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# Experimental Validation of the APOLLO2 code for High Burnup MOx Fuel. JEF2.2 Results and JEFF3.0 Improvements.

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This paper presents the APOLLO2 analysis of a destructive experiment of MOx fuels from the DAMPIERRE PWR reactor using the latest versions of the JEFF European nuclear data library. This analysis has demonstrated the ability of the current industrial code APOLLO2.5 and its associated library CEA93.V6 to predict actinide inventory in high-burnup MOx fuel up to 60GWd/tHM.

This interpretation confirms the major JEF2.2 nuclear data trends provided by the experiments performed in France and shows the improvement given by JEFF3.0 in the prediction of isotopic fuel contents, mainly $^{236}$U, $^{237}$Np and $^{242}$Pu. These MOx results are consistent with the satisfactory C/E comparison obtained with JEFF3.0 on UOx spent fuel inventory, except for $^{239}$Pu which remains overestimated: +4% $\pm$2% at 50GWd/tHM and +7% $\pm$3% at 60GWd/tHM. Furthermore, it is expected that the next JEFF3.1 evaluation will improve the $^{239}$Pu C/E agreement as the $^{238}$U effective integral resonance should be decreased by 1%.

***KEYWORDS : Validation,*** APOLLO2***, Post Irradiation Experiments, MOx Fuel, High Burnup, JEFF Project,*** DAMPIERRE

## 1. Introduction

Since 1987, plutonium is industrially recycled in French PWRs. The fuel management strategy in France includes recycling plutonium in the twenty 900MWe PWRs loaded with 30% MOx assemblies of the core. The current "hybrid" fuel management in mixed core loading is based on 3 irradiation cycles, leading to MOx 5.3% Pu fuel burnup of about 40 GWd/t. Fresh UOx assemblies are burnt for 4 irradiation cycles up to 50 GWd/t.

Industrial target in the distant future is to reach higher fuel burnups up to 80 GWd/t for UOx and up to 60GWd/t for MOx. In the near future, the objective is to burn Pu fueled assemblies up to 4 irradiation cycles like for current UOx assemblies. This industrial target called "MOx Parity" is a major program for the French industry. Therefore, MOx assemblies with high plutonium content and degraded Pu isotopic vector will be loaded into 900MWe PWRs. The current average Pu amount in MOx is 5.3% and will increase up to 7.7% in the MOx Parity design.

In this industrial context, it must be demonstrated that fuel lattice codes and data (and particularly the deterministic multigroup transport code APOLLO2 [1] developed at CEA and used by EdF and FRAMATOME) are able to predict the main neutronic parameters within the target accuracy. Hence, new experiments were carried out in order to extend the current experimental validation, particularly fuel depletion and Actinide-FP build-up, to larger Pu amount and higher burnups in MOx fuels.

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The Post Irradiation Experiment database is currently being extended up to 80GWd/t for UOx fuels and up to 60 GWd/t for MOx fuels. Fuel rod cuts have been extracted after 2 to 5 cycles in MOx assemblies from DAMPIERRE PWR, and up to 7 irradiation cycles in UOx assemblies from Gravelines PWR. Major and minor actinides, as well as main poisoning fission products, are analyzed.

In order to extend the MOx qualification range [2] of APOLLO2 (Version 5), and its associated library CEA93.V6 based on the european nuclear datafile JEF2.2, the interpretation of chemical assays from DAMPIERRE MOx fuels is presented in this paper. The experiment analysis based on the new JEFF3.0 European File (CEA2003.V1) is also presented.

In the fisrt two sections, we will present the experimental settings of DAMPIERRE Post Irradiation Experiment (P.I.E.) and the calculation route used. In the third part, we will discuss the experimental and the simulated irradiation follow-up.

The fourth section will present the APOLLO2 interpretation results of DAMPIERRE P.I.E. at the end of the 4 and 5 irradiation cycles using JEF2.2 and JEFF3.0 nuclear data file libraries.

This work allows us to conclude with respect to improvements in nuclear data. This information is summarised in the conclusion.

## 2. Experimental settings

The 900MWe PWR DAMPIERRE reactor is characterized by a 30% MOx fuel loading. 17x17 MOx assemblies (AFA type) include three zones with different plutonium enrichments in order to flatten the assembly power distribution. The central zone is characterised by a high plutonium content (6.7%) and the peripheral zone by a small plutonium content (3.4%).

