JEF2 FISSION PRODUCT QUALIFICATION BASED ON FRENCH INTEGRAL EXPERIMENTS

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1. INTRODUCTION

In order to allow for Fission Products and Actinides in Criticality-Safety analyses, a Burnup Credit (BUC) programme has been developed at CEA in the framework of the CEA-COGEMA collaboration. This programme[1] involves two kinds of experiments :

- Chemical analyses and microprobe measurements of PWR pins to obtain the fuel inventory (Actinides and FPs).

- Reactivity worth measurements of the various BUC nuclides by oscillation of specific samples in the Minerve reactor.

The analysis of these experiments was carried out using the APOLLO2 transport code[2] with the CEA93 library. This library is a 172-group data set (XMAS structure) derived from the European JEF2.2 file.

2. CHEMICAL ASSAYS

To validate APOLLO2 depletion and fuel inventory calculations[3], a large experimental programme based on spent fuel chemical analysis has been carried out since 1993. Uranium, Plutonium, Americium and Curium have been analysed in PWR samples. Furthermore, FP chemical elements (Sm, Rh, Nd, Cs, Mo, Eu, Gd, Ag, Ru) have been measured relative to ^{238}U as well as their isotopic composition. We have selected 14 FPs for BUC analyses[4] : these correspond to the most absorbing, stable and non volatile nuclides. In order to check ^{155}Gd build-up, the ^{155}Eu isotope was also investigated.

The chemical analysis programme for UO_2 assemblies is summarised below :

French PWR	Fuel assembly	Irradiation
BUGEY3	17 x 17, 3.1 %	1.5 cycle, 2 and 3 cycles
GRAVELINES3	17 x 17, 4.5 %	2 cycles to 5 cycles

he BUGEY3 assemblies correspond to the standard fuel management in French PWRs. The RAVELINES3 assemblies enable validation of the current trend : high burnups and long cycle lengths.

Chemical analyses are carried out on pin cuts (20 mm long) at various heights along the fuel pin in order to investigate the axial temperature and water density effects. Moreover, various pins are studied inside the assembly to emphasize the radial effect due to the local spectrum. Micropobe measurements along the radius of the GRAVELINES3 pellets enabled us to validate Uranium, Plutonium and FP radial profile calculations[5].

From these assays, we can deduce information on the main FPs. For example, Table 1 shows the C/E comparison on the Nd and Cs isotopics ; from these results some trends are pointed out on the absorbing isotopes, i.e. Nd143-Cs133-Nd145 which are respectively 3rd-5th-11th for their poisoning worth in LWRs.

Isotopic ratio	(C-E)/E in BUGEY3
Nd143/U238	+ 0.3 % ± 0.2 %
Nd144/U238	- 3.1 % ± 0.6 %
Nd145/U238	+ 0.0 % ± 0.3 %
Nd146/U238	- 0.3 % ± 0.8 %
Nd148/U238	+ 2.0 % ± 0.3 %
Nd150/U238	- 0.2 % ± 0.6 %
Cs133/U238	- 4.9 % ± 1.4 %
Cs134/U238	- 3.9 % ± 3.2 %
Cs135/U238	- 10 % ± 1.3 %
Cs137/U238	- 3.0 % ± 1.0 %

Table 1 : Calculation-Experiment comparison based on JEF2.2
Average disagreement on 5 assembly samples, and spread (1 s)

2.1 Trends on the JEF2.2 thermal fission yields

The satisfactory build-up of Nd143 and Nd145 in APOLLO2 demonstrates that their cumulative yields are well represented in JEF2.2 (they are consistent with Meek and Rider recommendation).

The underestimation of Cs133 points out that JEF2 thermal yields of Xe133 are underestimated. Meek and Rider values are more satisfactory : + 1.5 % and + 1.3 % on U235 and Pu239 fission yields respectively compared to JEF2 values.

d148 and Cs137 are not absorbing FPs, but they are used as burnup monitor. Thus, their calculated nounts must be obtained within 2 % accuracy. Unfortunately, the JEF2.2 Nd148 yield from Pu239 fission was increased, compared to the Meek and Rider values, leading to a - 2 % low burnup estimation. At the opposite, the Cs137 yields look underestimated in JEF2.2.

