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## UNCERTAINTY QUANTIFICATION OF ISOTOPIC DENSITIES IN DEPLETED FUEL

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### ABSTRACT

Uncertainty quantification of interest outputs based on the nuclear data is an important issue for nuclear safety, from operation of nuclear facilities to long term deposit management. To be more accurate, the Verification and Validation process must take into account the uncertainty quantification due to various sources: input data, design, models...

This work is part of the VVUQ process for CEA depletion codes on a PWR fuel rod application. We compare the results of DARWIN and MENDEL and experimental data. Experimental data used in this work consist of isotopic concentrations for four UO<sub>2</sub> fuel samples irradiated up to 64.7 GWd/t in a pressurized water reactor. Uncertainties used for nuclear constant data are JEFF-3.1.1 ones, when cross sections uncertainties data are from the CEA COMAC data base.

Concerning the choice of uncertainty quantification methods, DARWIN deterministic approach is based on sensitivity profiles computation by direct forward perturbation, thanks to INCERD module. MENDEL probabilistic method is based on Monte Carlo correlated sampling. Those samplings are achieved through the CEA uncertainty platform URANIE.

Nuclear data uncertainties are propagated and experimental data errors are taken into account. Modelisation biases or algorithm accuracy are supposed negligible. Output quantities of interest are the isotopic concentrations at given cooling times.

We show that uncertainty propagation results by both methods give similar results, compatible with experimental data. This work validates both the methods and code systems on this application of high burnup PWR fuel irradiation. We focus on the elaboration of a measure for sensitivity coefficients applicable to the stochastic approach, where thousands of eventually correlated uncertain input parameters are taken into account.

### 1. INTRODUCTION

Code system VVUQ (Verification, Validation and Uncertainty Quantification) process consists of a three step proof of the validity of the codes:

- **Verification** : code elementary tests and code to code comparisons;
- **Validation** : code to experiment confrontation;
- **Uncertainty Quantification** : propagating input data uncertainties to outputs of interest.

This work is part of the VVUQ process for MENDEL v1.2 [1], CEA new generation depletion code. It focuses on a high burnup pressurized water reactor (PWR) fuel rod application. Experiment data [2] give isotopic concentrations for four  $\text{UO}_2$  fuel samples irradiated up to 64.7 GWd/t in a PWR. We use two fuel cycle codes developed in CEA/DEN: DARWIN/PEPIN2.4.2 [3] and MENDEL v1.2. Nuclear data nominal values and uncertainties on decay data are taken from JEFF-3.1.1 [4], while uncertainties on cross section data are taken from the CEA COMAC data base [5].

From now on, DARWIN/PEPIN2.4.2 is referred as DARWIN, and MENDEL v1.2 as MENDEL. Both code systems use a different approach for uncertainty quantification. DARWIN propagates uncertainties through its INCERD module [6, 7] thanks to a deterministic approach: forward perturbation, one at a time method. MENDEL propagation is based on a stochastic approach, using Monte Carlo correlated sampling [8, 9]. Samplings are generated by the CEA uncertainty platform URANIE [10]. Both codes solve the depletion equations (Bateman equation) and there is a simple chaining with the flux solvers, with no retroaction of isotopic concentration to neutronic flux in the presented application. Both methods are classic methods largely used in fuel cycle applications [11, 12, 13, 14].

This article is an extension of a previously published work [15] where validation process had not taken into account uncertainty quantification. Uncertainty is propagated to isotopic concentrations at several cooling times.

For this study, only experimental measurement errors and nuclear input data uncertainties are taken into account. Indeed, algorithm accuracy can be considered as marginal in regards to input data uncertainties, as we use reference solvers for the Bateman equation resolution. For modeling biases, we use the official complete decay chain available in MENDEL (resp. DARWIN), which is the best effort chain, generally used for reference calculations.

We show that both uncertainty propagation results give similar results, compatible with experimental data and validating both the methods and the code systems. We focus on the elaboration of a measure for sensitivity coefficients applicable to the stochastic approach, where thousands of eventually correlated uncertain input parameters are taken into account.

Similar work for elementary fission pulse have already been achieved and published, for both nominal computations [15] and uncertainty quantification [16, 1].

