Sensitivity/uncertainty analysis of the validation of the EVOLCODE 2.0 burn-up system with PWR experimental data

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Abstract

The prediction of the isotopic composition of irradiated nuclear fuels is crucial for studies on burn-up credit, reactor physics and waste management. With this motivation, CIEMAT has been developing, upgrading and validating the burn-up simulation system EVOLCODE 2.0 over the last decade.

One of these validations involved the ICE experiment, a whole irradiation of some UO_2 fuel assemblies in a pressurised light-water reactor (PWR) for a total of around 30 GWd/tU and the later isotopic content experimental measure of U and Pu isotopes. The validation of EVOLCODE 2.0, consisting in the simulation of the PWR with a single pin model, provided very satisfactory results (with mass deviations between the simulation and the experimental data smaller than 3% for the main U and Pu isotopes), with generally smaller mass deviations than those obtained with the deterministic code KAPROS. However, for some actinides the obtained mass deviations are larger than the experimental uncertainties.

The aim of this work is to analyse the simulation uncertainties in the final isotope masses and to comprehend their possible sources. For this reason, an uncertainty analysis has been performed based on the sensitivity methodology, providing the uncertainties in the isotopic content propagated from the cross-sections uncertainties, and the energy ranges and isotopes responsible for the major contributions to these uncertainties as well. The COMMARA data library has provided us with both the covariance information and the cross-section uncertainties in 33 energy groups.

Preliminary results show that the cross-section uncertainties are an important contributor to the simulation uncertainties, sometimes even larger than the experimental and the simulation uncertainties. Besides, some simulation uncertainties for actinides with a very small amount are significantly larger than the other uncertainties and need further investigation.

This work has been carried out within the framework of the ANDES Project (Accurate Nuclear Data for nuclear Energy Sustainability, 7th Framework Programme of the European Union). Its results will allow us to give some guidelines in order to advise additional strategies for cross-section uncertainty reduction.

Introduction

Computer codes with the burn-up capability to simulate irradiation history have been extensively used, on the one hand, for understanding the behaviour of current reactors fuel pins under irradiation and, on the other hand, for evaluating the properties of advanced reactors still without worldwide implementation. These computer codes are of especial interest when dealing with high burn-up simulations achievable in the previous nuclear concepts for economic reasons (extending the cycle length) or for reactivity, safety or waste management issues.

The last generation simulation system EVOLCODE 2.0 was developed at CIEMAT [0] with the aim of providing a computer code capable of making simulations of current and future reactors in any range of operation and to provide detailed spatial distribution and time evolution of the isotopic composition of fuels and activated materials. In particular, the capability of making simulations of isotopic evolution in the fuel for nuclear systems with very diverse characteristics and reaching long fuel burn-ups was focused.

In previous works, the burn-up capabilities of this code were tested in international code benchmarks mainly for fast neutron systems. However, recently a set of experimental measurements on a pressurised light water reactors (PWR) irradiation has become available. This experiment is the isotopic correlation experiment (ICE) [0], and is intended for the measurement of some actinides and fission products generated after burn-up.

The ICE experimental measurements have been used by different institutions to validate several burn-up codes (with a generally high degree of success) [0-0], which is the case with MCNPX with the integration of CINDER [0], the deterministic code KAPROS and its burn-up module KARBUS [0] or preliminarily with EVOLCODE 2.0. Nevertheless, in these studies only a direct comparison between experimental and simulation data was performed without analysing the sources of the deviations.

In this paper, a sensitivity/uncertainty analysis (S/U) was performed to explore whether the deviations between the experimental data and the simulations results were caused by the propagation of the cross-section uncertainties or other uncertainty sources should be investigated. A summary of the main results found from the validation of EVOLCODE 2.0 using the ICE measurements were also included.

This work was performed within the framework of the ANDES Project (Accurate Nuclear Data for nuclear Energy Sustainability, 7th Framework Programme of the European Union). Its results provide some guidelines in order to advise additional strategies for cross-section uncertainty reduction.

