

Fission Products Experimental Programme

Nicolas Leclaire^{*1}, Jacques Anno^{**2}, Emmanuel Girault³, Eric Létang¹

¹ Institut de Radioprotection et de Sûreté Nucléaire (IRSN), BP 17 -
92262 Fontenay-aux-Roses Cedex, France

² Commissariat à l'Énergie Atomique (CEA)/Institut de Protection et de Sûreté Nucléaire
(CEA/IPSN),

³ Commissariat à l'Énergie Atomique (CEA), Centre de Valduc,
21120 Is-Sur-Tille, France

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Abstract

The “Fission Products (FPs)” experimental programme was carried out from 1998 to 2004 in CEA/Valduc Apparatus B in the framework of the IRSN-AREVA NC Common Interest Program dealing with “burnup credit” studies. It aims at compensating for the lack of critical experiments with FPs in the literature and validating a selection of 6 stable, non-volatile, fission products representing half of the irradiated fuel absorption of all fission products: ¹⁰³Rh, ¹³³Cs, ¹⁴³Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd. This paper provides a more exhaustive approach of the k_{eff} results, uncertainties and tendencies associated with the different steps of the “Fission Products” programme.

1. Introduction

In their safety analyses, nuclear fuel managers now take into account the fuel “burnup credit” of irradiated fuels. Given the anti-reactivity margin associated with the “burnup credit”, it became necessary to have a validation of the cross-sections for a selection of Fission Products responsible for the overall anti-reactivity caused by the FPs in the irradiated fuels. As a consequence, IRSN has, on one side, performed studies in a French working group with the aim to evaluate the best way to account for the burnup credit [1] in fuels and, on the other side, investigated the need of integral critical experiments to validate the cross-sections of major actinides (HTC programme [2]) and fission products. All this work contributes to the validation of the APOLLO2-MORET 4 calculation route of the CRISTAL V1 criticality package [4].

The “Fission Products” experimental programme which aims at validating the six following Fission Products, ¹⁰³Rh, ¹³³Cs, ¹⁴³Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd has been divided into three steps to have a progressive validation.

* Corresponding author, Tel: +33 1.58.35.91.66, Fax: +33 1.46.57.29.98, E-mail: nicolas.leclaire@irsn.fr

** Retired from CEA/IPSN

These three steps related to different interacting conditions between the FPs and the uranium and plutonium nuclei are the followings:

- the first step (“Physical” type experiments) deals with the study of FPs in low acidic solutions, alone or mixed, without interaction,
- the second step (“Elementary Dissolution” type experiments) deals with FPs in low acidic or depleted uranyl nitrate (DUN¹) solutions, alone or mixed, interacting with uranium and plutonium nuclei in arrays of UO₂ or HTC² rods,
- the third step (“Global Dissolution” type experiments) deals with FPs in DUN solutions, alone or mixed, interacting with the uranium of uranyl nitrate solutions or with uranium and plutonium nuclei in arrays of UO₂ or HTC rods.

2. Description of the experiments

2.1 Conditions of realization of the programme

The Fission Products experimental programme carried out at CEA Valduc research center aimed at validating the six aforementioned fission products. One of the required conditions to do so is to highlight a sufficient anti-reactivity worth (around 4%). The FPs concentrations were adjusted to account for that. Moreover, the FPs concentrations were also determined on the basis of the FPs dilution and solubility limits, as well as by the price and available quantities of FPs at Valduc research center. For instance, ¹³³Cs crystallizes around 130 g/l and the mass of ¹⁵²Sm available was limited to 180 g [3].

2.2 “Physical type” experimental programme

The first step (“Physical” type experiments) of the FP programme involves FPs solutions, in a Zircaloy central tank. This tank (6.2 cm × 6.2 cm internal dimensions) is positioned in the center of a driver array of UO₂ rods (4.738 wt. % ²³⁵U) at a 1.3-cm square pitch, moderated and reflected by water. The fissile zone under water height is the critical approach parameter. This series gathers 45 experiments with the followings FPs: ¹⁰³Rh, ¹³³Cs, ^{nat}Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd, a mixture of FPs (¹⁰³Rh, ¹³³Cs, ^{nat}Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd) at different concentrations. Experiments without FPs and with boron were also performed.