From the MOx assembly FXP0EG, one rod was extracted after 1 irradiation cycle and three rods after 3 cycles. From the MOx assembly FXP0EH (see Fig. 1), four rods were extracted after 2 cycles, three rods after 4 cycles and four rods after 5 cycles. As we can notice in Figure 1, the four fuel rods extracted after 2 cycles (positions G04, E13, D5 and B2) were replaced with four fresh $UO_2$ ($^{235}$U w/o: 3.7%) fuel rods. The three fuel rods (positions P16, M09 and E09) extracted after 4 irradiation cycles were replaced with three stainless steel rods.

This paper will present the interpretation of the destructive P.I.E. after 4 and 5 irradiation cycles on spent fuel rods (mid-active height) from the MOx assembly FXP0EH. Chemical assays have been performed to determine the concentrations in U, Np, Pu, Am, Cm isotopes, as well as Nd and Cs. Experimental uncertainties on isotopic ratios range from 0.3% to 1% ($1\sigma$).

## 3. Calculation route

The APOLLO2 depletion calculation scheme of MOx assemblies is based on the UP1 interface current method; the geometrical pattern, shown in Figure 1, involves 32 differentiated cells in the MOx assembly and is surrounded by depleted UOx assemblies. Indeed, the APOLLO2 multicell pattern enables the grouping of cells which have similar flux level, in order to improve computing efficiency; it is necessary in depletion calculation to optimise this flux discretization. A 1/8 symmetry is used in the cluster calculation.

To represent space-dependent self-shielding and nuclide concentration profile in the pellet, we subdivide the fuel pin into four radial rings. We use thin peripheral shells, 5% of the mass of the pellet at the outer ring, in order to account for the rim effect. We consider a detailed burnup chain with 20 actinides ($^{234}\text{U}\rightarrow^{247}\text{Cm}$) and 85 fission products. It was shown that actinide self-shielding must be recalculated at 4, 8, 12, 24, 38 GWd/tHM... The transport calculation, using accurate self-shielded cross-section, is performed in the XMAS 172-group structure.

In order to take into account the different fuel rod replacements during the irradiation (4 $UO_2$ rods after 2 cycles and 3 stainless steel rods after 4 cycles), we chose to replace in the calculation pattern, E05 (which corresponds to the 11th ”physical cell”) with the fresh $UO_2$ fuel rod after 20GWd/tHM, and to replace E09 (which corresponds to the 7th ”physical cell”) with the stainless steel rod after the 4th irradiation cycle. Thus, due to the 1/8 symmetry of the calculation pattern, we assume 4 replaced rods after 2 irradiation cycles and 4 replaced rods after 4 irradiation cycles.

