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EXPERIMENTAL VALIDATION OF THE FRENCH CRISTAL V1.0 PACKAGE

N. LECLAIRE, I. DUHAMEL
Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
BP 17 - 92262 Fontenay aux Roses Cédex, France
nicolas.leclaire@irsn.fr
isabelle.duhamel@irsn.fr

E. GAGNIER[1], Y.K. LEE[1], C. VENARD[2]
[1] Commissariat à l'Energie Atomique (CEA)
BP 28 - 91191 Gif sur Yvette Cédex, France
emmanuel.gagnier@cea.fr; yklee@cea.fr
[2] Commissariat à l'Energie Atomique (CEA)
13108 St Paul lez Durance Cédex, France
christophe.venard@cea.fr

ABSTRACT

CRISTAL criticality safety package new version, CRISTALV 1.0, has been developed by the Institut de Radioprotection et de Sûreté Nucléaire and the Commissariat à l'Energie Atomique, in collaboration with the COGEMA company, mainly to allow taking into account burn-up credit in criticality safety studies. Meanwhile, the validation database, which was made up of about 500 benchmarks for CRISTAL V0 package, was extended. CRISTAL includes two calculation routes, using nuclear data taken from the JEF2.2 library: a standard route based on multi-group cross-sections (APOLLO2 - MORET 4 or APOLLO2 Sn calculations) and a reference route using the pointwise data (TRIPOLI4 calculations). The CRISTAL V1.0 validation database is made up of 2132 critical experiments mainly taken from the OECD/ICSBEP Handbook or performed in French facilities. Most of them have already been investigated with the different CRISTAL routes. The first trends of the validation work point out that the calculation results are generally in good agreement with the benchmark k_{eff} and that the new calculation schemes improve the validation results.

Key Words: experimental validation, fission products, APOLLO2, MORET 4, TRIPOLI4

1 INTRODUCTION

The CRISTAL package [1] contains two calculation routes which use JEF2.2 nuclear data: on the one hand, the "standard route" involving the multi-group nuclear data library CEA93 (derived from JEF2.2 evaluation), the APOLLO2 cell code (used for self-shielding, flux calculations, collapsing and homogenisation) and the MORET 4 Monte Carlo code (for 3D calculation with a general P_n-like anisotropy treatment), and, on the other hand, the "reference route" using the TRIPOLI4 Monte Carlo code with a continuous energy JEF2.2 library [2].

In the framework of CRISTAL V1.0 [3], new versions of cross sections library and computer codes have been developed (see Table I.).

Table I. Versions of codes and libraries for CRISTAL routes

	CODES/LIBRARIES	VERSIONS
STANDARD ROUTE	CIGALES GUI*	3.0
	APOLLO2 CODE	5.4
	LIBRARY	CEA93 V6
	MORET 4 CODE	B.2
REFERENCE ROUTE	TRIPOLI4 code	4.3
	Library	JEF2.2

* Graphical User Interface

Regarding the different calculation options of the "standard route", an important validation work was performed in order to improve CRISTAL package accuracy and optimise calculation time. For example, a P3 anisotropy representation and a 20-groups energy structure were used for APOLLO2 Sn calculations (with S8 order or S16 for metallic systems), whereas a P5 anisotropy and a 172-groups energy structure were adopted in MORET 4 calculations. In fact, these approximations do not significantly increase the Monte Carlo calculation time and give more accurate results. Finally, the recommended calculation schemes were introduced in procedures libraries to make easier the use of the "standard route".

Meanwhile, an extensive validation work has been performed. Thus, the initial validation database of CRISTAL V0 [4], which was made up of more than 500 critical experiments, is being extended to 2132 benchmarks.

2 VALIDATION DATABASE

Validation is mainly based on calculation-experiment comparisons. The observed discrepancies are then interpreted and eventually transposed to actual configurations. Therefore critical experiments were selected to investigate all the operations encountered in the nuclear fuel cycle (including fabrication, transport, storage and reprocessing).

