A burnup credit approach for irradiated fast-neutron reactor fuels nuclides of interest and fuel storage application

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A BURNUP CREDIT APPROACH FOR IRRADIATED FAST-NEUTRON REACTOR FUELS: NUCLIDES OF INTEREST AND FUEL STORAGE APPLICATION

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ABSTRACT

The concept of taking into account the reduction of the reactivity of nuclear used fuel due to their burnup is referred to as "Burnup Credit" (BUC). Economic incentives offered by the implementation of a BUC methodology in criticality analyses of Light Water Reactor (LWR) used fuel facilities are nowadays well-demonstrated.
The current prospect of reprocessing the fissile and fertile subassemblies of the PHENIX reactor in France, and the development of the 4th-Generation Fast-neutron Reactors (FR) enable research projects in relevant fuel cycle operations. Under these circumstances, the CEA and AREVA-NC have decided to study a burnup credit approach for irradiated fuels of the PHENIX reactor.

This paper focuses on a preliminary investigation on the use of burnup credit for used FR fuel operations. The first step consists in analyzing and selecting the nuclides of interest for burnup calculation, on the basis of the 27 BUC nuclides (12 actinides and 15 fissions products) chosen for LWR-MOx fuels [1]. These nuclides are then involved in a criticality calculation (a used fuel pool) to highlight the interest of a BUC approach.

This study highlights the interest of a burnup credit approach for FR fuels and shows that the highest contribution to reactivity loss is due to only one fission product: the ^{149}Sm. The opportunity to take account of only one fission product is highly promising and will considerably simplify the way to determine a penalizing fuel inventory. Moreover, it is to notice that results for different configurations may differ from those obtained for a fuel storage application. Thus, specific analyses have to be carried out in order to study the applicability of the conclusions of this paper.

KEYWORDS
Burnup Credit, Fast-Neutron Reactors, Used fuel, Criticality

1. INTRODUCTION

The concept of taking into account the reduction of the reactivity of nuclear used fuel due to their burnup is referred to as "Burnup Credit" (BUC). Economic incentives offered by the implementation of a BUC methodology in criticality analyses of Light Water Reactor (LWR) used fuel facilities are nowadays well-demonstrated.
The current prospect of reprocessing the fissile and fertile subassemblies of the PHENIX reactor in France, and the development of the 4th-Generation Fast-neutron Reactors (FR) enable research projects in relevant fuel cycle operations. Under these circumstances, the CEA and AREVA-NC have decided to study a burnup credit approach for irradiated fuels of the PHENIX reactor.

This paper focuses on a preliminary investigation on the use of burnup credit for used FR fuel operations. The first step consists in analyzing and selecting the nuclides of interest for burnup calculation, on the basis of the 27 BUC nuclides (12 actinides and 15 fissions products) chosen for LWR-MOx fuels [1]. These nuclides are then involved in a criticality calculation (a used fuel storage pool) to highlight the interest of a BUC approach.
This study has been carried out using the latest version of the nuclear data library JEFF-3.1.1 [2], the latest version of the fuel cycle package DARWIN2.3 [3], recently validated for FR fuels [6], and the new version of the criticality-safety package CRISTALV2.0 [8].

It is to notice that the study does not use bounding values nor penalizing assumptions: depletion calculations use nominal conditions of irradiation, the fresh fuel inventory values corresponding to mean values as the average burnup used.

2. CALCULATION METHODS, ASSUMPTIONS AND MODELS

2.1. Calculation tools

The PEPIN depletion solver of DARWIN calculates the isotopic concentrations at the end of the irradiation or after a cooling time. Then, these concentrations are used as input data in the Criticality-Safety package CRISTAL providing the effective multiplication factor (k_{eff}) associated to the calculated configuration.

