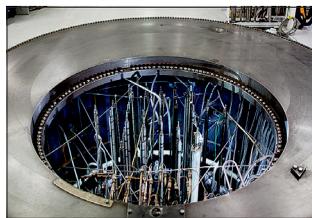
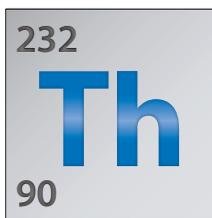


Introduction of Thorium in the Nuclear Fuel Cycle

Short- to long-term
considerations



Nuclear Science

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Cover photos: View of the Halden reactor hall (IFE, Norway); the CANDU fuel bundle (AECL, Canada).

Foreword

Since the beginning of the nuclear era, significant scientific attention has been given to thorium's potential as a nuclear fuel. Although investigations have been carried out on thorium-based fuels, and thorium-fuelled power reactors have operated in the past, the thorium fuel cycle as a whole has never been fully developed. Today, the opportunities and challenges that could arise from the use of thorium in the nuclear fuel cycle are still being studied in some countries and in the context of diverse international programmes around the world.

In parallel with the rise of various thorium advocacy fora in recent years, the use of thorium as a nuclear fuel has been gaining considerable attention in the media, where it is sometimes portrayed as a ready-to-use and risk-free solution to the world's energy problems. Such oversimplified representations of thorium's potential could however lead to misconceptions about thorium among decision makers and the general public. In reality, the thorium fuel cycle is a complex subject even for those familiar with nuclear technology. Any novel fuel cycle proposal must be assessed not only from a multidisciplinary, scientific perspective, but also from economic and industrial points of view, each within the broader context of well-established nuclear energy strategies.

Following discussions on these issues in the NEA Nuclear Science Committee (NSC) in June 2013, the NSC encouraged the drafting of a balanced and scientifically sound study of thorium's potential role in nuclear energy in the short to longer term. This report offers an assessment of the diverse options, opportunities and current impediments to be considered if thorium fuel cycles are to be pursued. It relies on contributions from a large NEA network of international experts, in particular members of the NEA Working Party on Scientific Issues of the Fuel Cycle (WPFC), the NEA Working Party on Scientific Issues of Reactor Systems (WPRS) and the Generation IV International Forum Molten Salt Reactor System Steering Committee (GIF/MSR).

The key findings of this assessment were discussed at the policy debate on thorium fuel cycles during the NEA Steering Committee meeting in October 2014.

Acknowledgements

The NEA would like to acknowledge all the contributors to this report and would especially like to thank Gregg Butler, Stephanie Cornet, Thierry Dujardin, Dominique Grenèche, Claes Nordborg, Gerald Rimpault and Luc Van Den Durpel for their useful input, which served to enhance the overall quality of this report. Franco Michel-Sendis offered valuable support in co-ordinating the report.

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List of abbreviations and acronyms

AECL	Atomic Energy of Canada Limited
AHWR	Advanced heavy water reactor
BWR	Boiling water reactor
CANDU	Canada Deuterium Uranium
CEA	French Alternative Energies and Atomic Energy Commission (Commissariat à l'énergie atomique et aux énergies alternatives)
CVR	Coolant void reactivity
FGR	Fission gas release
FR	Fast reactor
GIF	Generation IV International Forum
GIF/MSR	GIF Molten Salt Reactor System Steering Committee
HDBP	Di-butyl phosphoric acid
HEU	Highly enriched uranium
HTR	High-temperature reactor
HWR	Heavy water reactor
IAEA	International Atomic Energy Agency
IFE	Norwegian Institute for Energy Technology
LER	Linear element rating
LEU	Low-enriched uranium
LWR	Light water reactor
MA	Minor actinide
MOSART	Molten salt actinide recycler and transmuter
MOX	Mixed-oxide fuel
MSFR	Molten salt fast reactor
MSR	Molten salt reactor
NEA	Nuclear Energy Agency
OECD	Organisation for Economic Co-operation and Development
ORNL	Oak Ridge National Laboratory

PHWR	Pressurised heavy water reactor
PT-HWR	Pressure tube heavy water reactor
PUREX	Plutonium-uranium redox extraction
PWR	Pressurised water reactor
R&D	Research and development
REE	Rare earth element
SIMFUEL	Simulated high burn-up nuclear fuel
TBP	Tributyl phosphate
TD	Theoretical density
THOREX	Thorium-uranium extraction
Th/Pu	Thorium/plutonium
TMSR	Thorium molten salt reactor
TRU	Transuranic elements
UOX	Uranium oxide

1. Introduction

From the early days in the development of nuclear energy, thorium was considered a potential fuel that could possibly supplement or even replace natural uranium, which at the time was considered to be scarce. The New Piles Committee, created in April 1944 in the United States to explore a variety of reactor concepts (then called “piles”), recommended that “more work be done on the nuclear development of thorium because of its greater availability” (Lawson and Krause, 2004). The New Piles Committee also suggested experiments to develop reactors that would convert thorium to ^{233}U . However, in the years that followed, it was discovered that the supply of natural uranium was not as limited as projected. Also, thorium lacks a fissionable isotope: it is impossible to start any fission chain reaction purely on mined thorium, and therefore any nuclear system relying on thorium would be initially dependent on prior generation of fissile matter (extracted from uranium or bred in uranium systems). Faced with these two factors, interest in thorium declined or fell into abeyance.

The situation with uranium was entirely different; natural uranium, which contains 0.71% fissile ^{235}U , provided the fuel for heavy water and carbon moderated reactors which did not require any enrichment of ^{235}U . Moreover, the necessary enrichment capacity to provide the enriched uranium fuel required to operate light water reactors (LWRs) also existed. The plutonium generated in all these uranium-fuelled reactor types provided an obvious route for developing fast breeder reactors, which could use the plutonium as fuel and generate, from ^{238}U , more plutonium than they consumed. During the early days of nuclear energy, starting the uranium/plutonium fuel cycle was therefore the obvious route to “fuel independence” for the main nuclear developing countries. Today, the availability of fissile material (plutonium or enriched uranium) that arises from the well-established uranium/plutonium fuel cycle makes the implementation of thorium fuels feasible in principle, although the necessary economic drivers for devoting significant industrial resources to that end are not yet clearly established.

It is worthwhile to mention that the initial idea of “thorium converters” was submitted in early 1944 by Eugene Wigner, as an option for producing an atomic bomb using ^{233}U as the fissile material. The proposal of using thorium for nuclear reactors was then brought up again in the framework of the New Piles Committee, which included the most eminent physicists and chemists working in the area of nuclear reactors. It should also be noted that, at that time, nuclear reactors were an entirely new technology, since the first man-made fission chain reaction had occurred less than two years before.

