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► **To cite this version:**

A. Santamarina, C. Riffard. The MOX Powder Calculation Challenge OECD Benchmark Proposal and CRISTAL-RIB Results. ICNC 2015 - International Conference on Nuclear Criticality, Sep 2015, Charlotte, United States. cea-02489557

HAL Id: cea-02489557

<https://cea.hal.science/cea-02489557>

Submitted on 30 Mar 2020

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MOX POWDER CALCULATION IMPROVEMENT: OECD BENCHMARK PROPOSAL AND CRISTAL-RIB RESULTS

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ABSTRACT

In the framework of the WPNCs Expert Group on Uncertainty Analysis for Criticality Safety Assessment, we propose a blind numerical benchmark. Since the WPNCs Expert Group on Experimental Needs had highlighted the poor database of representative experiments of low-moderated MOX powders, this blind benchmark is focused on MOX wet powders. Three PuO₂ contents are proposed: 100%, 30% and 12.5%. Two Pu isotopic vectors are considered: a conservative one (0%, 71%, 17%, 11%, 1% for ²³⁸Pu up to ²⁴²Pu) and a realistic one obtained from reprocessed LWR 30 GWd/T fuels. For these 6 cases the powder moisture rate is 3%. The MOX fissile medium is a sphere surrounded by 20 cm water reflector. The required results are the calculated K_{eff} and the associated prior uncertainty. Using benchmark experiments, contributors should provide the projected-K_{eff} and the corresponding posterior uncertainty. CRISTAL-V2 results based on JEFF3.1.1 nuclear data are presented. As an example, in the 30% PuO₂ challenging case, the calculation uncertainty is decreased from 1160 pcm (1σ) down to 450 pcm using EOLE and MASURCA critical Pu experiments. This uncertainty depends slightly on 'conservative' or 'realistic' Pu because the representativity factor does not vary significantly with the Pu vector.

KEYWORDS

UACSA, Safety-Criticality, MOX powders, blind benchmark

1. INTRODUCTION

The Pu recycle from reprocessing plants, such as La Hague in France, through MOX assemblies introduced in commercial Light Water Reactors (LWR) and future GEN-4 reactors, has emphasized the need for experimental benchmarks dealing with MOX fuel [1]. Experiments are required for criticality-safety evaluation of fuel cycle facilities, particularly MOX assembly factories. The assessment of the available benchmarks, performed by the OECD Working Party on Nuclear Criticality Safety (WPNCs) Expert Group on Experimental Needs, had highlighted the poor database of representative experiments of low-moderated MOX powders [2] [3]. Therefore, some new targeted experiments were proposed in French [4] and Russian mock-up reactors, and were evaluated [5]. In 2005-2006, a series of experiments referred to as BFS/MOX was conducted at the BFS-1 experimental facility in IPPE, Russia. Nine critical configurations with heterogeneous compositions of plutonium metal, depleted uranium dioxide and polyethylene were assembled [6]. However, the strong heterogeneity of these experiments raised a representativity problem compared to homogeneous MOX powders, as well as some calculation difficulties in the analysis.

Furthermore, the WPNCs Expert Group on Uncertainty Analysis for Criticality Safety Assessment (UACSA) was created in December 2007 to assess calculation uncertainty in Criticality Safety studies.

Exercise Phase I was conducted in order to illustrate predictive capabilities of criticality validation approaches, which include similarity assessment, selection of relevant experiments, determination of k_{eff} bias and bias uncertainty [7]. Unfortunately, this numerical benchmark proposed application cases corresponding to experimental benchmarks of the OECD Experiment data base IHECSBE [8] : thus the projected calculated eigenvalue is known ($K_{\text{eff}}=1$), that limits the actual spread among contributor results.

