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## 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,  $\gamma$ ,  $\alpha$ ,  $\beta$  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 <sup>235</sup>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,  $\gamma$ ,  $\alpha$ ,  $\beta$  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.

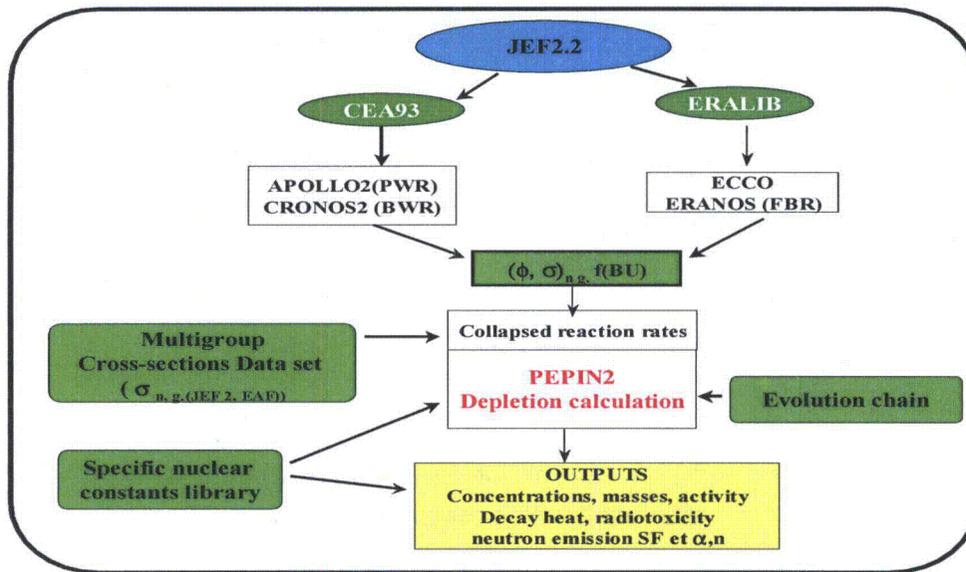


Figure 1 : the DARWIN Package

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  $(\alpha, \nu)$  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 UO<sub>2</sub> 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 UPI 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<sub>2</sub> 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% <sup>235</sup>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 <sup>236</sup>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  $^{145}\text{Nd}/^{238}\text{U} + ^{146}\text{Nd}/^{238}\text{U}$  sum for PWR fuel cut analyses and the  $^{235}\text{U}/^{238}\text{U}$  residual enrichment for the assembly dissolutions (the neodymium chemical analyses are not available).

In the following tables, the total uncertainties ( $1\sigma$ ) correspond to the combination of uncertainties on chemical assays and determination of the burnup of the assembly derived from Nd (or  $^{235}\text{U}$ ) 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)↓	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	$^{236}\text{U}/^{238}\text{U}$
UOx "BUGEY FESSENHEIM" 3.1 % $^{235}\text{U}$	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 % $^{235}\text{U}$	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 % $^{235}\text{U}$	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 % $^{235}\text{U}$	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 % $^{235}\text{U}$	35	-16.5 ± 2.2	-0.05 ± 2.8	-4.7 ± 1.0
URE "CRUAS" 3.5 % $^{235}\text{U}$	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  $^{235}\text{U}$  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  $^{235}\text{U}/^{238}\text{U}$  residual enrichment.

The abundance of  $^{236}\text{U}$  is underestimated in standard UOx fuel and in MOx calculations, principally due to the underestimation of the  $^{235}\text{U}$  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  $^{236}\text{U}$  build-up will be very well predicted : (C-E)/E = -1%  $\pm$  1.0% at 60 GWd/t for UOx fuels and a mean value of -3%  $\pm$  2.0% in Mox fuels.

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

The abundance of  $^{234}\text{U}$  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  $^{241}\text{Pu}$ . In the back-end cycle, the long-term toxicity is mainly linked to  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{242}\text{Pu}$ . Furthermore,  $^{242}\text{Pu}$  is the father of  $^{244}\text{Cm}$  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  $^{239}\text{Pu}$ , highly involved in criticality calculations.

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

Fuel↓	BU(GWd/t)↓	$^{236}\text{Pu}/^{238}\text{U}$	$^{239}\text{Pu}/^{238}\text{U}$	$^{240}\text{Pu}/^{238}\text{U}$	$^{241}\text{Pu}/^{238}\text{U}$	$^{242}\text{Pu}/^{238}\text{U}$
UOx "BUGEY FESSENHEIM" 3.1 % $^{235}\text{U}$	20	-12.3 $\pm$ 3.2	-0.6 $\pm$ 1.1	-0.9 $\pm$ 1.6	-4.0 $\pm$ 2.1	-6.6 $\pm$ 3.9
	25	-9.0 $\pm$ 4.6	-0.1 $\pm$ 1.5	-0.7 $\pm$ 2.1	-3.2 $\pm$ 2.9	-5.4 $\pm$ 5.0
	40	-6.4 $\pm$ 2.0	2.5 $\pm$ 0.9	-1.9 $\pm$ 0.8	-1.0 $\pm$ 1.2	-7.6 $\pm$ 2.2
	50	-4.2 $\pm$ 1.6	2.0 $\pm$ 1.1	-1.2 $\pm$ 0.6	-0.4 $\pm$ 1.2	-6.1 $\pm$ 1.8
	60	-8.7 $\pm$ 2.5	3.1 $\pm$ 2.4	-0.7 $\pm$ 0.9	-0.3 $\pm$ 2.6	-8.7 $\pm$ 2.9
UOx "GRAVELINES" 4.5 % $^{235}\text{U}$	30	-12.2 $\pm$ 5.3	-1.6 $\pm$ 0.6	-2.2 $\pm$ 2.3	-5.0 $\pm$ 2.7	-6.8 $\pm$ 5.5
	40	-10.0 $\pm$ 5.0	-0.4 $\pm$ 0.2	-1.9 $\pm$ 2.0	-4.2 $\pm$ 2.0	-6.9 $\pm$ 4.8
	50	-10.3 $\pm$ 4.5	+1.6 $\pm$ 0.1	-1.8 $\pm$ 1.5	-3.0 $\pm$ 1.2	-8.6 $\pm$ 4.3
	60	-9.9 $\pm$ 4.3	+1.9 $\pm$ 0.2	-1.5 $\pm$ 1.2	-2.6 $\pm$ 1.0	-7.2 $\pm$ 3.9
UOx "La HAGUE" 3.1 % $^{235}\text{U}$	30	-14.9 $\pm$ 5.0	+0.9 $\pm$ 0.2	-1.7 $\pm$ 2.0	-2.5 $\pm$ 2.0	-11.7 $\pm$ 4.8
	40	-15.4 $\pm$ 4.5	+0.7 $\pm$ 0.1	-1.0 $\pm$ 1.5	-6.7 $\pm$ 1.2	-11.9 $\pm$ 4.3
UOx "La HAGUE" 3.25 % $^{235}\text{U}$	35	-14.1 $\pm$ 5.0	1.1 $\pm$ 0.2	-1.7 $\pm$ 2.0	-1.9 $\pm$ 2.0	-11.1 $\pm$ 4.8
	45	-11.7 $\pm$ 4.5	2.1 $\pm$ 0.1	-0.5 $\pm$ 1.5	-0.5 $\pm$ 1.2	-9.5 $\pm$ 4.3
UOx "La HAGUE" 3.45 % $^{235}\text{U}$	35	-18.4 $\pm$ 5.0	-0.1 $\pm$ 0.2	-2.5 $\pm$ 2.0	-2.6 $\pm$ 2.0	-11.5 $\pm$ 4.8
URE "CRUAS" 3.5 % $^{235}\text{U}$	15	-2.8 $\pm$ 3.9	1.7 $\pm$ 1.2	-2.2 $\pm$ 2.6	0.5 $\pm$ 3.6	-5.1 $\pm$ 5.8
	25	-2.8 $\pm$ 3.4	1.0 $\pm$ 1.0	-0.6 $\pm$ 2.2	-1.6 $\pm$ 2.7	-4.9 $\pm$ 5.1
	35	-1.7 $\pm$ 3.3	1.9 $\pm$ 1.0	-0.5 $\pm$ 1.7	-0.6 $\pm$ 1.8	-4.2 $\pm$ 4.3
MOx SLB1	10	-6.3 $\pm$ 0.6	0.05 $\pm$ 0.7	0.6 $\pm$ 0.2	-5.2 $\pm$ 0.5	-1.4 $\pm$ 0.5
	30	-6.3 $\pm$ 0.5	-0.2 $\pm$ 1.1	0.9 $\pm$ 0.2	-3.0 $\pm$ 0.5	-2.7 $\pm$ 0.8
	42	-6.3 $\pm$ 0.3	-1.6 $\pm$ 1.4	0.7 $\pm$ 0.5	-3.5 $\pm$ 0.7	-3.9 $\pm$ 0.8