## 2. VVUQ PROCESS

### 2.1 Bateman Equation

In this work, we focus on irradiation process and cooling down of a homogenized radioactive material, where the atom density  $N_i(t)$  of nucleus  $i$  in a fissile material region is the solution of generalized Bateman [17] equation (1).

$$\frac{dN_i(t)}{dt} = -(\lambda_i + \tau_{i,i})N_i(t) + \sum_{j \neq i} (b_{j,i}\lambda_j + \tau_{j,i}^r)N_j(t) + \sum_k \gamma_{k,i}\tau_k^f N_k(t) \quad (1)$$

where

- $b_{j,i}\lambda_j$  is the partial decay rate of nuclide  $j$  to nuclide  $i$ ;
- $\tau_{i,i}$  is the total disappearance rate of nuclide  $i$  by its neutronic transmutations;
- $\tau_{j,i}^r$  is the transmutation rate of nuclide  $j$  to nuclide  $i$  by neutronic reaction  $r$ ;
- $\gamma_{k,i}$  is the fission yield of fissile nuclide  $k$  creating the fission product  $i$ ;
- $\tau_k^f$  is the fission rate of fissile nuclide  $k$ .

Multiple methods of resolution exist to solve the generalized Bateman equation [18]. The ones implemented in MENDEL have been validating in other works [19]. In the work presented here, both DARWIN and MENDEL uses a Runge-Kutta 4th order method, even if CRAM method [20] have also been implemented in MENDEL [19]. It will be released with MENDEL v2.0.

## 2.2 Correlations Between Uncertain Data

For the Uncertainty Quantification process, assumptions have been done on the choice of correlation between nuclear data. Uncertain data are of five types. No correlation is taken into consideration between data of different types.

- independent fission yields: default correlation assumes that the sum of yields for a given fissile system is constant (id est, no uncertainty on the mean number of fission products);
- radioactive decay branching ratio: correlation assumes that the sum of the decay branching ratios from a father isotope is equal to 1;
- radioactive decay energies: by defaults, considered independent;
- radioactive decay periods: considered independent;
- multigroup microscopic cross sections: correlations are given by COMAC [5] data base.

In the present work, DARWIN and MENDEL use uncertainty data from JEFF-3.1.1 [4] for independent fission yields, radioactive decay periods, radioactive decay branching ratios and radioactive decay energies.

## 2.3 Uncertainty Quantification Methods

We describe briefly in this section the propagation methods and hypotheses.

### 2.3.1 Deterministic Method Used in DARWIN

DARWIN propagates uncertainties from nuclear data to decay heat or isotopic concentrations using a direct forward first order perturbation method (one at a time method). With  $X$  the uncertain input variables (nuclear data) and  $Y$  the uncertain outputs (isotopic density), we can use the following formula:

$$\text{cov}(Y) = S_{Y/X} \text{cov}(X) S_{Y/X}^T \quad (2)$$

where  $\text{cov}(X)$  (resp.  $\text{cov}(Y)$ ) stands for the variance-covariance matrix for variable  $X$  (resp.  $Y$ ) and  $S_{Y/X}$  stands for the sensitivity matrix of  $Y$  regarding  $X$ .

The use of a direct forward first order perturbation method implies the hypothesis of linearity of the outputs (as a function of uncertain nuclear data). For small perturbations of the uncertain input parameters (limited to one standard deviation), this approximation is proved to be valid when comparing with propagation approaches not taking this linearity hypothesis, such as stochastic uncertainty propagation approach as the one used in MENDEL.

The uncertainty propagation in DARWIN needs  $(n + 1)$  depletion calculations where  $n$  is the number of uncertain parameters. Calculations are performed in parallel mode by DARWIN through its INCERD module, which establish the sensitivity matrix before computing the interest output covariance through equation (2).

Some parameters are associated to a zero value of uncertainty in the JEFF-3.1.1 library, due to the lack of knowledge. If we propagate a default uncertainty level for those parameters, the one at a time method will need to launch some 47039 calculations for decay data only (cross sections from COMAC will be added to this number): 7709 for decay energies, 3626 for decay periods, 2952 for decay branching ratios and 32752 for independent fission yields related to thermal and fast fissile systems. This level of accuracy is needed to obtain good precision on all isotopic concentrations and decay heat. Those calculations being highly scalable on perturbations, it is totally realistic in fuel cycle code systems, where flux are taken from a previous calculation. Calculation time for one perturbation lasts several seconds.

### **2.3.2 Stochastic Method Used in MENDEL**

MENDEL propagates uncertainty using a correlated sample method. The creation of the samples is performed by CEA/DEN URANIE platform [10]. Samples are generated by the use of a LHS method [8] based on the Inverse Density Function methodology. For correlated sampling, we use a correlation by rank algorithm on the samples.

For this study, 2000 realizations were propagated. Most random variables are chosen Gaussian if the relative standard deviation is lower than 50% and Log-Normal in the other cases. Fission yields are always considered log-normal. This choice for nuclear data probability density functions have been explicit in [21].

