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The Need for Integral Critical Experiments with Low-moderated MOX Fuels

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NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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FOREWORD

The use of MOX fuel in commercial reactors is a means of burning plutonium originating from either surplus weapons or reprocessed irradiated uranium fuel. This requires the fabrication of MOX assemblies on an industrial scale.

The OECD/NEA Expert Group on Experimental Needs for Criticality Safety has highlighted MOX fuel manufacturing, as an area in which there is a specific need for additional experimental data for validation purposes. Indeed, integral experiments with low-moderated MOX fuel are either scarce or not sufficiently accurate to provide an appropriate degree of validation of nuclear data and computer codes. New and accurate experimental data would enable a better optimisation of the fabrication process by decreasing the uncertainties in the determination of multiplication factors of configurations such as the homogenisation of MOX powders.

In this context, the OECD/NEA Nuclear Science Committee organised a workshop to address the following topics:

1) Expression and justification of the need for critical or near-critical experiments employing low-moderated MOX fuels.

2) Proposals for experimental programmes to address these needs.

3) Prospects for an international co-operative programme.

The workshop was held at OECD headquarters in Paris on 14-15 April 2004. Appendix 1 lists the composition of the organising committee and scientific secretariat. Forty-four participants from seven countries and two international organisations attended the workshop (see Appendix 2 for a list of participants). Those who attended the meeting belong to the main industrial companies involved in MOX fuel manufacturing, R&D organisations and regulatory bodies. Seventeen presentations were given in three technical sessions, and a panel discussion involving eight panellists concluded the workshop (see the meeting programme in Appendix 3).

This report presents the proceedings of the workshop. One paper from Session 1 was not submitted for publication. The summary record of the panel session is included in the proceedings. The main conclusions of the workshop are summarised in the last section of the Executive Summary. A CD-ROM containing the electronic version of the papers and slides presented during the workshop is appended to this report.

TABLE OF CONTENTS

EXECUTIVE SUMMARY

Introduction

Over the past several years, the NEA Working Party on Nuclear Criticality Safety (WPNCS) has been discussing the need for additional integral benchmark experiments. An Expert Group on Experimental Needs was formed in 1997 to investigate international experimental needs. This group has identified the MOX fuel manufacturing process as an area in which there is a specific need for additional integral benchmark data. In November 2003, the WPNCS issued a recommendation to the NEA Nuclear Science Committee (NSC) to promote the establishment of an international consortium that would perform the needed experiments. This recommendation was presented and discussed at the NSC bureau meeting in December 2003. The NSC bureau asked the NEA secretariat to organise a workshop at which the needs can be further verified and proposals for experimental programmes presented and discussed. The main objective of the workshop was to address the following questions:

1) Is there a real need for additional MOX experiments?

2) Which proposal or combination of proposals seems to best fill the need?

3) What are the prospects for an international co-operative programme?

Summary of the sessions

Forty-four (44) participants from 7 countries and 2 international organisations attended the workshop. Those who attended belong to the main industrial companies involved in MOX fuel manufacturing (Belgonucléaire, BNFL and COGEMA), to R&D organisations and to regulatory bodies. Seventeen (17) presentations were given in 3 technical sessions, and a panel discussion involving 8 panellists concluded the workshop. The summary of the sessions is given below.

Session I: MOX Fuel Fabrication – Design, Industrial Experience and Associated Criticality Safety Issues

Five presentations were given. It was emphasised that the lack of experimental data on MOX fissile media does not impact the safety of existing plants. Indeed, safety margins were taken into account in the design of the plants and these margins are sufficient to compensate for the lack of adequate data for the validation of current configurations. However, a reduction of margins would provide the operators of running MOX fuel fabrication facilities with added flexibility in the process, especially as concerns the preparation of the primary blend. Additional benchmarking may thus benefit application areas involving Pu and MOX powders by reducing the margins.

Session II: Experimental Data Needs

Six presentations were made during this session. Sensitivity/uncertainty analysis techniques were extensively used. These techniques provide interesting insights into code, microscopic and integral data validation. However, the comparison of uncertainty data, available in major evaluated files, showed large differences. This impacts the calculations of the effect of nuclear data uncertainties on integral parameters.

Old versions of nuclear data libraries (e.g. UKNDL, Hansen & Roach) lead to very conservative results. It was also demonstrated that high safety margins considerably impact MOX throughput and/or fault tolerance in some accident scenarios. Unfortunately, there is not sufficient experimental data available to derive accurate determination of biases with recent evaluated data files. In fact, the only applicable experiments, performed at Hanford in the 70s, were shown to be of a limited reliability due to the lack of accurate experimental information. The results from some completed experimental programmes (e.g. the ERASME/S experiments performed at CEA Cadarache and some of the BFS configurations) could provide valuable (though perhaps limited) information if released.

A study from JAERI showed that mixing conditions in the MOX fabrication process need to be studied in more detail (e.g. study of volume conservation when mixing the powder and additives and study of the effect of heterogeneous distributions of concentration).

Session III: Proposed Programmes and Presentation of Experimental Facilities

A wide range of experimental proposals was made including homogeneous and heterogeneous set-ups of low-moderated (tight pitches or voided) lattices. The possibility of using plutonium powder in a test zone or for oscillation measurements was also investigated. It was shown that the availability of fuel in the experimental facility impacts the cost of the programme. However, this also affects the possibilities of experimental set-ups since the available fuel is not always representative of the needs.

Panel Discussion: Prospects for an International Co-operative Programme

The following questions were debated:

1) Is there a real need for additional MOX experiments?

2) Which proposal or combination of proposals seems to best fill the need?

3) What are the prospects for an international co-operative programme?

There was a consensus among the panellists that there was a lack of reliable data for better code validation and that additional experimental data would be beneficial. However, the variety of interests expressed by the panellists together with a lack of detailed information on the experimental proposals were the main reasons why a consensus on a single or a combination of experimental programmes could not be reached. Most panellists expressed their interest in setting-up an international co-operative programme to perform the needed experiments. Indeed, this would allow efficient cost sharing and thus optimisation of available resources.

Conclusion

The recommendations of the workshop can be summarised as follows:

- Investigate the possibility of releasing and evaluating unpublished experimental data (especially the ERASME/S and BFS-49 data).

- Define a framework and method for the selection and performance of new experimental programme(s) of interest.

- Help disseminate and assess sensitivity/uncertainty methods.

SESSION I

MOX Fuel Fabrication – Design, Industrial Experience and Associated Criticality Safety Issues

Chairs: Y. Vanderborck, T. Yamamoto

MELOX PLANT MOX MANUFACTURING RESULTS AND PROSPECTS

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Abstract

The main industrial facilities involved in closing the back-end of the nuclear fuel cycle in France are La Hague reprocessing plants in which fission products are vitrified and recycle materials are recovered and purified, and MELOX plant where the recovered plutonium is used to fabricate MOX fuel. These facilities are operated by AREVA/COGEMA.

Since the opening of the first plutonium box at the MELOX plant in February 1995, a huge return of experience has been accumulated in the operation of the first large-scale industrial and highly automated MOX manufacturing plant in the world. The results which will be presented in this paper are the consequences of the permanent quality improvement policy implemented since start-up as a necessary condition to satisfy safety requirements as well as commercial, economical and social constraints.

In the first part of the paper, a detailed history of production since February 1995 up to 2003 will be presented, with special emphasis on 1997 (first year of 100 tHM production), March 1998 (a one-month demonstration period concluding with 19 t production), and since 1999 (beginning of multi-design productions within the limits of the decree).

The second part of the paper will be devoted to the results and performances of the plant since start-up:

- Description of management by objectives, aiming at customer satisfaction.
- Safety and security indicators history, such as gaseous and liquid waste releases, health physics, safety events related to INES scale, etc.
- Product quality, satisfaction of the technical specifications of the customers in association with the complete mastering of the fabrication process on the one hand, and the quality control process on the other hand.
- Productivity of the plant in relation to the high level of professionalism of the workers, and the permanent policy of improvement of quality and output, and the performances of the industrial tool. In illustration, some examples will be given, especially all the improvements enabling the plant to recycle the scraps generated by the production.
- Customisation of the productions, giving MELOX the opportunity to fabricate all types of PWR MOX fuels demanded or foreseen in the future by the conceptor-vendors.

Finally, the paper will present the prospects for the future, and especially in the development of the level of production up to 145 tHM within the limits of the new decree.

MELOX MOX fuel manufacturing plant history

Design, construction and start-up

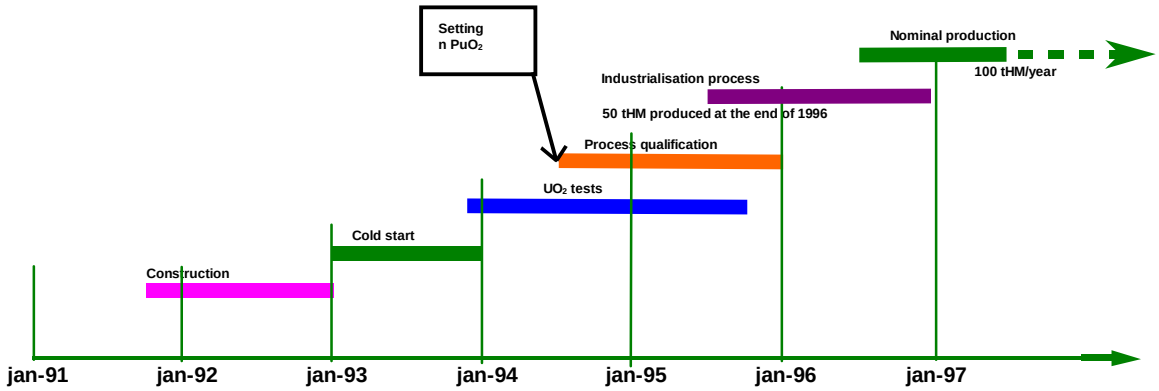
The very beginning of the project was the decision made in 1985 by EDF (the French utility company), COGEMA and FRAMATOME to promote the recycling of plutonium in the 900 Mwe French PWR reactors, and to design and build a high-capacity MOX fuel manufacturing plant in France.

The design studies, including the preliminary Safety Report issued in 1988, led to the "Decree of Authorisation of Construction" of the plant, delivered in May 1990,with a limit of 100 t HM of annual production.

The construction on the Marcoule site in 1991 was followed by inactive tests of the equipment in 1993, and a period for operational testing of the equipment with UO_2 powder.

The first introduction of one PuO_2 can in the production line was authorised in February 1995. The qualification period began with the very first batch of MOX manufactured. Over the year 1995, the qualification of the processes of the production line and of the 17 × 17 PWR EDF MOX fuel was satisfactorily performed. The first deliveries of MOX assemblies to EDF took place in mid-1996. The first year of 100 tHM MOX production and industrial maturity was 1997.

Figure 1. Start-up of the MELOX plant



Effective capacity of the plant

A continuous five-shift operation in March 1998 led to a 19-tonne production of MOX in one month, this demonstrating an annual production capacity of more than 200 t HM.

Accumulated experience

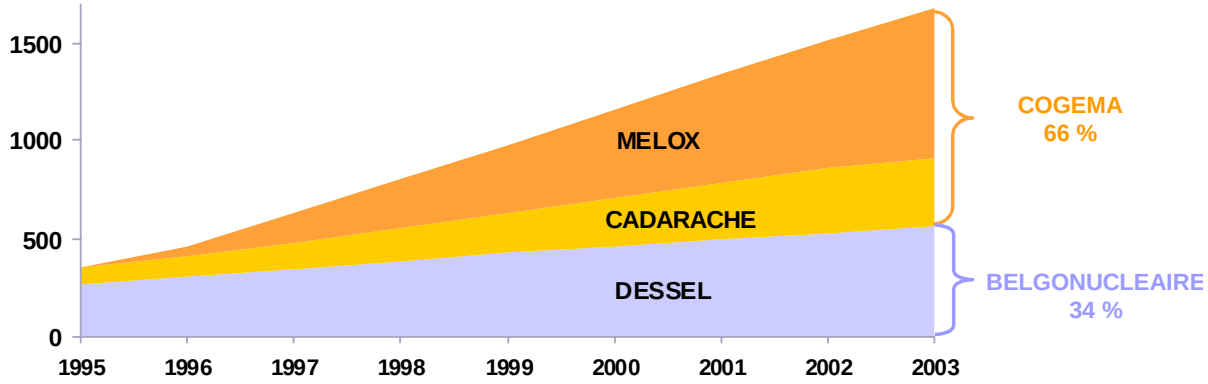
As of 1999, additional equipments were installed for multi-design production. New production of the PWR MOX design started in 1999, followed in 2001 by the first BWR production for Japanese utilities.

At the end of 2003, cumulated MELOX production represents 750 tHM, in 1 700 fuel assemblies.

Presently the contribution of the MELOX plant in the total of production of the three MOX plants (MELOX, CADARACHE and BN) is preponderant. This is illustrated in Figure 2, which also gives a good idea of the huge amount of experience in MOX manufacturing accumulated by the AREVA/COGEMA group.

Today, more than 1 000 persons are directly or indirectly involved in the operation of the MELOX plant.

Figure 2. 1 512 tHM of MOX fuel pellets produced by the end of 2002



Aiming total quality

Since the beginning of industrial activities, MELOX has developed a total quality management policy. This policy is based on:

- continuous improvement, with twin goals of complete customer satisfaction and sustainable development;
- encouragement for workers to remain permanently involved;
- successful integration in the local environment.

One of the main axes of this policy is “management by objectives”, which is implemented at every level of the MELOX structure. The MELOX steering committee is responsible for defining the general objectives related to three main fields (customer satisfaction, management safety/environment, local integration). These general objectives are then taken into account by every part of the MELOX organisation in order to define more than 200 action plans. Each plan is then piloted by a specific employee. This integrated quality system is certified under international ISO 9000 and 14001 norms.

This policy, which includes other axes such as permanent improvement of quality and constant involvement of employees, was awarded with the French National Quality Prize in 2001 (Figure 3).

Furthermore, MELOX is regularly evaluated by EDF as its best fuel supplier among the industrial fuel cycle companies (2000 rating: 91/100).

Figure 3. French National Quality Prize awarded to MELOX in 2001 for its total quality management policy

Product quality

MIMAS process

Through its impressive accumulated experience as concerns MOX fuel fabrication, MELOX has met with MIMAS the challenge of producing fuel meeting the most stringent requirements of quality and assuring this quality to the same level of confidence as UO_2 fuel. MOX fuel is indeed very similar to UO_2, since as many features as possible are adopted unchanged.

In order to benefit to a maximum extent from the statistically demonstrated quality level achieved by the UO_2 fuel, as many features as possible of the UO_2 fuel assemblies are adopted for the MOX fuel assemblies. The main difference is pellet itself.

The success of MIMAS is to make the MOX pellet resemble as closely as possible an industrially established UO_2 pellet with well-established quality records.

This process provides high scrap recycling rates and good performance in terms of Pu utilisation.

A successful start-up

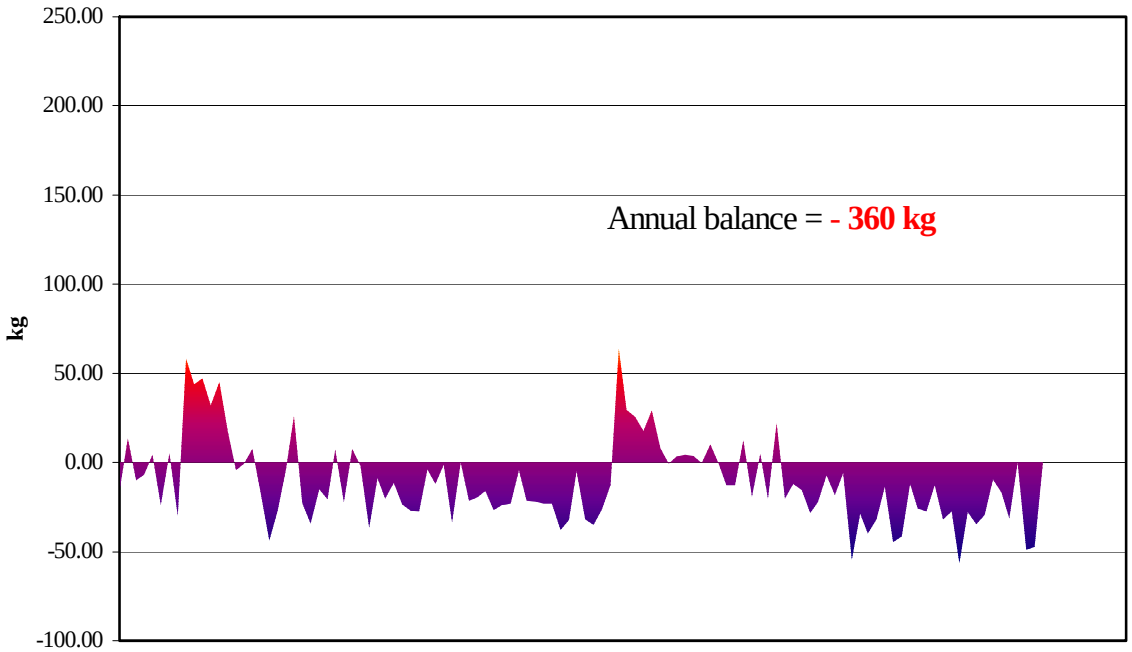
As a result of the optimised conditions of MELOX's start-up, conformity to customer requirements was assured from the beginning: the first production pellet and rod batches were fully demonstrated to comply with the technical file and FA delivered to the reactor.

During the first production year, the equipment was progressively adjusted in order to improve yield batch after batch, by reducing the main rejection sources, related to:

- complex circulation of pellets;
- pelletising cycles;
- reliability of electrical heating of furnaces;
- high automatisation of pellet quality aspect-checking devices, which required a significant period of time to be fully optimised.

During the following years, production was constant at the 100 tHM level, with continuous increase of productivity ratios and plant efficiency.

Figure 4. Extra rejected oxide for 2002



As an example, the qualified dry recycled powder incorporation rate moved from an initial 10% to 15%, and 18% was achieved in 2002. Concerning rod fabrication, the rate of conformity before repair was around 95% at plant start-up. Continuous improvement quickly raised this ratio to 98%.

As a result of this high quality level and high scrap recycling ability, the global ratio of the plant is now more than 99.5%, demonstrating an optimal use of Pu produced in the reprocessing step. Figure 4 displays pellet extra rejection for 2002.

Industrial efficiency

A main concern in a MOX plant is to establish and maintain a deep quality and safety/security culture involving all plant employees.

The first essential stage for MELOX was to hire and train employees before commencing UO_2 tests, intensively using existing plants of the AREVA/COGEMA group.

As UO_2 tests commenced, all safety and security rules related to Pu conditions were already in place, so as to be totally ready and trained for real Pu start-up.

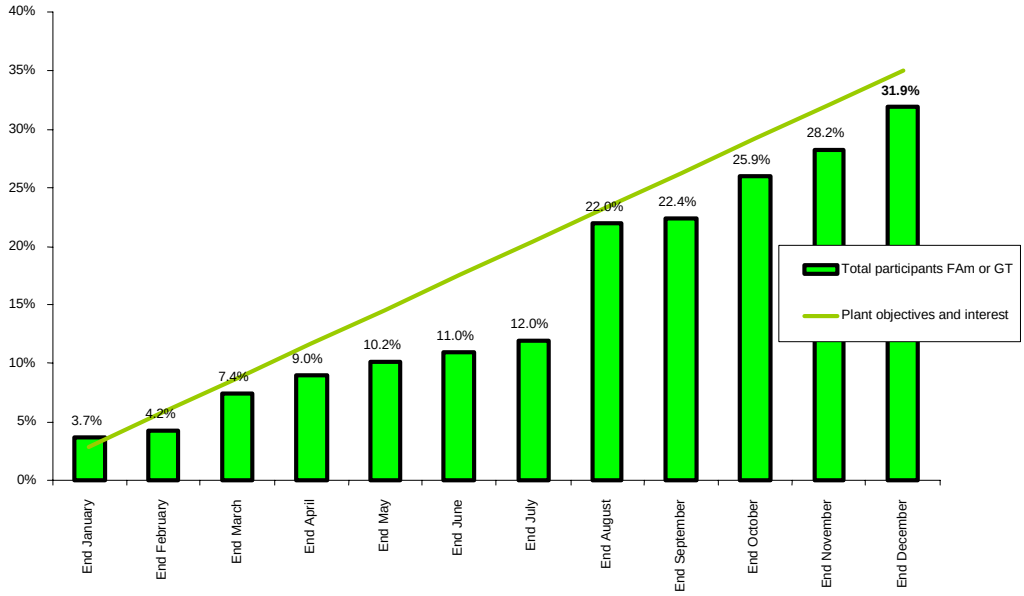
This high level of professionalism has been continuously sustained, leading to significant results in terms of industrial efficiency and safety management.

Continuous employee training

A significant training programme, corresponding to 7% of total wages, aims at the continuous improvement of professional skills. The main items of this programme deals with technical knowledge, polyvalence, quality awareness and cultural training as concerns foreign customers.

Workers are encouraged to bring individual and collective suggestions to improve any field relative to plant activity. They are also encouraged to be involved in working and project groups.

Figure 5. Personnel contribution to improvement actions in 2002



Fabrication tool productivity

The productivity of the most sophisticated parts of equipment was continuously increased by relying on the feedback of production teams. This feedback is tracked and analysed by specialised working groups in order to define technical, organisation or maintenance improvements. For example:

- The average grinding and sorting throughput was multiplied by two in 1997, and increased by 50% between 1998 and 2002.

- The average throughput of the cladding machines was improved by a factor of three between 1996 and 2002.

This high level of productivity made it possible to reduce the number of production shifts to two within the framework of the 100 t HM license.

Continuous safety and security improvement

Nuclear safety

Since plant start-up, no significant incident has occurred, which demonstrates the high technical level of safety systems and the workers' professionalism.

Table 1. INES events from 1999-2003

INES events	1999	2000	2001	2002	2003
Level O	5	0	3	6	2
Level 1	0	1	2	0	0
Level > 1	0	0	0	0	0

Dosimetry mastery

Despite increasing Pu content, workers' individual dosimetry is still very low with respect to legal requirements (limit: 20 mSv, see Table 2)

Table 2. Levels of worker dosimetry

	1999	2000	2001	2002	2003
0-10 mSv	99%	100%	98%	98%	>99%
10-15 mSv	1%	0%	2%	2%	<1%
> 15 mSv	0%	0%	0%	0%	0%

Local integration

Since the beginning, MELOX has been continuously concerned with its integration with respect to local social life. This has lead to implement a policy aiming at open-mindedness, information and involvement with communities around the plant.

For example, MELOX regularly sends information carriers to main contacts such as town, county and regional councils, associations, local media, and the Local Information Commission (CLI) of Gard prefecture. An annual report is dedicated to the impact of the plant on the environment.

MELOX is deeply involved in local life. An active partnership has been constructed with local organisations, in which many employees are involved. These include varied activities, ranging from the protection of cultural heritage to the promotion of sporting, cultural and social activities. Additionally, about 800 visitors are welcomed to the plant each year.

Customisation of MELOX productions

New customers

In order to increase its ability to satisfy its foreign customers, especially Japanese and German utilities, MELOX has built an additional set of equipment, connected to the original line. This enables the manufacture of any type of MOX fuel design for PWR and BWR reactors. A 1999 decree allows MELOX to operate this new equipment.

Qualifications

The qualification process for foreign customers began in 1998:

- Off-line and on-line qualifications for NFI 17 × 17 PWR design were completed in 1999.
- Off-line qualification for MHI 17 × 17 PWR design was obtained in 1999.
- On-line qualification for GNF-J 8 × 8 BWR design was obtained in 2001.
- KTA 14001 certification was obtained in July 2002.
- Qualification for FANP/FG for 16 × 16 and 18 × 18 PWR designs was obtained in 2003.

All these qualifications have been obtained based on feasibility studies and detailed reviews of customer specifications, inactive and active tests, workshop and quality system audits performed by each customer.

A new set of equipment

The additional set of equipment consists of:

- a fourth multi-design press;
- a third grinding and sorting line;
- a fourth sintering furnace;
- two additional pellet storages and associated transfer systems;
- a second rod inspection line;
- a second assembling bench;
- a third cladding line;
- additional fuel rod storages.

Fabrication experience

Japanese market

The first PWR batch was manufactured in 1999/2000. All pellets and rods were manufactured on the original MELOX line. A few FA were manufactured on the multi-design mounting line. Fabrication of fuel assemblies was stopped after the change of regulation concerning import of foreign fuel in Japan.

A first batch of 32 BWR 2/5 fuel bundles was manufactured on the multi-design line in 2001. A second batch of 60 BWR 6 fuel bundles was started in 2002. These two batches are stored in MELOX either in the form of fuel rods or in the form of fuel bundles.

German market

An initial batch of 24 16×16 FA was manufactured on the multi-design line. Production of a second batch of 32 18×18 FA started in the second half of 2003 at the MELOX plant.

Prospects for the future

In April 2001, COGEMA issued an application to increase the level of authorised production of MELOX, up to 195 tHM. This value was demonstrated to be the right level considering the real capacity of the plant without need for additional nuclear building or significant modification.

The safety requirements and limits defined in the previous 1990 decree (plutonium characteristics, hold up in the plant, and especially liquid and gaseous releases) remain unchanged. Concerning this major item, the excellent performances of MELOX must be pointed out:

- The gaseous releases remain under the limit of detection which itself corresponds to a very small part of the authorised limit. The limit itself was fixed by the authorities in order to result in a negligible impact on the environment.

- The liquid wastes, in a MOX plant in which the process is necessarily a dry one, are limited to very low values resulting in negligible impact on the environment.

Considering the urgent need to take into account (starting in 2003) the needs for German MOX fuel manufacturing coupled with the end of commercial MOX production in the Cadarache plant at the end of July 2003, as a first step in the licensing procedure COGEMA petitioned for a new decree which would authorise 145 tHM. The value of 145 tHM comes from the direct addition of the MELOX and Cadarache productions of MOX.

This petition was met with the official approval of the French government as of the end of September 2002, and the new decree of 145 tHM has been applicable since September 2003.

OPERATING A CRITICALITY SAFETY MANAGEMENT SYSTEM IN A MOX FUEL FABRICATION PLANT

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Abstract

Belgonucléaire operates a mixed plutonium/uranium oxide (MOX) fuel fabrication plant. MOX is manufactured by mixing plutonium oxide and depleted or natural uranium oxide powders. The whole production line is installed in dry glove boxes. The fabrication plant is divided into four main parts corresponding to different process steps. Due to the important throughput of fissile material, criticality prevention is one of the major concerns for the safe operation of the plant. The prevention of the criticality risk rests on two consecutive pillars: the determination of the breaking values of operation and determination of the procedures of operation. The paper presents these pillars and provides some examples of their use.

Introduction

Belgonucléaire operates a mixed plutonium/uranium oxide (MOX) fuel fabrication plant located in Dessel (Belgium). Since 1986, the plant has been working at an annual capacity of 35-40 tonnes of heavy metal, using the MIMAS fabrication process (cumulated production of more than 560 tonnes of heavy metal).

MOX is manufactured by mixing plutonium oxide and depleted or natural uranium oxide powders. The whole production line is installed in dry glove boxes. The fabrication plant is divided into four main parts corresponding to different process steps:

- primary blend [Pu/(U+Pu) between 25 wt.% and 30 wt.%];
- secondary blend [Pu/(U+Pu) ≤ 10 wt.%];
- pellet fabrication (pelletising and sintering);
- rod fabrication.

In addition to these four main fabrication steps, the storage areas and laboratories are also included in the criticality prevention procedures.

Due to the important throughput of fissile material, criticality prevention is one of the major concerns for the safe operation of the plant [1]. The prevention of the criticality risk rests on two consecutive pillars:

- determination of the breaking values of operation (“calculations” phase);
- determination of the procedures of operation (“analyses” phase).

Determination of the operational values

This phase of the prevention can be subdivided in two different approaches according to the situation: the “global approach” and the “work unit approach”.

Global approach

During the fabrication process, the fissile material can be found in various geometrical configurations, at various densities and at various moderation ratios. The “global approach” consists in determining the critical radius of a sphere containing the fissile material in homogeneous mixture with a moderator (generally H_2O). The sphere is supposed to be reflected by an outer water layer of 20 centimetres thickness, which is generally conservative from a safety point of view because water acts as a moderator and a reflector for the neutrons leaving the fissile mixture.

It is obvious that this method strongly depends on the design assumptions taken into account. It is appropriate to define a set of limiting values for the most important parameters (density, impurities, enrichment, isotopic composition...) which must not be exceeded during the production process.

In order to simplify the management of the impurities and the moisture presence, we use a factor (called the “metal factor”) which represents the ratio of heavy metal mass to total mass. For example, the “metal factor” for a pure U+Pu oxide is near to 88.19% = 239/(239+32). For a powder, the “metal

factor" must be greater than 85.5%, which corresponds to a blend for which 3% of the mass is due to moisture and impurities. After the sintering operation the "metal factor" must be greater than 87.8%. For the criticality calculation, the 3% will be considered as water.

From the critical radius of the sphere, acceptable limits are deduced by taking into account various safety coefficients. For example, in order to define the mass limit, it is necessary to apply the principle of double contingency, reducing the critical mass to half of its value. Finally, on account of the various uncertainties, a global multiplicative factor of 0.43 is applied to the critical mass obtained by calculation.

Approach by work unit

The "global approach" can be very penalising with respect to the production process or unsuitable for specific installations such as a storage. In this case, a direct calculation of the k_{eff} factor in a three-dimensional approach is preferable.

The three-dimensional calculation model can be simplified in a conservative way (i.e. disregarding the presence of structural materials, like steel). The characteristics of the fissile material and the quantity taken into account in the model must be conservative compared to those allowed in the installation. Calculations are generally performed with a Monte Carlo type code, which simulates the process history of the appearances and disappearances of the neutrons between the generations N and N+1 in the geometrical model.

The calculations are first performed in the normal situation, and then in various accidental situations [introduction of moderator (H_2O), partial loading, volume reduction…]. In each case, the requirement of the safety criterion $k_{eff} + 3\sigma < 0.95$, must be met.

The set of computer codes used for these two approaches must be checked and validated by the authorised inspection and licensing body (AVN) which is recognised by the National Safety Authority. The values obtained with the calculation scheme in its data-processing environment must be in agreement with the values obtained by other international organisations (OCDE/NEA), and also compared to results of experiments.

Determination of the operational procedures

In order to take the calculated limits into account, the operational procedures refer to separate "work units". In each "work unit" a specific type of matter (primary blend, secondary blend, pellet...) is accepted within the associated limits of mass, enrichment, humidity, isotopic composition...

The management of fissile material transfers between "work units" includes an effective control of the masses and characteristics of the various batches within all "work units". With this aim in view, a procedure describes in a specific way the operations to perform and the precautions to take, in order to transfer the fissile material from one "work unit" to another. This procedure describes precisely the five phases of a transfer:

1) The operator of a "work unit" requires from the safeguard responsible an authorisation in order to perform a transfer. This request specifies the quantity and the characteristics of the fissile material to be transferred and is simultaneously transmitted to the person in charge for the receiving "work unit".

2) The operator of the receiving "work unit" gives his agreement for the transfer via the encoding of a specific password.

3) The person in charge of the safeguard checks that the allowed limits for the "work unit" will not be exceeded, gives the authorisation for the transfer and records the quantities of fissile material within both "work units". These two "work units" are now "isolated" from the others (no other transfer is authorised).

4) The transfer is performed.

5) The operator of the receiving "work unit" indicates the end of the transfer via the encoding of his password.

In addition to this, the content of each "work unit" is regularly controlled in order to make sure that no maintenance failures occurred (presence of waste, results of composition analyses…). The presence of a moderator within the work units is also strongly controlled and must be approved by the safety department.

Moreover, the head of the safety department must approve any modification of the manufacturing process as well as any modification of the infrastructure. These are also submitted to the approval of the control organisation. The operational procedures are regularly checked and must be approved by the control organisation as well.

Examples

Result issued from the "global approach"

In the areas operating with primary blend, the following assumptions are taken:

- Pu/(U+Pu) = 35 wt.%, whereas Pu content is generally comprised between 25 wt.% and 30 wt.%.
- bounding isotopic composition of Pu (239/240/241/242): 70 wt.%/18 wt.%/10 wt.%/2 wt.%;
- isotopic composition of U (235/238): 0.72 wt.%/99.28 wt.%;
- density of 5.2, the presence of scraps being taken into account;
- water 3 wt.% ("metal factor" = 85.5%).

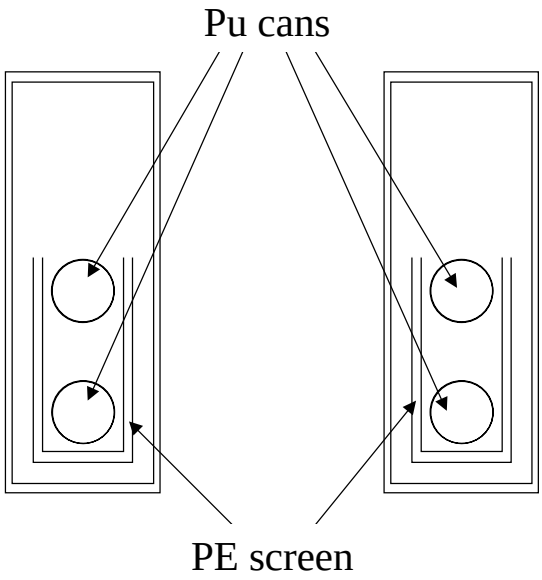
These assumptions give a critical radius of 21.6 cm or a critical volume of 42.2 l or a critical (^{235}U+Pu) mass of 67.8 kg. Safety coefficients (0.43×0.9) are applied and the resulting mass is 26.2 kg. The limit of this "work unit" has been set to 25.9 kg of (^{235}U+Pu).

Explicit geometry for infrastructure modification

A specific study was made in order to evaluate the impact of a shielding [polyethylene (PE) screen round the can] on the coupling of two glove boxes included in the "work unit" hereupon mentioned. We assume here that (2×2) cans of secondary blend are placed in these two glove boxes (see Figure 1).

We performed a k_{eff} factor calculation using a three-dimensional model. The characteristics of the fissile material and the quantity taken into account in the model were taken conservatively compared

Figure 1. 2 × 2 cans placed in two separate glove boxes



to those allowed in the installation. That is the reason why we decided to simulate an infinite-height can. The isotopic composition of Pu (239/240/241/242) remains the same. The powder moisture was set to 3 wt.% and its density to 5. The ambient air was considered dry.

Under these conditions the k_{eff} factor is near to 0.9. This value meets the requirement on k_{eff} but is considered as "high". Nevertheless, since the assumption to consider infinite-height cans is very conservative and that four cans in the glove boxes leads to exceed the limit of mass of the work unit, we finally concluded that the insertion of a PE shielding was acceptable. Moreover the introduction of PE shielding, while decreasing neutron dose to workers, also decreases the excess of reactivity brought by the possible presence of an operator near a glove box, since the neutrons are already thermalised in the PE material.

This study is a typical example of the limitations associated with the "global approach", which can be overcome when considering a more detailed approach, based on a 3-D model. It also shows that the "global approach" does not help very much when addressing the issue of interacting separate regions.

Result obtained from the "work unit approach"

For practical reasons, a temporary storage area for the rods has been built between the testing area and the storage place. This area is designed to receive more than 1 000 rods, and is enclosed in one "work unit".

To meet the safety requirements in this area, a three-dimensional approach was chosen. The following assumptions for the storage have been made:

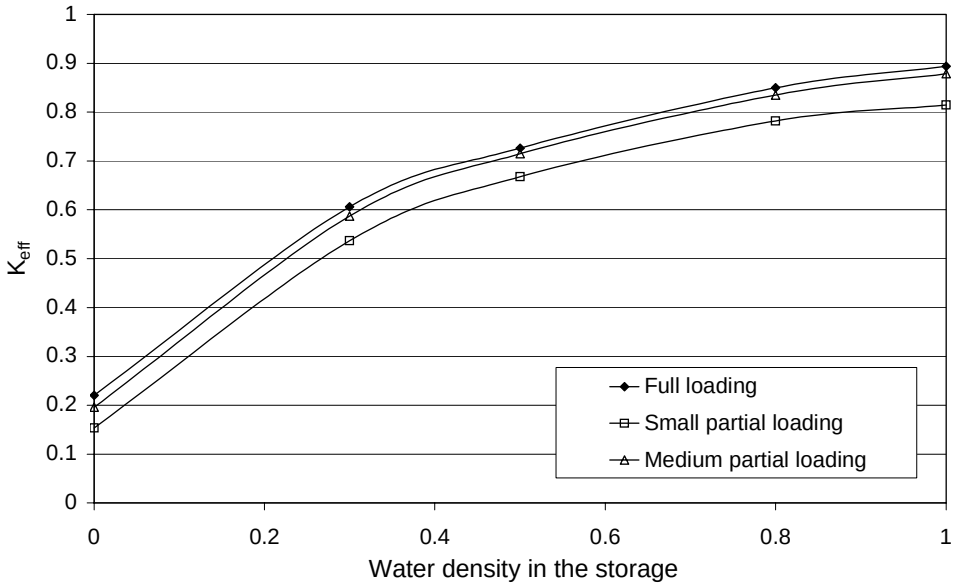
- infinite length rods;
- Pu/(U+Pu) = 12 wt.%, whereas the normal Pu content is below 10 wt.%;
- density of 11;

- isotopic composition of Pu (239/240/241/242) 70 wt.%/18 wt.%/10 wt.%/2 wt.%;
- isotopic composition of U (235/238) 0.72 wt.%/99.28 wt.%.

Due to the possible intrusion of water in the storage, we simulated water of different densities (from 0 to 1) between the rods. Structural materials such as steel were not taken into account.

The results show that within a full storage loading, a complete flooding of the rods leads to an under-moderated situation. It was thus necessary to complete the calculations with simulations of partial loading (Figure 2), which proved to be less penalising. In all cases, the k_{eff} remained below 0.9.

Figure 2. k_{eff} plot as a function of water density in a storage, fully and partly loaded with MOX rods



Conclusion

As it is seen, the choice of the starting assumptions is very important. Very conservative and penalising approaches are frequently used. Moreover, the cross-sections used by the various codes are probably conservative with respect to the fissile material powder [2].

The daily operating of a MOX plant points out two major issues:

- The use of improved cross-section libraries would lead to more realistic and less constraining limits with respect to the manufacturing process, while maintaining the required safety level.
- A better knowledge and the development of a practical tool for reactivity follow-up of the fissile material, according to moderation (moisture), density, heterogeneity (scrap), and to time-dependent mixing of the heavy nuclides occurring in a nuclear fuel [4].

These are among the reasons why effort should be devoted to develop critical experiments involving such fissile material [3] or to experiments simulating powders with the use of tight lattice of MOX rods.

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US MOX FUEL FABRICATION – DESIGN, INDUSTRY EXPERIENCE, CHALLENGES AND ASSOCIATED CRITICALITY SAFETY ISSUES

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AREVA and Duke Energy

This paper was unavailable at the time of publication. The full presentation is, however, available on the CD-ROM included with these proceedings.

CRITICALITY SAFETY ISSUES ASSOCIATED WITH MOX FUELS

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Abstract

The MELOX criticality safety analysis has been successively validated by the French Safety Authority, taking particular of account the operating feedback. An important issue in this analysis was to guarantee appropriate safety margins in order to balance some uncertainty on criticality code validation in the area of low-moderated MOX powder material. This paper presents the MELOX criticality safety margins analysis and the IRSN assessment. This analysis is mainly based on the identification and justification of intrinsic parameters, which, in addition to "operating" parameters, guarantee appropriate safety margins. Those criticality safety margins could be reduced by a better knowledge of the criticality calculation code uncertainties and with the agreement of the French Safety Authority. Therefore a work programme on the codes' validation in the area of the low-moderated MOX powder should be examined.

The MELOX facility produces MOX fuel for pressurised or boiling water reactors.

Key points on the criticality safety of MOX powders

For units dealing with UO_2-PuO_2 powders, the fissile media chosen by the operator are UO_2-PuO_2 with a PuO_2 weight content of 30% for the primary blend mixture and of 12.5% for the final blend mixture.

The isotopic composition in mass of plutonium is 71% in ^{239}Pu, 17% in ^{240}Pu, 11% in ^{241}Pu and 1% in ^{242}Pu; that of the uranium is 1.2% in ^{235}U and 98.8% in ^{238}U.

The control of criticality safety is based on the limitation of the mass and moderation. Thus, concerning the limitation of the moderation, the equivalent content of water of the powders covers the moisture resumption of the powders as well as the presence of additives (lubricants and pore-formers). In normal situations, the water content is equal to 3%. This value corresponds to a moisture level of 1% and a presence of additives equivalent to 2% of water. In abnormal situations, the water content retained in the criticality studies is equal to 5% (a more important moisture resumption or a double batch of additives).

Moreover, some abnormal situations take into account a leakage of oil in the fissile material. In this case, the calculation is performed with a heterogeneous distribution of oil in the fissile medium.

Initially, the criticality safety studies were performed with a maximum density of the powders of 3.5 g/cc.

As for the multiplication factor k_{eff}, the acceptability criterion (taking into account all uncertainties of the configurations modelled) is equal to 0.95 for normal situations and to 0.975 for some abnormal situations. Those criteria are usually retained in France. However, to compensate for the absence of data on the degree of confidence which can be granted to the computer codes for the low-moderated UO_2-PuO_2 media, pessimistic assumptions, detailed further on, were taken into account by the operator.

Examination of the final safety report

The final safety report of the MELOX plant was examined by the standing group of experts on 12 April 2000. The assessment of the safety report was performed and presented by IRSN (now IPSN).

The operator reported that the density of the powders was higher than expected (3.5 g/cc), which led to a review of the criticality safety studies with new assumptions concerning the density in accordance with the operating feedback. Indeed, a value varying from 3.5 g/cc to 5.5 g/cc according to the work unit (for example, 5.5 g/cc for primary blend mixture after crushing) was considered.

Using more realistic models, he proved that acceptability criteria for k_{eff} were still respected.

In its assessment, IRSN recommended that the operator show that safety margins not taken into account in the criticality safety studies are sufficient to compensate for the lack of experimental data (benchmarks) and consequently for the degree of confidence in the code validations in corresponding situations.

During the meeting of the standing group of experts, the operator presented an argumentation on the safety margins available for the units having the primary blend mixture as the fissile medium of reference, which presents the weakest margins. The operator thus presented safety margins relating to the quantities of hydrogenated fluid really present, to the masses really implemented, to the localisation of the hydrogenated fluids. Lastly, the operator highlighted that the studies retain a ^{240}Pu content of 17% whereas that really implemented is about 25%.

This approach having been considered to be acceptable by the standing group of experts, the French Safety Authority required of the operator a study of the safety margins available for the units implementing MOX powders or pellets with a limitation of the moderation and for which the criticality safety studies retain a maximum value of the multiplication factor of 0.975 to justify the sub-criticality of the unit in abnormal situations.

In addition, the standing group of experts considered that an additional reduction of the safety margins for the units implementing MOX powders or pellets with a control of moderation shall require a validation of the criticality codes used.

Study of the safety margins

Following the standing group, the operator transmitted an analysis of the safety margins of the units controlled by the mass associated with a control by the moderation. The operator listed different types of margins, some of which are detailed further on. IRSN classified these margins into the two following categories:

- those related to “intrinsic” characteristics, based mainly on design characteristics, not likely to evolve except by modifying the equipment or the glove boxes; among the margins presented by the operator, IRSN classified in this category the following ones:
  - only the oil capacities able to reach the equipments containing fissile materials are considered;
  - the distribution of the fissile material in the various modules constituting a unit although the criticality safety analysis considers the gathering of the entire fissile material authorised in a unit;
  - the density or the plutonium content of the fissile medium of reference, retained to the maximum value in a unit, but which can be lower for one of the modules of the unit concerned.
- those related to operating parameters and likely to evolve according to the activity of the plant; among the margins presented by the operator, IRSN classified in this category the following ones:
  - fissile material mass really present in the unit;
  - maximum content of hydrogenated products in the fissile material.

Another example for an “intrinsic” characteristic is the mass limit retained in the criticality safety analysis which is the same for every unit. But in some units, the volume of equipments is such that the mass of fissile materials contained cannot reach the mass limit.

When the mass is not limited by vessel capacity, the nominal mass is considered as an operating parameter. Today, the nominal mass provides a margin compared to the criticality mass limit. But this parameter is likely to evolve according to the activity of the plant. In actuality, this margin cannot be considered by IRSN as a sufficiently strong guarantee.

Illustration of the IRSN procedure for the assessment

The work unit concerned by this example is used for mixing PuO_2, UO_2 and "chamotte" (recycled materials) in order to obtain the primary blend mixture. Those products are collected by specific equipments and poured in a jar, which is permanently located in the work unit. The content of the jar is then transferred to a mixer, where additives are inserted. Finally, the primary blend mixture is poured in a new jar, which is transferred to the next work unit.

For this work unit, the fissile medium considered in the safety analysis is MOX powders, the density of which is 4.1. The PuO_2 content is inferior to 30%. The water content of the powders is 3%, which corresponds to a moisture level of 1% and a presence of additives equivalent to 2% of water.

The safety study takes into account the leakage of the maximum quantity of oil likely to be inserted in the entire quantity of fissile material authorised in the work unit, in case of seism. This assumption leads to the following case: 151 kg of fissile material, in which 5.3 litres of water are inserted in a heterogeneous manner. Given that the reactivity of 160 kg of fissile material in which 5.3 litres of water are inserted is 0.966, the operator analysed the safety margins for this work unit and presented two types of margins:

- The work unit is actually divided into two clearly separate modules. Thus, the maximum quantity of oil likely to leak in one of this module is 4.4 litres.

- The water content of the fissile medium is lower than assumed. Indeed, at this step of the process the quantity of additives inserted in the powders is 0.53%, which in addition to 1% due to the moisture resumption, leads to an amount of 1.53% water content.

The first margin is classified by IRSN as an "intrinsic" margin, as the capacity of oil tanks is fixed and not likely to evolve. Considering this margin reduces the reactivity by more than $1\,600 \times 10^{-5}$.

In addition to this first margin, taking into account the second one would result in a final k_{eff} of 0.935. Nevertheless, classifying this second margin as "intrinsic" would lead to a strict limitation on the quantity of additives inserted, whereas a small variation is acceptable.

Consequently, IRSN estimates the safety for this work unit is ensured all the more as the isotopic composition in mass of plutonium taken into account is penalising.

Conclusion of the assessment

IRSN estimates that the values of multiplication factors obtained with assumptions more realistic than those selected initially are acceptable taking into account the degree of the validation of criticality codes used. Indeed, considering the "intrinsic" parameters for abnormal situations, the values of the multiplication factors are lower than 0.95. However, IRSN estimates that operating parameters provide a degree of conservatism for the calculation assumptions and concludes that this degree of conservatism should remain the same in case of evolution of one of operating parameters.

This study of the safety margins confirms the former conclusion of the standing group of experts stating that the current dimensioning of the plant has reached the maximum degree of optimisation and that no reduction of the safety margins is possible without a better knowledge of the area of low-moderated MOX fuel on the validation of the codes used.

Conclusion

It may be concluded that a work programme on the code validation in the area of low-moderated MOX powder would be of interest, not only for scientific purposes, but also for future industrial purposes. Thus, one can imagine that, if the global uncertainties were better evaluated in the area of low-moderated MOX fuel, the criticality safety constraints on the MELOX plant could be released.

Even if a need for releasing these constraints is not identified today, it could be useful in the future of the MELOX plant or for a future MOX plant.

LICENSING ISSUES ASSOCIATED WITH PuO_2 AND MIXED OXIDE POWDER PROCESSES

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Abstract

The unique regulatory aspects of licensing processes involving low-moderated PuO_2 and mixed-oxide (MOX) powders are described. This is based on the approach taken in the proposed MOX Fuel Fabrication Facility (MFFF) at the Savannah River Site, as well as the La Hague and MELOX facilities in France. Anticipated normal and upset conditions for these processes are compared to existing low-moderated benchmark experiments. To assist in comparing benchmarks to anticipated applications, the TSUNAMI sequences of the SCALE 5 code package were used. These sensitivity/uncertainty codes quantitatively measure the sensitivity of the system k-effective to the underlying cross-section data, and compute an integral correlation coefficient for each experiment-application pair. The use of this code in addressing these issues is described.

Among the unique regulatory challenges for an MFFF are: the complexities of plutonium chemistry and isotopic composition, control of powder density for criticality control, and control of isotopic composition and homogeneity during the oxide blending process. The relative risks of the various process stages and anticipated conditions are discussed.

Once the highest-risk conditions were defined, the TSUNAMI codes were then used to compare them to the applicant's proposed benchmarks. The results of these codes were also confirmed using direct perturbation of system parameters. The application of these codes to address variations in powder density, moderator content and isotopic nature of the plutonium is described. The application to determining a bounding fissile medium to represent miscellaneous plutonium compounds is described. The application of PuO_2-polystyrene and Pu-metal benchmarks to low-moderated PuO_2 applications is discussed. Areas where additional critical benchmarks would be helpful are then discussed.

MFFF processes and regulatory issues

The introduction of plutonium into a historically uranium-based fuel raises several challenges in the area of nuclear criticality safety (NCS). The underlying physics concepts and basic phenomena are no different from those encountered in processes with low- or high-enriched uranium. However, there are important differences that need to be considered in the design of an MFFF.

The use of MOX with a plutonium content in the range of 2-6 wt.% Pu/(U+Pu) can result in fuel that is substantially more reactive than typical (i.e. < 5 wt.% ^{235}U) US light water reactor fuel. ^{239}Pu tends to have a much smaller minimum critical mass than ^{235}U. This is true even for plutonium nitrate vs. uranyl nitrate, despite a higher nitrogen-to-fissile ratio (Table 1). The larger volume and dimensional limits for plutonium nitrate indicate that optimal moderation for Pu tends to occur at a much higher H/X than for U – or equivalently, that a ^{239}Pu system exhibits a harder neutron spectrum than a ^{235}U at the same H/X. This becomes important in validation because, at moderation levels anticipated in MOX powder processes, there can be a strong component in the intermediate and fast neutron energy ranges.

Table 1. Single-parameter limits

From ANSI/ANS-8.1-1983

	LEU (< 5 wt.% ^{235}U)	**$^{235}UO_2(NO_3)_2$**	**$^{239}Pu(NO_3)_4$**
Mass	3.3 kg (7.3 lb)	0.78 (1.7 lb)	0.48 (1.1 lb)
Cyl. diam.	42.7 cm (16.8 in)	14.4 cm (5.7 in)	15.4 cm (6.1 in)
Slab thick.	23.4 cm (9.2 in)	4.9 (1.9 in)	5.5 (2.2 in)
Volume	111 l (29.3 gal)	6.2 l (1.6 gal)	7.3 (1.9 gal)

A second unique aspect is the complexity of the isotopic mixture. In a typical uranium-based fuel cycle facility, only the ^{235}U enrichment need be specified to fully characterise the fissile material. In an MFFF, both the plutonium and uranium isotopics must be specified, in addition to the ratio of plutonium to uranium. Incoming feed to the proposed US MFFF would be limited to 90-95 wt.% ^{239}Pu, 5-9 wt.% ^{240}Pu and < 1 wt.% ^{241}Pu. Depleted uranium would be limited to < 0.25 wt.% ^{235}U. PuO_2 would then be down-blended to MOX powder in two stages, first in a master blend with 20 wt.% Pu content and then in a final blend with 2-6 wt.% Pu content. The master blend would be analysed at 22 wt.% Pu content and the final blend at 6.3 wt.% Pu content. The applicant has proposed bounding assumptions for the isotopics of the incoming feed material. MOX isotopics would be controlled in the powder blend operation. A specified quantity of depleted UO_2 powder would be metered into the large geometry blend tank and then combined with a specified quantity of PuO_2 powder to produce MOX powder. The NRC staff considers this one of the key processes with regard to criticality safety in the entire MFFF, because the resultant isotopic mix is credited for the safety of all downstream processes. It is particularly important to obtain a highly uniform mix to avoid having “hot spots” with greater than allowed Pu content. This would be done by controlling the amount of moderator (both for reactivity and to prevent clumping), by mechanical stirring and by subsequent homogenisation of the powder. It is anticipated that in the blend tank there would be a significant contribution from the intermediate neutron energy range.

A third unique aspect is caused by the complexity of plutonium chemistry. Plutonium has up to six positive valence states and can form a large number of different chemical compounds. The valence state is dependent on process conditions (e.g. acidity, presence of oxidising/reducing agents, temperature). Though the electronic configuration has essentially no effect on the nuclear properties of plutonium, it can affect both the process flow and system reactivity. The efficiency of the purification cycle in

aqueous polishing depends strongly on the Pu valence state; Pu(IV) forms much stronger complexes with tributyl phosphate(TBP) than Pu(III) does. The presence of Pu in the Pu(IV) oxidation state is required in the extraction, scrubbing and stripping processes; the correct operation of these processes ensures there is very little plutonium left in the solution raffinate. In the stripping column, the plutonium is reduced to Pu(III) using hydroxylamine nitrate (HAN) and stripped from the solvent. The plutonium must then be oxidised back to Pu(IV) using nitrous oxide before being contacted with oxalic acid and precipitated as plutonium oxalate. In terms of reactivity, the valence state determines the chemical form [e.g. $Pu(III)(NO_3)_3$ versus the slightly less reactive $Pu(IV)(NO_3)_4$]. Because of this complex chemistry, a large variety of chemical forms must be considered in the criticality evaluation of the MFFF [PuO_2, PuO_2F_2, $Pu(NO_3)_3$, $Pu(NO_3)_4$ and $Pu(C_2O_4)_2$]. In the US MFFF design, PuO_2F_2 has been proposed as a bounding fissile medium; the validation of this material with existing benchmarks is treated below.

A fourth unique aspect is the important role that density plays in the criticality safety basis. The proposed US MFFF contains processes that credit several different values of PuO_2 and MOX density, ranging from 3.5-7 g/cm^3 (218.8-437.6 lb/ft^3), as well as full theoretical density of 11.46 g/cm^3 (716.4 lb/ft^3) for incoming powder and sintered material. A PuO_2 density of 7 g/cm^3 (437.6 lb/ft^3) is recognised as being very conservative even for tapped powder. However, the method of preparation, process conditions and any mechanical handling can all affect the density. Measurements have been conducted on PuO_2 powder prepared in several different chemical processes, resulting in densities of 1-2.3 g/cm^3 (62.5-143.8 lb/ft^3) [1]. The lowest reported densities were those for PuO_2 produced from oxalic precipitation, as in the proposed US MFFF. The situation regarding the density of MOX powder is more indefinite. When powders with two different densities are combined, the total volume may be less than the sum of their individual volumes (i.e. powder blending may be non-volume additive). There appears to be little available data in the literature for the density of MOX powders with different isotopic mixes. The measurement of actual in-process powder density is therefore very important whenever less than theoretical powder density is credited for criticality safety. The effect of density on applicability of existing benchmarks is treated below.

Perhaps the most significant unique aspect associated with processing plutonium occurs in the area of computer code validation. This is not due to any inherent complexities associated with plutonium, but rather with benchmark experiments available in the literature. With regard to validation, the Code of Federal Regulations (CFR) includes the following requirement for criticality safety at NRC-regulated fuel facilities [10 CFR 70.61(d)]:

> "…the risk of nuclear criticality accidents must be limited by assuring that under normal and credible abnormal conditions, all nuclear processes are subcritical, *including use of an approved margin of subcriticality for safety*…"

The determination of an appropriate "margin of subcriticality" for processes associated with MOX fuel manufacturing is not a trivial exercise. Traditionally, higher-enriched fuel facilities have been licensed with larger margins in k_{eff} than low-enriched facilities. Determination of an appropriate margin must take into account several considerations, including inherent risk of the various processes, the sensitivity of k_{eff} to changes in the underlying parameters, and the quality (i.e. number and similarity) of available critical benchmarks. This paper looks at each of these three considerations and applies them to operations in the proposed US MFFF that is currently under NRC review.

MFFF processes and risk perspective

In approaching criticality code validation, the proposed US MFFF divides operations into five different areas of applicability (AOA), as follows:

1. Plutonium nitrate solutions.

2. Mixed-oxide fuel pellets, rods and assemblies.

3. Plutonium oxide powder.

4. Mixed-oxide powder.

5. Miscellaneous plutonium compounds.

A separate validation was performed, documented and reviewed for each of the five AOAs above. The design of the proposed MFFF was based on Cogema's La Hague (aqueous polishing) and MELOX (MOX fuel fabrication process) facilities, with the exception that the US plant would use weapons-grade plutonium ($\leq$ 96 wt.% ^{239}Pu) as feed material instead of reactor-grade plutonium. This simplifies the plutonium isotopic vector, but also increases system reactivity and decreases any margin that may result from the fresh-fuel assumption for reactor-grade plutonium.

Based on the history of criticality accidents [2], the highest risk is expected to occur in the aqueous polishing process [covered by AOA(1) and AOA(5)]. The lowest risk is expected to be in the areas handling finished MOX fuel [covered by AOA(2)] due to the comparatively low Pu assay and the fixed material form. This qualitative comparison is true in terms of the unmitigated risk – that is, based solely on the form of material and its potential to reach a critical state. Solutions that have small critical masses and can readily conform to an unfavourable geometry have relatively high unmitigated risk. MOX fuel, with a large critical mass and stable form, has relatively low unmitigated risk. Intermediate in risk between these are the PuO_2 and MOX powders, which are dry under normal conditions (with an assumed bounding moisture content of 1 wt.% H_2O), vary in assay from high to low Pu, and are conformable to an unfavourable geometry container.

The picture is somewhat different when mitigated risk is considered – the design ensures that plutonium nitrate solutions are passively controlled, relying mainly on fixed neutron absorbers and favourable geometry vessels. Powder operations, conversely, rely mainly on mass and moderation control, and are much more hands-on operations. Therefore, these operations are expected to be much more administratively controlled than aqueous polishing. When all this is considered, PuO_2 and MOX powder processing operations may be the areas of highest overall mitigated risk.

These processes are also especially important due to their unique nature, reliance on isotopic blending and the higher uncertainties associated with bulk powder systems (uncertainties in density, nuclear data in the intermediate energy range and lack of benchmarks).

Parametric evaluation of MFFF processes

Ref. [3] contains a list of steps for validating calculational methods, and in particular for determining the method's AOA. The first step is the characterisation of the key parameters associated with normal and credible abnormal conditions.

The parameters traditionally associated with validation include the moderator-to-fissile ratio (e.g. H/X for homogeneous mixtures), isotopic character [e.g. ^{239}Pu assay, ^{235}U enrichment, Pu/(U+Pu) content], and energy parameters [e.g. thermal fission fraction, average energy group, energy of average lethargy causing fission (EALF)]. These continuous parameters are convenient for trending, and are among those thought to have the largest potential effect on system k_{eff}, and thus, on bias. The parameters to which k_{eff} has the highest sensitivity can be supposed to be the most likely to produce a trend in the bias (i.e. traditional trending parameters).

