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Impact of Fission Products Impurity on the Plutonium Content in PWR MOX Fuels

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CONTENTS

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1. INTRODUCTION

This report presents the results of a neutronics analysis in which the separation of the fission products (FP) during the reprocessing of UOX spent fuel assemblies (UOX SNF) is not perfect and that, consequently, a certain amount of FP goes into the Pu stream used to fabricate PWR MOX fuel assemblies. Only non-gaseous FP have been considered (see the list of 176 isotopes considered in the calculations in Appendix 1). This mixture of Pu and FP is called $\underline{Pu^{FP}}$. Note that, in this preliminary analysis, the FP losses are considered element-independent, i.e., for example, 1% of FP losses mean that 1% of all non-gaseous FP leak into the Pu stream.

The main objective of these calculations is to quantity the increase of the Pu content of a PWR MOX fuel necessary to maintain the same average burnup at discharge (51 GWd/tIHM) independently of the amount of FP in the Pu stream, i.e. independently of the Pu^{FP} composition. The calculations are performed with SCALE6.1 as well as, in order to assess its validity, with the internally developed fuel cycle tool FIT.

The amount of Pu and FP present in the reference 51.0 GWd/tIHM UOX SNF (4.3% U-235) after 10 years of cooling time is given in Table 1.1 below. It shows that 1 ton of UOX SNF contains 11.33 kg of Pu and 44.05 kg of non-gaseous FP, hence, for example, if the FP losses are equal to 1 w%, the Pu^{FP} mixture contains 11.33 kg of Pu and 0.4405 kg of non-gaseous FP, or put in differently, $Pu^{FP\text{-}1\%}$ = 96.26 w% Pu + 3.74 w% FP. The compositions of the Pu^{FP} mixture as a function of the FP losses during the reprocessing of UOX SNF are presented in Table 1.2.

Isotope	kg/tIHM
Pu238	0.295
Pu239	6.153
Pu240	2.930
Pu241	1.086
Pu242	0.864
Total Pu	11.33
Total FP	52.64
Total FP (no-gas)	44.05
Ratio FP/Pu	4.646
Ratio $FP_{no\text{-}gas}$ /Pu	3.888

Table 1.1. Pu and FP present in the reference 51 GWd/tIHM UOX SNF (10-year cooling time).

FP losses (w%)	FP in the Pu^{PF} mixture (w%)	Pu in the Pu^{PF} mixture (w%)
0.00 %	0.00%	100.00%
0.01 %	0.04%	99.96%
0.05 %	0.19%	99.81%
0.10 %	0.39%	99.61%
0.25 %	0.96%	99.04%
0.50 %	1.91%	98.09%
0.75 %	2.83%	97.17%
1.00 %	3.74%	96.26%
2.00 %	7.22%	92.78%
3.00 %	10.45%	89.55%
4.00 %	13.46%	86.54%
5.00 %	16.28%	83.72%

Table 1.2. Composition of the Pu^{FP} mixture as a function of the FP losses during the reprocessing of UOX SNF.

2. CALCULATION METHODOLOGY

The calculations needed by this study have been performed through two different codes:

- SCALE 6.1 code system;
- FIT (**F**uel-cycle **I**ntegration and **T**radeoffs) toolkit [7].

A small overview on the main capabilities and models used is reported in the following paragraphs.

2.1 SCALE 6.1 CODE SYSTEMS.

The reactor physics calculations have been performed by the lattice physics capabilities of the SCALE 6.1 code systems. The calculation flow consists of the use of several modules mutually coupled. The discrete-ordinates code NEWT (**N**ew **E**SC-based **W**eighting **T**ransport code) coupled to the depletion code ORIGEN [8] via the TRITON control module [4]. Using the discrete-ordinates approximation to the transport equation on an arbitrary grid, together with a 238-group neutron cross-section library based on ENDF/B-VII, NEWT provides a robust and rigorous deterministic solution for non-orthogonal configurations. The differencing scheme employed by NEWT, the Extended Step Characteristic Approximation, allows a computational two-dimensional mesh based on arbitrary polygons. Such a mesh can be used to closely approximate curved or irregular surfaces to provide the capability to model problems that were formerly difficult or impractical to model directly with discrete-ordinates methods.