## **3. VALIDATION OF MENDEL ON A PWR APPLICATION CASE**

### **3.1 Description of the Experiment**

The isotopic concentrations are calculated by MENDEL and DARWIN for a 3.8% enriched UOX PWR fuel cell, using JEFF-3.1.1 nuclear library data. Results are compared to the experimental data in [2]. This study consists of a chemical analysis of a high-burnup PWR-UO<sub>2</sub> fuel, whose four fuel samples (A to D) are analyzed. Burnup varies from 52.8 to 64.7 GWd/tHM. This burnup is obtained after 5 cycles, and the chemical isotopic concentrations are calculated after a cooling time from 3 years to 5 years. Four samples are extracted at four different axial positions of the high-burnup PWR-UO<sub>2</sub> fuel, with different moderator temperature which is supposed to be constant during the irradiation. Table 1 indicates the elapsed time from discharge to chemical isotopic analysis for each sample.

Sample	Nuclide	Cooling period (in days)
A	U, Np, Pu	1206
	Am, Cm, fission products	1234
B, C, D	<sup>134</sup> Cs, <sup>137</sup> Cs, <sup>154</sup> Eu	1625
	U, Pu	1649
	Np, other fission products	1696
	Am, Cm	1706

Table 1 – Cooling time from discharge to chemical isotopic analysis for each sample and isotope

MENDEL and DARWIN/PEPIN2.4 both compute isotopic densities with CEA-V512 nuclear data library [22] (based on JEFF-3.1.1) for all the isotopes on which the chemical analysis had been carried out. Reference chains with more than 2500 isotopes are used in both codes. Calculation of isotopic concentrations and decay heat in MENDEL and DARWIN uses neutronic data (reaction rates and neutronic flux) issued from a Saphyb file created by APOLLO2 [23]. Cooling period between the cycles is not taken into account during the modeling process in APOLLO2, but added during the DARWIN (resp. MENDEL) calculations. During the simulation in MENDEL and DARWIN, cooling periods of 7 weeks (49 days) between two cycles are taken into consideration. A fourth order Runge Kutta solver with parabolic interpolation of neutronic reaction rates during all the five cycles is used in MENDEL. In DARWIN, the fourth order Runge-Kutta method is implemented on constant by steps neutronic reaction rates.

In the first place, comparison will be done without uncertainty quantification. Discrepancy between calculated isotopic concentrations (by both MENDEL and DARWIN), as well as between calculated isotopic concentrations and the experimental values are analysed.

In a second step, uncertainty propagation is applied in order to study the influence of nuclear data values on isotopic concentration calculation and accuracy. In this part, computed uncertainties in MENDEL and DARWIN are taken into consideration for the sake of giving the confidential interval of calculation.

### 3.2 Isotopic concentrations without uncertainty propagation

#### 3.2.1 Comparison between MENDEL and DARWIN

In order to compare isotopic concentrations from MENDEL and DARWIN calculations, we use the same depletion chain, the same numerical method (Runge Kutta 4th order), the same assumptions of a constant by steps reaction rates. The discrepancy of calculated isotopic concentrations between MENDEL and DARWIN is shown in the figure 1. Discrepancy is calculated by  $\frac{N_{MDL} - N_{DAR}}{N_{MDL}}$ , where  $N_{MDL}$  (resp.  $N_{DAR}$ ) represents the isotopic concentrations in MENDEL (resp. in DARWIN). Isotopic concentration discrepancies are all in the -0.7% to 0.4% interval (where there are only 3 isotopes with discrepancies larger than 0.5%, while 44 isotopes whose discrepancies less than 0.2%). Hence, MENDEL and DARWIN give nearly the same result.

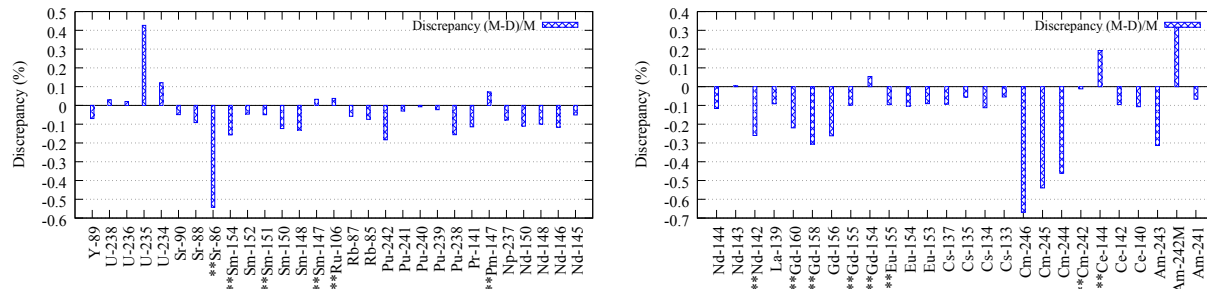


Figure 1 – Comparison of isotopic concentrations between MENDEL and DARWIN

### 3.2.2 Comparisons between experimental data and MENDEL results

MENDEL and DARWIN results were compared to experimental data and relative discrepancy for each nuclide. Reader can refer to mean values of figure 2 which gives the mean relative discrepancy between experimental data and numerical results over the available fuel samples (A to D) data. Those results are similar to reference paper [2] and to previous work [15].

