Experimental Validation of Plutonium Ageing by Monte Carlo Correlated sampling

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Abstract

Integral measurements of Plutonium Ageing were performed in two homogeneous MOX cores (MISTRAL2 and MISTRAL3) of the French MISTRAL Programme between 1996 and year 2000. The analysis of the MISTRAL2 experiment with JEF-2.2 nuclear data library highlightened an underestimation of ^{241}Am capture cross section. The next experiment (MISTRAL3) did not conclude in the same way. This paper present a new analysis performed with the recent JEFF-3.1 library and a Monte Carlo perturbation method (correlated sampling) available in the French TRIPOLI4 code.

KEYWORDS: Plutonium Ageing, Correlated Sampling, Monte Carlo

1. Introduction

The aim of this paper is the calculation of the reactivity effect of Plutonium Ageing with the TRIPOLI4 Monte Carlo code [1] and the European Nuclear Data Libraries JEF-2.2 and JEFF-3.1. Integral measurements of the Plutonium Ageing were performed during the MISTRAL experiment carried out in the EOLE zero-power reactor at Cadarache Center (France). The MISTRAL experimental programme [2], [3] was launched by the french Atomic Energy Commission (CEA) within the framework of a collaboration with its industrial partners and the japanese Nuclear Power Energy Corporation (NUPEC). It consisted in four configurations. The experiment which started in 1996 and ended mid-July 2000, was devoted to Advanced Light Water Reactor studies with high moderation lattices and loaded with 100% MOX fuel.

2. Experimental set-up

Two experimental programmes are studied in this paper: MISTRAL2 and MISTRAL3 which are 7% MOX homogeneous cores with 4×4 guide-tubes devoted to the safety cluster (enriched B_4C rods) and one pilot rod. Figure 1 shows the radial cross sections of these two cores obtained with the TRIPOLI4 code.
The first regular and homogeneous MOX core (MISTRAL2) consists in about 1600 UO_2PuO_2 fuel pins (7 % $w/o\ Pu$) with a lattice pitch of $1.32\ cm$, leading to a moderation ratio of about $V_{mod}/V_{fuel} = 1.7$ and $H/HM = 5.1$. Criticality is achieved by adjusting the number of peripheral fuel pins. Two core configurations were performed, seperated by 357 days, and

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76 fuel pins were added to achieve the criticality. This is a geometric perturbation so a Monte Carlo perturbation is not allowed in this (MISTRAL2) case.
MISTRAL3 is an homogeneous MOX core (7 % $w/o\ Pu$) involving about 1600 fuel pins and a square pitch of about $1.39\ cm$. The criticality of this core is achieved by modifying the boron concentration in the moderator (the number of pins is unchanged between each configurations). This is an atomic composition perturbation (only the boron concentration in the moderator and the Plutonium vector are modified) and the Monte Carlo Perturbation is allowed in this case concerning the six differents core configurations of MISTRAL3.
Plutonium Ageing is experimentally determined by measuring the residual reactivity of each core configuration (divergence method).

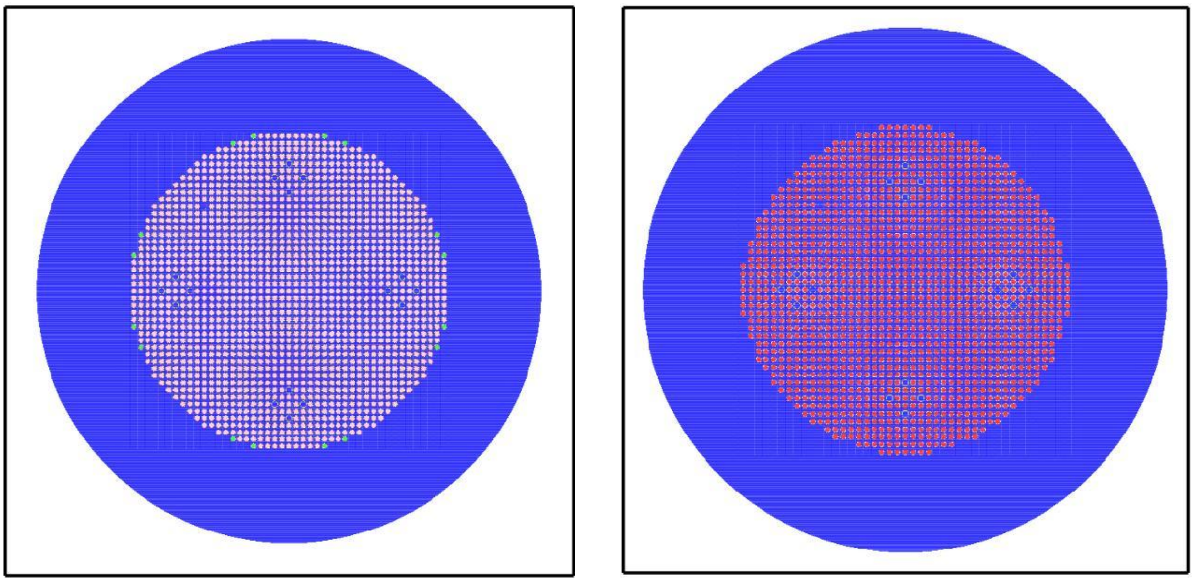

Figure 1: MISTRAL2 and MISTRAL3 radial cross sections

3. Analysis

Due to the β decay of ^{241}Pu and consequently the build-up of ^{241}Am poisoning nuclide, the reactivity of a MOX core decreases during time. In order to compensate this effect and to achieve the criticality, the number of peripheral fuel pins is increased in MISTRAL2 and the boron concentration is modified in MISTRAL3. The analysis of these experiments were performed in a near past with the French deterministic code APOLLO2 and a $S_N - XY/20$-group calculation scheme (the experimental axial buckling was applied in order to take into account the axial leakage). The results of the Monte Carlo perturbations and the JEF-2.2 library are very closed to those results.