Anticipated design applications for powder systems include spheres (when relying on mass control) and other simple geometrical configurations of uniform PuO_2- or MOX-water systems (with 100, 22 and 6.3 wt.% Pu content). The powder may be assumed to be at full theoretical density (and thus dry) or at some reduced density. The system may have various dimensions depending on the size of batches, and may have various reflector conditions (from one-inch tight-fitting nominal reflection to full water reflection, or reflection by materials such as concrete, steel or depleted uranium). The dimensions and reflector conditions primarily affect the neutron energy spectrum.

Among the most important parameters are those characterising the fissile medium. PuO_2 and MOX powders will be assumed to have a moisture content of 1 wt.% H_2O (corresponding to an H/X of ~0.3) under normal conditions. Some water moderation should always be assumed, due to the hygroscopic nature of this material. The energy spectra of these systems are expected to vary widely, depending on the total mass, dimensions and reflection. Mass will vary from effectively infinite for the blend tank down to a few kilograms for the incoming PuO_2 containers. The spectra of individual design applications and benchmarks were characterised by specifying the value of EALF. However, it should be recognised that the spectrum of a low-moderated, reflected system typically covers the entire range from very fast to thermal energies, as shown in Figure 1. Systems with different values of EALF can still have similar neutron spectra. Experiment NSE55T5-07 has an EALF = 43.5 eV (5.15×10^{-18} ft-lb), while PMF016-01 has an EALF = 11.7 keV (1.38×10^{-15} ft-lb), even though there is considerable overlap in much of the spectra.

Figure 1. Fission spectra for typical Pu-metal and MOX-polystyrene benchmark

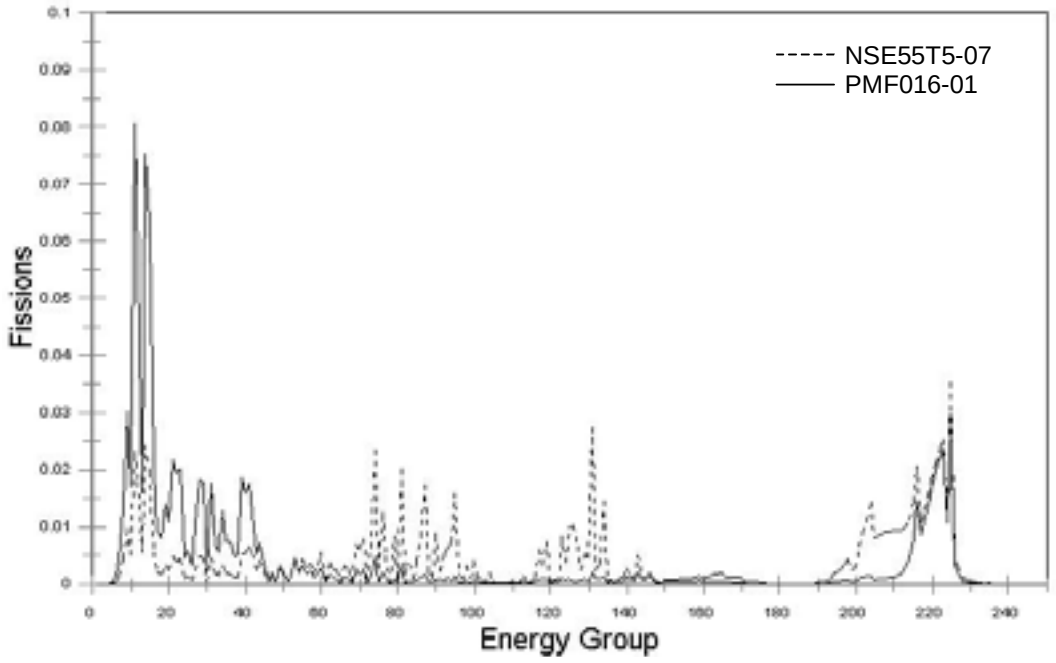


Table 2 below shows some of the more important parameters associated with MFFF processes under both normal and credible abnormal conditions. Under abnormal conditions, a broader range of parameters may be encountered, such as upsets involving the moderation of dry powders, which will account for the highest value of H/X.

Table 2. Parameters associated with powder handling operations

	PuO_2 powder – AOA(3)	**MOX powder – AOA(4)**
Material form	PuO_2-water mixture	MOX-water mixture
Geometric form	Parallelepipeds Arrays of cylinders Spheres	Parallelepipeds Spheres
Isotopics	96 wt.% ^{239}Pu 0.3 wt.% ^{235}U 100 wt% Pu/(U+Pu)	96 wt.% ^{239}Pu 0.3 wt.% ^{235}U 6.3, 22 wt.% Pu/(U+Pu)
Density	3.5-11.46 g/cm^3 (218.8-716.4 lb/ft^3)	5.5 g/cm^3 (343.8 lb/ft^3)
H/X	0-16.5	1.1-1.6
EALF	3 eV-266 keV (3.55×10^{-19}-3.14×10^{-14} ft-lb)	0.28 eV-850 eV (3.31×10^{-20}-1.00×10^{-16} ft-lb)
Reflector	Water, Cd, concrete	Water

A challenge is posed by the fact that there are few, if any, experiments similar to anticipated design applications for AOA(3) and AOA(4). Applications will consist largely of homogeneous masses of uncontained, slightly moderated PuO_2- and MOX-water mixtures. Experiments drawn from the *International Handbook of Evaluated Criticality Safety Benchmark Experiments* [4] were evaluated for their suitability for use in validating PuO_2 and MOX powder calculations [5,6]. The experiments can be lumped into two broad categories: (1) Pu-metal benchmarks, with H/Pu = 0, and (2) low-moderated PuO_2- or MOX-polystyrene compacts. The characteristics of these benchmarks – geometric shape, density, physical form, chemical composition, moderation and isotopic character –can vary significantly from those of anticipated design applications. Applying the traditional method of reliance on qualitative comparison of benchmarks to design applications does not yield enough applicable benchmarks to enable calculation without the use of a statistical margin of up to several per cent in k_{eff}.

It was therefore necessary to develop other means to determine whether benchmarks that do not appear applicable on the surface do indeed have sufficiently similar neutron physics to be useful for validation. This was done by developing a set of "screening criteria" that can be applied to a set of candidate benchmark experiments. The first decision faced by an analyst is which of the system parameters (including those in Table 2) are the most likely to affect the bias. Table 2.3 of Ref. [3] contains parametric criteria for determining the AOA, though this reference states that these are based on the consensus of a number of criticality experts and are therefore conservative.

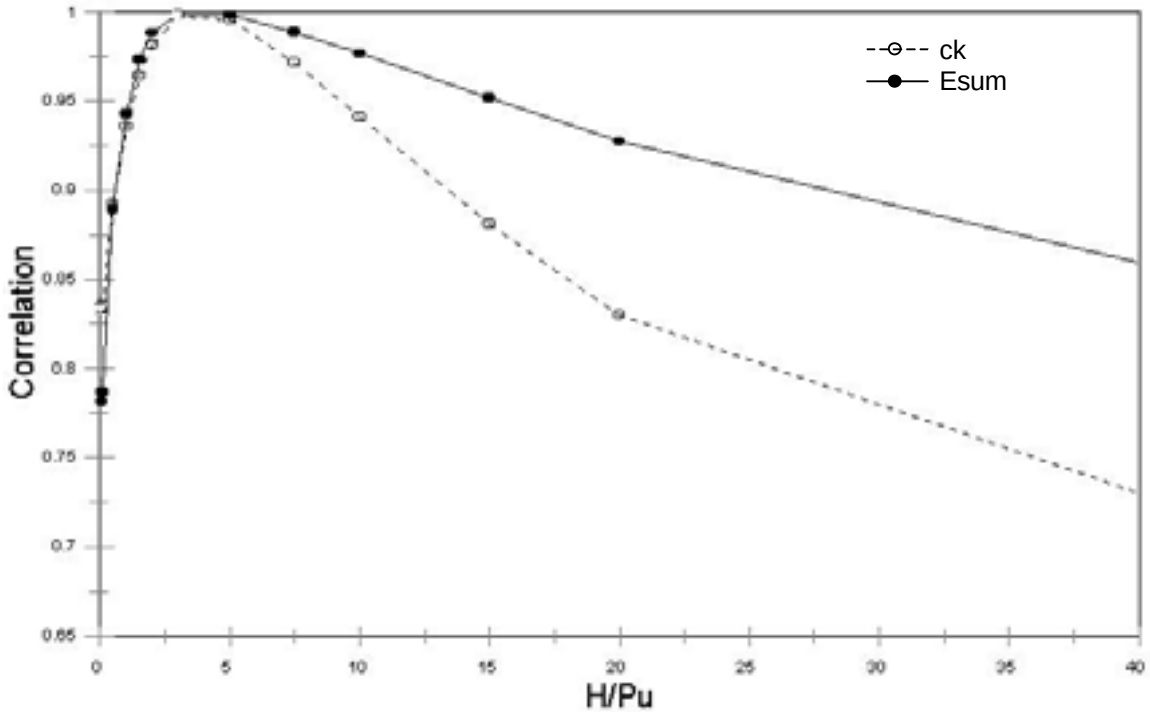
Refs. [5] and [6] used Oak Ridge's TSUNAMI sensitivity/uncertainty codes to compare a set of benchmark experiments directly to hypothetical design applications (uniform spheres of PuO_2- and MOX-water mixtures with various reflector conditions). This code provides powerful insights into the neutronic behaviour of fissile systems, but is still under development. One caveat is that care must be taken to select an appropriate cut-off value of c_k. The NRC staff experience has been that cases with a c_k/0.95 show a high degree of similarity in terms of traditional comparison, while some differences were noted for cases with c_k between 0.90 and 0.95. A working definition has evolved that agreement between systems with $c_k > 0.90$ is "good" and agreement between systems with $c_k > 0.8$ is "marginal."

In the present study, the TSUNAMI code was used to investigate the relative importance of different system parameters to aid in the development of a set of screening criteria, rather than to directly compare benchmarks to design applications. Once a hypothetical design has been defined, the key parameters can be varied to investigate the effect of parametric changes on the sensitivity and correlation coefficients. This results in a curve showing the value of a sensitivity or correlation coefficient as a function of the parameter. Implicit in this approach is the idea that if k_{eff} is relatively insensitive to a given parameter, then the correlation coefficient will be close to unity and will not exhibit much variation. If k_{eff} is highly sensitive to a given parameter, then the correlation coefficient will exhibit a strong dependency on this parameter and will drop off rapidly. In addition to providing information on the relative importance of various parameters, this also enables an analyst to develop a set of screening criteria without reference to specific benchmarks. Generally the more sensitive k_{eff} is to a given parameter, the narrower the screening range, and the higher the desired confidence (i.e. the higher the c_k cut-off), the narrower the range. The results of several sensitivity studies showing the effect of parameter variations on c_k are shown in the following sections.

Plutonium powder systems

Figure 2 shows the result of a sensitivity study in which a uniform PuO_2-water sphere was modelled with varying levels of moderator, so as to determine an appropriate screening criterion in H/X. Early investigations showed that the material properties (isotopic composition, moderation, absorbers) had a much more significant effect on c_k than did system geometry. Therefore, the comparisons were done on spherical models using the TSUNAMI-1D code, which relies on the XSDRNPM one-dimensional discrete ordinates code, instead of explicitly modelling a realistic process. Uniform spheres of the fissile medium were used in most cases, although cylinders or slabs were also modelled and showed very similar results.

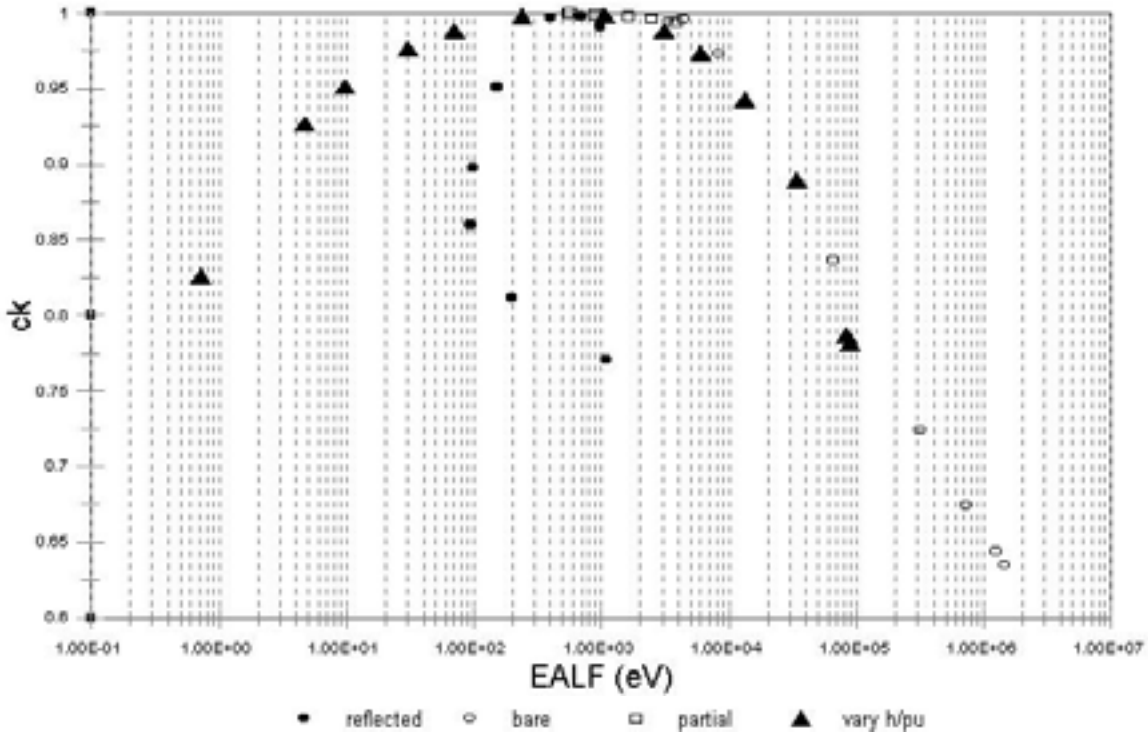
Figure 2. C_k dependence on H/Pu ratio (H/Pu = 3.79, reference case)



The reference case chosen consisted of a fully-reflected PuO_2-water sphere with H/Pu = 3.79 and EALF = 544 eV (6.43×10^{-17} ft-lb). These values were chosen because this was near the middle of the parametric range defining AOA(3) at an earlier stage in the proposed US MFFF design. The range over which c_k exceeded 0.9 was $0.9 \leq$ H/Pu ≤ 13.5. The range over which c_k exceeded 0.8 was $0.6 \leq$ H/Pu ≤ 28. The strong dependence of c_k on H/Pu is not unexpected, because most of the sensitivity in the PuO_2-water cases is consistently due to ^{239}Pu and ^{1}H.

Because there is a strong correlation between H/Pu and EALF, additional sensitivity studies were performed with spheres having different diameters and reflection conditions, but with H/Pu = 3.79. EALF for the hypothetical spherical cases ranged from 89 keV (1.05×10^{-14} ft-lb, at H/Pu = 0.05) down to 0.72 eV (8.51×10^{-20} ft-lb, at H/Pu = 50). The value of c_k was then graphed as a function of EALF, as shown in Figure 3. Also included on this graph are the results of the above study, in which H/Pu was varied from 0.05 to 50 with full water reflection.

Figure 3. C_k as a function of EALF [EALF = 544 eV (6.43×10^{-17} ft-lb), reference case]



The similarity between the curves when H/Pu was varied and when the diameter of the sphere was varied indicates that most of the strong dependence of c_k on H/Pu (in Figure 2) is due to changes produced in the neutron energy spectrum. Using the bounding curves, $c_k > 0.9$ for systems with EALF from ~2 eV-25 keV (2.36×10^{-19} – 2.96×10^{-15} ft-lb); $c_k > 0.8$ for EALF from ~0.4 eV-100 keV (4.73×10^{-20} – 1.18×10^{-14} ft-lb). This is qualitatively consistent with the results of Ref. [3], in which the neutron energy screening criteria require only that EALF be in the same energy range (i.e. thermal, intermediate or fast). This is perhaps explained by the fact that even systems with very different EALF can have neutron spectra with considerable overlap over much of the range from thermal to fast neutron energies.

Similar investigations were also undertaken with regard to other system parameters, including powder density, geometry and plutonium isotopics (^{240}Pu assay). These studies showed that there was relatively little effect on c_k from any of these parameters, so that they may be regarded as of secondary importance. The theoretical density of powder in the PuO_2-water system was gradually decreased from 11.46 to 3 g/cm^3 (716.4 to 187.5 lb/ft^3), while maintaining H/Pu = 3.79 (thus increasing the effective void space). This had a very small effect on the correlation coefficient, which dropped to ~0.97 for a system at full theoretical density compared to one at 3 g/cm^3 (187.5 lb/ft^3).

The effect of system geometry was similarly small. Because the geometric size and shape can affect the value of EALF, spheres, cylinders and slabs of varying sizes were modelled and compared to the reference (spherical) case. When c_k was graphed as a function of EALF, the curves corresponding to spheres, cylinders and slabs were almost coincident. This indicates that the material properties are of primary importance to benchmark applicability, and that the geometric form is much less important. Size appears to be significant only in that it affects the neutron energy spectrum.

The effect of ^{240}Pu assay was similarly found to be of secondary importance. The weight per cent of ^{240}Pu in the plutonium models was varied from 2 wt.% to 30 wt.% (MOX fuel will be analysed at 4 wt.% ^{240}Pu). A two-isotope ^{239}Pu-^{240}Pu mixture is assumed. Even at a ^{240}Pu content of 30 wt.%, the value of c_k when compared to a nominal 4 wt.% reference case was > 0.97. This was not unexpected, as the sensitivity of these systems to ^{240}Pu accounted for only a few per cent of the total sensitivity to the fissile mixture. Direct perturbation of number densities was employed as a spot check on the sensitivity calculations.

One application concerned miscellaneous Pu compounds present during the aqueous polishing process in the proposed US MFFF. Various compounds [including PuO_2, PuO_2F_2, $Pu(NO_3)_4$ and $Pu(C_2O_4)_2$] are to be modelled as PuO_2F_2. The optimum nature of PuO_2F_2 over the range of interest in H/Pu can be confirmed by a traditional sensitivity calculation. However, none of the benchmarks proposed in Refs. [5] or [6] contained PuO_2F_2 as the fissile medium. Curves provided in the validation report showed similar EALF values for subcritical (k_{eff} = 0.93), fully reflected cylinders containing PuO_2 to those containing PuO_2F_2, over a broad range in H/Pu of 0.3-30. However, the difference in EALF values gradually increased at low H/Pu. This raised a two-fold question: (1) whether a comparison of the EALF values is sufficient to demonstrate that two cases are closely related; and (2) if so, what difference in energy represents when two cases are not closely related. To test this methodology, the NRC staff ran several cases (at H/Pu = 0.3, 1, 3, 10 and 30) and, at each H/Pu, computed the c_k value for the PuO_2 system vs. the PuO_2F_2 system. The results showed that, across the range of H/Pu = 0.3-30, there was a very high degree of correlation ($c_k \exists 0.98$) between the PuO_2 and PuO_2F_2 systems. This can be explained by the low value of ^{16}O and ^{19}F sensitivity coefficients as compared to the total mixture sensitivity.

Another area in which TSUNAMI proved useful was the validation of calculations with strong neutron absorbers (such as borated steel, borated concrete and Cd). Frequently, available benchmarks did not contain these materials, so decisions had to be made with regard to when design applications that had strong absorbers were applicable to benchmarks that did not. Traditionally, benchmarks were required to have the same materials as design applications. This is not always feasible. Using the current approach, however, limited use of absorbers may be appropriate even when benchmarks do not contain these materials, provided that the sensitivity of k_{eff} to these materials is low.

MOX powder systems

Low-moderated MOX powder systems can be expected to have many of the same properties as low-moderated PuO_2 systems, in terms of relative sensitivity to various system parameters. The actual permissible ranges of fundamental parameters may be different, however, because very pure ^{235}U or ^{239}Pu systems tend to have greater sensitivity to parameter changes than low ^{235}U or ^{239}Pu systems. The most significant physical difference between PuO_2 and MOX powders is the presence of large quantities of ^{238}U (88 and 93.7 wt.% ^{238}U for the master and final blend respectively). ^{238}U capture can contribute a significant fraction of the total fissile mixture sensitivity in the thermal to intermediate energy range, while ^{238}U fission contributes in the fast energy range. Typical sensitivity profiles for ^{239}Pu fission and capture are shown in Figure 4, for a case with ~30 wt.% Pu content. Typical sensitivity profiles for ^{238}U fission and capture are shown in Figure 5.

Figure 4. ^{239}Pu total, fission and capture sensitivity profile for NSE55T5-07

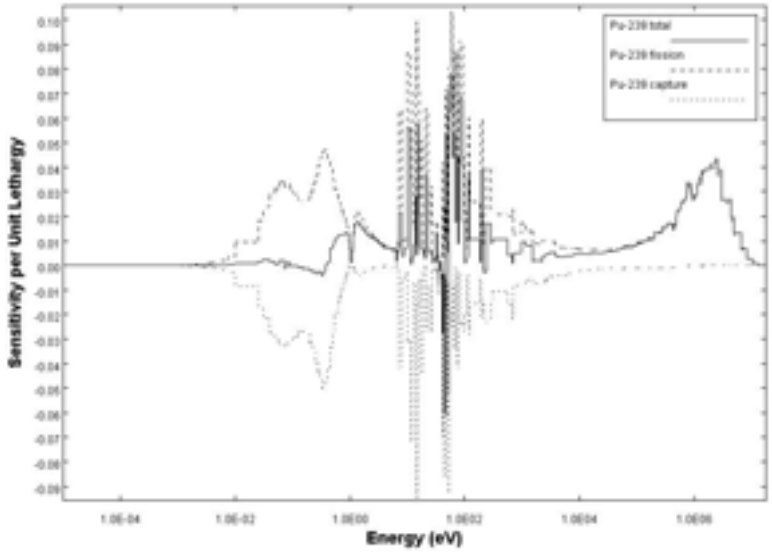
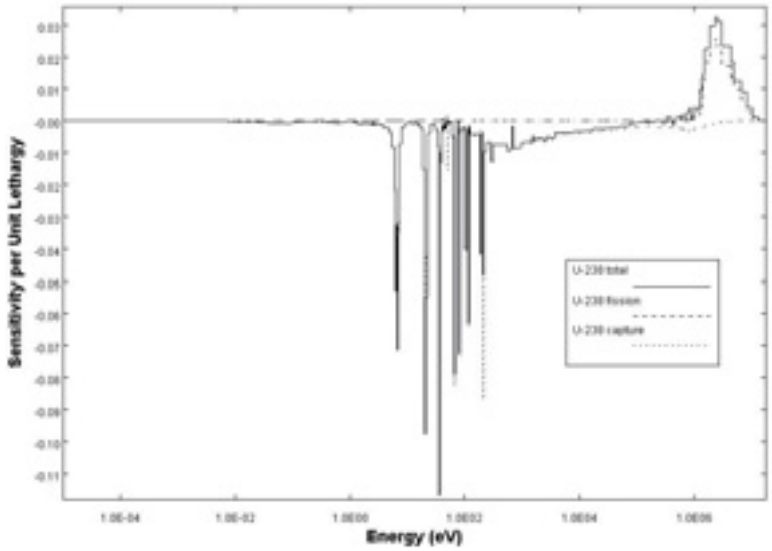


Figure 5. ^{238}U total, fission and capture sensitivity profiles for NSE55T5-07



The total sensitivity coefficients for the dominant reactions for this case are shown below in Table 3, in decreasing order. The subscripts (f), (c) and (s) refer to fission, capture and scattering. The total sensitivity for this fissile mixture was 0.306.

Table 3. Dominant sensitivity coefficients for NSE55T5-07

^{239}Pu(f)	^{239}Pu(c)	^{1}H(s)	^{238}U(c)	^{240}Pu(c)	^{238}U(f)
0.424	-0.209	0.115	-0.085	-0.054	0.029

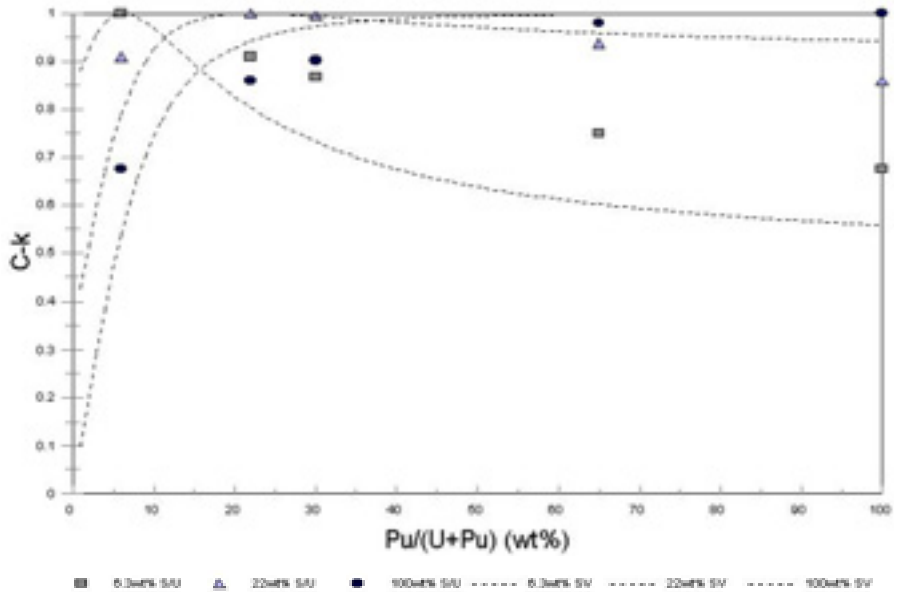
Note that ^{240}Pu was previously found to be of secondary importance, so anything with a coefficient less than that for ^{240}Pu capture is also a minor contributor.

What we see from Figure 4 is that in the fast energy range, almost all of the sensitivity of k_{eff} to ^{239}Pu (the dominant nuclide) is due to ^{239}Pu fission. In such a regime, it would not be unreasonable to base acceptance largely on a comparison of the ^{239}Pu fission spectra. However, in the thermal and intermediate ranges, ^{239}Pu capture becomes increasingly important. Figure 5 shows that ^{238}U fission is an important contributor at fast energies and ^{238}U capture becomes significant at intermediate energies, even rivalling the sensitivity of ^{239}Pu fission and capture in this regime.

Therefore, in systems in which there is a large amount of ^{238}U, the effect on this nuclide on the overall system k_{eff} cannot be discounted. ^{239}Pu fission dominates in the fast energy range but for systems with sizeable spectral contributions in the intermediate and thermal ranges, the relative sensitivity of k_{eff} to other nuclide-reaction pairs must be considered.

The results of a parametric analysis in which MOX-water spheres were modelled with varying Pu content are presented below in Figure 6. In addition to changing the number densities of ^{239}Pu and ^{238}U, increasing the Pu content of the mixture results in a hardening of the fission spectrum, which may be responsible for part of the drop-off in c_k with increasing Pu content.

Figure 6. Correlation of MOX-water spheres to 6.3, 22 and 100 wt.% cases



This graph shows that there is a considerable drop-off in c_k as the difference in Pu content between the cases compared grows. The points represent c_k values calculated using the TSUNAMI

code; the MOX-water spheres were modelled using the XSDRNPM discrete ordinates code. This shows that 100 wt.% Pu benchmarks have a high degree of applicability to applications with much lower Pu content, but that there is a sudden drop-off below ~30 wt.% Pu. The breadth of this range is thought to be due to the dominance of ^{239}Pu fission, which is responsible for almost all of the system reactivity. Removing the ^{238}U results in a net change in k_{eff} of at most a few per cent over the range of 30-100 wt.% Pu.

The dashed curves are the result of another type of analysis. Ref. [7] derives an analytical approach for the correlation between a pair of two-isotope systems. This was originally derived to investigate the behaviour of a partially-enriched system, but can be applied to a ^{239}Pu-^{238}U system as well. The dashed curves are the tabulated values of the following function for the correlation parameter:

$$\rho = \frac{\left\{\sigma_1\sigma_2\varepsilon_1\varepsilon_2\delta_P^2 + \sigma_1\sigma_2(1-\varepsilon_1)(1-\varepsilon_2)\delta_U^2\right\}}{\sqrt{\left\{\sigma_1^2\varepsilon_1^2\delta_P^2 + \sigma_1^2(1-\varepsilon_1)^2\delta_U^2\right\}}\sqrt{\left\{\sigma_2^2\varepsilon_2^2\delta_P^2 + \sigma_2^2(1-\varepsilon_2)^2\delta_U^2\right\}}}$$

In this application, σ_P and σ_U are the total ^{239}Pu and ^{235}U cross-sections, δ_P and δ_U their uncertainties, and ε_1 and ε_2 the Pu content of the two cases. The total macroscopic cross-sections of the two mixtures can be expressed as $\sigma_1 = \sigma_P\varepsilon_1 + \sigma_U(1-\varepsilon_1)$ and $\sigma_2 = \sigma_P\varepsilon_2 + \sigma_U(1-\varepsilon_2)$. This simple expression does not take into account energy- and reaction-dependent differences in the cross-sections, but only makes a gross comparison that depends on the ratios of the total cross-sections and their uncertainties. In the intermediate-to-fast energy range, the ratio of the ^{239}Pu to the ^{238}U total cross-section is ~1. In the thermal energy range, however, the ratio ranges from ~10 to ~100 due to the prevalence of thermal ^{239}Pu fission. The curves above were derived using the assumption that the uncertainties are in the same ratio as the cross-sections, with σ_P/σ_U (and thus δ_P/δ_U) = 100. Although a much less sophisticated treatment, this gives results qualitatively similar to that using the full TSUNAMI approach.

Based on the TSUNAMI analysis, the following range in Pu content is derived, based on a c_k cut-off of 0.90. For 100 wt.% Pu applications, the screening range is 30-100 wt.% Pu. For 22 wt.% Pu, the screening range is 4-82 wt.% Pu. And for 6.3 wt.% Pu, the screening range is ≤ 23 wt.% Pu.

Application to existing benchmarks

Based on the foregoing parametric analysis for low-moderated PuO_2 and MOX-water systems, the parameters of greatest significance to benchmark applicability are the H/X ratio and total Pu content. Other parameters, including geometric form, physical and chemical form, density, and plutonium isotopic (^{240}Pu content) appear to play only a minor role.

The set of screening criteria found in Tables 4 and 5 are based on the application ranges in Table 2, the results of TSUNAMI calculations, and the screening criteria in Table 2.3 of Ref. [3]. The parametric ranges covered by the experiments analysed in Refs. [5,6] are also included for comparison.

Note that TSUNAMI analyses were not run for every combination of parameters. For MOX powders, only the effect of Pu content was investigated; the effect of the other parameters is presumed roughly similar to that for PuO_2 powders.

Based on these tables, it appears that the primary limitation of existing benchmarks is their ability to cover the lowest energy powder systems, as well as low Pu content systems. The application of TSUNAMI results in general in less restrictive screening ranges than those derived using Table 2.3 of Ref. [3] (especially in regard to H/X, Pu content and ^{240}Pu content). The use of a different cut-off value for c_k would have a corresponding effect on these values.

Table 4. Screening ranges and benchmarks for AOA(3)

Parameter	Ref. [3]	TSUNAMI	Applications	Benchmarks
H/X	0-19.8	0-25	0-16.5	0-15.1
Pu-content	90-100 wt.%	30-100 wt.%	100 wt.%	29.3-100 wt.%
^{240}Pu	0-8 wt.%	0-30 wt.%	4 wt.%	2.2-18.4 wt.%
EALF	1-2 × 10^{7} eV (1.18×10^{-19} – 2.36×10^{-12} ft-lb)		3-2.66 × 10^{5} eV (3.55×10^{-19} – 3.14×10^{-14} ft-lb)	4-1.25 × 10^{6} eV (4.73×10^{-19} – 1.48×10^{-13} ft-lb)

Table 5. Screening ranges and benchmarks for AOA(4)

Parameter	Ref. [3]	TSUNAMI	Applications	Benchmarks
H/X	0.88-1.92		1.1-1.6	2.77-15.1
Pu content	3.8-37	≤ 82 wt.%	6.3, 22 wt.%	8.1-100 wt.%
^{240}Pu	0-8 wt.%		4 wt.%	2.2-11.5 wt.%
EALF	0-10^{4} eV (0-1.18×10^{-15} ft-lb)		0.28-850 eV (3.31×10^{-20} – 1.00×10^{-16} ft-lb)	0.6-92.6 eV (7.09×10^{-20} – 1.09×10^{-17} ft-lb)

There are two issues that are of particular interest: (1) the applicability of Pu metal benchmarks to validate MOX-power systems, and (2) the applicability of PuO_2 systems to validate low-Pu MOX systems. The dependence of c_k on Pu content has already been explored, and this reveals the need for additional low-Pu MOX benchmarks. With regard to Pu metals, replacing low-density PuO_2 powder with full-density Pu metal is expected to have a very slight effect on system c_k. This is expected based on the demonstrated low dependence on powder density, and on the low sensitivity to the unique element present in powder (i.e. the ^{16}O cross-sections). The primary issue with regard to use of Pu-metal benchmarks, then, would seem to be the fact that these benchmarks have H/Pu = 0.

Conclusions

The primary area of concern with regard to subcritical margin appears to be the low-energy (thermal to intermediate) range of PuO_2 and especially MOX powder design applications. This was based on a consideration of relative risks, sensitivity of k_{eff} to traditional trending parameters and a survey of existing benchmarks. In the fast energy range ^{239}Pu fission plays a dominant role, but other absorbing and moderating materials such as ^{238}U come to play a significant role in the lower-energy regime. The intermediate energy range is of particular interest for MOX systems, where the ^{239}Pu and ^{238}U cross-sections are in competition. This is expected to occur in the powder blending process, which is also an area of key criticality control.

The parameters of primary importance appear to be the H/X ratio and Pu content, based on parametric curves derived in this study and on the observation that ^{239}Pu and ^{1}H account for most of the system k_{eff} sensitivity in low-moderated PuO_2 and MOX systems. Density, geometry and neutron energy appear to have a much less significant effect. This study shows that use of the TSUNAMI codes, with a conservative c_k cut-off value can, together with traditional trending approaches and other analytical methods, provide valuable insights into which parameters and behaviour are important for validation of process in an MFFF.

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SESSION II

Experimental Data Needs

Chairs: M. Westfall, P. Cousinou

CRITICALITY CALCULATION CODES VALIDATION: EXPERIMENTAL NEEDS FOR LOW-MODERATED MOX MEDIA

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Abstract

This paper aims to highlight the need for integral critical experiments dealing with low-moderated MOX media, which are mainly encountered in MOX fuel fabrication plants, transport and dry storage.

For this purpose, the available experiments dealing with low-enriched uranium, plutonium and/or MOX powders and with low-moderated arrays (low-enriched uranium and mixed U-Pu) were investigated. These experiments were mainly taken from the ICSBEP handbook, but also from the French calculation codes validation database for fast and thermal reactors (ERANOS and APOLLO2 codes).

Several neutronics parameters were compared in order to confirm that new and accurate experiments are needed to significantly decrease the uncertainties linked to k_{eff} prediction for low-moderated MOX media of interest.

Introduction

In the framework of calculation codes' validation, the use of mixed-oxide fuel (MOX) in nuclear power plants to recycle LWR-Pu from reprocessing plants or convert surplus weapons-grade (WG) plutonium has created a need for benchmarks that deal with MOX fuel. Particularly in the field of criticality hazard in fuel cycle facilities, accurate critical experiments are a necessary means to decrease uncertainties linked to configuration multiplication factor prediction in order to optimise different processes.

The OECD/NEA Expert Group on Experimental Needs for Criticality Safety has highlighted MOX fuel manufacturing as an area in which there is a specific need for additional experimental data for validation purposes. This paper aims to contribute to the establishment of the technical basis for the stated need.

Low-moderated MOX fuel encountered in MOX fuel manufacturing

In the MOX fuel fabrication process, the fissile material is present as plutonium-oxide powders, mixed uranium-plutonium oxide powders with different plutonium contents (i.e. primary blend and final blend), pellets, MOX fuel rods and assemblies. During the different steps of the fabrication processes, low quantities of hydrogenated materials (lubricants, pore-formers) are involved.

MOX fuel fabrication plants must be designed to prevent the risk of a nuclear criticality accident under all normal and credible abnormal conditions. As the fabrication process cannot be conducted only using geometrically safe conditions, it is necessary to limit both the masses of the fissile media and their hydrogen content in the main units of the plant.

Typically, for MOX powder fabrication units, the following parameters have to be considered:

	Reactor-grade (RG) plutonium	**Weapons-grade (RG) plutonium**
PuO_2 content	30% (primary blend) 12.5% (final blend)	22% (primary blend) 6.5% (final blend)
Powder density	4.6 g/cm^3 5.5 g/cm^3	5.5 g/cm^3
Water content	3% (normal conditions) 5% (abnormal conditions)	1-5%
U enrichment	^{235}U wt.% ≤ 1.2%	^{235}U wt.% ≤ 1.2%
Pu composition	^{240}Pu wt.% ≥ 17%	^{240}Pu wt.% ~ 4%

Computer codes and calculation methods

The calculations were performed with the multi-group route of CRISTAL, which is the French package used for criticality safety studies.

This route deals with the APOLLO2 assembly code, the CEA93 172-group energy library, and the MORET4 Monte Carlo code. The CEA93 application library using the 172-group Xmas energy structure was derived from JEF-2.2 and was processed using NJOY-THEMIS. The assembly code APOLLO2 is used for self-shielding (using the generalised Livolant-Jeanpierre Formalism), for flux calculations (using the P_{ij} method) and for sensitivity studies. Then, self-shielded and homogenised cross-sections are used in the Monte Carlo code MORET4 for 3-D calculations.

So as to enable comparisons between the different presented papers, a five-group energy structure was proposed by the OECD. Taking into account the energy group boundaries in the 172-group energy mesh, the adopted five-group structure is as follows:

- Group 1: 111 keV < E < 19.6 MeV.
- Group 2: 11.14 keV < E < 111 keV.
- Group 3: 9.906 eV < E < 11.14 keV.
- Group 4: 0.1 eV < E < 9.906 eV.
- Group 5: E < 0.1 eV.

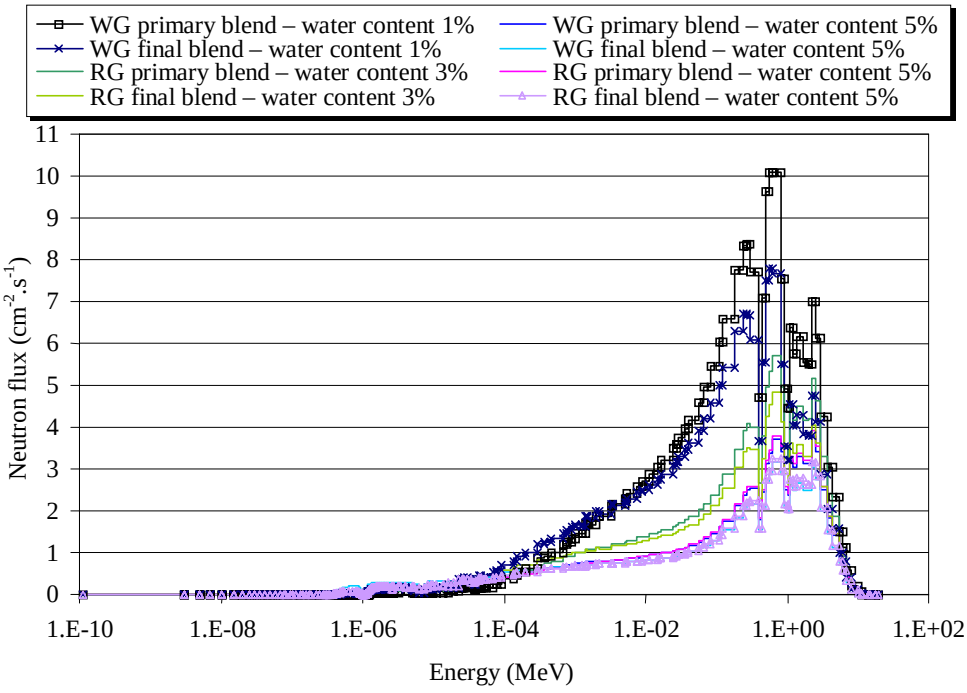
Main neutronic characteristics of MOX powders

Calculations were performed for reactor-grade (RG) and weapons-grade (WG) MOX powders with the different PuO_2 and water contents given previously. Maximal density (5.5 g/cm^3) and uranium enrichment (1.2% of ^{235}U) and minimal ^{240}Pu content were studied (4% for WG and 17% for RG plutonium).

Regarding the reactor-grade plutonium, the isotopic composition assessed in the MELOX French MOX fuel manufactory safety report (71% of ^{239}Pu, 17% of ^{240}Pu, 11% of ^{241}Pu, 1% of ^{242}Pu) was considered in this paper.

For all of these powders, the neutron spectrum varies between the fast and the intermediate energy range (see Figure 1).

Figure 1. Neutron spectra in infinite media for "MELOX" powders



In order to make a judgment upon the similarity between an existing or a planned experiment and the MOX powders of interest, several physic parameters were calculated:

- $q_{\infty\text{-}4eV}$: Slowing-down density, which is the number of neutrons per fission neutron that slow down past an energy of 4 eV. A value of 1 indicates a thermal spectrum in fissile media, a value of 0 a very fast media (metallic systems).
- GMF: Average group of neutrons causing fission in the 172-group energy structure and its corresponding energy boundaries.
- EALF: Energy of average lethargy causing fission.
- Five-group flux, fission and capture data (in per cent).
- Five-group detailed balance for the main isotopes.
- Sensitivity coefficients to the main nuclear reactions for uranium and plutonium isotopes.

Table 1. Spectra characterisation of MOX powders

Plutonium	PuO_2 (%)	Water (%)	H/(U+Pu)	H/Pu	$H/Pu_{fissile}$	GMF	Energy (MeV)	$q_{\infty\text{-}4eV}$	EALF
Reactor-grade	30	3	0.929	3.436	5.366	48	5^{E-3}, 5.53^{E-3}	0.062	6 116
	30	5	1.580	5.846	9.131	57	9.14^{E-4}, 1.01^{E-3}	0.130	1 648
	12.5	3	0.928	7.558	11.804	59	6.77^{E-4}, 7.49^{E-4}	0.135	1 281
	12.5	5	1.579	12.862	20.088	72	4.55^{E-5}, 4.83^{E-5}	0.237	298
Weapons-grade	22	1	0.303	1.382	1.439	38	3.70^{E-2}, 4.09^{E-2}	0.016	33 670
	22	5	1.579	7.200	7.499	70	5.16^{E-5}, 5.56^{E-5}	0.187	731
	6.5	1	0.303	4.677	4.871	54	1.43^{E-3}, 1.51^{E-3}	0.050	6 845
	6.5	5	1.578	24.369	25.380	89	5.35^{E-6}, 6.16^{E-6}	0.320	61

As shown in Table 4, less than 5% of the neutron flux occurs under 10 eV (Groups 4 and 5) and more than 50% above 100 keV. The main parts of absorption occur in Group 3.

Sensitivity calculations highlight that absorption in infinite fissile media is mainly due to the ^{238}U capture in the resonance range between 7 and 100 eV (see Figure 3), the ^{239}Pu fission (see Figure 4) and capture (see Figure 5) below 10 keV, and the ^{240}Pu capture in the first resonance and above 100 eV.

Available experiments

Experiments taken from the ICSBEP handbook [1]

Experiments involving low-moderated powders

Two experimental programmes involving low-enriched uranium powders, which could contribute to ^{238}U cross-sections validation, are available in the ICSBEP handbook:

- MARACAS experiments (LEU_COMP_THERM_049), which were performed at CEA Valduc (France) with low-moderated UO_2 powders (H/U = 2, 2.5, 3).
- Rocky Flat experiments (LEU_COMP_THERM_045), which involved low-moderated U_3O_8 powders (H/U = 0.77) with an additional interstitial moderation by Plexiglas.

Spectra calculations show that these experiments could not allow validation in the energy range of interest. In fact, more than 60% of fissions occur in thermal range (E < 0.625 eV) and EALF values ranging from 0.5 to 2.78, with $q_{\infty\text{-}4eV}$ varying from 0.3 to 0.4.

Regarding MOX powders, only one benchmark (MIX_COMP_THERM_012) dealing with 33 bare or Plexiglas reflected MOX-polystyrene compacts configurations with different plutonium contents and various moderation ratios, is available. These experiments allow MOX powder validation, but mainly in the thermal range; in fact, EALF and $q_{\infty\text{-}4eV}$ values respectively range from 0.07 to 0.26 and from 0.56 to 0.65, and most of fissions (more than 85%) occur under 0.625 eV.

Finally, only two sets of experiments dealing with plutonium oxide (PU_COMP_MIXED_001 and 002) occur in the energy range of interest. These experiments involve PuO_2-polystyrene compacts, bare or Plexiglas reflected, with different ^{240}Pu content (2.2 to 18.35 wt.%) and various H/Pu (0.04, 5, 15, 49.6). EALF values range from 0.75 to $1^{E\,6}$, $q_{\infty\text{-}4eV}$ from 0 to 0.53 and 5% to 60% of fissions occur between 0.625 eV and 100 keV.

Unfortunately, the experimental data are weak or missing (lack of information on the exact cube configurations, empirical corrections for cladding…). Therefore these experiments are highly uncertain and cannot be used as "benchmarks" for the purpose of code validation.

Other experiments

As most of fissions occur in Groups 1 (above 100 keV) and 3 (between 10 eV and 10 keV), configurations involving low-moderated MOX powders would be categorised as "mixed" or "intermediated" spectra systems in ICSBEP classification.

Until now, very few experiments have been performed in the intermediate energy range; two experimental programmes dealing with plutonium-metal systems are available in the ICSBEP handbook. Thus, they could contribute to plutonium cross-section validation in intermediate and fast spectra.

Finally, experiments performed at the BFS facility at Obninsk dealing with depleted UO_2 disks stacked with WG-Pu disks, which are not yet evaluated as benchmarks in the ICSBEP, could perhaps allow to cover the energy range of interest.

Experiments taken from the French validation database: The ERASME experiments [2]

The ERASME experiment was built up to reduce the uncertainties regarding fundamental lattice parameters associated with the use of MOX fuel in under-moderated spectrum. The fuel pins correspond to the HCPWR design with stainless steel clad and 11% PuO_2 content (Pu reprocessed from 33 GWd/t PWR). Three lattices were considered, with different moderation ratios: 0.5, 0.9 and 2.1, corresponding to the ERASME/S (serré = tight), ERASME/R (realistic) and ERASME/L (large) cores respectively.

These experiments allow covering the intermediate energy range (see Figure 2).

The integral lattice parameters investigated in these cores were:

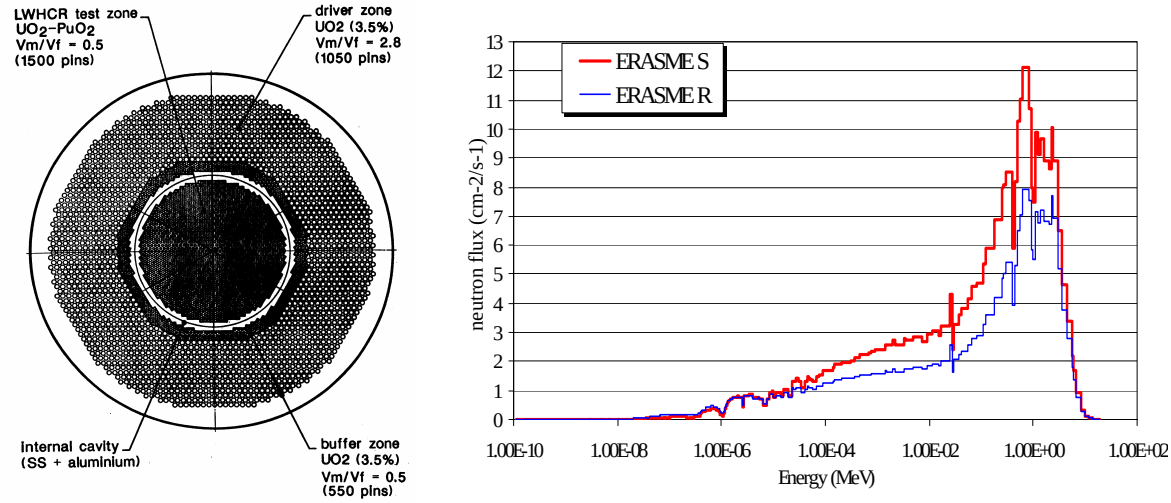
- The determination of the buckling through fission rate distributions.
- The conversion factor ($\sigma_c^{238}U/\sigma_f^{239}Pu$).

- The fission rates of the most relevant heavy isotopes (^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am) by using fission chambers.

- Absorber worth and cluster configurations (stainless steel, B_4C, Hf, AIC and Eu).

- Perturbations induced by adding HCR assembly heterogeneities (water holes, fertile UO_2, etc.).

Figure 2. Neutron spectra for ERASME experiments



Regarding the fission and capture balance for MOX powders' major isotopes (see Table 2), one can conclude that ERASME/S experiment is close to "MELOX" final blend with a water content of 5%: the large contributions of continuum and unresolved resonance range (respectively 8% and 10%) to ^{238}U capture is preserved, as well as the 60% predominant component of the 10 eV-10 keV domain to ^{239}Pu capture.

However, ERASME experiments do not mock-up powder configurations with high PuO_2 content and with water content lower or equal to 3% (see Figures 3 and 4). Consequently, to assess more precisely the actual safety margins in the "MOX" fuel facilities under normal conditions, it seems necessary to perform new experiments with low-moderated mixed-oxide fissile materials.

Proposal of "homogeneous" core experiments using new UO_2-PuO_2 rods (higher Pu content)

Obviously, the most representative experiments corresponding to the fabrication process would be experiments dealing with MOX powders with different moisture content (from 1 to about 5%) and a density close to 5.5 g/cm^3.

Unfortunately, with mixed oxide powders, such experiments rise many difficulties mainly due to the plutonium containment, the plutonium cooling, in order to assure a definite temperature and a given moisture content, and to the quantity of fissile material needed.

For these reasons, IRSN [3] has studied the possibility of performing experiments with low water-moderated MOX fuel rods arrays.

Table 2. Comparison of fission and capture neutron balance in the five-group energy structure

		ERASME/S experiment		RG MOX powder 12.5% PuO_2 - 5% H_2O		Discrepancies	
Isotopes	**Groups**	**Capture (%)**	**Fission (%)**	**Capture (%)**	**Fission (%)**	**Capture (%)**	**Fission (%)**
^{238}U	1	8.423	99.929	7.214	99.933	16.7	-0.004
	2	9.960	0.024	9.057	0.022	9.9	10.3
	3	65.145	0.047	67.347	0.045	-3.3	4.7
	4	16.138	0.000	16.228	0.000	-0.5	–
	5	0.3345	0.000	0.1535	0.000	117.9	–
^{239}Pu	1	1.474	13.779	1.532	15.151	-3.8	-9.0
	2	2.029	3.441	2.201	3.726	-7.8	-7.6
	3	58.916	43.798	64.512	48.350	-8.7	-9.4
	4	31.644	31.191	28.491	28.506	11.0	9.4
	5	5.936	7.791	3.263	4.269	81.9	82.5
^{240}Pu	1	0.727	89.350	0.669	89.452	8.6	-0.1
	2	0.971	1.881	0.927	1.797	4.7	4.7
	3	23.619	8.527	23.915	8.511	-1.2	0.2
	4	72.648	0.234	73.509	0.236	-1.2	-0.9
	5	2.035	0.008	0.980	0.004	–	–

Figure 3. Sensitivity to ^{238}U capture cross-section

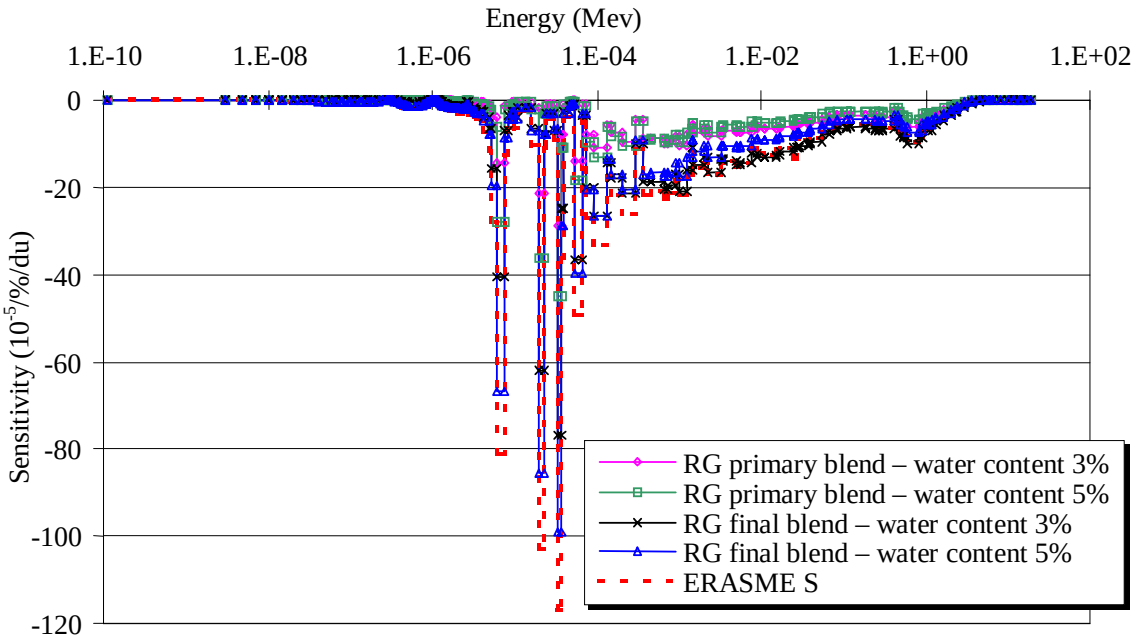
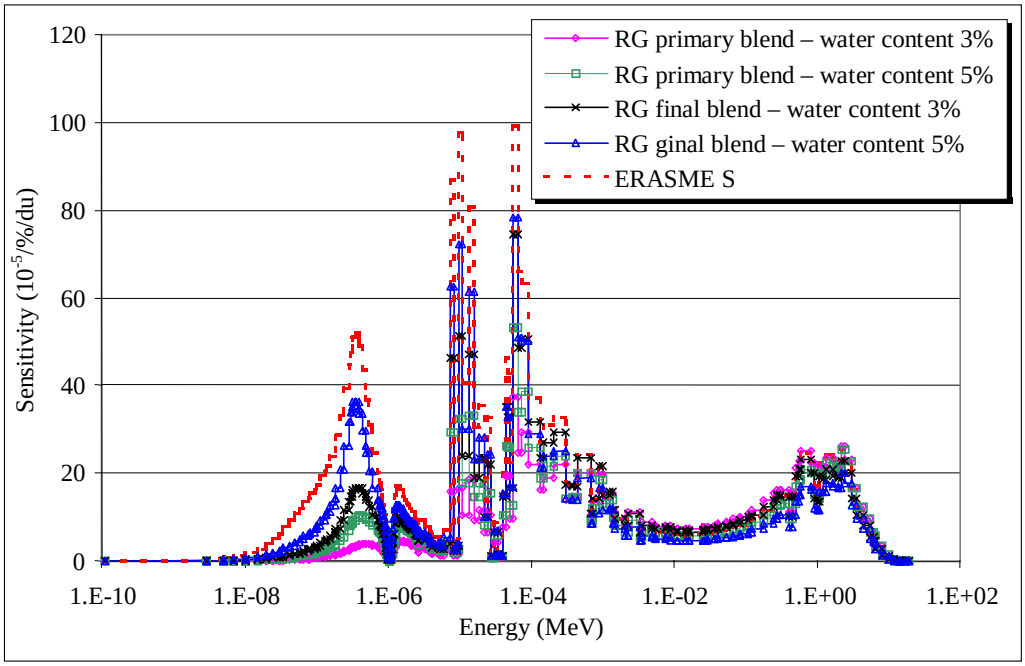


Figure 4. Sensitivity to ^{239}Pu fission cross-section



The preliminary design (pellets diameter, pitches, isotopic composition…) is being optimised with regard to the feasibility of the experiments, including the possibility of using the existing equipment, as concerns the fuel fabrication cost and the representativity of the experiments.

The experimental configuration involves a MOX fuel rod array inside a parallelepiped tank partially filled with water. Fuel rods contain UO_2-PuO_2 pellets composed of about 27.5% (wt) of PuO_2, which corresponds to the real plutonium content of the primary blend mixture in the French fabrication process. Mixed-oxide is obtained from reactor-grade plutonium (about 25% of ^{240}Pu). Several "homogeneous" cores with different tight triangular pitches are chosen, corresponding to MOX powders' water-to-fuel ratios, in order to cover the same spectral range in these "clean" and representative experiments.

As demonstrated previously, experiments with UO_2-PuO_2 fuel rods can be representative of MOX powders when a low moderation ratio is maintained. In fact, for water content lower than 5%, the use of homogeneous or heterogeneous fissile media does not have a great impact with regard to the neutron mean free path in this kind of spectra. Thus, the proposed experiments allow covering the same energy range as MOX powders, when keeping a similar $H/Pu_{fissile}$ ratio.

Table 3 presents the comparison of fission and capture balance in the five-group energy structure for the major isotopes between one of the proposed experiments and the "MELOX" primary blend with 3% water content. As can be seen, the representativity of the proposed experiment is satisfactory, since ^{238}U capture in unresolved resonance range is preserved as well as ^{239}Pu absorption in resolved and unresolved domains.

Thus, such experiments, added to the ERASME experimental programme, would enable to validate calculation codes for the different powders encountered in a French MOX fuel fabrication plant. Moreover, as they involve only one kind of fissile media, these experiments would be easy to investigate with all calculation codes.

Table 3. Comparison of fission and capture balance in the five-group energy structure

		Proposed experiment: 27.5% PuO_2 MOX rods		RG MOX powder 30% PuO_2 – 3% H_2O		Discrepancies	
Isotopes	**Groups**	**Capture (%)**	**Fission (%)**	**Capture (%)**	**Fission (%)**	**Capture (%)**	**Fission (%)**
^{238}U	1	12.710	99.939	11.876	99.938	-6.5	-0.0
	2	14.724	0.025	14.821	0.024	0.6	-2.8
	3	63.711	0.037	66.485	0.038	4.3	4.8
	4	8.825	0.000	6.806	0.000	-22.9	–
	5	0.030	0.000	0.011	0.000	-61.3	–
^{239}Pu	1	4.622	33.277	4.664	34.381	0.9	3.3
	2	6.168	8.351	6.628	8.860	7.4	6.1
	3	78.039	48.378	81.170	49.926	4.0	3.2
	4	10.083	8.870	7.090	6.374	-29.7	-28.1
	5	1.088	1.124	0.449	0.460	-58.7	-59.0
^{240}Pu	1	3.926	92.335	3.621	91.932	-7.8	-0.4
	2	5.109	1.989	4.988	1.995	-2.3	0.3
	3	50.675	5.649	54.352	6.048	7.2	7.1
	4	39.648	0.026	36.797	0.025	-7.2	-5.9
	5	0.642	0.000	0.241	0.000	-62.465	–

Conclusion

In the framework of calculation codes' validation, the use of mixed-oxide fuel in nuclear power plants has created a need for benchmarks that deal with MOX fuel.

