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VALIDATION OF THE UNCERTAINTY PROPAGATION METHOD FOR THE DECAY HEAT WITHIN THE DARWIN2.3 PACKAGE

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ABSTRACT

The DARWIN2.3 package is dedicated to spent fuel characterization in general and to decay heat computations in particular, and benefits from a Verification, Validation and Uncertainty Quantification process (VV&UQ). The current experimental results available are insufficient to accurately assess the biases and uncertainties due to nuclear data for the decay heat for the fuel managements of the French reactor fleet. This is the reason why an uncertainty propagation method has been recently implemented within the CYRUS tool (a mockup for the DARWIN2.3 package), and later on within the DARWIN2.3 package in order to compute a priori uncertainty due to nuclear data variance/covariance matrices. The method implemented is the quadratic summation method, which allows in-depth analysis of the uncertainty through sensitivity and variance analysis. Nevertheless, this method relies on a fundamental hypothesis of linearity, which is not straightforward in the case of the decay heat. Moreover, due to the specificity of the DARWIN2.3 package, approximations have been made when dealing with cross-sections, such as the collapsing of the multigroup covariance matrices, and the neglecting of the Boltzmann/Bateman coupling when computing the sensitivity profiles. These three items have been studied in details. This paper shows some of the results that accredit the legitimacy of the linearity hypothesis and collapsing method, and provide some recommendations in dealing with the Boltzmann/Bateman coupling.

1. INTRODUCTION

The decay heat (DH) is the thermal heat emitted after the reactor shutdown. Its value is about 6.5% of the nominal power directly after shutdown [1], and still 1.5% of the nominal power one hour later, that is to say about 40 MW for a 900 $MW_é$ French Pressurized Water Reactor (PWR). Therefore it is of paramount importance to provide accurate calculation of this quantity, associated with controlled biases and uncertainties over the whole domain of application. For the French reactor fleet, this domain is bounded to PWR 17x17 uranium oxide fuels (UOX) (1.0 < ^{235}U enr. < 5.0 wt%) and mixed oxide fuels (MOX) (4.0 < Pu content < 11.0 wt%) with discharge burn-up going from 0 to 90 GWd/t and cooling times going from 0 second (reactor shutdown) to 100,000 years (repository times).

The decay heat is computed with the DARWIN2.3 package at the CEA [2]. The decay heat's computation with the DARWIN2.3 package has been experimentally validated by comparing the calculation results to the available integral measurements. The problem is that these experiments cover a very small part of the DA for PWR, and only deals with UOX fuels (see Figure 1):

- The French experiment MERCI [3] (**3.7wt%** ^{235}U, **3.5 GWd/t**) provides DH measurements from **45 minutes to 42 days**,
- the Swedish CLAB experiments [4] (**2.1 wt%** < e_{235U} < **3.4 wt%, 20 GWd/t** < burn-up < **47 GWd/t**) provide measurements from **13 years** up to **23 years** of cooling,
- the American experiments of General Electric [5] (**3.4 wt%** < e_{235U} < **4.0 wt%**, **26 GWd/t** < burn-up < **39 GWd/t**) and Handford Engineering Development Laboratory [6] (**e_{235U} =2.5 wt%, 25 GWd/t** < burn-up < **30 GWd/t**) provide integral measurements from **2 to 8 years**.

The preliminary work conducted in [7] accredits the fact that the corpus of experimental data currently available is insufficient to extrapolate the uncertainties outside the experimental domain in order to cover the whole application domain. A propagation method of input data uncertainties is required to fill the blanks in the meantime.

The quadratic summation method has been implemented in the CYRUS tool [8]. The CYRUS tool is a mockup for the DARWIN2.3 package. The method itself, and its implementation taking into account the characteristics of the DARWIN2.3 package, need to be validated in order to increase the reliability of the results produced. The goal of this paper is to identify the hypotheses and approximations made, and propose a protocol to justify them.

