ELABORATION AND EXPERIMENTAL VALIDATION OF THE APOLLO2 DEPLETION TRANSPORT ROUTE FOR PWR PU RECYCLING

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ABSTRACT

In order to optimise the fuel cycle and to save uranium resources, France decided in June 1985 to recycle in PWRs the Plutonium produced in standard Nuclear Power Plants. Currently, sixteen 900 MWe PWRs are loaded with 30 % Mixed-Oxide (MOX) assemblies. The programme started in November 1987 with the first loading of 16 MOX assemblies in the Saint-Laurent B1 (SLB1) reactor.

Several neutronic parameters are significantly modified in this case, and the associated calculation uncertainties are increased when Plutonium is introduced in a UO_2 core.

So we have paid an important effort to the elaboration and the experimental validation of the depletion transport route for PWR Pu recycling using the French neutronic code APOLLO2.

This paper describes this study which has been carried out by the Commissariat à l'Energie Atomique (CEA) and has been supported by the French utility Electricité de France (EdF) and the French nuclear plant designer Framatome.

1. INTRODUCTION

Since the mid 80's, the French Atomic Commission (CEA) has developed a new lattice code for PWRs in the framework of a co-operation between the EDF and the French Designer Framatome. This code, called APOLLO2 /1/ is a user-friendly code and is completely modular. Nuclear Data libraries, called CEA93, were built using the JEF2.2 evaluations /2/. Specific improved modules for collision probability calculations or self-shielding models were implemented in the code and allow to investigate the physical phenomena in various types of situations.

An extensive programme of validation of APOLLO2 and its associated libraries involving the major neutronic parameters of Light Water Reactors was established. This paper presents the definition and validation of the APOLLO2 depletion transport route for PWR Pu recycling. Pu fuel introduced in a UO_2 core causes important local heterogeneities of the flux which is difficult to simulate. Owing to these strong UOX/MOX differences, mixed loading core designs and analyses have to be perform using specific calculational routes which must be able to treat accurately the physical phenomena occuring in such reactors.

Consequently, calculation schemes based on state-of-the-art methods in neutron transport must be used.

Nevertheless, complex and expansive schemes are not practical in design calculations and, to ensure reasonable execution time, approximations must be done.

The process used to define the APOLLO2 « reference » calculational route for MOX assemblies consists of investigating successively the different physical and numerical approximations made. The modelling errors are assessed by comparison with reference calculations, for example a TRIPOLI4 Monte-Carlo calculation /3/.

The APOLLO2 calculation route is defined by selecting the code options which yield known and acceptable errors. In this way, we obtain a 2D-APOLLO2 depletion transport route which allows accurate core calculations.

It addresses both local (pin by pin) and global problems, with a 1 % accuracy for Pu239 depletion prediction.

In the second section, we present the work done to establish the APOLLO2 « reference » calculational route. The third section presents the experimental programme used to validate the APOLLO2 depletion transport route for PWR Pu recycling.

In the fourth part, we compare the results obtained using the previous design route and our new reference route.

This work permits us to obtain tendencies on nuclear data. This information is summarised in the last section.

2. DEFINITION OF THE APOLLO2 DEPLETION TRANSPORT ROUTE FOR PWR PU RECYCLING

Stochastic resolution gives a reference solution of the problem, but a Monte Carlo code cannot yet be used in a LWR design route. Instead, we have to use approximate deterministic methods, allowing a faster resolution. CEA has developed a new lattice code for LWRs.. This modular code, called APOLLO2, allows heterogeneous assembly calculations using the Pij collision probability method, and core calculations using discrete ordinate techniques. APOLLO2 uses the CEA93 library, which was processed from the JEF2.2 evaluations. We used the European X-MAS 172 group-structure well suited for Oxygen elastic scattering resonance and self-shielding of Pu isotopes /4/.

The process implemented to define the APOLLO2-based « reference » calculation route for MOX assemblies assesses the different physical approximations by comparison with reference calculations using the same nuclear data file JEF2 : TRIPOLI4 Monte Carlo continuous-energy calculation or actual geometry Pij calculation in APOLLO2, i.e MARSYAS module for exact 2D Pij or TDT module for general geometry /5/.

To establish the recommended models and methods, we investigated different issues such as :

1) <u>The spatial treatment of the MOX assembly</u>

In PWR Pu recycle, the core is loaded with 30 % MOX assemblies. Therefore, a MOX assembly is surrounded by UOX assemblies. It is necessary to account for UO_2 environment in neutronics pattern : UOX surrounding provides a thermal neutron current towards the MOX assembly. Thus, the fission rate in boundary rods is increased, which requires a zoning of the MOX assembly. We have to notice that standard calculation based on infinite medium pattern for MOX assembly does not handle the peaking factor at the boundary. The error can attained 30 % in Pu239 concentration at 40 GWd/t for rods located at the peripheral zone. So, the geometrical model in APOLLO2 route represents one half MOX assembly surrounded by half UOX assemblies with reflective conditions (1 MOX per 3 UOX), as shown in Figure 3.