The TRITON control module performs the task of coordination of data transfer between various physics codes available within SCALE 6.1 and of invoking those codes in the proper sequence for a desired type of calculation. The high-fidelity nature of the NEWT solution in estimating angular flux distributions combined with the rigor of the ORIGEN depletion solver gives TRITON the capability to perform precise burnup-dependent physics calculations with few

implicit approximations, and limited primarily by the accuracy of nuclide cross-sectional data. Such rigor may be necessary to capture the unique attributes of MOX fuel behavior as well as that of advanced, highly heterogeneous fuel assembly designs being deployed in current-generation reactors. Cross-sectional self-shielding is carried out by BONAMI for unresolved-range resonance data; the resolved resonance processor module CENTRM performs a 1-D discrete-ordinates code that uses point-wise cross-section data to produce a set of continuous-energy fluxes at discrete spatial intervals for each unit cell. Following a CENTRM calculation, the code PMC uses the resulting flux to collapse the point-wise continuous-energy cross sections into multi-group cross sections for each nuclide in each material in a unit (e.g., pin cell). The result is a multi-group library in which point cross-sectional data are weighted using the explicit point-wise spectrum representative of the nuclides present in a pin cell. Effects from overlapping resonances, fissile material in the fuel and surrounding moderator, anisotropic scattering, and inelastic level scattering are explicitly handled by this approach.

For the physics calculations carried out during this study, a TRITON model of one fourth standard (17x17) fuel assembly has been used (Fig 2.1). All the MOX rods have the same U-235 enrichment and the same Plutonium content. The 0.5 mm water gap at the periphery is explicitly represented. The model uses three different burn-up zones to take into account the different local moderating ratios: 1 for the corner rods (blue), 1 for the other rods located at the periphery (green) and 1 for the internal rods (red).

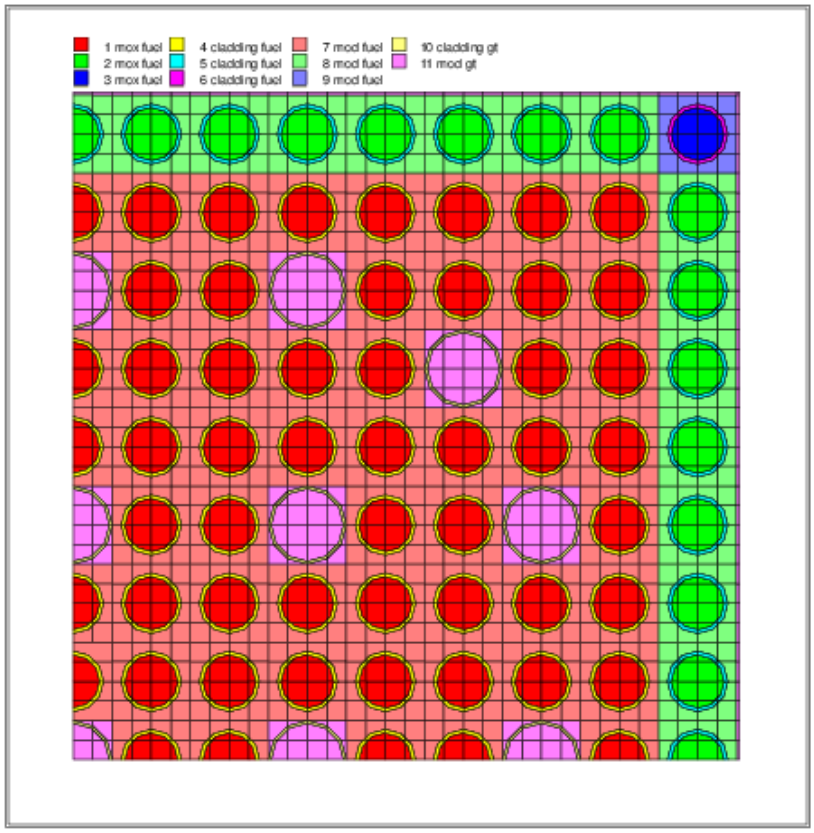

Fig 2.1. TRITON model (one fourth of a standard 17x17 fuel assembly)

The plutonium content in the MOX fuel assemblies must be increased to compensate for the absorption of the fission products and thus maintain the same average burnup at discharge of 51 GWd/tIHM independently of the amount of fission products in the plutonium stream. The calculations have been run considering a 3-batch core with an average discharge burn-up of **51.0 GWd/tIHM.** The model takes in account 1 year fuel aging (time needed by the fabrication and transport operations).

