French Fission Products Experiments Performed in Cadarache and Valduc. Results Comparison.

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Cofunded by Cogema, two complementary experimental programmes on burn up credit (BUC) related to fission products (FPs) are performed by CEA & IRSN at Cadarache and Valduc. After shortly recalling the main characteristics of each experiment, a first comparison of some results is presented, especially the energy range in which most part of cross section absorption are qualified. Both experiments exhibit great quality and accurate results, giving a high degree of confidence to the whole experimental French process of qualification devoted to BUC.

1. Introduction

As theoretical studies have shown the economical interest of using FPs in safety assessments at various stages of Fuel Cycle, France has been engaged since many years in an extensive programme related to BUC in order to qualify FPs absorption cross sections and calculation tools for criticality calculations in storage, transportation and dissolution of spent fuels. Cofunded by Cogema, two experimental complementary programmes have been separately performed in Valduc [1)] and in Cadarache [2)] on six (chosen by IRSN) or better, fifteen (chosen by CEA) main FPs, responsible for 50% [3)] and 80% [2)] of the total FPs absorption (see Table 1). This paper presents a short overview of the two French programmes. After recalling the main aspects of each programme, already published in ICNC or ANS meetings, some new results are given, completed by a comparison between the two experiments analysis.

2 Valduc Experiments

2.1 Description

The Valduc experiments are performed in three gradual steps at Valduc [1,3)] in so-called 'B Apparatus' by using the sub-critical approach technique based on the rising of moderating and reflecting water of a driver array up to $K_{eff} = 1 - \beta/10$. In the centre of the driver, FPs are in solution in a Zr tank, alone or mixed, with or without interactions with U, Pu, & Am.

The first series of experiments, called 'Physical' type experiments, is representative of storage and transportation conditions: the square pitch of the driver array (1.3 cm) leads to a thermal neutron spectrum, representative of the nominal square pitch (1.27 cm) of storage and transportation. The 1.3 cm square pitch is obtained by taking into account the water holes devoted for control and detection devices.

To improve the 'dissolution' qualification (whose neutron spectrum is more thermal than the previous ones), a second series of experiments, named 'Elementary Dissolution' type, has been performed. FPs are then in close interaction with the U, Pu & Am isotopes of inner array of UO_2 rods or HTC rods (with a square pitch of 1.272 cm) in the Zr tank. The UO_2 rods have an initial fuel enrichment of 4.738 wt% ^{235}U, and the HTC rods, so-called 'Haut Taux de Combustion', simulates U, Pu & Am composition for a UO_2 fuel with initial enrichment of 4.5 wt% ^{235}U irradiated at 37.5 GWd/t, without FP. This second series is itself divided in two cases: FPs in acid solutions (HNO_3 – 1N) or FPs in Depleted Uranyl Nitrate Solution (DUN). All these cases are completed.

Other experiments were performed on natural Gd solution, on ^{95}Mo in thin slices of CH_2/natural metallic Mo, and on F as polytetrafluorethylene (PTFE) solid block. In all, 156 experiments have been performed.

A third series of experiments, named 'Global or Advanced Dissolution' type, is planned. It consists of a large SS tank (70.4 x 70.4 cm^2) containing an 44 x 44 HTC rod array (square pitch 1.6 cm) steeping in a DUN solution poisoned with 6 FPs. The 1.6 cm pitch leads to an even more thermal neutron spectrum, more convenient to 'dissolution' qualification if necessary.

FP solutions are very carefully and accurately done by dissolving known masses of FPs in known quantities of nitric acid. A Saclay/CEA qualified laboratory independently checks concentrations and isotopic compositions of FPs. The adequacy and the accuracy of the model are checked on critical

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experiments performed with slightly acid water (without FP) solutions or with boron (well known cross sections) solutions.

2.2 Experiments Analysis

Description and analysis of experiments follows ICSBEP methodology, with special effort for uncertainty weight evaluation. Thus, clean benchmarks are available. Calculations are performed using the standard CRISTAL route [4] with APOLLO2-MORET4 and its CEA93 library (a 172 group library using the European JEF2.2 file [5]). TRIPOLI4.1, with pointwise JEF2.2 or ENDF BVI cross-sections is also used to perform some calculations. Uncertainty weights are obtained by keff difference of standard APOLLO2 Sn results (on related cylindrical geometry of the model) or by using MORET4 Perturbation [6] (correlated samples method), giving very accurate results.

