INVESTIGATION OF MINOR ACTINIDE TRANSMUTATION IN NEPTUNIUM FUELLED BFS67 ASSEMBLIES

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ABSTRACT

In order to reduce the growing amount of Neptunium produced in reactors, Russia and France are currently investigating the possibility of using fast reactors to burn Neptunium[1,2,3].

The BFS67 experimental programme, established in the framework of a joint Russia-France agreement, has been set up to confirm the validity of this possibility considering a fast reactor design where Np237 is homogenised uniformly in the fuel. Four cores have been built for this purpose.

The BFS67-1 core is a conventional fast reactor core mock-up which serves as a reference core for the Np237 burner core design. The core has a central enriched plutonium zone surrounded by a uranium driver zone. In the central part of the BFS-67-1 core, a zone with different Neptunium content (13.1% then 6.5%) was introduced to create the BFS67-2, 3 and 3B cores.

Measurements in these cores concern the critical masses, spectral indices, sodium void reactivity worth and the central reactivity worth of various materials. The measured values observed provide a method of validating the expected changes of the core characteristics due to the introduction of Neptunium.

The transposition of the experimental results to full power reactors requires calculation analysis, with particular attention being given to the modelling of both reactor and experimental devices. Several of the BFS67 experiments have been analysed using the JEF2[4]/ECCO[5]/ERANOS[6] calculational scheme which includes the algorithmic features required to perform such a refined analysis.

INTRODUCTION.

During recent years investigations have been carried out to find a way of performing the transmutation of Minor Actinides (MA) accumulated at the end of spent fuel reprocessing. Due to their high flux and hard energy spectrum, fast reactors have the most suitable properties for this purpose. Experiments have been defined at the IPPE BFS facility[7,8], in the framework of the SPRC/IPPE collaboration for the investigation of minor actinide transmutation in

Neptunium fuelled BFS67 assemblies. The purpose of experimental program is to determine the adequacy of the calculation for a reactor design with minor actinides (MA) in the fuel.

A new stage of experimental investigations has started in 1993 when about 250 pellets were manufactured from approximately 10 kg of neptunium dioxide This allowed for the first time the performing of mock-up type measurements for observing the influence on the main reactor characteristics of the addition of Neptunium.

Several types of measurements were carried out:

- critical mass;
- ratios of average fission and capture cross-sections at the core centre.
- reactivity coefficients of reactor material samples at the core centre.
- sodium void reactivity worth.
- control rod worth mock-ups using natural and enriched boron carbide (B_4C).

The transposition of experimental results to full power reactors requires refined calculation analysis, with particular attention being given to the modelling of the reactor and experimental devices.

The calculational scheme JEF2/ECCO/ERANOS includes the features to pursue such objectives:

- the recent JEF2 evaluated data file.
- the ECCO cell code with a 3D representation of the fuel pellets in the assembly tubes,
- the S4 P1 transport theory code BISTRO for a flux calculation in a 2D RZ geometrical representation of the cores.

In this paper, the description of the critical assemblies and the experimental results are presented. A precise analysis of the experiments has been performed using the JEF2/ECCO/ERANOS code and data system.

DESCRIPTION OF THE BFS-67 CRITICAL ASSEMBLIES.

The BFS-67-1 critical assembly is a reference core with a cell composition typical of internal core of Superphenix with a near 19% Plutonium content. The BFS-67-1 critical assembly has a plutonium fuelled central zone surrounded by an uranium driver zone. Axial and radial blankets contain depleted uranium dioxide.

Three other cores have been defined by introducing Np237 in a central core region:

- BFS67-2 with a central region of 31 drawers and a Neptunium enrichment of 13.1%,
- BFS67-3 with a central region of the same size but with a reduced Neptunium enrichment of 6.5%,
- BFS67-3B with an enlarged central region of 61 drawers and the same Neptunium enrichment (6.5%) as in BFS67-3.

The change of the BFS-67-1 core structure was carried out by the replacement of 31 central fuel rods (FR) containing UO_2 pellets by NpO_2 ones in a such way that the core cells had either two NpO_2 pellets (BFS-67-2 core, 13.1% neptunium content), or one pellet (BFS-67-3 and 3B cores, 6.5% neptunium content).

The layout of the BFS-67-1 assembly is presented in Fig.1.

Zone 1 includes 169 fuel rods with active core cells containing plutonium pellets (4.4 % of Pu^{240}) with an enrichment of 18.6 %.

Zone 2 (uranium driver zone) includes 417 fuel rods with active core cells containing uranium pellets with an enrichment of 21.3 %.

