THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY IN APPLIED PHYSICS

Thorium Fuels for Light Water Reactors

Steps towards commercialization

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Division of Nuclear Engineering

CHALMERS UNIVERSITY OF TECHNOLOGY

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Abstract

Thorium-containing nuclear fuel is proposed as a means of gaining a number of benefits in the operation of light water reactors, some related to the nuclear properties of thorium and some related to the material properties of thorium dioxide. This thesis aims to investigate some of these benefits and to widen the knowledge base on thorium fuel behaviour, in order to pave the way for its commercial use.

Part of the work is dedicated to finding ways of utilizing thorium in currently operating light water reactors which are beneficial to the reactor operator from a neutronic point of view. The effects of adding different fissile components to the fertile thorium matrix are compared, and the neutronic properties of the preferred alternative (plutonium) are more closely investigated. The possibility to use thorium as a minor component in conventional uranium dioxide fuel is also subject to study.

Another part of the work is related to the thermal-mechanical behaviour of thorium-containing nuclear fuel under irradiation. To assess this behaviour, an irradiation experiment has been designed and is ongoing in the Halden research reactor. Existing software for prediction of thermal-mechanical fuel behaviour has been modified for application to mixed thorium and plutonium oxide fuel, and the preliminary simulation output is compared with irradiation data.

The conclusion of the research conducted for this thesis is that the adoption of thorium-containing fuel in light water reactors is indeed technically feasible and could also be attractive to reactor operators in a number of different aspects. Some steps have been taken towards a more complete knowledge of the behaviour of such fuel and therewith towards its commercial use.

Keywords: Thorium, plutonium, light water reactors, neutronic simulations, Halden research reactor, fuel performance

To my father, a firm believer in the saving of humankind through technical development.

Preface

This thesis is the result of work performed within the framework of an industrial PhD program. I am employed by the company Thor Energy, based in Oslo, Norway, and the research subjects have been chosen to fit the research needs of Thor Energy. For this reason, a commercial focus has been held throughout the work, meaning that primarily aspects of interest to commercial actors in the nuclear field have been studied. The industrial context has also had the consequence that some of the work performed has been documented in the form of patent applications instead of academic journal articles. The groundwork forming the basis for one of the two patent applications is presented herein.

As a part of the research and development strategy of a commercial company, the work presented herein does not have the character of a single well-defined project, but rather a part, limited in time and scope, of a large and long-term research undertaking. For example, the irradiation experiment which has formed a large part of the work performed during the PhD project is at the time of writing still ongoing, and work to expand its scope is in progress. The results presented herein are thus only the first parts of the more comprehensive data set that will ultimately be generated.

There is currently considerable interest for thorium as a nuclear fuel also from parties not directly involved in the field of nuclear technology, and misperceptions are common. For this reason, the thesis begins with an introduction to thorium as a nuclear fuel, aimed towards explaining the basics of the thorium fuel cycle and relating it to comparable uranium utilization schemes.

It is my hope as the author of this thesis that it will be readable and interesting not only within academia but also to readers within the energy industry.

Klara Insulander Björk,
Göteborg 2015.04.07

List of Publications

Paper I

K. Insulander Björk, V. Fhager, and C. Demazière (2011). Comparison of thorium-based fuels with different fissile components in existing boiling water reactors. *Progress in Nuclear Energy* **53**, 618–625
The present author performed all simulations and the main part of the analysis and wrote the manuscript.

Paper II

K. Insulander Björk, S. Mittag, R. Nabbi, A. Rineiski, O. Schitthelm, and B. Vezzoni (2013a). Irradiation of a Thorium-Plutonium rodlet: Experiment and benchmark calculations. *Progress in Nuclear Energy* **66**, 73–79
The present author performed one out of four reported benchmark simulations, participated in the analysis of the results and wrote the manuscript.

Paper III

K. Insulander Björk and V. Fhager (2009). "Comparison of Thorium-Plutonium fuel and MOX fuel for PWRs". *Proceedings of Global 2009*. September 6-11. Paris, France
The present author performed all simulations and the main part of the analysis and wrote the manuscript.

Paper IV

K. Insulander Björk, C. W. Lau, H. Nylén, and U. Sandberg (2013b). Study of Thorium-Plutonium Fuel for Possible Operating Cycle Extension in PWRs. *Science and Technology of Nuclear Installations* **2013**. Paper 867561
The present author performed the lattice simulations, participated in the data analysis and wrote the manuscript.

Paper V

K. Insulander Björk (2013). A BWR fuel assembly design for efficient use of plutonium in thorium-plutonium fuel. *Progress in Nuclear Energy* **65**, 56 –63
The present author performed all simulations and analysis and wrote the manuscript.

Paper VI

K. Insulander Björk, S. S. Drera, J. F. Kelly, C. Vitanza, C. Helsengreen, T. Tverberg, M. Sobieska, B. C. Oberländer, H. Tuomisto, L. Kekkonen, J. Wright, U. Bergmann, and D. P. Mathers (2015). Commercial thorium fuel manufacture and irradiation: Testing (Th,Pu)O_2 and (Th,U)O_2 in the "Seven-Thirty" program. *Annals of Nuclear Energy* **75**, 79–86

The present author wrote the manuscript and is the manager of the described research program, a task which has comprised choosing and procuring materials for irradiation and deciding on instrumentation and irradiation conditions together with the co-authors associated with IFE. The author has also organized meetings collecting advice on the conduction of the research program from the co-authors associated with the collaboration partners Fortum, Westinghouse and NNL, and participated in the analysis of the irradiation data.

Paper VII

K. Insulander Björk and P. Fredriksson (2014). "Development of a fuel performance code for thorium-plutonium fuel". *Proceedings of PHYSOR 2014*. September 28 - October 3. Kyoto, Japan

The present author performed all thermal-mechanical modeling and programming, supervised the student performing the neutronic modeling and programming and wrote the manuscript.

Paper VIII

K. Insulander Björk and L. Kekkonen (2015). Thermal-mechanical performance modelling of thorium-plutonium oxide fuel and comparison with experimental data. Submitted to Journal of Nuclear Materials

The present author performed all simulations and programming, participated in the experiment data analysis and wrote the manuscript.

Other publications related to this thesis:

K. Insulander Björk, V. Fhager, and C. Demazière (2009a). "Method for investigating the applicability of thorium-based fuels in existing BWRs". *Proceedings of ICAPP '09*. May 10-14. Tokyo, Japan

K. Insulander Björk, V. Fhager, and C. Demazière (2009b). "Comparison of thorium-based fuels with different fissile components in existing BWRs". *Proceedings of ICAPP '09*. May 10-14. Tokyo, Japan

K. Insulander Björk, V. Fhager, and C. Demazière (2009c). "Comparison of thorium-based fuels with different fissile components in existing boiling water reactors". *Proceedings of Advances in Nuclear Fuel Management (ANFM) IV*. April 12-15. Hilton Head Island, South Carolina, USA

O. Schitthelm, R. Nabbi, B. Vezzoni, A. Rineiski, S. Mittag, and K. Insulander Björk (2011). *A thorium-plutonium pin benchmark based on the experimental results from the KWO irradiation*. Tech. rep. LWR-DEPUTY D11. Forschungszentrum Jülich

K. Insulander Björk (2012). "Thorium-plutonium fuel for long operating cycles in PWRs - preliminary calculations". *Proceedings of the SAIMM conference on Thorium and Rare Earths*. February 21-22. Cape Town, South Africa

K. Insulander Björk and S. Helmersson (Oct. 30, 2013). "A fuel assembly for a nuclear reactor". Patent application filed by the European Patent Organization.

C. W. Lau, H. Nylén, K. Insulander Björk, and U. Sandberg (2014a). Feasibility Study of 1/3 Thorium-Plutonium Mixed Oxide Core. *Science and Technology of Nuclear Installations* **2014**. Paper 709415

C. W. Lau, V. Dykin, H. Nylén, K. Insulander Björk, and U. Sandberg (2014b). Conceptual study on Axial Offset Fluctuations by Stepwise Power Changes in a Thorium-Plutonium Core to Improve Load-Following Conditions. *Annals of Nuclear Energy* **72**, 84–89

K. Insulander Björk and Ø. Asphjell (Oct. 17, 2014). "Fuel assembly for a nuclear power boiling water reactor". Patent application filed by the European Patent Organization.

S. S. Drera, J. F. Kelly, Ø. Asphjell, and K. Insulander Björk (2014a). "Overview of the Thor Energy thorium fuel development program". *ANS Winter Meeting and Nuclear Technology Expo 2014*. November 9-13. Anaheim, California, USA

S. S. Drera, H. Feinroth, J. F. Kelly, and K. Insulander Björk (2014b). "Development of LWR fuels utilizing thorium dioxide in conjunction with both zirconium-based and SiC cladding". *ANS Winter Meeting and Nuclear Technology Expo 2014*. November 9-13. Anaheim, California, USA

S. S. Drera, J. F. Kelly, Ø. Asphjell, K. Insulander Björk, M. Sobieska, and B. C. Oberländer (2014c). "Ceria-thoria materials testing and pellet manufacturing in preparation for thoria-plutonia Th-MOX LWR fuel production". *2014 Water Reactor Fuel Performance Meeting / Top Fuel / LWR fuel performance meeting*. September 14-17. Sendai, Japan

S. S. Drera, J. F. Kelly, Ø. Asphjell, K. Insulander Björk, M. Sobieska, and B. C. Oberländer (2015). Ceria-thoria pellet manufacturing in preparation for plutonia-thoria LWR fuel production. Submitted to Nuclear Technology

Contents

Nomenclature

Abbreviations

2RPu	Twice recycled plutonium
ABWR	Advanced boiling water reactor
AmPu	RGPu with an ingrowth of americium
BOL	Beginning of life
BWR	Boiling water reactor
CERN	European Center for Nuclear Research
DPC	Doppler power coefficient
EOL	End of life
FGR	Fission gas release
FTC	Fuel temperature (Doppler) coefficient
H/HM ratio	Hydrogen-to-heavy metal ratio
HCS	Hot-to-cold reactivity swing
HEU	High enriched uranium
IFBA	Integral fuel burnable absorber
IFE	The Norwegian Institute for Energy Technology
ITC	Isothermal temperature coefficient
JRC-ITU	Joint Research Center - Institute for Transuranium Elements
LEU	Low enriched uranium
LHGR	Linear heat generation rate
LWR	Light water reactor
MA	Minor actinides
MTC	Moderator temperature coefficient
MWd/kgHM	Mega-Watt days per kilogram of heavy metal (unit of burnup)
NatU	Natural uranium
PIE	Post-irradiation examination
PWR	Pressurised water reactor
RGPu	Reactor grade plutonium
TD	Theoretical density
Th-MOX	Mixed thorium and plutonium oxides
TMOL	Thermal-mechanical operating limit
U-MOX	Mixed uranium and plutonium oxides
UOX	Uranium oxide
USNRC	United States Nuclear Regulatory Commission
VC	Void coefficient
WGPu	Weapon's grade plutonium

Other notation

β_{eff}	Effective delayed neutron fraction
η	Average number of neutrons released per neutron absorbed in the isotope
T_C	Fuel centerline temperature
k_∞	Infinite multiplication factor

Glossary

actinide	Element with atomic number 89–103, e.g. thorium, uranium and plutonium
closed fuel cycle	Fuel cycle in which a major part of the spent fuel is recycled
cross section	Measure of the probability of interaction
epithermal neutron	Neutron with an energy intermediate between *fast* and *thermal*
fast neutron	Neutron with an energy in the order of MeV
fertile	Can be converted to a fissile isotope upon absorption of a neutron
fissile	Can undergo fission upon absorption of a thermal neutron
moderator	Medium that slows down neutrons from fast to thermal energies
neutronic	Related to neutron-induced nuclear reactions
open fuel cycle	Fuel cycle where a minor or no part of the fuel is recycled
thermal neutron	Neutron with an energy of about 0.025 eV

Part I

Extended Summary

CHAPTER 1

Introduction

This chapter describes the objectives and limitations of the work described in this thesis and outlines the contents of the following chapters.

1.1 Objective and limitations

Thorium based nuclear fuel has been subject to much research and a wide range of applications have been proposed. This thesis focuses on thorium based fuel for Light Water Reactors (LWRs), and in particular on two specific thorium oxide containing fuel types. In this context, Boiling Water Reactors (BWRs) and Pressurised Water Reactors (PWRs) are included in the notion of LWRs. The objective of this work is to identify and investigate some of the most important areas where more research is needed for thorium fuel to be considered as an attractive alternative for LWR operators and for the fuel to be licensable for use within the current nuclear regulatory framework. The goal of these investigations is to take some of the steps remaining before thorium based nuclear fuels can be commercially used in LWRs.

Even though the scope has been narrowed down to two fuel types in two closely related reactor types, several aspects will have to be left out of the current work. This thesis focuses on the area of most direct relevance to a reactor operator - the behaviour of the fuel during reactor operation. The first step in the value chain, i.e. thorium mining and thorium oxide powder production, is not investigated here. The subject of

fuel manufacture is very briefly touched upon, since it is a necessary part of realising an experiment where the thermal-mechanical behaviour is assessed. The properties of spent thorium based fuel, in terms of its isotopic content, are also touched upon, but the direct consequences for preliminary storage in spent fuel pools are not evaluated and final storage of all or parts of the spent fuel is only synoptically discussed. Chemical properties and possible reprocessing methods for thorium based fuel are not part of this work.

1.2 Outline of this thesis

The thesis commences with an introduction to thorium as a nuclear fuel, putting it into the context of nuclear fuel cycles. This chapter is written for a reader not familiar with nuclear technology, although some basic knowledge of physical concepts is required. The purpose is to motivate the choice of subject for this work, i.e. to explain why thorium fuel is relevant, and why it is worthwhile to investigate its behaviour in LWRs.

The remaining chapters will demand a certain level of familiarity with nuclear technology from the reader. Chapter 3 describes how the research area was narrowed down to two specific fuel types by scoping studies of the neutronic properties of a number of alternatives. The detailed investigation of the operational behaviour of the chosen fuel types is then presented in the following chapters. The operational behaviour of the fuel can be subdivided into two closely interrelated categories – The *neutronic* behaviour, related to the neutron-induced nuclear reactions taking place inside the reactor and the *thermal-mechanical* performance, i.e. how material properties like thermal expansion and conductivity interact to determine parameters such as the temperature and dimensional changes of the fuel.

Chapter 4 focuses on the neutronic behaviour. One particular area which is pointed out as important for licensing of any fuel type is that its neutronic behaviour can be accurately modeled by dedicated software. An attempt at validation of a particular computer code for neutronic modelling of thorium fuel is described, and the detailed properties of the fuel are then investigated using this software. The thermal-mechanical performance is discussed in Chapter 5. This behaviour has to be assessed experimentally and an experiment designed for this purpose is desribed. Also some results of this experiment are presented, including a comparison with theoretical predictions. Finally, the work is summarised in Chapter 6 and some future work, both ongoing and proposed, is outlined.

CHAPTER 2

Thorium as a Nuclear Fuel - Background

This chapter gives an introduction to the nuclear fuel cycle, and in particular to thorium as a nuclear fuel, based on open literature. The intention is to provide a background for a reader not familiar with nuclear technology.

2.1 History of thorium fuels

The history of thorium use for nuclear applications is almost as old as that of uranium. The possibility to convert thorium into fissile material, from which energy can be extracted, was discovered in 1941, and it was proposed that this should be utilized in the development of nuclear weapons. The relative simplicity and maturity of uranium-based technologies however made thorium a less attractive alternative (Seaborg 1976, 1977, 1978, 1979, 1980). When nuclear development later turned towards more peaceful purposes, research on the use of thorium based nuclear fuels was carried out in parallell with that on uranium, although with less intensity. The main reason for favouring the uranium based alternatives was, at that time, the fact that the still not fully developed breeder reactor concept[1] seemed to favour the use of a uranium based nuclear fuel cycle (Moir and Teller 2005),

[1] Breeder reactors can be described as reactors which produce their own fuel, enabling a closed fuel cycle where spent fuel is reprocessed and the main part of it is re-inserted into the reactor.

see also Section 2.3.1. The use of non-breeding reactors was at that time regarded as a transition phase towards the more sustainable era of breeder reactors. Nevertheless, the feasibility of using thorium based fuel was demonstrated in several research programs (Baumer and Kalinowski 1991; Gottaut and Krüger 1990; Haubenreich and Engel 1970; Price 2012; Walker 1978), most notably in the Shippingport reactor in the USA, where breeding was demonstrated in an LWR under very specifically tailored conditions (Clayton 1993).

Since the seventies, the main rationale for research into thorium based fuel has been to provide an alternative to uranium based fuel, would the uranium resources ever become depleted (Onufriev 1987). Consequently, the intensity of the related research has to some extent followed the uranium price. India has been an exception to this trend because the country has large thorium reserves and limited access to imported uranium[2]. For this reason, thorium has formed an important part of India's nuclear program since 1958 (Bucher 2009). The latest surge in thorium fuel research occurred in connection with the rapid increase in the uranium price in 2007.

At this time, several decades have gone by without the projected transition to large scale deployment of breeder reactors. The primary reasons for this delay have been that new uranium findings have reduced anxiety that these resources would become depleted in the near future, and that breeder reactors are not economical to operate at their current state of development and today's relatively low uranium prices. Meanwhile, non-breeding LWRs have become the dominating reactor type, providing almost 90% of the world's nuclear electricity generating capacity (World Nuclear Association 2015a). In this context, thorium fuel research is increasingly becoming directed towards LWR applications, and to some extent towards heavy water reactor applications in countries where this reactor type is more common (Dekoussar et al. 2005).

Although the uranium resources may be very large, it is recognized that the thorium content of the earth's crust is about three times larger, reflecting its longer half-life. In the long term, this means that the thorium resources can expand the nuclear fuel resource base significantly. Due to the low demand for thorium, no mining activities are being directed towards retrieval of thorium today, but it often occurs as a by-product of rare earth element mining. As a result, large thorium stockpiles are available over ground today. Only the US thorium stockpile of about 1500 metric tonnes (Hedrick 2004) would suffice for roughly 60 years of reactor operation using thorium based fuel.

