BEST ESTIMATE PLUS UNCERTAINTY ANALYSIS FOR THE ^{244}CM PREDICTION IN SPENT FUEL CHARACTERIZATION

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ABSTRACT

The estimation of uncertainties from various sources are presented for the calculation of isotopic concentrations for Post Irradiation Examination (PIE) data. The specific cases of Swiss irradiated samples during the PROTEUS program are considered, with an emphasis on important isotopes for spent fuel storage (such as ^{244}Cm). The calculations are performed with CASMO-5, and the irradiation history is based on realistic conditions extracted from core validated models. The considered samples are U1 and U2, with *reported* burnup values of 37.6 and 54.3 MWd/kgU, respectively. The method is simply based on Monte Carlo uncertainty propagation: repeating a large number of times the same calculation, each time changing the initial conditions.
Different sources of uncertainties are considered: from nuclear data, operating conditions & manufacturing tolerances (based on probability density functions, or pdf, from the literature), with a preliminary assessment of the burnup-induced technological changes.
Results are presented with a comparison to measured isotopic concentrations. It will be shown that (1) the impact of the different sources of uncertainties is relatively important (*e.g.* 10 % on ^{244}Cm), and (2) that a detailed irradiation history generally improves the agreement between calculation and measurement.

1. INTRODUCTION

The characterization of the spent nuclear fuel (SNF) with a reasonable level of confidence is of high importance in the context of its transport and intermediate- and long-term storage. For each assembly, such characterization is performed with the use of a few quantities (averaged or spatially distributed), such as its burnup, isotopic content, and source terms (neutron and gamma); the goal being of reaching transport and storage solutions which are both safe (from the point of view of criticality and radiation dose) and economically sound.

As many of these quantities cannot systematically be measured for the thousands or tens of thousands of SNF, the dependence on simulations is crucial. But the use of calculated quantities implies different levels of codes and systems validations, for instance with respect to existing measurements, such as from Post Irradiation Examination (PIE). Different PIE data are available in the literature, such as from the SFCOMPO database, or MALIBU and ARIANE campaigns [1, 2, 3]

and are for instance used to compare calculated and measured isotopic contents, knowing the irradiation history of the specific samples. At PSI, the LWR-PROTEUS Phase II experiments are used for such purpose [4]. These data are recognized to be of good quality, leading to trustful measured inventories. The LWR-PROTEUS results for specific isotopes are therefore unique candidates to provide a detailed comprehension and analysis of the number density predictions.

Still, as for any simulated quantities, the calculations can be influenced by specific assumptions and input choices. The estimation of these input parameters are of course determined by the history of the selected SNF, but a level of expert judgment is also playing a role. Such uncertainties are of importance for the later calculation of bias and safety margins for the handling of the SNF. A convenient way to quantity the impact of the selected parameters is to estimate the uncertainties on calculated values by randomly varying inputs. This simple Monte Carlo approach allows to assess the uncertainties (standard deviations) with confidence as long as the employed method is explained.

This is the subject of this paper: for two specific LWR-PROTEUS samples (called U1 and U2), the best estimate calculations will be presented (with a comparison between calculations and measurements for the ^{244}Cm isotopic content), together with calculated uncertainties due to various inputs: nuclear data, operating conditions, manufacture tolerances and burnup induced technological changes. For parts of these inputs, specific assumptions for their uncertainties and probability density functions are still based on *ad-hoc* expert judgment, but this work allows to quantify such effects, given these assumptions.

2. CALCULATION TOOLS

The simulation tool in this work is the lattice code CASMO, version 5MX. It is based on version 5M 1.07.01, with PSI modifications to allow for nuclear data variations [5]. Without nuclear data perturbation, this code version is equivalent to the version 5, with the ENDF/B-VII.1 nuclear data library (release “e7r1.201.586"). The version 5MX is included in SHARK-X tool, version 2.0. SHARK-X is a set of Perl scripts which allows to modify the nuclear data inside CASMO; further details on the SHARK-X tool for the PROTEUS sample U2 can be found in Ref. [6].