It is therefore perfectly accurate to say that the use of thorium has been considered as an option for nuclear fuel since the advent of nuclear energy. Thorium cannot be a real alternative to uranium, however, at least at the initial stage, because it has no fissile isotope. Thorium does, nonetheless, generate the fissile nuclide ^{233}U by neutron capture in a nuclear reactor. ^{233}U is an excellent fissile nuclide, with advantages over the ^{235}U or ^{239}Pu used in current reactors. This inherent “neutronic attractiveness” of ^{233}U , particularly in thermal neutron spectrum reactors, makes the use of thorium scientifically interesting, despite the multiple challenges yet to be resolved technically and industrially.

During the pioneering years of atomic energy, from 1950 to 1970, the thorium fuel cycle was the subject of numerous studies. The thorium option has never been fully discarded and “the thorium fuel cycle” has, albeit with fluctuating intensity, continuously been studied worldwide.

1.1 Past incentives and hindrances in the development of the thorium fuel cycle

The main initial motivation for the development of thorium fuel was to provide a fuel cycle that could, by replacing uranium fuel, avoid the potential of natural uranium shortage in the event of the expected rapid growth of nuclear power. Indeed, during the rapid expansion of nuclear power in the mid-1970s, the price of uranium reached a high point of around USD 40/lb U_3O_8 , reinforcing the fear of a uranium shortage. An additional factor was the likely abundance of thorium resources in nature, with the average concentration of thorium in the earth's crust estimated to be about three times that of uranium.

Apart from thorium's abundance in nature and the prospect of ^{233}U breeder systems, the essential reasons for the interest in thorium were the intrinsically good basic physical properties of both ^{232}Th and ^{233}U , including:

- good neutronic properties;
- generally good behaviour under irradiation of thorium-based fuels;
- the possibility of optimising the overall ratio and inventory of fissile matter produced in comparison to the fissile matter consumed by a reactor at equilibrium, which could make thorium-based fuels attractive from fuel cycle and waste management perspectives.

Nevertheless, several factors dimmed the enthusiasm for fuel cycles requiring the recycling of ^{233}U as would be required in the longer term for so-called pure "thorium fuel cycles". Since ^{233}U is associated with ^{232}U , whose decay chain includes high-energy gamma emitters, research, development and demonstration work on thorium fuel recycle must take place in remotely operated shielded facilities, which are far more expensive than the glovebox installations required for the uranium/plutonium cycle. This would have made thorium cycle operations prohibitively expensive for most sources of funding, especially in comparison to the processes associated with the uranium cycle. The picture becomes more balanced if the uranium/plutonium cycle moves to optimising waste management by investigation of partitioning and transmutation schemes of minor actinides (MAs), but these possibilities had not yet become mature technical options.

The early years of nuclear energy deployment were not, in general, limited by questions of natural uranium availability¹ or considerations in relation to the back end of the fuel cycle; specifically ultimate waste management schemes. In some countries, such as France, the deployment of the uranium/plutonium fuel cycle was in progress with the introduction of prototype fast reactors intended for multiple recycling of plutonium as well as the development of LWR mixed-oxide (MOX) fuel technology and fuel cycle facilities that would facilitate better management of fissile materials and ultimate waste.

In the late 1970s and 1980s, interest in the nuclear option weakened significantly, particularly in the United States, where public support for nuclear power was significantly affected by the Three Mile Island accident in April 1979. Public misgivings were then intensified in Europe after the Chernobyl disaster seven years later. Over this same period, the price of uranium had fallen to very low levels in the early 1980s, weakening the economic case for fuel cycles using recycling. This also applied to the thorium cycle, even though other potential benefits were envisaged. In the United States, the late 1970s saw the Ford and Carter administrations banning the reprocessing of commercial irradiated fuels due to the perceived risk of proliferation of the fissile

1. The lack of indigenous uranium resources greatly affected the Indian nuclear programme, as discussed in Section 1.1.1.

material separated during these operations. Most importantly, this reprocessing ban effectively blocked the possibility of closing any fuel cycle in the United States, with impacts on the decision making in other countries. Driven by the same proliferation fears, the use of enriched uranium beyond 20% in civilian reactors was abandoned throughout the world.

These situations and decisions discouraged the implementation of thorium cycles for three main reasons:

- The promoters of the thorium cycle had to abandon the reference cycle that was based on the use of highly enriched uranium (HEU), by replacing it with uranium enriched to a maximum of 20% (low-enriched uranium, or LEU). This complicated the implementation of the thorium cycle significantly and reduced its overall performance.
- The ban on reprocessing spent fuel prevented the recovery of the ^{233}U from thorium spent fuel for recycling, at least in the United States.
- The uranium/plutonium fuel cycle was under full development, with implementation of partial recycling in some countries such as France. At the time, there appeared to be no limitations on the uranium/plutonium fuel cycle.

1.1.1 The case of India

India was an exception to the general trend. It has almost no domestic uranium resources (only 1% of world uranium resources) and India's capacity to import enriched uranium fuel on the international market has been historically limited, although this situation has changed in recent years. However, India possesses relatively large amounts of thorium and from the beginning of its nuclear programme, it pursued the development of a thorium cycle that could supply indigenously sourced fuel for a long-term nuclear programme (Bhardwaj, 2013).

The Indian programme is structured around three stages, reflecting a strategy aimed at the optimal use of available fissile materials and neutrons within the Indian nuclear power programme:

- Initial deployment of heavy water moderated and cooled reactors, which use only natural uranium, do not require enrichment technology and can produce plutonium for later use. The opening of the uranium market to India also facilitates the deployment of LWRs, providing another source of plutonium for the next stages of fuel cycle development.
- The gradual start-up of fast neutron reactors using plutonium-based fuel with thorium blankets to generate ^{233}U . The construction of a 500 MWe prototype fast breeder reactor is underway at the Kalpakkam site and commissioning is expected in 2015, with other fast reactors (FRs) being scheduled for deployment from the 2020s onwards.
- The construction of a new type of advanced heavy water reactor (AHWR) using thorium/ ^{233}U fuel with recycle and having a very high conversion factor (close to one). In the longer term, dedicated molten salt reactors (MSRs) may also be considered for this stage.