From now on, both MENDEL and DARWIN use their own reference depletion chain.

Referring to heavy nuclides,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{239-242}\text{Pu}$  are overestimated by MENDEL results compared to experimental data, and this overestimation has been confirmed when comparing with CBZ code [24] results.

Detailed power history is unknown. We made an hypothesis of constant power history for each cycle, which causes the imprecise neutron flux as well as discrepancy of the concentration for  $^{235}\text{U}$ . Concerning to the concentration discrepancy of  $^{234}\text{U}$ , it is possibly due to the inaccuracy initial concentration.

Discrepancies for several nuclides – like plutonium, americium and curium – are outside the experimental error bars. To investigate more precisely, uncertainty quantification is decided.

### 3.3 Uncertainty propagation in MENDEL and DARWIN

#### 3.3.1 Comparison of experimental and computed concentration with uncertainty propagation

For one isotope, let's consider we have  $M_{nz}$  experimental data ( $M_{nz} \in [1, 4]$ ), corresponding to the available data for fuel samples A to D. Global discrepancy is computed as the mean discrepancy over available data. We note  $X$  the isotopic density.

We show that the mean standard deviation  $\sigma_{ABCD}(X)$  over  $N$  statistical samples reads:

$$\sigma_{ABCD}^2(X) = \frac{1}{M_{nz} * N - 1} \sum_{k=1}^{M_{nz}} \sum_{i=1}^N [(X_{k,i} - \bar{X}_k)^2] = \frac{1}{M_{nz} * N - 1} \sum_{k=1}^{M_{nz}} (N - 1) \sigma_k^2(X) \quad (3)$$

where  $X_{k,i}$  is the isotopic density calculated for sample  $i$  in case  $k$ ,  $\bar{X}_k$  is the mean value for isotopic densities in case  $k$  and  $\sigma_k(X)$  is the standard deviation for case  $k$ .

Hence, we obtain

$$\sigma_{ABCD}^2 = \frac{N - 1}{M_{nz} * N - 1} \sum_{k=1}^{M_{nz}} \sigma_k^2 \cong \frac{1}{M_{nz}} \sum_{k=1}^{M_{nz}} \sigma_k^2(X) \quad (4)$$

Figure 2 shows isotopic concentrations from MENDEL and DARWIN calculations. Ordinates correspond to one standard deviation when propagating all nuclear data uncertainties (fission yields, decay periods, decay branching ratios and microscopic cross sections).