The use of the CASMO simulation lattice code brings the advantage to either follow the irradiation history of the specific samples as given by the pre-calculations of the fuel vendors, or to use the complete irradiation history of the specific power plant as performed for the safety studies by the LRT laboratory (Reactor Physics and Thermal hydraulic Laboratory) for the Swiss regulator.

In the case of the Gösgen PWR power plant, where the LWR-PROTEUS U1 and U2 samples considered in this work were irradiated, the operational data (such as the fuel and moderator temperatures, the moderator and power density) can be obtained from the plant data as used in SIMULATE. These irradiation history data went through an extensive validation procedure over many years.

3. IRRADIATION HISTORY

The irradiation history of a specific sample has a high impact on the calculated inventory. In the case of ^{244}Cm, the dependence on the burn-up history is believed to follow a 4th power variation [7]). For the LWR-PROTEUS samples considered in this study, a large experience was gained over the past decades as presented in many publications and reports (see for instance Refs. [6, 7, 8]). In this paper, the irradiation history is defined by the following quantities which enter in the CASMO input file:

- PDE: sample power,
- TFU & TMO: fuel and moderator temperatures,
- BOR: boron concentration,
- DEP: burn-up steps.

Each of these quantities is repeated many times in the CASMO input file for different burn-up steps. The final calculated number densities for actinides and fission products will depend on these quantities as well as the considered cooling time. The number of burn-up steps follows the available information about the irradiation history of the assembly of interest. Traditionally, this information is obtained from public documentation, or is delivered as "pre-cycle calculation" (as in the case of the LWR-PROTEUS samples analyzed in the above references). Such information is often given on a rather coarse time scale, with often 4 or 5 steps per cycles. Examples of these step functions are presented in Figure 1 for the U2 sample.

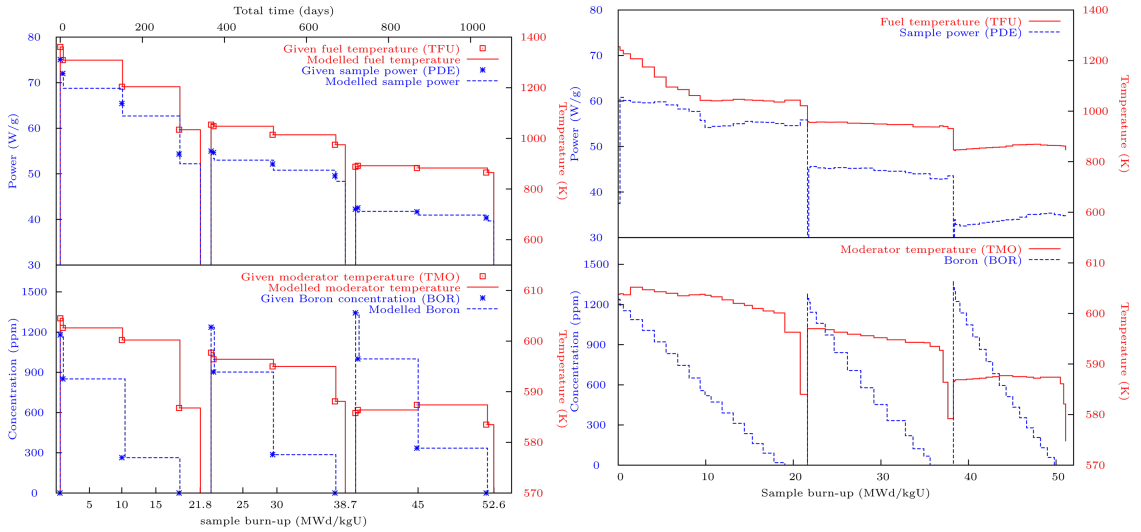

Figure 1 – Left: Four main CASMO parameters used in Ref. [7] for the U2 sample. Right: same parameters obtained with the detailed history, extracted from SIMULATE).