Note that calculations taking account of FP self shielding were made to estimate its effect: this one is very small in Valduc experiments (being generally in the thermal range), except for ^{133}Cs (effect ~ 40 10^{-5}).

2.3 Results

Keff were calculated by APOLLO2-MORET4. The benchmark keff results are close to 1, with an average reactivity weight of total uncertainty about 50 10^{-5}. The most important uncertainty comes from the outer clad diameter (on which we have done 300 measurements after the new cladding, following LEU-COMP-THERM-50 experiments, put in ICSBEP Handbook [7]).

Table 2 (A) presents calculation-experiment comparison (C/E-1) (%) on FP reactivity worth, depending on the reference (model or average Keff water cases).

3. Cadarache Experiments

3.1 Description

The Cadarache experiments consist in measuring the BUC nuclide reactivity worth by the oscillation technique in the MINERVE experimental reactor.

The oscillation technique consists in oscillating periodically the central pin of the MELODIE lattice through the core, so the sample under study is alternatively in and out of the core. A rotating control rod is automatically operated so as to maintain the count rate of a flux detector. The corrected rotation amplitude is in very close linear relationship with the sample reactivity.

By using various specific appropriate lattices in the test zone located at its centre, MINERVE can provide a large range of spectra. Two UOX configurations were implemented. The first one, R1-UO2 (Figure 1), is devoted to storage and transportation. The second one, R2-UO2 (Figure 2), aims to mock-up the softer spectrum corresponding to the optimum moderation-ratio in a fuel dissolver.

Each sample is measured at least three times in order to avoid systematic errors. The fission products poisoning worth is directly derived by subtracting the fission product sample reactivity from a reference sample reactivity that differs just by its lack of fission products.

Two kinds of PWR-type samples were manufactured:

- *calibration samples*: fresh UO_2 with increasing U enrichment and borated samples to relate experimental signal and calculated reactivity (^{235}U and ^{10}B are well-known isotopes),
- *separated fission product isotopes* added with different matrices (natural UO_2 or inert) to validate every CBU fission product poisoning worth.

For each sample, 13 pellets were manufactured. 10 of them constitute the sample, and the 3 others were used for chemical or mass spectrometer analysis. Each mass of fission product isotope by sample was optimized in order to obtain a similar reactivity worth corresponding to the maximum accuracy in MINERVE worth measurements. For the most resonant absorbers, i.e. ^{133}Cs-^{103}Rh-^{109}Ag, several samples have been manufactured with increasing fission product isotope amount, in order to investigate the resonance self-shielding effect.

3.2 Experiments Analysis

The MINERVE reactivity worth measurements were computed with the French criticality calculation package CRISTAL V1 [4], using the APOLLO2 code [8] (version V2.5) and its CEA93 library (version V6). Neutron fluxes in the MINERVE R1-UO2 and R2-UO2 Test Zones were obtained by a 2D transport calculation. The P_{ij} method was used in APOLLO2 in order to account for the exact heterogeneous geometry. The resonance self-shielding is rigorously calculated, for all fission products, through effective cross-section formalism.

3.3 Results

Calculation to experiment comparisons on 6 fission products (common to the Valduc programme) reactivity worth in R1-UO2 and R2-UO2 configurations are summarized in Table 2 (B): the thermal absorbers capture cross-section of ^{143}Nd, ^{149}Sm and ^{155}Gd are underestimated. ^{103}Rh (n,γ) cross-section tends to be overestimated by approximatively 12 %. It could be due to chemical analysis problem, investigation about it is in progress. The ^{133}Cs resonant capture is overestimated by + 4 %, while the ^{152}Sm capture cross-section seems to be well known.

The final uncertainty combines three independent uncertainties corresponding to:

- The knowledge of the fission product mass introduced in the sample,
- The reactivity measurement itself,
- The calibration of the signal.