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

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

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

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

The abundance of  $^{238}\text{Pu}$  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  $^{238}\text{Pu}$  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  $^{238}\text{Pu}$  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  $^{238}\text{Pu}$  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  $^{238}\text{Pu}$  in UOx and MOx fuels (at discharging)

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

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

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

In MOx fuels, table III shows that plutonium 238 is mainly formed by the decaying of  $^{242}\text{Cm}$ , coming from the  $^{241}\text{Am}(n,\gamma)$  reaction. The underestimation of  $^{238}\text{Pu}$  build-up is probably linked to an underestimation of  $^{241}\text{Am}$  capture cross-section. Using the PERTURBATION module of the DARWIN package, we can conclude that an increasing of 20% of  $^{241}\text{Am}$  cross-section will reduce the current -6% (C-E)/E discrepancy to -1%.

However this correction on  $^{241}\text{Am}$  cross-section must be confirmed by rigorous analyses from nuclear physicists.  $^{238}\text{Pu}$  build-up is also linked to the knowledge of  $^{242}\text{Cm}$  decay constant ; however this value is well known and its associated uncertainty can't explain the underestimation on  $^{238}\text{Pu}$  inventory :  $\lambda = 162.94 \text{ days} \pm 1.44 \text{ hours}$ .

$^{238}\text{Pu}$  disappeared by neutron capture ; an overestimation of  $^{238}\text{Pu}(n,\gamma)$  cross-section could explain the inventory underestimation and this cross-section must be investigated.

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

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

In order to evaluate the sensitivity of  $^{238}\text{Pu}$  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  $^{238}\text{Pu}$  build-up

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

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

In URE fuels, the  $^{238}\text{Pu}$  inventory is well calculated. It comes mainly from the depletion chain  $^{236}\text{U}(n,\gamma) \rightarrow ^{237}\text{U}(\beta^-) \rightarrow ^{237}\text{Np}(n,\gamma) \rightarrow ^{238}\text{Np}(\beta^-) \rightarrow ^{238}\text{Pu}$  as shown in Table III for URE fuels. This indicates that  $^{236}\text{U}(n,\gamma)$  and  $^{237}\text{Np}(n,\gamma)$  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)↓	$^{237}\text{Np}/^{238}\text{U}$
UOx "BUGEY FESSENHEIM" 3.1 % $^{235}\text{U}$	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 % $^{235}\text{U}$	30	-0.3 ± 4.7
	40	-2.4 ± 4.2
	50	-4.4 ± 3.4
	60	-3.0 ± 3.1
URE "CRUAS" 3.5 % $^{235}\text{U}$	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  $^{237}\text{Np}$  is underestimated. This underestimation could be due to the formation of  $^{237}\text{Np}$  ; the Table VI below gives the formation ways for the different fuels studied.

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

Taux de combustion (GWj/t) →	20	30	40	50	60
Formation ways in standard <b>UOx</b> ↓					
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	<b>20.2</b>	16.0	13.8	12.7	11.6

Taux de combustion (GWj/t) →	20	30	40	50
Formation ways in <b>URE</b> ↓				
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) →	10	30	45
Formation ways in <b>MOx</b> ↓			
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	<b>85.6</b>	<b>78.1</b>	<b>76.5</b>
Am241(α) → Np237	0.7	1.0	1.2

In MOx fuels  $^{237}\text{Np}$  is mainly produced (80% at 45 GWd/t) by  $^{238}\text{U}(n,2n)$  reaction as shown in Table VI suggesting that the (n,2n)  $^{238}\text{U}$  cross-section is underestimated in JEF2. This is confirmed by comparison between different evaluations [10].

In UOx fuels the underestimation of  $^{237}\text{Np}$  build-up is due on one hand to the underestimation of the  $^{235}\text{U}$  capture Resonance Integral and in the other hand to the underestimation of (n,2n) $^{238}\text{U}$  in JEF2, contributing to 20% in the  $^{237}\text{Np}$  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 their 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
$^{238}\text{Pu}$	-310 pcm	-760 pcm
$^{240}\text{Pu}$	-8300 pcm	-17500 pcm
$^{242}\text{Pu}$	-710 pcm	-1500 pcm
$^{241}\text{Am}$	<b>-1290 pcm</b>	<b>-4600 pcm</b>
$^{243}\text{Am}$	<b>-280 pcm</b>	<b>-1000 pcm</b>

The  $^{242m}\text{Am}$  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-UO<sub>2</sub> fuel ( $\rho = +27$  pcm at 40 GWd/t /10/); but in MOX spent fuel this fissile isotope,  $\sigma_f^{2200} = 6886$  barns in JEF2 library, must be accounted for:  $\rho_{\text{Am242m}} = +500$  pcm in the 20-50 GWd/t burnup range.

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

Fuel↓	BU (GWd/t)↓	$^{241}\text{Am}/^{238}\text{U}$	$^{242m}\text{Am}/^{238}\text{U}$	$^{243}\text{Am}/^{238}\text{U}$
UOx "BUGEY FESSENHEIM" 3.1 % $^{235}\text{U}$	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 % $^{235}\text{U}$	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 % $^{235}\text{U}$	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  $^{241}\text{Am}$ , formed by decaying of  $^{241}\text{Pu}$ , is reasonably estimated.

$^{242m}\text{Am}$  is underestimated by about -20%. This suggests increasing  $^{241}\text{Am}$  capture cross-section. We must also improve the knowledge of the branching ratio of  $^{241}\text{Am}$  to  $^{242m}\text{Am}$  in the epithermal range.

The underestimation in  $^{242}\text{Pu}$  generates the underestimation for  $^{243}\text{Am}$ . The increase of the  $^{241}\text{Pu}$  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 ( $\sigma_0^{2200} = 500$  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)↓	<sup>243</sup> Cm/ <sup>238</sup> U	<sup>244</sup> Cm/ <sup>238</sup> U	<sup>245</sup> Cm/ <sup>238</sup> U	<sup>246</sup> Cm/ <sup>238</sup> U	<sup>247</sup> Cm/ <sup>238</sup> U
UOX "BUGEY" FESSENHEIM" 3.1 % <sup>235</sup> 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 % <sup>235</sup> 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 % <sup>235</sup> 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 <sup>242</sup>Pu generates an underestimation for <sup>243</sup>Am, and consequently for <sup>244</sup>Cm and <sup>245</sup>Cm. Recent studies on <sup>241</sup>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 <sup>243</sup>Cm build-up needs also to be corrected. Studies on <sup>242</sup>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)↓	<sup>147</sup> Sm/ <sup>238</sup> U	<sup>149</sup> Sm/ <sup>238</sup> U	<sup>150</sup> Sm/ <sup>238</sup> U	<sup>151</sup> Sm/ <sup>238</sup> U	<sup>152</sup> Sm/ <sup>238</sup> U
UOX "BUGEY" 3.1 % <sup>235</sup> 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 % <sup>235</sup> 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 <sup>151</sup>Sm build-up, the samarium isotope calculations are satisfactory. However we can make some remarks on nuclear data.