The different criteria of the database selection were:

- various fissile media,
- diversity of configurations (to cover a wide moderation ratio range and to validate different materials),
- variety of laboratories (to detect experimental biases),
- quality of experimental data provided in benchmarks.

The main sources of the benchmarks selection are, on the one hand, the OECD/ICSBEP Handbook [5] and, on the other hand, experiments performed by IRSN in the Valduc facility (Apparatus B, MARACAS), some of them with the financial support of the COGEMA company [6], as well as experiments performed by CEA in Saclay (Alecto) and in Cadarache (Eole, Minerve).

The validation database, which was made up of more than 500 critical experiments for CRISTAL V0 package, is being extended for the new V1.0 version:

- to take into account the needs highlighted for the last few years (mainly configurations involving slabs arrays or interacting metallic systems),
- to validate burn up credit calculations (nuclear data of major Fission Products),
- to validate the new "multi-cell Pij flux calculation" options,
- to have a better statistics in terms of experiments number for the different categories, a larger diversity of laboratories,
- to investigate inconsistencies detected in the CRISTAL V0 validation studies (for example, two experimental programs dealing with mixed uranium and plutonium solutions [4] were added in the database because of observed inconsistencies [7]).

Finally, CRISTAL V1.0 experimental validation database is composed of 2132 critical experiments (see Table II), amongst which 1926 were selected for APOLLO2-MORET 4, 854 for APOLLO2-Sn and 874 for TRIPOLI4.

3 VALIDATION METHOD

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

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).

This initial work is completed from case to case by inter-code comparisons (KENO, MCNP, MONK…) to uncouple the different sources of code biases (nuclear data inaccuracies and models approximations).

Finally, a comparison with other available experimental programs in the same field (similar media and configurations) allows highlighting experimental biases.

4 MAIN RESULTS

Among the 2132 benchmarks saved in the validation database, 1318 have already been investigated with the "standard route" APOLLO2-MORET 4, 182 with the "standard route" APOLLO2-Sn and 423 with the "reference route" TRIPOLI4 (see Table II).

The main results of the validation are presented by medium type hereafter and summarized in Table III.

Table II. Number of experiments investigated with CRISTAL V1

Physical Form	Fissile material	APOLLO2-MORET 4		APOLLO2-Sn		TRIPOLI 4	
		Validation database	Experiments already investigated	Validation database	Experiments already investigated	Validation database	Experiments already investigated
SOLUTIONS	Low-enriched Uranium	**73**	**73**	**33**	26	**60**	13
	High-enriched Uranium	**140**	95	**156**	40	**70**	20
	Plutonium	**161**	133	**125**	18	**96**	36
	Mixte U-Pu	**52**	**52**	**32**	9	**39**	19
	^{233}U	**42**	**42**	**0**	0	**5**	2
POWDERS	Uranium	**117**	98	**59**	0	**77**	18
	Plutonium	**34**	**34**	**0**	0	**34**	34
	Mixte U-Pu	**33**	**33**	**33**	0	**33**	0
ARRAYS	UO_2 rods	**458**	217	**188**	20	**39**	34
	UO_2-PuO_2 rods	**205**	180	**60**	5	**59**	15
	Slabs	**127**	82	**28**	4	**63**	2
	Uranium metal	**37**	**37**	**0**	**0**	**43**	43
	Pu metal	**0**	**0**	**3**	**3**	**0**	0
ARRAYS IN FISSILE SOLUTIONS	UO_2 rods	**8**	**13**	**6**	**6**	**2**	2
	UO_2-PuO_2 rods	**73**	67	**0**	**0**	**12**	3
INTERACTION	Uranium	**58**	16	**0**	**0**	**58**	15
	Plutonium	**102**	45	**0**	**0**	**59**	59
	Mixte U-Pu	**49**	**49**	**0**	**0**	**0**	**0**
	Metal	**63**	14	**13**	0	**63**	63
METAL	Uranium	**43**	22	**58**	41	**35**	24
	Plutonium	**21**	15	**27**	10	**18**	18
	Mixte U-Pu	**28**	0	**33**	0	**5**	0
	^{233}U	**1**	**1**	**0**	**0**	**3**	**3**
	^{237}Np	**1**	**0**	**0**	**0**	**1**	0
TOTAL		**1926**	**1318**	**854**	**182**	**874**	**423**

