EXPERIMENTAL VALIDATION OF JEF2 FISSION PRODUCTS. REQUIRED IMPROVEMENTS IN THE JEFF3 EVALUATIONS

A. SANTAMARINA, C. CHABERT, B. ROQUE, N. THIOLLAY, G. WILLERMOZ

CEA - Cadarache DRN/DER/SPRC
13108 Saint Paul Lez Durance Cedex France
✆ 33 4 4225-7046, 33 4 4225-7009 FAX, alain.santamarina@cea.fr

1. THE FRENCH EXPERIMENTAL DATA BASE

In order to validate PWR depletion calculation routes and cycle length prediction, an extensive PIE Programme was launched in the framework of the CEA-EdF-Framatome collaboration[1]. The fuel inventory calculation was mainly qualified through the analysis of fuel irradiated 2 - 3 cycles in Bugey3 and 4 - 5 cycles in Fessenheim2 (3.1% U235 enrichment), and fuel irradiated for 2, 3, 4 5 cycles in Gravelines (4.5% enriched - maximum burn-up fraction : 62 GWd/t_m) [2]. Furthermore, to qualify Pu recycling in French PWRs, chemical assays were performed in MOX assemblies irradiated in SLB1 and Gravelines4 reactors [3].

To allow for Fission Products in Criticality-Safety analyses, a Burnup Credit (BUC) programme has been launched at CEA[4] in 1990. The experiments were first carried out within the CERES co-operation : oscillation of UO2 samples doped with a separated FP isotope at the center of a 800 PWR-UO2 fuel rod block in the Minerve reactor. The BUC programme was pursued in the framework of the CEA-COGEMA collaboration : oscillation of separated FP isotopes (UO2 matrix and inert Al2O3 matrix) in dissolver medium spectrum, BWR spectrum and MOX lattice[5].

These programmes involve three kinds of experiments[6] :

- Chemical assays and microprobe measurements[7] of LWR pins to obtain the fuel inventory (Actinides and FPs).

- Reactivity worth measurements of the various BUC nuclides by oscillation of specific samples in several lattice blocks located at the centre of the Minerve reactor.

- Reactivity worth measurements of PWR fuel cuts (up to 62 GWd/t_m) in the Minerve reactor.

2. CALCULATION ROUTE

The analysis of these experiments was carried out using the APOLLO2 transport code[8] with the CEA93 library. This library is a 172-group data set (XMAS structure) derived from the European JEF2.2 file.

The refined 172-group energy mesh allows the calculation to account for the self-shielding of the first resonances[9] : ^{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.

Concerning PIE analysis, a true master calculation covering all the physical parameters precisely, results in excessive calculation costs and cannot therefore be recommended for industrial uses. This is the reason behind the development of the APOLLO 2 calculation route for evolving 17x17 UO_2 assemblies. It enables the bias to be minimised between the "master" calculations, and it satisfies our target accuracy, namely 1% for Pu_{239} and power in each pin, up to 70 GWd/t_m. For instance, the recommendations for the flux calculation are as follows :

- The spatial calculation for the UOx assembly is achieved using the UP_1 HETE approximation. This enables the probability of leakage P_{IS} and the probability of transmission P_{SS} to be calculated for the true geometry, the interface currents are considered to be linearly anisotropic.
- The inter-assembly water gap is integrated within the peripheral cells of the assembly, constituting a row of rectangular fuel cells.
- The fuel pellet is split into 4 rings (50%, 30%, 15% and 5% of the total volume of the pellet) in order to give a faithful representation of the resonant absorption of U_{238} in the pin and of the actinide and fission product concentration profiles. This leads to the determination of 72 evolving media (18 physical cells x 4 rings).

The studies conducted for the calculation of self-shielding in 17x17 UO_2 assemblies, also enabled recommendations to be made for this :

- The isotopes to be self-shielded are : U_{238}, U_{235}, U_{236}, Pu_{239}, Pu_{240}, Pu_{241} and Pu_{242}.
- Self-shielding of the U_{238} large resonances are covered by the accurate model UP_1.
- It is essential to differentiate between various self-shielded pins for the U_{238} in order to adequately take into account the differences in the Dancoff effect :
    - 1 pin opposite the water hole ,
    - 1 pin at the corner of the water hole,
    - 1 peripheral pin.
- The self-shielding of U_{238}, U_{236} and Pu_{240} is considered in rings by the Background Matrix formalism[8].

Concerning oscillation experiment analysis, the Minerve Test Zones were calculated by a 2D transport model. 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 adjoint flux and the perturbed direct flux respectively.

