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STATUS AND PERSPECTIVES OF CRITICAL EXPERIMENTS

TEST POTENTIAL OF THE CEA VALDUC CRITICALITY LABORATORY STATUS AND PERSPECTIVES

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1. POTENTIALITY OF THE VALDUC CRITICALITY LABORATORY

1.1 General purpose

In order to control the criticality hazards in laboratories, plants or transportation where nuclear material in sufficient quantities is present, qualified calculation codes are needed, both for the design of these facilities and for safety assessment purposes. 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 this framework, the VALDUC Criticality Laboratory operated by the Criticality and Neutron Science Research Department (SRNC), is able to perform representative criticality experiments thanks to the major subcritical facility named APPAREILLAGE B.

The department also operates three nuclear research reactors, two metallic core reactors named CALIBAN and PROSPERO and one liquid fuel reactor named SILENE, two electrostatic accelerators, SAMES and ALVAREZ and the BISE bench, devoted to Airborne Release Fraction studies.

Furthermore it manages a large inventory of nuclear materials in various physicochemical forms: solution (uranium and plutonium), fuel rods (uranium oxides and mixed uranium and plutonium oxydes) and metals (High Enriched Uranium) and operates related equipment that is essential for the quality of experiments (analysis, preparation, reprocessing, neutronic and nuclear measurements...).

1.2 Criticality Experiment Facilities

The VALDUC Criticality Laboratory houses Criticality Experiment Facilities witches are known all over the world for nearly 45 years of experience in the field of criticality. It is today a unique set of human capabilities and experimental facilities and devices located and operated in the same place.

The Laboratory operates different facilities designed to meet specific objectives:

APPAREILLAGE B

The APPAREILLAGE B (figures 1, 2 and 3) is a sub-critical facility that can reproduce a great diversity of criticality configurations, representative of the storage, transport, and reprocessing of fissile materials in the form of rods or fissile solutions, with or without the presence of a moderator or neutron absorbers (boron, gadolinium, fission products, etc...) [1].

Implemented in 1963, APPAREILLAGE B was fully renovated in 1995-1997 (glove boxes, fuel solution storage, control system…) in compliance with the safety and security requirements of French nuclear facilities.

A great variety of experiments were performed, more than 3,000 so far, and the experimental device was adapted several times according to the programs.

APPAREILLAGE B consists of a core tank, transfer equipment (used to progressively introduce water or solution into the core tank), water and fissile solutions storage and glove boxes for radioactive solution management. The shape of the core tank can be annular, cylindrical or parallelepipedic (pool core).

The system reactivity is slowly increased, through the sequential introduction of small quantities of liquid moderator into the core. The coupled moderation and neutron reflection effects thus bring the fissile configuration being studied close to the critical state.

During the experiments, criticality is approached up to the value of $k_{eff} = 1$, more precisely around $k_{eff} = 1-\beta/10$ and the critical height is obtained by extrapolation. The sub-critical approach parameter, liquid level, is measured with a needle which follows the free upper surface.

Because it employs simple measurements means, APPAREILLAGE B allows performing highly accurate integral experiments, in accordance with the on-going and emerging requirements for the criticality codes and cross-section (reactivity worth experiments) validation.

These experiments contribute to:

- the qualification of French CRISTAL Criticality Safety Package [2],
- the production of a wide range of reference benchmarks integrated into the OECD-NEA ICSBEP Handbook (40 evaluations for 681 critical configurations)

Cylindical Core (solution)

Experimental determination of the temperature effect of plutonium dilute solutions

Pool Core (fuel rod array)

MIRTE Experimental Program

Figure 1 – APPAREILLAGE B / Different Recent Design Cores

Core confined in Glove Box
(fuel rod array immersed in solution)

Fission Products Experimental Program

Pool Core
(fuel rod array)

