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IRSN Projects for Critical Experiments "Low Moderated MOX Fuel Project" and Others

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This document gives an overview of the criticality experimental programs of interest for IRSN[1]. These experiments, which could be carried out after June 2004 on the "Apparatus B" CEA[2] facility concern:

- the qualification of the effect of plutonium decrease to americium and validation of the associated neutronics data,
- the qualification of structural material cross-sections,
- the determination of the temperature effect of diluted plutonium solutions,
- the low moderated MOX fuel qualification.

The major part of the present paper is devoted to the presentation of an IRSN proposal for an international collaboration on such an experimental program for MOX fuel qualification.

1. Foreword

In order to control the criticality hazard in laboratories, plants or transports where nuclear material in sufficient quantities are present, qualified calculation codes are needed, both for design of these facilities and for safety assessment purpose. Therefore, reference experiments are required, particularly since the nuclear industry is seeking to improve the economy of these facilities, while reducing the constraints linked to the criticality hazard.

In the framework of its mission on the assessment of the criticality risk, the IRSN conducts studies focused not only on preventing the risk of criticality, but also on assessing the consequences of a criticality accident. A lot of French experimental programs have been conducted in the past in the CEA Criticality Laboratory at the Nuclear Research Center of Valduc [1)]. A team of the IRSN "Criticality Studies Division" is in charge of the definition of the experimental programs to qualify computer calculation codes.

2. Criticality programs planned in the medium term

Three programs are planned in Valduc criticality facility Apparatus B with various objectives. This apparatus is presented in appendix.

2.1 Qualification of the decrease from ^{241}Pu to ^{241}Am

The period of ^{241}Pu is about 14.4 years and rods used for the "HTC" experimental program [2)], carried out from 1986 to 1991 in Valduc criticality facility Apparatus B, are more than 16 years old. The rods are available to plane the same experiments as the "HTC" ones, in order to evaluate the effect of radioactive decrease on critical water height. Preliminary calculations show that the effect should be not far from 3% on k effective. Half is due to decrease of ^{241}Pu and half to ^{241}Am absorption capacity.

COGEMA is a partner for this program planned in 2004.

2.2 Qualification of structural materials cross-sections

Reports presently prepared to demonstrate sub-criticality of nuclear facilities and transport packages exhibit a new trend. This trend is to perform calculations on configurations as close as possible to actual ones with models including the exact materials surrounding the fissile medium. This development is part of an optimization process: calculations are performed on bounded geometric models leading to less conservatism and taking also into account the surrounding material absorption ability which is far from being negligible. This smarter calculation approach may be a decisive step to demonstrate facilities sub-criticality with reasonable margins when studying current assemblies made of more enriched fissile media and resulting of optimized design.

Moreover, the improved criticality calculation tools and their associated computer programs are facilitating this process. The computer time needed to

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solve a problem simulated by complex geometry models is significantly reduced. The graphic computer interfaces evolve to include specific criticality know-how and user needs to solve problems of complex configurations, even with a great number of parameters. These new possibilities make easier the computing work with a minimum of preparation effort. Such last generation tools are so user friendly that they can be used by a larger panel of engineers beyond the traditional criticality expert skills.

This evolution puts a new emphasis on the qualification process associated with these improved tools. Previously, the qualification studies were mainly oriented towards the understanding of the behavior of fissile materials and other ones considered either as major neutrons absorbers or as neutrons reflectors. Now, the qualification required for improved criticality assessment methodologies, as previously described, involves characterization of an enlarged panel of materials to cover the ones used in facilities for structural purposes and not for any secondary criticality benefit. Indeed, such structural materials surrounding the fissile medium modify interactions between fissile media for a wide range of the neutronic spectrum.

2.2.1 Structural materials of interest

A large screening of studies, where structural materials had a significant impact when being considered to demonstrate facility sub-criticality, leads to select a first list of materials of interest:

- Copper (> 5 cm)
- Aluminum (> 10 cm)
- Steel, Stainless steel, Iron, Cast iron (few mms to few cms)
- Concrete (> 20 cm)
- PVC (< 1 cm) with better moderation than water
- Fire extinguishing powder with low hydrogenated material content (< 5 cm).

2.2.2 Summary of benchmarks available in ICSBEP[3]

A survey of experimental data available for the above selected materials, mainly collected by the ICSBEP working group, confirmed that these data do not cover the present needs because:

- available benchmarks concern either specific material reflection properties or absorption properties but not both. Moreover, in the collected data, the effect on the reactivity worth of considered materials is low;
- in experiments where the considered structural materials are installed between fissile media in interaction, their reactivity level is low.

