Resumen

1. Introduction

The European Commission has established on its long-term strategy that renewable and nuclear energies will constitute the backbone of a carbon-free European power system. The Lead-cooled Fast Reactor (LFR) is one of the three advanced reactor technologies selected by the Sustainable Nuclear Energy Technology Platform that can meet future European energy needs. However, significant drawbacks for the industrial deployment of LFR still exist, among which are the lack of operational experience and the little knowledge about the potential impact of uncertainties in the reactor design, operation and safety assessment.

Traditionally, expensive experimental facilities have been needed for the design of a new nuclear reactor in order to conduct dedicated experiments to cover a wide range of scenarios and aspects of the new system. Nowadays, considering the cost of such facilities, it would be desirable that the design of new systems could rely more on computer simulations. Accurate simulations can help to improve understanding and optimize the design and safety margins in any operational conditions. Hence, the capability to provide reliable predictions is recognized as a priority to support the development of LFR technologies.

The quantification of uncertainties associated with the computational outcomes is imperative to establish the credibility of the results and make robust decisions based on simulations. In nuclear reactor design, the uncertainties come from material properties, fabrication tolerances, operative conditions, simulation tools and nuclear data. The uncertainty in nuclear data is one of the most important sources of uncertainty in neutronics calculations and consequently in reactor design. Accurate and reliable tools are therefore needed to propagate nuclear data uncertainties to reactor key parameters (neutron multiplication factor, safety coefficients, …). Significant gaps between the estimated uncertainties in reactor core parameters and the target accuracies (i.e., maximum acceptable uncertainties for a certain parameter) have been systematically shown in the past, suggesting nuclear data weaknesses. Meeting the target accuracy is required not only to achieve the requested level of safety for this technology, but also to minimize the increase in the costs due to additional safety measures.

The main objective of this Thesis has been to analyse and improve the nuclear data required for the development, safety assessment and licensing of LFR reactors, reducing the uncertainties in the criticality safety parameters due to the uncertainties in nuclear data, in order to reach the target accuracies defined by scientific community, industry and regulators. This has been carried out by means of sensitivity and uncertainty (S/U) analyses to quantify the impact of nuclear data uncertainties in reactor integral parameters, and data assimilation to constrain uncertainties, with a S/U code, SUMMON, and a data assimilation code, DAWN, developed in the framework of this Thesis. In addition, LFR coolant materials in JEFF-3.3 state-of-the-art nuclear data library have been thoroughly analysed. LFR nuclear data needs have been identified, improvements have been proposed and recommendations have been given.

2. Sensitivity and Uncertainty Methodology for MONtecarlo codes

The Sensitivity and Uncertainty Methodology for MONtecarlo codes (SUMMON) system has been conceived as a tool to perform complete automated sensitivity and uncertainty analyses of the most relevant criticality safety parameters of detailed complex reactor designs from the neutronics point of view, i.e., k_{eff}, β_{eff}, Λ_{eff} and reactivity coefficients, using state-of-the-art nuclear data libraries and covariances.

SUMMON is based on the use of the KSEN card of MCNP code to perform the eigenvalue sensitivity calculations, although any code that can provide sensitivity coefficients can be used. The sensitivity coefficients of a reactivity response are calculated using the eigenvalue definition of reactivity, which is equivalent to applying the Equivalent Generalized Perturbation Theory. Moreover, the effective delayed neutron fraction sensitivity coefficients are derived from Bretscher's approximation or employing Chiba's modified method, whereas the sensitivity coefficients of the effective neutron generation time are obtained using the 1/v insertion method and the Equivalent Generalized Perturbation Theory. Uncertainties are propagated using the "Sandwich Rule" of the "Propagation of Moments" method employing state-of-the-art covariance libraries.

The sensitivity coefficient implementations in SUMMON have been validated and verified using integral benchmark experiments from the International Handbook of Evaluated Criticality Safety Benchmark Experiments (ICSBEP) and comparing SUMMON with the well-validated and consolidated codes such as SCALE, SUSD3D and SERPENT (e.g., Tab 1). Good agreement has been found in (n,f) and (n,γ) reactions and the average delayed, prompt and total neutron fission multiplicities, whereas differences are found in the (n,n) and (n,n') sensitivities due to differences in the k_{eff} sensitivity to nuclear data predicted by both codes for those particular reactions and the inherent difficulty of accurately estimating the sensitivity to elastic scattering.

