ISOTOPIC UNCERTAINTY ASSESSMENT DUE TO NUCLEAR DATA UNCERTAINTIES IN HIGH-BURNUP SAMPLES

[1,2)] O. Cabellos, [2)] J.S. Martinez and [1)] C.J. Diez

[1)] Department of Nuclear Engineering, [2)] Institute of Nuclear Fusion
Universidad Politécnica de Madrid
C/Jose Gutierrez Abascal, n2, 28006, Madrid, Spain
oscar.cabellos@upm.es

ABSTRACT

The accurate prediction of the spent nuclear fuel content is essential for its safe and optimized transportation, storage and management. This isotopic evolution can be predicted using powerful codes and methodologies throughout irradiation as well as cooling time periods. However, in order to have a realistic confidence level in the prediction of spent fuel isotopic content, it is desirable to determine how uncertainties affect isotopic prediction calculations by quantifying their associated uncertainties. The aim of this paper is to study the importance of nuclear data uncertainties in the prediction of high-burnup spent fuel content and its effect in the criticality safety assessment.

This work has been undertaken within the research activities between the Spanish Safety Council (CSN) and the Polytechnical University of Madrid (UPM) in the high-burnup spent fuel agreement. Calculations described in this paper were performed for a typical pin-cell in a water moderated infinite lattice configuration. This fuel rod with an initial ^{235}U enrichment of 4.5 wt% is irradiated to an average burnup of about 78 MWd/kgU following the operation description reported by ENUSA in the Spanish Vandellós 2 pressurized water reactor during cycles 7–11, between June 1994 and September 2000.

Key Words: Uncertainty, high-burnup, actinides, fission products

1   INTRODUCTION

With the intention of providing a base for the intercomparison of computer codes, methods and data applied in spent nuclear fuel analysis, well-defined calculational benchmarks were established in the past decades by the NEA Burnup Credit Working Group. For instance, the Phase I-B [1] was proposed in 1996 to provide a comparison of the ability of different best-estimate code systems and data libraries to predict isotopic concentrations. In addition to the computational Benchmarks, several international burnup credit experimental programs were initiated. Recently, a Spanish high-burnup fuel program coordinated by the Spanish Safety Council (CSN), ENUSA and ENRESA for High-Burnup PWR Spent Fuel has been performed [2], and the analyses carried out by ORNL have been reported in Ref. [3].

In a previous work [4], SCALE 6.0 and a revised MONTEBURNS 2.0 code were used to model the same reported geometries, material compositions and burnup history of the Spanish Vandellós II reactor cycles 7-11 [2, 3] and to reproduce measured isotopes after irradiation and decay times. We analyzed comparisons between measurements and each code results for several grades of geometrical modelization detail, using different libraries and cross-section treatment methodologies. Moreover, in this work [4] we performed a preliminary estimation of the uncertainties in the nuclide inventory assessing the potential impact of uncertainties in nuclear data. We presented a methodology to estimate uncertainty propagation to the isotopic inventory based on a Monte Carlo method. This technique is able to account for the impact of uncertainties in the basic nuclear data (cross-section, decay data and fission yields) and flux spectrum errors along the consecutive spectrum-depletion steps. A more extensive uncertainty analysis work was presented in Ref. [5] for the Phase I-B burnup credit Benchmark, with a maximum burnup of 44 GWd/TMU.

In this paper, we assess the impact of nuclear data uncertainties in some relevant actinides and fission products in high-burnup samples (up to 78 GWd/TMU) irradiated in LWR. A typical high-burnup Vandellós-II pin-cell has been chosen for this study. This activity has been undertaken within the framework of one the activities initiated by the Expert Group on Uncertainty Analysis Modeling (UAM) to address further developed and validated Uncertainty Analysis (UA) and Sensitivity Analysis (SA) methods suitable for depletion calculations [6].

2   PROBLEM DESCRIPTION AND UNCERTAINTY METHODOLOGY

All calculations described in this work were performed for a single pin-cell in a water moderated infinite lattice configurations, based on a 17x17 Vandellos II fabricated by ENUSA with an initial ^{235}U enrichment of 4.5 wt%. The burnup history corresponds to five operational cycles (7th to 11th) and 5 years of cooling time. The specific power is operated to reach a burnup of 78 GWd/TMU. In Ref. [7] a more extensive work is presented to elucidate what level of description is needed in order to reproduce isotopic content accurately and shed light on the real importance of the models precision, the involved magnitudes knowledge and, over all, the impact on isotopic calculations of position and surroundings of a specific fuel rod. Cycles 7th-11th are reproduced for several increasing burnups and compared to experimental measures.

The evolution of all the major and minor actinides is illustrated in Fig. 1, showing in the cooling time the generation of ^{241}Am from the β-decay of ^{241}Pu.

Figures 2 and 3 show the evolution of all fission products of interest taken from Ref. [2]. Fig. 2 shows fission products whose evolution does not change after 5 years of cooling time, as a consequence of their long half-lived and with negligible formation from decay of other fission products.
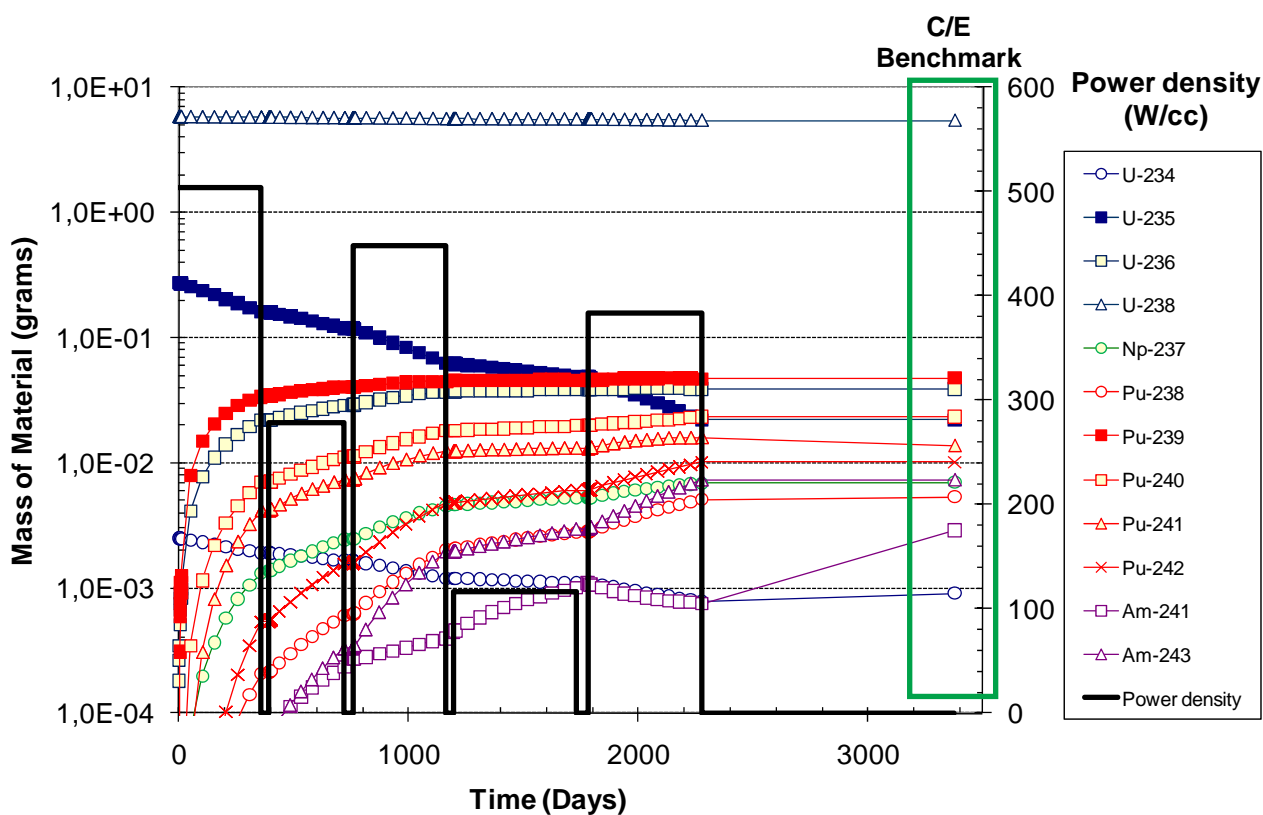