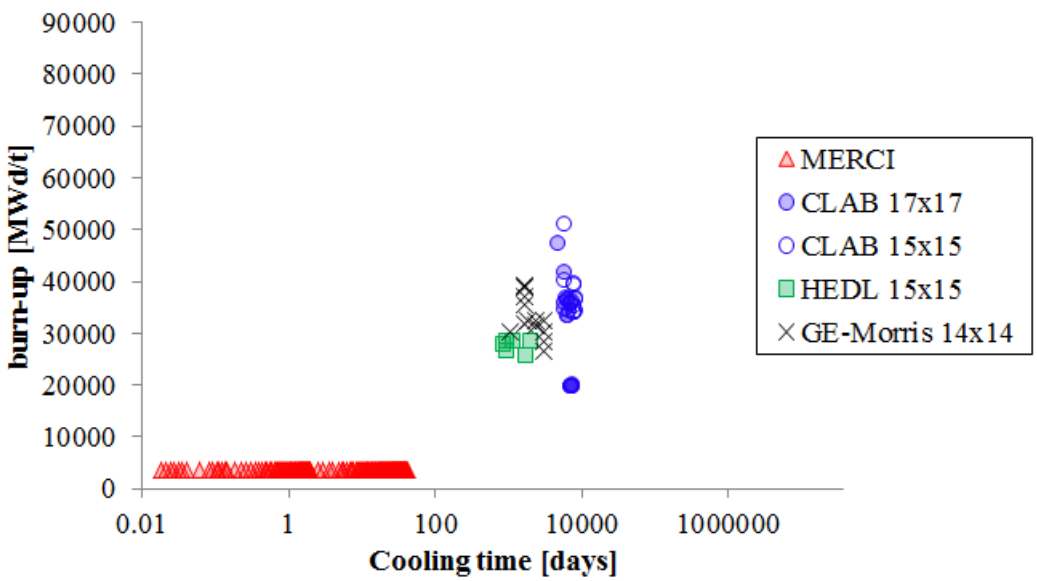

Figure 1 - Integral experiments of decay heat measurements

2. DESCRIPTION OF THE DARWIN2.3 PACKAGE FOR THE COMPUTATION OF THE DECAY HEAT OF PWR

The DARWIN2.3 package [9] [2] [10] is developed at the CEA (France) with support from its industrial partners EDF and AREVA. DARWIN2.3 is dedicated to spent fuel characterization from all kind of reactors. It computes the following physical quantities: fuel inventory, decay heat, activity, radiotoxicity, neutron, alpha, beta and gamma sources and spectra.

The DARWIN2.3 package involves neutron codes such as APOLLO2 (for PWR [11] [12] and BWR [13] calculations), ERANOS2 (for FNR [14] calculations), TRIPOLI4 [15] (Monte-Carlo) dedicated to the computation of self-shielded and collapsed cross sections and neutron spectra as a function of the burn-up of the fuel, and a depletion code PEPIN2, whose strength is to have almost complete filiation chains, describing more than 3800 radionuclides. Nuclear data libraries are associated with these codes, all based on the JEFF-3.1.1 European evaluation [16].

As this paper focuses on the uncertainty quantification of the decay heat of PWR fuels, only the calculation route of PWR fuels is going to be described here (see Figure 2).

In the PWR calculation route, the 2D lattice neutron code APOLLO2 is used [11] [17] in order to produce a SAPHYB file which gathers together the multigroup self-shielded cross-sections and neutron spectra as a function of the burn-up, for the studied fuel. The computation of the neutron data is done according to a calculation scheme called CYCLE2008, which has been optimized and validated for fuel cycle applications of PWR UOX and MOX fuels [2].

The PEPIN2 depletion code uses the SAPHYB file provided by APOLLO2 in order to produce a collapsed library with burn-up dependent cross-sections. This library is completed with cross-sections from JEFF-3.1.1 for the missing isotopes in the APOLLO2 filiation chains, and completed with decay data and fission yields data coming from the JEFF-3.1.1 library as well. A very precise depletion history can be given to the PEPIN2 code, with intra-cycles for instance; power variations and cooling periods (bore concentration, moderator and fuel temperatures tracking). The collapsed cross-sections and neutron flux are then interpolated according to this depletion history.

