

**QUALIFICATION OF THE APOLLO 2 ASSEMBLY CODE
USING PWR-UO₂ ISOTOPIC ASSAYS.**

**THE IMPORTANCE OF IRRADIATION HISTORY AND THERMO-MECHANICS ON
FUEL INVENTORY PREDICTION**

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ABSTRACT

The work presented in this paper is part of the qualification procedure of the APOLLO 2 neutronic code for calculating the depletion of 17x17 UO₂ PWR assemblies. The fuel inventory calculation was qualified through the analysis of fuel irradiated for 2 and 3 cycles in BUGEY 3 (3.9% enriched - maximum burnup fraction : 38.3 GWd/t_m) and fuel irradiated for 2, 3, 4 and 5 cycles in Gravelines (4.5% enriched - maximum burnup fraction : 60 GWd/t_m).

The determination and qualification of the APOLLO 2 calculation process for depletion, is an important stage in reproducing an accurate inventory of the fuel. This work also identifies the need to precisely describe the irradiation history (fuel temperature, moderator temperature, boron control, power).

The trends observed through the BUGEY 3 programme and the Gravelines programme are consistent and demonstrate the ability of APOLLO 2 to accurately reproduce the fuel inventories for 17x17 UO₂ PWR assemblies : the concentrations of the main actinides U₂₃₅, Pu₂₃₉, Pu₂₄₀ are reproduced with greater than 2% accuracy.

INTRODUCTION

The validation-qualification process for the APOLLO 2 calculation code [1] and its associated library of effective cross-sections CEA 93 [2] is a major task. The qualification, a point which must be stressed here, involves comparing the results of the "Industrial code + nuclear data library + calculation route" combination with the measured values. The qualification using experimental results, enables the error of the tool to be calibrated for calculating various parameters such as the initial reactivity, the loss of reactivity per cycle, the efficiency of the soluble boron, the material inventory... Similarly, the follow-up of PWR reactors on the basis of APOLLO 2 provides a guarantee of the overall quality of the major core calculations in operating conditions. This qualification process also enables any errors to be detected with respect to the nuclear data and the processing of the evaluations, and makes it possible to identify the trends in the CEA 93 multigroup library.

Among the PWR project parameters to be qualified, one is the subject of this study : the evolving material inventory for UO₂ PWR assemblies. To do this, there is a large amount of French experimental data : the analyses of irradiated fuels. This data base, which provides a wealth of information for qualification purposes, is regularly augmented with new experimental results. Amongst these, we selected two programmes concerning 17x17 PWR fuel for the qualification of the UO₂ material inventory : BUGEY 3, which is representative of the standard French nuclear power reactors, and Gravelines for high burnups. The description of these programmes is the subject of the first part of this paper. It then deals with the thermo-mechanical aspects of the problem, the choices and recommendations made in particular for the modelling of the irradiation process.

This qualification task cannot be performed correctly without first defining the APOLLO 2 calculation route. This definition is established by comparison with the "master" calculation results obtained using for example the TRIPOLI-4 code [3], also developed at the CEA. TRIPOLI-4 is a continuous energy Monte-Carlo code, and APOLLO 2 is a modular code which involves both the integral equation and the integro-differential transport equation. The APOLLO 2 datum calculation route for evolving 17x17 UO₂ assemblies previously developed and currently recommended, is presented in this paper.

In the second part, the results of the interpretation of the isotopic analyses of irradiated fuels are presented together with the major trends identified and their associated degrees of error.

The combination of this work enables the 'APOLLO 2 / CEA 93 / calculation route' to be calibrated on the fuel inventory up to 60 GWd/t_m in the calculation of major and minor actinides in 17x17 UO₂ PWR.