2.2 Basic nuclear reactions

Naturally occuring thorium has only one isotope, Th-232. This isotope is not *fissile*, meaning that a thermal neutron can not induce fission of a Th-232 nucleus. The probability of different nuclear reactions are quantified in terms of their *cross section*, which depends

[2]India has not signed the nuclear Non-Proliferation Treaty, for which reason uranium trading with India has been restricted. An agreement was however reached in 2008, loosening the restrictions.

on which reaction is considered, which nucleus is involved and on the energy of the incoming neutrons[3]. Thus, the fission cross section of Th-232 is very low in thermal reactors such as LWRs, where the neutrons are predominantly thermal.

The nuclear fission is the primary reaction by which energy is released in a nuclear reactor. It is thus not possible to generate energy directly from thorium in a thermal reactor, but it has first to be converted to a fissile isotope. Such conversion takes place when a thorium nucleus captures a neutron, a reaction which has a comparatively high cross section in a thermal reactor. Nuclei, such as Th-232, which are convertible into fissile nuclei are called *fertile*. The fissile isotope formed from Th-232 is U-233[4]. This isotope is not only fissile, but has also a high neutron yield per neutron absorbed (a parameter commonly denoted η) compared with other fissile isotopes.

2.2.1 Fissile components

The neutrons required for conversion to take place must be made available, and the most practical way to do this is to place the thorium in a nuclear reactor where neutrons are produced by fission reactions. This, of course, demands that not only thorium but also some fissile isotopes are present in the reactor. In the context of LWRs, the practical way to introduce these fissile isotopes is to include them in the fuel by mixing them with the thorium itself or by concentrating them in so-called seed zones, whereas the zones where the thorium resides are referred to as blanket zones. The number of fissile nuclei relative to the fertile nuclei must be high enough to sustain a fission chain reaction, i.e. that the number of neutrons generated by fission is equal to the number of neutrons being consumed, primarily by inducing fission in fissile nuclei or being captured by fertile nuclei.

In practice, there are only a few alternatives available for these fissile nuclei.

One alternative is U-235. Since natural uranium only contains 0.71% of the fissile isotope U-235 (the rest being fertile U-238), natural uranium has to be enriched in U-235 in order to provide sufficient fissile material for both maintaining a fission chain reaction and providing extra neutrons for the conversion of Th-232 into U-233.

Another alternative is to use plutonium. Plutonium does not occur naturally, but can be recovered from spent uranium-based nuclear fuel, where various plutonium isotopes are formed as a result of neutron captures in U-238. One neutron capture in U-238 leads to the formation of Pu-239[5], and consecutive neutron captures lead to formation of the higher plutonium isotopes Pu-240, Pu-241 and Pu-242. Also some Pu-238 is formed from a chain of neutron captures in U-235. Of these isotopes only Pu-239 and Pu-241 are fissile. However, isotopic separation of plutonium is not practiced, so these isotopes always occur together, although in different proportions, depending on the origin of the plutonium.

[3]The term is also used for other reactions than those induced by neutrons.
[4]This reaction goes via the intermediate isotopes Th-233 and Pa-233.
[5]This reaction goes via the intermediate isotopes U-239 and Np-239.

The third alternative is to use U-233, which may be obtained from reprocessing of spent thorium fuel. Since there is no use and reprocessing of thorium on an industrial scale, this option is currently not available except for laboratory or pilot scale operations.

Thorium, uranium and plutonium, together with a number of other heavy elements, are collectively called *actinides*, referring to their placement in the periodic table.

2.2.2 Reaction products

An often mentioned advantage of thorium fuels is the low amounts of long-lived nuclear waste formed during its irradiation. The reason for this is that the most long-lived and strongly radiotoxic isotopes, mainly plutonium, americium and curium, are formed in very low quantities, due to the long series of low cross section neutron captures required to form these heavy actinides from Th-232. This can be compared with the relatively few steps required to form these elements when the mother isotope is U-238. However, some other undesirable isotopes are formed, most importantly U-232, which will be further discussed in Section 2.4.

For a fuel cycle based exclusively on thorium and its fissile daughter product U-233, the radiotoxic inventory of the actinide waste to be disposed of is about a tenth of that pertaining to the closed uranium-plutonium fuel cycle, for the first 10 000 years after disposal (Gruppelaar and Schapira 2000). However, when a fissile component such as plutonium or enriched uranium is needed the matter is much more complicated, since neutron captures in these materials produce the aforementioned heavy actinides.

In addition to the actinides, the spent nuclear fuel also contains the fission products, i.e. the nuclei resulting from fission reactions. The fissioning nucleus generally splits in two fragments, each with an atomic weight approximately half that of the original nucleus[6]. The set of fission products is similar but not identical for different fissile nuclei. The fission products are often higly radioactive and are responsible for the main part of the waste radiotoxicity for the first few hundred years.

2.3 Fuel cycles and reactor types

The practical relevance of the above described nuclear reactions does not become apparent until it is put into a context - a nuclear fuel cycle. The description of a nuclear fuel cycle involves, of course, the nuclear fuel itself, the reactor in which it is used and also the scheme deployed to handle the spent nuclear fuel.

[6]In fact, the fission products very rarely have a mass exactly half that of the fissioning nucleus, but rather slightly above or below, i.e. about 100 or 140 u.

2.3.1 Reactor types

The most basic classification of nuclear reactors refers to the typical energy of the neutrons causing the fission reactions, which broadly divides reactor types into two classes: *Thermal* reactors, in which the neutrons inducing fission typically have relatively low energies (around 0.025 eV) and *fast* reactors, in which the neutrons have high energies (around 1 MeV). These classifications of neutron energies are somewhat arbitrary, but the numbers give an impression of the orders of magnitude. As noted above, the cross section of a nuclear reaction, such as fission or capture, depends on the energy of the neutron inducing it, which makes these two reactor types fundamentally different. However, there is of course a continuum of neutron energy spectra between the two extremes and reactors can also operate with intermediate neutron energy spectra, often referred to as *epithermal*. The energy dependence of the cross sections is different for different nuclei, which is the most fundamental reason for thorium based fuel being different to uranium based fuel.

There are several ways of constructing a thermal reactor but the thermal LWR fuelled with enriched uranium dominates the world of nuclear power production, accounting for 88% of the total capacity. The perhaps most important reason for this dominance is that ordinary water serves excellently both as a coolant, transporting the generated energy from the fuel to ordinary steam turbines, and a neutron *moderator*, slowing down the fast neutrons produced by a fission reaction to thermal energies[7]. This reactor type has the advantage of being comparatively cheap, well proven and relatively easy to operate, and, with the prevailing operation schemes, the disadvantage of using less than one percent of the energy stored in the fuel, leaving large volumes of radioactive waste to be handled.

By allowing the water to boil intensively, its density and thereby its neutron moderating effect can be reduced so that an epithermal spectrum is achieved, which opens a possibility for more efficient fuel usage, especially with thorium fuel. This can not be directly implemented in currently operating LWRs, but given the similarities, a reduced moderation core may be retrofitted into a sufficiently modern LWR such as the Advanced Boiling Water Reactor (ABWR) (Uchikawa et al. 2007).

Fast reactors are devised to make significantly better use of their fuel than thermal reactors do, an improvement mostly attributed to the fact that η is higher for fissions induced by fast neutrons, rendering an excess number of neutrons for conversion of fertile isotopes to fissile. Fast reactors can generally be designed to be breeder reactors creating more fissile material than they consume, i.e. the production of fissile material from fertile is high enough to compensate for the loss of fissile material through fission. An additional feature of fast neutrons is that they can induce fission not only in thermally fissile nuclei, but also in the heavier actinides, reducing the radiological problem that the production of these isotopes constitutes.

In principle, all reactor types may be fuelled with thorium, but some are more suited to thorium fuel than others. A "thorium reactor", as is sometimes discussed, is thus

[7]Heavy water serves the same purpose in heavy water reactors, which stand for another 6.5% of the world's nuclear power production.

not a well-defined concept. It should however be pointed out that U-233 has a high η value in the epithermal spectrum, compared with U-235 and Pu-239, making thorium an advantageous fuel for the reduced moderation epithermal LWRs. This option has been investigated by several researchers (Kim and Downar 2002; Lindley et al. 2014; Shaposhnik et al. 2013), concluding that breeding can indeed be achieved.

The reactor types most commonly referred to as "thorium reactors" are molten salt reactors and accelerator driven systems. Thorium fuelled molten salt reactors have been investigated both in theory (e.g. (Heuer et al. 2014)) and practice (Haubenreich and Engel 1970) and have received much recent attention in China where in 2011 a very large research program was launched, aiming to develop a thorium fuelled molten salt reactor for commercial power production starting in 2032 (World Nuclear Association 2015b). Thorium fuelled accelerator driven systems gained much attention in the 1990's due to Nobel laureate Carlo Rubbia's embracement of the concept and development of the so-called "Energy Amplifier" (Rubbia et al. 1995). Both concepts are promising in that they may provide a means of generating energy from the abundant thorium resources while simultaneously incinerating the long-lived radiotoxic waste generated by today's nuclear reactors. The two concepts can even be combined into an economical and safe system for nuclear waste reduction (Salvatores et al. 2001). The drawback of these technologies is that they are still comparatively immature and much research and development work remains until they are ready for deployment, as indicated by e.g. the projected (and probably quite optimistically so) commercialization time for the Chinese project.

2.3.2 Spent nuclear fuel handling

There are two major strategies for the handling of spent nuclear fuel and different strategies are adopted by different countries. The one adopted in Sweden and some other countries is direct disposal, i.e. the spent nuclear fuel (with over 99% of its potentially recoverable energy remaining) is disposed of, preferably in deep geological repositories, where it must remain separated from the biosphere for several hundreds of thousands of years. The other approach is reprocessing, which involves separating the constituents of the spent fuel, making use of some of them.

Despite the obvious drawbacks of the direct disposal option, reprocessing is not very widely practiced. This is partly due to the high costs connected with reprocessing activities and the manufacture of new fuel from their products, and partly to the perception that separation of material which is to some extent fissile (such as plutonium with its main fissile isotope Pu-239) constitutes a nuclear weapon proliferation risk. However, reprocessing is a necessary tool for improving fuel utilization. Irrespective of which reactor type is used, the fuel will degrade with burnup and will need to be reprocessed if the majority of the fissile isotopes produced are to come to use.

About 3% of the spent LWR fuel consists of fission products. These are often

radioactive, with mostly relatively short half-lifes compared with the actinides[8], and need to be disposed of. The rest of the spent fuel is made up of actinides, which may be re-used in nuclear fuel. Most of the actinide content is just the original fertile isotopes (U-238 or Th-232), which have not undergone any nuclear reaction. In addition, there are the new isotopes generated by neutron captures. These are e.g. U-233 or Pu-239 which may readily be re-used for nuclear fuel manufacture, but especially in the case of uranium based fuel, there are also small quantities of heavier actinides.

Reprocessing of uranium fuel is an established process which is being widely practiced in e.g. France, where the separated plutonium is mixed with natural uranium and re-used in so-called mixed oxide fuel[9], enabling uranium savings of up to 30%. Reprocessing of thorium based fuel has never been practiced on an industrial scale. A process for thorium based fuel recycling, THOREX, was devised already in the 1960's, and continued development is ongoing in India (Das and Bharadwaj 2013). The reprocessing of thorium based oxide fuel (which is the most commonly used chemical form of fuel material) is more difficult than that of uranium fuel, due to the fact that thorium dioxide is relatively chemically inert and hence difficult to dissolve.

Whereas reprocessing is optional in the current fuel management scheme, it is crucial for a breeding cycle to make sense, since it is required in order to make use of the produced fissile isotopes. A fuel cycle including reactors which produce enough fissile material to be self-sustaining and reprocessing of the spent fuel with recycling of the major part of the spent fuel is referred to as a *closed fuel cycle*. For the case of the uranium based fuel cycle, despite many decades of research, there are some unresolved technical issues with manufacturing and using fuel containing the complete set of actinides produced in this cycle. There are conceptual designs of closed fuel cycle systems in which all actinides are recycled, but they are still under development, see e.g. Somers (2011); Ikeda et al. (2014).

It is thus clear that a breeding reactor combined with reprocessing is required in order to achieve the closed, self-sustaining, low-waste producing thorium fuel cycle which is often referred to by thorium proponents. Although the technology to reprocess thorium based fuel is still immature, it is ultimately possible to establish.

2.4 Safety and security aspects

In addition to the central aspects of the nuclear fuel cycle - reactors and reprocessing - there are additional aspects that need to be discussed, related to safety and security concerns connected with nuclear activities.

[8]There are some long-lived exceptions to this, with the consequence that the radioactivity of the fission products as a collective decreases very rapidly for the first few hundred years and then settles at a low level, remaining there for a few million years.

[9]The term U-MOX will be used in this work to distinguish mixed uranium and plutonium oxides from mixed thorium and plutonium oxides (Th-MOX)

2.4.1 Safe reactor operation

The specific nuclear properties of the isotopes central to the thorium cycle, U-233 and Th-232, makes the fuel different from uranium based fuel in terms of reactor operation. Most importantly, the differences affect how the reactor responds to changes in operation conditions, i.e. the dynamic characteristics of the reactor. This directly affects the possibilities to safely operate a thorium fuelled reactor.

The dynamic characteristics are often described in terms of reactivity coefficients, i.e. how the reactivity changes when a certain parameter such as the fuel temperatures changes. The efficiency of the different mechanisms used to control the reactor, such as control rods, are also affected by the nuclear properties of the fuel. How the reactivity coefficients and the control mechanism efficiency change when torium fuel is introduced depends strongly on which fissile component is used.

Another important aspect for reactor safety is the delayed neutron fraction, i.e. the fraction of the total number of neutrons emitted with some delay after the fission event. This parameter determines how rapidly the reactor responds to changes in operational conditions. It depends on which isotope is fissioning, and is lower for U-233 than for U-235 and Pu-239, meaning that a reactor in which U-233 is the predominant isotope responds more rapidly and is hence more diffcult to control. The importance of this of course depends on how much U-233 is actually present in the core.

All of these aspects are much more thoroughly discussed in Chapters 3 and 4.

2.4.2 Radiotoxicity issues

One safety related topic has already been touched upon: The radiotoxicity of the materials to be handled. One part of this problem is the final waste to be disposed of. In the case of a closed fuel cycle, this waste consists of fission products and some actinides which inevitably are lost to the waste stream during reprocessing. The losses are usually of the order of 0.1 - 1%. The fission products dominate the waste radiotoxicity for the first few hundred years, after which the actinides take over as the major component. As stated earlier, thorium has an advantage of an order of magnitude lower radiotoxicity of the actinide waste up to about 10 000 years (Gruppelaar and Schapira 2000). However, it is important to note that thorium based fuel adds a complication to reprocessing and manufacture of fuel from reprocessed material. The reason is that a small quantity of radioactive U-232 is produced during irradiation, whose decay chain results in emission of high-energetic γ-rays (in particular from Tl-208). This makes it necessary to deploy remote-controlled manufacture of recycled fuel, whereas glove-box handling is sufficient in the uranium case. This makes recycled thorium fuel manufacture more expensive.

For fuel cycles with no reprocessing, the radiotoxicity of (and heat production in) spent thorium fuel depends heavily on the fissile component. The addition of a fissile

component inevitably brings U-238 or heavier actinides into the fuel cycle[10], meaning that long-lived radiotoxic isotopes are nevertheless produced in the fuel, although not primarily from the thorium itself.

In case of direct disposal of spent fuel, thorium oxide fuel also has an advantage due to its chemical inertness. This means that the thorium fuel matrix, which effectively encapsulates most of the fission products and heavy actinides, will remain intact for a longer time than a uranium matrix, even if groundwater would permeate to the fuel in the deep geological repository (Greneche and Chhor 2012; Gruppelaar and Schapira 2000).

Irrespective of whether the fuel is reprocessed or taken to final disposal, the emission of high energy γ-radiation adds a complication to the handling of spent fuel already at the reactor site. Additional shielding will be needed already at fuel discharge and intermediate storage (Ade et al. 2014).

2.4.3 Proliferation concerns

Resistance to nuclear proliferation, i.e. the proliferation of nuclear weapon related materials and technology, is often mentioned as a possible advantage of the thorium fuel cycle. This perception comes mostly from the fact that no plutonium is produced. In fact, if plutonium is used as the fissile component of thorium fuel, the plutonium is efficiently destroyed, which will also be discussed further in Chapter 4. This method of plutonium incineration has been judged to offer non-proliferation benefits (Gruppelaar and Schapira 2000; Trellue et al. 2011).

Although no plutonium is produced from thorium, so is U-233, from which nuclear weapons can also be manufactured if the fuel is reprocessed. It can however be argued that this uranium is extremely difficult to use for bomb making due to its contamination with γ-ray producing U-232, and it has also been suggested to "denature" the U-233 by adding some U-238 to the thorium fuel. It would in principle be possible to obtain pure U-233 by separation of its precursor Pa-233. This is not practical for spent solid fuel, but in principle possible for molten salt reactors in which the liquid fuel is continuously reprocessed during reactor operation. Considering all these aspects, it can nevertheless be concluded that the thorium cycle causes slightly less proliferation concerns than the uranium cycle (Dekoussar et al. 2005; Pellaud 2013).

2.5 Material properties

Also the material properties of thorium distinguishes it from uranium. The chemical form of uranium predominantly used in nuclear fuels is uranium dioxide, UO_2, and it

[10]This can almost completely be avoided by using high-enriched uranium (Rose et al. 2011), which however adds significant concerns related to nuclear weapon proliferation and enrichment costs.

is assumed that thorium will be used in the same way, i.e. as thorium dioxide, ThO_2. ThO_2 has some features which are beneficial for its use as a nuclear fuel. It has a much higher melting point compared with that of UO_2, a lower thermal expansion and a higher thermal conductivity. In particular the thermal conductivity is important, since it leads to a lower inner fuel temperature. The fuel temperature in turn drives many processes deteriorating the performance of the fuel. These properties are of course affected by the addition of the necessary fissile component (also assumed to be in oxide form) and will be more thoroughly discussed in Chapter 5. To a first approximation, though, these beneficial properties makes it reasonable to assume that ThO_2-based fuel should be able to operate at higher power levels and/or to a higher *burnup*, i.e. to a higher amount of energy released per unit mass of the fuel material.