Therefore, in the framework of the UACSA Expert Group, we propose a numerical blind benchmark. Due to the challenging calculation of Pu resonant reaction rates in intermediate spectra, we have selected MOX powder applications encountered in fuel cycle facilities. This new benchmark should allow the evaluation of the ability of validation methods based on mock-up experiments to determine the Bias on the predicted k_{eff} and the corresponding bias Uncertainty.

2. BLIND BENCHMARK ON MOX POWDERS

Three PuO₂ contents are proposed : 100%, 30% (maximum PuO₂ load in GEN4-SFR) and 12.5% (maximum PuO₂ content in LWR-MOX pellets). The corresponding dry MOX powder density is respectively: $\rho^{\text{MOX}} = 3.5, 5.5$ and 4.6 g/cm^3 .

Two Pu isotopic vectors are considered:

- 71%, 17%, 11%, 1% for ²³⁹Pu up to ²⁴²Pu, corresponding to a conservative Pu vector..
- 64%, 23%, 10%, 3%, corresponding to a realistic Pu vector obtained from reprocessed LWR 30 GWd/T fuels. ²³⁸Pu and ²⁴¹Am poison concentrations are neglected to increase conservatism.

Concerning, the UO₂ powder, a conservative uranium isotopic enrichment is set to 1.2%.

For these 6 cases the powder moisture rate is $M_{\text{H}_2\text{O}} / (M_{\text{H}_2\text{O}} + M_{\text{UO}_2\text{-PuO}_2}) = 3\%$.

The MOX medium is a sphere, surrounded by 20 cm water reflector. The fixed radius of the 6 spheres is:
 $R_{\text{MOX}} = 17.0 \text{ cm}, 22.5 \text{ cm}, 46.0 \text{ cm}$ respectively for the 100%, 30%, 12.5%PuO₂ cases with the conservative Pu.

$R_{\text{MOX}} = 17.7 \text{ cm}, 24.1 \text{ cm}, 52.5 \text{ cm}$ respectively for the 100%, 30%, 12.5% PuO₂ cases with the realistic Pu vector.

These quasi-critical radii were assessed using CRISTAL-V2 [9] calculations based on the JEFF3.1.1 Nuclear Data Library [10].

The benchmark specifications for the 6 application cases are summarized in Table I.

Table I. Specifications of the MOX wet powder benchmark (3% w/o H₂O)

Benchmark	MOX Radius	$M_{\text{PuO}_2}/M_{\text{MOX}}$	²³⁹ Pu/Pu	²⁴⁰ Pu/Pu	²⁴¹ Pu/Pu	²⁴² Pu/Pu
Case 1	17.0 cm	100%	71%	17%	11%	1%
Case 2	22.5 cm	30%				
Case 3	46.0 cm	12.5%				
Case 4	17.7 cm	100%	64%	23%	10%	3%
Case 5	24.1 cm	30%				
Case 6	52.5 cm	12.5%				

Atom densities for the six benchmark cases are given in Table II.

Atom densities in the surrounding water reflector are: $N_{\text{H}_2\text{O}} = 3.336\text{E-}02$ (in 10^{24} atom/cm³)

Table II. Atom densities (10^{24} atom/cm³) in the fissile spheres

Isotope	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
²³⁵ U	-	1.0434E-04	1.0909E-04	-	1.0434E-04	1.0909E-04
²³⁸ U	-	8.4823E-03	8.8679E-03	-	8.4823E-03	8.8679E-03
²³⁹ Pu	5.5222E-03	2.6033E-03	9.0722E-04	4.9800E-03	2.3475E-03	8.1816E-04
²⁴⁰ Pu	1.3167E-03	6.2073E-04	2.1632E-04	1.7863E-03	8.4186E-04	2.9337E-04
²⁴¹ Pu	8.4844E-04	3.9998E-04	1.3939E-04	7.6775E-04	3.6198E-04	1.2615E-04
²⁴² Pu	7.6812E-05	3.6211E-05	1.2619E-05	2.3034E-04	1.0859E-04	3.7845E-05
¹⁶ O	1.5528E-02	2.4494E-02	2.0505E-02	1.5528E-02	2.4494E-02	2.0505E-02
H ₂ O	3.6180E-03	5.6860E-03	4.7560E-03	3.6180E-03	5.6860E-03	4.7560E-03

