclear data (cross sections, spectra and neutron emission) using covariance matrices (uncertainties and correlations) from the latest release of the different libraries, namely ENDF/B–VIII.0 (Brown et al., 2018), JEFF-3.3 (Plompen et al., 2020) and JENDL-4.0 (Shibata, 2011). All actinides and important fission products are considered, including all relevant reactions (capture, fission, (n,2n), elastic and inelastic). In the case of fission yields, the same approach is followed, also using uncertainties from the ENDF/B–VIII.0 library. As correlations are not provided for fission yields, an in–house approach is applied, as presented in (Leray et al., 2016; Leray et al., 2014). Results will be presented in the next section and in the tables of the appendix.

The same approach for the three-dimensional calculations is applied here: for each perturbed nuclear data, the full calculation scheme based on the CMSYS/SNF database is performed. In order to be consistent with the two-dimensional case, the perturbation factors are used. For a given perturbed set of nuclear data, the 3D calculation is more computer intensive: if a single CASMO5 calculation (for one segment) is required in a 2D simulation for one perturbed nuclear data set, a total of 19 different CASMO5 calculations are needed, in order to cover all the different assembly segments used in the full core calculations for all cycles. This virtually multiply the calculation time by about 20, as SIMULATE3 and SNF calculations have relatively short running times. In such a case, parallel calculations are performed, as each CASMO5 segment calculation is independent of each other.

If this method can be applied to any of the above combination of codes, the requirement for the calculation time is not the same between SIMULATE3 and SIMULATE-5. In the case of SIMULATE-5, the necessary CASMO5 calculations are significantly longer than for SIMULATE3. Therefore, it was decided to perform the 3D uncertainty propagation only in the case of SIMULATE3, and with CASMO5 version 2.03.00, thus allowing an easier comparison with the 2D case. Results will be presented in Section 4.2.

2.4.2. Operating conditions

A number of operating conditions can be considered as not perfectly known in the present CASMO5 simulation. In the following, parameters for operating conditions include the fuel and moderator temperatures (and the coolant density), called TFU and TMO, respectively, boron concentration (called BOR), and the depletion steps of the irradiation history (called DEP). In order to calculate the impact of the variation of these parameters, they are separately (and independently) varied a thousand times, following uniform distributions with a 2% standard deviation for TFU, TMO and BOR, as in (Macian et al., 2007). The burnup steps, representing the irradiation history, have a large impact on isotopic concentrations, as well as on the sample burnup. A variation of 0.3% is applied in order to follow the experimental uncertainty of $^{148}$Nd (without the effect of the mass balance).
For the variation of one of these parameters, four random numbers are generated prior to a simulation, one for each cycle. A unique variation of the parameter is then used for the complete cycle (for instance increasing TFU by 1% at all burnup steps). Therefore all variations are correlated within a cycle, but they are different from a cycle to another. This method is applied for these four parameters and only for the 2D simulations with CASMO5 version 2.03.00. Results for the changes (or uncertainties) on isotopic concentrations due to the operating conditions can be found in Tables A.3 and A.4.

### 2.4.3. Manufacturing tolerances

Similar to the case for the operating conditions, a number of quantities related to the manufacturing of the assemblies can be varied. Such tolerances are often not from public domain and depend on fuel and assembly manufacturing companies. The following quantities and variations are therefore deduced from our specific experience, and shall not be generalized: taking into account uniform distributions, pin and guide tube radius (called RAD, standard deviation of 0.5%), pin pitch (called PITCH, standard deviation of 0.5%), fuel pin and guide tube position shifts in the X and Y positions (called X-Y, with standard deviation of 0.5 mm for the fuel rod and 0.1 mm for the guide tube), and the fuel density and $^{235}$U enrichment (called DEN + ENR, with standard deviations of 1.0 and 0.2%, respectively).

In the case of the RAD parameter, as a fuel pincell is defined with three radii (including fuel, gap and cladding), three random numbers are generated, using a uniform distribution with a standard deviation of 0.5% (for guide tubes, the same standard deviation is applied). This is performed once for the assembly, covering the four irradiation cycles. Due to the limitation of the number of different pincell which can be defined in a single CASMO5 input, radii variations are applied to 32 pincell around (and including) the rod containing the sample. Examples for the variations of RAD and X-Y are presented in Fig. 1. The uncertainties are increased for these figures, to highlight the effects. As observed, each fuel pin and guide tubes are randomly affected by the variations of their manufacturing parameters. The resulting isotopic uncertainties will be presented in the following.

Note that a number of variations are not considered in this work: bowing effect (necessitating the simulation of more than one assembly), and the burnup-induced technological changes (X-Y variations as a function of burnup, as well as radius variation during the irradiation). As for the operating conditions, uncertainties due to manufacturing tolerances are presented in Tables A.3 and A.4.

### 3. The GU1 sample

The GU1 sample is one of the samples from the ARIANE program. ARIANE consisted in the irradiation and analysis of various samples from the Dodewaard, Beznau and Gösgen reactors (ARIANE, 2000). This particular sample was irradiated in Gösgen over four consecutive cycles and stayed in the same assembly for the full irradiation time. It was located in the assembly labeled 12–40 in cycles 12 to 15. Fig. 2 presents the radial layout of this 15×15 assembly and the location of the GU1 sample is indicated in red (in rod C11, where C and 11 indicate the line and the raw in Fig. 2). The vertical segment containing GU1 is number 10 (0 being the bottom of the fuel, and 40 the top of the fuel), as indicated in the same figure (the vertical location is only important in the case of the 3D calculations). The assembly 12–40 contained UO$_2$ fuel rods with an enrichment of 3.5% in $^{235}$U. The total irradiation time was of 1335 days over the 4 cycles, each separated by a period of 30 to 33 days. The isotopic concentrations were measured at SCK-CEN in Belgium after a certain cooling time, varying from 3 to 6 years, depending on the isotopes (ARIANE, 2000).

The sample burnup was originally estimated to be about 60 MWd/kgU (ARIANE, 2000). As observed, the rod containing the GU1 sample is in between the center of the assembly and its side. The sample is therefore expected to be moderately influenced by the surrounding environment (surrounding assemblies), contrary to the U1 sample (located in position A12, but in a different assembly, see Rochman et al., 2020 for the impact of neighboring assemblies).

#### 3.1. Rod relocation

If the rod containing the GU1 sample stayed for the full irradiation periods in the same assembly, such assembly nevertheless went through a modification of a number of other rods during the four cycle irradiation. As noticed in ARIANE (2000), rod exchanges appear at the beginning of the third cycle (cycle 14), as well as at the beginning of the last cycle (cycle 15). These rods were adjacent to C11 and are potentially expected to modify the spectrum environment of the GU1 sample. An illustration is given in Fig. 3. In total, six rods were replaced, at five locations (the location D12 was subsequently filled with two relocated rods). No

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**Fig. 1.** Left: layout of a quarter of the assembly containing the GU1 sample. The rods close to the GU1 sample present random radii. The changes of radius is increased by a factor 10 to be noticeable. Right: same layout but for random rods x and y shifts.
details on the impact of these changes are given in the original ARIANE report (ARIANE, 2000), and it was noticed in Radulescu et al. (2010) that these relocations were nevertheless of potential influence for the GU1 sample. It was concluded in this reference that, due to the lack of information in the original ARIANE report, and to the apparent similar burnup values of the replaced rods with the ones from the symmetrical locations, the reconfiguration was not simulated.

