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# A MODEL FOR FISSION YIELD UNCERTAINTY PROPAGATION BASED ON THE URANIE PLATFORM

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## ABSTRACT

In the present work, we analyze how fission yields uncertainties can be propagated in a burn-up calculation. The first part of the work is dedicated to the fission yield covariances generation in CONRAD (COde for Nuclear Reaction Analysis and Data Assimilation) to be used in the neutronic code APOLLO2. Fission yield covariance files are in fact unavailable in present nuclear databases such as JEFF-3.2 and ENDF/B-VII. To propagate such uncertainties, we adopted a statistical method which has a solid theoretical base and a relatively simple implementation. Fission yields have been therefore treated as random variables to be sampled from a normal input parameter multivariate distribution, taking into account correlations. Successively, a statistical representative number of calculations are carried out with the different sampled input data. An output multivariate probability distribution for core characteristics is then inferred. Random variable sampling and statistical post-processing has been performed using URANIE, a sensitivity and uncertainty analysis platform based on ROOT. This methodology is applied on a simplified geometry, leaving further developments for more complicated layout to future works.

*Key Words:* Fission yields, covariances, URANIE, uncertainty propagation, latin hypercube sampling.

## 1 INTRODUCTION

Future innovative nuclear systems will require strict safety-by-design standards. Accurate uncertainty quantifications of engineering parameters are therefore needed and several efforts have being spent for cross section uncertainty estimation and improvement to upgrade the basic nuclear data knowledge to be applied in nuclear design calculation tools [1].

Fission products yields (FY) are fundamental nuclear data for burn-up and activation calculations, including those on decay heat, shielding, dosimetry, fuel handling, waste disposal and safety [2]. Nowadays full uncertainty information are not always available in the actual nuclear database and the nuclear community expressed the need for full fission yield covariance matrices

to be able to produce inventory calculation results that take into account complete uncertainty data [3, 4].

In this work, some FY best estimates and associated uncertainty evaluation methodologies are shown. The main goal is to generate reliable and physically consistent FY covariances and to propagate FY uncertainties in a coupled transport-depletion quasi-static calculation on a simplified geometry. In the present study, we will start talking about mass, isotopic, isomeric and cumulative FY, showing the models we adopted to generate covariances through a Bayesian methodology. Afterwards we describe the development of a straightforward methodology in order to achieve statistical uncertainty quantifications of neutronic parameters, relying on Monte Carlo random or quasi-random sampling of the FY which have been supposed random variables governed by Gaussian multivariate distributions.

The sampling procedures of the FY data, required to generate the input files to be used in the depletion calculation, have been performed through URANIE [5]. The "Sensitivity and Uncertainty" platform URANIE developed at CEA provides algorithms and methods to accomplish sensitivity and uncertainty analysis, statistical processing, large-scale data analysis and code validation. URANIE is based on the data analysis framework ROOT, an object-oriented and petaflop computing system developed at CERN.

In the final part of this work, some preliminary results will be shown, followed by a brief discussion on future developments and improvement of this methodology for more advanced applications.

## 2 FY MODELS AND COVARIANCE GENERATION

In this section, a brief description of the fission yield models we used in a Bayesian procedure to estimate covariances will be presented. Covariances can be in fact generated simultaneously together with best estimates using the Generalized Least Square Method (GLSM, see ref. [6] for further information). To generate covariances, we used the GLSM and the data analysis modules available in CONRAD (COde for Nuclear Reaction Analysis and Data assimilation) developed at CEA-Cadarache [7].

### 2.1 Fission Yield Basic Definitions

Generally it is good practice calling primary fission fragments the isotopes born just after the scission process, before the emission of prompt neutrons [8]. Secondary fission fragments are afterwards obtained once prompt neutrons are emitted. We talk about fission products when the radioactive decay comes into play.

Independent FY (IFY) are those referring to secondary fission fragments distribution. IFY are crucial for decay and elementary fission heat calculations since they allow to determine the initial concentration of the fission fragments inventory that will release energy through the radioactive decay. FY are usually differentiated in:

1. Mass FY, which give the distribution of the isobaric fission fragments;
2. Isotopic FY, which take into account the charge distribution for a fixed mass  $A$ ;
3. Isomeric FY, which consider the isomeric repartition of the fission fragments if metastable states are available.