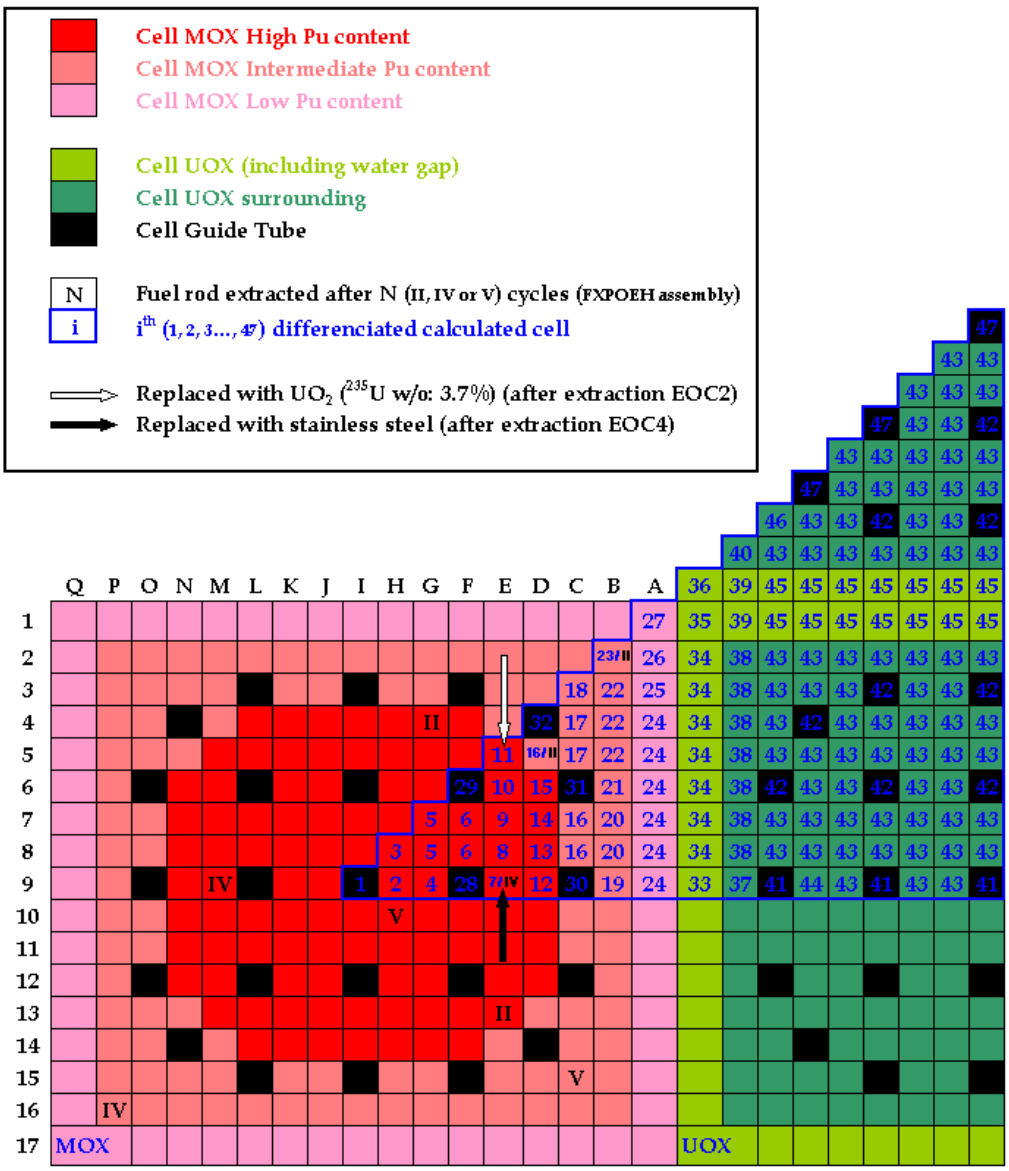


Figure 1: MOx assembly pattern

An experimental determination of the local burnup is deduced for each sample, using the $^{145}$Nd/$^{238}$U and $^{150}$Nd/$^{238}$U experimental isotopic ratios.

As an example, Figure 2 shows the burnup determination for the irradiation cycles 4 and 5 using the experimental neodynium concentration of the five experimental fuel rods. We can notice that the $^{145}$Nd/$^{238}$U concentration ratio versus burnup is quite linear and is therefore a good burnup indicator. The uncertainty on the inferred local burnup is about 1.5% ($1\sigma$).

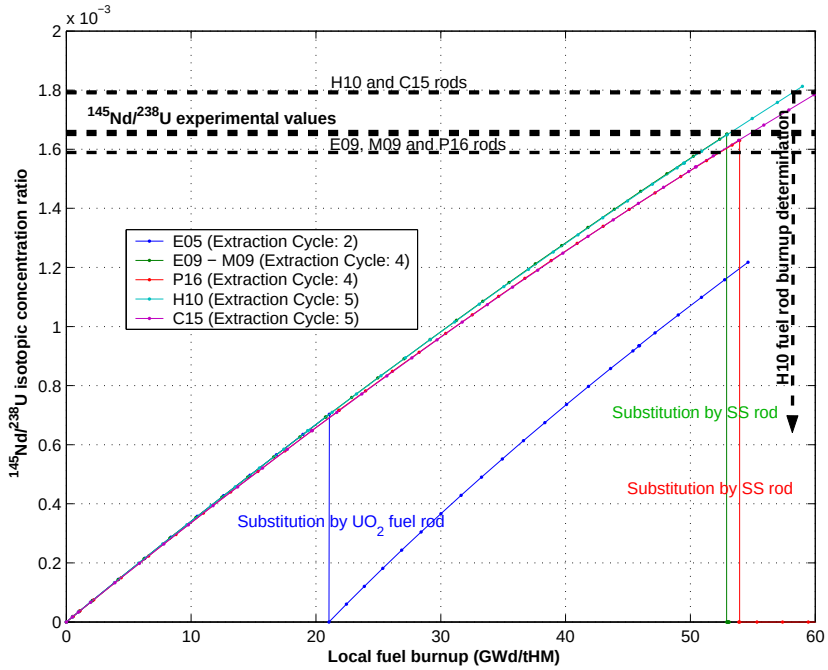


Figure 2: $^{145}$Nd/$^{238}$U isotopic ratio versus local burnup

## 4. Irradiation follow-up

Table 1 and Figure 3 show the main operating parameters (BOC and EOC dates, fuel specific energy) of the irradiation cycles.