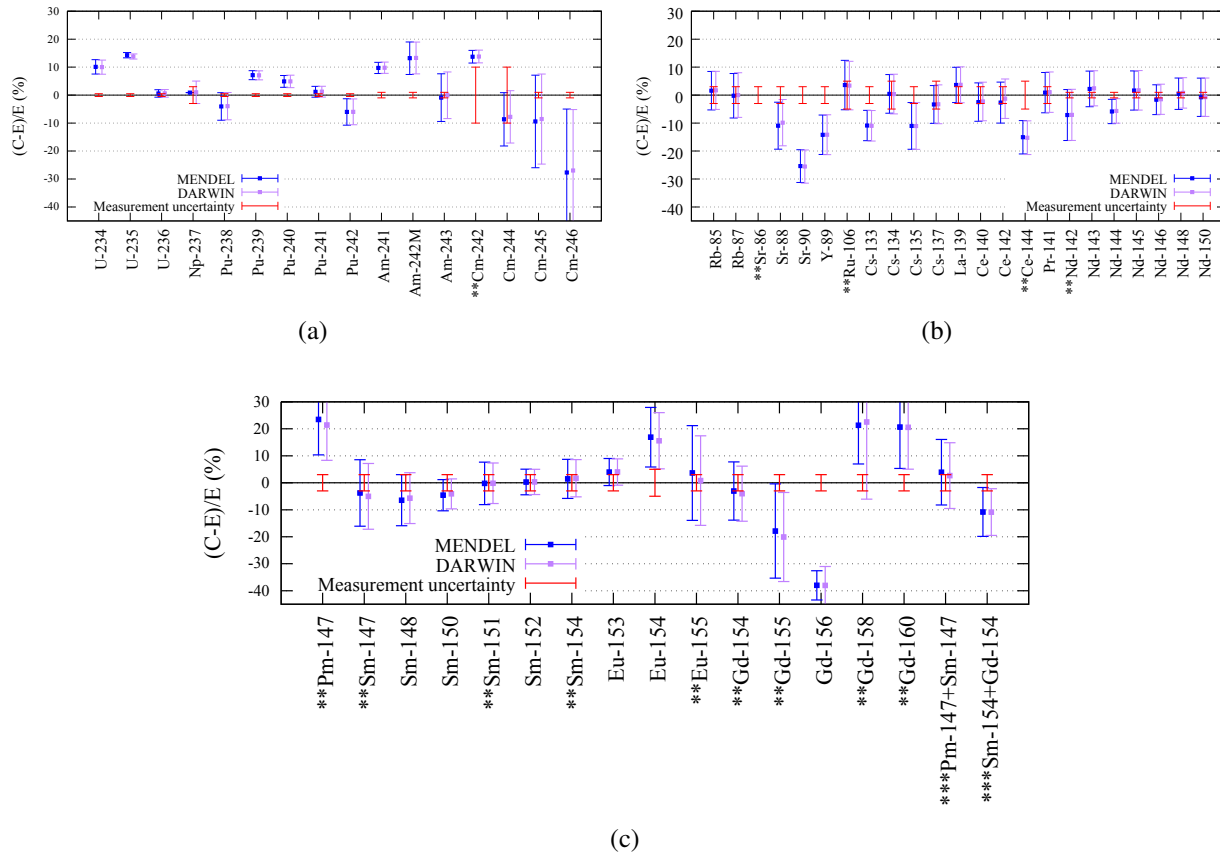


Figure 2 – C/E ratio of MENDEL and DARWIN with uncertainty propagation within  $1 \sigma$

It can be observed that for several isotopes, uncertainty propagation bars and experimental error bars at  $1 \sigma$  do not overlap. The seven isotopes for which it is still the case for  $2 \sigma$  are listed in table 2.

Isotope	$^{86}\text{Sr}$	$^{90}\text{Sr}$	$^{156}\text{Gd}$	$^{234}\text{U}$	$^{235}\text{U}$	$^{239}\text{Pu}$	$^{241}\text{Am}$
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Table 2 – List of isotopes out of experimental error bars with  $2 \sigma$

As shown in figure 3, uncertainty on isotopic concentration is mainly due to fission yields for fission products and to microscopic cross section for actinides.

A comprehensive study is needed for the 7 isotopes listed in table 2 (3 fission products and 4 actinides) will be studied particularly. For fission products, cross section effects can not be omitted, particularly for Europium, Gadolinium, Samarium,  $^{142}\text{Nd}$ ,  $^{147}\text{Pm}$ ,  $^{133}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{135}\text{Cs}$ .

### 3.3.2 Discrepancy between computed uncertainty for $^{158}\text{Gd}$ in MENDEL and DARWIN

Figure 2c shows a considerable different of uncertainties for  $^{158}\text{Gd}$  calculated by both codes. As shown by table 3 the main contributor to  $^{158}\text{Gd}$  concentration is the capture cross section of  $^{156}\text{Eu}$ , for which the equivalent one group standard deviation would be 135 %. On some energy groups,



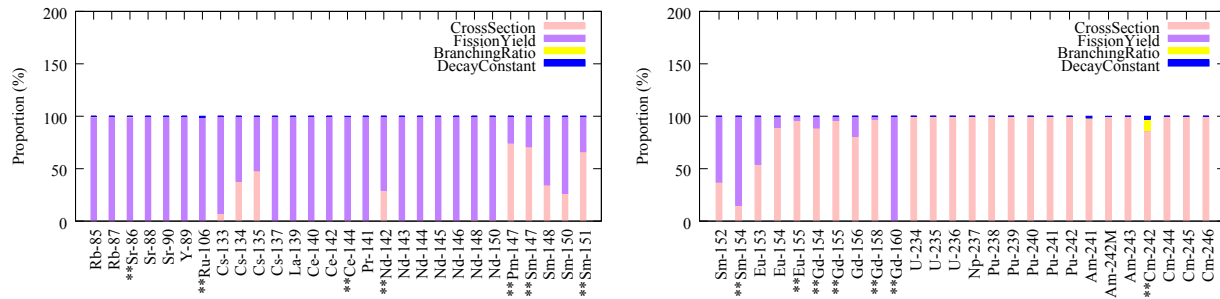


Figure 3 – Contributions of  $Y$   $\lambda$   $Br$  and  $\sigma$  on isotopic concentrations

relative standard deviation goes up to 1500 %. This discrepancy is due to the huge level of