Reaction	$S_{l_{eff},\alpha}$ **(%/%)**	
	SERPENT	**SUMMON**
(n,f)	-0.247	-0.250
(n,n)	0.227	0.237
(n,n')	0.202	0.181
(n,γ)	-0.037	-0.034
ν	0.010	0.009

Tab 1: SUMMON and SERPENT l_{eff} sensitivities to ^{239}Pu reactions for PU-MET-FAST-001 integral experiment

3. JEFF-3.3 Nuclear Data library

Before nuclear data can be used for practical applications, a thorough verification/validation must be performed in order to ensure that the data is consistent with energy dependent and integral experimental measurements. Consequently, a thorough validation, verification and benchmarking of the latest version of the JEFF library at the time, JEFF-3.3T1, released on March 2016, was performed in order to ensure the quality and accuracy of the data used in the posterior analyses. In particular, the coolant materials of an LFR, lead and bismuth, were chosen as the main objects of study since they are of vital importance for the neutronics performance and criticality safety and were not covered in the CIELO pilot project.

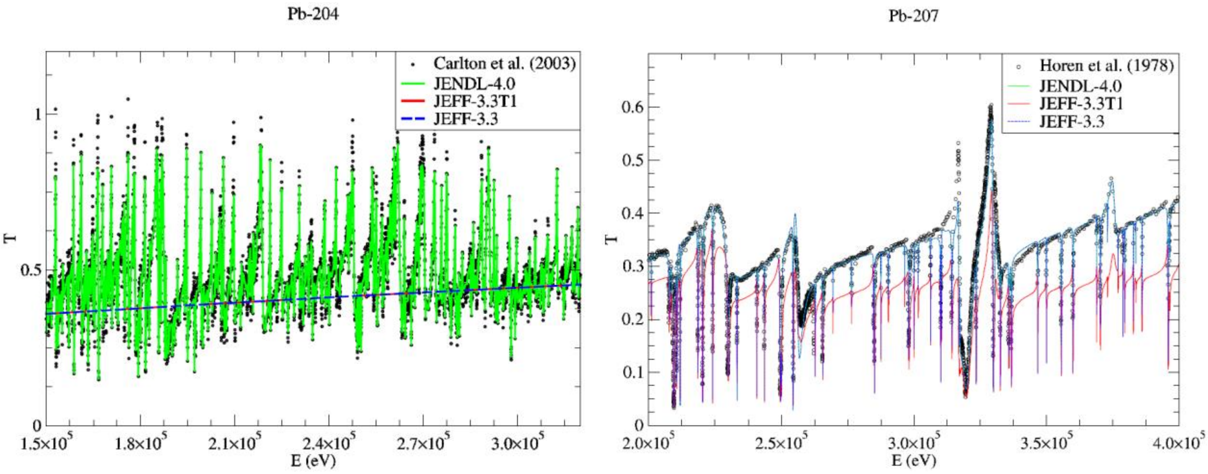

Fig 1. ^{204}Pb (left) and ^{207}Pb (right) transmission comparisons with experimental data

Several problems were found in the Resolved and Unresolved Resonance Ranges (RRR and URR) of JEFF-3.3T1 lead and bismuth files: in ^{204}Pb the total cross section is underestimated due to the lack of resonances (Fig 1); the total cross section of ^{207}Pb is severely overestimated (underestimation of the transmission); and ^{209}Bi overestimates the total cross section in the RRR (up to 100 keV) and has a clearly unphysical cross section shape from 100 to 250 keV, missing resonances at the upper end of the RRR (from 100 to 200 keV) and providing only background (Fig 2).

