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# Critical Experiments That Simulated Damp MOX Powders – Do They Meet The Need?

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J. Blair Briggs
Ali Nouri
Claes Nordborg

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# CRITICAL EXPERIMENTS THAT SIMULATE DAMP MOX POWDERS – DO THEY MEET THE NEED?

**J. Blair Briggs**
Idaho National Laboratory
2525 North Fremont
Idaho Falls, ID 83415-3860
U.S.A.
j.briggs@inl.gov

**Ali Nouri**
Institut De Radioprotection Et De Sûreté Nucléaire
BP 17
92262 Fontenay-aux-Roses Cedex
France
ali.nouri@irsn.fr

**Claes Nordborg**
OECD Nuclear Energy Agency
Le Seine Saint-Germain
12 boulevard des Iles
F-92130 Issy-les-Moulineaux
Claes.nordborg@oecd.org

## ABSTRACT (Abstract Head)

The OECD Nuclear Energy Agency (NEA) Working Party on Nuclear Criticality Safety (WPNCS) identified the MOX fuel manufacturing process as an area in which there is a need for additional integral benchmark data. The specific need focused on damp MOX powders. The WPNCS was ultimately asked by the NEA Nuclear Science Committee (NSC) to provide the framework for the selection and performance of new experiments that fill the identified need. A set of criteria was established to enable uniform comparison of experimental proposals with generic MOX application data. Criteria were established for five general characteristics: (1) neutronic parameters, (2) type of experiments, (3) financial aspects, (4) schedule, and (5) other considerations. Proposals were judged most importantly on their ability to match the neutronic parameters of predetermined MOX applications. The neutronic parameters that formed the basis for comparison included *core average values (not local values)* for flux, fission and capture rate; detailed balance data (fission and capture) for the main isotopes (Actinides, H and O); sensitivity coefficients to important nuclear reactions (fission, capture, elastic and inelastic scatter, nu-bar, mu-bar) for all uranium and plutonium isotopes, hydrogen, and oxygen; sensitivity profiles to the main nuclear reactions for uranium and plutonium isotopes; energy of average lethargy causing fission; and the average fission group energy. The focus of this paper is on the definition of the need; the neutronics criteria established to assess which, if any, of three proposed MOX experimental programs best meet the need; and the actual assessment of the proposed experimental programs.

*Key Words*: Critical-Experiment, Damp-Powder, International-Collaboration, MOX

# 1 INTRODUCTION

The OECD Nuclear Energy Agency (NEA) Working Party on Nuclear Criticality Safety (WPNCS) has been discussing the need for additional integral benchmark experiments for several years. An Expert Group on Experimental Needs was formed in 1997 to investigate international experimental needs. This group identified the MOX fuel manufacturing process as an area in which there is a specific need for additional integral benchmark data. This step in the MOX fuel cycle is one in which reliance solely on geometric favorable equipment is not possible. Controls on fissile material mass and hydrogen content are necessary in the main units of the plant.

In this context, critical experiments with low-moderated MOX fissile media that, at least in part, fill the identified need were proposed. However, the participants in the Expert Group were not in a position to make funding commitments to support the establishment of a new experimental program. At the 2003 Working Party meeting, the WPNCS concluded there was little more they could do without the support and direction of the NEA Nuclear Science Committee (NSC). After careful consideration, the WPNCS formally issued a recommendation to the Nuclear Science Committee to establish an international consortium among member countries to support the completion of the needed experiments.

In response to the WPNCS recommendation, the NSC authorized the organization of a Workshop to:

(1) Validate the need for additional experiments with low-moderated MOX fuel,
(2) Identify relevant experimental programs that have the potential to fulfill the need,
(3) Evaluate which experimental program or combination of programs best fill the need, and
(4) Evaluate the prospects for successful organization of an international co-operative program to complete the needed experiments.

The workshop was held in April of 2004 at which time the need for the experiments was validated, five experimental programs were proposed, and additional existing data were identified. However, due to the lack of uniform presentation and, in some cases sufficient detail, it was not possible to reach a consensus on which program or combination of programs would best address the need.

Based on the outcome of the Workshop, the NSC requested the WPNCS to provide the framework for the selection and performance of new experiments. Within this framework, criteria were established to enable uniform comparison of experimental proposals with generic MOX application data and an assessment team was formed to review and assess any MOX experimental proposals.

# 2 SELECTION CRITERIA

A set of criteria were established to enable uniform comparison of experimental proposals with generic MOX application data. Criteria were established for five general characteristics: (1) neutronic parameters, (2) type of experiments, (3) financial, (4) schedule, and (5) other considerations. Proposals were judged, most importantly, on their ability to match a set of neutronic parameters that correspond to predetermined generic MOX applications. The set of parameters for the MOX ***Application Configurations*** were generated independently for the WPNCS by scientists from the Institute of Physics and Power Engineering (IPPE) and were provided to the organizations that submitted proposals to the MOX workshop. A five-group energy structure was considered:

| | |
|---|---|
| Group 1 | E > 100 keV |
| Group 2 | 10 keV < E < 100 keV |
| Group 3 | 10 eV < E < 10 keV |
| Group 4 | 0.1 eV < E < 10 eV |
| Group 5 | E < 0.1 eV |

The following *core average values (not local values)* were compared:

- 5-group flux, fission and capture rate (in percent),
- 5-group detailed balance (fission and capture in %) for the main isotopes (Actinides, H and O),
- 5-group Sensitivity coefficients to the main nuclear reactions for all uranium and plutonium isotopes, Hydrogen, and Oxygen: Fission, Capture, Elastic, Inelastic, Nu-bar, and Mu-bar
- Sensitivity profiles (238-group) to the main nuclear reactions for main uranium and plutonium isotopes, and $C_k$ values obtained from the ORNL TSUNAMI code system [2].
- EALF: Energy of average lethargy causing fission,
- AFGE: Average Fission Group Energy.