3. TRENDS ON FP JEF2 DATA FROM INTEGRAL MEASUREMENTS

The analysis of the Minerve reactivity worth measurements of the CERES samples in the PWR, MOX and dissolver spectra gave consistent results with the worth measurements in Dimple[10] . The additional samples manufactured in the framework of the CEA/COGEMA Burnup Credit Programme confirmed the reported trends[6] , as well as the FP chemical assays in PWR spent fuels.

3.1 Rh103

The cumulative yield of Rh103 looks slightly overestimated by + 4 % ±3 % (1 σ).
The E_R = 1.3 eV resonance integral should be strongly decreased by 10 %±3 % (1 σ).

3.2 Sm149

The Sm149 equilibrium concentration is well predicted by APOLLO2 ; therefore, the Nd149 yields of U235 and Pu239 fissions are satisfactory in JEF2 file.
From Sm149 and Sm-nat samples oscillated in the three Minerve lattices, we can conclude that the thermal capture cross section is underestimated by - 5 %±2 % (1 σ). This trend is confirmed by the 4 % underestimation of the Sm150 build-up in PWR spent fuel.

3.3 Nd143

The satisfactory build-up of Nd143 in APOLLO2 demonstrates that the cumulative yields are well represented in JEF2.2 (they are consistent with Meek and Rider recommendation).
From Nd143 and Nd-nat samples oscillated in the three Minerve lattices, we can deduce that the thermal capture cross section is underestimated by - 4 %±2 % (1 σ).

3.4 Cs133

The Calculation - Experiment on the Cs isotopics is summarised in Table 1.

		CYCLE	BU (MWd/t)	Cs133/U238	Cs134/U238	Cs135/U238	Cs137/U238
Bugey 3	G11	2A	20400	-3.00±1.53	-8.48±2.92	20.44±6.20	-2.10±1.60
	K07	2B	24700	-4.17±1.44	-14.63±2.88	16.03±6.18	-4.25±1.51
	K11	3	38300	-6.83±1.42	-3.87±2.68	5.57±4.30	-5.15±1.62
Gravelines	K11	2	26900	-6.42±1.46	-15.02±2.83	-0.75±5.23	-7.43±1.61
	G07	3	38360	-3.83±1.41	-11.69±2.70	-1.63±4.29	-5.63±1.61
	G11	4	38360	-4.20±1.36	-8.61±2.57	-3.26±3.39	-6.47±1.79
	J07	5	59850	-4.98±1.35	-1.92±2.43	-4.42±2.57	-7.43±1.92
	J09	5	59850	-3.98±1.35	0.60±2.43	-3.12±2.57	-4.24±1.92
Fessenheim	H08	4	49100	-1.21±1.28	-0.06±2.54	4.58±3.38	-3.78±1.73

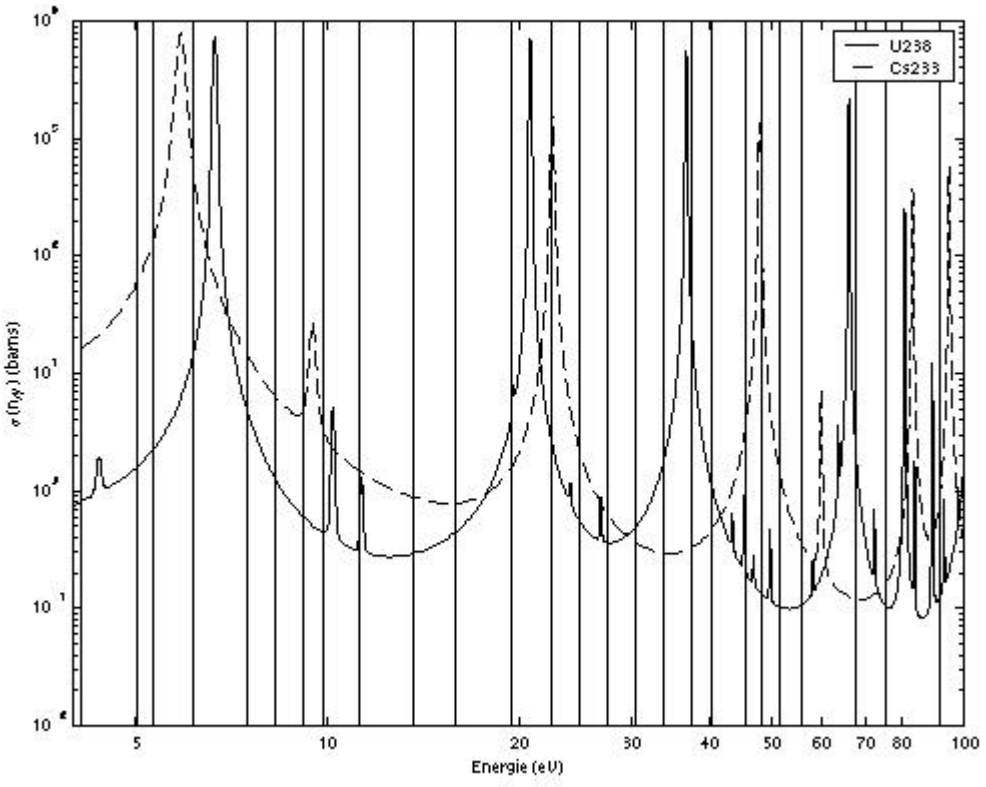
K11	5	58000	-4.84±1.28	-4.43±2.53	-0.35±2.56	-6.68±2.03