Table 1: DAMPIERRE**-2 Irradiation cycles.**

| **Irradiation cycles in** DAMPIERRE**-2** | **Beginning of cycle** | **End of cycle** | **Estimated length of cycle (MWd/tHMi)** |
|---|---|---|---|
| 1 | 14/11/1993 | 14/10/1994 | 9059 |
| 2 | 24/11/1994 | 15/02/1996 | 12699 |
| 3 | 06/04/1996 | 28/03/1997 | 12081 |
| 4 | 21/05/1997 | 16/05/1998 | 11282 |
| 5 | 27/07/1999 | 17/06/2000 | 10460 |

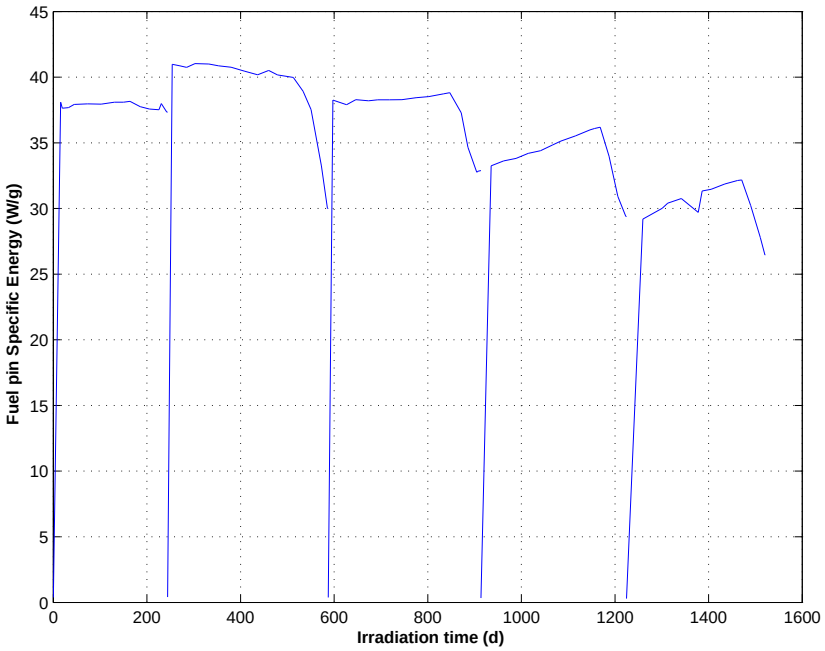


Figure 3: Estimation of the specific energy of the C15 fuel pin

The interpretation with APOLLO2 takes into account

- $^{238}$Pu and $^{241}$Pu concentration decay before the assembly loading,
- isotopic concentration modifications due to cooling during the 4-5 intercycle,
- the stretch-out with reactor power decrease, as well as moderator and fuel temperatures decrease, at the end of the 4th and the 5th cycle,
- cooling after experimental fuel pin irradiation up to the date of analysis.

The assembly average specific energy used for the depletion calculation is 32W/g and the diluted boron concentration is 600ppm, up to stretch-out characterized by $C_B$=0ppm.

## 5. JEF2.2 and JEFF3.0 results

### 5.1 Fuel depletion

The local perturbation due to the replacement of the 7 extracted fuel rods during the assembly irradiation was estimated on the C/E isotopic ratios in the fuel rods cuts.

Accounting for the fuel rod replacements in assembly depletion calculation implies:

- a decrease of the calculated $^{239}$Pu amount by about 0.5% at the end of the 4th cycle in the two analysed fuel pins and about 1% at the end of the 5th cycle for the two analysed fuel pins.
- a decrease of the calculated $^{235}$U amount by about 0.5% at the end of the 5th cycle in the two analysed fuel pins.

This is mainly due to the increase of thermal neutron flux linked to the replacement of a MOx fuel rod by a fresh UOx fuel rod at the end of the 2nd cycle.

The $^{239}$Pu isotopic concentration versus MOx assembly exposure is plotted in Figure 4.