The <sup>147</sup>Sm is underestimated. This isotope is formed by decaying of <sup>147</sup>Pm, itself formed by cumulative fissions, and disappeared by neutron capture.

The interpretation of <sup>147</sup>Sm samples oscillations in the MINERVE [17] reactor shows that the <sup>147</sup>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 <sup>147</sup>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 <sup>147</sup>Sm is due to its formation. The capture cross-section of <sup>147</sup>Pm, and <sup>147</sup>Nd cumulative fission yields must be investigated.

The  $^{149}\text{Sm}$  seems to be underestimated but the chemical uncertainty is very high and the  $^{149}\text{Sm}$  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  $^{150}\text{Sm}$  is underestimated, varying in the direct ratio of its father the  $^{149}\text{Sm}$ . The  $^{151}\text{Sm}$  is also overestimated. This is probably due to an overestimation of  $^{151}\text{Pm}$  cumulative fission yields.

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

#### 5.4.2 THE NEODYMIUM ISOTOPES

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

The Nd isotopes are very well calculated.

The  $^{143}\text{Nd}$  build-up is slightly overestimated for high burnups : +2%. At these burnups,  $^{143}\text{Nd}$  concentration is sensitive to its capture cross-section. This result confirms an under-estimation of the thermal capture cross-section of  $^{143}\text{Nd}$ . The same conclusion is drawn from the interpretation of  $^{143}\text{Nd}$  samples oscillated in Minerve lattices. A new evaluation of  $^{143}\text{Nd}$  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  $^{144}\text{Nd}$  concentration due probably to an underestimation of  $^{144}\text{Ce}$  cumulative fission yields.

#### 5.4.3 THE CESIUM ISOTOPES

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

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

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

Fuel ↓	BU (Gwd/t) ↓	<sup>133</sup> Cs/ <sup>238</sup> U	<sup>134</sup> Cs/ <sup>238</sup> U	<sup>135</sup> Cs/ <sup>238</sup> U	<sup>137</sup> Cs/ <sup>238</sup> U
UOX "BUGEY" FESSENHEIM" 3.1 % <sup>235</sup> 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 % <sup>235</sup> 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 <sup>133</sup>Cs in UOx fuels. This underestimation is reduced to 2% in MOx fuels. A study has shown that the <sup>133</sup>Cs inventory is not very sensitive to the <sup>133</sup>Cs capture cross-section, so we can conclude that this underestimated abundance is due to an underestimation of <sup>135</sup>Xe fission yields. The steady under-prediction in UOx fuels indicates that the increasing of the fission yields for <sup>235</sup>U is necessary but that the fission yields for <sup>239</sup>Pu and/or <sup>241</sup>Pu must be increased too. This last point is confirmed by the MOx results.

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

The <sup>135</sup>Cs isotope is well calculated when the irradiation history is accurately known (Gravelines and Saint Laurent B1).

The <sup>134</sup>Cs build-up is underestimated. This underestimation could be due to an underestimation of <sup>133</sup>Cs capture cross-section, or to an overestimation of <sup>134</sup>Cs capture cross-section itself or to an underestimation of the <sup>134</sup>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) ↓	<sup>109</sup> Ag/ <sup>238</sup> U	<sup>99</sup> Tc/ <sup>238</sup> U	<sup>95</sup> Mo/ <sup>238</sup> U	<sup>101</sup> Ru/ <sup>238</sup> U	<sup>103</sup> Rh/ <sup>238</sup> U
UOX "BUGEY" 3.1 % <sup>235</sup> 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 % <sup>235</sup> 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 <sup>95</sup>Mo, <sup>99</sup>Tc, <sup>101</sup>Ru and <sup>103</sup>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

<sup>153</sup>Eu isotope and <sup>155</sup>Gd isotope are important poisoning FPs in Burn up credit applications. In order to check <sup>155</sup>Gd build-up, the <sup>154</sup>Eu, <sup>155</sup>Eu and <sup>154</sup>Gd were also investigated. Furthermore the <sup>154</sup>Eu/<sup>137</sup>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)↓	<sup>153</sup> Eu/ <sup>238</sup> U	<sup>154</sup> Eu/ <sup>238</sup> U	<sup>155</sup> Eu/ <sup>238</sup> U	<sup>154</sup> Gd/ <sup>238</sup> U	<sup>155</sup> Gd/ <sup>238</sup> U	<sup>156</sup> Gd/ <sup>238</sup> U
UOX "BUGEY" 3.1 % <sup>235</sup> 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 % <sup>235</sup> 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	

<sup>153</sup>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 <sup>153</sup>Eu samples oscillations in the MINERVE reactor [17] shows that the <sup>153</sup>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 <sup>153</sup>Eu build-up ; so we can conclude that the overestimation of <sup>153</sup>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 <sup>153</sup>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 <sup>153</sup>Eu build-up of fission yields modification (corresponding in fact to the uncertainties).

Tableau XV: Modification of <sup>153</sup>Eu fission yields: impact on <sup>153</sup>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 <sup>153</sup> 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 <sup>153</sup>Eu concentrations. The fission yields for <sup>235</sup>U seem to be well known, on the other hand for <sup>239</sup>Pu and <sup>241</sup>Pu the uncertainties are huge.

One can notice that the ENDFB6 fission yield value for <sup>239</sup>Pu is 0.364% [18] that is to say 8% lower than the JEF2 value : the jef2 value must be decrease for <sup>239</sup>Pu as well as for <sup>241</sup>Pu.

<sup>155</sup>Gd is strongly over-predicted but agrees with its father, <sup>155</sup>Eu, which is produced by capture on <sup>154</sup>Eu and directly by fission. Furthermore the <sup>156</sup>Gd, daughter by capture of <sup>155</sup>Eu is under-predicted.

<sup>154</sup>Eu is also strongly overestimated and the same level of overestimation is found on its daughter <sup>154</sup>Gd. These remarks point out that modification of <sup>154</sup>Eu and <sup>155</sup>Eu capture cross-section is needed in JEF2 file.

Recent studies on  $^{154}\text{Eu}$  and  $^{155}\text{Eu}$  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  $\sigma_c$  from ENDFB6.7 for  $^{154}\text{Eu}$  and  $^{155}\text{Eu}$  in GRAVELINES UOx fuels

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

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

BU (MWd/t)	13200	29200	43000	46000
$^{155}\text{Gd}/^{238}\text{U}$ ; $^{154}\text{Eu}$ and $^{155}\text{Eu}$ <u>JEF2.2</u>	68.8 ± 3.7	53.9 ± 5.4	9.7 ± 6.4	19.4 ± 6.6
$^{155}\text{Gd}/^{238}\text{U}$ ; $^{154}\text{Eu}$ and $^{155}\text{Eu}$ <u>ENDFB6.7</u>	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  $^{155}\text{Sm}$  cumulative fission yields (leading to the overestimation of  $^{155}\text{Eu}$ ) and/or to an underestimation of the capture cross-section of the  $^{155}\text{Gd}$  itself.