The necessary Pu^{FP} (Pu + FP) enrichments are determined with a methodology that is standard for this kind of application: the k-infinity of the assembly at the average end of cycle burn-up (34 GWd/tHM), without soluble boron, is equal to that of the reference 4.3% UOX assembly calculated using the same code (SCALE 6.1), methods (SN), nuclear data (238 group library based on ENDF/B - VII), etc., i.e. **k-inf = 1.0365** in our case. This is the strategy ensures the different fuels will release the same amount of energy and thus allows relevant comparisons among them.

2.2 FUEL-CYCLE INTEGRATION and TRADEOFFS (FIT).

The **F**uel-cycle **I**ntegration and **T**radeoffs toolkit [7] is a tool internally developed at the Idaho National Laboratory in order to permit a systematic examination of the chemical behavior of a FC and analyze different cases.

FIT is a method to examine different fuel cycles using common bases; in particular, to determine how changes in one part of a fuel cycle (say, fuel burn-up, cooling, or separation efficiencies) affect other parts of the it. FIT provides the following:

- Rough estimate of physics and mass balance feasibility of combinations of technologies. If feasibility is an issue, it provides an estimate of how performance would have to change to achieve feasibility;
- Estimate of impurities in fuel and impurities in waste as function of separation performance, fuel fabrication, reactor, uranium source, etc.

The following figure illustrates the basic mass flows. For the first recycle iteration (recycle-1), there are only the streams from incoming used fuel via separation-1. There is no mass flow yet from separation-2. For subsequent recycle streams, the TRU-U-1 product mass flow from separation-1 remains as before and the TRU-U-2 product mass from separation-2 from the previous iteration is added to create a combined TRU-U product stream. FIT uses a single set of separation factors for the entire suite of technologies that may be combined together. The user selects whether to use RU-1, RU-2, or DU to blend with this combined TRU-U product stream. The model estimates the required ratio of TRU-U product to U feed for that recycles's iteration. Since the TRU-U product can contain uranium, and the RU streams will typically contain some TRU impurity, the ratio of TRU-U product to U product is not the same as the chemical TRU:U ratio in the final blended product.

The reactor simulation is performed by an internally developed burn-up code **MRTAU [**1], [6].

