

THE PRINCESS PROJECT: FROM DIFFERENTIAL TO INTEGRAL EXPERIMENTS

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ABSTRACT

Following the shutdown of the CEA Valduc experimental facilities, where, for more than 50 years, IRSN used to perform experiments related to criticality safety, IRSN initiated a new project named PRINCESS (PRoject for IRSN Neutron physics and Criticality Experimental data Supporting Safety). The objective is to continue collecting experimental data necessary for the IRSN missions in nuclear safety. For this purpose, collaborations with various national and international laboratories have been established. The PRINCESS project covers various nuclear physics fields from nuclear data to criticality-safety and reactor physics providing information to both differential and integral data improvements.

KEYWORDS: experiments, integral, differential, collaboration

1. INTRODUCTION

In the frame of its missions, IRSN performed various experimental programs related to criticality-safety in collaboration with the CEA Valduc. Following the shutdown of the Valduc facilities, IRSN has initiated a new project named PRINCESS [1] for “PRoject for IRSN Neutron physics and Criticality Experimental data Supporting Safety”. The aim is to continue gathering experimental data necessary for criticality safety by the mean of collaborations with various national and international laboratories and to extend them to reactor physics. The PRINCESS project covers the acquisition of new experimental data, but also old ones that are not freely available.

This paper presents an overview of the status of the IRSN PRINCESS project.

2. NUCLEAR PHYSICS FIELDS OF THE PRINCESS PROJECT AND ON-GOING COLLABORATIONS

The PRINCESS project covers various nuclear physics fields from nuclear data to criticality safety and reactor physics providing information to both differential and integral data improvements.

The following main technical domains related to the project have been identified according to the various needs.

2.1. Nuclear data

A precise knowledge of neutron cross sections is of great importance to accurately calculate reaction rates and detailed neutron flux distributions in many nuclear applications. Reducing uncertainties in the neutron cross-section data can result in an enhanced safety of present and future nuclear systems. Therefore, differential experiments with high energy resolution over the whole energy spectrum are required. These measurements are mainly obtained at neutron Time-Of-Flight (TOF) facilities.

2.1.1. Water thermal scattering cross-sections measurements

The water cross-sections and the distribution of neutrons in the thermal energy range are determined by the interatomic bindings of the hydrogen atoms in the molecular system. In the standard thermal scattering libraries, these effects are described by a $S(\alpha,\beta)$ function which is often termed as thermal scattering law (TSL). The differences between the most recent TSL evaluations and the lack of clear understanding of the temperature behavior of the TSL highlight the need of new experiments for light water to prepare and validate new TSL evaluations.

Thus, IRSN has carried out Inelastic Neutron Scattering measurements of light water with two time-of-flight spectrometers at the Institut Laue-Langevin (ILL) in France to generate the frequency spectrum at several high temperatures and pressures [2]. Additionally, series of light water inelastic neutron scattering experiments were also conducted at the Oak Ridge National Laboratory (ORNL) Spallation Neutron Source (SNS) covering temperatures ranging from 295 K to 600 K and pressures of 1 bar and 150 bar [3], which correspond to ranges of pressurized light water reactors. Considering the requirement of high resolution inelastic neutron scattering measurement, it appeared that the SEQUOIA spectrometer at the SNS is one of the best suited facilities to study the dynamical structure properties of light water at high temperature and pressure with a satisfactory signal-to-noise ratio.

2.1.2. Molybdenum cross section measurements

In reactor physics, molybdenum isotopes are mainly encountered in irradiated fuel as fission products or in molybdenum alloys in research or naval reactors. In the nuclear fuel cycle, ^{95}Mo is also taken into account in criticality safety studies considering burn-up credit for transport casks or irradiated fuel storage, ^{95}Mo being one of the 15 main absorbing fission products in Light Water Reactors (LWR) irradiated fuel assemblies. Besides, in reprocessing plants, during the dissolution process, some UPu-MoZr deposits appear in specific equipments. Thus, accurate nuclear data of molybdenum isotopes in a wide energy range are of great importance for nuclear safety.