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

The  $^{156}\text{Gd}$  calculation is also well improved by the  $^{155}\text{Eu}$  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%  $^{235}\text{U}$  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 the 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  $^{241}\text{Pu}$  and overestimation of europium and gadolinium isotopes. However, the introduction in the future JEFF3.0 file of new evaluations, such as  $^{235}\text{U}$ ,  $^{241}\text{Pu}$ ,  $^{153-154}\text{Eu}$  and  $^{155}\text{Eu}$  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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## The French Post Irradiation Examination Database for the validation of depletion calculation tools

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This paper presents the experimental programmes conducted in France by the Commissariat à l'Energie Atomique (CEA) in order to validate spent fuel inventory calculations for core studies as well as fuel cycle studies. This large experimental programme is obtained in collaboration with our French partners, Electricité de France (EDF), FRAMATOME-ANP and COGEMA. The experimental data are based on chemical analysis measurements from fuel rod cuts irradiated in French reactors for PWR-UOx and MOx fuels, then dissolved in CEA laboratories, and from full assembly dissolutions at the COGEMA/La Hague reprocessing plants for UOx fuels. This enables us to cover a large range of UOx fuels with various enrichments in <sup>235</sup>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 an initial Pu amount in the central zone of 5.6 % and a maximum burnup of 45 GWd/t. Uranium, Plutonium, Americium, Curium isotopes and some fission products were analysed. Furthermore, Fission Products involved in Burn up Credit studies were measured.

The experimental database contains also data for Boiling Water Reactor (BWR) with irradiated samples of BWR 9X9 and full BWR assemblies dissolutions. Furthermore some data exist for Fast Breeder Reactor (FBR) with small samples irradiated in the PHENIX reactor.

An overview of ongoing programmes is also presented.

**Keywords :** *Experimental database, PIE, spent fuel, UOx, MOx, PWR, BWR, FBR*

### 1. Introduction

In order to validate French neutronic codes 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 as well as some caesium and neodymium isotopes have been analysed in PWR and BWR samples. Furthermore the 15 fission products used in Burnup Credit (BUC) criticality calculations have been measured for this two types of reactor. The available experimental information consists of chemical assays from fuel rod cuts irradiated in reactors and of solution samples derived from full assembly dissolutions at the COGEMA/La Hague reprocessing plants.

For Fast Breeder Reactor the database contains also the results of measurements of actinide samples transmutation made in the PROFIL and TRAPU irradiation programme in the PHENIX reactor.

This paper describes this large experimental database, some comments on chemical process and an overview of ongoing programmes.

### 2. The experimental database of irradiated PWR fuels

We describe here the main experimental programmes carried out on irradiated UOX and MOx PWR fuels and used currently for code system validation.

The experimental information can be classified into two groups :

- **small fuel pin samples**, irradiated in French reactors, with positions in the assembly well characterized. The analysed samples are obtained from cuts of extracted pins. In order to investigate the local irradiation effect, various axial locations of cuts are selected with always a sample cut in the middle of the pin (height = 1900 mm). These time-consuming and expensive experiments provide very accurate results for a limited number of samples.
- **dissolution aliquots of entire assembly** sets obtained from reprocessing plant (COGEMA/La Hague). A lot of these measurements have been performed. However the irradiation histories of the assemblies are not very well known and the fact that dissolutions are performed over batches of several assemblies limit the usefulness of these data for code validation. These type of data is nevertheless used to extend the validation range in a statistical sense.

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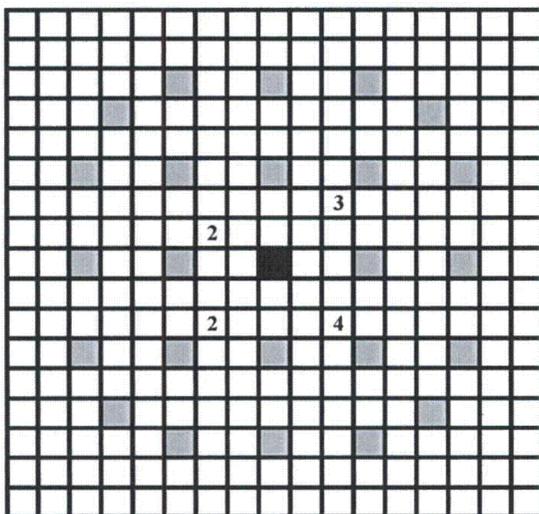
### 2.1 The UOx fuels experimental database

Four main programmes related to UOx fuel samples are used for the experimental validation of actinides and fission products inventory. They are named : BUGEY3, FESSENHEIM II, GRAVELINES, and CRUAS.

The experimental programme derived from the **BUGEY3** reactor was the first representative of the French nuclear power reactors. It involves standard PWR fuel, the assembly consists of 17X17 pins clad in zircaloy 4. The programme covers two assemblies with removable pins. One assembly, with 2.1% initial enrichment was irradiated for the start cycle, the other with 3.1% enrichment was irradiated for 3 cycles. The maximum burnup in this programme is 40 GWd/t.

The **FESSENHEIM II** programme (4 and 5 cycle irradiations) allows the study of UOx fuel with higher burnup than in BUGEY3. Using a UO2 fuel with an enrichment of 3.1%  $^{235}\text{U}$ , samples with burnup in the 45 GWd/t to 60 GWd/t range were analysed. Among the extracted fuel pins, the following ones have been selected : four pins from the FEC57 assembly after four cycles of irradiation and one pin from the FEC52 assembly after 5 cycles .

The programme performs from the **GRAVELINES** reactor is devoted to the validation of the calculation schemes for high burnup fuel and high cycle length operation. The initial UO2 fuel enrichment is 4.5%.



**Figure 1** : Extracted rods in the FF06E2BV assembly after 2, 3 and 4 irradiation cycles

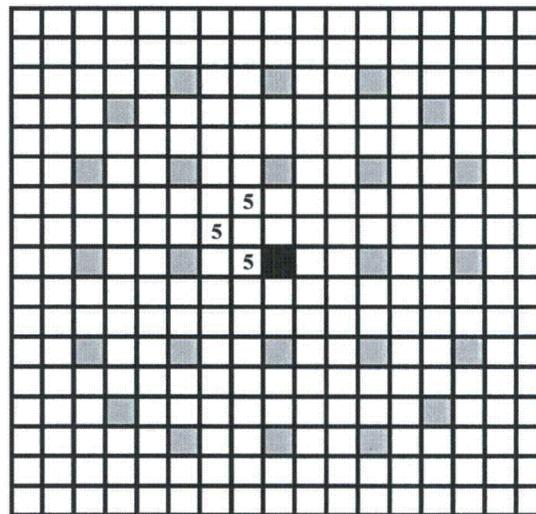
From **full assembly dissolutions** at COGEMA/La Hague reprocessing plants, we get uranium and plutonium chemical analyses. The involved assemblies are 900 MWe 17X17 PWR with 3.1%, 3.25% or 3.45% enrichments and with burnups between 25 to 45 GWd/t. One can notice that the

This experimental programme on a 900 MWe PWR is the most important being carried out in France up to now.

Two assemblies named FF06E2BV and FF06E3BV containing each 20 removable fuel pins were irradiated for 4 cycles in the Gravelines 3 reactor then in the Gravelines 2 reactor for the fifth cycle. On these twenty pins, for the FF06E2BV assembly, 2 pins were extracted at the end of the second cycle of the reactor, one pin at the end of the 3rd cycle and two pins at the end at the fourth cycle but only one was analysed. For the FF06E3BV, two fuel rods have been extracted after fourth cycles, one was analysed, then 4 pins were extracted after the fifth cycle, three were analysed.

The Figure 1 and 2 above show that we have prioritised the choice of pins located at the corner of a guide tube (filled with water the most part of the irradiation), position more representative of an "asymptotic" pin ; however a fuel pin with 5 irradiation cycles was selected facing a guide tube.

The **CRUAS** programme is devoted to the validation of the URE (Reprocessed Uranium) fuel, using reprocessed then enriched uranium. It permits validation of the  $^{236}\text{U}$  capture cross-section. This programme is characterized by a 3.5%  $^{235}\text{U}$  enrichment with an initial amount of 1.2% of  $^{236}\text{U}$ .



**Figure 2** : Extracted rods in the FF06E3BV assembly after 5 irradiation cycles

dissolution process involves batches of several assemblies, therefore we must select sets with very close irradiation histories and burnups.

## 2.2 The MOx fuels experimental database

Currently, in France, twenty-four 900 MWe PWRs are devoted to the Pu recycling in 30% mixed core loading. This increase of French MOX fuel cycle emphasizes the need for enlarge the experimental database to plutonium-fueled assemblies.

