EXPERIMENTAL VALIDATION OF THE CODE SYSTEM "DARWIN" FOR SPENT FUEL ISOTOPIC PREDICTIONS IN FUEL CYCLE APPLICATIONS

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ABSTRACT

The DARWIN package, developed by the CEA and its French partners (COGEMA, EDF and FRAMATOME) provides the required parameters in fuel cycle applications and characterizing the irradiated fuels from reactors: fuel inventory, decay heat, activity, neutron, γ, α, β sources and spectrum, radiotoxicity.
In order to validate this code system for spent fuel inventory calculations, a large experimental programme has been performed in France, based on spent fuel chemical assays.
Experimental data are based on chemical analysis measurements from fuel rod cuts irradiated in French PWR reactors and from full assembly dissolutions at the COGEMA/La Hague reprocessing plants. This enables us to cover a large range of UOx fuels with various enrichments in ^{235}U, 3.1% to 4.5%, associated with burnups from 10 GWd/t to 60 GWd/t. Recently, MOx fuels have also been investigated, with a initial Pu amount in the central zone of 5.6 % and a maximum burnup of 45 GWd/t. Uranium, Plutonium, Americium and Curium isotopes were analysed in PWR samples. Furthermore, Fission Products involved in Burn up Credit studies were measured.

1 INTRODUCTION

In order to validate the DARWIN code system for fuel inventory calculations, a large experimental programme based on spent fuel chemical analysis has been carried out in France since 1993. Uranium, plutonium, neptunium, americium and curium isotopes have been analysed in PWR samples. Furthermore the 15 fission products used in Burnup Credit (BUC) criticality calculations have been measured.
The available experimental information consists of chemical assays from fuel rod cuts irradiated in French PWR reactors and of solution samples derived from full assembly dissolutions at the COGEMA/La Hague reprocessing plants.
This paper describes the spent fuel inventory validation which is carried out making a comparison between the DARWIN calculation results and experimental results. The trends observed from experimental validation, analysis of isotopes formation ways and perturbation studies allow us to propose revisions on nuclear data ; some of them are presented in this paper.

2 PRESENTATION OF THE DARWIN PACKAGE

DARWIN [1] [2] is the reference calculation package for the fuel cycle of all types of reactors. It was developed by the CEA and its French partners (COGEMA, EDF and FRAMATOME) to estimate the physics quantities characterizing the spent fuels from reactors: material balance, decay heat, activity, neutron, γ, α, β sources and spectrum, radiotoxicity.

DARWIN is devoted to all cycle studies, with current fuels (UOx, MOx) or innovative fuels (MIX, APA, PuTh) and for every nuclear road (Pressured Water Reactor, Fast Breeder Reactor, Boiling Water Reactor, Advanced Reactors). DARWIN is also used in the back-end cycle for actinide incineration (SPIN) or long term interim storage studies.

The simplified DARWIN structure, based on new codes and libraries is described in the Figure 1.

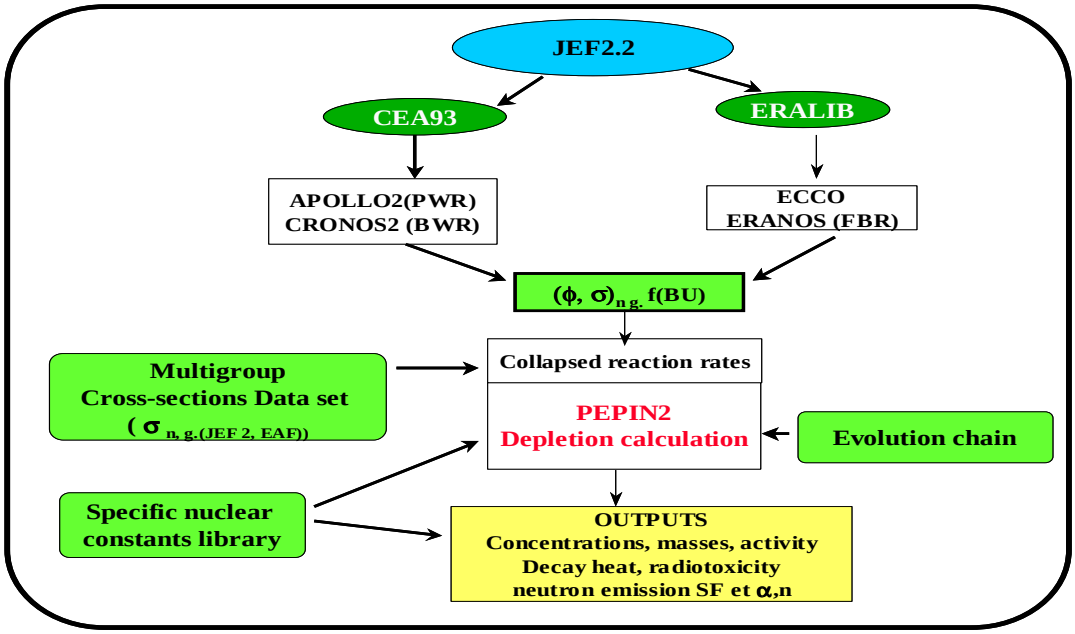

Figure 1 : the DARWIN Package

The PEPIN2 program performs the nuclide depletion calculations. Different libraries feed this module :

- neutronics data provided by French assembly transport codes APOLLO2 [3] (for PWR studies), ECCO-ERANOS system (for FBR studies) : these data are self-shielded cross sections and neutron spectra,
- nuclear data (decay data, fission and (α,ν) yields) and evolution chains
- complementary cross-sections, missing from the transport codes libraries, specially for activation products. They are included in the 'cycle library'.

The basic nuclear data comes from the JEF2.2 [4] European evaluation ; in the scope of our study on PWR assemblies, the neutronic data necessary to the depletion module are provided by the APOLLO2 code and its CEA93 library and benefits from its extensive experimental validation achieved in the framework of the PWR lattices.

DARWIN makes possible the retrieval of cumulated reaction rates during irradiation in order to give the origin of every isotope build-up. Furthermore, a "PERTURBATION" of main nuclear data, such as capture cross-section, initial isotopic composition, flux, is also available and allows sensitivity studies.

3 CALCULATIONAL MODELS

The accuracy of the DARWIN results depends mainly on the APOLLO2 assembly calculation. APOLLO2 solves the integral form of the Boltzmann equation through the collision probability method. We use an APOLLO2 reference calculation route devoted to depletion calculation of 17x17 UO2 and MOx PWR assemblies [5] [6]. This calculation route uses the CEA93 cross-section library in a 172 groups structure processed from the JEF2 evaluations. Among the themes studied in the optimisation of the assembly calculation, we can mention :

- the radial discretization of the fuel pellet in 4 rings in order to give a faithful representation of the resonant absorption of U238 in the pin and of the actinide and fission product concentration profile ;
- the spatial calculation achieved using the UP1 HETE approximation. This enables the probability of leakage PIS and the probability of transmission PSS to be calculated for the true geometry, the interface currents are considered to be linearly anisotropic ;
- a grouping of cells with similar flux within a unique 'physical cell' ;
- the self-shielding of resonant isotopes, with a differentiated treatment for each one depending on the physical characteristics of their cross-sections ;
- optimised evolution steps
- for the MOx assembly calculation, the UO_2 environment must be accounted for.