As the available nuclear data for molybdenum included in the nuclear data libraries are not of sufficient quality and information about uncertainties and covariance are missing, IRSN and the Japan Atomic Energy Agency (JAEA) performed experimental measurements on molybdenum at the J-PARC (Japan Proton Accelerator Research Complex) facility in Japan at the beginning of 2019 [4]. J-PARC is a proton accelerator facility operated by JAEA and KEK in Tokai-Mura. Neutron capture cross section and transmission measurements were performed on natural molybdenum with ANNRI (Accurate Neutron-Nucleus Reaction measurement Instrument) in MLF (Material Life and science Facility for the purpose of using neutrons to investigate material properties) of J-PARC. Five metallic natural molybdenum samples with various thicknesses of 0.1 mm, 0.5 mm and 2 mm for capture and 0.5 mm and 5 mm for transmission were considered. Additional measurements were performed to determine the background and the normalization factors. A NaI detector (flight length of about 28 m) was used for capture measurements and a Li-glass detector (flight length of about 28.7 m) for transmission measurements. After the data reduction process, the measured data are being analyzed in order to produce more accurate cross sections and associated uncertainties.

Additional measurements on ^{95}Mo , ^{96}Mo , and ^{97}Mo enriched samples are already envisioned for the next years.

The JAEA-IRSN collaboration in nuclear data field will also be strengthened with the measurements of neutron capture cross section of iron isotopes (using enriched samples of ^{56}Fe , ^{57}Fe and ^{54}Fe) that are planned to be conducted at J-PARC in the beginning of 2020.

2.2. Criticality risk prevention

The objective is to contribute to criticality calculation packages and nuclear data validation. The experiments of interest are mainly slightly sub-critical approaches extrapolated to critical conditions using split-tables, pool tanks or sub-critical experiments dealing with noise measurement techniques.

Within the PRINCESS project, IRSN continues its long-standing collaboration with US Department of Energy (DOE) laboratories on various experimental programs under the auspices of the US Nuclear Criticality Safety Program (NCSP). Additionally, the long term collaboration established with JAEA has been strengthened notably with the study of neutronic characteristics of fuel debris in the new STACY Facility.

2.2.1. Design of critical experiments dealing with Mo and Rh in SPRF/CX

In nuclear fuel cycle, molybdenum and rhodium are sometimes taken into account in criticality safety studies for transport casks, irradiated fuel storage or in reprocessing plant (use of Burn-up credit with fission products or UPu-MoZr deposits in reprocessing plants equipment after dissolution, for example). Thus, having accurate nuclear data of Mo and Rh isotopes validated for a wide energy range is important for criticality-safety practitioners. Taking into account that very few integral experiments are available in thermal and epithermal spectra and in order to have uncorrelated additional data for Mo and Rh, IRSN studied the benefit of performing some experiments in the SPRF/CX installation at Sandia National Laboratories (SNL). Integral Experimental Requests (IER) were submitted to the NCSP for both isotopes in 2015 and preliminary designs of experiments [5] were proposed by IRSN. For Molybdenum experiments (IER 305), it is envisioned to perform experiments using molybdenum metallic sleeves around SANDIA UO_2 fuel rods or molybdenum metallic foils, which would be inserted between UO_2 pellets in SANDIA specific fuel rods (known as BUCCX). Regarding Rhodium validation (IER 306), preliminary design shows that using rhodium foils in BUCCX fuel rods brings potential improvements to the sensitivity of k_{eff} to the total cross section of ^{103}Rh when comparing with former existing experiments and that experiments with UO_2 fuel rods in a rhodium nitrate solution offer the opportunity to cover the sensitivity at the first resonance peak of ^{103}Rh . Experiments involving a rhodium resin block pierced with holes hosting UO_2 rods are also being investigated.

2.2.2. TEX experiments

The need for epithermal and intermediate energy range critical benchmarks is an established international criticality safety data need. Indeed, a high sensitivity to the cross sections of interest in the intermediate energy range (0.625 eV – 100 keV) is very desirable to validate nuclear data in the resonances range.