In the following sections, we will refer to mass IFY by using the symbol  $Y(A)$ . Since no ternary fission are considered in the present treatment, we obtain

$$\sum_A Y(A) = 2 \quad (1)$$

Isotopic IFY, denoted as  $Y(A, Z)$ , are given once the charge distribution is considered through the fraction  $f(A, Z)$  such that

$$Y(A, Z) = Y(A)f(A, Z) \quad (2)$$

$$\sum_Z Y(A, Z) = Y(A) \quad (3)$$

Isomeric IFY, denoted as  $Y(A, Z, M)$ , where  $M$  is the isomeric state that identifies a metastable state ( $T_{1/2} > 1.0 \text{ ms}$ ), are similarly given by

$$Y(A, Z, M) = Y(A, Z)R(A, Z, M) \quad (4)$$

$$\sum_M Y(A, Z, M) = Y(A, Z) \quad (5)$$

where  $R(A, Z, M)$  is the isomeric ratio that describes the repartition into the available metastable states of the fission fragments.

After the prompt neutron evaporation, the fission fragments are still too rich in neutrons and  $\beta^-$  decays are necessary. Usually, a secondary fission fragments is located at 3 charge units far from the  $\beta^-$ -decay stable line. For nuclei very far from stability, the half-lives are reduced such that for a 6-units far isotope we find  $T_{1/2} = 1 \text{ s}$ . To take into account radioactive decay cumulative FY are used. Cumulative FY are in fact the summation of all the contributions to a given isotope overall the entire decay time, since the fission process happened. In formulas [2] we can write:

$$C_i = Y_i + \sum_{j=0}^N C_j b(j \rightarrow i) \quad (6)$$

where  $i$  indicates a generic triplet  $(A, Z, M)_i$  and  $b(j \rightarrow i)$  is the branching ratio which gives the probability that an isomer  $(A, Z, M)_j$  decays into  $(A, Z, M)_i$ . With  $N$ , we have indicated all our fission fragments inventory. In matrix form, the last equation can be written as

$$\vec{C} = \mathbf{Q}\vec{Y} \quad (7)$$

where

$$\mathbf{Q} = (\mathbf{I} - \mathbf{B})^{-1} \quad (8)$$

indicating with  $\mathbf{B}$  the branching ratio matrix, with  $\mathbf{I}$  the identity matrix and finally with  $\vec{C}$  and  $\vec{Y}$  respectively the cumulative and the independent FY vectors.

## 2.2 Isotopic Fission Yield Model

Unfortunately, a precise self-consistent physical model with good predictive capabilities for the fission process cannot be provided yet. The knowledge about fission still has large margins of improvement and many problems are challenging physicists. To generate covariances, we based the GLS analysis on semi-empirical models based on phenomenological postulations. In this section, we will give only a general overview on the models used in this work, without pretending to be exhaustive and leaving the interested reader to specialized references for further research on the topic.

Isomeric IFY can be expressed as:

$$Y_{post}(A, Z, M) = \left[ \sum_{\nu_i=0}^{\infty} Y_{pre}(A + \nu_i) \cdot p_{A+\nu_i}(\nu_i) \cdot f(A + \nu_i, Z) \right] \cdot R(A, Z, M) \quad (9)$$

where we took into account the probability for a primary fission fragment of mass  $(A + \nu_i)$  to evaporate  $\nu_i$  prompt neutrons. In the Eq. 9, we specified post and pre-neutron distributions to distinguish the yields before and after the prompt neutron emission.

For the mass pre-neutron yields, we used a five-Gaussian distribution representing the Brosa fission modes [9]. To represent yields after prompt neutron emission, so the independent fission yields, it is necessary to multiply this distribution by the prompt neutron emission probability  $p_A(\nu_i)$ . We supposed it based on Gaussian distributions [10]. The average number of prompt neutron emitted by a primary fragment follows the saw-tooth curve [8], responsible for the asymmetry of the post-neutron mass yield distribution.

The charge distribution  $f(A, Z)$  has been represented using the Wahl model [11]. Wahl developed a semi-empirical model based on the Unchanged Charge Density hypothesis, modified by a correction term to take into account polarization phenomena.

The isomeric ratio  $R(A, Z, M)$  has been calculated using the Madland and England model. Madland and England developed a simplified model to calculate branching isomeric ratio for FY prediction [12], proposing a one-parameter model able to directly provide the ratio  $R(A, Z, M)$ .

