Review of four decades dedicated to spent fuel analyses in france an incomparable database built for codes validation.

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REVIEW OF FOUR DECADES DEDICATED TO SPENT FUEL ANALYSES IN FRANCE: AN INCOMPARABLE DATABASE BUILT FOR CODES VALIDATION.

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This paper describes the experimental programs conducted in France by the Atomic and Alternative Energy Commission (CEA) in order to validate spent fuel inventory calculations and reactivity effects for core studies as well as fuel cycle studies. This large experimental program took almost 40 years to be completed and was defined within the framework of collaboration with our French partners, AREVA and Electricité de France (EDF).

The experimental database comprises chemical analysis measurements (UOX, URE, UO2-Gd2O3 and MOX fuels) from fuel pin samples irradiated in reactors built in France. Additional information comes from full assembly dissolutions at the AREVA/La Hague reprocessing plants for UOx fuels. Furthermore, for some of analyzed fuel rods, specific samples of about 10 cm long have been extracted in the neighborhood of the analyzed sample in order to be oscillated in dedicated lattices in the MINERVE facility of the CEA at Cadarache, allowing the determination of the total reactivity effect versus the burnup of spent fuel compared to fresh reference fuel.

This database is regularly updated and nowadays several hundreds of isotopic ratios are available.

I. INTRODUCTION.

Since more than four decades, the French nuclear energy actors (Atomic Energy and Alternative Energy Commission – CEA, Electricité de France – EDF and AREVA) have launched a large experimental program (Ref 1.) based on spent fuel chemical analyses for validation of neutronic codes aimed at fuel inventory calculations. Uranium, plutonium, neptunium, americium and curium isotopes as well as cesium or neodymium isotopes have been analyzed in PWRs and BWRs fuel. Furthermore, the 15 fission products used in burnup credit (BUC) criticality calculations (^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{143}Nd, ^{145}Nd, ^{153}Eu, ^{133}Cs, ^{155}Gd, ^{99}Tc, ^{95}Mo, ^{101}Ru, ^{103}Rh, and ^{109}Ag) and the isotopes involved in decay heat have been measured.

The experimental database comprises chemical analysis measurements (UOX, URE, UO_2-Gd_2O_3 and MOX fuels) from fuel pin samples irradiated in reactors built in France. Additional information comes from full assembly dissolutions at the AREVA/La Hague reprocessing plants for UOx fuels. Furthermore, for some of analyzed fuel rods, specific samples of about 10 cm long have been extracted in the neighborhood of the analyzed sample in order to be oscillated in dedicated lattices in the MINERVE facility of the CEA at Cadarache, allowing the determination of the total reactivity effect versus the burnup of spent fuel compared to fresh reference fuel.

This database is regularly updated and nowadays several hundreds of isotopic ratios are available. The experimental database covers a large range of ^{235}U enrichments (from 3.1% up to 4.5%) for UOX fuel, with a burnup range varying from 10 GWd/t up to 85 GWd/t. MOx fuels have also been investigated, with a Pu amount varying from 2.9% up to 9.8%; For the irradiated MOx fuel pins, the burnup varies from 10Gwd/t up to 56 GWd/t.

This paper describes the available experimental results used in France to validate fuel inventory calculations (Ref. 2). The first part gives an overview of the experimental database and the second part describes the several sequences from irradiation in power plant to chemical and isotopic analyses. Information related to oscillations of spent fuel samples in MINERVE are also given in the second section. The third part presents the typical uncertainty and the last section gives some highlights on the main validation results for the APOLLO-2 neutronic code.

II. THE EXPERIMENTAL DATABASE FOR LWR DEPLETION CODE VALIDATION.

This section describes the experimental database used for Light Water Reactor depletion code validation. Table 1 synthetizes the available experimental data. Thus, the database contains information about irradiated fuel in PWRs and BWRs, which can be classified into two groups:

- Small fuel pin samples, irradiated in French reactors, with positions in the assembly well characterized. The analyzed samples are obtained from cuts of extracted pins. In order to investigate the local irradiation effect, various axial locations of cuts are selected with always a sample cut in the middle of the pin. These time-consuming and expensive experiments provide very accurate results for a limited number of samples.
- Dissolution aliquots of entire assembly sets obtained from reprocessing plant (COGEMA/La Hague). A lot of these measurements have been performed. However the irradiation histories of the assemblies are not very well known and the fact that dissolutions are performed over batches of several assemblies limits the usefulness of these data for code validation. This type of data is nevertheless used to extend the validation range in a statistical sense.