The assessment of the available benchmarks has highlighted the lack of experiments dealing with low-moderated MOX powders, which are mainly encountered in MOX fuel fabrication plants.

For the different configurations assessed in MOX fuel manufactory safety report, the neutron flux varies in the fast and the intermediate-energy range. In such spectrum, the neutron mean free path is very high, thus the fissile media heterogeneity does not have a great impact.

Therefore, experiments taken from French reactor validation database (PWR and fast breeder) have been investigated. Comparison of neutronic parameters and sensitivity studies have highlighted that the ERASME/S experiment is similar to the "MELOX" final blend with a 5% water content. Unfortunately, this experiment is not representative enough of MOX powders with higher PuO_2 content or with lower moisture.

The next step of this study is to assess the nuclear data uncertainties' effect on k_{eff} uncertainty and to verify that this uncertainty can be decreased using integral experiments. Within this framework, adjustment techniques will be used to judge the relevance of different experiments.

Thus, it was verified that new and accurate experimental data would enable a better optimisation of the fabrication process by decreasing the uncertainties on the determination of multiplication factors of configurations involving MOX powders.

Table 4. Flux, fission and capture balance in the five-group energy structure for MOX powders

Pu	PuO_2 (%)	H_2O (%)	Flux (%)					Fission (%)					Capture (%)				
			1	2	3	4	5	1	2	3	4	5	1	2	3	4	5
RG	30	3	62.28	16.99	20.01	0.72	0.00	38.41	7.75	46.03	7.43	0.38	9.09	9.47	68.67	12.54	0.23
	30	5	61.53	15.26	21.75	1.45	0.01	31.09	5.53	48.31	14.02	1.04	6.29	6.04	64.66	22.44	0.57
	12.5	3	57.27	16.90	24.06	1.76	0.01	30.25	5.00	47.65	15.64	1.46	7.21	7.99	63.27	20.96	0.57
	12.5	5	57.19	14.98	24.80	2.99	0.05	24.05	3.34	43.73	25.39	3.49	5.13	5.09	56.35	32.12	1.31
WG	22	1	61.25	21.61	16.87	0.27	0.00	49.83	13.05	35.00	2.04	0.08	14.92	19.15	62.59	3.30	0.05
	22	5	59.65	15.02	22.81	2.51	0.02	28.30	4.74	45.26	19.59	2.11	5.74	5.73	62.56	24.99	0.98
	6.5	1	53.90	21.99	23.19	0.91	0.00	39.28	8.22	44.89	7.10	0.52	11.48	16.33	64.38	7.66	0.16
	6.5	5	54.71	14.51	25.75	4.88	0.15	20.21	2.17	33.69	36.13	7.79	4.92	4.96	53.32	34.22	2.58

Figure 5. Sensitivity to ^{239}Pu capture cross-section

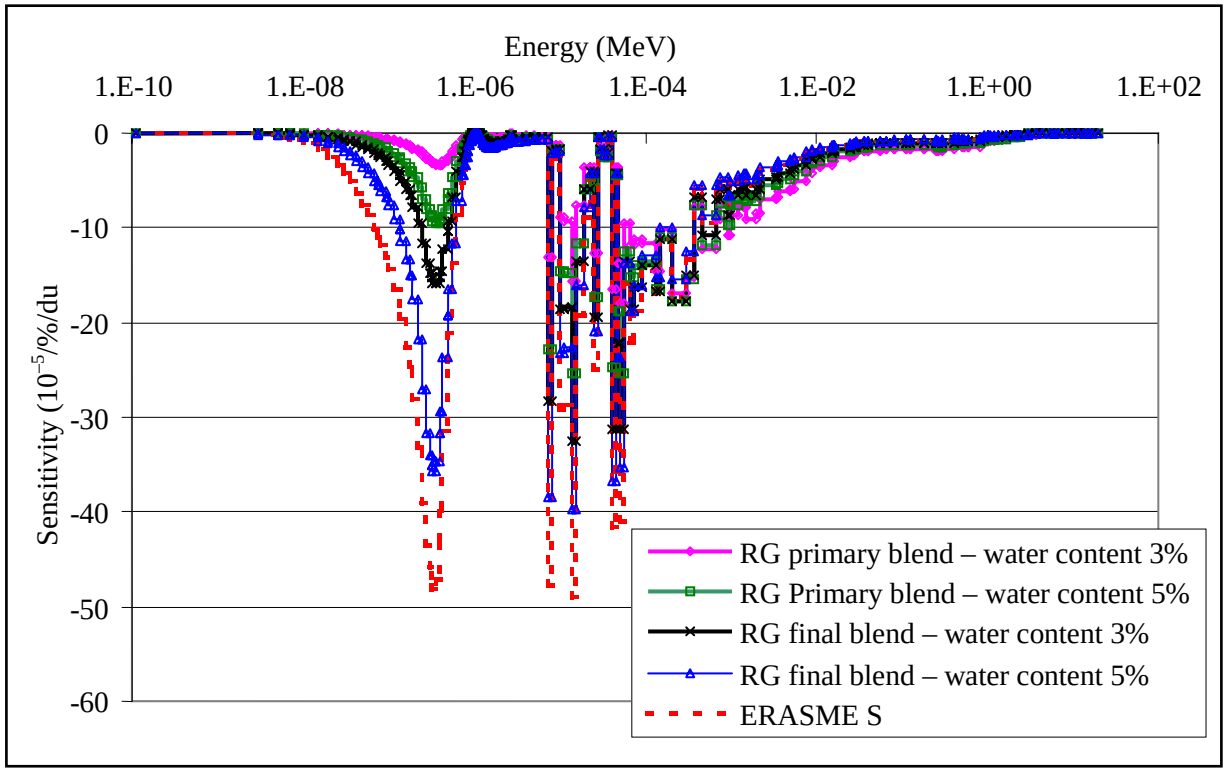


Table 5. Five-group detailed balance in weapons-grade and reactor-grade MOX powders

		"MELOX" powders								WG MOX powders							
PuO_2 (%)		30%				12.5%				22%				6.5%			
H_2O (%)		3%		5%		3%		5%		1%		5%		1%		5%	
Nuc.	Gr.	Cap.	Fis.	Cap.	Fis.	Cap.	Fis.	Cap.	Fis.	Cap.	Fis.	Cap.	Fis.	Cap.	Fis.	Cap.	Fis.
^{235}U	1	0.04	0.37	0.03	0.28	0.06	0.49	0.04	0.36	0.09	0.69	0.03	0.31	0.12	0.92	0.04	0.39
	2	0.05	0.17	0.04	0.11	0.08	0.24	0.05	0.15	0.12	0.39	0.04	0.13	0.19	0.61	0.05	0.17
	3	0.53	1.14	0.53	1.08	1.12	2.30	0.99	1.95	0.60	1.38	0.70	1.41	1.63	3.56	1.26	2.44
	4	0.06	0.13	0.09	0.20	0.20	0.46	0.25	0.62	0.04	0.09	0.18	0.41	0.19	0.45	0.45	1.33
	5	0.00	0.00	0.00	0.01	0.01	0.04	0.02	0.10	0.00	0.00	0.01	0.03	0.00	0.02	0.06	0.37
^{238}U	1	1.98	4.58	1.39	3.75	2.69	5.76	1.84	4.71	4.08	6.59	1.59	4.23	5.54	7.93	2.02	5.09
	2	2.47	0.00	1.70	0.00	3.55	0.00	2.31	0.00	5.69	0.00	1.95	0.00	8.65	0.00	2.55	0.00
	3	11.1	0.00	11.2	0.00	18.8	0.00	17.2	0.00	12.2	0.00	13.6	0.00	25.7	0.00	20.3	0.00
	4	1.14	0.00	1.90	0.00	3.02	0.00	4.15	0.00	0.54	0.00	3.04	0.00	2.07	0.00	6.22	0.00
	5	0.00	0.00	0.00	0.00	0.02	0.00	0.04	0.00	0.00	0.00	0.01	0.00	0.01	0.00	0.15	0.00
^{239}Pu	1	0.74	12.9	0.51	9.68	0.34	5.68	0.23	4.19	1.42	20.8	0.52	9.78	0.49	6.77	0.16	2.99
	2	1.05	3.33	0.71	2.25	0.50	1.59	0.33	1.03	2.20	7.07	0.73	2.31	0.84	2.68	0.24	0.75
	3	12.9	18.8	12.9	18.8	10.5	14.8	9.60	13.4	12.9	18.6	14.9	21.9	10.4	14.4	8.58	11.9
	4	1.13	2.39	2.32	4.71	2.36	4.58	4.24	7.87	0.44	1.08	4.67	9.68	1.14	2.39	7.71	13.9
	5	0.07	0.17	0.19	0.45	0.21	0.51	0.49	1.18	0.02	0.05	0.44	1.05	0.08	0.19	1.20	2.92
^{240}Pu	1	0.23	1.79	0.16	1.41	0.11	0.77	0.07	0.60	0.08	0.44	0.03	0.25	0.03	0.14	0.01	0.07
	2	0.32	0.04	0.22	0.03	0.16	0.02	0.10	0.01	0.12	0.01	0.04	0.00	0.05	0.01	0.01	0.00
	3	3.52	0.12	3.57	0.10	2.88	0.07	2.60	0.06	0.79	0.03	1.02	0.02	0.61	0.02	0.49	0.01
	4	2.39	0.00	4.91	0.00	4.89	0.00	8.00	0.00	0.38	0.00	4.13	0.00	1.15	0.00	5.14	0.00
	5	0.02	0.00	0.04	0.00	0.05	0.00	0.11	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.05	0.00
^{241}Pu	1	0.31	1.92	0.22	1.42	0.14	0.85	0.10	0.62	–	–	–	–	–	–	–	–
	2	0.18	0.84	0.12	0.57	0.09	0.40	0.06	0.26	–	–	–	–	–	–	–	–
	3	1.55	5.95	1.58	5.81	1.18	4.26	1.08	3.73	–	–	–	–	–	–	–	–
	4	0.40	1.67	0.65	2.58	0.54	1.98	0.74	2.59	–	–	–	–	–	–	–	–
	5	0.01	0.04	0.03	0.09	0.03	0.11	0.08	0.24	–	–	–	–	–	–	–	–
^{242}Pu	1	0.01	0.08	0.01	0.07	0.01	0.04	0.00	0.03	–	–	–	–	–	–	–	–
	2	0.02	0.00	0.01	0.00	0.01	0.00	0.01	0.00	–	–	–	–	–	–	–	–
	3	0.15	0.00	0.14	0.00	0.10	0.00	0.09	0.00	–	–	–	–	–	–	–	–
	4	0.31	0.00	0.51	0.00	0.45	0.00	0.58	0.00	–	–	–	–	–	–	–	–
	5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	–	–	–	–	–	–	–	–
H_2O	1	0.12	–	0.17	–	0.12	–	0.17	–	0.05	–	0.17	–	0.05	–	0.17	–
	2	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–
	3	0.03	–	0.04	–	0.04	–	0.06	–	0.01	–	0.05	–	0.02	–	0.07	–
	4	0.01	–	0.03	–	0.04	–	0.08	–	0.00	–	0.06	–	0.01	–	0.17	–
	5	0.00	–	0.00	–	0.00	–	0.01	–	0.00	–	0.00	–	0.00	–	0.03	–
O_{16}	1	0.51	–	0.43	–	0.51	–	0.43	–	0.62	–	0.43	–	0.62	–	0.43	–
	2	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–
	3	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–
	4	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–
	5	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–	0.00	–

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RESEARCH ACTIVITIES AT THE JAPAN ATOMIC ENERGY RESEARCH INSTITUTE FOR CRITICALITY SAFETY ISSUES ON MOX FUEL FABRICATION

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Abstract

The authors reviewed the calculation model of the experiments with homogeneous stacked MOX blocks performed by Bierman, *et al*. By constructing the benchmark model as exactly as possible and modifying the component of the cladding material, the calculated k_{eff} are found to be improved. The critical experiments have a potential to be used as benchmark data.

The main neutronic parameters are presented for proposed MOX powder conditions. The differences of calculated k_{eff} among JENDL-3.3, JEF-2.2 and ENDF/B-VI.8 are not large for the proposed MOX powder condition.

The assumption of a realistic mixing condition of MOX powder and additives could lead to a relaxation of the criticality safety limit values concerning MOX powder during the homogenisation process.

Introduction

Although validations of criticality safety evaluation methods for MOX fuel fabrication process are needed, only a very limited number of experiments have been performed thus far. On the other hand, sufficient criticality experiment data have been accumulated for reactor core design of MOX fuel utilisation in light water reactors. However, one of features of MOX fuel fabrication process in light of reactor physics is that the moderation of the fuel is much lower than that of light water reactors.

Some of criticality experiments using low-moderated homogeneous MOX fuel were performed at the Hanford Critical Mass Laboratory in the 1970s. These experiments were recently evaluated under an activity of the International Critical Safety Benchmark Evaluation Project and were published in the ICSBEP Handbook. However, calculated k_{eff} deviate quite a bit from the experimental values. In this paper, the authors re-evaluate the experiments and report the results.

During the homogenisation process of a MOX fuel fabrication facility, MOX fuels are handled in a powder state. MOX powder is mixed with UO_2 powder and additives. The mixture condition of these powders is important to make a reasonable evaluation of the criticality of the mixtures. It is not an easy task to identify the mixture condition because this requires aspects of the powder technology. The authors discuss the effect of the mixture condition on the criticality of MOX powder.

Re-evaluation of polystyrene-moderated mixed-oxide cube experiments

Criticality experiments performed at the Hanford Critical Mass Laboratory in the 1970s used low-moderated homogeneous MOX fuel blocks; these experiments are quite precious for validating criticality safety calculations. They were carefully evaluated and are documented in the ICSBEP Handbook [1]. Figure 1 shows the calculated k_{eff} in order of the critical heights of the homogeneous blocks, which were taken from the ICSBEP Handbook. The calculations were performed with MCNP and ENDF/B-V. As shown in Figure 1, the calculated k_{eff} largely deviate from the experimental k_{eff} (nearly 1.0). Furthermore, the calculated k_{eff} seem to correlate with the critical heights. These tendencies suggest that the benchmark model in this evaluation does not properly represent the experimental configurations in the light of criticality. These experiments mostly provide higher k_{eff}, with the exception of “Set 1”. If criticality safety limit values for MOX powder were determined using the experiments, the limit values would becomes less conservative.

Each block was encased in a single layer of plastic tape, specifically, MM&M (3M) #471. There existed void space between the blocks when they were stacked in a critical configuration. Dimensions for bare blocks, clad blocks and the effective dimension occupied by blocks when they were stacked in the experimental configurations are given in the original reports. The dimensions for individual blocks/claddings/voids, derived from the references, are provided in the ICSBEP Handbook. The actual configuration of the experiments is shown in the left of Figure 2 and the benchmark model adopted in the handbook is shown in the right of Figure 2. Internal voids and tapes were left out in the benchmark model, and the critical height was corrected to derive a critical slab height that would necessarily be composed of only polystyrene compacts without cladding or internal void contribution. The derived critical height correction factor was applied to all the configurations in each respective set.

Using the “reported average block dimensions in the stack [1]”, the authors constructed a “detailed model”, which includes voids and tapes, to reproduce the experimental configurations as exactly as possible. The 0.5-inch blocks did not fill the uppermost layer, and the arrangements of the 0.5-inch blocks are not presented in any literature [2]. Thus, the authors assumed that the blocks were laid from one of the corners in the uppermost layer.

Figure 1. Calculated k_{eff} values for homogeneous MOX compact blocks

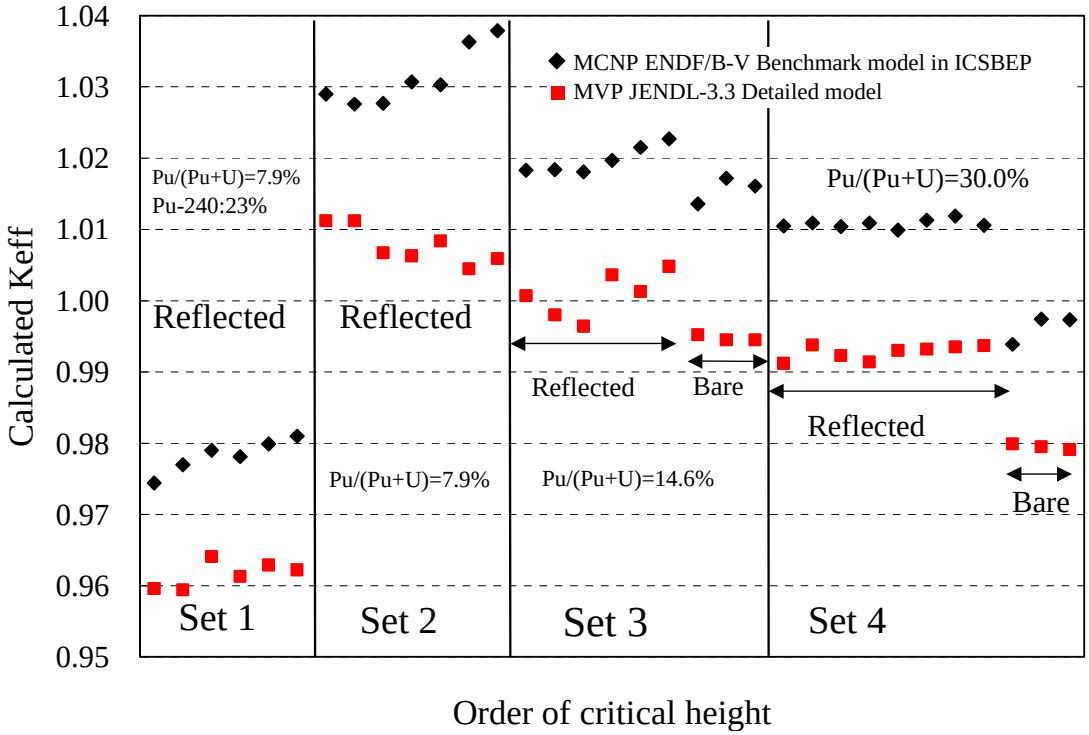
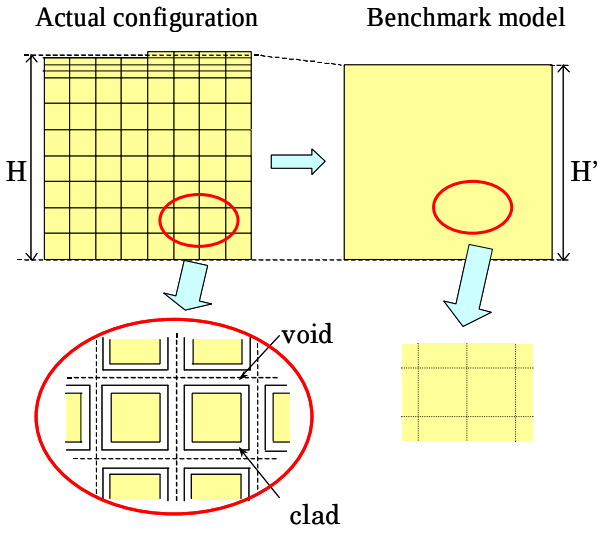


Figure 2. Actual configuration and benchmark model



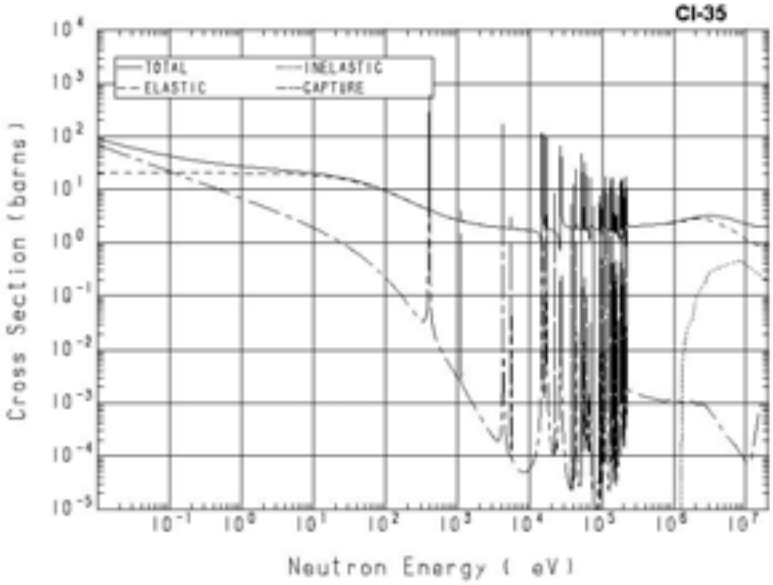
Weight fraction and atom density of each constituent element of the clad tape are given in Ref. [2], and are listed in Column A of Table 1. However, an inconsistency is found between the weight fraction and the atom densities. The weight fraction in Column B may be correct. The clad tape is made of polyvinylchloride. Its chemical formula is CH_2CHCl, and the atom ratio is H:C:Cl = 3:2:1. With this atom ratio, the weight fraction listed is derived as shown in Column C of Table 1. Comparing the weight fractions in Columns B and C, the weight fraction of chlorine in Column B may be that of carbon, and vice versa. Therefore, a true weight fraction and atom density of the clad tape may be as listed in

Table 1. Atom densities of clad tape

	A (reported data in [2])		B	C		D	
	wt.%	Atoms/barn·cm	wt.%	Atom ratio	wt.%	wt.%	Atoms/barn·cm
H	55.3	4.489×10^{-2}	6.7	3	4.84	6.7	4.483×10^{-2}
C	6.7	3.111×10^{-2}	55.3	2	38.44	38.0	2.134×10^{-2}
Cl	38.0	7.240×10^{-3}	38.0	1	56.73	55.3	1.052×10^{-2}

Column D. A density of the clad tape, 1.120 g/cm^3, given in Ref. [2], was used for the atom density calculation. Cross-section curves of ^{35}Cl, the most dominant isotope of chlorine, are shown in Figure 3. Since the cross-section of ^{35}Cl is not small in the thermal energy range, the effect of ^{35}Cl on criticality may not be negligible especially for low-Pu-content MOX fuels. K_{eff} were calculated for the "detailed model" with a continuous-energy Monte Carlo code MVP [3] and JENDL-3.3. The atom densities in the clad tape used for the calculations are listed in Column D of Table 1. The calculated k_{eff} are shown in Figure 1. The k_{eff} calculated by the "detailed model" are approximately 2% smaller than the original benchmark model in the ICSBEP Handbook. The reduction of the k_{eff} for 7.9% Pu-content fuel is mostly caused by adding Cl into the benchmark model. On the other hand, the reduction of the k_{eff} for 30% Pu-content fuel is mostly caused by changing the geometry of the benchmark model as shown in Figure 2. The calculated k_{eff}, which were overestimated by the original benchmark model, can possibly be improved by revising the calculation model. The k_{eff} using the "detailed model" without the Plexiglas reflector are too low by 2% for 30% Pu-content fuel. The "detailed model" of the bare systems does not have any surrounding structures around the stacked blocks. Taking into consideration the room return effect could improve the k_{eff} for the bare systems. For Set 1 (Pu/(Pu+U) = 7.9%, ^{240}Pu:23%), the calculated k_{eff}, which were underestimated in the ICEBEP Handbook's sample calculation, were worsened by revising the calculation model, and thus the calculated k_{eff} are about 0.96. Therefore, introducing the detailed calculation model dose not always improve the calculated k_{eff} of the critical experiments. There may still exist unknown reasons for the underestimation of k_{eff} in Set 1 and in the bare system of Set 4, which is a future subject. As a consequence, by revising the calculation model properly, these experiments would resurrect as precious benchmark experiments.

Figure 3. Cross-section curve of ^{35}Cl in JENDL-3.3



Neutronic parameters of MOX powder fuel

Several neutronic parameters were obtained using JAERI's standard deterministic calculation system SRAC [3] and JENDL-3.3. Fuel conditions are as follows:

PuO_2 content:	30% (corresponding to primary blend) 12.5% (corresponding to final blend)
Powder density:	5.5 g/cm^3
Water content:	1%, 5% (abnormal conditions)
Uranium enrichment:	^{235}U:1.2%
Plutonium composition:	^{239}Pu:^{240}Pu:^{241}Pu = 71/17/12 (usually used for criticality safety analysis for reactor grade plutonium in Japan)

Calculated parameters are:

- Critical cylinder diameter.
- Five-group neutron flux, fission, capture.
- Sensitivity coefficients to fission and capture reaction of heavy elements.

The geometry of the powder is cylindrical with infinite length, and the cylinder is surrounded by a 2.5-cm-thick water reflector. First, 107-group microscopic cross-sections were prepared by the SRAC code system and SRAC public library based on JENDL-3.3. After preparing the 107-group macroscopic cross-sections, k_{eff} were calculated with ONEDANT, and forward and adjoint neutron flux were obtained. Along with the forward and adjoint neutron flux and 107-group microscopic and macroscopic neutron cross-sections, sensitivity coefficients were calculated based on the generalised perturbation theory. The critical cylinder radii are shown in Table 2. In these calculations, it is assumed that all internal water is uniformly mixed with the MOX powder with the initial powder density (5.5 g/cm^3) unchanged. Table 3 shows per cent fraction of fission, capture reactions and neutron flux in the proposed five-energy group structure for each critical dimension. Table 4 shows the sensitivity coefficients of heavy isotopes to fission and capture reactions. For MOX powder with the PuO_2 content 30% and water content 5%, the sensitivity coefficients in the proposed five-energy group structure to capture and fission reactions of heavy isotopes are shown in Table 5.

K_{eff} were calculated for the critical infinite cylinders having 2.5-cm-thick water reflector with JEF-2.2 and ENDF/B-VI.8 as well as with JENDL-3.3. The differences of k_{eff} with JEF-2.2 and ENDF/B-VI.8 with respect to JENDL-3.3 are shown in Table 6. K_{eff} calculated with JEF-2.2, ENDF/B-VI.8, and JENDL-3.3 agreed within a difference of 0.004. The precise contribution of each isotope and each reaction obtained by perturbation calculations shows some conspicuous differences among cross-section libraries. However, the overall dependence of calculated k_{eff} on cross-section libraries is not large considering the conditions of the MOX powder handling process.

Table 2. Critical cylinder radii with 2.5-cm-thick water reflector (5.5 g/cm^3 powder density)

PuO_2 content	Water content	Critical cylinder radius (cm)
30%	1%	22.16
	5%	16.43
12.5%	1%	39.52
	5%	26.97

Table 3. Per cent fraction of fission, capture and flux in the proposed five-energy group structure

PuO_2 content	**30%**			**30%**		
Water content	**1%**			**5%**		
	Fission	**Capture**	**Flux**	**Fission**	**Capture**	**Flux**
> 100 keV	57.2	18.6	71.8	34.1	5.9	65.7
10 keV-100 keV	9.5	15.9	16.6	4.9	5.8	13.9
10 eV-10 keV	24.3	51.9	11.24	41.3	61.2	18.9
0.1 eV-10 eV	5.0	9.9	0.34	14.7	24.0	1.38
< 0.1 eV	4.0	3.7	0.02	5.0	3.1	0.04
PuO_2 content	**12.5%**			**12.5%**		
Water content	**1%**			**5%**		
	Fission	**Capture**	**Flux**	**Fission**	**Capture**	**Flux**
> 100 keV	47.1	14.6	63.3	25.5	4.8	59.6
10 keV-100 keV	8.5	16.3	19.2	3.1	5.0	14.3
10 eV-10 keV	34.8	59.1	17.1	40.1	55.7	23.2
0.1 eV-10 eV	6.0	8.2	0.47	24.5	31.7	2.81
< 0.1 eV	3.6	1.8	0.03	6.8	2.8	0.09

Table 4. Sensitivity coefficients of heavy isotopes in MOX powder

PuO_2 content	**30%**		**12.5%**	
Water content	**1%**	**5%**	**1%**	**5%**
$^{235}U(n,f)$	2.20E-2*	2.41E-2	6.31E-2	5.88E-2
$^{235}U(n,\gamma)$	-3.49E-3	-5.36E-3	-1.08E-2	-1.26E-2
$^{238}U(n,f)$	1.16E-1	7.84E-2	1.76E-1	1.14E-1
$^{238}U(n,\gamma)$	-8.41E-2	-1.10E-1	-2.11E-1	-2.03E-1
$^{239}Pu(n,f)$	6.49E-1	6.68E-1	5.78E-1	6.34E-1
$^{239}Pu(n,\gamma)$	-8.51E-2	-1.38E-1	-9.53E-2	-1.50E-1
$^{240}Pu(n,f)$	5.23E-2	3.29E-2	2.92E-2	1.66E-2
$^{240}Pu(n,\gamma)$	-2.23E-2	-4.66E-2	-2.66E-2	-6.39E-2
$^{241}Pu(n,f)$	1.55E-1	1.95E-1	1.53E-1	1.78E-1
$^{241}Pu(n,\gamma)$	-1.40E-2	-2.28E-2	-1.42E-2	-2.22E-2

* Read as 0.0220.

Table 5. Sensitivity coefficients of heavy isotopes in MOX powder in five-energy group structure

(PuO_2 content: 30%, water: 5%)

	$^{238}U(n,f)$	**$^{238}U(n,\gamma)$**	**$^{239}Pu(n,f)$**	**$^{239}Pu(n,\gamma)$**	**$^{240}Pu(n,f)$**	**$^{240}Pu(n,\gamma)$**	**$^{241}Pu(n,f)$**	**$^{241}Pu(n,\gamma)$**
> 100 keV	7.84E-2*	-1.03E-2	2.14E-1	-2.45E-3	3.77E-2	-2.65E-4	4.16E-2	-9.35E-5
10 keV-100 keV	1.75E-5	-1.11E-2	3.91E-2	-4.51E-3	7.73E-4	-1.21E-3	1.09E-2	-5.33E-4
10 eV-10 keV	7.44E-5	-7.50E-2	3.14E-1	-9.44E-2	1.92E-3	-2.25E-2	9.94E-2	-1.19E-2
0.1 eV-10 eV	-2.58E-5	-1.28E-2	8.27E-2	-2.61E-2	-4.28E-3	-2.11E-2	4.26E-2	-9.05E-3
< 0.1 eV	-1.29E-4	-2.75E-4	2.11E-2	-8.71E-3	-4.45E-3	-3.68E-3	3.67E-4	-3.73E-3

*Read as 0.0784.

Table 6. Difference of k_{eff} with JEF-2.2 and ENDF/B-VI.8 with respect to JENDL-3.3

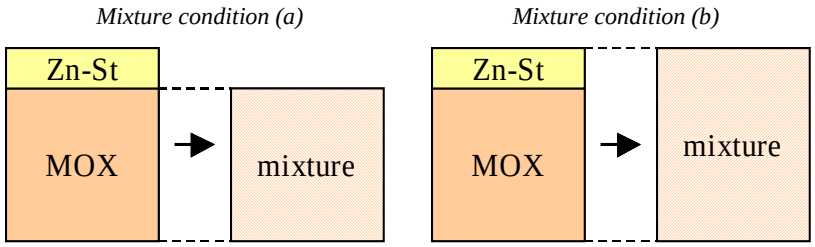
PuO_2 content	Water content	JEF-2.2	ENDF/B-VI.8
30%	1%	-1.51E-3*	-3.79E-4
	5%	-4.17E-3	-3.61E-3
12.5%	1%	-7.81E-4	2.77E-3
	5%	-3.38E-3	-3.41E-3

* Read as -0.00151.

Effect of mixture condition on criticality of MOX powder

Additives such as zinc stearate as well as moisture adsorbed in MOX powder act as neutron moderators. Additive content is one of the important control items for assuring the criticality safety of MOX powder during the homogenisation process. In a criticality safety evaluation of MOX powder containing additives, the additives are assumed to be mixed with MOX powder without changing the density of the powder if the additive is small enough in quantity to be adsorbed in a void space of the powder. For example, when zinc stearate is added to a MOX powder of 5.5 g/cm^3 and they are mixed homogeneously, the MOX density is assumed not to change from 5.5 g/cm^3. Since the theoretical density of MOX fuel is approximately 11 g/cm^3, the volume fraction of the void space in the powder is 50% (= 1-5.5/11). The particle size of zinc stearate is known to be much lager than that of MOX powder. Thus, additives mixed with MOX powder probably expand the volume of the powder instead of entering the void space. That is, the density of the MOX powder mixed with additives may decrease. Figure 4 shows two extreme assumptions concerning the mixture condition. Condition "(a)" assumes that MOX powder completely accommodates all zinc stearete in its void space. Condition "(b)" assumes that the volume of the mixture is equal to the sum of them before mixing. From the viewpoint of the powder technology and experiences of powder handling, a realistic mixture condition is closer to the condition "(b)". Here, it is discussed how the mixture condition affects the criticality of MOX powder.

Figure 4. Two assumptions of mixture condition



K_{eff} were calculated for a bare sphere of MOX powder of 600 kg with ONEDANT. The density of MOX powder before mixing was 5.5 g/cm^3. The weight fraction of hydrogen in zinc stearate is very close to that of water. Zinc stearate in MOX powder has almost the same reactivity effect with the same content of water. Therefore, the weight fraction of zinc stearate is dubbed as "water content", and the calculations were carried out by replacing zinc stearete with the same weight of water. Figures 5 and 6 show k_{eff} versus water content for two mixture assumptions. These results indicate that the mixture condition (a) gives much higher k_{eff} than the condition (b), especially for higher PuO_2 content. In most criticality safety evaluations, assumption (a) may be adopted. However, this assumption leads to a too-conservative design for the homogenisation process. Criticality safety design for the homogenisation process can be relaxed by introducing a realistic mixing state of MOX powder and additives. Besides criticality experiments for low-moderated MOX powder, experiments on mixture condition could be useful for reasonable criticality safety design of MOX powder.

Figure 5. k_{eff} vs. water content for PuO_2 content 30%

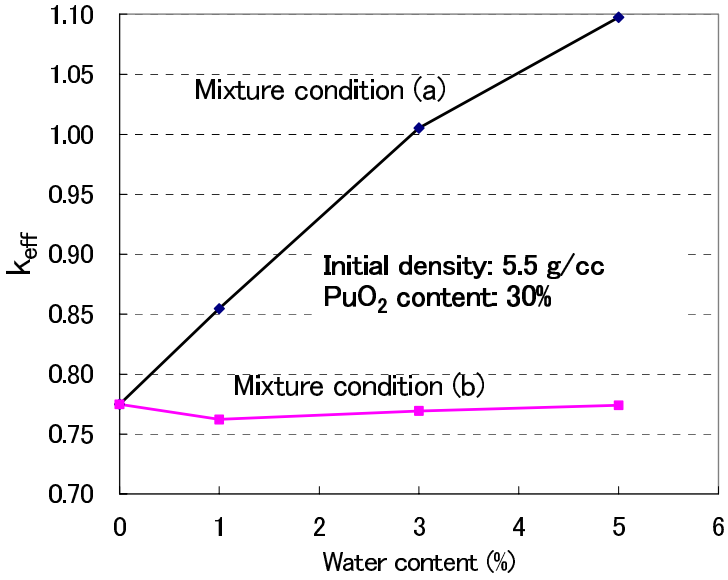
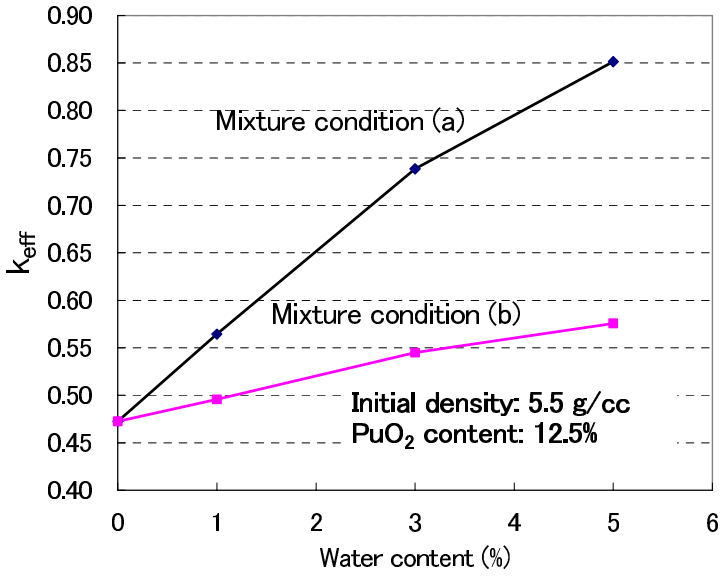


Figure 6. k_{eff} vs. water content for PuO_2 content 12.5%



Conclusions

- Bierman's experiments with homogeneous MOX blocks should be evaluated precisely; the experiments could revive as precious benchmark data.
- Dependence of calculated k_{eff} on cross-section libraries JENDL-3.3, JEF-2.2 and ENDF/B-VI.8 is not great for the MOX powder homogenisation process.
- Discussions from the aspect of the powder technology are important for the criticality safety evaluation of MOX powder.

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LINKS AMONG AVAILABLE INTEGRAL BENCHMARKS AND DIFFERENTIAL DATA EVALUATIONS, COMPUTATIONAL BIASES AND UNCERTAINTIES, AND NUCLEAR CRITICALITY SAFETY BIASES ON POTENTIAL MOX PRODUCTION THROUGHPUT

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Abstract

Through the use of Oak Ridge National Laboratory's recently developed and applied sensitivity and uncertainty computational analysis techniques, this paper presents the relevance and importance of available and needed integral benchmarks and differential data evaluations impacting potential MOX production throughput determinations relative to low-moderated MOX fuel blending operations.

The relevance and importance in the availability of or need for critical experiment benchmarks and data evaluations are presented in terms of computational biases as influenced by computational and experimental sensitivities and uncertainties relative to selected MOX production powder blending processes. Recent developments for estimating the safe margins of subcriticality for assuring nuclear criticality safety for process approval are presented. In addition, the impact of the safe margins (due to computational biases and uncertainties) on potential MOX production throughput will also be presented.

Introduction

For the past five or so years the Organisation of Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA) Working Party on Nuclear Criticality Safety (WPNCS) Expert Group on Experimental Needs has been considering the possible need for mixed plutonium and uranium oxide (MOX) experiments. During the course of these considerations various shortcomings in nuclear data have been highlighted, specifically the lack of critical experiment data for poorly-moderated fissionable material systems of low-enriched uranium and/or plutonium. The results of this lack of information are unknown computational biases and relatively large computational uncertainties. Industries and regulators have presumed to have accommodated these suspected biases and uncertainties by limiting the masses and geometries of process systems. However, new developments as concerns the use of weapons-grade plutonium and uranium oxide (WGMOX) in the United States and re-examination of production efficiencies in Europe for reactor-grade plutonium and uranium oxide (RGMOX) have prompted the OECD/NEA Nuclear Science Committee (NSC) to investigate and document the need for critical experiments in poorly-moderated regimes for WGMOX and RGMOX. As part of this investigation, the NSC has solicited various OECD member (and other) nations to review the issues of isotopic compositions and densities of WGMOX and RGMOX and to provide analyses of issues and proposals for critical experiment studies that will define the computational biases and uncertainties. Documentation of the computational biases and uncertainties will then provide the framework for needed OECD/NEA WPNCS technical basis reports that may be referenced in domestic and international standards, regulations and industrial safety analyses for MOX systems.

The Oak Ridge National Laboratory (ORNL) has been developing specialised sensitivity and uncertainty computational analysis capabilities as part of the US Department of Energy (DOE) Nuclear Criticality Safety Program (NCSP) task entitled *Applicable Ranges of Bounding Curves and Data* (AROBCAD). Within the AROBCAD task is a requirement to implement the developing capabilities to address DOE production/safety issues. The DOE has a vested interest in assuring cost effective – and safe – process operations with poorly-moderated plutonium and to stimulate/support efficient processes at the proposed US MOX Fuel Fabrication Facility undergoing US Nuclear Regulatory Commission licensing. Because of the AROBCAD task requirement to apply ORNL's developing technology and because of the relevance of the OECD initiative to DOE interests, ORNL has provided computational sensitivity and uncertainty evaluations in support of the OECD/NEA WPNCS and NSC investigative initiative by providing evaluations of some WPNCS specified WGMOX and RGMOX systems for comparison with proposed critical experiments and the identification of other potentially needed critical experiments. This report provides the results of those evaluations.

Background

The main characteristics of MOX powder applications are listed in Table 1. Due to the nature of the process, some important parameters such as powder density, etc., can assume a range of values. In order to better understand and analyse the similarity between the application processes and the

Table 1. MOX powder application characteristics

Parameter	RGMOX	WG MOX
PuO_2 content, wt.%	30-12.5	22-6.5
Powder density, g/cm^3	4.6-5.5	5.5
Water content, wt.%	3-5	1-5
Uranium enrichment, wt.%	$^{235}U \leq 1.2$	$^{235}U \leq 1.2$
Plutonium composition, wt.%	$^{240}Pu \geq 17$	$^{240}Pu \sim 4$

proposed experiment, several configurations with different parameter values have been created. These configurations are listed in Table 2 and are referred to as the "applications" for the rest of this paper. These configurations were generated by using bounding values as well as a mid-point value of the parameters that are given by a range of values. The first 27 applications in the table correspond to RGMOX, whereas the last 9 correspond to WGMOX. The applications are assumed to be powder-water mixture spheres of a given radius with a 20-cm water reflector. For a given PuO_2 content and powder density, the radius of the sphere increases as water is added to the system. The radii of the applications were selected to yield a calculated k_{eff} of approximately 1.0. The energy of average lethargy-causing fission (EALF), the average energy-causing fission (AEF) and the k_{eff} values for the applications were obtained using the XSDRN [1] module of the SCALE [2] code system with the 238-group SCALE cross-section library and are also listed in Table 2.

Table 2. MOX powder application configurations

Application number	PuO_2 content, wt.%	Powder density, g/cm^3	Water content, wt.%	Radius, cm	EALF, eV	AEF, eV	Calculated k_{eff}
1	12.5	4.6	3	57.4	311	654 600	0.9900
2	12.5	4.6	4	58.0	179	599 800	0.9987
3	12.5	4.6	5	58.6	109	552 300	1.0069
4	12.5	5.05	3	57.3	367	660 900	1.0130
5	12.5	5.05	4	58.0	205	605 100	1.0192
6	12.5	5.05	5	58.7	121	556 700	1.0254
7	12.5	5.5	3	57.3	422	665 900	1.0329
8	12.5	5.5	4	58.0	229	609 300	1.0369
9	12.5	5.5	5	58.7	133	560 300	1.0412
10	21.25	4.6	3	37.6	379	668 000	0.9734
11	21.25	4.6	4	38.0	248	616 000	0.9832
12	21.25	4.6	5	38.4	167	571 300	0.9909
13	21.25	5.05	3	37.6	472	676 500	1.0056
14	21.25	5.05	4	38.0	299	623 300	1.0128
15	21.25	5.05	5	38.4	196	577 600	1.0182
16	21.25	5.5	3	37.5	570	683 400	1.0347
17	21.25	5.5	4	38.0	350	629 200	1.0393
18	21.25	5.5	5	38.5	225	582 700	1.0423
19	30	4.6	3	29.9	482	695 300	0.9611
20	30	4.6	4	30.0	331	643 200	0.9716
21	30	4.6	5	30.3	234	598 300	0.9798
22	30	5.05	3	29.6	615	705 500	0.9976
23	30	5.05	4	30.0	410	651 900	1.0059
24	30	5.05	5	30.3	282	605 900	1.0119
25	30	5.5	3	29.6	759	713 800	1.0313
26	30	5.5	4	30.0	492	659 000	1.0371
27	30	5.5	5	30.4	330	612 100	1.0409
28	6.5	5.5	1	63.2	694	778 000	0.9232
29	6.5	5.5	3	65.0	133	607 600	0.9984
30	6.5	5.5	5	66.7	37	491 000	1.0563

Table 2. MOX powder application configurations (*cont.*)

Application number	PuO_2 content, wt.%	Powder density, g/cm^3	Water content, wt.%	Radius, cm	EALF, eV	AEF, eV	Calculated k_{eff}
31	14.25	5.5	1	37.0	659	757 300	0.9480
32	14.25	5.5	3	38.0	217	611 300	1.0018
33	14.25	5.5	5	39.0	83	509 800	1.0406
34	22	5.5	1	29.2	892	781 200	0.9658
35	22	5.5	3	30.0	335	638 200	1.0099
36	22	5.5	5	30.8	143	538 100	1.0423

Sensitivity and uncertainty analysis

The sensitivities of each application to the selected nuclide reaction pairs are calculated using the TSUNAMI-1D [3] sequence of the SCALE code system. TSUNAMI-1D utilises the XSDRN module to calculate the adjoint and forward fluxes that are then used to calculate the sensitivity coefficients. For the MOX powder applications considered in this paper, the most important nuclide reaction pairs are ^{239}Pu fission, ^{240}Pu capture, ^{235}U fission and ^{238}U capture. The sensitivity values for these nuclide reaction pairs are shown in Figures 1-4 for all applications. The applications are more sensitive to ^{239}Pu fission and ^{238}U capture than ^{240}Pu capture and ^{235}U fission. This is expected as there are more atoms of ^{239}Pu and ^{238}U per unit volume in the applications than there are ^{240}Pu and ^{235}U atoms.

The applications were analysed against the proposed experiment for similarity using the integral parameters g [4,5], c_k and E_{sum} [6]. The proposed experiment contains 30 wt.% PuO_2 in the RGMOX (25.2 wt.% ^{240}Pu) fuel rods that are placed in a 1.07-cm square-pitched 31×31 array inside a parallelepiped tank partially filled with water (characteristics of the proposed experiment have been defined in a white paper entitled "Project of Critical Experiments for Nuclear Criticality Codes Validation for Low-moderated 'MOX' Fissile Media" by the *Institut de Radioprotection et de Sûreté Nucléaire*, Department of Prevention and Studies of Accidents).

Figure 1. ^{239}Pu fission sensitivity

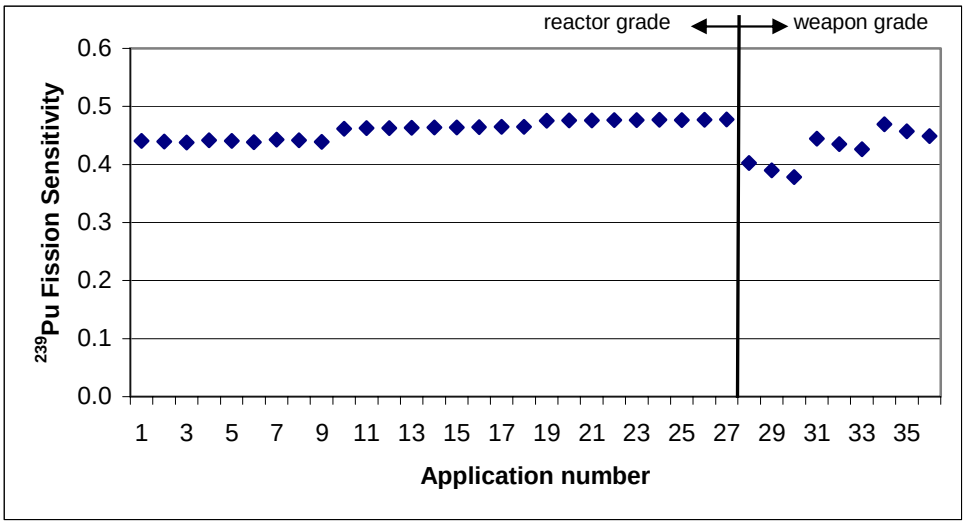


Figure 2. ^{240}Pu capture sensitivity

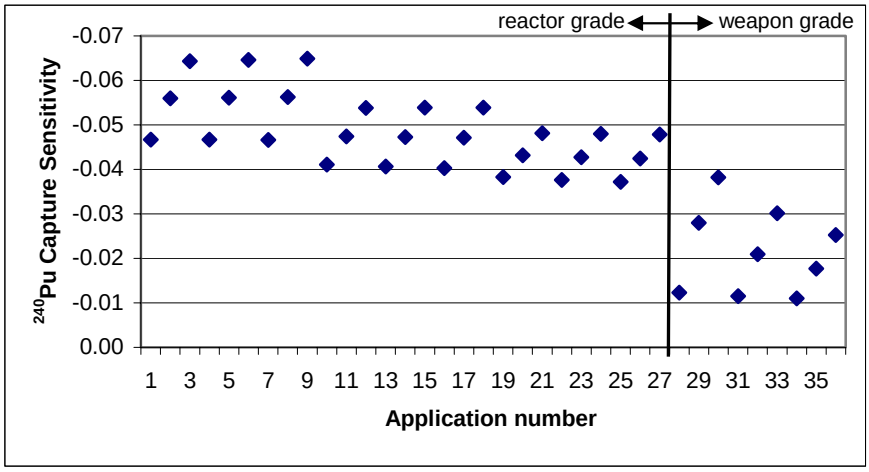


Figure 3. ^{235}U fission sensitivity

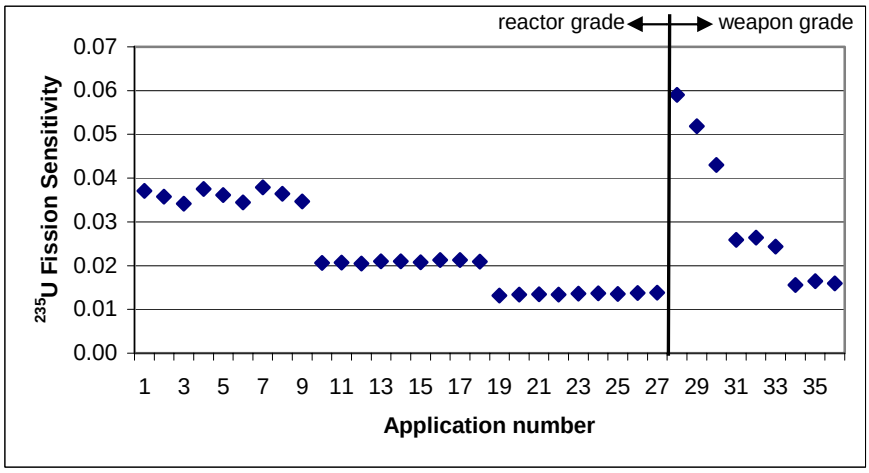
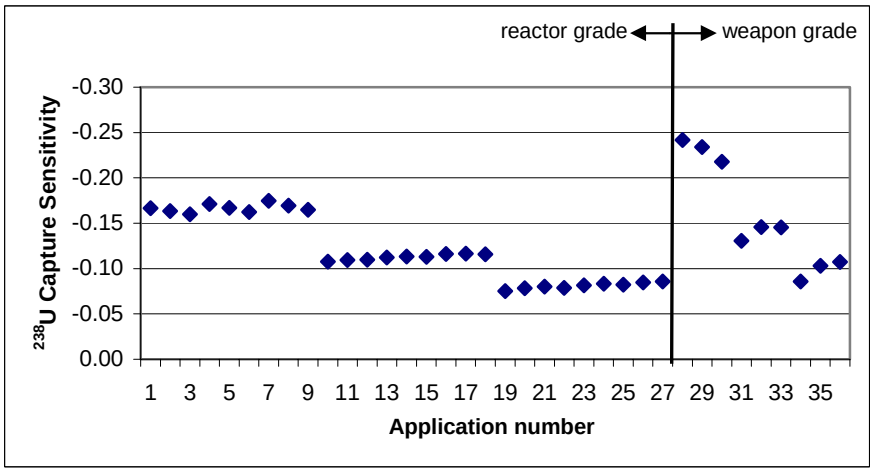


Figure 4. ^{238}U capture sensitivity



The differences between the application and benchmark sensitivities are indicative of how well the benchmark covers the application. Coverage is defined as having one or more benchmarks with sensitivities greater than the application's sensitivity for a specific nuclide, reaction and energy group. If the differences are small, then the benchmarks are considered adequate for the application for validation purposes, and the effect of the uncertainties in the cross-section data would be quantified by trending analysis. If the differences are large, then the benchmarks are not adequate for the application for validation purposes and the computational penalty that is generated is not appropriate. The penalty assessment methodology used in this work is based on the assumption that a benchmark with a greater sensitivity for the nuclide, reaction and energy group triplet of interest sufficiently covers the triplet in the application. The approach used in this method is to determine the differences between the application and benchmark sensitivities for all triplets that are not covered, and to quantify the importance of this non-coverage in terms of its final effect on the k_{eff} value of the application as a computational penalty after utilising the cross-section uncertainties.

The integral parameter *g* is defined as the normalised difference in sensitivities between the application and each benchmark. A large *g* value indicates that the covered part of the application's sensitivity for a specific nuclide reaction pair makes up the majority of the application's sensitivity for that nuclide reaction pair.

The E_{sum} parameter is a quantity that identifies similarity between two systems based only on the magnitude and shape of the sensitivity profiles for fission, capture and scatter. The integral parameter c_k is a quantity that relates the two systems and measures the similarity of the systems in terms of uncertainty, not just sensitivity.

In order to determine the integral parameters, a set of benchmarks has been created. This set of benchmarks is comprised of the proposed experiment and 95 selected benchmarks from the International Handbook of Evaluated Criticality Safety Benchmark Experiments [7]. The values of *g* between each application and the set of benchmark experiments have been calculated for ^{239}Pu fission, ^{240}Pu capture, ^{235}U fission and ^{238}U capture pairs. The maximum *g* values (maximum value calculated against all benchmarks) and the EALF values for each application are shown in Figures 5-8. For ^{239}Pu fission and ^{235}U fission, the *g* values indicate that the proposed experiment provides no additional

Figure 5. Maximum *g* values for ^{239}Pu fission

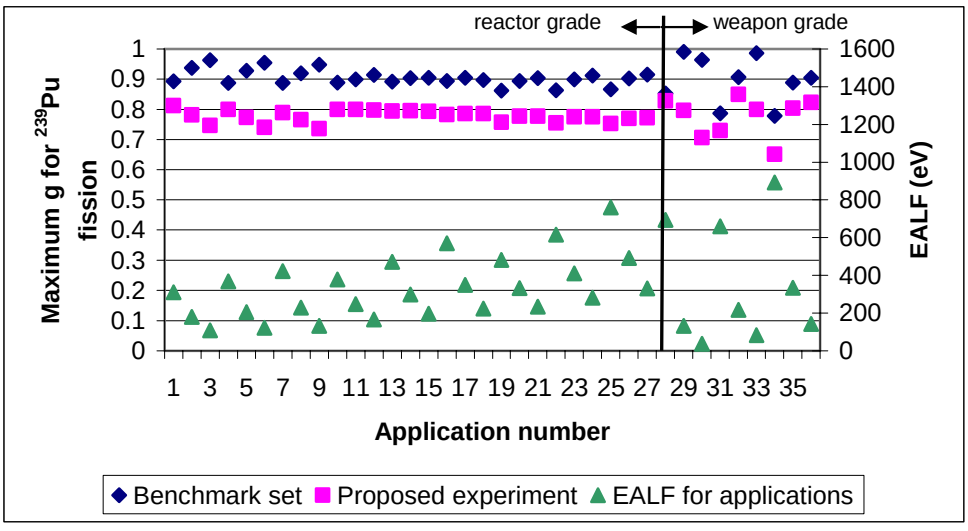


Figure 6. Maximum *g* values for ^{240}Pu capture

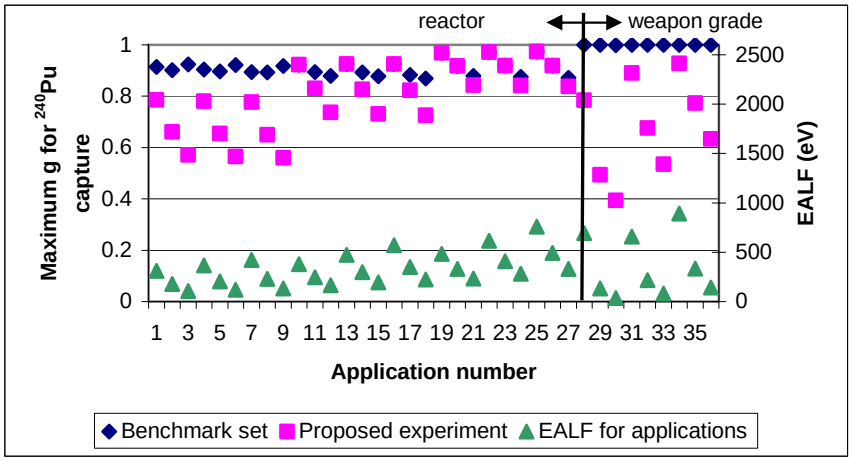


Figure 7. Maximum *g* values for ^{235}U fission

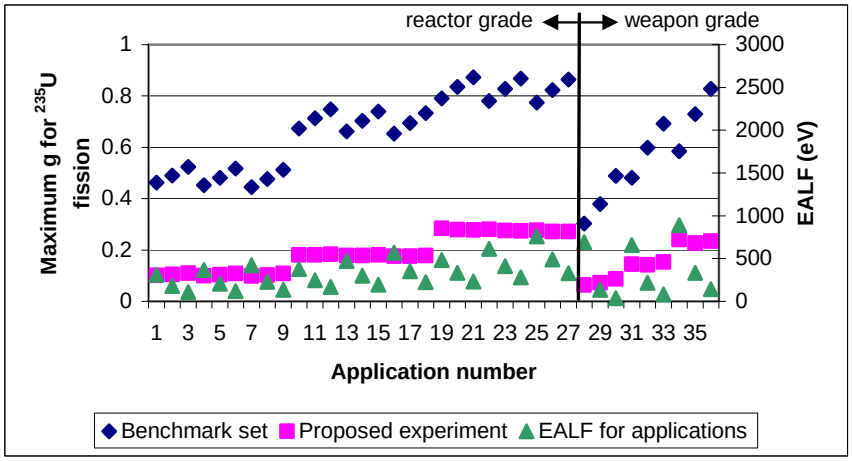
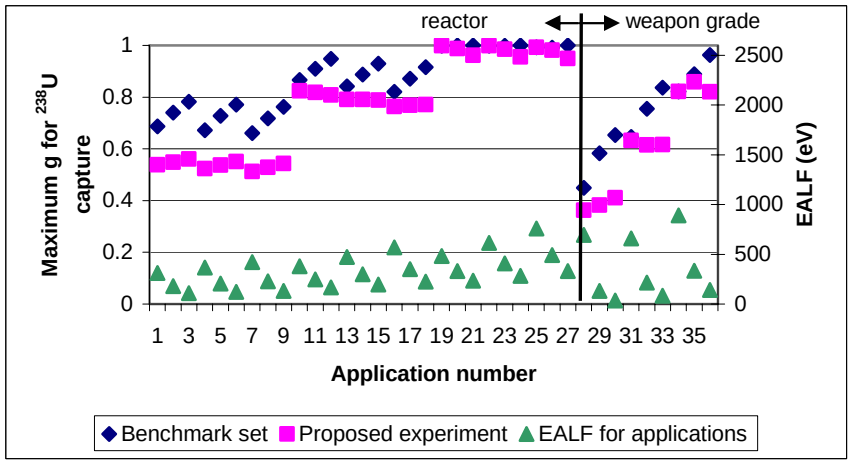


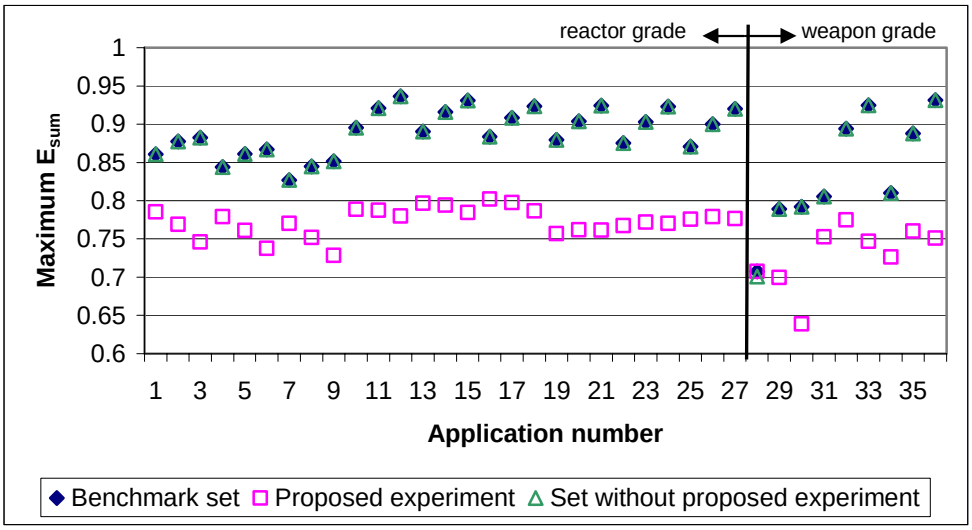
Figure 8. Maximum *g* values for ^{238}U capture



coverage beyond the coverage provided by the benchmark set. For ^{240}Pu capture and ^{238}U capture, the proposed experiment provides more coverage than the rest of the benchmarks in the set for some of the applications. When only the water content is varied, coverage with the benchmark set increases as the system becomes more thermal. On the other hand, the harder the neutron spectrum in the application, in general, the closer the coverage with the proposed experiment (EALF = 410 eV) to the coverage with the benchmark set. This indicates that although the proposed experiment does not provide better coverage than the benchmark set, it approaches the benchmark set as the neutron spectrum of the application becomes harder. Another observation that can be made is that as the neutron spectrum in the RGMOX applications become harder, the coverage for Pu and U isotopes with the proposed experiment generally gets better. This is expected since the experiment is designed to provide data for poorly-moderated systems. For WGMOX applications, no specific trend is observed.