Figure 1. Major and minor actinides evolution calculated in the pin-cell for 5 operational cycles and 5 years of cooling time. Calculation performed with MCNP+ACAB codes.
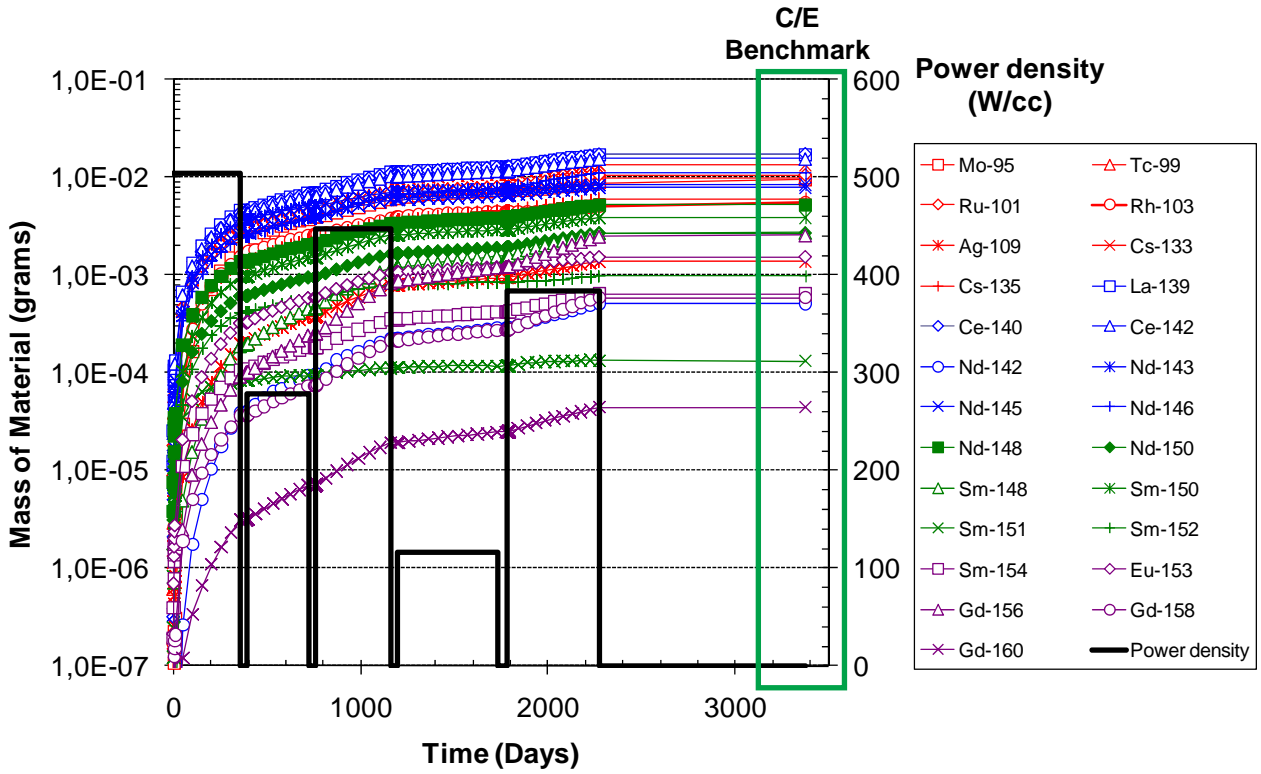
Figure 2. Fission products evolution calculated in the pin-cell for 5 operational cycles and 5 years of cooling time. Calculation performed with MCNP+ACAB codes.

Figure 3. Fission products evolution calculated in the pin-cell for 5 operational cycles and 5 years of cooling time. Calculation performed with MCNP+ACAB codes.
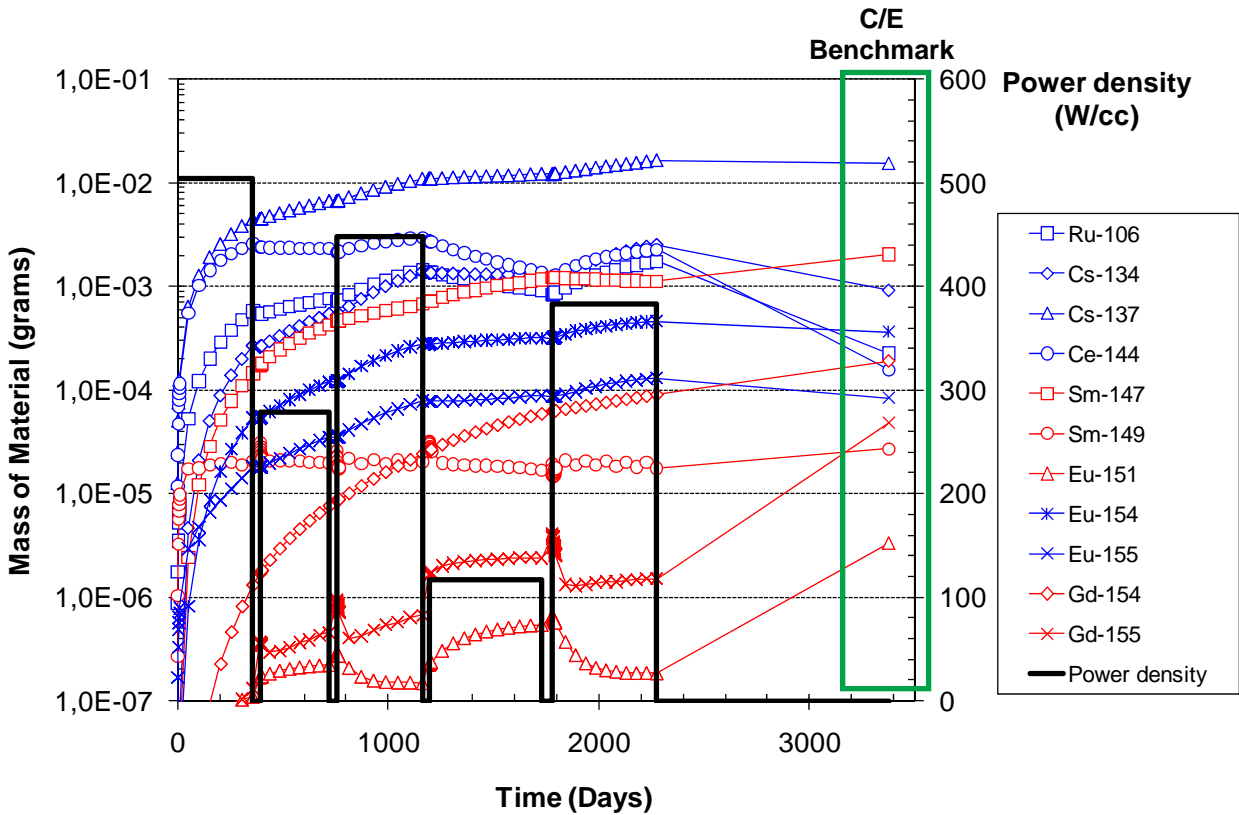
In Fig. 3, isotopes with a significant evolution after shutdown are showed. Concentrations of [106]Ru, [134,137]Cs, [144]Ce and [154,155]Eu are decreased due to itself decay process. The rest of the isotopes increase its concentration due to beta decay process: [147]Sm (~46% of the formation of [147]Sm in this cooling time period is due to β-decay of [147]Pm), [149]Sm(~37% due to [149]Pm), [151]Eu(~94% due to [151]Sm), [154]Gd (~68% due to [154]Eu)and [155]Gd(~99% due to [155]Eu).