The PEPIN2 evolution module then uses the results provided by APOLLO2, self-shielded cross-sections and multigroup spectra - to make up the collapsed library with burnup dependent cross-sections, required in order to characterize the isotopes described in the depletion chains.

4 THE EXPERIMENTAL DATABASE OF IRRADIATED PWR FUELS

We describe here the main experimental programmes carried out on irradiated UOX and MOx PWR fuels.

The experimental information can be divided into two categories :

- small samples of fuel pins, irradiated in French reactors, with positions in the assembly well characterized. These are sensitive to local irradiation conditions. These time-consuming and expensive experiments provide very accurate results for a limited number of samples
- dissolution aliquots of entire assembly sets ; these are numerous and very different as to the type of assemblies covered, but the information on the irradiation condition is limited.

Five programmes related to fuel samples are used for the experimental validation of actinides and fission products inventory. Four programmes are devoted to UOx fuels : BUGEY3, FESSENHEIM II, GRAVELINES, CRUAS and one to MOx fuels called Saint Laurent B1.

- **BUGEY3** uses standard fuel with 3.1% initial enrichment and the assembly consists of 17X17 rods with a zircaloy clad. The maximum burn-up is 40 GWd/t.

- **FESSENHEIM II** allows the study of UOx fuel (3.1% ^{235}U) with high burn-up up to 60 GWd/t.

- **GRAVELINES** is devoted to the extension of the calculation scheme validation for high burn-up (five irradiation cycles) with higher enrichment corresponding to 4.5%. This experimental programme on a 900 MWe PWR is the most important being carried out in France today.

- **CRUAS** is devoted to the validation of the URE (Uranium Reprocessed Enriched) fuel, using reprocessed then enriched uranium. It allows the ^{236}U capture validation.

- **Saint Laurent B1 (SLB1)** concerns French 17x17 MOx assemblies. The initial Pu amount in the central zone is 5.6 % and the maximum burnup 45 GWd/t.

From full assembly dissolutions at COGEMA/La Hague reprocessing plants, we get uranium and plutonium chemical analyses. The assembly involved are 17X17 PWR at 3.1%, 3.25% and 3.45% enrichment with burnup between 25 and 40 GWd/t.

5 RESULTS OF THE EXPERIMENTAL VALIDATION

The calculation-experiment comparison (C-E)/E (in %) is summarised below for actinides and important fission products, including the fission products involved in Burnup Credit studies.

The calculation burnup is adjusted using an experimental indicator, namely the $^{145}Nd/^{238}U+^{146}Nd/^{238}U$ sum for PWR fuel cut analyses and the $^{235}U/^{238}U$ residual enrichment for the assembly dissolutions (the neodymium chemical analyses are not available).

In the following tables, the total uncertainties (1σ) correspond to the combination of uncertainties on chemical assays and determination of the burnup of the assembly derived from Nd (or ^{235}U) isotopics. When more than one sample is available for the same burnup, the spread of results is also considered.

5.1 URANIUM ISOTOPE RESULTS

For the uranium isotope build-up calculation, a good accuracy is more important for UOx fuels than for MOx because in MOx fuels depleted uranium is used.

Table I : (C-E)/E (%) for 'uranium' inventory

Fuel↓	*BU (GWd/t)*↓	$^{234}U/^{238}U$	$^{235}U/^{238}U$	$^{236}U/^{238}U$
UOx "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	2.5 ± 0.6	0.4 ± 1.3	-3.6 ± 1.0
	25	5.2 ± 1.1	0.3 ± 2.4	-3.1 ± 1.2
	40	1.7 ± 0.7	0.04 ± 2.1	-3.2 ± 0.4
	50	-5.9 ± 1.1	0.1 ± 2.7	-4.2 ± 0.1
	60	-2.3 ± 2.3	3.3 ± 7.5	-4.1 ± 0.2
UOx "GRAVELINES" 4.5 % ^{235}U	*30*	-0.3 ± 2.0	-0.7 ± 1.7	-3.4 ± 1.3
	40	0.2 ± 2.2	-0.4 ± 2.8	-3.9 ± 1.0
	50	-4.7 ± 2.2	1.4 ± 4.1	-4.9 ± 0.6
	60	1.4 ± 2.5	0.8 ± 5.3	-4.3 ± 0.3
UOx "La HAGUE" 3.1 % ^{235}U	*30*	-16.6 ± 2.2	-0.01 ± 2.8	-1.4 ± 1.0
	40	-15.7 ± 2.2	0.03 ± 4.1	-1.2 ± 0.6
UOx "La HAGUE" 3.25 % ^{235}U	*35*	-12.5 ± 2.2	-0.01 ± 2.8	-2.4 ± 1.0
	45	-13.7 ± 2.2	-0.01 ± 2.8	-1.8 ± 1.0
UOx "La HAGUE" 3.45 % ^{235}U	*35*	-16.5 ± 2.2	-0.05 ± 2.8	-4.7 ± 1.0
URE "CRUAS" 3.5 % ^{235}U	*15*	0.2 ± 0.5	-0.2 ± 1.0	-0.5 ± 0.4
	25	0.2 ± 0.8	0.8 ± 1.7	-0.3 ± 0.3
	35	-0.5 ± 1.1	0.02 ± 3.1	-1.1 ± 0.2
MOx SLB1	*10*	4.4 ± 2.1	0.3 ± 0.5	-7.6 ± 1.0
	30	-5.0 ± 1.3	1.2 ± 0.9	-5.8 ± 0.9
	42	-3.1 ± 1.2	1.7 ± 1.4	-4.8 ± 0.6

Table I points out that uranium isotope concentrations are well predicted for fuel cycle applications.

The ^{235}U depletion is accurately simulated up to very high burnup, even if the quantities involved become very small, and the associated uncertainty and the sensitivity to the cross-section become greater.

For the "La Hague " results, the (C-E)/E is negligible because the calculation burnup is adjusted using the $^{235}U/^{238}U$ residual enrichment.

The abundance of ^{236}U is underestimated in standard UOx fuel and in MOx calculations, principally due to the underestimation of the ^{235}U capture Resonance Integral in JEF2. Many works have been done on this subject and a new evaluation, called Leal-Derrien-Wright-Larson will be introduced in the European JEFF3.0 file [7] [8] [9]. The ^{236}U build-up will be very well predicted : (C-E)/E =-1% ± 1.0% at 60 GWd/t for UOx fuels and a mean value of -3% ± 2.0% in Mox fuels.

In URE fuel, the underestimation is no more existing because the ^{236}U concentration is mainly linked, at low burnup, on the initial ^{236}U content and not on the ^{235}U capture.

The abundance of ^{234}U is well reproduced by calculation if the initial content in the fresh fuel is well known. This is the case, for example for the CRUAS experiment.

5.2 PLUTONIUM ISOTOPE RESULTS

In the plutonium series, all isotopes are important in fuel cycle studies. In the front-end cycle, during the PuO2 fuel fabrication, plutonium 238 is the major contributor to the alpha activity and to the neutronic emission. During this fabrication, plutonium 236 and its daughters contribute to the increasing of the gamma dose. However the most part of this gamma dose is due to americium 241, formed by decaying of ^{241}Pu. In the back-end cycle, the long-term toxicity is mainly linked to ^{239}Pu, ^{240}Pu, ^{242}Pu. Furthermore, ^{242}Pu is the father of ^{244}Cm well known for its contribution to the neutron dose and activity from spent MOx fuels for cooling time up to 100 years after discharge from the reactor. At last, we must mention the fissile ^{239}Pu, highly involved in criticality calculations.

