Full MOX recycling in ALWR : Lessons Drawn through the MISTRAL Program.

S. CATHALAU[1] – P. FOUGERAS[1] – P. BLAISE[1] – N. THIOLLAY[1] – A. SANTAMARINA[1] – O. LITAIZE[1] – C. CHABERT[1] – T. YAMAMOTO[2]– Y. IWATA[2]* – T.UMANO[2] — R.KANDA[2]
P. GIRIEUD[3] – D. BIRON[4] – M. TATSUMI[5] – T. KAN[6] – Y. ANDO[7] – K. ISHII[8]

[1] CEA/DEN/CAD/DER – C.E. Cadarache – Bldg. 238 –
13108 St.-Paul-lez-Durance – FRANCE.
Tel : ++33 4 42 25 45 94
Fax : ++33 4 42 25 70 25
e-mail : scathalau@cea.fr

[2]NUPEC – Systems Safety Dept – Fujita Kanko Bldg. 5F – Toranomon 17-3, 1-Chome – Minato-ku – Tokyo 105-001 – JAPAN.

[3] FRAMATOME/EPD – Tour FRAMATOME ; Paris – La Défense – FRANCE
[4] EDF/SEPTEN – Villeurbanne – FRANCE
[5] NFI - PWR Fuel Division - Osaka, JAPAN
[6] MHI - Nuclear Energy Systems Engineering Center - Yokohama - JAPAN
[7] TOSHIBA - Power & Industrial Systems Research & Development Center - Kawasaki - JAPAN
[8] HITACHI - Power & Industrial Systems R&D Laboratory - Hitachi - JAPAN
*Present Address: HITACHI - Nuclear Systems Division - Hitachi - JAPAN

ABSTRACT

From 1995 to 2000, CEA and NUPEC undertook an extensive experimental program, called MISTRAL, devoted to the study of advanced LWR loaded with 100% of MOX fuel. Four core configurations were implemented in the EOLE facility at Cadarache aiming at the measurement of the main neutronic physical phenomena arising in such lattices. This paper presents the main results obtained during these experiments in term of experimental techniques and calculation/experiment comparisons

INTRODUCTION.

In 1995, the French CEA and the Japanese Corporation NUPEC, in collaboration with their industrial partners, decided to launch an extensive experimental program, called MISTRAL [1,2,3,4,5], aimed at measuring the main neutronic parameters of full MOX ALWR. This program was planned for covering a representative range of higher moderation. Three regular lattices (one UOX and two MOX) having a moderation ratio (H/HM[1]) varying from 5 to 6, and a mock-up lattice (H/HM~6) simulating advanced 17×17-PWR assemblies (full MOX) were investigated :

- *MISTRAL-1* (fig. 1) : Regular enriched UO_2 (3.7 % in ^{235}U) core involving about 750 fuel pins in a square pitch of 1.32 cm (H/HM~5), used as a reference for the following MOX cores. The criticality was obtained by adjusting the soluble boron concentration. The experimental study was completed in November 1996.
- *MISTRAL-2* (fig. 2) : Regular 100% MOX (7 % of Pu) core involving about 1600 fuel pins in the same lattice as above (H/HM~5), and no boron in the moderator. The criticality was obtained by modifying the number of peripheral fuel pins. The experimental study was completed in April 1998.

[1] H/HM = [number of Hydrogen atoms in the moderator]/[number of Heavy Metal atom], H/HM is the “Atomic moderation Ratio”

- *MISTRAL-3* (fig. 3) : Regular 100% MOX (7 % of Pu) core involving about 1400 fuel pins in a square pitch of 1.39 cm (H/HM~6). The criticality is obtained by adjusting the soluble boron concentration. The measurements began August 1998 and were completed in April 1999.
- *MISTRAL-4* : Mock-up configurations simulating a 100 % MOX-fueled PWR involving 17x17 sub-assemblies in a square pitch lattice of 1.32 cm (H/HM~6.0) with and without absorber clusters. The study of these configurations was completed in July 2000. Figure 4 shows the reference mock-up MOX core.

The main neutronic parameters of such advanced lattices were measured with a high accuracy allowing the validation of calculation tools used for designing full MOX cores. Figure 5 gives an overview of the performed measurements and of the main scope of the program. In parallel, CEA and NUPEC performed the analyses using their calculation tools based on the deterministic and Monte-Carlo codes. This paper presents the main lessons drawn through these analyses. The first part describes the measurement techniques and the main information obtained during the MISTRAL program. The second section is devoted to the calculation/experiment comparisons.

MEASUREMENT TECHNIQUES AND MAIN EXPERIMENTAL RESULTS.