Supprimé : the APOLLO2-MORET 4

Supprimé : 459

Supprimé : 120

Supprimé : 61

Supprimé : 43

Supprimé : 58

Supprimé : -

4.1 Solutions

4.1.1 Low-enriched Uranium solutions

The investigated experimental programs involved low enriched $UO_2(NO_3)_2$ and UO_2F_2 solutions with an uranium concentration ranging from 170 g/l to 980 g/l. Different tanks and reflectors were studied.

Calculations pointed out a good k_{eff} prediction, except for LEU-SOL-THERM-001 experiment (UO_2F_2 with an uranium concentration of about 980 g/l), which leads to an over-prediction of about 1.5 %. Similar results were obtained with other codes (MONK and MCNP). Thus, this ensured that the discrepancy observed is code independent.

4.1.2 High-enriched Uranium solutions

The experiments involved mainly UO_2F_2 and $UO_2(NO_3)_2$ solutions. The uranium concentration ranged from 20 g/l to 730 g/l. Some of them were performed with soluble gadolinium.

A general trend to k_{eff}'s over-prediction was observed especially for high Uranium concentrations. This over-prediction of about 0.7 % is mainly due to the JEF2.2 ^{235}U cross-sections, the use of JEFF3.0 ^{235}U cross-sections allowing reducing these discrepancies [8].

4.1.3 Plutonium solutions

These experiments covered a broad range of plutonium concentration (10 g/l to 270 g/l) with various ^{240}Pu contents.

Compared with the experimental values, the calculated results led to a slight over-prediction. There is no obvious trend, neither with plutonium concentration nor with the ^{240}Pu content.

Given the diversity of the systems studied and their origin and the different code systems used in our analysis [7], it is concluded that this slight over-prediction is a code-independent commentary on the accuracy of the JEF2.2 files for plutonium data.

Finally, it can be noticed that the same trend is observed for plutonium solutions in interaction with APOLLO2-MORET 4 standard route.

4.1.4 Mixed Uranium and Plutonium solutions

The experiments were carried out in different tanks with homogeneous mixed U-Pu nitrate solutions. Different ^{240}Pu contents were studied.

On the whole, good results are obtained, except for MIX-SOL-THERM-003 benchmark, which leads to contradictory results.

4.1.5 ^{233}U solutions

42 critical experiments involving 98 % ^{233}U enriched $UO_2(NO_3)_2$ or UO_2F_2 solutions were investigated with APOLLO2-MORET 4. Amongst them, 2 experiments were also calculated with TRIPOLI4. The uranium concentration ranged from 49 g/l to 866 g/l.

A good calculation-experiment agreement was observed for uranyl nitrate solutions, which can let think that the ^{233}U nuclear data are relevant in the thermal energy range.

However, the experiments involving UO_2F_2 led to an important under-prediction of k_{eff}.

Regarding TRIPOLI4 results, with the two investigated experiments involving UO_2F_2, a more important under-prediction of k_{eff} was observed, which is due to the problem of Be reflector cross section in JEF2.2 library. This deficiency of Be cross section in JEF2.2 was discussed in previous studies ([2] and [9]). Using Be cross section from ENDF/B-VI.4 in TRIPOLI4 calculations, the under-prediction of k_{eff} involving ^{233}U uranyl fluoride solution is clearly improved and TRIPOLI4 (ENDF/B-VI.4) gives a very close k_{eff} comparing with MCNP4C (ENDF/B-VI.6).

Notice that the Beryllium nuclear data used in CEA93 V4 library for APOLLO2-MORET 4 calculations have already been replaced by data from ENDF/B-VI evaluation.

Several comparisons with VIM (ENDF/B-V), MCNP (ENDF/B-VI) and COG (ENDF-ENDL) codes showed that calculation-experiment discrepancies are code independent; thus it allows questioning one of the other sources of discrepancies (experimental biases or uncertainties and nuclear data inaccuracy in this energy range).