In the present work, three different approaches are pursued:

- **This work**. The CMSYS validated database can be used to obtain the segment burnup for these 6 rods. In this database, the relocation of rods is not taken into account, but these burnup values can still provide an indication with regards to the potential reconfiguration effect. These SIMULATE values are indicated in Fig. 3 with the name “This work”. The same values are used in the scheme (4) presented in Section 2.3 (scheme (5) and (6) are using slightly different burnup values).
- **This work**. In these calculations, the two-dimensional code CASMO5 alone is used without considering the relocation of other rods. These burnup values are obtained from the scheme (1) presented in Section 2.2, and are labeled “This work” in Fig. 3.
- **This work**. One can nevertheless notice that the rods in assembly 12–40 have been extensively replaced for the studies of other various high-burnup samples. The rod B10 in cycle 15 was in fact a rod already relocated once for the study of the U5 sample from the LWR-PROTEUS program (Grimm et al.,...
At the beginning of cycle 15, the U5 sample burnup was higher than the value given in (ARIANE, 2000). Taking into account this sample burnup and the difference in vertical segment (U5 was located in segment 12), the estimated B10 segment burnups are about 69.7 and 80.2 MWd/kgU for the beginning and end of cycle 15, respectively. These values are indicated in Fig. 3 under the label “This work3”. For the other relocated rods, no additional information was found, and the segment burnup provided in ARIANE (2000) are adopted. The assembly irradiation is performed with CASMO5 alone, using the procedure of rod relocation for the 6 rods of interest, at the segment burnup values indicated in ARIANE (2000). The prior irradiation of these rods was performed from assemblies similar to the assembly 12–40.

In the following, the results of these cases will be compared in terms of sample burnup values and isotopic contents. As it will be shown, the effect of the rod relocation on GU1 is relatively small.

3.2. Core position and irradiation

The position of assembly 12–40 is indicated in Fig. 4 with black crosses for the cycles of interest. The red colors are proportional to the calculated assembly burnup values at the end of cycles: light red for low burnup, dark red for high burnup. The values were calculated with SIMULATE3. As observed, the location of the assembly of interest is changing from one cycle to the next, as well as its surrounding assemblies. For these cycles, all loaded assemblies are made of UO2 fuel, and no assembly rotation is performed from one cycle to the next. As observed, all cycles present a strong radial quarter symmetry, but calculated burnup values are nevertheless slightly different in each quarter. The position of control rods are not indicated in the figures, but they are not inserted at any point of the cycle lengths to be close enough to the sample segment.

3.3. Sample burnup

As indicated, the sample burnup was originally expected to be about 60 MWd/kgU. It can be obtained from the 2D and 3D calculations, but not based on the same procedure. In the case of the 2D CASMO5 calculations, the sample burnup value is obtained after possible adjustment so that the calculated and measured 148Nd concentrations are in agreement, for instance within one standard deviation. Such an adjustment is done by increasing or decreasing the segment burnup. The 2D calculations are performed with the hypothesis of infinite lattice, implying that the assembly 12–40 is surrounded by itself in all directions. From such calculation and hypothesis, four actinides mainly contribute to the fission rate (integrated over the full irradiation time): 235U, 238U, 239Pu and 241Pu. Their relative contributions to the fission rate are respectively: 44%, 8%, 38% and 10%.

In the case of 3D calculations, the assembly and sample burnup values cannot be adjusted and are derived by the core simulator given the reactor conditions and the core loading (with heterogeneous assembly loading). These differences impact the burnup...
distribution for all rods, as presented in Fig. 5. As observed, the burnup distributions in the assembly segment are not the same. In the case of the 2D calculation, a symmetrical burnup distribution is obtained, whereas in the 3D one, the effect of the assembly location in the core and of the adjacent assemblies is visible. This effect on the rod of interest leads to variations of the burnup value between both types of calculations: 58.9 MWd/kgU for the 2D cases without rod relocation (as the CASMO5 input sample description is the same for cases (1) to (3), the same sample burnup is obtained), 59.5 MWd/kgU with the rod relocation, and values from 58.2 to 58.8 MWd/kgU for the 3D cases (cases (4) to (6)). Differences are nevertheless rather small, leading to an average value of 58.8 ± 0.5 MWd/kgU (corresponding to one standard deviation), the uncertainty being simply the standard deviation of the calculated values.

Regarding the calculated values reported in the literature, they are 59.7, 60.7 and 58.6 MWd/kgU (ARIANE, 2000; Gauld et al., 2011; Cousin et al., 2010). The burnup value of 60.7 MWd/kgU is reported in (Radulescu et al., 2010; Gauld et al., 2011; Ilas et al., 2010) as the simulations are based on similar input assumptions. The extreme burnup values are separated by 3.5%, reflecting the impact of the various simulation assumptions; the literature values are within two standard deviations of the value obtained in the present work.

3.4. Sample measurements and uncertainties

Isotopic compositions of a number of isotopes (actinides and fission products) were measured for the GU1 sample. The original ARIANE program planned to perform these measurements in three different laboratories: PSI (Switzerland), SCK-CEN (Belgium) and ITU (Germany), but only the measurements from SCK-CEN were kept for the final ARIANE report. In the case of the SCK-CEN measurements, the mass balance was 1.12 for the GU1 sample, leading to an increase of the calculated uncertainties (see next section).

In total, 50 isotopes were measured (18 actinides and 32 fission products). The isotopic concentrations used in this study are directly taken from the ARIANE report (ARIANE, 2000). As different units are reported (mg/gU, μg/g solution, atomic%, mg fuel/g solution), the values in mg/gU (relative to the total amount of measured uranium) are selected for comparison with the present calculations. In the case of concentrations from the residue solution, the sum of concentrations from the dissolved and residue solutions is simply taken; this concerns 99Tc, 55Mo, 101, 106Ru, 103Rh, 109Ag and 125Sb.

Experimental uncertainties are also provided in the ARIANE report. In the case of the SCK-CEN data, these uncertainties are equivalent to 95% confidence intervals, corresponding to a 2σ (standard deviation) if one assumes a Normal distribution. They are obtained from a combination of “manipulation” uncertainties and “measurement” uncertainties. In the following 1σ uncertainties will be reported, being simply half of the ones provided in the ARIANE report.

3.5. Reported mass balance and systematical error

The verification of the sample mass with the method of the “mass balance” allows to check the consistency of the reported experimental data. Detailed explanations and equations can be found in the appendix of the ARIANE report. Because all isotopic concentrations refer to the original solid fuel weight (in mg/gU, μg/g solution or mg fuel/g solution), it is crucial that this quantity is correctly known. An error in the original solid fuel weight will therefore introduce a systematic deviation for all concentrations (or bias). In the original ARIANE report, a tolerance of ±2% in the mass balance is accepted as being not significant.

The mass of the sample can be obtained in two independent ways, which theoretically lead to the same value: the “measured solid fuel weight” (called \( w^m \)), and the derived mass, using the sum of the weights of the measured actinides plus the calculated loss due to fission (called \( w^d \)). The sum \( w^d \), multiplied by the ratio of the fuel weight (approximated by \( ^{238}U + 2 \times ^{16}O \)) over the initial heavy metal weight (≈ 1.1345) can be compared with \( w^m \); it is therefore convenient to define \( R = 1.1345 \times w^d / w^m \) (as described in the ARIANE report). If a substantial difference between these two masses is observed, then \( R \neq 1 \), and it indicates a possible error at one of the stages of the measurements (in the solid fuel weighing and/or in the element concentrations).

**Fig. 5.** Rod burnup distributions for the assembly 12–40 (and neighbors) and segment 10 at the EOC for cycle 15. Left: CASMO5 calculation; Right: SIMULATE3 calculation. The rod C11 containing the GU1 sample is also indicated. For these calculations, the relocation of the other rods is not considered (corresponding to the approach called “This work” in Fig. 3).
The first mass is referred to as the “measured solid fuel weight”, defined as the difference between the initial total sample weight before dissolution (with the cladding) and the cladding weight after dissolution. In the case of the GU1 sample, the reported measured mass is \( w^m = 590.7 \pm 1.0 \mu g \) solution (the uncertainty corresponds to \( \sigma \)).

The mass derived \( w^d \) is simply the sum of the measured element masses for actinides (\( w_{\text{U}} + w_{\text{Pu}} + w_{\text{Cm}}, \text{MA being for minor actinides} \), plus the quantity \( \Delta w \), being the loss of initial uranium due to fission. The quantity \( \Delta w \) can be approximated by \( (w_{\text{Uat144}}/\Gamma) \times 238/148 \), where \( w_{\text{Uat144}} \) is the measured mass of \(^{148}\text{Nd}\), and \( \Gamma \) is the effective yield of \(^{148}\text{Nd}\). All these quantities are provided in the ARIANE report, where \( \Gamma \) is the only one being not measured. The amount of \( w^d \approx 465 \pm 1.0 \mu g \) solution can be obtained from the reported values (\( \Gamma \) is believed to be well known as the fission yields for the main four actinides contributing to the production of \(^{148}\text{Nd}\) are evaluated with small uncertainties).