4. Comparison of Cadarache and Valduc Experiments

4.1. Energy zone of absorption qualification

Table 3 gives the FP cumulative normalized absorption ratio in the neutron flux of the experiment cell for Cadarache and Valduc experiments, which are displayed in Figures 3.1, 3.2 and 3.3 for three PF, ^{103}Rh, ^{149}Sm and ^{152}Sm: the qualification energy zones are roughly the same, although the ratio values (q) from Cadarache experiments seems to define different energy zones.

4.2 Sensitivity

For comparing the VALDUC experiments sensitivity against MINERVE's one, calculations with MORET4 Perturbation [6] were made to obtain the impact of FPs concentration variation (10%), with a standard deviation $\sigma \sim 0$. Table 4 points out that a low overestimate or underestimate capture cross-section (about 4 %), detected through MINERVE experiments, will induce smaller differences (Δk = 32 to 72 10^{-5}). This shows the present sensitivity limits of VALDUC experiments in the field of nuclear data validation. It can be stated that, all data being constant, these experiments can surely detect absorption cross-section variations of 1.2% for $\Delta k = 20.10^{-5}$. This is pessimistic, because theoretically, MORET4 Perturbation can detect a smaller variation about $\Delta k = 5.10^{-5}$.

5. Conclusion

Till now, only so called VALDUC 'Physical' type experiments are evaluated, which are a little more in the thermal range than the MINERVE experiments. We intend to continue the comparison with other set of VALDUC Experiments (so called 'Elementary' and 'Advanced' Dissolution type, the first ones being now completed and evaluated).

Nevertheless, up to now, the comparison of the various VALDUC and CADARACHE experimental results gives a very high consistency of the whole complementary French qualification programmes on fission products for Burnup Credit applications: MINERVE experiments are mainly devoted to nuclear data validation, and VALDUC experiments to the CRISTAL route APOLLO2 MOTET4 qualification. They are complementary, both of a great quality, and their accurate results give a high degree of confidence to the whole qualification experimental French process devoted to BUC.

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Table 1 Fission products contribution to UOX spent fuel reactivity loss, ΔK/K (10^{-5})
PWR 17x17 - Initial enrichment = 3.5w% ^{235}U - Cooling Time = 5 years

N°	FP↓ BU→	20	40	60	FP↓ BU→	20	40	60	N°
1	^{149}Sm *	980	1030	1050	^{99}Tc, T=2,1.10^{5}a	240	440	610	**8**
2	^{103}Rh *	790	1360	1700	^{145}Nd	230	410	540	**9**
3	^{143}Nd *	530	900	1100	^{153}Eu	150	390	610	**10**
	^{131}Xe (g)	470	790	940	^{95}Mo *	150	290	400	**11**
4	^{133}Cs *	420	750	1010	^{147}Sm	150	230	270	**12**
5	^{155}Gd *	390	1550	2990	^{147}Pm, T=2,6a	120	140	130	
6	^{151}Sm, T=90a	350	500	600	^{150}Sm	120	270	380	**13**
7	^{152}Sm *	250	490	660	^{109}Ag	100	250	360	**14**
					^{101}Ru	100	220	330	**15**
15 FP ΔK/K (10^{-5}) (%)		4950 **(81)**	9080 **(79)**	12610 **(78)**	7 FP ΔK/K (10^{-5}) (%)	3120 **(51)**	6370 **(55)**	8910 **(55)**	
200 FP		**6120**	**11500**	**16200**					

BU (GWd/t), (g)= gaseous, * 7 = 6 initial IRSN choice + ^{95}Mo, recently added. 15 = OECD/CEA choice

Table 2 FP reactivity worth (C-E)/E in %

	A / Valduc Physical Type Experiments		B / Minerve Experiments	
Fission Products	E = Model Exp. Keff (C/E)-1 (%)	W = Average Water Keff (C/W)-1 (%)	R1-UO2 (C/E)-1 (%)	R2-UO2 (C/E)-1 (%)
^{143}Nd	0.155 (as Nd nat)	- 0.013 (as Nd nat)	-4.5 ± 2.5	-10.0 ± 3.0
^{149}Sm	0.182	- 0.013	-5.7 ± 2.1	-9.8 ± 2.5
^{152}Sm	0.061	- 0.0135	-0.2 ± 3.2	-1.2 ± 4.2
^{103}Rh	0.213	0.017	+13.3 ± 4.0	+11.9 ± 3.8
^{155}Gd	0.182	- 0.014	-1.9 ± 2.9	-11.0 ± 3.6
^{133}Cs	0.103	- 0.078	4.3 ± 1.9	2.0 ± 2.0