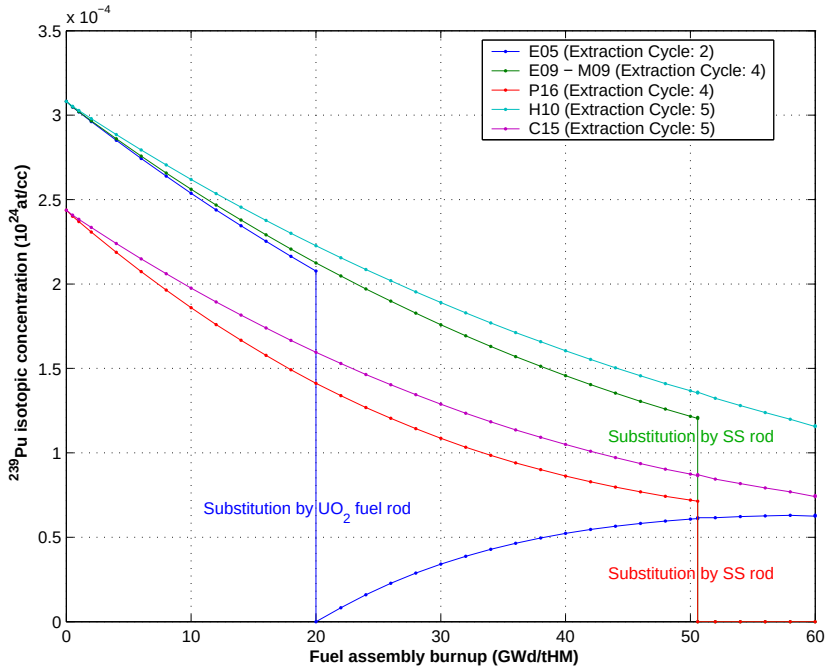


Figure 4: $^{239}$Pu depletion versus assembly burnup

We can notice that the $^{239}$Pu build-up in the replaced $UO_2$ fuel pin (at the end of the 2nd cycle) has reached the same level as the $^{239}$Pu residual amount in the intermediate MOx fuel zone at the end of the assembly irradiation.

## 5.2 JEF2.2 results

The Calculation-Experiment comparison for each P.I.E. fuel rod, corresponding to APOLLO2-CEA93.V6 industrial tool (CEA93 library is based on JEF2.2 file), are summarized in Table 2. The last two lines show the local burnup of the fuel samples determined from Nd isotopes, and the corresponding assembly burnup; the uncertainty on this sample exposure is estimated to 1.5% ($1\sigma$) mainly linked to the $^{145}$Nd and $^{150}$Nd fission yields. The total uncertainty on C/E value for $^{239}$Pu/$^{238}$U ratio amounts to $\pm$2% at 50GWd/tHM and $\pm$3% at 60GWd/tHM due to quadratic combination of burnup component uncertainty, chemical assay uncertainty, reactor history modelling, fuel and moderator temperatures.

Table 2: APOLLO2-CEA93.V6 results of DAMPIERRE MOx P.I.E. (C/E-1) in %

| JEF2.2 | 4 CYCLES | | | 5 CYCLES | |
|---|---|---|---|---|---|
| | E09 6.7% | M09 6.7% | P16 5.3% | H10 6.7% | C15 5.3% |
| U234/U238 | -3.0 | -4.2 | -0.5 | -5.1 | -0.6 |
| U235/U238 | 2.0 | 1.5 | 0.0 | 1.5 | 0.8 |
| U236/U238 | -6.9 | -6.9 | -8.3 | -7.2 | -7.1 |
| | | | | | |
| Np237/U238 | -12.9 | -10.3 | -14.0 | -10.9 | -8.3 |
| | | | | | |
| Pu238/U238 | -4.5 | -3.7 | -2.4 | -2.2 | -1.5 |
| Pu239/U238 | 4.0 | 3.4 | 4.3 | 7.1 | 8.0 |
| Pu240/U238 | 1.1 | 0.4 | 0.2 | 4.1 | 1.7 |
| Pu241/U238 | 0.4 | 0.0 | 1.7 | 3.3 | 2.5 |
| Pu242/U238 | -2.2 | -2.4 | -2.5 | -0.4 | -3.7 |
| | | | | | |
| Am241/U238 | 5.3 | 5.0 | 4.7 | 6.2 | 11.1 |
| Am241/U238 EOC | 16.1 | 16.8 | 13.0 | 8.9 | 22.6 |
| Am242M/U238 | -22.0 | -22.9 | -15.0 | -25.2 | -14.8 |
| Am243/U238 | 1.2 | 1.9 | 1.8 | 2.3 | 4.7 |
| | | | | | |
| Cm243/U238 | -12.3 | -11.6 | -6.1 | -12.4 | -8.4 |
| Cm244/U238 | -5.0 | -4.5 | -3.9 | -3.2 | -1.8 |
| Cm245/U238 | -1.3 | -0.2 | 1.8 | 1.7 | 7.5 |
| Cm246/U238 | -6.6 | -5.4 | -12.4 | -3.9 | -5.8 |
| Cm247/U238 | -12.4 | -10.8 | -19.0 | -9.1 | -8.6 |
| | | | | | |
| Nd143/U238 | 1.6 | 0.8 | 0.5 | 1.2 | 1.8 |
| Nd144/U238 | -1.6 | -2.2 | -2.5 | -2.1 | -2.0 |
| Nd145/U238 | 0.2 | -0.4 | -0.6 | -0.4 | -0.5 |
| Nd146/U238 | 1.1 | 0.7 | 0.3 | 1.1 | 1.4 |
| Nd148/U238 | 2.2 | 1.8 | 2.0 | 2.1 | 2.1 |
| Nd150/U238 | 0.4 | 0.4 | 0.6 | 0.7 | 0.8 |
| | | | | | |
| Cs133/U238 | -1.1 | nm* | 0.0 | nm | nm |
| Cs134/U238 | 0.8 | nm | 3.0 | nm | nm |
| Cs135/U238 | 7.7 | nm | 6.3 | nm | nm |
| Cs137/U238 | -1.7 | nm | -0.4 | nm | nm |
| | | | | | |
| Burnup i (MWd/t) | 52974 | 52974 | 51895 | 57927 | 59935 |
| | | | | | |
| Burnup ASS (GWd/t) | 50.0 | | | 59.6 | |