The MISTRAL program allows to complete the experimental data base devoted to Pu recycling in ALWR 100% MOX-fuelled cores. Several experimental techniques were used for measuring the neutronic parameters with an high accuracy.

In the regular lattices, the material buckling was determined using six integral γ-spectrometry measurements directly on the fuel pins and miniature fission chambers for both radial and axial fission rate distributions. The fission chambers were placed within a watertight tube in several positions in the cores. Four types of response functions were used in order to draw information about neutron fast spectrum (^{237}Np and ^{238}U deposits) and thermal spectrum (^{239}Pu and ^{235}U deposits). The axial fission rate distribution was obtained from the same fission chambers and completed by using a γ-spectrometry measurement. The axial measurements were performed for three radial positions and for the whole fissile height (80 cm) by 2 cm steps. These techniques enable us to determine the material buckling of the core at less than ± 0.9 % for MISTRAL-1, ± 0.6 % for MISTRAL-2 and MISTRAL-3.

Spectral indices were measured using miniature fission chambers placed in a watertight tube located in the central position of the core and at the mid fissile plane. ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{237}Np fission rates were investigated with such miniature devices. The Modified Conversion Ratio (MCR) defined as the ratio of the capture rate of ^{238}U to the total fission rate, was determined directly on the central fuel pin using the experimental technique proposed by NAKAJIMA [12] and adapted to our facility allowing an accuracy better than ±3%. It involves the specific γ-peak checks (the self γ-absorption effects in the pin were estimated by Monte-Carlo calculations and APOLLO-2 calculations of the reaction rates within the pellet) of representative γ-rays ::

- γ-ray at 277.6 keV associated to the β^- decay of ^{239}Np (~2.4 days of period) for the determination of the ^{238}U capture rate arising in the fuel.
- γ-ray at 293.27 and 537.31 keV associated to the β^- decay of ^{143}Ce and ^{140}Ba respectively for the determination of the total fission rate of the fuel.

A new thermo-regulation system was implemented in the EOLE facility in 1995 in order to measure very accurately the isothermal temperature coefficient (ITC) between 10°C and 80°C for the regular UO_2 and MOX lattices. The delayed neutron fraction was measured both UO_2 and MOX lattices using a neutron fluctuation technique. The experimental uncertainty is better ± 2%

The reactivity of the core was determined by the divergence technique (measurement of doubling time linked to the residual reactivity of the core through the Nordheim equations) and by counting in subcritical configurations (equation relying the counting obtained by a detector to the negative reactivity of the core). Considering the modifications between cores (insertion or withdrawing of absorbers, modification of the soluble boron concentration or of the critical size, temperature of the moderator, etc.), reactivity effects of absorbers, clusters, peripheral fuel pins, temperature variation, were determined with a high accuracy. For example, the integral boron efficiency ($\Delta\rho \pm 1\sigma$ in \$) measured in the regular lattices seem to follow a quite linear behaviour :

ΔCB (ppm)	MISTRAL-2	MISTRAL-3	MISTRAL-4
250	-5.6 ± 0.2	-2.5 ± 0.2	/
400	-9.0 ± 0.3	-4.7 ± 0.3	-5.0 ± 0.3
500	-11.1 ± 0.4	-9.4 ± 0.5	-9.7 ± 0.5
600	-13.3 ± 0.5	-13.9 ± 0.7	-13.8 ± 0.7

The reactivity of single absorbers placed in the centre of the regular lattices were checked in regular lattices :

Absorber	Relative effect		
	MISTRAL-1	MISTRAL-2	MISTRAL-3
UO_2 - Gd_2O_3	0.46 ± 0.03	0.11 ± 0.02	0.15 ± 0.03
Ag-In-Cd	0.70 ± 0.06	0.22 ± 0.05	/
Natural B_4C	0.75 ± 0.06	0.26 ± 0.06	/
Enriched B_4C	1.00 ± 0.05	0.37 ± 0.05	0.45 ± 0.05

*Normalization to enriched B_4C efficiency in MISTRAL-1 core

The MISTRAL-1 results give information on the thermal absorption of the absorbers, while the MISTRAL-2 results concern more the epithermal-energy range. The spectrum shift between MISTRAL-1 and MISTRAL-2 is shown in the UO_2-Gd_2O_3 effect. The UO_2-Gd_2O_3 is more efficient in the thermal spectrum of MISTRAl-1, this is due to the very large thermal capture cross sections of the ^{155}Gd and ^{157}Gd. The AIC efficiency covers the whole spectrum, but the Cd component has a large capture cross-section in the thermal range (~0.54 eV), this explains the efficiency decreasing between MISTRAL-1 and MISTRAL-2. Natural B_4C is more efficient in a UOX lattice due to a higher amount of thermal neutrons. The enriched B_4C efficiency decreases from 1.00 in MISTRAL-1 down to 0.37 in MISTRAL-2 and increases again up to 0.45 in MISTRAL-3 due to a harder spectrum in the MOX lattice and to a higher of pins loaded in core.