The experimental PIE programme devoted to MOx fuels is described here after.

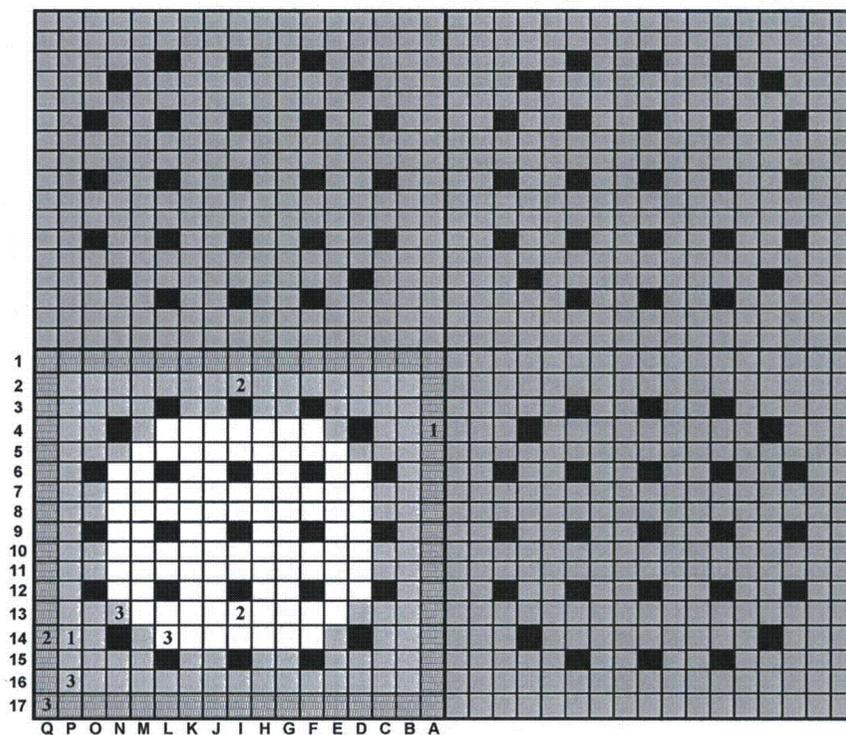
The first French reactor using MOx assemblies is the so-called **SAINT-LAURENT B1** reactor. The PIE analyses are carried out on fuel cuts coming from this reactor.

The standard MOx assemblies, used in Saint-Laurent B1 include three zones with different plutonium enrichments to flatten the within assembly power distribution and to attenuate fission rate discontinuities at the MOx-UOX interface. The central zone is

characterized by a high Pu content (5.6%) and the peripheral zone by a small Pu content (2.9%).

Two MOx fuel assemblies were selected for the SLB1 programme. The extracted and analysed MOx rods were irradiated for 1, 2 and 3 cycles, with burnups ranging from about 10 GWj/t to 45 GWj/t.

The figure 3 shows the position of the rods extracted from the MOx assembly. The associated number indicates the irradiation cycle length (the rods extracted after 1 cycle comes from another assembly but for simplification we put them on the same figure). On this figure, we represent the MOx surrounded by UOx assemblies, in order to point out that in the neutronic calculation this environment must be modeled to represent the thermal neutron current from the UOx toward the MOx assembly.



-  - MOX rods small Pu content
-  - MOX rods intermediate Pu content
-  - MOX rods high Pu content
-  - UO2 fuel rods (3.25w/o U-235/U Enrichment)

The extraction and analysis of MOx pins from the reactor **GRAVELINES 4** enables us to have a validation for the first fuel pins irradiated for 4 cycles. We dispose of analyses for two fuel rods extracted after 3 et 4 cycles in two different assemblies. The central zone is characterized by a high Pu content of 6.0% and the peripheral zone by a Pu content of 3.1%. The table 1 summarises the various programmes for PWR UOx and MOx fuel ; all these programmes are completed. The chemical analyses performed for each programme are also indicated. Uranium, plutonium, americium and curium nuclides are always analysed. Additionally for a few experiments (BUGEY, GRAVELINES and Saint Laurent B1) fission product chemical analyses elements (Sm, Rh, Nd, Cs, Mo, Eu, Gd, Ag, Ru), have been measured relatively to  $^{238}\text{U}$ , as well as their isotopic composition. These fission products are involved in French Burnup Credit studies.

In this table 1, the following abbreviations are used and the corresponding analysed isotopes are listed :

**MJA** = Major Actinides :  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$

**MA** = Minor Actinides :  $^{237}\text{Np}$ ,  $^{241}\text{Am}$ ,  $^{242\text{m}}\text{Am}$ ,  $^{243}\text{Am}$ ,  $^{243}\text{Cm}$ ,  $^{244}\text{Cm}$ ,  $^{245}\text{Cm}$ ,  $^{246}\text{Cm}$ ,  $^{247}\text{Cm}$

**BUC** = Burnup Credit :  $^{147}\text{Sm}$ ,  $^{149}\text{Sm}$ ,  $^{150}\text{Sm}$ ,  $^{151}\text{Sm}$ ,  $^{152}\text{Sm}$ ,  $^{95}\text{Mo}$ ,  $^{99}\text{Tc}$ ,  $^{101}\text{Ru}$ ,  $^{103}\text{Rh}$ ,  $^{153}\text{Eu}$ ,  $^{109}\text{Ag}$ ,  $^{155}\text{Gd}$  (plus  $^{155}\text{Eu}$ ,  $^{154}\text{Eu}$  and  $^{154}\text{Gd}$  to check  $^{155}\text{Gd}$  build up)

**Nd** : neodymium isotopes :  $^{143}\text{Nd}$ ,  $^{144}\text{Nd}$ ,  $^{145}\text{Nd}$ ,  $^{146}\text{Nd}$ ,  $^{148}\text{Nd}$ ,  $^{150}\text{Nd}$

**Cs** : caesium isotopes :  $^{133}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{135}\text{Cs}$ ,  $^{137}\text{Cs}$

**$\gamma$  spectrometry**:  $^{154}\text{Eu}/^{137}\text{Cs}$ ,  $^{106}\text{Ru}/^{137}\text{Cs}$ ,  $^{134}\text{Cs}/^{137}\text{Cs}$ ,  $^{144}\text{Ce}/^{137}\text{Cs}$

**Table 1** : The experimental database for PWR-UOx and MOx fuels

PROGRAMME name	Fuel	Enrichment	Burnup range	Experimental data
<b>Bugey 3</b> 1 to 3 cycles	REP 17x17 UOx	2.10% 3.10%	19 to 38 GWd/t	MJA, MA, Nd, Cs, BUC
<b>Fessenheim 2</b> 2 cycles	REP 17x17 UOx	2.60%	27 to 30 GWd/t	MJA, MA, Nd
<b>Fessenheim 2</b> 4 & 5 cycles	REP 17x17 UOx	3.10%	45 to 60 GWd/t	MJA, MA, Nd, Cs,
<b>Gravelines 3+2</b> 2 to 5 cycles	REP 17x17 UOx	4.50%	25 to 62 GWd/t	MJA, MA, Nd, Cs, BUC, $\gamma$ spectro
<b>Cruas 4 / URE</b> 1 to 3 cycles	REP 17x17 UOx coming from URT	3.61% U6 /U8 = 1.2%	11 to 34 GWd/t	MJA, MA, Nd
<b>Saint Laurent B1</b> 1 to 3 cycles 3 zones	REP 17x17 MOx 3 Pu content	U app. 0.22% $(\frac{\text{Pu}}{\text{U} + \text{Pu}})_{\text{moy}} \cong 4.5\%$	10 to 45 GWd/t	MJA, MA, Nd, Cs BUC
<b>Gravelines 4</b> 3 & 4 cycles central zone & intermediate zone	REP 17x17 MOx	U app. 0.22% $(\frac{\text{Pu}}{\text{U} + \text{Pu}})_{\text{moy}} \cong 4.5\%$	40 to 50 GWd/t	MJA, Nd
<b>La Hague</b> Fuel dissolution	REP 17X17 UOx	3.1%; 3.25%, 3.45%	25 to 45 GWd/t	MJA

### 3. The experimental database of irradiated BWR fuels

The experimental database contains chemical analysis results coming from samples irradiated in reactor and from dissolution of various assemblies.