2.1.1. Depletion calculations with DARWIN2.3

DARWIN2.3 [3] is the French reference calculation package for fuel cycle applications, such as fuel inventories and decay heat. DARWIN2.3 performs the nuclide depletion calculation (PEPIN solver), fed by libraries on the one hand, and neutronic data on the other hand (Fig. 1). All the decay data and fission yield values come from the JEFF-3.1.1 evaluation [2], whereas the self-shielded cross-sections and neutron spectra are provided by deterministic neutron transport codes: ERANOS-2 for Fast-Neutron Reactor studies [4] resolving the Boltzmann equation on the whole reactor core; APOLLO2 for Light Water Reactors studies [5]. Complementary cross-sections, missing from the transport code libraries are taken from JEFF-3.1.1 evaluation. It is to notice that DARWIN2.3 has been experimentally validated for LWR fuels [7] and recently for FR fuels [6].

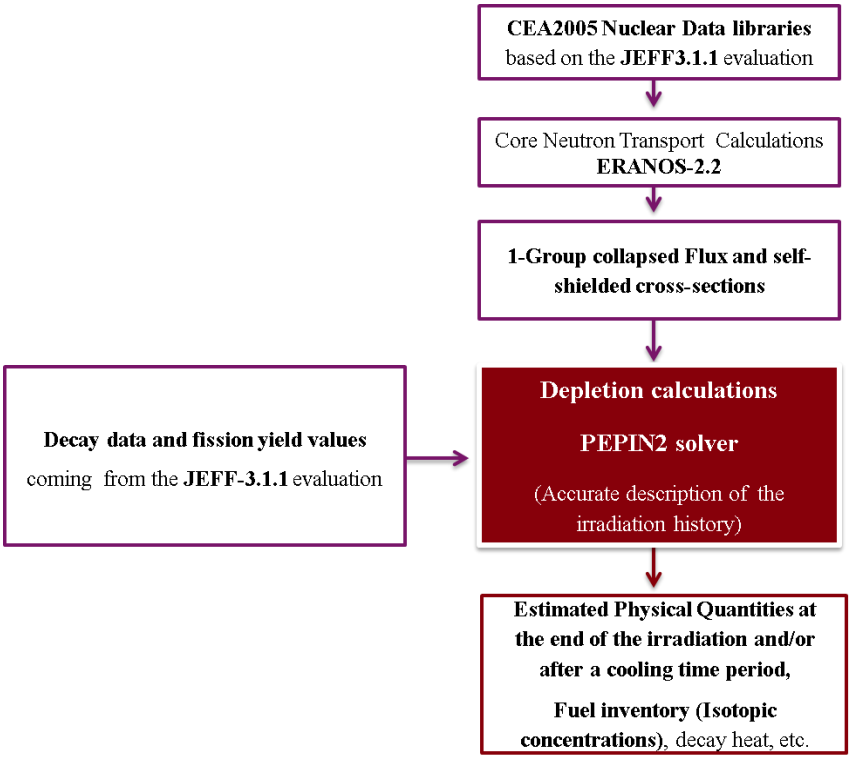

Figure 1 DARWIN2.3 calculation route for FR fuels depletion calculations

2.1.2. Criticality calculations with CRISTALV2.0

The CRISTAL package uses nuclear data libraries, well established transport codes and a graphical user interface. It has been developed by IRSN and CEA with the support of AREVA & EDF.
The new version 2 of the criticality-safety package CRISTAL [8] is based on the latest Joint European nuclear datafile JEFF-3.1.1 and four calculation routes: two multi-group routes based on multi-group (281 groups) cross-sections (APOLLO2 – MORET 5 or APOLLO2 Sn calculations), a pointwise Monte Carlo route (TRIPOLI-4) and a criticality standard calculation route, based on the latest version of neutronic codes APOLLO-2.8 [5] MORET 5 [9] and TRIPOLI-4.8 [10].

In this study, the criticality calculations providing the effective multiplication factor (k_{eff}) associated to the calculated configuration are performed with the pointwise Monte-Carlo TRIPOLI4.8 route, in order to minimize computing approximations.