Consequently \( R = 1.1345 \times 465/591 = 0.89 \), as indicated in the ARIANE report. Such deviation from \( R = 1 \) indicates that at least \( w^n \) or \( w^o \) are not correct. Considering that a tolerance of 2% is acceptable on \( R \), the isotopic compositions can be corrected using a factor between 1.10 and 1.15 (being \( 1+R \pm 2\% \); the value of 1.12 was originally used in the ARIANE report). Because such correcting factor is applied to all measured isotopic concentrations used in this work, it is concluded that a systematical error of 2% exists for all reported data. Such systematical error will be later compared to the reported uncertainties for isotopic concentrations.

4. PIE Results

The measured isotopic compositions for the GU1 sample are compared in the following with calculated values. The effect of nuclear data is also quantified for each isotope, and their partial contributions to the total uncertainties are also presented in this section.

4.1. Nominal calculations

As presented earlier, different types of calculations are performed in this study: 4 with CMS05 only (3 without rod relocation, and one including it), and 3 with the combination of CMSYS/SNF (CMS05, SIMULATE and SNF). The four first ones are lattice 2D calculations, whereas the 3 last ones are full core 3D calculations. Results in terms of C/E-1, in percent, are presented in Figs. 6, 7 and Tables A.1, A.3 and A.4 in Appendix A, C being the calculated values and E the experimental ones. In the case of SNF calculations, only the SNF version 1.07 can provide values for all isotopes. The first remark is that for all CMS05 calculations without rod relocation (same applies for all CMSYS/SNF calculations), differences due to the code version or due to the nuclear data library are small. For almost all isotopes, differences are marginal, with one noticeable exception: \(^{244}\text{Pu}\). The increase of its calculated concentration is due to the change from the e7r1.201.586.bin to the e7r1.202.586.bin library. One difference between the two library versions is the addition in the latest one of the possibility to produce \(^{244}\text{Am}\) from the neutron capture on \(^{243}\text{Am}\). The e7r1.201.586.bin library version only contained the production of \(^{244}\text{Am}\) (ground state). Subsequently, \(^{244}\text{Am}\) can decay by electron conversion to \(^{244}\text{Pu}\), therefore increasing its production compared to the e7r1.201.586.bin library. In the case of the CMSYS/SNF calculations, the main difference (apart from the difference in the \(^{244}\text{Pu}\) concentrations) is that SNF version 1.7 provides more isotopic concentrations compared to SNF version 1.6.4. In all calculation types, similar calculated concentrations are obtained and a poor agreement with the measured values is observed for the same isotopes (\(^{244}\text{Pu}, \ 243, 246\text{Cm}, 241\text{Pu}, 235\text{Pu}, 239\text{Pu}\), \(^{243}\text{Cm}\), \(^{147}\text{Pm}\)). This indicates that for this sample, the 2D calculations provide similar results compared to the 3D ones.

Considering all isotope concentrations, the average C/E-1 is +17.4% for CMS05 version 2.13 and +15.6% for SNF version 1.7. By removing all C/E-1 values above \(+60\% \), the average C/E-1 becomes +0.4% and –0.9%, respectively (this only indicates that a limited number of extreme cases is degrading the average C/E value).

Regarding the effect due to the relocation for adjacent rods, the differences compared to the C/E values obtained without relocation are also limited (see Fig. 7 and Table A.1). The relocated rods differ from the original ones by their burnup rates, but the impact on the GU1 sample concerns mainly a limited number of isotopes: \(^{239}\text{Pu}, \ 242\text{Cm}, \ 109\text{Ag}\) and some Cs isotopes (their relative changes are higher than 50%). It is interesting to note that the Cs and Cm isotopes are mostly affected by the replacement in the last cycle, whereas the effect on \(^{239}\text{Pu}\) is shared between the replacements in cycle 14 and 15. Consequently, the simplification due to assuming no rod relocation moderately affects the isotopic concentrations.

The present calculations can also be compared to the values reported in (Cousin et al., 2010; Azzaoui et al., 2010) for SCALE6/TRITON 2D calculations, see Fig. 8 (only data from Cousin et al. (2010) are presented here as similar trends are observed in both references). In the case of Cousin et al. (2010), the average C/E values are equal to +447% and +20% with all C/E values and only the ones including in \( \pm 60\% \), respectively. The high value of +447% is mainly due to the extreme disagreement for \(^{109}\text{Ag}\). For Azzaoui et al. (2010), the average C/E values are +90% and +26%, under the same conditions. The same outliers are observed in the present calculations and Cousin et al. (2010) and Azzaoui et al. (2010), namely \(^{243}\text{Cm}, \ 109\text{Ag}\) and the other metallic products (C/E values for \(^{244}\text{Pu}\) are not presented in these studies). This would either indicate a common error in the CASMO5 and TRITON calculations (possibly from nuclear data), or a common measurement error with this set of isotopes. The term “error” is used here, as the reported experimental uncertainties are considerably smaller than the differences between C and E (see Tables A.3 and A.4). Additional remarks on experimental uncertainties are given in subSection 5.2.

4.2. Uncertainties due to nuclear data

The effect of changes in nuclear data, both for transport and depletion, is calculated for this sample with SHARK-X using perturbations of cross sections, spectra, neutron emission and fission yields, based on the three major libraries: ENDF/B-VIII.0, JENDL-4.0 and JEFF-3.3. The energy group structure adopted for the perturbation factor is 19, from 0 to 20 MeV, as in Rochman et al. (2020). In the following, the fission yields are called FY and the other nuclear data XS. The perturbation factors are applied to the nominal library (ENDF/B-VII.1). There is therefore an inconsistency between the nominal nuclear data and the covariance libraries. This cannot be avoided as the current versions of the processed CASMO5 libraries are all based on ENDF/B-VII.1.

The total effect is presented in Table A.2 in the columns labeled “2D” and “3D”. The label “2D” refers to the uncertainties from the CASMO5 calculations, using the calculation scheme (1), whereas the label “3D” corresponds to the CMSYS/SNF calculations from the scheme (4). The 3D case was performed with ENDF/B-VIII.0 only. For the case (1), 5000 calculations were performed based on random perturbation factors; for case (4), a
smaller number of 1400 calculations were realized, as they necessitate a larger number of segments (all cycles from 1 to 15 were recalculated with the perturbed nuclear data). The errors of the standard deviations from these two sets of runs are 1.4 and 2.7%, respectively. Such large numbers of perturbations can be used later to extract correlations between XS and FY, and will be presented in a separate study.

One can first notice that for a number of isotopes, their uncertainties can strongly vary, depending on the choice of library. These isotopes are highlighted in red in Table A.2 (see for instance the differences for $^{134}$Cs, which will also have an impact for the decay heat uncertainties). These differences are kept in the following as the impact of nuclear data uncertainties will be presented as range between the minimum and maximum uncertainties. The origin of such differences lies in the individual isotope covariances and is not analyzed here. It is nevertheless important to be sensible to the fact that nuclear data uncertainties as well as their impacts on isotopic concentrations are not unique.

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**Fig. 6.** Comparisons between calculated and measured values (C/E-1) for each measured isotopes from different calculations for the GU1 sample; top: CASMO5 calculation only (This work$^2$), bottom: CMSYS/SNF (This work$^3$). For these calculations, the relocation of the other rods is not considered. Vertical arrows indicate that the C/E-1 values are out of scale.

**Fig. 7.** Comparisons for the same CASMO5 version, but with and without the relocation of rods adjacent to GU1 (see Section 3.1 for details).
One can then notice that in both calculations (1) and (4) (in 2D and 3D), the calculated uncertainties are relatively similar. No strong differences appear for all isotopes. This was rather different in the study of the U1 sample (Rochman et al., 2020), where the case (4) lead to somewhat smaller uncertainties. The U1 study cannot be directly compared with the present work without a detailed analysis, as different nuclear data covariance matrices were used. Another difference between U1 and GU1 is the positioning of the sample: U1 is at the border of the assembly of interest, whereas GU1 is located in an inside row.