In the **full assembly dissolutions** at COGEMA/La Hague reprocessing plants, the involved assemblies are 7X7, 8X8 and 9X9 BWR with various enrichments and burnups ranging between 20 to 40 GWd/t. The

amounts of depleted U and Pu are only available.

An important experimental programme was launched concerning chemical analyses of **samples of fuel rods** irradiated in the GUNDREMMINGEN reactor. The Table 2 summarises the chemical analyses performed in this programme.

**Table 2 : Summary of analyses for the BWR GUNDREMMINGEN programme**

Fuel rod	Sample position /bottom of fuel (mm)	Estimated Burnup (GWd/t)	Estimated Void fraction (%)	Analyses							$\gamma$ Spectro and neutron emission	Other analyses
				U	Pu	Nd	Am	Cm	Np	<sup>238</sup> Uabs		
A	200	26.5	1.5	X	X	X			X	X	X	-
	600	45.4	15.9	X	X	X			X	X	X	-
	845	47.2	24.2	X	X	X			X	X	X	-
B	200	26.3	1.5	X	X	X	X	X	X	X	X	-
	600	45.0	15.9	X	X	X	X	X	X	X	X	-
	685	46.0	18.7			Z				X	X	Cs
	815	46.9	23.2	X	X	X	X	X	X	X	X	BUC
B	1170	46.3	34.5	X	X	X	X	X	X	X	X	-
	1440	46.3	42.0	X	X	X	X	X	X	X	X	-
	1680	45.0	47.2			X					X	Cs
	1815	45.0	50.1	X	X	X	X	X	X	X	X	BUC
	2300	43.8	57.9	X	X	X	X	X	X	X	X	-
B	2780	41.9	63.3	X	X	X	X	X	X	X	X	-
	3290	35.4	67.3			X					X	
	3420	32.0	68.2	X	X	X	X	X	X	X	X	BUC

\*BUC = Burnup Credit isotopes (see page 4)

### 4 The experimental database for Fast Breeder Reactors

Two types of irradiation experiment have been carried out in the French PHENIX reactor : fuel rod irradiations (the TRAPU experiment) and irradiations of pure isotopic samples (the PROFIL 1 and 2 experiments). After irradiations, chemical analyses are carried out for cuts of fuel rods and for the irradiations samples. The irradiation of pure samples is more devoted to giving information concerning capture and (n,2n) cross-sections.

The TRAPU experiment was a six cycle irradiation of mixed oxide fuel pins containing plutonium of three different isotopic compositions. These compositions are detailed in the Table 3. TRAPU1 and TRAPU2 are quite similar in isotopic composition, TRAPU3 contains a higher proportion of <sup>240</sup>Pu. The chemical analyses focused on Uranium and Plutonium isotopes.

**Table 3 : Pu isotopic composition of the TRAPU pins**

	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu
TRAPU-1	0.1	73.3	21.9	4.0	0.7
TRAPU-2	0.8	71.4	18.5	7.4	1.9
TRAPU-3	0.2	34.0	49.4	10.0	6.4

In the PROFIL-1 experiment, the irradiated isotopes were : <sup>235,238</sup>U, <sup>238,239,240, 241,242</sup>Pu, <sup>241</sup>Am. In PROFIL-2 they were : <sup>232</sup>Th, <sup>233,234,235,238</sup>U, <sup>237</sup>Np, <sup>238,239,240, 241,242</sup>Pu, <sup>241,243</sup>Am, <sup>244</sup>Cm.

### 5 Some comments on chemical analyses

After cutting in the CEA/CADARACHE/LECA laboratory, the samples of irradiated fuel rods are dissolved in nitric acid by the CEA in the COMIR or ATLANTE Laboratories. The dissolved solution are sent in CEA/SACLAY in order to be analysed. Uranium and plutonium are first separated with an ion exchange resin and are then ready for isotopic

characterization by mass spectrometry. The remaining fraction of solution is used in order to separate by liquid chromatography the minor actinides Americium and Curium (interaction between masses 242 and 243), the Neodymium, Cerium, Promethium and Samarium (interaction between masses 142, 144, 147, 148 and 150) and Caesium and Barium (interaction between masses 134, 135 and 137).

The isotopic content of the various fractions and their amounts are then measured by isotopic dilution mass-spectrometry using a thermo-ionization-type mass spectrometer (TIMS). For Am, Cm and Cs isotopes a

specific method of 'total consumption' in TIMS has been developed and give very accurate results. The isotopic dilution uses double or triple isotopic tracers such as  $^{242}\text{Pu}/^{233}\text{U}/^{145}\text{Nd}$ ,  $^{133}\text{Cs}/^{233}\text{U}$ ,  $^{243}\text{Am}/^{233}\text{U}$ ,  $^{248}\text{Cm}/^{233}\text{U}$  (used for the determination of Cm/U ratios). The  $^{237}\text{Np}/^{238}\text{U}$  is measured by ICPMS (Inductively Coupled Plasma Mass Spectrometry) and the  $^{242}\text{Cm}$  by alpha spectrometry.

Some percentage uncertainties of the analytical value, depending on the nuclides, are given in maximum values in the Table 4.

**Table 4 : Uncertainties of analytical values for some actinides and FPs**

	$^{234}\text{U}$ , $^{236}\text{U}$	$^{235}\text{U}$	$^{238}\text{Pu}$	$^{239,240,241,242}\text{Pu}$	Np	Am	Cm(243 to 246)	Nd	Cs
Uncert.	≈ 1%	< 0.2%	< 1%	< 0.4%	< 4%	< 3%	< 2%	≈ 0.5%	< 1%

## 6 Ongoing programmes

Four main programmes are currently in progress.

For metallic BUC fission products such as  $^{95}\text{Mo}$ ,  $^{99}\text{Tc}$ ,  $^{101}\text{Ru}$ ,  $^{103}\text{Rh}$  and  $^{109}\text{Ag}$  the chemical dissolution process in the COMIR laboratory is not yet optimised and our first calculation-experiment comparisons have shown inconsistency due to the experimental results. In fact, for these metallic fission products, the dissolution is not total and non-soluble deposits yield to inaccurate analyses. New results, should be obtained at the end of 2003 with a new process of dissolution, performed in ATALANTE laboratory, for both MOx and UOx fuels.

The P.I.E. database is currently being extended to higher burnups for PWR-UOX and MOx fuels. Fuel rod cuts extracted from French reactors after 5, 6 and 7 irradiation cycles, up to 80 GWd/t for UOx fuels and after 4 and 5 cycle, up to 60 GWd/t, for MOx fuels. This experimental programme, called HTC (for Hauts Taux de Combustion, Higher Burnups), will be completed in 2005. In this programme, new experimental results such as  $^{232}\text{U}$ ,  $^{236}\text{Pu}$  and  $^{242}\text{Cm}$  will be obtained.

Another programme, entitled PRECCI, deals with the long term evolution of spent nuclear fuel in all types of scenarios for the back-end of fuel cycle. It is therefore necessary to extend the experimental database to the most significant isotopes involved in long term studies. The selected isotopes, long life fission products and activation products, are the following :  $^{129}\text{I}$ ,  $^{126}\text{Sn}$ ,  $^{90}\text{Sr}$ ,  $^{93}\text{Zr}$ ,  $^{107}\text{Pd}$ ,  $^{79}\text{Se}$ ,  $^{14}\text{C}$ ,  $^{36}\text{Cl}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ ,  $^{93}\text{Mo}$ ,  $^{94}\text{Nb}$ . This programme implies also the research and development of accurate chemical analysis methods. Some of them are completed and first experimental results will be available in 2004.