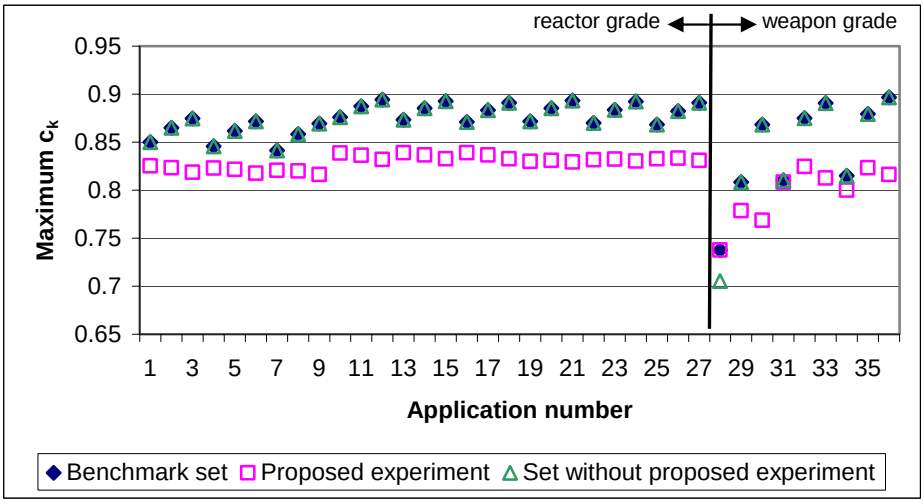
The E_{sum} values for all applications against the benchmark set as well as the values against the proposed experiment are shown in Figure 9. The E_{sum} values for RGMOX applications against the benchmark set are generally greater than 0.85, indicating that the applications are similar to the experiments in the benchmark set. The E_{sum} values for RGMOX applications against the proposed experiment, however, are generally lower than 0.8 indicating that the proposed experiment does not add any value to the existing benchmark set. This assessment is confirmed by plotting the E_{sum} values against the benchmark set without the proposed experiment in the same figure. It is clear from the figure that the set without the proposed experiment essentially yields the same E_{sum} values with the set with proposed experiment. Similar behaviour is observed for the WGMOX applications except the E_{sum} values are lower against both the benchmark set and the proposed experiment. Hence, neither the proposed experiment nor the other experiments in the benchmark set are sufficiently similar to the WGMOX applications.

Figure 9. Maximum E_{sum} values



The c_k values for all applications against the benchmark set as well as the values against the proposed experiment are shown in Figure 10. The c_k values for both the RGMOX and WGMOX applications against the benchmark set are generally greater than 0.85, indicating that the applications are similar to the experiments in the benchmark set. For RGMOX applications, the c_k values against the benchmark set without the proposed experiment are identical to the values against the benchmark set with the proposed experiment. This is an indication that the proposed experiment does not add any value to the existing benchmark set for RGMOX applications. This assessment is also confirmed by

Figure 10. Maximum c_k values



plotting the c_k values against the benchmark set without the proposed experiment in the same figure. It is clear from the figure that the proposed experiment yields lower c_k values than the set with or without proposed experiment. For WGMOX applications, similar behaviour is observed except the c_k values are lower for some of the WGMOX applications.

In calculating the c_k values that were used in the analysis, a standard deviation value of 0.15 has been assumed for all nuclides/isotopes that are not on the covariance data file with the exception of using a standard deviation value of 0.05 for χ for ^{238}U, ^{241}Pu and ^{242}Pu because these χ values are assumed to be known within 5%. To determine the effect of assumed standard deviation value of χ for ^{238}U, ^{241}Pu and ^{242}Pu on calculated c_k values, the calculations were repeated assuming a standard deviation value of 0.01. The c_k values from the calculations with a standard deviation of 0.01 for χ for ^{238}U, ^{241}Pu and ^{242}Pu were higher than the c_k values reported above. In addition, the c_k values with a standard deviation of 0.01 indicated that the proposed experiment was more similar to the applications than any other experiment in the benchmark set.

The reason behind different conclusions based on whether E_{sum} or c_k values with varying assumed standard deviation values are used can be better understood by considering their definitions. The E_{sum} values indicate the systems are not similar based only on their sensitivities. However, when the uncertainty in the cross-sections is included (i.e. c_k values), some of the nuclide reaction pairs that are important (high sensitivity) but not common between the two systems do not result in a smaller similarity index if they have small uncertainty values. Hence, although the g values indicate that the proposed experiment does not provide better coverage for important nuclide reaction pairs, and in terms of system-wide similarity the proposed experiment is not similar to the applications as implied by both the E_{sum} and c_k values, assuming small uncertainties for important nuclide reaction pairs may render the application systems similar to the proposed experiment.

Detailed analyses

To provide a more detailed analysis, six applications were selected. These include RGMOX applications 5, 14 and 23, and WGMOX applications 30, 32 and 34. Some selected nuclide reaction sensitivities as well as the EALF and AEF from these applications are listed in Table 3. The five-group, per cent flux, fissions and absorptions for the selected applications are listed in Table 4. A close look

at the per cent flux data reveals that almost half of the neutrons in the selected application systems are in the lowest neutron group (highest energy). Epithermal and fast neutron flux together make up approximately 60-80% of the neutrons.

Table 3. Sensitivity and spectral data for selected MOX powder application configurations

Nuclide	Reaction	Application number					
		5	14	23	30	32	34
^{1}H	Total	0.052	0.062	0.072	0.150	0.096	0.038
^{1}H	Scatter	0.054	0.063	0.072	0.154	0.097	0.039
^{1}H	Elastic	0.054	0.063	0.072	0.154	0.097	0.039
^{1}H	Capture	-0.001	-0.001	-0.001	-0.003	-0.001	0.000
^{16}O	Total	0.011	0.033	0.043	0.002	0.032	0.044
^{16}O	Scatter	0.016	0.038	0.047	0.008	0.037	0.048
^{16}O	Elastic	0.017	0.038	0.047	0.008	0.037	0.048
^{16}O	Capture	-0.005	-0.004	-0.004	-0.006	-0.005	-0.004
^{235}U	Total	0.023	0.014	0.009	0.024	0.016	0.011
^{235}U	Scatter	0.000	0.000	0.000	0.000	0.000	0.000
^{235}U	Elastic	0.000	0.000	0.000	0.000	0.000	0.000
^{235}U	Fission	0.036	0.021	0.014	0.043	0.026	0.016
^{235}U	Capture	-0.013	-0.008	-0.005	-0.020	-0.011	-0.005
^{235}U	Nubar	0.068	0.041	0.027	0.095	0.054	0.032
^{235}U	Chi	0.068	0.041	0.027	0.095	0.054	0.032
^{238}U	Total	-0.059	-0.006	0.014	-0.111	-0.018	0.039
^{238}U	Scatter	0.029	0.043	0.044	0.030	0.049	0.047
^{238}U	Elastic	0.032	0.031	0.027	0.032	0.036	0.031
^{238}U	Fission	0.077	0.062	0.051	0.075	0.076	0.077
^{238}U	Capture	-0.167	-0.113	-0.082	-0.218	-0.146	-0.086
^{238}U	Nubar	0.124	0.097	0.078	0.124	0.119	0.116
^{238}U	Chi	0.122	0.096	0.077	0.121	0.117	0.115
^{239}Pu	Total	0.266	0.298	0.319	0.161	0.260	0.339
^{239}Pu	Scatter	0.001	0.004	0.008	0.000	0.004	0.008
^{239}Pu	Elastic	0.001	0.003	0.005	0.000	0.002	0.005
^{239}Pu	Fission	0.441	0.464	0.477	0.378	0.435	0.469
^{239}Pu	Capture	-0.175	-0.170	-0.165	-0.217	-0.179	-0.138
^{239}Pu	Nubar	0.791	0.837	0.863	0.782	0.823	0.844
^{239}Pu	Chi	0.792	0.837	0.863	0.782	0.823	0.845
^{240}Pu	Total	-0.042	-0.026	-0.016	-0.037	-0.017	-0.005
^{240}Pu	Scatter	0.002	0.003	0.004	0.000	0.000	0.001
^{240}Pu	Elastic	0.002	0.003	0.003	0.000	0.000	0.000
^{240}Pu	Fission	0.013	0.018	0.023	0.001	0.003	0.006
^{240}Pu	Capture	-0.056	-0.047	-0.043	-0.038	-0.021	-0.011
^{240}Pu	Nubar	0.019	0.027	0.034	0.002	0.005	0.008
^{240}Pu	Chi	0.019	0.027	0.033	0.002	0.005	0.008
Spectral data		Application number					
		5	14	23	30	32	34
EALF (eV)		204.9	298.7	409.9	36.9	216.6	891.9
AEF (keV)		605.1	623.3	651.9	491.0	611.3	781.2

Table 4. Per cent flux, fission, and absorption data for selected MOX powder application configurations

Per cent flux							
Group	Upper energy boundary (eV)	Application number					
		5	14	23	30	32	34
1	2.0E+07	49	45	42	49	45	44
2	1.0E+05	12	10	9	12	10	9
3	9.5E+03	18	15	13	20	15	10
4	10.0	6	7	7	7	7	7
5	0.1	15	23	28	12	23	30
Per cent fissions							
1	2.0E+07	27	30	33	20	29	42
2	1.0E+05	3	4	5	2	4	6
3	9.5E+03	36	34	31	30	32	20
4	10.0	21	17	15	34	18	12
5	0.1	13	16	17	15	17	21
Per cent absorptions							
1	2.0E+07	13	13	14	10	13	17
2	1.0E+05	4	4	3	3	4	5
3	9.5E+03	36	30	25	35	29	15
4	10.0	22	17	15	30	16	11
5	0.1	26	36	42	22	39	52

Sensitivity profiles for selected nuclide reaction pairs are plotted against the composite profiles for the same nuclide reaction pairs for each selected application in Figures 11-16. A composite profile is one that is created by comparing, on a group-wise basis, the sensitivity value of an application with all benchmarks, and selecting the sensitivity value of the benchmark that is closest to the application's value. If there exist some benchmarks that have larger sensitivity values than the application, then the sensitivity value of the composite profile is set equal to the application's sensitivity value. Thus, the composite profile shows the energy groups where none of the benchmarks have sensitivities as great as the application, and how the sensitivity of the most sensitive benchmark for this energy group compares to the sensitivity of the application.

In all six selected applications, the composite sensitivity profiles for ^{239}Pu fission and ^{240}Pu capture are almost identical to the sensitivity profile of the application except ^{239}Pu fission for application 34. This implies that the benchmarks in the benchmark set all together provide complete coverage for the plutonium reactions of interest. In terms of ^{235}U fission and ^{238}U capture, except for ^{238}U capture in application 23, the opposite is true in that none of the composite profiles indicate good coverage. The total sensitivity values (summed over all neutron energy groups) for ^{239}Pu fission, ^{240}Pu capture, ^{235}U fission and ^{238}U capture from the selected applications are listed in Table 5 along with the composite sensitivity values from the benchmark set with and without the proposed experiment. The last column in the table is the difference in the composite profile sensitivity values between the complete benchmark set and the benchmark set without the proposed experiment. The absolute value of the difference values are essentially zero with three-digit precision. This is an indication that the proposed experiment does not add value to the set for purposes of determining the composite profile and thereby determination of a computational penalty.

Figure 11. Comparison of composite and actual sensitivities for application 5

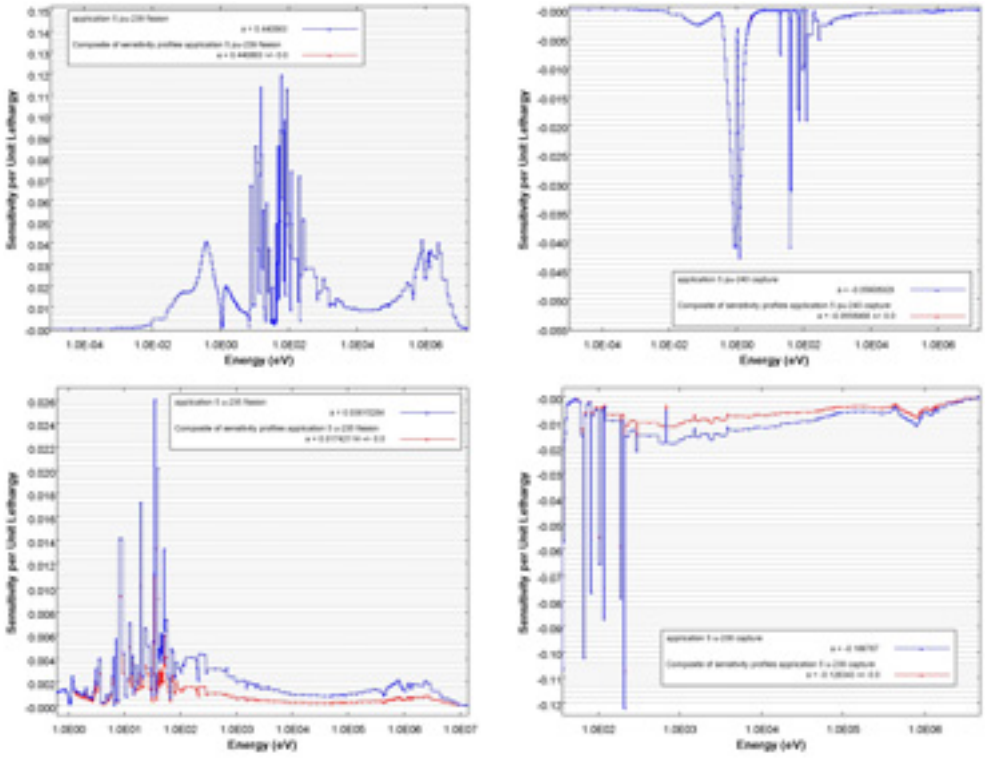


Figure 12. Comparison of composite and actual sensitivities for application 14

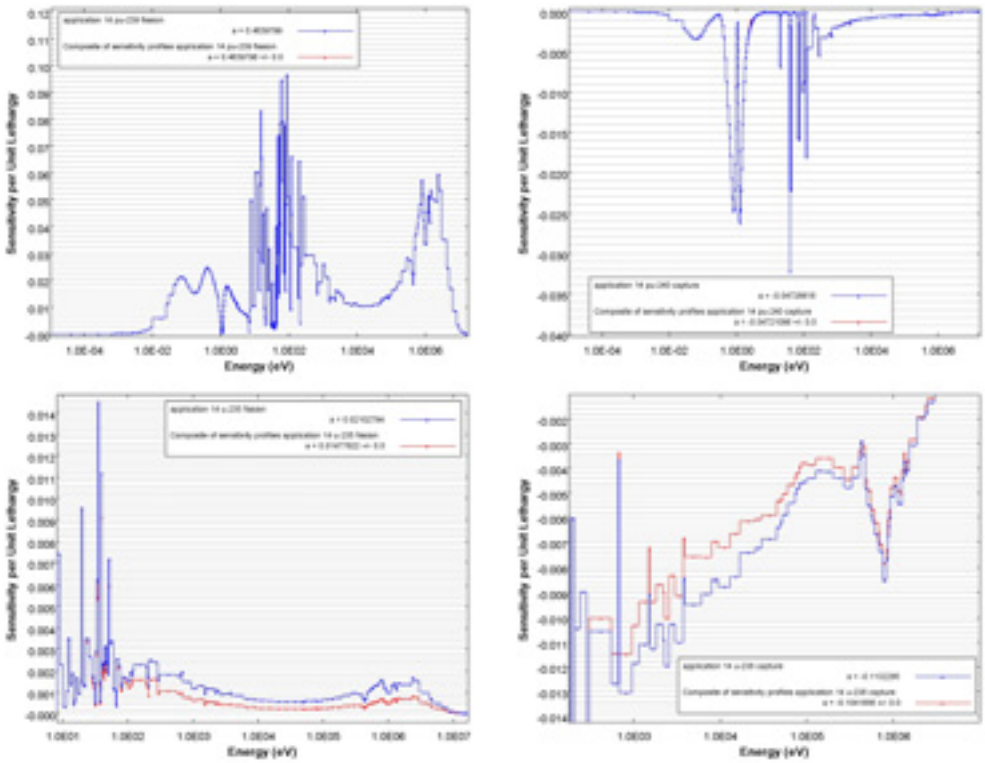


Figure 13. Comparison of composite and actual sensitivities for application 23

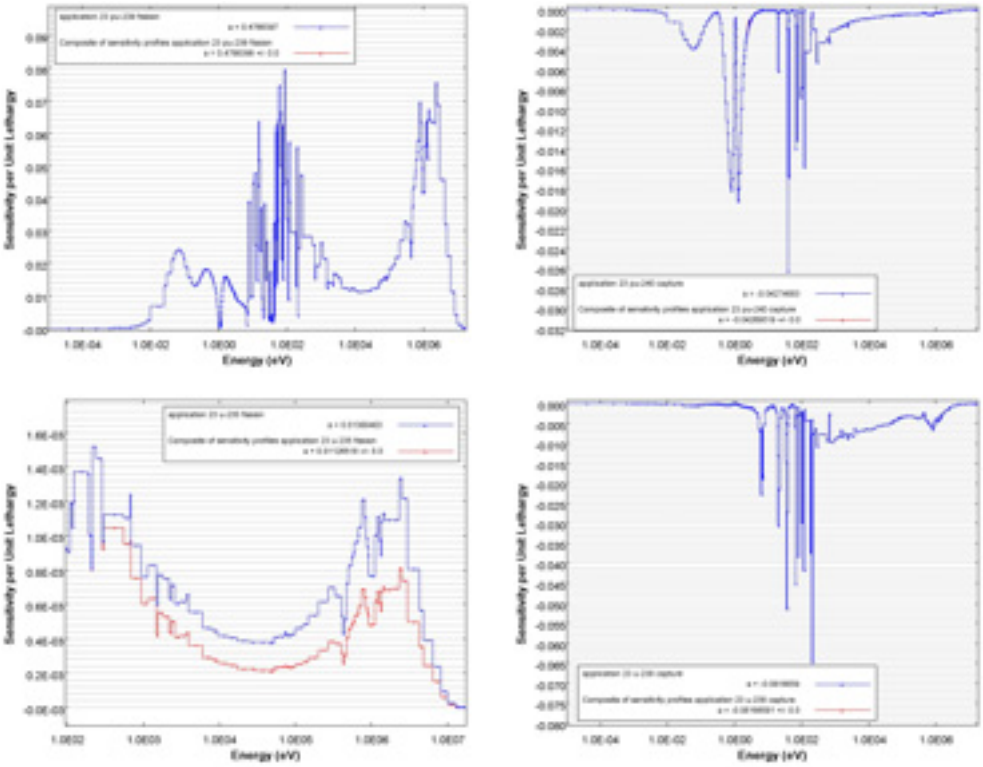


Figure 14. Comparison of composite and actual sensitivities for application 30

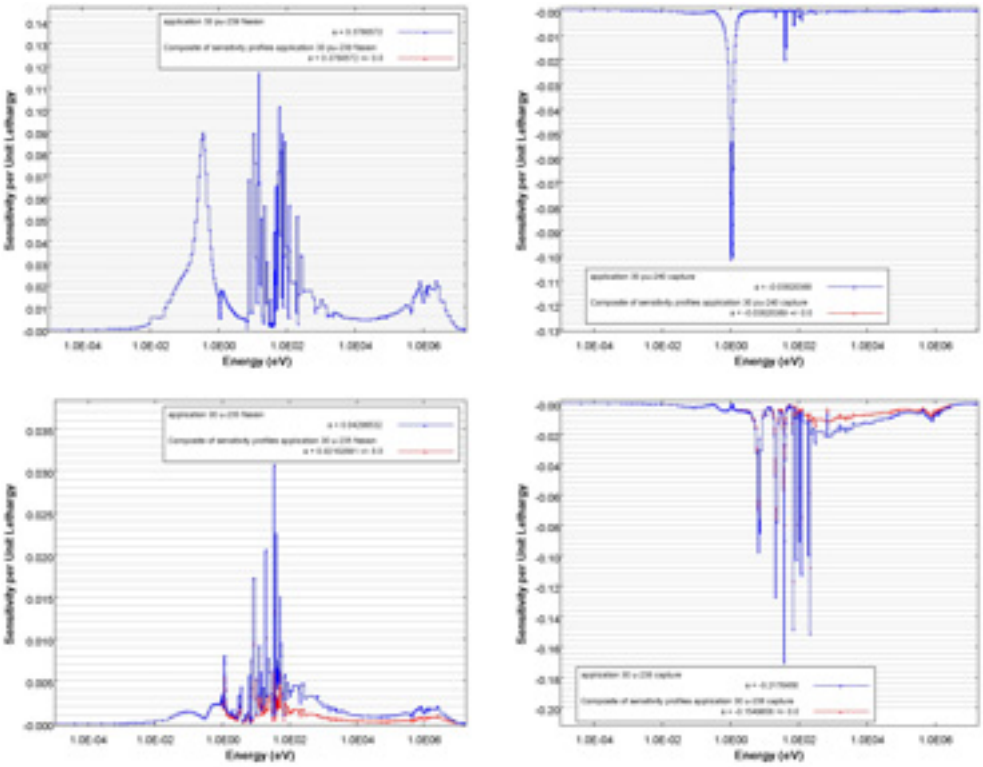


Figure 15. Comparison of composite and actual sensitivities for application 32

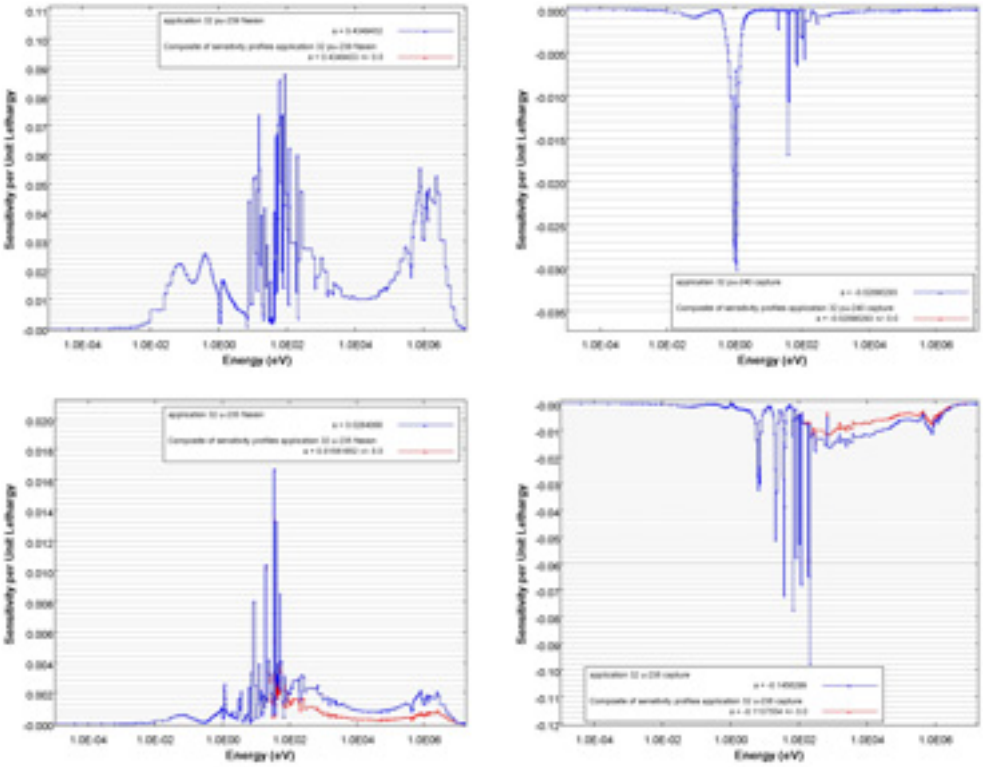


Figure 16. Comparison of composite and actual sensitivities for application 34

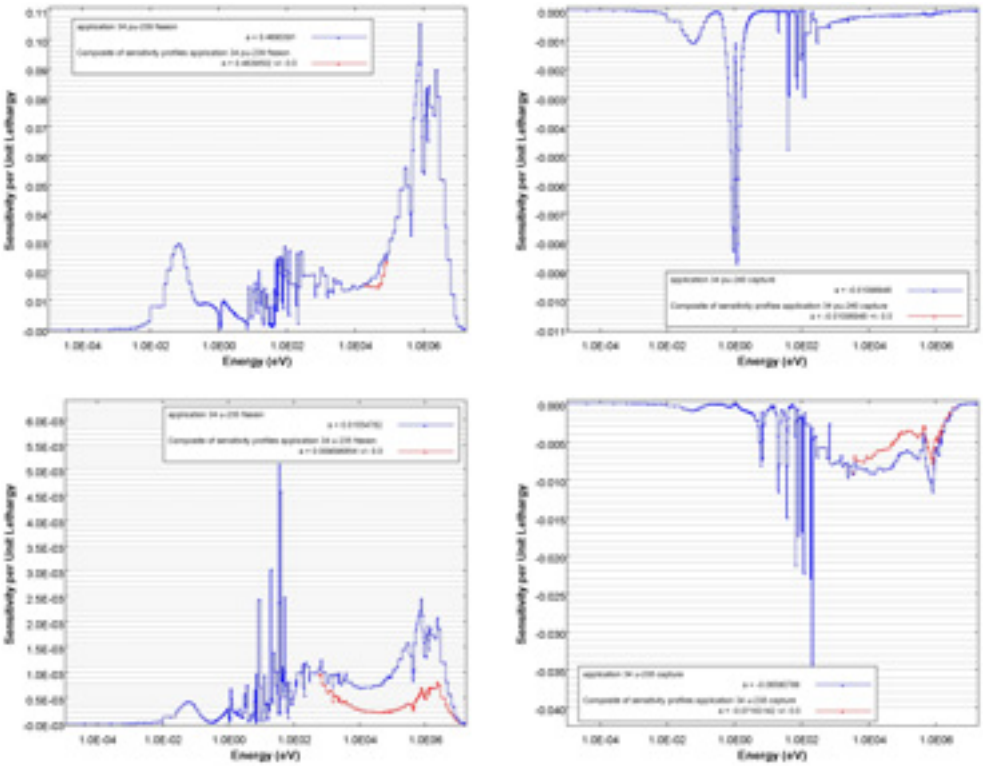


Table 5. Comparison of composite profiles with and without proposed experiment

Application 5				
Nuclide-reaction	**Application**	**Composite with complete set**	**Composite without proposed experiment**	**Difference**
^{235}U	0.036	0.017	0.017	0.000
^{238}U	-0.167	-0.126	-0.123	-0.003
^{239}Pu	0.441	0.441	0.441	0.000
^{240}Pu	-0.056	-0.056	-0.056	0.000
Application 14				
^{235}U	0.021	0.015	0.015	0.000
^{238}U	-0.113	-0.104	-0.101	-0.003
^{239}Pu	0.464	0.464	0.464	0.000
^{240}Pu	-0.047	-0.047	-0.047	0.000
Application 23				
^{235}U	0.014	0.011	0.011	0.000
^{238}U	-0.082	-0.082	-0.082	0.000
^{239}Pu	0.477	0.477	0.477	0.000
^{240}Pu	-0.043	-0.043	-0.042	-0.001
Application 30				
^{235}U	0.043	0.021	0.021	0.000
^{238}U	-0.218	-0.155	-0.151	-0.003
^{239}Pu	0.378	0.378	0.378	0.000
^{240}Pu	-0.038	-0.038	-0.038	0.000
Application 32				
^{235}U	0.026	0.016	0.016	0.000
^{238}U	-0.146	-0.114	-0.110	-0.003
^{239}Pu	0.435	0.435	0.435	0.000
^{240}Pu	-0.021	-0.021	-0.021	0.000
Application 34				
^{235}U	0.016	0.009	0.009	0.000
^{238}U	-0.086	-0.072	-0.068	-0.003
^{239}Pu	0.469	0.464	0.464	0.000
^{240}Pu	-0.011	-0.011	-0.011	0.000

The computational penalty for the selected application configurations are listed in Table 6 as a function of the standard deviation for the cross-section values that do not have any covariance data. These values are per cent relative calculated penalty values with the benchmark set that is used and represent the additional computational bias as percentage of the calculated k_{eff} values. Note that the computational penalty is in addition to any computed or experimental bias due to the method, cross-sections, etc. It has been introduced to allow for lack of coverage, i.e. inadequacy of the benchmark set to sufficiently represent the application. The values in the last column are calculated by using a standard deviation value of 0.15 for all nuclides/isotopes that are not on the covariance data file with the exception of using standard deviation value of 0.05 for χ for ^{238}U, ^{241}Pu and ^{242}Pu since these values are assumed to be known within 5%. When the benchmark set contains benchmark experiments that are very similar to the application, the calculated penalty due to non-coverage is small. The non-coverage data for the selected applications are listed in Tables 7-12. These data indicate that for RGMOX, the non-coverage of ^{235}U fission is greatest among the nuclide reaction pairs of interest. For WGMOX applications, however, the non-coverage of ^{238}U capture is the greatest.

Table 6. Per cent computational penalty for selected MOX powder application configurations

Application	Default standard deviation			
	0.0	**0.1**	**0.25**	**0.15***
5	0.31	0.41	0.73	0.34
14	0.18	0.19	0.24	0.18
23	0.25	0.25	0.26	0.25
30	0.30	0.42	0.81	0.33
32	0.23	0.32	0.61	0.25
34	0.63	1.07	2.26	0.77

* Covariance data for χ for ^{238}U, ^{240}Pu, and ^{241}Pu was set equal to 0.05 rather than default value of 0.15 since these values are assumed to be known with less than 5% error.

Table 7. Non-coverage for application 5

	^{239}Pu fission	**^{240}Pu capture**	**^{235}U fission**	**^{238}U capture**
Number of groups not covered	0	7	169	91
Sum of sensitivities not covered	0.00E+00	-1.97E-04	1.87E-02	-4.04E-02
Group with largest sensitivity	74	194	74	74
Largest sensitivity group value	1.66E-02	-2.35E-03	1.84E-03	-8.99E-03
Best experiment for max group	pci001	pcm001-05sen	mct002-02sen	mct002-02sen
Group sensitivity for best exp	3.91E-02	-6.67E-03	6.17E-04	-5.91E-03

Table 8. Non-coverage for application 14

	^{239}Pu fission	**^{240}Pu capture**	**^{235}U fission**	**^{238}U capture**
Number of groups not covered	0	5	106	69
Sum of sensitivities not covered	0.00E+00	-5.31E-05	6.25E-03	-9.04E-03
Group with largest sensitivity	74	81	74	74
Largest sensitivity group value	1.74E-02	-1.90E-03	1.06E-03	-6.23E-03
Best experiment for max group	pci001	french_exp	mct002-02sen	mct002-02sen
Group sensitivity for best exp	3.91E-02	-2.21E-03	6.17E-04	-5.91E-03

Table 9. Non-coverage for application 23

	^{239}Pu fission	**^{240}Pu capture**	**^{235}U fission**	**^{238}U capture**
Number of groups not covered	0	5	76	0
Sum of sensitivities not covered	0.00E+00	-5.01E-05	2.34E-03	0.00E+00
Group with largest sensitivity	74	74	74	74
Largest sensitivity group value	1.67E-02	-1.95E-03	6.62E-04	-4.51E-03
Best experiment for max group	pci001	french_exp	mct002-02sen	mct002-02sen
Group sensitivity for best exp	3.91E-02	-2.66E-03	6.17E-04	-5.91E-03

Table 10. Non-coverage for application 30

	^{239}Pu fission	^{240}Pu capture	^{235}U fission	^{238}U capture
Number of groups not covered	0	0	195	156
Sum of sensitivities not covered	0.00E+00	0.00E+00	2.20E-02	-6.27E-02
Group with largest sensitivity	131	190	133	74
Largest sensitivity group value	1.24E-02	-2.39E-03	2.01E-03	-1.07E-02
Best experiment for max group	pci001	pu-8-2sen	mct002-02sen	mct002-02sen
Group sensitivity for best exp	3.36E-02	-4.57E-03	1.09E-03	-5.91E-03

Table 11. Non-coverage for application 32

	^{239}Pu fission	^{240}Pu capture	^{235}U fission	^{238}U capture
Number of groups not covered	0	0	119	81
Sum of sensitivities not covered	0.00E+00	0.00E+00	1.06E-02	-3.19E-02
Group with largest sensitivity	74	194	74	74
Largest sensitivity group value	1.62E-02	-1.06E-03	1.35E-03	-7.74E-03
Best experiment for max group	pci001	pcm001-05sen	mct002-02sen	mct002-02sen
Group sensitivity for best exp	3.91E-02	-6.67E-03	6.17E-04	-5.91E-03

Table 12. Non-coverage for application 34

	^{239}Pu fission	^{240}Pu capture	^{235}U fission	^{238}U capture
Number of groups not covered	10	0	73	58
Sum of sensitivities not covered	5.09E-03	0.00E+00	6.45E-03	-1.43E-02
Group with largest sensitivity	11	194	74	60
Largest sensitivity group value	1.86E-02	-4.29E-04	5.80E-04	-3.63E-03
Best experiment for max group	pcm001-01sen	pcm001-05sen	mct002-02sen	mct002-02sen
Group sensitivity for best exp	5.73E-02	-6.67E-03	6.17E-04	-3.26E-03

Effect of ^{238}U cross-sections

Since non-coverage is greatest with ^{238}U capture, the effect of ^{238}U cross-sections are investigated in more detail for applications 1 (RGMOX) and 28 (WGMOX). These applications have been selected because they had some of the highest sensitivity values for ^{238}U capture. Note that except for ^{238}U cross-sections, all other cross-sections are from the 238-group SCALE cross-section library, which is based on ENDF/B-V evaluation except for ^{16}O, which is from ENDF/B-VI evaluation. In addition to the standard 238-group ^{238}U SCALE cross-sections that are based on ENDF/B-V, two additional calculations were performed with 1) ^{238}U cross-sections were replaced with ENDF/B-VI cross-section data that was evaluated by ORNL[8], and 2) ^{238}U cross-sections were replaced with ENDF/B-VI cross-sections. The new evaluation by ORNL was performed for ^{238}U cross-sections in the neutron energy range of 10^{-5}–$2\cdot10^{4}$ eV only.

The total and capture sensitivity values of ^{238}U from these calculations are listed in Table 13. The sensitivity values calculated using ENDF/B-V and ENDF/B-VI values are essentially identical. The newly evaluated ENDF/B-VI cross-sections, however, result in more than 3% difference (less sensitive) in ^{238}U total cross-section sensitivities and about 0.5% difference (less sensitive) in ^{238}U capture cross-section sensitivities. The difference in calculated k_{eff} values between the ORNL evaluated ENDF/B-VI values and ENDF/B-V values is less than 0.3%. However, there is no difference in calculated k_{eff} values between the ENDF/B-V values and the ENDF/B-VI values.

Table 13. Effect of ^{238}U cross-sections and evaluations for applications 1 and 28

	^{238}U total cross-section sensitivity				
	ENDF/B-V	ENDF/B-VI[1]	% Difference[2]	ENDF/B-VI[3]	% Difference[4]
Application1	-0.0470	-0.0454	-3.5	-0.0470	0.0
Application 28	-0.1089	-0.1054	-3.2	-0.1089	0.0
	^{238}U capture cross-section sensitivity				
Application1	-0.1665	-0.1656	-0.6	-0.1665	0.0
Application 28	-0.2419	-0.2406	-0.5	-0.2419	0.0

[1] ^{238}U cross-sections based on ENDF/B-VI and evaluated by ORNL.
[2] Per cent difference between ORNL evaluated ENDF/B-VI results and ENDF/B-V results.
[3] ^{238}U cross-sections based on ENDF/B-VI.
[4] Per cent difference between ENDF/B-VI results and ENDF/B-V results.

The sensitivity profiles for ^{238}U total and capture reactions for applications 1 and 28 are shown in Figures 17-20. From these figures it is clear that most of the differences are in the resonance energy range. Analysis of the maximum values of g for ^{238}U capture, E_{sum} and c_k indicate that the new evaluation results in improved similarity indices for the applications 1 and 28. Maximum g values are as much as 1.2% higher, while maximum E_{sum} and c_k values are as much as 0.4% higher. However, as the sensitivities for the benchmarks were not calculated using the newly evaluated ^{238}U cross-section values, the improvement in the integral indices can not be relied upon until more extensive work and subsequent analysis of the effects of ^{238}U evaluation can be performed.

Safe limits

The effect of computational bias and uncertainty as well as the computational penalty on MOX throughput was investigated. Radii of initially critical configurations (calculated $k_{eff} \sim 1.0$) for RGMOX and WGMOX were modified to asses the effect of magnitude of per cent safety limit on potential MOX throughput, i.e. maximum volume of MOX powder that would be allowed in a batch. Per cent decrease in volume of MOX powder as a function of per cent safety limit (in units of calculated k_{eff}) is shown in Figure 21 for selected RGMOX (application 2) and WGMOX (application 28) applications. For these applications that are initially critical, computational penalty due to non-coverage is 0.3% and 1.0%, respectively, assuming the standard deviation of the data that are not in the covariance data file that has been used is 0.15 and standard deviation of χ for ^{238}U, ^{241}Pu and ^{242}Pu is 0.05. Corresponding reduction in MOX batch volumes for RGMOX and WGMOX are 3.9% and 11.5%, respectively. For a standard deviation value of 0.25, the reduction in MOX batch volumes for RGMOX (application 2) and WGMOX (application 28) are 7% and 30%, respectively. From these results, it is clear that the bias and uncertainty in the cross-sections, calculated values, or penalty due to non-coverage considerably affect the volume of safe MOX powder batch. The effects are greater for WGMOX as they are more sensitive to changes in cross-sections due to higher ^{239}Pu content.

Figure 17. Comparison of ^{238}U total cross-section sensitivities for application 1

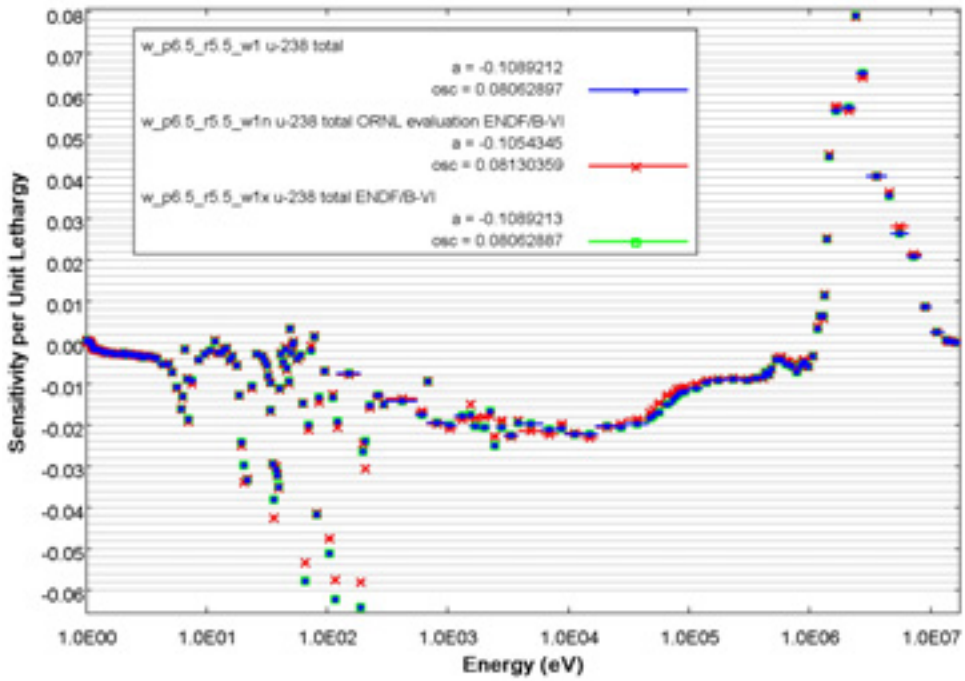


Figure 18. Comparison of ^{238}U capture cross-section sensitivities for application 1

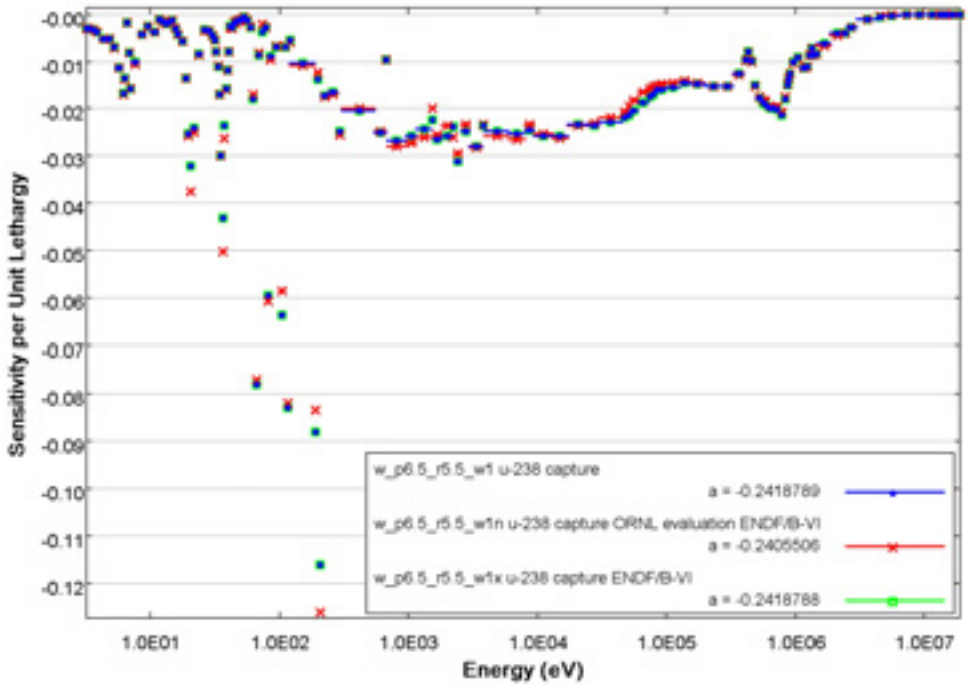


Figure 19. Comparison of ^{238}U total cross-section sensitivities for application 28

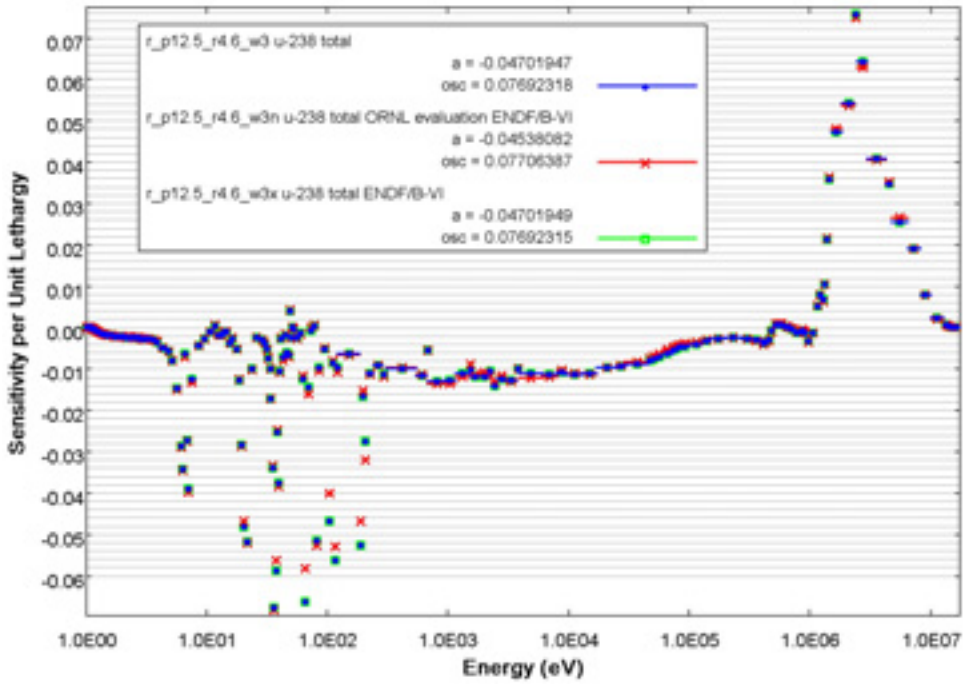


Figure 20. Comparison of ^{238}U capture cross-section sensitivities for application 28

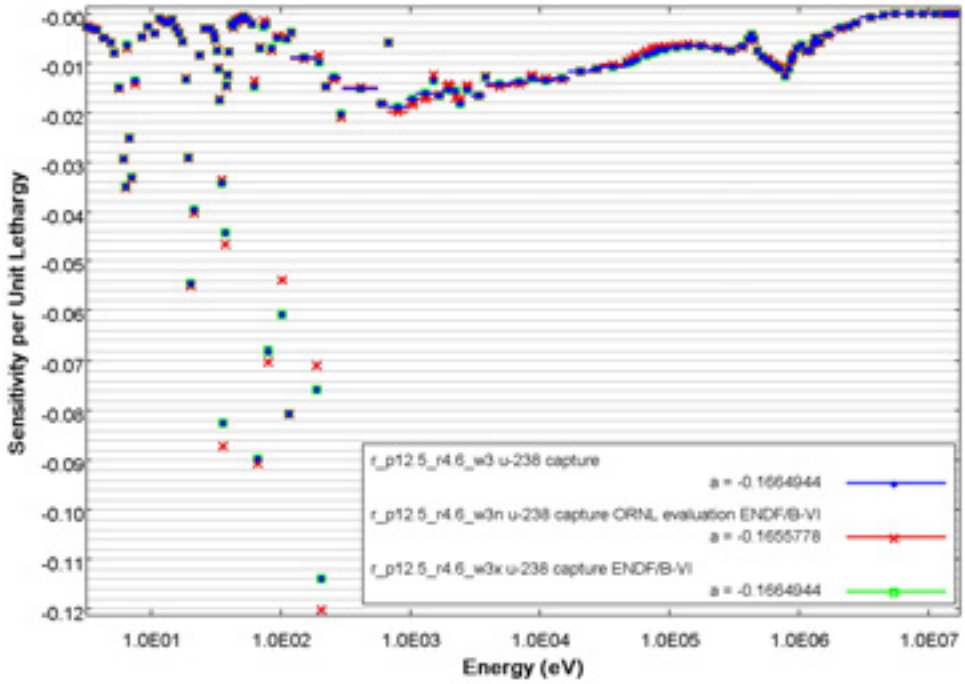
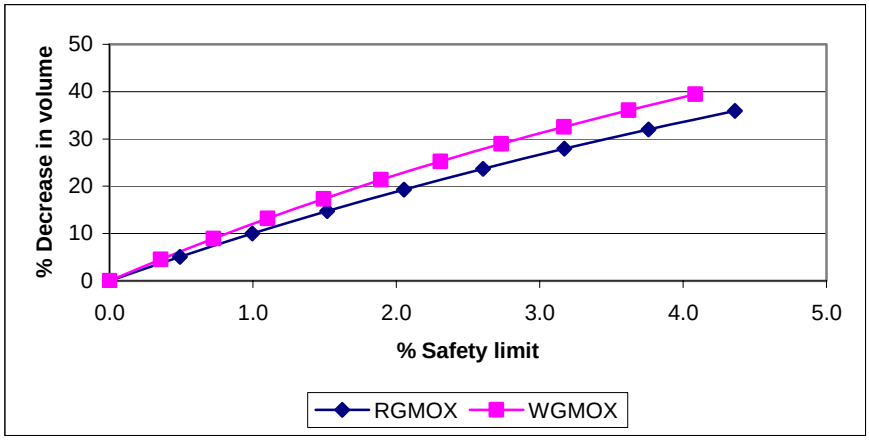


Figure 21. Per cent decrease in volume as a function of per cent safety limit



Summary and conclusions

Based on the range of system parameter values that are characteristic for RGMOX and WGMOX, a total of 36 application configurations were investigated to determine the applicability of the proposed experiment to the validation of low-moderated MOX fuel blending operations. Analyses of the application configurations were performed using state-of-the-art analysis tools that were developed at ORNL utilising sensitivity/uncertainty methodology. The similarity of the proposed experiment to the configurations that might be encountered during the MOX fuel blending process was assessed using integral parameters g, E_{sum} and c_k. Although system-wide similarity was observed between the proposed experiment and most of the application configurations, on a nuclide reaction level the proposed experiment was found to be insufficient for providing an additional basis for validation of the nuclide reaction pairs of interest, specifically ^{239}Pu fission, ^{240}Pu capture, ^{235}U fission and ^{238}U capture. It was also observed that the proposed experiment did not improve the similarity indices compared to the existing benchmarks. Although assuming smaller standard deviation for χ for ^{238}U, ^{241}Pu and ^{242}Pu improves the similarity index c_k against the proposed experiment, the contradictory indications by E_{sum} and g values imply that actual χ for ^{238}U, ^{241}Pu and ^{242}Pu is not known to within 1%.

The status of existing benchmarks for validation of MOX fuel blending processes was assessed by determining the computational penalty due to non-coverage of the nuclide reaction sensitivities of the MOX powder systems. Even the highest computational penalty is <1% of the calculated k_{eff} value, which implies the effect of inadequacy of the benchmark set to sufficiently represent the application systems is <1% because most of the cross-sections for the nuclides in these systems are known with small uncertainty.

The effect of ^{238}U cross-section evaluations was also investigated. Although some differences in the calculated values were observed, the true cause of these differences (i.e. due to new evaluation or an artefact of using new evaluations for the application cross-sections only) needs to be determined by further analysis.

High safe limits affect allowable safe MOX powder volumes considerably. Experiments that can provide a good basis for similarity can reduce the computational penalty due to non-coverage as well as provide an additional source for reducing bias and uncertainty associated with the calculation of the k_{eff} of MOX powder applications.

The tools that were used to assess the utility of the proposed benchmark can also be used to improvise the proposed experiment or evaluate other experiment designs in order to better fit the applications for validation purposes. However, due to limited time available, no effort was extended towards improving the proposed benchmark for applicability to the validation of MOX fuel blending processes.

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EXPERIMENTAL NEEDS FOR ESTIMATION OF CRITICALITY PREDICTION ACCURACY FOR SYSTEMS WITH MOX FUEL

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Abstract

The reliability of critical safety evaluation of MOX fuel management is determined by ^{239}Pu and ^{238}U neutron data accuracy. A qualitative analysis of up-to-date knowledge on these data, important for nuclear safety assurance, is given in the paper.

The analysis of existing experimental information shows a need for additional integral benchmark experiments for low-moderated MOX media. Experiments performed and planned at the BFS facility might provide valuable information on the quality of nuclear data.

^{239}Pu neutron data

The neutron data for ^{239}Pu from various libraries of many countries are in a good agreement. Table 1 displays averaged over fission neutron spectrum $\bar{\nu}$ values, fission and inelastic scattering cross-sections taken from the ABBN-93 library and root-mean-square deviations, determined using the mentioned values from the ENDF/B-V, ENDF/B-VI, JENDL-3.0, JENDL-3.3, JEF-2.2 and JEFF-3.0 libraries. These deviations would be the measure of uncertainty if all the mentioned evaluations were independent. In reality it is not so, and in the line below, the maximum discrepancy between the data of various libraries is indicated: it is two to three times higher. Such large discrepancies manifested for all three values under consideration using a low number of data show that the spread of these data is far from statistical. The uncertainties estimated on the basis of covariance data presented in the ENDF/B-V and ABBN-78 libraries are also given in the table. They exceed by two to three times even the maximum deviation and by about five times the square-root deviations. It should be noted that independently estimated uncertainties of neutron data from ABBN-78 are only slightly more optimistic than those from ENDF/B-V.

Table 1. Cross-sections averaged on fission spectrum and their uncertainties

	Nu-bar	**Fission**	**Capture**
ABBN-93 data	3.172	1.796	1.458
Sq.-root deviation, %	0.2	0.3	4.4
Maximum deviation, %	0.6	1.0	13
Uncertainty according to **ENDF/B-V**, %	1.3	2.9	–
Uncertainty according to **ABBN-78**, %	0.7	2.8	15

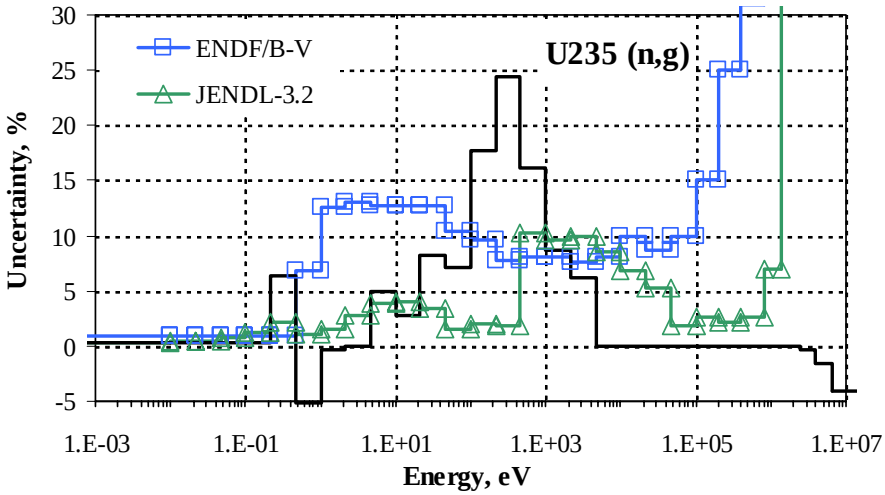
Table 2 provides radiation capture and fission cross-sections averaged over Fermi spectrum (resonance integrals), spectra of critical systems with dry and dump MOX fuel and spectrum of fast reactor with sodium coolant. Similarly to Table 1, square-root deviations of these values determined on the basis of data from the mentioned libraries, maximum discrepancy of these data and uncertainty estimated on the basis of covariance files from ENDF/B-V and ABBN-78 are presented in the table. Apparently, various evaluations are in the best agreement for spectra formed in MOX fuel systems, i.e. spectra, in which a maximum fraction of neutron interactions occur in the keV energy range, which has not been sufficiently studied. As regards the neutron spectrum of LMFBR, for which the neutron data were verified very well, the differences in the data are lower or equal to the differences for MOX fuel systems. The uncertainties according to the ENDF/B-V and ABBN-78 libraries are even larger.

Table 2. Average capture and fission cross-sections on various spectra and their uncertainties

	Fission				**Capture**			
	1/E	**MOX 10-5**	**MOX 10-0**	**LMFBR**	**1/E**	**MOX 10-5**	**MOX 10-0**	**LMFBR**
ABBN-93 data	308.6	11.18	3.175	1.885	193.9	6.078	1.064	0.606
Sq.-root deviation, %	0.6	0.2	0.3	0.6	2.0	1.4	0.9	1.2
Maximum deviation, %	1.6	0.5	0.8	1.7	6.4	4.1	3.0	4.0
Uncertainty according to **ENDF/B-V**, %	1.4	2.2	2.0	3.0	3.7	5.4	4.6	8.5
Uncertainty according to **ABBN-78**, %	3.4	3.1	1.8	2.4	7.5	6.2	4.4	6.9

It should be noted that for the resonance energy range, which is very important for critical safety analysis, all researchers take one data estimate made by H. Derrien (ORNL) [1] and T. Nakagawa (JAERI), which is undoubtedly the most reliable. However, some doubt subsists concerning the reliability even of these data. Resonance parameters of ^{239}Pu were estimated independently in three energy ranges, namely: 1.0 keV, from 1.0-2.0 keV and from 2.0-2.5 keV. In the same manner, L. Leal, *et al*. [2] (ORNL) made evaluation of ^{235}U cross-sections, which was placed in ENDF/B-VI (Rev. 2). In 1997, the expansion of calculation capabilities made it possible to perform a joint overview of the same experimental data over the whole range of resolved resonances. This evaluation was adopted in the up-to-date versions of almost all libraries. Figure 1 shows the difference between ^{235}U cross-sections from the ENDF/B-VI.2 and ENDF/B-VI.5 libraries, and cross-section uncertainties obtained from the ENDF/B-V and JENDL-3.2 libraries' covariance files.