2.2. Models and assumptions

This study is performed for a fissile sub-assembly of the French Sodium Fast-Reactor PHÉNIX (Fig. 2). Only two of the three vertical parts of the assembly have been described: the central fissile part (C) and the Lower Axial Blanket (fertile). Indeed, before the transport of the assemblies, the Upper Axial Blanket is separated of the other part of the assembly to be transported, stored and reprocessed separately. This study focuses on the fissile part of the assembly.

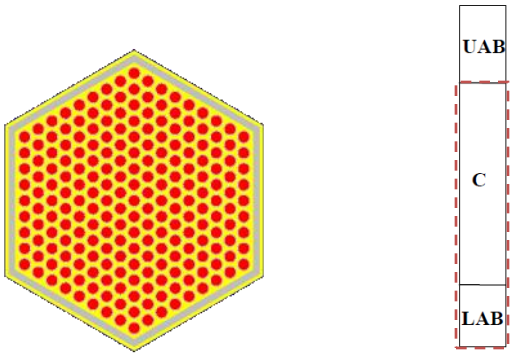

Figure 2 Design of a fissile sub-assembly of the PHÉNIX reactor

2.2.1. Fuel inventory

The central fissile part is made up of Mixed Oxide (MOX) fuel, with a weight ratio Pu/(U+Pu) equal to 22.5% and the U- Pu vectors presented on the Table I.

Table I. Composition of the fissile fresh fuel of PHÉNIX

Nuclides	%
^{238}Pu	0,18
^{239}Pu	72,38
^{240}Pu	22,21
^{241}Pu	3,82
^{242}Pu	0,87
^{241}Am	0,54

Nuclides	%
^{238}U	99,68
^{235}U	0,32

The Lower Axial Blankets are made up of depleted Uranium (Table II). These blankets give possible the uranium to plutonium conversion.

Table II. Composition of the fresh LAB of PHÉNIX

Nuclides	%
^{238}U	99,68
^{235}U	0,32

In order to perform a standard configuration, the used fuel inventory has been determined for a total average burnup of 92 GWd/t and a 5-year cooling time.

2.2.2. Criticality calculation model

The criticality simulations are performed for an infinite array of the fissile assemblies, separated with 5 cm of non-borated water and reflected with 1.5 meters of non-borated water at the top and the bottom.

3. SELECTION OF THE NUCLIDES OF INTEREST AND BURNUP CREDIT ESTIMATION

In order to take into account the burnup of the FR fuel, through the depletion of fissile content and the build-up of absorbent isotopes, the nuclides that contribute significantly to the negative burnup reactivity worth have to be selected.
Many studies have been carried out to determine which isotopes contribute significantly to the negative reactivity worth for LWR-UOX and MOX fuels. For these fuels major actinides, some minor actinides and 15 fission products (FP) have been selected for the BUC applications. These 15 FPs correspond to the most absorbent nuclides, stable and non-volatile.
This selection of nuclides was carried out at CEA in 1991 and has been approved by OECD/Expert group on BUC Criticality Safety [1].
The SFR PHÉNIX involves MOX fuels, so in a first time it seems relevant to involve the same nuclides as LWR-MOX fuels although the main absorbent isotopes will be necessarily different because of the fast spectra irradiation.

3.1. Reactivity worth of the absorbent nuclides

Table III presents the negative reactivity worth of the absorbent isotopes for the fissile part of a FR assembly in the configuration describe on the previous section 2.

Table III. Negative reactivity worth of the absorbent isotopes for a FR fuel of PHÉNIX (92 GWd/t BU, 5 years CT)

	Isotopes	Reactivity worth - Δk_{eff} (pcm) [σ = ±14 pcm]
16 ACTINIDES	^{234}U	-40
	^{235}U	
	^{236}U	-60
	^{238}U	
	^{238}Pu	-40
	^{239}Pu	
	^{240}Pu	-3030
	^{241}Pu	
	^{242}Pu	-60
	^{241}Am	-150
	^{242m}Am	
	^{243}Am	-50
	^{243}Cm	
	^{244}Cm	-20
	^{245}Cm	
	^{237}Np	-40

