Improvements of Isotopic Ratios Prediction through Takahama-3 Chemical Assays with the JEFF3.0 Nuclear Data Library

Arnaud Courcelle*[1] Alain Santamarina[1] and Stéphane Mengelle[2]
[1] *CEA-Cadarache, DEN/DER/SPRC, 13108 St-Paul-Lez-Durance, France*
[2] *CEA-Saclay, DEN/DM2S/SERMA, 91191 Gif-sur-Yvette, France*

This paper presents the interpretation of a destructive experiment of PWR fuels from the Takahama-3 reactor with the latest versions of the JEFF European nuclear data library. Such experiments, widely used in the JEF2.2 testing process, provide meaningful information to test specific cross-sections (or fission yields) in the thermal and resonance range. It is demonstrated in this work that the new JEFF3.0 library greatly improves main actinides and fission products isotopic ratio predictions compared with JEF2.2 results. The incorporation of JEFF3.0 evaluations within the multigroup nuclear library CEA2003V1 contributes towards the improvement of the APOLLO2 neutronic prediction capabilities. In particular, the new U235, U238 and Pu241 files adopted in JEFF3.0 remove longstanding discrepancies observed in U236, Np237 and Pu242 build-up prediction with JEF2.2 and reduce C/E discrepancies of minor actinides. Despite these improvements, further differential measurements and evaluation work are suggested to achieve a better accuracy in the prediction of Americium and Curium isotopes.

KEYWORDS: Nuclear data, Validation, JEF2.2, JEFF3.0, isotopic ratios, CEA2003, APOLLO2, Post-Irradiated Experiments, Takahama

1. Introduction

In the framework of the JEFF project and during the elaboration process of the new library JEFF3.0 released in April 2002, destructive analyses of irradiated fuel from PWR (UO_2 and UO_2-PuO_2 fuel) have reliably contributed to the improvements of actinide and fission product nuclear data (see [1] and [2]). Some of the JEF2.2 cross-sections of major isotopes (U235, Pu241, U238) have been revisited to take into account this experimental information and improve inventory calculations.

The aim of this article is twofold : the Post-Irradiated Experiment (PIE) carried out in Japan in the Takahama-3 reactor is analysed with JEF2.2 in order to confirm in an independent manner the trends provided by the French experiments. Besides, the whole JEFF3.0 library is tested and its performances are compared with JEF2.2.

2. The Takahama-3 Post-Irradiated Experiment

Takahama-3 is a Pressurized Water Reactor operated by the Kansai Electric Power Company in Japan. From the 17×17 assemblies called NT3G23 and NT3G24, several samples were cut and isotopic compositions were measured for irradiated UO_2 and UO_2-Gd_2O_3 fuel rods. The specification of this benchmark is taken from the document [3] issued in the framework of co-operation between CEA and Japanese University Association. Furthermore, the Takahama-3 characteristics are now included in the promising SFCOMPO database originally developed at the JAERI and now maintained at the NEA Data Bank.

* Corresponding author, E-mail: arnaud.courcelle@cea.fr

The main features of the Takahama-3 reactor are summarized in Table 1.

Table 1 ; Main features of the Takahama-3 facility

Reactor type	**PWR**
Pellet Diameter (mm)	8.05
Clad Thickness (mm)	0.64
Clad Material	Zry-4
Rod Array	17×17
Number of Rods	264
Fuel Rod Pitch (mm)	1.265
Eq. Diameter (mm)	about 3040
Active Height (mm)	about 3660
Number of Assemblies	157

Some important physical data are missing in [3] and in the SFCOMPO database so that additional assumptions have to be made regarding the experimental conditions (geometry, temperature, boron contents). Present nuclear data library testing is performed with one sample extracted from the NT3G24 assembly (displayed in Figure 1) which was loaded with 248 UO_2 pins (4.1%wt U235), 16 UO_2-Gd_2O_3 pins (2.6% wt U235 and 6%wt Gd) and 25 water holes. This assembly has been irradiated 3 cycles and the SF97 experimental fuel pin reached an average burn-up around 45 GWj/t. The SF97 UO_2 pin is located in the peripheral row of the assembly (Figure 1). The SF97-4 sample, located at the mid-height of the fuel pin was chosen in this study (see Figure 2).