Figure 1. Difference between ^{235}U cross-sections obtained from ENDF/B-VI.2 and ENDF/B-VI.5 libraries, and cross-section uncertainties obtained from the ENDF/B-V and JENDL-3.2 libraries' covariance files



It is shown that the revision has provided a more significant change in the capture cross-section value than the cross-section uncertainties obtained even on the basis of the ENDF/B-V covariance file. There is no guarantee that a similar effect would not appear for ^{239}Pu.

Thus, estimated neutron data for ^{239}Pu appear much more correct as compared to that justified by the correctness of measurement results.

Below, a comparison of different specialists' opinions is given as concerns the up-to-date accuracy of neutron data. Figures 2, 3 and 4 present uncertainties of fission and capture cross-sections and $\bar{\nu}$ value for ^{239}Pu obtained from the covariance files of ENDF/B-V and JENDL-3.2 libraries and cross-section uncertainties covariance matrix ABBN-78. The opinions of specialists regarding the uncertainties are apparently quite different, and the probability of having a proven opinion on the true accuracy of up-to-date neutron data on the basis of available estimated data on the uncertainties is low.

It follows from the above considerations that the real uncertainties of computed k_{eff} values due to nuclear data uncertainties cannot be obtained only on the basis of the information on uncertainties in nuclear data libraries because of the considerable spread among them.

Figure 2. Uncertainties of ^{239}Pu fission cross-section adopted in various libraries

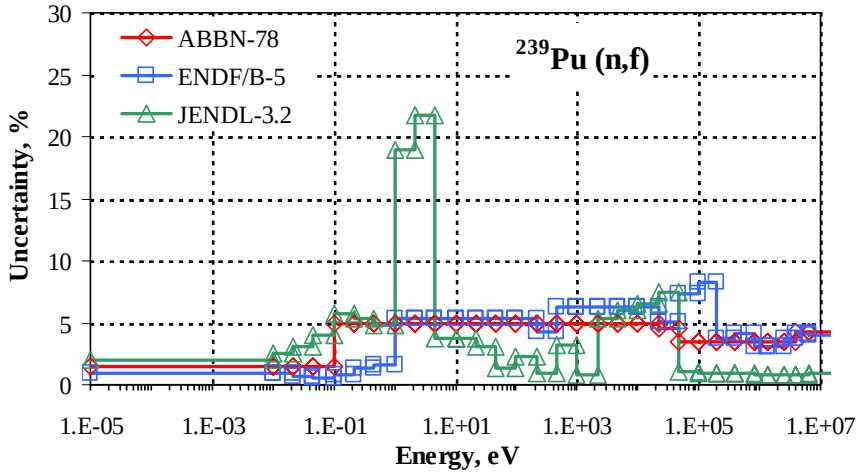


Figure 3. Uncertainties of ^{239}Pu radiation capture cross-sections from various libraries

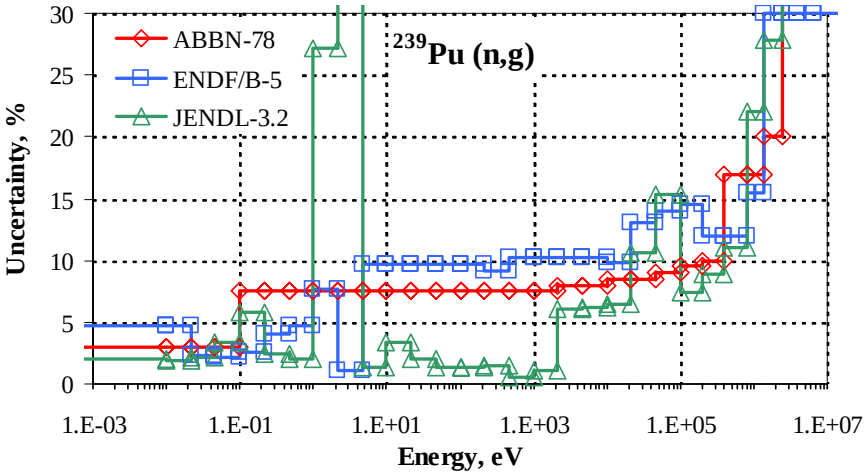
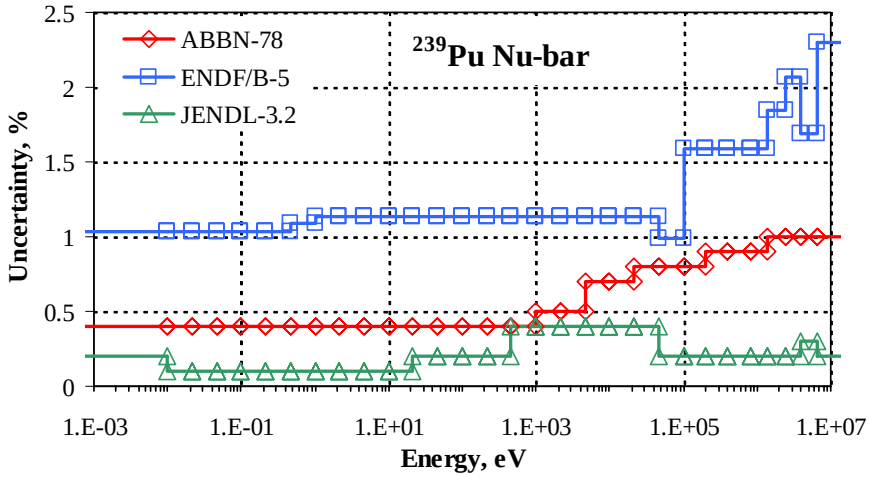


Figure 4. Uncertainties of $\bar{\nu}$ value for ^{239}Pu from various libraries



Roughly 100 experimental configurations containing the solutions of plutonium were selected. These experiments describe the application. Here it is expedient to consider as an application the interpolation curve characterising the dependence of critical mass of an idealised sphere, filled with an aqueous solution of plutonium and surrounded by a thick water reflector, on the plutonium concentration. The minimum concentration (10 g/l) corresponds to $k_{\infty} = 1$; the maximum concentration corresponds to metallic plutonium. Figure 5 shows that there are no experiments covering the range of fuel concentration 1-10 g/cc.

Figure 5. Sensitivity coefficient of k_{eff} to ^{239}Pu neutron data (1 group)

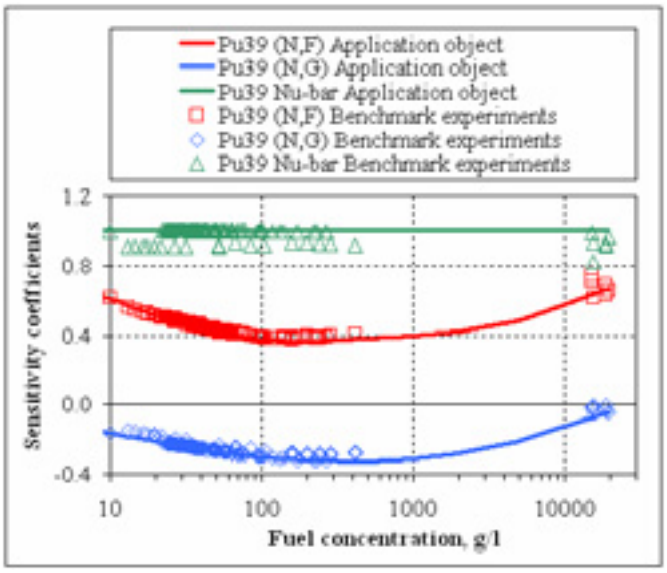


Figure 6 shows the criticality prediction uncertainties and its main components for the application as a function of fuel concentration before the adjustment. On Figure 7 the comparison of the uncertainty before and after the adjustment is given.

Figure 6. k_{eff} uncertainty and its components for application before the adjustment

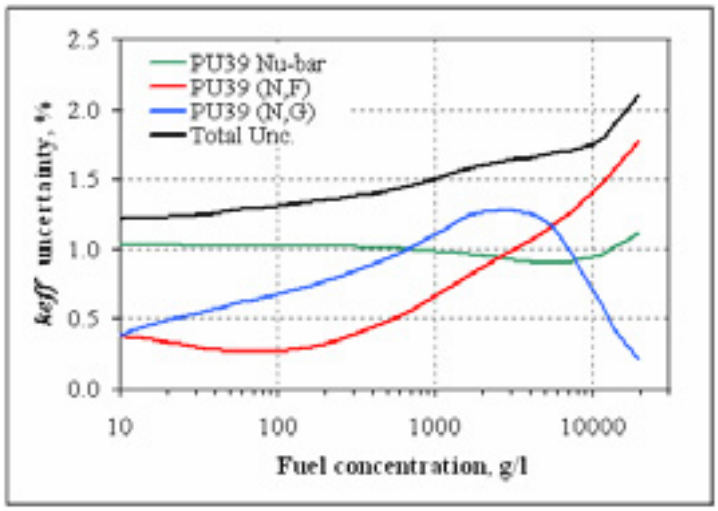
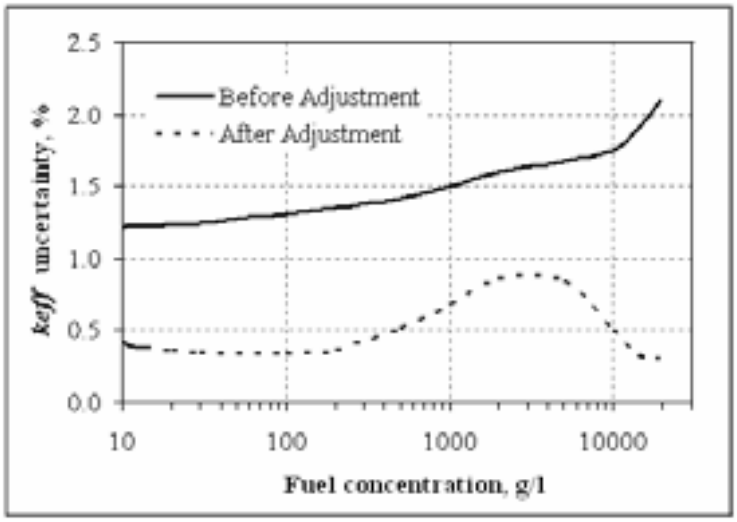


Figure 7. k_{eff} uncertainty for application before and after the adjustment



Conclusions

- The spread of ^{239}Pu neutron data adopted in various evaluations does not show the level of their uncertainties.
- Available estimates of uncertainties in the neutron data for ^{239}Pu are not acceptably reliable.
- In order to obtain reliable estimates of uncertainties in the criticality forecast, it is necessary to validate neutron constants in critical experiments sensitive to the keV energy range.
- There are no experiments which cover the range H/Pu value from ~1 to 25.

^{238}U neutron data

Table 3 presents averaged over fission spectrum values of ν, fission and inelastic scattering cross-sections, cross-section of removal below fission threshold of ^{238}U (removal cross-section), as well as the average cosine of elastic scattering angle. Removal cross-section is determined as follows:

$$\sigma_{rem} = \frac{\int_0^\infty dE \cdot \chi(E) \cdot \sigma_f^8(E) \cdot \left[\int_0^\infty dE' \cdot \sigma_{in}^x(E \to E') \cdot \left(1 - \frac{\sigma_f^8(E')}{\sigma_f^8(E)} \right) \right]}{\int_0^\infty dE \cdot \chi(E) \cdot \sigma_f^8(E)}$$

The ABBN-93 cross-section library was used for obtaining all these values. In the line below, the root-mean-square deviation from these values is displayed, obtained using other estimated data (ENDF/B-V, ENDF/B-VI, JENDL-3.2, JENDL-3.3, JEF-2.2, JEFF-3.0 and recent estimates by Maslov, *et al*.). The next line gives maximum discrepancies obtained using the above estimates. Finally, in the bottom line, uncertainties estimated on the basis of the covariance data file in ENDF/B-V are shown.

Table 3. Average cross-sections in the fission spectrum and estimates of their uncertainties

	Nu-bar	Fission	Inelastic	Removal	Mu-avg.
ABBN-93 data	2.830	0.3045	2.364	2.111	0.4777
Sq.-root deviation, %	0.4	0.5	3.0	5.0	4.9
Maximal deviation, %	1.2	1.3	11	11	16
Uncertainty according to **ENDF/B-V**, %	0.9	2.0	–	–	–
Uncertainty according to **ABBN-78**, %	0.8	2.4	14	17	–

Similarly to the case of ^{239}Pu, the root-mean-square spread of data is 2 to 3 times lower than the maximum spread, and this undoubtedly means that the estimated data spread is not subordinate to Gaussian distribution. In contrast to ^{239}Pu, the maximum spread of ν and inelastic scattering cross-section values is close to that expected from uncertainty estimates. Review of the deviations themselves reveals that inelastic scattering cross-section is higher by 8-10% than that adopted in ABBN-93, while removal cross-section caused by inelastic scattering alternately turns out to be lower than that in the ABBN-93 (in the case of ENDF/B-VI or JENDL-3.3, the difference is over 10%). This difference has a rather strong effect on the ^{238}U/^{239}Pu fission number ratio in MOX fuel critical systems. In order to validate the estimate of the removal cross-section, it is necessary to use the results of integral experiments, primarily, measurements of the ^{238}U/^{239}Pu fission cross-section ratio in uranium-plutonium medium or ^{238}U/^{235}U fission cross-section ratio in the medium with enriched uranium with k_∞ close to one (it is the removal cross-section that is required for a correct evaluation of these cross-section ratios). Unfortunately, estimated integral experiments of this type are not always in a good agreement. In order to extract information from the results of experiments with $k_\infty > 1$ (such as Bigten), neutron leakage (competing with inelastic removal) should be correctly taken into account. A large spread of estimated data on the average cosine of elastic scattering angle means that it is also a problem to correctly take into account neutron leakage from the uranium systems.

Figure 8 shows uncertainties of capture cross-sections for ^{239}Pu and ^{238}U appearing from the covariance files of the ENDF/B-V and JENDL-3 libraries and covariance matrix of constants ABBN-78. Similar to the case of ^{239}Pu, uncertainties in the neutron data obtained by specialists in various countries are quite different.

Figure 8. Uncertainties of radiation capture cross-section of ^{238}U adopted in various libraries

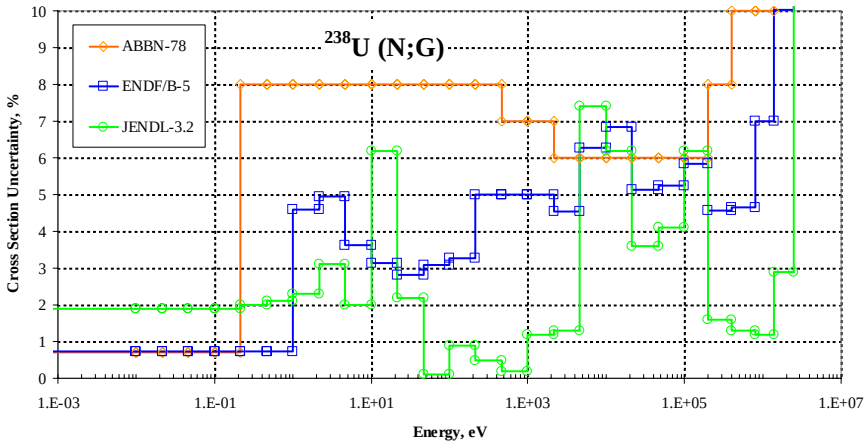


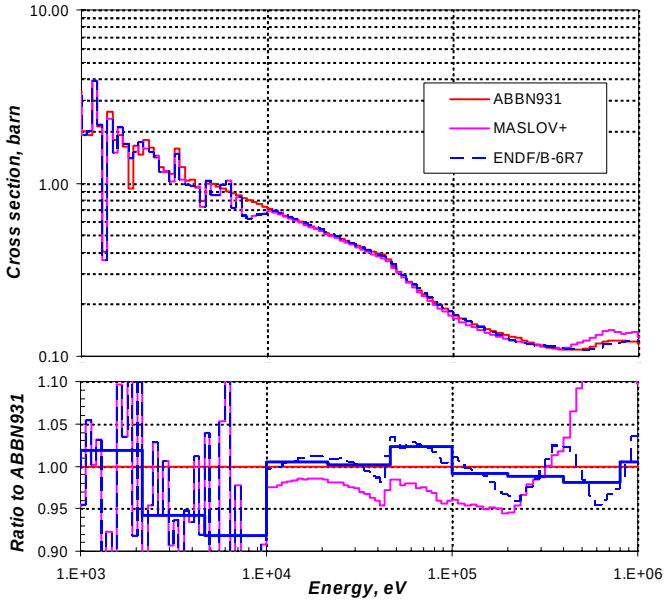
Table 4 shows the radiation capture cross-sections averaged over Fermi spectrum (resonance integrals), spectra of critical systems with dry and wetted MOX fuel and spectrum of power fast reactor with sodium coolant. Various estimates of their uncertainties are also presented.

Table 4. ^{238}U capture cross-sections averaged on various spectra and their uncertainties

	Capture			
	1/E	**MOX-10-5**	**MOX-10-0**	**LMFBR**
ABBN-93 data	193.9	6.078	1.064	0.606
Sq.-root deviation, %	0.3	0.3	0.6	1.1
Maximal deviation, %	1.2	1.1	1.5	3.3
Uncertainty according **ENDF/B-V**, %	4.1	4.0	3.4	4.2
Uncertainty according **ABBN-78**, %	7.9	7.6	5.3	4.3

Figure 9 shows radiation capture cross-section on ^{238}U and various estimates of this cross-section with respect to the ABBN-93 estimate vs. energy. The most significant difference is observed in the 2-10 keV range, for which the estimate is adopted in ABBN-93 based on the average resonance parameters describing both characteristics of resonances below 2 keV and data on the average neutron cross-sections and scattering anisotropy up to 200 keV. In all other libraries of estimated data, a later estimate made by M.G. Sowerby [3] and M.C. Moxon (Harwell, 1988) is used. This estimate takes into account the results of an experiment performed by Moxon, *et al.*, in which resonances were resolved up to 10 keV. This estimate is actually validated to a better extent, and adopted estimates of average resonance parameters describing cross-sections for the over 10 keV energy range are still to be brought into agreement with this estimate and data on average cross-sections and scattering anisotropy.

Figure 9. Various estimations of ^{238}U capture cross-section



For ^{238}U cross-section validation it is necessary to select the informative experiments. First, these are the experimental configurations with uranium fuel and uranium-plutonium fuel including MOX fuel where the k_∞ were measured. The most pressing task is to carry out a joint analysis of experiments with uranium and plutonium and to eliminate contradictions between the calculation-experimental deviations (see Table 5).

Table 5. k_∞ calculation-experimental deviation for experiments with uranium and uranium-plutonium fuel

Fuel	Enrichment, %	Experiment identifier	MCNP-4C, ABBN-93 subgroups	MCNP-4C, ENDF/B-VI.5 prob. tables
U (met.)	5.6	SCHERZO-5.56	0.3	1.8
		BFS-35-1	0.2	1.8
		BFS-35-2	-0.2	1.3
		BFS-35-3	-0.1	1.4
		SNEAK	-0.2	1.1
U+Pu (met.)	4.0	BFS-38-4	0.0	1.6
		BFS-38-5	-0.6	1.0
UOX	7.7	BFS-33-1.1	-1.1	-0.1
		BFS-33-2.1	-0.7	0.3
		ERMINE-OU-10	-1.1	-0.1
MOX	7.0	BFS-31-4	-1.1	-0.2
		BFS-31-5	-0.2	0.5
		ERMINE-OP-10	0.7	1.7
MOX+H	6.6	BFS-42	-0.3	0.5

Conclusion

- ^{238}U neutron data in the range above fission threshold of this isotope requires additional validation in the integral experiments.

- The joint evaluation of experiments with uranium and uranium-plutonium fuel and elimination of contradictions is required.

MOX issue

On some stages of MOX fuel fabrication, wet MOX powders with about several weight per cent water content are used. In this view, there is a need to validate the codes and nuclear data used for the analysis of critical safety of these systems. Below, the analysis of the information content of available data of macro experiments suitable from the standpoint of this problem solution is given. Also, a series of additional tests is proposed to perform on the BFS critical facility at the SSC RF/IPPE, which would provide informative experimental data for those values of MOX fuel plutonium enrichment and water concentration, for which there is a lack of data.

The effective multiplication factor of the MOX fuel sphere with some water impurity surrounded by an infinite reflector was considered an application object. The content of ^{240}Pu in plutonium is 4 wt.% (weapons-grade plutonium). Plutonium enrichment of the uranium-plutonium mixture ranged from 5-100 wt.%. The water content in the water-MOX fuel mixture varied from 1-5 wt.%. Theoretical density was adopted for MOX fuel. All options of enrichment/water content ratio are indicated in Table 6, where there are also relevant H/Pu ratio values. The information content of experiments (both performed and planned at the BFS critical facility) were evaluated with respect to the above application object.

Table 6. H/Pu ratio values for the cases under consideration (application)

Water content, wt.%	Plutonium content (primary blend), wt.%					
	5	**10**	**15**	**20**	**30**	**100**
0	0	0	0	0	0	0
1	6.9	3.4	2.3	1.7	1.1	0.3
2	13.9	6.9	4.6	3.5	2.3	0.7
3	21.0	10.5	7.0	5.3	3.5	1.1
4	28.3	14.2	9.5	7.1	4.7	1.4
5	35.8	17.9	11.9	9.0	6.0	1.8

Figure 10 gives values of critical plutonium mass referring to various points of application object (analysis made using ABBN-93 data). Figure 11 also gives data on critical volumes.

Figure 10. Pu critical mass of for the application

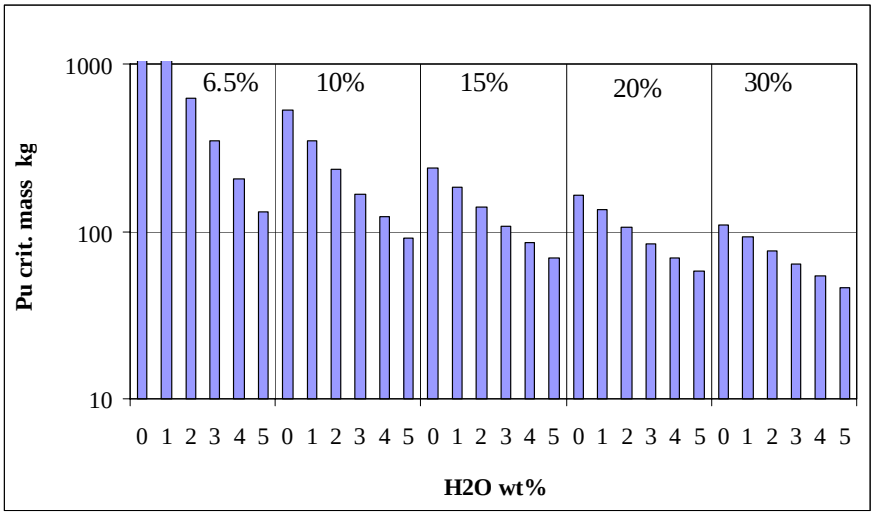
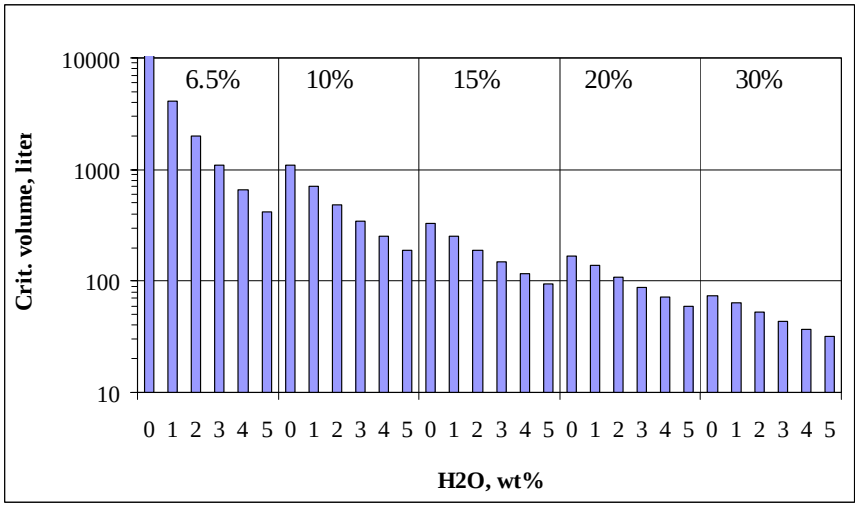


Figure 11. Critical volumes for the application



The analysis of the informative nature of existing and planned experiments with respect to the application was conducted on the basis of the generalised least squares method. In this approach the following data were used: sensitivity coefficient of k_{eff} to nuclear data, the nuclear data uncertainty covariance matrices and the experimental uncertainty covariance matrices for selected configurations.

About 800 kg of plutonium are available at the BFS facilities (about 100 kg plutonium with a high ^{240}Pu content). The plutonium fuel is stored and used in the form of stainless-steel-clad cylindrical pellets of 47-mm diameter. Polyethylene in the form of pellets and long sticks placed between the tubes, located in a hexagonal lattice is used for the simulation of water.

A lot of critical experiments with plutonium have been carried out at the BFS facility. Six of them were selected for this analysis as the most informative. Table 7 gives the plutonium content values and hydrogen-to-plutonium nuclide (H/Pu) ratios corresponding to the selected assemblies.

Table 7. Main characteristics of the selected experiments performed at the BFS facility

Experiment	Pu, %	H/Pu, %
BFS-31	6.6	0
BFS-42	6.6	0.7
BFS-49-1	12.5	0
BFS-49-3	12.5	0.7
BFS-57	6.7 (U)	31 (U)
BFS-59	8	23.6

The similarity between the selected experiments and the application was estimated according to the degree of the computation uncertainties' decrease after the attraction of the macroscopic experiments in comparison with the level, which corresponds to the accuracy of microscopic data. The results of the uncertainty estimations using only microscopic data and both microscopic and macroscopic data are shown in Figures 12 and 13, respectively.

Figure 12. Uncertainty of criticality prediction for the application before the adjustment

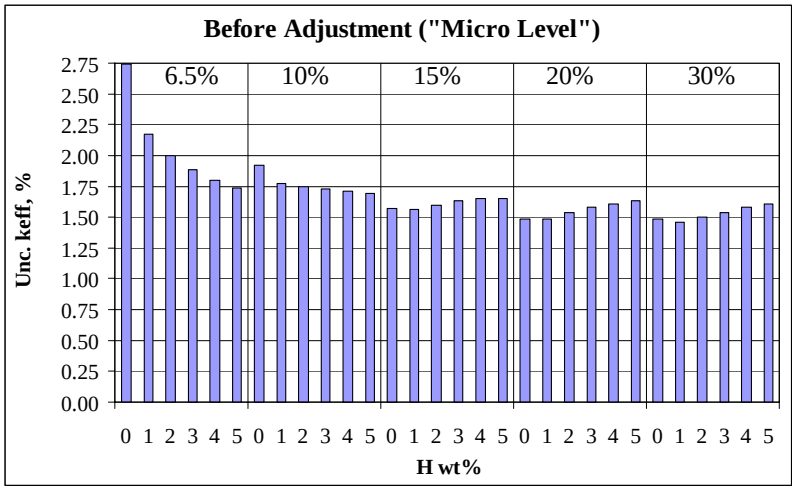


Figure 13. Uncertainty of criticality prediction for the application after the adjustment using data of the selected experiments (see Table 7)

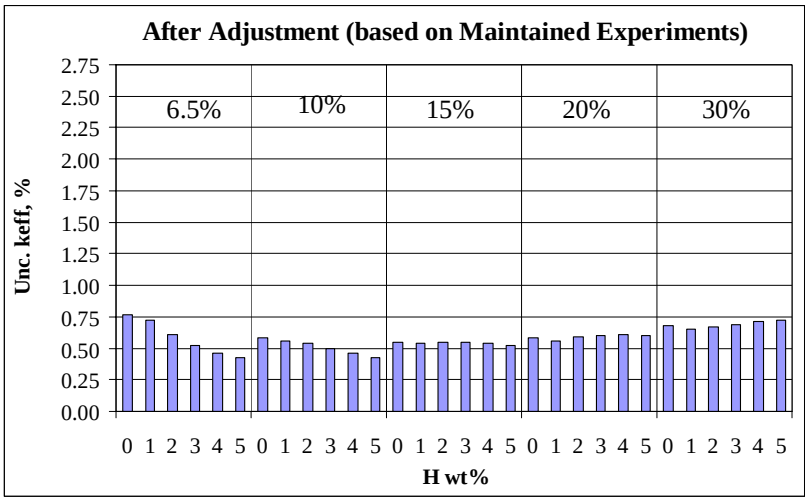
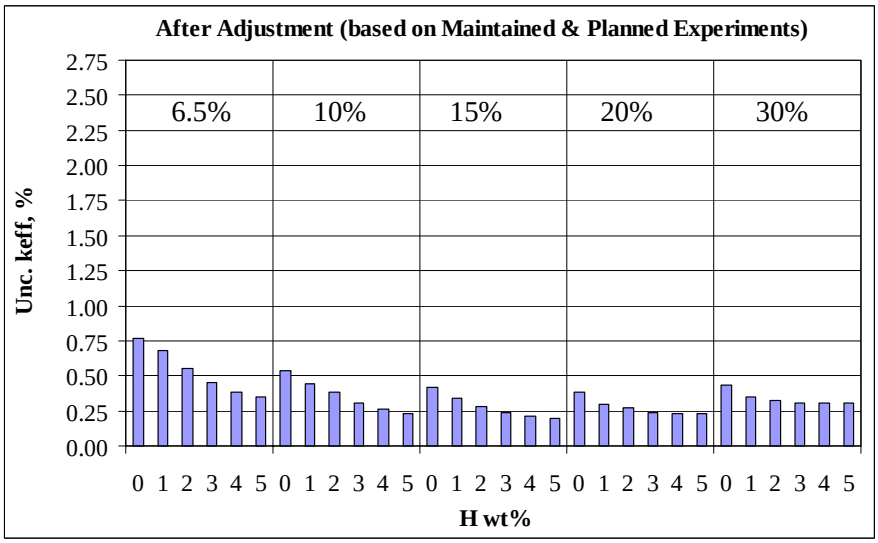


Table 8. The main characteristics of the planned experiments

Experiment BFS	Pu, %	H/Pu, %
MOX-30-5	30	6.3
MOX-22-0	22.5	0
MOX-15-5	15	11.5
MOX-15-3	15	6.5
MOX-10-5	10	17.3

Figure 14. Uncertainty of criticality prediction for the application after the adjustment using data of both performed and planned experiments on the BFS



Conclusion

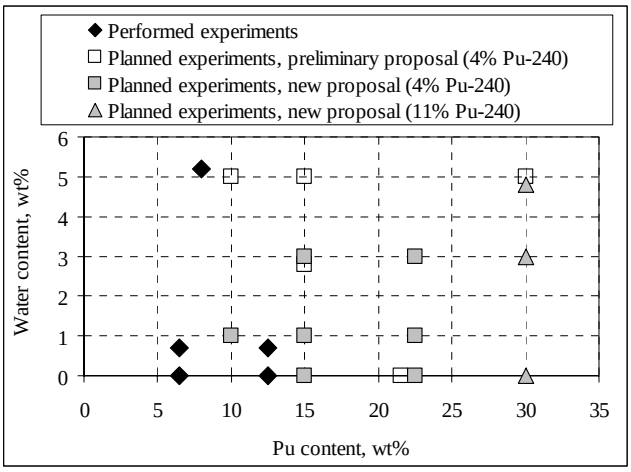
It is demonstrated in the paper that experiments for low-moderated MOX media are needed.

The existing and planned experiments at the BFS facility increase the accuracy of the criticality prediction for systems with plutonium and MOX fuel.

To increase the reliability of the nuclear data validation used for criticality calculations the following steps are suggested:

- To perform a joint international analysis of the experiments with $k_\infty \sim 1$ so as to eliminate contradictions between the evaluations.
- To evaluate experiments performed at the BFS-57, -59, 49(1, 2, 3) critical assemblies.
- To perform the proposed experimental programme at the BFS facility.

Figure 15. The main characteristics of the experiments performed and planned on the BFS facility



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VALIDATION FOR INTERMEDIATE SPECTRUM MOX APPLICATIONS AND ESTIMATES OF THE IMPACT OF NUCLEAR DATA UNCERTAINTIES ON ASSESSMENT MODELLING

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Abstract

In the UK predictions of critical mass have, historically, relied upon validated UKNDL nuclear data used within the MONK Monte Carlo code. More recent application of JEF-2.2 data to MOX fuel showed significant differences (several per cent in k_{eff}) for "intermediate spectrum" cases.

Initial review indicated a lack of critical benchmarks for MOX systems with intermediate spectra. Further review of the ICSBEP and MONK validation database indicated that assembly 8F of the Winfrith Zebra reactor (ZEBRA8F) provides information on intermediate spectra MOX systems. UKNDL calculations show an over-prediction of about 10% in k_∞ for ZEBRA8F, while JEF-2.2 agrees to within the experimental uncertainty. However this assembly has a harder spectrum than the MOX fuel assessment cases having minimal fission events below 0.625 eV compared with up to 50% for some moist MOX powder systems

Further searches for suitable data include consideration of measurements of spectral indices in the BIZET "salt and pepper cores" (BZD/1 and BZC/1) at ZEBRA. The spectra in these regions may be sufficiently representative to make the indices useful as supporting evidence of code/data accuracy for the MOX assessment models.

In addition to searches for other potential benchmark data, sensitivity studies have been made to determine the effect of cross-section uncertainties on calculated k_{eff}. E. Fort has tabulated JEF-2.2 cross-section uncertainties in JEF/DOC-439. Those for the most important reactions (capture in ^{238}U, capture and fission in ^{239}Pu, and capture in ^{240}Pu) are coupled with sensitivities for the models to derive the uncertainty in k_{eff} or k_∞ due to nuclear data uncertainties. This route is applied to the ZEBRA8F model and also to three intermediate spectrum models used in MOX fuel assessment, typifying the spread in neutron spectra for this type of system. These analyses indicate an uncertainty of about 0.7% in k_{eff} for the assessment cases.

A summary of the conclusions of the review of alternative benchmarks and on the results of the sensitivity studies is presented.

Introduction

The Sellafield MOX Plant (SMP) has been operational since 2001 processing plutonium from THORP, blending with UO_2 to produce MOX fuel elements. In handling the quantities of fuel associated with a commercial fuel fabrication plant, it is necessary to impose criticality controls.

Assessment of accident conditions includes consideration of accumulations of powders or pellets at various stages in the manufacturing process. Moisture content changes as various agents are added and removed and as the pellets are passed through the furnace for sintering. A set of material specifications has been identified as bounding for each stage. The corresponding safe mass limits were derived using the MONK [1] criticality code. These predictions of critical mass have, historically, relied upon validated UKNDL nuclear data, but more recent application of JEF-2.2 data to MOX fuel showed significant differences (several per cent in k_{eff}) for "intermediate spectrum" cases.

Investigations of the likely causes of these differences and a review of experimental benchmark data have been made. It was concluded that the significant differences in calculated k_{eff}, and hence safe mass, occur for low-moderated MOX systems with an intermediate neutron spectrum. Review of the criticality validation data set shows a lack of experimental benchmarks for these types of system. Appraisal of other types of experiment (fuel substitution, spectral indices) is ongoing and may provide supporting evidence of code accuracy. Analysis has also been made to quantify the effect of uncertainties in the basic nuclear data on the accuracy of calculations for low-moderated MOX systems.

This paper presents a summary of the review of experiments and analyses of the impact of nuclear data uncertainties.

Outline of fabrication process

The main stages of the fabrication process are summarised below.

The prescribed quantity of feed material is transferred from the appropriate dispensing station in purpose-built containers, which are then individually fed into the Homogenisation Attritor Mill. The Homogenisation Attritor Mill is a vessel with sloping sides, rotating mixing rods and a charge of metal balls. When milling is complete the powder is gravity fed to the Homogenisation Blender. The blender is a shallow cylindrical vessel, positioned on its side below the mill, which is fed with a quantity of ZnSt before receiving a batch of MOX powder.

MOX powder is then metered from the blender by a vibro-feeder into the Conditioning Mill Feed Hopper and from there to the Conditioning Attritor Mill. The Conditioning Attritor Mill is similar in design to the Homogenisation Attritor Mill. A pre-determined quantity of Conpor, a pore-forming agent, is added to the mill with the batch of homogenised and blended powder. After the milling operation the mill discharges its contents into the Spheroidiser, pre-lubricated with ZnSt like the blender. The Spheroidiser is another end-on cylindrical vessel and feeds the granules it produces to the Press Feed Hopper.

The powder is then pressed into pellets which are loaded into "boats" for transfer through a furnace in order to be sintered. This sintering process also drives off any intrinsic moderator present in the form of water and the fuel additives ZnSt and Conpor. The pellets are then transferred downstream for incorporation into rods used for the manufacture of fuel assemblies.

Criticality safety considerations and safe masses

Normal operations

Plutonium dioxide (PuO_2), uranium dioxide (UO_2) and recycled MOX are mixed together in batches. An Engineered Protection System (EPS) prevents the production of MOX powder in excess of 20 w/o Pu(fissile)/(Pu+U). This is achieved through the combination of a "weight-based" system and a diverse "neutron monitoring" radiometric system. The "neutron monitoring" component of the EPS determines the fissile enrichment of the batch of MOX powder, based on pessimistic isotopic requirements of the PuO_2 feedstock powder. Guaranteeing the maximum MOX enrichment of 20 w/o Pu(fissile)/(Pu+U) at an early stage of the fuel manufacturing process enables the criticality safety assessor to demonstrate that normal operations are deterministically safe. Typically the MOX powder is bounded by about 10 w/o Pu(fissile)/(Pu+U) for normal conditions.

Accident conditions

Two key accident conditions are excess moderation and accumulations of excess fissile material. In the case of excess moderation the differences between UKNDL and JEF-2.2 are small. For these systems neutron reactions at thermal energies are most important. In the context of code accuracy, it is accumulations of powders or pellets (with normal moderating conditions) that are most significant. These scenarios fall within the intermediate spectrum range and are typified by three assessment material types described below. These cover MOX powder, un-sintered ("green") pellets and sintered pellets.

Table 1 summarises the material specifications used in criticality assessment modelling, along with an indication of the range of values for moisture and density. In all cases the Pu isotopics are selected so that safe masses are bounding with respect to MOX with 17 w/o ^{240}Pu.

Table 1. MOX material types modelled in SMP criticality assessment

Description	Moisture content	Density	Pu(fissile)/(Pu+U)
MOX powder	<8 w/o, typically 0.3 w/o	3.5-5.5 g/cc	10 w/o (typical), 20 w/o (bounding)
Green pellets	<5 w/o, typically 2 w/o	~ 8 g/cm^3	10 w/o (typical), 20 w/o (bounding)
Sintered pellets	Trace	~ 11 g/cm^3	10 w/o (typical), 20 w/o (bounding)

Safe masses for cylinders of these materials have been calculated using MONK. Table 2 compares the results for UKNDL and JEF-2.2 nuclear data.

Table 2. Comparison of UKNDL and JEF-2.2 MONK calculations of safe mass

Material type	MOX enrichment [w/o Pu(fissile)]	Density	Moisture content	Safe mass (kg)		Δ mass
				JEF-2.2	UKNDL	
MOX powder	10	High	High	370.8	343.4	8%
MOX powder	10	High	Low	1 100	990.3	11%
MOX powder	20	High	Low	251.5	235.3	7%
Green MOX pellets	10	Normal	High	858.4	648.9	32%
Green MOX pellets	10	Normal	Normal	999	755	32%
Green MOX pellets	20	Normal	Normal	275.3	224.6	23%
Sintered MOX pellets	20	Normal	0	250.1	231.8	8%

It may be seen that the effect of switching from UKNDL to JEF-2.2 data produces substantial increases in calculated safe mass (equivalent to several % in k_{eff}), particularly for the un-sintered pellets.

Review of validation data

In resolving the differences between the calculated safe masses, a review of validation results is needed. Inspection of the MONK validation set [1] revealed a lack of critical MOX experiments with intermediate neutron spectra. A wider search for other sources of data, including the ICSBEP Handbook [2] failed to reveal any other well-matched critical benchmarks. However, the MONK validation set, and ICSBEP, do include measurements of k_∞ on a range of Pu, U and MOX assemblies in intermediate spectra. These were carried out in Core 8 of the ZEBRA zero-power fast reactor experimental series [3]. In particular assembly ZEBRA8F provides a measure of k_∞ for MOX fuel plates mixed with graphite. This exhibits an intermediate neutron spectrum. Comparisons with MONK for this experiment show a large over-prediction for UKNDL data, (by about 10% in k_∞), while calculations using JEF-2.2 agree to within the experimental uncertainty. This difference is consistent with the differences seen for the SMP assessment models and indicates that the UKNDL calculations are conservative.

In order to provide spectra for comparison of the SMP scenarios with benchmark cases it was necessary to re-run a selection of the SMP MONK calculations for MOX powder, green MOX pellets and sintered MOX pellets using the ICSBEP GROUP option in MONK8B. This option allows output of the flux, fission and capture rates in 3- and 30-group schemes. These spectra can then be compared with ICSBEP benchmark and validation data. The group bounds of the spectra are shown below (Table 3) along with the 15-group bounds used for the sensitivity studies (see section below entitled *Sensitivity to basic nuclear data uncertainties*).

Table 3. Energy group schemes* used for spectral comparison and scoring sensitivities

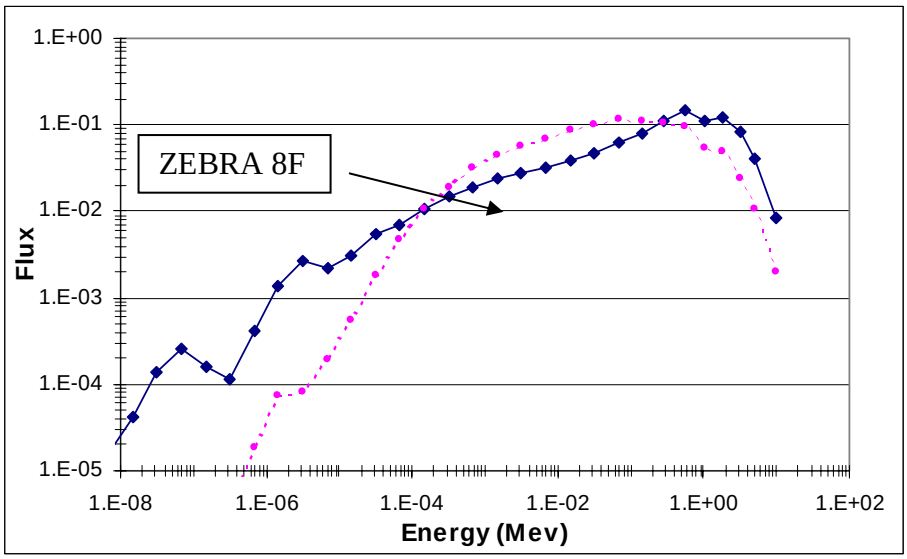
30-energy-group scheme (ICSBEP)				15-energy-group scheme (sensitivity)		3-energy-group scheme (ICSBEP)	
Group no.	**Upper energy (MeV)**	**Group no.**	**Upper energy (MeV)**	**Group no.**	**Upper energy (MeV)**	**Group no.**	**Upper energy (MeV)**
1	1.50E+01	16	4.64E-04	1	1.50E+01	1	1.50E+01
2	6.50E+00	17	2.15E-04	2	6.07E+00	2	1.00E-01
3	4.00E+00	18	1.00E-04	3	2.23E+00	3	6.25E-07
4	2.50E+00	19	4.64E-05	4	1.35E+00		
5	1.40E+00	20	2.15E-05	5	4.98E-01		
6	8.00E-01	21	1.00E-05	6	1.83E-01		
7	4.00E-01	22	4.64E-06	7	6.74E-02		
8	2.00E-01	23	2.15E-06	8	2.48E-02		
9	1.00E-01	24	1.00E-06	9	9.12E-03		
10	4.64E-02	25	4.64E-07	10	2.03E-03		
11	2.15E-02	26	2.15E-07	11	4.54E-04		
12	1.00E-02	27	1.00E-07	12	2.26E-05		
13	4.64E-03	28	4.64E-08	13	4.00E-06		
14	2.15E-03	29	2.15E-08	14	5.32E-07		
15	1.00E-03	30	1.00E-08	15	1.00E-07		

* The upper boundary of MONK nuclear data is 15 MeV and the lower boundary is 1E-11MeV.

Table 4 compares calculated neutronic parameters for seven of the SMP assessment models with the ZEBRA 8F assembly. The values are derived from the MONK and JEF-2.2 models. The modelling, including the sensitivity analyses reported below, was carried out by Serco Assurance and is reported in detail in Ref. [4].

From these comparisons it is clear that the SMP cases and the ZEBRA experiment have a large proportion of reactions in the resonance region, but that there are significant differences in the neutron spectra. This is illustrated in Figure 1, which plots the fluxes in 30 groups for the green MOX pellet case and for ZEBRA 8F.

Figure 1. Comparison of calculated 30-group fluxes for green MOX pellets [20 w/o Pu(fissile)] and for ZEBRA 8F



In this regard it is difficult to draw a firm conclusion on the accuracy of JEF-2.2 calculations for SMP on the basis of the ZEBRA results alone.

Further investigations for suitable benchmarks have centred on the possibility of using ZEBRA "BIZET" cores BZC/1 and BZD/1. In these fast reactor benchmarks performed in the 1970s at the ZEBRA Zero Energy Experimental Reactor were located central regions of uranium breeder cells within the plutonium-fuelled core. These may produce the neutron spectrum conditions suitable for comparison. Experimental foil traverses exist for these experiments as well as reaction rate ratio measurements in the fuelled region. It would be possible to derive a ^{238}U capture to ^{239}Pu fission ratio along these traverses. Consideration is being given to study of these core measurements, including modelling the core/breeder regions using MONK with JEF-2.2 data. At this stage though it is apparent that there is a lack of directly applicable integral benchmark data.

Sensitivity to basic nuclear data uncertainties

Given the lack of integral benchmarks, a sensitivity study has been made to assess the impact of nuclear data uncertainty on calculated k_{eff}, (and k_∞ for the ZEBRA 8F assembly). This covers uncertainty in the JEF-2.2 cross-sections for ^{239}Pu fission, ^{239}Pu capture, ^{240}Pu capture and ^{238}U capture. These four reactions are expected to be the most important. The analysis uses uncertainties and covariance data provided in a 15-group energy scheme, reported in Ref. [5].

Table 4. Neutronics parameters for SMP assessment models

Description	Density	Moisture	MLENCE*		Percentage group contributions								
			Capture	Fission	Flux			Fission			Capture		
					Fast	Resonance	Thermal	Fast	Resonance	Thermal	Fast	Resonance	Thermal
Powder 10%	High	High	1.80E-05	2.91E-05	59.6	39.4	0.9	18.34	39.24	42.42	4.26	75.48	20.26
Powder 10%	High	Low	4.12E-03	3.13E-02	67.7	32.2	0.1	57.98	33.75	8.26	24.38	69.99	5.63
Powder 20%	High	Low	2.36E-03	5.00E-02	77.9	22.0	0.0	65.72	26.34	7.94	28.89	61.47	9.64
Green pellets 10%	Normal	High	7.99E-05	5.44E-04	61.6	38.1	0.3	29.79	53.07	17.13	7.12	83.40	9.48
Green pellets 10%	Normal	Normal	2.82E-04	2.39E-03	62.6	37.1	0.3	32.75	46.70	20.54	9.42	78.58	12.00
Green pellets 20%	Normal	Normal	3.64E-04	6.53E-03	70.1	29.8	0.1	46.32	43.77	9.91	13.24	78.16	8.60
Sintered pellets 20%	Normal	0	3.17E-03	7.14E-02	81.3	18.6	0.0	70.77	21.46	7.77	35.31	54.14	10.55
ZEBRA 8F/2				1.51E-02	45.0	55.0	0.0	41.00	59.00	0.00	12.00	88.00	0.00

* MLENCE = Mean log energy of neutrons causing event.

Sensitivities in k_{eff} have been calculated using MONK with JEF-2.2. For the purposes of comparison this has been made in 15 groups and the 3-group ICSBEP scheme. The sensitivities for the green MOX pellet model are listed in Table 5.

Table 5. Sensitivity of k_{eff} to JEF-2.2 cross-sections: Cylinder of green MOX pellets with 2.5 cm of water reflector

Sensitivity of k_{eff}									
Group	**Upper energy (MeV)**	**^{239}Pu fission**	**sd**	**^{239}Pu capture**	**sd**	**^{240}Pu capture**	**sd**	**^{238}U capture**	**sd**
1	1.96E+01	0.0023	11.1%	0.0000	5.3%	0.0000	3.9%	0.0000	3.9%
2	6.07E+00	0.0223	4.0%	-0.0001	2.5%	-0.0001	2.5%	-0.0008	2.5%
3	2.23E+00	0.0150	4.6%	-0.0001	2.5%	-0.0001	2.5%	-0.0020	2.5%
4	1.35E+00	0.0296	3.5%	-0.0006	2.4%	-0.0005	2.4%	-0.0083	2.4%
5	4.98E-01	0.0189	4.1%	-0.0010	2.4%	-0.0006	2.4%	-0.0070	2.4%
6	1.83E-01	0.0138	4.6%	-0.0011	2.5%	-0.0007	2.5%	-0.0072	2.5%
7	6.74E-02	0.0100	5.6%	-0.0013	2.5%	-0.0009	2.5%	-0.0107	2.5%
8	2.48E-02	0.0089	5.4%	-0.0020	2.5%	-0.0012	2.5%	-0.0134	2.5%
9	9.12E-03	0.0153	4.7%	-0.0061	2.5%	-0.0027	2.5%	-0.0214	2.6%
10	2.03E-03	0.0257	3.9%	-0.0114	2.6%	-0.0049	2.8%	-0.0214	2.8%
11	4.54E-04	0.0700	2.9%	-0.0327	2.8%	-0.0140	3.3%	-0.0354	2.9%
12	2.26E-05	0.0242	4.0%	-0.0150	3.5%	-0.0007	8.4%	-0.0144	3.4%
13	4.00E-06	0.0126	4.7%	-0.0019	4.9%	-0.0158	4.1%	-0.0016	3.3%
14	5.32E-07	0.0198	4.4%	-0.0188	3.5%	-0.0045	3.8%	-0.0005	3.6%
15	1.00E-07	0.0307	3.9%	-0.0221	3.1%	-0.0109	3.1%	-0.0020	3.1%
Sensitivity of k_{eff}									
1	1.50E+01	0.1005	2.6%	-0.0025	2.3%	-0.0017	2.3%	-0.0222	2.3%
2	1.00E-01	0.1725	2.4%	-0.0709	2.5%	-0.0383	2.7%	-0.1211	2.5%
3	6.25E-07	0.0525	3.3%	-0.0410	2.9%	-0.0171	3.0%	-0.0026	3.0%

It is seen that all reactions show relatively high sensitivity in the resonance group. Using these sensitivities and combining them with the nuclear data uncertainties and covariance the overall uncertainty in k_{eff} and k_{∞} for the three SMP assessment material types and for the ZEBRA 8F assembly have been derived. The results are listed in Table 6.

Table 6. Values of k_{eff} and uncertainties

Case	k_{eff}	Stochastic uncertainty (σ)	JEF-2.2 cross-section uncertainty (σ)
MOX powder	0.9278	0.0010	0.0065
Sintered MOX	0.9273	0.0010	0.0070
Green MOX	0.9308	0.0010	0.0062
ZEBRA8F	0.9709	0.0010	0.0120

It is seen that the uncertainty for the SMP models is about 0.7%, rising to 1.2% for ZEBRA 8F. Since the UKNDL results are at least 2% higher than the JEF-2.2 results, this provides further evidence that the MONK-UKNDL derivation of safe mass for intermediate spectrum MOX systems is conservative.

Conclusions

The following conclusions have been drawn:

- There are significant differences in calculated safe mass when switching from UKNDL data to JEF-2.2.
- There is a lack of critical benchmarks to validate calculations for intermediate spectrum MOX systems.
- Some evidence for conservatism in MONK-UKNDL calculations is provided by the ZEBRA 8F experiment. MONK-JEF-2.2 calculations agree well with ZEBRA 8F.
- There are significant differences in neutron spectra between ZEBRA 8F and MOX assessment models for SMP.
- Analysis of the impact of JEF-2.2 nuclear data uncertainties on calculated k_{eff} for the SMP assessment models yields an uncertainty of less than 1%, which provides additional evidence that the UKNDL results are conservative.
- Significant increases in safe mass might be realised if better experimental validation were available to underpin the application of JEF-2.2 data.

Acknowledgements

The assistance of Christopher Dean, Bernard Franklin and George Wright of Serco Assurance in support of the studies presented here is gratefully acknowledged.

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CRITICALITY CODES VALIDATION ON SPHERICAL PLUTONIUM SYSTEMS

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Abstract

In an attempt to update their 1-D qualified scheme and for QA reasons, Belgonucléaire (BN) undertook an internal study of different criticality codes. It was divided into two parts: (i) comparison between calculations and experiments, on the basis of ICSBEP database, and (ii) comparison between the selected computing schemes and with the values available in the literature. The paper focuses on the spread observed between the calculated results, for both the critical radius and the critical mass. The spread is rather high and the general trend could induce an increase of the maximum allowable mass in the work units. However it is felt that critical experiments involving PuO_2 and MOX powders with and without moderation could advantageously complete a database such as ICSBEP.

Introduction

Criticality safety in a MOX fabrication plant often relies on calculations of fissile spherical and reflected systems. This simple approach is conservative and permits to deduce the maximum allowable mass of Pu, PuO_2 or MOX for each work unit. Such a mass is obtained from the calculated critical mass, corrected by various safety coefficients. This procedure was applied for the design of the Belgonucléaire (BN) MOX plant, using the 1-D calculation scheme Hansen & Roach (16 groups, P_1 cross-sections) – ANISN (transport S_n).

In an attempt to update their 1-D qualified scheme and for QA reasons, BN undertook an internal study of different criticality codes. It was divided into two parts: (i) comparison between calculations and experiments on the basis of the ICSBEP database [1], and (ii) comparison between the selected computing schemes and with the values available in the literature [2-4].

The paper focuses on the spread observed between the results for both the critical radius and the critical mass. Both *minimum* critical value *and* critical value *at low moderation ratio* (m = H/(U+Pu) ~ 0) are considered, since the two sets of values correspond to two different criticality control levels, according to the fact that the process in the plant includes or not the possibility of moderator ingress.

Calculation schemes

The criticality calculation schemes were the following:

- Hansen & Roach [5] – ONEDANT [6].
- Hansen & Roach – KENOVa [7].
- WIMS8a [8] (cross-section processing) – ONEDANT.
- WIMS8a – KENOVa.
- ENDF-B6 – MCNP4c2 [9].

Both Monte Carlo (statistics method) and transport (deterministic) approaches were chosen in order to separately assess the effects of the cross-section libraries and of the calculation method. The Monte Carlo method is not thought to be a practical tool for the determination of critical values within a 1-D geometry, mainly for the CPU time reason. Alternatively, this method remains the most valuable for 3-D calculations, e.g. for storage rooms.

For Monte Carlo calculations, neutron per generation and total number of generations were chosen so that the statistical deviation was comprised between 50 and 200 pcm. For the transport method, calculations were performed with the angular discretisation S_{32}, after cross-section processing at the anisotropy order P_1.

All criticality calculations, except those with MCNP, were performed after cross-section condensation to 16 energy groups, in a group structure that fits the Hansen & Roach library.

Calculation/measurement comparisons

Selected experiments

For the first part of the work, a set of ~ 30 critical spheres experiments were selected, varying the fissile nuclides and the moderation ratio [m = H/(U+Pu), through the physical state]. The detailed list is reported in an appendix, with their main characteristics, but we restrict the current discussion to the experiments involving plutonium.

Results

Each experiment was evaluated by the five criticality calculation schemes and individual bias ($b = k_{cal} - k_{exp}$) were obtained. This information was then summarised by grouping the experiments according to the group to which they belong, thus calculating a weighted average bias:

$$\bar{b} = \frac{\sum_i b_i / \sigma_{b,i}^2}{\sum_i 1/\sigma_{b,i}^2}$$

where σ_b accounts for the weight attributed to the individual bias (experimental and calculation uncertainties). The resulting bias are given in Table 1, as well as the maximum spread between the various calculation schemes, Δ_{MAX}.

Table 1. Bias (pcm) obtained with the various calculation schemes for each group of ICSBEP experiments (negatives values are non-conservative)

Group	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (pcm)
PU-FAST	-694	289	-3 085	-249	-2 088	3 374
PU-THERM	581	841	623	2 053	1 008	1 472
MIX-FAST	93	102	-1 922	21	-595	2 024

Some trends are clearly observed, such as the conservative impact induced by the Hansen & Roach library. Cross-section processing with WIMS generally leads to the least conservative results, especially for plutonium systems within a fast spectrum, which is not really surprising since WIMS is based on a multi-group cross-sections library suitable for LWR spectrum conditions. In counter part, the Hansen & Roach library can be too conservative for plutonium systems within a thermal spectrum.

The bias has been used to deduce the upper safety limits (USL) per group of experiments, through the relation:

$$USL = 1 + \bar{b} - 3 \times \sigma_b$$

and it was verified that the USL are in agreement with the usual criterion $k_{eff} + 3 \times \sigma_{stat} \leq 0.95$.

The maximum spread Δ_{MAX} between the calculation schemes is in any case considered to be important, ranging from ~ 1 500 to ~ 3 000 pcm, according to the group of experiments.

Calculation/calculation comparisons

Selected fissile systems

The experiments selected in the firstpart of the work did not cover a sufficient moderation range (m = 0 or m > 350, see list in appendix), and neither considered homogeneous mixtures of resonant nuclides Pu and U, as for the MOX fuel. Such mixtures are however important from the point of view of a MOX producer, so that a second part of the work was needed, dealing with an intercomparison of criticality codes calculations at various moderation ratios and for different mixtures of UO_2, PuO_2 and MOX diluted and reflected by water.