Table 1 : Calculation-Experiment comparison on Cs isotopics ratios

The underestimation of Cs133 concentration 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.
Cs137 is not an absorbing FP, but it is used as burnup monitor ; its yield seems underestimated in JEF2.2.
The measured concentration of the daughter of a poisoning FP enables the qualification of the (n, gamma) cross sections : the Cs134/Cs133 ratios indicate that Cs133 capture is calculated within 5% accuracy (in one standard deviation) ; however, this tendency is not enough accurate due to the short T = 2 years half period of Cs134.

The Minerve measured reactivity worth of the various Cs133 samples were reanalysed using a reference TRIPOLI4/JEF2.2 Monte Carlo calculation. This continuous energy calculation enables to overcome mutual shielding (E_{U238} = 6.67 eV/E_{Cs133} = 5.9 eV and E_{U238} = 21 eV/E_{Cs133} = 22.5 eV resonance overlapping) and self-shielding problem in the large 5.9 eV resonance, as shown in fig. 1.
The TRIPOLI4 analysis of the two UO2-Cs133 samples (3 grams and 2.2 g) stresses an overprediction of the Cs133 worth by + 6 %±3 % (1 σ). Thus the JEF2 resonance integral should be reduced in agreement with the JENDL3.2 evaluation, as shown in Table 2.

Figure 2 : Cs133 and U238 capture cross sections (XMAS 172 group limits)



Bibliothèque	(barns)
JEF2.2	438.6
ENDF/B-VI	382.7
JENDL3.2	395.9
CEA93-V6	438.7

Table 2 : Cs133 Resonance Integral in the various library

3.5 Eu153

The Eu153 build up is slightly overestimated using JEF2 file : + 6 % at 20 GWd/t up to + 13 % at 60 GWd/t. This trend is consistent with the 4 % underestimation of the measured poisoning worth of the Eu153 sample in the Minerve lattices. The use of the JENDL3.2 evaluation cancels the C/E disagreement in reactivity worth experiments.

3.6 Mo95

The Mo95 content is well predicted in PWR spent fuel. Furthermore, the reactivity worth of the Mo95 sample in Minerve is perfectly reproduced using JEF2. On the contrary, the use of the JENDL3.2 evaluation would worsen the C/E comparison by + 5 %.

3.7 Sm152

The Sm152 build up is correctly calculated : + 1 % ± 2 % (1 σ). The reactivity worth of the Sm152 is underestimated by -2 % ± 2 % (1 σ) in Minerve lattices.

3.8 Nd145

The Nd145 concentration is perfectly assessed in depletion calculation. Reactivity worth of the sintered UO2-Nd145 sample pointed out that Nd145 capture is nicely mocked-up in JEF2.2.

3.9 Tc99

The Tc99 build up calculation is satisfactory : C/ E = 0 % (± 3 % spread among PWR cuts).
The Tc99 reactivity worth looks overestimated by + 4 % ± 3 % (1 σ) in hard spectra.

3.10 Eu155

The Eu155 concentration is overestimated from + 8 % at 20 GWd/t up to + 14 % at 60 GWd/t. This trend is consistent with the too low JEF2 Resonance Integral I = 2170 barns, instead of 15 500b in JENDL3.2 (recommended Mughabghab value : 23 200 ± 300 b)[11].

3.11 Gd155

Unlike the Eu155 overestimation, the Gd155 content (mainly linked to the decay of Eu155 T = 5.0 years) is predicted by APOLLO2/JEF2 with a reasonable accuracy : + 3 % ± 3 % (1 σ).
The Gd155 worth calculation is satisfactory in the Minerve oscillations : - 3 % ± 3 % (1 σ).