Table II : (C-E)/E (%) for 'plutonium' inventory

Fuel↓	*BU(GWd/t)*↓	$^{238}Pu/^{238}U$	$^{239}Pu/^{238}U$	$^{240}Pu/^{238}U$	$^{241}Pu/^{238}U$	$^{242}Pu/^{238}U$
UOx "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	-12.3 ± 3.2	-0.6 ± 1.1	-0.9 ± 1.6	-4.0 ± 2.1	-6.6 ± 3.9
	25	-9.0 ± 4.6	-0.1 ± 1.5	-0.7 ± 2.1	-3.2 ± 2.9	-5.4 ± 5.0
	40	-6.4 ± 2.0	2.5 ± 0.9	-1.9 ± 0.8	-1.0 ± 1.2	-7.6 ± 2.2
	50	-4.2 ± 1.6	2.0 ± 1.1	-1.2 ± 0.6	-0.4 ± 1.2	-6.1 ± 1.8
	60	-8.7 ± 2.5	3.1 ± 2.4	-0.7 ± 0.9	-0.3 ± 2.6	-8.7 ± 2.9
UOx "GRAVELINES" 4.5 % ^{235}U	*30*	-12.2 ± 5.3	-1.6 ± 0.6	-2.2 ± 2.3	-5.0 ± 2.7	-6.8 ± 5.5
	40	-10.0 ± 5.0	-0.4 ± 0.2	-1.9 ± 2.0	-4.2 ± 2.0	-6.9 ± 4.8
	50	-10.3 ± 4.5	+1.6 ± 0.1	-1.8 ± 1.5	-3.0 ± 1.2	-8.6 ± 4.3
	60	-9.9 ± 4.3	+1.9 ± 0.2	-1.5 ± 1.2	-2.6 ± 1.0	-7.2 ± 3.9
UOx "La HAGUE" 3.1 % ^{235}U	*30*	-14.9 ± 5.0	+0.9 ± 0.2	-1.7 ± 2.0	-2.5 ± 2.0	-11.7 ± 4.8
	40	-15.4 ± 4.5	+0.7 ± 0.1	-1.0 ± 1.5	-6.7 ± 1.2	-11.9 ± 4.3
UOx "La HAGUE" 3.25 % ^{235}U	*35*	-14.1 ± 5.0	1.1 ±0.2	-1.7 ± 2.0	-1.9 ± 2.0	-11.1 ± 4.8
	45	-11.7± 4.5	2.1 ± 0.1	-0.5 ± 1.5	-0.5 ± 1.2	-9.5 ± 4.3
UOx "La HAGUE" 3.45 % ^{235}U	*35*	-18.4 ± 5.0	-0.1 ± 0.2	-2.5 ± 2.0	-2.6 ± 2.0	-11.5 ± 4.8
URE "CRUAS" 3.5 % ^{235}U	*15*	-2.8 ± 3.9	1.7 ± 1.2	-2.2 ± 2.6	0.5 ± 3.6	-5.1 ± 5.8
	25	-2.8 ± 3.4	1.0 ± 1.0	-0.6 ± 2.2	-1.6 ± 2.7	-4.9 ± 5.1
	35	-1.7 ± 3.3	1.9 ± 1.0	-0.5 ± 1.7	-0.6 ± 1.8	-4.2 ± 4.3
MOx SLB1	*10*	-6.3 ± 0.6	0.05 ± 0.7	0.6 ± 0.2	-5.2 ± 0.5	-1.4 ± 0.5
	30	-6.3 ± 0.5	-0.2 ± 1.1	0.9 ± 0.2	-3.0 ± 0.5	-2.7 ± 0.8
	42	-6.3 ± 0.3	-1.6 ± 1.4	0.7 ± 0.5	-3.5 ± 0.7	-3.9 ± 0.8

The ^{239}Pu is well predicted within 2% accuracy. The slight overestimation increasing with burnup could indicate an underestimation of its (n,γ) cross-section in JEF2.

The prediction of ^{240}Pu is very well estimated, confirming the correct modelling of Doppler/self-shielding resonance for ^{240}Pu at 1 eV. The negative reactivity worth of this most poisoning isotope will be very well represented in our BUC calculations.

^{241}Pu is slightly underestimated. Its prediction is highly sensitive to ^{239}Pu capture. This result could point out again a slight under-estimation of ^{239}Pu (n,γ) cross-section in JEF2 evaluation.

^{242}Pu is underestimated. This result shows that ^{241}Pu capture cross-section in JEF2.2 is under-estimated. Hence, a new evaluation of Pu^{241} will be introduced in JEFF3.0 : the increase of the capture will correct the ^{242}Pu, ^{243}Am and ^{244}Cm build-up [10] [11] [12].

The abundance of ^{238}Pu is underestimated by about 10% in standard UOx fuels and by -6% in MOx fuels.
In BUC application, this underestimation is not very penalizing because the ^{238}Pu is not a great contributor to the actinide BUC negative reactivity worth (10% of the total minor actinide reactivity worth at 40 GWd/t). However ^{238}Pu is responsible of the main part of alpha activity and neutron emission during the PuO2 fuel fabrication, so its build-up must be well known [13].
In order to understand this large underestimation, we have investigated the ^{238}Pu formation ways in UOx and MOx fuels thanks to the INVERSION module of the DARWIN package. The results are presented in Table III for UOx, URE and MOx fuels respectively.

Table III : Formation ways (in %) of ^{238}Pu in UOx and MOx fuels (at discharging)

Burnup (GWd/t) → Formation ways in **standard UOx fuels**↓	20	30	40	50	60
^{235}U(n,γ) → ^{236}U(n,γ) → ^{237}U(β–) → ^{237}Np(n,γ) → ^{238}Np(β–) → ^{238}Pu	66.0	71.0	73.0	73.8	75.6
^{236}U(n,γ) → ^{237}U(β-) → ^{237}Np(n,γ) → ^{238}Np(β-) → ^{238}Pu	6.7	4.8	3.8	3.0	2.6
^{238}U(n,γ) → ^{239}U(β-) → ^{239}Np(β-) → ^{239}Pu(n,γ) → ^{240}Pu(n,γ)→ ^{241}Pu(β-)→ ^{241}Am(n,γ)→ ^{242m}Am(β-) →^{242}Cm(α) → ^{238}Pu	2.6	5.4	7.7	9.6	9.7
^{238}U (n,2n) → ^{237}U (β-) → ^{237}Np(n,γ) → ^{238}Np(β-)→ ^{238}Pu	24.7	18.8	15.5	13.6	12.1