	Isotopes	Reactivity worth - Δk_{eff} (pcm) [σ = ±10 pcm]
15 FISSION PRODUCTS	^{95}Mo	≤ 10
	^{99}Tc	≤ 10
	^{101}Ru	≤ 10
	^{103}Rh	-70
	^{109}Ag	-40
	^{133}Cs	-170
	^{143}Nd	-100
	^{145}Nd	-20
	^{147}Sm	-50
	^{149}Sm	**-10950**
	^{150}Sm	≤ 10
	^{151}Sm	-320
	^{152}Sm	-20
	^{153}Eu	≤ 10
	^{155}Gd	-190

The two most absorbent actinides are ^{240}Pu and ^{241}Am. The reactivity worth of the ^{240}Pu depends first on its initial rate in the fresh fuel inventory.

Moreover, the results shown that the reactivity worth of the other actinides: ^{234}U, ^{236}U, $^{242m, 243}$Am, curium and neptunium isotopes are insignificant.

Regarding the fission products, the most striking point is the reactivity worth of the ^{149}Sm. The reactivity loss due to the Samarium 149 represents 90% of the reactivity worth of the 15 FPs. Finally, these results highlight that the other fission products are insignificant.

These results could be explained because during all the irradiation in a fast-neutron reactor, the fission products have been built up in the core (Fig.3).

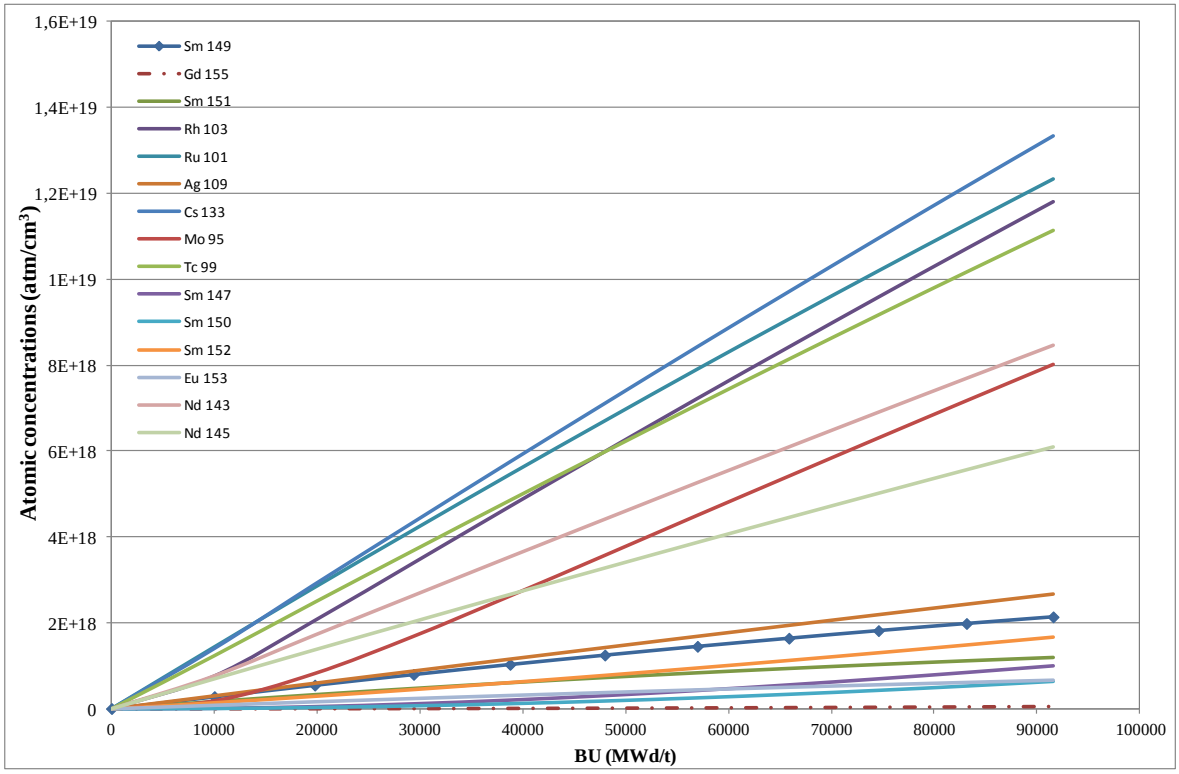

Figure 3 Fission product concentrations of a FR fuel as a function of the burnup

For example at the end of the irradiation, there is one hundred times more ^{149}Sm in a FR-MOX fuel than in a LWR-MOX fuel (BU=45GWd/t).