Another issue is the definition of the best multicell method to calculate the lattice. The Pij calculation in the exact geometry can be performed by MARSYAS model. However, this calculation is expensive, therefore design depletion calculation cannot be based on such exact 2D resolution. Then, it is necessary to investigate interface current Pij calculations. Two main options of interface current method have been tested here : UP0 (isotropic angular flux assumption) and UP1 (linear anisotropic flux model). The results obtained with UP1 model are much more accurate than UP0 model, as shown in Figure 1 and Figure 2. The discrepancies associated with UP0 model amounts to 15 % in MOX assembly, instead of 1 % in UP1 model.

In order to improve computing efficiency, it is necessary in depletion calculation to optimise flux calculation discretization. The APOLLO2 multicell pattern enables the grouping of cells which have similar flux level (‘physical cell’). We integrated the inter-assembly water gap inside the neighbouring fuel cells, which leads to rectangular physical cells. The optimised multicell geometry is shown in Figure 3.

To represent space dependent self-shielding and nuclide concentration profiles in the pellet, we subdivide the fuel pin into four radial rings. We use thin peripheral shells, 5 % of the mass of the fuel pellet at the outer ring, in order to account for the rim effect.

2) <u>Self-shielding calculation</u>

Isotopes considered for self-shielding are : U238, Pu239, Pu240, Pu241, Pu242, U235 and U236. We pointed out that Pu238 and Am241 self-shielding is not required in the 172-group structure, as pointed out in Table 1 and Table 2.

The Background Matrix formalism allows space-dependent self-shielding calculation. For a strong resonant isotope, such as U238, this method allows the self-shielding differentiation in the various fuel pins and concentric pellet rings.

It was demonstrated that 4 rings are needed to account for the space dependence of U238 self-shielding. Furthermore, it was pointed out that 3 different rod types (one for each Pu enrichment zone) are required for Pu239, Pu240, Pu241 and Pu242 self-shielding ; the efficiency of this self-shielding grouping is shown in Figure 4 for Pu240, where the bias in the resonant capture rate is limited to 0.09 % in any MOX fuel rod. Moreover, 2 different self shielded rods (in front of the water holes and at the angle) are needed for U238 to account for the difference in Dancoff effect. On the other hand, only one rod type is requested for U235 and U236 (averaged self-shielding).

Reference continuous-energy computation pointed out that accurate resonance absorption calculation requires 2D-Pij /6/ : we use sophisticated UP1 model for U238 (cf. Table 3), however UP0 model is satisfactory for other isotopes, even for Pu240 E>4eV (cf. Table 4).

Figure 1 - F_{tot} - UP0 ROTH/Pij 2D bias (in %) -

											-0.12
										0.88	-0.12
									1.37	0.48	0.02
								0.95	1.37	0.48	0.02
							4.24	2.55	0.80	0.88	-0.12
						7.37	5.73	3.36	-	-	-
					10.50	8.61	6.68	3.92	-	-	-
				13.20	11.68	9.68	7.31	4.36	-	-	-0.12
			15.69	14.35	12.58	10.55	7.75	4.47	-	-	-1.21
		17.49	16.51	15.33	13.42	11.01	8.27	4.76	-	-	-0.12
	18.56	17.99	17.01	15.75	13.85	11.34	8.47	4.87	-	-	-0.12
18.80	18.63	18.11	17.26	15.78	13.90	11.53	8.33	4.80	0.80	0.88	-1.21

Figure 2 - F_{tot} - UP1/Pij 2D bias (in %) -

											0.08
										0.26	0.08
									0.24	0.40	0.36
								0.11	0.24	0.40	0.36
							-0.34	-0.02	0.06	0.26	0.08
						-0.55	-0.29	-0.14	-	-	-
					-0.77	-0.65	-0.30	-0.13	-	-	-
				-0.66	-0.66	-0.61	-0.32	0.02	0.06	0.26	0.08
			-1.16	-0.81	-0.80	-0.85	-0.48	-0.16	-	-	-0.18
		-0.86	-0.93	-0.70	-0.72	-0.68	-0.30	-0.08	-	-	0.08
	-0.88	-0.88	-0.93	-0.77	-0.72	-0.68	-0.31	-0.11	-	-	0.08
-1.30	-0.99	-0.96	-1.16	-0.91	-0.81	-0.89	-0.56	-0.30	0.06	0.26	-0.18