2.2.3 Nuclear data assessment

Firstly, a preliminary assessment of the available nuclear data has been done based on a review of the work performed by OECD to summarize the validation of JEF2.2 data bank [3)].

Secondly, the cross-sections of various materials as given by JEF2.2 were compared with the values obtained while using CRISTAL standard route library. This comparison leads to highlight the nuclides of importance for neutron scattering and absorption for each material (Fe, Mn, Na, Cl) and the energy range of importance for qualification purpose.

2.2.4 Feasibility study and selection of critical configurations to be tested in Apparatus B

A feasibility screening study was done in order to select the critical configurations to be tested in Apparatus B. The proposed configurations for this feasibility study have the following general specifications:

- simple design, for installation in the "HTC" experimental program tank or equivalent[2)],
- use of rods available in Valduc laboratory (UO_2 enriched to 4.78% and/or UO_2-PuO_2 with a PuO_2 content of 1%,
- 80 cm of water height target for critical state,
- thickness of the selected structural materials as per criteria specified in above paragraph 2.2.1,
- development of an intermediate spectrum in the tested material (more than 50% of the neutrons with energy level greater than 1 keV).

The result of this screening work led to propose two main configurations families for further testing :

- structural materials can be spred out in rods lattice arrays when considering low thickness arrangements (1 cm maximum) in order to test material absorption ability (stainless steel, PVC),
- structural materials installed as a screen between two half cores when considering high thickness arrangement (a few cm up to 20 cm) in order to test material (aluminum, copper, concrete, fire extinguishing powder) interaction ability. An intermediate spectrum in concrete could be envisaged if some rods are installed inside the central concrete structural device.

2.3 Measurement of the temperature coefficient of diluted plutonium solutions

Calculations made by several countries (US, Japan, UK, France) have highlighted a possible positive reactivity temperature coefficient in the case of low concentration (<30 $g.l^{-1}$) plutonium solutions. Considering the importance of this effect, especially in case of criticality excursion, IRSN has designed a core to be installed in the "Apparatus B" to perform, initially in 1994, sub-critical experiments with Pu concentrations of about 14 to 20 $g.l^{-1}$ at various temperatures [4)]. This program has been postponed and, because of the evolution of the isotopic composition

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[3] International Criticality Safety Benchmark Experiments Program

of the plutonium during 11 years, new calculations were performed with CRISTAL [5].

At the present day, the calculations demonstrate that:

- the solutions of plutonium are less reactive and, because of the maximum dimensions of the core available, only concentrations above 14.4 $g.l^{-1}$ should be used eventually with a low acidity;
- the temperature effect may be increase with the proportion of ^{238}U in the solution. In order to confirm calculations, some measurements of density for mixed uranium and plutonium solutions must be done to define a precise correlation between concentrations, density and temperature.

Considering these first results, the program is on standby until additional studies will demonstrate the interest of this phenomena with mixed uranium and plutonium solutions for accident consequences. Therefore, the initial design of the core could be modified if this program is confirmed.

3. Programs to be scheduled and defined – Qualification of criticality codes for low moderated "MOX" fissile media

3.1 The needs

The use of MOX fuel in nuclear power plants, to convert surplus Reactor-Grade (~ 20 % ^{240}Pu) or Weapons-Grade (~ 4 % ^{240}Pu) plutonium, has created a need for neutron physics benchmarks that deal with MOX fuel configurations. The assessment of the available results on MOX fuel shows that there is a lack of experimental results for low moderated MOX fuel. For example, in MOX fabrication plants, the postulated possible contents of hydrogenated materials are limited to a low value, even for the postulated accidental conditions. For these fissile media, the neutron spectrum varies between the fast and the intermediate energy range (10 MeV to 1 keV). Up to now, very few experiments have been performed in this energy range. Moreover, the experimental data are not well known. This lack of qualifying experiments for low moderated fissile material has been emphasized by the expert group on experimental needs of the OECD Working Party on Nuclear Criticality Safety (WPNCS).

3.2 The proposed experiments

The most representative experiments corresponding to the fabrication process would be experiments with MOX powders with different moisture content. But the use of plutonium powder causes three main difficulties: maintain a definite temperature, guarantee a safe containment of the plutonium and assure the accuracy of the moiture content. Besides a large mass of MOX powder is needed. For this reason, the proposed experiments involve low water moderated MOX fuel rods arrays dealing with various lattices pitches in order to cover different moderation ratios. These experiments would be carried out at C.E.A/Valduc in the criticality facility Apparatus B without special difficulties. The preliminary design has been optimized taking into account the feasibility of the experiment, the possibility of using the existing equipment, the cost of fabrication of the fuel and the representativity of the experiment in terms of neutron spectrum.