The goals of the TEX (Thermal and Epithermal eXperiments) program [6], performed under the auspices of the NCSP, are to address these needs by executing critical experiments with NCSP fissile assets that span a wide range of fission energy spectra, from thermal (below 0.625 eV), through the intermediate energy range (0.625 eV to 100 keV), to fast (above 100 keV). Three test bed assemblies that will be assembled on a vertical lift machine (namely PLANET or COMET), have been designed by Lawrence Livermore National Laboratory (LLNL): a ^{239}Pu assembly that uses Zero Power Physics Reactor (ZPPR) plutonium metal plates, a ^{235}U assembly that uses highly enriched uranium (HEU) metal Jemima plates, and a ^{235}U assembly that uses ^{235}U oxide ZPPR plates. The assemblies were designed to be easily modified and consist of layers of fuel interspersed with varying amounts of polyethylene moderator, which is used to tune the neutron fission spectrum, and a thin polyethylene reflector to reduce the effects

of room return. Additional plates could be interspersed to test materials of interest (Tantalum, Hafnium, Iron, etc.).

Five TEX-Pu baseline experiments, containing only Pu ZPPR plates and polyethylene moderators, and five complimentary experiments that tested tantalum as a diluent material in the assemblies were performed in 2017-2018 on the PLANET vertical lift machine at the National Criticality Experiments Research Center (NCERC). The baseline cases are currently undergoing evaluation for inclusion in the International Criticality Safety Benchmark Evaluation Project (ICSBEP) handbook [7], IRSN being the independent reviewer.

Since the TEX kick-off meeting, IRSN is collaborating with NCSP in experiments design and evaluation and is currently leading the design of the TEX-MOX experiments [8].

The aim of the TEX-MOX program is to obtain critical experiments representative of $\text{UO}_2\text{-PuO}_2$ powder mixtures (11wt% - 30wt% PuO_2 content) with a varying isotopic content of ^{240}Pu (5wt% - 25wt%) and different water contents (between 0wt% and 5wt%). The set of the TEX-MOX experiments should also cover different energy ranges from thermal to fast neutrons.

Considering some experimental constraints and using optimization algorithms implemented in the IRSN PROMETHEE tool [9], preliminary critical configurations that satisfy the goals were found. Mixing available fuel plates allows varying the plutonium content and the isotopic ^{240}Pu content and the use of different materials as moderator and reflector allows covering different parts of the intermediate energy range. Thus, using Al_2O_3 as moderator allows covering the 1 keV–100 keV energy range, whereas polyethylene, borated polyethylene and graphite plates allow increasing the k_{eff} sensitivity to cross sections for energies ranging from 5 eV to 1 keV. Additionally, it was shown that critical thermal and fast configurations can be achieved.

It is important to mention that the decay heat of these configurations is higher than those of the TEX-Ta configurations. Therefore, thermal and radiation dose issues associated to the strong decay heat should be analyzed in further studies.

2.2.3. Fuel debris experiments in the new STACY Facility

In case of severe accident, it is important to evaluate the neutronic characteristics of molten-core-concrete-interaction (MCCI) products, which are composed of a mixture of fuel, concrete, and alloys (steel, zircaloy, etc.) to facilitate criticality risk assessments during retrieval operations from the reactors.

Following the Fukushima accident, the Japan Atomic Energy Agency (JAEA) has developed activities on the criticality control of fuel debris within a contract with National Regulation Authority (NRA). For that purpose, it was decided to re-start and renew the Static Critical Experiment Facility (STACY), whose core is being converted from a solution-fuel type to a fuel rod and water-moderator type [10]. The first criticality of the new STACY is planned in the beginning of 2021.

For many years, IRSN and JAEA have renewed regularly their general collaboration agreement and have had technical exchanges on various subjects in the frame of criticality safety. Therefore, in the frame of the PRINCESS project, IRSN and JAEA are collaborating in the core design and performance assessment of the fuel debris experiments [11]. Preliminary calculations were performed to optimize the design of core configurations of the new STACY to measure the criticality characteristics of pseudo fuel debris focused on Molten Core Concrete Interaction (MCCI) debris. The design method applied to define preliminary configurations is based on the optimization of k_{eff} sensitivity to the cross section using the IRSN PROMETHEE tool [9]. It was proposed that, for the first experimental configurations, pseudo fuel debris rods would be composed only of concrete. To have experiments adapted to the diversity of possible concretes, experiments for different concrete compositions could be considered. These compositions could be based on the samples taken from Fukushima Daiichi NPP.