Based on the provided values for the quantities of interest (DEP, TMO...) and for specific assemblies, one can start to adjust these parameters in order to match the reported "measured sample burn-up" value, or in a more direct way, the measured number densities for burn-up indicators

such as ^{148}Nd. This way, the sample burn-up can be determined with the CASMO calculation, and once the burn-up values are set, other number densities can be calculated.

Another approach is now available at PSI within the detailed database of the Swiss reactor cycle histories. The LRT group is analyzing the irradiation cycles for all the Swiss power plants and has access to many plant irradiation histories. These histories are used to analyze the different cycles with the core simulator SIMULATE, and contain data on a denser time grid compared to the "simplified history" as used before. Additionally, this information was validated over many years and a large amount of confidence was built over time. A method, called "BOHR", was then developed to extract such irradiation history from SIMULATE and to produce CASMO input files with the full cycle information using fine burn-up steps for each assembly, node and considered cycles. Therefore, all irradiated samples in a Swiss power plant can now be re-analyzed with an irradiation history directly coming from the core simulator. An example is presented in Figure 1 (right).

The use of these data helps to be closer to a realistic irradiation history and will lead to different assembly burn-up, rod burn-up, node burn-up (a fuel assembly is divided in 40 vertical sectors, called nodes) and finally different sample burn-up values. Such irradiation history does not exempt from the final stage which is the adjustment of the burn-up steps with the use of the DEP parameter to match the ^{148}Nd content. With such detailed irradiation history, the calculation over experiment ratio (C/E) for all isotopes is expected to be improved compared to the ones obtained from the simplified irradiation history. As presented later, the improvement is related to the experimental and calculated uncertainties: the goal being that the C/E for ^{148}Nd is within the calculated and experimental uncertainties. Other criteria could have been chosen (such as the global agreement for all C/E and not only ^{148}Nd), and the impact of such choices can be analyzed in follow-up studies.

4. U1 and U2 SAMPLES

Many experimental data followed by specific simulations exist in the open literature. As mentioned, two samples from the LWR-PROTEUS program (U1 and U2) are selected in this work. These samples were irradiated in the Swiss PWR Gösgen power plant and analyzed in the past at PSI. Table 1 presents these samples and their C/E values in the case of ^{244}Cm. The explanations for these values are given in the following subsections.

Table 1 – Summary of the results obtained in this work for ^{244}Cm and for the considered samples (U1 and U2). See text for the terms "simplified" and "detailed history".

Sample name	Enrichment fissile (wt %)	Burn-up			^{244}Cm C/E-1	
		simplified history (MWd/kgU)	detailed history (MWd/kgU)	variation %	simplified history (%)	detailed history (%)
U1	4.1	37.6	35.8	-5	+25	-3.4
U2	3.5	54.3	52.3	-4	+14	-0.7

As a preliminary remark, it is of prime importance in the case of ^{244}Cm to use a realistic irradiation history with a trusted set of irradiated samples. As indicated in Ref. [7], the inventory for ^{244}Cm is extremely sensitive to the irradiation history, as it is built-up after many neutron captures.

4.1 U2

This sample was irradiated at Gösgen (KKG) for 3 cycles. Samples of 200 mm length were cut from these rods for reactivity worth measurements in the PROTEUS facility, and small samples from adjacent axial locations were used for the chemical assays. The chemical measurements for the Cm isotopes were realized 3575 days after the end of the irradiation (9.79 years). For comparison, the half-life of ^{244}Cm is 18.1 years.

As explained in section 3., two different models for the irradiation history are available: (1) based on Ref. [7] (simplified) and (2) based on the power plant history (detailed). In the first case, the irradiation data originate from Framatome-ANP and are based on pre-calculations of the KKG cycles, using Framatome three-dimensional core simulator and a reconstruction of pin-wise power and power distributions. These irradiation data are labeled "simplified history" in Table 1. Examples for the 4 major quantities (power density, moderator temperature, fuel temperature, boron concentration) are presented in Figure 1. The final C/E-1 values for all measured isotopes are presented in Figure 2 (see Ref. [9] for details on the experimental uncertainties).