2.2 Trends on the radiative capture cross sections

The measured concentration of the daughter of a poisoning FP enables the qualification of the (n, gamma) cross sections. Table 2 shows the APOLLO2/Exp. comparison on the Nd144/Nd143, Nd146/Nd145 and Cs134/Cs133 ratios.

Isotopic ratio	BUGEY3 (3 rods)	GRAVELINES
Nd144/Nd143	- 3.4 % ± 0.6 %	- 5.6 % ± 1.8 %
Nd146/Nd145	- 0.3 % ± 0.6 %	- 0.2 % ± 0.4 %
Cs134/Cs133	+ 0.9 % ± 3.4 %	- 5.0 %± 3.1 %

Table 2 : Calculation-Experiment comparison based on JEF2.2

These results stress a - 4 % ± 1 % underestimation of the thermal (n, gamma) cross-section of the Nd143 poisoning isotope.

Table 2 shows that Nd145 capture is perfectly mocked-up in JEF2.2. The resonant capture of the Cs133 fission product (Eo = 5.9 eV large resonance) seems satisfactory.

3. INTEGRAL MEASUREMENTS OF FP CROSS-SECTIONS IN MINERVE REACTOR

FP capture cross-sections were obtained from reactivity measurements of separated FP samples oscillated at the center of MINERVE test lattices.

We manufactured 3 kinds of PWR-type samples within the framework of the Burnup Credit Programme. The 3 types of oscillation samples (UO_2 and PuO_2, separated FP isotopes in UO_2 pellets, and irradiated samples) had identical geometries, specifically 100 mm-long sections of PWR pins contained in thin, water tight zircaloy sleeves with an external length of 103.5 mm.

3.1 Separated FP oscillation samples

The goal of these FP samples is to supply the poisoning reactivity worth of each BUC-FP. The separated FP is homogeneously mixed inside the fuel pellet (natural UO_2)

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he characteristics of these FP samples are summarized in Table 3. The mass of each FP isotope by ımple was optimised in order to obtain a similar reactivity worth corresponding to the maximum accuracy in Minerve worth measurements.For the most resonant absorbers, i.e., ^{133}Cs-^{103}Rh-^{109}Ag, we manufactured several samples with increasing FP isotope amount, in order to investigate the resonance self-shielding effect.

Sample UO_2+X_2O_3	Additive X	Mass X (ppm)
UF		0
Sm	Smnat	460
Sm9	^{149}Sm	69.3
Sm2	^{152}Sm	10750
Sm7	^{147}Sm	18500
Nd	Ndnat	69900
Nd3	^{143}Nd	10700
Nd5	^{145}Nd	59500
Gd5	^{155}Gd	121
Eu3	^{153}Eu	9000
Rh	^{103}Rh	5350
Sample non-sintered UO_2	Additive	Mass Additive (g)
UC		0
Cs3 % 8	Cs_2CO_3	3.77
Cs3 % 6	Cs_2CO_3	2.76
Cs3 % 1	Cs_2CO_3	0.46
Mo5	$Mo95O_3$	5.66
Tc9	KTcO4	4.32
Ag9 % 2	$AgNO_3$	1.07
Ag9 % 0.3	$AgNO_3$	0.117
Rh3 % 0.8	Rh	0.372
Rh3 % 0.1	Rh	0.045

Table 3 : Separated FP samples

To obtain complementary information on metallic FPs, we manufactured simulant samples containing a natural element mixed in a Al_2O_3 matrix : Ag, Ru, Rh, Mo and Cs samples. The measured reactivity worth of the Ag, Ru and Mo elements is mainly due to the BUC nuclides : ^{109}Ag, ^{101}Ru and ^{95}Mo, respectively.

The sample content in both types of FP samples was obtained from chemical or mass spectrometer analysis on pellets from the same batch.

.2 Reactivity worth measurements

The samples were oscillated in the centre of the PWR lattice (MELODIE block) implemented at the Minerve reactor.