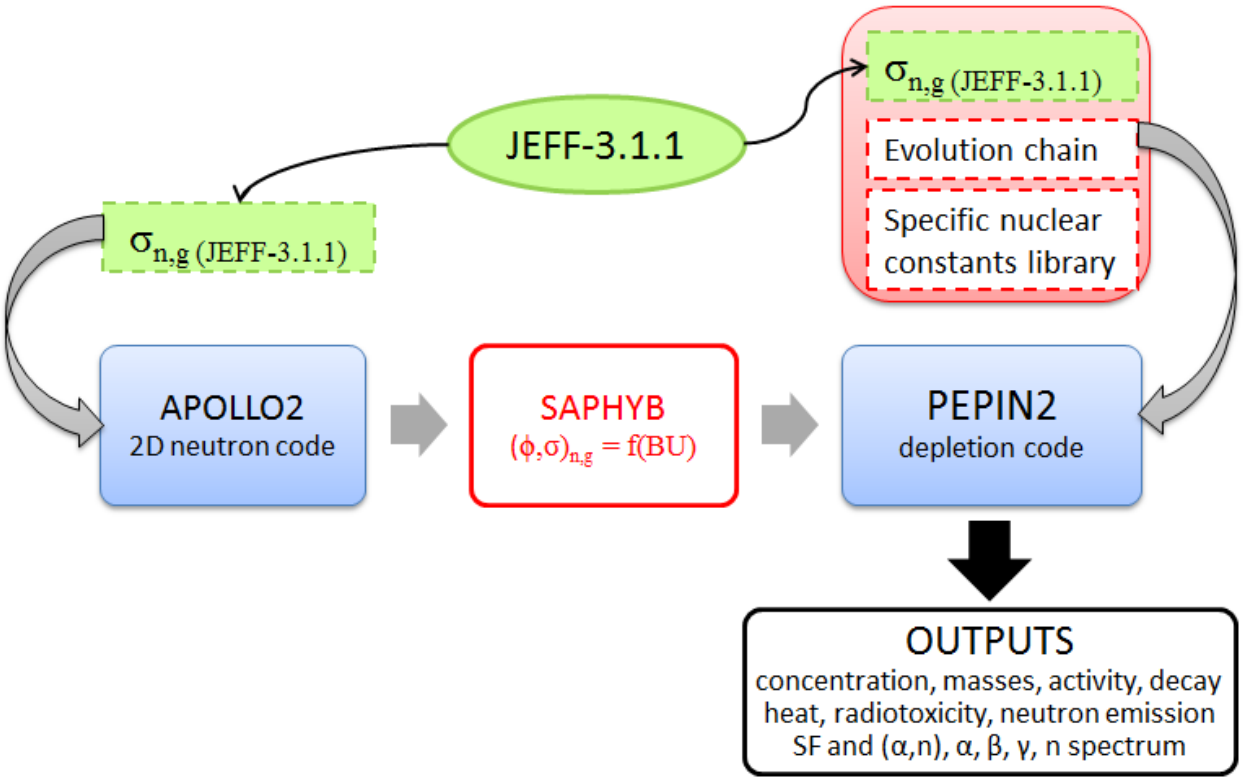

Figure 2 – The DARWIN2.3 package in the PWR calculation route

3. THE UNCERTAINTY PROPAGATION METHOD AND ITS IMPLEMENTATION WITHIN THE DARWIN2.3 PACKAGE

3.1 Choice of the uncertainty propagation method

The choice of the uncertainty propagation method depends on the following considerations: dimension of the input and output vectors, simulation time, linearity of the model and nature of the expected results.

For the particular case of the decay heat, the vector of outputs is of dimension 1, whereas the vector of input parameters is about 40,000. The calculation time (neutron computation + depletion calculation) is less than an hour, and the linearity of the output (the DH) to the inputs (nuclear data) is not straightforward. The expected results are the variance of the output, associated with a confidence interval, and sensitivity and variance analysis. The computation of the DH within the DARWIN2.3 package is performed in two steps: first, solving of the Boltzmann equation, and then solving of the Bateman equations with a constant neutron flux during each burn-up step of calculation.

Given that, the most appropriate method should be the adjoint method [18] [19] [20] [21] but this method cannot be used at the moment because the adjoint calculation of the fuel inventory is not available in the APOLLO2 code. Therefore the quadratic summation method has been implemented within the CYRUS tool [8], working with the DARWIN2.3 package.

3.2 Description of the quadratic summation method

The quadratic summation method is based on two results of probability theory which give the expected value (1) and the variance (2) of a linear function of uncertain parameters. If Y is a linear model of random variables $X_1, \dots, X_d$ and $a_1, \dots, a_d$ real numbers:

$$E(Y) = \sum_{i=1}^{d} a_i E(X_i) \tag{1}$$

$$var(Y) = \sum_{i=1}^{d} \sum_{j=1}^{d} a_i a_j cov(X_i, X_j) \tag{2}$$

The covariance terms $cov(X_i, X_j)$ are related to the linear correlation coefficients $\rho_{i,j}$and to the variances of the uncertain parameters according to the relationship (3):

$$cov(X_i, X_j) = \rho_{i,j} var(X_i) var(X_j) \tag{3}$$

Then the idea is to linearize the decay heat thanks to a Taylor series expansion at first order, as shown in (4), where $\overline{X_i}$ is the expected value of X_i and $[\partial DH/\partial X_i]_{X_i=\overline{X_i}}$ is the sensitivity coefficient of the DH to the random variable X_i.

$$DH = DH(\overline{X}) + \sum_{i=1}^{d} \left[\frac{\partial DH}{\partial X_i}\right]_{X=\overline{X}} (X_i - \overline{X_i}) + o(\|X_i - \overline{X_i}\|) \tag{4}$$

The use of the formula (2) to (4) gives the relative variance of the decay heat (6), which can be written in a matrix form as the product of a covariance matrix C with the sensitivity vector S (5). In the formula (6), the superscript T refers to the transpose. The matrix form is known as the "sandwich rule".

$$S = \left[\frac{\partial DH}{\partial X_i}\frac{X_i}{DH}\right]_{X=\bar{X}} \tag{5}$$

$$\text{rel_var(DH)} = \frac{var(DH)}{DH(\bar{X})^2} = \sqrt{S^T * C * S} \tag{6}$$

The quadratic summation method relies on the hypothesis of linearity of the decay heat to the nuclear data (cross-sections, decay data and fission yields data). If this hypothesis is true, then the computation of the sensitivity coefficients is given by a direct perturbation of the DH around the expected value of X_i, and besides, the sensitivity coefficients are independent from the perturbation rate applied.