II.A. The UOx fuels experimental database

Originally, four main programs related to UOx fuel samples in PWRs were used for the experimental validation of actinides and fission products inventory. They are named: BUGEY3, FESSENHEIM II, GRAVELINES, and CRUAS. The experimental data coming from the BUGEY3 reactor were the first representative experiment of the French nuclear power reactors. It involves standard PWR Zy4-clad fuel pins irradiated in two 17×17 assemblies (2.1% initial ^{235}U enrichment irradiated during the first two cycles, and a 3.1% enrichment irradiated for 3 cycles). The maximum burnup reaches 40 GWd/t. The FESSENHEIM II program (4 and 5 cycle irradiations) completes the BUGEY3 program with UOx fuel in a higher burnup range from 45 up to 60 GWd/t.

The program performed with the fuel originating from the GRAVELINES reactor is devoted to the validation of the calculation schemes for high burnup fuel and high cycle length operation (initial UO_2 fuel enrichment is 4.5%). This experimental program on a 900 MWe PWR is the most important being carried out in France up to now. Two 17x17 assemblies containing each 20 removable fuel pins were irradiated for 4 cycles in the Gravelines 3 reactor then in the Gravelines 2 reactor for the fifth cycle. Out of these twenty pins, two pins were extracted at the end of the second cycle of the reactor, one pin at the end of the 3rd cycle and two pins at the end at the fourth cycle but, in the latter case, only one was analyzed. For the other assembly, two fuel rods have been extracted after four cycles, one was analyzed, then 4 pins were extracted after the fifth cycle, three among them were analyzed. Figure 1 shows the radial cross-section of a GRAVELINES assembly. The associated number indicates the irradiation cycle length (the rods extracted after 1 cycle comes from another assembly but for simplification we put them on the same figure).

The CRUAS program is devoted to the validation of the URE (Reprocessed Uranium) fuel, using reprocessed then enriched uranium. It permits validation of the ^{236}U capture cross-section. This program is characterized by a 3.5% ^{235}U enrichment with an initial amount of 1.2% of ^{236}U.

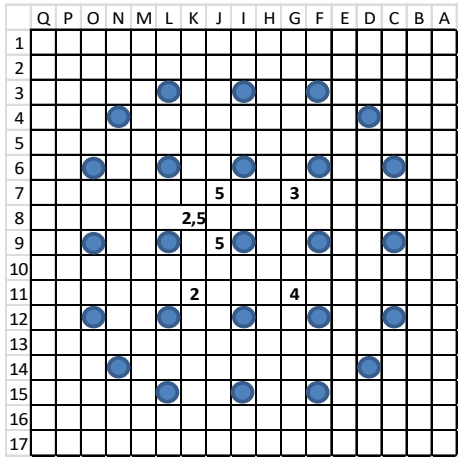

Fig. 1. Experimental UOX-5 cycles fuel pins in GRAVELINES.

To complete the preceding experiments, additional information for irradiated UOX fuel come from the MALIBU program (Ref. 3) which consisted of analyzing one sample located in the medium axial plane of a pin (4.3% ^{235}U) of a 15x15 assembly loaded in the Swiss GOSGEN reactor. For high burn-up, an additional program called “ALIX” was undertaken at the beginning of the 2000’ and aimed at increasing the burn-up for UOX fuel in the GRAVELINES-5 power plant. In this program, four UOX fuel pins (4.5% ^{235}U enrichment) were irradiated during 5, 6 and 7 cycles reaching thus about 85 GWd/t for the most irradiated sample.

For the validation of gadolinium poison rods calculation a series of two experiments, called GEDEON (Ref. 4, 5), has been undertaken in the 80’ and consisted in irradiating UO_2-Gd_2O_3 poison rods in the center of the former MELUSINE reactor in GRENOBLE. For the experimental pins, the burn-up varies in a range of 2 up to 12 GWd/t.