Experimental programmes have been established to prepare for the implementation of the above-mentioned concepts. Thorium oxide pellets have been irradiated in research reactors, including fast neutron reactors, and have been processed using a simplified method for recovering the ^{233}U , which has then been used in other research facilities. Thorium fuels have also been introduced in heavy water power reactors. The Kakrapar 1 and 2 reactors were loaded with 500 kg of thorium and operated for 300 and 100 days respectively with this load. The use of thorium fuel is planned in the Kaiga 1 and

2 reactors as well as in the Rajasthan 3 and 4 reactors, which are now in commercial operation.

A 300 MWe prototype AHWR is also under development. It will be fuelled by a mixture of thorium oxide and plutonium oxide and also a mixture of thorium and ^{233}U (Sinah et al., 2000).

In this long-term strategy, reactors entirely fuelled by thorium/ ^{233}U would be deployed only in the third and final stage. Although time frames for this long-term strategy are rarely announced, deployment of the third phase is foreseen beyond 2070 (Vijayan, 2013), but could be much later.

In spite of this thorium-based strategy, a recently signed international agreement has given India access to natural uranium supplies, and India has purchased light water-cooled reactors on the global market.

1.2 Past and recent efforts in thorium research

Various international reviews have been undertaken on research conducted, or experiences with, thorium use in reactors. The following sections, while not exhaustive, highlight past work on thorium and recent international co-operation developments.

1.2.1 Thorium utilisation in thermal reactors

It is interesting to note that some of the first commercial LWRs developed in the late 1950s and early 1960s in the United States were initially operated with thorium-based fuels; notably, the Elk River boiling water reactor (BWR) and the Peach Bottom high-temperature reactor (HTR), which started operations in 1964 and 1967 respectively. The Fort St. Vrain HTR, which was a larger-scale commercial successor to the Peach Bottom reactor, also used thorium-based fuel from 1976 to 1989.

The Edison Indian Point-1 pressurised water reactor (PWR) started up in 1962 with an initial core of thorium seed commingled with fully enriched ^{235}U . The used fuel was reprocessed at the West Valley reprocessing plant in 1968 (Benedict, Pigford and Levi, 1981), using the Interim-23 process for recovery of the mixed uranium product solution (mostly ^{233}U , ^{235}U and ^{232}U), which was shipped to Oak Ridge National Laboratory (ORNL) for conversion to oxide, encapsulation and subsequent disposal.

Demonstration of the feasibility of an LWR breeder was carried out at the Shippingport PWR reactor in the 1970s and 1980s using a ^{233}U /thorium cycle (IAEA, 2005), where a conversion factor of 1.0139 (IAEA, 2000) was achieved. This was the only power reactor to use ^{233}U as fissile material and to operate at approximately 70 MWe. It was thus experimentally demonstrated that it is possible to produce more ^{233}U than is consumed in an LWR. However, this performance was only achieved by using a sophisticated reactor core design, which would be difficult to extrapolate to commercial reactors (e.g. absence of neutronic poisons or absorbers) and detrimental to other performance parameters (e.g. low burn-up).

Germany also studied the thorium fuel cycle from the early days of its nuclear industry, as an interesting alternative to the established uranium/plutonium fuel cycle. Several test programmes were initiated aiming at closing the knowledge gap, and focusing on the use of thorium in LWRs and HTRs, such as the thorium HTR (THTR-300). This latter reactor, which had its first criticality in 1983, started up using thorium/uranium fuels.

There have been four phases of investigations on thorium utilisation in LWRs in Germany:

- The early phase, from 1968 to 1977, with the co-operation of industry and research institutes in the investigation of possibilities for plutonium recycling as thorium/plutonium MOX in PWRs.
- Various research and development (R&D) programmes funded by the German Federal Ministry for Research and Development on the irradiation of thorium/plutonium fuel in the BWR at Lingen (KKL).
- Co-operation between Kraftwerk Union AG, KFA-Jülich, NUKEM and NUCLEBRAS/CDTN, Brazil from 1979 to 1988 and financed by the German Federal Ministry for Research and Development.
- The irradiation of a test rod with thorium/plutonium (Th/Pu) fuel in the Obrigheim Power Station from 2000-2005. This project was performed within the Fifth Framework Programme of the European Union and continued in the following framework programme.

More details about these programmes are provided in Section 4.1.

1.2.2 Recent R&D or international co-operation programmes on thorium fuels

The feasibility of introducing thorium-based fuels into established, readily licensable platforms (LWR, heavy water reactors [HWRs]) has been a key motivating factor for a number of R&D and irradiation programmes that have either been recently carried out, are ongoing, or planned. In the context of near-term use of thorium in existing reactors, the following recent R&D efforts are summarised, and further details are given in Chapter 4:

- In 2013, the Norwegian technology company, Thor Energy, started an irradiation programme in the Halden reactor aimed at qualifying thorium/plutonium fuel for LWRs. This programme is overseen by the Norwegian Institute for Energy Technology (IFE) at Halden, host of the NEA Halden Reactor Project. Instrumented test-rods containing candidate thorium fuel specimens are currently undergoing irradiation trials in the reactor, which is able to emulate PWR or BWR operating conditions. Key properties of thoriated fuels such as thermal conductivity, or swelling and fission gas release (FGR) as a function of burn-up are to be determined in these irradiation tests. The trial operation of a commercial prototype thorium-MOX fuel in the Halden test reactor is a significant step towards the broader use of thorium fuel for achieving “near-term” fuel cycle goals, most notably with respect to minimisation of transuranics in spent fuel.
- In late 2013, Areva and Solvay/Rhodia announced a common agreement to develop an R&D programme on the use of thorium as a potential complementary fuel to the present uranium/plutonium cycle in nuclear plants. Both companies have inventories of thorium in France originating from uranium or rare-earth extractions. The R&D programme is geared towards the development of qualified thorium fuels for use in generation III reactors and plans for irradiation testing in commercial NPPs. In addition, the programme includes R&D for the back end of fuel cycle operations, ultimately allowing recycling of the fissile materials.
- Canada has for many years been involved in R&D of thorium fuels with added fissile components, such as uranium or plutonium. To further identify and address present knowledge gaps in the understanding of thorium fuel technology, Atomic Energy of Canada Limited (recently renamed Canadian Nuclear Laboratories, CNL) has initiated a “Thorium Roadmap Project” covering most aspects of the thorium fuel cycle. The Thorium Roadmap Project includes a phase in which activities identified in the roadmap will be undertaken.

- In the context of international co-operation, the IAEA has an existing Co-ordinated Research Project (CRP) on *Near-Term and Promising Long-Term Deployment of Thorium Energy Systems* (IAEA, n.d.).