Reaction type	$\frac{\Delta X}{X}$	Sensitivity	Uncertainty (%)
$^{239}\text{Pu} (n, \gamma) \sigma^{158}\text{Gd}$	2.29E-02	3.40E-02	0.08
$^{239}\text{Pu} (n, f) \rightarrow^{158}\text{Gd}$	1.28E-02	1.40E-01	0.18
$^{239}\text{Pu}$ fission $y_i^{157}\text{Sm}$	1.86E-01	1.30E-01	2.41
$^{241}\text{Pu} (n, \gamma)$	1.48E-02	1.10E-01	0.16
$^{153}\text{Eu} (n, \gamma)$	4.86E-02	1.56E-01	0.76
$^{156}\text{Eu} (n, \gamma)$	1.35E+00	1.65E-01	22.39

Table 3 – Cross sections sensitivity to  $^{158}\text{Gd}$  concentration

uncertainty, URANIE sampler is not able to correctly sample this level of uncertainty with only 2000 samples, and needs millions of samples to do it. The same result have been shown with python native Gaussian and log normal distributions. Samplers specifically written for high level of uncertainties should be use, which is not yet available in URANIE. Thanks to this study, the high level of uncertainty in COMAC data base has been reported to COMAC team, for eventual corrections.

### 3.3.3 Sensitivity analysis

A sensitivity analysis has been carried out in DARWIN for all isotopes listed in table 2. All main contributors to their concentrations, with related sensitivities, are given in table 4.

We analyze in this paper the production of  $^{90}\text{Sr}$  and  $^{86}\text{Sr}$ .

- Production of  $^{90}\text{Sr}$  and  $^{86}\text{Sr}$

From figure 2b, discrepancy between experiment and computation is 25.5 % for  $^{90}\text{Sr}$  concentration, while propagated uncertainty is 5.93 %. It should be around 12 % to match the experimental value for a  $2\sigma$  error bar. The main production chain for  $^{90}\text{Sr}$  is from  $^{90}\text{Kr}$  as shown in table (5a).

JEFF-3.1.1 uncertainty data do not include any branching ratio uncertainty for the  $^{90}\text{Kr}$  decay to Rubidium. To know the influence of this branching ratio uncertainties on isotopic concentration uncertainty, a sensitivity analysis was achieved, giving 1 % to 10 % uncertainty to this parameter (cf. table (5b)). From DARWIN results, it is proven that uncertainty on this branching ratio can

Table 4 – Main sensibilities to table 2 isotope concentrations

Isotope	Principle reaction	$\frac{\Delta X}{X}$ %	Sensitivity	Uncertainty %	$\frac{\Delta C_c}{\Delta C_{c_{total}}}$
<sup>86</sup> Sr	<sup>86</sup> Rb β <sup>-</sup>	0.107	5.55E-04	3.57E-05	
	<sup>85m</sup> Kr → <sup>85</sup> Rb	0.509	9.22E-01	0.465	
	<sup>85</sup> Br → <sup>85m</sup> Kr	2.069	-8.64E-06	1.79E-05	72.0 %
	<sup>235</sup> U (n,f <sub>thermal</sub> ) <sub>Yi</sub> <sup>85</sup> Sr	9.884	4.96E-01	5.160	
	<sup>235</sup> U (n,f <sub>fast</sub> ) <sub>Yi</sub> <sup>85</sup> Sr	12.514	1.21E-02	0.145	
<sup>90</sup> Sr	<sup>239</sup> Pu (n,f <sub>fast</sub> ) <sub>Yi</sub>	36.694	2.31E-05	8.46E-04	
	<sup>239</sup> Pu (n,f <sub>thermal</sub> ) <sub>Yi</sub>	36.500	4.96E-03	0.181	
	<sup>235</sup> U (n,f <sub>fast</sub> ) <sub>Yi</sub>	36.927	3.84E-05	1.42E-03	
	<sup>235</sup> U (n,f <sub>thermal</sub> ) <sub>Yi</sub>	37.677	3.93E-03	0.148	74.9 %
	<sup>90m</sup> Rb β <sup>-</sup>	0.411	2.48E-01	0.102	
	<sup>90</sup> Rb β <sup>-</sup>	3.160	-3.77E-06	1.19E-05	
	<sup>235</sup> U (n,f <sub>thermal</sub> ) <sub>Yi</sub> <sup>90</sup> Kr	8.130	5.65E-01	4.592	
	<sup>235</sup> U (n,f <sub>fast</sub> ) <sub>Yi</sub> <sup>90</sup> Kr	10.200	1.16E-02	0.118	
<sup>156</sup> Gd	<sup>155</sup> Gd (n,γ)	3.836	4.66E-004	1.79E-3	
	<sup>154</sup> Eu (n,γ) <sup>155</sup> Eu	11.897	1.57E-01	1.862	
	<sup>239</sup> Pu (n,f <sub>thermal</sub> ) <sub>Yi</sub> <sup>151</sup> Pr	17.584	4.50E-02	0.791	
	<sup>239</sup> Pu (n,f <sub>fast</sub> ) <sub>Yi</sub> <sup>151</sup> Pr	18.160	5.15E-04	9.35E-03	47.0 %
	<sup>155</sup> Eu (n,γ) <sup>156</sup> Eu	19.319	4.92E-02	0.951	
	<sup>152</sup> Sm (n,γ) <sup>153</sup> Sm	2.830	2.58E-01	0.732	
	<sup>153</sup> Eu (n,γ) <sup>154</sup> Eu	4.860	4.13E-01	2.010	
<sup>239</sup> Pu	<sup>239</sup> Np β <sup>-</sup>	0.169	-1.32E-02	2.24E-03	58.6 %
	<sup>238</sup> U (n,γ) <sub>σ</sub> <sup>239</sup> U	0.856	9.60E-01	0.821	
<sup>241</sup> Am	<sup>239</sup> Pu (n,γ) <sup>240</sup> Pu	2.295	6.71E-01	1.540	
	<sup>240</sup> Pu (n,γ) <sup>241</sup> Pu	1.931	2.21E-01	0.427	79.8 %
	<sup>238</sup> U (n,γ) <sup>239</sup> U	0.856	9.90E-01	0.847	
	<sup>241</sup> Pu β <sup>-</sup>	0.028	8.82E-01	0.025	
<sup>245</sup> Cm	<sup>244</sup> Cm (n,γ)	14.691	8.75E-01	1.285	77.2 %
<sup>246</sup> Cm	<sup>245</sup> Cm (n,γ)	16.242	9.07E-01	1.473	64.8 %