HTC Experimental Program

Figure 2 – APPAREILLAGE B / Different Former Design Cores

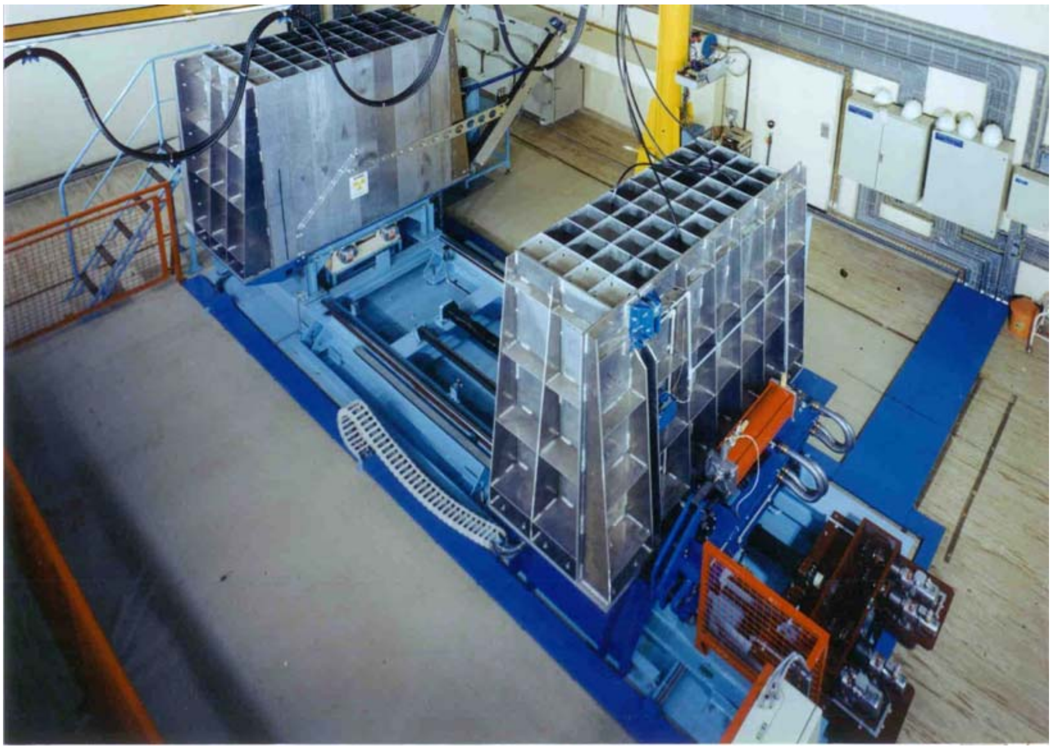


Figure 3 – APPAREILLAGE B / MARACAS Split Table (dismantled 20 years ago)

SILENE REACTOR

The SILENE experimental reactor (figure 4) was constructed in 1973-1974. More than 2,000 divergences and 1,000 subcritical experiments have been performed to study criticality accidents and their radiological consequences and for industrial uses (material and component irradiations).The major results and conclusions regarding the physics and dosimetry of the criticality excursions have been reported in [3].

The SILENE core is an annular tank of 36 cm in diameter fueled by a 71 g/L solution of Uranyl Nitrate (HEU) and located in a large concrete test room. The solution is introduced up to a super-critical level in the presence of an absorber rod. Divergence is achieved by ejection of the rod. The reactor is shutdown by draining the solution into a storage tank.

Steel Shield

Lead Shield

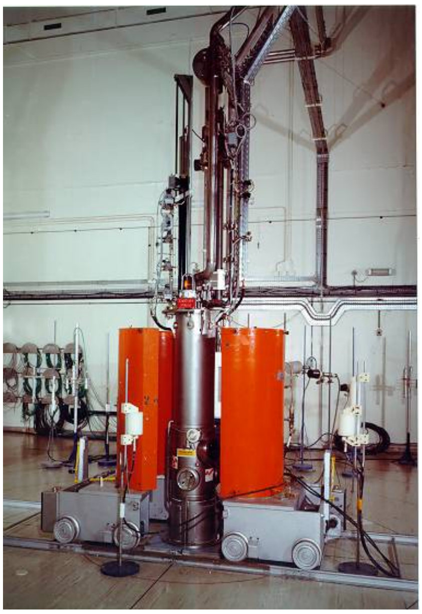


Figure 4 – SILENE Reactor / Different shielded configurations

The kinetics of the power excursions that can be reproduced vary from a few minutes to a millisecond following various modes:

- Free evolution.
- Pulse.
- Steady state.

To operate SILENE in pulse mode, 3β reactivity insertion step is authorized whereas in “reactivity ramp” operation, up to 7β can be inserted.

Experiments performed with SILENE regarding criticality deal with the following objectives:

- Studies of criticality accident phenomena in solution media, intended to better define and model accident mechanisms and phenomenology and provide data for safety assesments, relevant to surveillance and prevention policies.