2.6 Commercial viability of thorium fuel

With this background information on the historical, physical, technical, safety and security related aspects of thorium as a nuclear fuel, the utility of using it commercially can now be discussed.

It is clear that a long-term sustainable nuclear fuel cycle must involve breeding reactors and reprocessing of spent fuel. It seems plausible that a transition to such a cycle must eventually occur, although several decades of research on closed uranium based fuel cycles have not yet resulted in an economically competitive alternative. It can however be stated, based on the above, that thorium could potentially provide a faster and cheaper way to such a cycle, through deployment of the mentioned thorium fuelled reduced moderation LWRs. Due to the similarity to existing LWR technology, the transition to such systems would be evolutionary rather than revolutionary, moving gradually to higher conversion systems. In addition, such a fuel cycle would lead to lower production of radiotoxic waste compared with its uranium counterpart, provide some non-proliferation benefits and make use of an abundant resource.

From the economical perspective, it is clear that the establishment of a closed fuel cycle based on either thorium or uranium requires very large investment in both research and fuel cycle infrastructure. The question then becomes how to realise the first step in this evolutionary development, i.e. how thorium can offer economical advantages in the current reality of the nuclear industry, while simultaneously working towards the goal of a self sustaining thorium fuel cycle by gaining operational experience and also by generating U-233 for the next generations of thorium fuelled reactors.

As mentioned, thorium can be used in almost any reactor type, including existing LWRs. Given the dominance of LWRs in current nuclear power production and the potential for a transition to a breeding cycle using closely related technology, the development of thorium based fuel for LWRs makes sense both from the perspective of near-term commercial viability and eventual sustainability. Thus, the objective of the following chapters is to investigate the potential advantages of thorium fuel usage in currently operating LWRs.

2.7 Research needs

The United States Nuclear Regulatory Commission (USNRC) released a report in 2014 titled "Safety and Regulatory Issues of the Thorium Fuel Cycle" (Ade et al. 2014). The regulatory framework formulated and enforced by the USNRC is the reference framework in many countries, at least in the western world, and the regulatory framework in many countries is similar to that in the US. This report is thus a useful starting point for outlining the research needs.

The report states that the most likely near-term application of thorium in the US is in currently operating light water reactors. The report then outlines the research needs that are most important from a regulatory point of view. The related issues have been divided into six categories: Physical properties, nuclear data, fuel performance, reactor safety, front-end issues and back-end issues. The physical properties (thermal conductivity, melting point etc.) of thorium oxide and mixtures with uranium or plutonium oxides are partially known and research is ongoing, in particular at the Institute for Transuranium elements (ITU), see e.g. Cozzo et al. (2011), Vălu et al. (2014) and Böhler et al. (2015). The nuclear data relevant for thorium fuel cycles are generally known, although not with the same high level of confidence as the corresponding data related to the uranium fuel cycle. Measurement of nuclear data is a discipline of basic research generally undertaken by universities and research institutes with access to large and expensive machinery, and research is being carried out at e.g. CERN (Belloni et al. 2012). As previously noted, front- and back-end issues are not considered in this work.

In the field of fuel performance, the USNRC report states that "it is likely that fuel irradiation experiments would be needed to generate and validate fuel performance data and codes". Such an irradiation experiment forms a major part of the current work. With respect to reactor safety, the report comprises a computational assessment of neutronic safety parameters, and concludes that "Further study should be initiated when realistic fuel design information becomes available" and that "Whole core analyses are required". This work aims to find such realistic fuel design information, with the interpretation that a realistic fuel design is a design which is acceptable both from economic and safety perpectives. A whole core analysis is also performed. In addition, it is noted that these analyses must be carried out with validated software, an issue which is also adressed in this work.

Neutronic analysis of thorium based nuclear fuels in existing LWRs has indeed been carried out in several contexts. Most of these propose unconventional features, such as heterogeneous fuel assemblies with partly metallic fuel (Galperin et al. 2000), micro-heterogeneous fuel (Shwageraus et al. 2004a) or annular fuel (Caner and Dugan 2000). Many of these features are primarily intended to improve conversion of Th-232 to U-233, which is beneficial for the fuel economy but adds an aspect that could be considered challenging by both reactor operators and regulatory bodies. With the view to make the initial step of thorium fuel adoption as small as possible, the proposed solution should only entail replacement of uranium with thorium while not altering any other parameters.

Other proposed concepts are based completely or partly on U-233 as the fissile component (Baldova et al. 2014a,b; Rose et al. 2011), an alternative which is currently not available.

Quite a few researchers have simulated PWRs with homogeneous thorium oxide fuel with U-235 or Pu-239 as the fissile isotope (Ade et al. 2014; Arkhipov 2000; Gruppelaar and Schapira 2000; Herring et al. 2001; Puill 2002). The reports are basically similar in that they conclude that the introduction of thorium in current PWRs is indeed feasible, that thorium-plutonium mixed oxide fuel (Th-MOX) is similar to uranium-plutonium mixed oxide fuel (U-MOX) and that for unaltered reactor operation conditions, replacing some of the uranium with thorium does not lead to U-235 savings.

Only few studies have been made of thorium fuelled BWRs: Núñez-Carrera et al. (2008) studied thorium fuelled BWRs with a heterogeneous fuel assembly concept involving metallic fuel, and some similar studies have been published after the publication of the papers included in this thesis by Galahom et al. (2015). This imbalance is probably due to the relative dominance of PWRs over BWRs (273 PWRs and 81 BWRs were in operation in 2015 (World Nuclear Association 2015a)). However, BWRs could potentially offer an advantage due to their slightly faster spectrum, which emphasizes the good breeding properties of thorium fuels.

CHAPTER 3

Choice of Fuel Types

This chapter describes scoping studies providing a basis for the continued work, by investigating the basic properties of a range of thorium-containing fuel types. The aim is to determine which fuel types are particularly promising and which have inherent properties making them ineligible for further study. This chapter is based on the content of Paper I and calculations made in preparation for a patent application (Insulander Björk and Asphjell 2014).

3.1 Comparison of fissile components - Paper I

The basic properties of four different thorium based fuel types were investigated using CASMO-5 (J. Rhodes et al. 2007), a multi-group transport theory code for depletion simulations of nuclear fuel assemblies in a two-dimensional geometry. The suitability of this code for the purpose will be further discussed in Section 4.1. The infinite multiplication factor k_∞ of the fuel assembly can be calculated for different burnup and operational conditions, so information on the fuel behaviour can be deduced from these simulations, which can be used for comparing the basic properties of different fuel types. The operational parameters were modeled on the Swedish BWR Forsmark 3 and the fuel assembly design chosen for this study was GE14-N, a modern design used in Forsmark 3 as well as in many other BWRs worldwide. The operational parameters can be found in Paper I.

As explained in Section 2.2.1, a fissile component must be added to the thorium in order to make a fuel capable of sustaining a fission chain reaction. The four thorium-containing fuel types investigated in this study all consist mainly of thorium and are distinguished by the respective fissile component used, namely:

- *Reactor grade plutonium (RGPu)* - plutonium with an isotope vector typical for reprocessed LWR fuel with a burnup of 40-50 MWd/kgHM. The one used in this study is 2.5% Pu-238, 54.2% Pu-239, 23.8% Pu-248, 12.6% Pu-241 and 6.9% Pu-242. The feature distinguishing RGPu from weapons' grade plutonium (WGPu) is that it has a much lower content of the fissile isotope Pu-239, which makes it unsuitable for nuclear weapon production.
- *Low Enriched Uranium (LEU)* - uranium enriched to 20% in U-235. Enrichments up to 20% count as low, whereas uranium with higher enrichments (High Enriched Uranium, HEU) is subject to much harder restrictions due to proliferation concerns.
- *U-233* - Isotopically pure U-233 is not available in practice, but this alternative is included to understand the basic characteristics of this fissile isotope and the consequences of its introduction in the fuel in relation to the alternatives.
- *Recovered uranium and RGPu (UPu)* - The uranium recovered from one spent Th+RGPu-assembly (containing 89% U-233 and 11% U-234) is used for one assembly of this type and combined with RGPu to provide sufficient reactivity.

In addition to the four thorium based fuel types, two reference cases were also included in the study - one uranium oxide (UOX) fuel assembly and one assembly with mixed uranium and plutonium oxides (U-MOX). In order to compare equivalent fuel assemblies, the amount of the fissile component in each assembly was adjusted to yield equal amounts of energy, determined by the linear reactivity model as explained in Paper I. Reactivity coefficients were estimated by perturbation calculations and the control rod worth by comparing k_∞ with or without control rods inserted in the simulated infinite lattice. Pin power distributions, delayed neutron fractions and decay heat are routinely calculated by CASMO-5.

3.1.1 Results

The composition of each fuel type at the beginning of life (BOL), as determined by the condition of equal energy yield, is given in Table 3.1, in terms of kg per fuel assembly. The plutonium and U-233 content of each fuel type is also given at the end of life (EOL). Already this table offers some information relevant for the choice of fuel type. It is seen that the total U-235 mass needed, and therewith the natural uranium requirements, is higher for Th+LEU than for the UOX reference. The U-MOX and Th+RGPu cases can be compared from the point of view of plutonium destruction. It is then clear that Th-RGPu offers an advantage both in terms of total plutonium mass reduction (8.3 kg for

Th+RGPu versus 4.6 kg for U-MOX) and in terms of the fissile fraction of the discharged plutonium (39% for Th+RGPu versus 48% for U-MOX). The Th+UPu case also offers a small advantage in terms of plutonium destruction compared with U-MOX. It is also clear that the system is far from breeding, given the large reduction of the U-233 mass for the Th+U-233 case. The Th+UPu case is close to self-sustaining in terms of U-233, but requires the addition of RGPu.

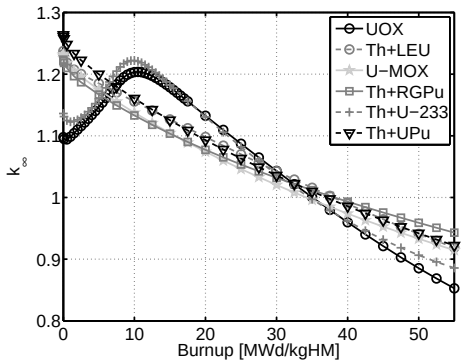

Figure 3.1: The infinite multiplication factor for the investigated fuel types.

The multiplication factor, plotted in Figure 3.1 in itself is not directly relevant to reactor operation, but offers some insight into the fundamental processes leading to the differences between the fuel types. Firstly, it is clear that the slope of the k_∞ curve is steeper for the UOX and Th+U-233 cases, compared with the other cases. For this reason, the burnable absorber gadolinium is routinely used in UOX fuel, and it was chosen to use this also for the Th+U-233 case. The use of gadolinium causes a gradually decreasing suppression of k_∞ at the beginning of life in these two cases, so that k_∞ is actually increasing while the gadolinium is burnt out.

The differences in the rates of decrease of k_∞ depends on the balance between consumption of fissile material through fission and production of fissile material through the neutron captures in the fertile material. Efficient conversion gives a continuous buildup of new fissile isotopes, sustaining k_∞ at a higher level. Conversion is improved in the Th+RGPu and U-MOX cases by the presence of Pu-240, having a capture cross section two orders of magnitude larger than U-238. Also in the Th+LEU case, conversion is improved by the simultaneous presence of Th-232 and U-238, providing a double set of absorption resonances and in addition the high thermal absorption in Th-232.

Reactivity coefficients, control rod worth, delayed neutron fractions and short term decay heat are central to the safe operation of a reactor. The most important reactivity coefficients for a BWR is the fuel temperature coefficient (FTC), the moderator temperature coefficient (MTC), the void coefficient (VC) and the isothermal temperature coefficient (ITC). All these need to be negative in order to avoid positive feedback loops

leading to uncontrollable reactivity increase. They may however not be too negative, i.e. negative and too large in amplitude, since this may have an adverse impact in some transient scenarios. The estimates based on lattice simulations are only indicative for the fuel behaviour, and are in this context only used for comparison of the different cases. Full core simulations of cases similar to the Th+U-233, Th+LEU and Th+RGPu cases have been carried out in a previous study (Insulander Björk 2008), providing a useful reference. The safety parameters, also listed in Table 3.1, were similar for all cases, and acceptable as far as can be judged based on the present simulations, with some important exceptions:

- The MTC and VC are positive for the Th+U-233 case. The reason for this is further discussed in Section 4.3. Full core simulations, taking e.g. the presence of control rods and leakage effects into account, show a negative VC for Th+U-233 under normal operational conditions, but significantly closer to zero than the reference, yielding unacceptably low margins.
- The FTC is negative and very large in amplitude in the Th+LEU case, because the Doppler effect, i.e. the broadening of the capture resonances with increased temperature, is amplified by the simultaneous presence of Th-232 and U-238.
- The control rod worth (CRW) is strongly reduced for the plutonium containing fuel types U-MOX and Th+RGPu and somewhat reduced also for Th+UPu. This is a known phenomenon from U-MOX usage and is attributed to the hardening of the neutron spectrum caused by the presence of the large thermal absorption resonances in several of the plutonium isotopes.
- The effective delayed neutron fraction β_{eff} for the U-MOX, Th+RGPu, Th+U-233 and Th+UPu cases is significantly lower than for the UOX reference. This mirrors directly the lower delayed neutron yields for fission of U-233 and Pu-239 compared with U-235. U-MOX fuel is routinely used in LWRs and full U-MOX core loadings are considered for most Generation III+ LWRs, so this is not expected to be an insurmountable problem since the delayed neutron fractions calculated for the thorium fuel types are not significantly lower than that for U-MOX fuel.

3.1.2 Conclusions

Based on these results and the background information provided in the previous chapter, an informed choice can be made about which fuel type should be subject to continued research.

The Th+LEU option adds many complications compared with standard UOX fuel. More enrichment work is needed to enrich uranium up to 20%, and in addition, commercial fuel factories are in general not licensed to handle uranium with an enrichment above 5%. The manufacture of a two-component fuel can also be expected to be more expensive

Table 3.1: Key safety parameters and balance of the most relevant elements.

Fuel type	UOX	Th+LEU	U-MOX	Th+RGPu	Th+U-233	Th+UPu
Fuel composition data, in terms of kg per fuel assembly						
Initial U-235	7.2	8.0	0.5	-	-	-
Initial RGPu	-	-	13.2	15.6	-	8.3
Initial U-233	-	-	-	-	5.8	2.9
Final total Pu	1.8	0.56	8.6	7.3	≈ 0	2.7
Final fissile Pu	1.0	0.34	4.1	2.8	-	0.76
Final U-233	-	1.8	-	2.1	2.5	2.8
Burnup averaged safety parameters						
MTC [pcm/K]	-10.2	-7.5	-17.3	-7.1	14.3	-2.6
VC [pcm/%void]	-56	-50	-52	-38	6	-34
FTC [pcm/K]	-2.4	-3.6	-2.5	-2.9	-2.7	-3.0
CRW [arbitrary]	20.0	20.1	17.2	17.2	21.5	18.2
β_{eff} [pcm]	555	547	395	357	333	360

than standard UOX fabrication. This alternative is in practice only justifiable if it can offer natural uranium savings, which it according to this study does not.

The alternatives involving U-233, i.e. Th+U-233 and Th+UPu, are, as stated earlier, not practically available at present. There are however two conclusions to be drawn from this study. The fact that the VC and MTC of the Th+U-233 fuel is positive means that it has a higher reactivity at lower moderation. This indicates that the fuel is over-moderated also in a BWR and would perform better at lower moderation, which is in line with previous studies e.g. by Kim and Downar (2002). For the normal BWR operational conditions simulated, it is clearly far from being self-sustaining, given that the U-233 content decreases during life even with the initial addition of RGPu.

This leaves us with the option of RGPu as the fissile component. The present study does indeed not indicate any showstoppers, but rather a big similarity with U-MOX. Although RGPu was the plutonium type investigated in this work, other plutonium vectors may also be considered, such as for example WGPu, which is becoming available when nuclear weapons are decommissioned. Having excluded the other alternatives involving mixtures of thorium oxide with uranium oxide, the mixture of thorium oxide with plutonium oxide will in the following be referred to as Th-MOX, to underline its analogy with U-MOX.

Th-MOX as an option for management of plutonium (and other transuranic elements) has been considered many times before as a means to reduce the large and growing stockpiles of these elements resulting from the operation of the current generation of uranium fuelled LWRs. A comprehensive report on the potential of this concept in PWRs was compiled by Gruppelaar and Schapira (2000), and it has also been investigated by other researchers (Dziadosz et al. 2004; Shwageraus et al. 2004b). These studies show,

just like the BWR-calculations presented here, that Th-MOX offers clear benefits over U-MOX in terms of plutonium destruction. The reason for this is that new plutonium is generated from the U-238 mixed with the plutonium in U-MOX fuel, whereas no new plutonium is produced when Th-232 is the fertile component. As mentioned, the fissile isotope U-233 is produced instead, but the Th-MOX concept nevertheless has some merits over U-MOX from a non-proliferation point of view (Trellue et al. 2011).

Seen from another viewpoint: if the long term goal of thorium fuel usage is deployment of breeding, thorium-fuelled reduced-moderation LWRs, U-233 should be viewed as an asset rather than a proliferation concern. With this perspective, plutonium as the fissile component offers another advantage over LEU in that no U-238 is present in the fuel that would make the uranium isotope vector unsuitable for further use as a fissile component in this context.

However, both the perspectives of proliferation safety and long-term nuclear strategy are relevant rather on national or global level, but not so much for nuclear reactor operators. There is of course a possibility that a national-level decision will be made in some country to deploy thorium based fuels for reasons of e.g sustainability, proliferation, waste management or resource self-sufficiency (as has been done in India), but a shorter way to adoption of thorium based fuel may be to appeal directly to the reactor operators. In order to do so, it must first of course be demonstrated that Th-MOX indeed provides an advantage, and an advantage than can be converted into an economic benefit for the operator, while in no way compromising the safe operation of the reactor. The advantage must also, in most situations, be not only over U-MOX but also over UOX fuel.