Table 5 – Sensitivity analysis of <sup>90</sup>Sr

(a) Production of <sup>90</sup> Sr		(b) Branching ratio sensitivity analysis		
<sup>90</sup> Kr	$\begin{array}{l} \nearrow \\ \searrow \end{array} \begin{array}{l} \text{\textsuperscript{90m}Rb} \\ \text{\textsuperscript{90}Rb} \end{array} \begin{array}{l} \searrow \\ \nearrow \end{array} \text{\textsuperscript{90}Sr}$	$\frac{\Delta Br}{Br}$ (%)	Sensitivity	Uncertainty %
		1 %	0.108	0.108
		10 %	0.108	1.08

only contribute to up to 1 % to <sup>90</sup>Sr concentration (with the hypothesis of a 10% uncertainty on the branching ratio). In conclusion, completion of the library would be a plus, but would not alone explain the discrepancy on this isotope.

From table (4), sensitivities of β decay of nuclide <sup>90m</sup>Rb and thermal fission of <sup>235</sup>U to <sup>90</sup>Kr are

respectively  $S_{\beta_{90mRb}, 90Sr} = 0.248$  as well as  $S_{\gamma_{90Kr}, 235U, 90Sr} = 0.565$ . Therefore, the amount of  $^{90}Sr$  depends on that of  $^{90m}Rb$  and the reevaluation of  $^{235}U$  ( $n, f_{fast}$ )  $Y_i$   $^{90}Kr$  fission yield is necessary.

Concerning to  $^{86}Sr$ , it is shown in the table (4) that its concentration is sensitive to fission yield related to the  $\gamma$  decay of  $^{85}Kr \rightarrow ^{85}Rb$  as well as that associated to the fast fission of  $^{235}U$  to  $^{85}Sr$ , where sensitivities are respectively 0.922 and 0.496. Reevaluation of those data or a more precise distribution power in time would be a plus to fully validate concentration on this PWR case.

### 3.4 Sensitivity analysis in MENDEL

In this section, we introduce a way to compute sensitivity coefficients from MENDEL output distributions as shown in equation (5). The derivation is considered as the mean value of all linear derivations.

$$\begin{aligned}
 S_{Conc/\lambda} &= \frac{\partial Conc}{\partial \lambda} \frac{E(\lambda)}{E(Conc)} \\
 &= \frac{1}{\frac{N(N-1)}{2}} \sum_{i=1}^N \sum_{j=i+1}^N \frac{Conc_i - Conc_j}{\lambda_i - \lambda_j} \frac{\frac{1}{N} \sum_{i=1}^N \lambda_i}{\frac{1}{N} \sum_{i=1}^N Conc_i} \\
 S_{Conc/\lambda} &= \frac{1}{\frac{N(N-1)}{2}} \frac{\sum_{n=1}^N \lambda_n}{\sum_{m=1}^N Conc_m} \sum_{i=1}^N \sum_{j=i+1}^N \frac{Conc_i - Conc_j}{\lambda_i - \lambda_j} \quad (5)
 \end{aligned}$$

$N = 2000$  decay constant samples are used in MENDEL. Uncertainty propagation was done on all parameters (yields, decay parameters and cross sections). The number of computed sensitivity is  $N_s = \frac{N(N-1)}{2} = 19990000$ .