3.3 Description of the covariance data

The accuracy of the calculation results depends on two kinds of input data uncertainties: nuclear data uncertainties and descriptive data. Descriptive data gathers together geometrical tolerances, uncertainties in the composition of the materials involved, uncertainty on operating parameters and timelines. In this paper, only nuclear data uncertainties are taken into account. Numerical biases and uncertainties are not taken into account.

The variance/covariance data associated with independent fission yields, decay periods, branching ratios and mean decay energies are read in the JEFF-3.1.1 library [16].

The covariance matrices associated with neutron cross-sections are taken from the COMAC database [22]. This database provides covariance matrices in a multigroup format (26 or 33 groups) for the main radionuclides involved in reactor physics and back end cycle.

3.4 Implementation of the uncertainty propagation method in the DARWIN2.3 package

The decay heat computation of a depleted fuel with the DARWIN2.3 package requires a two-steps calculation: first the neutron calculation of multigroup cross-sections and neutron spectra as a function of the burn-up (Boltzmann equation), and then the solving of the Bateman equations with collapsed cross-sections and neutron flux.

Therefore the implementation of the quadratic summation method in the DARWIN2.3 package requires two approximations that need to be studied in details. The multigroup covariance matrices associated with cross-sections have to be collapsed into one-group variances, and the Boltzmann/Bateman coupling is neglected when computing the sensitivity profiles of the decay heat to cross-sections.

4. VALIDATION OF THE HYPOTHESIS OF LINEARITY

The decay heat is only linear to the mean decay energies. The nuclide densities result from the coupled solving of the Boltzmann and Bateman equations, which are non-linear equations.

Therefore it is compulsory to check if the hypothesis of linearity is a reasonable approximation or not for the decay heat of PWR UOX and MOX fuels.

To do so, it has been proposed to compare the results obtained with the quadratic summation method to those produced with a different uncertainty propagation method that does not need a linearity hypothesis. This is the case of the sampling approach, implemented in the URANIE/MENDEL code system [23] [24]. URANIE is the uncertainty platform of the CEA and MENDEL is a depletion code similar to PEPIN2, reading the same SAPHYB input files, using the same libraries coming from JEFF-3.1.1 and with similar filiation chains.

The sampling approach consists in selecting distribution laws for each random input (more often, Gaussian laws, but sometimes, in order to respect the positiveness, truncated Gaussian laws or log-normal laws are used) and to sample them with a Latin Hypercube Sampling technique in order to have n realization of each variable. Then, the MENDEL code is called n times with n different sets of input data according to the results of the sampling step. Eventually, the distribution of the decay heat is build and the moments are extracted.

The comparison has been made for a PWR UOX fuel with 3.7wt% enriched uranium and a MOX fuel with a mean plutonium content of 9.5 wt%, at a discharge burn-up around 50 GWd/t. The results are summarized in Figure 3. Both methods lead to coherent values over the whole range of cooling times. These results substantiate the hypothesis of linearity.