4.2 Powders

The investigated experiments concerned:

- low-enriched (5 %) uranium oxide powder with an atomic ratio H/U of 2, 2.5 or 3, reflected by polyethylene,
- high-enriched (93 %) uranium oxide powder reflected by polyethylene,
- low-enriched (2 and 3 %) UF_4 powder moderated by paraffin with an atomic ratio H/U ranging from 4 to 20 and reflected by hydrogenous materials (paraffin, polyethylene, plexiglas),
- plutonium oxide powder or UO_2-PuO_2 powder moderated by polystyrene (with an atomic ratio H/Pu or H/U+Pu ranging from 0 to 50), bare or plexiglas reflected.

Regarding UO_2-PuO_2 and PuO_2 powders, the results showed an important dispersion of the calculated results as for other codes (MCNP,…). Nevertheless, these discrepancies can be attributed to weak or missing experimental data.

Considering the experiments involving uranium powders (UO_2 and UF_4), the calculations are in good agreement with experimental k_{eff}; in fact, most of the calculation-experiment discrepancies are in the uncertainty range.

4.3 Fuel rods arrays

4.3.1 UO_2 and UO_2-PuO_2 rods

The investigated experiments involved arrays of low-enriched UO_2 or UO_2-PuO_2 (with different PuO_2 and ^{240}Pu contents) pins. Different square and triangular lattices of water-moderated were considered in order to cover a wide range of moderation ratio.

Some of these configurations involved soluble poisons (boron or gadolinium), absorbing canisters (borated steel, boral, hafnium, cadmium…) and different kinds of reflectors (water, polyethylene, concrete, steel, lead…).

The calculations showed good agreements with experimental k_{eff} for the well-thermalised UO_2 and UO_2-PuO_2 lattices; in fact, the discrepancies were within the uncertainty ranges, without any discernible trend with moderation ratio.

However, differences between the standard route (APOLLO2-MORET 4 and APOLLO2-Sn) and the reference route (TRIPOLI4) were noticed for rod arrays with high flux heterogeneity (for example cases involving canisters). These discrepancies were tracked down to the approximations used in APOLLO2 code for array calculations (cell model and homogenisation), which leads to over-predicted k_{eff} for APOLLO2-MORET 4 calculations. New schemes involving APOLLO2 multi-cell calculations (based on the interface current method) are currently being studied; first results seemed to show an improvement in the k_{eff} prediction for such configurations.

Finally, an overestimation could be noticed (up to 1.5 %) with the standard route, for steel reflected experiments, which increases when the distance between the reflecting walls and the assemblies decreases. Further analyses [10] have highlighted that this over-prediction is mainly linked to the multi-group treatment of the iron isotopes (predominantly ^{56}Fe). In fact, the point-wise Monte Carlo code TRIPOLI4 gives satisfactory results without any trend with core-reflector distance.

4.3.2 Slabs arrays

With regard to the standard route, the investigated experimental programs involved water-moderated highly enriched (93 % ^{235}U) UAl plates, or Uranium metal slabs moderated with polyethylene, plexiglas or teflon.

Regarding uranium metal slabs, a good agreement between the experimental k_{eff} and the benchmark k_{eff} can be pointed out, except for cases involving teflon as moderator.

Regarding fuel plates assemblies, two cases can be considered: when the spacing between fuel elements is low, the calculations led to satisfactory results. However, for larger spacing, an under-prediction could be observed. These discrepancies might be reduced when using the new calculation scheme for plate lattices, which is currently being developed in collaboration with CEA in the framework of CRISTAL project.

4.4 Fuel rods arrays in fissile solution

These experiments concerned UO_2 or UO_2-PuO_2 fuel rods in uranium, plutonium or mixed U-Pu nitrate solutions. Different lattices pitches and various solutions concentrations were tested.

These configurations allowed validating the APOLLO2 calculation options for self-shielding, taking into account the resonant isotopes in both the fissile solution and the fuel rods.