## 3 GENERIC APPLICATIONS

Rather than specifying actual configurations encountered in the MOX fabrication plants, simplified geometries were considered and the range of important parameters was defined (moderation ratio, plutonium content, and origin of the plutonium – weapon grade or reactor grade). Specifications for generic ***Application Configurations*** were established as a basis for comparing experimental proposals and assessing the ability of each proposal to provide meaningful data for the MOX fuel manufacturing process. Two types of generic ***Application Configurations*** were established – MOX powders with Reactor Grade Plutonium and MOX powders with Weapon Grade Plutonium. For each of these application types, the $^{240}$Pu, $^{241}$Pu, $^{235}$U content and the MOX powder density were fixed and the total $PuO_2$ and water contents were varied. Specifications for the two types of applications are given in Table I.

**Table I. Compositions for the generic MOX a*pplication configurations***

| Reactor Grade Applications | Weapon Grade Applications |
|---|---|
| Pu-240 = 20% | Pu-240 = 4% |
| Pu-241 = 12% | Pu-241 = 0% |
| U-235 = 0.7% | U-235 = 0.7% |
| Density = 5.0 g/cc | Density = 5.0 g/cc |
| $PuO_2$ Content: 10%, 20%, and 30% | $PuO_2$ Content: 10%, 20%, and 30% |
| Water Content: 0%, 1%, 3%, and 5% | Water Content: 0%, 1%, 3%, and 5% |

Twenty-four different compositions were derived from two different plutonium compositions (reactor or weapon grade) with three $PuO_2$ contents (10%, 20%, and 30%) and four different water contents (0%, 1%, 3%, and 5%). Applications were represented as simple homogeneous mixtures of MOX material in spherical geometry. Both water-reflected (20 cm thickness) and un-reflected systems were evaluated, making a total of 48 ***Application Configurations***.

## 4 EXPERIMENTAL PROPOSALS

Of the five proposals presented at the MOX Workshop in April 2004, the requested data were provided in time for full evaluation by only two institutes – the Institute of Physics and Power Engineering (IPPE) in the Russian Federation and the Commissariat à l'Energie Atomique (CEA) / Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in France. A third proposal was submitted by Studie Centrum voor Kernenergie (SCK) /Centre d'Etude de l'Energie Nucléaire (CEN) in Belgium after the evaluation process had begun. A short description of each proposal follows.

### 4.1 IPPE Proposal

The IPPE proposal consists of nine configurations that would be assembled on the BFS-1 Facility at IPPE. Damp MOX powders are simulated by assembling tightly packed cylindrical aluminum tubes containing various ratios of plutonium and depleted uranium dioxide fuel pellets. The specified water content is achieved by periodically including polyethylene pellets. The composition of the fuel material is more representative of weapon grade MOX. Several spectral indices would also be measured. The IPPE proposed experimental program can be completed in a relatively short time frame (~1.5 years) at a relatively low cost (~$0.6M).

**Table II. Summary of the proposed IPPE experiments**

| Experiment | Designator | Pu-240 Content (%) | Pu Content (%) | $H_2O$ Content (%) |
|---|---|---|---|---|
| **BFS-97-1** | **W22 – 0%** | 4.5 | 22.5 | 0 |
| **BFS-97-2** | **W22 – 0.6%** | 4.5 | 22.5 | 0.6 |
| **BFS-97-3** | **W22 – 2.5%** | 4.5 | 22.5 | 2.5 |
| **BFS-97-4** | **W22 – 3.4%** | 4.5 | 22.5 | 3.4 |
| **BFS-99-1** | **R22 H2O – 2.5%** | 10 | 22.5 | 2.5 |
| **BFS-99-2** | **R22 H20 – 3.4%** | 10 | 22.5 | 3.4 |
| **BFS-101-1** | **W10 – 0.7%** | 4.5 | 9.6 | 0.7 |
| **BFS-101-2** | **W10 – 1.3%** | 4.5 | 9.6 | 1.3 |
| **BFS-101-3** | **W10 – 2.1%** | 4.5 | 9.6 | 2.1 |

## 4.2 CEA/IRSN Proposal

The CEA/IRSN proposal consists of five configurations that would be assembled on the Apparatus B Facility at Valduc Criticality Laboratory and in the EOLE Zero Power Reactor at Cadarache. Damp MOX powders are simulated with assemblies of low-moderated MOX fuel rods. The composition of the fuel material is mainly representative of reactor grade MOX. The combination of the two facilities allows the determination of various parameters (critical mass, material buckling, spectral indices and reactivity temperature coefficient from 5°C up to 80°C). Fuel manufacture has the major impact to the cost and schedule for the CEA/IRSN proposed experimental program. The time required to manufacture the fuel and complete the experiments is approximately 4 years and the total cost could exceed $14M.

**Table III. Summary of the proposed CEA/IRSN experiments**

| Pitch | Designator | Pu Content (%) | Pu-240 Content (%) | $H_2O$ Content (%) |
|---|---|---|---|---|
| **0.96** | **MOX096** | 27.5 | 25.08 | ~2.4 |
| **0.98** | **MOX098** | 27.5 | 25.08 | ~3.2 |
| **1.00** | **MOX100** | 27.5 | 25.08 | ~3.9 |
| **1.02** | **MOX102** | 27.5 | 25.08 | ~4.7 |
| **1.04** | **MOX104** | 27.5 | 25.08 | ~5.4 |

## 4.3 SCK•CEN Proposal

Details of the SCK•CEN proposal were not made available until late in the evaluation process. The SCK•CEN proposal consists of two different approaches using tightly packed MOX fuel rods to simulate damp MOX powders. The first approach includes two main configurations of tightly packed MOX fuel rods. The second approach includes one main configuration with a tightly packed test zone of MOX fuel rods surrounded by driver zones. The composition of the fuel material used in both approaches is mainly representative of reactor grade MOX. In addition to criticality, the reactivity coefficient $\delta\rho/\delta h$ (all MOX); axial and horizontal fission rate distributions; infinite neutron multiplication factor, $k_\infty$; spectral indices; mean neutron generation time and effective delayed neutron fraction (all MOX cores only) will also be measured. All of these measurements can be carried out on the VENUS critical facility. The time required to manufacture the fuel and complete the experiments is approximately 3 years. Cost information was incomplete.