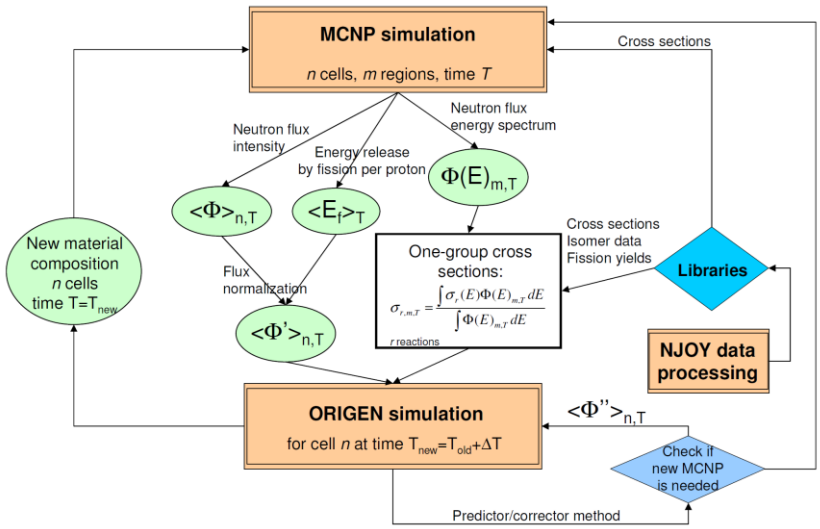
Computational tool

EVOLCODE 2.0 is an in-house development to solve the burn-up problem, that is, the coupled problem of neutron transport and isotopic evolution. Figure 1 shows its cycle data flow.

The neutron transport stage is solved by the MCNPX code [0], allowing an important degree of the heterogeneity description in the reactor core model. The isotope evolution stage is solved by the ORIGEN code [0]. The user selects some geometrical regions or cells inside which neutronic properties and material composition are considered constant. For each of these cells, EVOLCODE 2.0 creates one-group effective cross-section libraries, using the neutron flux energy spectrum provided by MCNPX in such a way that the whole calculation is faster than allowing MCNPX to achieve the desired result. Then, one ORIGEN execution is made independently for each cell providing the isotopic evolution of

the materials. This procedure is repeated for several successive irradiation steps (or cycles) to reach the total burn-up specified by the user.

Figure 1: EVOLCODE2 cycle data flow scheme



Sensitivity/Uncertainty analysis

The principles of sensitivity/uncertainty (S/U) analysis are well known and documented and have been applied to studying the impact of the nuclear data uncertainties on performance parameters such as the irradiated fuel isotopic composition [0,0]. Some information is summarised as a starting point of the analysis. Let $\vec{X}(t) = (X_1(t), X_2(t), ..., X_n(t))$ be the nuclide composition of a material at time t. The net balances of each isotope X_k can be described by the Bateman Equations which expressed in a matrix notation are:

$$\frac{d\vec{X}}{dt} = \mathbf{B}\Phi\vec{X}, \qquad (1)$$

where **B** is the matrix involving the cross-sections and decay values), Φ is the neutron flux and t is the irradiation time. The solution of this equation, for constant flux and effective cross-sections, is:

$$\vec{X}(t) = e^{\mathbf{B}\Phi t}\vec{X}(0). \qquad (2)$$

This work refers to only a small group of *q* nuclides. The evolution of these isotopes depends only on a particular set of cross-sections that can be grouped as $\vec{\sigma} = (\sigma_1, \sigma_2, ..., \sigma_m)$, considering isotopes, and reactions. It should be noted that the other parameters in the problem (other cross sections, decay values,) are known and constant, and that the analysis is made for a period of time between 0 and t. Each element X_k is hence a function of the cross-section vector $\vec{\sigma}$, $X_k = X_k(\vec{\sigma})$, where t will no longer be included for simplicity. The goal is then to analyse how the uncertainties in the cross-sections are transmitted to the nuclide composition $\vec{X}$.