From full assembly dissolutions at COGEMA/La Hague reprocessing plants, we get uranium and plutonium chemical analyses. The involved assemblies are 900 MWe 17×17 PWR with 3.1%, 3.25% or 3.45% enrichments and

with burnups ranging between 25 up to 45 GWd/t. However, the dissolution process involves batches of several assemblies: therefore sets of assemblies with very close irradiation histories (and consequently burnups) are especially selected to be integrated in the database.

Table 1: Overview of the whole experimental program dedicated to spent fuel measurements.

Fuel	Power Plant	Enrichment	Burn Up (GWd/t)	Isotopic Analyses	BUC FP	Oscillations in MINERVE
UOX-Gd	« GEDEON »		2-12	U, Pu, MA, FP		
UOX	BUGEY & FESSENHEIM	2.1% & 3.1%	20	U, Pu, MA, FP	x	x
			25	U, Pu, Am, FP		x
			40	U, Pu, Am, FP	x	x
			50	U, Pu, Am, FP		x
			60	U, Pu, Am, FP		x
UOX	GRAVELINES	4.5%	25	U, Pu, Am, FP		x
			40	U, Pu, Am, FP	x	x
			50	U, Pu, Am, FP	x	x
			60	U, Pu, Am, FP	x	x
ERU	CRUAS	3.5%	15	U, Pu, Am, FP		
			25	U, Pu, Am, FP		
			35	U, Pu, Am, FP		
UOX	BWR « GUNDREMINGEN »	4.0%	26	U, Pu, Am, FP	x	
			46	U, Pu, Am, FP	x	
UOX	GOSGEN « MALIBU »	4.3%	70	U, Pu, Am, FP	x	
UOX	GRAVELINES ALIX	4.5%	65	U, Pu, Am, FP		x
			75	U, Pu, Am, FP		x
			85	U, Pu, Am, FP		x
MOX	SLB1	2.9-5.6%	10	U, Pu, Am, FP		
			28	U, Pu, Am, FP		
			40	U, Pu, Am, FP		
MOX	DAMPIERRE	6.7%	10	U, Pu, Am, FP		x
			40	U, Pu, Am, FP	x	x
			52	U, Pu, Am, FP		x
			58	U, Pu, Am, FP	x	x
MOX	TRICASTIN	9.8%	43	U, Pu, Am, FP	x	planned
			56	U, Pu, Am, FP		planned

MA: Minor Actinides – BUC: Burn-Up credit – FP: Fission Products – ERU : Enriched Retreated Uranium

II.B. The MOx fuels experimental database

Currently, in France, twenty-two PWRs are devoted to the Pu recycling in 30% mixed core loading. This increase of French MOX fuel cycle emphasizes the need to enlarge the experimental database to plutonium-fueled assemblies. The experimental database includes information coming from three reactors SLB1, DAMPIERRE and TRICASTIN.

The first French reactor using MOx assemblies is the SAINT-LAURENT B1 reactor. The post-irradiation examinations are carried out on fuel cuts coming from this reactor (Ref. 6).

The standard MOx assemblies used in Saint-Laurent B1 include three zones with different plutonium content. The central zone is characterized by a high Pu content (5.6%) and the peripheral zone by a small Pu content (2.9%). Two MOx fuel assemblies were selected for the SLB1 program. The extracted and analyzed MOx rods were irradiated for 1, 2 and 3 cycles, with burnups ranging from about 10 to 45 GWd/t. Figure 2 shows the position of the experimental fuel pins.

The DAMPIERRE program involves nine rod cuts and is devoted to the extension of the MOX calculation scheme validation for high burnup up to 58GWd/t The information concern fuel pins irradiated between 1 cycle (10 GWd/t) to 5 cycles (58 GWd/t) for a Pu enrichment of 5.3% and 6.7%.

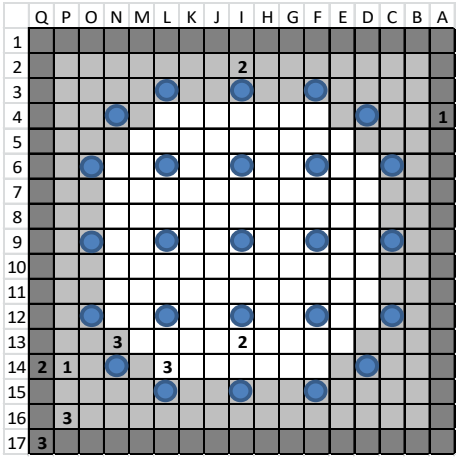

Fig. 2. Experimental MOX fuel pins in SLB1 program.