- Enhancing detection instruments and qualifying them in various criticality accidents scenarios (Criticality Accident Alarm System and radioprotection detectors qualification and testing).
- Research and development of dose measurement methods.
- Radiation Protection studies.
- Biological dosimetry and studies on physiopathology of complex irradiations.
- National and international comparison exercises of criticality accident dosimetry (figure 5).
- Experimental validation of calculations codes in the fields of Criticality Accidents or Health Physics Protection.
- Training of teams for emergency preparadness (organizing actions such as personnel evacuation, rapid medical screening, training of teams to deal with criticality emergency situations) as mentionned in ANSI/ANS 8.23-1997:
    - *Emergency response personnel training*
    - *Re-entry team personnel training*
    - *Technical staff training*
- Development of the hands-on training and training for operators proficiency (International training collaboration).
- Information Preservation and Dissimination to sustain knowledge in criticality safety.
- Reactivity worth measurements.
- Sub-critical measurements.

Figure 5 – SILENE Reactor / Criticality Accident Dosimetry Applications

Figure 6 – SILENE Reactor / CAAS Tests

CALIBAN REACTOR

The CALIBAN reactor, in operation since 1970, is an experimental super prompt-critical reactor with a metallic core (figure 7). Cylindrical in shape, its diameter is 19.5 cm and its height is 25 cm. It is composed of two blocks: a fixed block and a moveable block. Each block is made of five metallic plates of a molybdenum and HEU alloy. Four cylindrical control rods, made of the same alloy, allow operating the reactor following two modes: a steady state power mode and a pulsed one. CALIBAN reactor includes a central irradiation cavity situated inside the two blocks. The whole core is covered by a steel and boron carbide hood.

With its hood **Without its hood**

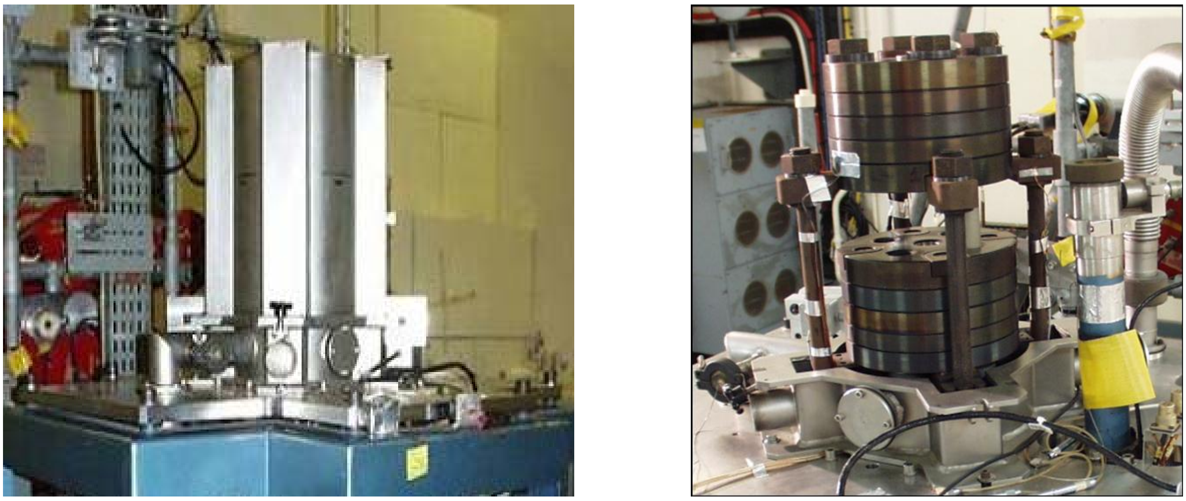
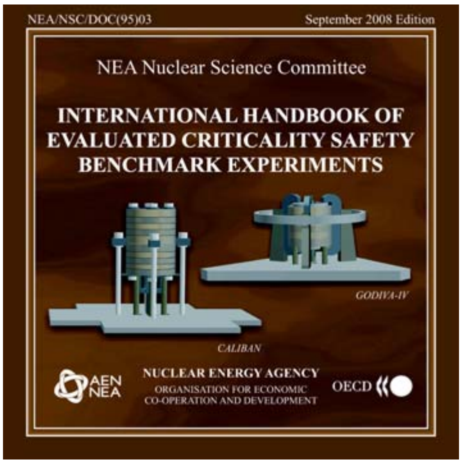