The uncertainty in $\vec{X}$ due to the uncertainties in $\vec{\sigma}$ can be expressed as $\Delta X_k = \sum_{j=1}^{m} S_{kj} \Delta\sigma_j^l$, where the sensitivity coefficients S_{kj} are given by $S_{kj} = \frac{\partial X_k}{\partial \sigma_j^l}$. In these expressions, a superscript l has been introduced, which takes into account that nuclear data uncertainties $\Delta\sigma_j^l$ are usually provided only for a small amount of G energy groups. In order to state the dependence of the sensitivity coefficient on the one-group cross-section, it is considered that $\sigma_j^{eff} = \sum_{l=1}^{G} \sigma_j^l \Phi^l \Big/ \sum_{l=1}^{G} \Phi^l$ so

$$S_{kj} = \frac{\partial X_k}{\partial \sigma_j^l} = \frac{\partial X_k}{\partial \sigma_j} \frac{\Phi^l}{\sum_{l=1}^{G} \Phi^l}. \tag{3}$$

Furthermore, the cross-sections corresponding to different energy groups are usually correlated and even the cross-sections of different reactions are correlated in some cases. Taking into account these correlations, the uncertainties can be represented in the form of a variance-covariance matrix,

$$C = \begin{pmatrix} c_{11} & c_{12} & \dots & c_{1m} \\ c_{21} & c_{22} & \dots & c_{2m} \\ \dots & \dots & \dots & \dots \\ c_{m1} & c_{m2} & \dots & c_{mm} \end{pmatrix}, \tag{4}$$

where each c_{ij} represents a GxG matrix accounting for the G energy groups.

Once the sensitivity coefficients and the uncertainties in the variance-covariance matrix are known, the variance of the nuclide composition $\vec{X}$ can be evaluated as follows:

$$\Delta^2 X = S^T C S \tag{5}$$

The dependence of X_k on the cross-sections comes, on the one hand, from their explicit appearance in the solution of the Bateman Equations for isotope k and, on the other hand, from the implicit dependence of the neutron flux on the cross-sections. The implicit dependence has been usually neglected in previous works related to uncertainties propagation in fast systems [10] [12] considering that the feedback from flux and spectrum changes along irradiation is very low. In this work, the same hypothesis was made, although the possibility that the implicit dependence of the neutron flux normalisation could significantly impact the final uncertainty of some isotopes should be tested.

The isotope correlation experiment

The European Safeguards Research and Development Association (ESARDA) working group on Isotopic Correlation Techniques (ICT) and Reprocessing Input Analyses performed in the 1970s an experiment to check the feasibility of the ICT. This experiment was developed at the Obrigheim Nuclear Power Plant and was called the Isotopic Correlation Experiment (ICE) [2].

The objectives of this experiment were the determination of the accuracy of the measurement technique, the identification of additional required information and

safeguard-related issues. In order to achieve these objectives, several fuel assemblies were chosen from the spent fuel after the normal operation of the Obrigheim Power Plant (KWO).

The experiment was conducted at normal operation of the plant. Ten consecutive dissolution batches, each making up half of the fuel assembly, were chosen for the experiment. The geometrical data of the pins used for the experiment were obtained from the bibliography [13]. The UO_2 pin (with a stoichiometry of 2.00 ± 0.01%) had an external radius of 0.465 cm. Its average operation temperature was 1 028 K. The cladding material was zirconium with an outer radius of 0.535 cm and an average operation temperature of 605 K. The moderator was water at a temperature of 572 K averaged over the fuel channel.

The uranium content (approximately 311 kg of UO_2 per assembly with an uncertainty of 0.4%) was supplied by KWO, as well as the irradiation history and the boron concentration in water [13].