As show on Fig.4, during an irradiation under a thermal spectrum (LWR-MOX), there is a competition between build-up and capture phenomena: the latter becomes predominant above a concentration and eventually leads to the decrease of ^{149}Sm concentration at the end of the irradiation. This is not the case for a FR, as thermal capture by ^{149}Sm is negligible during all the irradiation: all the ^{149}Sm is then available in thermal spectra configurations (transport, storage, etc.)[1]. This, combined with one of the higher FPs cross-section [σ_c (^{149}Sm -2200 m.s^{-1}) = 41731 barns], explain the important gain in terms of reactivity worth in our configurations.

[1] Samarium 149 is a thermal absorbent: more than 98 % of its reactivity worth is due to its capture cross-section under 0.6eV

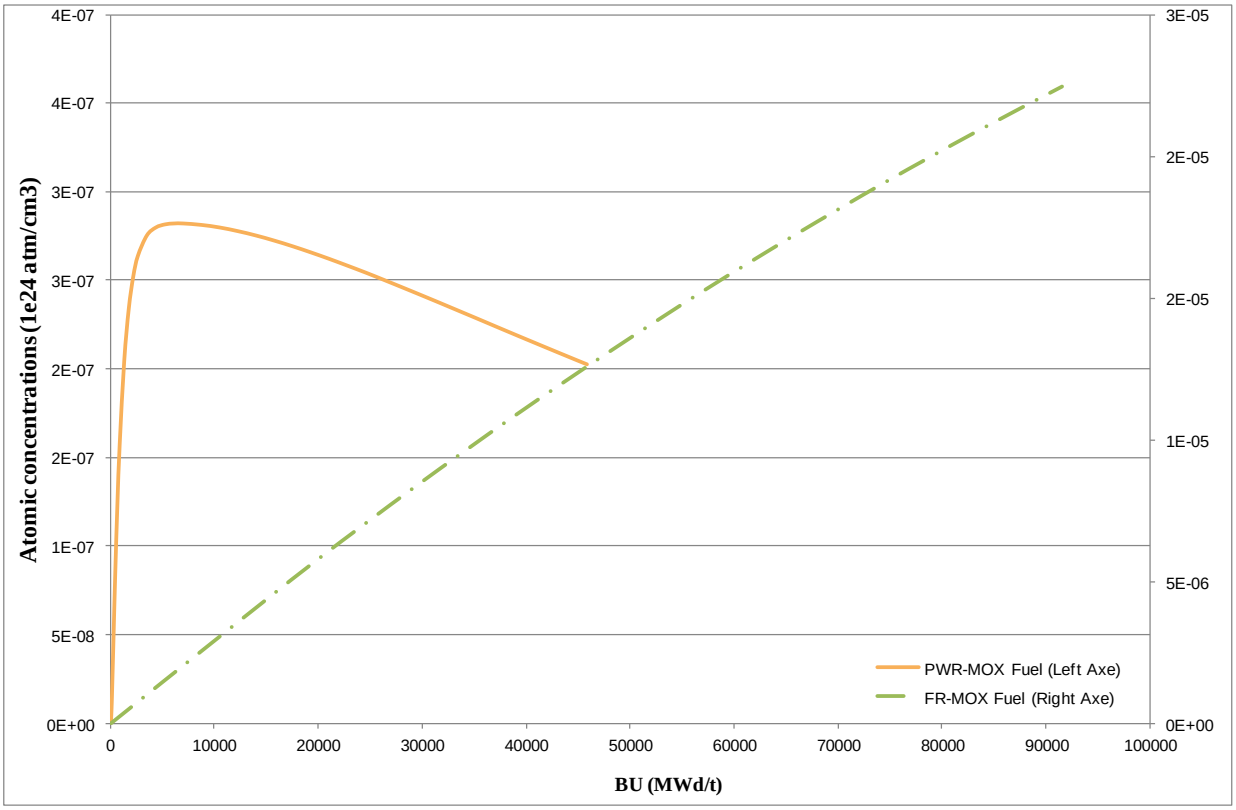

Figure 4 ^{149}Sm concentrations for a FR-MOX fuel and a LWR-MOX fuel as a function of the burnup

3.2. Burnup credit estimations

Traditionally for LWR used fuels, two approaches are described in criticality studies taking account for burnup credit:

- The "Actinide-Only" approach, taking only into account the major and some minor actinides (the 16 actinides presented in the previous section 3.1);
- The "Actinide and fission products" approach, taking account of all the actinides and fission product described in the previous section (27 isotopes).

Table IV. Comparative burnup credit gain values for PWR-UOx, PWR-MOx and FR-MOX

	PWR-UOX	**PWR-MOX**	**FR-MOX**
BUC ACTINIDES	**17300 pcm**	6200 pcm	**5334 pcm**
BUC 15 FPs	7700 pcm	**7800 pcm**	**11870 pcm**
BUC ACTINIDES+FPs	25000 pcm	14000 pcm	**17204 pcm**
	BU = 45 Gwd/t - CT= 5 y		*BU = 92 GWd/t -CT = 5 y*

Table V. Burnup credit gain as function as the nuclides involved in the fuel inventory

	Burnup credit gain (Δk_{eff}, pcm)
16 Actinides only	5300
Samarium 149 only	11000
8 Actinides and Samarium 149	16300
16 Actinides and Samarium 149	16300
16 Actinides and 15 PF	17200
16 Actinides and all FPs[2]	17500