2.3 “Elementary Dissolution type” experimental programme

The second step (“Elementary Dissolution” type experiments) of the FP programme involves FPs solutions, in slightly acidic solutions, alone or mixed, in a Zircaloy central tank (14.2 cm × 14.2 cm internal dimensions) containing an array of UO₂ or HTC rods at a 1.271-cm square pitch. This tank is positioned in the center of a driver array of UO₂ rods (4.738 wt. % ²³⁵U) at a 1.3-cm square pitch, moderated and reflected by water. The fissile zone under water height is the critical approach parameter.

This series of experiments can be divided into two sub-groups:

- a first one gathering 68 experiments involving the following FPs: ¹⁰³Rh, ¹³³Cs, ^{nat}Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd and a mixture of FPs (¹⁰³Rh, ¹³³Cs, ^{nat}Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd) at different concentrations, in nitric acid solution,
- a second one gathering 21 experiments involving ¹⁴⁹Sm and a mixture of FPs (¹⁰³Rh, ¹³³Cs, ^{nat}Nd, ¹⁴⁹Sm, ¹⁵²Sm and ¹⁵⁵Gd) at different concentrations, in DUN (400 g/l).

¹ Depleted uranyl nitrate

² HTC: simulated high burnup fuel rods with U and Pu vectors consistent with burnup of 37.5 GWd/MTU, but without FPs

The HTC rods are composed of (U-Pu)O₂ (1.1% wt.% PuO₂), with uranium oxide enriched at 1.57% ²³⁵U and ²⁴⁰Pu content equal to 24.335%. The plutonium isotopic vector is given in Table 1.

Table 1: Plutonium isotopic vector of HTC rods (November, 7th 1987).

Element	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴¹ Am
Wt. %	1.304	59.227	24.335	10.076	5.056	1.432

2.4 “Global Dissolution type” experimental programme

The third step (“Global Dissolution” type experiments) of the FP programme involves:

- in a first series of experiments, an array (26 × 26) of UO₂ fuel rods (4.738 wt. % ²³⁵U), at a 1.3-cm square pitch, moderated and reflected by DUN,
- in a second series of experiments, an array (44 × 44) of HTC fuel rods (see aforementioned composition), at a 1.6-cm square pitch, moderated by DUN poisoned or not by FPs and reflected by water.

The first set of experiments involving UO₂ rods in DUN solutions (300 g/l and 100 g/l) comprises 7 cases. The second set of experiments involving HTC rods in DUN solutions (300 g/l and 92 g/l) containing or not a mixture of FPs also comprises 7 cases. In both cases, the fissile zone under DUN is the critical approach parameter. These experiments aim at validating the treatment of uranium, plutonium and americium self-shielding.

3 Results of APOLLO2-MORET 4 and TRIPOLI4 calculations

The benchmark configuration k_{eff} was calculated using CRISTAL V1.0 [4] criticality safety package. The validation of the six FPs is based on the difference between the benchmark k_{eff} (1.00000) and the calculated k_{eff} .

The validation work consists in calculating the C-E value [5], which is given by calculated k_{eff} minus benchmark's k_{eff} , and its combined standard deviation ($\sigma = \sqrt{\sigma_{\text{calculation}}^2 + \sigma_{\text{benchmark}}^2}$).

Calculations are considered in good agreement with the benchmark when the discrepancies are in the uncertainties ranges (depending on the combined standard deviation and on the confidence interval).

The calculations were performed using the standard route (APOLLO2-MORET 4) associated with the CEA93 V6 172-energy group cross-section library based on JEF2.2 evaluation and the point-wise reference route (TRIPOLI4 [6]) of the CRISTAL criticality package. The reactivity worth of FPs in solution has been evaluated by the difference between the k_{eff} of the configuration with the fission product and the configuration without the fission product. All the results are provided in the following Tables. It should be noted that for confidentiality reasons the FPs concentrations have been intentionally rounded.