1.2.3 Thorium and molten salt reactor-related research

The Molten Salt Reactor System Steering Committee (MSR/SSC) was established in 2010 to serve as an information exchange framework so as to foster international collaboration around research conducted on MSRs under the Generation IV International Forum. France (Alternative Energies and Atomic Energy Commission – CEA) and Euratom were official MSR/SSC founding members, with the Russian Federation joining officially in 2013. The committee also welcomes the United States, the People’s Republic of China, Korea and Japan as regular observers. The MSR/SSC currently carries out viability studies centred on MSR concepts that utilise thorium in the composition of the liquid salt fuel, but it is not exclusively focused on liquid fuel systems. A recent review of these studies can be found in Boussier et al. (2012), and an overview of current status of MSR research is given in Chapter 6.

In 2011, China announced the start of an ambitious R&D programme led by the Chinese Academy of Sciences with the creation of the thorium molten salt reactor (TMSR) Research Centre at the Shanghai Institute of Applied Physics. In particular, the TMSR centre has announced the construction of two small pilot demonstrators: a 2 MW TMSR-SF (“solid fuel”: a pebble-bed reactor that will use molten salts only as a coolant, initially announced for 2017) and a 2 MW pilot TMSR-LF (“liquid fuel”: a reactor which will use molten salt as fuel), currently still in a pre-conceptual design phase.

1.3 Renewed interest in thorium

In the mid-1990s, thorium became the subject of renewed interest at an academic research level in several countries, albeit with diverse resources and technical focuses. Many of these research efforts were carried out in the broader context of research on innovative reactors, in particular MSRs, where the use of thorium in liquid fuel form brings significant theoretical advantages, as well as HTRs with thorium fuels, including the option for Pu-burning. These programmes have in the past been limited to academic research, with limited technological development and have had little impact on movement towards significant industrial applications.

Interest in the thorium cycle was renewed in the so-called “nuclear renaissance” period during 2000 to 2010. In the context of expected growth of nuclear power worldwide, new questions emerged on the sustainability of nuclear energy, driven in particular by doubts concerning the long-term availability of cheap uranium and the sustained management of used fuels and radioactive waste. However, this nuclear renaissance was seriously weakened by the Fukushima Daiichi accident in March 2011, resulting in nuclear power programmes being re-examined worldwide. Notwithstanding, the fundamental driver of this renaissance, the decarbonising of the world’s energy production to limit climate change, still remains valid.

Increased importance has been given to the back end of the nuclear fuel cycle, in particular with regard to plutonium management and especially ultimate waste management. Given the delays and uncertainties occurring in the realisation of geological repositories, a growing socio-political debate on nuclear energy since around 2000 has largely focused on the longevity of the ultimate waste forms. In this context, increased interest has been generated in advanced schemes for the back end of the fuel cycle, aimed at drastically reducing the quantity and longevity of the waste to be ultimately disposed. In the United States, such schemes have gained particular attention, with the cancellation of the Yucca Mountain deep geological repository. Given thorium’s ability to serve as matrix material and its intrinsic potential for reduced MA production, thorium has increasingly emerged as an additional enabler for some of these advanced

fuel cycle options. Such options would build on the previously mentioned HTR thorium/plutonium fuel studies undertaken during the 1990s in the EU, which were seeking increased Pu-burning capabilities for the incineration of plutonium inventories.

In the past 15 years, there has also been a focus on thorium-based fuels for nearer-term thorium utilisation in present or evolutionary reactors. Some research groups in universities, laboratories and industry investigated options for evolutionary progress towards more sustainable nuclear energy systems in the wake of possible delays in transitioning to generation IV systems; specifically, FRs and thus providing additional options in the management of uranium and plutonium. In addition, considering the very good physico-chemical characteristics of thorium, the potential role for thorium in so-called accident-tolerant fuels is also being investigated.

Thorium's strengths include:

- The possibility of reaching higher burn-ups with thorium-based fuels.
- The higher melting temperatures and improved thermal characteristics of thorium fuels, considered in the context of accident tolerant fuels.
- Lower production of plutonium and MAs in thorium/uranium fuels in general, and thorium's use as a robust fertile fuel matrix for consuming surplus plutonium (and possibly other actinide elements) from used-fuel inventories.
- The possibility of reaching higher conversion factors in thermal or epithermal neutron spectra, using thorium-based fuels in evolutionary systems (such as AHWRs), with a view to favouring the *in situ* consumption of ^{233}U in long residence time fuel assemblies, but also recognising future reprocessing scenarios.
- The use of thorium as an additive to uranium oxide (UOX) LWR fuels in order to improve fuel ceramic properties and in-core operation characteristics, and therefore improve operational safety margins.
- The synergies that arise from the above points with the uranium/plutonium fuel cycle and that can contribute to the recycling of fissile materials in thermal neutron reactor systems, with higher conversion factors made possible by introduction of thorium.

These potential advantages may explain the current interest in thorium in several academic and R&D institutions, as well as among industrial reactor designers and fuel vendors.

In recent years, thorium has also received significant media attention due to various thorium-advocacy fora, some of which portray thorium as a ready-to-use, problem-free solution to the world's energy problems. Independently of thorium's potential uses or advantages, the often unbalanced and unscientific representation of thorium in the media can only further obscure the issue of thorium's authentic potential and challenges to decision makers and the non-specialist public in general.

It is therefore important to demystify thorium by bringing the discussion on thorium's potential back into a scientific and technologically sound assessment of thorium's role in nuclear energy both in the short to long term, and by examining the various options and impediments that need to be considered. The main objective of this report is to provide a contribution to such an assessment.

References

- Benedict, M., T. Pigford and H. Levi (1981), *Nuclear Chemical Engineering*, 2nd Edition, Mcgraw Hill, New York, pp. 514-527.
- Bhardwaj, S.A. (2013), "Indian Nuclear Power Programme: Past, present and future", *Sadhana*, Vol. 38, Part 5, Indian Academy of Sciences, pp. 775-794.
- Boussier, H., et al. (2012), "The molten salt reactor in generation IV: Overview and perspectives", *Proceedings of "Generation IV International Forum Symposium"*, San Diego.
- IAEA (n.d.), Nuclear Fuel Cycle and Materials (web page), www.iaea.org/OurWork/ST/NE/NEFW/Technical_Areas/NFC/advanced-fuel-cycles-crp-thorium-2011.html.
- IAEA (2000), *Thorium Based Fuel Options for the Generation of Electricity: Developments in the 1990s*, IAEA TECDOC-1155, International Atomic Energy Agency, Vienna.
- IAEA (2005), *Thorium Fuel Cycle-Potential Benefits and Challenges*, IAEA-TECDOC-1450, International Atomic Energy Agency, Vienna.
- Lawson, C.G. and C. Krause (2004), "Documenting history: minutes of the new piles committee meetings", *Nuclear News*, www2.ans.org/pubs/magazines/nn/docs/2004-11-3.pdf.
- Sinah, R.K., et al. (2000), "Design and development of AHWR: The Indian thorium fueled innovative reactor", Status, Strategies and Directions INSAC, *Proceedings Ann. Conf.*, Mumbai, India, 1, pp. 81-106.
- Vijayan, P.K. (2013), "Overview of thorium programme in India", International Thorium Energy Conference, October 2013, CERN, Geneva.