Burnup (GWd/t) → Formation ways in **URE fuels**↓	20	30	40	50
^{235}U(n,γ) → ^{236}U(n,γ) → ^{237}U(β–) → ^{237}Np(n,γ) → ^{238}Np(β–) → ^{238}Pu	7.1	13.5	16.8	19.1
^{236}U(n,γ) → ^{237}U(β-) → ^{237}Np(n,γ) → ^{238}Np(β-) → ^{238}Pu	87.2	78.6	73.8	70.5
^{238}U(n,γ) → ^{239}U(β-) → ^{239}Np(β-) → ^{239}Pu(n,γ) → ^{240}Pu(n,γ)→ ^{241}Pu(β-)→ ^{241}Am(n,γ)→ ^{242m}Am(β-) → ^{242}Cm(α) → ^{238}Pu		2.4	4.0	5.0
^{238}U (n,2n) → ^{237}U (β-) → ^{237}Np(n,γ) → ^{238}Np(β-)→ ^{238}Pu	5.7	5.5	5.4	5.3

Burnup (GWd/t) → Formation ways in **MOx fuels**↓	10	30	45
^{235}U(n,γ) → ^{236}U(n,γ) → ^{237}U(β–) → ^{237}Np(n,γ) → ^{238}Np(β–) → ^{238}Pu			1.7
^{238}U(n,γ) → ^{239}U(β-) → ^{239}Np(β-) → ^{239}Pu(n,γ) → ^{240}Pu(n,γ)→ ^{241}Pu(β-)→ ^{241}Am(n,γ)→ ^{242m}Am(β-) →^{242}Cm → ^{238}Pu			2.2
^{238}U (n,2n) → ^{237}U (β-) → ^{237}Np(n,γ) → ^{238}Np(β-)→ ^{238}Pu	1.0	4.1	7.4
^{239}Pu(n,γ) → ^{240}Pu(n,γ) → ^{241}Pu(β-) → ^{241}Am(n,γ) → ^{242m}Am(β-) → ^{242}Cm(α) → ^{238}Pu		2.0	7.9
^{240}Pu(n,γ) → ^{241}Pu(β-) →^{241}Am(n,γ) → ^{242m}Am(β-) → ^{242}Cm(α) → ^{238}Pu		5.3	12.6
^{241}Pu(β-) → ^{241}Am(n,γ) → ^{242m}Am(β-) → ^{242}Cm(α) → ^{238}Pu	1.2	6.5	8.9
^{241}Am(n,γ) → ^{242m}Am(β-) → ^{242}Cm(α) → ^{238}Pu	18.8	39.1	35.2
^{238}Pu (initial)	79.0	43.0	24.1

In MOx fuels, table III shows that plutonium 238 is mainly formed by the decaying of ^{242}Cm, coming from the ^{241}Am (n,γ) reaction. The underestimation of ^{238}Pu build-up is probably linked to an underestimation of ^{241}Am capture cross-section. Using the PERTURBATION module of the DARWIN package, we can conclude that an increasing of 20% of ^{241}Am cross-section will reduce the current -6% (C-E)/E discrepancy to -1%.
However this correction on ^{241}Am cross-section must be confirmed by rigorous analyses from nuclear physicists.
^{238}Pu build-up is also linked to the knowledge of ^{242}Cm decay constant ; however this value is well known and its associated uncertainty can't explain the underestimation on ^{238}Pu inventory : $\lambda = 162.94$ days $\pm$ 1.44 hours.
^{238}Pu disappeared by neutron capture ; an overestimation of ^{238}Pu (n,γ) cross-section could explain the inventory underestimation and this cross-section must be investigated.

In UOx fuels, a lot of parameters are involved in ^{238}Pu inventory calculation. The underestimation could be due to an underestimation of ^{241}Am and/or an underestimation of ^{235}U capture cross-sections and/or an

underestimation of ^{237}Np capture cross-section and/or to an underestimation of (n,2n) ^{238}U reaction (contribution of 25% to the ^{238}Pu formation at low burnup) and/or to an overestimation of ^{238}Pu (n,γ) cross-section.

In order to evaluate the sensitivity of ^{238}Pu build-up to all those possible origins of underestimation, we made some cross-section perturbations presented in Table IV.

Table IV : Results of perturbation studies in order to improve ^{238}Pu build-up

Perturbed Reaction	Modification on nuclear data (%)	Impact on C-E/E (%)	
		20 GWd/t	60 GWd/t
^{241}Am (n,γ)	+20	+1.2	+1.6
(n,2n) ^{238}U	+15	+3.0	+1.9
^{235} U (n,γ)	+10	+2.5	+2.2
^{237}Np (n,γ)	+5	+4.0	+2.6
^{238}Pu (n,γ)	-10	+1.0	+ 3.0

We already know that in JEF2.2, there is an underestimation of the ^{235}U capture Resonance Integral and an underestimation of (n,2n) ^{238}U reaction. The correction of these data in the JEFF3.0 future nuclear data file will improved the ^{238}Pu inventory calculation leading to a quite satisfactory (C-E)/E ≈ -5% ± 5%. However we have to progress on the other nuclear data knowledge, ^{241}Am (n,γ), ^{237}Np (n,γ) and ^{238}Pu (n,γ).

In URE fuels, the ^{238}Pu inventory is well calculated. It comes mainly from the depletion chain ^{236}U(n,γ) → ^{237}U(β-) → ^{237}Np(n,γ) → ^{238}Np(β-) → ^{238}Pu as shown in Table III for URE fuels. This indicates that ^{236}U(n,γ) and ^{237}Np(n,γ) cross-sections are quite satisfactory.

5.3 MINOR ACTINIDES RESULTS

5.3.1 THE NEPTUNIUM ISOTOPE

The neptunium 237 is an important minor actinide because of its contribution to the long-term toxicity. Furthermore, it could be interesting in BUC studies : its poisoning lead to a -620 pcm antireactivity in UOx fuel at 40 GWd/t.

Table V : (C-E)/E (%) for 'neptunium' inventory

Fuel↓	*BU (GWd/t)*↓	^{237}Np/^{238}U
UOx "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	-10.5 ± 4.4
	25	-7.2 ± 4.3
	40	-0.6 ± 2.1
	50	-3.2 ± 2.5
	60	3.0 ± 3.1
UOx "GRAVELINES" 4.5 % ^{235}U	*30*	-0.3 ± 4.7
	40	-2.4 ± 4.2
	50	-4.4 ± 3.4
	60	-3.0 ± 3.1
URE "CRUAS" 3.5 % ^{235}U	*15*	-4.1 ± 3.2
	25	-0.2 ± 3.1
	35	3.9 ± 3.0
MOx SLB1	*10*	-11.3 ± 2.5
	30	-11.1 ± 2.1
	42	-7.4 ± 1.9

The abundance of ^{237}Np is underestimated. This underestimation could be due to the formation of ^{237}Np ; the Table VI below gives the formation ways for the different fuels studied.