3.1 “Physical type” experimental programme

The k_{eff} results are reported on Fig.1 and in Table 2. It should be noticed that:

- the anti-reactivity worth of FPs depends on the nature of the FP (1.5% to 4%); it is limited for ¹³³Cs and ¹⁴³Nd (1.5% to 2%) for solubility reasons and for ¹⁵²Sm due to a restricted available quantity,
- a good agreement between the benchmark k_{eff} (1.00000) and the calculated k_{eff} is obtained,

- the same agreement is obtained with point-wise TRIPOLI4 reference route using JEF2.2 evaluation; however, the use of ENDF/B-VI cross-sections library provides an underestimation of the k_{eff} .

A TRIPOLI4 calculation performed with a slightly acidic solution without FPs in the central tank shows that the discrepancy between the two libraries used is of the same order of magnitude. As a consequence, it seems that the difference could not be attributed to the FP cross-sections but rather to the uranium or plutonium cross-sections (driver array).

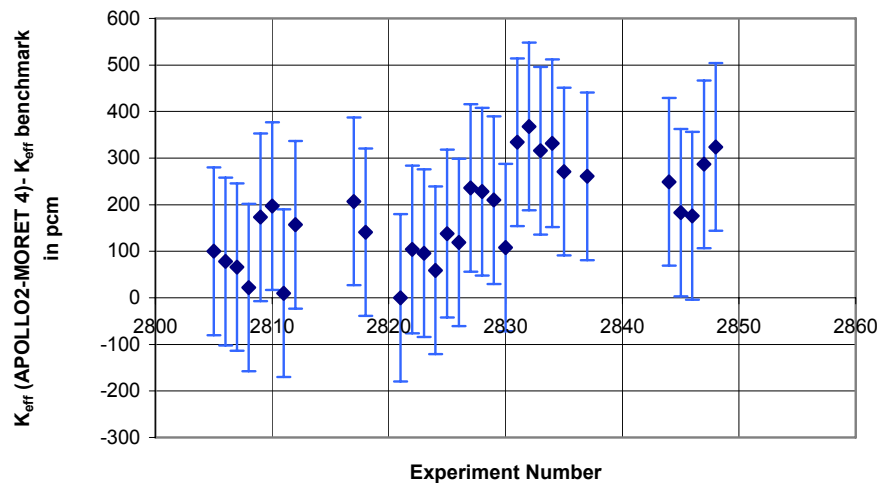
A more precise analysis of the influence of the FP nature on the k_{eff} shows that the difference between the calculated k_{eff} and the benchmark k_{eff} , for the majority of configurations, is lower than the combined standard deviation.

Finally, given that the flux level in the central tank and in the driver array is not necessarily the same, it was considered to be worth calculating the FPs cross-sections in the central tank with the appropriate flux weighting. This work is still in progress.

Table 2: k_{eff} results and FP anti-reactivity worth of “Physical type” experiments.

Case	Exp. N°	C(FP) g/l	FP or solution type	APOLLO2-MORET 4 Calculated k_{eff}	FP worth in 10^{-5}	TRIPOLI4 Calculation k_{eff}	
						Library	
						JEF2.2	ENDF/B-VI
2	2834	40	¹⁰³ Rh	1.00332	2798	1.00109	0.99634
3	2835	20	¹⁰³ Rh	1.00271	1793		
8	2811	140	¹³³ Cs	1.00010	2169	0.99990	0.99536
11	2809	70	¹³³ Cs	1.00173	1448	0.99910	0.99346
13	2812	70	¹³³ Cs	1.00157	1314	0.99929	
16	2821	130	Ndnat	1.00000	1470	0.99948	0.99547
18	2823	50	¹⁵² Sm	1.00096	4474	0.99987	0.99604
21	2826	20	¹⁵² Sm	1.00119	3361		
23	2828	0.2	¹⁵⁵ Gd	1.00228	2705		
26	2831	0.1	¹⁵⁵ Gd	1.00334	1932		
28	2844	Rh 7 Cs 30 Gd 0.05 Sm2 1.5 Sm9 0.04 Ndnat 25	Mixt.	1.00249		1.0000	0.99562
31	2847	0.2	¹⁴⁹ Sm	1.00287	3592		
32	2848	0.1	¹⁴⁹ Sm	1.00324	2246		