Figure 7 – CALIBAN Reactor

CALIBAN is able to meet various needs in the criticality field, some applications [4, 5, 6 and 7]:

- Neutron transport code improvement.
- Various material neutron property studies.
- Nuclear reaction cross section databases testing (figure 8).
- Fissile element critical mass studies.
- Studies of criticality accident phenomena in metallic media (dosimetry and kinetic aspects as SILENE) and so:
  - *Training of teams for emergency preparedness.*
  - *National and international comparison exercises of criticality accident dosimetry.*
- Development of the hands-on training and training for operators proficiency (International training collaboration).
- Information Preservation and Dissimination to sustain knowledge in criticality safety.
- Reactivity worth measurements.
- Sub-critical measurements.



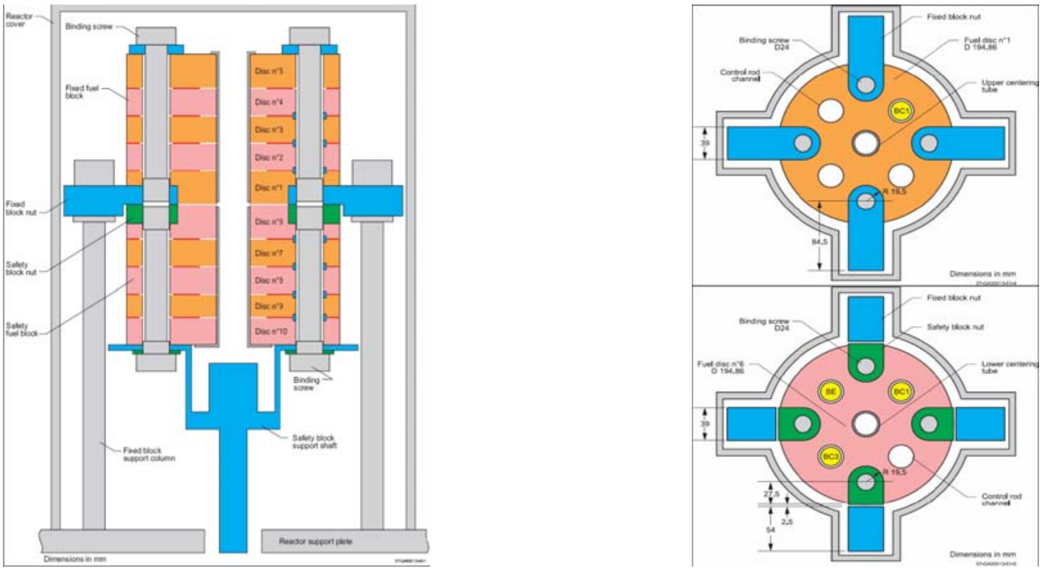


Figure 8 – CALIBAN Reactor OECD-NEA ICSBEP Benchmark [4]

1.3 Related equipment

In order to perform high quality experiments that can then serve as benchmarks, and meet the requirements of regulations and the management of nuclear materials, the following specific equipment is operated in the VALDUC Criticality Experiment Facilities:

- Physico-chemical analysis laboratories for uranium and plutonium.
- Counting laboratory (Dosimetry and α, β, γ radiation measurement).
- Chemical recycling laboratories for uranium and plutonium solutions.
- Electronics and nuclear instrumentation laboratory.
- Storage for Nuclear Fuels and Special Nuclear Materials.

Moreover for performing experiments, a broad range of fuel types is available:

- Plutonium and uranium nitrate solutions.
- PWR type rods representing fresh or high burn up fuel rods (without Fission Products).
  - *1260 UO_2 rods (4.75 % ^{235}U).*

- *2500 HTC UO_2-PuO_2 rods (1.1 % plutonium and 98.9 % uranium with 1.57 % ^{235}U).*

HTC « Haut Taux de Combustion » is the French acronym of High Burn up Fuel.
Both types of rods are zircaloy cladd with a fissile column height of 900 mm.

2. STATUS ON CRITICALITY PROGRAMS

The Criticality and Neutron Science Research Department manages and operates experimental devices APPAREILLAGE B, SILENE and BISE house in Building 010, for the benefit of French Institute for Radiological protection and Nuclear Safety (IRSN). Thus the criticality programs recently and currently conducted by the CEA with APPAREILLAGE B are funded by IRSN.