THE EXPERIMENTAL DATA BASE OF IRRADIATED PWR FUELS

The spent fuel analyses form the basis for the qualification of the calculations for the neutronic evolution in the fuel [4]. Additionally, these analyses constitute a data base which provides a wealth of information for the qualification of basic nuclear data. The chemical assays of irradiated fuel provide information on the integrated reaction rates involved in depletion chains and on isotopes formed only during the irradiation process. The interest of this is therefore highly complementary to the experimental zero power reactor studies. It is possible, for example, to obtain information on the capture of U_{238} , through a critical experiment, whereas a complete study of the build up of Pu_{239} , necessarily involves studying irradiated fuels.

The French experimental data base for spent fuels, which is used for the qualification of neutronic codes, covers a wide range of irradiation situations and of types of fuels. This data base extends from FBR to PWR via BWR. With respect to PWR, two programmes are used primarily for the qualification of APOLLO 2 depletion calculation, these are described below.

The BUGEY 3 experiment

BUGEY 3 is a 900 MWe pressurised water reactor. The experimental programme which was conducted on it was for the first time representative of the French nuclear power reactors. It involves standard fuel, the assembly consists of 17x17 pins clad in zircaloy 4, with inconel spacer grids.

The programme covers two assemblies with removable pins. The first, FGA54, with 2.1% initial enrichment was irradiated for a start cycle and the second, FGC53, with 3.1% initial enrichment was irradiated for 3 cycles.

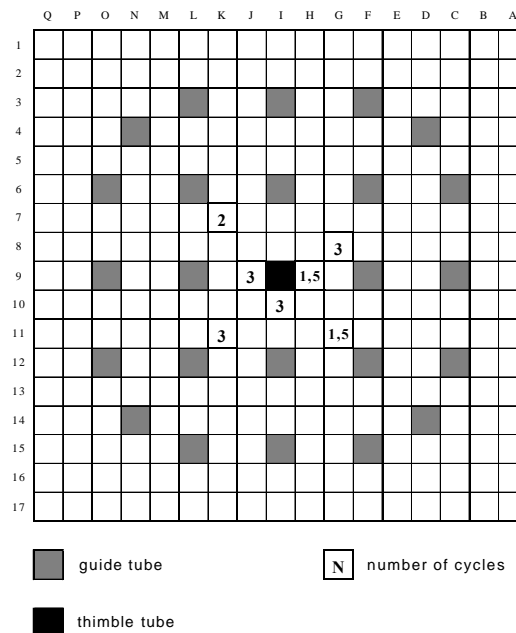


Figure 1. Extracted pins in the FGC53 assembly

The essential part of the neutronic experimental programme was conducted on the FGC53 pin assembly. The positions of the pins extracted from this assembly for chemical analysis, are represented in Figure 1.

The GRAVELINES experiment

This experiment, which is part of a vast EDF - Framatome - CEA co-operative programme, is devoted to the study of high burnup irradiated fuels in the French nuclear power reactors. This programme is dedicated to the extension of the calculation route qualification to the high burnups (5 irradiation cycles) and high enrichment values (4.5% initial enrichment). The fuel loaded in this reactor is type AFA; the essential difference relative to "standard" assemblies being the composition of the spacer grids. These are made of zircaloy, only the pin retaining springs being made of inconel (whereas in standard assemblies, they are entirely made of inconel).

The irradiated fuel programme covers 2 assemblies. The pin extractions were performed after 2, 3, 4 and 5 irradiation cycles, in the central zone of removable pins. The positions of the pins extracted for chemical assays are shown in Figures 2 and 3.