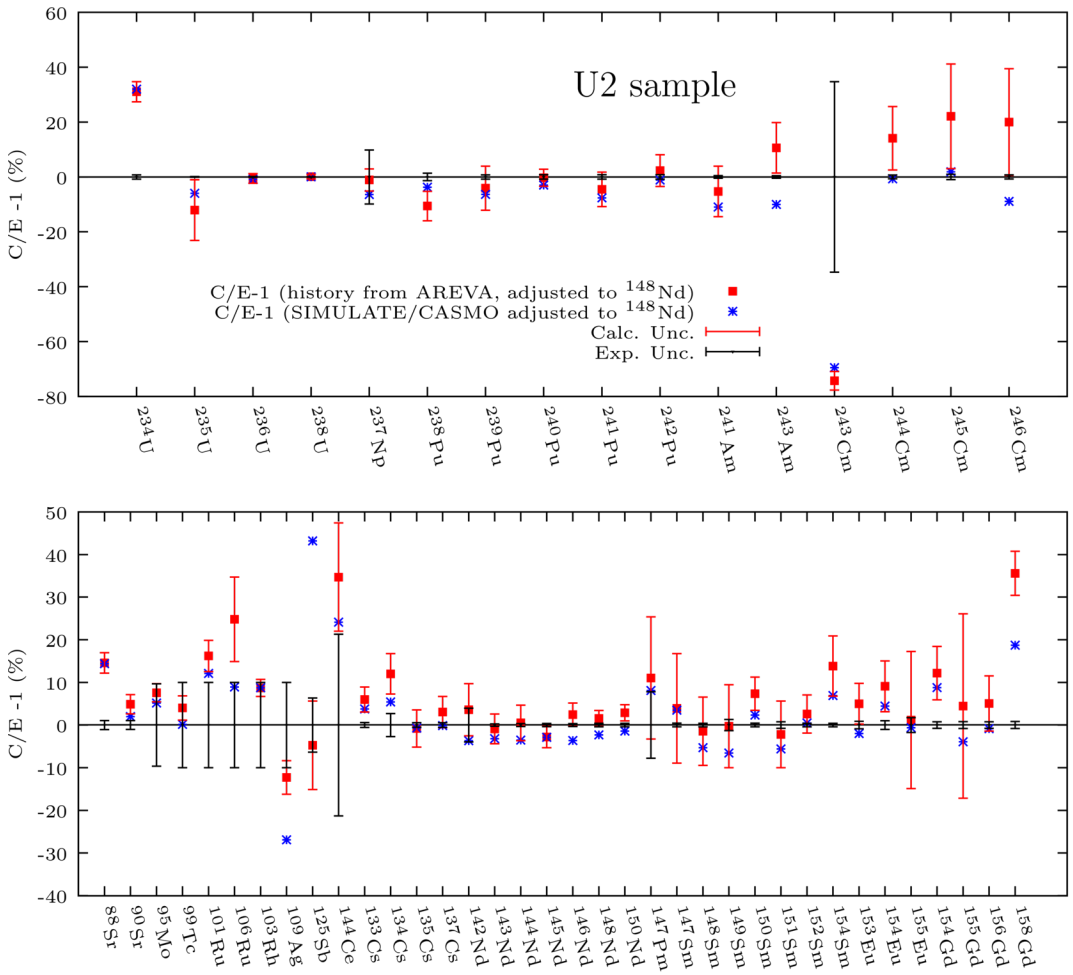

Figure 2 – C/E-1 for the U2 sample considering the two irradiation histories available (simplified and detailed). The calculated uncertainties (red) are presented in the next sections.

As observed, the C/E-1 values in the case of the detailed history are globally better. Some differences remain and a detailed analysis of the calculations and experimental values would be necessary. Nevertheless the ^{244}Cm C/E-1 value is greatly improved.