Post-irradiation analyses performed at the reprocessing plant WAK at Karlsruhe included the measurement of the concentration of uranium isotopes (^{235}U, ^{236}U and ^{238}U), plutonium isotopes (^{238}Pu to ^{242}Pu), some fission products and minor actinides (Am, Cm). ^{148}Nd was used as an indicator of the burn-up. There were no measurements of head-end losses.

ICE simulation details

The main geometrical and thermal hydraulic settings used by EVOLCODE 2.0 to simulate ICE are included in Table 3 [2] [4] [13]. The simulation model consisted in a pin design of an Obrigheim 14x14 PWR, with reflecting boundary surfaces. The fuel pin was divided radially into 25 evolving cells with different thicknesses to take into account the larger creation of plutonium at the pin periphery (shown in the previous sections). No axial divisions of the pin were implemented.

Table 1: ICE simulation settings

Parameter	Value
Pin radius	0.465 cm
Cladding thickness	0.07 cm
Pin side length	1.498 cm
Pin height	1 cm
Fuel density	0.0681 at/cm·barn
Cladding density	0.0432 at/cm·barn
Moderator density	0.0721 at/cm·barn
Fuel temperature	1 200 K
Cladding temperature	600 K
Water temperature	600 K
Initial U-235 enrichment	3.1%

The pin is surrounded by a Zr cladding. The moderator is water. Thermal S(α,β) tables for hydrogen in light water at 600 K were used in the moderator. The volume ratio of the moderator to fuel was set to $V_m/V_f = 1.978$.

A total of 53 EVOLCODE cycles were performed for an adequate simulation of the ICE experimental irradiation history, including 11 cycles without irradiation (decay only). MCNPX version 2.7.a was used with the JEFF3.1.1 library.

For the S/U analysis, the COMMARA-2.0 neutron cross-section covariance library [14] was used including the information of the relative uncertainty in the cross-sections and the correlations (for the 33 energy groups from 10^{-5} eV to 19.6 MeV, obtained by processing with code NJOY using 1/E flux) for the following isotopes and reactions: ^{235}U (n,f), ^{235}U (n,γ), ^{235}U fission-capture cross-correlations, ^{236}U (n,γ), ^{238}U (n,γ), ^{238}Pu (n,γ), ^{238}Pu (n,f), ^{239}Pu (n,f), ^{239}Pu (n,γ), ^{239}Pu fission-capture cross-correlations, ^{240}Pu (n,γ), ^{241}Pu (n,γ), ^{241}Pu (n,f) and ^{237}Np (n,f). The neutron flux energy spectrum and the reaction rates for the 33 energy groups were obtained by MCNPX.

Results

This section includes the validation results of the implementation of EVOLCODE 2.0 for the ICE experiment simulation and the S/U analysis. Isotopic composition results are shown as a comparison of the number density (atoms per initial metal atoms, IMA) of each isotope with experimental data. The evolution with burn-up of the number density of the isotopes of interest can be found in Figure 2 to Figure 5. For this validation, the burn-up was estimated by means of the ASTM standard procedure [15], using ^{148}Nd as a burn-up indicator.

Figure 2: Comparison of ^{235}U (left) and ^{238}U (right) number densities obtained with EVOLCODE 2.0 and experimental data as a function of burn-up