Showing an advantage is however not sufficient. It must also be possible to provide a very high degree of confidence in any predictions made regarding the fuel behaviour. This is a big challenge. The very small experience base regarding thorium fuel operation is probably one of its largest drawbacks, which has followed it since the uranium cycle got a headstart of a few years in the 1950's. The following chapters will deal with both potential advantages of Th-MOX, and with a few steps taken towards providing the required level of confidence.

3.2 Thorium as an additive to uranium fuel

Although the simulations described above seemed to disfavour the combination of thorium with LEU, another promising option for thorium usage together with uranium in PWRs was proposed by Lau et al. (2012). This option involves only a minor fraction of thorium, so low that the uranium enrichment does not need to exceed the 5%-limit held by commercial fuel manufacture plants. It was found that in a fuel assembly operated under typical operational conditions in the Swedish reactor Ringhals 3, a thorium fraction of 7% would allow for similar discharge burnup as the currently used UOX assemblies in that reactor. Full core simulations (Lau et al. 2013; Lau et al. 2014c) indicated some promising features, such as improved core stability and favourable power distributions. The original intention

was to utilize thorium's absorbing properties to reduce the need for burnable absorbers, a goal which was indeed reached. However, it was also found that the natural uranium requirements were higher than for the pure UOX alternative, and also the enrichment costs would increase due to the higher enrichment level required.

In spite of the results with respect to the economic prospects of this concept being discouraging, it was clear that the margins to becoming economically feasible were much smaller than for the obviously un-economical Th-LEU case described in the previous section. It was also recognized that this concept would constitute an even smaller first step in the evolutionary approach to thorium fuel adoption. For these reasons, simulations were performed for a similar concept for usage in BWRs. Due to the status of thorium as an additive in this concept, it is hereafter referred to as thorium-additive fuel or Th-Add for short. The initial investigations by Lau et al. (2012) also aroused enough interest in the concept so that it was decided to include two rods of a representative material, i.e. UO_2 with with 7% ThO_2, in the irradiation experiment described in Chapter 5.

The neutronic calculations performed have not been reported in an academic publication. Instead, a patent application was filed with the European Patent Organization. Being written for a legal rather than an academic context, the patent application is not suitable for inclusion in this thesis.

3.2.1 Assembly design with thorium additive

Nuclear fuel design in a BWR is a complicated matter, as compared with the PWR case. Since cruciform control rods are inserted between the fuel assemblies, there are large water gaps to acommodate for these, giving an uneven moderation in the assembly. Due to this fact, the enrichment must be low in the well-moderated rods closest to the water gaps to prevent high power peaks there, whereas the less moderated rods must have a higher enrichment to be able to sustain their power share. In the PWR assembly proposed by Lau et al., all rods had the same enrichment and the same thorium fraction (7%), a design which cannot be used in a BWR assembly. Instead, the design strategy was to start from a reference BWR fuel assembly design with only UOX fuel with varying enrichments, let the U-235 fraction remain unchanged and thorium only introduced to replace some of the U-238. In practice, this could be done by enriching all uranium up to e.g. 4.95%[1] and mixing it with thorium in different fractions. In a BWR assembly, the enrichment typically varies from about 1% in the most well-moderated areas of the fuel assembly to 4.95% in the least well-moderated areas. In the rods which have a 4.95% enrichment in the reference UOX assembly, no thorium can be introduced without lowering the U-235 fraction and hence the reactivity. In the rods with a lower enrichment in the reference UOX assembly, however, the corresponding rods in a Th-Add assembly may contain some thorium while maintaining the same U-235 fraction and still using uranium with an enrichment below the maximum allowed 4.95%. The maximum allowable thorium fraction

[1] Commercial fuel factories are designed and licensed for handling uranium with a maximum enrichment of 5%, and a margin of 0.05 percentage points to the maximum is usually held.

f_{Th} for a rod with the desired U-235 fraction $x\%$ can be calculated as $f_{\mathrm{Th}} = 1 - \frac{x}{4.95}$.

There are some exceptions to be made to this simple recipe. Firstly, there exists no experience at all with mixtures of thorium, uranium and gadolinium oxides, and their properties would be difficult to predict even theoretically. In order to avoid the introduction of such an exotic mixture, it was chosen to let the burnable absorber rods consist of only gadolinium and uranium oxides. Secondly, preliminary simulations showed that high thorium fractions (over 40%) in the well-moderated corner rods resulted in too efficient breeding of U-233, resulting in very high peaking factors towards the end of life. A lower thorium content was therefore used in these rods, and the uranium enrichment consequently reduced.

Just as for the PWR concept investigated by Lau et al., the need for burnable absorbers is quite significantly reduced by the introduction of thorium (a reduction of 20 - 30% seems possible, depending on the application). The most important effect of this is that the number of burnable absorber rods can be reduced. This makes it easier to achieve low power peaking factors early in the life of the fuel assembly, since the number of rods running at a low power level due to their burnable absorber content is reduced, so that more rods can share the power load. Also the fraction of gadolinium in the remaining burnable absorber rods could be reduced. Finally, the power peaking factor could be further reduced by altering the thorium content in some of the rods slightly, optimising the design. All these considerations resulted in a fuel assembly design for which a number of preliminary studies have been carried out, comparing it with a reference assembly with an equal content of U-235. Preliminary full core simulations have also been carried out.

3.2.2 Neutronic properties of the Th-Add BWR fuel

The k_∞ curve plotted in Figure 3.2 already gives the key to the benefits of the Th-Add concept. Most importantly, it is noted that the lifetime energy yield of this assembly, as calculated by the methodology outlined in Paper I, is equal to that of the UOX reference. This means that the addition of thorium in this case does not increase the U-235 requirements.

The presence of thorium in the fuel makes the k_∞ curve slightly more level, which enables gadolinium savings. As can be seen, the initial period of suppressed reactivity is shorter for the Th-Add fuel than for the reference, and the peak k_∞ is still lower. The reactivity cost of the thorium at the beginning of life is paid back towards the end of life, where the reactivity remains at a higher level. This feature gives a smaller reactivity difference between the most and least reactive fuel assemblies, which is beneficial for the power balance in the core. This will reduce power peaking in the core as a whole, and also improve the shutdown margin (SDM), since clusters of highly reactive fuel can be avoided.

The effect of the reduced gadolinium requirements is also seen in the assembly power peaking factors, plotted in Figure 3.3. The power peaking is significantly reduced

throughout the lifetime of the fuel, except for the very last period when it increases because of one of the well-moderated corner pins gaining in power due to U-233 breeding, despite having a reduced initial Th-232 content.

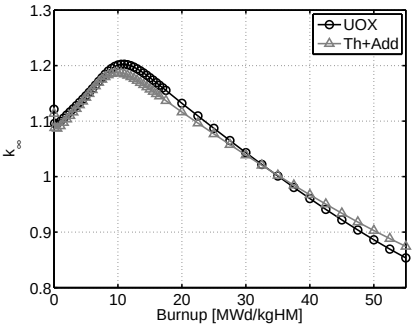

Figure 3.2: k_∞ development with burnup for Th-Add and a reference UOX fuel assembly.

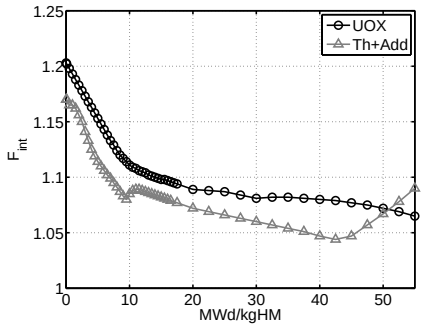

Figure 3.3: The assembly internal power peaking factor.

The core loading patterns and control rod sequences used in the full core simulations have not been optimised, for which reason the absolute values of shutdown margins and power peaking factors are not within acceptable ranges. Also, an unrepresentatively long coast-down period had to be modeled at the end of the cycle for the fuel to reach the target discharge burnup, also due to the loading scheme not being optimised. However, since identical core loading patterns have been used for both fuel types, relevant comparisons can nevertheless be made. The achievable cycle length was slightly shorter for the Th-Add core. However, the difference (5 days) is small enough to be rectifiable with only slight changes of the loading pattern. It can be expected that the larger thermal margins provided by the Th-Add fuel can allow for more "agressive" loading patterns, i.e. where more fresh fuel is loaded centrally in the core, reducing neutron leakage.[2]

Figure 3.4 shows the peak linear heat generation rate for the two cores, which is lower for the Th-Add case throughout the cycle. The shutdown margin is negative for both cores due to the non-optimised loading pattern, but significantly better for the Th-Add case. The delayed neutron fraction is very slightly reduced by the addition of thorium, which was expected since the in-bred U-233 only stands for about 7% of the fissions in the Th-Add core and has only a slightly lower delayed neutron yield than Pu-239. The control rod worth, as estimated from the lattice simulations, is also insignificantly affected. The VC and MTC are virtually unaffected, whereas the amplitude of the (negative) FTC, shown in Figure 3.5, is increased by about 17% by the thorium additive.

[2]Preliminary studies for a currently operating BWR show that this is indeed the case, and that the total costs of uranium and enrichment can indeed be slightly reduced by use of Th-Add fuel.

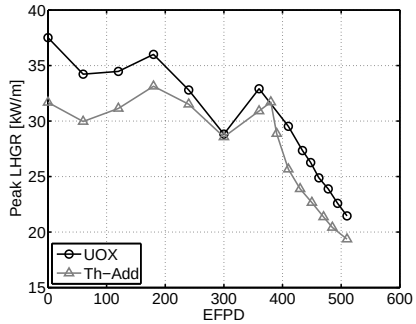

Figure 3.4: The peak LHGR in the Th-Add and reference UOX cores.

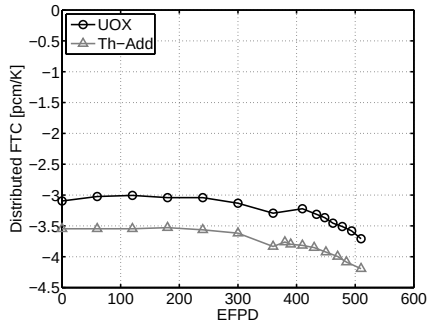

Figure 3.5: The FTC in the Th-Add and reference UOX cores.

CHAPTER 4

Neutronic Properties of Thorium-Plutonium Fuel

This chapter describes the aspects of the neutronic code CASMO-5 related to simulation of Th-MOX fuel and a benchmark study for Th-MOX modelling. Thereafter, studies performed with CASMO-5 and the closely related code CASMO-4 are described. The first two studies address the possible advantages of Th-MOX usage in PWRs, and the objective of the third study is to investigate possible improvements in performance of Th-MOX fuel for BWRs. This chapter is based mainly on the content of Papers II - V.

4.1 Th-MOX modelling with CASMO-5

For detailed studies of the neutronic behaviour of Th-MOX fuel, it is important to investigate the applicability of the software used for this purpose. Due to its widespread use in the nuclear industry, the neutronic simulation code CASMO has been used. Most simulations have been carried out using CASMO-5 whereas the older version CASMO-4E was used for the simulations performed for Paper IV, since this is the code used in normal operation of the simulated reactor. The specificities of CASMO-5 are outlined below whereas the differences to CASMO-4E are briefly discussed in Section 4.2.

Validation is not only necessary in order to make sure that the calculations carried out in this work are relevant. It is also a prerequisite for loading of this fuel in a commercial

reactor. A regulatory body will generally demand that the in-core neutronic behaviour of a fuel can be predicted with "sufficient" accuracy. What is regarded as sufficient is generally different between different countries, having different regulatory systems, and also depends on how much fuel will be loaded, i.e. whether only a few rods, a small number of assemblies or partial or full core loadings are being considered. Whereas the benchmarking carried out within this work is certainly not sufficient for full core loadings, it may be sufficient for loading of lead test rods.

In general, Th-MOX and U-MOX fuels behave similarly, compared with UOX fuel, as concluded by several studies performed with different neutronic software (e.g. Ade et al. 2014; Gruppelaar and Schapira 2000), and also indicated by the study presented in Section 3.1. The reason for this is that the main fissile isotopes are the same. It is thus relevant to know that CASMO-5 has been benchmarked against some U-MOX experiments (Lee et al. 2008; J. Rhodes et al. 2010; Xu et al. 2010) with good results. This provides confidence that plutonium-containing fuels are generally well modelled by CASMO-5. It then remains to assess how well thorium and related isotopes are modelled.

4.1.1 CASMO-5

CASMO-5 is a deterministic neutronic simulation code, based on a numerical method known as the method of characteristics. Also the method of collision probabilities is used in the pin cell calculations. The computational methodology in itself is not specific to the type of fuel being simulated. However, a number of decisions has been made in the implementation of the method, and also in the preparation of the cross section library, which are based on the assumption that the fuel may be either UOX or U-MOX, i.e. that the fuel mainly consists of U-238.

The energy group divisions in the 586-group cross section library are chosen to give the best possible results for simulation of UOX or MOX-fuelled LWRs. Due to the dominance of thermal neutrons in such reactors, the library is very fine-meshed in the thermal region, i.e. 0 – 10 eV, a feature which is equally adequate for thorium and uranium based fuels. In the resonance region, the group boundaries are chosen so that as far as possible, the U-238 absorption resonances are uniformly distributed within each group, in order to improve the accuracy in the calculation of group average cross sections. This choice is of course then not as adequate for Th-232. Since the U-238 and Th-232 resonances are randomly distributed with respect to one another, it is not obvious whether this will lead to an under- or overprediction of the resonance absorption, or in general what effect this will have on the calculated results. Comparison with experiments is thus necessary, and adaptations of the code for thorium modelling in this respect would be beneficial to provide confidence in the accuracy of the results.

A second choice of group boundaries is made for the two-dimensional transport calculation. The 586 micro-group cross sections are then condensed to 35 larger energy groups. In particular in the thermal spectrum, these boundaries are specifically chosen to

resolve some resonances typical for U-MOX fuel[1], i.e. the large absorption resonances of Pu-240 (1 eV), Pu-242 (2.7 eV) and U-238 (6.6 eV). Some finer group boundary spacing is also present close to the broad resonance of Pu-239 at 0.3 eV. This choice of group boundaries is of course equally good for Th-MOX and U-MOX with respect to the plutonium isotopes and the coverage of the U-238 resonance is merely unimportant for the Th-MOX case. Th-232 does not have any low-energy resonances, so its cross section is adequately modeled with the present thermal group structure. To the small extent that U-233 is produced in the fuel, its comparatively broad and small cross section resonances are not very well covered. However, given that the U-233 number density generally remains below that of any of the Pu isotopes in Th-MOX fuel and that its resonance peaks are more than an order of magnitude below those of the plutonium isotopes, this is not expected to have any large consequences for the modeling accuracy. It could be noted that the distribution of energy groups does not take into account any features of the U-235 cross section either.

Finally, the cross section library has a slightly lower degree of precision with respect to thorium related isotopes in that cross sections for these isotopes are only evaluated at four temperatures whereas the cross sections for U-235, U-238 and the major plutonium isotopes are evaluated at ten temperatures. Larger interpolation errors can thus be expected.

With regards to the methodology, some special adaptations are being made to U-238 as the normally dominant isotope. An important one is that a resonance scatter model has been applied to correct for large scattering resonances in U-238, which gives an improvement of the modeling if the scattering cross section is large compared with the absorption cross section. This enables the modelling of upscattering in the resonance region, and is estimated to give a 10% more negative Doppler coefficient compared with codes which do not have this model (J. Rhodes et al. 2008). This model is however not applied to Th-232, the consequences of which are unknown. Trellue et al. (2011) made a comparison of reactivity coefficients and boron efficiency of Th-MOX with different plutonium vectors, calculated by CASMO-5 and the Monte Carlo-based code Monteburns. The conclusion is that there were indeed differences in the predictions on the order of 25% but there was no clear tendency towards under- or overprediction compared with Monteburns, so other modelling differences with respect to e.g. data libraries or pin cell/lattice/full core calculations can be assumed to play a larger role.

In addition to these considerations, it is noted that U-233 and Th-232 are treated as resonance absorbers by default, adding to the confidence in the calculated results. A correction is routinely applied for the presence of multiple resonance absorbers. Another beneficial feature of CASMO-5 for the modeling of thorium-based fuels is that the effective energy release per fission, Q_{eff} (and also the fission neutron spectra) are calculated for all fissioning isotopes, meaning that the smaller Q_{eff} of U-233 compared with e.g. U-235 and Pu-239 is taken into account.

[1] A less fine 18-group division is made for UOX cases, however the 35 group division is automatically activated by the presence of Pu isotopes, so also for Th-MOX simulations.

The general conclusion that can be drawn from the above outlined considerations is that none of the underlying assumptions in CASMO-5 are in direct conflict with the modelling of Th-MOX fuel, but since it is optimized for U-238-dominated fuel, the confidence in the results for Th-MOX is somewhat lower. Benchmark calculations are ultimately the only way to assess the fidelity of calculated results. Such a benchmark calculation has been undertaken and is adressed in the Section 4.1.3.

4.1.2 Cross section libraries

Although the calculation methodology can be more or less adapted to modelling of a certain isotope, as outlined above, it is ultimately the cross section data used by the code that are of most importance to the accuracy of the predictions. The cross section library integrated in CASMO-5 and used in this work builds on the Evaluated Nuclear Data File ENDF/B-VII, release 0 (Chadwick et al. 2006). It should be noted that some changes were made to the Th-232 and U-233 cross sections in ENDF/B-VII.0 short before its release (Mosteller 2008). These data sets have not been subject to the same level of testing as the other data sets in the library. Benchmarks for U-233 cross sections in the thermal spectrum show some improvement over the last ENDF/B-VI version. For Th-232 cross sections, there are not many well-vetted benchmarks. The cross section library in CASMO-5 has been subject to some change since the studies presented here were made e.g. in connection with the release of ENDF/B-VII.1. New simulations will be carried out to assess the impact of these changes on thorium fuel modeling.