The Madland-England model is based on the following assumptions [12]:

1. Fission fragments are formed with a density distribution,  $P(J)$ , of total angular momentum,  $J$ , given by

$$P(J) = P_0(2J + 1)e^{\left[ \frac{-(J+\frac{1}{2})^2}{J_{rms}^2} \right]}, \quad (10)$$

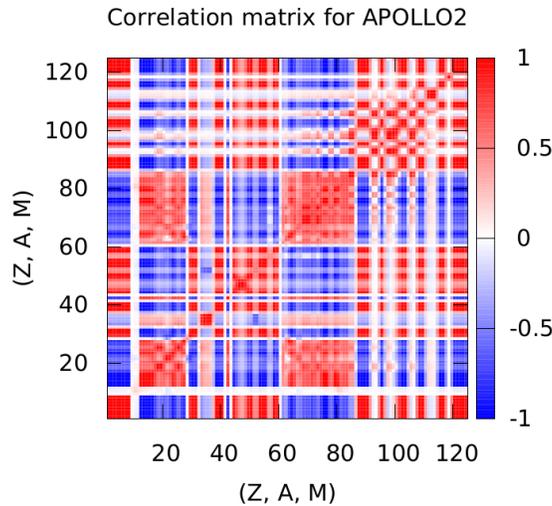
which is predicted by the statistical model [13].

2.  $J_{rms}$  is constant for all the fragment masses but varies with the incident neutron energy.

3. The branching mechanism is quite straightforward: fragments with  $J$  closer to the angular momentum  $J_m$  or  $J_g$  decays to the metastable or to the ground state respectively. Fragments with  $J$  exactly in the middle between  $J_m$  and  $J_g$  follows an equal repartition between both.

### 2.3 APOLLO2 Fission Yields Library Generation

The neutronic code APOLLO2 has the capability to perform burn up calculations in a quasi-static formulation for the Boltzmann and the Bateman equations. The input file is characterized by a list of about 120 fission products for the fast and thermal fission of several actinides. In this work we considered only the thermal fission of  $^{235}\text{U}$ , generating its correlation matrix for APOLLO2 using CONRAD (see Fig. 1).



**Figure 1. Fission yield correlation matrix for the thermal fission of  $^{235}\text{U}$  for the fission products used in APOLLO2 calculations.**

The APOLIB (APOLLO2 library) used in this work is based on the European nuclear data library JEFF-3.1.1. As previously mentioned, no fission yields covariance files are available in JEFF. Our main goal was to reproduce as much as possible the JEFF library for fission yields, including associated covariances. Therefore JEFF files were used as pseudo-experimental data in the adjustment procedure and covariances were produced through the GLSM applied to the models briefly described in the paragraphs before (see ref. [10]).

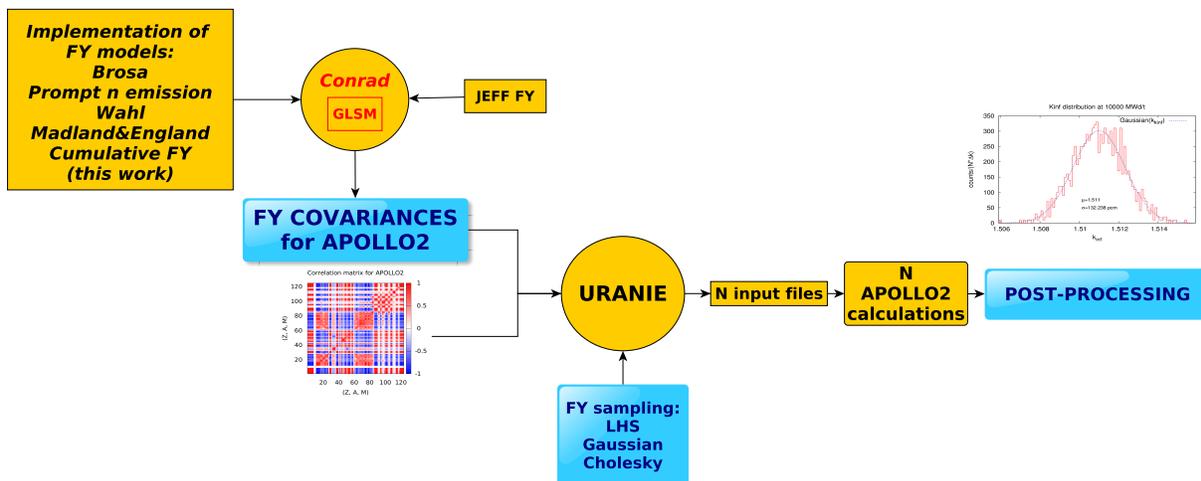
The reported fission yields in APOLLO2 are partial cumulative fission yields. Long-lived isotopes are in fact allowed to decay in the evolutionary part of the calculation, when the elapsed time becomes significant compared to the half lives of the considered isotopes. In the next section, the method used in URANIE to sample the fission yields preserving the correlations calculated by CONRAD is explained.