The first configuration, R1-UO_2, corresponds to a uniform PWR-type lattice, 1.26 cm square pitch, made of 800 3 %-enriched UO_2 rods. This R1-UO_2 is devoted to BUC investigation in storage pool and PWR-assembly transportation. The slowing-down density at thermal cut-off, which is representative of the spectrum hardness, amounts to q = 0.65 in the R1-UO_2 lattice.

The second experimental configuration R2-UO_2 aims to mock-up the softer spectrum corresponding to the optimum moderation-ratio in a fuel dissolver (q = 0.8).

The R1-UO_2 experiment was carried out in 3 successive experimental campaigns : in 1993, 1994 and 1995, respectively, for the separated FP samples, the spent fuel samples, and the natural FP in Al_2O_3 matrix. The R2-UO_2 experiment was performed in 1996-1997.

The samples were oscillated at the centre of the Minerve Zone. Moreover, borated UO_2 and variable enrichment UO_2 samples were oscillated for reactivity worth calibration.

The central pin of the R1-UO_2 lattice is periodically oscillated through the core, so the sample under study is alternatively in and out of the core. A rotating control rod is automatically operated so as to maintain the count rate of a flux detector. The corrected rotation amplitude is in a very close linear relationship with the sample reactivity.

One measurement for each sample corresponds to 30 oscillations with a period of T = 60 seconds. Each sample is measured at least 3 times. The FP poisoning worth is directly derived by subtracting the FP sample reactivity from the reactivity of the reference sample (natural UO_2). The uncertainty on the FP measured worth is $\pm$ 0.7 % in one standard deviation. Effective FP cross-sections are obtained in terms of the boron effective cross-section, from the comparison between the measured poisoning worth for the FP and boron samples ; thus a supplementary uncertainty must be added due to this reactivity worth calibration ($\pm$ 1 % systematic uncertainty).

3.3 FP reactivity worth qualification

The neutron fluxes in the Minerve R1-UO_2 and R2-UO_2 Test Zones were obtained by a 2D transport Calculation. We used the P_{IJ} method in APOLLO2 in order to account for the exact heterogeneous geometry. We checked that a sophisticated multicell assumption based on the interface current method is accurate enough. Therefore, the sample reactivity worth is obtained from the exact perturbation formula :

$$\rho = \left\langle \phi^{+}, \Delta H \phi^{*} \right\rangle \Big/ \left\langle \phi^{+}, P \phi^{*} \right\rangle$$

where ΔH stands for the modification of the Boltzmann operator, ϕ^{+} and ϕ^{*} stand for the adjoin flux and the perturbed direct flux respectively.

he Calculation-Experiment comparison on the separated FP worth in the R1-UO_2 and R2-UO_2 onfigurations is summarised in Table 4.

FP	Mass (g)	R1-UO_2		R2-UO_2	
		(C-E)/E in %	exp. uncert. 1 s (%)	(C-E)/E in %	exp. uncert. 1 s (%)
Sm	0.026	- 4.5	2.9	- 3.3	3.6
^{149}Sm	0.004	- 6.0	2.9	- 4.9	3.6
^{147}Sm	1.008	+ 1.3	4.3	+ 2.7	4.7
^{152}Sm	0.586	- 1.6	2.9	- 1.8	3.7
Nd	3.602	+ 0.4	3.0	- 3.3	3.7
^{143}Nd	0.574	- 7.1	3.1	- 8.5	3.8
^{145}Nd	2.325	+ 0.4	3.8	+ 1.1	4.4
^{155}Gd	0.008	- 2.5	2.9	- 6.1	4.0
^{153}Eu	0.431	- 4.2	4.0	- 1.3	4.6
^{99}Tc	2.142	+ 4.1	3.8	+ 3.4	3.5
^{95}Mo	3.650	- 3.1	3.4	- 3.7	3.8
^{133}Cs	3.076	+ 8.5	3.2	+ 7.6	3.8
^{133}Cs	2.200	+ 7.6	3.5	+ 9.3	3.8
^{133}Cs	0.378	- 11.0	7.3	- 0.4	5.7
^{103}Rh	0.376	+ 11.0	4.0	+ 8.0	4.2
^{103}Rh	0.045	-	-	+ 14	9.0
^{109}Ag	0.640	- 3.6	4.3	- 4.5	4.3
^{109}Ag	0.073	- 4.6	9.0	+ 2.8	6.9
Ag	1.105	- 4.7	4.2	+ 0.3	4.7
Ru	5.850	+ 10	3.8	-	-
Mo	19.740	+ 1.5	3.2	+ 2.1	3.8
^{133}Cs	2.290	- 0.6	3.8	- 2.4	4.3
^{133}Cs	0.290	+ 4.1	8.5	+ 9.1	7.3