4.1.3 Benchmark based on irradiated Th-MOX rodlet - Paper II

With the aim to create an experimental benchmark for Th-MOX fuel, a Th-MOX rodlet was irradiated in a commercially operating reactor in Obrigheim, Germany, during its last four operating cycles, 2001-2005. This activity was part of the Thorium Cycle Project (Klaassen et al. 2008). The rodlet was of 14.4 cm length and was mounted in one of the inner guide tubes of a U-MOX assembly located at the core center. After the four cycles, the rod was discharged and subjected to post-irradiation examination (PIE), as a part of the LWR-DEPUTY project (Verwerft et al. 2011). One of the analyses performed was a radiochemical analysis, determining the concentration of 28 isotopes in the pin. It should also be mentioned that the general conclusion of the PIE, including profilometry, fission gas analysis and gamma scanning, was that the Th-MOX fuel had withstood the irradiation very well.

A benchmark was formulated such that the irradiation history of the rodlet and its carrier assemblies was simulated. Four different codes or code systems participated in the study; CASMO-5, HELIOS (Villarino et al. 1992), MCBurn (Schitthelm et al. 2010) and a combination of the codes Ecco/Eranos (Rimpault 2002) and Train (Rineiski 2008) (EET). At first, the power history was simulated as measured by an aeroball system located in a next-to-neighbouring fuel assembly, although the uncertainties in the extrapolation of

these measurements to the power at the carrier assembly were known to be large. In addition, the plutonium composition in the carrier U-MOX assemblies was unknown, adding to the uncertainty in the neutronic surroundings of the Th-MOX rodlet. As a test, all participants calculated the content of the burnup indicator Cs-137 present in the fuel, resulting in large overpredictions by all codes. As a result, it was decided that all participants should recalibrate the power history with an individual factor resulting in the correct Cs-137 content being calculated by each code. Thus, the calculated results with respect to the isotopic content of the fuel rod are merely relative to the Cs-137 content.

Whereas this individual recalibration made calculated values of k_∞ and pin power distributions within the assembly difficult to compare, the calibration factors used by the respective benchmark participants gives an indication of how the power of the Th-MOX pin relative to the surrounding U-MOX pins was predicted by the respective codes. For three of the participating codes, the calibration factor was very similar; 0.778 (HELIOS), 0.759 (CASMO-5) and 0.791 (EET), whereas for MCBurn, the calibration factor was 0.663, deviating significantly from the others. This indicates that the three former codes agree reasonably (within 2%) on the relative power of Th-MOX and U-MOX fuel, which is useful for comparative studies. [2]

For the eight elements thorium, protactinium, uranium, plutonium, americium, curium, neodymium and europium, radial distributions were calculated by each code. The profiles for plutonium and uranium are shown in Figures 4.1 and 4.2 as examples. The CASMO-5 prediction of the shape of the radial distribution of plutonium, americium and curium agreed well with the other codes, whereas for thorium, protactinium and uranium, the profile as predicted by CASMO-5 was almost level, disagreeing strongly with the results calculated by the other codes. It is noted that the latter isotopes are the ones most strongly related to the thorium cycle. The present protactinium and uranium isotopes are in this case direct or indirect products of neutron reactions with Th-232. For the fission products europium and neodymium, the profiles predicted by CASMO-5 are slightly more level compared with the other predictions. This is explained by the fact that they stem from fissions of both Pu-239, the distribution of which is strongly periphery-peaked, and U-233, for which the profile, as modelled by CASMO-5, is almost level.

[2] A possible explanation for the deviating result of the MCBurn calculations could be that a reduction of the moderator boron fraction that took place during the last of the four irradiation cycles was not taken into account in the MCBurn simulations. The k_∞-curve shown in Paper II indicates that this could be the case. A higher boron content gives a harder spectrum which in turn gives a higher reactivity of the Th-MOX rod compared with the U-MOX rods, since the Th-MOX rod at this late stage of the irradiation contains significant amounts of U-233 which is favoured by a harder spectrum as shown in Paper V. A higher relative power of the Th-MOX rod would explain why the Th-MOX reaches the same burnup in all simulations, despite the lower assembly power level in the MCBurn calculations. This theory is strengthened by the fact that the production of U-233 is overestimated by MCBurn and the consumption of Pu-239 is underestimated, since a harder spectrum tends to favour captures (producing U-233) over fissions (consuming Pu-239).

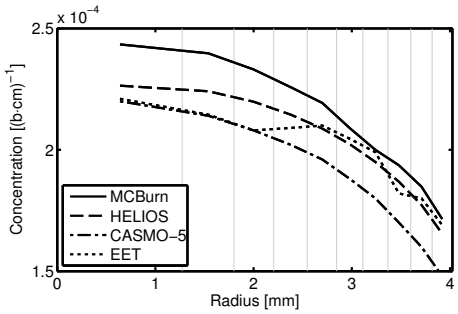

Figure 4.1: Modelled radial plutonium distribution in the Th-MOX rod.

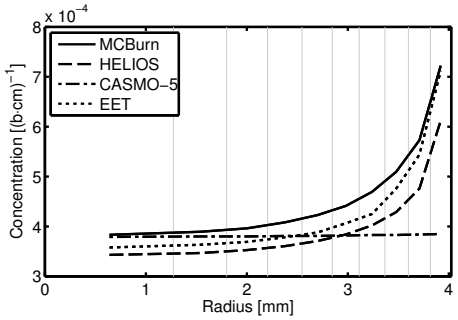

Figure 4.2: Modelled radial uranium distribution in the Th-MOX rod.

Unfortunately, technical problems with the electron-probe micro-analysis instrument at the JRC-ITU (Joint Research Center - Institute for Transuranium Elements) have delayed the experimental assessment of these profiles, so there is no experimental data to support any of the predictions. However, the architects of CASMO-5, Studsvik Scandpower, have confirmed that the shielding effects of Th-232 (which are more thoroughly discussed in Paper VIII) are not accounted for in the used CASMO-5 release. CASMO-5 relies on an empirical radial distribution function for the U-238 resonance integral for generating appropriate power profiles in UOX pellets, when requested by user input (Xu et al. 2009). No such function is implemented for Th-232, which can explain the present results. Xu et al. (2009) state that the radial distribution function applied to the U-238 resonance integral only affects the power profile within the fuel pellet, and not the global results with respect to k_∞ or generation of Pu-239. While this is not necessarily true for the Th-232 case, it at least lends some confidence in the global results calculated by CASMO-5, despite the inadequate representation of the rod internal radial distribution of the reaction rates. It is noted that a similar radial distribution function was implemented for Th-232 in a later release of CASMO-5 (J. D. Rhodes 2014).

The calculated average contents of the investigated isotopes are generally in line with the experimental results, also for CASMO-5. Most deviations could be explained by uncertainties in the cross section libraries or experimental uncertainties. The most important deviation in the CASMO-5 results is that the production of U-234 and U-235 are underestimated, and to some extent also that of U-233. This would indicate that the capture rates in Th-232 are underestimated, which could be related to the discussed issue with the radial distribution of the reaction rates.

Based on the above, it can be concluded that CASMO-5 can be used for theoretical studies of Th-MOX fuel behaviour with some confidence, since the used methodology is generally applicable to thorium. Further validation is of course required before CASMO-5 can be used for safety evaluations of actual core loadings in commercially operating reactors.

4.2 Th-MOX fuel in PWRs

A study was made of Th-MOX in PWRs. In a first phase, lattice simulations were made of Th-MOX with several different plutonium isotope vectors, whereafter full core simulations were performed, based on the experience gained.

4.2.1 Lattice simulations - Paper III

Lattice simulations were performed with CASMO-5 for PWR conditions, comparing Th-MOX and U-MOX fuels with a range of different plutonium types constituting the fissile component. The plutonium types were RGPu, WGPu, plutonium recovered from spent U-MOX fuel (2RPu) and RGPu with an in-growth of americium (AmPu). All Pu vectors represent specific plutonium management scenarios, and the purpose of the study was to investigate whether Th-MOX could offer particular benefits in some specific scenario. The two plutonium types AmPu and 2RPu can be referred to as "low quality", and higher plutonium fractions are necessary to yield the same amount of energy compared with RGPu. A UOX case was also simulated for reference. The operating conditions and the used methodology are described in Paper III. Equivalent fuel compositions were determined and k_∞, safety parameters and mass balances for plutonium, U-233 and minor actinides (MA) were calculated. Burnup averaged safety parameters and mass balance data are listed in Table 4.1, whereas plots showing the burnup dependence of the safety parameters are provided in Paper III. The results for one of the cases - where RGPu was the fissile component - could be compared with similar cases reported by Gruppelaar and Schapira (2000). The conclusions with respect to the reactivity coefficients and plutonium mass balances were in good agreement whereas no reference was provided for the delayed neutron fraction or the short term decay heat. The comparison between the Th+WGPu case and the UOX case could also be referenced against an earlier study by Dziadosz et al. (2004), and the conclusions were also in this case in agreement.

The following conclusions could be drawn from the study:

- The most notable differences between the UOX reference and the Th-MOX and U-MOX fuel types were decreased efficiency of reactivity control systems (soluble boron and control rods), a decreased delayed neutron fraction and a larger (less negative) ITC. It has been found (Shwageraus et al. 2004b) that the decreased efficiency of the reactivity control, which is due to strong thermal absorption in some plutonium isotopes, can be mitigated by increasing the moderation.

- In all cases, more plutonium was required for generating an equal amount of energy using Th-MOX fuel compared with U-MOX fuel, under current burnup constraints. The initial reactivity is reduced by the higher thermal capture cross section of Th-233, and the payback in form of more fissions in U-233 does not compensate for that in the modelled burnup period. It was estimated that one would have

to go to significantly higher burnup (almost 100 MWd/kgHM in the Th+RGPu case) to achieve plutonium savings. This way of using thorium is thus not a way to maximise the energy value of the plutonium stockpile. Also in this context, increased moderation can improve the plutonium utilization (Shwageraus et al. 2004b).

- Most of the safety parameters depend more strongly on which plutonium vector is used, or rather on the fraction of plutonium in the fuel, than on whether the fertile component is Th-232 or U-238. Differences between the different U-MOX and Th-MOX fuel types were in most cases small compared with the difference between the UOX reference and the MOX fuel types as a collective.

- An important exception to this rule was the coolant void reactivity (CVR), estimated as the k_∞ difference between normal operational conditions and a fully voided core. For the low-quality plutonium vectors AmPu and 2RPu, the coolant void reactivity was significantly closer to zero than it was for the UOX reference, but also significantly lower for Th-MOX fuel than for U-MOX fuel, representing a larger margin to a positive CVR. Th-MOX could thus be used to incinerate low-quality plutonium with larger safety margins than U-MOX.

- The slope of the k_∞ curve shown in Figure 4.3 was smaller than the reference for all plutonium types except for WGPu, in which case it was similar. This indicates that some improvement with respect to the power distribution between fuel assemblies in a core can be expected by introduction of Th-MOX or U-MOX fuel with any of the plutonium types RGPu, AmPu or 2RPu. This feature, however, is not improved by an increased moderation ratio, which generally increases fission rates at the cost of capture rates, thus making the slope of the k_∞-curve steeper.

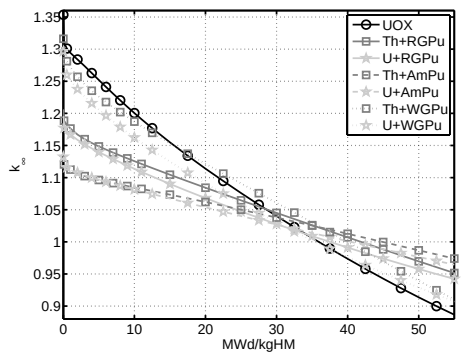

Figure 4.3: k_∞ development with burnup for the UOX reference and six of the investigated Th-MOX and U-MOX fuel types. The k_∞ curves for the 2RPu cases are omitted for clarity. They are very similar to those of the RGPu cases.

Table 4.1: Key parameters for safety and fuel design, and balance of the most relevant elements in terms of kg per fuel assembly. The units of the CRW and the CVR are arbitrary but comparable for the different cases.

Fuel type	UOX	U-RGPu	Th-RGPu	U-AmPu	Th-AmPu	U-WGPu	Th-WGPu	U-2RPu	Th-2RPu
Burnup averaged safety parameters									
CRW [arbitrary]	30	21	22	19	20	25	25	19	20
CVR [arbitrary]	26	15	16	8	12	24	24	8	11
BW [pcm/ppm]	-7.1	-3.0	-3.2	-2.4	-2.6	-4.2	-4.3	-2.5	-2.6
FTC [pcm/K]	-2.9	-3.2	-3.4	-3.0	-3.2	-3.1	-3.3	-3.3	-3.4
MTC [pcm/K]	-45	-58	-48	-50	-46	-62	-46	-48	-42
ITC [pcm/K]	-10.6	-6.4	-6.1	-6.2	-6.2	-6.9	-6.3	-6.0	-5.9
β_{eff} [pcm]	565	399	367	378	343	360	316	417	389
Fuel composition data									
Initial Pu mass [kg]	-	38.2	41.0	49.0	49.6	22.6	26.7	54.2	55.2
Final Pu mass [kg]	5.8	27.5	18.9	37.8	26.7	14.8	7.4	41.3	31.3
Pu consumption [%]	-	28	54	23	46	35	72	24	43
Final Pu quality [%]	68	57	43	56	44	64	47	50	38
Final MA mass [kg]	0.57	1.90	1.97	3.12	3.05	0.494	0.499	3.10	3.12

With these conclusions in mind, new reflections can be made on how to use Th-MOX fuel in a sensible way. There appear to be two paths to follow: One is to leave the moderation ratio and the assembly design unaltered, and try to draw benefits from the flatter k_∞-curve for other plutonium compositions than WGPu. This approach is held in the work presented in Section 4.2.2. The other path would be to increase the moderation ratio in order to improve plutonium utilization and possibly also safety parameters. This path is investigated in Section 4.3.

4.2.2 Full core simulations - Paper IV

An even power distribution in a core is not a goal in itself, but is rather necessary for maintaining a high average power without exceeding thermal limits in any assembly. Introducing Th-MOX to improve the power distribution compared with the current UOX fuel could thus give a possibility to increase the average power, i.e. a power uprate. A major power uprate would however also necessitate a number of large and expensive modifications to the whole reactor system. Another implication of the flatter k_∞ curve is that the fuel can be designed to reach a higher discharge burnup without having to bear with an excessively high initial k_∞. This is an interesting option, since the aforementioned good material properties of thorium oxide makes it plausible that Th-MOX could sustain higher burnups than U-MOX.

A higher burnup capacity can be utilized either by decreasing the number of fuel assemblies replaced in each reload, or by extending the operating cycles. Of these two, the option of operating cycle extension was chosen. Full core simulations were performed for the Swedish PWR Ringhals 3, using CASMO-4E (J. Rhodes et al. 2009) and SIMULATE-3 (Dean 2007) since these are the codes normally used for core management in this reactor. CASMO-4E operates essentially in the same way as CASMO-5, but with fewer energy groups in the pin cell calculations and a different cross section library. In addition, the mentioned correction for multiple resonance absorbers is not implemented in CASMO-4E. The differences between CASMO-5 and CASMO-4E can be expected to affect the modeling of thorium based fuel to some extent, but CASMO-4E has previously been benchmarked for simulation of Th-MOX fuel with good results (Shwageraus et al. 2004b), so it is expected that the modeling is reasonable. The results should be regarded as indicative. SIMULATE-3 uses the homogenized macroscopic cross sections provided by CASMO-4E for two-group full core diffusion simulations, and is thus less dependent on the specific isotopes present in the fuel.

A normal UOX reload, designed for a one-year operating cycle, was used as a reference case and compared with a Th-MOX core which was designed for an 18-month operating cycle, i.e. a 50% cycle extension. As previously noted, the reactivity worth of soluble boron and control rods is reduced by the presence of plutonium, and for this reason, the Th-MOX core was modelled assuming the soluble boron to be enriched in B-10, and stronger control rods were also modelled. Another difference to the reference case was that Gd_2O_3 mixed with the fuel matrix was not used, since ternary mixtures of ThO_2,

PuO_2 and Gd_2O_3 are difficult both to manufacture and to model. Instead, a thin layer of zirconium boride applied to the outer surface of the fuel pellet was modelled, a concept referred to as Integral Fuel Burnable Absorber (IFBA).

The conclusion of the study is that operating cycle extension using Th-MOX fuel is indeed possible, provided the above mentioned reinforcements of the reactivity control systems and that the fuel material can sustain high burnups. Despite the fuel assemblies being designed for higher burnup, the initial k_∞ is lower than that of Th-MOX as seen in Figure 4.4, partially due to the use of IFBA. The different indicators of the power distribution are in general well within the safety margins and the margin to departure from nucleate boiling is even improved as shown in Figure 4.5.

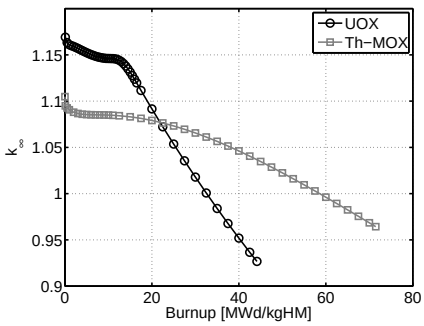

Figure 4.4: k_∞ development with burnup for the Th-MOX and the reference UOX fuel assembly.

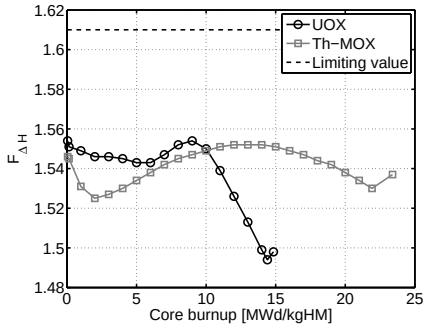

Figure 4.5: $F_{\Delta H}$, i.e. maximum rod power relative to core average rod power as a function of burnup, for the Th-MOX and the reference UOX core.

The calculated safety parameters are listed in Table 4.2 along with the limits for these applied at Ringhals 3. As can be seen, three of the safety parameters were outside the pre-defined limits: β_{eff}, the Doppler power coefficient (DPC) and the FTC. Transients involving these three safety parameters were qualitatively assessed, indicating that these transients would have an acceptable course with Th-MOX fuel. Details are given in Paper IV. Since Ringhals 3 is a typical PWR with no features making it especially suitable for MOX fuel, it seems that operating cycle extension using Th-MOX fuel should be possible in PWRs in general.