A last remark is that the amplitude of the calculated uncertainties are still small compared to some of the C/E values; nuclear data uncertainties are therefore not enough to explain differences between measured and calculated values and biases could be taken into account to explain some deviations (see Section 5.2 for more discussion on this subject).

Apart from the global effect of nuclear data on isotopic compositions, one can assess the separated effects of each type of nuclear data by perturbing only specific ones and keeping other ones constant. This is performed in the next section based on covariances from ENDF/B-VIII.0 only.

4.3. Partial effects of nuclear data

The partial effects of individual nuclear data can be assessed in different manners. One possibility is to perform sensitivity calculations (for instance by modifying one by one cross sections in each specific energy group) and to multiply the sensitivity vectors by the nuclear data covariance matrix. A different method is applied here: perturbing a specific reaction (for instance the $^{235}$U(n,f) cross section) n times and repeating the CASMO5 calculations with each variation. This is similar to the assessment of the global impact of nuclear data, simply applied to specific groups of nuclear data. For the estimation of the partial effects, only the 2D calculations (CASMO5 alone) were considered. For this work, the covariance data from the ENDF/B-VIII.0 are used.

Such method only allows to estimate the importance of each independently varied input quantities, and the (quadratic) sum of their effects is not necessary equal to the total uncertainties, mainly because of the cross correlations due to simultaneous variations of different inputs (this was observed in Rochman et al., 2020 for specific reactor quantities). In practice, varying individual cross sections and fission yields and considering different energy groups requires a large amount of calculations, notwithstanding the normalization issues in the case of fission yields or neutron emitted spectra. Such variations are only partially performed here, for groups of nuclear data and for all energies at once.

First, the XS and FY are varying separately to globally assess the importance of the first group of data compared to the second one. The fission yields from thermal neutron-induced fission are considered for $^{235}$U, $^{239,241}$Pu, whereas for $^{238}$U, the fission yields from fast neutron-induced fission are taken into account. The results are presented in Fig. 9 (top) for 200 perturbations of XS and FY, separately. On this figure, the “relative contribution” of each nuclear data...
Data type is defined as the ratio (in percent) of the partial effect over the total effect. As expected, the variation of FY has a limited impact on actinides, contrary to fission products. XS are major contributors to the uncertainties on actinides, but can also significantly affect some fission products (see the Nd isotopes, $^{155}$Eu and $^{155}$Gd). In fact, FY strongly affect about half of the fission products: metallic fission products (Mo, Ru, Tc and Rh), Cs, $^{109}$Ag, $^{125}$Sb, $^{150}$Nd and $^{154}$Sm. For a number of them, both FY and XS have a non-negligible impact ($^{153}$Sm, Rh, Pm, $^{153}$, $^{154}$Eu). These results are not intuitive, and depends on the nuclear data covariance files which can vary from one nuclear data library to another. This is nevertheless the results obtained with the ENDF/B-VIII.0 covariance files.

Regarding the FY, only a limited number of actinides significantly contributes to the uncertainties of isotope concentrations. Generally speaking, with variations due to the sample burnup, the main contributors to the fission rates are $^{235}$U, $^{239}$Pu, $^{241}$Pu and $^{238}$U. As mentioned, for the GU1 sample, their contributions to the integrated fission rates are 44, 38, 10 and 8%, respectively (as calculated with CASMO5). For the contributions to the isotope uncertainties, the FY for all actinides were also separately varied (with 200 random perturbations) and the results are presented in Fig. 9 (middle) for contributions higher than 50% to the total uncertainties due to FY. It can be observed that the uncertainties are mainly due to $^{235}$U and $^{239}$Pu, with the noticeable exception of $^{109}$Ag (due to $^{241}$Pu). To mention one example, it is interesting to observe that the uncertainties on $^{148}$Nd are firstly due to the $^{239}$Pu (thermal) fission yields. This is an important indication for nuclear data evaluators if one wants to decrease calculated uncertainties on sample burnup (in practice, such uncertainty reduction

Fig. 9. Top: Relative contributions of the cross sections (called XS) and fission yields (called FY) to the total uncertainties. Middle and bottom: same but for FY only and XS only. See text for the definition of the relative contribution.
can be challenging due to the already low uncertainties on the concerned fission yields). The relative contributions of $^{235}$U and $^{239}$Pu will vary as a function of the sample burnup, but as these two actinides are the main fissioning isotopes in UO$_2$ samples, the above remark is likely to be applicable to a large number of UO$_2$ PIE samples. Details for specific fission products are presented in Appendix B.

Finally, the same method is applied to the variations of specific XS (also 200 random perturbations per reaction, e.g. $^{235}$U(n,$f$) or $^{238}$U(n,$\gamma$) reactions) and the main contributors are presented in Fig. 9 (bottom). In this case, it is less straightforward to extract general trends, but two reactions affect a large number of isotopic concentrations: $^{238}$U(n,$\gamma$) and $^{239}$Pu(n,$\gamma$). As indicated in the figure, other capture reactions can be of importance for specific concentrations, such as $^{242}$Pu(n,$\gamma$), $^{235}$U(n,$\gamma$) or $^{242}$Cm(n,$\gamma$). A general observation is that neutron capture cross sections (on actinides or fission products) have a higher contribution to the total XS uncertainties compared to fission cross sections.

5. Summary of PIE C/E and total calculated uncertainties

The effect of the variations of nuclear data and other parameters are presented in Tables A.3 and A.4. For nuclear data, because the three considered libraries do not provide the same isotopic uncertainties, a range is proposed corresponding to the smallest and largest uncertainties. For the other parameters, unique values are proposed, corresponding to one standard deviation. The sum of uncertainties is obtained by performing a simple quadratic sum of all effects, followed by the calculation of the square root. Because of the range of nuclear data uncertainties, two sums are obtained (called “min” and “max”). Additionally, the effect of the correction due to the mass balance (evaluated at 2%) is also presented, resulting in an increase of the total calculated uncertainties.

5.1. Comparison of uncertainties

It can be observed that generally, calculated uncertainties are larger than measured uncertainties. More details are given in the next section. Among the calculated uncertainties, the effect of nuclear data is often the predominant one, but with a number of exceptions: $^{235}$U, $^{239}$Pu, Nd isotopes, $^{149}$Tm, $^{151}$Sm and $^{154}$Eu. Note that these Sm and Eu isotopes are present in small concentrations (less than 0.05 mg/g).

The calculated amounts of fissile isotopes are sensitive to the moderator temperature and density, which is related to a neutron spectrum shift, thus affecting the fission rates. In the case of $^{235}$U, non negligible effects also come from the location of the fuel rods, which is also linked to the thermalization of neutrons and fission rates. As previously indicated, the burnup step (DEP, which also define the sample burnup) also affect all concentrations, as it can be used as a adjustment factor to match the $^{148}$Nd content.

Regarding the effects on the Nd isotopes, the sums of calculated uncertainties are still reasonably small. One can nevertheless note that $^{148}$Nd is considered as the main burnup indicator (following the ASTM recommendations ASTM Standard C1769-15, 2003), and acts as a normalization factor for the simulation of the irradiation. It was recently noticed that its cumulative yields for the thermal fission of $^{235}$U and $^{239}$Pu were increased in the JEFF-3.3 library by more than 0.5 and 1%, respectively. Such changes, not reflected in the present work, can also be added to the effect of nuclear data (although it cannot be estimated by considering the cumulative yield uncertainties alone).

Regarding $\Delta E$, some reported uncertainties are noticeably small (less than 1%). This is not uncommon for reported measured isotopic concentrations, as they represent quantities such as the efficiency of the dissolution of the samples, solution handling, standards, homogenization, element separation, etc. It was nevertheless noted in Gauld et al. (2011) that “Uncertainties were not used quantitatively in this study because the values reported by several laboratories appear to be unrealistically small and did not account for uncertainties in all phases of the measurement process. As noted in many publications, the issues related to the full dissolution of metallic fission products (e.g. Ag, Mo, Tc, Ru, Rh) render the estimation of experimental uncertainties difficult.”. The present study also tends to indicate that the experimental uncertainties do not cover all aspects of the process and need to be reassessed. Similar remarks can be found in (Spent Nuclear Fuel Assay Data for Isotopic Validation, 2011).