4.2 U1

As for the U2 sample, the U1 sample comes from the Gösgen nuclear power plant. The irradiation of the fuel rod lasted for 2 cycles up to a calculated sample burn-up of about 37 MWd/kgU. The procedure followed for this sample is similar to the one for the U2 sample, and the simplified and the detailed histories with the BOHR method are used with CASMO. For both the U1 and U2 samples, an improved C/E for ^{244}Cm (and globally the other isotopes) is obtained with a reduction of the calculated sample burn-up, as too high amounts of ^{244}Cm were originally calculated compared to the measured values.

5. UNCERTAINTY ON ISOTOPIC INVENTORY

Following a better assessment of the irradiation history, the uncertainties from the calculation method and its inputs can be evaluated. Depending on the sample and the knowledge of the parameters used in the calculations, different assumptions can be made. In this section, details for the uncertainty calculations are presented, together with the results. These uncertainties are already presented in Figure 2. The method applied for all the different inputs is based on Monte Carlo sampling and repeating many times the same calculation, with the varied inputs. About 300 samples were performed, and the quoted uncertainties refer to the standard deviation of the obtained distributions. A summary table is presented below (Table 2) in the case of ^{244}Cm , with explanations in the subsequent sections. Additionally, the uncertainty propagation procedure was applied in the case of the simplified irradiation history in order to save calculation time. A similar method can nevertheless be applied with the detailed irradiation history.

Table 2 – Uncertainties in % on the ^{244}Cm concentration at the time of the PIE measurements, due to specific inputs. The total uncertainties are calculated as a simple quadratic sum with correlation terms.

Sample name	Nuclear data	Operating conditions	Manufacturing tolerances	Burn-up induced	Total
U1	8.6	3.8	2.7	5.0	10.2
U2	9.6	2.1	1.6	2.5	11.1

These uncertainties depend on the assumptions made for many CASMO input quantities and are presented in Ref. [6]. The value of 10.2 % is the one reported in Figure 2 together with the U2 C/E value as vertical uncertainty. Details are given below.
It is emphasized here that apart from nuclear data, many unknowns still exist for the probability density functions of the other sources of uncertainties, as the literature is very scarce on this subject.

5.1 Nuclear data

Nuclear data such as microscopic cross sections or emitted spectra are now routinely considered by uncertainty propagation methods. Different possibilities exist, with their advantages, drawbacks and conditions of applicability. In the present case, the SHARK-X methodology [10] is used, based on nuclear data covariances included in libraries using the ENDF-6 format. The ENDF-6 format covariance libraries are first processed into the COVERX format using NJOY99.396 [11]. NJOY is used to reconstruct cross sections based on the ENDF-6 formatted files, perform Doppler broadening and calculate group-averaged cross sections. The 44-energy-group structure is used in NJOY and then converted to 19 groups for CASMO-5 with a module of SHARK-X.

All the major isotopes are considered with the most important cross sections (elastic, inelastic, capture, fission) and emitted spectra (neutron spectra and prompt fission neutrons). The origin of the covariance matrices is the ENDF/B-VII.1 library. A list is given below:

- Major actinides: ^{235,238}U and ^{239}Pu,

- Light elements: ^{1}H, ^{10,11}B, ^{16}O and ^{nat}C,
- Structural materials: $^{28-30}$Si, ^{59}Co,^{58}Ni, ^{54,56,57}Fe, ^{50,52,53}Cr, ^{90,91,92,94,96}Zr,
- Heavy isotopes: $^{152,154-158,160}$Gd, ^{182,183,184,186}W,
- Minor actinides: ^{234,236}U, $^{235-239}$Np, $^{236,238,240-242,244}$Pu, ^{241,243}Am, $^{242-244}$Cm,
- Fission products: ^{95,98,100}Mo, ^{99}Tc, $^{101-103,106}$Ru, ^{109}Ag, ^{127,129}I, ^{131,134}Xe, ^{133,135}Cs, ^{135}La, ^{139}Ce, ^{141,143,145}Nd, ^{148}Pm, ^{153,155}Eu.
- Fission Yields: In the case of the fission yields, two sources of simple covariance information exist in the ENDF/B-VII.1 and JEFF-3.2 library, without correlation terms between the fission products. In this work, the covariances from JEFF-3.2 are used, together with the option of "advanced normalization" in SHARK-X which uses simple conservation rules, leading to some correlation values between fission products (see Refs. [12, 13] for more details). In total, the fission yields for 13 actinides are randomly changed: $^{234-236,238}$U, $^{239-242}$Pu, ^{241,243}Am and $^{243-245}$Cm.

It is worth mentioning that the thermal scattering data are not considered, neither the decay data.

5.2 Operating conditions

In this work, the operating conditions are randomly modified with a constant relative uncertainty for all cycles until the end of irradiation. It does not take into account the burnup induced variations on these quantities. This is an important approximation, as the uncertainty on the power and other quantities will vary with increasing burn-up. For these quantities (fuel and moderator temperatures, reactor pressure, boron concentration, power and moderator density) and based on the literature data, the uncertainties presented in Table 3 are used in this work.

Table 3 – Uncertainties and probability density functions for the operating conditions considered in this work ("U" means uniform and "N" normal probability density function). σ is the standard deviation (uncertainty) of the distribution.

	Fuel Temp.	Moderator Temp.	Reactor Pressure	Boron Conc.	Irradiation history
CASMO keywords	TFU	TMO	PRE	BOR	DEP
Standard deviation (σ)	2 % U	2 % U	1 % U	2 % U	1 % U

The power (PDE) and the moderator density (MOD) are not independently varied within CASMO, but are fully correlated with the fuel and moderator temperatures
Depending on how many times one of these keywords is defined in the CASMO input file, the parameter can be changed at different burn-up steps. For instance, "PRE" is defined only once at the beginning of the calculation and is therefore changed (once) for the complete burn-up scheme. "TFU", "TMO" and "BOR" are defined for 13 different groups of burn-up steps and are therefore changed 13 times for one calculation (from 0 MWd/kgU to the end of cooling time), but the

changes are random only at the beginning of each cycle. In between, the same relative variation is applied compared to the beginning of cycle.

The power PDE is defined for many burn-up steps, but is changed in correlation with the fuel temperature and the moderator temperature. In Ref. [14], the correlations between the power and TFU and TMO is obtained from irradiation histories of different samples. Strong correlation (larger than $\rho = 0.5$) are reported. In this work, correlations for (PDE,TFU) and (PDE,TMO) are taken equal to 1.

5.3 Manufacture tolerances

Similar to the operating conditions, the manufacturing tolerances for the characteristics of the pin cells and the assembly are randomly modified, see Table 4).

Table 4 – Uncertainties and probability density functions for the 7 manufacturing conditions considered in this work ("U" means uniform and "N" normal probability density function).

	Pin radius	Fuel pin position shift	Guide thimble shift	Fuel density	^{235}U enrich.	^{234}U enrich.	^{238}U enrich.
CASMO keywords	R	LDX/Y	LDX/Y	FUE D	W5	W4	W8
	0.5 % U	1 mm	0.2 mm	1.5 % U	0.2 % U	correlated with W5	correlated with W5

5.4 Burnup induced Technological Changes

During the burn-up of the assemblies in a reactor, the mechanical material and structural characteristics of the fuel rod pellet/gap/cladding will evolve due to several phenomena including *e.g.* densification, swelling, fission gas release, clad creep-down/creep out, oxidation, hydrogen uptake etc. All these phenomena will thus introduce sources of uncertainties in the CASMO calculations that should be accounted for. Because the quantification of these uncertainty sources is perhaps one of the most challenging task, only a limited number of uncertainties in relation to geometrical characteristics are considered here:

- Fuel Pin position shift: 0.2 mm/burn-up groups (uniform),
- Moderator Pin position shift: 0.05 mm/burn-up groups (uniform),
- Pellet diameter increase: 150 μm at 53 GWd/t.