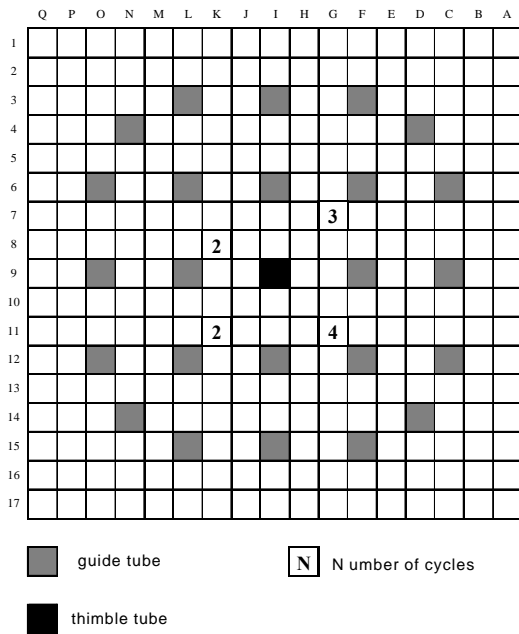


Figure 2. Extracted rods in the 4.5% enriched FF06E2BV assembly.

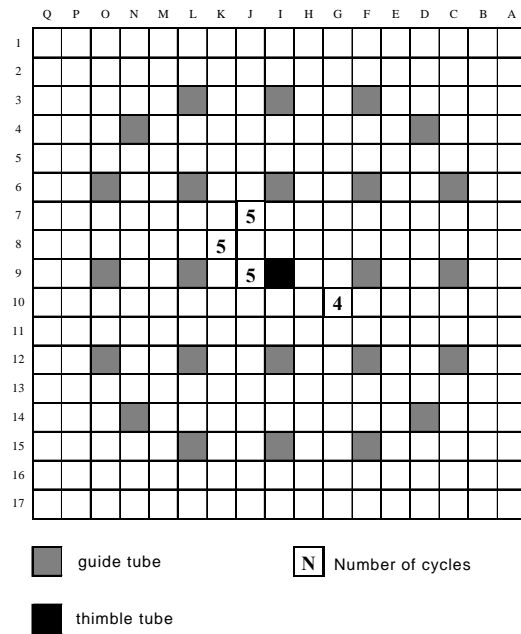


Figure 3. Extracted rods in the 4.5% enriched FF06E3BV assembly.

MODELLING OF THE IRRADIATION AND TEMPERATURE HISTORIES

Several studies have shown that the quality of calculation results is also dependent on the modelling of operating parameters [4] and [5].

For example, an important parameter is the temperature variation of the fuel during irradiation, and its radial profile within the pellet. Other essential parameters are the variation in boron concentration during irradiation, the cooling time, the stretch-out, the operating power, the moderator temperature.

1) Operating power

One of the parameters which must be specified for the APOLLO 2 evolution calculation, is the operating power, for which the calculated flux is normalised. For the cycle monitoring in APOLLO 2 calculation, the parameter retained with respect to the history record provided by EDF, is the irradiation duration.

As the burnup increase for the assembly is known for each cycle, the mean specific power of the assembly can be determined for each cycle.

Knowledge of the burnup of the irradiated fuel samples is crucial, since this determines the precision with which the variation between the experimental and calculated results can be obtained for each of the isotopes analysed. The degree of uncertainty in determining the fluence τ , may become predominant for the isotopes at the end of the chain, such as Pu_{242} (as well as Pu_{238}) for which the abundance is constructed initially in relation to τ^4 . A variation of $\pm 1.5\%$ for example in the burnup, leads to an uncertainty respectively of $\pm 3.6\%$ and $\pm 3.4\%$ at 40 GWd/t_m for Pu_{238} and Pu_{242} .

As the burnup of the sample cannot be measured directly, it is generally deduced from certain fission products considered as fluence indicators, such as the ratios :

$$\frac{Nd_{238}}{U_{238}}, \frac{Nd_{235}}{U_{238}}, \frac{Nd_{240}}{U_{238}}.$$

These fission products are stable, additionally, the cross-sections of the dominant fissile isotopes and their fission yields are well known. As the number of U_{238} nuclei varies very little during the evolution, the degree of uncertainty associated with its determination by calculation is very small.

The experimental analysis of these indicators for various samples leads back to the assembly burnup used in the APOLLO 2 calculation.

2) Fuel temperature

Temperature is an essential parameter in neutronic calculations. It appears both in the initialisation of the cross-sections and fluxes, and in calculating resonance self-shielding. The temperature variations within a nuclear reactor result in variations in the cross-sections, which need to be taken into account.