nm*: Not yet Measured.

The C/E biases obtained in the three different MOx pins, after four irradiation cycles, are very consistent. Chemical assays in fuel samples irradiated during five cycles (BU=59GWd/t) show similar C/E trends. Calculation-Experiment biases are generally within target accuracy. However, in agreement with previous studies [2-3], $^{236}$U build-up is underestimated by 7%, due to a strong underestimation of the $^{235}$U radiative capture resonance integral in JEF2.2 evaluation; $^{239}$Pu and $^{241}$Pu are overestimated, partly due to a rough description of the actual irradiation history; $^{242}$Pu concentration is underestimated by 0.4% to 3.7%.

Figure 5 illustrates the increase of the $^{239}$Pu amount overestimation versus MOx fuel burnup, from previous SLB1 P.I.E. [3] and from this recent High-Burnup DAMPIERRE experiment.

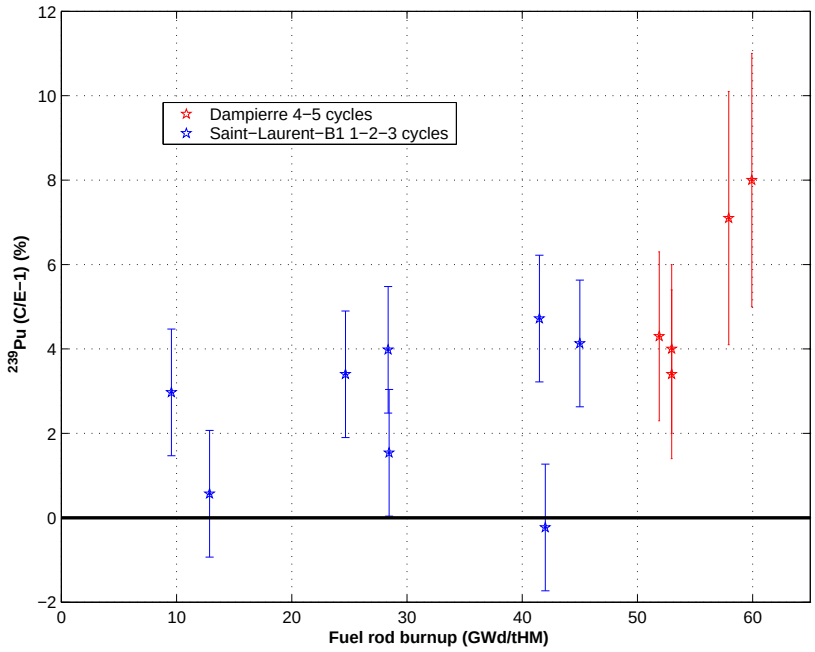


Figure 5: (C-E)/E bias in % on $^{239}$Pu depletion versus MOx burnup

At the time of the chemical analysis, 70% of the $^{241}$Am amount comes from the $^{241}$Pu decay in the 4-cycle samples, and 50% in the 5-cycle samples. We notice a calculation trend to $^{241}$Am overprediction: +5% in the 50GWd/tHM fuel samples after 3-years cooling ($^{241}$Pu decay), corresponding to +15% at EOC; this result is in agreement with Belgo-Nucléaire trends obtained from ARIANE P.I.E. using WIMS/JEF2.2 calculation [5]. The strong $^{241}$Am overestimation by about 15% and the underestimation of the $^{242M}$Am amount could be due to the underestimation of the $^{241}$Am$(n,\gamma)$ cross-section.