The fissile systems were the following:

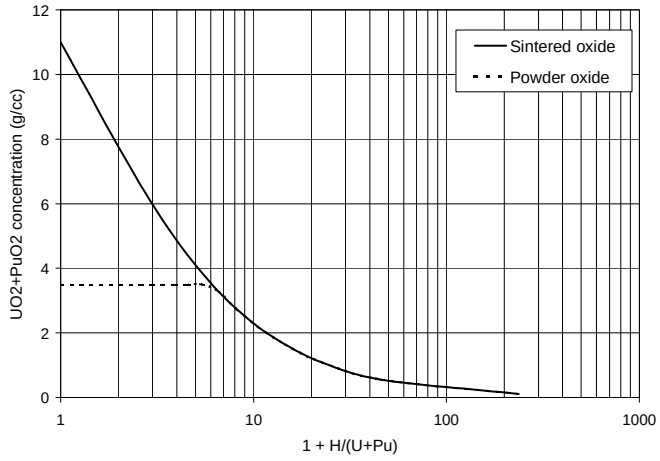
- Sintered oxide UO_2 3 wt.%.
- Sintered oxide UO_2 20 wt.%.
- Sintered oxide PuO_2 (100 wt.% ^{239}Pu).
- Sintered oxide PuO_2 (Pu 71/17/11/1).
- Powder oxide MOX (Pu 35 wt.%).
- Sintered oxide MOX (Pu 12.5 wt.%).

The paper focuses on the three last fissile systems, which are the most relevant ones with respect to the MOX fabrication plant.

Results

For each fissile system and each calculation scheme, the moderation ratio has been varied through the simulation of a progressive dilution of the sintered oxide or of the powder oxide in water, as shown in Figure 1. The critical radius is then computed as a function of the moderation ratio or of the heavy metal dilution (see Figures 2 and 3). This allows deducing the minimum critical values and the values at very low moderation (m ~ 0), as reported in the following tables.

Figure 1. Sketch of the two dilution laws used in the framework of this study



Mixture of sintered PuO_2 with H_2O

The detailed results are summarised in Tables 2 and 3 for, respectively, the minimum values and the values obtained at $m \sim 0$.

Table 2. Minimum critical values for the mixture PuO_2-H_2O

Parameter	*m*	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (%)
R_c^{min} (cm)	0	7.1	7.0	7.4	7.0	7.6	**8.6**
M_c^{min} (kg)	500-1 000	1.01	1.07	1.15	0.93	1.07	**24**

Table 3. Critical values for the mixture PuO_2-H_2O at moderation ratio $m = 0$

Parameter	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (%)
R_c^0 (cm)*	7.1	7.0	7.4	7.0	7.6	**8.6**
M_c^0 (kg)	16.5	15.8	18.7	15.8	20.2	**28**

* Corresponding to the minimum critical radius for this fissile system.

At low moderation, one recovers the less conservative trend of WIMS, as compared to the use of H&R cross-sections or the use of the scheme ENDF-MCNP. For the minimum critical mass, at moderation *m* between 500 and 1 000, the agreement between WIMS and ENDF-MCNP is better, whereas the use of Hansen & Roach probably leads to over-conservative results.

Mixture of powder MOX with H_2O

In this section we present the detailed critical curves in Figures 2 and 3, along with the values obtained by IRSN and ORNL in [3,4].

Figure 2. Critical radius as a function of the moderation ratio for the mixture of powder MOX with H_2O

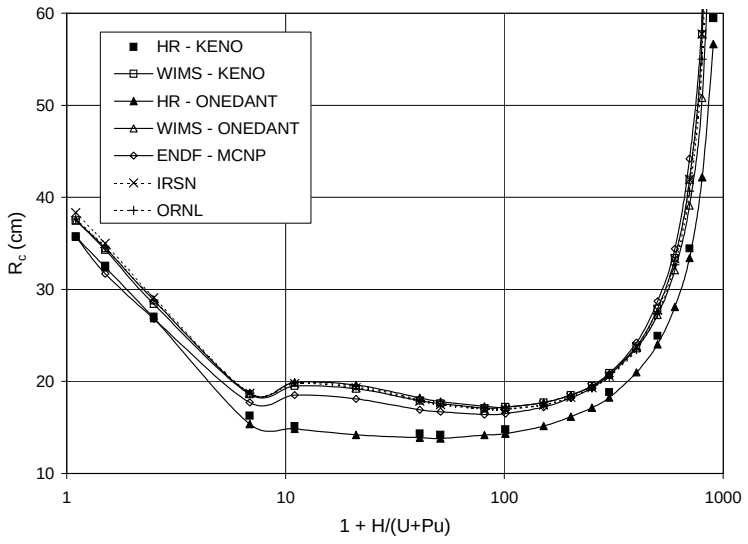
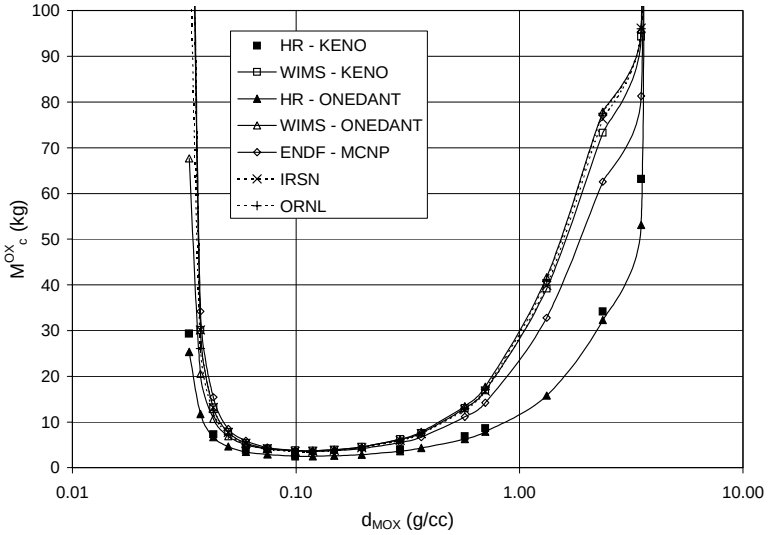


Figure 3. Critical mass as a function of the MOX density for the mixture of powder MOX with H_2O



The point to which the use of the Hansen & Roach library leads to over-conservative results is striking, especially for moderated mixtures. On the other hand we observe a good agreement between WIMS and the values reported by IRSN (APOLLO code) and ORNL (SCALE code). It is the signature of the fact that resonance treatment by modern codes tends to reduce the dispersion between the resulting k_{eff}. The solution obtained with ENDF-MCNP is also slightly more conservative than using WIMS, APOLLO and SCALE calculation schemes.

The results for the minimum values and the values at $m \sim 0$ are given in Tables 4 and 5. For low moderation, one observes a good agreement between ENDF-MCNP and the use of H&R cross-sections. It can be questioned whether the values obtained by WIMS, APOLLO and SCALE on the one hand, or the values obtained by ENDF-MCNP on the other hand, are the most reliable at zero moderation ratio.

Table 4. Minimum critical values for the mixture MOX-H_2O

Parameter	*m*	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (%)
R_c^{min} (cm)	50-100	14.2	16.4	17.1	13.8	17.2	**28**
M_c^{min} (kg)	250-300	2.77	3.58	3.69	2.51	3.69	**47**

Table 5. Critical values for the mixture MOX-H_2O at moderation ratio $m = 0$

Parameter	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (%)
R_c^0 (cm)	35.8	35.7	37.5	35.7	37.5	**5**
M_c^0 (kg)	671	667	773	667	773	**16**

The resulting dispersion between the codes is much more important than for the previous fissile system, on account of the homogeneous mixture of resonant nuclides of uranium and plutonium, more complicated to calculate.

Mixture of sintered MOX with H_2O

The results for the minimum values and the values at $m \sim 0$ are displayed in Tables 6 and 7. The observations are the same as for the MOX powder: for the minimum critical values, ENDF-MCNP is in agreement with WIMS and the use of H&R library leads to a strong over-conservatism, especially for the mass. At zero moderation, ENDF-MCNP is in agreement with the H&R library and no definitive conclusion can be given on the most reliable scheme.

Table 6. Minimum critical values for the mixture MOX-H_2O

Parameter	*m*	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (%)
R_c^{min} (cm)	30-40	15.3	18.9	18.8	15.0	19.2	**28**
M_c^{min} (kg)	80-100	8.1	12.1	12.7	7.7	12.5	**65**

Table 7. Critical values for the mixture MOX-H_2O at moderation ratio $m = 0$

Parameter	H&R KENOVa	ENDF MCNP4a	WIMS KENOVa	H&R ONEDANT	WIMS ONEDANT	Δ_{MAX} (%)
R_c^0 (cm)	27.6	27.7	30.0	27.9	29.6	**8.7**
M_c^0 (kg)	935	945	1 200	965	1 153	**28**

Conclusions

This work has been performed on the basis of calculation to experiment comparisons, as well as on the basis of calculation to calculation intercomparisons, since neither the range of the moderation ratios nor the number of fissile systems were sufficiently covered in the ICSBEP database.

The dispersion between the various calculations can be attributed partly to resonance treatment, well accounted for in modern codes like APOLLO, SCALE and WIMS.

However, the reduced conservatism of those criticality codes is questionable at low moderation, all the more since ENDF-MCNP is in a good agreement with the use of Hansen & Roach library, which was developed for fast and intermediate spectra.

As a consequence, we conclude that it would be desirable to complete the existing databases, such as ICSBEP, with criticality experiments involving Pu fissile systems (PuO_2, MOX, etc.) homogeneously mixed with a moderator at low moderation ratios ($m \sim 0$), but also at higher moderations, up to $m \sim 300$.

Such an issue could be addressed using powders or pellets under low moderation conditions. The practical consequence would be a possible revision towards an increase of the maximum allowable mass in the work units of a MOX fabrication plant, from 20-50%, while retaining the implementation of various safety coefficients, as is done today. Such an increase would make the operation of the plant more comfortable and flexible. An alternative impact could be an increase in the number of work units or a reduction of their size.

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Appendix

LIST OF EXPERIMENTS USED IN THE FIRST PART OF THE WORK – C/E COMPARISONS

#	ICSBEP label	Description of the sphere	$B^2\ 10^{-3}$ (cm^{-2})	FWE (MeV)	*m* = H/ (U+Pu)
1	PU-MET-FAST-001	Non-reflected Pu (95.2/4.5/0.3/0.0)	24.2	1.82	0
2	PU-MET-FAST-010	U_{nat} reflected Pu (94.8/4.9/0.3/0.0)	117	1.27	0
3	PU-MET-FAST-011	H_2O reflected Pu (94.5/5.2/0.3/0.0)	11.3	1.73	0
4	PU-MET-FAST-029	Non-reflected and hollowed sphere Pu (88.6/9.9/1.5/0.0)	345	1.80	0
5	PU-MET-FAST-031	CH_2 reflected and hollowed sphere Pu (88.9/9.7/1.4/0.0)	142	1.72	0
6	PU-MET-FAST-036	CH_2 + Cd reflected and hollowed sphere Pu (98.2/1.8/0.0/0.0)	142	1.72	0
	PU-SOL-THERM-009	Non-reflected solution of nitrate Pu			
7	#3	Pu (97.4/2.5/0.1/0.0)	2.59	0.503	2730
8	#3a			0.502	2734
	PU-SOL-THERM-020	Non-reflected solution of nitrate Pu			
9	#10	Pu (95.0/4.6/0.3/0.0)	4.34	0.773	617
10	#11		4.34	0.743	754
11	#12	Various Pu contents	4.34	0.816	466
12	#13		4.32	0.820	452
13	#14		4.32	0.821	540
14	#15		4.30	0.883	341
15	#16		4.30	0.740	759
16	HEU-MET-FAST-001	Non-reflected HEU (97.7 w/o)	129	1.57	0
17	HEU-MET-FAST-004	H_2O reflected HEU (97.7 w/o)	8.81	1.54	0
18	HEU-MET-MIXED-003	Three spheres: CH_2-HEU (89.9 w/o)-CH_2	15.8	1.48	0
	HEU-SOL-THERM-013	Non-reflected solution of nitrate HEU			
19	#1	U (93.2 w/o)		0.510	1282
20	#2		8.10	0.548	1094
21	#3	Various U contents		0.578	960
22	#4			0.591	905
23	HEU-SOL-THERM-012	H_2O reflected solution of nitrate HEU U (93.2 w/o)	5.30	0.508	1186
24	IEU-MET-FAST-003	Non-reflected IEU (36.8 w/o)	42.0	1.09	0
25	IEU-MET-FAST-009	CH_2 reflected IEU (36.8 w/o)	30.5	0.355	0
	LEU-SOL-THERM-002	H_2O reflected solution of nitrate LEU			
26	#1	U (4.9 w/o)	4.00	0.540	54
27	#2		4.00	0.571	50
28	MIX-MET-FAST-001	HEU reflected Pu (94.8 / 4.9 / 0.3 / 0.0)	220	1.73	0
	MIX-MET-FAST-002	Pu (94.9 / 4.7 / 0.3 / 0.0) surrounded by			
29	#1	HEU (93.3 w/o) and reflected by U_{nat}		1.34	0
30	#2		16.9	1.34	0
31	#3			1.33	0

SESSION III

Proposed Programmes and Presentation of Experimental Facilities

Chairs: P. D'Hondt, J. Gulliford

THE CRITICALITY LABORATORY OF VALDUC (FRANCE) AND ITS ABILITY TO MEET THE EXPERIMENTAL NEEDS FOR LOW-MODERATED MOX FISSILE MEDIA

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Abstract

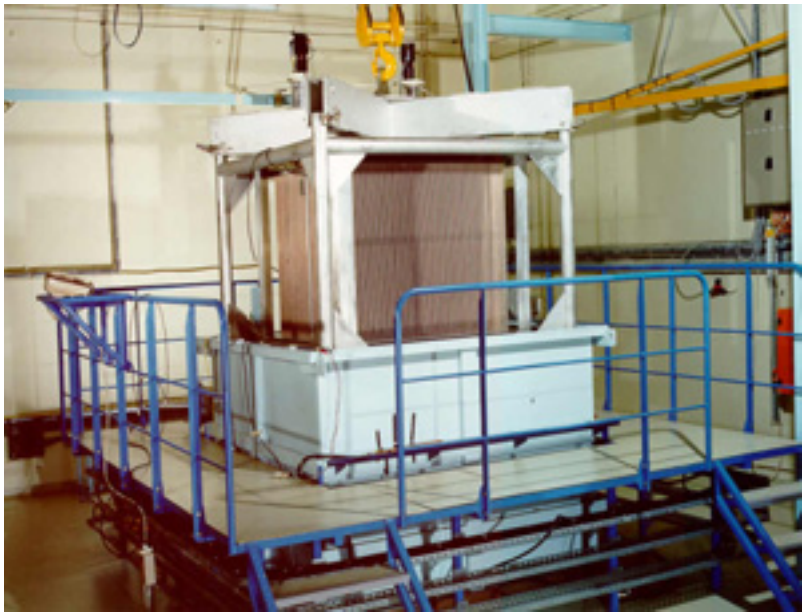
This paper briefly presents the main assets and potential of the French criticality laboratory at Valduc from the technical and human know-how point of view. The main part is devoted to the presentation of the experiments proposed by the CEA/DAM of Valduc which could be carried out with Apparatus B to meet the experimental needs for low-moderated MOX fissile media. Two types of experiment involving fuel rods and PuO_2 powders are studied and discussed.

Assets and potential of the Valduc criticality laboratory

The criticality laboratory of Valduc [1] was built in the sixties. Over its 40-year span of operation, a large amount of technical know-how has been accumulated.

The major facility of the laboratory today, Apparatus B (Figure 1), is a multi-purpose subcritical assembly that enables reproducing a great variety of criticality configurations, representative in particular of the fabrication, storage, transport and reprocessing of fissile materials in the form of rods or solutions. Moderating materials, absorbers, reflectors or shields (lead, concrete, polyethylene…) can be added under various forms.

Figure 1. Apparatus B – general view of fuel rods array setting up in the core tank



Apparatus B is mainly an experimental tank which contains the fissile core to be studied and the associated instrumentation. The core can be made with rods immersed in an aqueous moderator, fissile or not, or a fissile solution. The subcritical approach is carried out through the increase of the liquid in the core. The neutron amplification technique is used in this approach.

A broad range of various types of fuels are available in the laboratory, including plutonium and uranium solutions and fuels rods: 1 300 UO_2 rods with 4.74% U^{235} enriched and 2 500 high burn-up UO_2 rods with 1.1% plutonium and uranium with 1.57% U^{235} enriched. Both type are zircaloy clad with a fissile column height of 900 mm.

In order to perform high-quality experiments that can serve as benchmarks and meet requirements with respect to regulations and the management of nuclear materials, specific equipments are operated in the installation, such as physico-chemical analysis laboratories for nuclear materials.

Valduc criticality laboratory contribution to address experimental needs – proposed experiments

The most representative experiments corresponding to the fabrication process would be integral experiments with MOX powders of different densities and moisture contents.

Designs of experiments addressing the validation needs associated with MOX fabrication process were studied at CEA Valduc in the late eighties with cans of low-moderated (H/Pu from 0 to 6) and low-density (3.5 g/cc) plutonium-grade reactor powders arranged on a split table (up to 210 kg). The programme is comparable to the MARACAS programme performed in the same facility with uranium oxide powder.

However, the use of plutonium powder presents three main challenges: maintaining a definite temperature, guaranteeing a safe containment of the plutonium and assuring the accuracy of the moisture content and of the density. In addition, a large mass of MOX powder is required.

These potential difficulties, combined with the fabrication cost, were such that this programme was not undertaken.

Today, new experiments involving low-water-moderated MOX fuel rod arrays, dealing with various lattice pitches in order to cover different moderation ratios, are proposed. This design has been optimised taking into account the feasibility of the experiment, the possibility of using existing materials and equipment, the cost of fabrication of the fuel and the representativity of the experiment in terms of neutron spectrum (Tables 1, 3, 4, 5 and 6).

In addition to this programme, the Valduc laboratory is working on complementary experiments. Thus, driver UO_2 fuel rod arrays and PuO_2 powders (plutonium weapons-grade) using Apparatus B and some perturbation experiments with small amounts of PuO_2 powders have been studied using the SILENE and CALIBAN reactors. Only the most interesting experiments investigated are described and commented hereafter.

Experiments on Apparatus B with fuel rod arrays

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 the French fabrication process.

In order to reduce the number of rods needed and the fabrication cost, the MOX fuel pellets could be obtained from a blend of different existing powders in the COGEMA plant at Cadarache. Thus about 2 200 new fuel rods must be manufactured with the following characteristics:

- MOX fuel isotopic content:

Isotope	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{235}U	^{238}U
Weight %	0.26	69.31	25.08	1.88	1.01	2.46	0.25	99.75

- MOX rod characteristics:

Pellet diameter (cm)		0.735
Clad Zr_4	**Outside diameter (cm)**	0.94
	Inside diameter (cm)	0.82
Column height (cm)		100
Fuel density (g/cm^3)		10.4
Pu/(U+Pu+Am) weight ratio (%)		27.5

Different tight pitches (triangular and square) are considered to cover different moderation ratios close to powder (or fuel assembly) ratios.

Neutronic characteristics and similarity study with MOX powder

The neutronic characteristics are displayed in Table 1.

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

In this way, the neutronics characteristics of the proposed experiments were compared to those of MOX powders with water content varying from 1-5% in weight (Tables 2 and 3); these experiments cover the same energy range, when keeping a similar $H/Pu_{fissile}$ ratio.

This qualitative comparison has been quantified with the methodology used by the Oak Ridge National Laboratory, for slightly different fuel rods [2].

The study results show:

- A very high degree of similarity. The situations correspond to low-moderated reactor-grade MOX powders. This is confirmed using the ORNL sensitivity/uncertainty methodology; $c_k = 0.97$ for reactor-grade MOX powder containing 3% H_2O.

- A moderately high degree of similarity. The situations correspond to low-moderated weapons-grade MOX powders. This is confirmed using the ORNL S/U methodology; $c_k = 0.84$ for weapons-grade MOX powder containing 3% H_2O.

For this purpose, c_k is thus a correlation coefficient to assess the similarity between two configurations, with $c_k = 1$ indicating systems are identical and $c_k = 0$ indicating that systems are totally dissimilar. A value between 0.8 and 1 indicates similar systems.

Integral experiments with MOX fuel rods

The integral experiments consist of either a hexagonal or a square array of MOX fuel rods set up in the Apparatus B core tank (Figure 2). During the subcritical approach the configuration tested is reflected by 20 cm of water (on lateral and bottom sides). Table 2 shows some corresponding k_{eff} for these experiments; calculations are performed with the French CRISTAL criticality safety package [3] for a constant fissile column height immersed in water (h = 90 cm).

Table 2. K_{eff} for different cases

Hexagonal array a and rod numbers	**Triangular pitch (cm)**	**K_{eff} σ =100 pcm**	**Square array n × n**	**Square pitch (cm)**	**K_{eff} σ = 100 pcm**
27 – 2 107	0.96	1.01839	41 × 41	1.05	1.00856
26 – 1 951	0.98	1.00857	41 × 41	1.06	1.00866
26 – 1 951	1.0	1.01011	39 × 39	1.07	1.00580
26 – 1 951	1.02	1.02668	38 × 38	1.08	1.00559
25 – 1 801	1.04	1.00274	38 × 38	1.09	1.00770
			37 × 37	1.10	1.00950

Table 1. Neutronic characteristics (APOLLO2 calculations)

Media	^{240}Pu (%)	PuO_2 (%)	D (G/cc)	H_2O (%)	H/ U+Pu	H/Puf	GMF*	Q†	EALF	Fission (%)				
										1	2	3	4	5
MOX powder (reactor-grade)	17	30	5.5	3	0.93	5.37	48	0.062	6 116	38.4	7.7	46.0	7.4	0.4
		30	5.5	5	1.58	9.13	57	0.130	1 648	31.1	5.5	48.3	14.0	1.0
		12.5	5.5	3	0.93	11.8	59	0.135	1 281	30.2	5.0	47.6	15.6	1.5
		12.5	5.5	5	1.58	20.09	72	0.27	298	24.0	3.3	43.7	25.4	3.5
MOX powder (weapons-grade)	4	22	5.5	1	0.30	1.44	38	0.016	33 670	49.8	13.0	35.0	2.0	0.1
		22	5.5	5	1.58	1.50	70	0.187	731	28.3	4.7	45.3	19.6	2.1
		6.5	5.5	1	0.30	4.87	54	0.05	6 845	39.3	8.2	44.9	7.1	0.5
		6.5	5.5	5	1.58	25.38	89	0.32	61	20.2	2.2	33.7	36.1	7.8
Triangular pitch 0.98 cm	25.08	27.5	10.4	–	0.944	4.831	69	0.067		42.7	7.1	41.3	8.0	0.9

Media	^{240}Pu (%)	PuO_2 %	D (G/cc)	H_2O (%)	Capture (%)					Fission (%)				
					1	2	3	4	5	1	2	3	4	5
MOX powder (reactor-grade)	17	30	5.5	3	9.1	9.5	68.7	12.5	0.2	62.3	17.0	20.0	0.7	0.00
		30	5.5	5	6.3	6.0	64.7	22.4	0.6	61.5	15.3	21.7	1.4	0.01
		12.5	5.5	3	7.2	8	63.3	21	0.6	57.3	16.9	24.0	1.8	0.01
		12.5	5.5	5	5.1	5.1	56.3	32.1	1.3	57.2	15.0	24.8	3.0	0.05
MOX powder (weapons-grade)	4	22	5.5	1	14.9	19.1	62.6	3.3	0.05	61.2	21.6	16.9	0.3	0.00
		22	5.5	5	5.7	5.7	62.6	25	1	59.7	15.0	22.8	2.5	0.02
		6.5	5.5	1	11.5	16.3	64.4	7.7	0.2	54.0	22.0	23.2	0.9	0.00
		6.5	5.5	5	4.9	5	53.3	34.2	2.6	54.7	14.5	25.7	4.9	0.15
Triangular pitch 0.98 cm	25.08	27.5	10.4	–	8.99	9.25	64.4	16.8	0.53	62.6	16.7	19.6	1.09	0.01

* GMF, average group causing fission in the Xmas 172-group energy structure of neutronic library; the corresponding energy given is the midpoint of the energy group.

† Q, slowing down density is the number of neutrons that slow down past 4 eV per fission neutron.

137

Figure 2. Hexagonal array configuration

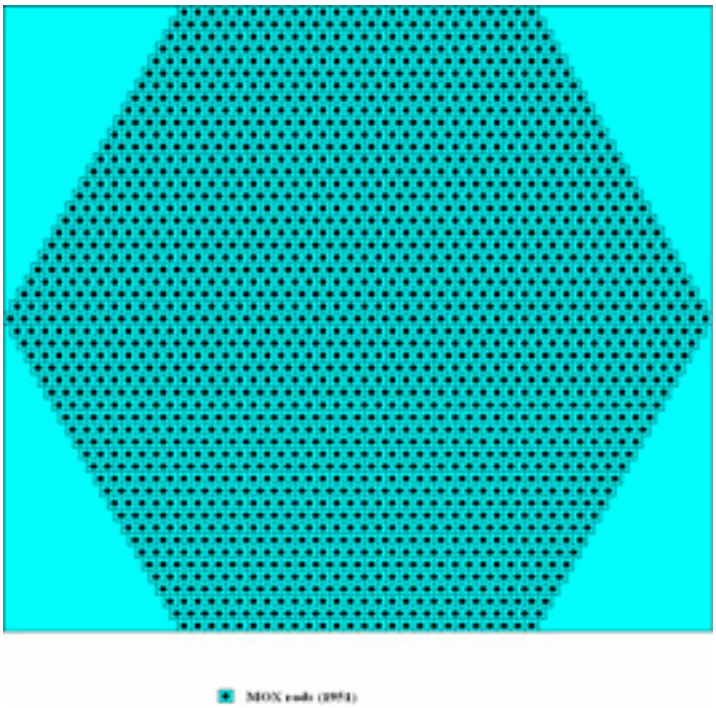


Table 3. Neutronic characteristics range of some integral experiments

	Pitch		% water content equivalent	Vmod/Vox	H/Puf	GMF	Average energy (eV)
	Δ	□					
Experiments proposed	0.96		4.52	0.2455	3.626	65.30	142.75
	0.98		5.98	0.3297	4.795	68.36	71.785
	1.0		7.25	0.4055	5.988	70.59	53.59
	1.02		8.60	0.488	7.207	73.40	42.845
	1.04		9.94	0.572	8.447	76.96	32.115
		1.05	15.66	0.9628	14.219	89.05	5.753
		1.06	16.34	1.0126	14.954	89.77	5.753
		1.07	17.01	1.0628	15.696	91.96	4.586
		1.08	17.68	1.1134	16.443	93.79	3.6905
		1.09	18.34	1.1646	17.199	94.56	3.3405
		1.1	19.00	1.2162	17.961	96.27	2.744
		>					
Reactor-grade			1 to 5		3 to 15.5	68 to 86	71.8 to 8.75 eV
Weapons-grade			1 to 5		5 to 26	75 to 100	35.5 to 2.24 eV

Δ – triangular pitch, □ – square pitch.

Mixed UO_2 and MOX fuel rods arrays

In order to limit the number of fuel rods (five less than initially) and thus to reduce the total cost of the programme (fabrication, transportation, storage, etc.) another type of experiment is being studied. Based on the principle of "mixed" fuel rods arrays, experimental configurations consist of an internal array of MOX rods (21 × 21) surrounded by an array of UO_2 rods (41 × 41) used as a driver core (Figure 3). The square pitch of both these lattices is 1.05 cm.

Figure 3. Mixed arrays configuration

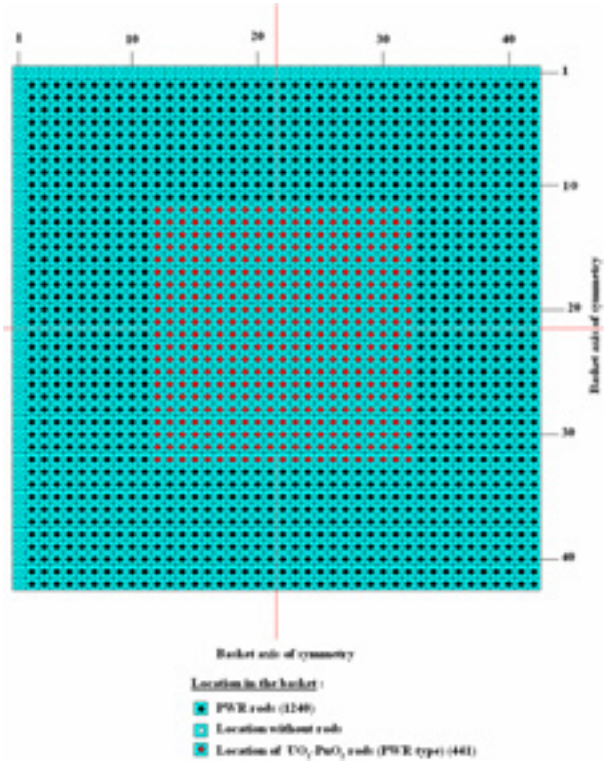


Table 4. Neutronic characteristics of a mixed array

	Square pitch	% water content equivalent	Vmod/Vox	H/Puf	GMF	Emoy (eV)
Experiments proposed	1.05	15.66	0.9628	14.219	83.13	12.46
Reactor-grade		1 to 5		3 to 15.5	68 to 86	71.8 to 8.75 eV
Weapons-grade		1 to 5		5 to 26	75 to 100	35.5 to 2.24 eV

Experiments on Apparatus B with driver UO_2 fuel rods arrays and PuO_2 powders

These experimental configurations involve cans filled up with PuO_2 powders placed inside a cylindrical zircaloy container. This assembly is set up at the centre of a UO_2 fuel rod driver core [1.05 cm square pitch and 1 258 rods (Figures 4 and 5)].

Figure 4. UO_2 driver array and PuO_2 powder cans section

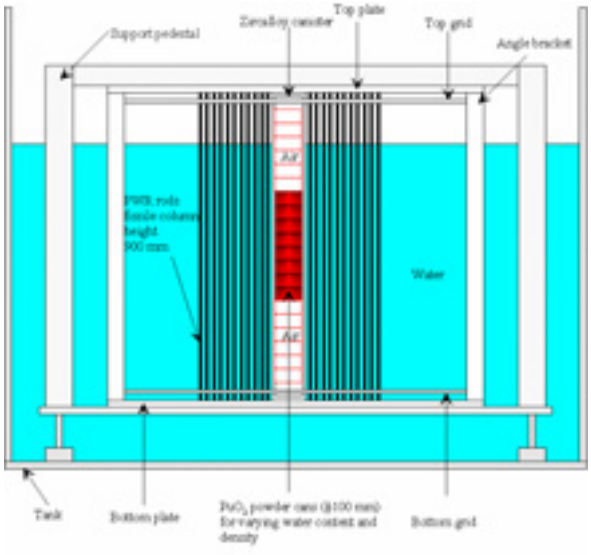
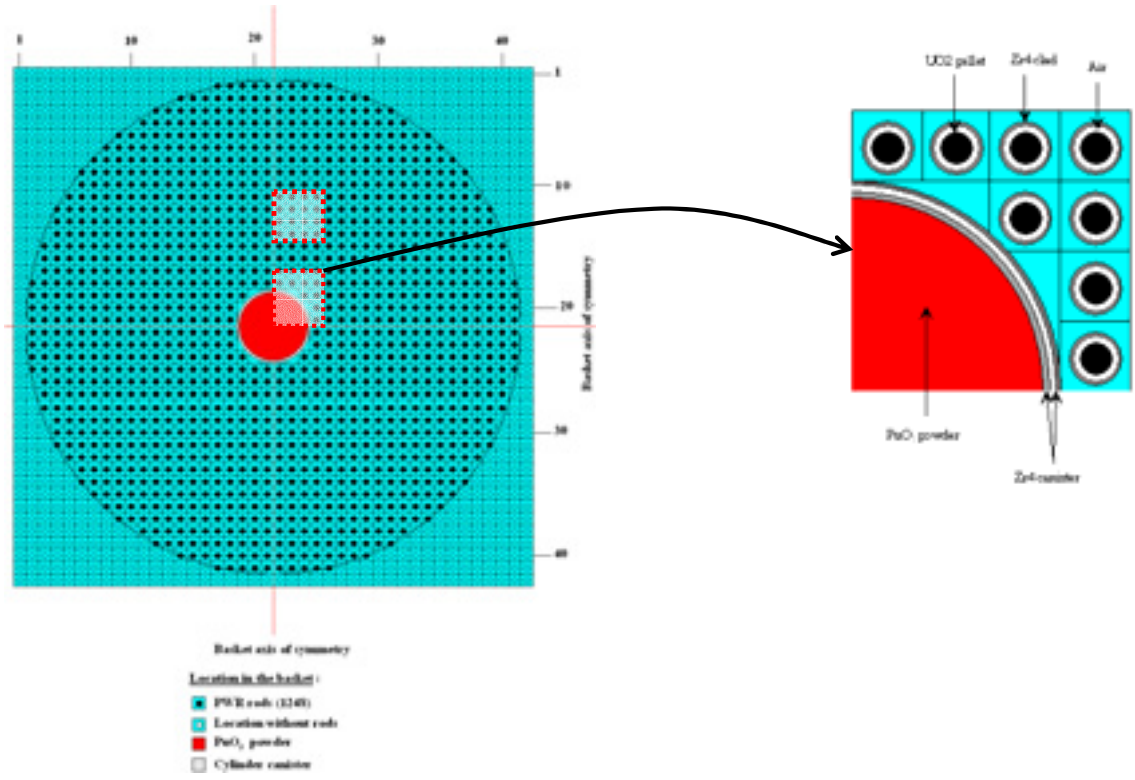


Figure 5. Bird's eye view of the UO_2 driver array and PuO_2 powder can



The minimum PuO_2 powder mass required for performing representative experiments is about 22 kg, for the following plutonium weapons-grade:

Isotope	Weight %
^{239}Pu	92
^{240}Pu	8

Table 5. Example of neutronic characteristics of experiments with PuO_2 powders

Experiments	Powder density	% water content	H/Puf	GMF	Average energy (eV)
	5.5	0	0	40.35	28 300
		1	0.330	42.34	20 705
		2	0.667	43.72	15 825
		3	1.012	45.51	10 129
		4	1.363	46.94	8 292
		5	1.721	48.81	5 268

Moreover, some additional and complementary experiments are proposed on the CALIBAN and SILENE reactors operated by the division. This means that it is possible to test small quantities of this weapons-grade plutonium powder (about one hundred grams) inside the central cavity of these reactors. This perturbation experiment concept enables highly accurate reactivity worth measurements for various neutron energy spectra:

- Fast neutron energy spectrum: CALIBAN, “metal” reactor.
- Thermal neutron energy spectrum: SILENE, “solution” reactor.

Preliminary neutronic studies related to these experiments are not fully achieved and feasibility is not demonstrated. However, the final results and the potential interest of such experiments will be presented and discussed during the workshop.

Conclusion

Integral experiments with MOX or PuO_2 powders are not achievable without technological difficulties, so in the framework of this project Valduc laboratory has worked in the following two directions:

- Feasibility of the experimental programme proposed by IRSN [2] involving MOX fuel rods arrays.
- Proposition of other experiments with PuO_2 powders which could be carried out with Apparatus B, CALIBAN or SILENE reactors.

Even though the laboratory has not performed the full study required (neutron balance data and sensitivity data, etc.) it is shown that the neutronic characteristics range covered by the integral experiments proposed with MOX fuel rods meet the validation needs for low-moderated fissile MOX media. Thus in conclusion, these experiments could be carried out at CEA Valduc in the criticality facility Apparatus B without particular difficulties if the health physics protection aspects are taken into

Table 6. Neutronic characteristics synthesis

			PuO_2 %	D G/cc	Vmod/Vox	Water content %	H/U+Pu	H/Puf	GMF*	Fission (%)					Capture (%)					Flux (%)				
										G1	G2	G3	G4	G5	G1	G2	G3	G4	G5	G1	G2	G3	G4	G5
										E > 111 keV	11.1 keV < E < 111 keV	9.91 eV < E < 11.1 keV	0.1 eV < E < 9.91 eV	E < 0.1 eV	E > 111 keV	11.1 keV < E < 111 keV	9.91 eV < E < 11.1 keV	0.1 eV < E < 9.91 eV	E < 0.1 eV	E > 111 keV	11.1 keV < E < 111 keV	9.91 eV < E < 11.1 keV	0.1 eV < E < 9.91 eV	E < 0.1 eV
MOX powder (reactor-grade)			30	5.5	–	3	0.93	5.37	48	38.41	7.75	46.03	7.43	0.38	9.09	9.47	68.67	12.54	0.23	62.28	16.99	20.01	0.72	0.002
			30	5.5	–	5	1.58	9.13	57	31.09	5.53	48.31	14.02	1.04	6.29	6.04	64.66	22.44	0.57	61.53	15.26	21.75	1.45	0.01
			12.5	5.5	–	3	0.93	11.8	59	30.25	5.00	47.65	15.64	1.46	7.21	7.99	63.27	20.96	0.57	57.27	16.90	24.06	1.76	0.01
			12.5	5.5	–	5	1.58	20.09	72	24.05	3.34	43.73	25.39	3.49	5.13	5.09	56.35	32.12	1.31	57.19	14.98	24.80	2.99	0.05
MOX powder (weapons-grade)			22	5.5	–	1	0.30	1.44	38	49.93	13.05	35.00	2.04	0.08	14.92	19.15	62.59	3.30	0.05	61.25	21.61	16.87	0.27	0.0004
			22	5.5	–	5	1.58	7.50	70	28.30	4.74	45.26	19.59	2.11	5.71	5.73	62.56	24.99	0.98	59.65	15.02	22.81	2.51	0.02
			6.5	5.5	–	1	0.30	4.87	54	39.28	8.22	44.89	7.10	0.52	11.48	16.33	64.38	7.66	0.16	53.90	21.99	23.19	0.91	0.004
			6.5	5.5	–	5	1.58	25.38	89	20.21	2.17	33.69	36.13	7.79	4.92	4.96	53.32	34.22	2.58	54.71	14.51	25.75	4.88	0.15
Experiments proposed	MOX arrays	Δ pitch 0.96	27.5	10.4	0.246	–	0.713	3.626	65	36.95	6.22	31.44	10.46	14.93	8.10	8.83	55.37	19.52	8.17	64.26	16.54	17.87	1.12	0.21
		□ pitch 10.5	27.5	10.4	0.963	–	2.797	14.22	89	21.17	2.72	32.01	25.63	18.47	3.31	3.21	43.86	41.16	8.46	60.12	13.14	22.19	4.01	0.54
	Mixed array	□ pitch 10.5	27.5	10.4	0.963	–	2.797	14.22	83	22.08	3.01	35.49	28.23	11.19	3.30	3.30	45.44	43.15	4.80	59.08	13.48	22.92	4.21	0.30
	Driver array	PuO_2 powder	100	5.5	–	1	0.304	0.330	42	58.18	7.67	22.35	9.61	2.19	10.83	8.56	51.81	25.56	3.23	77.08	11.59	10.31	1.013	0.009

* GMF, average group causing fission in the Xmas 172-group energy structure of neutronic library; the corresponding energy given is the midpoint of the energy group.
Δ – triangular pitch, □ – square pitch.

account at the earlier stage of the designing (transportation, storage and handling). We should mention that the experimental core tank is available and the Apparatus B control system and instrumentation are adapted but fuel rod containers and storage equipment adaptations are necessary. This programme could easily be extended to higher pitches and various configurations: arrays in interaction, rod array heterogeneities, poisoning moderator (B, Gd, fission products, etc.), as well as various reflector and/or absorber solid materials.

If experiments proposed with PuO_2 powders (plutonium weapons-grade) present complementary relevant interest for validation purposes, CEA/DAM Valduc could produce specific powder batches (about 22 kg) with the adapted technology to assure the accuracy of the moisture content and the density.

Obviously after these preliminary designs, all experiments retained should be optimised as regards fuel rod number, pitches of arrays and total mass, density or water content for powders.

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EXPERIMENTAL STUDY PROGRAMME ON CRITICALITY OF MOX FUEL SYSTEMS AT THE BFS FACILITY

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Abstract

In the paper, there is a description of the BFS facility, on which an experimental study programme is planned concerning the criticality of MOX fuel-hydrogen moderator systems. The programme implies the creation of a series of critical assemblies with MOX fuel (Pu content ranging from 10-30%) and polyethylene (0-10%). A brief description of the earlier-tested critical assembly BFS-59 is presented.

BFS critical facilities

The complex of fast critical facilities of the SSC RF-IPPE, which includes two critical facilities (BFS-1 and BFS-2) and the MI-30 electron accelerator is a unique experimental base for studies on fast reactor neutronics, safety, core optimisation, incineration of actinides and utilisation of weapons-grade plutonium.

The reactor materials used in the critical facilities (including 9 tonnes of high-enrichment uranium and plutonium, 280 tonnes of fertile materials, 250 tonnes of coolant and 120 tonnes of structural materials) make it possible to assemble both full scale complicated models of fast reactors and simple ("pure") core models and carry out tests of these models for the correction of nuclear data and modification of analytical techniques.

The BFS-1 facility is used for studies on neutronics of the simplest assemblies (benchmarks), as well as design studies of the models of research and power fast reactors up to 1 000 MWth with various core and blanket arrangements using assorted fuels, fertile materials and coolants (sodium and lead).

This facility is also used for experimental studies on LWR and other types of reactors.

Experimental studies on the BFS-59 assembly

Experimental studies on the assemblies using polyethylene were performed earlier. As regards the problem under consideration, additional data can be obtained using the BFS-41 and BFS-42 assemblies (Pu content in the fuel is 6.6%, and H/Pu ratio is equal, respectively, to 0 and 0.7), BFS-49 assembly (Pu content in the fuel is 12,5%, and H/Pu ratio is equal, respectively, to 0 and 0.7) and "tight grid" models on BFS-57 and BFS-59 assemblies.

Description of BFS-59 assembly is given as an example as well as brief information on the experimental studies.

The central insert of the BFS-59 critical assembly with plutonium fuel contains reactor-grade plutonium disks (^{240}Pu weight content is 9.94%), depleted uranium oxide and polyethylene disks. Fuel enrichment in the insert is 8.12% and the hydrogen ratio is p^{H}/p^{Pu239} = 26.2. The central insert has a 31-cm radius (134 fuel tubes) and is 76.65 high; the insert was surrounded by the driver containing plutonium of different isotope compositions and a larger overall content of ^{239}Pu (95%). The driver thickness was 9.9 cm (91 fuel tubes). A 30-cm thick layer of UO_2 was used as radial and axial reflector. The condition of critical facility with all control rods on the upper limit position is +11 cents. Aluminium tubes were used within the core with the exception of the control rods, which were steel tubes. The elementary cell of the central insert contained one disk of reactor-grade plutonium, five disks of depleted uranium oxide (1 cm thickness) and six polyethylene disks of 4 mm thickness. The total number of such cells in the driver rod was 16 (see Annex). A description of the experimental studies performed using the critical assembly are given below:

- *Measurements of reaction rates*. Measurements of fission reaction rate ratios F238/F235 and F239/F235 were made using section fission chambers in the central area of the assembly and small fission chambers in the inter-tube space in the vicinity of the assembly central area. Results of measurements are presented in Table 1.

Table 1. Results of the measurements of reaction rates

H, mm	F238/F235	F239/F235
-30	0.00244 ± 0.00005	2.09 ± 0.02
0	0.00256 ± 0.00005	2.09 ± 0.02

H – distance from the assembly midplane.

Absolute technique was used for measurements of capture integrals in the uranium dioxide layers and absolute rate of ^{239}Pu fission in the inter-tube space. The results of measurements are as follows:

$$C238/F239 = 0.0118 \pm 0.00003$$

Measurements of capture cross-sections in ^{238}U were made using 0.1-mm thick uranium metal foil.

- *Normalised fission integral in the cell was measured.* It was used as a denominator for obtaining K^+ value on the basis of the results of cell reactivity measurements. Measurements were made using the following values for assembly subcriticality: -10, -15, -20, -38 and -58 cents. The absolute fission rate of ^{239}Pu was measured in the inter-tube space, where the neutron source (^{252}Cf) of certain activity was also placed. The result of the measurements was $K^+ = 1.13 \pm 0.02$. The K^+ value obtained by normalisation to the fuel reactivity (+6.6 cents) turned out to be 1.12 ± 0.02.

- *Reactivity coefficients of some materials were measured in the central area of the BFS-59 assembly*. Among these materials there are fuel, standard scattering and absorber materials: ^{235}U, ^{239}Pu, CH_2, ^{10}B. The results of the measurements are presented in Table 2.

Table 2. Results of measurements of reactivity coefficients of various materials

Samples	Weight (g)	Worth, mcents
^{235}U (88.6%)	16.22(2)*	4214 ± 5†
^{239}Pu (95.96%)	6.023(2)	1396 ± 6
	28.44(4)	4856 ± 7
PuO_2 (67.6% ^{239}Pu)	6.243(4)	1499 ± 5
CH_2	13.352(4)	2637 ± 5
	2.658(4)	179 ± 5
	4.659(4)	335 ± 5
	6.867(2)	603 ± 4
^{10}B (82.4%)	0.4604(4)	-6144 ± 9
C	23.39(4)	-71 ± 6
Zr	73.63(4)	-277 ± 5
	86.3(4)	-332 ± 5

* The number in brackets is the number of simultaneously oscillated samples.
† Statistical errors.

- *Neutron spectrum was studied by time-of-flight method* (0-1 keV energy range).

- *Measurement of the reactivity of the elementary cell*. This was done by measuring the reactivity of the system before and after cell introduction into the central area of the critical assembly. Reactivity of the cell and fuel (plutonium disk) caused by the cell introduction is, respectively, equal to + 4.45 ± 0.05 cents and + 6.6 ± 0.1 cents.

New series of tests

Preparation work has recently begun on the BFS-97-1 assembly at the BFS-1 critical facility. The main parameters of this assembly are as follows:

Pu/Pu+4	22.5%
H/Pu	0%
Cell composition	UO_2, Pu, UO_2, Pu, UO_2
Material of radial and axial reflectors	Uranium oxide
Diameter	~ 60 cm
Height	~ 40 cm

In addition to criticality detection and certification of the assembly, the experimental programme implies measurements of ratios of fission and capture cross-sections of ^{238}U to fission cross-section of ^{235}U.

Further steps would include the replacement of the radial reflector with polyethylene and the introduction of polyethylene to the core.

Proposals have been prepared as concerns the following critical assemblies:

- Assembly with ~30% plutonium enrichment, H/Pu ~6.3.

- Assembly with ~15% plutonium enrichment, H/Pu ~6.5.

Planned critical assemblies

Parameters of a series of critical assemblies are presented in Table 3. The first column provides the experiment label, which is composed of the word “MOX” followed by plutonium enrichment values and then a value referring to water content. Ratios of plutonium and depleted uranium dioxide disk numbers are given in the second column.

Table 3. Planned critical assemblies

BFS	PU:UO_2	H/PU	R, cm	H, cm	V, l	M_{PU}, kg
MOX-10-5	1 : 4	17.3	50.4	39.3	314	130
MOX-15-5	2 : 5	11.5	29.5	36.9	101	63
MOX-15-3	2 : 5	6.5	39.4	32.2	157	114
MOX-22-0	2 : 3	0.0	28.0	35.6	87	114
MOX-30-5	1 : 1	6.3	25.8	27.6	58	57
MOX-10-1	1 : 4	4.7	50.4	70.3	562	279
MOX-15-0	2 : 5	0.0	46.4	38.6	262	220
MOX-15-1	2 : 5	2.3	58.0	29.9	316	245
MOX-15-3	2 : 5	6.5	39.4	32.2	157	114
MOX-22-0	2 : 3	0.0	25.5	35.6	73	95
MOX-22-1	2 : 3	2.3	24.2	36.3	67	77
MOX-22-3	2 : 3	4.7	21.9	40.5	61	63

R, H and V are, respectively, radius and height of the assembly and core volume.
M_{Pu} is the plutonium mass.
H/Pu is the hydrogen/plutonium nuclei ratio.

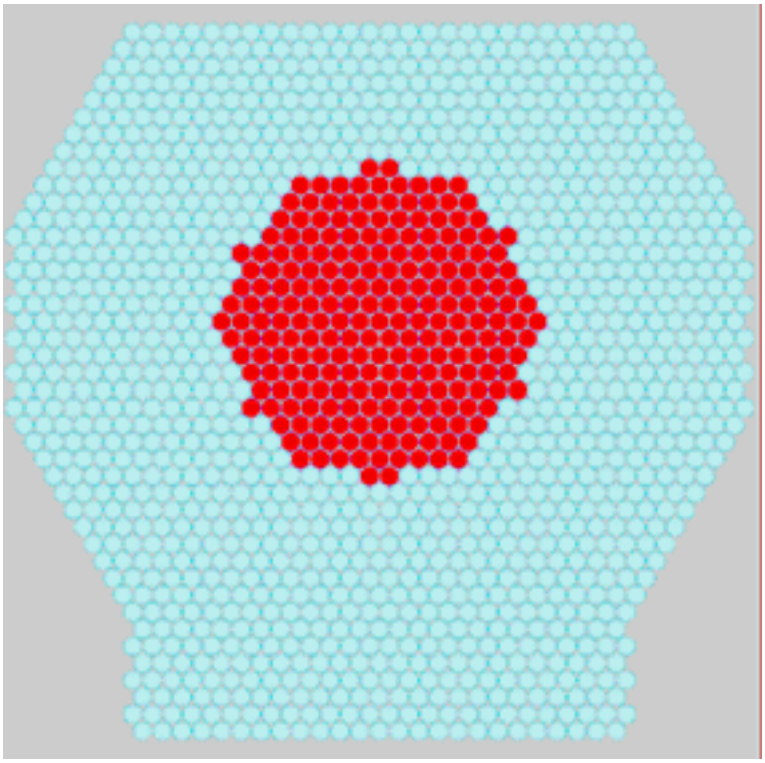
REFERENCE

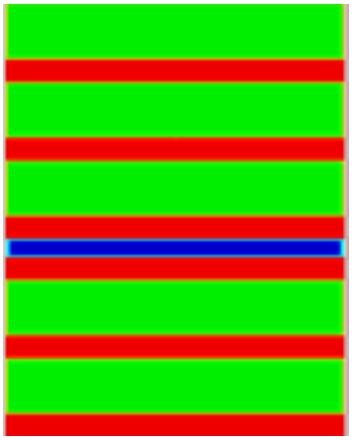
[1] Belov, S.P., *et al*., *Results of Tests Performed on Critical Facilities BFS-57 and BFS-59*, Preprint FEI-2681, Obninsk (1998).