A brief review of the last IRSN and CEA programs carried out is presented here.

2.1 Qualification of the decrease from ^{241}Pu to ^{241}Am ("Aging" Effect Program)

The HTC rods were manufactured in 1987. Since then ^{241}Am has increased because of ^{241}Pu decay (half-life 14.4 years), therefore, the rod reactivity has decreased. In order to quantify this loss of reactivity, some experiments originally carried out from 1986 to 1991 in the framework of the "HTC experimental program" [8], have been redone in 2004-2005, more than 15 years later.

Calculations have shown that the effect is about 3% on k_{eff}. Half is due to the decrease of ^{241}Pu and half is due to the increase of ^{241}Am absorption.

2.2 Measurement of the temperature coefficient of dilute plutonium solutions (B-Pu Program)

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 decided in 2004 to lead an experimental program with APPAREILLAGE B to perform sub-critical experiments with plutonium concentrations of about 14 to 20 $g.L^{-1}$ at various temperatures ranging from 21°C to 40°C [9].

Between 2006 and 2007, thirteen experiments have been performed and some of them demonstrated a positive temperature effect (isotopic % mass ^{238}Pu / ^{239}Pu / ^{240}Pu / ^{241}Pu / ^{242}Pu / 0.19 / 76.80 / 20.77 / 1.12/1.10).

2.3 Qualification of structural materials cross-sections (MIRTE Program)

In 2005, IRSN decided to perform new integral experiments with APPAREILLAGE B to address the need of criticality calculation codes and nuclear data validation dealing with various structural materials that are important to criticality safety – The MIRTE program [10].

In 2007, the program evolved into an international collaboration involving IRSN, the French energy group AREVA, the French National Radioactive Waste Management Agency ANDRA and the United States Department of Energy (US-DOE / NNSA).

This program mainly aims to study interacting configurations. Nevertheless, some reflected configurations have been also designed. The experimental configurations which are being tested, involve water-moderated, low enriched UO_2 rods lattices separated or reflected by different structural material plates.

Three main configurations are under testing:

- Two interacting UO_2 rod arrays separated by an absorbing screen of thicknesses up to 30 cm.
- Four interacting UO_2 rod arrays separated by thin absorbing plates for thicknesses less than 2 cm.
- One UO_2 rod array reflected by structural material (5 to 20 cm).

After a preliminary feasibility study, following materials have been obtained by IRSN:

- Iron, Nickel, Lead, Aluminum, Copper, Titanium, Zircalloy.
- Silicon dioxide (glass).
- Concrete, with various water contents.

The first experiment phase of MIRTE program will be completed in the middle of the year 2010, 42 configurations are planned and 20 have been already tested since December, 2008.

2.4 Integral measurement of ^{235}U isomer with CALIBAN reactor

The integral measurement of the ^{235}U isomer activation cross section in a fission-like spectrum was performed in 2005 to 2006 using the neutron leakage of the pulsed reactor CALIBAN. The µg activation foils were counted with a dedicated electron detector. Sample preparations, efficiency measurements, irradiations and isomer decay measurements were performed at the CEA VALDUC Criticality Laboratory by a multinational team of researchers coming from LOS ALAMOS (US-DOE / NNSA – LANL), BRUYÈRES LE CHÂTEL and VALDUC (CEA-DAM).

The results of this experiment have been compared to the last evaluated cross section at CEA-DAM BRUYÈRES LE CHÂTEL and LOS ALAMOS NATIONAL LABORATORY and presented in the 5th NEMEA Workshop [5].

2.5 Reactivity worth measurements by perturbation method with CALIBAN and SILENE Reactors

Reactivity worth measurements of material samples placed in the central cavities of nuclear reactors allows the testing of cross section nuclear databases or to infer information about the critical masses of fissile elements.

Such experiments have been already completed on CALIBAN using the perturbation measurement technique.

Calculations have been recently performed to prepare future experiments on new materials, such as light elements, structural materials, fission products and actinides [6].

2.6 Calculation of kinetic parameters of CALIBAN Reactor from stochastic neutron measurements

Knowing the fundamental kinetic parameters of the reactor is very useful, indeed necessary, to the operator. The purpose of this study was to develop and perform experiments allowing the determination of these parameters.

The prompt neutron decay constant and particularly its value at criticality can be measured with reactor noise techniques such as the interval-distribution, the Feynman variance-to-mean, and the Rossi-α method. By introducing the Nelson number, the effective delayed neutron fraction and the average neutron lifetime can also be calculated with the Rossi-α method.