Zone 3 is the reflector zone which consists of 827 rods of depleted uranium dioxide located inside stainless steel pins (1.9 pins per tube).

In the BFS-67-2, 3 and 3B assemblies, the Neptunium fuel is found in zone 5, located in the central part (Fig.2). In the BFS-67-2 and BFS-67-3 cores, zone 5 includes 31 fuel rods with a radius is 14.91 cm. In the BFS-67-3B core, this zone includes 61 fuel rods with a radius of 20.91 cm.

In the BFS-67-2 assembly, zone 2 (uranium driver) consists of 420 fuel rods with a thickness of 30.18 cm. In the BFS-67-3 assembly, it consists of 419 fuel rods with a thickness of 30.12 cm, and in the BFS-67-3B it consists of 421 fuel rods, its thickness 30.23 cm.

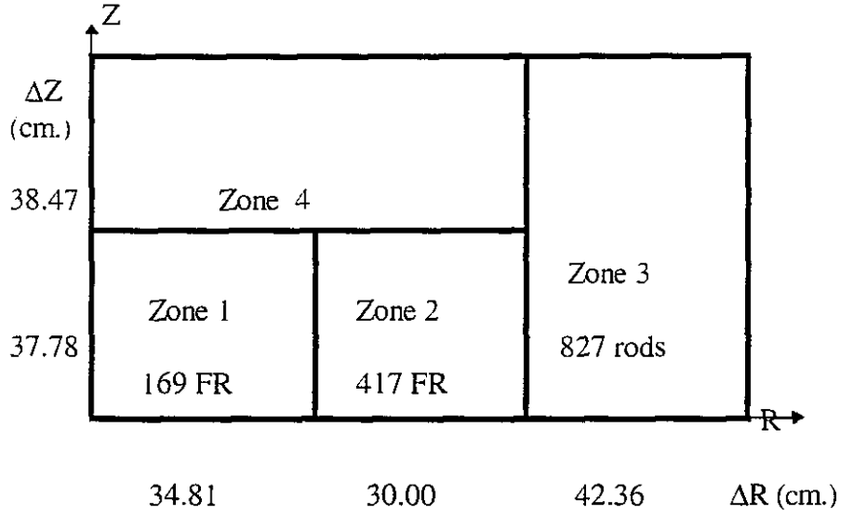

FIGURE 1 R-Z model of BFS-67-1 assembly (1/4 of reactor).

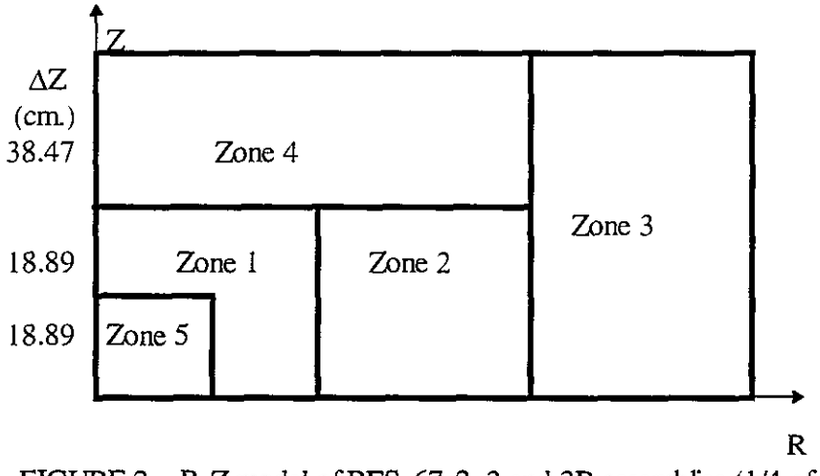

FIGURE 2 R-Z model of BFS-67-2, 3 and 3B assemblies (1/4 of reactor).

EXPERIMENTAL RESULTS

Critical Mass

The critical masses of the BFS-67 assemblies are presented in Table 1. Loading is using Fuel Rods (FR) to get the critical masses. Criticality is achieved by rod insertion. Rod insertion reactivity is obtained by the S curve calibration.

TABLE 1. The critical masses of BFS-67 assemblies.

Core	Number of FR with Np	Number of FR with Pu	Number of FR in the driver	Reactivity, % β_{eff}
BFS-67-1	-	169	417	+8
BFS-67-2	31	138	420	+7
BFS-67-3	31	138	419	+8
BFS-67-3B	61	108	421	+10

Central neutron cross-sections average ratios

Spectral indices have been measured in the central region of the three BFS cores (BFS67-1, BFS67-3B and BFS67-2).The average fission cross-section ratios are measured by three types of ionisation fission chambers: small-size chambers (SSC), segment chambers (SC) and absolute chambers (AC). For some isotopes the measurements were performed with solid state detectors (SSD).