### 3 MONTE CARLO SAMPLING OF FISSION YIELDS AS RANDOM VARIABLES

In the last years, many statistical methods have been employed to estimate the uncertainty propagation of cross section uncertainties [14–18].

In this section, a similar Monte Carlo sampling generally used for cross sections and now applied to fission yields (considered as random variable) is described. Sampled fission yields are successively used in APOLLO2 input files describing a burn-up problem for a highly enriched LWR UOX cell.

The procedure for uncertainty estimation of core characteristic such as  $k_{inf}$  using the Monte-Carlo based sampling method is presented in Fig. 2.



**Figure 2. Flow chart for the procedure we adopted to estimate the uncertainty associated to core characteristics in a burn-up calculation.**

This procedure can be summarized as follows:

1. Evaluation of fission yield covariance matrix for the fast and thermal fission of several actinides.

2. Monte Carlo based sampling of fission yields using URANIE and generation of corresponding input files for APOLLO2 with randomly perturbed FY.
3. Performing of core analysis using each perturbed FY library and then post-processing of the obtained core characteristics.

Let us now describe in the following paragraphs the Monte Carlo sampling procedure performed by the code URANIE and applied in the present work.

### 3.1 FY Monte Carlo Sampling

FY have been considered as random variables following a multivariate normal distribution. Thanks to the URANIE libraries, FY are sampled based on the covariance data previously calculated by CONRAD, and then subsequent burn-up calculations are carried out using the sampled fission yields.

A random variable vector  $\vec{y}$ , which is described by a multivariate normal distribution, can be expressed as:

$$\vec{y} = \vec{\mu} + \mathbf{L}\vec{s} \quad (11)$$

where  $\vec{\mu}$  is the mean vector for  $\vec{y}$  which represents the FY vector and  $\vec{s}$  is the multivariate standard normal random vector [14]. The matrix  $\mathbf{L}$  is expressed by

$$\mathbf{L} = \mathbf{T}\sqrt{\mathbf{D}} \quad (12)$$

where  $\mathbf{T}$  and  $\mathbf{D}$  derive from the Cholesky decomposition of the covariance matrix  $\mathbf{C} = \mathbf{T}\mathbf{D}\mathbf{T}^t$ , since the matrix  $\mathbf{C}$  is in principle a Hermitian matrix.  $\mathbf{T}$  is the lower triangular and  $\mathbf{D}$  is diagonal matrix. The matrix  $\mathbf{L}$  should give:

$$\mathbf{C} = \mathbf{L}\mathbf{L}^t \quad (13)$$

### 3.2 Latin Hypercube Sampling

In this paragraph a brief introduction on the latin hypercube sampling (LHS) method is provided. In the LHS, each input parameter domain is subdivided into  $N$  intervals, where  $N$  is the number of samples [14]. Such intervals have an identifying number  $i = 1 \cdots N$ . The  $N$  interval numbers for each parameter domain are independently shuffled in order to form  $P$  random interval number sequences, where  $P$  is the number of parameters.

For example, supposing to have two fission yields  $y_{1,2}$  that must be sampled  $N = 10$  times, shuffling the interval numbers we generate  $P = 2$  arrays of 10 integers. Each pair formed by elements with the same array index  $j$  provides the indices for the sub-domain in the  $(N \times P)$ -dimensional space where perform the  $j$ -th uniform random sampling for the parameters.

Once all the samples are performed, a  $N \times P$  matrix  $\mathbf{R}_{LHS} = (\vec{r}_1 \ \vec{r}_2 \ \dots \ \vec{r}_N)$  of random numbers is generated. To generate the multivariate standard normal random vector  $\vec{s}$ , the inverse transform is used [14]. Once the cumulative function is given,

$$F(s) = \int_{-\infty}^s f(t)dt \quad (14)$$

where  $f(t)$  is the normal distribution, the multivariate standard normal vector can be obtained by solving  $\vec{s} = F^{-1}(\vec{r})$  for each of the  $N$  column of  $R_{LHS}$ .