Table VI : Formation ways of ^{237}Np in UOx and MOx fuels

Taux de combustion (GWj/t) → Formation ways in standard **UOx**↓	20	30	40	50	60
U235(n,γ) →U236(n,γ)→ U237(β–) → Np237	74.6	80.2	83.1	84.8	86.2
U236(n,γ)→ U237(β–) → Np237	5.2	3.8	3.1	2.5	2.2
U238(n,2n) → U237(β–) → Np237	**20.2**	16.0	13.8	12.7	11.6

Taux de combustion (GWj/t) → Formation ways in **URE**↓	20	30	40	50
U235(n,γ) →U236(n,γ)→ U237(β–) → Np237	9.9	18.3	22.4	25.0
U236(n,γ)→ U237(β–) → Np237	84.4	76.1	72.1	69.4
U238(n,2n) → U237(β–) → Np237	5.7	5.6	5.5	5.6

Taux de combustion (GWj/t) → Formation ways in **MOx**↓	10	30	45
U235(n,γ) →U236(n,γ)→ U237(β–) → Np237	8.3	16.2	20.1
U236(n,γ)→ U237(β–) → Np237	5.4	4.7	2.2
U238(n,2n) → U237(β–) → Np237	**85.6**	**78.1**	**76.5**
Am241(α) → Np237	0.7	1.0	1.2

In MOx fuels ^{237}Np is mainly produced (80% at 45 GWd/t) by ^{238}U(n,2n) reaction as shown in Table VI suggesting that the (n,2n) U^{238} cross-section is underestimated in JEF2. This is confirmed by comparison between different evaluations [10].
In UOx fuels the underestimation of ^{237}Np build-up is due on one hand to the underestimation of the ^{235}U capture Resonance Integral and in the other hand to the underestimation of (n,2n)^{238}U in JEF2, contributing to 20% in the ^{237}Np build-up at low burnup (see Table VI).

5.3.2 THE AMERICIUM ISOTOPES

The main americium isotopes are significant in fuel cycle studies due to their radioactive characteristics and also to the antireactivity introduced in spent fuel.
Americium 241 is a gamma emitter embarrassing for fuel transport, fabrication or storage and americium 243 is a great contributor to long-term toxicity (half-life of de 7360 years).
Furthermore, it could be very interesting to take into account 241 and 243 americium isotopes in burnup credit applications because of there important contributions to the total antireactivity. This is emphasized in MOx fuels as shown in the Table VII below, giving the reactivity effect of absorbing actinides in UOx and MOx fuels at 40 GWd/t for 5 years cooling time.

Table VII : reactivity worth of plutonium and americium isotopes in spent fuels(in pcm 10^{-5} Δk/k)
(40 GWd/t, cooling time 5 years)

Isotope	UOx	MOx
^{238}Pu	-310 pcm	-760 pcm
^{240}Pu	-8300 pcm	-17500 pcm
^{242}Pu	-710 pcm	-1500 pcm
^{241}Am	**-1290 pcm**	**-4600 pcm**
^{243}Am	**-280 pcm**	**-1000 pcm**

The ^{242m}Am build-up must be well calculated because on one hand the critical mass of this isotope is very weak, and on the other hand, in burnup credit studies, the small positive reactivity introduced by Am242m build up is neglected in spent LWR-UO2 fuel (ρ= +27 pcm at 40 GWd/t /10/) ; but in MOX spent fuel this fissile isotope, σ_f^{2200} = 6886 barns in JEF2 library, must be accounted for : ρAm242m = +500 pcm in the 20-50 GWd/t burnup range.

Table VIII : (C-E)/E (%) for 'americium' inventory

Fuel↓	*BU (GWd/t)*↓	^{241}Am/^{238}U	^{242m}Am/^{238}U	^{243}Am/^{238}U
UOx "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	-6.3 ± 3.4	-25.2 ± 10.1	-10.6 ±7.8
	25	-2.9 ± 3.1	-9.9 ± 3.7	-10.6± 7.5
	40	-3.0 ± 1.2	-17.3 ± 3.0	-4.5 ± 3.4
	50	-13.2 ± 2.6		-10.5 ± 2.9
	60	-3.9 ± 2.4	-21.1 ± 11.4	-8.8 ± 4.6
UOx "GRAVELINES" 4.5 % ^{235}U	*30*	-5.0 ± 2.8	-33.0 ± 5.0	-10.3 ± 7.7
	40	-3.5 ± 2.1	-25.0 ± 3.2	-7.9 ± 7.0
	60	+1.3 ± 1.0	-16.2 ± 2.4	-4.8 ± 5.6
URE "CRUAS" 3.5 % ^{235}U	*35*	-1.3 ± 2.3	-17.0 ± 2.8	-4.8 ± 6.4
MOx SLB1	*10*	-4.3 ± 1.5	-27.7 ± 1.9	-11.1± 2.1
	30	2.3 ± 1.3	-23.3 ± 2.0	-7.1 ± 1.9
	42	1.6 ± 1.6	-22.7 ± 2.1	-6.0 ± 1.6

The ^{241}Am, formed by decaying of ^{241}Pu, is reasonably estimated.
^{242m}Am is underestimated by about -20%. This suggests increasing ^{241}Am capture cross-section. We must also improve the knowledge of the branching ratio of ^{241}Am to ^{242m}Am in the epithermal range.
The underestimation in ^{242}Pu generates the underestimation for ^{243}Am. The increase of the ^{241}Pu capture in the future European file JEFF3 will partially correct this underestimation.

5.3.3 THE CURIUM ISOTOPES

The importance of curium isotopes, in fuel cycle applications, is linked to their long radioactive decay. Curium 244 has not really a long half-life (18 years) but is the major contributor to the neutron dose and activity from spent MOx fuels for cooling time up to 100 years after discharge from the reactor.

Concerning the contribution to the curium isotopes in criticality studies, one can notice that these isotopes are neglected in UOX burnup studies due to their small concentrations (curium worth : -30 pcm in 40 GWd/t spent UOX assemblies [14]). However, due to the increasing amount of Curium in MOX fuel, particularly of the fissile isotopes Cm243 and Cm245 (σ_ϕ^{2200} = 500 barns and 2130 barns respectively), the BUC component of Cm isotopes is non negligible ; it becomes positive beyond 30 GWd/t due to Cm245 build up at high burnups. The Cm243, Cm244 and Cm245 nuclides must be accounted for in MOx fuels BUC calculations. The Cm242 (T1/2 =163 d) and Cm246 absorbing isotopes are rejected due to their negligible poisoning worth.

Table IX : (C-E)/E (%) for 'curium' inventory

Fuel↓	*BU (GWd/t)*↓	$^{243}Cm/^{238}U$	$^{244}Cm/^{238}U$	$^{245}Cm/^{238}U$	$^{246}Cm/^{238}U$	$^{247}Cm/^{238}U$
UOX "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	-42.7 ± 11.3	-33.7 ± 8.0	-27.5 ± 10.4	-81.2 ± 8.7	/
	25	-34.5 ± 8.9	-19.9 ± 9.1	-19.2 ± 10.9	-7.7 ± 17.8	/
	40	-9.6 ± 5.8	-12.1 ± 5.4	-2.3 ± 7.8	-27.8 ± 7.2	/
	50	-28.3 ± 10.2	-21.6 ± 8.1	-18.2 ± 13.4	-31.0 ± 11.3	/
UOX "GRAVELINES" 4.5 % ^{235}U	*30*	-28.1 ± 8.0	-18.5 ± 10.4	-20.0 ± 12.4	-25.0 ± 15.5	-25.6 ± 18.2
	40	-26.5 ± 8.0	-15.4 ± 9.8	-16.8 ± 11.5	-23.0 ± 15.2	-30.2 ± 18.0
	60	-24.5 ± 8.0	-12.4 ± 8.4	-11.4 ± 9.3	-23.1 ± 13.4	-30.2 ± 16.3
URE "CRUAS" 3.5 % ^{235}U	*35*	-5.4 ± 6.3	-2.03 ± 9.3	-0.05 ± 11.2	-12.2 ± 12.8	-12.9 ± 31.7
MOx SLB1	*10*	/	-39.8 ± 2.1	-29.5 ± 2.2	-30.6 ± 3.2	-59.4 ± 4.5
	30	/	-22.3 ± 2.1	-6.6 ± 2.3	-8.3 ± 3.0	-16.6 ± 4.3
	42	/	-16.5 ± 1.5	-5.6 ± 2.1	-7.6 ± 2.4	-14.6 ± 3.7