The fuel rods are composed of MOX pellets containing about 27.5 % (wt) of PuO_2. This plutonium content corresponds to the primary blend mixture in French fabrication process. The MOX fuel pellets are obtained from Reactor-Grade plutonium. In order to reduce the number of rods needed and the fabrication cost, "BWR type" diameters currently available could be chosen. Different small pitches (square and triangular) can be considered to cover different moderation ratios close to powder or fuel assemblies' ratios.

3.2 Neutronics characteristics

In the ICSBEP characterization, these experiments could be classified as intermediate spectra systems. In fact, as can be seen in the following table giving the results of preliminary calculations, less than 50% of the fissions occur in any one of the three energy ranges (fast, intermediate and thermal).

Table 1 Neutronics characteristics

(%)	Energy range		
	Fast (>100 KeV)	Intermediate (0.625 eV to 100 keV)	Thermal (<0.625 eV)
Fission	35 to 21	About 41	23 to 28
Neutron flux	64 to 60	35 to 38	0.4 to 1.6
Absorption	21 to 12	About 57	20 to 32

Thus, these experiments could allow validating nuclear data libraries and computer codes in the fast and intermediate energy range.

3.3 Similarity study with MOX powder

This similarity study between MOX powders and proposed experimental configurations with rods is an important validation step. Like in MOX powders, the experimental configurations involve mixed uranium and plutonium oxide but with a higher density (which conducts to compact configurations).

In this way, the neutronics characteristics of the proposed experiments were compared with those of MOX powders with water content varying from 1 to 5% in weight (see table 2): these experiments cover the same energy range, when keeping a similar $H/Pu_{fissile}$ ratio.

Table 2 Neutronics characteristics

* Slowing down density is the number of neutrons that slow down past 4 eV per fission neutron.

**GMF: Average Group causing Fission in the Xmas 172-group energy structure; the corresponding energy given is the midpoint of the energy group.

	Proposed experiments	Reactor Grade MOX powder	Weapon Grade MOX powder
PuO_2 content	27.5 %	12.5 %	6.3%
Water content	3 to 10%	1 to 5%	1 to 5%
$H/Pu_{fissile}$	5 to 20	3 to 15.5	5 to 26
Slowing-down density*	0.06 to 0.27	0.017 to 0.183	0.052 to 0.29
GMF** (corresponding energy)	70 to 94 (53.6 to 3.34 eV)	68 to 86 (71.8 to 8.75 eV)	75 to 100 (35.5 to 2.24 eV)

This qualitative comparison has been quantified using the methodology used by the Oak Ridge National Laboratory [6)].

The study results show:

- a very high degree of similarity with the situations corresponding to Reactor-Grade low moderated MOX powders, which is confirmed by the ORNL [4] Sensitivity/Uncertainty methodology; $c_k = 0.97$ for Reactor-Grade MOX powder containing 3 % H_2O,
- a moderately high degree of similarity with the situations corresponding low moderated Weapons-Grade MOX powders which is confirmed by the ORNL S/U methodology; $c_k = 0.84$ for Weapons-Grade MOX powder containing 3 % H_2O,

where c_k is a correlation coefficient to assess the similarity between two configurations. $c_k=1$ indicates systems are identical, $c_k=0$ indicates systems totally dissimilar and value between 0.8 and 1 indicates similar systems.

4. Conclusion

At the beginning of 2005, the criticality facility Apparatus B of CEA Valduc will be ready to start a new experimental program. IRSN first priority is the program "qualification of structural materials cross-sections". The program devoted to measurement of the temperature coefficient of diluted plutonium solutions needs to be confirmed. At last, the proposed experimental program devoted to validate criticality calculation codes and nuclear data libraries for low-moderated mixed oxide fissile media needs to be worked out through an international collaboration.

[4] Oak Ridge National Laboratory

Appendix – Valduc Criticality facility Apparatus B

The Apparatus B criticality facility is to day the only European reseach tool available to conduct representative experiments, from the neutronics point of view, of the highly varied conditions met during storage, fabrication, transport or reprocessing of fuel elements. Implemented in 1963, the Apparatus B has been fully renovated in 1997 after performing more than 10,000 criticality experiments.

The Apparatus B enables experiments to determine the criticality conditions of a core consisting of fissile materials (solid and/or liquid) mixed or not with structural materials and surrounded or not by a reflector (liquid and/or solid).