3.1 MISTRAL2: direct calculations

As the reactivity worth of the peripheral fuel pins $\Delta\rho^{exp}_{pins}$ is acurately calculated in MISTRAL2, the reactivity effect of Plutonium Ageing $\Delta\rho^{exp}_{Pu\ ageing}$ can be estimated from the total change in residual reactivity $\Delta\rho^{exp}_{res}$ between two states.

$$\Delta\rho^{exp}_{res} = \Delta\rho^{exp}_{pins} + \Delta\rho^{exp}_{Pu\ ageing} \tag{1}$$

In this experiment, two configurations separated by 357 days were analysed. The critical size was adjusted by adding 76 peripheral fuel pins. The difference between these two experimental residual reactivities is about $(28 \pm 3)\ pcm$. The fuel pin worth was experimentally determined for 8, 24 and 48 additional pins. The mean ractivity worth is about 0.0204 \$, that is to say $7.5\ pcm/pin$ (the effective delayed neutron fraction is about $370\ pcm$). The reactivity worth of the 76 additional pins is: $\Delta\rho^{exp}_{76pins} = 570\ pcm$ and the calculated value is exactly the same. The experimental Plutonium ageing is directly obtained through equation (1):

$$\rho^{exp}_{Pu\ ageing} = -1.52\ pcm/d$$

The TRIPOLI-4 calculation of the reactivity difference between the two core configurations is about $(+97 \pm 23)\ pcm$ with JEF-2.2 and $(+84 \pm 11)\ pcm$ with JEFF-3.1. The TRIPOLI-4 estimation of the Plutonium ageing goes from $-1.32\ pcm/d$ with JEF-2.2 to $-1.36\ pcm/d$ with JEFF-3.1 and consequently the difference between calculation and experiment is a little bit better:

$$C/E - 1 = -13\% \pm 4\% \quad (\text{JEF-2.2})$$

$$C/E - 1 = -10\% \pm 2\% \quad (\text{JEFF-3.1})$$

Due to the statistical uncertainty ($\pm 4\%$ in one standard deviation for JEF-2.2 calculation), we can not conclude if this better agreement between calculation and experiment is mainly due to the cross section improvement in JEFF-3.1.

3.2 MISTRAL3: correlated sampling based calculations

Criticality of each core in the MISTRAL3 experiment is achieved by adjusting the boron concentration. Six configurations with different boron concentrations were measured. The boron reactivity worth measured by the doubling time technique is equal to: $\Delta\rho^{exp}_{boron} = -8.4 \pm 0.4\ pcm/ppm$. In this way, a Monte Carlo perturbation based on correlated sampling can be performed to estimate the reactivity change between two states at two different dates.

With the well known correlated sampling techniques [4], the difference between the pertubed and the unperturbed collision densities is simply equal to the unperturbed game multiplied by a weight factor.

Table 1 points out the residual reactivity and table 2 points out reactivity effect due to Pu Ageing in MISTRAL3 experiment. Previous results obtained with APOLLO2 code are recalled here in the first table for comparison.

Table 1: Residual Reactivity in MISTRAL3

time interval (days)	ρ^{exp}_{res} (pcm)	$\rho^{ap2/J2}_{res}$ (pcm)	$\rho^{T4/J2}_{res}$ (pcm)	$\rho^{T4/J3}_{res}$ (pcm)
49	14.5 ± 8.6*	5.9	6.5 ± 0.8	4.5 ± 0.8
160	19.5 ± 8.8	15	15.9 ± 2.1	12.5 ± 2.1
179	10.9 ± 8.7	0.9	0.0 ± 2.0	-2.0 ± 2.0
195	22.8 ± 8.0	22.9	23.4 ± 2.1	19.9 ± 2.2
228	-3.6 ± 7.9	-13.3	-13.1 ± 2.1	-18.5 ± 2.1

* Experimental uncertainties from residual reactivity and boron concentration measurements.

Table 2: Plutonium Ageing in MISTRAL3

time interval (days)	$\Delta\rho^{exp}_{Pu\ ageing}$ (pcm/d)	$\Delta\rho^{T4J2}_{Pu\ ageing}$ (pcm/d)	$\Delta\rho^{T4J3}_{Pu\ ageing}$ (pcm/d)
160	-1.07	-1.09	-1.11
179	-1.05	-1.11	-1.12
195	-1.09	-1.09	-1.11
228	-1.06	-1.10	-1.12

The difference between calculation and experiment for MISTRAL3 are:

$$C/E - 1 = +2\% \pm 3\% \quad (\text{JEF-2.2})$$

$$C/E - 1 = +4\% \pm 3\% \quad (\text{JEFF-3.1})$$

4. Conclusion

The analysis of the various 100% MOX critical cores in EOLE reactor has demonstrated that the Pu ageing worth is reasonably calculated using the new JEFF-3.1 library. The poisoning reactivity worth is predicted within $\pm 5\%$ (1σ) accuracy. JEFF-3.1 and JEF-2.2 Plutonium Ageing are similar even though the worth component are strongly different : in JEFF-3.1, the ^{241}Am component is increased by $+10\%$ due to an improved resonant capture evaluation [5] and the ^{241}Pu is less reactive due to an increase of the Γ_γ value, particularly in the $0.3eV$ resonance [6]. Therefore, a new integarl experiment is needed to validate more accurately the Plutonium Ageing worth calculation.

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