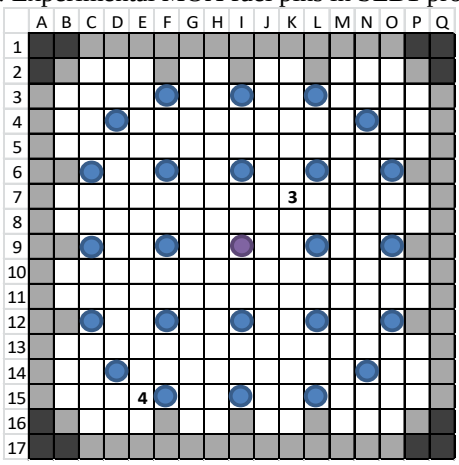

Fig. 3. Experimental MOX fuel pins in TRICASTIN program. New MOX loading pattern.

The TRICASTIN program involves two 9.8%-MOX fuel pins (3 and 4 cycles) loaded in a new loading pattern MOX assembly. The reached burn up are 42 and 56 GWd/t respectively. Figure 3 shows the location of the experimental pins in the assembly.

II.C. The experimental database of irradiated BWR fuels.

The experimental database for BWR contains chemical analysis results coming mainly from full dissolution of various assemblies completed with data related to samples irradiated in reactor then dissolved. In the full assembly dissolutions at AREVA/La Hague reprocessing plants, the involved assemblies are 7×7, 8×8 and 9×9 types with various enrichments and burnups ranging between 20 up to 40 GWd/t. Only the amounts of depleted U and Pu are available.

In order to improve the experimental information, an important experimental program was also launched by the former COGEMA Company (nowadays AREVA-NC), concerning chemical analyses of samples of fuel rods irradiated in the GUNDREMMINGEN reactor. Two fuel pins with cuts at several heights were investigated.

II.D. Oscillations in the MINERVE facility.

For both UOX and MOX programs, specific additional cuts were selected in the vicinity of some analyzed cuts in order to manufacture samples to be oscillated in the MINERVE facility, located at the Cadarache center.

MINERVE is a pool-type reactor operating at a maximum power of 100 watts. The core is submerged under 3 meters of water and is used as a driver zone for the different experimental lattices located in a central square cavity with a size of about 70×70 cm². The coupled lattices in this cavity are built such that they can reproduce the typical neutron spectrum of various reactors. The core is built in a parallelepiped pool of stainless steel containing about 100 m^3 of water. The moderator is distilled water insuring also the cooling by natural convection. The driver zone consists of enriched metallic uranium/aluminum MTR-type plate cladded with aluminum and gathered in elements of 9, 12, and 18 plates. About 30 elements comprise the driver zone which is surrounded by a graphite reflector**.** Figure 4 shows view of the MINERVE facility.

Fig. 4. View of the MINERVE facility

The oscillation experiments performed in the center of the central zone, allow determining the reactivity loss due to fission product and minor actinide build up linked to the irradiation**.** Several lattices can be loaded in MINERVE (Cf. Figure 5), allowing oscillating the samples in representative neutronic spectra of either standard UOX PWR, or MOX PWR, and also La Hague reprocessing plant dissolver characteristics for criticality studies (Burn-Up Credit experiments – Ref. 6, 7). Two High Conversion Reactor lattices were also designed at the middle of the 80's and used during 10 years (Ref. 8, 9).

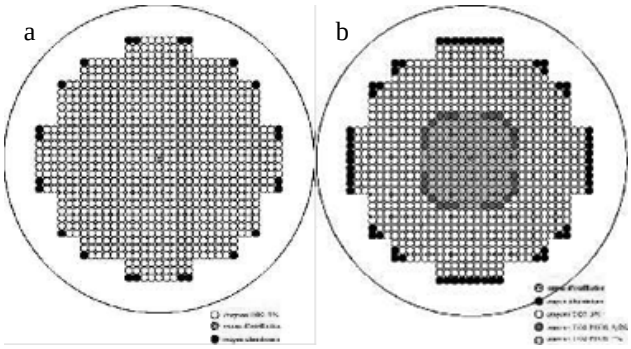

Fig. 5. Typical lattices to be implemented in the MINERVE facility ('a': UOX-PWR, 'b': MOX-PWR)

Thus, the experimental database contains data related to the fuel samples (material balance) and the reactivity effects of these samples compared to well-known calibrated samples containing boron (absorbing samples) or ^{235}U (fissile samples) quantities.