Later studies by Lau et al. (2014b) also found that the axial offset, i.e. the power imbalance between the upper and lower halves of the core, was reduced for the Th-MOX core. Moreover, the fluctuations in axial offset induced by rapid power changes were almost eliminated, greatly improving the core stability.

As a final remark, it is interesting to note that some but not all of the predictions made by the lattice calculations proved to be in agreement with the full core situation. The decreased delayed neutron fraction and boron and control rod worths were similarly

important. The ITC was predicted by the lattice simulations to be higher for Th-MOX, but was found to be lower in the full core simulations. Given that the full core simulations take several effects into account which are not present in the lattice simulations, most importantly presence of control rods, the full core results are likely to be more representative in this remark. The Doppler power coefficient was not calculated in the lattice simulations, but the closely related FTC was, showing no big deviation from that of UOX fuel. In the full core simulations however, these coefficients were outside of the specified limits. Referencing the results to those of Shwageraus et al. (2004b), we see that results from the full core simulations are confirmed rather than those of the lattice simulations.

Table 4.2: Key safety parameters for the Th-MOX core and the UOX reference.

	Limit	UOX reference	Th-MOX
Min boron worth [pcm/ppm]	-15	-8.3	-8.7
Max boron worth [pcm/ppm]	-5	-6.2	-5.2
Min SDM [pcm]	2000	2559	3895
Max rod worth [pcm]	-	6505	7808
Max MDC [$(\Delta$ k/k)/(g/cm^3)]	0.50	0.41	0.34
Max MTC [pcm/K]	-	-1.3	-4.0
Max ITC [pcm/K]	0.0	-4.4	-7.9
Min DTC [pcm/K]	-4.00	-3.72	-4.53
Max DTC [pcm/K]	-1.70	-2.10	-2.36
Min DPC [pcm/%power]	-21.0	-15.5	-21.5
Max DPC [pcm/%power]	-6.5	-9.8	-14.6
Min β_{eff} EOC [pcm]	430	502	350
Max β_{eff} BOC [pcm]	720	647	370

4.3 BWR fuel assembly design for efficient use of plutonium - Paper V

Since thorium based fuel is suggested as an efficient means to manage stockpiles of plutonium and MA, it is relevant to investigate how this can be done in the most efficient way, while staying within the limits drawn by the condition of safe reactor operation. It is widely recognized (Puill 2002; Shwageraus et al. 2004b) that conventionally designed fuel assemblies loaded with plutonium-containing fuels are strongly undermoderated. A higher H/HM ratio increases the initial reactivity of the fuel by favouring fission over neutron capture, which has the additional consequence in Th-MOX fuel that the initially added plutonium is more thoroughly consumed, since less U-233 is generated to compete for the thermal neutrons.

The moderation ratio can be increased by several means. The most straightforward is

perhaps to increase the moderator density, which can be done by decreasing its temperature or by increasing the reactor pressure. Both these operations demand a change in how the reactor is operated, and quite possibly to the thermohydraulic system of the reactor. Another method which limits the change to the reactor core is to decrease the fuel volume and, consequently, increase the moderator volume. The BWR context was chosen for trying this, since there is a high degree of design flexibility within the box surrounding the BWR assembly. The consequences for different fuel types of varying the H/HM ratio are illustrated in Figure 4.6. The normalized curves represent the beginning of life k_∞ at different H/HM ratios, showing that the maxima occur at very different H/HM ratios. Also shown are the void coefficients at different H/HM ratios for each case, which are in effect the derivatives of the respective k_∞-curves. Since a negative void coefficient is a prerequisite for safe reactor operation, the H/HM ratio must be slightly below the optimum. As shown in Table 4.3, the H/HM ratio in a standard BWR assembly is just below 5 (assuming 40% void in the coolant), varying slightly depending on the density of the fuel material. The optimum H/HM ratio for Th+U-233 fuel is below that, which explains the positive void coefficient encountered for this fuel type in Paper I. It also indicates that the H/HM ratio for Th-MOX fuel must have a reasonable margin to the optimum seen in Figure 4.6, since the increasing U-233 content of this fuel will shift the optimum towards lower values during burnup.

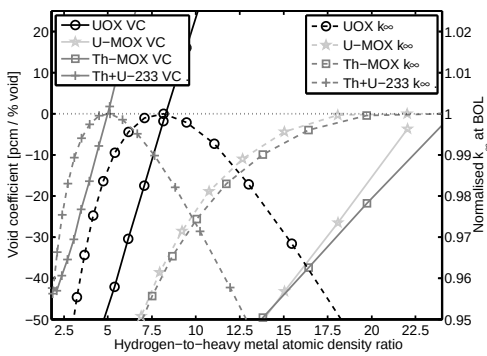

Figure 4.6: Normalized k_∞ and void coefficients for four different fuel types.

The goal of the exercise is once again to introduce as little change as possible for the reactor operator. Therefore, the new fuel assembly is designed to yield an equal amount of energy throughout its lifetime as a normal UOX BWR fuel assembly. Decreasing the fuel volume then means that the burnup must be increased, i.e. more energy must be generated per unit fuel mass. Once again, it has to be assumed that Th-MOX fuel can sustain high burnups.

A fuel assembly design was created with an almost doubled H/HM ratio compared with the reference BWR assembly, GE14-N. This increase in the H/HM ratio was realised both by removing some rods from the lattice and by decreasing the diameter of those remaining.

The results were compared with a UOX and a U-MOX reference of normal (GE14-N) design, and also with a Th-MOX assembly with unaltered design. Key parameters for all references and the modified assembly are listed in Table 4.3 and the general conclusions were the following:

- The energy generated per loaded kilogram of plutonium was increased by 19% compared with an unmodified Th-MOX assembly. The achieved value is similar to that of an unmodified U-MOX assembly.

- The fissile fraction of the discharged plutonium was only 24%, to be compared with the 49% for discharged U-MOX fuel.

- The discharged masses of U-233 and of MA were lowered.

- The reactivity coefficients were higher (less negative) but not widely different from those of the reference.

- The control rod worth was not improved relative to the unmodified Th-MOX assembly, despite the increased moderation.

- The hot-to-cold reactivity swing (HCS), i.e. the k_∞ difference between hot full power conditions with all rods out and cold zero power conditions with all rods in, was nevertheless improved. This is an indicator of the shutdown margin that can be expected.

It can be concluded that plutonium incineration by Th-MOX fuel in BWRs can be made even more efficient by modification of the fuel assembly design. This strategy is most suitable where there is a stockpile of plutonium to be disposed of and there is no intention to reprocess the fuel, since the amounts of fissile material in the spent fuel is minimised. In this case, the chemical inertness of the ThO_2 matrix is beneficial, improving the fuel's suitability for final storage.

Table 4.3: Key parameters for safety and fuel design, and balance of the most relevant elements.

Fuel type	UOX ref.	U-MOX ref.	Th-MOX ref.	New Th-MOX
Burnup averaged safety parameters				
CRW [arbitrary]	16	14	14	13
HCS [arbitrary]	-14	-12	-13	-14
VC [pcm/% void]	-55	-52	-37	-41
FTC [pcm/K]	-2.4	-2.5	-2.9	-2.1
ITC [pcm/K]	-28	-26	-21	-12
β_{eff} [pcm]	551	399	365	369
Design parameters				
H/HM ratio	4.6	4.4	4.8	9.0
Fuel mass [kg]	186	192	172	108
B_D [MWd/kgHM]	53	52	58	91
Fuel composition data				
Initial Pu mass [kg]	-	13.4	15.8	13.4
Pu consumption [%]	-	34	54	67
Final Pu quality [%]	59	49	40	24
Final ^{233}U mass [kg]	-	-	2.3	1.4
Final MA mass [kg]	0.20	0.74	0.75	0.62

CHAPTER 5

Thermal-Mechanical Properties of Thorium Fuel Materials

This chapter describes an irradiation experiment designed to provide data on how Th-MOX and Th-Add fuel performs under irradiation with respect to its thermal-mechanical properties. Theoretical simulations of the thermal-mechanical performance of Th-MOX fuel are also described and comparisons made between the experimental results and the theoretical predictions. This chapter is based mainly on the content of Papers VI - VIII.

5.1 Instrumented irradiation experiment

In the preceding chapters, it has been necessary to make some assumptions for the thermal-mechanical operating limits. In general, it has been assumed that Th-MOX fuel can withstand the same linear heat generation rates (LHGR) as uranium fuel, but that thorium fuel can operate to higher burnup. These assumptions rest upon the fact that thorium oxide has a number of beneficial material properties. However, exactly how these properties interact is difficult to foresee, especially in the presence of another component in the fuel; plutonium oxide. Furthermore, the properties change as the fuel is irradiated, making the predictions even more difficult. Experimental assessment is thus crucial to

build the necessary degree of confidence in the predictions.

5.1.1 Rig IFA-730 in the Halden research reactor - Paper VI

The research reactor located in Halden offers the possibility to measure the properties of nuclear fuel under irradiation. Information on fuel centerline temperature (T_C), dimensional changes of fuel and cladding, the internal pressure in the fuel rod and a number of other parameters is conveyed on-line, during operation, from dedicated instruments located inside the core. Experimental irradiation rigs comprising up to about twelve nuclear fuel rodlets can be loaded in up to 30 locations in the reactor core and be irradiated for approximately six months per year. The reactor is the world's only boiling heavy water reactor, operating at a pressure of 33.6 bar and a coolant temperature of 240 °C. It is operated by the Norwegian Institute for Energy Technology, IFE, which also runs nuclear material laboratories at Kjeller in Norway, where all the driver fuel and some of the test fuel for the reactor is manufactured.

The experimental rig IFA-730 comprises six rods and is depicted in Figure 5.1. The rods consist of the pellet types listed in Table 5.1. All materials except for the Th-MOX material were manufactuerd at IFE Kjeller. As seen in Figure 5.1, there are only two short segments of Th-MOX pellets in two rods (rods one and three – *ThMOX-1* and *ThMOX-3*). These segments comprise four pellets each of the eight Th-MOX fuel pellets which were available at the start of the experiment. These were manufactured by JRC-ITU, and being originally designed for a previous experiment (Verwerft et al. 2007), they have a small diameter, not representative for LWR fuel. The remaining pellets in rods *ThMOX-1* and *ThMOX-3* are composed of the material referred to as "ThUOX" in Table 5.1. This material was designed to provide a neutronic environment for these pellets that would minimise the effects of discontinuities in the macroscopic cross sections at the ends of these short segments. Since the IFE Kjeller labs routinely handle 20% enriched uranium, this was chosen as the fissile component and was mixed with thorium in proportions chosen to give the two materials approximately equal power throughout the irradiation. One rod (rod five – *ThUOX-5*), made of only ThUOX material, was also inserted to provide a reference to rods *ThMOX-1* and *ThMOX-3*. The density of these pellets was initially measured to 94% of the theoretical density (TD), however it underwent very high densification during the early phases of the irradiation and so the density was re-measured on remaining un-irradiated pellets, and found to be about 80% of TD. A possible explanation is the low pressing force used for these pellets.

The remaining two materials are the UOX reference material (rod six - *UOX-6*) and a material representative for the Th-Add concept as modelled by Lau et al. for use in PWRs (rods two and four – *ThAdd-2* and *ThAdd-4*). The diameter of these pellets was chosen as typical for BWR fuel, i.e. significantly larger than that of the Th-MOX pellets. The uranium enrichment in these pellets was chosen so that their power would not be too high compared with that of the Th-MOX, but at the same time high enough to allow for reasonable burnup accumulation rates.

Table 5.1: Basic characteristics of the fuel pellets being irradiated in rig IFA-730. Compositions are given as oxide weight percentages.

Fuel type	Th-MOX	ThUOX	Th-Add	UOX
Compositions				
Th content [%]	92.1	42	7	-
Pu content [%]	7.9	-	-	-
U content [%]	-	58	93	100
U enrichment [% U-235]	-	19.8	9.0	8.4
Manufacture parameters				
Pressing force [t/cm^2]	4.1	2.5	4.0	4.0
Sintering temperature [°C]	1650	1680	1680	1680
Sintering time [ht]	6	4	4	4
Sintering atmosphere	Wet Ar/H_2	Dry H_2	Dry H_2	Dry H_2
Diameter [mm]	5.9	5.9	8.48	8.48
Density [% of TD]	97	80	95	97

As shown in Figure 5.1, all rods are instrumented with thermocouples, measuring the T_C of the pellet stacks. For the rods containing the Th-MOX pellets, these are located so that the thermocouple tip extends to the center of the Th-MOX segment. Pressure bellows with direct communication with the rod interior are mounted on rods *ThAdd-2*, *ThMOX-3*, *ThUOX-5* and *UOX-6*. Changes in pressure cause lateral movement of the bellows, which is converted to an electric signal by transducers mounted in the rig. Similarly, rods *ThMOX-1* and *ThAdd-4* are instrumented with transducers converting the lateral movement of the cladding, caused by its extension and contraction, to electric signals.

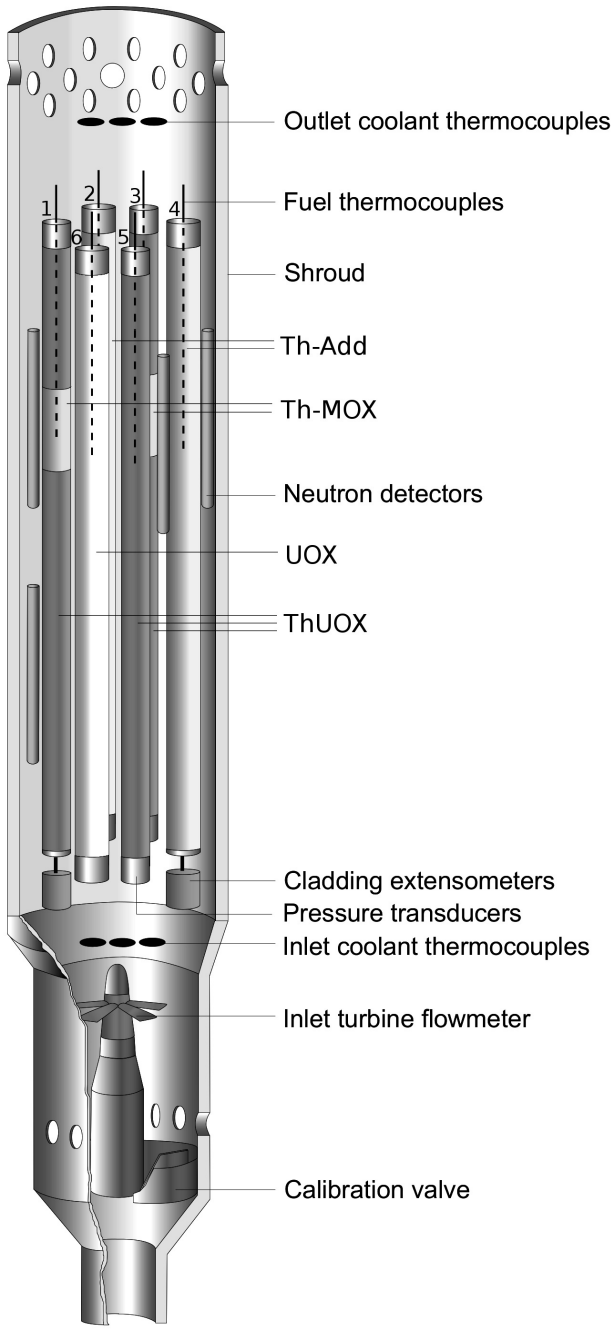

Figure 5.1: Schematic image of the testrig IFA-730.

5.1.2 Results of the irradiation

At the time of this writing (March 2015), IFA-730 has been under irradiation for 350 days. The rods with BWR dimensions, *ThAdd-2*, *ThAdd-4* and *UOX-6*, have accumulated a burnup of about 17 MWd/kgHM and the small-diameter rods *ThMOX-1*, *ThMOX-3* and *ThUOX-5*, have accumulated about 25 MWd/kgHM. Most of the instrumentation has been working well, with three exceptions. The pressure signal from rod *ThUOX-5* was lost early in the irradiation due to water penetration into the cable. The pressure bellows on rod *UOX-6* initially gave spurious signals, followed by a sudden drop, after which the instrument responded as expected to power changes but showed high absolute values of the rod internal pressure. It can however be expected that the instrument will show when fission gas release (FGR) occurs. Finally, the signal from the thermocouple on rod *ThAdd-2* made an unexpected jump during an early episode when it operated close to its operating limit of 1300°C and has since then given signals difficult to interpret.

The pin powers used for normalization of the measured data are calculated using a rig-internal pin power distribution obtained from simulations with HELIOS (Casal et al. 1991). The input power level of the rig is calibrated using measured values of the coolant flow and temperature at the rig inlet and outlet during the first startup. The calculated radial power distribution between the pins is corrected for a possible flux tilt over the rig quantified by four neutron detectors located in the same horizontal plane as the thermocouples. Finally, the axial power distribution can be calculated using a signal from a fourth neutron detector located at a different axial level. The location of the neutron detectors is shown in Figure 5.1.

The T_C for the rods with BWR dimensions are plotted in Figure 5.2, normalized to 30 kW/m, which is close to their actual operating LHGR. During the first power ramp, the two Th-Add rods had a lower normalized T_C than their UOX reference by 20 K and 35 K respectively. After this, cracking, relocation and possibly some damage to the *ThAdd-2* thermocouple caused the normalized temperatures to drift relative to each other. After a shutdown at about 90 days, during which the rig was moved and pellet fragments maybe relocated, the original relation with the Th-Add rods below the UOX reference was re-established, although the very low temperature indicated by the possibly faulty *ThAdd-2* thermocouple may be questioned. Since this result was highly unexpected it was suspected that the power predictions were not accurate enough to resolve such small differences as the ones observed. To assess this, the rig was rotated 180° after 290 days of irradiation. After this, the relation between the normalized temperatures of *ThAdd-4* and *UOX-6* was reversed. The rapid drop of the normalized temperatures of rods *ThAdd-2* and *UOX-6* is unexpected and suggests that the power levels may not be correctly calculated. This implies that the measured normalized temperature difference lies within the experimental error, if the probably erratic *ThAdd-2* signal is disregarded.