A extensive study was devoted to 2D void simulation if the MISTRAL regular lattice using aluminium clad and block. Thus, void fractions of 40%, 60% and 100% were checked for reactivity.

Void Fraction	ASM	Boron Eq.* (CEA)
40 %	0.50 ± 0.02	0.49 ± 0.03
60 %	0.76 ± 0.04	0.76 ± 0.05
100 %	1.00 ± 0.05	1.00 ± 0.06

*Normalization to 100% of void efficiency.

This table indicates that the void effects do not seem to be linear with the void fraction : a detailed analysis is currently performed specially for the effects between 40%→60% and 60%→100% of void, for which the neutron spectrum modifications are very strong.

Absorber clusters of 12 stainless steel rods and 24 Ag-In-Cd, metallic Hf and enriched B_4C rods were tested in MOX mock-up core enabling to obtain the following results in term of reactivity effects (synthesis of boron concentration modification and critical size modifications) :

Cluster	Δρ in \$ (CEA)
12 S.S.	-0.95 ± 0.04
24 Hf	-12.67 ± 0.63
24 AIC	-13.39 ± 0.67
24 enr. B_4C	-15.96 ± 0.80

Thus, the metallic Hf cluster is less efficient than the "classical" Ag-In-Cd alloy in advance 100% MOX lattices and the higher antireactive cluster is the 24 enriched B_4C.

Radial and axial fission rate were measured in reference regular cores and in configurations involving heterogeneities to check the perturbations. Figure 6 compares the radial fission rate in the MISTRAL-3 reference core and with absorbers placed in the center : the reference curves follows the theoretical behaviour and for absorbers, after 3 or 4 rings the measured fission rate is very near the fundamental mode. Figure 7 presents the same results for the 2D void studies.

In the MISTRAL-4 mock-up cores, a particular care was taken to measure the radial power distribution within assemblies, specially when the 24 absorber clusters were introduced in the central part. The criticality of the cores was obtained by adding UO_2 fuel pins at the periphery. Figure 8 gives an example of the radial fission rate profile measured in the MOX-AIC mock-up configuration. For the cores involving an UO_2 assembly in the center, a specific γ-ray test was implemented in order to allow the inter-calibration of power delivered by MOX and UO_2 pins. For such a measurement, ^{140}Ba-^{140}La and ^{92}Sr individual peaks at 1596 MeV and 1384 keV respectively were checked. In addition, fission rates obtained in fission chamber placed in the center of each core were determined in order to improve the normalization results. The peak check investigation leads to the conclusion that the fission yields of ^{92}Sr and ^{140}Ba are not consistent whatever the nuclear data library chosen for analysis. A new evaluation seems to be necessary.

C/E COMPARISONS.

The calculation routes used by CEA and NUPEC are both based on deterministic codes APOLLO-2 [6] and SRAC [7] respectively for assembly spectrum and 2D core calculations. In addition, reference calculations are performed by 3D continuous-energy Monte-Carlo codes TRIPOLI4 [8] in France and MVP [9] in Japan. The nuclear data files are JEF2.2 [10] in 172 groups and JENDL3.2 [11] in 107 groups. Both deterministic routes used for assembly spectrum calculations are based on collision probability methods coupled with specific self-shielding techniques and homogenisation/collapsing modules. APOLLO-2 applies some equivalence methods allowing the consistency between the 172 group spectrum calculation and the core models. Core calculations are performed by APOLLO-2 in the CEA route and by CITATION or TWOTRAN for NUPEC.

Using the lattice codes (APOLLO-2 [13] and SRAC) associated with their nuclear data library, the multiplication factors are well estimated for the regular lattices ; in the same way, core calculations show rather good agreement (C-E in pcm) :

Experiment	Cell calculations	Core calculations	
	CEA	CEA	NUPEC
MISTRAL-1	-256 ± 500	+420	-190
MISTRAL-2	+58 ± 350	+679	+410
MISTRAL-3	+297 ± 350	+750	+420

The analysis of MISTRAL-1 confirmed that the multiplication factor of CEA is overestimated linked to an underestimation of the ^{235}U capture cross-section in the resonance range for JEF2.2. In MOX lattices, reactivity seems to be correctly estimated. The criticality of the different cores show a rather good consistency with the experiment showing that reflector model is sufficiently accurate.

The delayed neutron fraction is very well reproduced by both analyses (C/E varying in a range 0.975 to 1.024 with a S.D. of ± 2% at 1σ). A more detailed analysis is presented during this conference [14].