2. Perspectives on the use of thorium in the nuclear fuel cycle

Improving the efficiency of utilisation of mineral resources (whether it is uranium or thorium) while reducing ultimate waste streams are among the major challenges that the nuclear energy industry must address if nuclear energy is to develop significantly and become a sustainable source of energy for the long term.

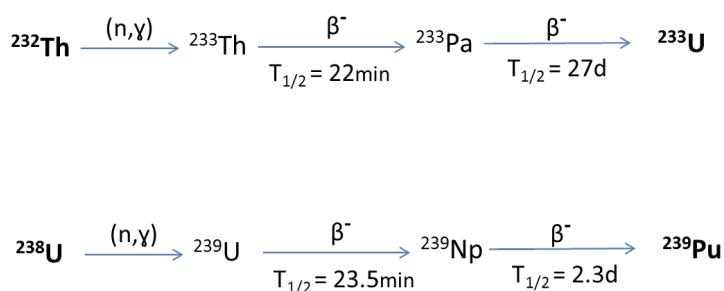
2.1 Basic considerations

2.1.1 Fertile/fissile cycles

Today, water-moderated reactor technologies are dominant in the world. According to IAEA statistics (IAEA, 2013), of the 437 power reactors in operation in 2013, more than 90% were water moderated: 273 are PWRs, 84 are BWRs and 48 are pressurised heavy water reactors (PHWRs). These technologies rely on the extraction of energy released by fissions triggered by thermalised neutrons. Heavy nuclides that fission under thermal neutron interaction are called “fissile”. Others are called “fertile”, because they are more likely to absorb a neutron instead of undergoing fission and thereby transmute into a heavier nuclide, which may or may not be fissile.

When irradiated by thermal neutrons, ^{232}Th and ^{238}U follow similar processes. ^{232}Th breeds ^{233}U in a completely analogous way to that in which ^{238}U breeds ^{239}Pu . These two processes of neutron “radiative capture” (n,γ) reactions, followed by consecutive radioactive decays (β^-) represent the two practical “fertile/fissile fuel cycles”; the $^{238}\text{U}/^{239}\text{Pu}$ fuel cycle and the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle (see Figure 2.1).

Figure 2.1. The thorium and uranium fertile/fissile cycles



2.1.2 Conversion ratios and breeding

Although requiring an initial fissile “seed”, a thorium fuel cycle may ultimately become autonomous in terms of fissile inventory as creation of the fissile nuclide ^{233}U occurs in fertile thorium, provided the retrieval of this ^{233}U is viable through reprocessing and separation. Under certain conditions, it is possible to create (or breed) more fissile material than is consumed by the reactor.

The conversion ratio, also called breeding ratio, is defined as the ratio of the rate of production to the rate of consumption of fissile nuclei in the fuel of an operating reactor.¹ If the conversion ratio is greater than or equal to 1, the system is called a “breeder”. Achieving a conversion ratio of one allows a reactor in a closed fuel cycle to function independently² from any continuous external supply of fissile material (assuming negligible losses at the reprocessing stage).

In terms of mineral utilisation per unit energy produced, there is much room for improvement. Today, in order to produce 1 GW-year of electrical energy in existing PWRs (in once-through utilisation), approximately 200 tonnes of natural uranium are used.³ The use of recycle plutonium in PWR-MOX fuels improves this ratio, but does not change the order of magnitude. HWRs such as CANDUs are intrinsically more efficient in their use of natural uranium (per unit energy produced) by approximately 30% to 40%, due to a better neutron economy with less neutron capture by the heavy water coolant/moderator. This facilitates the direct use of natural uranium in the fuel, without the tails produced during enrichment for LEU LWR fuel and, importantly, without ²³⁵U losses in the enrichment tails. However, the shorter, lower burn-up cycles of CANDU fuel bundles lead to an increase in volume of spent fuel production per unit energy produced.

Conversion ratio values of conventional PWR reactors today range from 0.5 to 0.6, with implementation of plutonium MOX recycle improving this value to around 0.7. Even if breeding is not achieved, moving towards higher conversion ratios in evolutionary designs is desirable in terms of better utilisation of natural resources. Hardening the neutron spectrum by decreasing the moderator-to-fuel ratio as much as possible without affecting cooling (water is both the moderator and coolant) is the principle behind high conversion in reduced-moderation water reactors concepts.

Thorium’s basic properties and, in particular, the neutronic properties of ²³³U in a thermal or epithermal spectrum (it has the highest number of spare neutrons available for breeding at these neutron energies) have led to the use of thorium being proposed and studied for many types of reactor concepts as they may allow higher conversion ratios. These concepts, often referred to as “evolutionary designs” of PWRs, BWRs, HTRs or HWRs generally seek higher conversion ratios with tighter fuel lattices, such as, for example, in modified CANDUs (Bromley, 2014; Bromley and Hyland, 2014) or potentially in modified BWRs (Ganda et al., 2012).

If the neutron spectrum is fast (no moderation), breeding is more easily achievable (in terms of neutron economy), in uranium/plutonium systems, due to the larger number of neutrons available for breeding generated by ²³⁹Pu (than those generated by ²³³U) in fast neutron induced fission. The concept of breeding is by no means exclusive to the thorium cycle and can therefore be achieved with the uranium cycle in fast neutron reactors. However, in this respect, thorium has an advantage over uranium cycles since it can in

1. The conversion ratio depends, therefore, on effective cross-sections, which vary during burn-up as the composition of the fuel evolves. A usual indicator of the conversion ratio used is the Fissile Inventory Ratio, which can be defined as the ratio of final over initial fissile mass established at discharge of the fuel.
2. A functioning breeder reactor would, in principle, only require the initial fissile inventory to reach equilibrium. Since fissile matter is consumed and produced at the same average rate in the core of a breeder reactor, the actual consumption of matter in the reactor would then only rely on the input of fertile matter into the fuel cycle. This fertile matter, either uranium or thorium, is much more abundant than fissile matter (i.e. in the case of uranium, ~140 times more ²³⁸U than ²³⁵U) and more readily available.
3. This roughly corresponds to the quantity needed to produce the 20 tonnes of conventional UOX fuel enriched to around 5% that will produce 1 GW of electrical energy continuously during one year before the fuel is discharged in once-through utilisation.

principle achieve conversion ratios close to or slightly in excess of unity in thermal (slow) neutron spectra due to the favourable inherent neutronic properties of ^{233}U in this neutron energy range.⁴

Tables 2.1 and 2.2 provide indicative⁵ average cross-section values of main nuclides in both thorium and uranium fuel cycles for two reference neutron spectra, thermal and fast.