Sub-critical, critical, and even super-critical experiments were performed. With the Rossi-α method, it was found that the prompt neutron decay constant at criticality was $6.02\times10^5\ s^{-1} \pm 9\%$.

Experiments also brought out the limitations of the used experimental parameters [7].

3. FACILITIES AND PROGRAMS PERSPECTIVES

3.1 APPAREILLAGE B

APPAREILLAGE B programs are planned until the end of 2012 (phase 2 of the MIRTE Program, to be confirmed by IRSN) and the facility uses are currently "available" until 2014.

APPAREILLAGE B licensing, in compliance with the new safety and security requirements of French nuclear facilities, will have to be done before 2015.

3.2 SILENE

SILENE experiments are "possible" until the end of 2010.

SILENE licensing, in compliance with the new safety and security requirements is probably impossible without a complete refurbishment:

- Chemical uranium recycling facility is difficult to maintain in safe and efficient conditions due to its old design dating from the time of the CRAC [3] facility (glove boxes for example).
- The control system is outdated (1974) and also very difficult to maintain.

3.3 Building 010 perspectives

As a result, the proposal of the CEA is to dismantle the experimental devices and the related chemical recycling facilities.

This could be done with the following time frame:

- **2009-2011**: Dismantling of the Chemical Plutonium Recycling Laboratory.
- **2011-2014**: Dismantling of SILENE (Reactor and Chemical Uranium Recycling Laboratory).
- **2012-2014**: Dismantling of BISE.
- **2013-2014**: Dismantling APPAREILLAGE B.
- **2014-2015**: Dismantling of the uranium and plutonium Radiochemistry Analysis Laboratories.

3.4 CALIBAN

CALIBAN programs are planned until after 2020.

CALIBAN licensing, in compliance with the new safety and security requirements of French nuclear facilities, will probably have to be done after 2015.

4. CONCLUSION

Over the 45 years of experimentation, APPAREILLAGE B has been used to study a wide set of nuclear materials, such as uranium and plutonium nitrate solutions, UO_2 powders, UO_2 or MOX rods, neutronic poisons, fission products and various moderators, reflectors and shields. Moreover, as it is flexible and easy to operate, it is well adapted to validate a wide range of critical configurations even for large scale geometry (split table machine as MARACAS [11] for example).

In the same way, SILENE has been used since 1974 as a unique tool to study the physics of criticality accidents involving fissile solution, to develop and qualify CAAS, to train teams for emergency preparedness and to perform comparison exercises of criticality accident dosimetry in a known mixed radiation field of neutrons and gamma photons as we do with CALIBAN since few years.

Nevertheless, taking into account that VALDUC Criticality Laboratory was built in the 1960's and that the existing Criticality Experiment Facilities are beyond their usable life, CEA must shut down and dismantle the aging uranium solution SILENE reactor and its related equipment but also the chemical recycling uranium and plutonium laboratories (2010-2015).

However, there is a need for new precise integral data utilizing plutonium, uranium, and minor actinides in various combinations to support advanced reactor fuel cycles. Additionally, there are some major improvements needed in the understanding of the physics of large, dilute systems that can only be achieved with a large multipurpose horizontal assembly device.

Moreover, the detection of a criticality accident and the emergency management thereof is of major interest. Indeed, critical assemblies are used for the qualification of detection and alarm systems and allow the training of biological analysis laboratories for the determination of the received dose during a criticality accident.

To maintain the capability to meet the future emerging needs for the next 30 years "Building 010" shall be refurbished, in compliance with the new safety and security requirements of French nuclear facilities, if construction of new experiment assemblies is envisioned.

This new facility should be designed to combine the capability to conduct sub-critical, critical, and transient integral experiments in complex configurations using all fuel types (solid, liquid or combination).

Time schedule for such a new facility could be:

- **2010-2011**: Approval of the General Specifications.
- **2010-2013**: Feasibility Studies
  Definition of General Purpose for Safety and Security Objectives
  Preliminary Design.
- **2012-2018**: Final Design
  Construction
  First Tests

These experiments should be designed to address configurations studied in safety assessments and physical basis experiments for cross section qualification and criticality excursion studies. In addition, the new facility would be used to train people in the physical behavior of nuclear systems at, near, and above the critical state.

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