However, there is a second way to assess the thermal conductivity of the irradiated materials. When the reactor is scrammed, T_C exponentially approaches that of the coolant and the time constant for this process gives an independent indication of the conductivity. A high time constant corresponds to a low conductivity and a high T_C. As

can be seen in Figure 5.3, the time constants for rods *ThAdd-4* and *UOX-6* have the same relation as indicated by the normalized temperatures whereas the comparatively high time constants for rod *ThAdd-2* contradict the very low T_C measured for this rod, underlining that this thermocouple may indeed be unreliable. These measurements thus reconfirm the previous indication that the Th-Add material initially has a higher conductivity than the UOX rod, whereas the relation is later reversed.

Whereas the matter must of course be further investigated, it should be noted that a higher thermal conductivity for the Th-Add material would be an interesting phenomenon. Assuming a homogeneous mixture of ThO_2 and UO_2, the thermal conductivity should be lower than for pure UO_2, since the ThO_2 acts as a phonon-scattering impurity in the lattice, just like PuO_2 in a ThO_2 matrix, as discussed in Section 5.2.2. Unfortunately, no direct measurement of the thermal conductivity of $(Th,U)O_2$ with such low ThO_2 fraction could be found in the literature. A hypothesis that may explain the observed behaviour is that the mixture of ThO_2 and UO_2 in this fuel is very inhomogeneous, possibly leading to a "network" of more well-conducting ThO_2-rich regions between less well-conducting UO_2-rich regions.

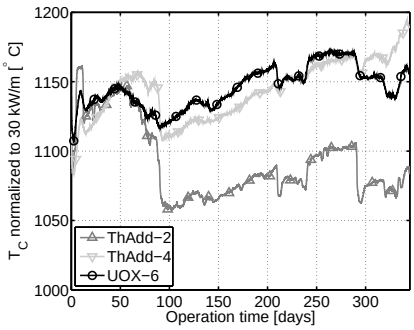

Figure 5.2: Measured T_C for rods *ThAdd-2*, *ThAdd-4* and *UOX-6*.

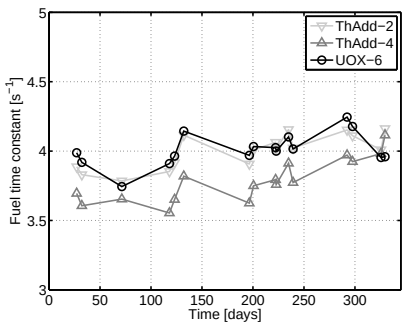

Figure 5.3: Scram time constants for rods *ThAdd-2*, *ThAdd-4* and *UOX-6*.

The Th-MOX rods and their reference operate at a lower LHGR than the Th-Add and UOX pins, about 20 kW/m, due to their smaller diameter. Their respective T_C, normalized to this power level, are plotted in Figure 5.4. This figure shows a simpler burnup dependence of T_C compared with Figure 5.2, possibly indicating less cracking and relocation. For the Th-MOX rods, T_C increases almost linearly, an effect of the slow decrease of the thermal conductivity caused by irradiation damage to the fuel matrix. For rod *ThUOX-5*, the normalized T_C starts at a much higher temperature due to the low conductivity of this material caused by its low density. T_C then increases rapidly as the low density material resinters during the first months of operation, increasing the pellet-cladding gap size. After this, it increases linearly at approximately the same rate as the Th-MOX rods. The scram time constants shown in Figure 5.5 confirm the recorded behaviour.

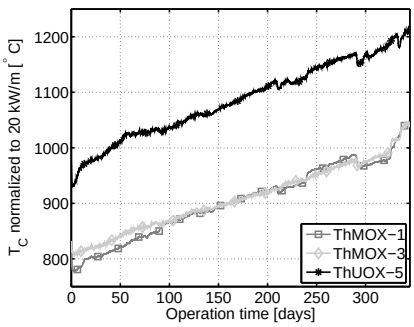

Figure 5.4: Measured T_C for rods *ThMOX-1*, *ThMOX-3* and *ThUOX-5*.

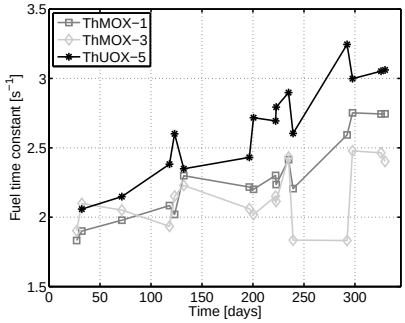

Figure 5.5: Scram time constants for rods *ThMOX-1*, *ThMOX-3* and *ThUOX-5*.

The cladding elongation registered for rods *ThMOX-1* and *ThAdd-4* is plotted in Figure 5.6. The slow and approximately linear elongation is due to normal irradiation swelling of the cladding material. The discontinuity at about 90 days coincides with the shutdown period during which the rig was moved, and is probably a consequence of the forces acting on the rig in connection with this. The rapid increase seen for rod *ThAdd-4* at about 300 operation days indicates that there is some contact between the pellet and the cladding, causing the cladding to expand at the same rate as the fuel during the power ramps that took place at this point in the irradiation. The rapid elongation was partly reversed by relaxation of the cladding material in connection with a shutdown at 340 days, indicating that the pellet-cladding gap is not firmly closed yet. The low pressure at which the Halden reactor operates does not cause the cladding to creep inwards towards the fuel to the same extent as it does in most commercial reactors. The current burnup is quite typical for detecting the first signs of impending gap closure for UOX fuel irradiated in the Halden reactor.

The fact that the first indications of gap closure for rod *ThAdd-4* almost coincide with the rig rotation at 290 days further complicates the interpretation of the temperature readings. Given the similarity between rods *ThAdd-4* and *UOX-6*, it is reasonable to assume that gap closure would occur at approximately the same time. Gap closure generally gives a drop in the normalized T_C, similar to what was recorded in connection with the rig rotation.

Finally, the pressure recorded for rods *ThAdd-2*, *ThMOX-3* and *UOX-6* is plotted in Figure 5.7, normalized to zero power and room temperature. The pressure for rod *ThMOX-3* behaves as expected, increasing slowly as the fuel material swells with increasing irradiation exposure. Similarly to the temperature recordings for rods *ThAdd-2* and *UOX-6*, the pressure signals from these rods drift relative to each other in an unexpected manner, but settles for a more predictive behaviour after the shutdown at 90 days. As mentioned, the pressure bellows on rod *UOX-6* showed erratic behaviour at the beginning of the irradiation, so the absolute value of the recorded pressure is likely not accurate.

However, all three rods almost simultaneously show indications of FGR at about 320 days. This coincides with a power ramp, a situation which is known to trigger FGR. It also occurs at a burnup and temperature typical for FGR, so once again, normal behaviour is confirmed. It should be recalled that the FGR in rod *ThMOX-3* is dominated by the low-density ThUOX material, and is thus not representative of Th-MOX behaviour.

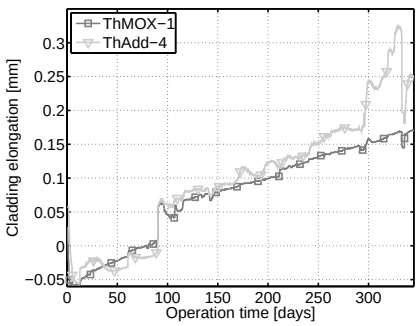

Figure 5.6: Cladding elongation measured for rods *ThMOX-1* and *ThAdd-4*.

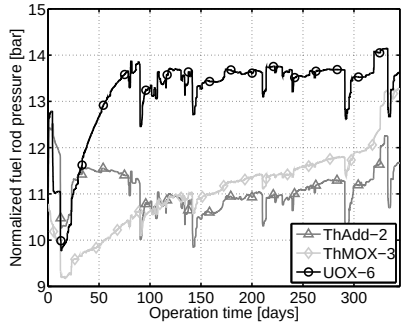

Figure 5.7: Fuel rod pressures measured for rods *ThAdd-2*, *ThMOX-3* and *UOX-6*.

The most interesting indication so far from this irradiation is that the temperature of the Th-Add rods may be lower than that of the UOX rod. It also seems, from the pressure readings, that the Th-Add behaviour with regards to FGR is similar to that of UOX fuel. The first indications of gap closure at rod *ThAdd-4* also occurs at a typical burnup. For the Th-MOX rods, the most useful result is the recorded T_C. This can be compared with theoretical prediction, which is the subject of the following section. The fact that the Th-MOX material only constitutes a minor part of the rods in which it is irradiated makes it impossible to draw any conclusions from the pressure and cladding elongation data, since these signals are dominated by the filler material.

5.2 Fuel performance modelling and benchmarking

A crucial aspect in the licensing of a nuclear fuel for use in a commercial reactor is that its thermal-mechanical behaviour can be accurately predicted for all operation conditions and at any burnup up to the projected discharged burnup. Such predictions are routinely made by fuel performance codes, making use of established correlations for the fuel material properties and their dependence on most importantly temperature, but also on burnup and the composition of the fuel. The material behaviour is also closely related to the neutronic behaviour of the fuel through the spatial distribution of the power generation within the fuel. Several well established codes exist for the prediction of UOX fuel behaviour, and most of them can also be used for U-MOX fuel. A version of the well-established code FRAPCON has also been written for prediction of $(Th,U)O_2$ fuel performance (Long et al.

2004) and recently several efforts have been directed towards the modelling of Th-MOX, e.g. by Mieloszyk et al. (2014). An earlier effort to model Th-MOX was also made within the OMICO project (Verwerft et al. 2007).

Given the uncertainties in many of the material property correlations, in particular regarding their burnup dependence, and the simplifications that are necessarily made in the codes, thorough validation through comparison with experimental data is necessary. The described irradiation experiment is intended to form part of the basis for such a validation. The efforts made to write and validate a fuel performance code for modeling of Th-MOX fuel are described in this section.

5.2.1 FRAPCON 3.4

The fuel performance code FRAPCON 3.4 (Geelhood et al. 2011a) has been developed for the USNRC and is used for fuel performance simulations for regulatory control. This code was used as a basis for the development of a fuel performance code for Th-MOX. FRAPCON 3.4 calculates temperature, pressure and deformation of an LWR fuel rod for steady-state operation, up to the highest burnups typical for LWR fuel. The state of the fuel rod is determined for each time step by iterative calculations until the fuel-cladding gap temperature difference and the rod internal gas pressure converge (Geelhood et al. 2011a).

FRAPCON 3.4 has a modular structure with a specific subroutine for calculation of each material property. The subroutines related to the fuel material properties have been modified as described in Paper VIII. Dependence of the material properties on density, stoichiometry and burnup has been included to the largest possible extent. Where data are lacking for Th-MOX, a best estimate has been made based on the available literature and corresponding correlations for U-MOX. The subroutine related to FGR has been left unmodified due to the lack of relevant experimental data to support a new model, but its importance is recognized and briefly discussed in Section 5.2.3. The subroutine for prediction of the radial distribution of the power generation within the fuel pin has been updated as discussed in Paper VII and outlined in Section 5.2.4. Other subroutines, related to the properties of water, the gas mixture in the fuel-cladding gap and the cladding material are left unmodified.

FRAPCON 3.5 was released during the course of the development work. The differences between the results as calculated by FRAPCON 3.5 and FRAPCON 3.4 were negligible after the same material property subroutines were modified for both codes. Since the modified subroutine for radial power profile prediction could not be directly implemented in FRAPCON 3.5, it was decided that FRAPCON 3.4 would be kept as the base for the development work, for the time being.

5.2.2 Material properties

The good material properties of thorium oxide have formed the basis for the assumption that thorium based fuel should be able to operate to higher discharge burnup. Whereas many investigations on the material properties of UOX and to some extent also U-MOX fuel have been carried out, much less information is available about thorium based fuel. There is some information available on the properties of $(Th,U)O_2$, although not for mixtures with ThO_2 fractions below 50%, which is unfortunate given the compositions typical for the proposed Th-Add concept. The knowledge base of Th-MOX fuel was until recently very small (Bakker et al. 1997), but has been significantly expanded by measurements performed at the JRC-ITU in the last years (Böhler et al. 2015; Cozzo et al. 2011; Vălu et al. 2014). The details of the material property correlations can be found in Paper VIII, and the most important features, related to those of UO_2 are:

- Lower thermal expansion, putting less strain on the cladding during temperature increases when the pellet-cladding gap is closed, but also leaving a larger pellet-cladding gap early in life. The used correlation is plotted in Figure 5.8.
- A higher melting point; approximately 700 K higher for a composition with 8% plutonium, see Figure 5.9.
- Lower heat capacity, giving a smaller stored heat to be cooled off in transient scenarios, but also a faster fuel temperature increase in a reactivity insertion accident, see Figure 5.10.
- Higher thermal conductivity for compositions up to about 8% plutonium, see Figure 5.11.

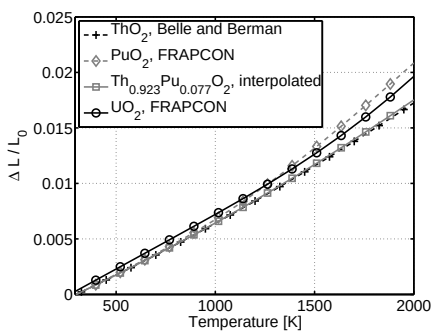

Figure 5.8: Thermal expansion of ThO_2, PuO_2, UO_2 and the present Th-MOX material.

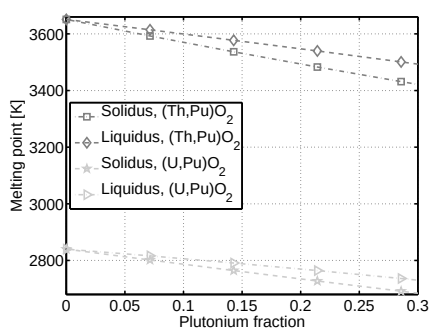

Figure 5.9: Solidus and liquidus temperatures for $Th_{1-y}Pu_yO_2$ and $U_{1-y}Pu_yO_2$ for PuO_2 fractions lower than 30%.

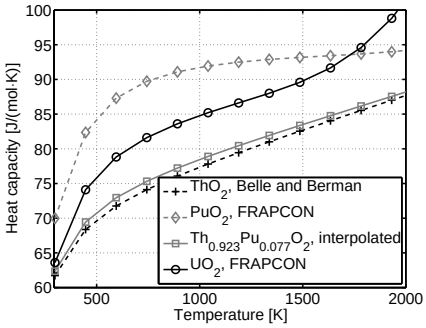

Figure 5.10: Heat capacity for ThO_2, PuO_2, UO_2 and the present Th-MOX material.

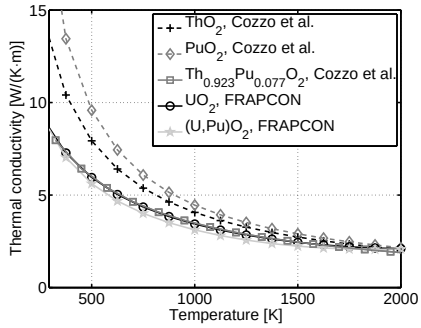

Figure 5.11: Thermal conductivity for ThO_2, PuO_2, UO_2, U-MOX fuel and the present Th-MOX material.

Equations for these curves are given in Paper VIII, further discussed in Section 5.2.5.

The thermal conductivity deserves a more thorough discussion since it is the most important material property, having a strong influence on the fuel temperature. It is noted that the correlation used in FRAPCON for the thermal conductivity of U-MOX fuel includes no dependence on the PuO_2 fraction, whereas it can be seen that already a small addition of PuO_2 to the ThO_2 matrix causes a strong reduction of the thermal conductivity. This is primarily due to the fact that the reduction of the thermal conductivity depends on the relative mass difference and the relative lattice parameter difference between the two components (Cozzo et al. 2011). These differences are larger for the (Th,Pu)O_2 case than for the (U,Pu)O_2 case. The general conclusion is that the thermal conductivity of thorium based fuels is higher than that of uranium based fuels only under certain conditions.

5.2.3 Fission gas release

The fuel temperature is the driving force behind the processes, most importantly FGR, that limit the lifetime of the fuel. The onset of FGR depends largely on the diffusion rate of fission gases in the fuel matrix. The diffusion rates depend on the fuel material and are inherently lower for ThO_2 than for UO_2. There is also a strong dependence on the fuel temperature, and the lower FGR seen in Th-MOX fuels (Karam et al. 2008) is mostly attributed to the lower temperature caused by the higher thermal conductivity.

Fission gases are generally modeled as being released in a stepwise process. First, the fission gas atoms are formed by fission reactions after which they migrate through the fuel matrix, ultimately being trapped at grain boundaries. There, fission gas bubbles are formed, which eventually interconnect. When a direct connection is formed with the pellet surface, the fission gases are released. This often happens in connection with power ramps,

during which the heated gases expand, putting additional strain on the fuel material. The many processes involved depend not only on the diffusion rates and fuel temperature, but also on the fission gas generation rates, the fuel microstructure and other factors. The complexity of the process makes it necessary to benchmark against experimental data. Given the very scarce data available, and in particular since no Th-MOX FGR data can be obtained from IFA-730, the development of a FGR model for Th-MOX fuel has been left for later. A new FGR model has been developed for (Th,U)O_2 fuel and benchmarked against existing irradiation data for this fuel type by (Long et al. 2002). The model employed for Th-MOX fuel will most likely be similar.

5.2.4 Radial power profiles - Paper VII

The neutronic differences between Th-MOX and UOX fuels have a direct impact on the fuel performance simulations through the radial variations of the power production within the fuel pin. This necessitates an adaptation of the subroutine determining the radial power profile in FRAPCON 3.4.

The original model for radial power profile prediction in FRAPCON 3.4, called TUBRNP (TransUranus BuRnuP equations, adopted from the fuel performance code TRANSURANUS), uses a one-group diffusion approximation for the neutron flux, solving the equation analytically assuming a homogeneous distribution of all isotopes within the fuel. This approximation is judged to hold sufficiently well for UOX fuel, and is also used for U-MOX fuel. Isotopic concentrations are determined for each radial node and local power generation is calculated from the flux and the macroscopic fission cross sections. The radial distribution of the Pu-239 production is described by an empirically derived shape function which is multiplied with the average U-238 concentration when calculating local concentrations of U-238 and Pu-239 for the power profile determination.