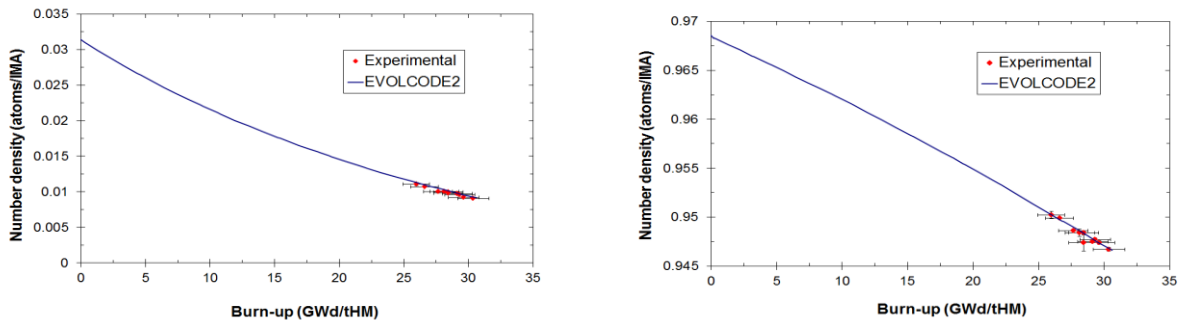


Figure 3: Comparison of ^{236}U (left) and ^{238}Pu (right) number densities obtained with EVOLCODE 2.0 and experimental data as a function of burn-up

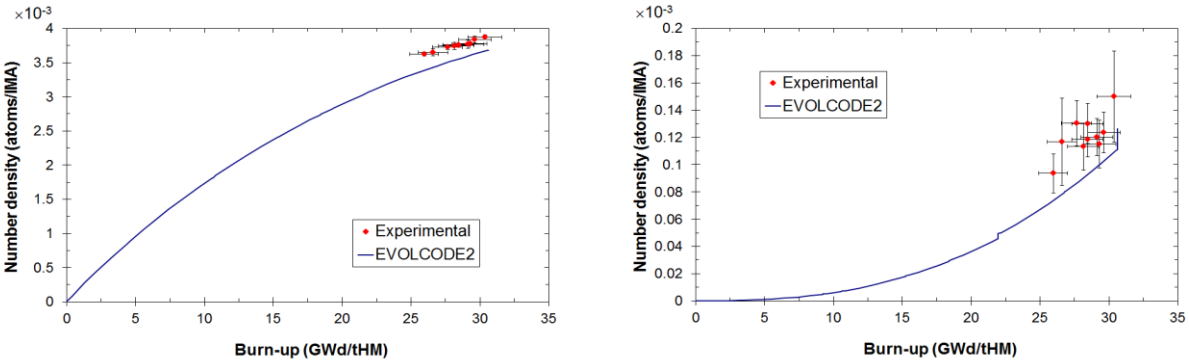


Figure 4: Comparison of ^{239}Pu (left) and ^{240}Pu (right) number densities obtained with EVOLCODE 2.0 and experimental data as a function of burn-up

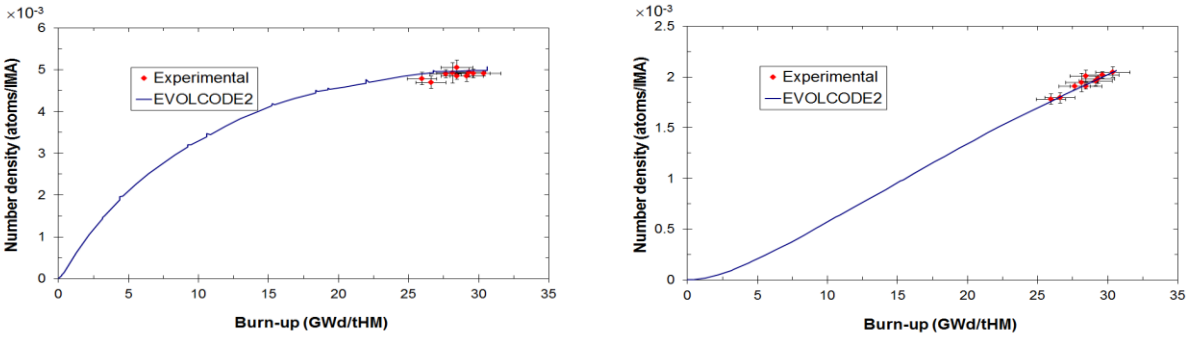
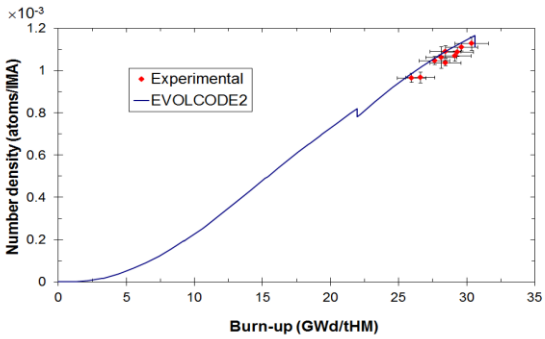