III. EXPERIMENTAL PROCESSES AND MAIN INFORMATION.

This chapter describes the several processes performed to obtain valuable data about irradiated fuel characteristics for code validation.

III.A. Nondestructive examinations of fuel pins.

For the French PWRs, the irradiated fuel pins are shipped from the power plant to the LECA/STAR facility at Cadarache center for non-destructive examinations and. especially a γ-spectrometry for axial burn-up profile measurement through ^{134}Cs and ^{137}Cs γ-rays counting.

Figure 6 shows an example of a typical axial γ-spectrometry result (^{137}Cs) obtained for a MOX fuel pin. The fuel pin is then cut in several samples; some sections of about 2 cm long, located far from any local perturbation such as grids, selected to be transferred to the ATALANTE facility in the Marcoule center for dissolution.

In the LECA-STAR facility, the gaseous fission products (He, Xe and Kr) are also characterized using the MERARG device which was recently (Ref. 10) up-graded with γ-spectrometry allowing on-line measuring fission gas release. The objective is to provide irradiated fuel specialists with an isotopic content of each gaseous fission products.

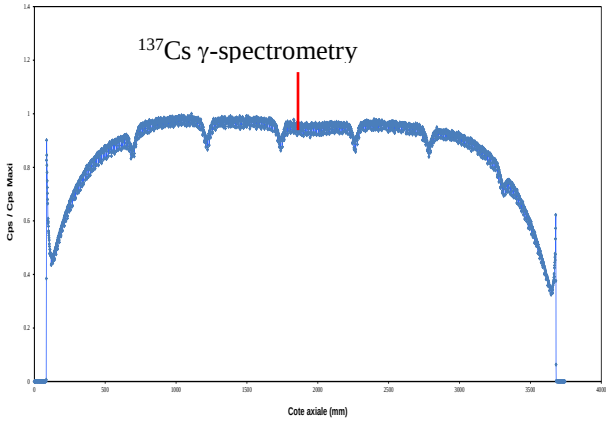

Fig. 6. Example of an axial γ-spectrometry of a fuel pin

III.B. Dissolutions processes at ATALANTE facility.

The selected sample is dissolved in acid solutions after having been weighted and geometrically characterized in a dedicated hot cell. Depending on the type of investigated chemical element in the irradiated fuel, either simple or more complex dissolutions are performed. For the classical heavy nuclides and fission products (U, Pu, Np, Am, Cm and Nd or Cs), a simple dissolution involving nitric acid is required. If one is interested in metallic fission products such as Ag, Tc, Rh or Ru (most of them are important for the Burn-Up credit), several processes are performed aimed at chemically stabilizing the dissolution and analyzing the parts which were not dissolved during the first run: in that case, four different processes are needed. Figure 6 show pictures of some devices used in hot cell for dissolution and filtration.

III.C. Radiochemical analyses at Saclay center.

The nitric solutions are then shipped to the chemical labs located in Saclay for radiochemical analyses. After chemical separation (U, Pu, minor actinides, Fission products), simple (Ref. 11) or double dilutions involving calibrated solutions are performed by mass spectrometry for measuring isotopic ratios such as $^{237}Np/^{238}U$, $^{239}Pu/^{238}U$ or $^{148}Nd/^{238}U$ (Ref. 12). Isotopic concentration ratios for the same element (for example $^{i}U/^{238}U$ or $^{i}Pu/^{239}Pu$) are measured directly by mass spectrometry.

The last measurements concerned MOX fuel pin (9.8% of Pu) which was irradiated during 4 cycles (56 GWd/t) in the TRICASTIN NPP; The analyses of two samples were performed in order to evaluate the influence of a single dissolution compared to a total dissolution involving 4 different phases of chemical treatment in the ATALANTE facility on the final isotopic ratios. The neighboring samples were chosen in the same part of the fuel pin in order to limit strong variations on the material balance due to irradiation conditions. (Figure 7 shows approximatively the location of the samples in red).