The CEA is also involved in an international programme, MALIBU (radiochemical analysis of Mox And Uox Lwr fuels Irradiated to high Burn up), conducted par BELGONUCLEAIRE. The aim is to perform radiochemical measurements on fuel samples which have reached about 75 GWd/t. The fuel concerns is coming from a PWR 15X15 reactor. UOx and MOx fuel samples will be analysed in different

chemical laboratories. The CEA will participate in chemical analyses.

## 6 Conclusion

The French PIE experimental database presented in this paper offers valuable data with very low experimental uncertainties. These data are currently used in the validation of nuclear data and French depletion calculation tools.

In order to take up new nuclear challenges the experimental database is always in progress.

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Enclosure 29 to TN E-29128

NRC Safety Evaluation Report, "Safety Evaluation of Topical Report BAW-10228P, *SCIENCE*," USNRC, October 26, 1999, TAC NO. MA4599, associated with RAI P6-7



UNITED STATES  
NUCLEAR REGULATORY COMMISSION

WASHINGTON, D.C. 20555-0001

October 26, 1999

Mr. T. A. Coleman, Vice President  
Government Relations  
Framatome Cogema Fuels, Inc.  
3315 Old Forest Road  
P.O. Box 10935  
Lynchburg, Virginia 24506-0935

SUBJECT: SAFETY EVALUATION OF TOPICAL REPORT BAW-10228P, "SCIENCE"  
(TAC NO. MA4599)

Dear Mr. Coleman:

We have completed our review of the subject topical report that Framatome Cogema Fuels, Inc., (FCF) submitted by letter dated February 5, 1998. The report is acceptable for referencing in licensing applications to the extent specified, and under the limitations delineated in the reports and in the associated U.S. Nuclear Regulatory Commission (NRC) safety evaluation, which is enclosed. The safety evaluation defines the basis for acceptance of the reports.

When the report appears as a reference in license applications, we do not intend to repeat our review of the matters described in the report that we found acceptable, except to ensure that the material presented is applicable to the specific plant involved. Our acceptance applies only to the matters described in the report.

In accordance with procedures established in NUREG-0390, it is requested that FCF publish accepted versions of this report, proprietary and non-proprietary, within 3 months of receipt of this letter. The accepted version shall incorporate this letter and the enclosed evaluation between the title page and the abstract. The accepted versions shall include an "A" (designating accepted) following the report identification symbol.

If our criteria or regulations change so that our conclusions about the acceptability of the report are invalidated, FCF and the licensees referencing the topical report will be expected to revise and resubmit their respective documentation or to submit justification for the continued effective applicability of the topical report without revision of the respective documentation.

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Mr. T. A. Coleman

- 2 -      October 26, 1999

This concludes NRC review activity for this report (TAC MA4599). If you have any questions regarding this matter please contact me at (301) 415-1321, or by email at [snb@nrc.gov](mailto:snb@nrc.gov).

Sincerely,



Stewart N. Bailey, Project Manager, Section 2  
Project Directorate III  
Division of Licensing and Project Management  
Office of Nuclear Reactor Regulation

Project No. 693

Enclosure: Safety Evaluation

cc w/encl:

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UNITED STATES  
NUCLEAR REGULATORY COMMISSION  
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SAFETY EVALUATION BY THE OFFICE OF NUCLEAR REACTOR REGULATION

TOPICAL REPORT BAW-10228P, "SCIENCE"

FRAMATOME COGEMA FUELS, INC.

1.0 BACKGROUND

BAW-10228P describes the SCIENCE code package submitted for review by Framatome Cogema Fuels, Inc., (FCF) for use in nuclear analysis of pressurized-water reactor (PWR) cores (Reference 1). SCIENCE is an integrated system of codes specifically designed for performing nuclear analysis of PWRs. The SCIENCE code package consists of core physics tools that are two-dimensional (2D) lattice calculations and three-dimensional (3D) core calculations and data manipulation codes. The SCIENCE code package consists of the codes APOLLO2-F, SMART, and COPILOTE.

The APOLLO2-F and SMART codes contain the physical description (models) of the SCIENCE code package, while COPILOTE serves as the interface between the user and the two physics codes, permitting sequencing and submittal of the calculations through interactive graphical interface.

2.0 TECHNICAL EVALUATION

2.1 Model Description

The underlying function of the SCIENCE code package is the linking of the major core physics codes, APOLLO2-F and SMART. APOLLO2-F calculates the parameters that are required by the SMART code. These parameters are the cross sections and the discontinuity factors, as well as the pin-to-pin reconstruction parameters (Reference 2).

For each type of composition, the parameters generated by the APOLLO2-F code are placed in data files referred to as "data libraries." The data libraries contain information regarding the dependence of these parameters on feedback system variables, such as burnup, xenon, soluble boron, moderator density, fuel temperature, and spectral effects (Reference 3). These libraries are generated in three steps:

- First, APOLLO2-F performs fuel depletion calculations and stores the data as depletion files. The stored data from the fuel depletion calculations account for the heterogeneity of the assembly under normal (reference) conditions and perturbed conditions. The perturbed condition is signified in this case by a change in the water density, thus indicating the spectral differences between actual conditions in the core and the normal reference core depletion.

- Second, APOLLO2-F takes the results of the depletion calculations and uses them to initialize the isotopic concentrations for new calculations in which one or more physical state variables of the assembly are modified with respect to their initial value(s). These calculations are typically referred to as restart calculations. Some of the variables are xenon level, moderator density, fuel temperature, and a variable that is representative of the control rod presence. All this generated data, representing numerous conditions that the assembly may encounter during the cycle depletion in the core, are placed in "restart" files for use by the SMART code.
- Third, APOLLO2-F creates two data files for each type of fuel. One file contains cross sections and discontinuity factors and the other file contains reconstruction data (pin-by-pin power distribution and burnup). These files form the libraries that contain parameters that are a function of the state variables, such as boron and xenon concentrations, burnup, moderator density, spectral history, fuel temperature profile, and control rod locations.

The fuel assembly is numerically fitted and geometrically represented by dividing the fuel assembly into cubic regions to account for all the possible variations that go into making up the assembly. These variations are represented by polynomial expansions, utilizing determined polynomial coefficients to reproduce the assembly parameters calculated by the APOLLO2-F restart calculations at the fitting point.

## 2.2 Description of Codes

APOLLO2-F is an assembly lattice code developed by the Commissariat a l'Energie Atomique and modified by FCF for its design needs. It solves the 99-group transport equation for an assembly geometry and furnishes the homogenized two-group cross sections for the SMART code. The transport equation is solved using the integral-differential equation form that is discretized based on the collision probability method. FCF pointed out that a coupling of the regrouped multicell calculation and the six-group homogeneous calculation permits a good compromise between accuracy and calculation cost.

This coupling is provided by heterogeneous/homogeneous equivalence functions contained in the code. The assembly calculations can be carried out on various geometries (one-eighth of an assembly, one-fourth of an assembly) with different boundary conditions and symmetry. A sophisticated self-shielding model is applied to the cross sections in order to correctly take resonances into account. The flux calculations can be performed with a search for critical buckling to obtain proper spectral weighting. APOLLO2-F contains a fuel depletion module. The reflector constants (radial or axial) are generated from one-dimensional (1D) APOLLO2-F calculations using the code's  $S_N$  option.

The SMART code solves the two-energy group diffusion equation for the core geometry under static or kinetic conditions. It solves a neutron balance equation using the average flux and provides the core power distributions for assemblies and also for each pin (on a pin-by-pin basis) in every assembly. The nodal expansion method is used to solve the neutron balance equation. It is based on a coupling between a coarse mesh finite difference calculation and a calculation of the neutron current at each interface.