The curium isotopes are underestimated. The underestimation in ^{242}Pu generates an underestimation for ^{243}Am, and consequently for ^{244}Cm and ^{245}Cm. Recent studies on ^{241}Pu JEF2 capture cross-section enable to partially correct this problem. However an underestimation remains and investigations on cross-sections are needed.
The underestimation in ^{243}Cm build-up needs also to be corrected. Studies on ^{242}Cm capture cross-section are in progress.

5.4 FISSION PRODUCTS RESULTS

5.4.1 THE SAMARIUM ISOTOPES

The samarium isotopes studied are mainly involved in burnup credit studies.

Table X : (C-E)/E (%) for 'samarium' inventory

Fuel↓	*BU (GWd/t)*↓	$^{147}Sm/^{238}U$	$^{149}Sm/^{238}U$	$^{150}Sm/^{238}U$	$^{151}Sm/^{238}U$	$^{152}Sm/^{238}U$
UOX "BUGEY" 3.1 % ^{235}U	*20*	-4.9 ± 1.5	4.5 ± 4.7	-3.7 ± 2.6	-3.1 ± 1.2	0.4 ± 1.9
	40	-5.9 ± 1.0	-3.4 ± 23.3	-3.6 ± 2.2	7.3 ± 1.5	1.1 ± 1.7
UOX "GRAVELINES" 4.5 % ^{235}U	*40*	-6.5 ± 1.1	-8.6 ± 8.4	-3.2 ± 2.3	3.6 ± 1.1	1.4 ± 1.8
	50	-7.0 ± 1.0	5.6 ± 4.8	-4.8 ± 2.1	7.9 ± 1.1	3.5 ± 1.6
	60	-7.4 ± 0.9	-3.5 ± 10.0	-4.1 ± 1.8	12.8 ± 1.0	5.6± 1.7
MOx SLB1	*10*	-5.7 ± 1.6	-7.1 ± 0.6	7.1 ± 2.2	-8.1 ± 0.7	-5.3 ± 2.0
	30	-3.9 ± 1.1	2.6 ± 1.2	-6.3 ± 2.2	-1.2 ± 0.8	-1.4 ± 1.5
	42	-3.8 ± 0.5	-7.7 ± 0.6	-5.7 ± 0.6	1.9 ± 0.3	2.9 ± 0.8

Except the slight overestimation of ^{151}Sm build-up, the samarium isotope calculations are satisfactory. However we can make some remarks on nuclear data.

The ^{147}Sm is underestimated. This isotope is formed by decaying of ^{147}Pm, itself formed by cumulative fissions, and disappeared by neutron capture.
The interpretation of ^{147}Sm samples oscillations in the MINERVE [17] reactor shows that the ^{147}Sm capture cross-section is well calculated (C-E)/E = +1.9% ± 3.2% in UOx lattice and (C-E)/E = -0.3% ± 3.2% in MOx lattice ; furthermore the sensitivity of ^{147}Sm build-up is very low because of the low value of the cross-section (57 barns at 2200m/s) ; so we can conclude that the underestimation of ^{147}Sm is due to its formation. The capture cross-section of ^{147}Pm, and ^{147}Nd cumulative fission yields must be investigated.

The ^{149}Sm seems to be underestimated but the chemical uncertainty is very high and the ^{149}Sm build-up is very sensitive to the power history mesh. However this slight underestimation is confirmed by rigorous APOLLO2 calculation describing precisely the irradiation history.

The ^{150}Sm is underestimated, varying in the direct ratio of its father the ^{149}Sm. The^{151}Sm is also overestimated. This is probably due to an overestimation of ^{151}Pm cumulative fission yields.

^{152}Sm is well predicted but we can notice a slow drift toward overestimation when the burnup increases. This could be due to ^{151}Sm calculation overestimation but also to an underestimation of the ^{152}Sm capture cross-section as suggested by the MINERVE oscillation experiment. More studies are needed to conclude.

5.4.2 THE NEODYMIUM ISOTOPES

The ^{143}Nd is the unique FP involved in BUC studies, the others neodymium isotopes are used as burnup indicator.

Table XI : (C-E)/E (%) for 'neodymium' inventory

Fuel↓	*BU (GWd/t)*↓	^{143}Nd/^{238}U	^{144}Nd/^{238}U	^{148}Nd/^{238}U	^{150}Nd/^{238}U
UOX "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	0.3 ± 1.3	-2.0 ± 1.8	0.2 ± 1.7	-6.7 ± 1.8
	25	0.3 ± 1.7	-2.2 ± 2.5	1.5 ± 2.2	0.7 ± 2.5
	40	/	/	0.2 ± 1.7	/
	50	2.4 ± 1.8	0.2 ± 2.9	-0.6 ± 2.6	-0.8 ± 3.2
UOX "GRAVELINES" 4.5 % ^{235}U	*40*	0.5 ± 1.4	-1.9 ± 2.5	1.6 ± 2.1	0.7 ± 2.4
	50	1.6 ± 1.0	-2.6 ± 2.6	1.5 ± 2.1	0.5 ± 2.4
	60	1.9 ± 0.4	-2.4 ± 1.5	1.4 ± 1.2	0.4 ± 1.4
URE "CRUAS" 3.5 %	*15*	0.0 ± 1.8	-1.8 ± 2.1	0.7 ± 2.1	-0.3 ± 2.3
	25	0.6 ± 1.6	-2.1 ± 2.2	0.7 ± 2.1	-0.1 ± 2.2
	35	0.7 ± 1.2	-2.3 ± 2.3	1.3 ± 2.1	0.6 ± 2.3
MOx SLB1	*10*	0.0 ± 1.9	-1.8 ± 1.5	0.3 ± 2.0	0.3 ± 1.5
	30	0.2 ± 1.7	-2.5 ± 1.4	0.2 ± 2.0	0.9 ± 1.3
	42	1.1 ± 0.8	-2.2 ± 1.4	0.4 ± 1.0	0.4 ± 1.3

The Nd isotopes are very well calculated.

The ^{143}Nd build-up is slightly overestimated for high burnups : +2%. At these burnups, ^{143}Nd concentration is sensitive to its capture cross-section. This result confirms an under-estimation of the thermal capture cross-section of ^{143}Nd. The same conclusion is drawn from the interpretation of ^{143}Nd samples oscillated in Minerve lattices. A new evaluation of ^{143}Nd will be introduced in the future JEFF3 file, in order to increase by 4% the (n,γ) cross-section in the 0-0.2eV energy range [7] [15] [16].

One can notice a slight underestimation of ^{144}Nd concentration due probably to an underestimation of ^{144}Ce cumulative fission yields.