# 5 ASSESSMENT OF EXPERIMENTAL PROPOSALS

An assessment team was assembled to review the three experimental proposals. A list of team members and their affiliation is given below.

| | |
|---|---|
| B. Briggs | INEEL, USA |
| I. Duhamel | IRSN, France |
| J. Gulliford | BNFL, UK |
| C. Hopper | ORNL, USA |
| R. McKnight | ANL, USA |
| T. McLaughlin | LANL, USA |
| A. Nouri | IRSN, France |
| B. Ponsard | SCK•CEN, Belgium |
| C. Venard | CEA, France |
| T. Yamamoto | JAERI, Japan |

The experiments were assessed according to the five previously established criteria namely, neutronic criteria, the type of experiments, financial aspects, anticipated schedule, and other aspects. Detailed sensitivity / uncertainty analyses were performed on the IPPE and CEA/IRSN proposals by scientists at Oak Ridge National Laboratory (ORNL). The SCK•CEN proposal was not provided in time to allow this detailed assessment.

### 5.1 Comparison of the Neutronic Criteria

Technically, the most significant criterion is the ability of the proposed experimental proposals to match the neutronic parameters of the ***Application Configurations***. The MOX application configurations are relatively large homogeneous systems of damp MOX powder. In order to conclude that an experiment meets the stated need, the neutronic parameters of the experiment must adequately match the neutronic parameters of the application. Core average values, not local values, were compared in order to appropriately assess how well the experiments represent the applications. Each of the parameters noted in Section 2 were compared and are discussed in the following subsections.

#### 5.1.1 Five-group flux, fission rate and capture rate

An attempt was made to quantify the similarity between these spectra by supposing that the spectrum (for fission or capture) in a given application is characterized by ($A_1$, $A_2$, $A_3$, $A_4$, $A_5$) where the subscripts 1,…5 represent the energy group number. Similarly, the spectrum in a given experiment is characterized by ($E_1$, $E_2$, $E_3$, $E_4$, $E_5$). Both spectra are normalised to 100.

A distance between a pair of application and experiment spectra was defined as:

$$D^2 = \sum_{i=1}^{5} \left(A_i - E_i\right)^2 \qquad (1)$$

Obviously, the smaller the value of $D^2$ the closer the spectrum of the application is to the spectrum of the experiment. This quantity does not have a physical meaning but it enables a reviewer to judge how close the experiments are to a given application. It also enables a reviewer to rate experiments according to their applicability. Figures 1 - 4 display the distance between pairs, (experiment, application), for capture and fission spectra. Unreflected and water-reflected applications are plotted on different graphs. The applications are represented on the xaxis and labelled using the main characteristics (e.g. "Reactor-Pu – 1% water" and "WG – 1% water", WG standing for Weapon Grade). The y-axis gives the distance from a given application, as defined above, for each experiment. Experiments are also labelled with abbreviations (e.g. "W22 H20 – 0.6% (Ru Exp)" for a Russian proposed experiment with Weapon Grade plutonium containing 22% Pu and 0.6% water). French experiments (labelled with the extension "Fr Exp") are all made with Reactor Grade Plutonium with a 27.5% Pu content. French experiments are plotted using a black line and Russian experiments with a red line. Only graphs for 20% Pu are provided. However, all graphs may be found in Reference 1.

Several interesting observations can be made:

- The graphs show that the spectra (both capture and fission) in weapon-grade and reactor-grade-based configurations are extremely similar, at least as far as the present comparison using the broad 5-group structure allows.

- The graphs also show that the reflection conditions (bare or water reflection) have an important effect on the spectrum. While the comparisons show clearly that the "W22-H20 0% (Ru Exp)" experiment is the most representative to the application case with 0% water content in the powder, the situation is not so clear for water reflected cases. Even for configurations with water mixed with the powder, the effect of reflection is important. If one considers the 3 closest experiments to each configuration, it appears that the French experiments are generally selected for the water reflected experiments while the Russian experiments are selected for the bare configurations.