The calculation of the nodal currents is performed by solving a 1D diffusion equation at each calculation node interface. The solution for the adjustment of the nodal currents is obtained by making assumptions of the fast and thermal neutron flux shapes on both sides of the interface and building a system of equations that can be solved directly. This solution is referred to as the "two-node" problem. The accuracy of the process is improved by using discontinuity factors, quadratic transverse leakage, and burnup gradients within the node. The burnup and spectral effects are modeled using a microscopic fuel depletion model.

The main depletion chains for the heavy nuclei and the main fission products are explicitly treated by SMART. The two energy group microscopic cross sections required to calculate isotopic depletion are obtained from data generated by the APOLLO2-F code. The microscopic cross sections and the isotopic densities resulting from the depletion calculations provide the macroscopic cross sections for the flux solution. The microscopic cross sections are stored in "multi-parameterized" data libraries from which the core calculation interpolates, depending on the local node conditions. A set of seven parameters is selected for cross section dependency: burnup, boron concentration, xenon, moderator density, fuel temperature, a spectral history parameter, and a control rod presence parameter. The SMART code calculates fuel pin information for power and burnup and reaction rates in the instrument tube by means of a pin reconstruction algorithm. The SMART code also solves the time-dependent two-energy group diffusion equation for 3D core geometry.

COPILOTE is an operating environment rather than a conventional calculational computer code. It is the graphical user interface by which the user processes input and output, controls the flow of data from one code to another, and displays the status of the calculations.

### 2.3 Measurement Comparisons

In Section 4 of the submittal, FCF provided numerous examples comparing the results of APOLLO2-F and the SMART code with measurement data. The data were collected from six reactors (Three Mile Island Unit 1, Oconee Units 1 and 2, McGuire Unit 1, Gravelines Unit 5, and Sequoyah Unit 1). These cores were selected on the basis of obtaining a wide variety of conditions, such as the type of burnable poison, fuel enrichment, loading patterns, and control rod patterns. Reactivity predictions versus core burnup, control rod worth, reactivity coefficients, and power distributions were provided (on a local and global basis) and compared to measured data from operating PWRs.

The agreement between measured data and SCIENCE prediction is generally very good. The SCIENCE results were just as good or better than prior FCF licensed codes predictions. The uncertainties for single bank worths and total bank worths that are supported by the data presented are 15 percent and 10 percent, respectively. Previous FCF methodology (Reference 1) contained a bias difference between the 15x15 and the 17x17 bank worth results. This bias difference was again observed in the SCIENCE methodology. The cause of the bias is due to the different sources of the measured results. The 15x15 data were obtained exclusively from B&W plants. Nearly all of the 17x17 data were obtained from other vendors using various measurement techniques (Reference 4).

Good agreement was achieved for ejected rod worths, critical boron concentrations, temperature coefficients, and power Doppler coefficients. In addition, review of the data shows that SCIENCE accurately predicts the core total peak and radial power peak. The nuclear reliability factors (NRFs) that SCIENCE uses to adjust the predicted local and global power distribution, were found to be less than those previously established NRFs provided in Reference 1. The staff agrees with the presented results.

#### 2.4 Qualification Methods for Future Modifications to the Science Code Package

FCF intends to periodically update the SCIENCE code package to incorporate the latest analytics and computation techniques. Consequently, any code development or improvement of the SCIENCE code package would necessitate benchmarking and validating the SCIENCE code package to ensure that any new feature(s) implemented will produce results in keeping with a standard set of qualification criteria as stated in the submitted Topical Report BAW-10228P.

The method to be used to qualify SCIENCE for future changes is similar to the method presented in this submittal and previous topical report submittals. This method will require that neutron code qualification be based on the ability of the modified SCIENCE code package to predict several key neutronic parameters. Some of these parameters are critical boron or k-effective at hot zero power, critical boron or critical k-effective at hot full power, individual bank rod worths, total rod worths, ejected rod worths, isothermal temperature coefficients, power Doppler coefficients, hot pin power, and hot pellet power (see Table 5.1 of References 1 and 3). These parameters will be recalculated with the modified SCIENCE code package and compared to measured results and new statistics generated along with their associated uncertainties. Subjecting the modified code package to the listed criteria will emphasize the contributions of the implemented features to the code package rather than highlight the differences between the two code packages. Consequently, any modifications to the SCIENCE code package that meet the listed criteria in Tables 5-1 and 5-2 of this submittal will validate the modifications made to the SCIENCE code package. If the changes to the SCIENCE code package meet the criteria, FCF will internally document the changes to the code package and the associated results without notifying the NRC since there were no changes to the uncertainties or their application. However, if changes to the uncertainties occur, FCF will submit supporting documentation to the NRC whenever the method changes affect the uncertainties to be applied in licensing applications.

The methodology and the data provided in this submittal, Topical Report BAW-10228P, form the basis for the current SCIENCE code package. Future application of the current SCIENCE code package to data not provided in this topical report (such as new fuel designs) will require revalidation of the SCIENCE code package.

#### 2.5 Range of Applicability of Benchmarking

The chosen benchmarks in this topical report include the types of fuel and poison that are typically licensed. The data presented in this topical report are sufficient to qualify the SCIENCE code package for the typical fuel types listed in this submittal. If a new fuel design is used that contains materials (poison, mixed oxide fuel and/or hafnium control rods, etc.) outside this collection of benchmarks, additional benchmarks will have to be established. In

accordance with its agreement, (Reference 4), FCF will submit to the NRC staff a description of the new design feature, the new benchmarks, and any impact on the current uncertainty factors.

### 3.0 CONCLUSION

The staff has reviewed the analyses in Topical Report BAW-10228P, "SCIENCE," and finds it acceptable for licensing applications, subject to the following conditions in accordance with FCF's agreement (Reference 4) :

1. The SCIENCE code package shall be applied in such a manner that predicted results are within the ranges of the validation criteria presented in Table 5-1 and the measurement uncertainties presented in Table 5-2.
2. Fuel designs to which the SCIENCE code package will be applied shall be within the validation bases of BAW-10228P. The bases of BAW-10228P are considered valid for the following conditions:
  - 15x15 or 17x17 UO<sub>2</sub> fuel designs.
  - U235 enrichments less than or equal to a maximum of 5.0 w/o.
  - Gadolinia loadings less than or equal to 8.3 w/o (nominal 8.0 w/o).
3. The following uncertainties shall be applied to the SCIENCE code package results:
  - Maximum pin peaking uncertainty of 3.8 percent.
  - Maximum pellet peaking uncertainty of 4.8 percent.
  - Total rod worth uncertainty of 10~~0~~ percent.
  - Bank rod worth uncertainty of 15 percent.
4. The SCIENCE code package shall only be used for PWR licensing analyses by FCF unless approved by the NRC for use by other organization.

### 4.0 REFERENCES

1. Letter from T. A. Coleman, Vice President of Government Relations, Framatome Cogema Fuels, Inc., to the NRC, regarding the submittal of the SCIENCE code package, dated October 12, 1998.
2. R. Sanchez, J. Mondot, Z. Stankowski, A. Cossic, and I. Zmijarevic, "APOLLO2-F: A User-oriented, Portable Modular Code for Multi-group Transport Assembly Calculations." International Topical Meeting on Advances in Reactor Physics, Mathematics and Computation, Paris, France, April 27-30, 1987.
3. R. Sanchez and M. Vergain, "An Acceleration Procedure for the Iterative Solution of the Flux Current Equations in the APOLLO2-F Code." International Topical Meeting on the Physics of Reactors: Operation and Design Computation, Marseilles, France, PHYSOR 90, April 23-27, 1990.

4. Letter from T. A. Coleman, Vice President of Government Relations, Framatome Cogema Fuels, Inc., to the NRC, regarding the submittal of Topical Report BAW-10228P, "SCIENCE", dated September 23, 1999.

Principle Contributor: T. Attard

Date: October 26, 1999