Table (6) shows MENDEL and DARWIN sensitivities associated to  $^{242}Cm$  caused by uncertain decay constants.

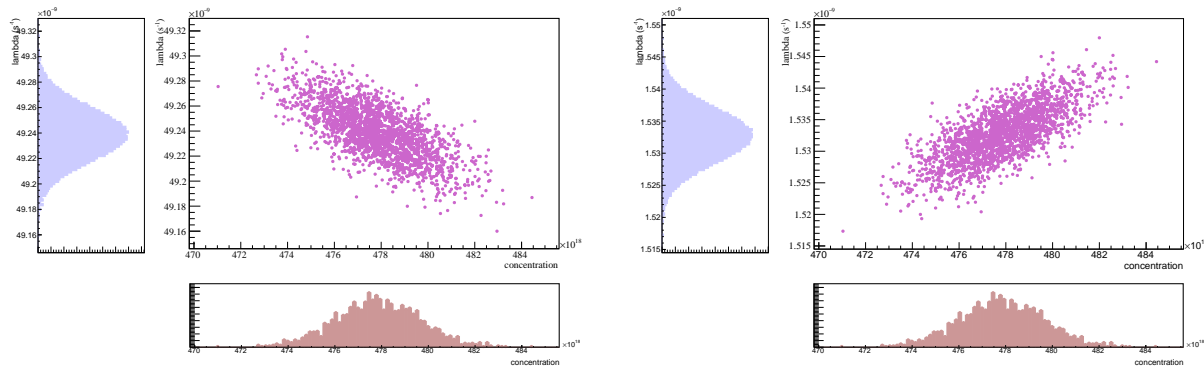
For high enough sensitivities, computation of an accurate sensibility through a 2000 samples Monte Carlo approach is possible. Nevertheless, discrepancies occur when sensitivity is lower. The best criteria found during this study to validate the sensitivity computed from samples is the use of the relative standard deviation  $\frac{\sigma_{sens}}{\mu_{sens}}$  of all sensitivity coefficients  $\left( \frac{Conc_i - Conc_j}{\lambda_i - \lambda_j} \frac{E(\lambda)}{E(Conc)} \right)$ .

We observe that the concordance is good when  $\frac{\mu_{sens}}{\sigma_{sens}}$  is less than  $1.1 \cdot 10^4$ , which also correspond to biggest sensitivities in DARWIN. Figures 4a and 4b show the sensitivity to  $\lambda^{242}Am$  and  $\lambda^{241}Pu$ .

Table 6 – Comparison of sensitivities of  $^{242}Cm$  between MENDEL and DARWIN

Isotope	$\lambda(^{242}Cm)$	$\lambda(^{241}Pu)$	$\lambda(^{242m}Am)$	$\lambda(^{242}Am)$	$\lambda(^{239}Np)$
Sens_DAR	-5.95283E+00	9.75182E-01	1.59265E-02	3.44186E-03	-2.35120E-03
Sens_MDL	-5.97778E+00	9.64403E-01	1.99953E-02	-1.10771E-01	9.212803E-02
$\frac{\sigma_{sens}}{\mu_{sens}}$ (%)	-5933.6	4736.6	101647.8	-115733.0	142980.6

In conclusion, sensitivities computed after a Monte Carlo propagation method can be used when  $\frac{\sigma_{sens}}{\mu_{sens}} < 10^5 \%$ .



(a)  $\lambda(^{242}\text{Cm})\text{-Conc}(^{242}\text{Cm})$  (b)  $\lambda(^{241}\text{Pu})\text{-Conc}(^{242}\text{Cm})$   
 Figure 4 – Decay constant  $\lambda\text{-}^{242}\text{Cm}$  concentration correlation

#### 4. CONCLUSION

On a PWR depletion and cooling case, MENDEL has given results fully compatible with already validated DARWIN code system. Furthermore, comparison to experiment data on 54 isotopes have shown validated concentrations with a  $2\sigma$  uncertainty propagation for 47 isotopes. Origin of the main uncertainty has been established, showing a preponderant effect of fission yields for fission products, and cross sections for heavy nuclides.

Sensitivity analysis have been carried out to stress out the main contributors to discrepancy for the 7 isotopes not matching the  $2\sigma$  uncertainty bars.

Sensitivity coefficients established from MENDEL coefficients for a relatively small number of realisations can be taken into account with regard of a criteria descibed in section 3.4.

In conclusion, MENDEL version can be considered as validated for PWR calculation with high burnups.

Work in undergoing to improve the computation of a statistical sensitivity coefficient, and deterministic way to propagate uncertainties in MENDEL is under development.

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