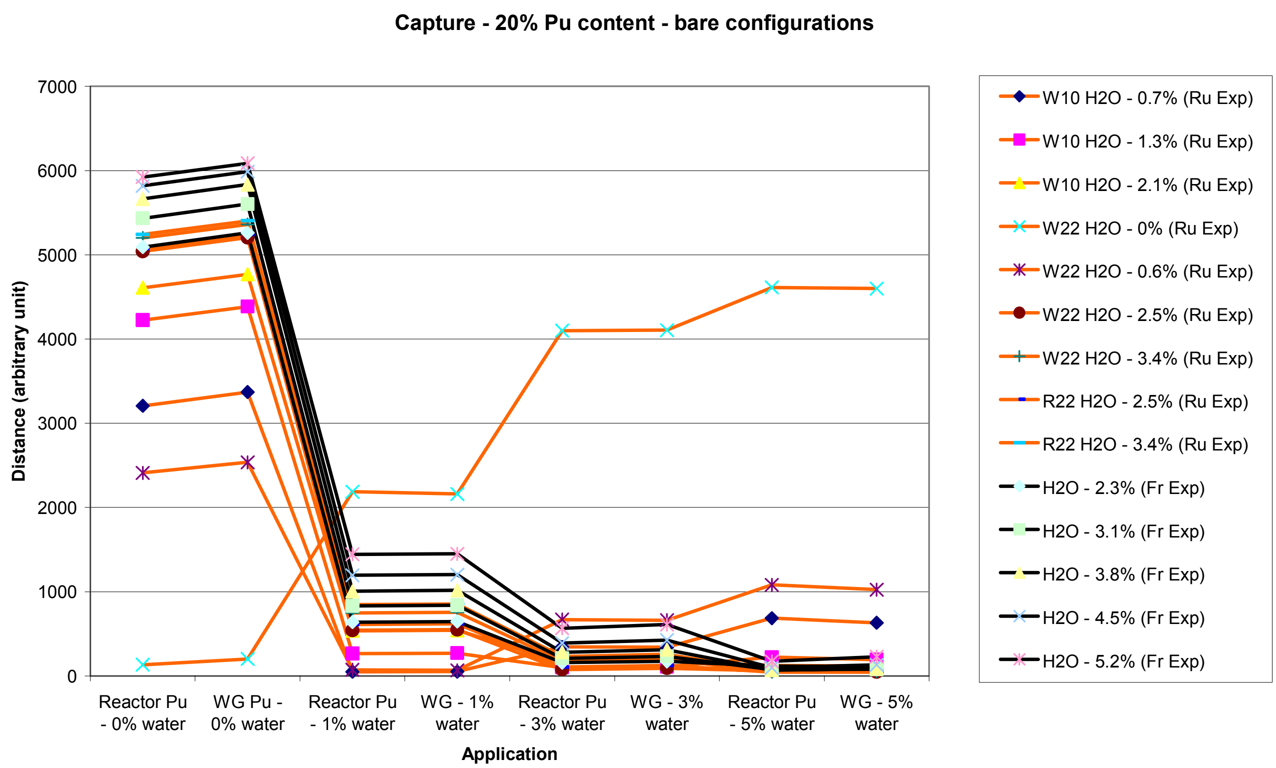


**Figure 1. Comparison of experiments with applications (capture – 20% Pu – un-reflected configurations)**

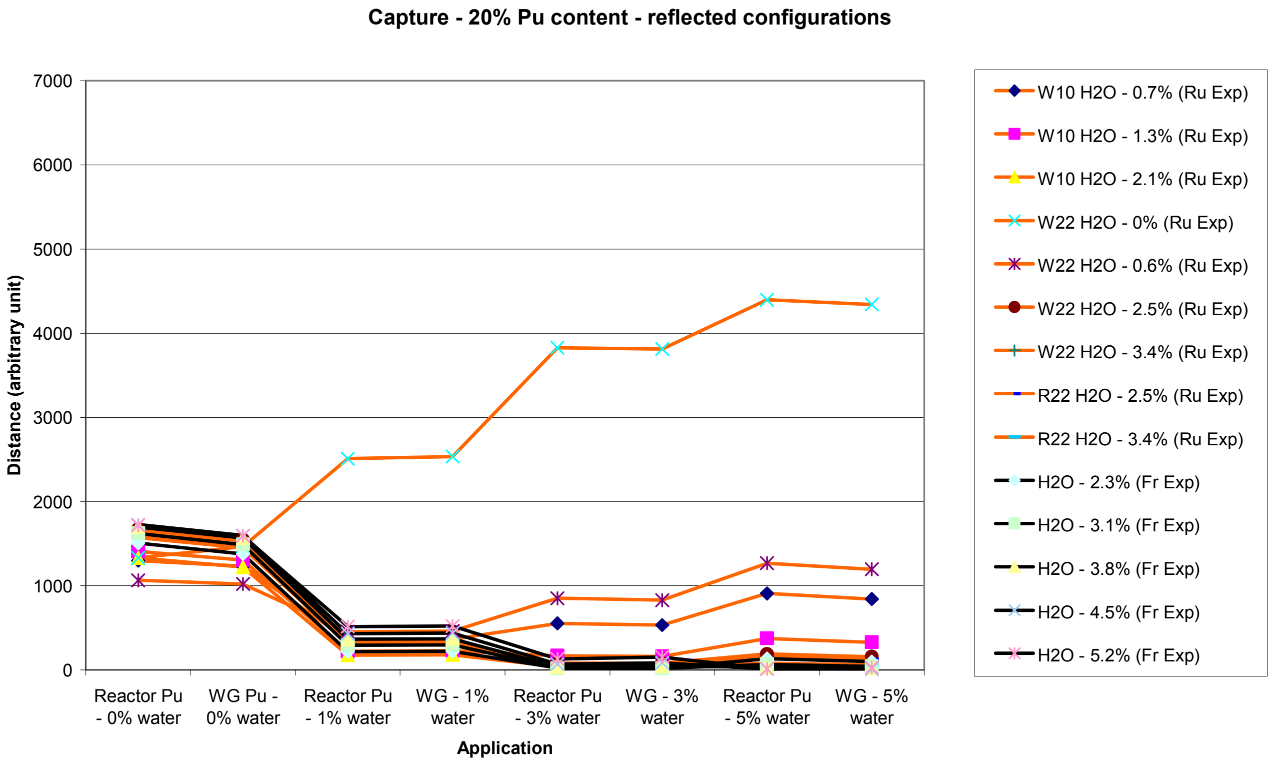


**Figure 2. Comparison of experiments with applications (capture – 20% Pu –reflected configurations)**

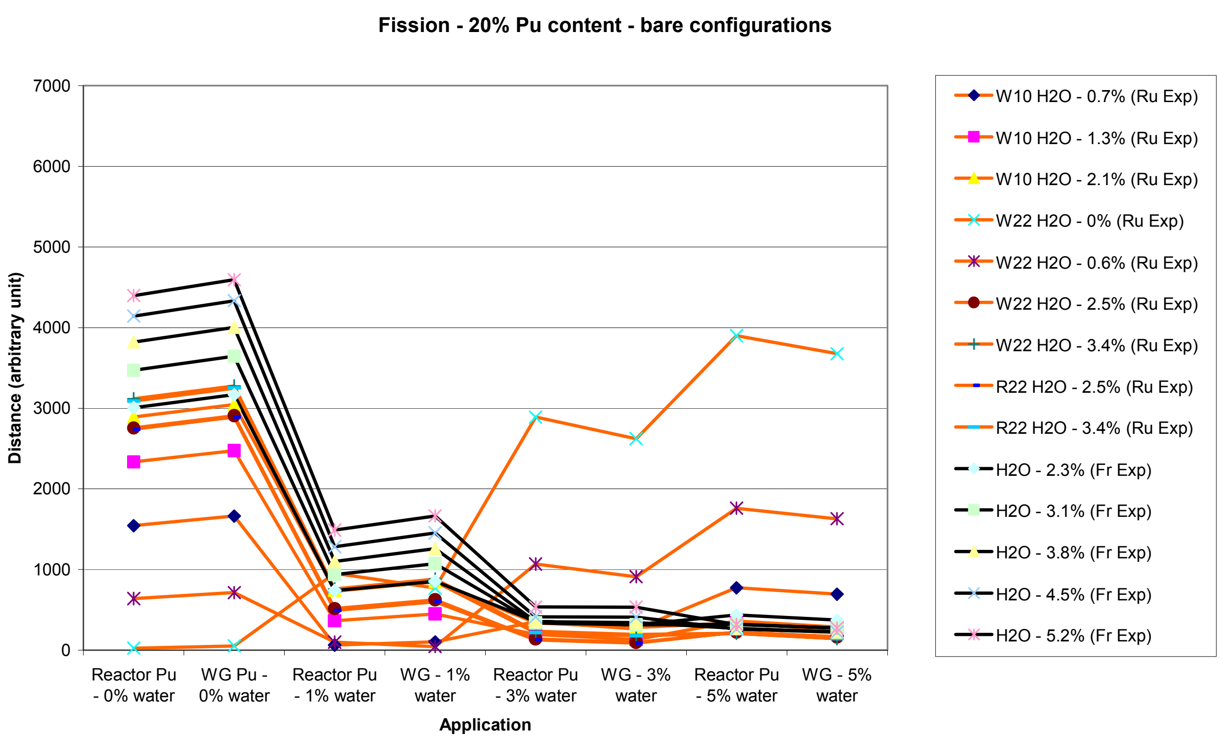


**Figure 3. Comparison of experiments with applications (fission – 20% Pu – un-reflected configurations)**

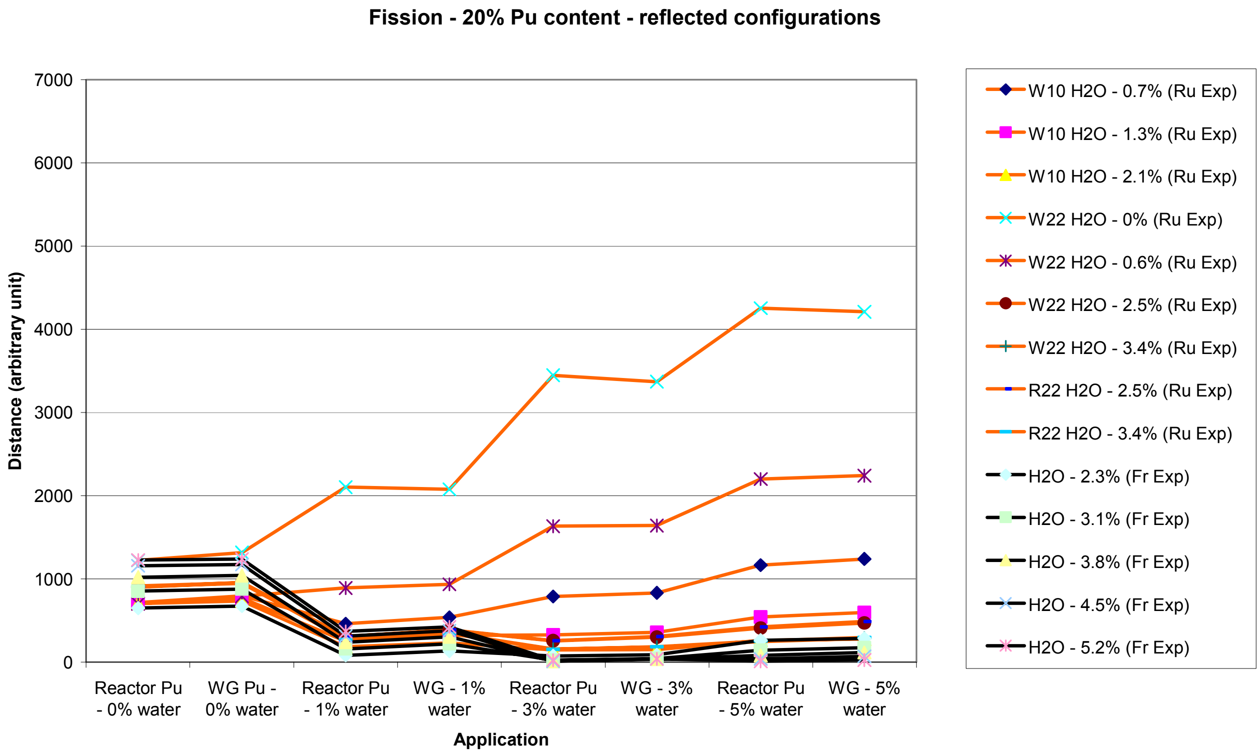


**Figure 4. Comparison of experiments with applications (fission – 20% Pu –reflected configurations)**

### 5.1.2 Five-group detailed balance data

Five-group detailed balance data (fission and capture in %) for the main isotopes (Actinides, H and O) for the proposed IPPE and the CEA/IRSN experimental program were reviewed and compared. For all cases, the major contribution to the neutron balance comes from $^{239}$Pu and $^{238}$U. For configurations with weapon-grade plutonium, these two isotopes are responsible for about 90% of the absorption in the core. For configurations with reactor-grade plutonium; however, about 20% of the absorption occurs in Pu-240 and Pu-241.