Figure 3 - The optimised multicell geometry

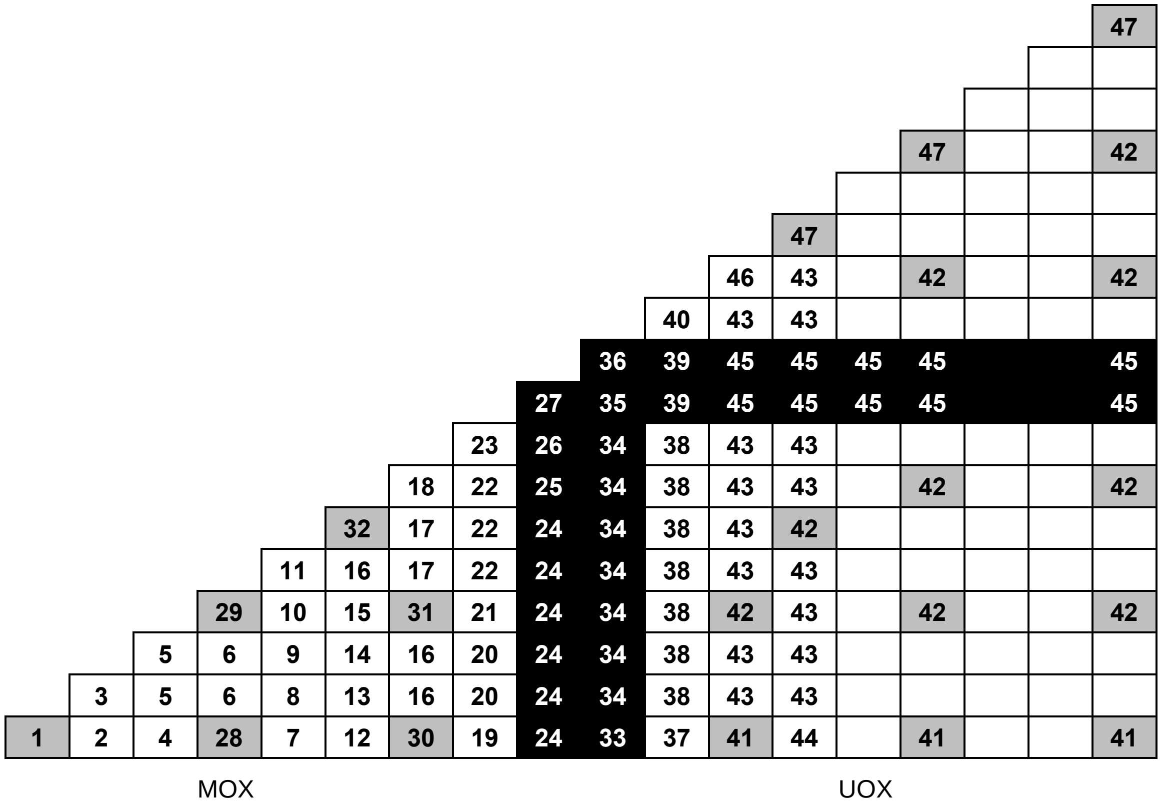


Table 1 - Pu238 self-shielding effect on Pu concentrations (in %)

	10 GWd/t	30 GWd/t	60 GWd/t
Pu238	+1.73E-02	+5.17E-02	+1.04E-01
Pu239	-8.85E-05	+1.10E-04	-3.82E-03
Pu240	-1.53E-03	-5.45E-03	-2.58E-03
Pu241	+2.14E-03	+1.22E-03	-1.93E-03
Pu242	-1.48E-03	+1.14E-03	-3.29E-03

Table 2 - Am241 self-shielding effect on Actinide concentrations (in %)

	6 GWd/t	18 GWd/t	60 GWd/t
Am241	0.05	0.12	0.14
Pu242	0.00	0.00	0.00
Am242m	-0.18	-0.08	0.02
Cm242	-0.18	-0.10	0.01

Table 3 - U238 self-shielded cross section versus Pij model

Pij model	Capture X-section (barns)	Bias against Pij 2D (in %)
Pij 2D exact	2.4226	
UP1 HETE	2.4292	+0.27
UP0 ROTH	2.4453	+0.94

Table 4 - Pu240 self-shielded capture versus Pij model

Pij model	Capture X-section (barns)	resonant capture rate
Pij 2D exact	115.44	9.2001E-02
UP1 HETE	115.46	9.2015E-02
UP0 HETE	115.48	9.2037E-02
UP0 ROTH	115.45	9.2010E-02

Figure 4 - Bias in Pu240 capture using only 3 self-shielded rod types (in %)

											-
										-	-
									0.00	0.00	-0.00
								-0.08	0.00	0.00	-0.00
							-0.06	0.01	-	-	-
						0.04	-0.01	0.02	-	-	-
						0.09	-0.01	-0.00	-	-	-
				-0.02	0.08	0.05	-0.02	0.01	-	-	-
				-0.03	-0.05		0.04	-0.01	-	-	
		0.03	-0.02	0.02	0.02	0.12	-0.01	0.00	-	-	-
	0.03	0.04	-0.02	0.03	0.01	0.09	-0.02	0.02	-	-	-0.00
	-0.02	-0.02		-0.02	-0.03		0.04	0.01	-0.00	-0.00	

3) Depletion calculation

In APOLLO2, we consider a detailed burn-up chain with 20 actinides (U234 $\rightarrow$ Cm247) and 85 fission products.