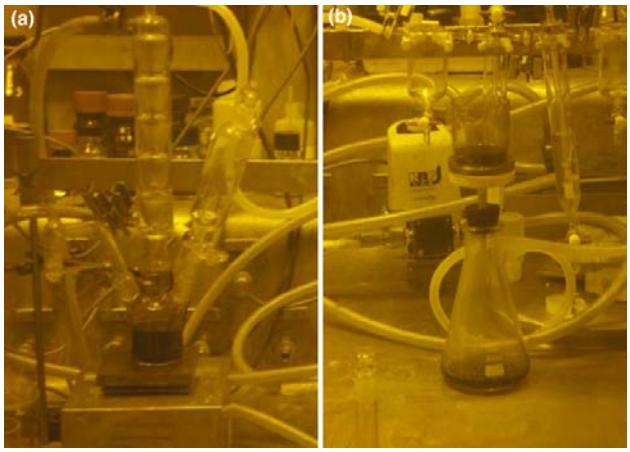

Fig. 7. Pictures of the first dissolution step (a) and first filtration (b).

The analyses show a very good consistency between the two samples as shown in Table 2. The variation of the isotopic ratios is less than the experimental uncertainty. This result seems to show that the dissolution technique used in ATALANTE could be used for the dissolution of irradiated MOX fuel assemblies in La Hague if the reprocessing of such fuel would be decided.

Table 2: Observed Discrepancy between analyses depending on dissolution processes

Nuclide Ratio	Discrepancy (%)
$^{235}U/^{238}U$	0.2 ± 0.3
$^{238}U/^{239}Pu$	0.1 ± 0.6
$^{240}Pu/^{239}Pu$	0.1 ± 0.3
$^{241}Am/^{238}U$	0.4 ± 0.8
$^{148}Nd/^{238}U$	0.1 ± 1.2
$^{135}Cs/^{238}U$	0.2 ± 1.1

IV. EXPERIMENTAL UNCERTAINTIES.

The experimental uncertainties come from: the chemical or radiochemical analyses, the dissolution processes and the material balance of the fuel sample. The typical uncertainty due to radiochemical analysis on an isotopic ratio is generally less than $\pm 0.2\%$ or 0.3% (1σ) depending on the considered isotope. For ratios involving isotopes of different elements the chemical uncertainty (1σ) can vary in a range of $\pm 0.1\%$ (typically for $^{148}Nd/^{238}U$) up to $\pm 1.2\%$ for $^{237}Np/^{238}U$ ratio.

The dissolution processes generate additional uncertainties (Ref. 13) which must be taken into account in the total experimental uncertainty for the depletion code validation. These additional uncertainties can come for example from losses during the dissolution (evaporations, residual drop during transfers, etc.) and also from the parts which are not dissolved during the first phase.

Generally, the non-dissolved part concerns mainly metallic fission products (^{109}Ag, ^{99}Tc, ^{95}Mo, ^{101}Ru, ^{103}Rh, etc.) but also a small part of initial U or Pu. A current study, from which the results will be published at the end of 2016, shows that these non-dissolved materials and losses induce additional uncertainties (1σ) of about $\pm 0.3\%$ up to $\pm 0.6\%$ depending on the sample and on the used dissolution process.

Finally, the total uncertainty is less than $\pm 1.5\%$ for $^{237}Np/^{238}U$ but for Pu or main fission products the uncertainty is less than $\pm 0.6\%$ (1σ).

For the MINERVE oscillation technique, the uncertainty can be separated in three independent parts : about 1% comes from the mass of irradiated sample, 2.5% associated to calibration of reactivity versus reference samples and less than 0.3% due to the measurement itself. Finally, the total uncertainty is $\pm 3.5\%$ (1σ).

V. MAIN VALIDATION RESULTS.

The depletion code used in France is the APOLLO-2 (Ref. 14) lattice code which solves the Boltzmann equation for the flux and after convergence, the Bateman equations for determining the isotopic concentrations of heavy nuclides, fission products and burnable poisons if necessary. The nuclear data library comes from on the JEF3.1.1 evaluation file and the recommended calculation scheme is the “SHEM-MOC” package (Ref. 15).