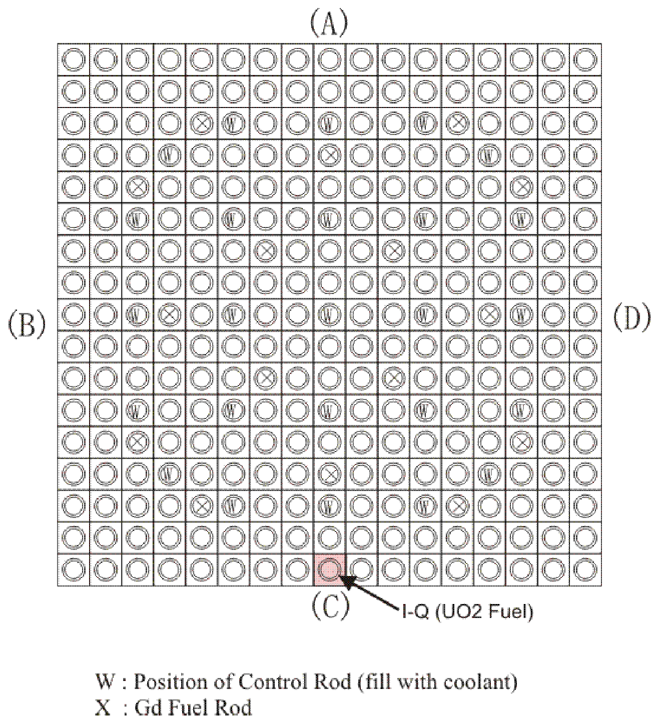

Fig. 1 : Takahama NT3G24 Assembly geometry

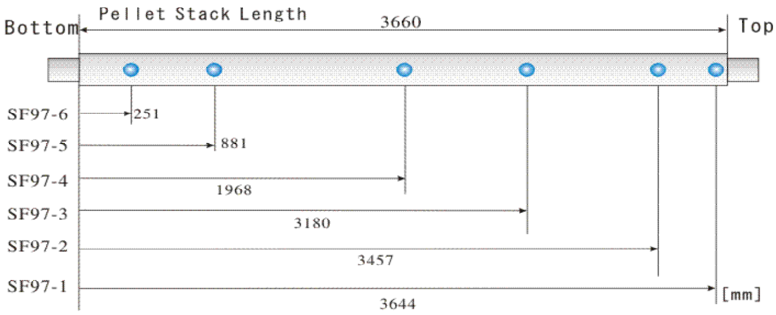

Fig. 2 : SF97 fuel pin. The sample SF97-4 has been analyzed in the present study.

3. Calculation methodology

Neutronic assembly calculations are performed with the deterministic code APOLLO2 [4] developed at CEA. The calculation route follows the main recommendations defined in previous analyses of Post-Irradiated Experiments (see [1] and [2] for a detailed description). The CEA93 JEF2.2 nuclear data library and the newly released CEA2003 (Version 1) based on JEFF3.0 are both processed consistently with the NJOY99 code (using tight tolerance in the reconstruction procedure) within the XMAS energetic structure (172 energy groups). It should be mentioned that fission yields and decay data are the same in our JEF2.2 and JEFF3.0 calculations.

APOLLO2 features an efficient self-shielding formalism [4] for the major resonant isotopes avoiding calculation biases in the resonant reaction rate within 172 energy groups. Continuous Energy Monte Carlo calculations have checked the accuracy of the present approach. Usual biases in energy groups, where resonances from two different isotopes overlap, are noticed and work is still in progress to improve the modeling of mutual shielding.

The Probability collision method (Pij) is used for the resolution of the integral form of the Boltzmann transport equation in self-shielding and flux calculations. Collision probabilities are computed in the so-called 2D-UP1 approximation which assumes linearly anisotropic interface angular fluxes. 4 rings in UO_2 and 6 rings for UO_2-Gd_2O_3 ensure a good representation of the radial profile of the neutron flux, as well as an accurate space-dependent self-shielding and depletion calculation.