5.2. Expanded uncertainties

One of the goals of this study is to compare calculated quantities (C) with experimental ones (E), in order to quantify the power of prediction for isotopic compositions of the different calculation schemes (noted here (1) to (6), together with the simulation including the adjacent rod relocations). Another goal of this analysis is to extract trends to provide a reliable feedback for nuclear data improvements (as presented in Rizzi et al., 2018; Rocchi et al., 2017). Such comparison can be expressed in simple C/E ratios (one for each isotope), but if one considers many PIE samples, a more global quantity can become convenient: for instance average C/E, or a generalized $\chi^2$; these quantities can be expressed per sample, per isotope, or simply over all samples and isotopes (see for instance Gauld et al., 2011). A requirement for such analysis is that, in the case of inconsistency between C and E values, there is an understanding of its origin. Such analysis takes into account $\Delta C$ and $\Delta E$, representing standard deviations (also called uncertainties), as well as biases $\delta$ between C and E (quantified in this work by the values called $\delta = C/E - 1$). In the following, the term “bias” is abusively used: it expresses here the difference between calculated and measured values, which is different from the usual definition referring to the difference between the true and estimated values. With these three quantities $\Delta C$, $\Delta E$ and $\delta$, it is possible to provide so-called “expanded uncertainties”, as introduced in the guidelines of the National Institute of Standards and Technology (Phillips et al., 1997) with three different definitions. Such uncertainties are larger than the three individual components, and helps in resolving discrepancies between E and C. By using $u^2 = (\Delta C)^2 + (\Delta E)^2$, the definitions can be summarized as follows (the reader is referred to Phillips et al. (1997) for detailed descriptions):

- $\delta$ can be treated as another uncertainty source and one can perform the usual root-sum-of-squares (RSS). This method is called RSS$_U$, and the expanded uncertainty $\Delta$expanded is equal to $k \sqrt{\delta^2 + u^2}$ ($k = 1$ in the case of 1σ uncertainty for a Normal distribution);
- the second method (called RSS$_D$) adds the bias $\delta$ to a RSS sum: $\sqrt{\delta^2 + \delta^2 u^2}$;
- finally, the third method called SUM/U proposes an asymmetric uncertainty band $[+U_{+}, -U_{-}]$ defined as

\[
\begin{align*}
U_{+} &= \left\{ \begin{array}{ll}
ku_0 - \delta & \text{if } ku_0 - \delta > 0 \\
0 & \text{if } ku_0 - \delta \leq 0
\end{array} \right.
\end{align*}
\]

and

\[
\begin{align*}
U_{-} &= \left\{ \begin{array}{ll}
ku_0 + \delta & \text{if } ku_0 + \delta > 0 \\
0 & \text{if } ku_0 + \delta \leq 0
\end{array} \right.
\end{align*}
\]

Such quantities $\Delta$expanded are presented in Table A.5 with $k = 1$ for expanded uncertainties being 1σ of a normal distribution. Only
the cases with the largest nuclear data uncertainties are presented. The RSSu, and RSSU provide equal and symmetric uncertainties, whereas SUMU is expressed in terms of asymmetric ranges. These values allow to combine the specific knowledge for the GU1 sample with the current simulations. As noted in Phillips et al. (1997), all expanded uncertainties are greater or equal to uncertainties that would be quoted if biases were corrected or ignored. They also represent the degree of prediction that one can obtain in the specific case of the GU1 sample and the present simulations. Of possible higher interest for the spent fuel characterization is a generalization of the present results and their impact on decay heat. The generalization can only be obtained by analyzing more PIE samples with a similar approach, and the relevance of the present results for decay heat is presented in the next section.6. Decay heat and decay heat uncertainties

Based on the previous full core model (CMSYS/SNF) and the variations of quantities mentioned in the PIE study, the assembly decay heat and its uncertainties can be calculated based on Monte Carlo sampling. The results therefore were obtained from the full 3D assembly model (expressed in Watts per tons of initial heavy metal), but are expected to be similar for the rod containing GU1.

6.1. Calculated decay heat and uncertainties

This approach presents the advantage to provide consistent nominal values and variations between the assembly decay heat and the sample isotopic compositions. Such calculation scheme is nevertheless performed under the assumption that no deformations due to the long-term assembly storage happen.

Results are presented in Table A.6 for the decay heat values and uncertainties due to all considered parameters. In the case of nuclear data from the ENDF/B-VIII.0 library, the main contributors to the uncertainties are presented in Fig. 10. Note that experimental uncertainties on decay data are not considered in this study. Important isotopes such as $^{90}$Sr, $^{90}$Y and $^{137}$Cs do not appear in this figure as they are not main contributors to the total decay heat uncertainty (between 1 and 100 years of cooling time, the main contributors are $^{134}$Cs, $^{137}$mBa, $^{244}$Cm and $^{239}$Pu). From Table A.6, one can see that the main source of uncertainties are from nuclear data. The ENDF/B-VIII.0 and JENDL-4.0 libraries provide relatively similar uncertainties, whereas uncertainties from JEFF-3.3 are smaller, especially for the first 50 years of cooling time. The other important quantities are the moderator temperature, and the assembly pitch. The total uncertainties presented in Table A.6 are obtained with a simple quadratic sum, considering all variables independent. Such values do not consider possible biases, such as coming from the PIE measurements, e.g. between the measured and calculated $^{239}$Pu concentration.

In Fig. 10, the ten most important isotopes in term of decay heat uncertainties are presented. The separation between fission products and actinides can be observed, with a transition period between 10 and 100 years (as already noticed in Rochman et al. (2018)). Considering the ENDF/B-VIII.0 library, the main source of uncertainty below 10 years is the $^{134}$Cs concentration. As presented in Fig. 9, this isotope is mainly influenced by the fission yields of $^{235}$U and $^{239}$Pu, and not by cross sections. Such remark can explain the difference of decay heat uncertainties with the JEFF-3.3 library (lower by more than a factor 2 below 10 years of cooling time), as the uncertainty for the thermal neutron-induced independent fission yield of $^{133}$I (leading to $^{133}$Cs by beta decay, then to $^{134}$Cs by neutron capture) is almost a factor 2 lower in JEFF-3.3 than in ENDF/B-VIII.0 for both $^{235}$U and $^{239}$Pu.

In the case of the contributions from the actinides, Fig. 10 does not represent possible (anti-) correlation, for instance between $^{239}$Pu and $^{240}$Pu. Such anti-correlations imply that the total uncertainties is not obtained from a simple quadratic sum of the independent contributions of each isotope. The importance of the plutonium isotopes, as well as $^{241}$Am is also explicit from this figure.

6.2. Expanded uncertainties

Similar to the case of isotopic concentrations, expanded uncertainties can be obtained for the decay heat as a function of cooling time. As such decay heat is not measured, the experimental uncertainty $\Delta C$ is not known, and is therefore not considered in this study. If known, the expanded uncertainties would then be greater. Two other quantities can nevertheless be deduced: $\Delta C$ is obtained from Table A.6 by calculating the RSS, and $\delta_{\text{decay heat}}$ can be approximated from the $\delta$ for the isotopic compositions: as the decay heat is obtained from the isotopic concentrations (plus the decay data information), a bias in a specific isotopic concentration will be transferred to the decay heat, proportionally to this isotope contribution to the total decay heat. For instance, $^{240}$Pu contributes to 50% of the total decay heat for 5 000 years of cooling time (such contribution is provided by the SNF code), followed by $^{239}$Pu up to 40%. Therefore the bias on 90% of the total decay heat is equal to $-3.5 \times 0.5 - 3.2 \times 0.4 = -3.1\%$, as reported in Table A.7. This approach is directly applied for various cooling times, considering the isotopes contributing the most to the decay heat and with a significant bias: $^{134}$Cs, $^{244}$Cm, $^{106}$Ru, $^{239}$Pu. For other important isotopes which are not measured (e.g. $^{90}$Y, $^{106}$Rh), their direct parent or daughter isotopes are considered: in the case of $^{106}$Rh, the isotope $^{106}$Ru is measured, which decays at 100% to $^{106}$Rh; for $^{90}$Y, $^{90}$Sr is considered instead.