Table 2.1. Neutron reaction cross-section values (in barns) averaged for a thermal (PWR) reference neutron spectrum

Thermal spectrum	^{232}Th	^{233}U	^{235}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu
Neutron capture (n,g) (b)	5.010	11.270	14.380	14.898	37.052	83.656	479.200	51.600
Fission (n,f) (b)	0.009	79.710	53.310	0.040	2.767	142.644	0.370	139.300

Note: Data computed with JANIS-4 using the JEFF-3.2 Neutron Data Library.

Table 2.2. Neutron reaction cross-section values (in barns) averaged for a reference fission neutron spectrum

Fast spectrum	^{232}Th	^{233}U	^{235}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu
Neutron capture (n,g) (b)	0.093	0.068	0.085	0.067	0.096	0.057	0.099	0.117
Fission (n,f) (b)	0.078	1.893	1.229	0.321	1.997	1.793	1.376	1.649

Note: Data computed with JANIS-4 using the JEFF-3.2 Neutron Data Library.

Achieving a conversion ratio of 1 allows a reactor using a closed fuel cycle to be in principle autonomous from any continuous external supply of fissile material, provided that the necessary initial fissile inventory is available to start and operate the system (also considering the necessary out-of-core inventories to ensure continuous operation during fuel reprocessing and re-fabrication). These are major hypotheses that should not be taken for granted, as closing fuel cycles (thorium- or uranium-based) still present major technical and commercial challenges, particularly for thorium as the reprocessing and re-fabrication of thorium fuels has yet to be demonstrated industrially.

Though simple neutronics may indicate breeding can be attained for a given reactor/fuel system, reaching this state in a real safety-assured reactor may be more of a challenge. For instance, the lower delayed neutron fraction offered by thorium fuels

4. It must, however, be noted that achieving breeding in thorium-fuelled light water moderated thermal systems is an extremely complex task, in particular for solid fuels. The Shippingport 60 MWe light water breeding reactor (LWBR) used Th/ ^{233}U fuel and demonstrated in the late 1970s the possibility of reaching breeding ratios slightly greater than one (1.014). The LWBR was indisputably an engineering success made possible by having very low burn-ups, which favours high conversion ratios, but sacrifices reactor performance, and implemented a costly and sophisticated operation of the core, notably by removing all neutronic poisons and control rods. Such a complexity is deemed unrealistic today for the safe or economical operation of reactors for electricity generation.
5. The averaged cross-section values in Tables 2.1 and 2.2 must be interpreted as indicative of the orders of magnitude only. The average is computed using a generic neutron spectrum which does not take into account, in particular, the modifications of the spectrum which would depend on the specific composition of the fuel considered and of its evolution over burn-up. This has a large impact on the shape of the spectrum, particularly in the thermal region, as caused by, for example, the presence of ^{240}Pu , a large resonant absorber.

(nevertheless comparable to plutonium MOX fuels), already points to the need for detailed transient safety assessments.

2.2 A necessarily progressive process for thorium fuels

Before identifying the different types of general strategies that could be conceived for thorium use in nuclear energy systems, it must be highlighted that the introduction of a new fuel into the industrial fuel cycle is a necessarily progressive process for a variety of reasons.

The development of qualified fuels (even considering evolutionary changes only) is a resource-intensive effort that necessitates a series of different steps, all of which are very time-consuming:

- the characterisation of basic physico-chemical data at laboratory scale;
- the qualification of reactor-physics and safety codes;
- the testing and qualification of fuel fabrication technology;
- the execution of irradiation experiments and integral testing;
- the post-irradiation analyses aimed at fuel rod/assembly characterisation;
- the re-evaluation of safety documents of reactors and fuel cycle facilities.

The licensing of new fuels, as for any other nuclear technology being introduced industrially, demands time, resources and a cost/benefit assessment, especially in the context of ensuring that nuclear power remains cost competitive relative to other energy technologies. Any transition, even in the well-known uranium/plutonium fuel cycle, demands a step-by-step phased approach and is likely to be decades in duration. The need for major changes leading to new fuel cycle facilities would result in even longer timescales.

The introduction of thorium into nuclear energy systems, if it occurs, will therefore have to happen progressively and none of the scenarios envisaging a full transition towards a “100%” thorium/²³³U fuel cycle in the near term are realistic, both for scientific and for industrial reasons.

Any industrial application of thorium as a nuclear fuel would continue to require the input of fissile material from the existing uranium/plutonium cycle until the required amounts of ²³³U could be produced and ultimately make the thorium cycle self-sustaining.

Until that point is reached, an important factor governing the rate at which ²³³U could be produced from the introduction of thorium/plutonium or thorium/uranium/plutonium cycles would be plutonium availability. The limitations imposed by fissile plutonium availability already point to rather long transition periods between thorium/plutonium and Th/²³³U systems, which are likely to be of the order of many decades.

2.3 Motivations for a change in the established fuel cycle

Overall, industry can only consider embracing thorium fuels in the context of a well-motivated strategic vision on the future of nuclear energy and, in this case, will need to assess the realignment of the whole supply chain towards the selected thorium fuel options and the industrial risks that may differ from those of the current uranium/plutonium cycle.

It is difficult to postulate that the perceived advantages of mitigation of uranium scarcity and/or reduced higher actinide production will emerge as immediate strong drivers to justify a transition towards a full thorium cycle in the short term. However, a variety of strategies and scenarios that consider a progressive introduction of thorium

can be regarded as being of interest today. These strategies combine a variety of possibilities stemming from current and future uranium/plutonium fuel cycle and future thorium fuel cycle options.