Table 3 Normalized cumulative absorption ratio from Upper Energy Limit down to 0.625 eV
(Fast and Epithermal range)

	CADARACHE		VALDUC		CADARACHE		VALDUC
	R1-UO2	R2-UO2	PF PHYS		R1-UO2	R2-UO2	PF PHYS
^{103}Rh	5.56 10^{-1}	3.91 10^{-1}	3.01 10^{-1}	^{155}Gd	1.16 10^{-2}	5.58 10^{-3}	2.80 10^{-3}
^{152}Sm	4.78 10^{-1}	3.28 10^{-1}	4.08 10^{-1}	^{149}Sm	1.41 10^{-2}	7.73 10^{-3}	5.04 10^{-3}
^{143}Nd	1.15 10^{-1}	6.10 10^{-2}	2.99 10^{-2}	^{133}Cs	7.24 10^{-1}	5.78 10^{-1}	4.25 10^{-1}

Table 4 Example of MORET4 Perturbation Calculation Results

Experiment		Perturbation PF			Experiment		Perturbation PF		
PF	C PF (g/l)	C + 10%	ΔC (%) or cross section variation at C = constant		PF	C PF (g/l)	C + 10%	ΔC (%) or cross section variation at C = constant	
		ΔK 10^{-5}	for 20 10^{-5}	for 5 10^{-5}			ΔK 10^{-5}	for 20 10^{-5}	for 5 10^{-5}
^{103}Rh	~39,0	183	1,05	0,247	^{152}Sm	~47,3	191	1,04	0,268
^{103}Rh	~19,5	138	1,46	0,364	^{152}Sm	~23,6	184	1,09	0,273
^{133}Cs	~130	148	1,35	0,339	^{155}Gd	~0,214	157	1,28	0,320
^{133}Cs	~76,5	111	1,81	0,452	^{155}Gd	~0,107	126	1,57	0,393
Ndnat	~119,6	120	1,68	0,419	^{149}Sm	~0,215	179	1,12	0,272
Average on 32 values [9)]			1,2	0,3	^{149}Sm	~0.101	143	1,40	31