The first step in the adaptation to Th-MOX fuel was to extend the list of isotopes being accounted for in the evaluation with the isotopes relevant for Th-MOX fuel i.e. Th-232, U-233, U-234, U-236 and Pu-238. The Monte Carlo-based neutronic code *Serpent* (Leppänen 2012) was used to calculate power profiles for Th-MOX fuel, and it was found that these were not well reproduced by the current solution model. The radial power profiles in Th-MOX pins differ significantly from those of UOX fuel in two respects:

- The high absorption cross section for thermal neutrons in Th-232 gives a strong shielding effect for thermal neutrons, resulting in a power profile early during irradiation which is more strongly peaked towards the pellet periphery, compared with that of UOX fuel. Also the plutonium isotopes, most notably Pu-240, have large resonances around 1 eV, enhancing this effect.
- The smaller resonance capture in Th-232 gives a weaker shielding effect for epithermal neutrons, compared with UOX fuel. This gives a more homogeneous buildup of new fissile material (U-233) in the fuel pin, giving a less periphery-peaked power profile at higher burnup.

The differences between the power profiles of the respective fuel types are illustrated in Figure 5.12.

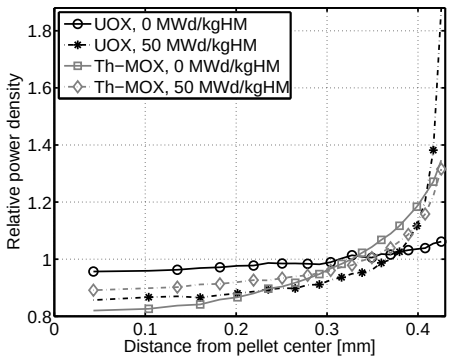

Figure 5.12: Radial pellet power profiles for UOX and Th-MOX fuel at 0 and 50 MWd/kgHM. The UOX fuel is enriched to 5% and the Pu concentration in the Th-MOX fuel is 8% with 80% Pu-239.

In order to calculate more representative power profiles while maintaining the simplicity of the TUBRNP subroutine, it was decided to stay with the one-group diffusion approximation, but to solve it numerically, accounting for the radial variation of the concentration of each isotope. The adopted solution scheme was the finite difference method with the boundary conditions of zero neutron current at the pellet center and the scalar flux at the pellet periphery equal to a normalization constant.

The one-group approximation cannot account for the fact that the presence of one isotope affects the flux which in turn affects the reaction rates for other isotopes. For example, the presence of Pu-239 hardens the spectrum since many thermal neutrons are absorbed by Pu-239, and the harder spectrum makes the one-group capture cross section of Th-232 lower than it would have been with no Pu-239 present. Neither can the approximation account for the radial variation in the neutron spectrum which is the direct consequence of the thermal shielding effect of Th-232, absorbing thermal neutrons already at the periphery of the pin and leaving less thermal neutrons to cause fissions at the center of the pin. The shielding effect thus cannot be directly modelled by the one-group diffusion approximation.

However, the application of TUBRNP for Th-MOX is fairly narrow. Only a small range of pin radii, a relatively small set of fuel compositions and neutron spectra typical for either LWRs or heavy water reactors need to be considered[1]. A sensitivity study was made, concluding that the pellet radius and composition and the neutron spectrum were the only parameters significantly affecting the power profile (Fredriksson 2014). This

[1]The type of reactor, LWR or HBWR (The Halden research reactor) is specified by a switch in the FRAPCON input.

means that the solution can be fine-tuned to fit the specific application, which was done by two means. Firstly, the numerical values of the one-group cross sections were adapted to give the best possible results. Furthermore, new shape functions were derived for the most significant isotopes, Pu-239 and Th-232. Since the diffusion equation was solved numerically, allowing for different cross sections in different nodes, the shape functions were multiplied directly with the macroscopic cross sections. THis differs from the original scheme in FRAPCON 3.4, where the shape functions were only used in the step where the isotopic concentrations were updated.

The adaptation of the numerical values of the one-group cross sections was made by means of a genetic algorithm, as described in Paper VII. The process is described in more detail by Fredriksson (2014). Reference power profiles at burnups up to 70 MWd/kgHM were calculated with *Serpent*, for a set of 61 different pin configurations representing a realistic range of plutonium isotope vectors, plutonium fractions and pin radii, in LWR conditions. A second reference set was generated for the conditions in the Halden research reactor, given that test irradiation data from this reactor will be used for benchmarking. The genetic algorithm was then used for adjusting the one-group cross sections and the shape function parameters used in TUBRNP, so that the reference data were reproduced as closely as possible.

The initial guess for the one-group cross sections was based on the corresponding values used by Long (2002) in the THUPS subroutine, which is a version of TUBRNP adapted for (Th,U)O_2 fuel performance simulations. It has to be noted that the differences between the adjusted set of cross sections and this initial guess were expected to be small, but this was not the case. Whereas some of the cross sections remained similar, others, such as the capture cross section of Pu-240, were widely different from their original values, indicating that these simplified neutronic calculations did not correctly model isotopic concentrations or reaction rates. Nevertheless, the reference power profiles generated by *Serpent* were well reproduced, so the new model and modified cross sections were adopted. However, the new TUBRNP version is to be regarded as empirical, so only the calculated power profiles and no the isotopic concentrations are to be used.

5.2.5 Comparison with irradiation data - Paper VIII

The modified FRAPCON 3.4 version was used in its current state of development to calculate the fuel centerline temperatures of the Th-MOX rods currently being irradiated in IFA-730. Input files were generated using all the data available for the irradiated pellets and their cladding. As a reference, also the *UOX-6* pin was modelled, using the original FRAPCON 3.4. The power histories were taken from the corrected and calibrated HELIOS calculations made by IFE, but increased by 12%, in order for the calculated power of the *UOX-6* pin to agree with the measured data. There are two justifications for this adjustment:

Firstly, there is an uncertainty of about 5% in the calibration of the total rig power (not affecting the power distribution between the individual pins in the rig). The systematic

underprediction of the temperatures for all fuel rods when using the power levels given by IFEs calculations indicated that the total rig power is indeed higher than assumed, which motivates the use of a higher power for the temperature calculations.

Secondly, FRAPCON 3.4 is known to underpredict fuel temperatures for Halden research reactor conditions by up to about 7% (Geelhood et al. 2011b). The exact reason for this is unknown, but it is likely that a systematic underprediction at Halden irradiation conditions would affect UOX rods and Th-MOX rods similarly, which is why the input power is adjusted in order to eliminate this factor.

The fuel temperatures calculated with the adjusted power history are plotted in Figure 5.13 together with the measured data. It can be seen that the calculated temperatures for the Th-MOX rods are underpredicted by about 30 K, which could be explained by the fact that the ThUOX filler material located above and below the Th-MOX pellets has a considerably higher temperature, heating also the Th-MOX pellets through heat conduction in the axial direction. It can also be noted that the temperature predicted by FRAPCON 3.4 for the UOX fuel remains good until about 300 irradiation days, after which the temperature is clearly overpredicted. A possible explanation is that the pellet-cladding gap has closed, greatly improving heat conductance from the pellet to the coolant. FRAPCON 3.4 does not predict gap closure yet for this fuel pin, but the indications that the gap is nearly closed for rod *ThAdd-4* strengthen this hypothesis.

Initially, a trend of increasing underprediction of the Th-MOX rods was clearly seen, growing to almost 100 K within the current irradiation period, that is to a burnup of 25 MWd/kgHM. Since the agreement between the calculated and measured temperatures was good at the beginning of life, causes were sought among the burnup dependent factors affecting the fuel centerline temperature. A sensitivity analysis was performed, including the effects of swelling, relocation, power profile prediction and the burnup dependent term in the expression for the thermal conductivity. Causes were also sought in the neutronic modelling used for the power predictions, since an error in the predicted power of the Th-MOX segments relative to the other materials would give an error in the calculated temperature which is based on the predicted power history. It was concluded that none of the three former factors could account for the observed underprediction, whereas an increase of the burnup dependent factor of the thermal conductivity resulted in the fair agreement shown in Figure 5.13. No error could be found in the neutronic simulations, and the scram time constants plotted in Figure 5.5 strengthen the hypothesis that the increasing temperature is a real phenomenon, since these are independent of the power levels. As can be seen, the time constants for the Th-MOX rods are increasing with burnup, indicating a decreasing thermal conductivity (the two unexpectedly low time constants for rod *ThMOX-3* at 240 and 292 days have very large uncertainties). However, it cannot be excluded that a gradual increase in the temperature contribution from the axial heat conduction from the ThUOX material causes part of the oserved increase of the underprediction, so a good estimate of the burnup dependent term in the expression for the thermal conductivity cannot be made based on only this set of experimental data.

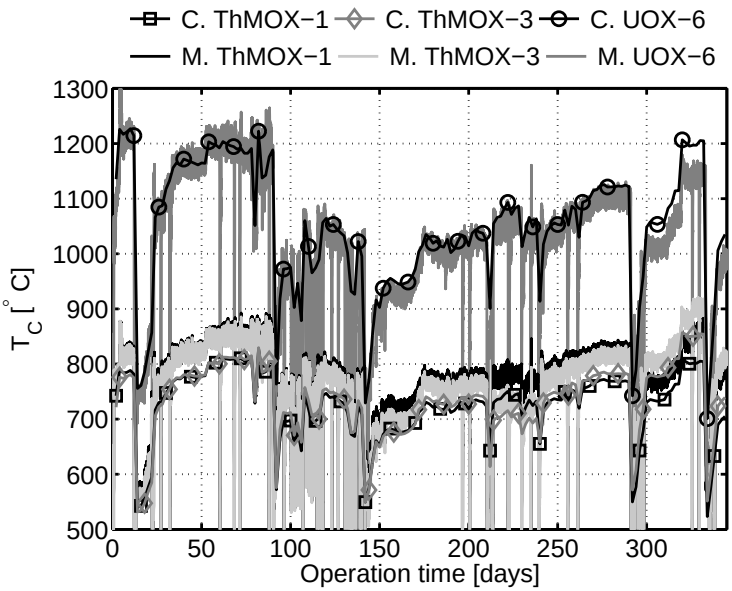

Figure 5.13: Calculated (C) and measured (M) fuel centerline temperatures for rods *ThMOX-1*, *ThMOX-3* and *UOX-6*.

The conclusion of the work so far is that existing data for fresh Th-MOX material[2], combined with the new model for radial power profile predictions and the simulation methodology of FRAPCON 3.4 as a whole, are adequate for predicting the temperature of fresh Th-MOX material. The current results indicate that an increase of the burnup dependence of the thermal conductivity of Th-MOX gives adequate predictions also at higher burnups. The implication that the thermal conductivity of the Th-MOX material decreases more rapidly with burnup than that of UOX material is reinforced by the time constant measurements. This would cast some doubt on the previously held assumption that Th-MOX fuel can indeed sustain higher burnups than UOX and U-MOX fuel. However, this is only one data set and, as noted, the experimental uncertainties are large. Additional test irradiation data sets, preferably on full Th-MOX rods, are required in order to disentangle the many different factors affecting the fuel temperature. As previously noted, several other types of behaviour such as FGR and dimensional changes also remain unadressed. Hence, much more validation work is needed before the code can be used for fuel performance predictions in a commercial context.

[2]It should be noted that the irradiated pellets are from the same batch that was used for the determination of the implemented thermal conductivity correlation, i.e. in the work by Cozzo et al. (2011). The calculated thermal conductivity can thus be expected to correspond very well to the conductivity of the irradiated pellets, but possibly worse for other pellets.

CHAPTER 6

Summary and Future Work

In this final chapter, the previous chapters are summarised and the results are discussed from the perspective of how well the goal stated in Chapter 1 is fulfilled. Finally, some relevant research activities are outlined which would be suitable for complementing and extending the work presented.

6.1 Summary

The initially stated goal of the work reported in this thesis was to take some of the steps remaining before thorium based nuclear fuels can be commercially used in LWRs. Two research areas were identified as most relevant in order to both provide motivation for using thorium based fuel and to expand the knowledge base for thorium fuel where needed to provide a basis for nuclear fuel licensing. These were the neutronic behaviour and the thermal-mechanical behaviour of thorium-containing fuels. The whole process and the conclusions are summarised below.

6.1.1 Choice of fuel types

At the outset, several different thorium containing fuel types were considered. An initial scoping study singled out thorium-plutonium fuel (Th-MOX) as the most interesting alternative (Paper I). One reason for making this decision was that the fuel seemed to

have neutronic properties similar to those of commonly used U-MOX fuel, i.e. using it in LWRs seemed feasible. The other reasons were rather related to considerations of nuclear waste management and possible transitions to purely thorium based breeding fuel cycles in LWRs.

Another approach, to use thorium as an additive in BWR UOX fuel (Th-Add), was also investigated, inspired by corresponding calculations carried out for PWRs. This alternative has to date been investigated to a much lesser extent, but preliminary indications are that the requirements for burnable absorbers can be significantly reduced, improving bundle-internal power peaking factors. The high conversion in Th-232 leads to smaller reactivity differences between fresh and irradiated fuel and consequently smaller core power peaking factors and larger shutdown margin.

6.1.2 Neutronic properties of Th-MOX fuel

A necessary prerequisite for the licensing of thorium based fuels is to provide confidence that the neutronic behaviour of the fuel can be adequately modeled (Paper II). A step in this direction was taken through the participation in a benchmarking activity, where predictions of four different codes were compared with experimental data. Due to experimental uncertainties, the benchmark exercise could not fully satisfy its goal to provide confidence in the modelled results through experimental confirmation. Nevertheless, two conclusions could be made regarding the performance of CASMO-5. One is that the power of Th-MOX fuel relative to that of U-MOX fuel was predicted similarly by three of the codes, including CASMO-5, which provides some confidence in this respect. Furthermore it was concluded that the radial distribution of the reaction rates characteristic to the thorium chain were not well modeled, an issue which has been attended to in later releases of CASMO-5.

The neutronic properties of Th-MOX fuel were further investigated using CASMO-5. PWR lattice simulations confirmed its similarity with U-MOX, with the exception of the coolant void reactivity, which was significantly more negative for Th-MOX than for U-MOX at high plutonium contents (Paper III). It was also noted, for both MOX fuel types, that the high conversion could give rise to advantages in the context of operating cycle extensions. Thus, full-core simulations were performed investigating this possibility, and the conclusions were that this application of Th-MOX fuel seems indeed to be feasible (Paper IV).

It is generally known that plutonium containing fuel benefits from a higher moderation ratio than UOX fuel, and the possibility to increase the moderation in a BWR fuel assembly has been investigated. It was concluded that both the energy generated from a fixed amount of plutonium and the incinerated fraction of the loaded plutonium could be increased in this way. Acceptable safety parameters, as evaluated by neutronic lattice simulations, were obtained, but the discharge burnup of the fuel assembly had to be significantly increased to reach this result (Paper V).

6.1.3 Thermal-mechanical properties of thorium fuel materials

Th MOX pellets and material representative of the Th-Add application have been irradiated in the Halden research reactor (Paper VI). The conclusions for the Th-Add material is that it behaves similarly to UOX fuel and may, surprisingly, exhibit lower fuel temperatures. The recorded centerline temperatures of the Th-MOX pellets have been used for initial validation of a thermal-mechanical fuel performance code for Th-MOX fuel.

The fuel performance code FRAPCON 3.4 has been modified, introducing new algorithms for calculations of pellet power profiles (Paper VII) and Th-MOX material properties. The initial validation shows good reproduction of fresh fuel temperatures, and assuming a faster decrease of the thermal conductivity of the Th-MOX material the predictions are also good for higher burnups (Paper VIII).

6.1.4 Conclusion

It can be concluded that the stated goal has been fulfilled since some steps have indeed been taken towards the commercial use of thorium fuels in LWRs:

- The experience base for neutronic modelling of thorium based fuel has been extended and some conclusions could be drawn which have lead to improved modeling of thorium based fuel in CASMO-5.
- Detailed full-core simulations of Th-MOX fuel have been performed, indicating the feasibility of Th-MOX fuel use for operating cycle extension, providing a motivation for reactor owners to consider this alternative.
- A novel concept for thorium use as an additive to UOX fuel for BWRs has been developed, providing another potentially attractive option for thorium fuel usage.
- The scarce irradiation experience of thorium based fuel has been extended through an instrumented irradiation experiment performed in the Halden research reactor.
- The work towards establishing a fuel performance code for prediction of thermal-mechanical performance of thorium based fuel has been initiated.

6.2 Future work

Some of the work described herein is currently being extended with continued research. These projects include:

- The irradiation of IFA-730 is ongoing, accumulating more data and higher burnup information. The project is scheduled to continue at least until 2017.
- Th-MOX fuel manufacture is being prepared at the IFE Kjeller, with the first Th-MOX trial run scheduled for March 2015.
- A second phase of the irradiation program is being planned, including Th-MOX pellets manufactured at IFE Kjeller and two more (Th,U)O_2 compositions representative of the BWR application.
- The reasons for the unexpected signals from the irradiated Th-Add rods in IFA-730 are being sought by more detailed modelling.
- The detailed properties of Th-Add fuel is being evaluated by full-core simulations, using a model of a currently operating BWR.

In addition to these ongoing projects, the following research activities would be highly relevant for taking the next steps towards thorium fuel adoption:

- The thermal conductivity of (Th,U)O_2 with low ThO_2 fractions should be measured directly, since there is a paucity of data for these compositions.
- The development of a fuel performance code for Th-MOX must be finished, and complemented with a code for transient fuel performance simulations.
- The detailed properties of spent thorium fuels must be evaluated from the viewpoint of intermediate and possibly also final storage.
- The argument that thorium fuel usage in today's LWRs is a step towards later adoption of advanced breeding LWRs is of course only valid if reprocessing technology for thorium based fuel is being developed. Research on the THOREX process is being done in India, but parallell efforts may be relevant.

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