Annex A

CONFIGURATION OF BFS ASSEMBLIES

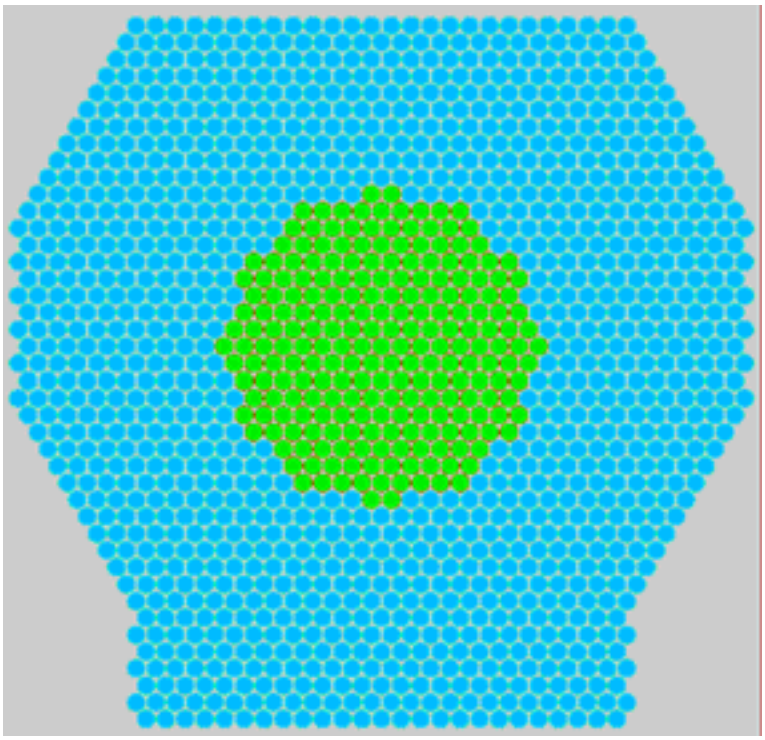
BFS-59 (MOX fuel)

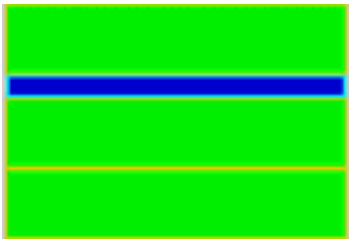




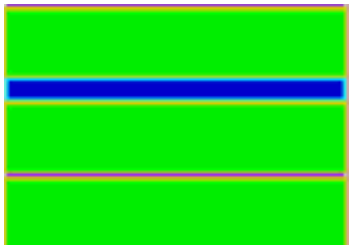
PU/(PU+U) – 8%
H – 5 wt.%

BFS-49 Series





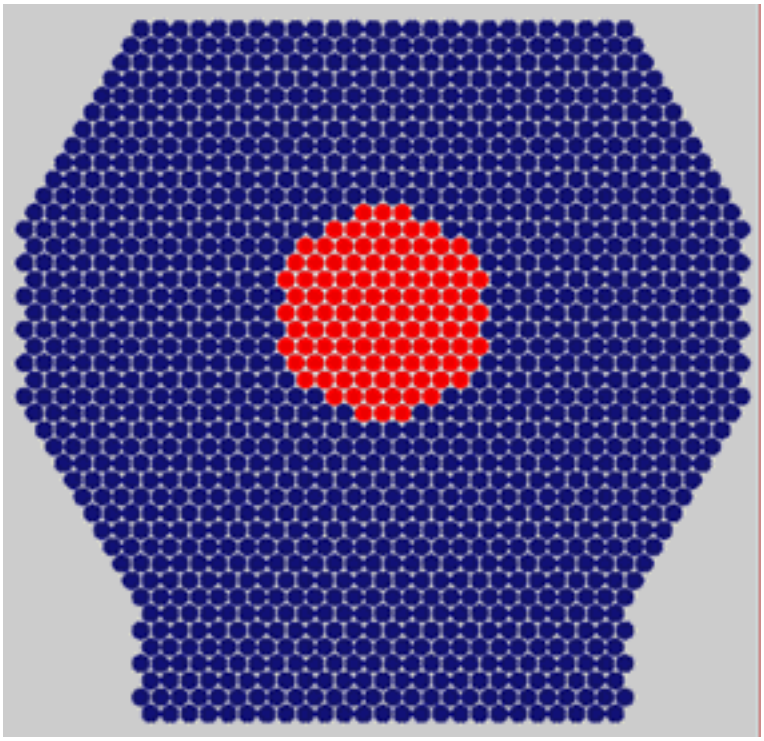
BFS-49-1



BFS-49-3

PU/(PU+U) – 12.5%
H – 0 wt.% (BFS-49-1), ~0.5 wt.% (BFS-49-3)

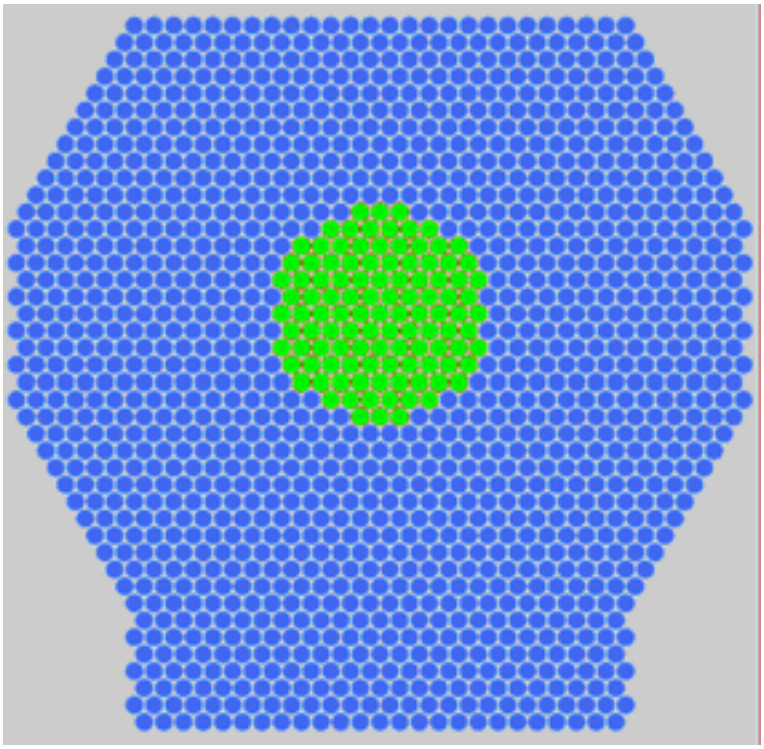
BFS-MOX planned



PU/(PU+U) – 30%
H – 5 wt.%

Ng	E_{max}	Balance * 100			Sensitivity * 100		
		F9	C9	C8	F9	C9	C8
1	10 MeV	7.3	.3	.8	16.7	-.4	-.8
2	100 keV	1.1	.4	.7	2.3	-.5	-.8
3	**10 keV**	**7.4**	**4.7**	**3.7**	**11.1**	**-6.8**	**-4.4**
4	10 eV	6.7	3.2	1.4	7.9	-5.5	-1.3
5	0.1 eV	8.5	3.4	0.8	6.3	-6.0	-1.2
∑		**31.1**	**12.0**	**7.3**	**44.3**	**-19.2**	**-8.4**

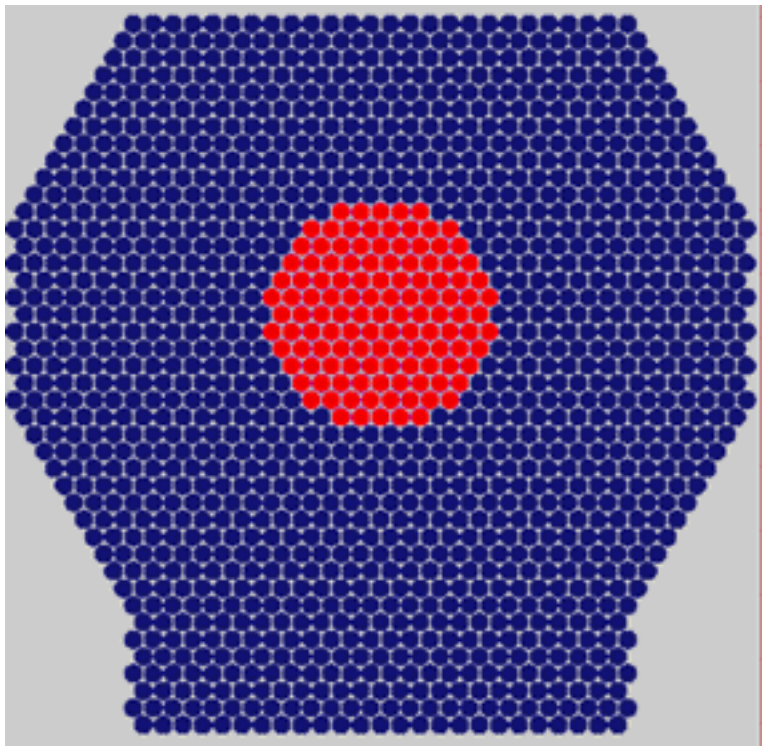
BFS-MOX planned

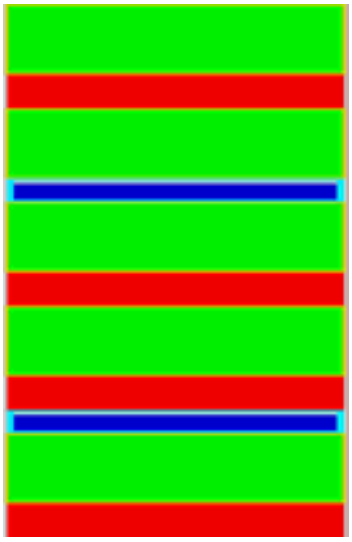


PU / (PU+U) – 22.5%
H – 0 wt.%

Ng	E_{max}	Balance * 100			Sensitivity * 100		
		F9	C9	C8	F9	C9	C8
1	10 MeV	13.6	.8	2.8	32.3	-.9	-2.8
2	100 keV	2.3	.7	1.9	5.6	-.7	-1.9
3	**10 keV**	**2.8**	**1.8**	**2.1**	**3.6**	**-2.0**	**-1.8**
4	10 eV	3.0	1.4	.7	2.6	-1.9	-.6
5	0.1 eV	6.3	2.5	1.0	4.4	-4.2	-1.5
∑		**28.1**	**7.3**	**8.4**	**48.5**	**-9.7**	**-8.6**

BFS-MOX planned

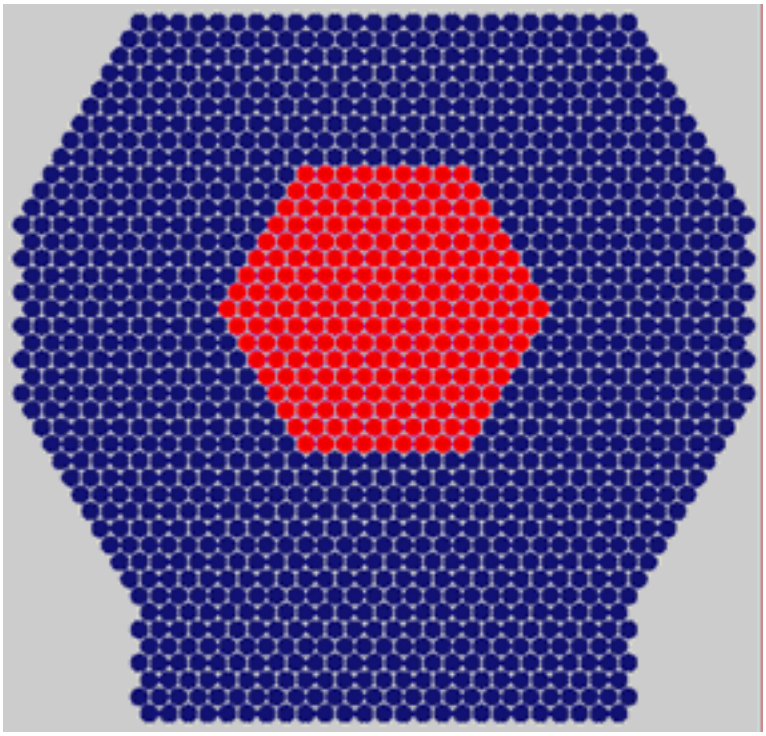


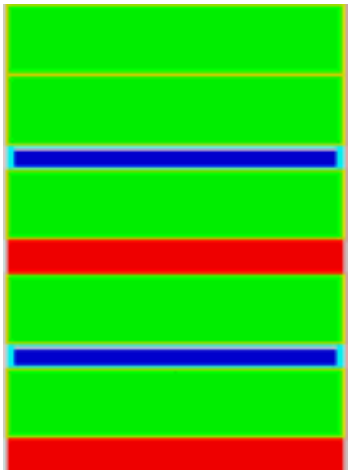


PU/(PU+U) – 15%
H – 5 wt.%

Ng	E_{max}	Balance * 100			Sensitivity * 100		
		F9	C9	C8	F9	C9	C8
1	10 MeV	5.7	.2	1.4	12.5	-.3	-1.4
2	100 keV	.9	.3	1.3	1.6	-.4	-1.5
3	**10 keV**	**6.5**	**4.1**	**7.8**	**11.5**	**-5.8**	**-8.5**
4	10 eV	7.3	3.6	3.1	9.6	-6.1	-3.1
5	0.1 eV	7.5	3.1	2.4	6.8	-4.9	-3.0
∑		**27.8**	**11.2**	**15.9**	**42.0**	**-17.5**	**-17.5**

BFS-MOX planned

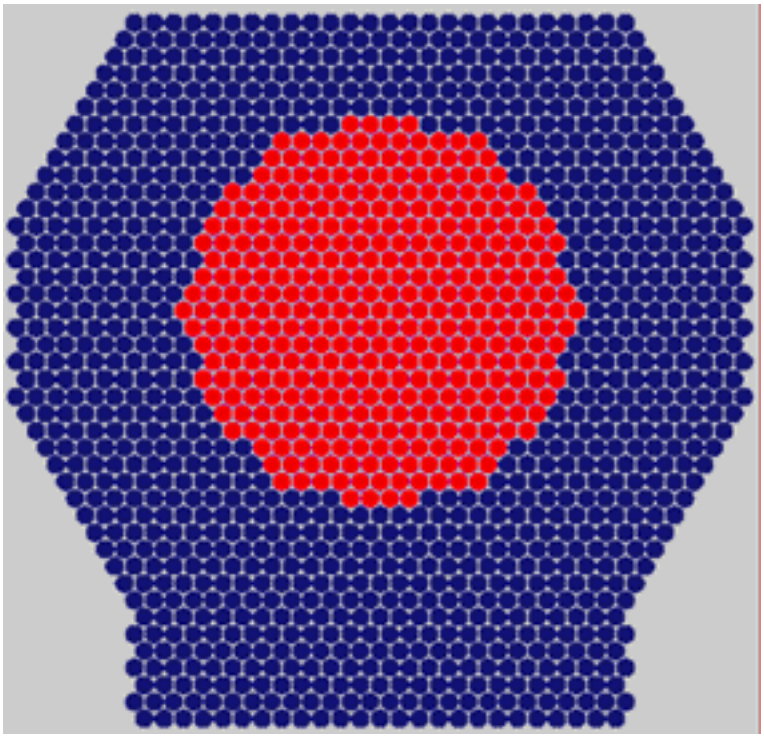


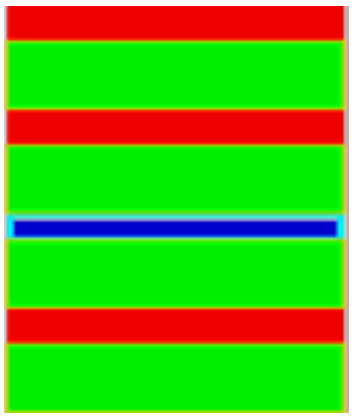


PU/(PU+U) – 15%
H – 3 wt.%

Ng	E_{max}	Balance * 100			Sensitivity * 100		
		F9	C9	C8	F9	C9	C8
1	10 MeV	7.1	.4	1.9	16.8	-.4	-1.9
2	100 keV	1.2	.4	1.8	2.8	-.4	-1.9
3	**10 keV**	**7.2**	**4.6**	**8.6**	**11.8**	**-6.0**	**-8.6**
4	10 eV	5.9	2.8	2.5	7.1	-4.6	-2.2
5	0.1 eV	6.3	2.6	1.8	4.7	-4.4	-2.3
∑		**27.7**	**10.7**	**16.5**	**43.3**	**-15.8**	**-17.0**

BFS-MOX planned





PU / (PU+U) – 10%
H – 5 wt.%

Ng	E_{max}	Balance * 100			Sensitivity * 100		
		F9	C9	C8	F9	C9	C8
1	10 MeV	4.7	.2	1.7	10.5	-.2	-1.7
2	100 keV	.7	.2	1.6	1.4	-.2	-1.7
3	**10 keV**	**5.5**	**3.4**	**10.8**	**9.0**	**-4.5**	**-10.3**
4	10 eV	7.0	3.5	4.8	9.0	-6.1	-4.2
5	0.1 eV	7.5	3.1	4.2	6.1	-5.7	-5.3
∑		**25.4**	**10.4**	**23.1**	**36.1**	**-16.8**	**-23.3**

Annex B

DESCRIPTION OF DISKS USED IN BFS-59

Table 1. Parameters of disks

Disks	Thickness of disk with clad (cm)	Diameter of disk with clad (cm)	Density of the core material (g/cm^3)	Density of cladding (g/cm^3)
UO_2 – 1	0.975	4.67	9.80	2.7
Pu (two types)	0.316	4.642	15.60	7.9
UO_2 – 2	0.173	4.67	9.80	7.9
CH_2 (3 items)	0.071	4.67	0.83	–
CH_2	0.386	4.67	0.91	–
Al	0.120	4.681	2.70	–

Table 2. Weight characteristics (g)

	Disk total weight	Cladding weight	Notes*
UO_2 – 1	143.91 ± 0.35	4.37(Al)+0.009% wt. H	See Table 3
Pu – 1	66.95 ± 0.15	12.38(SS)+0.50(Ni)	See Table 4
Pu – 2		11.57 (SS)	See Table 4
UO_2 – 2	22.90	5.73 (SS) + 0.17 (Al)	See Table 3
CH_2 (3 items)	1.004	–	–
CH_2	6.022 ± 0.05	–	–
Al	5.27 ± 0.08	–	0.4% wt. Fe

* Total disk weight includes: UO_2 – 1: "thick" disk of depleted uranium dioxide.
UO_2 – 2: "thin" disk of depleted uranium dioxide.

Table 3. Composition* (wt.%) of Pu disks

	Pu – 1	Pu – 2
^{239}Pu	86.81	93.55
^{240}Pu	9.94 ± 0.15	4.55 ± 0.10
^{241}Pu	1.13 ± 0.05	0.25 ± 0.05
Ga	2.12 ± 0.15	1.65 ± 0.15

* Data are given for the average fabrication date, May 1967.

Weight characteristics of extended elements (tubes and displacers) (g/cm):

Al tube	4.103 ± 0.010
SS tube	12.56 ± 0.030
Al displacer	1.042 ± 0.005
SS triangle seat	8.64

There are small amount of impurities in the aluminium tubes and displacers (0.4% Wt-Fe). Composition of SS structural elements is presented in Table 4.

Table 4. Composition of SS items (wt.%)

Isotopes	C	Al	Ti	Cr	Mn	Fe	Ni
%	0.10	0.85	0.62	18.00	1.50	69.43	9.50

CRITICAL EXPERIMENT WITH LOW-MODERATED MOX RODS IN VENUS

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Abstract

Although critical experiments involving PuO_2 and MOX powders are highly desirable, such Pu powders are difficult to model due to uncertainties linked to the moisture content, the density and the temperature. This is why powder experiments are sometimes substituted by well-characterised low-water-moderated lattices of MOX rods. The possibility of loading such a tight lattice test bundle in the VENUS critical facility and performing an experiment at low cost and over a short time period is considered and demonstrated. The paper is organised into two main parts: 1) VENUS measurement capabilities and 2) preliminary design calculations of critical configurations accommodating a central test bundle with low-moderated MOX pins.

Experimental techniques used in the VENUS critical facility

Description of the VENUS critical facility

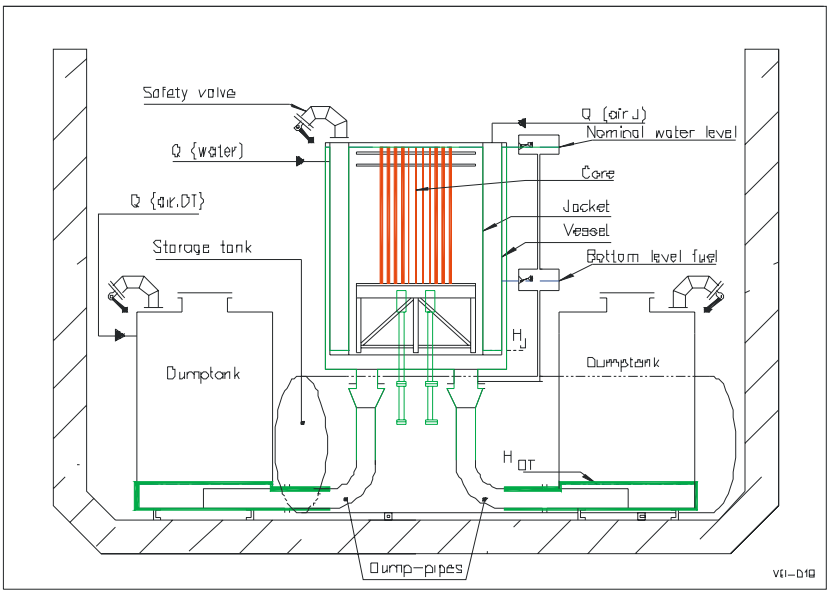
The VENUS critical facility is a water-moderated zero-power reactor. It consists of an open (non-pressurised) stainless steel cylindrical vessel including a set of grids (square lattice pitches of 1.26 and 1.31 cm available) which maintain fuel rods in a vertical position.

After a fuel configuration has been loaded, criticality is reached by raising the water level within the vessel. Via a series of pipes and pumps, water is led to the vessel from a storage tank that contains water when the reactor is not in operation. Reactivity control is obtained by controlling the water level in the vessel. With the help of the pumps the water level can be controlled in a rough way (about 0.5 mm). Fine-tuning is achieved by the so-called water level control rods (WLCR), which are thick aluminium bars. By regulating their insertion into the water the water level can be controlled (in principle) in steps of 0.0001 mm, sufficiently well for all purposes. Reactivity is sometimes controlled by means of absorbing rods if experiments are performed at a fixed nominal water level.

Since the neutron flux is very low, no water circulation is needed to keep the fuel rods at low (room) temperature.

Whilst the water level goes up in the vessel, air is injected simultaneously into the jacket (a closed stainless-steel circular cavity located around the vessel) and the two dump tanks (i.e. two closed tanks each as large as the vessel itself) connected to the vessel. The dump tanks and the jacket are kept under pressure, allowing the water level in the open vessel to go up whilst maintaining the water level constant both in the jacket and in the two dump tanks (see Figure 1).

Figure 1. Schematic layout of the VENUS critical facility



Reactor shutdown is induced by emptying the vessel. This can be done by opening the “safety valves” and allowing the air to escape from the jacket and the dump tanks; water is thereby transferred very rapidly into those two places.

The VENUS reactor is characterised by a high experimental flexibility, featured by:

- Direct access and manual handling of individual fuel pins.
- Easy loading of reactor.
- Special (dismountable) pins and removable grid parts (central 17 × 17 region).
- Easy water density simulation (down to 50% of density at room temperature).
- Reactor control through water level variation or absorber rod.
- Boron poisoning (up to 2 000 ppm).
- Special nuclear measuring devices compatible with the grids (e.g. fission chambers).

Moreover, the reactor allows for experiments to be carried out with a high accuracy due to:

- Very well characterised fuel composition, fuel geometry and reactor materials.
- Strict tolerance for fuel pin geometry.
- Fuel pins with the same dimensional characteristics as LWR fuel.
- Strict tolerances of the reactor grid geometry (0.05 mm on pitch).
- No perturbation of the investigated configuration.
- No local power perturbation due to fuel pin scanning.
- No local perturbation of the fine structure due to the use of dismountable fuel pins.

Table 1 shows some characteristics of the different fuel types available at the VENUS facility.

Main experimental parameters measured at VENUS

The most important parameters that are measured at the VENUS reactor are:

- The critical water level $h_{c.}$
- The reactivity coefficient $\delta\rho/\delta h$.
- The axial and horizontal fission rate distribution.
- Spectrum indices F5/F9, F8/F9, C8/F9.
- Kinetic parameters: mean neutron generation time and effective delayed neutron fraction.

Table 1. Characteristics of fuel rods available at the VENUS critical facility

Fuel type	^{235}U/Pu (w/o)	Isotopic composition Pu (w/o) 238/239/240/241/242	Fabrication process	Number of rods
Uranium	3.3/0		Pelleted	636
Uranium	4/0		Pelleted	865
L-MOX A	0.6/4.8	1/63/24/8/4	MIMAS	75
L-MOX B	0.3/5.4	1/63/24/8/4	MIMAS	66
M-MOX	0.3/9.7	1/61/24/9/5	MIMAS	215
H-MOX	0.4/14.3	1/62/24/9/4	MIMAS	229
MOX (50 cm)	3/1	0/92/7/1/0	Vibrated	408
MOX (50 cm)	2/2.7	0/79/17/3/1	Vibrated	470
MOX (50 cm)	1.7/3	0/79/17/3/1	Pelleted	65
MOX (50 cm)	0.7/4.3	0/96/4/0/0	Pelleted	25
MOX (50 cm)	0.7/5.1	0/79/17/3/1	Pelleted	62
U/gadolinium	3.5/7		Pelleted	60
AgInCd			Rodlets	20
B_4C			Pelleted	25
Pyrex			Rodlets	25

The critical water level

The critical water level is measured so as to determine the critical mass of the loaded configuration. Recent configurations had a critical height of about 60-80 cm. This height was chosen for the reason that the intermediate grid of the reactor would not interact with the neutrons (the upper water level had to be at least one extrapolation length of about 7 cm below the grid), while on the other hand the level should not be too low for reasons of precision of the measurements.

The random uncertainty of the value of the critical level is 0.02 cm, and the systematic uncertainty is about 0.07 cm. The random uncertainty is due to the reading of the water level and the temperature correction. The systematic uncertainty is mainly due to the determination of the lower level of the active part of the MOX and Gd fuel. The comparison of the same or similar configurations will only be affected by the random uncertainty. This is in particular important when reactivity effects of small changes of the configuration are measured.

With the help of the critical level measurements, changes in reactivity can also be measured. These changes can be due to:

- Am effect, by reloading previously measured MOX configurations.
- Changes in moderator density, by (un)loading aluminium water density reduction rods.
- Replacements of absorber rods (Gd, B_4C, AgInCd).
- Boron worth, by solving boron acid in water moderator.
- Overmoderation/water holes effect by unloading MOX rods from MOX assembly and/or loading Al rods.
- Void effect by introducing a void box (watertight Plexiglas or Al box).

Reactivity effect of the water level

The reactivity coefficient $\delta\rho/\delta h$ is measured to establish a link between the uncertainty of the critical level measurement and the uncertainty of the reactivity. Normally a value for $\delta\rho/\delta h$ is measured at about 0.15 to 0.08%/cm for a core height of 60-80 cm. Given the random uncertainty of the critical water level of 0.02 cm, the uncertainty of the reactivity is about 0.003% or 3 pcm. In case the systematic uncertainty plays a role as well, the uncertainty of the reactivity is about 11 pcm.

The reactivity coefficient is determined by the measurement of positive periods, created by making the reactor supercritical with an increment of the water level. The reactivity effect is calculated on the basis of measured periods T along with some calculated parameters like the delayed neutron fraction, the prompt neutron lifetime, the relative fraction of each delayed neutron group and the corresponding decay constant.

The uncertainty on the value of the reactivity coefficient due to the experimental uncertainties (period, water level increment) is estimated to be between 3-5%.

Axial and horizontal fission rate distribution

The axial and horizontal fission rate distribution can be used directly for validation purposes by comparing the measured and calculated fission rate distribution. Moreover, the axial fission rate distribution is used to determine the axial buckling by fitting the results with a cosine function. In this way a measure of the axial leakage is obtained, which is used for validation of two-dimensional codes. The axial buckling is measured with an uncertainty between 2-4%. For the fission rate distribution the random uncertainty for one pin is estimated to be 1% for UO_2 rods and 1.5% for MOX rods.

Both the axial and horizontal fission rate distribution are measured by gamma-scanning. The rods are irradiated during a sufficiently long period to permit the measurement of the fission product ^{140}La by a gamma-scanning installation using two similar NaI crystals. Between the fuel rod and the NaI crystal a collimator is placed with a variable slit width. Normally the slit width is 1 cm. The rods can be positioned in front of the collimator with an accuracy of much less than 0.1 mm.

The axial fission rate distribution is determined by measuring 30-40 positions on one fuel rod. Between every 3 to 5 points a so-called monitor is measured, which consists of a few irradiated pellets. These monitor measurements are used for a check of the decay correction applied to the measurements. All measurements are related to the monitor activity.

The horizontal fission rate distribution is determined by measuring about 50-100 rods. On each rod five positions are measured around the midplane, together with a measurement of the monitor and the background. The measurement results are corrected for decay, axial fission rate distribution, gamma self-shielding and fission yield.

An alternative to using gamma-scanning is the use of fission chambers. Fission chambers are often used as a check of the gamma-scanning results of the axial fission rate distribution, but sometimes they are used for a stand-alone measurement. Fission chambers can be loaded either at the position of a fuel rod or diagonally in between two fuel rods. In both cases there is a perturbation of the fuel configuration due to the lack of one fuel rod or the introduction of the fission chamber; therefore the gamma-scanning method is preferred. The horizontal fission rate distribution can also be verified using fission chamber measurements.

Spectrum indices

In some validation cases pin-to-pin fission rate distributions do not provide sufficient information. In these cases more information is requested about the contribution to the fission (or another nuclear reaction) rate by the different isotopes. Therefore some spectrum indices can be measured. A spectrum index is the ratio of two different reaction rates. Spectrum indices that are measured at VENUS are F5/F9, F8/F9 and C8/F9. F and C stand for fission and capture reactions, respectively, while 5, 8 and 9 stand for ^{235}U, ^{238}U and ^{239}Pu. Random uncertainties on the measured values are between 2-2.5%.

A spectrum index is measured by placing two foils in a dismountable fuel rod at the same distance from midplane. The foils are chosen in function of the spectrum index that is measured, e.g. a ^{235}U and a ^{239}Pu foil for the F5/F9 spectrum index. After irradiation the foils are unloaded and measured by gamma spectrometry (Ge detector). Dependent on the reactions that are considered, the activities of fission products or activation products are measured and corrected for e.g. foil mass, decay, gamma self-shielding, fission yield, branching ratio, etc. Since both foils have been irradiated under the same conditions, no corrections for the position of the foils are necessary.

For some spectrum indices fission chambers can be an alternative to foil measurements by performing measurements with e.g. both ^{235}U and ^{239}Pu fission chambers.

Kinetic parameters

The kinetic parameters are measured with the RAPJA method, which combines two well-known techniques, the measurement of a Rossi-alpha distribution and a Prompt Jump Analysis. By performing a prompt jump, the reactivity (in dollars) can be derived and can then be used in the analysis of the Rossi-alpha distribution to yield β_{eff} and l. Since the RAPJA technique assumes the same subcritical level during the prompt jump and the measurement of the Rossi-alpha distribution, the Rossi-alpha distribution will be measured at the stationary subcritical level driven by the intrinsic source and the anti-reactivity that was introduced during the prompt jump.

The following methodology is therefore applied in the RAPJA technique:

- Stabilisation at stationary critical level.
- Performing a prompt jump by lowering the water level and measuring the prompt jump fraction.
- Stabilisation at subcritical level driven by the intrinsic driving source and the reactivity of the prompt jump.
- Measuring a Rossi-alpha distribution at this stationary subcritical level.

The RAPJA technique allows determining β_{eff} and l in an absolute way without the need for calculations.

Preliminary design calculations

A neutronic study is performed in order to propose realistic and interesting critical configurations involving low-moderated H-MOX rods. The four options, considered hereunder, rely upon two features: 1) no MOX rods fabrication work and 2) use of a central test region incorporating the low-moderated MOX rods, surrounded by a driver zone.

Selected options

Several options are considered according to the characteristics of the core (Table 2) one wants to achieve and to some adaptation work needed.

Table 2. Characteristics of the considered critical configurations

Parameter	Option 1	Option 2	Option 3	Option 4
Driver zone lattice	Hexagonal	Hexagonal	Square	Square
Driver zone pitch (cm)	1.275	1.275	1.26	1.26
Central test region lattice	Hexagonal	Hexagonal	Square	Hexagonal
Central test region pitch (cm)	1.275	1.275	1.26	1.095
Channel void box?	No	Yes	Yes	No
Micro-rods diameter (cm)	0.21	0.21	0.55	–
Micro-rods material	Aluminium	Plexiglas	Plexiglas	–
Moderation ratio Vm/Vf	1.016	0.260	0.447	0.577
H/Pu-fiss	25	8	14	18
# H-MOX rods	217	217	225	229
# M-MOX rods	180	180	216*	215
# U 4/0 rods	744	744	784	800
# U 3/0 rods	–	–	624	592
# low-moderated MOX rods	217	217	289	444**
% low-moderated rods	19	19	16	24

* M-MOX rods inventory exceeded by one unit for computation purpose.
** All H- and M-MOX rods low moderated.

Option 1

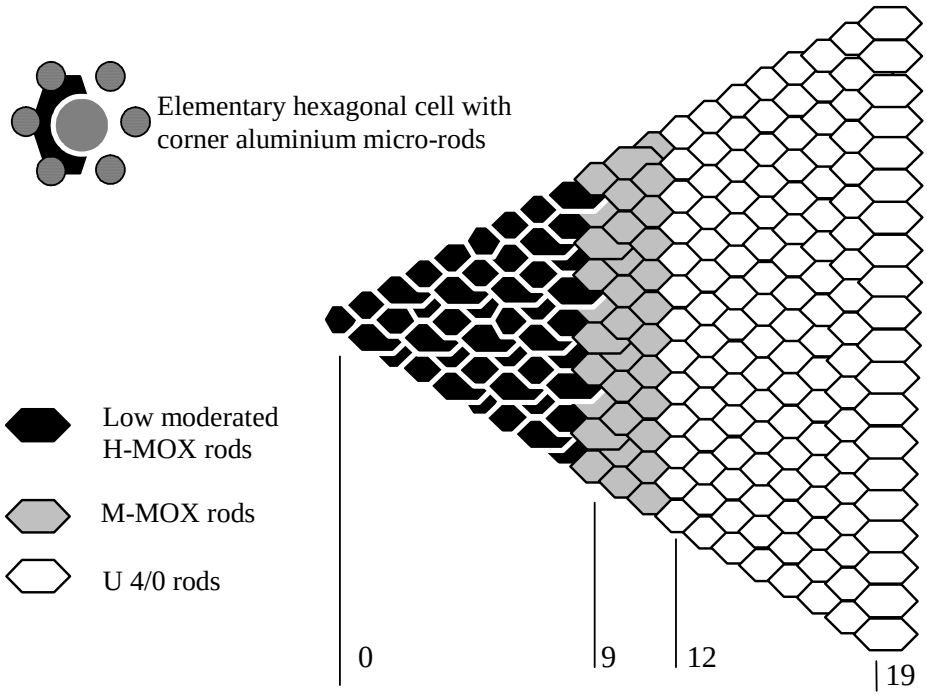
In the first option, one considers a full hexagonal lattice in the VENUS vessel. The pitch of the lattice is 1.275 cm so that a moderation ratio Vm/Vf = 1.276 is obtained with the VENUS MOX rods. This ratio can be reduced using aluminium micro-rods at the corner of each hexagonal cell, in the same way as is usually done for the square VENUS lattice and as is indicated in the zoom of Figure 2. With micro-rods of diameter = 0.21 cm, a moderation ratio of 1.016 could be attained, which is quite close to the highest ERASME moderation ratio [1].

The critical configuration could be as shown in Figure 2: a central low-moderated H-MOX region (eight rows), surrounded by M-MOX rods and then by U 4/0 rods. M-MOX and U 4/0 rods are preferred for the driver zone due to the requirement of spectrum hardening near the test region and because one tries to endow the central test zone with a significant reactive weight. The main fabrication work needed for such a scenario concerns the grids (hexagonal lattice) and the aluminium micro-rods.

Option 2

The second option also consists of a full hexagonal lattice in the VENUS vessel, however a central channel void box is used to completely remove the water moderator from row 0 to row 8. The moderation is then obtained using Plexiglas micro-rods 0.21 cm in diameter, such that a local moderation ratio Vm/Vf of 0.260 is achieved.

Figure 2. Schematic layout of core configuration 1 (1/6th of the core)



The fuel rods used in the test region and in the driver zone are considered here to be the same as in the previous option (see Figure 2 for the core layout). Such a scenario would imply the additional fabrication of a channel void box and Plexiglas micro-rods. The fabrication of a channel void box is an issue already addressed by SCK•CEN and BN in the framework of the VIPO experiments [2].

Option 3

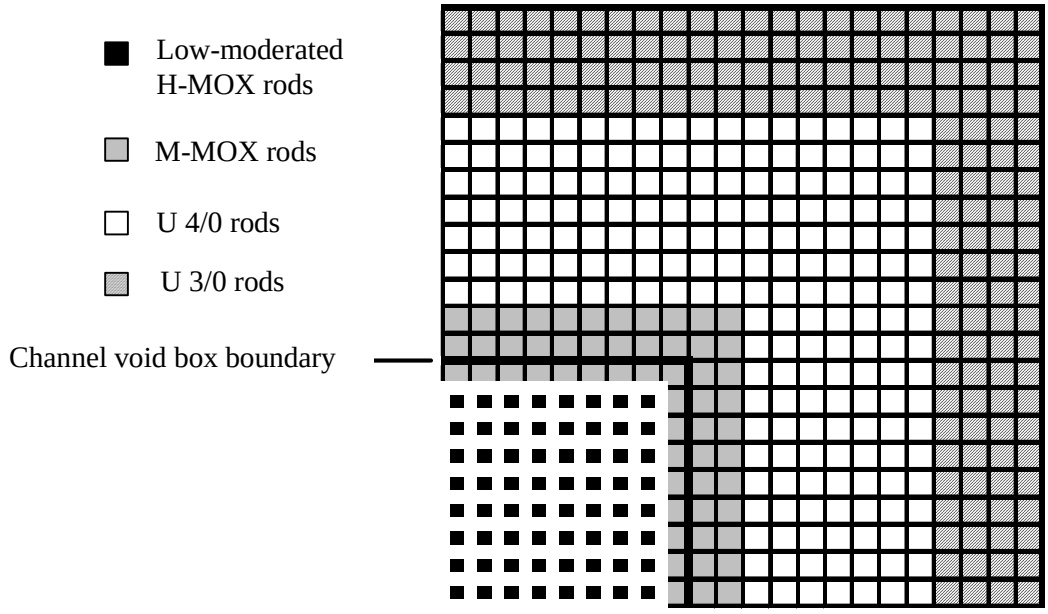
In the third option, one considers a full square lattice of PWR pitch = 1.26 cm, available at the VENUS facility. A channel void box is then used in the central 17 × 17 test region. As in the previous option, moderation is then controlled by inserting Plexiglas micro-rods in the corner of each elementary square cell. The Plexiglas micro-rod diameter considered here is 0.55 cm, giving Vm/Vf = 0.447, but a smaller diameter could be envisaged to address lower moderation.

The configuration should consist (Figure 3) of a central low-moderated region of dimensions 17 × containing 225 H-MOX rods (15 × 15) and an additional row of M-MOX rods. The driver zone is then made up of M-MOX, U 4/0 and finally some U 3/0 rods. The ordering of the rod types is still motivated by spectrum condition and reactive weight of the central test region. The external square shape of the driver zone is obviously not optimised with respect to the leakage but this first orientation calculation provides the main neutronic behaviour of the configuration.

Option 4

In the last option, the PWR square lattice grids of VENUS are still considered, but the central test region of dimensions 17 × 17 is filled with MOX rods in a triangular tight lattice. A pitch of 1.095 cm has been considered here, allowing to obtain a moderation ratio of Vm/Vf = 0.577 without using any

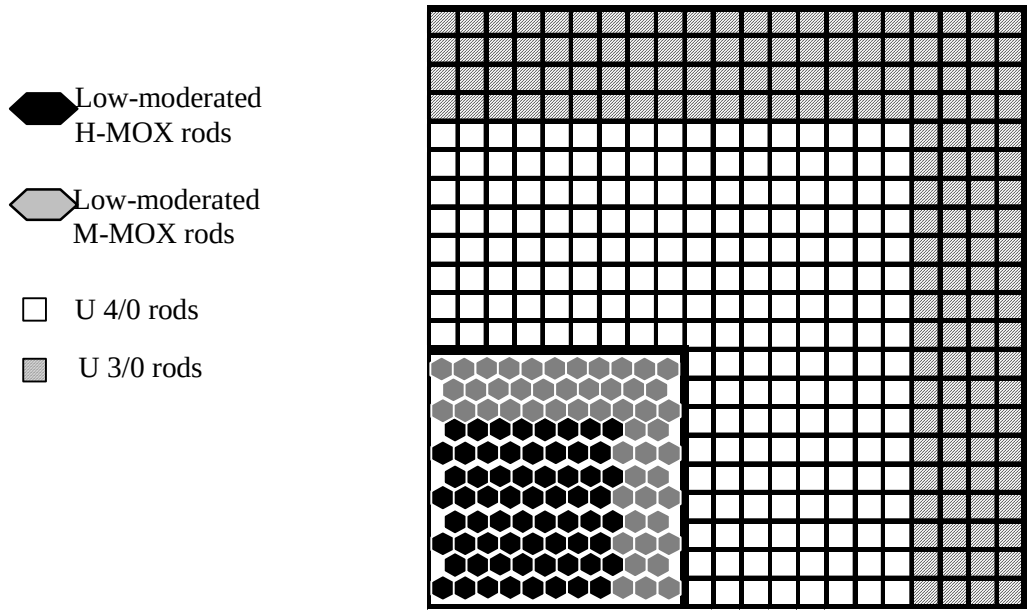
Figure 3. Schematic layout of core configuration 3 (1/4th of the core)



channel void box. The use of a very tight pitch implies that a higher number of MOX rods can fill the central region, giving the central test region a higher reactive weight. In fact, within the pitch 1.095 cm, the total amount of the H- and M-MOX rods can be set in the central test region and the only fabrication work concerns the test bundle.

The configuration defined here (Figure 4) is a central test bundle 17 × 17 filled by ~440 MOX rods (highest Pu content in the centre), surrounded by a driver zone made of U 4/0 and U 3/0 rods. Once again we did not optimise the external shape of the driver zone.

Figure 4. Schematic lay-out of core configuration 4 (1/4th of the core)



Results of the calculations

The calculations were performed with the WIMS8a multi-purpose neutronic code, as described in more detail in another paper of this workshop. These are 2-D calculations with an axial critical buckling option.

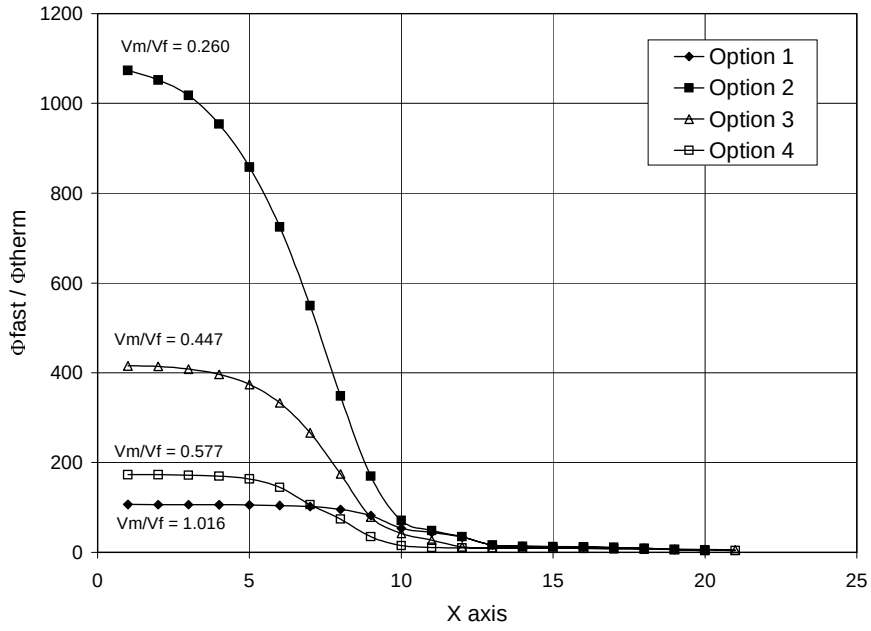
The main results are gathered in Table 3; they concern the critical water level (active length + extrapolation length) and the moderation conditions obtained in the central test region (fast, epithermal and thermal fluxes and the spectrum index Φ_{FAST}/Φ_{THERM}). The fission contribution of the test zone is also indicated, to give an insight of its reactive weight in the critical configuration.

Table 3. Main results with regard to the critical configurations considered

Parameter	Option 1	Option 2	Option 3	Option 4
$H_c + \lambda_{tot}$ (cm)	79	92	86	74
Test zone fission contribution (%) $\sum_f \Phi_{MOX}^{low\ mod} / \sum_f \Phi_{TOTAL}$	14	7.7	9.8	17
Central Φ_1 (> 100 KeV) (%)	57.5	60.2	58.6	56.8
Central Φ_2 (100 KeV ← 0.625 eV) (%)	41.6	39.8	41.2	42.6
Central Φ_3 (< 0.625 eV) (%)	0.9	0.1	0.2	0.6
Central Φ_{FAST}/Φ_{THERM} (cut-off 0.625 eV)	106	1 080	415	173

The results show that appropriate spectrum conditions [3] can be met and that spectrum hardening can be tuned according to the way the test bundle is designed, going from Φ_{FAST}/Φ_{THERM} = 100 to 1 000. The latter value is directly linked to the local moderation ratio Vm/Vf achieved by the use of a tight lattice, a channel void box and/or micro-rods, as demonstrated in Figure 5.

Figure 5. Radial distribution of the spectrum index, according to the various scenarios and the related local moderation ratio



All scenarios are realistic with respect to the active flooded height H_c and the relative weight of the test bundle is comprised between 8-17%. This reactivity weight is a function of the number of rods and the moderation ratio in the bundle.

Conclusions

This paper presents the VENUS facility as a flexible tool to address the issue of MOX powder criticality data through the use of low-moderated MOX pins. The parameters measured in the VENUS facility as well as candidate critical configurations are described.

The adaptation work needed to achieve the programme remains reasonable as all scenarios rely on the use of a central test bundle filled with MOX rods available at SCK•CEN.

Variant of the four scenarios studied can be considered, e.g. a triangular tight lattice in a full hexagonal lattice, variation of micro-rods diameter, measurement of a couple of critical configurations associated to two different moderation conditions, etc. As a result, it is possible to propose an experiment at low cost which could be performed within a short time.

REFERENCES

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[2] D'Hondt, P., *et al*., "Benchmark Experiments in VENUS: A Nuclear Data Package for LWR Pu-recycle", *PHYSOR'96*, Mito, Japan, 16-20 Sept. 1996.

[3] Rouyer, V., *et al*., "IRSN Projects for Critical Experiments – Low-moderated MOX Fuel Projects – And Others", *ICNC 2003*, Tokai-mura, Japan, 20-24 Oct. 2003.

KEOPS: A CRITICAL EXPERIMENT USING PuO_2 POWDER IN THE VENUS REACTOR

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Abstract

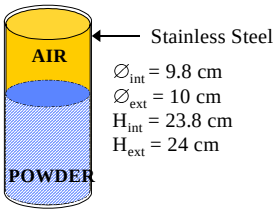
The safe handling of Pu powders is a major issue for MOX fabrication plants. Furthermore, the lack of experimental data, needed for code validation, imposes important safety margins for criticality prevention. The validation of criticality computer codes through the updating of the experimental databases on Pu-containing powders thus represents an important issue which should peak the interest of all organisations concerned by the handling of such powders. In this paper, we present a feasibility study of an experimental programme called KEOPS (Experimental Determination of *K-e*ffective *o*n Various *P*uO_2-containing *S*ystems) devoted to the criticality of PuO_2 powder. The project considers the direct loading of PuO_2 powder cans inside the critical facility VENUS, using a Plexiglas tube in the central test region of the core.

General data

The PuO_2 powder cans

The present section describes the type of plutonium powder cans this study was based on. Figure 1 provides a geometrical description of a typical plutonium powder can handled in a MOX fabrication plant.

Figure 1. Geometric description of a typical plutonium powder can handled in a MOX fabrication plant



For this feasibility study, a plutonium powder density of 2.2 g/cc was considered. This is a typical value used for criticality safety analysis. The plutonium isotopic vector is:

^{238}Pu	2 w/o
^{239}Pu	55 w/o
^{240}Pu	26 w/o
^{241}Pu	9 w/o
^{242}Pu	8 w/o

The typical powder humidity is assumed to be at 2.5 wt.% and the powder temperature 20°C. This temperature is obviously underestimated since plutonium systems are heated by their α emissions (especially ^{238}Pu). Considering a lower temperature would be conservative, however, and this assumption is usually made in criticality safety calculations.

Although the results presented in this paper are given for the above data, the influence of several parameters such as the powder density, humidity and temperature has been studied as well and is discussed in a sensitivity study hereunder.

The VENUS facility

A detailed description of the VENUS reactor is given in Ref. [2]; the focus here is thus only on the loading aspect of the can.

The purpose of introducing a powder can in the VENUS reactor is to achieve critical conditions by using a driver zone, and to perform critical measurements so as to determine the reactive weight of a plutonium powder can.

VENUS is a thermal reactor, which means that a core region with fast and epithermal spectrum conditions must be created in order to reproduce real can storage conditions near a can placed at the centre of the reactor. Various means considered for spectrum hardening in the VENUS central region included:

- The use of different types of driver zones.
- The insertion of a voided column 17 × 17 at the centre of the reactor.
- The insertion of additional rows of UO_2 or H-MOX rods at the periphery of the voided column.
- The insertion of a cadmium layer at the periphery of the voided column.

The driver zone

Two types of driver zones were selected for the present study, both accounting for the available rods at the VENUS facility. The first (Figure 2) comprises only UO_2 rods, and the second (Figure 3) includes two rows of M-MOX rods (medium-enriched MOX).

Figure 2. Driver zone comprising only UO_2 rods

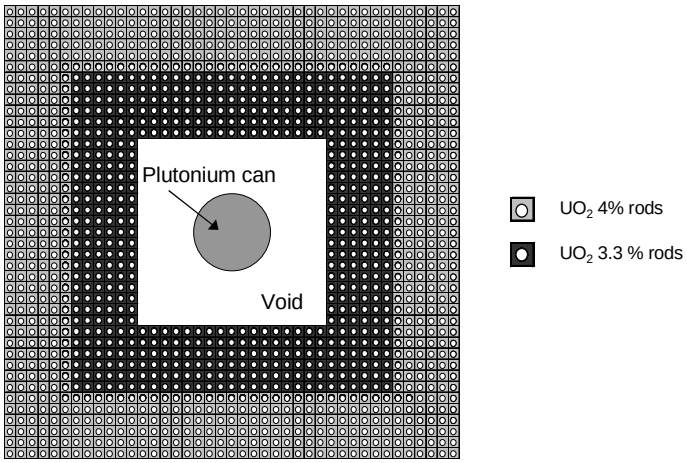
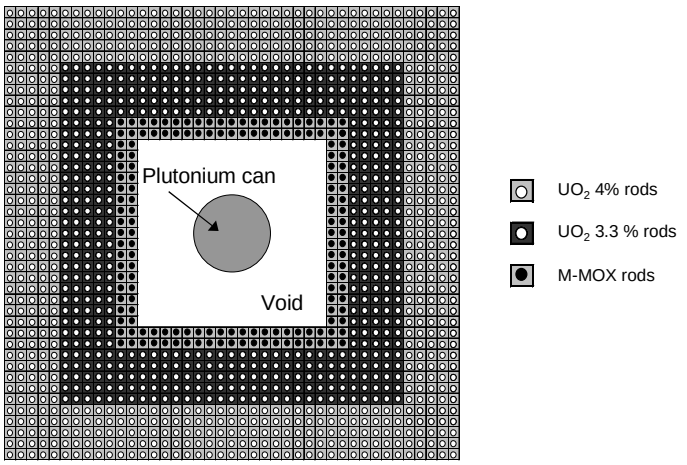


Figure 3. Driver zone comprising UO_2 and M-MOX rods



Insertion of additional rows of UO_2 or H-MOX rods or a cadmium layer

Figures 4 and 5 illustrate, respectively, the insertion of additional UO_2 or MOX rods and the insertion of a 2-mm thick cadmium layer at the internal periphery of the voided zone.

Figure 4. Insertion of additional UO_2 or MOX rods

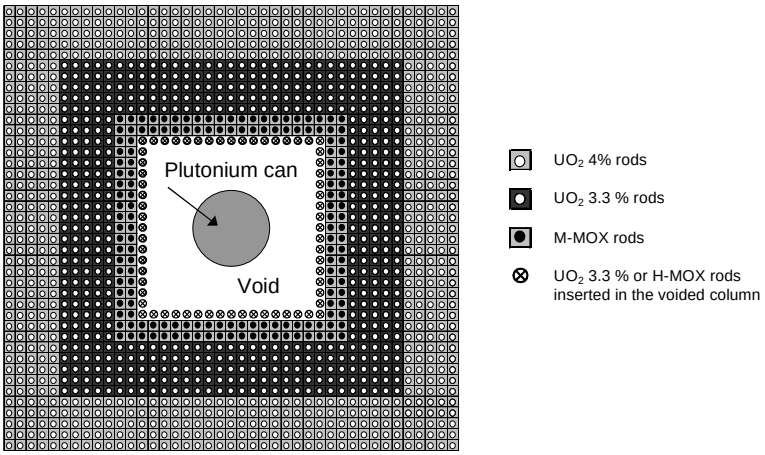
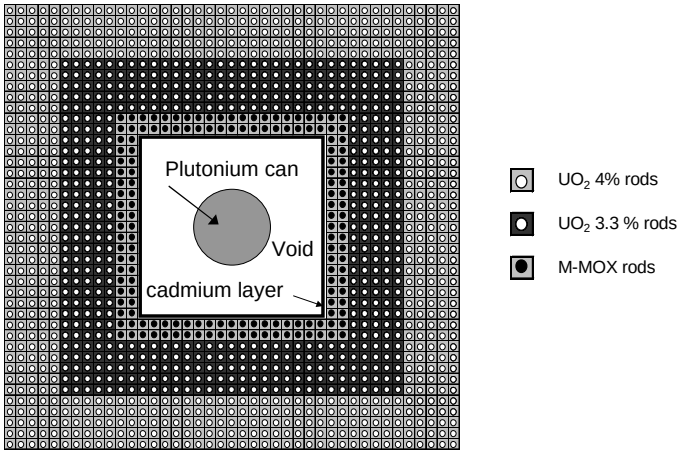


Figure 5. Insertion of a 2-mm thick cadmium layer at the internal periphery of the voided zone



Calculations

Calculation tools

All calculations have been performed using WIMS8a [3] modular codes fed with their own WIMS'97 library based on JEF-2.2. This cross-section library is composed of 172 energy groups that are condensed into seven energy groups prior to the main transport calculation. The energy boundaries of these seven groups are (eV): 820 850/111 090/9 118/4/0.625/0.14.

The initial approach consisted in performing three-dimensional diffusion calculations using the WSNAP module of WIMS. This method is not, however, appropriate for such heterogeneous systems. The results obtained with this diffusion model were often inconsistent and presented convergence difficulties.

A two-dimensional transport calculation method performed by the WCACTUS module of WIMS, which is reliable regarding the flux distributions and gives good estimations of Δk_{eff} values when comparing critical configurations, was thus the alternative method employed.

Main VENUS calculations results

Method description

The main idea of the VENUS calculations is to determine the Δk_{eff} between configurations with and without powder can at the centre of the core (all other parameters remaining identical) while analysing the spectrum conditions inside the powder can.

The two-dimensional model of the VENUS reactor is based on a square lattice with a pitch of 1.26 cm and a buckling corresponding to an 80 cm height. This means that the simulated powder can is also 80 cm high. The number of meshes and the shape of the powder can model were defined in order to have a symmetric model with a can surface as close as possible to the real can surface. Figures 6 and 7 illustrate the models with the UO_2 driver zone and with the MOX driver zone.

Figure 6. Illustration of the 2-D model of the VENUS reactor with the UO_2 driver zone

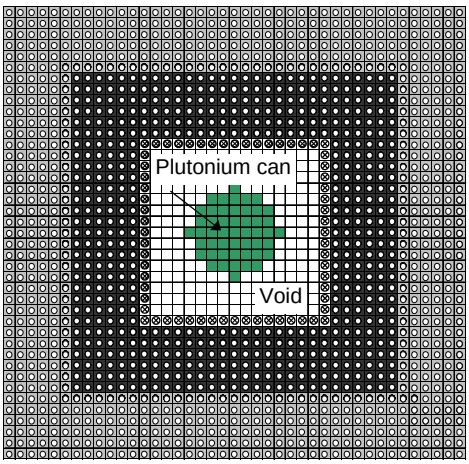
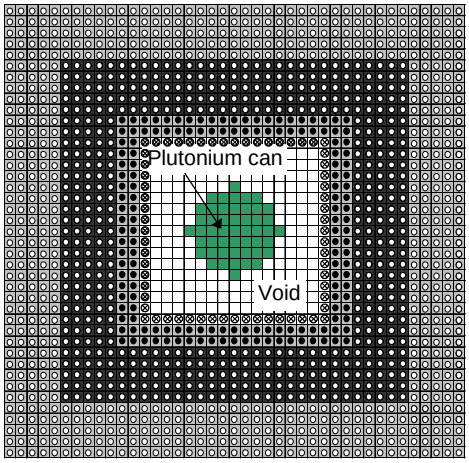


Figure 7. Illustration of the 2-D model of the VENUS reactor with the MOX driver zone



Numerous calculations were performed with the WIMS code, varying the following parameters:

- Type of driver zone.
- Number of additional rows (UO_2, MOX or cadmium) inside and at the periphery of the voided cluster.
- The humidity in the powder can.
- The temperature in the powder can.
- The density of the powder.

Results

Tables 1 and 2 show results for the particular case of a 2.2 g/cc powder density with 2.5% humidity and at 20°C inside the plutonium can for both UO_2 and MOX driver zones.

Figure 8 shows the ϕ_{FAST}/ϕ_{THERM} radial profile with and without can for the configuration with a MOX driver zone and two additional H-MOX rows at the periphery of the voided region.

Comments

With the different configurations considered here, ϕ_{FAST}/ϕ_{THERM} in a range of 5 to 500 can be obtained in the surrounding region of the powder can.

The insertion of UO_2, MOX or cadmium at the periphery of the voided column causes a thermal neutron fraction diminution corresponding to an important increase of ϕ_{FAST}/ϕ_{THERM}. One also observes also that the Δk_{eff} increases with ϕ_{FAST}/ϕ_{THERM} especially with the cadmium insertion.

The average spectrum conditions inside the can are not very sensitive to the external configuration. This is due to the fact that most of the neutrons coming from the driver zone are absorbed in the periphery of the powder can.

Sensitivity study

The reactivity weights of the powder density, temperature and moisture have been assessed. Some calculations were repeated, changing the powder temperature uniformly from 20°C to 350°C, which of course exaggerates the potential effect. The resulting reactivity effect does not exceed 10 pcm, which corresponds to an uncertainty of $\pm$ 0.3% Δk_{eff}.

For the powder moisture, variations between 0-5 wt.% have been computed and lead to reactivity effects between 80-200 pcm corresponding to a maximum uncertainty of $\pm$ 0.3% Δk_{eff}.

Finally, to deduce the reactivity weight of the powder density, variations from 2.2 g/cc to 3.5 g/cc have been simulated within the two-dimensional VENUS model by comparing three configurations (Figure 9 A, B and C) keeping the total PuO_2 mass constant.

Table 1. Main results with a UO_2 driver zone

With can – fluxes in PuO_2 powder			
	No rows	**1 row**	**2 rows**
ϕ_1/ϕ_{tot} (%)	67.45	70.45	71.21
ϕ_2/ϕ_{tot} (%)	32.05	29.20	28.44
ϕ_3/ϕ_{tot} (%)	0.50	0.35	0.35
ϕ_{FAST}/ϕ_{THERM} (%)	197	284	284
Without can – fluxes in central void			
	No rows	**1 row**	**2 rows**
ϕ_1/ϕ_{tot} (%)	44.70	49.80	53.19
ϕ_2/ϕ_{tot} (%)	38.98	39.96	39.39
ϕ_3/ϕ_{tot} (%)	16.32	10.24	7.42
ϕ_{FAST}/ϕ_{THERM} (%)	5.1	8.8	12.5
Δk_{eff} (with – without can) (pcm)	1 095	1 215	1 274

ϕ_1: ϕ > 111 090 eV.
ϕ_2: 0.625 eV < ϕ < 111 090 eV.
ϕ_3: ϕ < 0.625 eV.
ϕ_{FAST}/ϕ_{THERM}: Cut-off at 0.625 eV.

Table 2. Main results with a MOX driver zone

With can – fluxes in PuO_2 powder					
	No rows	**1 row**	**2 rows**	**3 rows**	**Layer Cd**
ϕ_1/ϕ_{tot} (%)	69.79	70.20	70.73	70.95	69.47
ϕ_2/ϕ_{tot} (%)	30.00	29.72	29.23	29.03	30.48
ϕ_3/ϕ_{tot} (%)	0.21	0.08	0.04	0.02	0.04
ϕ_{FAST}/ϕ_{THERM} (%)	464	1 324	2 796	5 347	2 226
Without can – fluxes in central void					
	No rows	**1 row**	**2 rows**	**3 rows**	**No rows**
ϕ_1/ϕ_{tot} (%)	52.31	58.00	60.68	62.56	56.41
ϕ_2/ϕ_{tot} (%)	42.51	40.79	38.84	37.24	43.11
ϕ_3/ϕ_{tot} (%)	5.18	1.20	0.48	0.20	0.49
ϕ_{FAST}/ϕ_{THERM} (%)	18.3	82.1	208.3	492.7	204.8
Δk_{eff} (with – without can) (pcm)	1 705	1 750	1 891	1 844	3 792

Figure 8. ϕ_{FAST}/ϕ_{THERM} with a MOX driver zone and two additional H-MOX rods rows inside and at the periphery of the voided region

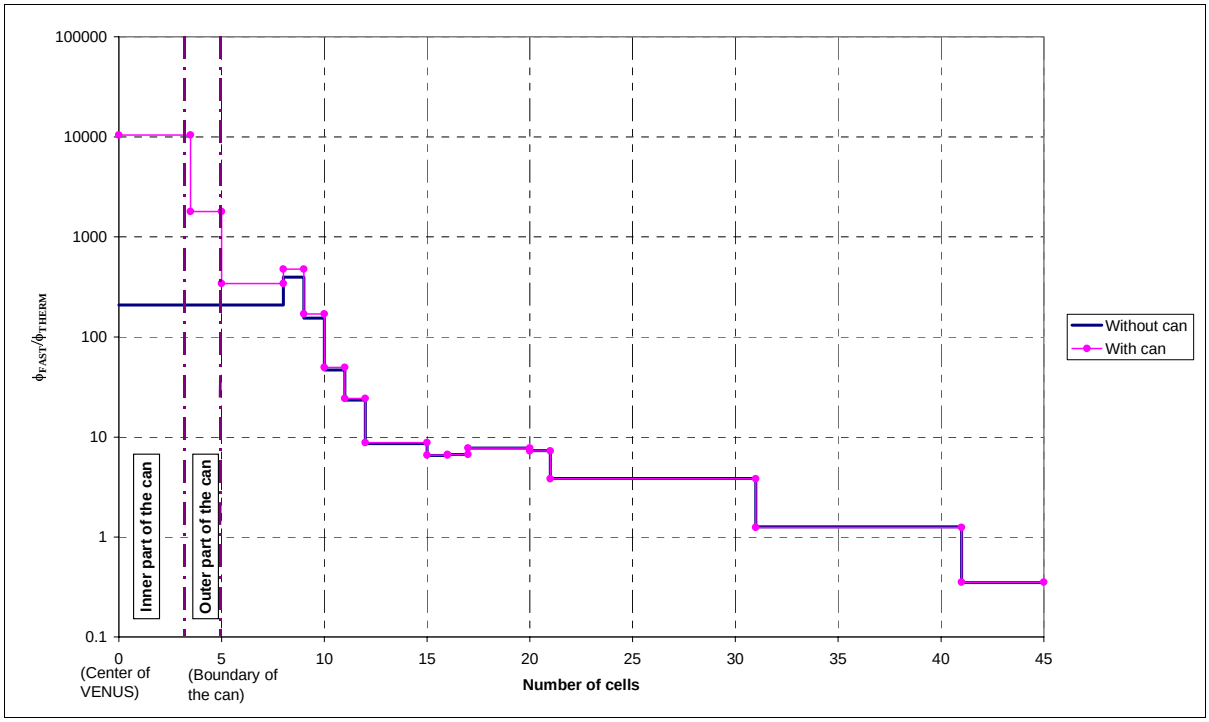
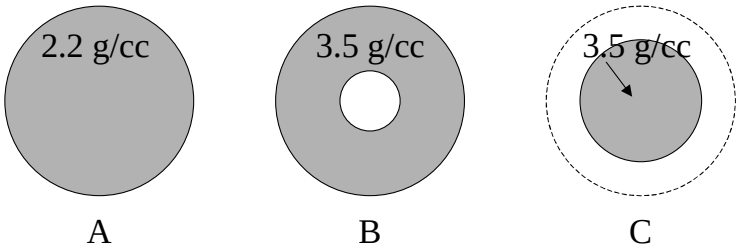


Figure 9. Can configurations studied for powder density reactivity weight



It appears that Δk_{eff} does not exceed 150 pcm, giving a maximum uncertainty of ± 4% Δk_{eff}.

The maximum total uncertainty estimated for powder temperature, moisture and density is:

$$\text{Total uncertainty} = \sqrt{(0.3\%\Delta k_{eff})^2 + (3\%\Delta k_{eff})^2 + (4\%\Delta k_{eff})^2} = 5\%\Delta k_{eff}$$

2-D to 3-D extrapolation

All VENUS calculations have been performed in a 2-D model with an axial buckling corresponding to an 80 cm height. As the real can height is 24 cm, a gross approximation is to divide the reactivity effects by a factor $\frac{80}{24} = 3.3$. The uncertainties are expressed here as percentages of Δk_{eff}, which means that their absolute values must also be divided by 3.3.

Example of criticality calculations

Method

In addition to the VENUS calculations, we also simulated an infinite lattice of 80-cm-high powder cans with a distance of 60 cm (border to border) between them. The humidity in the air surrounding the cans was then modified over a range of 0 g/cc (dry air) to 1 g/cc (water).

The WIMS code was used with a two-dimensional model (Figure 10), with reflective boundary conditions and with an infinite axial length. The results are given in Table 3.