	(C-E)/E ± σ (1σ) (%)	
β_{eff}	NUPEC	CEA
MISTRAL-1	-0.8 ± 1.5	+2.4 ± 1.5
MISTRAL-2	-2.5 ± 1.6	+0.1 ± 1.6

The following table shows the interpretation results related to spectral indices and modified conversion ratio.

Spectral Index	(C-E)/E ± σ (1σ) (%)	
MISTRAL-1	NUPEC	CEA
$^{239}Pu/^{235}U$	+ 1 ± 2.4	+ 0.2 ± 2.0
$^{241}Pu/^{239}Pu$	- 1 ± 2.7	+ 1.2 ± 2.0
MCR	+ 2 ± 3.0	+ 1.5 ± 3.0
MISTRAL-2	NUPEC	CEA
$^{238}U/^{235}U$	- 10 ± 6.7	- 2 ± 5
$^{239}Pu/^{235}U$	+ 4 ± 2.4	+ 2 ± 2
$^{238}Pu/^{239}Pu$	- 6 ± 13.7	/
$^{240}Pu/^{239}Pu$	- 17 ± 5.9	- 3 ± 5
$^{241}Pu/^{239}Pu$	- 2 ± 2.7	+ 0 ± 2
$^{242}Pu/^{239}Pu$	- 2 ± 7.6	+ 12 ± 7
$^{237}Np/^{239}Pu$	- 10 ± 3.2	- 5 ± 3
MCR	+ 1 ± 1.4	+ 2.1 ± 1.4
MISTRAL-3	NUPEC	CEA
MCR	+ 2 ± 1.4	+ 3.1 ± 1.4

The C/E for the modified conversion ratio (MCR) in the UOX lattice calculations is about 1.02 (±3%) for NUPEC and 1.015 for CEA. For MOX lattices, it increases with the moderation ratio from 1.01 (MISTRAL-2) up to 1.02 (MISTRAL-3) for NUPEC and from 1.02 (±1.4%) up to 1.03 (±1.4%) for CEA. The ^{238}U capture rate seems to be slightly overestimated in MOX lattices by the CEA route due to self-shielding of ^{238}U resonances and mutual self-shielding with other nuclides. The main fissile actinides (^{239}Pu and ^{241}Pu) spectral indices are well predicted by both calculation routes in UOX and MOX lattices. Concerning the threshold reactions (^{238}U, ^{240}Pu, ^{242}Pu and ^{237}Np) the results show that the fission rate is less satisfactory indicating that the flux near the threshold is not well reproduced.

The ITC is very well calculated by CEA : C-E = 0.0 ± 0.2 pcm/°C in UOX. In MOX cores, the ITC is under-estimated below 40°C and well reproduced between 40°C and 80°C.

The following table summarizes the results of absorber efficiency [(C-E)/E ± σ (1σ) in %] by CEA and NUPEC for the UOX and MOX lattices.

Lattice	MISTRAL-1		MISTRAL-2	
Absorber	CEA	NUPEC	CEA	NUPEC
UO_2-Gd_2O_3	+0.2 ± 3.6	+5 ± 3.6	-1.7 ± 1.3	+8 ± 6.6
Ag - In - Cd	-0.2 ± 2.3	+4 ± 2.5	-0.1 ± 1.2	+3 ± 7.6
B_4C nat	-1.5 ± 2.1	+4 ± 2.3	+0.5 ± 1.3	+0 ± 7.4
Enriched B_4C	-1.3 ± 1.4	+5 ± 1.6	+0.6 ± 1.3	-2 ± 7.1

This table indicates that the CEA route reproduces almost perfectly the absorber efficiency tested in MISTRAL. For NUPEC, some discrepancies are observed, mainly due to the lack of equivalence procedures in its code system.

The differential boron efficiency [$\Delta\rho/\Delta CB$ and 1 S.D.] comparison is given below :

(C-E)/E in %	NUPEC	CEA
MISTRAL-1	9 ± 11	3.4 ± 7.0
MISTRAL-2	8 ± 4.3	7 ± 5.0

Both NUPEC and CEA calculations are in rather good agreement with the experimental values.

For the 2D void studies in the MISTRAL-3 configuration, the following table summarizes the calculation/experiment comparison results (C/E ± 1σ).