Figure 1: Difference between the calculated k_{eff} and the benchmark k_{eff}



Finally, a good agreement calculation/experiment can be concluded.

3.2 “Elementary Dissolution type” experimental programme

The k_{eff} results are reported on Fig.2 and in Tables 3 and 4. It should be noticed that:

- the anti-reactivity worth of FPs depends on the nature of the FP (1.5 to 6%); it is limited for ^{133}Cs and ^{143}Nd (1.5 to 2.8%) for solubility reasons and for ^{152}Sm due to a restricted available quantity,
- a good agreement between the benchmark k_{eff} (1.00000) and the calculated k_{eff} is obtained with a slight tendency to overestimate k_{eff} for all FPs and mixture of FPs except for ^{103}Rh ,
- the same agreement is obtained with point-wise TRIPOLI4 reference route using JEF2.2 evaluation; however, ENDF/B-VI cross-sections library underestimate the k_{eff} .

A TRIPOLI4 calculation performed with UO_2 rods in the central tank in a slightly acidic solution without FP shows that the discrepancy between the two libraries is of the same order of magnitude. As a consequence, it seems that the difference could not be attributed to the FPs cross-sections but rather to the uranium or plutonium cross-sections. Moreover, the k_{eff} difference between the APOLLO2-MORET 4 and TRIPOLI4 code systems for the cases with and without FPs in the central tank being the same, it seems that the self-shielding and homogenization of FPs cross-sections with APOLLO2 code is properly processed.

A more precise analysis of the influence of the FP nature on the k_{eff} shows that the difference between the calculated k_{eff} and the benchmark k_{eff} is higher than the combined standard deviation for the following cases: mixture of FPs, ^{133}Cs , ^{149}Sm , ^{152}Sm and ^{155}Gd , except for ^{103}Rh , allowing to conclude to a slight overestimation tendency.

Table 3: k_{eff} results and FP anti-reactivity worth of “Elementary Dissolution type” experiments – UO₂ rods in the central tank.

Case	Experiment N°	Concentration (g/l)	FP or solution type	APOLLO2-MORET 4 Calculated k_{eff}	FP worth in 10 ⁻⁵	TRIPOLI4 Calculation k_{eff}	
						Library	
						JEF2.2	ENDF/B-VI
1	2875	110	¹³³ Cs	1.00360	2823	1.00033	0.99699
6	2883	50	¹⁰³ Rh	0.99906	6241		
7	2884	50	¹⁰³ Rh	0.99971			
10	2925	20	¹⁰³ Rh	0.99925	3626	0.99664	0.99293
11	2880	90	¹⁴³ Nd	1.00508	1568	1.00337	0.99750
12	2881	90	¹⁴³ Nd	1.00467			
13	2897	0.4	¹⁴⁹ Sm	1.00382	6594		
14	2898	0.4	¹⁴⁹ Sm	1.00407			
16	2929	0.15	¹⁴⁹ Sm	1.00412		1.00124	0.99609
18	2901	15	¹⁵² Sm	1.00365	3522		
20	2900	0.8	¹⁵⁵ Gd	1.00528	6933		
21	2926	0.4	¹⁵⁵ Gd	1.00329	3762	1.00042	0.99628
23	2943		Mixt1	1.00345			
25	2939		Mixt2	1.00546			
27	2937		Mixt3	1.00688		1.00389	0.99955

Figure 2: Difference between the calculated k_{eff} and the benchmark k_{eff} – UO₂ rods in the central tank