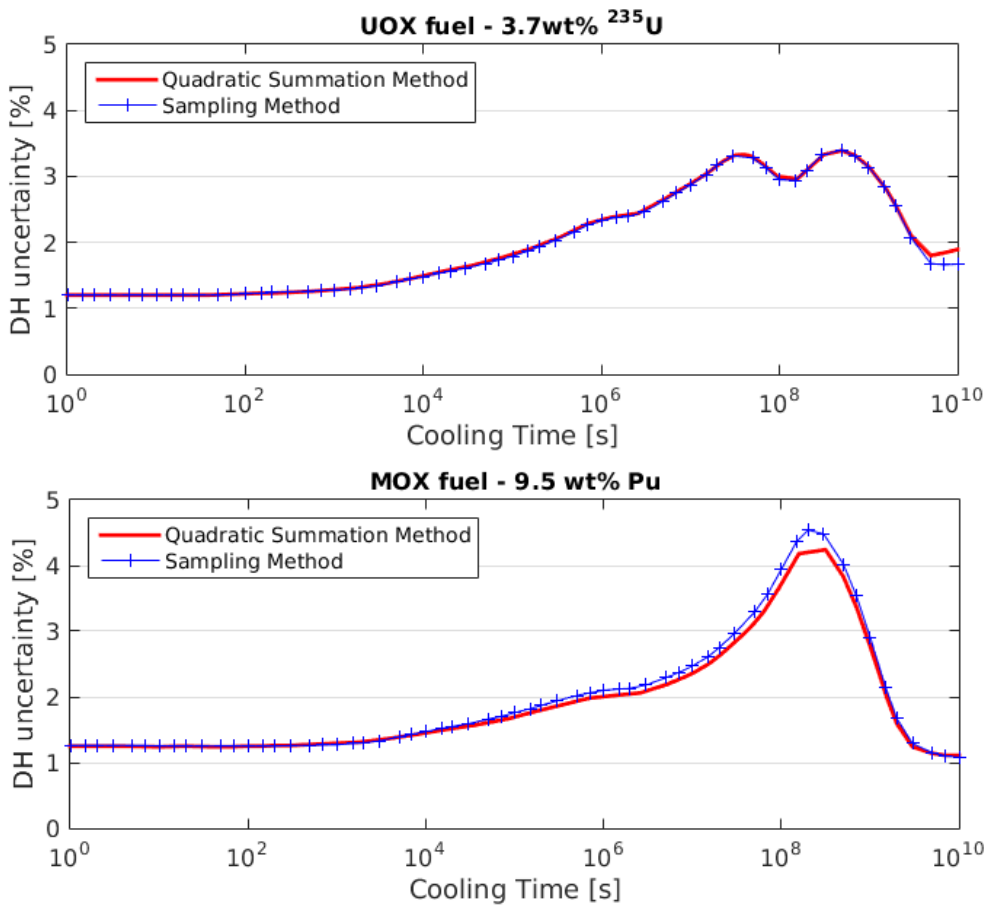

Figure 3 – Comparison of DH uncertainty results computed with the sampling method and the quadratic summation method

5. VALIDATION OF THE METHOD FOR COLLAPSING MULTIGROUP COVARIANCE MATRICES

5.1 Description of the collapsing method of multigroup covariance matrices

The method used to collapse multigroup covariance matrices for microscopic cross-sections is based on the conservation of the uncertainty, propagated in a deterministic way [25]. In other words, for a given isotope and reaction, the variance of the reaction rate τ should respect the equality (7).

$$var(\tau) = S^T_{\tau/\sigma_{1g}} * cov(\sigma_{1g}) * S_{\tau/\sigma_{1g}} = S^T_{\tau/\sigma_{ng}} * cov(\sigma_{ng}) * S_{\tau/\sigma_{ng}} \quad (7)$$

Where σ is the microscopic cross-section, τ is the reaction rate, knowing that $\tau = \sum_{i=1}^{n} \sigma_i \Phi_i$, with n the number of groups in the energy mesh, and Φ the neutron flux, $cov(\sigma_{ng})$ is the relative covariance matrix of the multigroup cross-section, $S_{\tau/\sigma_{ng}}$is the sensitivity vector of the reaction rate to the multigroup microscopic cross-section, collapsed to n energy groups (see formula (8)).

$$S_{\tau/\sigma_i} = \frac{\partial \tau}{\partial \sigma_i}\frac{\sigma_i}{\tau} = \frac{\partial\left(\sum_{j=1}^{n} \sigma_j \Phi_j\right)}{\partial \sigma_i}\frac{\sigma_i}{\tau} = \Phi_i \frac{\sigma_i}{\tau} = \frac{\tau_i}{\tau} \quad (8)$$

Using (7) in (8) leads to the expression (9) of the microscopic cross-section variance:

$$var(\sigma_{1g}) = \left(\frac{\tau_i}{\tau}\right)^T_{1\le i\le n} * cov(\sigma_{ng}) * \left(\frac{\tau_i}{\tau}\right)_{1\le i\le n} \quad (9)$$

A development similar to what has been done for (7) leads to the formula (10) for the collapsing of the multigroup covariance matrix $cov(\sigma_a, \sigma_b)_{ng}$ between two partial cross-sections a and b of a given isotope into a one-group covariance term $cov(\sigma_a, \sigma_b)_{1g}$. [25].

$$cov(\sigma_a, \sigma_b)_{1g} = {}^t\left(\frac{\tau_{a,i}}{\tau_a}\right)_{1\le i\le n} * cov(\sigma_a, \sigma_b)_{ng} * \left(\frac{\tau_{b,i}}{\tau_b}\right)_{1\le i\le n} \quad (10)$$

5.2 Validation of the method

For decay heat uncertainty propagation calculations, covariance matrices are taken in the COMAC library [22]. The matrices are in a multigroup format (26 or 33 groups). For decay heat purposes, only radiative capture, fission and (n,xn) covariance matrices are taken into account at the moment, as well as cross-correlation matrices between these reactions.