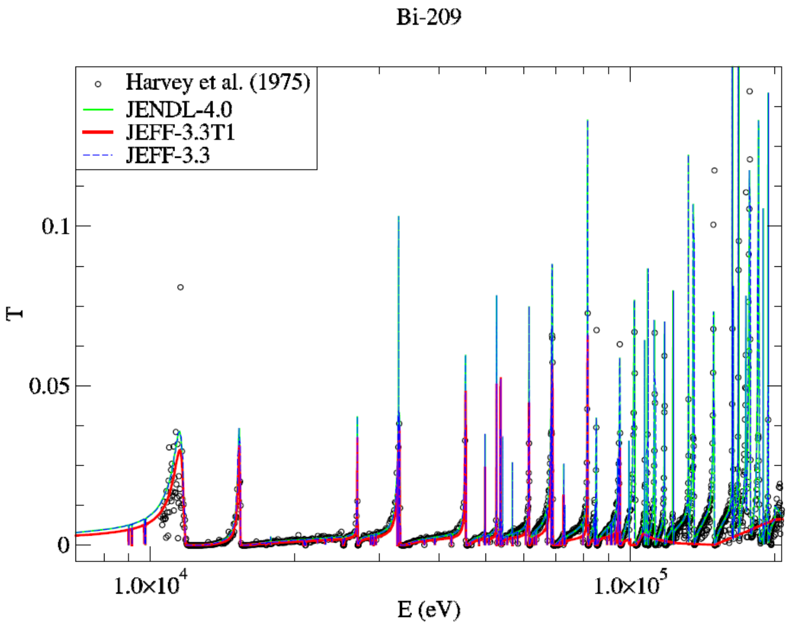

Fig 2. ^{209}Bi transmission comparison with experimental data in the RRR and URR

These problems, as well as the recommendation to employ the RRR from JENDL-4.0 for these isotopes in the final version of JEFF-3.3, were communicated to the JEFF collaboration and, as it can be seen in Fig 1 and Fig 2, the recommendations were adopted in the production version of JEFF-3.3 nuclear data library, excepting for ^{204}Pb which is still based on JEFF-3.2 evaluation.

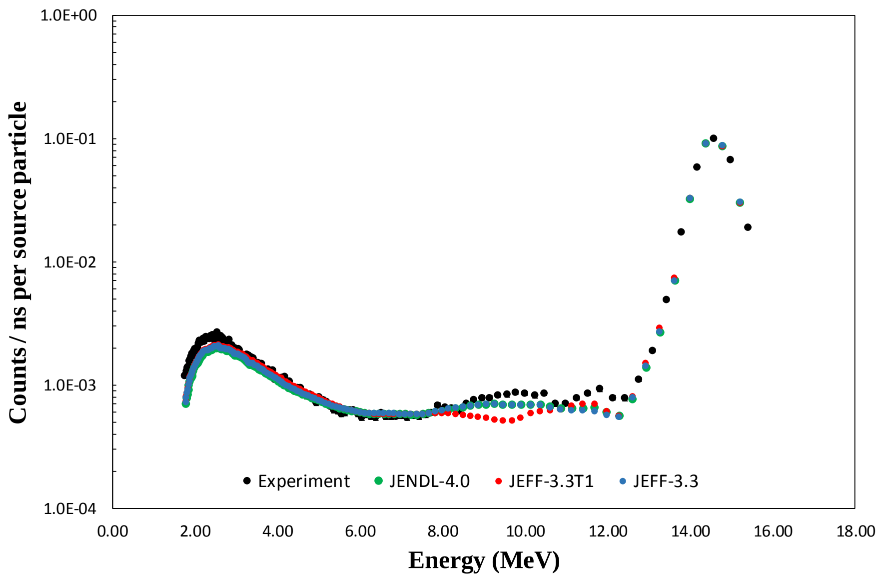

Fig 3. Neutron spectrum for LLNL Pb pulsed sphere

While numerous integral benchmarks sensitive to lead isotopes have been found in the criticality and reactor databases, a lack of integral benchmarks with enough sensitivity to bismuth cross sections has been observed and only one benchmark is currently available for bismuth data validation. JEFF-3.3T1 lead evaluation disagrees with all lead integral benchmarks in the RRR and URR energy ranges, whereas bismuth evaluation is not compatible with the transmission benchmark experimental data from 100 to 250 keV due to ^{209}Bi unphysical evaluation. Better agreement is observed for JENDL-4.0 and JEFF-3.3 lead and bismuth evaluations with integral experiments, as it can be seen in Fig 3.