The required results for each application case are:

- 1/ a brief description of codes and Nuclear Data library used
- 2/ the calculated k_{eff}
- 3/ the one-group integrated sensitivities to multiplicity, fission, capture and scattering of each isotope
- 4/ the k_{eff} prior uncertainty
- 5/ the selected relevant experiments
- 6/ the calculation bias (thus the projected k_{eff}) and the corresponding posterior uncertainty, using experiment information.

3. CRISTAL AND RIB RESULTS

3.1. Calculation codes and nuclear data library

The benchmark calculation was carried out with the new criticality package CRISTAL-V2. The multiplication factor K_{eff} was calculated both with the deterministic route APOLLO2.8 [11] using the *SHEM-MOC* calculation scheme [12] (based on the SHEM 281-group [13]) and the reference continuous-energy Monte Carlo route TRIPOLI4 [14]. Both codes use the JEFF3.1.1 library particularly validated for safety-criticality calculations [15] and LWR calculations [16]. Associated with the CRISTAL package, the RIB code [17] allows the selection of representative experiments and Bias plus Uncertainty calculation.

3.2. K_{eff} calculation results

JEFF3.1.1-based results in Table III are consistent between deterministic and Monte-Carlo calculations.

Table III. Calculation results for the six MOX-powder cases

K_{eff} CRISTAL	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
TRIPOLI4	0.99859	0.99854	0.99889	0.99778	1.00010	0.99930
APOLLO2.8	1.00056	1.00019	1.00015	0.99951	1.00092	1.00000
AP2 – T4 (pcm)	197±35	165±11	126±18	173±36	82±11	70±29

3.3. Sensitivity profiles

K_{eff} sensitivity profiles to nuclear data have been obtained from the First Order Perturbation Theory implemented in APOLLO2. Sensitivity profiles to the cross sections and multiplicities were calculated on the SHEM-281g mesh. The fine mesh below 23eV in SHEM allows the fine structure calculation of the adjoint flux in the first resonances of actinides. Afterwards, the sensitivity coefficients were derived on the European JEF 15-group structure. Sensitivities to $^{239}\text{Pu}(n,f)$ data are plotted in Fig. 1 (cases 1, 2, 3) : K_{eff} of the 12.5% PuO_2 powder is highly sensitive to ^{239}Pu fission at intermediate energy (resolved resonance range), while the sensitivity becomes predominant in the fast energy range for the 100% PuO_2 .