Three primary objectives can be identified that may be considered for thorium use in future nuclear energy systems:

- Continuous improvements in the management of fissile materials: in current and future fuel cycles that create opportunities from the short-term onwards, although with varying degrees of impact and benefits.
- Ultimate waste management objectives: thorium fuels may lead to less MAs production per energy generated; thorium may thus be used as an inert matrix for burning plutonium or even in a thorium fuel cycle leading to less MA-loaded ultimate waste streams, due also to the possibility that thorium fuels may allow longer burn-ups.
- The mitigation of natural resources and natural uranium scarcity: although this is not likely to happen in the near term, it can be considered a long-term issue (post-2050).

The scenarios or strategies that can be devised can then be classified under the main objective that motivates these scenarios and the timescale at which this motivation could come into play, as is schematically shown in Table 2.3 below.

Table 2.3. Different potential strategies of thorium utilisation at different time frames

Objective Time frame	Economy of natural resources	Management of fissile material	Management of ultimate waste	
Short term (pre-2030)	-	Th as “additive” or using separated natural thoria “fuel” rods		
Medium term (2030-2050)	-	RU-recycling Pu-recycling	Pu-burning	Deep Pu-burning
(Very) Long term (post-2050)	100% Th-cycle		Reduced MA-loaded waste	

The dark-shaded options explicitly require recycling schemes of the thorium-fuels. The light-shaded options may optionally implement recycling of the thorium fuels. RU stands for recovered uranium, uranium coming from reprocessing of spent uranium oxide fuel.

It is important to distinguish the need for reprocessing and recycling for the different thorium utilisation options that could be envisaged, given the technological challenges that these steps currently pose (see also Chapter 7 on spent fuel reprocessing). The recycling of bred ^{233}U , accompanied by the strong gamma-radiating ^{232}U , poses technological challenges associated with the industrialisation of any recycling thorium/ ^{233}U fuel cycle. Additional shielding measures for spent fuel containing ^{232}U will be required in subsequent fuel cycle steps to recycle ^{233}U into new fuel. This additional shielding will demand a technological step-change in today’s industrial fabrication technology, from glovebox fabrication to significantly shielded fabrication lines. It must nevertheless be noted that these shielding requirements are likely to be comparable to what would be needed in more advanced nuclear energy systems in the uranium/plutonium fuel cycle seeking recycling of MAs or other transuranics.

These technological and industrial challenges warrant the consideration of options where reprocessing of thorium-based fuel can be delayed. Given the very long half-life of ^{233}U ($T_{1/2} \sim 160\,000$ years), this delay would represent a smaller penalty than in the uranium/plutonium cycle where part of the fissile material bred (^{241}Pu , $T_{1/2} \sim 14$ year) is “lost” during the cooling of spent fuel. This is not the case for ^{233}U , though accompanied by significant levels of ^{232}U . As an order of magnitude, concentrations of around 2 ppb of ^{232}U occur in conventional UOX spent fuel of LWRs, with this figure increasing to around

6 ppb of ^{232}U present in the enriched reprocessed uranium coming from such spent fuel. In comparison, in a thorium/ ^{233}U fuel, concentrations of about 65 ppm of ^{232}U can be expected, a figure that represents an increase of ~30 000 times relative to current UOX fuels.

In October 2014, the United States Department of Energy (DOE) published a substantial report entitled *Nuclear Fuel Cycle Evaluation and Screening* (DOE, 2014), which is the result of a large study aimed at assessing a wide range of generic nuclear reactor fuel cycles, from mining to disposal, both for once-through and recycle fuel cycles options in thermal, intermediate and fast neutron spectra.⁶ According to the methodology and criteria used, which includes reduction of waste generation and improvement in the utilisation of resources, the study categorised as *most promising fuel cycles* a number of different families of uranium/plutonium fuel cycles which implement the *continuous recycle of U/Pu or U/TRU with new natural – U fuel in fast and/or thermal reactors*. It was noted that fuel cycles with U/Pu recycling pose “relatively lower development and deployment challenges” than those implementing U/TRU recycle.

Under that same study, fuel cycles using thorium were categorised under *additional potentially promising fuel cycles*. Thorium fuel cycles studied included the fuel cycle families which implement the *continuous recycle of $^{233}\text{U}/\text{Th}$ with new Th fuel or enriched U/Th fuel in thermal, fast or both fast and thermal reactors*. The study highlights that *none of these fuel cycles (promising or potentially promising) are ready to be deployed today and R&D is required to develop the appropriate implementing technologies*. It should be noted that if thorium fuel technologies are to develop in the longer term, the required R&D around these options must occur in the near term.

Based on these technological and industrial challenges, time frames which can be considered with different strategies for thorium introduction in time-evolving nuclear reactor fleets are briefly described here.

2.4 Short term (before 2030): Thorium as an additive to the uranium/plutonium fuel cycle

The introduction of thorium in smaller quantities, i.e. as an “additive”, in uranium/plutonium fuels can be considered, although will have a very limited impact on the overall perspective of natural uranium or waste management purposes. Such additive introduction, with typically 5 to 10% of thorium in uranium/plutonium oxide fuels, could be considered as a means to further improve the neutronics in such fuel assemblies and allowing improved core power flattening, as well as allowing a reduction in the use of burnable poisons such as gadolinium. Here, the introduction of thorium is justified from the inherently favourable neutronic and material properties of thorium and the resulting ^{233}U .

While this additive use of thorium does not address global objectives as uranium savings, fissile material management or ultimate waste management improvement, it may well be part of an initial step towards thorium use in generation III reactors and can contribute to further augmenting the irradiation experience towards higher burn-ups with thorium in LWRs.

It is, however, to be acknowledged that a fundamental economic disincentive associated to many thorium fuel designs using LEU is the fact that some fuel concepts

6. The objective of this study was to provide DOE decision makers with a set of standardised metrics and criteria for comparison of different fuel cycle options that could aid in identifying those with more promising potential improvement over the current nuclear fuel cycle in the United States.

may involve operating a reactor core at lower power than the nominal power rating for the facility. This power derating, sometimes described as an “opportunity cost”, is essentially aimed at favouring neutron absorption in thorium nuclei (and the subsequent production of ^{233}U) over the fission of ^{235}U at the start of the lifetime of thorium fuels. This is found unacceptable by most utilities from a commercial point of view, as the power output difference (compared to a conventional use) will be seen as lost revenue. Overcoming this issue in the longer term, by clearly identifying compensating drivers, e.g. waste management and/or safety benefits, will be crucial for thorium fuel developers seeking a nuclear utility partner.

Today, the lack of clear drivers and economic incentives to change provide a real barrier to the significant licensing work which would need to be undertaken for the implementation of thorium fuels and no clear specific developments are being considered industrially in this direction.