Regarding the benchmarking, the neutron multiplication factors of ALFRED and MYRRHA differ with respect to JEFF-3.2 using JEFF-3.3T1 evaluation, due to an increase in the fission probability and a decrease in the capture probability, which are partially compensated by a decrease in the average number of neutrons. The differences and inconsistencies found in lead and bismuth evaluations have a direct impact in the criticality and neutronics of both reactors.

Once the production version of the JEFF-3.3 nuclear data library was released, JEFF-3.3 lead and bismuth evaluations exhibited better agreement with differential and integral experimental data than JEFF-3.3T1; therefore, they can be considered as a significant improvement over the first beta evaluations for these materials in JEFF-3.3T1.

4. Target Accuracy Assessment of MYRRHA

Target accuracy assessments allow identifying nuclear data needs and requirements (by isotope, nuclear reaction and energy channel), in order to reduce margins, achieving the requested level of safety and minimizing the increase in the costs due to additional safety measures, in the preliminary conceptual design and in later design phases of selected reactor and fuel cycle concepts. In Tab 2, target accuracies established by researchers and industry for fast reactors at Beginning of Life are presented.

Multiplication Factor	300 pcm
Delayed neutron fraction	3%
Reactivity coefficients (coolant void and Doppler)	7%

Tab 2: Fast reactor target accuracies. No target accuracy for Λ_{eff} has been found in bibliography

SUMMON system has been used to perform a complete target accuracy assessment of an advanced LFR-representative, employing the state-of-the-art JEFF-3.3 nuclear data library. MYRRHA experimental reactor has been chosen as the main object of study, due to the key role it will play in the development of the Pb-alloy technology needed for the LFR Generation-IV concept as an LFR pilot plant.

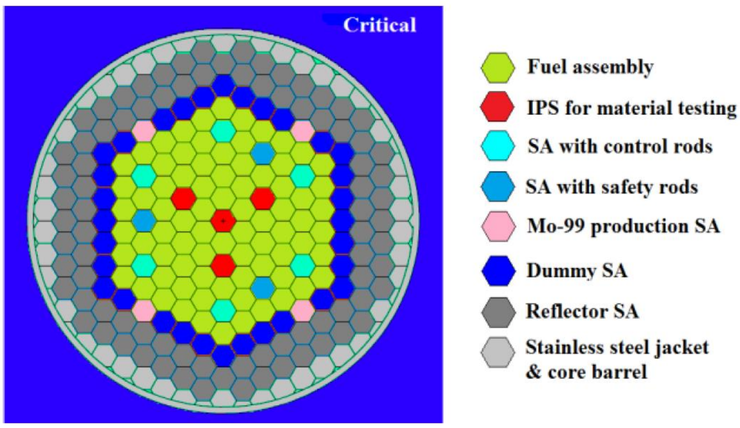

Fig 4. Critical MYRRHA core layout

Furthermore, strong support has been given by the Belgian government and numerous countries and, in fact, MYRRHA's project phase 1 has already started with the construction of the 100 MeV particle accelerator and with the pre-licensing of the reactor. For the analyses a simplified model, homogenised on fuel assembly level, of the critical core configuration in nominal conditions at Beginning of Life has been used. The layout of the core is shown in Fig 4.

Integrated Sensitivity Coefficients (ISC), total uncertainties and main contributors to the uncertainty in the criticality safety coefficients (i.e., k_{eff}, β_{eff}, Λ_{eff}, change in fuel temperature, change in coolant density and control rod worth) have been obtained. In Tab 3, global results from the uncertainty quantification analyses are presented.

Parameter	**Uncertainty (%)**
k_{eff}	0.8
β_{eff}	1.1
Λ_{eff}	20.8
Doppler coefficient (+400 K)	9.1
Coolant density coefficient (-5%)	20.3
Control rod worth	1.8

Tab 3: Uncertainty quantification for MYRRHA with JEFF-3.3

As it can be seen, target accuracies are exceeded for k_{eff}, change in fuel temperature and change in coolant density scenarios. ^{238}U, ^{239,240}Pu, ^{10}B(n,n'), ^{54,56,57}Fe(n,n) and ^{208}Pb(n,n) have been identified as major contributors to the uncertainty in the integral safety-related parameters, due to the high sensitivity of the parameters to these isotopes (e.g., Λ_{eff} sensitivities in Tab 4) and/or due to the uncertainty in the cross sections (e.g., uncertainty in elastic scattering cross section for JEFF-3.3 iron nuclides in Fig 5).