Figure 5: Comparison of ^{241}Pu number densities obtained with EVOLCODE 2.0 and experimental data as a function of burn-up



Sharp decreases (mainly seen at 22 GWd/tHM) correspond to the decay of ^{241}Pu in periods of radioactive decay only.

The experimental measurements of the burn-up values were determined radiochemically and involved an inaccuracy of ±(3-5)% [16]. Due to this, we assigned a 4% error bar to the experimental burn-up points when comparing the experiment with the numerical results in the figures. In addition, experimental data were obtained as averaged values coming from measurements made by four different institutions [2]. The variation coefficients of the averaged data were calculated by means of the variance and are also provided in this reference. These values were taken as uncertainties in the experimental number density.

It should be noted that, for reasons of comparison, the experimental results were corrected to the date of the reactor shut-down. This is the case with ^{241}Pu, ^{238}Pu, ^{242}Cm and some (unidentified) fission products. In-pile decay corrections, however, were not applied). For this reason, we will neglect the final 53rd cycle (365 days of cooling decay) in our analysis.The preliminary results are shown in Table 2 for the actinides of our study. The second column of this table shows the uncertainty of the experimental data for these nuclides, calculated as an average of the variation coefficients described above and taken as uncertainties in the figures. The third column of this table shows the results of the S/U analysis and will be described in detail below. The fourth column contains the averaged deviation between the experimental data and the simulation with EVOLCODE 2.0.

The agreement between the simulation results and the experimental data is very good for the main isotopes, that is, for those with larger amounts (number density larger than 10^{-3}), with an averaged deviation smaller than 3%, although it can be seen that Pu isotopes generally have a smaller deviation than U isotopes.

For those isotopes with very small presence (^{236}U and ^{238}Pu), the averaged deviations between EVOLCODE 2.0 and the experimental data are significantly larger than the other uncertainties. The case of ^{236}U can be seen in Figure 3 (left). For this nuclide, deviations are around 5% beyond experimental uncertainties. However, our simulation provided results very similar to the numerical simulations obtained in the past with MCNPX/CINDER [6]. Regarding ^{238}Pu, a high dispersion in the experimental data can be seen in Figure 3 (right). Considering this, the deviation between the simulation and experimental results would range from 1.5 to 2 times the experimental data fluctuations (rms). Similar results could be found for this isotope in other simulations involving MCNPX/CINDER or KAPROS [7].

Table 1: Preliminary results of the application of the COMMARA covariance data to the ICE simulation with EVOLCODE 2.0

Isotope	Experimental data uncertinty	Uncertainty due to cross-sections	Averaged deviation between EVOLCODE and experiment
^{235}U	0.65%	0.35%	2.49%
^{236}U	0.92%	1.84%	5.45%
^{238}U	0.0276%	0.0283%	0.0228%
^{238}Pu	15.36%	6.01%	25.52%
^{239}Pu	2.65%	1.20%	2.22%
^{240}Pu	2.51%	1.94%	1.23%
^{241}Pu	2.44%	2.45%	2.80%