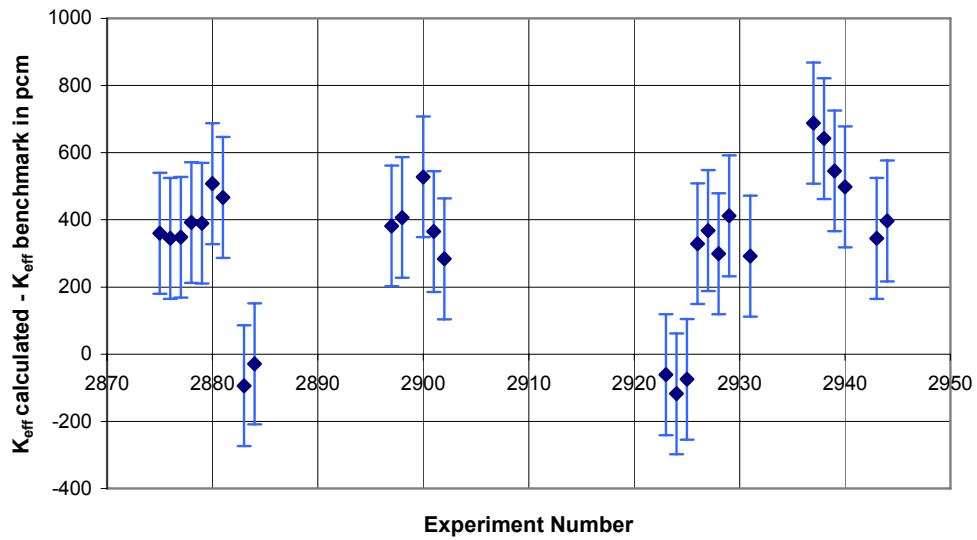


Table 4: k_{eff} results and FP anti-reactivity worth of “Elementary Dissolution type” experiments – HTC rods in the central tank.

Case	Experiment N°	Concentration (g/l)	FP or solution type	APOLLO2-MORET 4 Calculated k_{eff}	FP worth in 10^{-5}
55	2910	30	¹⁰³ Rh	1.00198	3206
57	2912	0.4	¹⁵⁵ Gd	1.00281	3436
59	2915	0.2	¹⁴⁹ Sm	1.00224	3514
61	2917		Mixt4	1.00253	

3.3 “Global Dissolution type” experimental programme

This series of experiments can be divided into two sub-series:

- one that can be used as a test series carrying out UO₂ rods at a square pitch of 1.3 cm in a DUN solution,
- another one with HTC rods in a DUN solution with FPs.

The k_{eff} results are reported on Fig.3 and in Tables 5 and 6. It should be noticed that:

- a good agreement between the benchmark k_{eff} (1.00000) and the calculated k_{eff} is obtained for the experiments with UO₂ rods at a 1.3-cm square pitch in the DUN solution without FPs,
- a tendency to overestimation is obtained for the experiments with HTC rods at a 1.6-cm square pitch and with FPs as well as without,
- the same agreement is obtained with point-wise TRIPOLI4 (JEF2.2 evaluation) reference route; however, ENDF/B-VI cross-section library underestimates the k_{eff} .

Table 5: k_{eff} results and FP anti-reactivity worth of “Global Dissolution type” experiments – UO₂ rods in a uranyl nitrate solution without FPs.

Case	Experiment N°	C(DUN) (g/l)	FP or solution type	APOLLO2-MORET 4 Calculated k_{eff}	TRIPOLI4 Calculation k_{eff}	
					Library	
					JEF2.2	ENDF/B-VI
1	2967	300	DUN	0.99962	0.99699	0.99198
5	2970	92	DUN	0.99872		

TRIPOLI4 calculations were run with the JEF2.2 and ENDF/B-VI libraries on the “Global Dissolution” experiments with HTC rods and FPs. A very good agreement is obtained between the two libraries. It seems that the difference observed in the previous paragraphs has been compensated by the fact that the pitch between rods is not the same (greater) and that the whole tank is filled with HTC rods (the reactivity worth being attributed only to these rods).