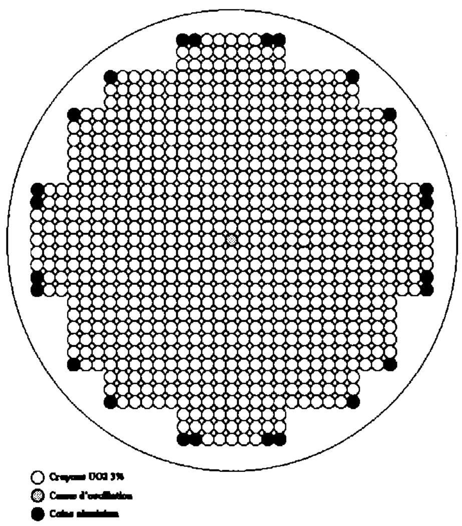

Fig. 1 MINERVE Test Zone - R1-UO2 lattice

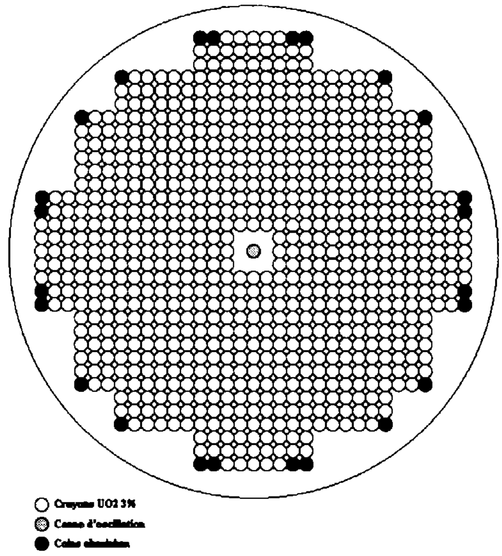

Fig. 2 MINERVE Test Zone - R2-UO2 lattice

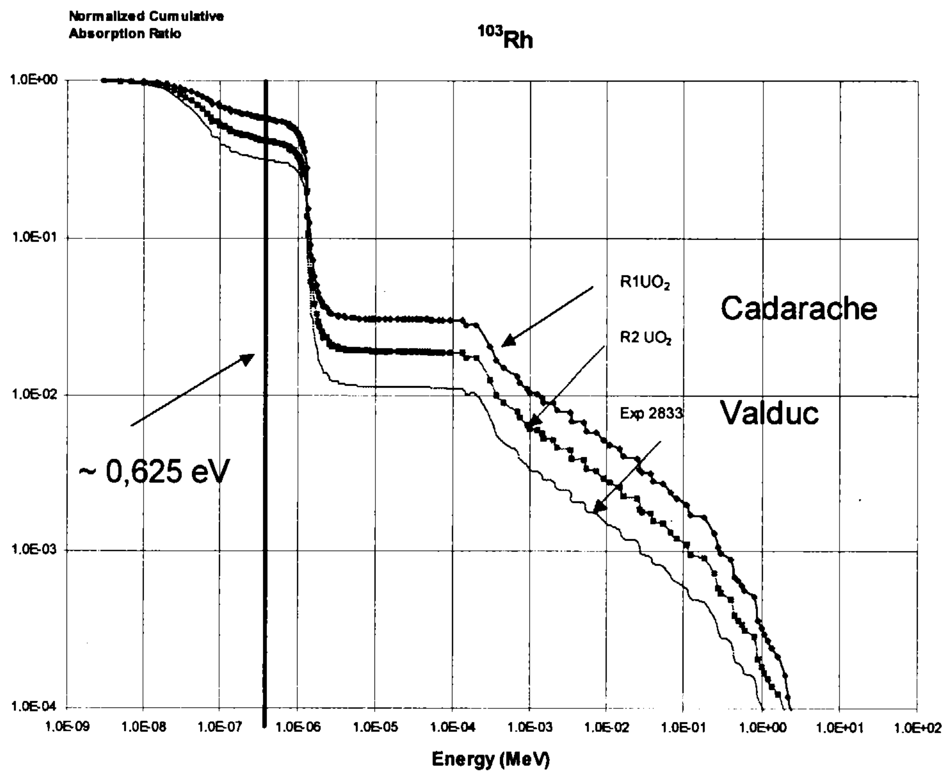

Fig. 3.1 – Variation of the Normalized Cumulative Absorption Rate from Upper Energy Limit

$$\frac{\sum_{1}^{i} \sigma_{ai} * n * \varphi_i}{\sum_{1}^{172} \sigma_{ai} * n * \varphi_i}$$ *with σ_{ai} = absorption cross section in group i,*

and n = atomic density of the fission product, φ_i = neutron flux in group i.