### 5.3 JEFF3.0 results

The fuel inventory C/E values are compared in Table 3 for APOLLO2 calculations based on the CEA93 "industrial library" processed from JEF2.2 and the recent library CEA2003.V1 processed from JEFF3.0. Clear improvements are obtained using JEFF3.0 evaluations [6]: the longstanding $^{236}$U, $^{237}$Np and $^{242}$Pu underpredictions are corrected within the $2\sigma$ uncertainty margins. Poisoning fission products, $^{143}$Nd and $^{133}$Cs (Burnup Credit nuclides) are satisfactory predicted, which confirms the improvement of their (n,$\gamma$) cross-section in the JEFF3.0 evaluations [4].

Table 3: C/E biases (in %) using JEF2.2 and JEFF3.0 libraries

| | JEF2.2 | | JEFF3.0 | |
|---|---|---|---|---|
| | 4 CYCLES | 5 CYCLES | 4 CYCLES | 5 CYCLES |
| U234/U238 | -2.6 | -2.9 | -3.1 | -3.4 |
| U235/U238 | 1.2 | 1.1 | 0.2 | -0.1 |
| U236/U238 | -7.4 | -7.1 | -3.1 | -2.7 |
| | | | | |
| Np237/U238 | -12.4 | -9.6 | 1.1 | 4.1 |
| | | | | |
| Pu238/U238 | -3.5 | -1.9 | -4.1 | -2.4 |
| Pu239/U238 | 3.9 | 7.6 | 3.1 | 6.9 |
| Pu240/U238 | 0.6 | 2.9 | 1.1 | 3.5 |
| Pu241/U238 | 0.7 | 2.9 | 0.8 | 3.1 |
| Pu242/U238 | -2.4 | -2.0 | 1.7 | 2.4 |
| | | | | |
| Am241/U238 | 5.0 | 8.6 | 4.8 | 8.3 |
| Am241/U238 EOC | 15.3 | 15.7 | 14.1 | 14.7 |
| Am242M/U238 | -20.0 | -20.0 | -21.0 | -20.8 |
| Am243/U238 | 1.7 | 3.5 | 4.3 | 6.4 |
| | | | | |
| Cm243/U238 | -10.0 | -10.4 | -14.6 | -14.7 |
| Cm244/U238 | -4.5 | -2.5 | -1.9 | 0.3 |
| Cm245/U238 | 0.1 | 4.6 | 7.0 | 13.0 |
| Cm246/U238 | -8.1 | -4.9 | -11.4 | -7.6 |
| Cm247/U238 | -14.0 | -8.9 | -2.3 | 4.7 |
| | | | | |
| Nd143/U238 | 1.0 | 1.5 | 0.2 | 0.6 |
| Nd144/U238 | -2.1 | -2.0 | -1.4 | -1.2 |
| Nd145/U238 | -0.2 | -0.4 | -0.3 | -0.5 |
| Nd146/U238 | 0.7 | 1.2 | 0.9 | 1.5 |
| Nd148/U238 | 2.0 | 2.1 | 2.1 | 2.2 |
| Nd150/U238 | 0.5 | 0.7 | 0.5 | 0.8 |
| | | | | |
| Cs133/U238 | -0.5 | nm* | 0.2 | nm |
| Cs134/U238 | 1.9 | nm | -0.3 | nm |
| Cs135/U238 | 7.0 | nm | 6.7 | nm |
| Cs137/U238 | -1.1 | nm | -0.9 | nm |

## 6. Discussion on MOx reactivity versus burnup

Using JEFF3.0 instead of JEF2.2, the calculated multiplication factor $k_\infty$ for the mixed MOx-UOx pattern (1 MOx assembly surrounded by 3 UOx assemblies) decreases by about 300pcm up to 600pcm from the Beginning up to the End of Irradiation (EOI) (see Figure 6).