The average capture cross-section ratios (by comparison with the fission of U-235) are measured by means of foils placed in the FR between pellets and inside a special depleted uranium dioxide pellet (U-238).

For the BFS67-2 experiments, measurements were carried out by both French and Russian teams The different techniques used with different fission chambers have given very consistent experimental results. In particular, the small fission chambers used by the French team (SSCFR) give satisfactory results.

The measured values of the average cross section ratios are given in Table 2.

TABLE 2. The measured values of the average cross section ratios for the BFS-67 assemblies.

Parameter	Detector type	BFS-67-1	BFS-67-3B	BFS-67-2
$\sigma_f U238/\sigma_f U235$	ssc	0.0322±0.0006	0.0358±0.0007	0.0380±0.0007
	sc	0.0328±0.0006	0.0361±0.0006	0.0380±0.0007
	sscfr			0.0389±0.0007
$\sigma_f Pu239/\sigma_f U235$	ssc	1.003±0.010	1.024±0.011	1.038±0.010
	sc	-	1.02±0.02	1.035±0.020
	sscfr			1.051±0.010
$\sigma_f Np237/\sigma_f Pu239$	ssc	0.236±0.005	0.248±0.006	0.258±0.006
	ac	0.232±0.005	0.241±0.005	0.254±0.005
	sc	0.234±0.007	0.252±0.008	0.259±0.008
	sscfr			0.256±0.006
$\sigma_f Pu238/\sigma_f Pu239$	ac	0.644±0.019	0.661±0.020	0.670±0.020
	sc	0.620±0.028	0.633±0.028	0.648±0.029
	ssd	0.634±0.023	0.660±0.025	0.651±0.023
$\sigma_f Pu240/\sigma_f Pu239$	ssc	0.246±0.004	0.260±0.004	0.267±0.004
	ac	0.248±0.007	0.261±0.007	0.272±0.007
	sc	-	0.259±0.008	0.272±0.008
	sscfr			0.269±0.004
$\sigma_f Pu241/\sigma_f Pu239$	ssc	1.314±0.017	1.285±0.013	1.273±0.017
	sc	-	1.284±0.007	1.274±0.017
	sscfr			1.288±0.007
$\sigma_f Pu242/\sigma_f Pu239$	ssc	0.181±0.003	0.193±0.003	0.199±0.003
	ssd	0.174±0.007	0.186±0.007	0.190±0.008
$\sigma_f Am241/\sigma_f Pu239$	ssc	0.203±0.004	0.217±0.004	0.224±0.004
	ac	0.206±0.007	0.217±0.007	0.231±0.007
	sscfr			0.205±0.004
$\sigma_f Am243/\sigma_f Pu239$	ssc	0.160±0.006	0.162±0.006	0.171±0.006
	ac	0.156±0.004	0.164±0.004	0.172±0.004
	sc	0.143±0.005	0.156±0.005	0.161±0.005
	ssd	0.150±0.006	0.158±0.006	0.169±0.006
	sscfr			0.162±0.004
$\sigma_f Cm244/\sigma_f Pu239$	ssc	0.259±0.006		0.284±0.007
	ac	0.258±0.011	0.285±0.013	0.292±0.013
$\sigma_c U238/\sigma_f U235$	Foils	0.134±0.003	0.139±0.003	0.135±0.003
$\sigma_c Au197/\sigma_f U235$	Foils	0.317±0.010	-	0.280±0.010
$\sigma_c Np237/\sigma_c U238$	Foils	5.52±0.22	5.50±0.21	5.35±0.21
$\sigma_c Np237/\sigma_f U235$	Foils	0.75±0.02	-	0.71±0.02

From these values, one can note the consequences of a spectrum hardening (on the F28/F25 for example) due to the Neptunium introduction in the core.

Sodium Void Reactivity Effect (SVRE)

The integral SVRE is measured by the replacement of sodium pellets by SS-cans in the four central cells for 31 FR (BFS - 67) . The initial reactivity state of the critical assembly (all control rods inserted in the core over the entire height) as well as the reactivity state after the change of the pellets by cans was measured by a digital reactimeter. The reloading were performed four times, and the experimental values are presented in Table3

TABLE 3. The Sodium Void Reactivity Effect.