It was shown that Actinide self-shielding must be re-calculated at 4 000, 8 000 , 12 000, 24 000, 38 000 MWd/t ...

3. EXPERIMENTAL VALIDATION OF THE APOLLO2 DEPLETION TRANSPORT ROUTE

Irradiations in SLB1 Reactor

The French Atomic Energy Commission (CEA), Electricité de France (EdF), and Framatome are jointly performing an extensive experimental programme of fuel irradiations and analyses in French power reactors. This programme has a high priority because it provides direct results concerning spent fuel characteristics such as nuclide inventory, activities and residual power in actual reactor operating conditions. The APOLLO2 reactivity loss with burnup is validated through spent fuel reactivity worth measurements by the oscillation technique in the MINERVE reactor /8/.

So, to validate our depletion transport route, chemical analyses of French PWR spent fuel provide a powerful tool with broad scope. These assays have been extended to high burnups, and MOX fuels with the addition of data from the SLB1 assemblies.

The SLB1 reactor, a 900 MWe PWR, is characterised by a 30 % MOX fuel loading. MOX assemblies 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 characterised by a high Pu content (5.6 %) and the peripheral zone by a small Pu content (2.9 %). These MOX fuel assemblies have been irradiated in SLB1 from 1987 to 1995. Figure 5 shows the MOX assembly with the different Pu zones and guide-tubes.

Two MOX fuel assemblies (FFPOOJJX and FFPOOMJX) were selected for the SLB1 programme. The MOX rods of interest belonging to those two assemblies were extracted and analysed after 1, 2 or 3 reactor cycles, with burnups ranging from about 10 to 45 GWd/t.

Figures 6 and 7 show the MOX rod positions in FFPOOJJX and FFPOOMJX assemblies, respectively.

Analyses have been performed to determine the concentrations in U, Pu, Am, Cm, Np isotopes, as well as Nd and Cs. Generally, the analytical results are obtained by mass spectrometry measurements. An experimental determination of burnup is deduced for each sample, either from the Nd148/U238-Nd145/U238-Nd250/U238 ratios, or from the Cesium ratio measured by gamma spectrometry.