This method provides only an underestimation of the decay heat bias, as not all isotopes are measured. The proposed expanded uncertainties are presented in Table A.7 for the RSSu, and SUMU methods. As observed, because of the large bias on $^{106}$Ru, the expanded uncertainties for low cooling times are rather large. As a limited comparison, these indirect biases can be compared to
the ones obtained between CASMOS calculations and decay heat measurements from the CLAB and GE-Morris campaigns (Sturek and Agrenius, 2006; Gauld et al., 2010). Our comparisons for 60 measurements between 3 and 20 years of cooling time lead to a spread of \( C/E \sim 1 \) of 1.8% (1\( \sigma \)) with extreme values of -4 and +4\% (similar values were presented in Yamamoto and Iwahashi (2016)). Such spread is somehow lower than the bias presented in Table A.7 for the same cooling periods, but is of the same order of magnitude.

As a final remark, it is clear from Table A.7 and Fig. 10 that the decay heat can be considered as an integral quantity, possibly correctly estimated due to compensation effects from biases of isotopic concentrations. It is equivalent to the \( k_{\text{eff}} \) value for criticality-safety benchmarks which is often well estimated, even if it is recognized that this is due to compensation effects from partially wrong contributions of various nuclear data reactions.

7. Conclusion

This work is part of a validation effort of the lattice code CASMOS and the depletion code SNF at PSI, considering samples from post-irradiation examination and their isotopic compositions. Presently, the isotopic compositions for 50 isotopes from the GU1 sample from the ARIANE program were calculated and compared to the measured values. Different calculation schemes were used: lattice calculations based on CASMOS, and full core calculations based on CMSYS/SNF. It was found that a similar agreement with the measured concentrations was observed between the two and three dimensional calculations. Additionally, the effect of relocation of adjacent rods on the GU1 burnup and isotopic concentrations is rather small. The effects of nuclear data uncertainties, manufacturing tolerances and operating conditions were also quantified and separated effects for nuclear data were presented. The combination of biases and uncertainties was performed under so-called “expanded uncertainties”. Finally, the assembly decay heat was calculated, together with uncertainties, in a consistent way with the isotopic compositions. In a similar manner, expanded uncertainties were proposed.

This study will be complemented by similar analysis for other PIE samples, within the goal of improving the spent fuel characterization by providing simulation prediction power for isotopic concentrations, decay heat and their uncertainties.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was partly supported by swissnuclear, the association of the Swiss nuclear power station operators, with the COLOSS project.

It was also partly funded by the European Union’s Horizon 2020 Research and Innovation Programme under grant agreement No 847593 (the EURAD project, Work Package 8).

The authors are grateful to G. Ilas from ORNL for sharing her experience on the GU1 sample and providing constructive remarks.

Appendix A. Tabulated isotopic compositions and uncertainties

Table A.1
Values of the C/E-1 ratios (in%), comparing the effect of the relocation of adjacent rods (see Section 3.1), based on CASMOS v2.03. Isotopes showing the largest differences are highlighted in “bold” (see Section 4.1 for comments).

| Isotope   | Without Relocation | With Relocation | Without Relocation | With Relocation |
|-----------|---------------------|-----------------|--------------------|-----------------|
| ^{243}Am | +20.7               | +20.2           | +24.4             | +20.8           |
| ^{241}Am | +1.2                | +1.3            | +0.0              | +0.0            |
| ^{242}Pu | -3.1                | -2.5            | -1.8              | -3.2            |
| ^{244}Pu | -3.0                | -3.5            | -4.3              | -5.3            |
| ^{242}Cm | -4.4                | -4.6            | -67.9             | -68.4           |
| ^{241}Am | +1.7                | +0.5            | +24.4             | +20.8           |
| ^{243}Am | -5.7                | -6.0            | -3.4              | -7.2            |
| ^{243}Cm | +219                | +219            | -7.9              | -7.7            |
| ^{245}Cm | -5.1                | -5.0            | -27.4             | -26.0           |
| ^{90}Sr  | -21.3               | -21.2           | +14.2             | +14.6           |
| ^{99}Tc  | +27.9               | +27.8           | +103              | +103            |
| ^{106}Ru | +77.5               | +79.4           | +91.9             | +91.6           |
| ^{109}Ag | +523                | +522            | +47.4             | +48.2           |
| ^{33}Cs  | +4.8                | -0.3            | -3.4              | +4.7            |
| ^{135}Cs | -1.1                | -2.6            | -1.4              | +0.2            |
| ^{44}Ce  | -1.9                | -1.3            | -1.7              | -1.2            |
| ^{43}Nd  | +3.7                | +3.5            | -1.6              | -1.5            |
| ^{45}Nd  | +1.7                | +1.7            | -0.7              | -0.6            |
| ^{48}Nd  | -0.4                | -0.4            | -0.7              | +0.3            |
| ^{147}Sm | +50.8               | +51.0           | -1.9              | -1.8            |
| ^{148}Sm | -5.8                | -5.5            | +1.0              | +1.6            |
| ^{151}Sm | -1.5                | +1.6            | -7.1              | -7.8            |
| ^{154}Sm | -4.9                | -5.0            | -2.1              | -2.2            |
| ^{153}Eu | -3.0                | -3.2            | +9.8              | +9.6            |
| ^{155}Eu | -4.9                | -4.9            | +1.9              | +1.9            |
Table A.2
Comparison of uncertainties (in %) for the isotopic concentrations from the three different libraries. Isotopes highlighted in "bold" present large differences (factor 5 or more). "2D" and "3D" results refer to CASMO5 (v2.03) and SNF, respectively. For JENDL-4.0 and JEFF-3.3, results correspond to the CASMO5 v2.03 calculations.

| Isotope | ENDF/B-VIII.0 2D | ENDF/B-VIII.0 3D | JENDL-4.0 2D | JENDL-4.0 3D | JEFF-3.3 2D | JEFF-3.3 3D |
|---------|------------------|------------------|---------------|---------------|-------------|-------------|
| 246Cm   | 0.6              | 0.7              | 9.9           | 9.9           | 2.8         | 2.8         |
| 245Cm   | 0.5              | 0.5              | 1.3           | 1.3           | 1.6         | 1.6         |
| 244Cm   | 2.7              | 2.7              | 9.4           | 9.4           | 13.2        | 13.2        |
| 242Pu   | 4.2              | 4.2              | 4.9           | 4.9           | 2.5         | 2.5         |
| 242Pu   | 5.0              | 5.0              | 8.7           | 8.7           | 8.6         | 8.6         |
| 241Am   | 3.5              | 3.5              | 4.4           | 4.4           | 3.6         | 3.6         |
| 241Am   | 7.5              | 7.5              | 7.0           | 7.0           | 8.7         | 8.7         |
| 241Am   | 11.4             | 10.8             | 12.2          | 12.2          | 16.9        | 16.9        |
| 245Cm   | 12.7             | 12.4             | 14.9          | 14.9          | 14.0        | 14.0        |

Table A.3
Summary of all uncertainties in % (or 1σ) considered in this study for actinides. For nuclear data, the range from Table A.2 is presented here. The "Sum" column corresponds to $\sqrt{\sum_i \Delta(C_i)}$ for calculated uncertainties. See text for the description of the columns. *The relocation of rods is considered in the CJE values. The systematic effect from the mass balance is 2%.