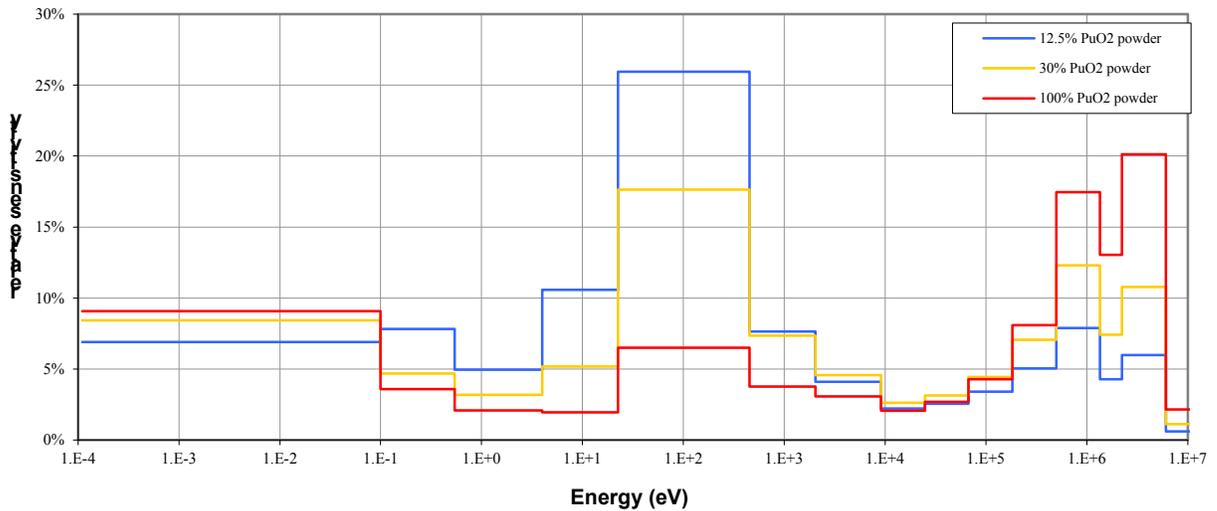


Figure 1: Sensitivity profiles to ^{239}Pu fission for MOX Applications

The integrated one-group sensitivities are summarized in Tables IV and V, respectively for the ‘conservative’ and ‘realistic’ Pu. They are quite similar for the two different isotopic vectors.

It is worth noting that JEFF3.1.1-based sensitivities in Table IV are consistent within 5% with previous JEF2-based sensitivities [18].

Table IV. Integrated sensitivity coefficients in pcm/% (cases 1, 2, 3 with conservative Pu)

Application case		1: 100%PuO ₂	2: 30%PuO ₂	3: 12.5%PuO ₂
Fission	²³⁹ Pu	422	376	350
	²⁴⁰ Pu	45	24	13
	²⁴¹ Pu	90	96	95
	²⁴² Pu	2	1	1
	²³⁵ U	-	12	33
	²³⁸ U	-	50	76
ν	²³⁹ Pu	758	687	631
	²⁴⁰ Pu	66	35	19
	²⁴¹ Pu	174	175	164
	²⁴² Pu	3	2	1
	²³⁵ U	-	24	63
	²³⁸ U	-	78	121
Capture	²³⁹ Pu	-105	-128	-135
	²⁴⁰ Pu	-29	-37	-45
	²⁴¹ Pu	-20	-21	-20
	²⁴² Pu	-2	-2	-3
	²³⁵ U	-	-5	-12
	²³⁸ U	-	-86	-180
	¹⁶ O	-2	-3	-4
	¹ H_H ₂ O	-84	-66	-45
Scattering	²³⁹ Pu	6	1	-1
	²⁴⁰ Pu	2	0	0
	²⁴¹ Pu	1	0	0
	²⁴² Pu	0	0	0
	²³⁵ U	-	0	0
	²³⁸ U	-	6	-14
	¹⁶ O	6	5	-4
	¹ H_H ₂ O	39	72	71

K_{eff} of the MOX powder configurations is particularly sensitive to variations of ²³⁹Pu and, to a less extent, ²⁴¹Pu nuclear data. Tables IV and V show a significant sensitivity to ²³⁸U resonant capture cross-sections, due to an intermediate neutron spectrum (demonstrated by the weak thermal slowing-down density : q_{∞} = 0.14 and 0.06 respectively for 12.5% and 30% PuO₂ powders).

Table V. Integrated sensitivity coefficients in pcm/% (cases 4, 5, 6 with realistic Pu)