MYRRHA – Λ_{eff} ISC		
Quantity		**JEFF-3.3**
^{56}Fe	(n,n)	$-1.712 \pm 3.6\cdot10^{-2}$
^{16}O	(n,n)	$1.567 \pm 3.8\cdot10^{-2}$
^{209}Bi	(n,n)	$-1.086 \pm 4.4\cdot10^{-2}$
^{238}U	(n,n)	$-0.772 \pm 3.3\cdot10^{-2}$
^{239}Pu	(n,f)	$-0.721 \pm 1.3\cdot10^{-2}$

Tab 4: Λ_{eff} ISC to the 5 most important neutron induced nuclear data for MYRRHA with JEFF-3.3 library

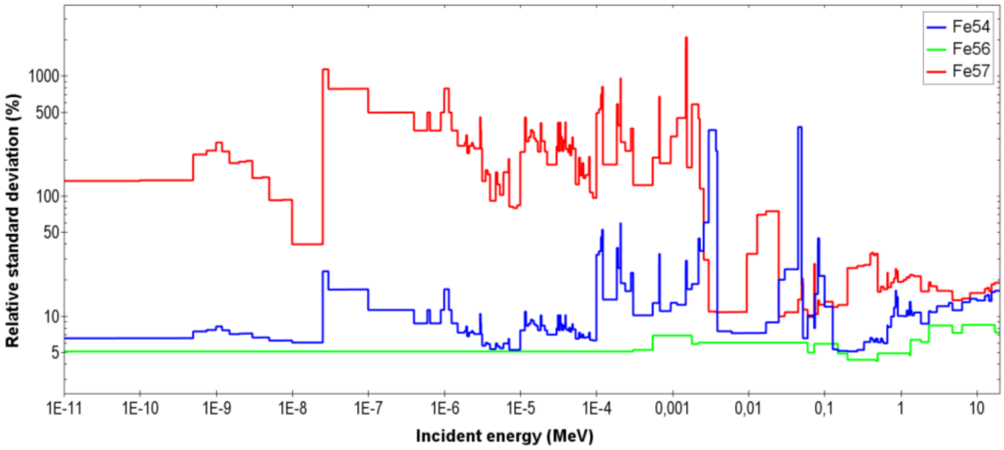

Fig 5. Relative standard deviation of elastic scattering for iron nuclides in JEFF-3.3 evaluations

5. Data Assimilation

To achieve the goal of reducing nuclear data uncertainties in LFR safety coefficients, a reduction of nuclear data uncertainties in JEFF-3.3 for the quantities identified in the S/U analyses is needed. Thus, Data Assimilation With summoN (DAWN) module has been developed to perform data assimilation using integral experiments from public databases, with the aim of providing adjusted nuclear data, not only capable of predicting reactor properties within the target design accuracy, but also statistically consistent with the various differential measurements.

The implemented methodology is based on the GLS technique and the use of integral experiments from ICSBEP database. Individual adjustments using a single experiment or complete adjustments, using up to 93 criticality mass experiments for with reported final experimental uncertainties in the ICSBEP Handbook, can be performed. Furthermore, the REWIND methodology has been implemented in DAWN in order to rank experiments to avoid compensation effects.

It must be stressed that, from a general-purpose library, the adjustment procedure generates a specific-application library, only usable for the targeted reactor concept, or for a system with a representativity factor close to one with that targeted reactor or experiments used in the adjustment.

DAWN was then used to perform data assimilation for MYRRHA with JEFF-3.3 nuclear data library on ^{238}U, $^{239,240}Pu$, ^{10}B, Fe and Pb isotopes. In Tab 5, the representativity factors, f_{RE}, between MYRRHA and the six experiments selected from ICSBEP are shown. ALFRED's representativity factor has also been included in order to evaluate the similarity between MYRRHA and ALFRED regarding nuclear data.

f_{RE}	ALFRED	HMF010	PMF028	HMF064	PMF006	PMF001	PMF002
MYRRHA	0.97	0.39	0.42	0.56	0.81	0.82	0.94

Tab 5: Representative factor of ALFRED and the selected integral experiments

Values greater than 0.9 demonstrate similarity between two experiments and values between 0.8 and 0.9 demonstrate moderate similarity. ALFRED and MYRRHA have a correlation factor of nearly one, therefore, from the point of view of nuclear data, they can be treated as the same system and, consequently, the adjustment will be valid for both reactors. In fact, any other LFR design that uses MOX fuel should have a similar representativity factor.