The method employed in order to validate the collapsing of the covariance matrices is also to compare the results to those obtained with the sampling method. Indeed, with URANIE/MENDEL, multigroup cross-section covariances are sampled without collapsing, taking into account cross-correlations between partial cross-sections.

The MOX fuel case is relevant to illustrate the collapsing method, because the cross-section contribution to the total decay heat uncertainty is strong, especially at long cooling times. Indeed, in the MOX fuel case, when propagating the COMAC covariance matrices, it appears that cross-

section uncertainties are responsible for about 20% of the total decay heat uncertainty at 1.0 second and for more than 90% of the total uncertainty after 10^8 seconds (see Figure 4).

At a cooling time of one second after the reactor shutdown, the three main cross-sections contributing to the decay heat uncertainty are the radiative capture of ^{238}U and ^{240}Pu and the fission cross-section of ^{239}Pu (see Table 1). After 10^8 seconds, the two main cross-sections responsible for almost 4.0% of uncertainty are the radiative capture of ^{242}Pu and ^{243}Am, involved in the build-up of the ^{244}Cm, main contributor to the total decay heat at this cooling time.

The results showed in Figure 5 confirm that this method gives correct results.

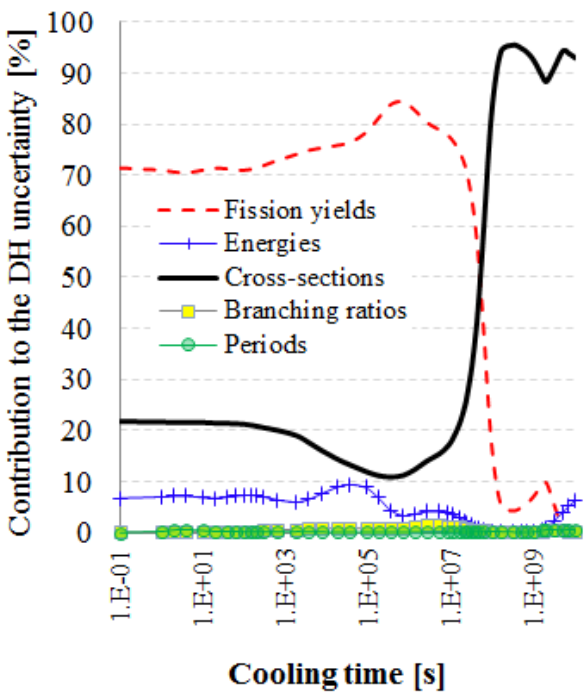

Figure 4 – MOX fuel case: relative contributions to the total DH uncertainty

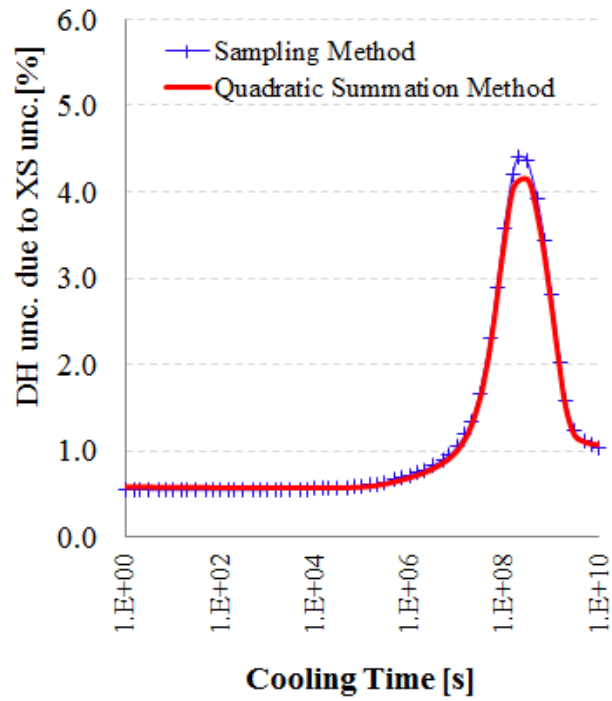

Figure 5 – MOX fuel case: DH uncertainty due to the propagation of cross-section covariances

Table 1 – Sensitivity coefficients and uncertainty values of the main cross-sections involved in the total decay heat uncertainty after 1 second and 3 10^8 seconds

Cooling time [s]	Cross-section	Sensitivity coefficient [%/%]	Uncertainty [%]
1	^{238}U, capture	0.31	0.85
	^{240}Pu, capture	0.15	1.60
	^{239}Pu, fission	0.13	1.20
3 10^8	^{242}Pu, capture	0.34	11.5
	^{243}Am, capture	0.26	3.80
	^{244}Cm, capture	-0.06	14.7
	^{238}Pu, capture	-0.07	9.50