A variation of $\pm 50^\circ\text{C}$ in the fuel temperature leads to a degree of uncertainty of about 1% at 60 GWd/t_m for the prediction of Pu₂₃₉.

In fact, an increase in temperature causes an increase in the resonant absorption. The phenomenon responsible is the Doppler broadening of resonances.

The radial distribution of the fuel temperature within the pellet, and its variation during irradiation are determined via a thermo-mechanical calculation, using the METEOR code developed by the CEA [6]. This code enables the behaviour of a fuel pin to be studied in relation to its geometry, the nature of its constituent materials and the irradiation characteristics.

As the self-shielding model which was used for our calculations cannot take into account the high temperature gradient present in a fuel pin, use is made of an "effective temperature" which preserves the absorption reaction rate of U₂₃₈.

The expression for the effective temperature, which is recommended for use for APOLLO 2 calculations is as follows [17] :

$$T_{eff} = \overline{T_{therm}} - \frac{1}{18}(T_C - T_S)$$

METEOR provides the following temperatures :

$$\overline{T_{therm}} = \frac{\int_0^R T(\tilde{r}) \cdot d\tilde{r}}{\int_0^R d\tilde{r}}, T_C \text{ and } T_S.$$

3) Cladding temperature

It has been shown that a variation in the cladding temperature has no influence on the evolution of the isotopic inventory.

In fact , tests have to be conducted beyond the physically plausible range ($T < 200^\circ\text{C}$ and $T > 600^\circ\text{C}$) in order to begin to perceive a variation of about 0.1%. For our study, the temperature chosen was 400°C.

4) Moderator-coolant temperature

The temperature variation of the moderator results both in a thermal spectrum shift and in a variation in density. As the temperature decreases, the density increases, and the moderation of neutrons is therefore enhanced. This results in a higher absorption of Pu₂₃₉ relative to the capture of U₂₃₈ (conversion) and therefore a deviation in the material inventory with respect to the quantities of Pu₂₃₉ and Pu₂₄₁.

The sensitivity at 40 GWd/t_m around T_{H₂O} = 306°C attains + 0.24% / °C for the ratios

$$\frac{Pu_{239}}{U_{238}} \text{ and } \frac{Pu_{241}}{U_{238}}.$$

The determination of the density of the moderator is therefore an important factor in the calculation of an accurate material inventory. The moderator temperature is calculated by METEOR, after which the density of the water is obtained by interpolation in the thermodynamic tables at 155 bar in our particular case.

5) Concentration of soluble boron

Monitoring of the decrease in reactivity due to burnup of the fuel, is performed through the soluble boron present in the moderator. For the evolution calculations, one usually chooses a mean content considered as being constant throughout the cycle ($C_B = 456$ ppm of boron in our 17x17 UO_2 PWR case). This choice is the previous modelling employed in our cycle and/or qualification calculations. In order to improve this representation (particularly Pu_{239} equilibrium concentration), a level portion has been introduced at the end of the cycle, with a zero boron concentration.

We have additionally conducted a comparative study between irradiation with close boron control, incorporating about 10 level portions per cycle (EDF data), and an irradiation sequence with 1 flat. The table below clearly shows that the modelling employed does not induce any significant bias of the isotopes which are important to the physics of reactors.

	Irradiation with 10 flats / Irradiation with 1 flat
Assembly burnup (MWd/t _m)	38 430
U_4 / U_8	- 0.17%
U_5 / U_8	- 0.43%
U_6 / U_8	0.17%
Np_7 / U_8	0.41%
Pu_8 / U_8	0.75%
Pu_9 / U_8	0.02%
Pu_0 / U_8	0.35%
Pu_1 / U_8	0.26%
Pu_2 / U_8	0.73%

6) Campaign extension

Campaign extension or "stretch-out" is a special operating mode frequently employed by EDF. It consists in continuing irradiation beyond the reactivity reserves controlled through the boron diluted in the moderator by reducing the water coolant temperature and the power level. This effect is modelled in our calculations.