The results shown in Tables IV and V highlight the interest of a BUC approach for FR used fuels. The reactivity loss due to the burnup ranges from 0.053 Δk (taking account for 16 actinides only) to 0.175 Δk (taking account for all actinides and fission products).
As expected for the FR fuels and contrary to PWR-UOx trend, the highest contribution to reactivity loss is due to fission products: the major fission products and the major actinides respectively contribute around 70% and 30% of the total reactivity loss due to the burnup.
Moreover, it is relevant to underline that the 15 BUC fission products represent about 95% of the reactivity loss due to all FPs[3] and the Samarium 149 represent 90% of the reactivity worth of these 15 FPs.
These results are really auspicious and give us the possibility to simplify the list of the BUC fission products to the ^{149}Sm only. The opportunity to take account of only one fission product is highly promising and will considerably simplify the way to determine a penalizing fuel inventory.

4. CONCLUSIONS

The concept of taking into account the reduction of the reactivity of nuclear used fuel due to their burnup is referred to as "Burnup Credit" (BUC). Economic incentives offered by the implementation of a BUC methodology in criticality analyses of Light Water Reactor (LWR) used fuel facilities are nowadays well-demonstrated.

The current prospect of reprocessing the fissile and fertile subassemblies of the PHENIX reactor in France, and the development of the 4th-Generation Fast-neutron Reactors (FR) enable research projects in relevant fuel cycle operations. Under these circumstances, the CEA and AREVA-NC have decided to study a burnup credit approach for irradiated fuels of the PHENIX reactor.

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[2] 157 fission products

The first results highlight the interest of a burnup credit approach for FR fuels and show that the potential interest of a burnup credit approach for FR fuels is possible because the back-end fuel cycle involve used fuels in neutron thermal spectra configurations.