The third column of Table 2 shows the uncertainties in the final isotopic composition propagated from the cross-sections. These uncertainties are generally smaller than the experimental data for the Pu isotopes (excepting ^{238}Pu), meaning that the simulation data discrepancies must be caused by the dispersions in the experimental measurements. The U isotopes show a different behaviour depending on the isotope. The averaged deviation between the simulated and the experimental data for ^{238}U is smaller than the other uncertainty sources. In contrast, for ^{235}U, the uncertainty propagated from the cross-section has a very low value, 0.35%, smaller than the experimental data uncertainty. This indicates that the deviation between the EVOLCODE results and the experimental data must be caused by other reasons, possibly the 4% experimental uncertainty in the burn-up, or an underestimation of the ^{235}U (n,γ) value and uncertainty. The other isotopes, ^{236}U and ^{238}Pu, have a larger averaged deviation between EVOLCODE and experiment. ^{236}U has a large simulation discrepancy that cannot be explained by the propagation of the cross-section uncertainties unless the ^{235}U (n,γ) error has been underestimated. According to [6], one of the possible factors inducing this discrepancy in the ^{236}U might be the inconsistency and inaccuracy in the ^{235}U capture cross-section databases in the energy range of 30 keV to 1 MeV above the resonance range, showing a 10% discrepancy in the capture cross-section between libraries. Nuclide ^{238}Pu has a large discrepancy but simulation and experimental data are compatible, taking into account their uncertainties.

Table 3 shows the contribution to the variance in the concentration of each actinide due to the uncertainty in each potentially relevant cross-section after burn-up. This result provides some guidelines in order to advise additional strategies for cross-section uncertainty reduction inside the ANDES Project. Moreover, the methodology applied in this work allows estimating the contribution of each energy group to the total uncertainty.

Table 2: Contribution to the variance in the main actinides due to uncertainties in each contributing cross-section

Variance (%) due to	Isotope						
	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu
^{235}U (n,f)	85.0	0	0	-0.07	0	0	0
^{235}U (n,γ)	15.0	91.6	0	1.72	0	0	0
^{236}U (n,γ)	0	8.4	0	14.1	0	0	0
^{238}U (n,γ)	0	0	100	0	101	19.1	59.6
^{237}Np (n,γ)	0	0	0	16.5	0	0	0
^{238}Pu (n,γ)	0	0	0	67.8	0	0	0
^{238}Pu (n,f)	0	0	0	0	0	0	0
^{239}Pu (n,f)	0	0	0	0	1.2	-1.9	-6.4
^{239}Pu (n,γ)	0	0	0	0	-2.2	2.8	11.6
^{240}Pu (n,γ)	0	0	0	0	0	79.9	7.5
^{241}Pu (n,γ)	0	0	0	0	0	0	5.60
^{241}Pu (n,f)	0	0	0	0	0	0	22.1

Conclusions

The ICE experiment was satisfactorily simulated, using the EVOLCODE 2.0 system as an on-going effort to validate this tool. Despite the very simple geometrical model of the reactor, the agreement between calculated and experimental data is excellent for actinides, with an accuracy of 3% or better for the main actinides of the study.

A sensitivity/uncertainty analysis of the impact of cross-section uncertainties on the isotopic composition uncertainties after burn-up was also performed, using the COMMARA 2.0 uncertainty data library. These uncertainties are generally smaller than the experimental data for the Pu isotopes with a larger amount, meaning that the simulation data deviations regarding the experimental data might be caused by the dispersions in the experimental measurements. Besides, for ^{235}U, the uncertainty propagated from the cross-section has a very low value, smaller than the experimental data uncertainty, indicating that the relatively small deviation between the EVOLCODE results and the experimental data must be caused by other reason, possibly the 4% experimental uncertainty in the burn-up or an underestimation in the ^{235}U (n,γ) value and uncertainty.

Although the S/U analysis indicates that the simulation with EVOLCODE 2.0 is rather satisfactory, this work is being carried out with the aim of estimating the contribution of each energy group to the final uncertainty. In addition, it should be noted that the implicit dependence of the neutron flux normalisation on the cross-sections is negligible. Estimating this implicit dependence and its possible impact on the uncertainties is also planned as future work.

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