Sodium Void Reactivity Effect, ($\%\beta_{eff}$)						
1	Number of FR voided	Position of voided cells	Mass of sodium (kg.)	BFS-67-1	BFS-67-3B	BFS-67-2
1	31	3+4+5+6	6.920	6.2±0.3		18.8±0.8
2	1	4+5	0.112	0.157±0.010	0.324±0.017	0,3980.008
3	1	3+6	0.112	0.081±0.010		0.259±0.008
4	1	2+7	0.112	-0.049±0.010		0.049±0.015
5	1	1+8	0.112	-0.214±0.010		-0.152±0.015
6	1	3+4+5+6	0.224	0.241±0.010		0.668±0.012

The experimental results presented here show the evidence of a significant increase of the sodium reactivity worth when introducing Neptunium in the fuel.

Absorber reactivity worth measurements in the core centre

Two types of absorber pellets were used: natural boron carbide and enriched (81.7% B-10) boron carbide. The measurement procedure of the absorber reactivity worth is a dynamic movement of a "long rod" from a position where the lower part (sodium) is in the core to a position where the upper part (boron) is inserted in the core. The reactor power is decreasing from a high level, and decreasing rapidly with the insertion of the rod. The reactivity worth is obtained by following the power history using the Carpenter method.

The measurement results are presented in Table 4.

TABLE 4. Absorber reactivity worth.

		Absorber reactivity worth (β_{eff})			
Absorber type	Height of absorber column (mm.)	BFS-67-1	BFS-67-3B	BFS-67-3	BFS-67-2
B_4C enr.	381.3	-2.16±0.12	-2.13±0.13	-2.10±0.12	-2.06±0.12
B_4C enr.	191.6	-1.26±0.06	-1.20±0.06	-1.20±0.06	-1.15±0.06
B_4C nat.	380.8	-0.879±0.039	-0.825±0.036	-0.809±0.035	-0.787±0.035
B_4C nat.	190.5	-0.493±0.021	-0.455±0.020	-0.455±0.020	-0.433±0.017

The associated uncertainties of the delayed neutron parameters provide a very important contribution to the estimated errors of the experimental results. The absorber worth ratios are determined with an improved accuracy (1.5 - 2%) when compared to the individual absorber worth. A decrease of the absorber reactivity worth with the Neptunium content can be noted.

Central reactivity coefficients (CRC)

The central reactivity coefficients of a conventional set of material samples, with the addition of Np237 dioxide and Am241 dioxide, have been measured in the centre of the core using an oscillation method. The reactivity change in the core associated with the introduction of the samples is presented in Table 5.

TABLE 5 Central Reactivity Worth Ratios $R_i(l=0)/R_{235}(l=0)$ [10^3]
(l - mean chord of a sample)

Isotope (i)	BFS-67-1	BFS-67-3B	BFS-67-2
U238	-67±2	-54±2	-46±1
Li6	-406±4	-	-
B10	-996±15	-784±15	-716±10
C12	-4.4±0.2	-9.7±0.2	-12.5±0.3
H	45±2	-66.0±1.3	-
Pu239	1330±20	1370±20	1380±20
Np237	-250±10	-112±4	-52±10
Am241	-238±10	-80±20	-28±15
Na	-4.7±0.4	-9.1±0.3	-13.1±0.5

These results indicate the main variations from the nominal values expected from a change in the density of one element.

EXPERIMENTAL ANALYSIS

The transposition of experimental results to full power reactors requires refined calculation analysis, with particular attention being given to the modelling of the reactor and experimental devices.

The JEF2/ECCO/ERANOS calculational scheme includes the features to pursue such objectives:

- the recent JEF2 evaluated data file[4],
- the ECCO cell code[5] with a self shielding of cross sections and a slowing down treatment calculated in 1968 groups in a 3D representation of the fuel pellets in the assembly tubes ,
- the S4 P1 transport theory code BISTRO for a flux calculation in a 2D RZ geometrical representation of the cores.

A precise analysis of the experiments has been performed using these specific features of the JEF2/ECCO/ERANOS[6] code and data system.

The BFS67-1 core has with a central enriched plutonium zone surrounded by an uranium driver zone. The calculated β_{eff} for this core using Tuttle's data is 481 pcm. Three successive cores are obtained by introducing Np237 in a central region (BFS67-2, BFS67-3 and BFS67-3B). The analysis with the JEF2/ECCO/ERANOS scheme of the critical masses of the four cores give the following results:

TABLE 6 Measured and Calculated Keff and discrepancies on the critical masses

Assembly	BFS67-1	BFS67-2	BFS67-3	BFS67-3B
Experiment	1.00038	1.00034	1.00038	1.00048
Calculation	0.99720	0.99647	0.99735	0.99736
Discrepancy (E-C)/C (pcm)	320	387	303	312

Calculations are in excellent agreement with experiments showing a nearly constant and small discrepancy of around 300 pcm for all of the configurations studied here. It can be noted that a significant heterogeneity effect has been observed (around 600 pcm) which is taken into account by a fully 3 dimensional heterogeneous cell description.