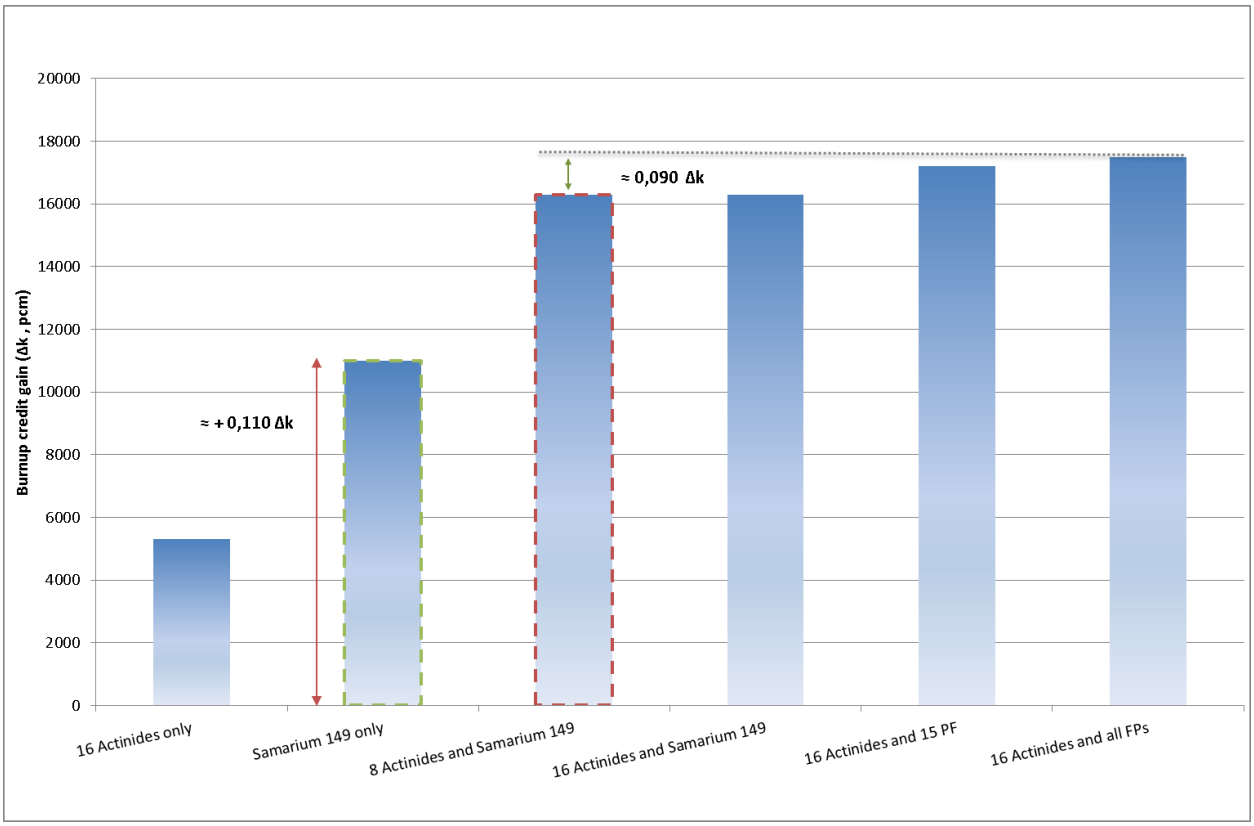

Figure 5 Burnup credit gain for a storage pool of FR fuels as a function of the nuclides involved in the fuel inventory

As illustrated on Fig.5, the main issues of this work are as follow:

- The reactivity loss due to the burnup has been calculated for a burnup of 92GWd/t and a 5-year cooling time and it ranges from 0.163 Δk (taking account for 12 actinides and ^{149}Sm only) to 0.175 Δk (taking account for all actinides and fission products) ;
- As expected for the FR fuels, the highest contribution to reactivity loss is due to fission products: the major fission products and the major actinides respectively contribute around 70% and 30% of the total reactivity loss due to the burnup ;
- The 15 BUC fission products represent about 95% of the reactivity loss due to all FPs and the Samarium 149 represent 90% of the reactivity worth of these 15 FPs; These results are really auspicious and give us the possibility to simplify the list of the BUC fission products to the ^{149}Sm only.

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