6. STUDY OF THE BOLTZMANN/BATEMAN COUPLING IMPACT ON UNCERTAINTY PROPAGATION RESULTS

Currently, cross-section's sensitivity coefficients are evaluated by direct perturbation of the nominal value stored in the SAPHYB file, after the neutron calculation. It is recalled that the SAPHYB file is a tabulated file of the multigroup cross-sections and spectra as a function of the burn-up steps of the neutron calculation. The 1-group reaction cross-section are perturbed as described in (12), where n is the total number of burn-up steps, $\sigma^*_{[BU_i;BU_{i+1}[}$ and $\sigma_{[BU_i;BU_{i+1}[}$ the perturbed and nominal values of the cross-section during the burn-up step I, and Δσ the perturbation rate.

$$\forall i \in \{1..n\}\ \sigma^*_{[BU_i;BU_{i+1}[} = \sigma_{[BU_i;BU_{i+1}[} * (1 + \Delta\sigma) \tag{11}$$

Therefore the retroaction of the perturbation of cross-sections on the neutron spectrum is not taken into account. In order to quantify the impact of this approximation, the rigorous perturbation of the cross-section at the beginning of the neutron calculation has been studied for different cases of UOX and MOX fuels. The results corresponding to an UOX fuel (2.5wt% ^{235}U) and a MOX fuel (7.0wt% plutonium content) at 50 GWd/t are detailed here because they are the ones with the higher discrepancies.

From this study it appears that only five sensitivity coefficients are not accurately evaluated when the Boltzmann/Bateman coupling is neglected. The involved cross-sections are the radiative capture cross-sections of ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu and the fission cross-section of ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu.

Figure 6 shows how much it may impacts the sensitivity profiles of the decay heat to these particular cross-sections. This figure refers to the sensitivity profile of the ^{238}U capture cross-section of the UOX fuel as a function of the cooling time. It can be seen that up to 10^9 seconds, the sensitivity coefficients are very close to 0 when taking into account the coupling whereas they are around 0.6 without coupling. This is due to the fact that a perturbation of the ^{238}U cross-section of a PWR UOX fuel has an incidence on the whole neutron spectrum and has a negative effect on the thermal reaction rates of the other isotopes, leading to a reduction of the total fission rate, and therefore a fewer production of fission products. The ^{239}Np is also less sensitive to the ^{238}U capture cross-section, and so is the build-up of ^{239}Pu.

At t=10^{10} seconds the sensitivity coefficient with coupling is higher than the one without. At this cooling time, ^{241}Am is the main contributor to the total decay heat (about 70% of the total decay heat is due to ^{241}Am). The retroaction on the neutron spectrum after the perturbation of the ^{238}U capture cross-section leads to a reduction of the ^{241}Pu capture rate, which favors the production of ^{241}Am by radioactive decay.

The impact on the decay heat uncertainty of using more correct sensitivity profiles for the cross-section of the main actinide is shown in Figure 7. As expected from the study of the sensitivity profile of ^{238}U, to neglect the Boltzmann/Bateman retroaction leads to an over-estimation of the decay uncertainty inferior to 0.2% before 10^6 seconds of cooling, and to a slight under-estimation (0.5% at the maximum) between 10^9 and 10^{14} seconds of cooling.