Table 4 : FP reactivity worth . (C-E)/E in %

he refined 172-group energy mesh allows the calculation to account for the self-shielding of the rst resonances[6] : ^{103}Rh (E_R = 1.3 eV), ^{99}Tc (E_R = 5.6 eV), ^{133}Cs (E_R = 5.9 eV), ^{109}Ag (E_R = 5.2 eV). However, the separated isotope load in some FP samples can reach 50 times the actual FP amount in a LWR spent fuel. Therefore, the resonance self-shielding of Mo95, Tc99, Ag109, Cs133, Nd145, Eu153 was rigorously calculated through an effective cross-section formalism.

Table 4 shows a quite satisfactory calculation-experiment agreement using JEF2 library in APOLLO2 calculation. However, the (n, gamma) cross-section of Nd143 absorber is underestimated in the thermal energy range by - 7 % ± 3 % (1 σ). The thermal capture of Sm149 is also underestimated by - 5 % ± 2 % (1 σ). The Rh103 capture inside the 1.3 eV resonance looks overestimated by + 10 % ± 4 % (1 σ). These trends are confirmed by the AEA work[7].

The Cs133 resonant capture is overestimated by + 7 % ± 3 % (1 σ) in UO_2 matrix samples, and is well reproduced in Al_2O_3 matrix samples (0 % ± 3 %). This disagreement could be due to U238/Cs133 approximate mutual shielding calculation in UO_2 matrix samples : in this case the E_{U238} = 6.67 eV/E_{Cs133} = 5.9 eV resonance overlapping requires an ultrafine energy mesh calculation. The Cs inert matrix samples give consistent result with isotopic ratio measurements (- 1 % ± 3 %).

CONCLUSION

The qualification of the radioactive capture cross-sections of the main FPs was achieved through sample worth measurements and PWR spent fuel assays. The calculation-experiment comparison based on JEF2 data is summarized below (experimental uncertainty in 1 σ) :

Mo95	(C-E)/E =	- 2 % ± 3 %
Tc99		+ 3 % ± 3 %
Rh103		+ 10 % ± 3 %
Ag109		- 3 % ± 3 %
Cs133		+ 1 % ± 3 %
Nd143		- 5 % ± 2 %
Nd145		0 % ± 1 %
Sm147		+ 2 % ± 3 %
Sm149		- 5 % ± 2 %
Sm152		- 1.6 % ± 2.5 %
Eu153		- 3 % ± 3 %
Gd155		- 3 % ± 3 %

From these results on the 12 main poisoning FPs (except the decaying Xe135 and Sm151 isotopes, and the gaseous Xe131 isotope), we can draw the following conclusions on the JEF2 capture cross-sections :

- Mo95, Tc99, Ag109, Cs133, Nd145, Sm147, Sm152 and Gd155 evaluations are satisfactory.

The Eu153 absorption rate is acceptable, however the JENDL3.2 evaluation improves the C/E agreement. Additional integral benchmarking would be useful.

- The E_R = 1.3 eV resonance integral of the Rh103 nuclide should be strongly decreased by 10 %. The Rh103 evaluations in the current international data files are no more suited.
- New evaluations of Sm149 and Nd143 are needed in JEFF3, in order to increase by 5 % the (n, gamma) cross-section in the 0 - 0.2 eV energy range. The JENDL3.2 file slightly improves the Nd143 reactivity worth (+ 0.3 %), but gives worse results on Sm149 poisoning.

These FP cross-section improvements in JEFF3 will allow more accurate reactor cycle lengths in LWR calculations, and less conservatism penalties in criticality calculations using burnup credit. Furthermore, cumulative Cs133 yields of U235 and Pu239 thermal fissions must be increased.

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