Exact Perturbation Theory shows that the 300pcm decrease of a mixed core reactivity at BOI has the following components:

- in the UOx spent assembly (-300pcm), the reactivity decrease is mainly due to $^{235}\mathrm{U}(n,\gamma)$ cross-section increase (-200pcm) and to the Pu cross-sections re-evaluations (-100pcm), and
- in the MOx assembly (0pcm), the contribution by about -100pcm of the increase of $^{241}\mathrm{Pu}(n,\gamma)$ cross-section in JEFF3.0 [6] is cancelled with +50pcm due to the $^{239}\mathrm{Pu}(n,f)$ increase and with +50pcm due to the $^{240}\mathrm{Pu}(n,\gamma)$ decrease.

At the EOI, the JEFF3.0 reactivity modification in the MOx assembly contributes now to -300pcm, and is equivalent to the UOx spent assembly contribution.

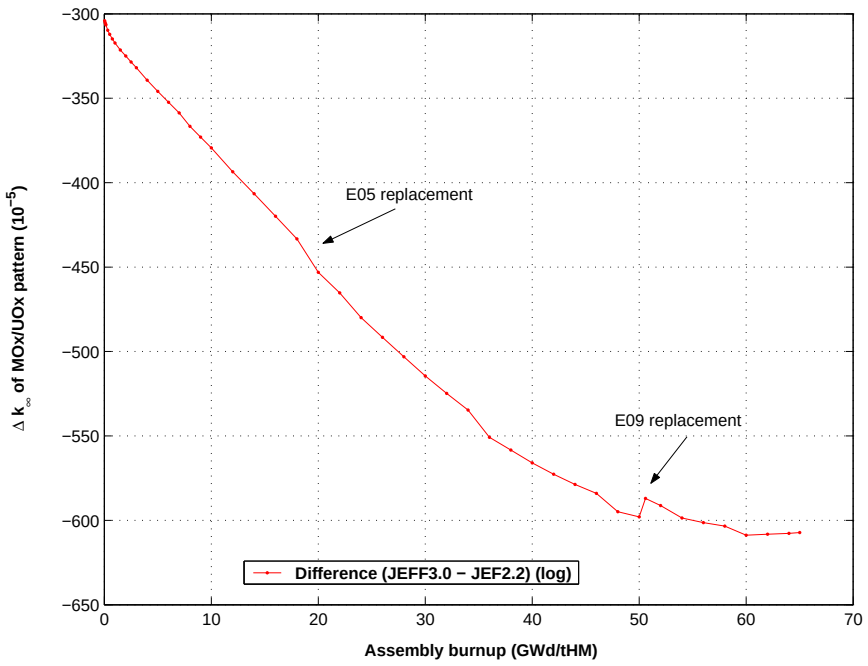


Figure 6: MOx reactivity improvement between JEF2.2 and JEFF3.0

The JEF2.2 reactivity overestimation in MOx lattices, ranging from +100pcm to +500pcm in French critical experiments [7-8] should be slightly improved using JEFF3.0. However, the reactivity loss with burnup in MOx fuels should be validated: the reactivity worth measurement of irradiated MOx fuel cuts, in the framework of an oscillation experiment in Minerve, is currently under analysis using both JEF2.2 and JEFF3.0.

## 7. Conclusion

This interpretation of the DAMPIERRE Post-Irradiation Experiment confirms the major JEF2.2 nuclear data trends given by the experiments performed in France and shows the good improvement given by JEFF3.0 in the prediction of isotopic fuel contents, mainly $^{236}$U, $^{237}$Np and $^{242}$Pu. These MOx results are consistent with the satisfactory C/E comparison obtained with JEFF3.0 on UOx spent fuel inventory [9]. Therefore, incorporation of JEFF3.0 evaluations within multigroup nuclear library CEA2003.V1 contributes towards the improvement of the APOLLO2 neutronic prediction capabilities.

The overestimation of the $^{239}$Pu content, already observed in SLB1-MOx P.I.E., is confirmed by this high-burnup MOx fuel interpretation (+4% at 50GWd/tHM and +7% at 60GWd/tHM). A more accurate interpretation, which details the irradiation history, is in progress; the $^{239}$Pu C/E comparison should be improved, mainly at EOC 5 due to a refined description of the 5th cycle discoutinuous power history. Furthermore, it is expected that the next JEFF3.1 evaluation will improve the $^{239}$Pu C/E agreement as the $^{238}$U effective integral resonance should be decreased by 1%.


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