Configuration	NUPEC (XY calc.)	CEA (XY calc.)	CEA (RZ calc.)
40% void	1.08 ± 0.06	1.08 ± 0.02	/
60% void	1.03 ± 0.06	1.03 ± 0.01	/
100% void	1.02 ± 0.06	1.02 ± 0.01	1.01 ± 0.01

The CEA and NUPEC results are very consistent. The values of C/E of NUPEC show good consistency between the analysis and the measured values taking into account the experimental errors that include the error of boron worth estimation necessary to obtain the experimental data. On the other hand, the interpretation of the 40% void experiment by CEA shows that the calculation over-estimates the partial void reactivity effect from about 8% in the calculations. In the configuration void 60%, the over-estimation of the void coefficient is about 3%. The variation calculation-experiment seems to decrease with the increase of the void rate. For the case void 100%, the calculation over-estimates very slightly the void effect reactivity (~1%). The RZ model of CEA allows to account for the axial leakage effects, and to consequently improve the result.

CONCLUSIONS

The MISTRAL experimental program was undertaken within a framework of a cooperation between the French CEA and the Japanese organization NUPEC and aimed at providing the reactor physicists with valuable and precise experimental results obtained in representative lattices of ALWRs loaded with 100% of MOX fuel. The program, shared in four phases, allowed to complement the experimental data base devoted to LWRs. New experimental techniques (modified conversion ratio) and new devices (thermo-regulation system) were developed and implemented for this program allowing to obtain very precise experimental results reaching thus the main target defined at the beginning of the CEA/NUPEC collaboration. Most of experimental results have been checked by the reference and industrial calculation routes leading to obtain valuable information about the validity of the codes for treating the physical phenomena occurring in full-MOX ALWRs. Further calculations are currently in progress in CEA to evaluate the ability of the codes to reproduce the experimental results obtained in the mock-up cores with and without absorber clusters.

ACKNOWLEDGEMENT

NUPEC has been supported by the Japanese Ministry of Economy, Trade and Industry (METI). Authors thank Dr. K. Kanda, Director of Japan Energy Policy Institute, and the other members of the implementation committee of "Study of Advanced LWR Cores for Effective Use of Plutonium" of NUPEC for their technical advice and suggestions.

REFERENCES.

[**1**] . S. Cathalau – J.C. Cabrillat – J.P. Chauvin – P.J. Finck – P. Fougeras – G. Flamenbaum – H. Matsu-ura – M. Ueji and T. Yamamoto, ***« MISTRAL : an Experimental Programme in the EOLE facility devoted to MOX core Physics »*** Int. Conf. On the Physics of Reactors - Physor'96, MITO - Japan - 1996

[**2**] T. Yamamoto – H Matsu-ura – M. Ueji – S. Cathalau – J.C. Cabrillat – J.P. Chauvin – P.J. Finck - P. Fougeras and G. Flamenbaum, ***« Core Physics Experiment of 100% MOX core : MISTRAL »*** Int. Conf. on Future Nuclear Systems - « GLOBAL'97 » p 395 ; AESJ - ANS ; Yokohama (Japan) 1997

[**3**] S. Cathalau – P. Fougeras and A. Santamarina ***« First Validation of Neutronic Lattice parameters of overmoderated 100% MOX fueled PWR cores on the basis of the MISTRAL Experiment. »*** Int. Conf. On the Physics of Nuclear Science and Engineering - Long Island (USA) 1998

[**4**] P. Fougeras – P. Blaise – J.P. Chauvin – R. Girieud ***« MISTRAL-4 : AN EXPERIMENTAL MOCKUP IN THE EOLE FACILITY DEVOTED TO HIGH MODERATION 100% MOX CORE PHYSICS ».*** Int. Conf. GLOBAL'99 - Jackson-Hole (Wyoming -USA) – 1999

[**5**] K. Hibi and Al. ***«Analysis of MISTRAL and EPICURE Experiments with SRAC and MVP Code Systems»*** Int. Conf. On the Physics of Reactors - Physor2000, Pittsburgh - USA – 2000

[**6**] R. Sanchez and J. Mondot ***"APOLLO-2 : A user-friendly Code for Multigroup Transport Calculations,"*** Topl. Mtg. On Advances in Nuclear Engineering Computation and Radiation Shielding, Santa-Fe, New-Mexico, April 9-13, 1989

[**7**] K. TSUCHIHASHI, et al. ***"Revised SRAC Code System"*** JAERI-1302 Report (1986)

[**8**] J.P. Both - M. Derriennic - B. Morillon and J.C. Nimal ***"A survey of TRIPOLI-4,"*** Proc. of 8th Conference on Radiation Shielding - April 24-28, 1994 Arlington, Texas (USA)

[**9**] T. MORI, et al. ***"Vectorization of Continuous Energy Monte Carlo Method for Neutron Transport Calculation"*** J. Nucl. Sci. Technol., 29, pp325 (1992)

[**10**] C. Nordborg and M. Salvatores, ***"Status of the JEF Evaluated Data Library,"*** *Proc. Int. Conf. Nuclear Data for Science and Technology*, Gatlinburg, Tennesse, May 9-13, 1994, p. 680, American Nuclear Society (1994).