It consists on a tank (about 190 cm x 190 cm x 140 cm) and transfer equipment used to progressively introduce water or liquid into the core tank containing the fuel. The coupled moderation and neutron reflection effects thus bring the studied configuration close to the critical state, in successive steps. During the experiments, the criticality is approached up to the value of 1 . The system reactivity slowly increases, through sequential introduction of small quantities of liquid moderator into the core (see Fig. 1). The sub-critical approach parameter, the liquid level, is measured with a needle which follow the free upper surface.

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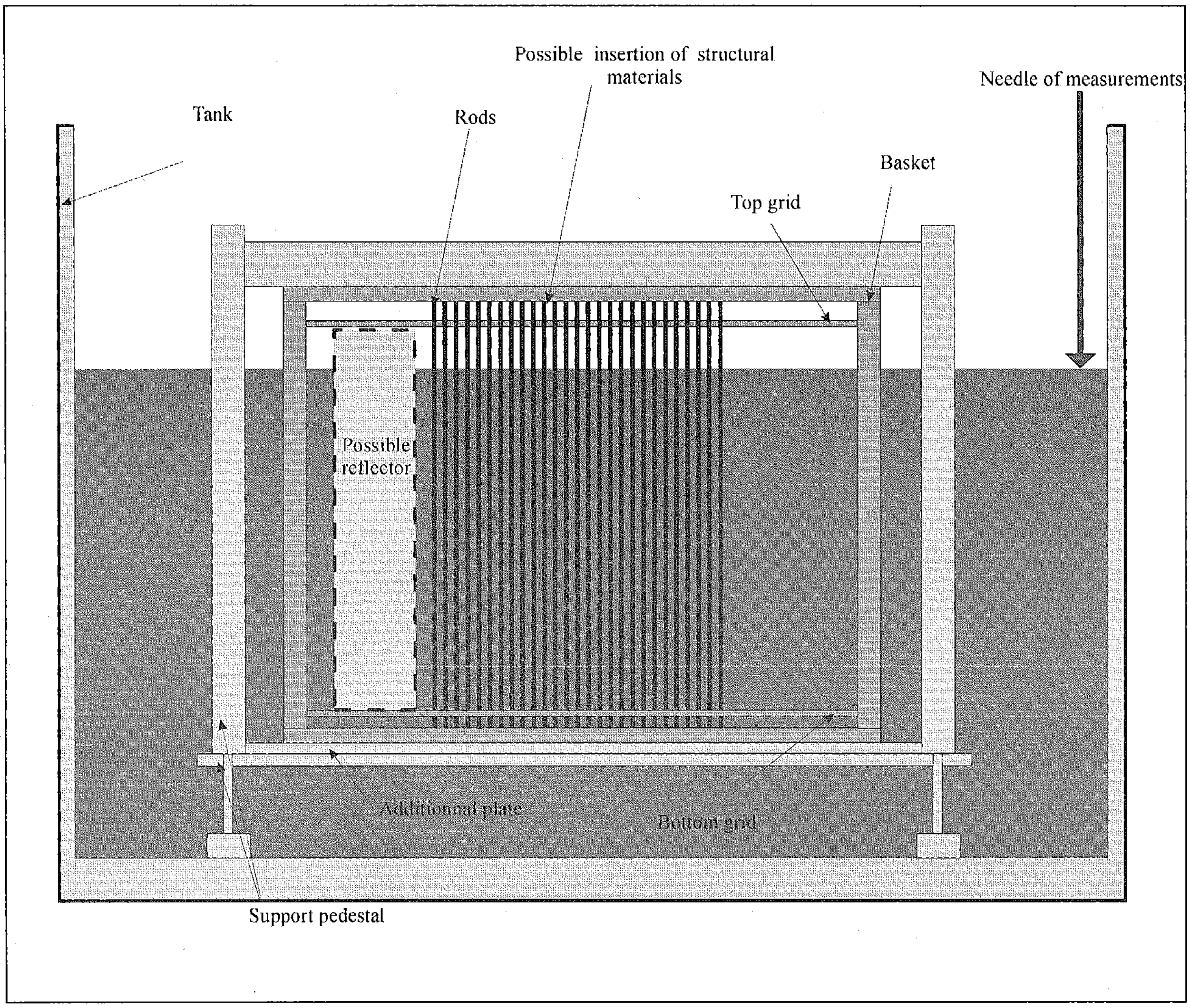

Fig.1 Apparatus B – Principle design