Figure 5 - MOX assembly zoning

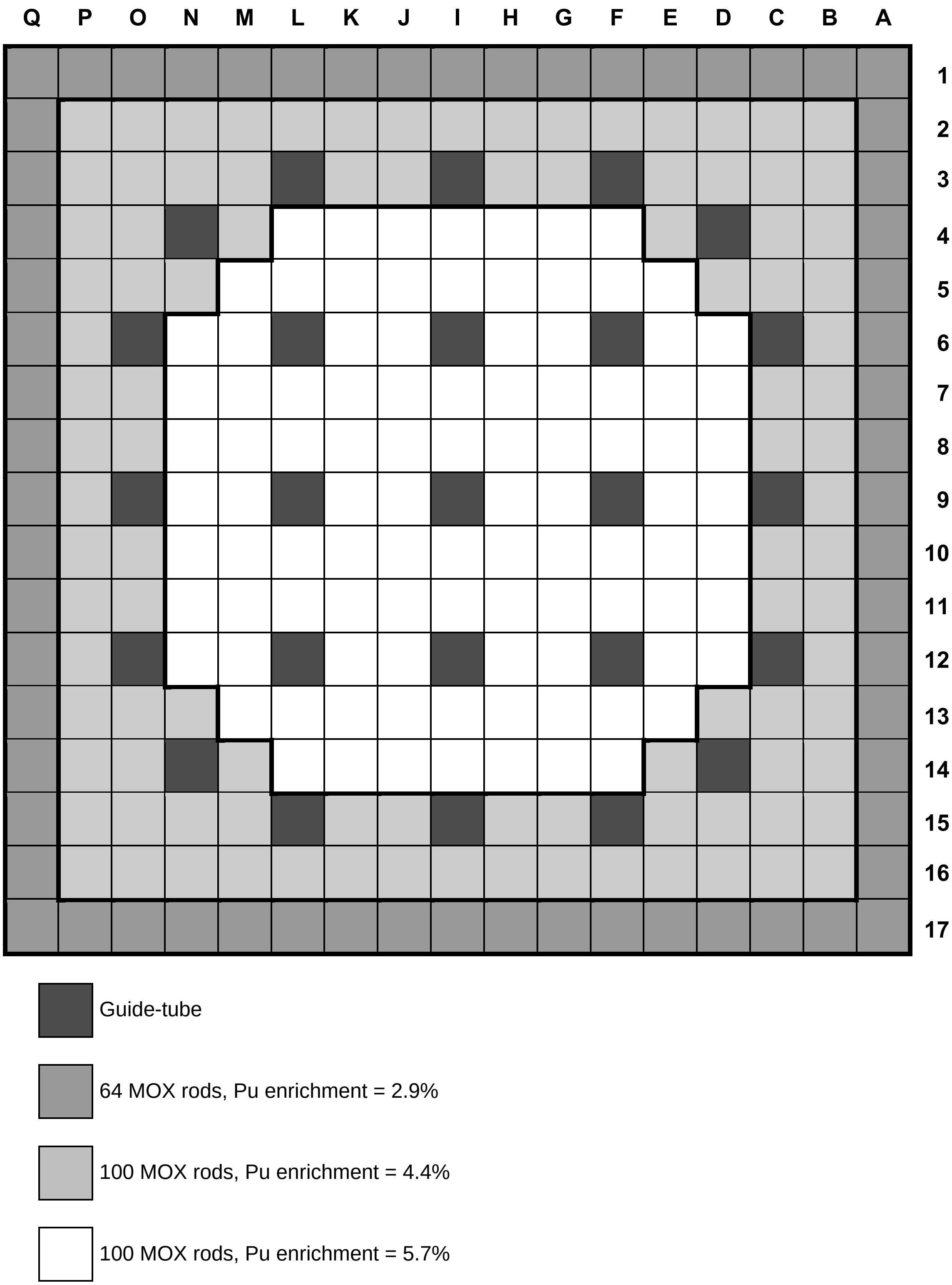