The bowing effect (or assembly deformation) has been widely observed in many commercial PWR. It is an effect which affects the control rod insertion and bring handling difficulties. Measurements of the bow indicated effects up to almost 10-15 mm. It is affecting the calculated quantities such as the number densities and needs to be taken into account. In CASMO, one possibility is to use the MxN card for 2D calculations, which allows to simulate a number of assemblies together and to change some of their characteristics during the burn-up steps (such as the position of the central

assembly if a 3x3 geometry is considered). Two calculations are performed (with burnup): one without the central assembly being shifted, and a second one with a shifted central assembly. In this work, a displacement of 2 mm is considered, affecting the ^{244}Cm concentration by about 2.5 %. Recent studies can be found (see for instance Ref. [15]) with more advanced methods showing the evolution of the impact on the ^{244}Cm concentration at the function of the burnup.

6. RESULTS

Taking into accounts all the sources of uncertainties and bias, it is possible to calculate the total uncertainties for the number densities and other quantities. The total uncertainties are assumed to be the quadratic sum of all the partial uncertainties (without correlation between different uncertainties). This method of obtaining the total uncertainties is based on the analysis of individual components (differential uncertainty quantification). An alternative approach is to randomly change all parameters together (integral uncertainty quantification). In a complete uncertainty quantification work, both methods would be applied.

In the case of the U2 sample, the total uncertainties for many isotopes are presented in Fig. 2. In the case of ^{244}Cm, the detailed uncertainties are presented in Table 2. It can be observed that these uncertainties are relatively large (about 10 %) at the time of measurements, many times larger than the reported measured uncertainty. This indicates the importance of presenting C/E ratios with uncertainties coming from the simulation procedure and inputs. As mentioned in many references, the nuclear data play an important role in this uncertainty quantification, showing that a better knowledge for specific cross sections would decrease the uncertainties on the C/E for isotopic compositions.

A second important point is presented in Table 1. It can be observed that the C/E for ^{244}Cm (and generally for other isotopes) is greatly improved by considering a detailed irradiation history. The simplified approach which uses only 4 variations of the important parameters during a cycle leads to large differences between C and E, larger than the calculated and experimental uncertainties. Such observation is statistically significant only once the calculated uncertainties are known.

To conclude, the results for both U1 and U2 samples are summarized as follows for ^{244}Cm:

- Original results: C/E-1(U1)= +25 % and C/E-1(U2)= +14 %,
- Best estimate plus uncertainties: C/E-1(U1)= -3.4 % with a calculated uncertainty of 10.2 %; and C/E-1(U2)= -0.7 % with a calculated uncertainty of 11.1 %,

indicating that the C/E values do not show significant disagreement between the measurements and calculations ^{244}Cm concentrations, given the calculated uncertainties.

7. CONCLUSION

In this work, it is demonstrated that the effect of the irradiation history is rather important when comparing calculated isotopic number densities with PIE measurements. An improved agreement is obtained with a detailed irradiation history compared to a simplistic one: the ^{244}Cm $C/E-1$

is reduced by a factor 7 and 20 for U1 and U2 samples, respectively. Additionally, a method to estimate the uncertainties for isotopic number densities is proposed, taking into account various sources of calculation assumptions and inputs. In the case of ^{244}Cm, the uncertainties on the calculated isotopic number densities at the time of the PIE measurements are about 10 % for both studied samples, covering the previous $C/E-1$ values obtained with the detailed irradiation history. This study of ^{244}Cm isotopic number densities for two LWR-PROTEUS samples is therefore a relevant example of best estimate calculation (with the improved irradiation history) plus uncertainties, allowing to go from apparent discrepancies between calculations and measurements to statistically compatible results.

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