To study how much room for k_{eff} is present in order to accommodate in the adjustment the discrepancy between the measured and calculated values, the adjustment margin of each experiment has been quantified. The HMF010 experiment has a negative adjustment margin. Negative values of adjustment margin indicate possible inconsistencies in the covariance matrix due to low nuclear data uncertainties, underestimation of experimental or calculation errors, missing experimental correlations and/or the presence of an undetected systematic error in the experiment. If the experiment is kept in the adjustment, unphysical changes in the cross sections may be produced and therefore HMF010 has been discarded.

After the adjustment was performed, the posterior uncertainties obtained using the adjusted covariance matrix were re-assessed in a first step for the integral experiments (Tab 6). For each experiment, the uncertainty in k_{eff} due to uncertainties in nuclear data has been reduced by at least 50%, meeting all experiments the target accuracy established for k_{eff}. The significant reductions in uncertainty are mainly caused by the appearance of strong negative correlations between reaction channels that were not present *a priori* in the covariance matrix, such as $^{239}Pu\ \chi$ – $^{239}Pu\ \nu_p$ or $^{239}Pu\ \nu_p$ – $^{239}Pu(n,f)$. Nevertheless, the uncertainties in the major contributors *a priori*, such as ^{239}Pu ν, have also been constrained, contributing to the uncertainty reduction.

Integral experiment	U_σ (%)	U_σ' (%)
PMF028	0.41	0.21
HMF064	0.33	0.10
PMF006	0.75	0.28
PMF001	0.66	0.11
PMF002	0.84	0.19

Tab 6: Prior and posterior uncertainties due to nuclear data in the experiments used for adjustment

The consistency of the assimilation was assessed against experimental data. An example of the consistency checks is given in Fig 6, where the prior and posterior ^{240}Pu(n,f) cross sections after the assimilation with PMF002 integral experiment are represented. It can be seen that the central value of the cross section has been modified in the fast energy range and is compatible with the various experimental differential data.

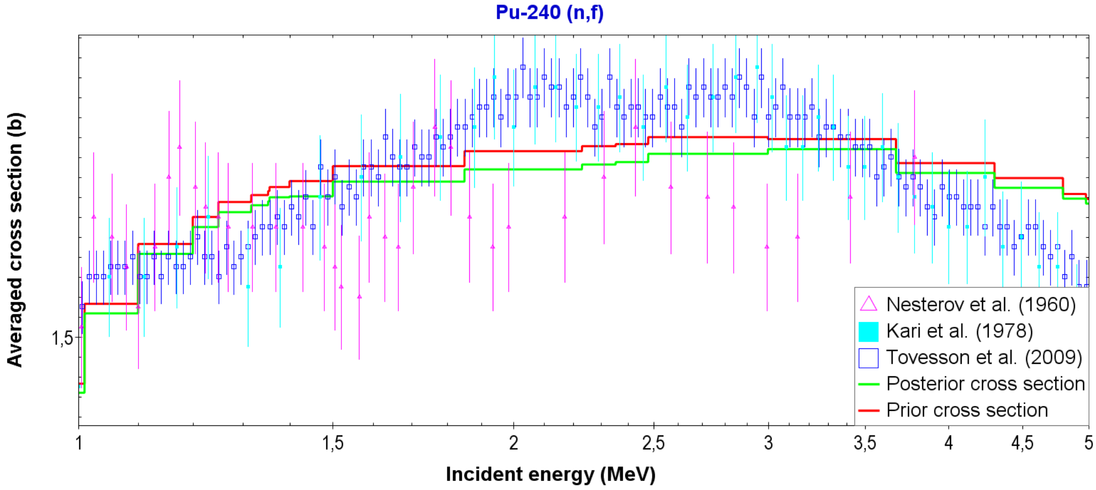

Fig 6. ^{240}Pu(n,f) prior and posterior cross sections compared with experimental data