The whole experimental results included in the database have been calculated and were taken into account in the validation report. Several hundreds of isotopic ratios and about one hundred oscillation measurement results are available.

Table 3 show the example of calculation/experiment comparisons obtained with a reference calculation scheme “SHEM-MOC” on isotopic ratios for UOX fuel. The same type of (C-E)/E are obtained for MOX fuel.

The MINERVE oscillation measurements contributed to show that APOLLO-2 is slightly overestimating the total reactivity loss with burn-up of about 1.7% to 3.6% ($\pm 2.3\%$ uncertainty at 1σ) for both

UOX and MOX fuels. This discrepancy does not significantly vary with the burn-up increase.

Figure 8 shows the comparison of calculated concentrations of odd Gd isotopes to the experimental data coming from the GEDEON-2 experiment (Ref. 16). It demonstrates the satisfactory accuracy of the APOLLO2.8 calculations (with the JEF3.1.1 library and the SHEM-MOC recommended scheme) for the depletion of ^{155}Gd and ^{157}Gd poisoning isotopes. With the *REL2005* calculation scheme, averaged C-E biases on their consumption throughout irradiation are -0.5% ± 1.0% (1σ-rms) and -0.7% ± 1.5% (1σ-rms) respectively.

Table 3: (C-E)/E for main isotopic ratios (1σ uncertainty)

Nuclide	25 GWj/t	60 GWj/t	85 GWj/t
^{235}U	0.4 ± 1.0	3.5 ± 4.0	9.5 ± 7.0
^{236}U	-0.1 ± 1.1	-1.1 ± 0.8	0.3 ± 1.2
^{237}Np	-2.8 ± 3.4	-1.3 ± 3.0	-2.6 ± 2.6
^{239}Pu	0.6 ± 0.9	2.3 ± 1.3	-1.5 ± 1.8
^{240}Pu	0.9 ± 1.7	2.1 ± 1.1	5.3 ± 1.0
^{241}Pu	-1.9 ± 2.0	-1.4 ± 1.6	-2.1 ± 1.8
^{242}Pu	-0.4 ± 3.8	-2.5 ± 2.8	-0.3 ± 2.6
^{241}Am	-2.2 ± 4.0	0.6 ± 4.5	0.4 ± 5.0

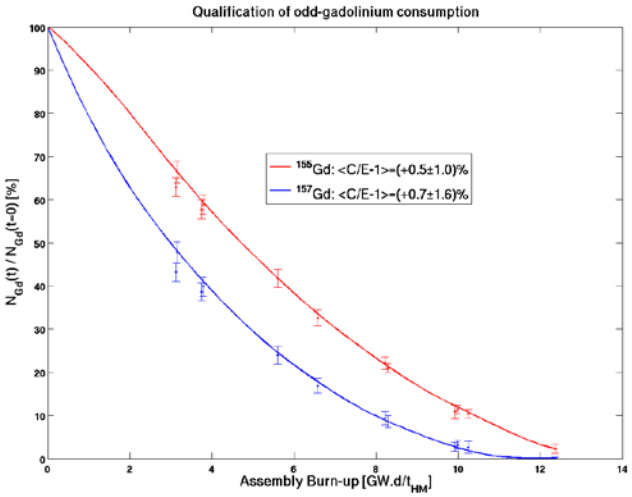

Fig. 8. Profile of odd Gd isotopes consumption versus burn-up. Experiment/calculation comparison.

VI. CONCLUSIONS AND PERSPECTIVES

This paper gives an overview of the experimental database of spent fuel built during forty years aimed at validating the French depletion codes.

This database is regularly enriched by new experimental results concerning current loading strategies for French PWRs. These data are also completed by information coming from the La Hague reprocessing plant.

Spent fuel analyses performed on more than fifty samples irradiated between 2 GWdt up to 85 GWd/t constitutes an incomparable database for validating the depletion codes used for LWR calculations.

The database comprises also data related to decay heat and fast breeder reactors covering thus almost the whole neutronic phenomena involved in irradiated fuel domain.

In the near future, it is planned to analyze UO_2-Gd_2O_3 fuel pin irradiated one cycle in a NPP in order to complete experimental results obtained through the GEDEON program.

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