| Isotope | C+1/E-1 (ÅE) | ND min – max | TFU | TMO | BOR | RAD | ENR +DEN | (X-Y) | DEP | PITCH | With Mass balance | Sum min – max |
|---------|--------------|--------------|-----|-----|-----|-----|----------|-------|-----|-------|------------------|---------------|
| 234U    | +20.2/2.5    | 0.6 – 9.9    | 0.1 | 0.1 | 0.0 | 0.0 | 0.1      | 0.3   | 0.2 | 0.6   | 2.2 – 10.1       | 0.6 – 9.9     |
| 235U    | +7.2/1.3     | 2.3 – 4.1    | 0.3 | 2.8 | 0.1 | 0.1 | 0.8      | 1.7   | 1.1 | 3.3   | 5.7 – 6.7        | 2.3 – 4.1     |
| 236U    | +1.3/0.4     | 0.5 – 1.6    | 0.0 | 0.3 | 0.0 | 0.0 | 0.2      | 0.0   | 0.3 | 0.1   | 2.1 – 2.6        | 0.5 – 1.6     |
| 238U    | +0.0/0.3     | 0.0 – 0.0    | 0.0 | 0.0 | 0.0 | 0.0 | 0.0      | 0.0   | 0.0 | 0.0   | 2.0              | 0.0 – 0.0     |
| 239Pu   | +2.5/1.5     | 2.7 – 13.2   | 0.0 | 1.2 | 0.0 | 0.0 | 0.0      | 0.3   | 0.0 | 1.7   | 4.0 – 13.5       | 2.7 – 13.2    |
| 235Pu   | +3.2/0.3     | 1.9 – 2.6    | 0.2 | 1.3 | 0.1 | 0.0 | 0.2      | 0.4   | 0.0 | 1.5   | 3.4 – 3.9        | 1.9 – 2.6     |
| 239Pu   | +3.5/0.3     | 2.5 – 4.9    | 0.1 | 0.7 | 0.0 | 0.0 | 0.1      | 0.1   | 0.1 | 0.5   | 3.3 – 5.4        | 2.5 – 4.9     |
| 241Pu   | +5.3/0.3     | 3.4 – 3.9    | 0.2 | 1.6 | 0.0 | 0.0 | 0.3      | 0.3   | 0.0 | 1.4   | 4.5 – 4.9        | 3.4 – 3.9     |
| 242Pu   | +4.6/0.3     | 5.0 – 8.7    | 0.0 | 0.4 | 0.0 | 0.0 | 0.0      | 0.5   | 0.5 | 0.4   | 5.6 – 9.0        | 5.0 – 8.7     |
| 244Pu   | –68.4/25     | 8.1 – 8.7    | 0.1 | 0.6 | 0.0 | 0.0 | 0.8      | 1.3   | 1.0 | 2.6   | 8.9 – 9.5        | 8.1 – 8.7     |
| 241Am   | +0.5/1.8     | 3.5 – 4.4    | 0.2 | 1.6 | 0.1 | 0.0 | 0.3      | 0.4   | 0.0 | 1.7   | 4.7 – 5.4        | 3.5 – 4.4     |
| 242mAm  | +20.8/5.3    | 4.0 – 4.4    | 0.3 | 3.2 | 0.1 | 0.1 | 0.5      | 0.8   | 0.0 | 3.6   | 6.6 – 6.9        | 4.0 – 4.4     |
| 243Am   | +6.0/1.8     | 7.4 – 8.7    | 0.1 | 0.5 | 0.0 | 0.0 | 0.6      | 0.7   | 0.7 | 1.8   | 8.0 – 9.2        | 7.4 – 8.7     |
| 242Cm   | +7.2/3.6     | 3.0 – 3.5    | 0.1 | 1.1 | 0.0 | 0.0 | 0.3      | 0.1   | 0.2 | 1.2   | 4.0 – 4.4        | 3.0 – 3.5     |
| 243Cm   | +219/37      | 11.4 – 16.2  | 0.1 | 1.7 | 0.0 | 0.0 | 0.4      | 0.1   | 0.4 | 2.0   | 11.9 – 16.5      | 11.4 – 16.2   |
| 244Cm   | –7.7/1.5     | 8.6 – 9.7    | 0.0 | 1.1 | 0.0 | 0.0 | 0.1      | 0.7   | 1.1 | 2.8   | 9.5 – 10.5       | 8.6 – 9.7     |
| 245Cm   | –5.0/3.0     | 12.7 – 14.9  | 0.1 | 4.0 | 0.1 | 0.1 | 0.7      | 0.8   | 1.3 | 3.9   | 14.1 – 16.1      | 12.7 – 14.9   |
| 246Cm   | –26.0/10.1   | 13.5 – 24.0  | 0.1 | 1.2 | 0.0 | 0.1 | 1.0      | 1.7   | 1.8 | 4.3   | 14.6 – 24.6      | 13.5 – 24.0   |
Table A4
Same as in Table A3, but for fission products.

| $^{150}$Sm | 0.56 | +1.6 | 0.3 | 2.6 |
| $^{145}$Nd | 1.14 | +1.7 | 0.3 | 3.1 |
| $^{241}$Am | 0.27 | +0.5 | 1.8 | 5.4 |
| $^{149}$Sm | 2.6 | 1.5 | 20.0 | 12.5 |
| $^{150}$Nd | −1.5 | 0.3 | 1.9 | 3.0 |
| $^{149}$Nd | 0.7 | 2.5 | 0.3 | 2.6 |
| $^{146}$Nd | −0.6 | 0.3 | 2.0 | 3.1 |
| $^{47}$Pt | 2.6 | 2.5 | 0.3 | 2.6 |
| $^{154}$Eu | 3.8 | 2.5 | 4.4 | 5.1 |
| $^{151}$Sm | 1.3 | 0.3 | 1.9 | 2.6 |
| $^{155}$Cd | 1.9 | 2.5 | 0.3 | 2.6 |

Table A5
Expanded estimated uncertainties $\Delta_{\text{expanded}}$ on isotopic concentrations, taking into account the calculation bias and the experimental and calculated uncertainties. $\Delta_{\text{max}}$ come from the nuclear data library leading to the highest nuclear data uncertainty (either ENDF/B-VIII.0, JEFF-3.3 or JENDL-4.0). Three combination methods are presented: RSS$_{\text{ui}},$ RSS$_{\text{SU}}$ (being equal to RSS$_{\text{SU}},$ for 1), and SUMU.

| Calculation | Bias (mg/g) | $\Delta_\text{E}$ | $\Delta_{\text{max}}$ | RSS$_{\text{ui}}$/RSS$_{\text{SU}}$ | SUMU | Calculation | Bias (mg/g) | $\Delta_\text{E}$ | $\Delta_{\text{max}}$ | RSS$_{\text{ui}}$/RSS$_{\text{SU}}$ | SUMU |
|-------------|-------------|-------------------|----------------------|-----------------------------|------|-------------|-------------|-------------------|----------------------|-----------------------------|------|
| $^{234}$U   | 0.16        | +20.2             | 25                   | 10.1 $\pm 22.7$             | [0.1] | $^{235}$U   | 2.44        | +7.2             | 1.3                   | 6.7 $\pm 9.9$            | [0.4 - 14]         |
| $^{236}$U   | 5.28        | +1.3              | 0.4                  | 2.6 $\pm 2.9$               | [1.3 - 3.9] | $^{238}$U   | 99.19       | +0.0             | 0.3                   | 2.0 $\pm 2.0$            | [+2.0 - 2.0]        |
| $^{238}$Pu | 0.47        | −2.5              | 1.5                  | 13.5 $\pm 13.8$             | [−16.1] | $^{239}$Pu | 5.17        | −3.2             | 0.3                   | 3.9 $\pm 5.1$            | [+7.1 - 0.7]        |
| $^{240}$Pu | 3.33        | −3.5              | 0.3                  | 5.4 $\pm 6.4$               | [−8.9 - 1.9] | $^{241}$Pu | 1.49        | −5.3             | 0.3                   | 4.9 $\pm 7.2$            | [+10.0]           |
| $^{242}$Pu | 1.60        | −4.6              | 0.3                  | 9.0 $\pm 10.1$              | [−13.4 - 4.4] | $^{244}$Pu | 1.01E-4     | −68.4            | 25.9                  | 9.5 $\pm 73$             | [+95.0]            |
| $^{244}$Am | 0.27        | +0.5              | 1.8                  | 5.4 $\pm 5.7$               | [−5.2 - 6.2] | $^{242}$Am | 9.20E-4     | +20.8            | 5.3                   | 6.9 $\pm 22.5$           | [+0.0 - 30]        |
| $^{234}$Am | 0.41        | −6.0              | 1.8                  | 9.2 $\pm 11.1$              | [−15.3 - 3.4] | $^{242}$Cm | 3.2E-4      | −7.2             | 3.6                   | 4.4 $\pm 9.2$            | [+13.0]           |
| $^{234}$Cm | 1.16E-3     | +2.19             | 37                   | 16.5 $\pm 22.3$             | [0.0 - 260] | $^{244}$Cm | 0.24        | −7.7             | 1.5                   | 10.5 $\pm 13.1$          | [+18.9 - 29] |
| $^{178}$W  | 1.78E-3     | −5.0              | 3.0                  | 16.1 $\pm 21.1$             | [−22.1 - 12] | $^{246}$Cm | 4.15E-3     | −26.0            | 10.1                   | 24.6 $\pm 37$           | [+53.0 - 64]       |