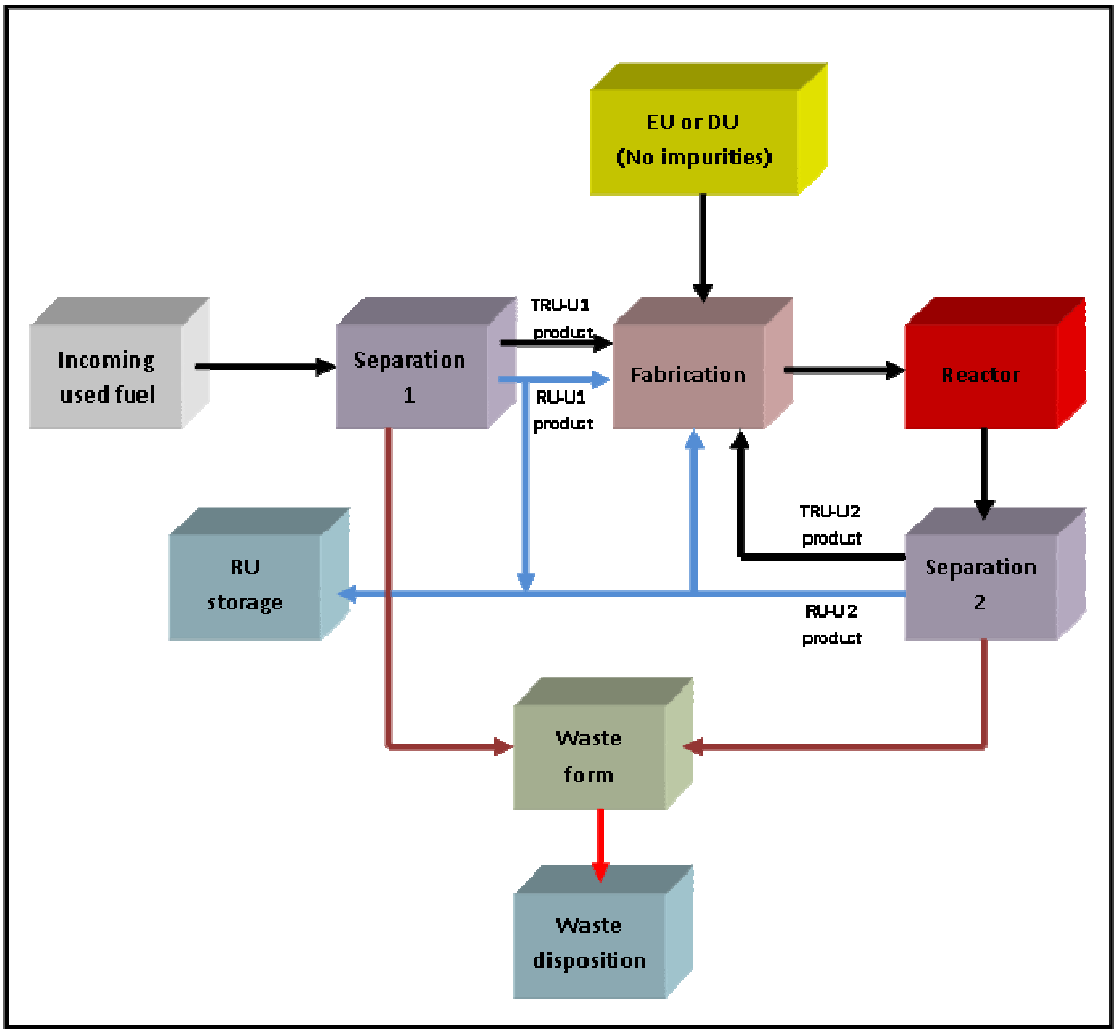

Fig 2.2. FIT basic mass flow.

The code needs two fuel compositions – that of the incoming stream and that for the reactor in the recycle loop. The composition of the incoming stream is used, as is. FIT adjusts the composition of the reference fuel in the recycle loop according to what happens in the simulation. The fuel adjustment in FIT is performed through a standard methodology. The code compares the BOC k-infinitive of the initial recipe (fuel without impurities) with that of the fuel coming from the separation stages (impurity content depending on the separation method [3]), adjusting the TRU:U ratio in order to get the same initial k-infinity. In order to perform this adjustment, FIT uses 1-group not-tabulated cross sections (the cross sections taken from reactor physic calculations performed outside the code).

As mentioned before, the separation stages are simulated through separation factors for each isotope or element families. In order to reproduce the fuel cycle cases (Table 2.1 and Fig. 2.2), analyzed through the reactor physic calculations, the separation matrices have been appropriately modified obtaining the same FP composition for each case.

3. RESULTS

3.1 SCALE6.1

The table and figure below show the calculated Pu^{FP} and Pu content in MOX fuel assemblies necessary to maintain the same average burnup at discharge of 51 GWd/tIHM independently of the amount of the FP losses. It shows, in particular, that the mass of Pu in the MOX assembly increases by approximately 3.5% per % of FP losses.

NB: The calculations of the necessary Pu^{FP} contents showed the presence of what looks like an artifact which seems to be caused by the large number of FP (176) in the initial fuel composition. This issue is currently being addressed by the ScaleHelp team. The values presented in the table below are obtained by subtracting the observed 0.17% bias on the Pu^{FP} content calculated with SCALE. We gratefully acknowledge our colleague Brent Dixon for pointing out the slight irregularities of some results which led us to find this artifact. More information can be found in Appendix 2.

FP losses (w%)	***Pu*^{FP} enrichment (w%)**	**Pu enrichment (w%)**
-	10.17%	10.17%
0.25	10.37%	10.27%
0.50	10.55%	10.35%
0.75	10.76%	10.45%
1.00	10.94%	10.53%
2.00	11.73%	10.88%
3.00	12.59%	11.28%
4.00	13.53%	11.71%
5.00	14.49%	12.13%

Table 3.1. Pu^{FP} and Pu content in MOX fuel assemblies for different FP losses.