7) Inter-cycle shut-downs

These shut-downs are taken into account in our calculations through a zero power cooling module in APOLLO 2. This modelling is essential to clearly take into account the decrease in radioactivity such as for example :

$\text{Pu}_{241} \rightarrow \text{Am}_{241}$ (β^- , T = 14.4 years) or $\text{Cm}_{242} \rightarrow \text{Pu}_{238}$ (α , T = 163 days)

APOLLO 2 REFERENCE CALCULATION ROUTE FOR DEPLETION OF 17X17 EVOLVING UO₂ PWR ASSEMBLIES

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 UO₂ assemblies. It enables the bias to be minimised between the "master" calculations, and it satisfies our target accuracy, namely 1% for Pu₂₃₉ and power in each pin, up to 60 GWd/t_m. The APOLLO 2 calculation route uses the CEA93 cross-section library in the European X-MAS group structure (172 groups), processed from the JEF 2.2 evaluations.

The recommendations for the flux calculation are as follows :

- The spatial calculation for the UOx assembly is achieved using the UP₁ 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. A single flux calculation point is adopted for the moderator.
- The inter-assembly water gap is integrated within the peripheral cells of the assembly, constituting a row of rectangular fuel cells.
- With a view to reducing the calculation cost, a grouping of cells with similar flux within a unique 'physical cell' is recommended. It consists of 18 physical fuel cells, 2 physical "tube-guide" cells and one physical "instrumentation tube" cell (cf. Figure 4).
- 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 U₂₃₈ 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 UO₂ assemblies, also enabled recommendations to be made for this :

- The isotopes to be self-shielded are : U₂₃₈, U₂₃₅, U₂₃₆, Pu₂₃₉, Pu₂₄₀, Pu₂₄₁ and Pu₂₄₂.
- Self-shielding of the U₂₃₈ large resonances are covered by the accurate model UP₁. The UP₀ ROTH approximation, which considers a linearly isotropic interface flux, is adequate to ensure the quality of the self-shielding results for the other isotopes.

								18
							15	17
						13	14	16
					21	10	13	16
			8	7	9	11	12	
		20	7	7	21	10	12	
	4	3	6	6	9	11	12	
	2	4	3	6	6	9	11	12
19	1	3	20	5	5	21	10	12

Figure 4. Multicell pattern of a 17X17 UOx assembly

- It is essential to differentiate between 2 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 corner + peripheral pin.
- The self-shielding of U_{238} , U_{236} and Pu_{240} is considered in rings by the Direct Method formalism[8]. A mean pellet self-shielding (Livolant formalism) is adequate for the other isotopes.

The following recommendations may be added with respect to the evolution :

- The evolution steps are : 0, 37.5, 75, 112.5, 150, 325, 500, 750, 100, 2 000, 2 500, 3 000, 4 000, 5 000, 6 000, 7 000, 8 000, 10 000 MWd/ t_m , then in 2 000 MWd/ t_m steps.
- The self-shielding calculations are conducted at the following steps : 0, 4 000, 8 000, 12 000, 24 000, 36 000, then in steps of 12 000 MWd/ t_m .

QUALIFICATION OF THE PWR FUEL INVENTORY CALCULATIONS

The total experimental uncertainties correspond to the combination of the following components obtained from sensitivity calculations :

- the evaluation of the temperature of the fuel,
- the evaluation of the temperature of the moderator,
- the precision of the cycle and power history. Table I shows for example the deviation observed between accurate cycle follow-up, such as that modelled by our calculations, and a simpler model (P, T and C_B constant throughout all the irradiation cycles).
- the burnup of the assembly derived from Nd and Cs isotopics,
- the chemical analyses.