5.4.3 THE CESIUM ISOTOPES

The two most contributing cesium isotopes in fuel cycle studies are ^{137}Cs and ^{135}Cs. ^{137}Cs, and its daughter product ^{137}Ba, are emitters of a high energy gamma radiation. Furthermore, ^{137}Cs, with an half-life of 30 years, is greatly participating, with strontium 90 in the harmful effect of βγ emitter waste. ^{134}Cs is involved in residual heating for cooling time up to 10 years after discharge from the reactor.

Finally, ^{135}Cs is one of the long life fission products contributing to the long-term radiotoxicity and ^{133}Cs, the most absorbing cesium isotope, is involved in burnup Credit studies.

Table XII : (C-E)/E (%) for 'cesium' inventory

Fuel↓	*BU (GWd/t)*↓	$^{133}Cs/^{238}U$	$^{134}Cs/^{238}U$	$^{135}Cs/^{238}U$	$^{137}Cs/^{238}U$
UOX "BUGEY FESSENHEIM" 3.1 % ^{235}U	*20*	-2.7 ± 2.0	-8.1 ± 4.6	18.8 ± 0.6	-1.8 ± 2.3
	25	-3.9 ± 1.9	-14.8 ± 4.8	15.2 ± 0.9	-4.0 ± 2.2
	40	-8.5 ± 1.8	-9.3 ± 4.9	3.9 ± 0.7	-1.2 ± 2.3
	50	-1.3 ± 1.6	-3.7 ± 5.6	5.1 ± 0.9	-4.4 ± 2.2
	60	-2.2 ± 1.5	2.1 ± 6.1	1.8 ± 0.9	-3.6 ± 2.3
UOX "GRAVELINES" 4.5 % ^{235}U	*30*	-4.9 ± 2.0	-11.7 ± 4.0	-1.9 ± 1.9	-5.7 ± 2.2
	40	-3.3 ± 2.0	-10.4 ± 3.8	-2.4 ± 2.1	-5.0 ± 2.3
	50	-3.9 ± 1.8	-8.2 ± 3.5	-3.9 ± 2.2	-6.2 ± 2.3
	60	-3.4 ± 1.2	-8.7 ± 2.9	-3.3 ± 1.6	-5.2 ± 1.5
MOx SLB1	*10*	--2.9 ± 1.4	-10.4 ± 2.6	3.1 ± 0.8	-3.6 ± 1.4
	30	-0.2 ± 1.1	-7.2 ± 2.5	4.7 ± 0.5	-1.8 ± 1.2
	42	-0.6 ± 1.0	-6.4 ± 2.7	1.7± 0.9	-2.6 ± 1.2

A slight and steady under-prediction by -4% is noticed for ^{133}Cs in UOx fuels. This underestimation is reduced to 2% in MOx fuels. A study has shown that the ^{133}Cs inventory is not very sensitive to the ^{133}Cs capture cross-section, so we can conclude that this underestimated abundance is due to an underestimation of ^{133}Xe fission yields. The steady under-prediction in UOx fuels indicates that the increasing of the fission yields for ^{235}U is necessary but that the fission yields for ^{239}Pu and/or ^{241}Pu must be increased too. This last point is confirmed by the MOx results.

^{137}Cs is used as a burnup monitor and must be calculated with about 2% accuracy. Our calculation leads to an under-prediction of the ^{137}Cs concentration; higher in UOx fuels than in MOx ; this indicates that there is an underestimation of ^{137}Cs fission yields in JEF2.2 for ^{235}U and also for ^{239}Pu and ^{241}Pu.

The ^{135}Cs isotope is well calculated when the irradiation history is accurately known (Gravelines and Saint Laurent B1).

The ^{134}Cs build-up is underestimated. This underestimation could be due to an underestimation of ^{133}Cs capture cross-section, or to an overestimation of ^{134}Cs capture cross-section itself or to an underestimation of the ^{134}Cs decay constant. Analyses of all these nuclear data have shown that the discrepancies obtained are not due to an error on these data and we can't conclude today on this underestimation.

5.4.4 THE METALLIC FISSION PRODUCT ISOTOPES

These fission products are mainly analysed for Burn up Credit applications.

Table XIII : (C-E)/E (%) for 'metallic fission products' inventory

Fuel ↓	*BU (GWd/t)*↓	$^{109}Ag/^{238}U$	$^{99}Tc/^{238}$	$^{95}Mo/^{238}$	$^{101}Ru/^{238}U$	$^{103}Rh/^{238}U$
UOX "BUGEY" 3.1 % ^{235}U	*20*		-7.6 ± 3.1	-3.7 ± 2.7	2.9 ± 2.9	3.5 ± 2.8
	40		4.8 ± 3.0	8.4 ± 2.6	15.2 ± 2.9	13.9 ± 2.6
UOX "GRAVELINES" 4.5 % ^{235}U	*40*		-0.6 ± 3.1	5.5 ± 2.7	5.0 ± 2.9	3.4 ± 2.7
	50		2.6 ± 3.1	2.6 ± 2.6	6.4 ± 2.3	4.6 ± 2.5
	60		3.6 ± 3.0	2.1 ± 2.6	0.9 ± 2.9	3.0 ± 2.3
MOx SLB1		87.8 ± 9.0	15.4 ± 6.6	3.8 ± 5.7	20.4 ± 6.1	28.0 ± 6.2
		36.35 ± 6.6	34.9 ± 7.0	16.2 ± 6.1	53.1 ± 7.3	57.2 ± 5.7
		22.8 ± 3.6	46.2 ± 4.1	27.3 ± 3.6	70.4 ± 4.4	67.8 ± 3.3

Metallic fission products isotopic prediction is quite satisfactory in UOx spent fuels since they are in agreement with the experimental standard deviation. However, it should be noted that the dissolution of metallic fission products such as ^{95}Mo, ^{99}Tc, ^{101}Ru and ^{103}Rh could lead to non-soluble deposits. This phenomena is highlighted in MOx fuels. A new programme of dissolution, more accurate, is planned in order to confirm the C/E discrepancies.

5.4.5 THE EUROPIUM AND GADOLINIUM ISOTOPES

^{153}Eu isotope and ^{155}Gd isotope are important poisoning FPs in Burn up credit applications. In order to check ^{155}Gd build-up, the ^{154}Eu, ^{155}Eu and ^{154}Gd were also investigated. Furthermore the ^{154}Eu/^{137}Cs ration can be used as a burnup indicator for large cooling time.