2.1 Uncertainty Methodology

The set of differential equations which describe the evolution of N in a neutron field may be written in matrix notation

$$\frac{dN(t)}{dt} = AN = \left[\lambda\right]N + \left[\sigma^{eff}\right]\Phi N + \left[(\gamma\sigma_{fiss})^{eff}\right]\Phi N \tag{1}$$

where A is the transition matrix involving the M-by-M matrix for the one-group effective cross sections [σ_{eff}], one-group effective fission yield cross section [$(\gamma\sigma_{fiss})^{eff}$] and decay values [$\lambda$]. Φ is the space-energy integrated neutron flux. Given the initial nuclide density vector as $N_0 = N(0)$, the solution is $N(t) = \exp(At)N_0$. ACAB computes the isotopic concentrations at the end of each burn step, taking the fluxes halfway through each burn step determined in the best-estimated calculation. (see Fig. 4)

The uncertainty methodology presented in this work is based on two steps [8]. In a first step, a coupled neutron-depletion calculation is carried out only once, taken the best-estimated values for neutron spectra. That is, when solving the transport equation to calculate the flux distribution for each time step, nor uncertainties in the input parameters nor statistical fluctuations are taken into account. This is called the best-estimated multi-step calculation. In a second step, the uncertainty analysis to evaluate the influence of the uncertainties in nuclear data involved in the transmutation process on the isotopic inventory is accomplished by the ACAB [9] code.

In this work, we assume no uncertainties in: (i) the initial nuclide density, (ii) the integrated neutron flux, (iii) and the flux spectrum. In summary, the sources of uncertainty in this transmutation calculation are only due to basic input nuclear data. We define a random vector $\alpha=$ (σ_{eff}, λ, γ_{eff}) containing all the cross sections, decays and fission yields involved in the problem. Each concentration at time t, N(t), is a function of the random vector α. To perform the uncertainty analysis we have used a random simulation or Monte Carlo (MC) method. Different assumptions can be made about the probability distribution; the simplest and more usual (in many other areas) is the normal distribution. An alternative distribution is the log normal, that is, $\log(\alpha/\hat{\alpha}) \to N(0,V)$, where V is the variance matrix of the nuclear data relative error. Then, ACAB code is used to propagate the overall nuclear data set. A 300 histories sample size is found appropriate for this application. A statistical analysis of the results allows assessing the uncertainties in the calculated number densities.

2.2 Nuclear Data Uncertainties

The nuclear data basic libraries used in this work are the following: (i) the multigroup activation neutron cross-section basic library EAF_N_XS-2007 [10], (ii) the decay data basic library JEFF-3.1.1 [11], and (iii) the fission yield basic library, JEFF-3.1.1[11].

The decay and fission yield uncertainty data have been taken and processed directly from JEFF-3.1.1. The neutron cross-section uncertainty data have been taken from the EAF-2007-2010/UN [12] library and SCALE6.0/COVA-44G [13]. EAF2007 and 2010/UN contain uncertainty information for all the reactions and isotopes potentially present in the irradiated fuel. Its main characteristics are: (i) below 20 MeV, the energy spectrum is divided in three energy groups for nuclear reactions without threshold and in one group for reactions with threshold; (ii) neglecting all type of correlations between different isotopes and reactions, (iii) errors between all bands inside a certain energy group are 100% correlated, and errors between different energy groups are 0%