0.5, 1.0, and 1.5 ns for the calculation bias and the experimental and calculated uncertainties. $\Delta_{\text{max}}$ come from the nuclear data library leading to the highest nuclear data uncertainty (either ENDF/B-VIII.0, JEFF-3.3 or JENDL-4.0). Three combination methods are presented: RSS$_{\text{ui}},$ RSS$_{\text{SU}}$ (being equal to RSS$_{\text{SU}},$ for 1), and SUMU.
combination methods are presented: RSSuc, RSS. Uncertainties for the fission product concentrations (see Fig. 9),

As mentioned before, only four actinides cover more than 99.7% of the fission products presented in Table A.2 can be explained. Examples are presented in Table B.1 in the case of the ENDF/B-VIII.0 library.

Appendix B. Details of uncertainty sources for specific fission products

When uncertainties of fission yields are the dominant sources of uncertainties for the fission product concentrations (see Fig. 9), their concentration uncertainties can be explained by considering the actinides contributing the most to the integrated fission rate. As mentioned before, only four actinides cover more than 99.7% of such rate: 235U (44%), 239Pu (38%), 241Pu (10%) and 238U (8%). Depending on the values of their fission yields and on the values of their fission yield uncertainties, the total uncertainty on the fission products presented in Table A.2 can be explained. Examples are presented in Table B.1 in the case of the ENDF/B-VIII.0 library.

As a representative case, the uncertainties for 90Sr are detailed. The amount of 90Sr measured is close to the 90Sr uncertainty is sensibly smaller than the FY contributions. As presented in Fig. 9, the contributions of XS to the 90Sr=0.7%. As presented in Fig. 9, the contributions of XS to the 90Sr uncertainty is sensibly smaller than the FY contributions. As presented in Fig. 9, the contributions of XS to the 90Sr uncertainty is sensibly smaller than the FY contributions.

Table A.5

| Cooling time (years) | Decay heat (W/t) | END/F/B-VIII.0 | JEFF-3.3 | JENDL-4.0 | TFU | TM0 | B0R | RAD | ENR | X-Y | DEP | PITCH | SUM min–max |
|----------------------|-----------------|----------------|-----------|----------|-----|-----|-----|-----|-----|-----|-----|--------|---------------|
| 0.01                 | 134 143         | +6.9           | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.02                 | 95 722          | +6.5           | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.03                 | 49 089          | +13            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.04                 | 36 625          | +16            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.05                 | 23 099          | +19            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.06                 | 14 787          | +22            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.07                 | 7 760           | +25            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.08                 | 2 297           | +28            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.09                 | 1 409           | +31            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.10                 | 976             | +34            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.11                 | 713             | +37            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.12                 | 488             | +40            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.13                 | 362             | +43            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |
| 0.14                 | 26              | +46            | -         | -        | -   | -   | -   | -   | -   | -   | -   | -      | -             |

Table A.6

Summary of all uncertainties in % (or 1σ) considered in this study for the calculated decay heat at the end of irradiation of the GU1 sample. The min – max of the sum (being equal to RSSuc for 175U) expanded for the decay heat, taking into account the main calculation biases for isotopic concentrations from Table A.5 and calculated uncertainties for the decay heat, taking into account the main calculation biases for isotopic concentrations from Table A.5 and calculated uncertainties from Table A.6. ΔCmax come from the nuclear data library leading to the highest nuclear data uncertainty (either ENDF/B-VIII.0, JEFF-3.3 or JENDL-4.0). Three combinations methods are presented: RSSu, RSSu (being equal to RSSu, for 1σ), and SUMU.
Details of the uncertainties for specific fission products provided in Table A.2. “Cont” means percent contribution to the isotope production from CASMO5 for the main four actinides. “Calculated” means the derived values from cumulative yields and their uncertainties, and the “Observed” values correspond to the uncertainties from the sampling method.

| Isotope  | Cont. \(\Delta C_{U235}\) | Cont. \(\Delta C_{Pu239}\) | Cont. \(\Delta C_{Pu241}\) | Cont. \(\Delta C_{U238}\) | Total \(\Delta C_{obs}\) |
|----------|------------------|------------------|------------------|------------------|------------------|
| \(^{90}\)Sr | 68% | 0.6 | 21% | 0.3 | 4% | 0.1 | 7% | 0.1 | 0.7 |
| Calculated | Observed | | | | |
| \(^{95}\)Mo | 52% | 0.6 | 34% | 0.4 | 7% | 0.2 | 8% | 0.1 | 0.7 |
| Calculated | Observed | | | | |
| \(^{99}\)Tc | 25% | 0.5 | 25% | 1.4 | 24% | 0.3 | 26% | 0.2 | 1.5 |
| Calculated | Observed | | | | |
| \(^{235}\)U | 42% | 0.6 | 39% | 2.9 | 11% | 0.6 | 8% | 0.3 | 3.0 |
| Calculated | Observed | | | | |
| \(^{239}\)Pu | 0% | 0.6 | 29 | 2.9 | 0% | 0.6 | 3% | 0.3 | 3.2 |
| Calculated | Observed | | | | |
| \(^{103}\)Ru | 3% | 0.1 | 32% | 1.2 | 46% | 0.6 | 19% | 0.1 | 1.3 |
| Calculated | Observed | | | | |
| \(^{109}\)Ag | 13% | 0.1 | 29% | 1.2 | 30% | 0.6 | 28% | 0.6 | 1.5 |
| Calculated | Observed | | | | |
| \(^{133}\)Cs | 1% | 1.5 | 20% | 21 | 72% | 21 | 7% | 1.8 | 30 |
| Calculated | Observed | | | | |
| \(^{137}\)Cs | 25% | 20 | 25% | 17 | 25% | 5 | 25% | 4 | 27 |
| Calculated | Observed | | | | |
| \(^{148}\)Nd | 25% | 5 | 24% | 4 | 27% | 0.5 | 24% | 0.2 | 6.5 |
| Calculated | Observed | | | | |
| \(^{181}\)Ta | 23% | 0.2 | 22% | 0.25 | 26% | 0.10 | 29% | 0.07 | 0.3 |
| Calculated | Observed | | | | |

\(\Delta C_{Pu239}=0.3\%\), \(\Delta C_{Pu241}=0.06\%\) and \(\Delta C_{U238}=0.1\%\). Note that the square root of the quadratic sum of these independent contributions corresponds to the uncertainty obtained from the variations of all FY together (0.7\%). Such partial contributions can be explained with the following considerations.

For the mass 90, the isotope considered by CASMO5 with the highest nuclear charge is \(^{90}\)Sr (\(^{90}\)Ru, \(^{90}\)Kr or \(^{90}\)Br isotopes have half-lives too short to contribute to neutron reactions). Therefore its cumulative yield is used instead of its independent yield in the CASMO5 library. For the four actinides of interest (\(^{235}\)U, \(^{239}\)Pu, \(^{241}\)Pu and \(^{238}\)U), the \(^{90}\)Sr cumulative yields considered in this study are 5.79 ± 0.8\%, 2.10 ± 1.7\%, 1.53 ± 1.6\% and 3.2 ± 1.4\%, respectively, corresponding to 46\%, 17\%, 12\% and 25\% of the sum of the yields.

Regarding the total production of \(^{90}\)Sr by fission, \(^{235}\)U contributes to 68\% (44\% fissile x 46\% yields = 0.202 which is 68\% of the total \(^{90}\)Sr produced), which in terms of contribution to the uncertainties is: 68\% x 0.8 = 0.6\%. This value is reported in Table B.1 as “calculated,” and is equal to \(\Delta C_{U235}\) obtained from the Monte Carlo sampling method. Similar results are obtained for the other contributions and the final results are presented in Table B.1.

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