For the cases involving UO_2 rods, the benchmark performed showed a trend to under-estimation (about 0.8%). Notice that these experiments, which were the only experiments investigated, had not been re-evaluated yet (no experimental uncertainties are available). So, it was difficult to conclude about models or nuclear data accuracy.

The experiments performed with UO_2-PuO_2 rods in plutonium or mixed U-Pu solutions showed a good agreement. Notice that two experimental programs have not been re-evaluated yet and that some uncertainties subsist on experimental data.

4.5 Metallic systems

High-enriched uranium and plutonium metallic systems were studied. Different reflectors with varying thickness were investigated (none, water, polyethylene, steel, graphite, aluminium).

Main results have pointed out:

- a slight k_{eff} under-estimation of about 0.5% for bare, water-reflected or CH_2-reflected uranium and plutonium metallic systems for all the different routes;
- an important over prediction for steel reflected cases (0.7% for a 4.49 cm-thick steel reflector and 4.5% for a 19.65 cm-thick steel reflector), which increases with the reflector thickness, for the standard route; as said previously, the over-estimation with steel reflector is mainly linked to the multi-group treatment of the iron isotopes.
- an over prediction for graphite reflected cases, which also increases with the reflector thickness for APOLLO2-MORET 4 calculations; the discrepancies observed, which were not shown with TRIPOLI4 code, are being studied. First conclusions engaged the graphite anisotropy treatment (presently in P1 order); it should be noticed that APOLLO2-Sn calculations did not show correlation between reflector thickness and C/E discrepancy.

New calculations were performed for the metallic media using the new CEA93 cross-section library, which contains major actinides cross sections with a P9 anisotropy order. Preliminary results with APOLLO2-MORET 4 route revealed a statistically insignificant impact on k_{eff} (less than 0.3 %) for metallic homogeneous media. The influence of cross-section anisotropy seemed to be more important for the APOLLO2-Sn route.

Moreover, configurations with several metallic systems in interaction have been studied. They involved arrays of uranium or plutonium metal cylinders reflected or not by water or paraffin. These configurations showed good calculation-experiment agreement.

Finally, one case with ^{233}U was investigated with APOLLO2-MORET 4 standard route and three with TRIPOLI4 code. A high under-prediction of about 1.2 % was observed with APOLLO2-MORET 4; according to TRIPOLI4 calculation results, the cross section of ^{233}U from JEF2.2 could under-predict k_{eff} of 2.8% for U233-MET-FAST systems compared with ENDF/B-VI.4.

4.6 Fission products

These experiments were performed by IRSN in Valduc with the financial support of the COGEMA company [6]. IRSN and COGEMA being the only owners, they have only been investigated by IRSN with the APOLLO2-MORET 4 standard route.

The first series of experiments, called “Physical” type experiments, which involved a solution of fission products, alone or mixed, in a Zr tank placed in the centre of a rods array driver is representative of storage and transportation conditions. Some of them (with ^{149}Sm) are available in the ICSBEP handbook (LEU-COMP-THERM-050).

To improve the “dissolution” qualification, a second series of experiments, named “Elementary Dissolution” type, has been performed. Fission products were then in close interaction with the U, Pu and Am isotopes of inner array. This second series is itself divided in two cases: fission products in acid solutions or in depleted uranyl nitrate solution.

Other experiments were performed to validate natural Gd, ^{95}Mo (with thin slices of CH_2/natural metallic Mo) and fluorine (with polytetrafluorethylene solid block).

A third series of experiments, named “Global or Advanced Dissolution” type, was achieved last year. It consists in a UO_2-PuO_2 rod array steeping in a depleted uranyl nitrate solution poisoned with 6 fission products.

The 6 selected fission products for these experiments are: ^{103}Rh, ^{133}Cs, ^{143}Nd, ^{149}Sm, ^{152}Sm, ^{155}Gd. The results of the first series “Physical” and “Elementary dissolution” showed good calculation-experiments agreement (< 0.4%) and low experimental uncertainties (< 0.1%).