Application case		4: 100%PuO ₂	5: 30%PuO ₂	6: 12.5%PuO ₂
Fission	²³⁹ Pu	409	374	352
	²⁴⁰ Pu	64	34	19
	²⁴¹ Pu	89	97	98
	²⁴² Pu	7	4	2
	²³⁵ U		13	37
	²³⁸ U		52	79
ν	²³⁹ Pu	728	667	613
	²⁴⁰ Pu	93	50	28
	²⁴¹ Pu	169	171	161
	²⁴² Pu	10	5	3
	²³⁵ U		26	68
	²³⁸ U		81	127
Capture	²³⁹ Pu	-101	-122	-128
	²⁴⁰ Pu	-37	-46	-55
	²⁴¹ Pu	-19	-20	-19
	²⁴² Pu	-4	-5	-6
	²³⁵ U		-5	-13
	²³⁸ U		-91	-188
	¹⁶ O	-3	-4	-5
	¹ H_H ₂ O	-81	-60	-35
Scattering	²³⁹ Pu	4	0	-1
	²⁴⁰ Pu	2	0	0
	²⁴¹ Pu	1	0	0
	²⁴² Pu	0	0	0
	²³⁵ U		0	0
	²³⁸ U		2	-24
	¹⁶ O	4	2	-9
	¹ H_H ₂ O	32	58	49

3.4. Uncertainty on nuclear data and K_{eff} prior uncertainty

The uncertainty $\varepsilon = \Delta I / I$ on a neutronic parameter I due to nuclear data is given at the first order by the “sandwich” rule [19]:

$$\varepsilon_A = \left(S_A^+ D S_A \right)^{1/2} \quad (1)$$

D: nuclear data multigroup Covariance matrix

$$S_A = \frac{\sigma^j}{I_A} \cdot \frac{dI_A}{d\sigma^j} : \text{Sensitivity vector of the Application parameter } I_A \text{ to nuclear data}$$

$S_{K_{\text{eff}}}$ sensitivity vector to nuclear data are obtained from CRISTAL calculations, as described in the previous section. Then, the K_{eff} prior uncertainty is automatically determined by the RIB code [17].

In the new CRISTAL-V2 package, the JEFF3.1.1 library is used. Covariances for this library are more reliable because they were obtained from a rigorous process : for each major actinide, the JEFF3.1.1 covariance matrix was obtained from the Nuclear Data re-estimation based on targeted experiments [20]. Prior and posterior ND Uncertainties (diagonal term of covariance matrices) are compared in Table VI. These covariance matrices are stored in the library COMAC (COVariance MATrix Cadarache [21]).

Table VI. ^{235}U Prior and Posterior uncertainty (%)

Group/Energy	$(n,\gamma)^{\text{prior}}$	$(n,\gamma)^{\text{post}}$	$(n,f)^{\text{prior}}$	$(n,f)^{\text{post}}$	v^{prior}	v^{post}
Gr12 / 23-4 eV	10	3.6	3.0	2.3	0.8	0.7
Gr13 / 4-0.5 eV	10	3.7	3.0	2.4	0.8	0.7
Gr14 / 0.5-0.1eV	2	1.6	1.0	0.8	0.8	0.5
Gr15 / E<0.1eV	1	0.8	0.4	0.3	0.4	0.2

The RIB results for the K_{eff} prior uncertainty are summarized in Table VII. Prior uncertainties are reduced in JEFF3.1.1-based calculations. The uncertainty level remains similar in cases characterized by either ‘conservative’ or ‘realistic’ Pu vector, due to the small uncertainty component linked to ^{242}Pu .

Table VII. Prior uncertainty (1σ) on the K_{eff} of the six fissile spheres

Application case	1: 100%PuO ₂	2: 30%PuO ₂	3: 12.5%PuO ₂	4: 100%PuO ₂	5: 30%PuO ₂	6: 12.5%PuO ₂
JEF2 calculation	1590 pcm	1340 pcm	1310 pcm	1600 pcm	1350 pcm	1330 pcm
JEFF3.1.1 calculation	1380 pcm	1150 pcm	1060 pcm	1390 pcm	1160 pcm	1080 pcm

3.5. Selected representative experiments

In order to select Experiments relevant for the industrial Application we use the Representativity r_{AE} [23] :

$$r_{\text{AE}} = \frac{(S_A^+ DS_E)}{\varepsilon_A \cdot \varepsilon_E} \quad (2)$$

This correlation coefficient r_{AE} represents the share of information provided by the experiment E common with the parameter $K_{\text{eff},A}$. A value of 0.0 means that the Experiment and the Application are not correlated. Information provided by the experiment E will not be of any utility for the validation of the application. On the contrary, a value of 1.0 indicates a full correlation between Experiment and Application that allows a strong calculation uncertainty reduction.