correlated; iv) the uncertainty values stored in the library are $\Delta^2_{j,EAF}$ (j- is the energy group), where can be interpreted as the uncertainty (or relative error) in the standard or best-estimate cross section, stored in the corresponding standard activation library. For this analysis, the values of $\Delta_{j,EAF}$ are taken as three times the experimental uncertainty, that is, $\Delta_{j,EAF}=3*\Delta_{j,EXP}$.
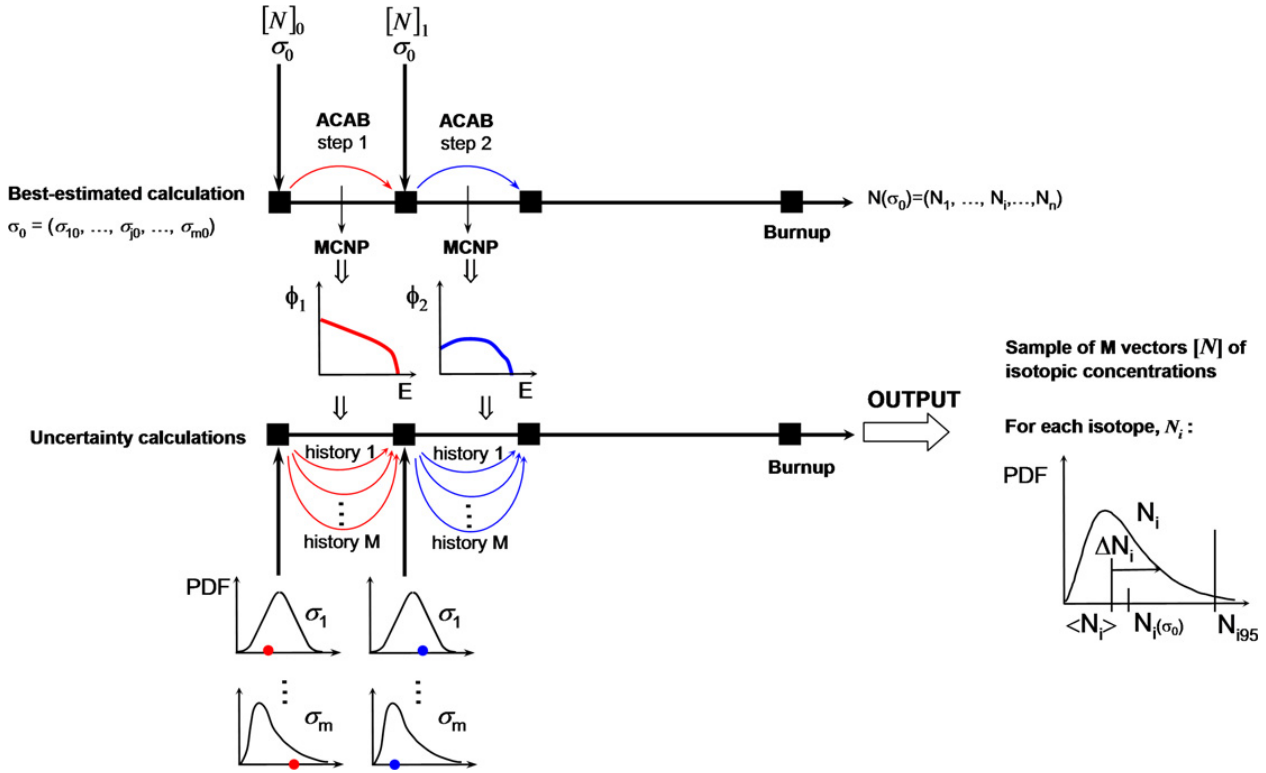

Figure 4. Monte Carlo method scheme implemented in MCNP-ACAB system to propagate uncertainties in final concentrations [Ref. 8].

SCALE6.0/COVA-44G is a 44-group cross section covariance matrix library retrieved from the SCALE-6.0 package. This covariance library is based on several different uncertainty approximations with varying degrees of "fidelity" to the actual ND evaluation. The library includes evaluated covariances obtained from ENDF/B-VII, ENDF/B-VI, and JENDL3.3. Correlations between different isotopes and reactions are included.

3   UNCERTAINTY ANALYSIS RESULTS

The Monte Carlo technique for nuclear data uncertainty propagation will allow estimating the uncertainties at different burnup/cooling times for the isotopic inventory. The importance of this effect for criticality safety analysis will be assessed with the uncertainty prediction of k_{eff}. In this case TSUNAMI [14] code will be used to predict the sensitivity coefficients of different isotope and reactions. The combination of the isotopic uncertainties with these sensitivity coefficients will permit to evaluate the total uncertainty contribution in k_{eff}.

3.1 Uncertainty isotopic prediction

Tables I and II show the calculated uncertainties in actinides and light-elements after 5 years of cooling time for this high-burnup sample irradiated at 78 GWd/TMU. And, taking advantage of some previous works [15] a list of design target accuracies can be established for high burn-up PWRs, assuming a maximum relative error in transmutation of 5% and for k_{eff} a maximum relative

error of 0.5%. As Ref. [15] suggests, these target accuracies should reflect the perceived state of the art from an R&D point of view.

Table I shows the actinide uncertainty for samples irradiated up to 78 GWd/TMU and after 5 years of cooling time. It can be seen that actinide concentration uncertainties due to decay data uncertainties remain very low. Only [243]Cm reaches 0.4% due to the 6.7% half-life relative error of [243]Cm. Regarding cross-section data uncertainties, it can be seen that EAF2007/UN does not fulfill accuracy requirements for [243,247,248]Cm and [250,251,252]Cf. EAF2010/UN and SCALE6/COVA do not fulfill for [250,251]Cf. In general, EAF2010/UN improves its value against EAF2007/UN, except for [238]Pu and [242]Cm. For major actinides, SCALE6.0/COVA fulfils better requirements, and some minor actinides are better fulfilled with EAF2010/UN.

Table I. Calculated uncertainties in actinides due to cross-section and decay data uncertainties for high-burnup.

Isotope	Decay	Cross-sections uncertainties		
		EAF2007	EAF2010	SCALE6.0
233U	0.1	2.5	1.9	2.1
234U	0,1	4,6	2,6	3,1
235U	0,0	1,1	1,1	0,2
236U	0,0	0,7	0,6	0,3
238U	0,0	0,3	0,3	0,1
237Np	0,0	1,3	1,5	0,7
238Pu	0,0	1,3	0,9	0,4
239Pu	0,0	1,6	1,7	0,5
240Pu	0,0	3,0	2,2	0,7
241Pu	0,0	2,1	1,6	0,5
242Pu	0,0	2,1	1,0	1,6
241Am	0,2	2,0	1,5	0,5
243Am	0,0	2,8	1,4	2,8
242Cm	0,2	2,1	1,5	0,6
243Cm	0,4	6,9	4,0	2,9
244Cm	0,1	2,5	1,1	2,1
245Cm	0,0	3,4	2,4	4,2
246Cm	0,0	4,5	1,9	2,9
247Cm	0,0	5,4	2,4	3,8
248Cm	0,0	7,4	3,0	4,0
250Cf	0,1	8,8	5,5	5,2
251Cf	0,1	9,4	6,5	6,0
252Cf	0,2	7,8	3,4	4,6

Fig. 5 shows that the uncertainty raises with burnup for [234,235,238]U, for other actinides the level of uncertainty decreases with burnup and it is maintained constant in the cooling time period, except for [241]Am and [234]U.

Table II shows the light-elements uncertainty for samples irradiated up to 78 GWd/TMU and after 5 years of cooling time. For decay data uncertainties, the isotope [151]Eu reaches a maximum uncertainty of 7.3% as a consequence of the 6.7% relative error in the half-life of [151]Sm. The uncertainties due to fission yields remain below 5%, [95]Mo with 4.5% (with high sensitivity to [95]Zr fission-yield [16]) and [149]Sm with 4.8% (with high sensitivity to [149]Pm fission-yield [16]).

Table II. Calculated uncertainties in light-elements due to cross-section, fission-yields and decay data uncertainties for high-burnup.