In the present analysis, a constant irradiation was considered. The consequence of this assumption (neglecting actual power history and stretch-out) has been previously studied in similar PWR configurations [2] and is significant only for Am241/U238 (2%), Cm242/U238 (5%), Am242m/U238 (10%) and Pu238/U238 (2%) isotopic ratios. The main sources of experimental uncertainties (fuel and moderator temperature, burn-up determination through Nd145/U238 indicator, geometry, chemical assay) have been also analysed for similar PWR through a sensitivity study in [2] and are merely combined in a "total uncertainty" quoted in the following Tables 2 and 3. The comparison of this uncertainty to C/E discrepancy allows the detection of meaningful trends on nuclear data.

4. Results and discussion

C/E values for JEF2.2 and JEFF3.0 are compared in Table 2 (actinide) and in Table 3 (fission products). The JEF2.2 results generally confirm the previous conclusions drawn from the experiments performed in Gravelines, Bugey and Fessenheim reactors [5].

Table 2 : C/E-1 in % for actinides with JEF2.2 and JEFF3.0 for the SF97-4 sample (Burn-up = 45 GWd/t).

isotope	**C/E-1 JEF2.2**	**C/E-1 JEFF3.0**	**total unc. 1σ**
Uranium			
U235/U238	+1.0%	-0.4%	±2.7%
U236/U238	**-4.9%**	**-1.5%**	**±0.7%**
Neptunium			
Np237/U238	**-8.1%**	**-3.7%**	**±3.4%**
Plutonium			
Pu238/U238	**-18.9%**	**-14.1%**	**±3.8%**
Pu239/U238	-4.7%	-4.8%	±2.0%
Pu240/U238	+1.5%	+2.9%	±1.3%
Pu241/U238	-8.5%	-7.3%	±2.3%
Pu242/U238	**-10.2%**	**-3.8%**	**±3.1%**
Americium			
Am241/U238	+4.0%	+4.7%	±3.2%
Am242m/U238	-32.2%	-31.5%	±12.0%
Am243/U238	**-14.8%**	**-7.7%**	**±4.7%**
Curium			
Cm242/U238	+6.7%	+8.9%	±6.2%
Cm243/U238	-31.8%	-35.2%	±4.9%
Cm244/U238	**-26.5%**	**-19.3%**	**±6.4%**
Cm245/U238	-33.4%	-22.9%	±7.7%
Cm246/U238	-36.9%	-32.9%	±10.2%
Cm247/U238	-48.8%	-35.6%	±13.0%

The present results demonstrate the overall improvements brought about by JEFF3.0 in the prediction of isotopic fuel contents during the irradiation. The increase of the U235 capture resonance integral [6] and the higher Pu241 capture cross-section in the 0.26 eV resonance [7] are the main sources of correction of the prediction of U236-Np237-Pu238 and Pu242-Am243 respectively. The increase of (n,2n) in the JEFF3.0 evaluation of U238 contributes to the improvement of Np237 and Pu238 calculation [8].
The underestimation of Pu239 is not seen in the French experiments and may be explained by the simplified modeling of irradiation and the uncertainties on geometry of the surrounding assemblies. For Pu239 the experimental uncertainty quoted in Table 2 is probably underestimated. The large discrepancies observed in Curium build-up (which are nevertheless very sensitive to the knowledge of Burn-up) are consistent with previous work [5].

The results for the fission products are displayed in Table 3. Note that the large discrepancy observed with JEF2.2 in the Eu154 prediction is removed with the new Europium evaluations introduced in JEFF3.0. The C/E comparison on the absorbing Fission Products is also consistent with previous experimental validation using JEF2.2 [8]. Sm149, Nd143, Sm150 and Sm147 which are neutronic poisons in LWRs are calculated within 3% with both JEF2.2 and JEFF3.0. There is room for improvement in JEFF3.0 for Sm151 and Sm152 which are still overestimated.

Table 3 : C/E-1 in % for fission products with JEF2.2 and JEFF3.0 for the SFP7-4 sample (Burn-up = 45 GWd/t).