Figure 10

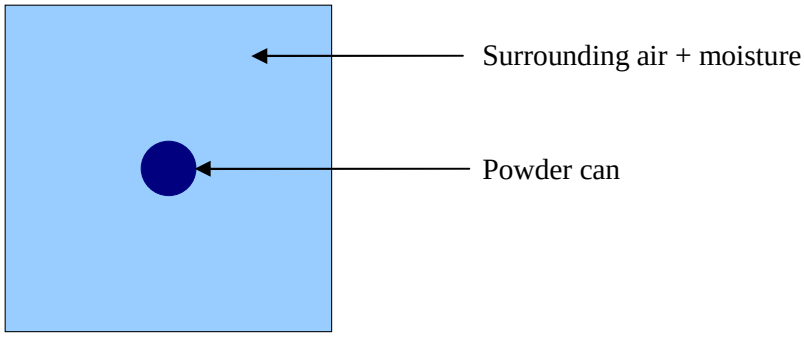


Table 3. Results for a lattice of powder cans of 80 cm height

Moisture density (g/cc)	0.0	0.05	0.07	0.1	0.15	0.2	0.4	0.6	0.8	1.0
Powder										
ϕ_1/ϕ_{tot} (%)	67.76	65.72	67.94	72.00	76.61	78.42	78.36	77.38	76.64	76.10
ϕ_2/ϕ_{tot} (%)	31.75	33.69	31.22	27.13	22.75	21.09	21.28	22.27	23.00	23.54
ϕ_3/ϕ_{tot} (%)	0.49	0.59	0.84	0.87	0.64	0.48	0.36	0.36	0.36	0.36
ϕ_{FAST}/ϕ_{THERM} (%)	203	167	118	114	154	206	277	280	279	276
Surrounding air + moisture										
ϕ_1/ϕ_{tot} (%)	62.47	63.41	65.69	69.86	74.74	76.72	76.84	75.91	75.20	74.68
ϕ_2/ϕ_{tot} (%)	36.61	35.54	32.84	28.62	24.12	22.41	22.52	23.46	24.16	24.68
ϕ_3/ϕ_{tot} (%)	0.92	1.05	1.47	1.52	1.14	0.87	0.64	0.63	0.63	0.64
ϕ_{FAST}/ϕ_{THERM} (%)	108	94	67	65	86	114	154	157	157	155
k_∞	0.15498	0.47362	0.56647	0.53609	0.42063	0.36785	0.38516	0.42938	0.46081	0.48497

Comments

From these scenario calculations, we verify that the conditions met in a real storage of cans and at least in a criticality study of such storage are not that far removed from the spectrum conditions simulated in VENUS. This tends to demonstrate that the proposed experiment is quite representative of operating or accidental conditions with real fissile media handled in a MOX fabrication plant.

Further options

Other possibilities involving fissile media commonly used in a MOX fabrication plant could be envisaged. A gross feasibility study confirms that we could, for instance, confine powders in standard claddings. The advantages of such a procedure would be:

- Exact characterisation of powder geometry (no more free top surface).
- Exact characterisation of powder density (use of X-radiography, gamma scans and fabrication of small segmented rodlets to avoid a too-high axial density gradient).
- No more temperature effect in the powder, since the radius of rodlet containing the powder is much smaller than the can radius.
- Same spectrum conditions in all test rodlets, also in account of the smaller size of the powder system (no more radial spectrum variation as is the case with a can).

Thus, fabrication of test rodlets at the BN MOX plant is considered feasible and opens a real new field of experiments. An extended programme could consider the loading of several kinds of test rodlets in the VENUS critical facility. Indeed, the test rodlets could be filled by various fissile media, according to the various steps encountered in a MOX fabrication process:

- PuO_2 powder rodlets, including (or not) scraps.
- MOX powder (primary blend) rodlets, including (or not) scraps.
- MOX powder (secondary blend) rodlets.
- Rodlets filled with scraps (average density ~ 5).
- Rodlets filled by "green" pellets (density ~ 6), i.e. before sintering.
- Rodlets partly sintered (density ~ 8), characterised by a density slightly less than sintered oxide (density ~ 10.5).

Conclusions

The KEOPS proposal takes advantage of the collaboration between SCK•CEN (owner of the VENUS critical facility) and Belgonucléaire (operating the nearby MOX manufacturing plant). The main idea is to load PuO_2 powder cans inside the critical facility or even to manufacture "powder rodlets" so as to increase the representativity of several parameters encountered in the MOX fabrication process.

The project has been studied neutronically and it has been shown how to design the part of the driver zone nearby the test core region, in order to assess the reactivity weight of the cans according to the spectrum conditions. The results show that the reactivity effect induced by one can (as compared to a situation without any can) should be around ~400 to ~750 pcm according to the spectrum tuning. But increasing step by step the number of cans inside VENUS could lead to a total reactivity effect of 1 800-3 500 pcm.

Uncertainties arising from the moisture in the powder, the density and the temperature distribution have been investigated. Doppler effect in the powder can be considered negligible, whereas an uncertainty of the moisture content (from 0-5 wt.%) and of the density can have a cumulated impact of 5% Δk_{eff}. Such a value is however overestimated:

- The uncertainty related to the powder density can easily be reduced by weighting and measuring the top powder surface.

- Methods exist to measure the moisture of the powder that can be implemented in the frame of this project.

As a result, one can estimate that an overall uncertainty of 2.5% Δk_{eff} could be achieved.

With regard to such uncertainties, it can be argued that calculation benchmarking against the experiment could be performed from the point of view of safety, for example search of the bounding moisture and density conditions that will bring the criticality calculation in the conservative side.

In any case, the handling and loading of PuO_2 cans inside VENUS remains representative of a daily process in a MOX fabrication plant, all the more in that the spectrum conditions offered by the core central test region can be tuned in a broad range, so that this integral experiment should bring an added value to criticality databases.

At the present time, the so-called KEOPS experiment could be performed using four available PuO_2 cans with slightly different compositions as compared to the cans considered in the neutronic study:

Isotopic vector
^{238}Pu: 0.210 w/o ^{239}Pu: 77.503 w/o ^{240}Pu: 18.323 w/o ^{241}Pu: 3.255 w/o ^{242}Pu: 0.709 w/o
Powder density
Apparent density ~ 2.2 g/cc Packed density ~ 3 g/cc

The definition and the size of a final scope will depend on the interest and on the support of the organisations concerned by the handling of plutonium powders. Such scope would have to consider the various critical configurations one wishes to investigate: 1) tuning of the spectrum inside the central test region, 2) step-by-step increase of the number of cans in the VENUS vessel, or other aspects such as the “powder rodlets” option.

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MIXED-OXIDE EXPERIMENTS AT THE LOS ALAMOS CRITICAL EXPERIMENTS FACILITY: A PROPOSAL

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Abstract

Disposal of excess weapons plutonium by means of producing mixed-oxide (MOX) fuel for use in commercial reactors necessitates integral critical experiments to support safe fuel production process design. The Los Alamos Critical Experiments Facility (LACEF) proposes a programme of mixed-oxide experiments using the uranyl fluoride solution assembly known as SHEBA.

Proposed experiment

The Los Alamos Critical Experiments Facility (LACEF) proposes to conduct integral measurements using the low-enriched uranyl fluoride solution assembly SHEBA. The experiments consists of determining the worth of various mixed-oxide (MOX) pin configurations in the experiment well of SHEBA.

The proposed experiments employ low-polyethylene-moderated MOX pins in various pitched lattices to achieve varying moderation ratios. These experiments may be shown to meet the requirements of the actual conditions requested.

Similar experiments have been performed using other fissile material samples. Critical conditions are established with the "sample" in and out of the experiment well. The difference in critical fuel height corresponds to a known reactivity worth. The configurations can then be modelled in computer codes for validation purposes.

Preliminary design

The experiment has been designed to provide pertinent data using existing materials and equipment. The design can easily incorporate future material types. The schematics of the design are shown in Figures 1-3.

Figure 1. Cross-section of SHEBA with 21-pin MOX bundle in experiment well

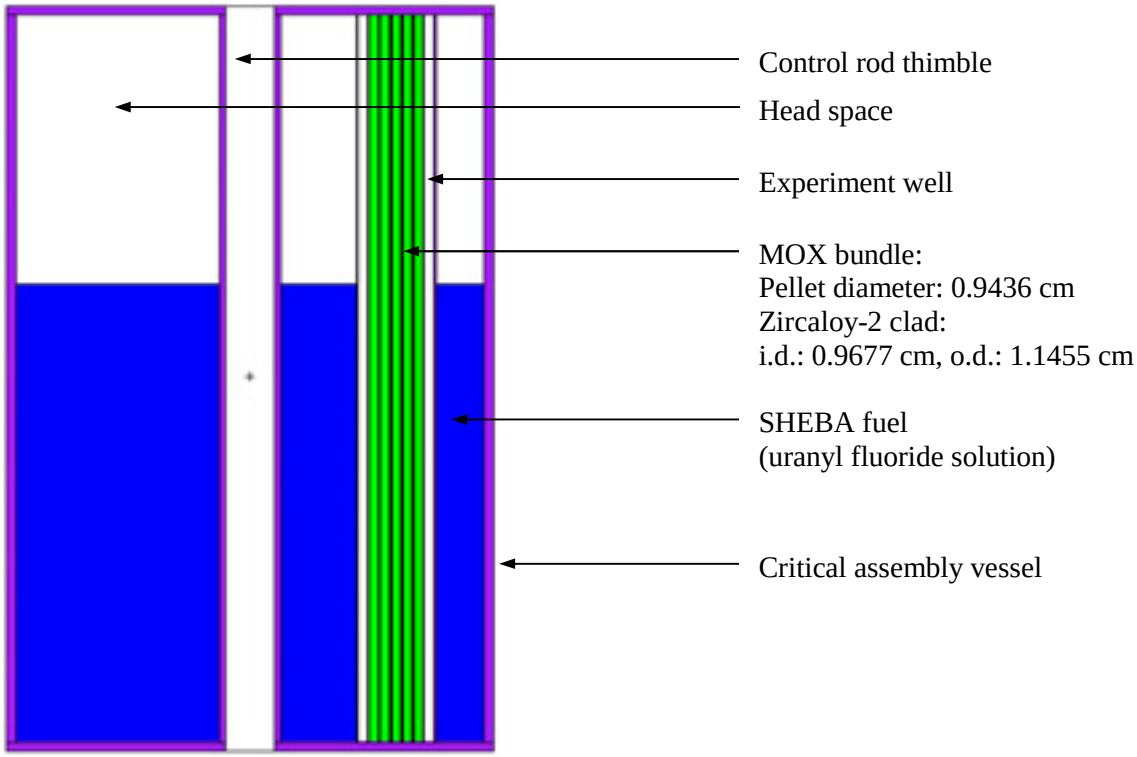


Figure 2. Top view of SHEBA with 21-pin MOX bundle in experiment well

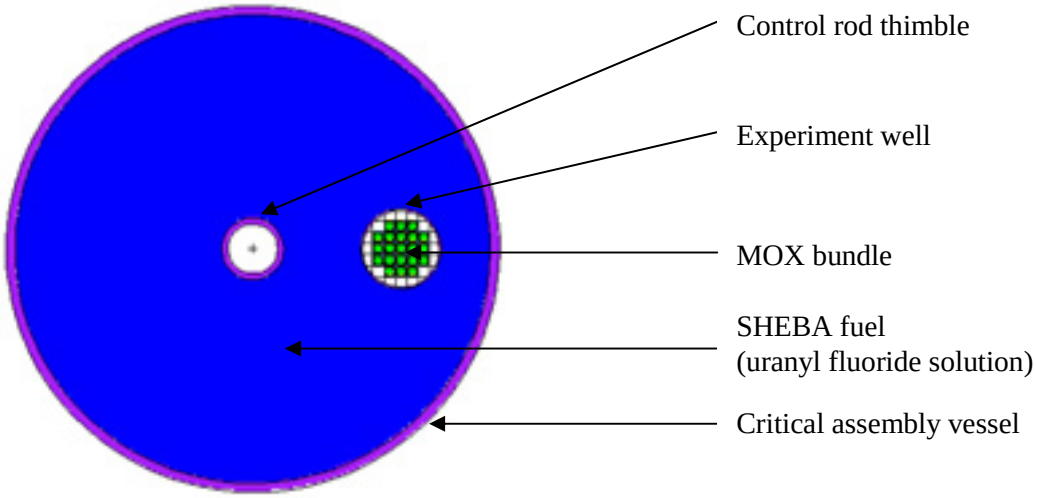
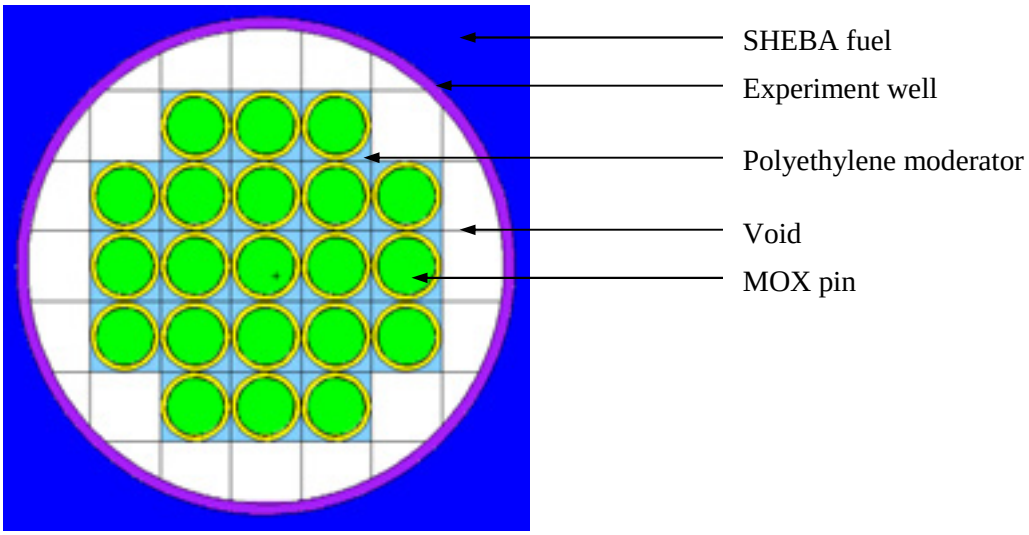


Figure 3. Top view of experiment well with 21-pin MOX bundle



Available material

LACEF has 530 MOX fuel pins available for use in critical experiments. Nominally, the fuel consists of 4.4 wt.% plutonium combined with natural uranium. The fuel was separated in 1975, resulting in some ^{241}Am in-growth. Characteristics of the pins are provided in Tables 1-3.

Table 1. MOX fuel pin physical characteristics

Active length	177.8 cm
Pellet diameter	0.9436 cm
Zircaloy-2 clad inner diameter	0.9677 cm
Cladding outer diameter	1.1455 cm

Table 2. Plutonium isotopics

	Isotopic Pu/total Pu (%)		
	Type 1	**Type 2**	**Type 3**
^{238}Pu	0.84	0.85	0.1
^{239}Pu	77.20	78.79	78.89
^{240}Pu	17.82	16.46	19.65
^{241}Pu	2.06	2.05	0.79
^{242}Pu	2.08	1.85	0.57

Table 3. Isotopic content (wt.%) of LACEF MOX fuel pins

	^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Total Pu	^{241}Am
Type 1	0.008	0.596	0.002	83.026	0.037	3.367	0.777	0.090	0.091	4.361	0.191
Type 2	0.008	0.597	0.002	83.076	0.037	3.424	0.715	0.089	0.080	4.346	0.190
Type 3	0.001	0.596	0.002	82.926	0.004	3.653	0.910	0.037	0.026	4.630	0.078

Table 4. Proposed experiment and MOX powder parameters

	Proposed experiments	**Reactor-grade MOX**	**Weapons-grade MOX**
PuO_2 content	4.4-4.6%	5.0-30.0%	5.0-22.0%
Lattice pitch	1.17 (square)	–	–
Water content	2%	1-5%	1-5%
H/Pu	2.4	1.1-12.7	1.4-25
$H/Pu_{fissile}$	3.1	1.3-15.5	1.5-26

Spectral characteristics and neutron balances

Calculations were performed with MCNP™, a general Monte Carlo n-particle transport code, to obtain parameters for comparison with the desired data. All calculations were performed with Type 1 pins. Neutron fluxes in the MOX bundle are shown in Figures 4 and 5 without a cadmium liner and with a cadmium liner, respectively. The data was tallied using 100 equal lethargy bins. Corresponding five-group fluxes and neutron balances are provided in Table 5.

As indicated by the data here, the SHEBA experiment compares favourably with a full lattice experiment. Based on the sensitivity discussion in Ref. [1], the SHEBA experiment should also compare favourably to configurations of low-moderated MOX powders. Five-group detailed balances by isotope are provided in Tables 6 and 7 for both the unlined and cadmium-lined experiment well cases.

Conclusion

Based on the discussion, experiments pertinent to data needs with “low-moderated mixed-oxide fissile materials” may be conducted in the near term with existing MOX and the SHEBA assembly. Future MOX mixtures may be utilised in the same manner, requiring substantially less material than a full lattice experiment. LACEF proposes that these experiments be conducted as soon as possible to support the design of full-scale critical experiments using the actual MOX constituents.

Figure 4. Average neutron energy spectrum in the MOX pin bundle without cadmium liner

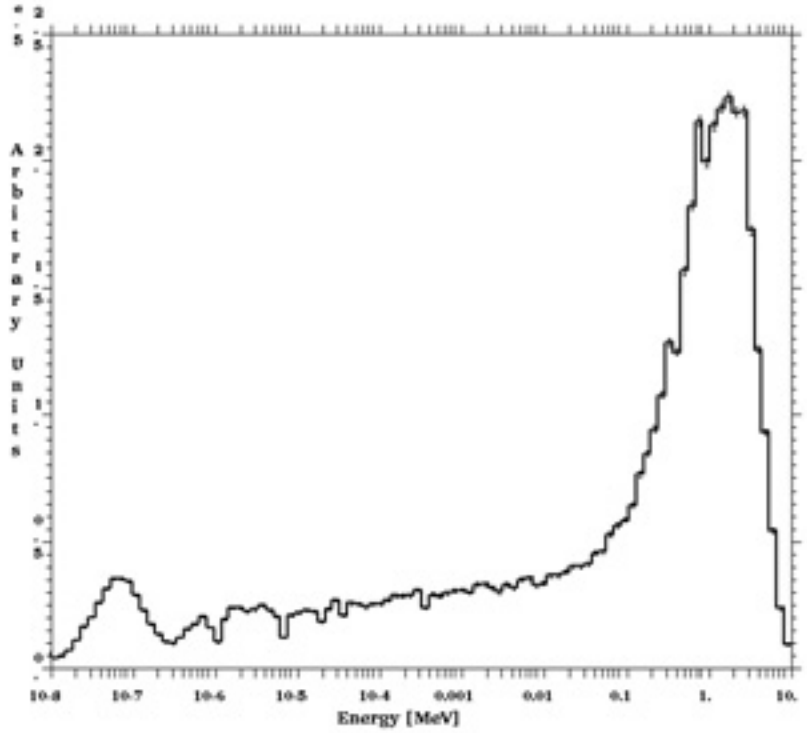


Figure 5. Average neutron energy spectrum in the MOX pin bundle with cadmium liner

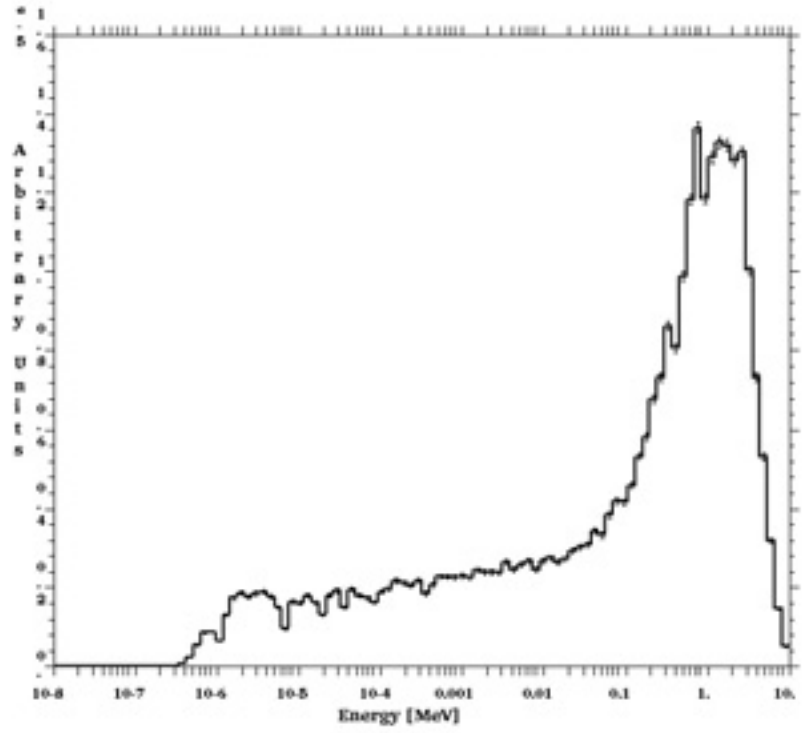


Table 5. Five-group neutron flux and balances

	Average neutron flux in MOX pins, %				
	< 0.1 eV	**0.1 eV-10 eV**	**10 eV-10 keV**	**10 keV-100 keV**	**> 100 keV**
No liner	4.7	8.1	18.4	9.6	59.1
Cd liner	0.0	6.4	22.3	11.5	59.7
	Average fissions in MOX pins, %				
	< 0.1 eV	**0.1 eV-10 eV**	**10 eV-10 keV**	**10 keV-100 keV**	**> 100 keV**
No liner	50.0	36.7	6.3	0.3	6.7
Cd liner	0.7	23.7	39.6	1.8	34.2
	Average captures in MOX pins, %				
	< 0.1 eV	**0.1 eV-10 eV**	**10 eV-10 keV**	**10 keV-100 keV**	**> 100 keV**
No liner	19.2	50.1	28.1	1.0	1.5
Cd liner	0.1	53.0	42.8	1.8	2.3

Table 6. Fissions by isotope

Average fissions per isotope per group, %						
Isotope	**Liner**	**< 0.1 eV**	**0.1 eV-10 eV**	**10 eV-10 keV**	**10 keV-100 keV**	**> 100 keV**
^{234}U	None	0.0	0.0	0.0	0.0	0.0
	Cd	0.0	0.0	0.0	0.0	0.0
^{235}U	None	11.0	4.7	14.2	17.6	2.8
	Cd	10.6	13.2	14.6	17.6	2.9
^{236}U	None	0.0	0.0	0.0	0.0	0.0
	Cd	0.0	0.0	0.0	0.0	0.0
^{238}U	None	0.0	0.0	0.0	0.2	69.6
	Cd	0.0	0.0	0.0	0.2	68.7
^{238}Pu	None	0.0	0.0	0.1	0.3	0.2
	Cd	0.0	0.0	0.1	0.3	0.3
^{239}Pu	None	86.0	92.8	82.2	77.5	22.4
	Cd	86.3	80.2	81.7	77.5	23.3
^{240}Pu	None	0.0	0.0	0.1	1.0	3.2
	Cd	0.0	0.1	0.1	1.0	3.2
^{241}Pu	None	3.0	2.4	3.4	3.3	0.5
	Cd	3.0	6.2	3.4	3.3	0.6
^{242}Pu	None	0.0	0.0	0.0	0.0	0.3
	Cd	0.0	0.0	0.0	0.0	0.3
^{241}Am	None	0.0	0.0	0.0	0.0	0.8
	Cd	0.0	0.2	0.0	0.0	0.8

Table 7. Captures by isotope

Average captures per isotope per group, %						
Isotope	**Liner**	**< 0.1 eV**	**0.1 eV-10 eV**	**10 eV-10 keV**	**10 keV-100 keV**	**> 100 keV**
^{16}O	None	0.0	0.0	0.0	0.0	16.4
	Cd	0.0	0.0	0.0	0.0	15.3
^{234}U	None	0.0	0.1	0.0	0.0	0.0
	Cd	0.0	0.1	0.0	0.0	0.0
^{235}U	None	3.3	0.6	1.2	1.1	0.9
	Cd	3.2	0.5	1.4	1.1	0.9
^{236}U	None	0.0	0.0	0.0	0.0	0.0
	Cd	0.0	0.0	0.0	0.0	0.0
^{238}U	None	13.0	28.1	86.4	91.2	78.6
	Cd	12.6	42.5	84.6	91.2	79.4
^{238}Pu	None	1.1	0.1	0.1	0.1	0.1
	Cd	1.0	0.0	0.1	0.1	0.1
^{239}Pu	None	61.1	28.4	9.2	4.8	2.4
	Cd	62.4	3.3	10.3	4.8	2.6
^{240}Pu	None	13.0	37.1	2.0	1.4	0.9
	Cd	12.7	48.8	2.2	1.4	0.9
^{241}Pu	None	1.8	0.4	0.2	0.1	0.1
	Cd	1.7	0.2	0.2	0.1	0.1
^{242}Pu	None	0.1	1.1	0.1	0.1	0.1
	Cd	0.1	1.7	0.1	0.1	0.1
^{241}Am	None	6.6	4.2	0.9	1.1	0.6
	Cd	6.3	2.9	1.0	1.1	0.6

REFERENCE

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INTEGRAL NEEDS FOR MOX POWDERS: STATE OF THE ART AT CEA CADARACHE ON MOX FUEL EXPERIMENTS

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Abstract

Several experimental programmes have been conducted at CEA connected with the use of MOX in commercial PWR or innovative concepts, such as high conversion light water reactors (HCLWRs) and at various moderation ratios. Measurements of integral parameters of interest were performed using fission chambers and gamma-scanning techniques, or by oscillation techniques. This paper presents an overview of the experimental programmes performed at the EOLE and MINERVE facilities located at CEA Cadarache.

Introduction

For about thirty years, the CEA has been conducting neutronic experiments in the MOX fuel field at two complementary facilities:

- EOLE for the analysis of the main neutronic parameters of regular or mock-up lattices in thermal and epithermal spectra.

- MINERVE for the basic study of integral cross-sections by oscillation technique in any kind of spectrum, from dissolver to fast ones.

Several programmes have been conducted at both facilities, connected to the use of MOX in commercial PWR or innovative concepts, such as high conversion light water reactors (HCLWRs) and in various moderation ratios[1], measuring integral parameters of interest using fission chambers and γ-scanning techniques, or by oscillations techniques.

The CEA experience with MOX integral experiments began during the 70s in the framework of a EURATOM CEA/EDF/EEC programme on Pu recycling in PWRs [1]. It was followed in 1984 with the ERASME programme on EOLE devoted to high-conversion LWRs (under-moderated MOX lattices) [2], together with the MORGANE oscillation programme in MINERVE [3] and ICARE irradiation in MELUSINE [4] (experimental reactor at Grenoble's CEA Research Centre, now closed and dismantled). These HCLWR studies were followed in 1989 by the EPICURE 30%MOX recycling programme in standard PWR [5,6]. Then in 1996, the MISTRAL2 programme devoted to 100%MOX recycling in standard PWR [7] and the MISTRAL4 experiment which is a mock-up of a RMA-MOX reactor (over-moderated PWR 100%MOX) [8] were undertaken. Finally, the BASALA programme during the 2000-2003 period aimed at studying 100%MOX behaviour in ABWR-type assemblies [9].

The validation of those programmes by CEA using both deterministic and stochastic models, especially concerning key parameters associated with the safety of the fuels used, showed that most of the target accuracies in term of criticality, power distribution, control rod worth, kinetics parameters and reactivity coefficients were met in any kind of spectrum.

The main concern of the present topic, i.e. the behaviour of low-moderated MOX fuels, should be partially covered by some integral results from several CEA programmes, mainly the ERASME programme.

In the following paragraphs, we present the EOLE and MINERVE facilities. An overview of each experimental programme is given, extracting the key information susceptible to cover – at least partially – the present area, and what could be covered by additional experiments.

The CEA Cadarache facilities

The EOLE facility

The EOLE facility is a critical facility operating up to a maximum power of 100 W. It is dedicated to the neutronic study of moderated lattices like light water reactors. EOLE is an easily adaptable facility composed of a concrete structure offering biological shielding for flux levels up to 10^9 n.cm^{-2}.s^{-1},

[1] For Pu contents ranging from 3-11% (including 4.3%, 7% and 8.7%) for standard 33 GWd/T first recycling UOX assemblies.

a cylindrical vessel in AG3 with an over-structure in stainless steel, able to contain various types of core structures, four control rods and a pilot rod, and a water circuit coupled with a thermoregulation station in order to analyse the temperature coefficients of the lattices ranging from 5-90°C.

The EOLE's flexibility enables one to analyse the safety parameters of new types of fuels and/or core/assembly designs by using special structures (void boxes, mini-grids for local pitch modification) or by adding various absorbers (single or clusters), as natural or enriched B_4C, aluminium-indium-cadmium (AIC) or Hf. Critical and subcritical measurements are used for the reactivity effects, the reaction rates arising in the lattice being measured by both miniature fission chambers (for fission) and/or γ-scanning techniques (for fission and capture).

It is noteworthy to mention that this facility is not restricted by its Pu content (i.e. vector or amount) since the safety criteria of lattices included in the experimental programme agree with the main prescriptions of the facility's General Safety Report.

The MINERVE facility

The MINERVE facility is a pool-type reactor operating with a maximum power of 100 W. The peripheral driver zone is composed of material testing reactor (MTR) assemblies with fuel having enrichments of 90 or 93% in ^{235}U. These fuel assemblies, submerged under 3 meters of light water, surround a central square cavity containing a watertight cylinder. Within this receptacle, any kind of configuration representative of any reactor lattice (spectrum) can be inserted, from very hard spectrum lattices (fast reactors) to very soft ones (dissolvers). The size of the experimental lattice is large enough to avoid perturbations from the driver zone and to achieve fundamental mode in its centre. The reactivity worth of oscillation samples is measured through the response of a rotating pilot rod to the insertion of the sample. Additional reaction rate measurements in the experimental zone can be made by using the same methodology as for EOLE.

The experimental programmes

The ERASME programme

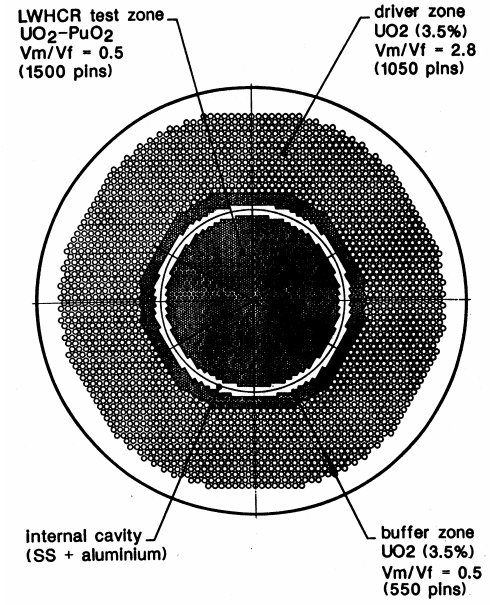
The ERASME programme was achieved in the EOLE facility, as part of a collaboration between CEA and the French electricity supplier EdF. The ERASME experiment was built up to reduce the uncertainties on fundamental lattice parameters associated to the use of mixed-oxide fuel in under-moderated spectrum. Three lattices were considered, with different moderation ratios: 0.5, 0.9 and 2.1, corresponding to the ERASME/S (serré = tight), ERASME/R (realistic) and ERASME/L (large) cores respectively.

The integral lattice parameters investigated in these cores were:

- The determination of the buckling through fission rate distributions.
- The conversion factor ($\sigma_c^{238}U/\sigma_f^{239}Pu$).
- The fission rates of the most relevant heavy isotopes (^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am) by using fission chambers.
- Absorber worth and cluster configurations (stainless steel, B_4C, Hf, AIC and Eu).
- Perturbations induced by adding heterogeneities in assembly (water holes, fertile UO_2, etc.).

An important emphasis was placed on the measurements of safety parameters relevant to this kind of lattice. In ERASME/S in particular, an extensive campaign was realised to water void in the central region. This was of primary importance, as this void coefficient can be positive in very tight plutonium lattices, and then limit strongly the under-moderated concept. The ERASME/S lattice was composed of a central triangular pitch of 11% MOX fuel pins of average volumetric moderation ratio of 0.5, surrounded by a 3.5% tight UO_2 buffer zone. The parameters measured and analysed covered were critical sizes, buckling, reactivity worths and spectral indices in the central region, to characterise this type of lattice from a neutronic point of view and validate French codes for HCLWR calculations. Figure 1 reproduces the radial cross-section of the ERASME/S lattice.

Figure 1. Radial cross-section of the ERASME/S core



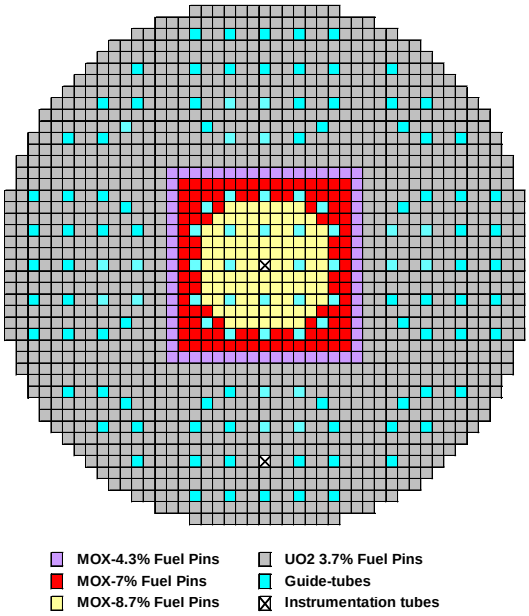
The EPICURE programme

EPICURE was conducted in the EOLE facility and supported the 30% MOX loading strategy built by the French electricity supplier EdF in five of its NPPs. The main objective of the EPICURE experimental programme was to evaluate the uncertainties of reactor physics calculations of plutonium recycling PWRs and to reduce these uncertainties significantly to reach the accuracy level obtained for traditional UO_2 fuel calculations. The EPICURE programme was optimised in order to make a representative mock-up of industrial plutonium recycling cores and to measure the associated safety parameters:

- Measurement of fundamental parameters in UO_2 and MOX.
- Reactivity worth and power distribution in presence of absorber clusters, water holes, local 2-D voids, of MOX 17 × 17 assembly with 7%MOX and one MOX 17 × 17 zoned assembly.
- Pin bowing measurement of UO_2 and MOX.
- Three-dimensional (3-D) void in UO_2 and MOX.

Special attention was brought to the study of power distribution measurements in a configuration typical of plutonium-recycling PWR cores, and in particular on the complex phenomena that occur at the interface between two neutronically very different media (neutron spectrum change between the MOX and UO_2 subassemblies and a strong flux gradient at the interface). Figure 2 reproduces the radial cross-section of the UM-ZONE configuration of the EPICURE programme. The measurement were realised between 1989 and the end of 1994, and was divided into three main phases. The EPICURE enabled the CEA teams and its partners to create a unique database and associated calculation tools for 30% MOX loading [6].

Figure 2. Radial cross-section of the EPICURE UM-ZONE configuration



The MISTRAL programme

MISTRAL followed the EPICURE programme in the EOLE facility. This programme was the first ever 100% MOX ALWR programme in over-moderated lattices. It was also the starting point of a French-Japanese collaboration in the reactor physics field. Four cores were investigated:

- MISTRAL-1 was an over-moderated UO_2 homogeneous core dedicated to reference measurements of various lattice fundamental parameters: critical masses, bucklings, local heterogeneities effects, reactivity worths of single absorbers, etc.

- MISTRAL-2 (Figure 3) and MISTRAL-3 were two 100% MOX homogeneous cores devoted to the analysis of the previous parameters behaviour in over-moderated lattices with two different lattice pitches (1.32 cm and 1.39 cm for MISTRAL-2 and -3 respectively).

- MISTRAL-4 was a mock-up core (Figure 4), particularly devoted to the study of control clusters (natural and enriched B_4C, AIC, Hf) and to the analysis of the UO_2-MOX interface for this kind of moderation ratio.

MISTRAL started in 1996 and ended in 2000.

Figure 3. Radial cross-section of homogeneous 100%MOX(Pu7%) core MISTRAL-2

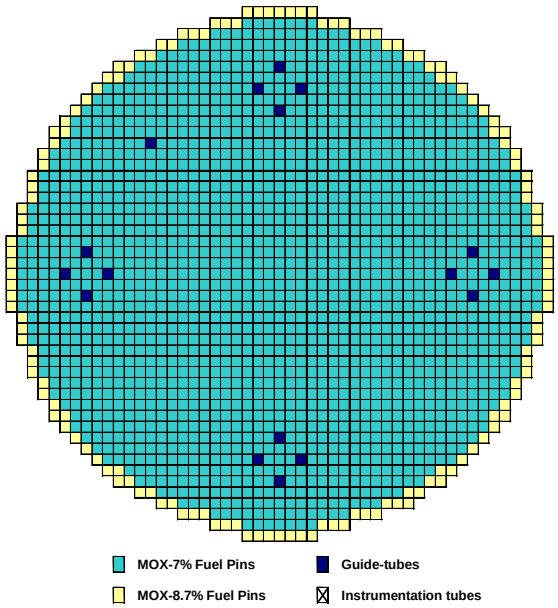
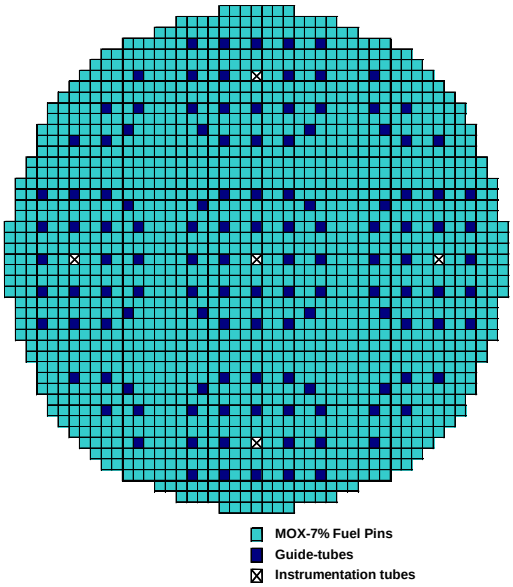


Figure 4. Radial cross-section of the 100%MOX mock-up core MISTRAL-4/REF



The MORGANE programme

The MORGANE experiments in MINERVE were a part of the HCLWR programme launched by the partners. The two main topics were to oscillate at the centre of a regular under-moderated MOX lattice, on the one hand various samples with increasing Pu content, and on the other hand irradiated samples from PWR spent assemblies. The main goal of this oscillation experiment was to validate the reactivity variation versus fuel burn-up in a HCLWR spectrum; furthermore, Pu worth measurement enabled the uncoupling of the actinide component from the fission product component in the fuel reactivity loss. MORGANE was split into two parts: MORGANE/S, which was the pendant of

ERASME/S on EOLE with a central triangular pitch zone of volumetric moderation ratio Vm/Vf ~ 0.5, and MORGANE/R characterised by a triangular pitch with Vm/Vf ~ 0.9. Figures 5 and 6 reproduce the radial and axial cross-sections of the MORGANE core. The main information brought by this experimental programme was the Pu worth and the neutron capture of global fission products in tight lattices.

Figure 5. Radial cross-section of the MORGANE/S core

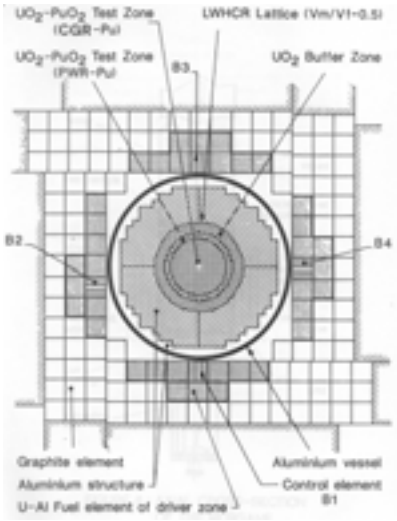
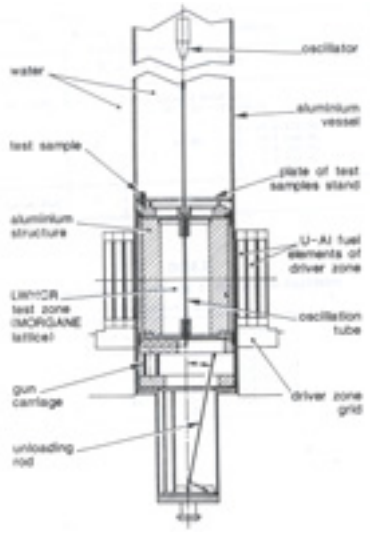


Figure 6. Axial cross-section of the MORGANE/S core with the sample oscillator



The OSMOSE programme

OSMOSE is a future programme that will start mid-2005 in the MINERVE facility. Samples of separated actinide isotopes will be oscillated in different spectra, in particular in the MORGANE/R block around 2007. This will lead to accurate information on the capture cross-sections of poisoning isotopes such as ^{238}Pu, ^{240}Pu, ^{242}Pu and ^{241}Am, and on the "Eta" neutron reproduction factor of fissile isotopes such as ^{239}Pu and ^{241}Pu in intermediate spectra.

Codes and nuclear data validation

The French criticality safety package CRISTAL involves the newest accurate codes APOLLO2, MORET4 and TRIPOLI4; the JEF-2.2 European file is used. The design calculation schemes for the APOLLO2-MORET4 industrial route were defined, accounting for the specificity of under-moderated Pu media [10]. However, the experimental validation of CRISTAL [10,11] concerns mainly Pu and U+Pu nitrate solutions (PU-SOL-THERM and MIX-SOL-THERM of the ICSBEP experiment database), and MOX fuel pin lattices (mainly the MIX-COMP-THERM-004 from JAERI experiments with pin lattice pitch ranging from 2.4-5.5). Unfortunately, these experiments correspond to well-thermalised Pu media.

Furthermore, APOLLO2 and TRIPOLI4 were validated against EOLE experiments, particularly the MISTRAL-2 and MISTRAL-3 100% MOX regular cores using Pu 7% MOX pins [12]. More generally, the MOX reactivity prediction by APOLLO2.5/CEA93 was qualified for moderation ratios V_{MOX}/V_{H2O} ranging from 0.51 (ERASME/S HCLWR experiment) up to 2.1 (MISTRAL-3 RMA experiment), as summarised in Table 1 [13]: these C/E results confirm the ability of the CRISTAL package based on JEF-2 nuclear data to satisfactorily predict the reactivity of MOX lattices for a large range of neutron spectra.

Table 1. Material buckling analysis of MOX lattices

Experiment	Pitch (cm)	Rmod	Slow-down density Q	C_B (ppm)	$K_{eff}^{AP2} - 1$ (pcm)	δk_{eff} (1σ) (pcm)
ERASME/S	0.945 (◊)	0.51	0.24	0	+ 345	(400
ERASME/R	1.035 (◊)	0.90	0.36	0	+ 736	(400
EPICURE MH1.2	1.26 (◊)	1.25	0.52	224	- 368	± 350
MISTRAL2	1.32 (□)	1.75	0.57	0	+ 73	± 350
ERASME/L small	1.19 (□)	2.1	0.50	0	- 628 pcm	± 500
ERASME/L large	1.19 (□)	2.1	0.53	1 656	- 696 pcm	± 400
MISTRAL3	1.39 (□)	2.11	0.60	235	+ 308 pcm	± 300

Δ – triangular pitch, □ – square pitch.

New integral experiments

A new critical mock-up could be implemented in EOLE based on an under-moderated "homogeneous" MOX core concept, provided that a thousand of new highly enriched MOX fuel pins (25-30% Pu content) become available. In this experiment, the main reaction rates could be measured in order to check neutron balance calculation of under-moderated MOX media in criticality-safety code packages.

Concerning MINERVE, different Pu samples on various supports (metallic Pu or oxide powder) could be envisaged and oscillated in intermediate spectra. This would lead to direct information on the integral cross-sections of Pu isotopes.

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PANEL DISCUSSION

Prospects for an International Co-operative Programme

Moderator: J.B. Briggs

Panellists

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Jean-Christophe Niel (IRSN, France)
Hervé Toubon (COGEMA, France)
Toshihiro Yamamoto (JAERI, Japan)
Anatoli Tsiboulia (IPPE, Russia)
Jim Gulliford (BNFL, UK)
Calvin Hopper (ORNL,USA)
Christopher Tripp (NRC, USA)

SUMMARY RECORD OF THE PANEL DISCUSSION

Introduction

J. Blair Briggs opened the session and asked the panellists to introduce themselves. He then recalled the three questions which would be debated during this session:

1) Is there a real need for additional MOX experiments?

2) Which proposal or combination of proposals seems to best fill the need?

3) What are the prospects for an international co-operative programme?

Is there a real need for additional MOX experiments?

In view of the discussions held during the technical sessions, this question was split up in three parts and re-worded in the following manner:

1a) Is there a lack of accurate/reliable experimental data for better code validation?

1b) Would additional experimental data be beneficial?

1c) How urgent/important is the need?

There was a consensus among the panellists that the answer to questions 1a) and 1b) is affirmative. For question 1c), the answer was not so definitive and depended on the perspective. It was agreed that the interest was strong enough to justify debating the remaining two main questions [2) and 3) above].

Which proposal or combination of proposals seems to best fill the need?

It was noted that existing unpublished experiments could partially address the need. However, as some of these experiments were proprietary, it was not possible to comment on their availability within the framework of this panel debate. It was agreed that one of the recommendations of the workshop would clearly state the desire of the international community to access these data and the Nuclear Science Committee would be asked to help investigate their availability. The rest of the panel discussion was therefore restricted to a debate on the opportunity of performing new experiments that could complement those already available.

J. Blair Briggs asked the panellists to express their views on question 2), i.e. among the five proposals presented in Session III, which proposal(s) seem(s) to fill the need? The following nomenclature will be used to refer to the proposals:

- BFS-MOX (see the presentation of Igor Matveenko in Session III).
- KEOPS (see the presentation of Paul Van den Hende in Session III).
- LANL-MOX (see the presentation of David Hayes in Session III).
- VALDUC-MOX (see the presentation of Patrick Fouillaud in Session III).
- VENUS-MOX (see the presentation of Peter Baeten in Session III).

Hervé Toubon (COGEMA, France) stated that he considered the uncertainties for this application area (MOX fuel fabrication) higher than in other areas of the fuel cycle. However, appropriate safety margins were taken into account in the licensing of the MELOX plant to accommodate the lack of code validation. The available safety margins and the corresponding uncertainties in the calculation models had never been thoroughly assessed. It would thus be beneficial to investigate and reduce these margins.

COGEMA does not have any urgent need in this area since the capacity of the MELOX plant was increased two years ago, and there are no plans at the present for a further increased capacity.

The possibility of accessing the ERASME/S experiment would address the need of validation for the secondary blend (12.5% Pu content and 5% water content). Consequently, Hervé Toubon stated that he is more interested in experiments with higher plutonium contents (up to 30% as in the primary blend) and lower water content (1-3%).

Jean-Christophe Niel (IRSN, France) explained that he sees five reasons to support an experimental programme on MOX fuel:

- From the industrial viewpoint, there is an interest in optimising the process. MOX fuel manufacturers expressed this interest in their presentations during the first session.
- From the viewpoint of regulators, it is important to review and precisely assess the available safety margins. Likewise, the tools used in the safety assessment need to be validated for a wide range of applications.
- From the viewpoint of public acceptance, plutonium handling has always been more problematic than for other fuels. Thus, it is important to demonstrate that all the operations with Pu and MOX fuels adhere to strict safety standards and that these standards are based upon the most reliable tools and data.
- It is important to keep an active level of research in the safety area. This is a key point for ensuring an appropriate level of competence and skill.
- It is also important to maintain research facilities. A recent OECD study highlighted the decrease of the number of experimental facilities world-wide and recommended support of the remaining infrastructure.

Concerning the choice of experiments, Jean-Christophe Niel stated that there was not sufficient information to carry out an objective comparison of the proposals. He suggested that a method be developed to this end. The application area should be precisely defined and an analysis grid should be developed with parameters such as:

- What are the needed data and corresponding target accuracy?

- How easily can the experiments be interpreted with the tools used in criticality safety assessment?

- Are the cost and schedule reasonable?

Concerning the application area, he highlighted that there was a need for data regarding high plutonium content and low moderation ratios.

Jacques Basselier (Belgonucléaire, Belgium) stated that Pu handling remained an important issue in the fuel cycle. As he worked at a fuel manufacturing company, he expressed his interest in further code validation for Pu and MOX fuel fabrication processes. He also felt that the main parameters to be considered in the choice of experimental programmes were the technical scope (mainly related to the applicability of experiments), the time schedule and the cost[1].

Christopher Tripp (NRC, USA) stated that the two main categories of fuel (weapons-grade plutonium and reactor-grade plutonium) deserve equal importance. As concerns the weapons-grade plutonium, the BFS-MOX experiments covered the widest range of important parameters (plutonium content and moderation). He noted that most of the proposals considered lattices of MOX rods while the need was related to MOX powders.

Jim Gulliford (BNFL, UK) said that in his opinion there was a sufficient justification for new experiments. He recognised that to continue using old nuclear data libraries just because they lead to conservative results was not a satisfactory approach. For his organisation, the need was not urgent, as there was no immediate plan to increase the design capacity of the Sellafield plant. However, the new experiments would provide increased flexibility that might be beneficial in the future.

Jim Gulliford then established a list of criteria that should be considered for the selection of experiments. These criteria included moderation, Pu content, Pu isotopics, the possibility of performing diagnostic measurements and the applicability of experiments.

More information was needed before establishing a definitive statement. However, the BFS-MOX experiments seemed to adequately address all these criteria. The VALDUC-MOX experiments, mainly limited to reactor-grade plutonium, respond correctly to the criteria on moderation, Pu content and applicability.

Toshihiro Yamamoto (JAERI, Japan) informed the participants that the operation of a MOX fabrication plant in Japan was planned for 2010. The licensing of this plant would take place in 2005. The need for experiments was thus quite urgent. However, since the schedule of the licensing was very close, he didn't see how any of the experimental proposals could be used in the licensing of this plant. Therefore, as in other countries, conservative assumptions would be used for the design of the fabrication plant. The experiments would still be needed before plant operation in order to provide increased confidence and to address questions of public acceptance.

[1] Note: J. Basselier added the following sentence to the summary record: *Concerning the KEOPS programme proposed by Belgonucléaire, J. Basselier stated that powder experiments would probably give the possibility to investigate various parameters and situations encountered in the MOX process. Critical experiments with pure PuO_2 powder, primary blend, secondary blend or scraps could be performed in order to provide a recent database to existing actors and newcomers in the MOX manufacturing business.*

As for the criteria of selection, he quoted the following:

- It is important to choose an experimental configuration which is as simple as possible to allow direct use in the licensing process.
- Experiments should preferably be performed with a homogeneous core (only one type of fuel material).
- Reactor-grade plutonium (content of ^{240}Pu higher than 17%) should be used, since this is the only application envisaged in Japan.
- Ideally, the experiments should use MOX powders, since additional justification must be provided during the licensing process if lattice experiments are used to demonstrate the validation of codes.

Anatoli Tsiboulia (IPPE, Russia) pointed out that nuclear data uncertainties were high in the energy range corresponding to the neutron spectra of low-moderated MOX powders. The situation was similar several years ago in the area of fast breeder reactors and integral experiments helped to decrease these uncertainties. Some unpublished experiments may help in the benchmarking of these media but they have not been properly evaluated and their added value in terms of decreasing the uncertainties was not proven. This was why he considered that new experiments were needed. As for the choice of experiments, he believed that the VALDUC-MOX experiments were the most promising. He finally stated that unless these experiments were performed in the framework of an international consortium, the results would likely remain proprietary.

Calvin Hopper (ORNL, USA) explained that it was very important to evaluate code biases and subcritical margins. The experiments may not seem important for a static mind (reflecting on the needs of the present situation only). However, their importance is clearly understood in a dynamic thinking process, as they would certainly help address the needs associated with technology improvements and optimisation. As far as the choice of experiments was concerned, Calvin Hopper explained that the BFS-MOX and VALDUC-MOX experiments offered the greatest flexibility for varying parameters such as moderation ratio and plutonium content. If one considered the following criteria: applicability of the experiments, cost of the programme and the corresponding schedule, he rated the BFS-MOX programme first and the VALDUC-MOX programme second. He considered the other programmes as less attractive, as they considered heterogeneous set-ups in which the weight of the test zone was not large enough compared to the driver zone. Concerning the question of performing experiments with homogeneous powder as opposed to tight pitch lattices, he explained that uncertainties associated with powder experiments were probably going to be high since it is difficult to control the moderation ratio and density. Lattice experiments were representative of the application area, as long as the moderation was low enough to ensure that the neutron free path was much larger than the pitch of the lattice.

The variety of interests expressed by the panellists together with the lack of detailed information on the experimental proposals were the main reasons why a consensus on a single or a combination of experimental programmes could not be reached. The proposal of Jean-Christophe Niel to develop a method of selection and iterate with the experimental facilities was considered as the appropriate way to proceed.

What are the prospects for an international co-operative programme?

Most panellists expressed their interest in setting-up an international Co-operative programme to perform the needed experiments. Indeed, this would allow an efficient cost sharing and thus

optimisation of available resources. Jacques Basselier informed the participants that an introductory meeting on the KEOPS programme would tentatively be held at Belgonucléaire (Brussels) on 24-25 June 2004 and he invited the workshop participants to this meeting. Anatoli Tsiboulia mentioned the International Science and Technology Center (ISTC) mechanism as a suitable means for carrying out the BFS-MOX experiments. Indeed, the ISTC has about 12 years of successful experience in co-ordinating co-operation between organisations around the world and laboratories in Russia. The BFS-MOX project could be performed as an extension of ISTC Project 815 for a total cost of 0.5 M$ over a period of 1.5 years.

Mike Westfall commented that the statement of Jean-Christophe Niel concerning the maintenance of the activity of experimental facilities was very important. He asked the experimentalists to highlight the significance of the MOX programme in this regard.

Conclusion

The recommendations of the panel can be summarised as follows:

- Investigate the possibility of releasing and evaluating unpublished experimental data (especially the ERASME/S and BFS-49 data).

- Define a framework and method for the selection and performance of new experimental programme(s) of interest.

- Help disseminate and assess sensitivity/uncertainty methods.

Appendix 1

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Appendix 3

WORKSHOP ON THE NEED OF INTEGRAL CRITICAL EXPERIMENTS WITH LOW-MODERATED MOX FUELS

14-15 April 2004
OECD Headquarters
Château de la Muette
2, rue André Pascal, Paris 16e

Final Programme

Wednesday, 14 April 2004

SESSION I **MOX Fuel Fabrication – Design, Industrial Experience and Associated Criticality Safety Issues**

Chairs: Y. Vanderborck (Belgonucléaire, Belgium), T. Yamamoto (JAERI, Japan)

M. Arslan, J-P. Bariteau, Y. Couty, D. Favet (COGEMA, France)
MELOX plant – MOX manufacturing – Results and prospects

H. Libon, J. Moerenhout, B. Lance, P. Van den Hende (Belgonucléaire, Belgium)
Operating a Criticality Safety Management System in a MOX Fuel Fabrication Plant

T. Doering, R. Foster, K. Niemer (AREVA and Duke Energy, USA)
US MOX Fuel Fabrication – Design, Industry Experience, Challenges and Associated Criticality Safety Issues

A. Devita[1], S. Evo[2], S. Perrin[1], V. Rouyer[2], J-L. Voitellier[2] ([1]MELOX, [2]IRSN, France)
Criticality Safety Issues Associated with MOX Fuels

C. Tripp (NRC, USA)
Licensing Issues Associated with PuO_2 and Mixed-oxide Powder Processes

SESSION II **Experimental Data Needs**

Chairs: M. Westfall (ORNL, USA), P. Cousinou (IRSN, France)

I. Duhamel[1], V. Rouyer[1], A. Santamarina[2], C. Venard[2] ([1]IRSN, [2]CEA, France)
Criticality Calculation Code Validation: Experimental Needs for Low-moderated MOX Media

T. Yamamoto, Y. Miyoshi (JAERI, Japan)
Research Activities in Japan Atomic Energy Research Institute for Criticality Safety Issue on MOX Fuel Fabrication Process

S. Goluoglu, C. Hopper (ORNL, USA)
Links Among Available Integral Benchmarks and Differential Data Evaluations, Computational Biases and Uncertainties, and Nuclear Criticality Safety Bases on Potential MOX Production Throughput

A. Tsiboulia (IPPE, Russia)
Experimental Needs for Estimation of Criticality Prediction Accuracy for Systems with MOX Fuel

J. Gulliford, J. Edge (BNFL, UK)
Validation for Intermediate Spectrum MOX Applications and Estimate of the Impact of Nuclear Data Uncertainties on Assessment Modelling

B. Lance, P. Van den Hende, J. Moerenhout, H. Libon (Belgonucléaire, Belgium)
Criticality Codes Validation on Spherical Plutonium Systems

Thursday, 15 April 2004

SESSION III Proposed Programmes and Presentation of Experimental Facilities

Chairs: P. D'Hondt (SCK•CEN, Belgium), J. Gulliford (BNFL, UK)

P. Fouillaud, E. Girault, P. Grivot, J. Legendre (CEA Valduc, France)
The Criticality Laboratory of Valduc (France) and Its Ability to Meet the Experimental Needs for Low-moderated "MOX" Fissile Media

I. Matveenko, A. Tsiboulia (IPPE, Russia)
Experiments with MOX Fuel Planned on the BFS facility

P. Baeten[1], H. Aït Abderrahim[1], P. D'Hondt[1], B. Lance[2], P. Van Den Hende[2], D. Marloye[2], J. Basselier[2] ([1]SCK•CEN, [2]Belgonucléaire, Belgium)
Critical Experiments with Low-moderated MOX Rods in VENUS

P. Van den Hende[1], B. Lance[1], D. Marloye[1], J. Basselier[1], P. Baeten[2], H. Aït Abderrahim[2], P. D'Hondt[2] ([1]Belgonucléaire, [2]SCK•CEN, Belgium)
KEOPS: A Critical Experiment Using PuO_2 Powder in the VENUS Reactor

D. Hayes (LANL, USA)
Mixed-oxide Fuel Experiments and the Los Alamos Critical Experiments Facility

P. Blaise, P. Fougeras and A. Santamarina, S. Cathalau (CEA, France)
Integral Needs for MOX Experiments: State-of-the-art at CEA Cadarache on MOX Experiments

SESSION IV Panel Discussion: Prospects for International Co-operative Programme

Moderator: Blair Briggs (INEEL, USA)

Panellists: J. Basselier (Belgonucléaire, Belgium)
Jean-Christophe Niel (IRSN, France)
Hervé Toubon (COGEMA, France)
Toshihiro Yamamoto (JAERI, Japan)
Anatoli Tsiboulia (IPPE, Russia)
Jim Gulliford (BNFL, UK)
Calvin Hopper (ORNL, USA)
Christopher Tripp (NRC, USA)

Workshop Summary – Ali Nouri (OECD/NEA)

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