Isotope	Fission-Yields	Decay	Cross-sections uncertainties		
			EAF2007	EAF2010	SCALE6.0
95Mo	4.9	0.0	0.6	0.5	0.2
99Tc	1.4	0.0	0.5	0.6	0.2
101Ru	1.8	0.0	0.5	0.5	0.3
106Ru	2.4	0.6	0.6	0.6	0.2
103Rh	1.7	0.0	4.0	1.1	0.6
109Ag	1.5	0.0	5.6	4.9	0.6
133Cs	1.1	0.0	0.7	0.4	0.5
134Cs	1.1	0.0	2.2	1.5	1.3
135Cs	1.1	0.0	1.5	1.0	0.5
137Cs	1.4	0.0	0.4	0.4	0.2
139La	1.5	0.0	0.4	0.4	0.2
140Ce	1.5	0.0	0.4	0.4	0.2
142Ce	1.4	0.0	0.4	0.4	0.2
144Ce	2.4	0.2	0.6	0.7	0.2
142Nd	1.6	0.0	2.2	1.8	1.2
143Nd	1.5	0.0	0.7	1.1	0.6
145Nd	1.5	0.0	0.7	0.5	0.5
146Nd	1.0	0.0	0.6	0.4	0.4
148Nd	1.2	0.0	0.4	0.4	0.2
150Nd	1.5	0.0	0.4	0.4	0.2
147Sm	1.7	0.0	1.8	0.6	1.7
148Sm	1.5	0.0	1.2	0.6	1.0
149Sm	4.8	0.0	13.8	3.5	6.6
150Sm	1.1	0.0	1.3	0.6	1.3
151Sm	2.8	0.2	4.0	3.0	5.0
152Sm	0.9	0.0	3.2	1.1	1.6
154Sm	1.5	0.0	0.5	0.5	0.2
151Eu	2.7	7.3	4.0	3.0	5.0
153Eu	0.9	0.0	8.6	6.0	1.2
154Eu	0.9	0.0	17.3	9.8	4.9
155Eu	1.5	0.1	29.4	9.1	7.3
154Gd	0.9	0.0	10.0	5.7	3.0
155Gd	1.5	0.2	29.1	9.0	7.2
156Gd	0.8	0.0	4.6	2.3	0.8
158Gd	1.1	0.0	9.0	1.7	1.2
160Gd	3.3	0.0	0.9	0.6	0.2

As we have seen for actinides, EAF2007/UN has the highest uncertainties, reaching values above 10% relative error for [149]Sm, [154,155]Eu and [154,155]Gd, between 5 to 10% for [109]Ag, [153]Eu and [158]Gd. EAF2010/UN improves uncertainties against EAF2007/UN, showing a dramatic reduction for *Eu* and *Gd* elements. SCALE6/COVA does not fulfill 5% target for: [149,151]Sm, [151,155]Eu and [155]Gd. In the case of [155]Gd (generated by β-decay of [155]Eu), it shows higher sensitivities to [153,155]Eu(n,γ) reaction and [155]Eu fission-yield [16]. For [149]Sm (important contribution generated by β-decay of [149]Pm) the higher sensitivities are due to [149]Sm(n,γ) and [149]Pm fission yield [16].

Figures 6 and 7 show the relative error for several fission-products with the irradiation time and cooling time. It can be seen an increase of the relative error in ^{155}Gd induced by β-decay of ^{155}Eu.
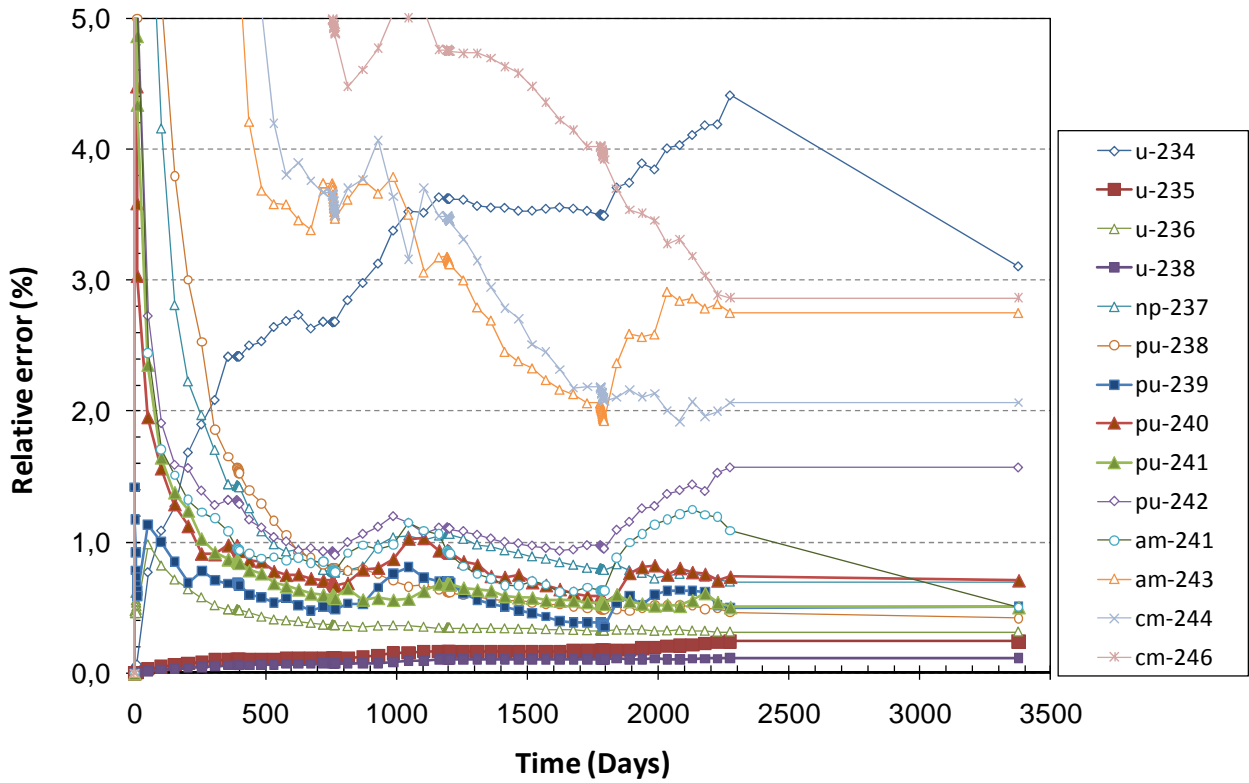
Figure 5. ΔN/N (%) for actinides predicted with Monte Carlo Method due to uncertainties in the cross-section data taken from SCALE6.0/UN.