Table I. Uncertainties due to cycle/power history (in %)

Isotopic ratio	30 GWj/t	40 GWj/t	50 GWj/t	60 GWj/t
U234/U238	±0.1	±0.2	±0.5	±1.0
U235/U238	±0.1	±0.2	±0.3	±0.4
U236/U238	±0.1	±0.1	±0.1	±0.1
Pu238/U238	±1.2	±1.4	±1.7	±2.0
Pu239/U238	±0.4	±0.4	±0.4	±0.4
Pu240/U238	±0.2	±0.2	±0.2	±0.2
Pu241/U238	±0.4	±0.5	±0.6	±0.7
Pu242/U238	±0.1	±0.1	±0.2	±0.3
Am241/U238	±1.8	±2.0	±2.2	±2.5
Am242/U238	±8.0	±10.0	±12.0	±15.0
Am243/U238	±0.1	±0.2	±0.2	±0.3
Cm243/U238	±0.4	±0.5	±0.6	±0.9
Cm244/U238	±0.5	±0.6	±0.8	±1.0
Cm245/U238	±0.6	±0.7	±0.9	±1.1
Cm246/U238	±0.6	±0.6	±0.6	±0.7
Cm247/U238	±0.8	±0.9	±1.0	±1.1

The trends observed in the analysis of both programmes were similar, and the results are summarised in Tables II and III relative to Burnup and Enrichment of the fuel, together with their associated uncertainties (in one standard deviation).

Table II. (C-E)/E in %. Major Actinides.

Isotope		20GWd/t	40GWd/t	50GWd/t	60GWd/t
U234	e=3.1%	3.0	2.0		
	e=4.5%	0.4	0.2	-2.0	1.0
	Uncertainty	±1.1	±1.4	±1.6	±2.0
U235	e=3.1%	-0.8	-2.0		
	e=4.5%	0.9	1.0	0.5	2.5
	Uncertainty	±1.1	±2.0	±2.7	±3.5
U236	e=3.1%	-3.2	-3.0		
	e=4.5%	-4.3	-3.7	-4.4	-4.2
	Uncertainty	±1.3	±0.9	±0.7	±0.6
Np237	e=3.1%	-12.0	-3.3		
	e=4.5%	-3.7	-3.8	-4.3	-5.0
	Uncertainty	±3.0	±2.8	±2.8	±2.7
Pu238	e=3.1%	-10.7	-6.0		
	e=4.5%	-10.3	-8.0	-7.7	-6.9
	Uncertainty	±4.0	±3.9	±3.8	±3.7
Pu239	e=3.1%	-3.2	-1.3		
	e=4.5%	-1.8	-0.2	1.2	2.4
	Uncertainty	±0.9	±1.1	±1.2	±1.3
Pu240	e=3.1%	-2.6	1.4		
	e=4.5%	-3.0	-1.0	-0.6	-0.4
	Uncertainty	±1.9	±1.5	±1.3	±1.1
Pu241	e=3.1%	-7.2	-6.4		
	e=4.5%	-6.3	-4.9	-2.8	-1.8
	Uncertainty	±2.3	±1.8	±1.6	±1.6
Pu242	e=3.1%	-8.4	-8.8		
	e=4.5%	-10.0	-9.0	-8.1	-8.2
	Uncertainty	±4.0	±3.4	±3.1	±2.8

- The U_{235} depletion is accurately simulated up to very high burnup fractions, even if the quantities present become very small, and the associated uncertainty and the sensitivity to the cross-section becomes greater.
- The abundance of U_{236} is underestimated by $-4\% \pm 0.6\%$, principally due to the approximately 10% underestimation of the U_{235} capture Resonance Integral in JEF2. Validation tests have moreover shown [9] that the new evaluation of U_{235} by Leal, Derrien and Larson, which is introduced in JEFF3 and ENDF/B6 release 5, improves the build-up of U_{236} and overcomes the underestimation for this isotope.
- The abundance of Pu_{238} is underestimated by about 8%, partly due to poor formation of U_{236} .
- Compared to previous APOLLO1 results [10], Pu_{239} is now evaluated within 2% accuracy, thanks to the APOLLO2 space-dependent self-shielding in pellet

concentric rings (Matrix Dilution theory)) and the accurate Dancoff calculation (2D lattice collision probabilities through UP₁. model).