2.2.4. Sub-critical experiments using neutron noise techniques

Because sub-critical experiments are time-dependent they are helpful to validate fission multiplicity treatments in calculation packages. Besides, validating neutron noise techniques measurements are also of great interest for characterization of multiplying systems, which could be useful to the nuclear counterterrorism and non-destructive analysis communities.

In the frame of the PRINCESS project, IRSN is collaborating with LLNL and Los Alamos National Laboratory (LANL) on the design, evaluation and analysis of various sub-critical experiments involving different fissile materials and reflectors.

Thus, IRSN contributed to the ICSBEP evaluation [7] and the analysis (using the IRSN MORET code [12]) of the 5 subcritical benchmark experiments conducted by LLNL with the Inherently Safe Subcritical Assembly (ISSA)[13], a highly enriched uranium (HEU) system moderated and reflected by water. Additional experiments using HEU fuel elements with lower uranium content are planned in 2019. Moreover, IRSN and LLNL will collaborate on the design of new experiments mixing fuel assemblies of different uranium content in order to study the feasibility of detecting a fuel loading error with neutron noise techniques.

In recent years, LANL has performed subcritical benchmark evaluations that comprise many configurations with beryllium-reflected plutonium (BERP) ball, referred as to FUND-NCERC-PU-HE3-MULT benchmarks in the ICSBEP handbook [7]. Some configurations include Cu (known as SCRaP experiments [14], for Subcritical Copper-Reflected Alpha-phase Plutonium), Ni and W reflectors. These experiments were performed at the National Criticality Experiments Research Center (NCERC) at the Nevada National Security Site (NNSS). IRSN and LANL collaborated on the evaluation and the analysis of these experiments using their own codes.

To expand the range of subcritical benchmarks, a new experimental program, named MUSIC [15], will be performed by LANL in collaboration with IRSN using highly enriched uranium shells, known as the Rocky Flats shells. This experiment will be performed using the Planet vertical lift machine, which will combine an upper and lower subassembly together to assemble the full experimental configurations. The aim is to validate subcritical measurement and simulation methods for deeply subcritical to critical configurations.

2.3. Criticality accident detection and consequences assessment

In the frame of its missions in nuclear safety, IRSN should evaluate the expected radiological consequences of a potential criticality accident, in order to verify that preventive measures planned by the operators (detection, evacuation, mitigation of the criticality accident, etc.) can limit as much as possible these consequences to the staff, the population and the environment. Besides, in case of a criticality accident, doses to the victims should be estimated within a short delay to contribute to the diagnosis and to help to define the best therapeutic strategy. For that purpose, it is necessary to understand the phenomenology of the criticality accidents and to validate the Radiation Protection Instrumentations and doses estimation. It is also tremendous to maintain competences by practicing criticality accident dosimetry regularly and to train staff to perform such dosimetry.

Since 2014, IRSN has contributed to many experiments and dosimetry inter-comparison exercises [16] with DOE laboratories, the first one being organized by IRSN with the CALIBAN and PROPSERO reactors at the CEA Valduc research center [17]. This was followed by criticality accident dosimetry exercises that occurred in the US Nevada National Security Site using GODIVA-IV and FLAT-TOP reactors under the auspices of the NCSP. One of the main conclusions of these exercises is that they need to be performed regularly (ideally every one or two years) in order to maintain skills of the labs for this very specific dosimetry.

Further collaboration is envisioned with LLNL in the next years using a well characterized ^{252}Cf source, in order to investigate of Criticality Accident Alarm System (CAAS) response. IRSN would perform the neutron kerma and photon dose measurements and neutron spectrum using various detectors.

A full dosimetry exercise that would be performed under the auspices of the NCSP is also under discussions. The aim is to play an exercise in conditions as close as possible of a real accident in order to test not only the classical criticality dosimetry systems, but also retrospective accident techniques in order to estimate the dose distribution in the body. Several phantoms (with various orientations and distances from the reactor) may be placed around GODIVA or FLATTOP reactor depending on their availability. These phantoms will be equipped with various “items” and samples (such as hairs, nails, teeth, blood sample, etc.), which will be measured and used for the estimation of doses.