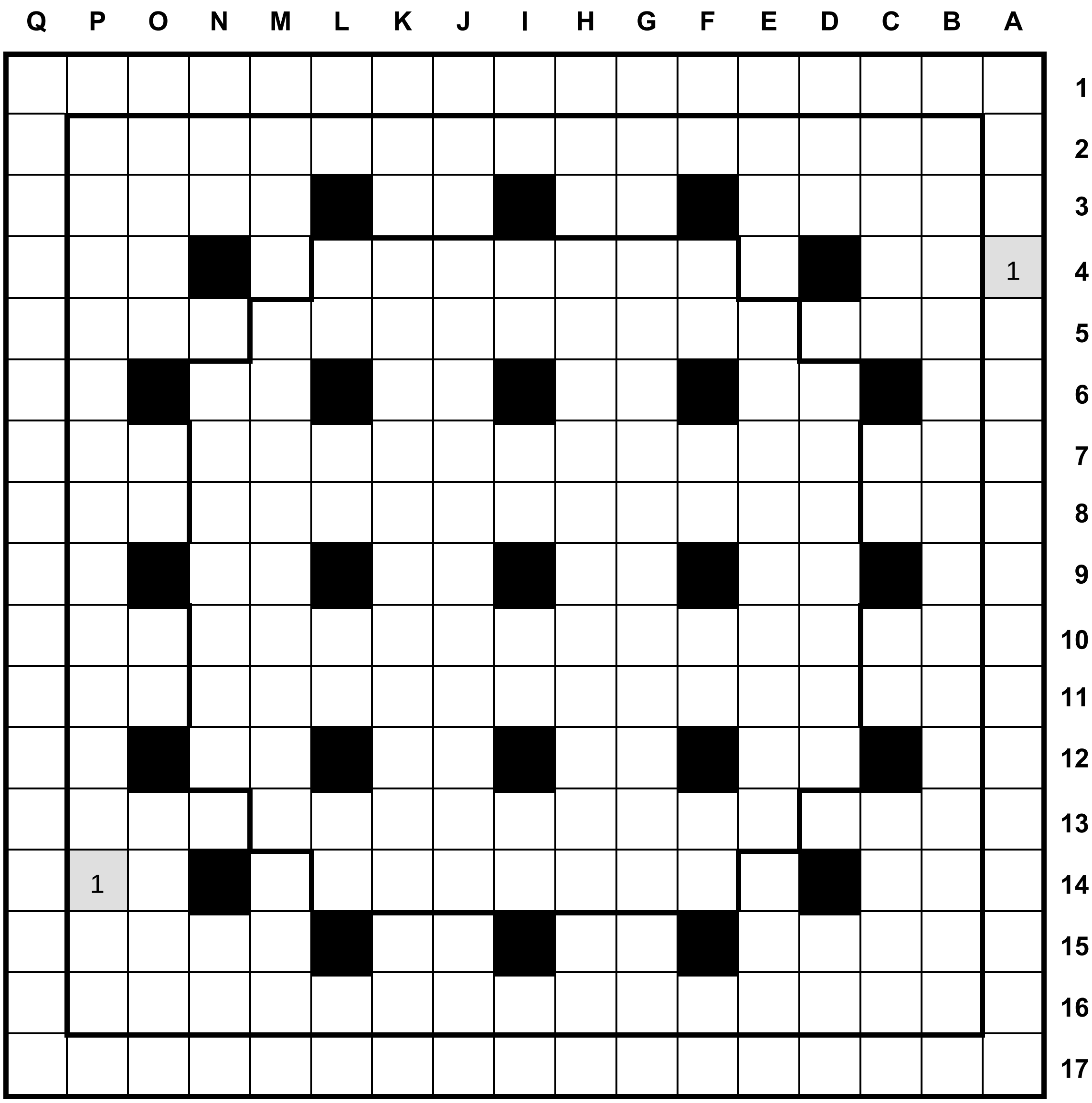
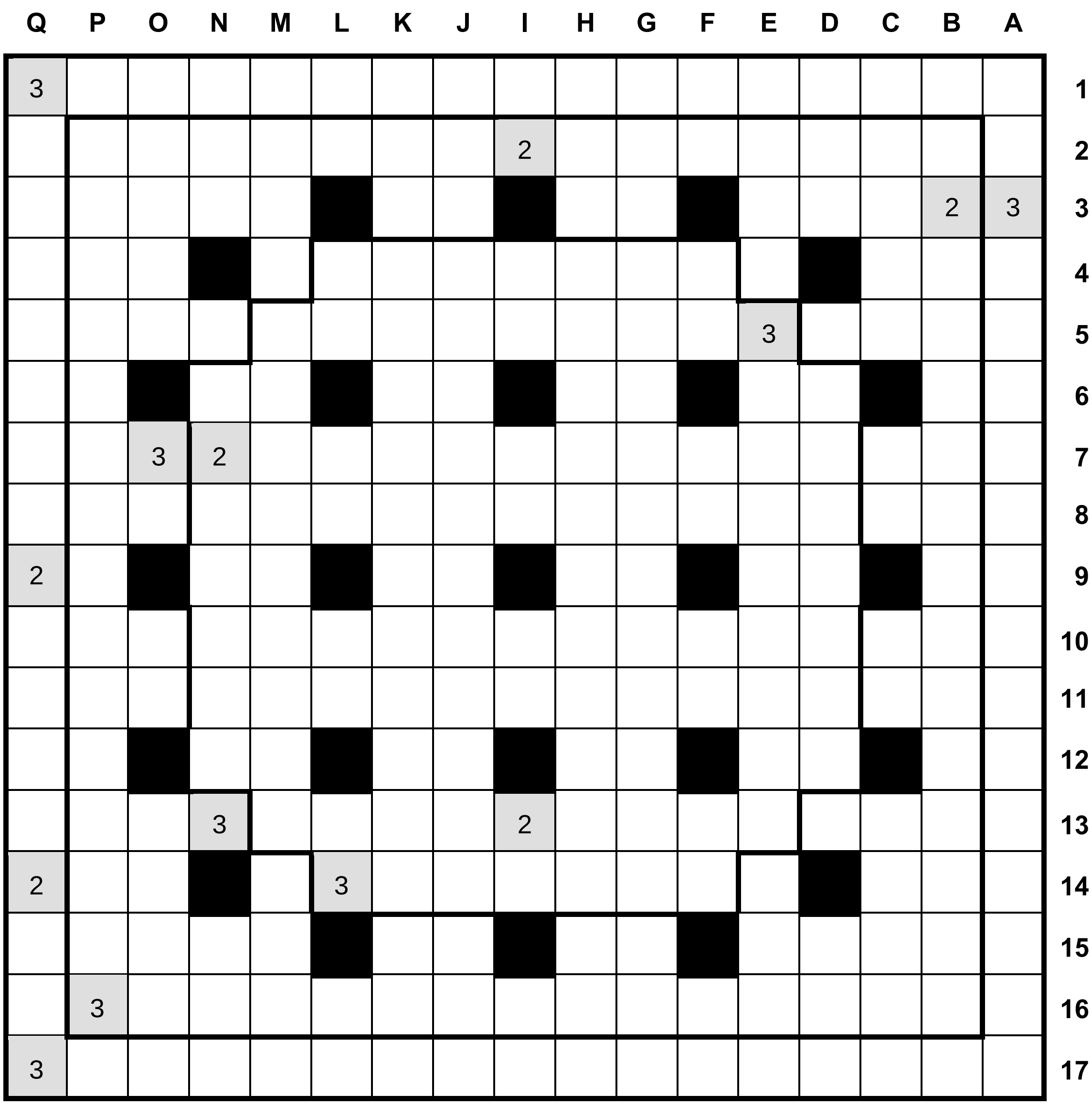