In this preliminary work, only critical experiments from the French experimental database were selected:
 - The ERASME experiments [22], achieved in the EOLE facility at Cadarache in the 1984-1986 period, was built up to reduce the uncertainties on High Conversion PWR parameters. MOX 11%Pu fuel rods are

used in tight lattices ($V_{\text{mod}}/V_{\text{MOX}}$ ranging from 0.5 up to 2.1). The tightest configuration ERASME/S is available in the Experiment data base IHECSBE as MIX-COMP-INTER-005.

- The 1A' experiment has been carried out in the MASURCA facility at Cadarache in 1969: it was conceived within the framework of the Fast Breeder Reactor design. The core was constituted of U-Pu metallic fuel assemblies. The moderator was graphite.

The main characteristics of these two uncorrelated experiments are summarized in Table VIII.

Table VIII. Selected U-Pu experiments in EOLE and MASURCA reactors

Experiment	Pu (%)	²⁴⁰ Pu (% Pu)	Lattice pitch (cm)	Moderator	$V_{\text{mod}}/V_{\text{MOX}}$	Slowing-down density q_{∞}	$\epsilon_{\text{exp}} (1\sigma)$ (pcm)
1A'	25	8.4	□ 1.39	Graphite	4.1	0.002	500
ERASME/S	11	18.6	△ 0.945	Water	0.5	0.28	400
ERASME/R			△ 1.035		0.9	0.42	400
ERASME/L			□ 1.19		2.1	0.66	310

△ - triangular pitch, □ - square pitch

Table IX gives respectively the Representativity factor r of these critical experiments to the various low-moderated MOX powders ('conservative' Pu vector). ERASME/S presents the highest correlation coefficient with respect to 12.5% PuO₂ powder with $r = 0.987$ while experiment 1A' is more representative of the powders with 30 and 100% PuO₂ content (respectively $r = 0.964$ and 0.929).

Table IX. Representativity factor

Experiment	12.5% PuO ₂ powder	30% PuO ₂ powder	100% PuO ₂ powder
ERASME/S	0.987	0.907	0.711
ERASME/R	0.949	0.866	0.666
1A'	0.882	0.964	0.929

3.6. Calculation Bias and bias Uncertainty

Due to the same fuel pins, experiments ERASME/R and ERASME/S are strongly correlated. Therefore, only ERASME/S and 1A' experiments are taken into account in the RIB assessment of the CRISTAL-V2 calculation bias and associated posterior uncertainty.

Each measured parameter $I_{E_i} \pm \delta_i$ affects the computation parameter I_A and its posterior uncertainty accordingly to its weight w_i :

$$w_i = \frac{1}{1 + \delta_i^2 / \epsilon_{E_i}^2} \quad (3)$$

The lower its measurement uncertainty relatively to computation uncertainty, the more the experiment will influence the transposition from Experiment to Application.

When using only two experiments, the ΔI_A computation error and the posterior uncertainty ϵ_A^* due to nuclear data are obtained respectively by formula (4) and (5):

$$\frac{\Delta I_A}{\varepsilon_A} = \frac{1}{1 - \hat{r}_{1,2}^2 \cdot w_1 \cdot w_2} \left[r_{A,1} \cdot w_1 \cdot \frac{\Delta I_{E_1}}{\varepsilon_{E_1}} + r_{A,2} \cdot w_2 \cdot \frac{\Delta I_{E_2}}{\varepsilon_{E_2}} - \hat{r}_{1,2} \cdot w_1 \cdot w_2 \left(r_{A,2} \cdot \frac{\Delta I_{E_1}}{\varepsilon_{E_1}} + r_{A,1} \cdot \frac{\Delta I_{E_2}}{\varepsilon_{E_2}} \right) \right] \quad (4)$$