Table XIV : (C-E)/E (%) for 'europium and gadolinium' inventory

Fuel↓	*BU (GWd/t)*↓	^{153}Eu/^{238}U	^{154}Eu/^{238}U	^{155}Eu/^{238}U	^{154}Gd/^{238}	^{155}Gd/^{238}U	^{156}Gd/^{238}U
UOX "BUGEY" 3.1 % ^{235}U	*20*	6.5 ± 3.0	40.9 ± 4.6	8.9 ± 3.8	40.3 ±4.5	-3.1 ± 3.4	
	40	11.8 ± 2.6	80.4 ± 4.2	8.9 ± 4.2	82.4 ±3.8	0.1 ± 3.7	
UOX "GRAVELINES" 4.5 % ^{235}U	*40*	8.9 ± 3.0	54.2 ± 6.4	13.9 ± 4.2	/	4.3 ± 3.8	
	50	11.7 ± 2.8	74.8 ± 6.6	15.4 ± 4.5	72.6 ± 6.0	8.1 ± 3.9	-22.0 ± 3.9
	60	16.4 ± 2.5	94.0 ± 6.2	18.2 ± 4.4	/	11.9 ± 4.0	-15.0 ± 4.0
MOx SLB1	*10*	-2.7 ± 5.3	-0.9 ± 8.7	83.8 ± 4.0	-0.3 ± 9.0	68.8 ± 3.7	
	30	4.0 ± 4.7	13.2 ± 9.9	61.6 ± 7.6	14.2 ± 9.8	53.9 ± 5.4	
	42	8.7 ± 2.7	41.5 ± 7.4	24.4 ± 4.3	38.5 ± 6.7	14.4 ± 4.6	

^{153}Eu is over-predicted up to 16% in UOx fuels at 60 GWd/t. This isotope disappeared by neutron capture and is formed by decaying of 151, 152 and 153 isobars, themselves formed by cumulative fissions.
The interpretation of ^{153}Eu samples oscillations in the MINERVE reactor [17] shows that the ^{153}Eu capture cross-section is well calculated (C-E)/E = -2.9% ± 3.0% in UOx lattice and (C-E)/E = -1.6% ± 4.1% in MOx lattice, with a slight underestimation. However this underestimation can't explain the large overestimation on ^{153}Eu build-up ; so we can conclude that the overestimation of ^{153}Eu is due to its formation depending on fission yields accuracy. The table XV above presents the JEF2.2 fission yields values and associated uncertainties.

Table XV : Cumulative fission yields in JEF2.2 for ^{153}Eu

Fissile isotope	U235	Pu239	Pu241
JEF2 fission yields	0.151E-2	0.395E-2	0.5165E-2
Uncertainties (%)	4.0	8.2	26.3

The table XVI present the impact on ^{153}Eu build-up of fission yields modification (corresponding in fact to the uncertainties).

Tableau XV: Modification of ^{153}Eu fission yields; impact on ^{153}Eu concentrations

Fissile isotope		U235		Pu239		Pu241	
JEF2 fission yields (%)		0.151		0.395		0.5165	
Modification applied on fission yields (in %)		-4.0		-8.2		-26.3	
Impact obtained (in %) on ^{153}Eu concentration	BU	UOx	MOx	UOx	MOx	UOx	MOx
	10 GWd/t	-2.0	-0.03	-3.0	-5.7	-0.7	-6.0
	40 GWd/t	-0.8	-0.03	-4.4	-4.6	-4.6	-9.8
	50 GWd/t	-0.5	-0.03	-4.5	-4.4	-6.0	-10.4

We can see that fission yields uncertainties lead to large effects on ^{153}Eu concentrations. The fission yields for ^{235}U seem to be well known, on the other hand for ^{239}Pu and ^{241}Pu the uncertainties are huge.
One can notice that the ENDFB6 fission yield value for ^{239}Pu is 0.364% [18] that is to say 8% lower than the JEF2 value : the jef2 value must be decrease for ^{239}Pu as well as for ^{241}Pu.

^{155}Gd is strongly over-predicted but agrees with its father, ^{155}Eu, which is produced by capture on ^{154}Eu and directly by fission. Furthermore the ^{156}Gd, daughter by capture of ^{155}Eu is under-predicted.
^{154}Eu is also strongly overestimated and the same level of overestimation is found on its daughter ^{154}Gd. These remarks point out that modification of ^{154}Eu and ^{155}Eu capture cross-section is needed in JEF2 file.

Recent studies on ^{154}Eu and ^{155}Eu capture cross-section confirm that data used in JEF2.2 file are not satisfactory and will be replaced by ENDFB6 evaluations in the new JEFF3.0 file. The use of these evaluations leads to the results on Eu and Gd build-up presented in Table XVI for GRAVELINES UOx fuels and in Table XVII for Saint Laurent B1 MOx fuels.

Table XVI : New (C-E/E) (%) results using σ_c from ENDFB6.7 for ^{154}Eu and ^{155}Eu in GRAVELINES UOx fuels

Burnup (GWd/t)	*40*	*50*	*60*
^{154}Eu/^{238}U	-7.1%	-4.1%	0.8%
^{155}Eu/^{238}U	13.6%	13.5%	15.5%
^{154}Gd/^{238}U		-3.9%	
^{155}Gd/^{238}U	4.0%	6.2%	9.1%
^{156}Gd/^{238}U		-3.8 %	2.4 %

Table XVII : ^{155}Gd (C-E/E) (%) results using σ_c from ENDFB6.7 for ^{154}Eu and ^{155}Eu in MOx fuels

BU (MWd/t)	*13200*	*29200*	*43000*	*46000*
^{155}Gd/^{238}U ; ^{154}Eu and ^{155}Eu *JEF2.2*	68.8 ± 3.7	53.9 ± 5.4	9.7 ± 6.4	19.4 ± 6.6
^{155}Gd/^{238}U ; ^{154}Eu and ^{155}Eu *ENDFB6.7*	19.3 ± 3.7	11.3 ± 5.4	2.1 ± 6.4	2.0 ± 6.6

From the results presented in table XVI and XVII, it is shown that the use of both europium 154 and europium 155 capture cross-section from the ENDFB6.7 evaluation leads to a better gadolinium 155 calculation build-up but a residual overestimation remains, specially in UOx fuels for high burnups ; this could be due to overestimation of ^{155}Sm cumulative fission yields (leading to the overestimation of ^{155}Eu) and/or to an underestimation of the capture cross-section of the ^{155}Gd itself.

The ^{154}Eu concentration, used as a burnup monitor for long cooling time, is well improved with a slight underestimation ; the conclusion is the same for ^{154}Gd.

The ^{156}Gd calculation is also well improved by the ^{155}Eu capture cross-section correction.

CONCLUSION

This paper has described the experimental validation of the fuel cycle package DARWIN, based on the powerful code APOLLO2. The qualification range extends up to 4.5% ^{235}U enrichment and high burnup fractions up to 60 GWd/t for UOx fuels. MOx fuels with 5.6% Pu content and burnup up to 45 GWd/t were also investigated.

The depletion code, DARWIN, has shown its capability to simulate the fuel inventory versus burnup for most of nuclides involved in fuel cycle applications. Furthermore, experimental validation results associated with the formation ways determination and perturbation studies enables us to propose some revision on nuclear data.

The DARWIN package is also well suited for most of burnup credit nuclides inventory, except a slight underestimation of ^{241}Pu and overestimation of europium and gadolinium isotopes. However, the introduction in the future JEFF3.0 file of new evaluations, such as ^{235}U, ^{241}Pu, $^{153\text{-}154}$Eu and ^{155}Eu will highly improved our C/E discrepancies. Nevertheless, isotopic measurement has to be improved to get precise values for metallic fission products.

The P.I.E. data base is currently being extended to higher burnups with PWR rod cuts extracted after 5, 6 and 7 irradiation cycles, up to 80 GWd/t for UOx fuels and after 4 and 5 cycles (up to 60 GWd/t) for MOx fuels.

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