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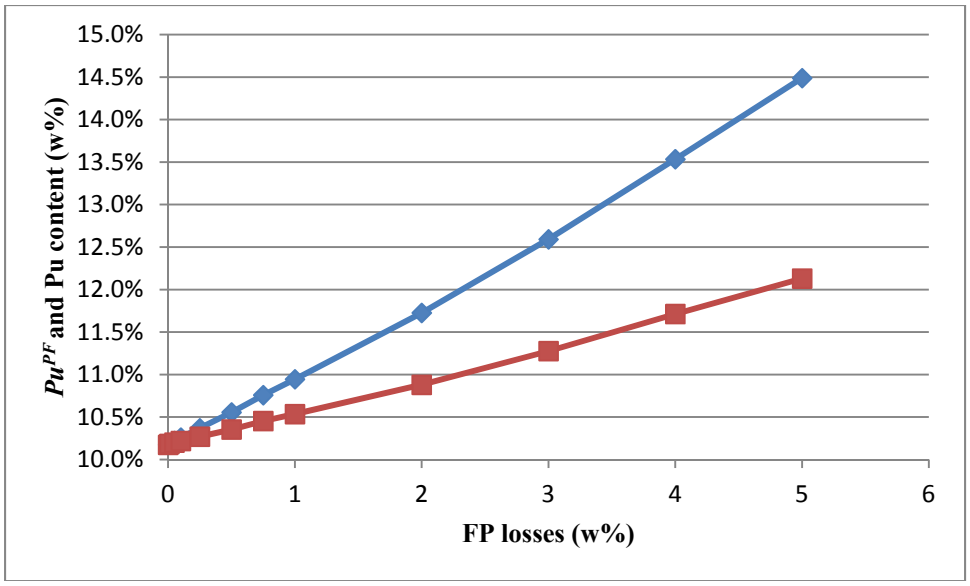

Figure 3.1. Pu^{FP} (blue) and Pu (red) content in PWR MOX fuel assemblies calculated with SCALE6.1 for different FP losses.

3.2 FIT

As mentioned earlier, FIT uses 1-group cross-sections generated for a reference fuel (in our case the 10.17% MOX fuel without FP) and estimates the effects of variations of this reference fuel composition using the same 1-group cross-section set. The figure below compares the Pu^{FP} contents calculated with FIT with those calculated with SCALE6.1 for FP losses up to 5%.

The discrepancies have been shown to come not from the constant 1-group cross-section set hypothesis used by FIT but instead from the difference in choosing the reference k-infinity. With SCALE, the necessary Pu^{FP} enrichments are determined so that the k-infinities of the assemblies at the average end-of-cycle burn-up are all the same, independently of the FP losses. This is the standard approach used everywhere for these kinds of analyses and it has proved its worth. FIT uses a similar approach but considering the k-infinity of the reference fresh fuel assembly as the target k-infinity that the other fuels must satisfy. This approach is not as valid as the other one because depending on their internal conversion ratio and on their FP reactivity worth, different fuels will necessitate different initial k-infinity to reach the same burnup. For example, a HEU fuel has a higher initial k-infinity than a LEU fuel and a LEU fuel has a higher initial k-infinity than a MOX fuel.

This was demonstrated by calculating the Pu^{FP} contents with SCALE but using the FIT "same fresh fuel k-infinity" approach (the green curve on the figure below) instead of the "same end-of-cycle fuel k-infinity" approach and observing that, in this case, both SCALE and FIT gave the same (wrong) Pu^{FP} contents. Modifying FIT to take this into account should be doable and it should make it more reliable.