- Pu₂₄₀ closely follows the evolution of Pu₂₃₉ which confirms the validity of the Pu₂₃₉ capture cross section and correct handling of Doppler/self-shielding resonance for Pu₂₄₀ at 1 eV.
- Pu₂₄₁ is slightly underestimated. Its prediction is highly sensitive to Pu₂₃₉ and therefore to the temperature of the fuel. Moreover, when the Pu₂₃₉ is at equilibrium, Pu₂₄₁ is quite accurately calculated.
- Pu₂₄₂ is underestimated by about 8% ± 3%.

Table III. (C-E)/E in %. Minor Actinides

Isotope		20GWdj/t	40GWd/t	50GWd/t	60GWd/t
Am241	e=3.1%	-9.3	-10.9		
	e=4.5%	-7.9	-4.2	-10.0	-0.5
	Uncertainty	±4.2	±3.4	±3.2	±3.0
Am242m	e=3.1%	-25.0	-39.0		
	e=4.5%	-37.0	-25.0	-35.0	-22.0
	Uncertainty	±8	±11	±12	±15
Am243	e=3.1%	-17.0	-11.0		
	e=4.5%	-18.0	-11.0	-19.0	-7.0
	Uncertainty	±6.2	±5.2	±4.7	±4.4
Cm243	e=3.1%				
	e=4.5%	-32.0	-27.0	-	-17.0
	Uncertainty	±7.1	±5.7		±4.3
Cm244	e=3.1%				
	e=4.5%	-27.0	-19.0	-	-15.0
	Uncertainty	±8.0	±6.9		±5.9
Cm245	e=3.1%				
	e=4.5%	-30.0	-20.0	-	-15.0
	Uncertainty	±9.6	±8.4		±7.0
Cm246	e=3.1%				
	e=4.5%	-37.0	-28.0	-	-28.0
	Uncertainty	±11.3	±10.7		±9.6
Cm247	e=3.1%				
	e=4.5%	-39.0	-35.0	-	-35.0
	Uncertainty	±13.8	±12.8		±11.8

- Am₂₄₁ is reasonably estimated (-5% ± 3%) through cycle follow-up which improves its estimation, as this isotope is formed by the decaying of Pu₂₄₁.
- Am₂₄₂ is underestimated by -25% ± 10%. This result is probably due to 2 factors : underestimation of the branching ratio of Am₂₄₁ to Am₂₄₂ and a high degree of uncertainty with respect to σ for Am₂₄₂, which is extremely high (σ = 6900 barns).
- The underestimation in Pu₂₄₂ generates an underestimation for Am₂₄₃ , which in turn generate errors for Cm₂₄₄ and Cm₂₄₅. (-20% at 40 GWd/t).

CONCLUSION

This study enabled the qualification of the evolving material inventory for 17x17 UO₂ PWR assemblies calculated using the APOLLO 2 reference calculation route and the associated CEA93 nuclear data library based on JEF2 evaluations

Two programmes were used : BUGEY 3 (3.1% enrichment in U₂₃₅) and Gravelines for high burnups (4.5% enrichment).

The METEOR thermo-mechanical code enabled the fuel and moderator temperatures to be obtained in relation to the irradiation. The choice of irradiation modelling in the form of cycle follow-up was performed and showed its advantage.

This work therefore showed that APOLLO 2 correctly determines the fuel inventory : the major actinide content -U₂₃₅, Pu₂₃₉, Pu₂₄₀ - is reproduced within 2% accuracy.

The depletion qualification range for APOLLO 2 as now for UO₂, up to 4.5% enrichment with U₂₃₅ and high burnup fractions up to 61 GWd/t_m.

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