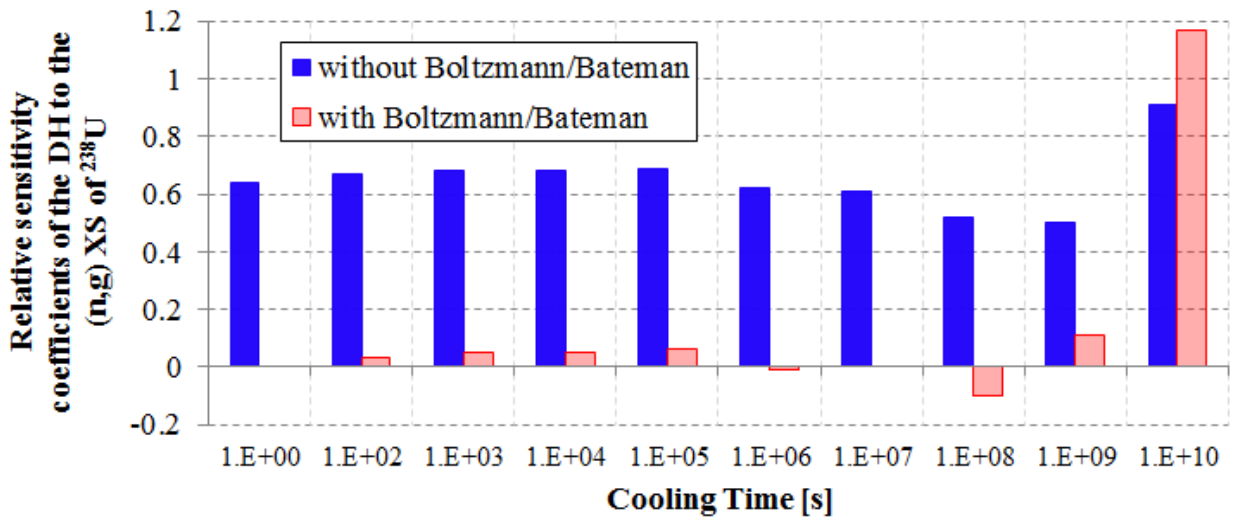

Figure 6 – Sensitivity profile of the decay heat to the ^{238}U capture cross-section for the UOX fuel

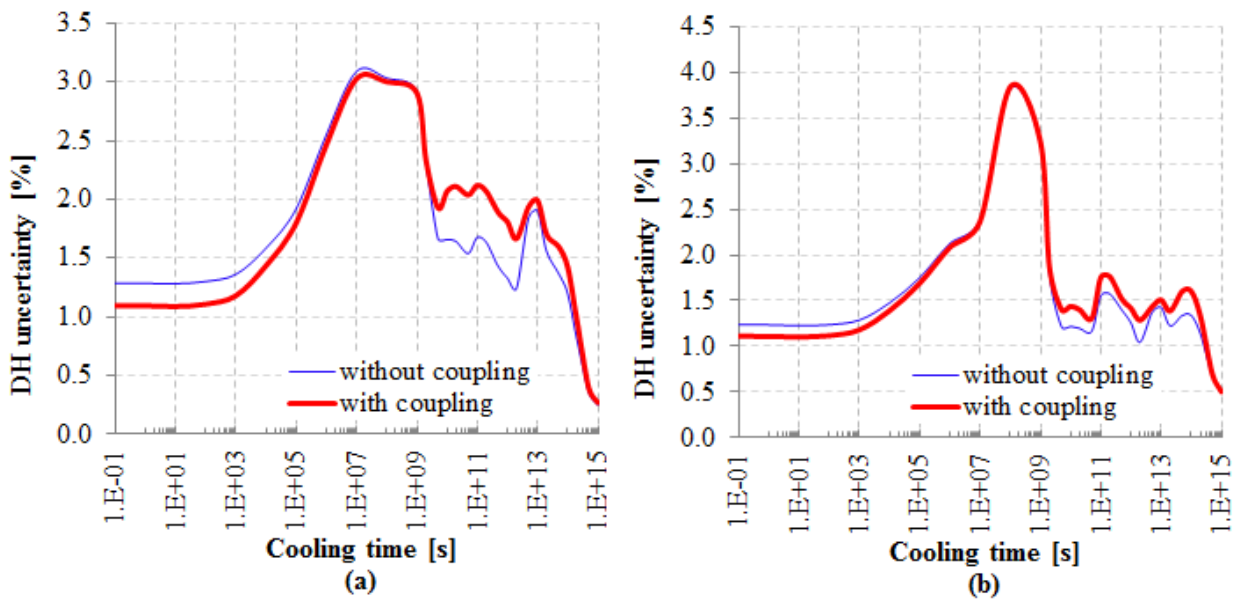

Figure 7 – Impact of the Boltzmann/Bateman coupling on the total decay heat uncertainty at one standard deviation for the UOX fuel (a) and the MOX fuel (b)

7. CONCLUSION

The goal of this paper is to explain what has been done in order to accredit the use of the quadratic summation method for decay heat uncertainty calculations and to check that the approximations made to implement this method within de DARWIN2.3 package do not bring about additional biases.

The results discussed in this paper give credence to the choices made. The linearity of the decay heat to nuclear data has been checked, as well as the method for collapsing multigroup covariance matrices for cross-sections and covariance matrices between partial cross-sections. Eventually, the fact that a decay heat calculation performed with the DARWIN2.3 package is

done in two separated steps: first the neutron calculation and then the depletion calculation makes it difficult to take into account properly the Boltzmann/Bateman coupling when computing the sensitivity profiles by direct perturbation. It has been found that this approximation is conservative until cooling times of about 300 years and that a few number of cross-sections are concerned.

These results, obtained with the CYRUS mockup, were useful for the development and the implementation of an uncertainty propagation method for the decay heat within the DARWIN2.3 package. This work is part of the global project of providing accurate and controlled biases and uncertainties for the decay heat over the whole application domain.

8. ACKNOWLEDGMENTS

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