### 3.3 Practical Considerations on the FY sampling

As previously mentioned, we chose to model FY random variables as normal distributions. The first problem we had to face using a symmetric distribution was the negative tails that can be significant for very low FY. In the present work, we did not sample yields smaller than  $\leq 10^{-9}$ . Since we were interested mainly on core characteristics, the effects of such FY have been considered negligible. Furthermore URANIE gives the possibility to set bounds in random variable sampling, then negative values were discarded. Positive defined probability distributions can eventually give better results and future developments of this work will analyze the application of such distributions (e.g. the log-normal distribution).

Another interesting point in this statistical uncertainty analysis is the determination of the minimum number of calculations needed to infer a reliable coverage of the output parameter population, with the related confidence. The theory of designing the sample size for non-parametric multivariate tolerance regions [19] has been proposed and used in several works on uncertainty propagation in nuclear reactor field [16, 20], and its complete description goes beyond the scope of this section. However, the essential aspects will be here presented for the sake of completeness.

Tolerance limits give a statistical interval where, with a given confidence, a proportion of a sampled population belongs to. Wilks, in his work [21], calculates the sample size in order to guarantee the coverage of a population (more generally: of a domain), with the required confidence. As also reported in [16], supposing just one output parameter, for the one-sided tolerance limit case the minimum sample size  $n$  is given by:

$$1 - \alpha^n = \beta \quad (15)$$

to infer the  $\alpha \times 100(\%)$  of the output population with a  $\beta \times 100(\%)$  of confidence. The coverage of

the output population from the  $(1 - \alpha) \times 100(\%)$  to  $\alpha \times 100(\%)$  with a  $\beta \times 100(\%)$  of confidence is obtained with a sample size given by:

$$1 - \alpha^n - n(1 - \alpha)\alpha^{(n-1)} = \beta \quad (16)$$

H. Ackermann and K. Abt [19], in their work "Designing the sample size for non-parametric, multivariate tolerance regions", provided approximate formulas and tables for the determination of the sample size constructing inner and outer limits for a multivariate tolerance region such as:

$$n = m \frac{\left(\frac{\chi_{\alpha,2m}^2}{2m} - 1\right)\sqrt{\beta} + 1}{1 - \beta} \quad (17)$$

where  $m$  is a parameter indicating the number of blocks (i.e. the portion of the hyperspace defined by the variables to be sampled) left out the tolerance limits.  $\chi_{\alpha,2m}^2$  is the  $\alpha$  percentile of the  $\chi^2$  distribution with  $2m$  degrees of freedom.

As previously discussed, to set up our methodology of FY uncertainty propagation in burn-up calculations, we tested a simplified geometry performing a statistical processing on core characteristics (see Sec. 4). We chose the several  $k_{inf}$  at different burn-up steps as output parameters, subsequently used to calculate the reactivity loss. Eq. 17 allows to construct tolerance regions provided with inner or outer limits, depending on  $\alpha$ .

We decided to adopt a sample size of 1000, a value that reasonably showed convergence for the output multivariate distribution. Furthermore, as previously mentioned, we relied on sample size optimization techniques such as the LHS, whose better efficiency against traditional random sampling has been already shown [14]. It has been demonstrated that for highly correlated output parameters the minimum sample size is lower than for independent non-correlated ones, and we will look for a correlation analysis for the output parameter results as part of the further developments of the method. In the next section a simple burn-up calculation for a PWR-UOx-cell has been set up in order to perform 1000 calculations with different sampled FY data sets.

#### 4 PRELIMINARY RESULTS ON A HIGHLY ENRICHED PWR CELL

In this section, we present some preliminary results on a simplified geometry implemented in APOLLO2 we decided to use as a test case to develop the present method of FY uncertainties propagation in burn-up calculations. In future developments using this methodology, we will study the possibility to include more complex geometry such as the Jules Horowitz Reactor (JHR), the high flux experimental reactor under construction at CEA-Cadarache.

**Table I. Geometry of the PWR-cell used in the APOLLO2 burn-up calculation.**

Cell Characteristics	(cm)
$R_{fuel}$	0.4098
$R_{i,cladding}$	0.4180
$R_{e,cladding}$	0.4750
Square Pitch	1.2620

#### 4.1 Geometry Description and Calculation Set-Up

Without entering to much into the details of the neutronic problem, considering that it has just the scope to be a test case for the feasibility of the present methodology, the geometry is mainly constituted by a zircalloy-cladded UOX cell moderated by light water. In Table I the main measures are reported.