In general, the contribution to capture reaction rates increases with the moderation ratio while the fission contribution decreases with the moderation ratio. This is due to a spectrum shift. Analysis of the change of the energy dependence of Pu-239 fission with moderation ratio reveals that fission at thermal energies does not change significantly. When water is added to the system, the major change concerns the shift of fission from high energy (E>100 keV) to intermediate energy (10 eV < E < 10 keV).

### 5.1.3 Sensitivity coefficients to the main nuclear reactions

Sensitivity coefficients (238-group) to the main nuclear reactions (fission, capture, elastic, inelastic, nu-bar, mu-bar) for nine configurations from the IPPE experimental program and one of the five configurations from the CEA/IRSN experimental program were calculated by Calvin M. Hopper, Bradley T. Rearden, and Karla Elam of Oak Ridge National Laboratory (ORNL). Correlation Coefficients between the proposed IPPE and CEA /IRSN experimental configurations and the MOX ***Application Configurations*** were also calculated. Details of this work are given in Reference 2. The conclusions based on the calculated correlation coefficients are similar to those based on analysis of the flux, fission, and capture rates and on a detailed neutron balance. Correlation Coefficients between the proposed IPPE and CEA /IRSN experimental configurations and the MOX ***Application Configurations*** are shown graphically in Figures 5 through 8.