4.7 Perspectives

The validation work is still in progress and will concern 2132 experiments for the CRISTAL V1.0 package.

The results of further investigations will concern more particularly:

- the slabs arrays to define a more precise calculation scheme,
- the critical experiments whose configurations involve high heterogeneity of flux, allowing validating the new “multi-cell Pij flux calculation” option developed in the V1.0 version (first results show a better k_{eff} prediction),
- the “global dissolution” fission products classified experiments performed in Valduc facility,
- ^{233}U cross-sections validation in high energy spectra,
- the anisotropy of graphite cross-sections.

Finally, the results of the 1926 critical experiments calculated with APOLLO2-MORET 4 will be investigated in MACSENS, the new MORET tool [11], in order to assist end-users in the estimation of the code bias.

5 CONCLUSION

In the framework of CRISTAL V1.0 development, an important validation work is being performed.

The extension of the validation database allows covering nearby all the different kinds of configurations encountered in the nuclear fuel cycle.

Validation studies highlighted the effect of the approximations used in the standard route and the nuclear data accuracy. New calculation procedures and models are being validated thanks to critical experiments and reference calculations.

The first trends of the validation work, which currently involve experiments, point out that the calculation results are generally in good agreement with the benchmark k_{eff} and that the improvements in the calculation schemes allow obtaining better validation results.

Finally, it must be emphasized that the validation work is still in progress and will concern 2132 experiments for the CRISTAL V1.0 package.

Table III. Validation main results – main trends with regard to the type of fissile material

MEDIA	CATEGORY	APOLLO2-Sn	APOLLO2-MORET 4	TRIPOLI4 [12]
SOLUTIONS	Uranium lowly enriched	Good results and coherent (36 exp./73 exp./13 exp.)		
	Uranium highly enriched	Over-prediction of k_{eff} (~0.7%) especially for uranium high concentrations (^{235}U of JEF2.2) (40 exp./95 exp./20 exp.)		
	Plutonium	Slight over-prediction (~0.6%) of k_{eff} (Sections JEF2.2) (18 exp./133 exp./36 exp.)		
	Mixed U + Pu	-0.41% < C-E <0.48% (9 exp.)	Good results (46 exp.)	No coherence between the 3 Exp. prog. (19 exp.)
	U233		$UO_2(NO_3)_2$ Good results UO_2F_2 large under-prediction – coherence with other codes (42 exp.)	Good results with ENDF/B-VI* (2 exp.)
POWDERS	Uranium		Good results (98 exp./18 exp.)	
	Plutonium		Over-prediction and high dispersion (34 exp./34 exp.)	
	Mixed U + Pu		High dispersion (33 exp.)	
ARRAYS	UOX	Regular arrays: good results		Good results (34 exp.)
		Storage and transport configurations: high flux heterogeneities Difference in comparison with the reference route (cell model)		
		Under-estimation (20 exp.)	Over-prediction (217 exp)	
	Mixed U + Pu	Good results (5 exp./180 exp./15 exp.)		
	Slabs		Good results Arrays of assemblies: under-estimation with the distance array-array (82 exp.)	
	Metal		One series: under-estimation (37 exp.)	One series: no visible trend (43 exp.)
ARRAYS IN FISSILE SOLUTIONS	Mixed U + Pu		Good results (67 exp.)	Standard deviation (3 exp.)
	UOX	Good results (6 exp.)	Slight under-estimation - coherence with other codes (13 exp./2 exp.)	
INTERACTION	Solutions of U			
	Solutions of Pu		Over-prediction (Pu JEF2.2) (45 exp.)	Good results (59 exp.)
	Mixed arrays		Good results (49 exp.)	
	Metal		Good results (14 exp.)	
METAL	Uranium	Slight under-estimation (~ 0.6%) (51 exp./37 exp./42 exp.)		-0.71% < C-E < 0.35%
	Plutonium			-0.72% < C-E < 1.07%
	Mixed U+Pu			
	U233		High under-estimation (1 exp./3 exp).	

*** Notice that ENDF/B-VI is not included in CRISTAL**

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