2.4. Validation of depletion calculation codes

Depletion codes are used in reactor physics to determine the composition of the spent fuel assemblies and also in criticality-safety studies for burn-up credit calculations. In order to validate the calculation packages (including codes, nuclear data and calculation schemes), Post-Irradiated Examinations (PIE), which consist of chemical analyses of irradiated fuel samples, are needed.

On November 2014, SCK•CEN (the Belgian Nuclear Research Centre) and IRSN signed a new framework cooperation agreement in the field of nuclear safety, radiation protection and nuclear waste management, which was actually the renewal for another 5 years of an on-going longstanding collaboration that was initiated in 2008. At the same time, a bilateral collaboration agreement was signed under this framework agreement that outlines the collaboration between SCK•CEN and IRSN in the REGAL (Rod Extremity and Gadolinia AnaLysis) program [18]. It aims investigating two challenging issues in terms of characterizing the isotopic inventory of nuclear fuel: rod extremity effects and the use of gadolinium-doped fuel. Post-Irradiated Examinations, including destructive radiochemical analyses, are being performed on UO₂ (initially enriched at 4.25% in ²³⁵U) and UO₂-Gd₂O₃ (with initial ²³⁵U enrichment of 2% and Gd₂O₃ content of 10%) PWR samples of various burn-ups. UO₂-Gd₂O₃ fuel samples are taken from a gadolinium-doped fuel rod that failed during its first cycle in the Tihange 1 reactor. Its average burn-up was around 12 MWd/kg_{HM}. This Program will provide valuable experimental data for the validation of the IRSN VESTA 2.2.0 [19] depletion code for fuel assemblies containing burnable poisons.

2.5. Reactor physics

The aim is mainly to contribute to neutron physics codes validation and to instrumentation and measurements techniques validation. The experiments considered are mainly reactor mock-up experiments. In the frame of the PRINCESS project, measurements are being performed in order to support safety assessment of reactors start-up and to study material ageing.

Simulations have shown that, at low power levels in a critical system, neutrons may start to cluster. Therefore, measurements were performed in 2017 at the Rensselaer Polytechnic Institute (RPI) Walthousen Reactor Critical Facility (RCF) in the frame of LANL, IRSN, and RPI collaboration in order to measure this phenomenon in a real reactor [20]. Measurements were conducted at different reactor powers, from less than 1 mW to 0.85 W at various reactivity states. Neutron counting was performed using two NOMAD detectors placed one on the other but also ³He tubes and RCF detectors (uncompensated ion chambers). ³He tubes were used to evaluate correlations versus distance in the core and NOMAD detectors for the analysis of spatial correlations versus the reactor power. Measurement times varied from 30 seconds to 2 hours long. Experimental results are being analyzed taking into account experimental uncertainties.

2.6. Density laws

In criticality-safety studies, density laws are often used to determine the composition of fissile media as a function of the moderation ratio. Chemical analyses of fuel solutions at various concentrations, acidities and temperatures are thus required in order to generate and validate density laws.

Some measurements on plutonium solutions are envisioned in the frame of the collaboration with the DOE-NCSP.

IRSN also takes part to the European Horizon 2020 GENIORS project [21], which focuses on improving current recycling of spent nuclear fuel and future multiple recycling strategies to be implemented in the 4th generation of reactors. In this frame, chemical analyses of fuel solutions using solvents envisioned for new reprocessing process at various densities and concentrations are planned to be performed.

3. CONCLUSIONS

IRSN initiated a project named PRINCESS (PROject for IRSN Neutron physics and Criticality Experimental data Supporting Safety). The objective is to collect experimental data necessary for its missions in nuclear safety. The PRINCESS project covers various nuclear physics fields from nuclear data to criticality-safety and reactor physics providing information to both differential and integral data improvements. It concerns the acquisition of new experimental data, but also old ones that are not freely available.

Strong bilateral collaborations have already been put in place with DOE/NCSP, JAEA and SCK-CEN, allowing IRSN to maintain its capabilities in nuclear criticality-safety and reactor physics. Collaborations with other institutes and other countries are foreseen in the next years.

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