Moreover, on the same “Global Dissolution” programme, a 0.4% discrepancy (slight overestimation) between APOLLO2-MORET 4 (JEF2.2 library) and TRIPOLI4 (JEF2.2 library) with and without FPs in the DUN solution can be noticed. The self-shielding and multi-group treatment of cross-sections seems to be responsible for this small k_{eff} difference.

Table 6: k_{eff} results and FP anti-reactivity worth of “Global Dissolution type” experiments – HTC rods in a uranyl nitrate solution with or without FPs.

Case	Experiment N°	C(DUN) g/l	FP or solution type	APOLLO2-MORET 4 Calculated k_{eff}	TRIPOLI4 Calculation k_{eff}	
					Library	
					JEF2.2	ENDF/B-VI
1	2974	300	DUN	1.00404	1.00040	1.00039
2	2975	300	DUN	1.00410		
3	2976	300	DUN	1.00413		
4	2977	92	Mixt.	1.00541	1.00221	1.00134
5	2978	92	Mixt.	1.00534		
6	2979	92	Mixt.	1.00537		
7	2980	92	Mixt.	1.00481		

3.3 FP absorption efficiency

The normalized cumulated absorption rates have been plotted against energy range in Fig.4 for the six FPs of the “Elementary Dissolution type” experimental programme. The so-called normalized cumulated absorption rate is used to determine and to observe in which energy zone the absorption rate of the FPs is 1%, 10%, or 90% of the total (Table 7). The related curve against energy shows steps more or less high, related to cross-sections peaks and flux level in the central tank. The absorption energy range of each FP can be clearly defined: ^{149}Sm , ^{155}Gd and ^{143}Nd absorb neutrons only in the thermal energy range; ^{103}Rh , ^{152}Sm and ^{133}Cs absorb neutrons on a larger energy range.

Figure 4: Mixture validation

Validation of mixture 1 absorption cross-sections

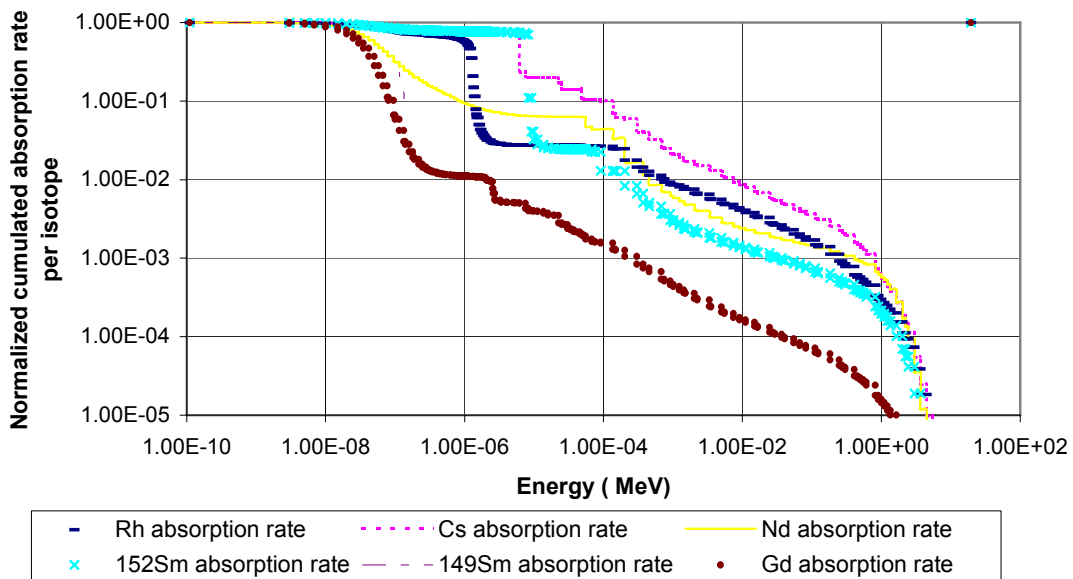


Table 7: Absorption energy zone of FPs.