Isotope	**C/E-1 JEF2.2**	**C/E-1 JEFF3.0**	**total unc. 1σ**
Neodymium			
Nd143/U238	-1.8%	-3.1%	±1.6%
Nd144/U238	-5.5%	-4.2%	±2.5%
Nd145/U238	0.0% (norm)	0.0% (norm)	±1.3%
Nd146/U238	-1.4%	-1.1%	±1.8%
Nd148/U238	+0.3%	+0.5%	±1.6%
Nd150/U238	-1.1%	-0.7%	±1.8%
Cesium			
Cs134/U238	-4.3%	-4.0%	±3.0%
Cs137/U238	-10.0%	-11.9%	±1.7%
Europium			
Eu154/U238	**+60.1%**	**-10.2%**	**±3.1%**
Cerium			
Ce144/U238	+3.4%	+3.2%	N.E
Ruthenium			
Ru106/U238	+0.9%	+1.6%	N.E
Samarium			
Sm147/U238	-1.3%	-1.5%	± 1.8%
Sm148/U238	-14.4%	-14.0%	N.E
Sm149/U238	+1.3%	-1.2%	± 8.0%
Sm150/U238	-0.2%	+0.2%	± 1.9%
Sm151/U238	+6.2%	+7.2%	± 2.0%
Sm152/U238	+11.5%	+11.5%	± 1.4%
Sm154/U238	+0.7%	+1.5%	N.E

5. Conclusion

This interpretation of the Takahama-3 Post-Irradiation Experiment confirms the major JEF2.2 nuclear data trends derived from French experiments and shows the large improvement provided by JEFF3.0 in the prediction of LWR isotopic fuel contents. The incorporation of JEFF3.0 evaluations within multigroup nuclear library CEA2003V1 contributes towards the improvement of the APOLLO2 neutronic prediction capabilities. Further work will be devoted to the experimental validation of Keff prediction during depletion for UOX and MOX fuels.

Since evaluations from other nuclear data libraries such as JENDL3.3 or ENDF/BVI.8 do not help removing discrepancies on Americium and Curium isotopes, evaluation work and high resolution differential measurements are required to achieve better accuracy on those isotopes. Resonance parameters of Am241, Am242m and Am241→Am242m isomeric ratio need to be investigated in priority.

Acknowledgements

This work was carried out in the framework of the CEA-JUA (Japanese University Association) Co-operation. The authors are indebted to Christine Chabert and Bénédicte Roque from the Commissariat à l'Energie Atomique for contributing strongly to this analysis.

References

1) C. Chabert, A. Santamarina, P. Bioux, "Elaboration and experimental validation of the APOLLO2 depletion transport route for PWR Pu Recycling," PHYSOR-2000 International Top. Meeting on Advances in Reactor Physics and Mathematics, Pittsburg, USA, May 7-11 (2000).
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," PHYSOR-2000 Int. Meeting on Advances in Reactor Physics and Mathematics, Pittsburg, USA, May 7-11 (2000).
3) T. Iwasaki, "Evaluation of Burn-up Calculation Code on Destructive Experiment of Spent PWR fuels," personal communication (2000)
4) S. Loubiere, R. Sanchez, M. Coste, A. Hebert, Z. Stankovski, C. Van Der Guth and I. Zmijarevic, "APOLLO2 Twelve Years Later," Proc. Int. Conf. On Mathematics and Computation, Madrid, Spain, September (1999)
5) C. Chabert, A. Santamarina, "Trends in Nuclear Data Derived from Integral Experiments in Thermal and Epithermal Reactors," International Conference on Nuclear Data, ND2001, Tsukuba, Japan, October 7-12 (2001)
6) L.C. Leal, H. Derrien, N.M. Larson, R.Q. Wright, "R-Matrix analysis of U235 neutron transmission and cross section measurements in the 0 to 2.25 keV Energy Range", Nucl. Sci. and Eng. **131**, pp. 230-253 (1999)
7) H. Derrien, L.C. Leal, A. Courcelle, A. Santamarina, "Re-evaluation and validation of Pu241 resonance parameters from thermal to 20 eV," Submitted to Nucl. Sci. and Eng.
8) A. Courcelle, C. Chabert, O. Litaize, B. Roque, A. Santamarina, O. Serot, "Experimental Validation of Main Fission Product and Actinide Nuclear Data. Improvements for JEFF," PHYSOR-2002, Int. Conf. on Reactor Physics, Seoul, Korea, October 7-10 (2002).