Figure 6 - SLB1 - Assembly FFP00JJX



N Fuel rod extracted after N irradiation cycles

Figure 7 - SLB1 - Assembly FFP00HJX



N Fuel rod extracted after N irradiation cycles

4. COMPARISON BETWEEN PREVIOUS DESIGN ROUTE RESULTS AND NEW REFERENCE ROUTE RESULTS

The Experiment-Calculation comparison (C/E - 1 in %) for some spent fuel rods (mid-height), derived from the APOLLO2 analysis of SLB1 programme, is summarised in Table 5. We compare the results obtained using the previous design route and the new reference route.

Table 5. Calc/Exp discrepancy (in %) using design route and reference route.

	Previous design route	Reference route	Previous design route	Reference route	Previous design route	Reference route
Fuel rod position	I02 - Interm. zone		Q17 - Periph. zone		P16 - Interm. zone	
Cycle	2		3		3	
fuel rod Burnup (MWd/t)	28 453		37 683		42 013	
U235/U238	+ 2.59	+ 2.92	+ 18.21	+ 5.79	+ 7.88	+ 2.04
Pu239/U238	+ 5.90	+ 1.54	+ 27.16	- 1.28	+ 20.76	- 0.23
Pu240/U238	- 2.72	+ 0.01	+ 8.33	+ 4.07	+ 0.25	+ 1.14
Pu241/U238	+ 1.46	- 3.22	+ 13.79	- 3.86	+ 7.66	- 3.17
Pu242/U238	- 5.14	- 4.60	- 10.54	- 5.73	- 9.53	- 4.87
Am241/U238	+ 4.79	- 0.09	+ 19.40	- 1.84	+ 7.77	- 3.35
Cm245/U238	+ 15.46	- 13.00	+ 20.83	- 14.44	+ 24.31	- 13.67

Because of the UO_2 environment representation, the reference route provides a good prediction of the Major and Minor Actinides in any MOX fuel pin, even in the corner of the assembly such as Q17 and P16 pins.

The satisfactory agreement observed on U235 depletion demonstrates that thermal flux map is well mock-up in the MOX assembly by the APOLLO2 reference route.

An appreciable improvement in the calculation of Pu239 build-up is also raised with the new route, due to the UO_2 environment and to the use of the accurate UP1 model (improvement in U238 self-shielding calculation).

The Pu240 concentration is also well predicted in our reference route. The CEA93 library is used in both routes, however the use of a 99-group structure in the previous route induces an inaccurate capture rate in the large 1 eV resonance. With twice groups in this resonance, the X-MAS 172-group structure of the reference route is more suited.

The reference route also improves the C/E comparison for Am and Cm isotopes. Hence, the Cm244 and Cm245 build-up is now slightly underestimated, consistently with the Pu242 trend.

Therefore, the reference route provides a good prediction of the local spent fuel inventory, particularly for the Pu239 major actinide. It is currently recommended in PWR mixed core calculations based on the APOLLO2 code.

5. DETAILED RESULTS OF SLB1 IRRADIATION USING THE APOLLO2 REFERENCE ROUTE - DERIVATION OF TRENDS

The APOLLO2 reference route results for the SLB1 irradiation are reported in Table 6 for the major actinides and in Table 7 for the minor actinides.

The burn-up is deduced from an average on the fluency indicators, mainly Nd isotopes build-up. The experimental uncertainty on burn-up normalisation amounts to 1.5 % in one standard deviation, mainly due to fission yields (1 %) and F.P isotopics (0.5 %).

For the perturbed rods near the UO_2 zone, like P16 that is in a corner, the satisfactory results indicate that the calculation route which takes into account the MOX-UO_2 interface, is adequate.

Table 6 - SLB1 evaluation with APOLLO2 Reference Route - Deviations (C/E)-1 in %
Major Actinides

	Rods located in Asymptotic neutron spectrum				
Fuel rod position	P14	I13	I02	N13	L14
Cycle	1	2	2	3	3
Zone	Intermediate	Central	Intermediate	Intermediate	Central
Fuel rod burnup (MWd/t)	12 868	28 368	28 453	41 493	45 005
U234/U238	- 4.42	-11.52	- 6.87	- 4.87	- 9.41
U235/U238	1.13	1.89	2.92	4.70	4.28
U236/U238	- 10.13	- 7.10	- 8.24	- 7.84	- 6.45
Pu238/U238	- 8.10	- 9.05	- 8.56	- 6.17	- 8.57
Pu239/U238	0.57	3.98	1.54	4.72	4.13
Pu240/U238	- 0.45	0.62	0.01	1.91	1.28
Pu241/U238	- 2.20	- 1.25	- 3.22	- 1.42	-1.83
Pu242/U238	- 2.19	- 3.37	- 4.60	- 6.21	- 5.64

	Rods placed in Peripheral zone			
Fuel rod position	A04	Q14	Q17	P16
Cycle	1	2	3	3
Zone	Peripheral	Peripheral	Peripheral	Peripheral
Fuel rod burnup (MWd/t)	9 556	24 664	37 683	42 013
U234/U238	15.75	1.12	2.09	- 3.94
U235/U238	1.20	3.96	5.79	2.04
U236/U238	- 7.76	- 8.00	- 5.63	- 5.18
Pu238/U238	- 9.78	- 8.33	- 7.56	- 6.76
Pu239/U238	2.97	3.40	- 1.28	- 0.23
Pu240/U238	1.05	2.75	4.07	1.14
Pu241/U238	- 6.97	- 2.87	- 3.86	- 3.17
Pu242/U238	- 3.06	- 5.85	- 5.73	- 4.87

The following trends for the major actinides are observed :

- U234
  It is difficult to conclude because of measurement uncertainties due to negligible amount of this isotope in the depleted uranium matrix of the MOX fuel.