FP	Rate = 1%	Rate = 10%	Rate = 90 %
¹⁰³ Rh	~ 100 eV	~ 1.5 eV	~ 0.01 eV
¹³³ Cs	1 keV	~ 20 eV	~ 0.01 eV
¹⁴³ Nd	~ 60 eV	~ 0.1 eV	~ 0.01 eV
¹⁵² Sm	~ 0.2 keV	~ 9eV	~ 0.01 eV
¹⁵⁵ Gd	~ 0.15 eV	~ 0.07eV	~ 0.01 eV
¹⁴⁹ Sm	~ 0.2 eV	~ 0.15 eV	~ 0.01 eV

4 Evaluation of experimental uncertainties

4.1 Evaluation method

The experimental uncertainties were evaluated for the three main steps of the Fission Products experimental programme. The Fission Products experimental programme was carried out from 1998 to 2004. The Pré-FP programme [7], which aimed at providing first data to improve the “Physical type” experimental programme, used dimensions issued from tolerances (especially for the rods, clads, etc...) for uncertainty calculations. The UO₂ rods initially clad with AGS were re-clad in 1994. It was the opportunity to perform measurements on the rods. As a consequence, the uncertainties were assessed much more precisely.

In the case measurements were performed, the experimentalists provide the level of confidence at which the measurement is known. The 1 σ uncertainty is directly derived.

In the case only tolerances are available, the evaluator must do assumptions on the distribution of measurements. Most of the time, the equiprobable distribution is assumed and the tolerance must be divided by square root of three to obtain the 1 σ uncertainty.

An APOLLO2-MORET 4 Monte-Carlo perturbation calculation or deterministic APOLLO2 calculation using the S_n method is then performed to evaluate the reactivity worth of the 1 σ uncertainty, the parameter being varied from 1 σ . A Δk_{eff} corresponding to the parameter variation is obtained.

A specific treatment has been applied on impurities:

- the measured detected ones are modeled in UO₂ fuel,
- the impurities are not modeled in FPs and in HTC fuel and are considered to account as an uncertainty,

These Δk_{eff} are combined quadratically (making the assumptions that the uncertainties are uncorrelated) to obtain the general uncertainty.

4.2 Results

It can be noted that the global uncertainty is lower than 0.06%, which is low. Therefore, it can be concluded that the methodology introduced by the experimentalists led to small uncertainties. In particular, the impact of the chemical uncertainties on global uncertainty is very low ($< 5.10^{-5}$). The main uncertainties are those on isotopic content (15 to 30.10⁻⁵), rod positioning (20.10⁻⁵) and oxide impurities in UO₂ rods (35.10⁻⁵). The oxide impurities in UO₂ and HTC rods have a paramount effect.

The global uncertainty value is approximately the same for the three steps of the programme (40 to 60.10⁻⁵) and coherent with what was assumed in the Pré-FP programme.

The low value of global uncertainties enables to use the experiments for validation purpose of the APOLLO2-MORET 4 calculation route.

5. Conclusion

From 1998 to 2004, around 150 critical experiments (some are test experiments without FPs) with Fission Products in various configurations have been carried out and evaluated. These configurations aim at covering the main steps of reprocessing and fuel management of the French fuel cycle. They aimed at validating the FPs cross-sections in the thermal energy range to clearly evaluate the anti-reactivity margins delivered by the burnup credit practice. For this reason, a progressive validation of 6 FPs has been performed successfully: ^{103}Rh , ^{133}Cs , ^{149}Sm , ^{152}Sm , ^{155}Gd and ^{143}Nd . These experiments are all the more valuable as they show very small uncertainties and as their calculated k_{eff} is very close to the benchmark k_{eff} . Only a small tendency to overestimation ($< 0.5\%$) has been noticed for “Elementary Dissolution” and “Global Dissolution” type experimental programmes and especially with ^{143}Nd and FPs mixtures.

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