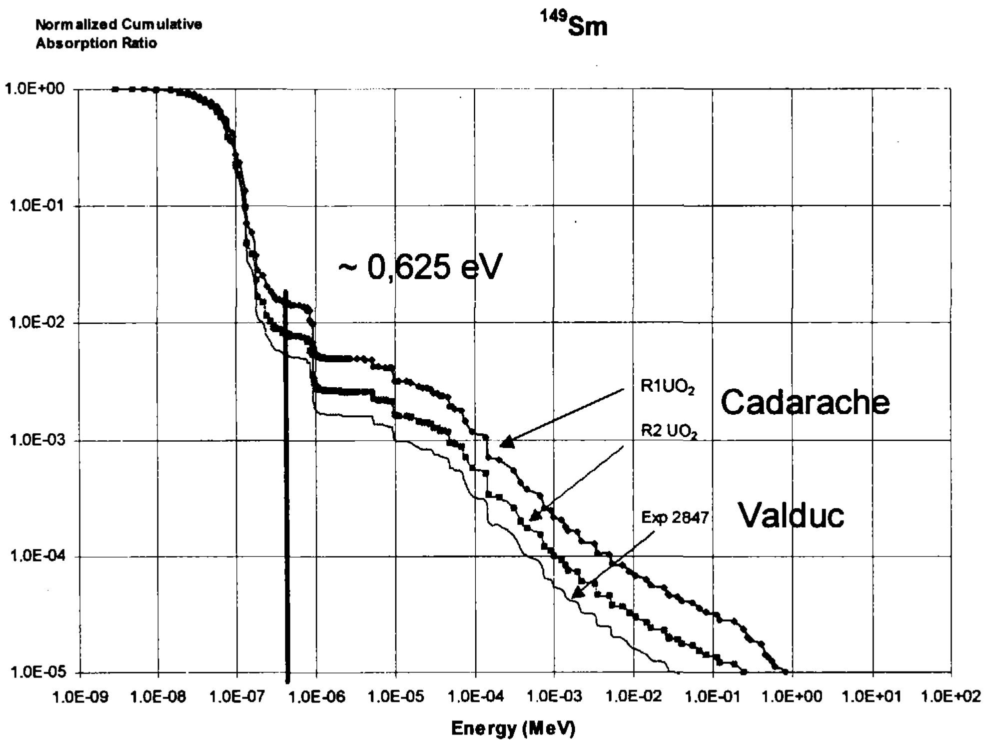

Fig. 3.2 – Variation of the Normalized Cumulative Absorption Rate from Upper Energy Limit

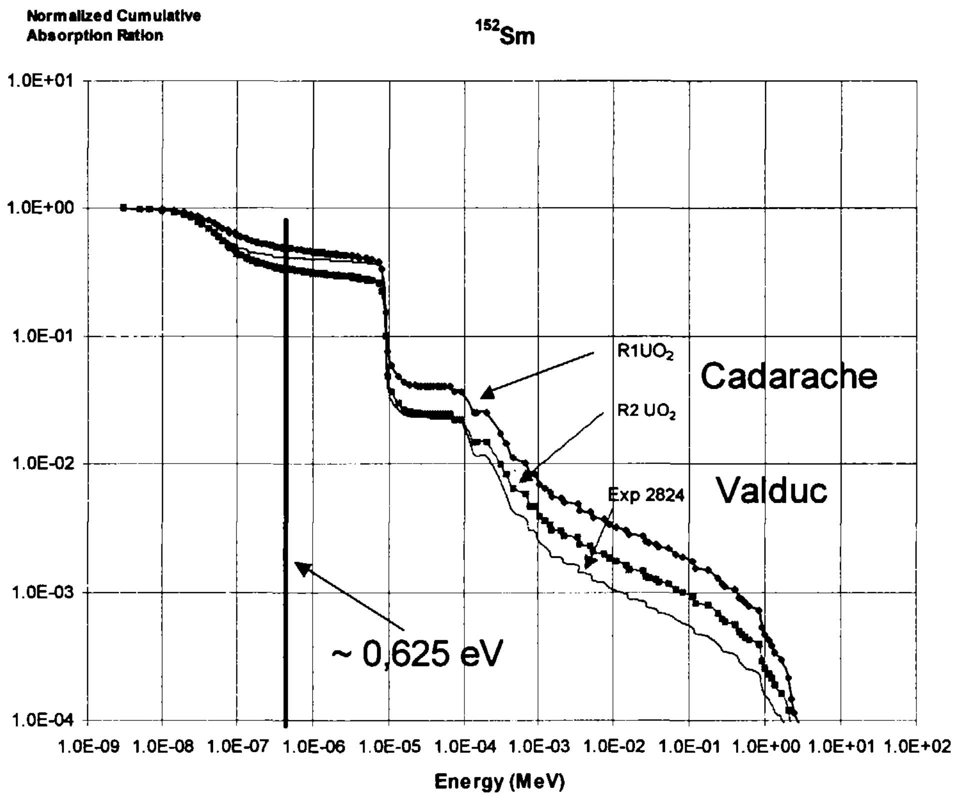

Fig. 3.3 – Variation of the Normalized Cumulative Absorption Rate from Upper Energy Limit