[**11**] T. NAKAGAWA, et al. "***JENDL-3.2***" J. Nucl. Sci. Tech., 32(12), pp1259 (1995)

[**12**] K. Nakajima - M. Akai and T. Suzaki, ***"Determination of Modified Conversion Ratio of Light-Water-Moderated Uranium-Plutonium Mixed-Oxide-Fuel Lattice,"*** Nuc. Sci. Eng., **119**, 175-181 (1995).

[**13**] O. Litaize – A. Santamarina and C. Chabert. "***Analysis of the MISTRAL experiments with APOLLO-2. Qualification on neutronic parameters of UOX and MOX cores."*** To be presented at this conference.

[**14**] O. Litaize and A. Santamarina . "***Experimental Validation of Effective delayed Neutron fraction in LWR-MOX cores."*** To be presented at this conference.

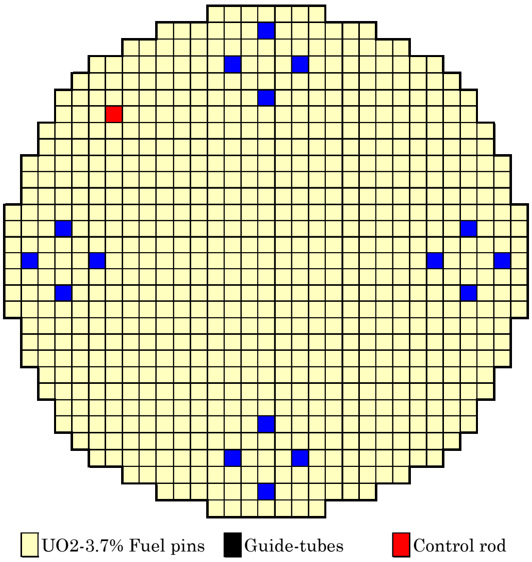

Figure 1 : Radial cross-section of the MISTRAL-1 core

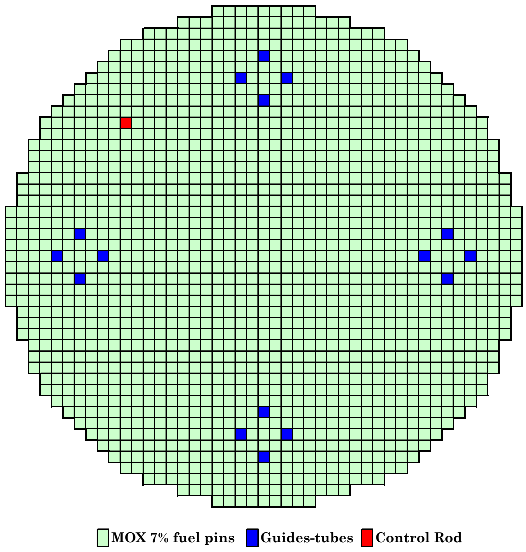

Figure 2 : Radial cross-section of the MISTRAL-2 core

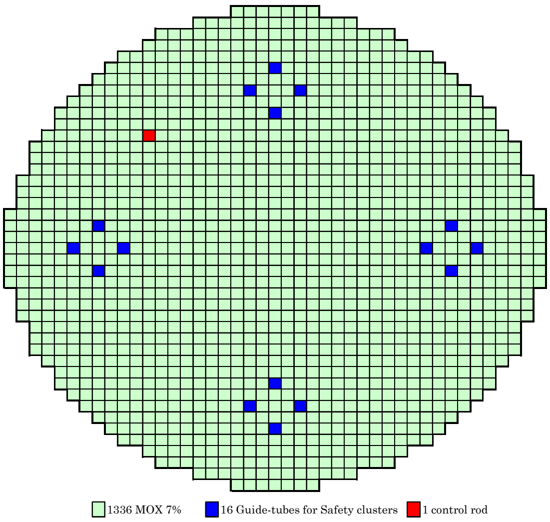

Figure 3 : Radial cross-section of the MISTRAL-3 core

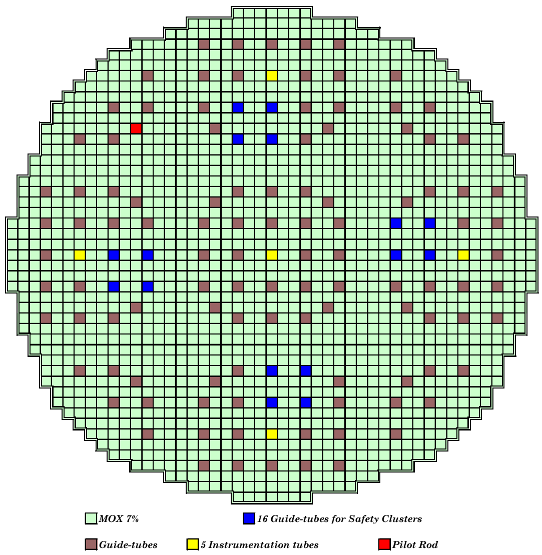