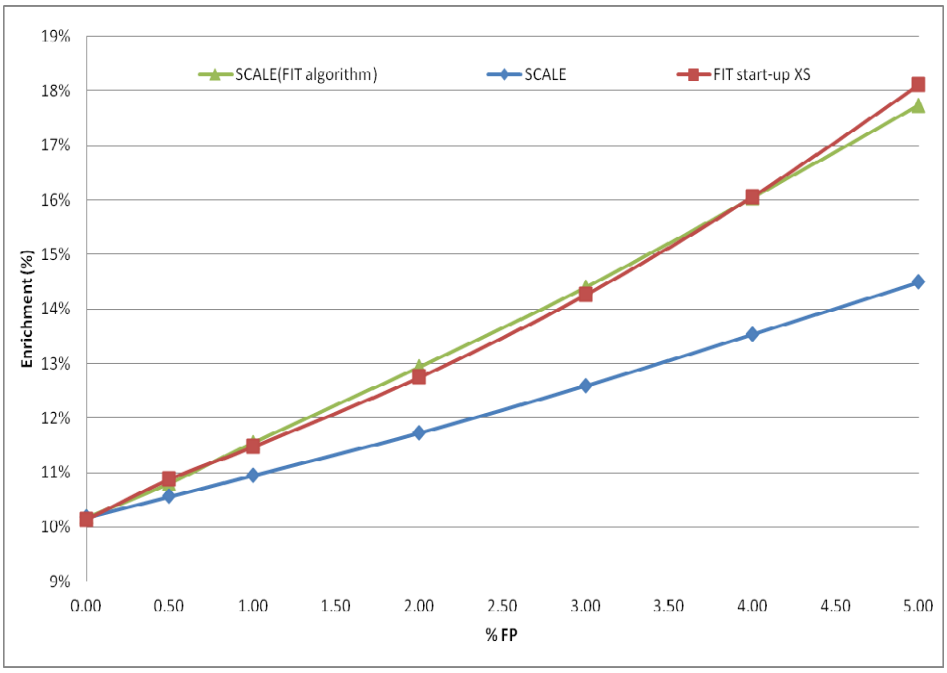

Figure 3.2. Comparison of the Pu^{FP} content calculated with FIT and SCALE6.1 for different FP losses.

4. CONCLUSIONS.

This report presents the results of a neutronics analysis in which the separation of the fission products (FP) during the reprocessing of UOX spent fuel assemblies (UOX SNF) is not perfect and that, consequently, a certain amount of FP goes into the Pu stream used to fabricate PWR MOX fuel assemblies. In this preliminary analysis, the FP losses are considered element-independent, i.e. for example, 1% of FP losses means that 1% of all non-gaseous FP leak into the Pu stream.

The main objective of these calculations is to quantity the increase of the Pu content of a PWR MOX fuel necessary to maintain the same average burnup at discharge (51 GWd/tIHM) independently of the amount of FP in the Pu stream. The calculations are performed with SCALE6.1 as well as, in order to assess its validity, with the internally developed fuel cycle tool FIT.

The two main conclusions are:

1) The mass of Pu in the MOX assembly increases by approximately 3.5% per % of FP losses.

2) The "same fresh fuel k-infinity" hypothesis used by FIT causes relatively large errors in the calculation of the Pu contents and should be modified.