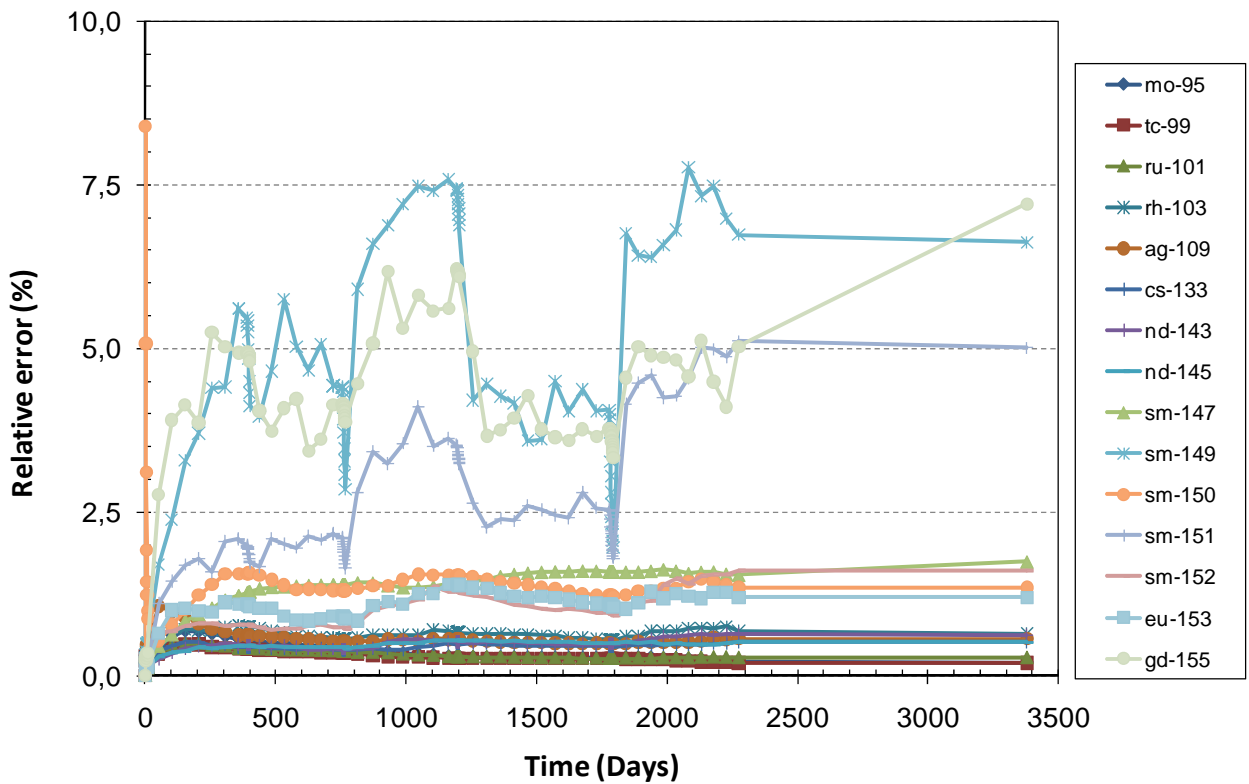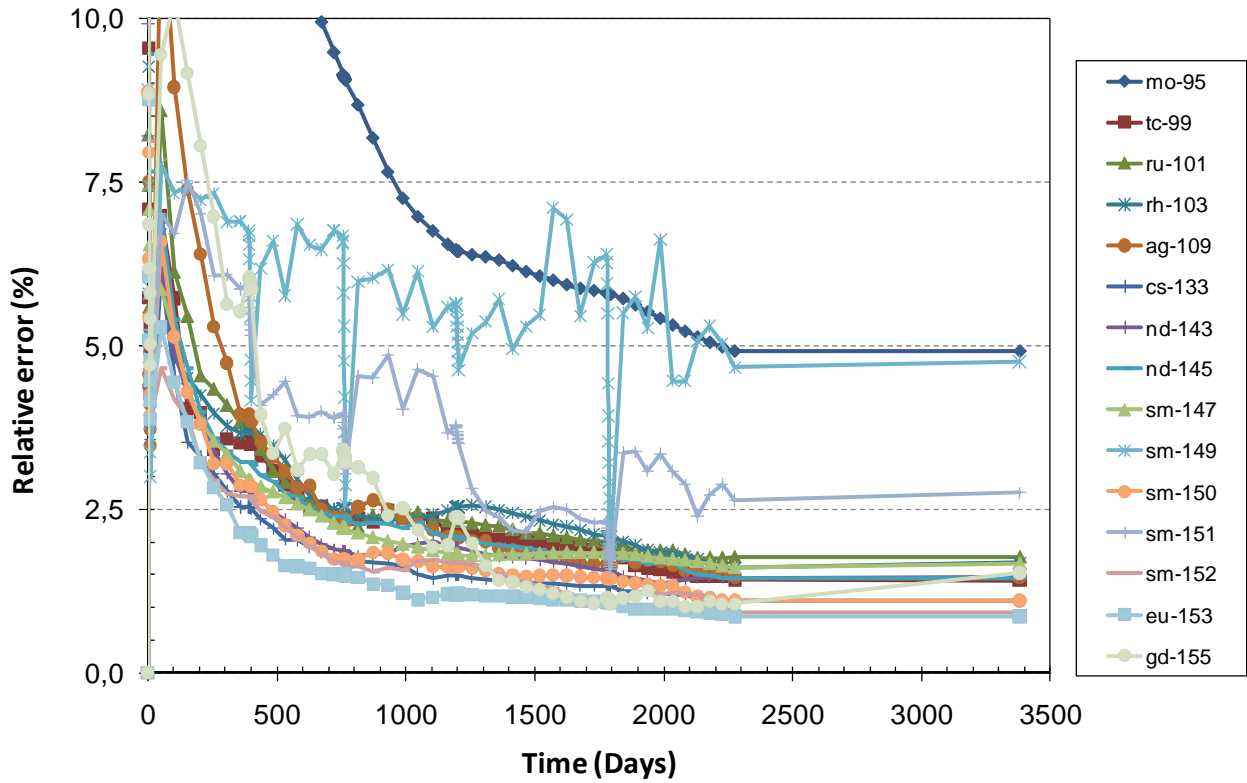**Figure 6. ΔN/N (%) for fission-products predicted with Monte Carlo Method due to uncertainties in the cross-section data taken from SCALE6.0/UN.**

Figure 7. ΔN/N (%) for fission-products predicted with Monte Carlo Method due to uncertainties in the Fission Yields taken from JEFF-3.1.1/FY.

3.2 Uncertainty criticality safety evaluation.

In this section, the assessment of uncertainty criticality safety evaluation in the prediction of k_{eff} is performed taking into account the nuclear data uncertainties. Being k_{eff} the magnitude to be analyzed where it is *explicitly* dependent on the nuclear data (e.g. cross-sections, nu-bar, …) and *implicity* dependent on the number densities which characterize the system. The relative error in k_{eff} can be defined as follows

$$\frac{\Delta k}{k} = \sum_j S_j \frac{\partial \sigma_j}{\sigma_j} + \sum_k \left[\frac{\Delta k / k}{\Delta N / N} \right]_k (\Delta N / N)_k \qquad (2)$$