Figure 4 : Radial cross-section of the MISTRAL-4 mock-up reference core

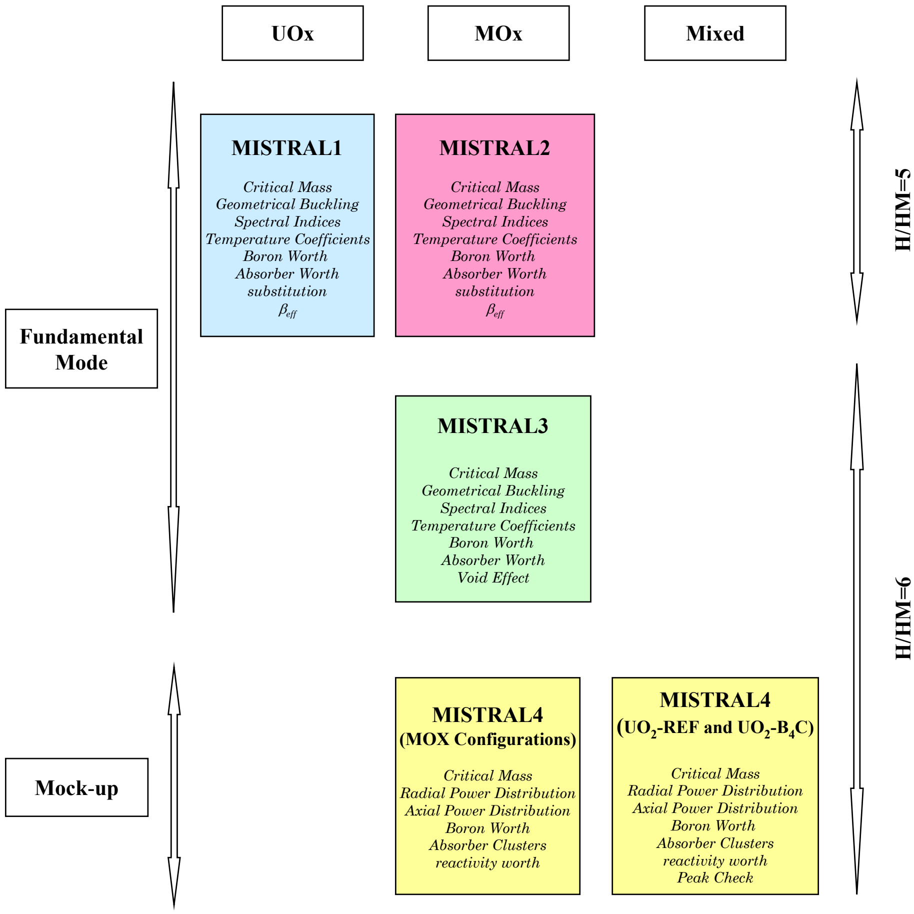

Figure 5 : MISTRAL PROGRAM : configurations and performed measurements

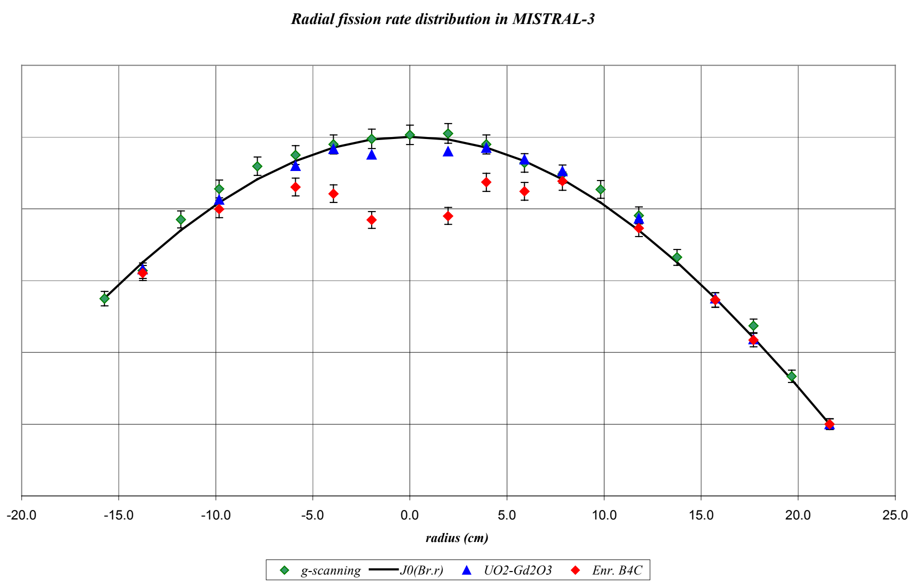

Figure 6 : Fission rate measurements around absorbers - Example

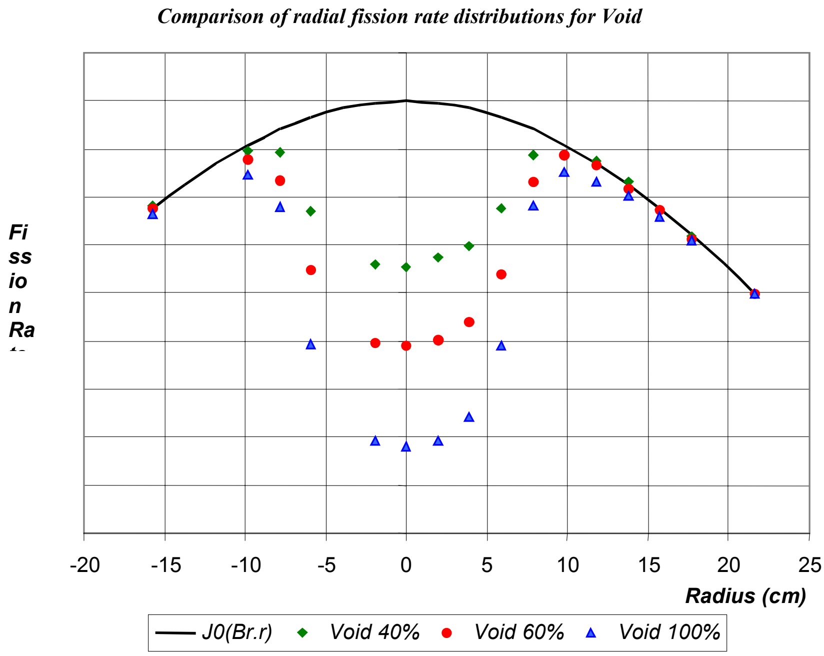

Figure 7 : Fission rate measurements in void studies

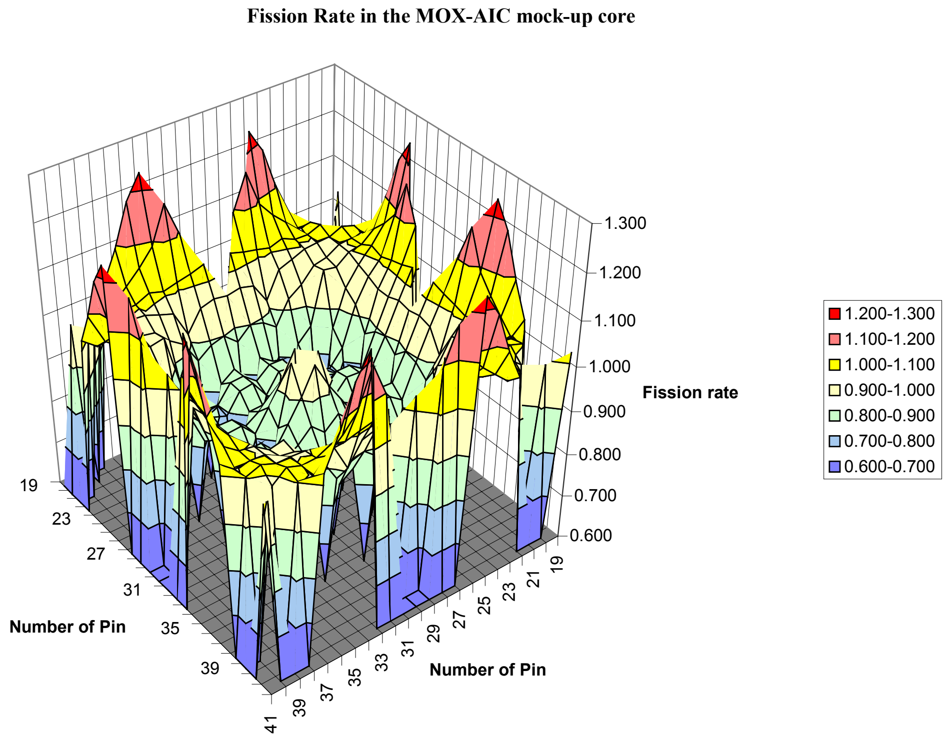

Figure 8 : Radial Fission rate profile in the MOX-AIC mock-up core