3.12 Sm147

The Sm147 is slightly underestimated : - 6 % ± 2 % (1 σ). The measured reactivity worth is well predicted : + 1 % ± 3 % (1 σ).

3.13 Ag109

The measured reactivity worth is correctly reproduced : - 3 % ± 3 % (1 σ).

4. CONCLUSION

The qualification of the (n,gamma) cross-sections of the main FPs was achieved through sample worth measurements and PWR spent fuel assays. From these results on the 13 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, Nd145, Sm147, Sm152 and Gd155 evaluations are satisfactory.
- The Eu153 absorption rate is acceptable, however the JENDL3.2 evaluation would improve the C/E agreement.
- The Eu155 Resonance Integral must be corrected in JEF2 evaluation.
- The E_R = 1.3 eV resonance integral of the Rh103 nuclide should be strongly decreased by 10 %.
- New evaluation of the Sm149 is needed in JEFF3, in order to increase by 5 % the peak level in the 0.1 eV resonance. Notice that the JENDL3.2 file gives worse results on Sm149 poisoning.
- New evaluation of the Nd143 is required in JEFF3, in order to increase by 4 % the thermal (n, gamma) cross-section in the 0 - 0.2 eV energy range. The JENDL3.2 file could improves the Nd143 reactivity worth by + 0.3 % only.
- New evaluation of the Cs133 is required in the resonance range, mainly for the large first resonance E_{Cs133} = 5.9 eV : its resonance integral should be decreased by 6 % in agreement with the JENDL3.2 evaluation.

Concerning FP yields, Cs133 yields of U235 and Pu239 thermal fissions must be increased. Nd148 and Cs137 are not absorbing FPs, but they are used as burnup monitor. Thus, their calculated amounts 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. To the contrary, Cs137 yields seem to be underestimated in JEF2.2.

REFERENCES

1. C. CHABERT, P. MARIMBEAU, A. SANTAMARINA, C. POINOT, J.C. LEFEBVRE
« Experimental validation of UOX and MOX spent fuel isotopics predictions »
Proc. of Int. Conf. on the Physics of Reactors PHYSOR'96. Mito (Japan), Sept. 16-20, 1996

2. C. CHABERT, A. SANTAMARINA, R. DOREL, D. BIRON, C. POINOT
« Qualification of the APOLLO2 assembly code using PWR-UO2 isotopic assays. The importance of irradiation history and thermo-mechanics on fuel inventory prediction ».
Proceedings Int. Conference PHYSOR2000, Pittsburgh(USA), 7-11 Mai 2000.

3. C. CHABERT, A. SANTAMARINA, P. BIOUX
« Elaboration and experimental validation of the APOLLO2 depletion transport route for PWR Pu recycling ». Proceedings Int. Conference PHYSOR2000, Pittsburgh(USA), 7-11 Mai 2000.

4. A. SANTAMARINA et al.
« Experimental Validation of Burnup Credit Calculations by Reactivity Worth Measurements in the Minerve Reactor », Proc. Int. Conf. on Nucl. Criticality ICNC'95, Albuquerque (USA), September 17-21, 1995

5. A. SANTAMARINA, N. THIOLLAY, C. HEULIN, J.P. CHAUVIN
« The French Experimental Programme on Burnup Credit », Proc. ANS Top. Meeting on Criticality Safety Challenges. Lake Chelan (USA), September 7-11, 1997

6. A. SANTAMARINA, N. THIOLLAY, C. CHABERT
« JEF2 Fission Product qualification based on French Integral experiments », JEF-DOC-784, Issy-les-Moulineaux, 12-14 April, 1999

7. A. SANTAMARINA, J.C. ESTIOT
« Effect of the Nuclide Radial Distribution in LWR pins on BUC Calculations », OECD BUC Working Group Meeting, Albuquerque (USA), Sept. 27-27, 1995

8. R. SANCHEZ et al
« APOLLO2 : a User-Oriented, Portable, Modular Code for Multigroup Transport Assembly Calculations », Nucl. Sci. Eng. 100, 352-362 (1988)

9. B. ROQUE, A. SANTAMARINA, C. MATTERA, E. LEJEUNE

« Validation of the New French Criticality-Safety Package CRISTAL », Proc. ANS Top Meeting on Criticality Safety Challenges. Lake Chelan (USA), September 7-11, 1997

10. N.T. GULLIFORD
« Notes in Support of Discussions at JEF Specialists Meeting on Fission Products ».
JEF Meeting, 18-19 June 1997, NEA Databank (JEF-DOC-732).

11. C. DEAN
JEF Meeting, NEA Databank, JEF-DOC-794