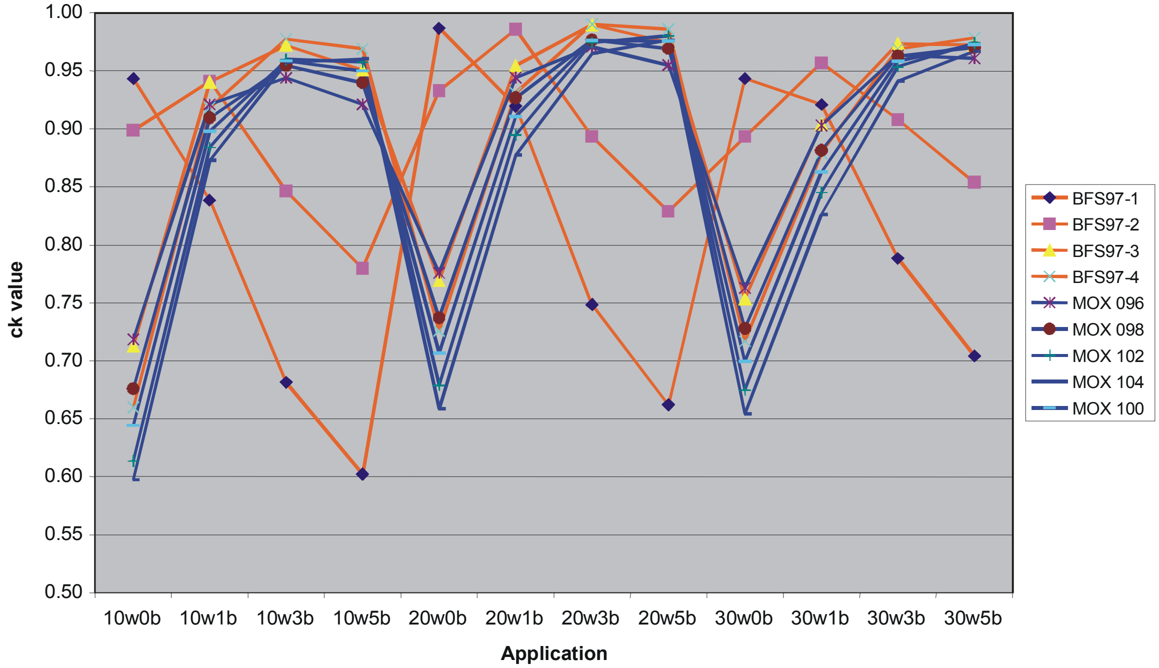


**Figure 5. Comparison of $c_k$ values with un-reflected weapon-grade *application configurations***

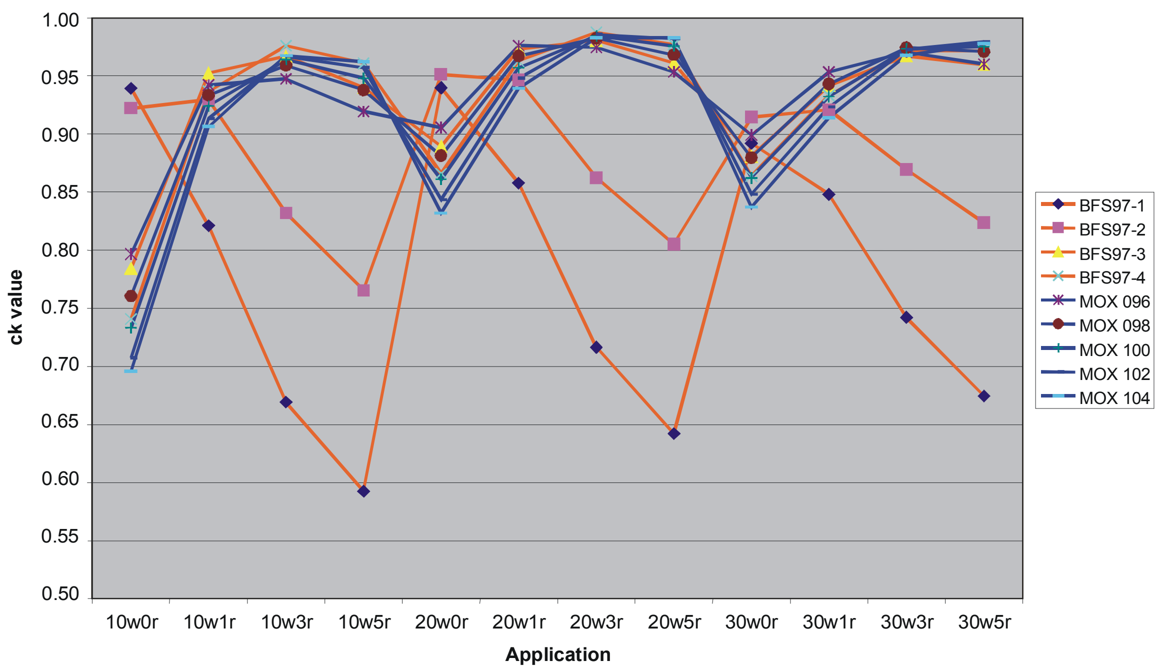


**Figure 6. Comparison of $c_k$ values with reflected weapon-grade a*pplication configurations***

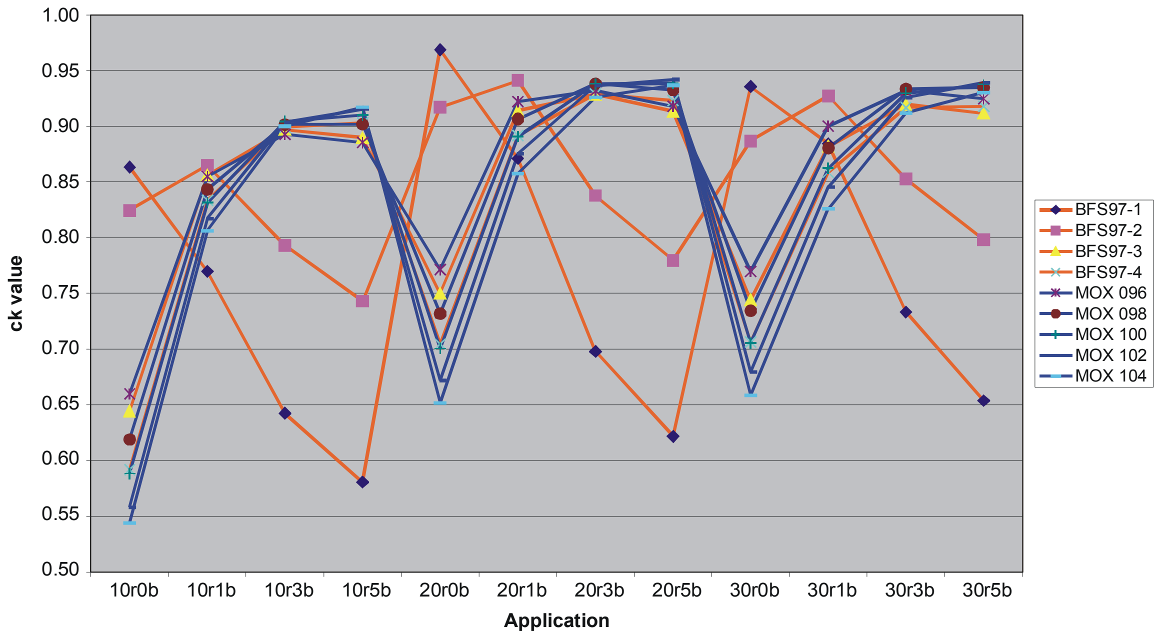


**Figure 7. Comparison of $c_k$ values with un-reflected reactor-grade *application configurations***


**Figure 8. Comparison of $c_k$ values with reflected reactor-grade *application configurations***

### 5.1.4 Energy of average lethargy causing fission (EALF) and the average fission group energy (AFGE)

Comparisons of the integral parameters, EALF and Average Fission Group Energy (AFGE), for the IPPE and CEA/IRSN experiments, for both water-reflected and un-reflected applications were made and the results are summarized in Figure 9. The conclusions drawn from these data are essentially the same as from comparison of flux, fission, and capture rate data and comparison of correlation coefficient data. Specifically, differences between weapon grade and reactor grade-based configurations are similar and the configurations are very sensitive the reflector condition. IPPE data are more representative of the un-reflected ***Application Configurations*** and the CEA/IRSN data are more representative of the water-reflected ***Application Configurations***. Also evident in Figure 9 is the complementary nature of the two series of experiments. The IPPE experiments are most applicable to the lower degrees of moderation while the CEA/IRSN experiments are most applicable to the higher degrees of moderation.