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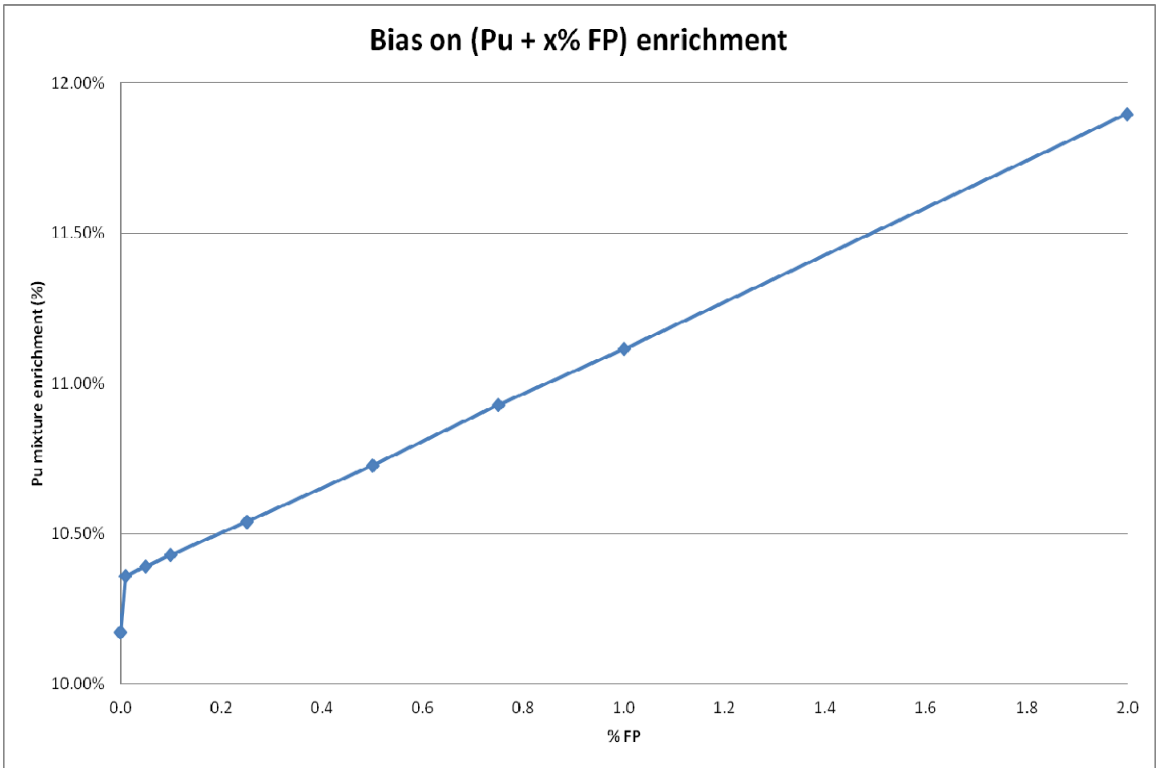
8. L.Gauld, O. Herman, et al., "*Origen-S: Scale system module to calculate fuel depletion, Actinide transmutation, fission product buildup and decay, and associated radiation source terms*", Oak Ridge National Laboratory (2009).

Appendix 1: List of non-gaseous fission products considered in the calculations.

Number of isotopes = 176							
Li6	Rb87	Mo96	Ag109	Sn123	Cs134	Nd143	Eu156
Li7	Sr86	Mo97	Ag110m	Sn124	Cs135	Nd144	Gd152
Be9	Sr87	Mo98	Ag111	Sn125	Cs136	Nd145	Gd153
N14	Sr88	Mo100	Cd106	Sn126	Cs137	Nd146	Gd154
Ga69	Sr89	Tc99	Cd108	Sb121	Ba132	Nd147	Gd155
Ga71	Sr90	Ru98	Cd110	Sb123	Ba134	Nd148	Gd156
Ge70	Y89	Ru99	Cd111	Sb124	Ba135	Nd150	Gd157
Ge72	Y90	Ru100	Cd112	Sb125	Ba136	Pm147	Gd158
Ge73	Y91	Ru101	Cd113	Sb126	Ba137	Pm148	Gd160
Ge74	Zr90	Ru102	Cd114	Te122	Ba138	Pm148m	Tb159
Ge76	Zr91	Ru103	Cd115m	Te123	Ba140	Sm147	Tb160
As75	Zr92	Ru104	Cd116	Te124	La138	Sm148	Dy160
Se76	Zr93	Ru106	In113	Te125	La139	Sm149	Dy161
Se77	Zr94	Rh103	In115	Te126	La140	Sm150	Dy162
Se78	Zr95	Pd102	Sn114	Te127m	Ce140	Sm151	Dy163
Se79	Zr96	Pd104	Sn115	Te128	Ce141	Sm152	Dy164
Se80	Nb93	Pd105	Sn116	Te129m	Ce142	Sm154	Ho165
Se82	Nb94	Pd106	Sn117	Te130	Ce143	Eu151	Ho166m
Br79	Nb95	Pd107	Sn118	I127	Ce144	Eu152	Er166
Br81	Mo92	Pd108	Sn119	I129	Pr141	Eu153	Er167
Rb85	Mo94	Pd110	Sn120	I131	Pr143	Eu154	Er168
Rb86	Mo95	Ag107	Sn122	Cs133	Nd142	Eu155	Er170

Appendix 2: SCALE 6.1 issue.

In our simulations we noticed the presence of a bias on the requested Pu^{PF} (Pu + x% FP) enrichment moving from the MOX reference case (0.0% FP) to the impurity cases. Indeed, the slightest addition of FP (we used 176 non-gaseous FP) in the initial MOX fuel composition causes a jump of about 0.17% on the necessary enrichment followed by a linear increase as shown on the figure below.