the first *explicitly* term is calculated using TSUNAMI code [14] showing in Fig. 8 the evolution with burn-up. TSUNAMI predicts ~0.50% relative error in k_{eff} for fresh fuel, and ~0.90% at EOC (78 GWd/TMU). This Figure shows also the most important reactions and nuclear data, at BOC: ^{235}U(nu-bar), ^{238}U(n,n') and ^{235}U(n,γ), and at EOC: ^{239}Pu-nubar, ^{238}U(n,n'), ^{238}U(n,γ), ^{239}Pu(n,fission) and ^{239}Pu(fission-capture). The second term of equation 2 indicates the *implicitly* term. These sensitivity coefficients for isotopes in spent fuel are showed in Fig. 9. For simplicity, only the most important sensitivities for actinides are shown: ^{235,238}U and ^{239,240,241}Pu. For fission products, only ^{149}Sm and ^{103}Rh are illustrated, the rest on the light-elements has lower sensitivities. Consequently, these isotopes will have a dominant effect in the total *implicit* uncertainty term. As we have seen in Table I, isotopic uncertainties of ^{235}U, ^{238}U and ^{239}Pu will have a negligible effect due to its low uncertainty; and ^{240}Pu and ^{241}Pu will induce the main contribution. In Fig. 10, the total *explicitly* and *implicitly* uncertainty is shown. For *implicitly* uncertainty, the importance of different source of nuclear data uncertainties is identified. At EOC, it can be concluded that decay data and fission yield data uncertainties have a negligible effect in k_{eff} uncertainty, with 6 pcm and 74 pcm respectively. Larger uncertainties are induced by cross-section uncertainties, EAF2007/UN and EAF2010/UN with 590 and 400 pcm, respectively. SCALE6/COVA leads to an uncertainty of 182 pcm.
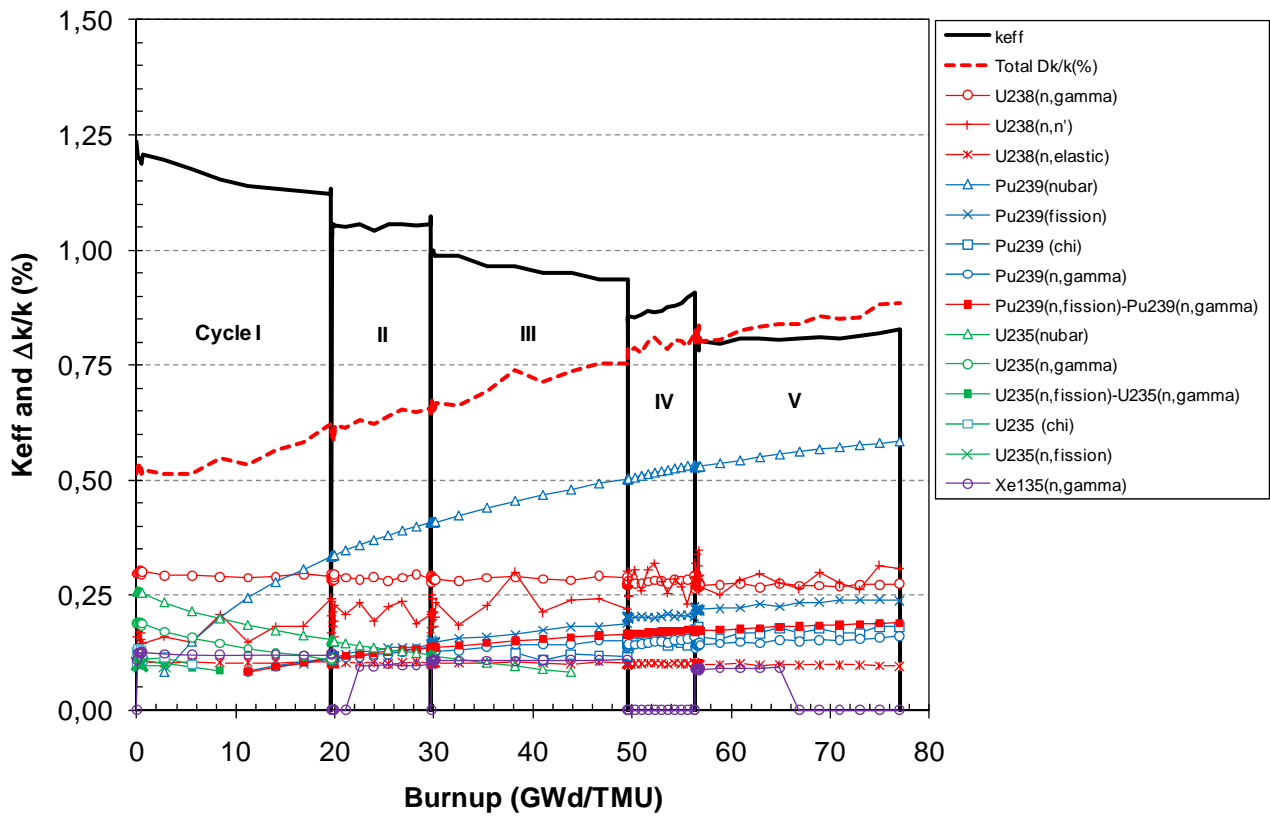
Figure 8. k$_{eff}$ and *explicitly* term of Δk/k (%), including the most important contributions calculated with TSUNAMI.
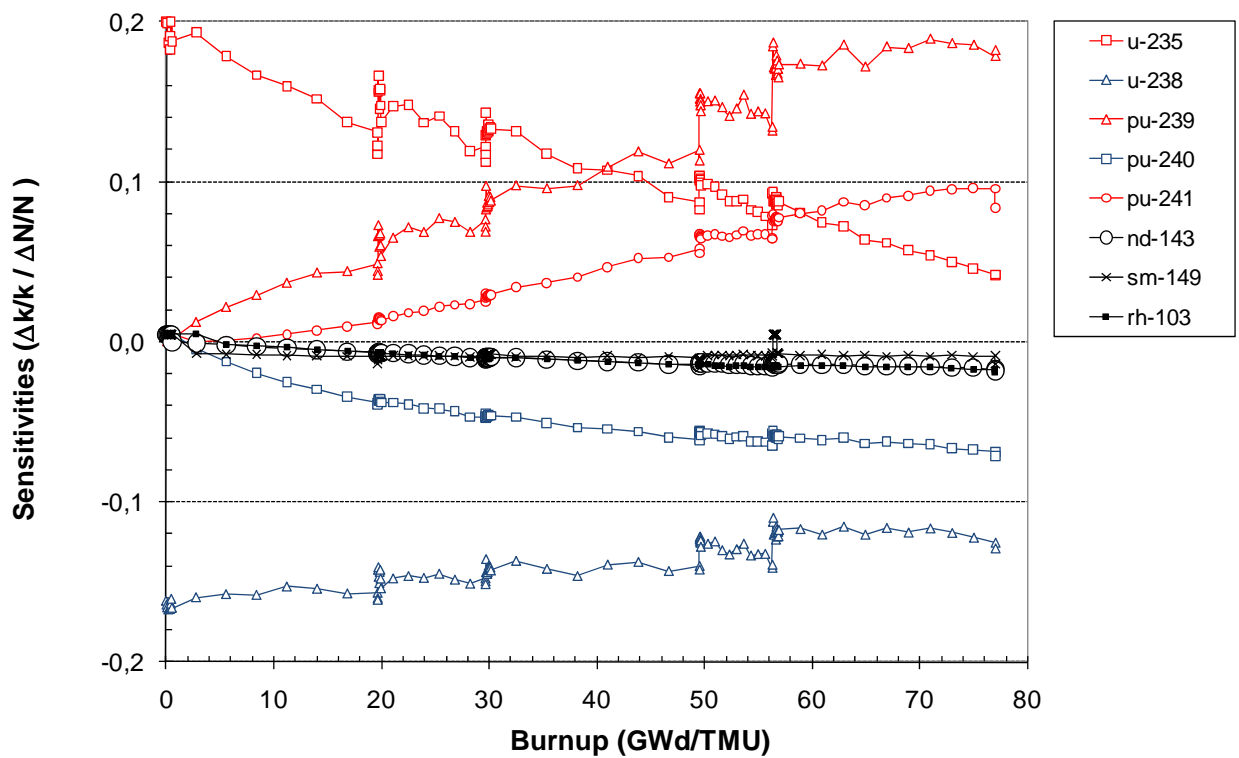