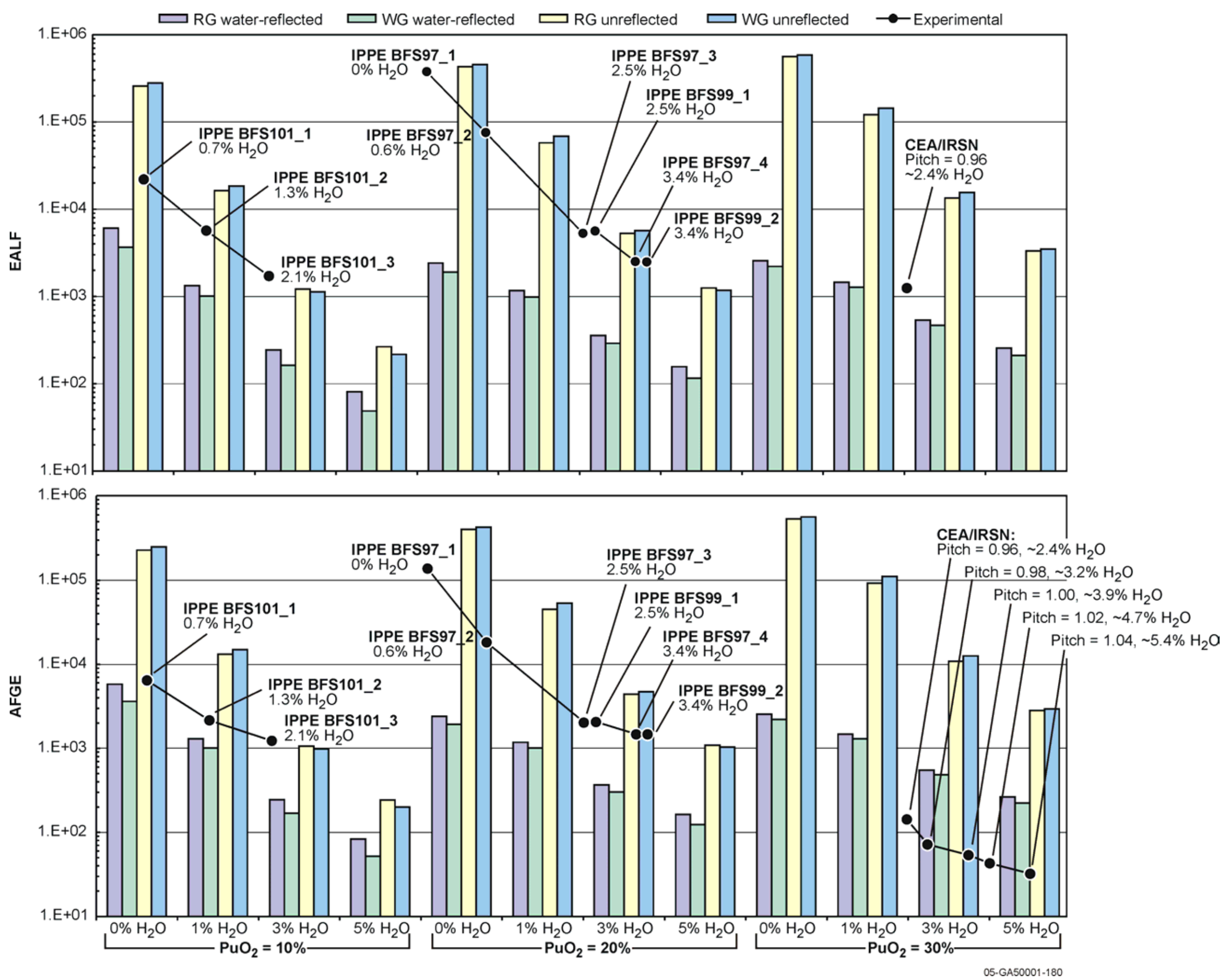


**Figure 9. Comparison of AFGE and EALF values for all *application configurations* with values for the IPPE and CEA/IRSN experiments**

### 5.1.5 Summary

Experimental proposals were evaluated, primarily, using four types of neutronics data or methods: reaction rate data (flux, fission rate, capture rate), ORNL sensitivity analysis (correlation coefficients), integral parameters (EALF and AFGE), and neutron balance data. Conclusions based on comparison of five-group reaction rate data, ORNL sensitivity analysis, and the integral parameters are essentially the same: weapon grade and reactor grade-based configurations are very similar, results are very sensitive the reflector condition, IPPE data are more representative of the un-reflected ***Application Configurations,*** and the CEA/IRSN data are more representative of the water-reflected ***Application Configurations***. Comparison of the neutron balance data indicates that, the major contribution to the neutron balance comes from $^{239}$Pu and $^{238}$U in all cases. These two isotopes are responsible for about 90% of the absorption in the core for configurations with weapon-grade plutonium. However, about 20% of the absorption occurs in Pu-240 and Pu-241 for configurations with reactor-grade plutonium. The complementary nature of the two series of experiments is evident in all the data. The IPPE experiments are most applicable to the lower degrees of moderation while the CEA/IRSN experiments are most applicable to the higher degrees of moderation.

## 6 CONCLUSIONS

Proposals for three different experimental programs were submitted; namely, the Institute of Physics and Power Engineering (IPPE) in the Russian Federation, the Commissariat à l'Energie Atomique (CEA) / Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in France, and the Studie Centrum voor Kernenergie / Centre d'Etude de l'Energie Nucléaire (SCK•CEN) in Belgium. Of the three proposals, only the proposal from IPPE and the proposal from CEA/IRSN were complete and submitted in time for full assessment.

Conclusions are based on the comparisons of spectral, balance, and sensitivity data. In general, the IPPE program provides the best coverage for low water concentrations, while the CEA/IRSN program provides the best coverage for higher water concentrations and higher $^{240}$Pu content. Except for higher water concentrations, the IPPE program provides better coverage for applications containing weapon grade plutonium. Except for lower water concentrations, the CEA /IRSN program provides better coverage for applications containing reactor grade plutonium. There is overlap between the two programs and if both are completed, there would be the opportunity to identify and assess systematic uncertainties.

The SCK•CEN proposal included two options. The data for the first option resembles, in large part, the CEA/IRSN proposal. These experiments would therefore offer the same degree of coverage as the CEA/IRSN proposal. The second option was comprised of a MOX test zone surrounded by a driver core. However, the SCK•CEN proposal was not finished and submitted in time for a complete assessment, including the detailed sensitivity analysis at ORNL. There were concerns shared by several members of the assessment team that the proposal with the driver core would not adequately represent the MOX configurations. While some of the spectral data compare favorably with the ***Application Configurations***, it was not demonstrated in the proposal how the experiments, which are dominated by low enriched fuel rods could adequately represent the MOX applications. It was stated in the proposal that approximately 15.6% of the total fissions and 21.3% of the total captures occur in the test zone, which tends to confirm the concern that the experiments do not adequately represent the ***Application Configurations***.

With one exception, the opinion of the assessment team was that the proposed IPPE and CEA/IRSN experimental programs are complementary and together fill the identified need. The recommendation given to the Nuclear Science Committee was that the IPPE proposal should be pursued independently of the CEA/IRSN experiments. This was due to the low cost of the proposal and the significantly different funding alternative (ISTC) that is not available to the other experimental programs. Since both programs are needed to fill the need, the CEA/IRSN experimental program should be the primary focus of the Nuclear Science Committee. Financial commitments could not be made by the members of the assessment team, but definite interest was expressed by representatives from France, Japan, Russian Federation, the United Kingdom, and the United States.

Since the assessment was completed, DOE and IRSN have agreed to collaborate and fund the IPPE experiments through the International Science and Technology Center. Planning for the CEA/IRSN experiments have essentially ceased because financial resources are presently not available.

## 7 ACKNOWLEDGMENTS

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