The adjusted covariance matrix was used to re-assess the total uncertainty in k_{eff}, Doppler and change in coolant density criticality safety coefficients (Tab 7). The total uncertainty in the reactor parameters has been reduced but target accuracies are still not met. Since the most significant adjustments and anticorrelations were generated for ^{240}Pu and ^{239}Pu, the strongest decrease in uncertainty using the adjusted covariance data concerned k_{eff} (nearly ~300 pcm). On the other hand, the reduction in the reactivity coefficients uncertainty due to the adjustment of iron and lead cross sections can be considered negligible.

Parameter	Posterior uncertainty (%)
k_{eff}	0.5
Doppler coefficient	8.7
Coolant density coefficient	19.5

Tab 7: Uncertainty quantification for MYRRHA with JEFF-3.3

6. Conclusions

The aim of this Thesis was to analyse and improve the nuclear data required for the development, safety assessment and licensing of lead cooled fast reactors, reducing the uncertainties in the criticality safety parameters due to the uncertainties in nuclear data, in order to reach the target accuracies defined by industry, experts and regulators. For this purpose, SUMMON system and DAWN module have been developed to perform sensitivity and uncertainty analyses of reactor criticality safety parameters and constrain uncertainties by means of data assimilation.

JEFF-3.3 state-of-the-art nuclear data library (both beta and production versions) has been thoroughly assessed in order to validate data for coolant materials of lead-cooled fast reactors. Several problems were found in the RRR of JEFF-3.3T1 lead and bismuth. These problems, as well as the recommendation to employ the RRR from JENDL-4.0 for these nuclides in the final version of JEFF-3.3, were communicated to the JEFF collaboration and the recommendations were adopted in the production version of JEFF-3.3 nuclear data library.

A target accuracy assessment of the latest MYRRHA design has been performed with the SUMMON system and JEFF-3.3 nuclear data library and target accuracies defined by researchers, industry and experts for k_{eff}, change in fuel temperature and change in coolant density scenarios are exceeded.

Aiming to produce a new covariance data set capable of predicting the criticality safety coefficients with uncertainties lower than the established target accuracies, SUMMON's data assimilation module, DAWN, has been used to perform data assimilation with JEFF-3.3 nuclear data library on ^{238}U, ^{239,240}Pu, ^{10}B, Fe and Pb nuclides. While a significant reduction in k_{eff} uncertainty was observed, k_{eff}, Doppler and coolant voiding coefficients target accuracies were still not met. However, it was proved that the combination of experimental covariance data and integral experiments together with Generalised Least Squares technique, can provide adjusted nuclear data capable of predicting reactor properties with lower uncertainty and consistent with differential data.

The following nuclear data needs in JEFF-3.3 nuclear data library have been identified during the course of this Thesis: i) adoption of JENDL-4.0 or re-evaluation of ^{204}Pb in the RRR and URR; ii) new evaluation of ^{57}Fe inelastic scattering cross section; iii) covariance evaluations for important quantities, such as ^{209}Bi elastic scattering cross section; and iv) reduction of the uncertainty in cross sections, such as ^{57}Fe elastic scattering.

To achieve uncertainties lower than the established target accuracies for Generation-IV advanced lead-cooled fast reactors several recommendations have been given: i) elemental experiments highly sensitive to a single isotope and reaction channel should be preferably used for data assimilation; ii) when using integral benchmarks for data assimilation, experimental measurements of other integral parameters, such as β_{eff}, Λ_{eff} or reactivity effects should be used in the adjustment to provide further complementary information about isotopes and reaction channels that is missing when only k_{eff} is assessed; iii) correlations between the experimental uncertainties of integral experiments representative of innovative reactor designs that can be used in nuclear data validation, adjustment, and assimilation, should be determined; and iv) public integral benchmarks for bismuth data validation are needed.

Finally, as a result of this Thesis, measurements to produce experimental transmission data that can be used to validate evaluated cross sections for neutron induced reactions on ^{nat}Pb and ^{209}Bi in the resonance region have been proposed.