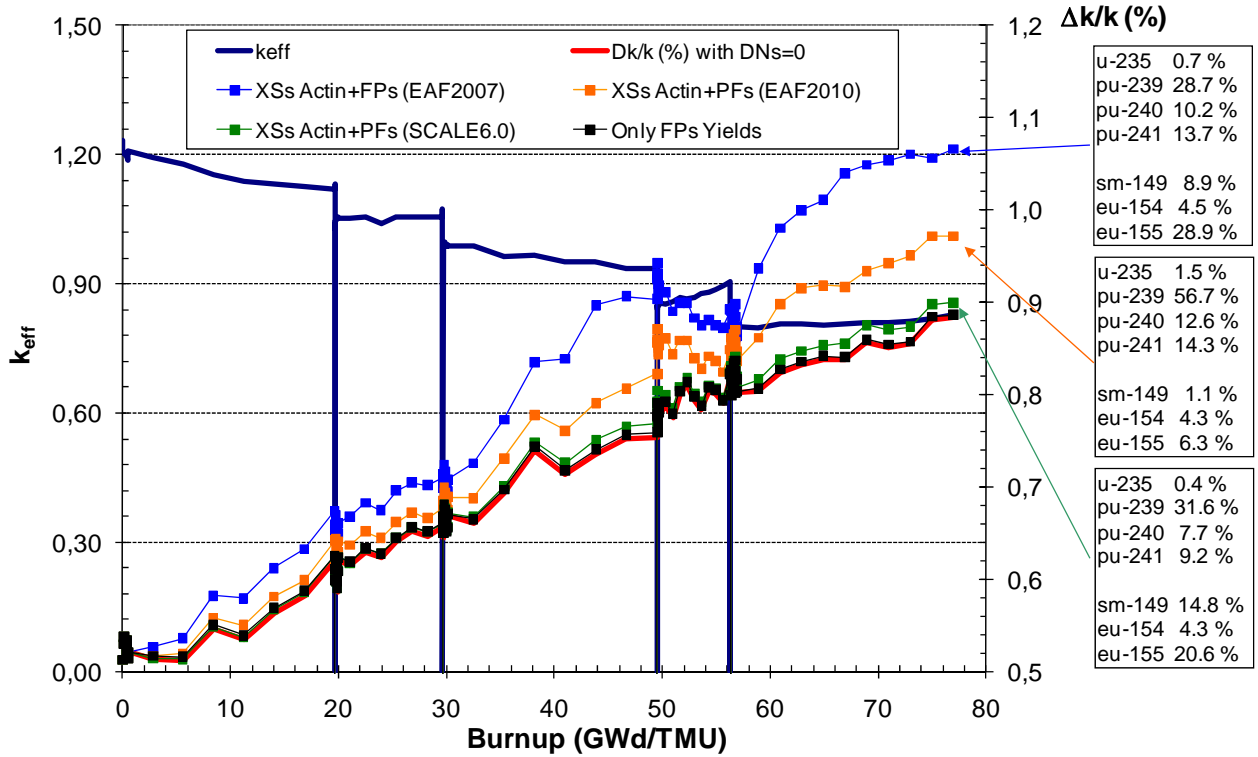**Figure 9. Sensitivities (Δk/k /ΔN/N) predicted with TSUNAMI for the most important contributions by isotopes.**

Figure 10. Total term of Δk/k (%) due to uncertainties in the isotopic inventory and nuclear data.

In addition, Fig. 10 shows the most important contributors of this *implicitly* uncertainty term, for actinides ^{239,240,241}Pu, and for light-elements ^{154,155}Eu and ^{149}Sm.

4   CONCLUSIONS

We have presented a methodology based on Monte Carlo method to deal with uncertainty propagation in transmutation calculations. The impact of nuclear data uncertainties (cross-section, fission-yields and decay data) is assessed in a high-burnup LWR sample irradiated up to 78 GWd/TMU. We conclude for this prediction that: (i) decay data uncertainties have a negligible effect on the isotopic prediction both in actinides and fission products, the maximum relative errors are found for ^{155}Eu (7.3%), ^{106}Ru (0.6%) and ^{243}Cm (0.4%) (ii) fission yield data uncertainties induce relative errors in fission products below 5%, the maximum relative errors are found for ^{95}Mo (4.9%) and ^{149}Sm (4.8%); and (iii) larger uncertainties in concentrations were found due to cross section data uncertainties. Regarding these uncertainty data for cross sections, EAF2007/UN seems to be very conservative and EAF/UN is really improved with EAF2010; SCALE6.0/COVA reaches lower uncertainties for major actinides, with similar level of uncertainties than EAF2010/UN for minor actinides and fission products. Larger uncertainties than 4% were found for actinides: ^{243}Cm, ^{250,251}Cf in EAF2010/UN and ^{245}Cm, ^{250,251,252}Cf in SCALE6.0/C0VA. And for fission-products: ^{109}Ag, ^{153,154,155}Eu, ^{154,155}Gd in EAF2010/UN and ^{149,151}Sm, ^{151,154,155}Eu, ^{155}Gd in SCALE.6/COVA.

We have performed a burnup criticality uncertainty analysis for this high-burnup LWR pin-cell with TSUNAMI code showing that relative error in k^{eff} at EOC can reach ~900 pcm, being the most important source of uncertainty: ^{239}Pu(nu-bar), ^{238}U(n,n'), ^{238}U(n,γ) and ^{239}Pu(n,fission). At BOC, with low relative error in k^{eff} ~500 pcm, being ^{235}U(nu-bar), ^{238}U(n,n') and ^{235}U(n,γ) the most important reactions. The contribution of the concentration uncertainty of major-minor actinides and fission products is assessed. We have obtained: i) negligible effect due to decay data uncertainty, ii) small contribution of 74 pcm due to fission yields uncertainty data, iii) significant contribution due to cross-section uncertainty data, 400 pcm from EAF2010/UN and 182 pcm from SCALE6/COVA.

Calculations with other uncertainty depletion methodologies based on Total Monte Carlo techniques would be valuable to assess both the importance of nuclear data uncertainties and the coupling neutron-depletion in this problem. TMC, NUDUNA and XSUSA methodologies should be candidates to deal with this problem in future works.

5   ACKNOWLEDGMENTS

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