- U235, U236
  U235 is over-estimated with JEF2 evaluation and U236 build-up is underestimated. Different works have shown that the underestimation of the U235 capture resonance integral in JEF2 and ENDF/B6.4 was responsible for this effect to a large extent /7/. The new Leal-Derrien-Larson implemented JEFF3.0 and ENDF/B6.5 cancels this C/E discrepancy

- Pu238
  We observe a deviation of about - 8 %. This isotope is present from the beginning of the irradiation. Its formation is associated with the Np237 capture (after U236 capture), and mainly with Cm242 decay in MOX fuel. The trend for Pu238 is being investigated and might be caused by the underestimation of U236 and Cm242 build-up.

- Pu239, Pu240, Pu241
  An appreciable improvement in the calculation of Pu239 concentration can be observed with the new scheme.
  The Pu240 calculation is also well predicted : this satisfactory result confirms the accuracy of the 1 eV resonance Doppler broadening and of the thermal self-shielding method used in the APOLLO2 reference route.
  The Pu241 is also well predicted. So, Pu239 (0.3 eV resonance) and Pu240 capture cross sections are satisfactory.

- Pu242
  This isotope is underestimated. We observe a deviation of - 5.6 % after 45 GWd/t. This could indicate that the (n,γ) cross-section of Pu241 is too low.

Among the minor actinides :

- Am241
  It is quite well predicted for a measurement after 2 years cooling time. It follows the evolution of its parent nuclide, Pu241. Its prediction is very sensitive to the modelling of the reactor irradiation history.

- Am242m, Cm243
  They are underestimated. To conclude on Cm243, we need information on Cm242, which was not available here. The Am242m results could indicate that the Am241 branching ratio toward this isotope is underestimated.

- Am243
  The agreement is good, so we can conclude that the Pu242 capture is correct, particularly in the large 2.7 eV resonance.

- Cm244
  Cm244 is underestimated by about 10 %, as a consequence of the Am243 low underestimation.

Table 7- SLB1 evaluation with APOLLO2 Reference Scheme . Deviations (C/E)-1 in % Minor Actinides

	Rods located in Asymptotic neutron spectrum				
Fuel rod position	P14	I13	I02	N13	L14
Cycle	1	2	2	3	3
Zone	Intermediate	Central	Intermediate	Intermediate	Central
Fuel rod burnup (MWd/t)	12 868	28 368	28 453	41 493	45 005
Am241/U238	- 8.28	0.85	- 0.09	1.31	- 1.62
Am242m/U238	- 38.64	- 32.50	- 28.70	- 21.40	- 22.85
Am243/U238	- 15.20	- 6.61	- 7.81	- 7.10	- 6.11
Cm243/U238	- 23.35	- 29.30	- 21.47	- 14.35	- 17.78
Cm244/U238	- 17.99	- 7.90	- 9.69	- 6.40	- 7.08
Cm245/U238	- 16.01	- 7.93	- 13.00	- 6.36	- 8.03

	Rods placed in Peripheral zone			
Fuel rod position	A04	Q14	Q17	P16
Cycle	1	2	3	3
Zone	Peripheral	Peripheral	Peripheral	Peripheral
Fuel rod burnup (MWd/t)	9 556	24 664	37 683	42 013
Am241/U238	- 5.90	1.28	- 1.84	- 3.35
Am242m/U238	- 36.22	- 25.89	- 23.44	- 28.75
Am243/U238	- 9.93	- 13.71	- 8.04	- 12.04
Cm243/U238	-	- 19.55	- 18.76	- 14.68
Cm244/U238	-	- 10.57	- 9.63	- 9.19
Cm245/U238	-	- 14.48	- 14.44	- 13.67

CONCLUSION

This work is devoted to the improvement of the APOLLO2 depletion calculation route, and its associated library CEA93, for Pu recycling in PWR mixed cores.

In the first step, we have selected the best suitable methods and models for simulating the complex physical heterogeneous met in PWR Pu recycling. This new route was checked against reference results like TRIPOLI4 M.C continuous energy, exact Pij 2D and Sn calculations, with specific concern for computing efficiency improvement :

- it was demonstrated that it is necessary to account for the UO_2 environment
- to reproduce reference results, we must use accurate UP1 interface current model
- recommendations for mutual and self shielding calculation were also established.

Both local and global problems were found to be solved by this 2D-APOLLO2 depletion transport route allowing accurate core calculations, such as for instance 1 % accuracy for Pu239 depletion prediction.

In the second step, we have started the qualification process of this route with the analysis of the SLB1 experimental programme. The satisfactory results confirmed the quality of the theoretical approach and the capability of the APOLLO2 calculation to reproduce the fuel inventory at the same accuracy level that found in standard UOX assemblies /9/ : for example +2% $\pm$ 2% and +1% $\pm$ 2%(1σ) respectively for Pu239 and Pu240 at 40 GWd/t.

In the next future, the qualification will be extended to high burnups, i.e 65 GWd/t, with chemical assays performed in MOX assemblies irradiated 4 and 5 cycles in French PWRs.

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