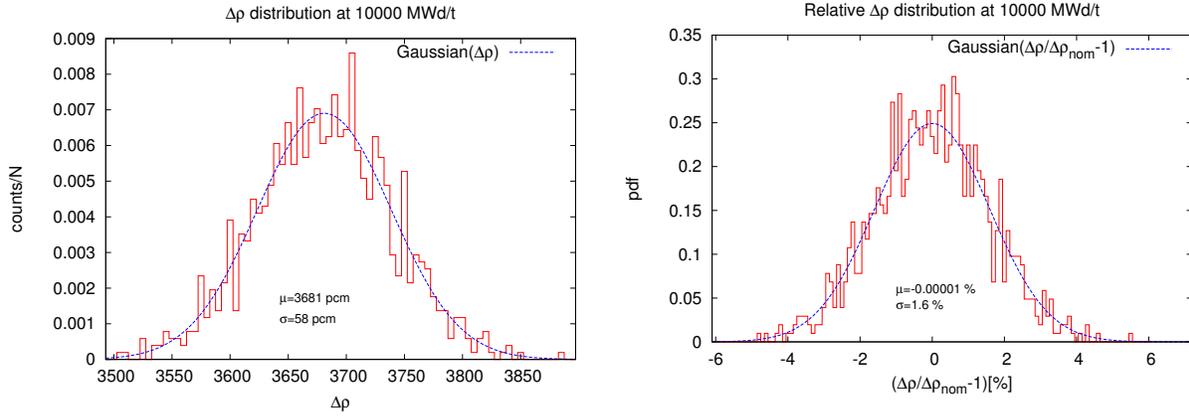
The UOX fuel has been set with an enrichment of 19.75 %, because we wanted to start already with some of the JHR reference conditions. The calculations have been performed using the collision probabilities solver available in APOLLO2 with 26 burn-up steps, from 37.5 MW.d.t<sup>-1</sup> to 10000 MW.d.t<sup>-1</sup>, and a specific power of 100 W.g<sup>-1</sup>.

For each sampled data sets of FY coming from  $^{235}\text{U}(n_{th}, f)$  reaction, a quasi-static problem has been solved with APOLLO2 using a URANIE complementary C++ interface able to generate samples, create APOLLO2 input files and collect  $k_{inf}$  at each burn-up step during the depletion calculations.

#### 4.2 Results

In Fig. 3 the normalized counts for  $\Delta\rho = \rho_{EOC} - \rho_{BOL}$ , the reactivity loss at the burn-up step of 10000 MW.d.t<sup>-1</sup>, and the relative reactivity loss compared to the nominal case are reported together with a Gaussian distribution. The post-processing has been performed using the estimators for mean and standard deviation to check if the averages were equal to the nominal case and to calculate the associated uncertainties on core characteristics. The  $\Delta\rho$  are nicely reproduced by a multivariate Gaussian distribution, which shows how the uncertainties on  $^{235}\text{U}$  FY can have considerable effects on the reactor neutronics. The relative reactivity loss distribution shows as the method converges (mean value equal to zero) and gives an estimation of the impact which the fission yield uncertainties, generated in this work, have on cycle length ( $\sim 1.6\%$ ).

This study has shown the feasibility and the relevance of FY uncertainty propagation in burn-up calculations. The simplified geometry has allowed us to perform calculations on single processors and, for future developments of this methodologies in more complex geometries, multi-processing is surely needed.



**Figure 3. PDF for the  $\Delta\rho$  at the burn-up step of 10000 MW.d.t<sup>-1</sup> (EOC) and representation of the relative  $\Delta\rho$  compared to the nominal case.**

## 5 CONCLUSIONS

In the present work, we have shown a model to propagate FY uncertainties in burn-up calculations. FY covariances, still missing in the actual nuclear databases, have been generated using CONRAD for the thermal fission of <sup>235</sup>U and used in a Monte Carlo Latin Hypercube Sampling to generate several APOLLO2 input files with different FY data sets sampled using URANIE. Performing the statistical post-processing on core characteristic for a simplified geometry, the methodology has given promising results and has shown the importance of FY uncertainty propagation to estimate the uncertainty associated to neutronic parameters.

In future developments of this methodology, more complex geometries will be treated, such as the experimental high flux reactor Jules Horowitz. Besides, more fissioning systems, at thermal and fast incident neutron energies, will be considered in FY covariances generation.

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