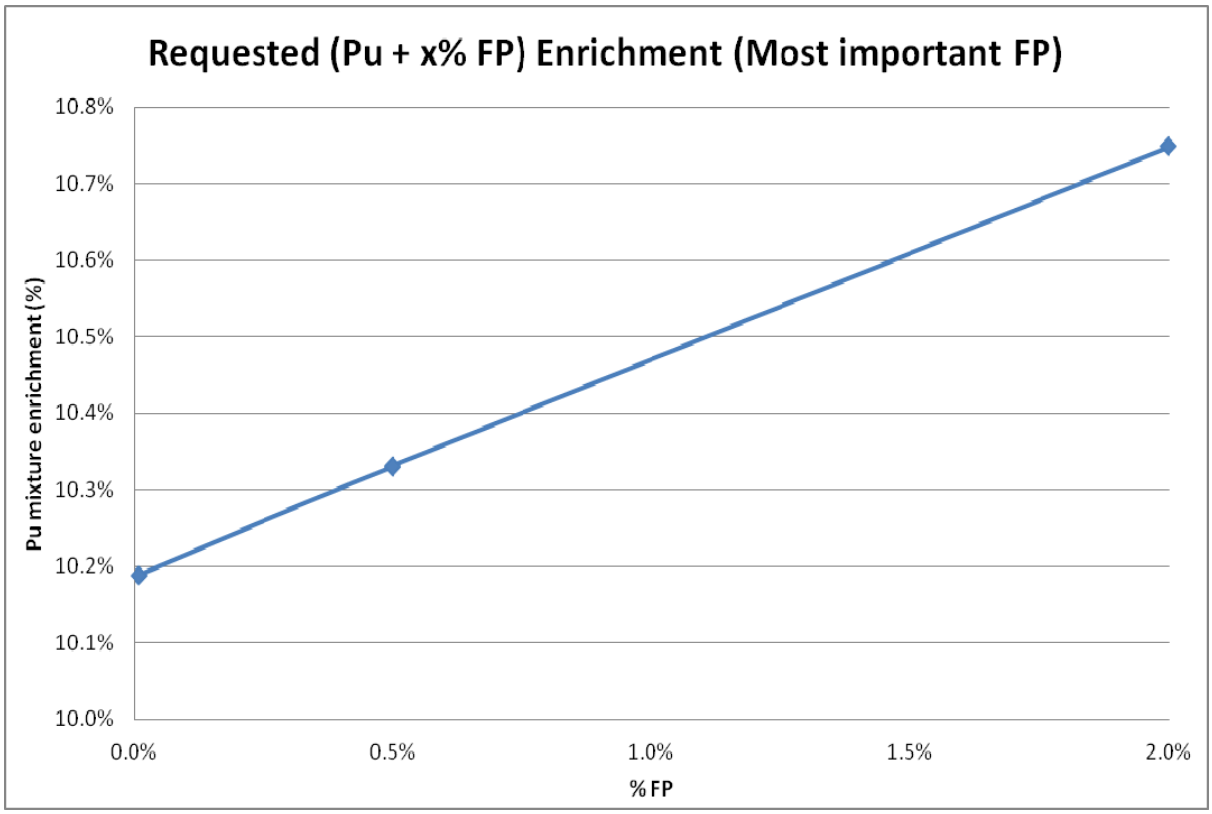
Necessary Pu^{FP} content calculated using 176 FP

In order to prove that the artifact was really present, we needed to perform a few more simulations. We decided to reduce the number of isotopes input in SCALE 6.1, keeping the 12 most important, in terms of neutron poisoning, fission products (see table below). These isotopes represent a contribution on the poisoning of about 60% even if the FP total mass is only ~15% of the full isotope set cases.

Twelve most important neutron poisons					
Tc99	Cs133	Pm147	Sm150	Sm152	Eu152
Rh103	Nd143	Sm149	Sm151	Sm154	Eu153

Most important FP

The requested Pu^{FP} contents computed for these cases (see figure below) tend to show that the jump observed is indeed an artifact caused by the large number of isotopes input in the code. This gave us the possibility to simply subtract the bias of **0.17%** from the enrichment curve of the full FP set cases.



Necessary Pu^{FP} content calculated using only the 12 most important FP