Central neutron cross-sections average ratios

As the spectral indice measurements of the small size chambers and the foils in the cells are performed between tubes and because of the very large heterogeneity effects, the calculations are performed using a 3D cell description. Calculated values are taken in the outer regions of the plates. The results of the analysis are presented in the following table with revised uncertainties to take into account systematic errors:

TABLE 7 C/E on Spectral Indices with JEF2/ECCO/ERANOS data and code system

Assemblies	BFS-67-1	BFS-67-3B	BFS-67-2
σ_fU238/σ_fU235	0.984±0.025	1.035±0.025	1.027±0.025
σ_fPu239/σ_fU235	0.986±0.015	0.995±0.015	1.003±0.015
σ_fNp237/σ_fPu239	0.936±0.030	0.980±0.030	0.979±0.030
σ_fPu238/σ_fPu239	1.032±0.035	1.042±0.035	1.046±0.035
σ_fPu240/σ_fPu239	1.032±0.030	1.061±0.030	1.067±0.030
σ_fPu241/σ_fPu239	1.022±0.015	1.017±0.015	1.008±0.015
σ_fPu242/σ_fPu239	1.011±0.020	1.046±0.020	1.053±0.020
σ_fAm241/σ_fPu239	0.926±0.030	0.962±0.030	0.964±0.030
σ_fAm243/σ_fPu239	0.955±0.040	1.026±0.040	1.007±0.040
σ_fCm244/σ_fPu239	1.127±0.030	1.115±0.030	1.110±0.030
σ_cU238/σ_fU235	1.074±0.025	1.035±0.025	1.004±0.025
σ_cAu197/σ_fU235	0.968±0.045	-	0.890±0.045
σ_cNp237/σ_cU238	0.960±0.050	0.913±0.035	0.960±0.050
σ_cNp237/σ_fU235	1.017±0.035	-	0.980±0.035

Results of the analysis on the indices show a reasonable agreement with systematic discrepancies appearing in all cores. Cm244 is giving a large discrepancy, the origin of which is not known, measurements for this reaction being difficult. The introduction of Np237 in cores does not enlarge the discrepancies on any of the reactions measured.

Sodium void reactivity measurements have been performed on various heights of the central part of the core. Calculated results are obtained by perturbation theory for BFS67-1 and BFS67-2 sodium void reactivities with a 3D cell description of the voided cell. Values are given in pcm:

TABLE 8 Sodium Void Reactivity Worth

	BFS67-1		BFS67-2	
voided parts	Experiment (in pcm)	C/E	Experiment (in pcm)	C/E
3+4+5+6; 31 fuel rods	29.8±1.4	0.89	90.4±3.9	1.00

The introduction of Np237 in the core fuel increases significantly the Na void reactivity worth, an effect which is correctly reproduced by the calculation. However, the calculated result overestimates the experimental one, a trend which is fortunately acceptable for safety considerations.

Central reactivity worth have also been measured in the central part of the reactor by oscillating samples of different sizes. Extrapolation to Zero size sample reactivity worth has led to values to which calculated values are compared. The trend due to the Neptunium introduction have been satisfactorily reproduced by the calculation but more refined analysis need to be defined to reproduce the absolute values.

CONCLUSIONS

As part of the France/Russia fast reactor collaboration, a series of experimental configurations have been established at the BFS facility with the aim of validating the concept of using fast reactors to burn Neptunium. A variety of Neptunium fuel contents (6.5%, 13.1%) have been studied to see the influence of the introduction of Neptunium on the various parameters.

The BFS67 experimental programme has also provided a valuable opportunity for the validation of the JEF2/ECCO/ERANOS calculational scheme enabling the transposition to full power reactor. It has been shown that the introduction of Neptunium introduces no significant discrepancies when comparing the experimental and calculated results, as long as a 3 dimensional cell description is used to reproduce the precise experimental conditions.

It has been shown that the introduction of Neptunium leads to a significant increase in the sodium void reactivity worth, as well as a decrease in the control rod worth of up to 15%. Both of these trends have been satisfactorily reproduced by the JEF2/ECCO/ERANOS scheme, and central reactivity worth calculations have shown a good agreement with the measured absolute reactivity variations. The dependence of the measured parameters on the Neptunium content has been shown to be close to linear.

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