$$\frac{\varepsilon_A^{*2}}{\varepsilon_A^2} = 1 - \frac{1}{1 - \hat{r}_{1,2}^2 \cdot w_1 \cdot w_2} \left(r_{A,1} \cdot \sqrt{w_1} - r_{A,2} \cdot \sqrt{w_2} \right)^2 - \frac{2}{1 + \hat{r}_{1,2} \cdot \sqrt{w_1 \cdot w_2}} \cdot r_{A,1} \cdot r_{A,2} \cdot \sqrt{w_1 \cdot w_2} \quad (5)$$

with $\Delta I_E = \frac{I_E^m - I_E}{I_E}$: Calculation-Experiment bias on the measured parameter

$\hat{r}_{1,2}$: correlation coefficient between experiment 1 and experiment 2.

Correlation coefficient $\hat{r}_{1,2}$ gathers similarity between experiments ($r_{1,2}$) and technological/measurement correlation. As we choose 2 independent measurements, $\hat{r}_{1,2}$ becomes $r_{1,2}$: $\hat{r}_{ERASME,1A'} = r_{ERASME,1A'} = 0.89$.

The transposition of C/E discrepancies accordingly to (4) supplies the TRIPOLI4 calculation error for each MOX powder application. Results shown in Table X allow the conclusion that CRISTAL-V2 error of wet MOX powders is small : the scaling correction $-\Delta I_A$ ranges between -156 pcm and -422 pcm.

The posterior uncertainty derived from formula (5), associated to the projected- K_{eff} , is reduced at least by a factor 2 thanks to the selected U-Pu experiments. This uncertainty does not depend on 'conservative' or 'realistic' Pu vector because the representativity factor does not vary significantly with the Pu vector.

Table X. Prior and posterior $K_{eff} \pm 1\sigma$ of TRIPOLI4-JEFF3.1.1 calculation

CRISTAL-V2 K_{eff}	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
Scaling correction	-158 pcm	-251 pcm	-420 pcm	-156 pcm	-249 pcm	-422 pcm
Prior T4 results	0.99859 ± 0.01380	0.99854 ± 0.01150	0.99889 ± 0.01060	0.99778 ± 0.01390	1.00010 ± 0.01160	0.99930 ± 0.01080
Projected T4 results	0.99701 ± 0.00721	0.99603 ± 0.00445	0.99469 ± 0.00395	0.99622 ± 0.00728	0.99761 ± 0.00447	0.99508 ± 0.00394

4. CONCLUSIONS

In the framework of the WPNCs Uncertainty expert Group, we propose a numerical blind benchmark on MOX wet powders. Three PuO₂ contents are investigated : 100%, 30% and 12.5%. Two Pu isotopic vectors are considered : a conservative one (0%, 71%, 17%, 11%, 1% for ²³⁸Pu up to ²⁴²Pu) and a realistic one obtained from reprocessed LWR 30 GWd/T fuels. For these 6 cases the powder moisture rate is 3%. The MOX fissile medium is a sphere, surrounded by 20 cm water reflector. The radius of the three quasi-critical spheres is : $R_{MOX} = 17, 22.5, 46$ cm respectively for 100%, 30%, 12.5% PuO₂ (conservative Pu).

We have presented the CRISTAL-V2 results based on the JEFF3.1.1 library. The K_{eff} prior uncertainty amounts from 1080 pcm for the 12.5%PuO₂ up to 1380 pcm for the 100%PuO₂ load. Using only EOLE and MASURCA benchmark experiments in the RIB automated tool, the projected- K_{eff} were obtained for the six application cases and the associated posterior uncertainties were reduced by about 60%. This preliminary study will be improved by the use of additional relevant experiments such as BFS/MOX.

ACKNOWLEDGMENTS

This work was supported by CEA and AREVA-NC.

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