2.5 Medium term (2030-2050)

While the longer-term options more explicitly aim at the deployment of the so-called “thorium fuel cycle”, i.e. replacement of uranium/plutonium into a thorium/ ^{233}U -based fuel cycle, none of these longer-term options can occur “at once” and will need to transition through a period where thorium and subsequently produced ^{233}U are gradually introduced into the fuel cycle.

While uranium scarcity is not a main driver in the short to medium term to justify a shift to thorium use, various uncertainties in nuclear energy deployment will need to be addressed within the short- to medium-term time frame.

In spite of the Fukushima Daiichi accident, which had an immediate impact on nuclear energy use or deployment in some countries, the medium- and longer-term forecasts still envisage an increasing reliance on nuclear energy (NEA, 2012) in the context of:

- growing needs to decarbonise the energy vector;
- increasing importance of energy independence;
- the availability of baseload low-cost energy supply;
- other future potential uses of nuclear energy such as process heat.

Nevertheless, any projections on nuclear energy’s market share in the future must be interpreted with care and will inherently depend on the answers nuclear energy can provide to societal concerns relating to its use; namely, safety, spent fuel, ultimate waste management and non-proliferation.

Ensuring a high level of safety is the prime objective of nuclear industry and the Fukushima Daiichi accident has strengthened its importance. New options are being considered including, for instance, the development of “accident-tolerant fuels”. Among the various options that could be considered for such fuels, thorium introduction could be one of the possibilities, based on the favourable physico-chemical characteristics offered by thorium. However, these characteristics need to be fully assessed on a case by case, as they will depend on the final thorium fuel form mixed with uranium and/or plutonium.

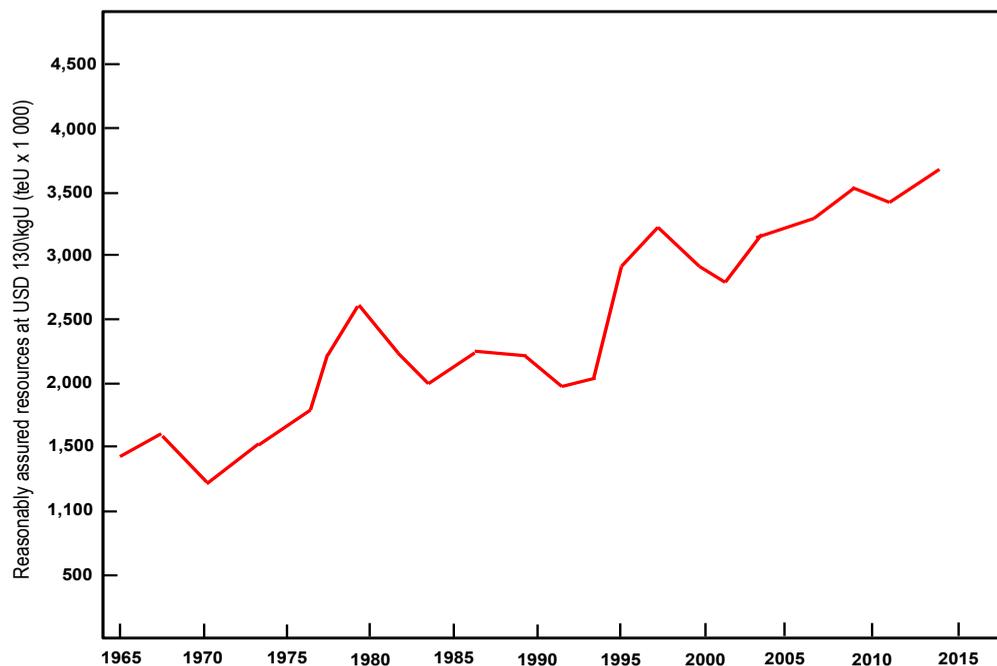
With most countries currently relying on once-through uranium/plutonium fuel cycles, the delayed deployment of geological repositories to dispose of spent fuel (as well as for vitrified waste) has introduced additional impediments to the deployment of nuclear energy. The increase in stored spent fuel may lead to technical and proliferation-risk concerns in the longer term. Some countries such as France have embarked on partially closed fuel cycles where fissile materials contained in spent UOX fuels are recycled in MOX fuels.

Irrespective of the option chosen in the past or used today (once-through or partially closed fuel cycles), virtually all countries envisioning an important role for nuclear energy in a sustainable energy future are examining a long-term transition towards generation IV nuclear energy systems, such as fast neutron spectrum reactor systems (FRs) using uranium/plutonium fuels. Such FRs will facilitate improved utilisation of uranium and a significant reduction in natural uranium use. FRs have the potential to close the fuel cycle with multiple recycle of uranium, plutonium and MAs, greatly decreasing the amount of high-level waste.

The effectiveness of reduced uranium consumption and/or reduced waste production as drivers for change is doubtful in the short to medium term. In spite of nuclear power having consumed some 3 million tonnes of uranium in the last 60 years, the trend in uranium resources has been generally upwards. This is illustrated in Figure 2.2, the data from which has been compiled from various IAEA/NEA *Uranium Production and Demand* report series.

Various studies have shown⁷ that the fuel cycle represents a relatively small part of the levelised cost of electricity from nuclear power – of the order of 10-20%. The proportion of fuel cycle cost due to uranium will vary with uranium price; but at a spot price of USD 50/lb U₃O₈, the World Nuclear Association calculated a typical June 2013 fuel cost⁸ as being 49% due to uranium, as shown in Table 2.4. Thus an increase of, say, 20% in the levelised cost of electricity would require a sustained rise in uranium prices by around a factor of 5, which is consistently higher than that reached by the uranium price spike in 2007, when uranium prices briefly rose to USD 135/lb U₃O₈.

Figure 2.2. Reasonably assured uranium resources at USD 130/kgU



Note: Compiled from IAEA/NEA *Uranium: Resources, Production and Demand* (1966-2014).

7. See for example *Projected Costs of Generating Electricity*, International Energy Agency/Nuclear Energy Agency, OECD, 2010.
8. www.world-nuclear.org/info/Economic-Aspects/Economics-of-Nuclear-Power/.

Table 2.4. Cost of 1 kg of LWR fuel as at June 2013

Uranium:	8.9 kg U ₃ O ₈ x USD 130	USD 1 160
Conversion:	7.5 kg U x USD 11	USD 83
Enrichment:	7.3 SWU x USD 120	USD 880
Fuel fabrication:	per kg	USD 240
Total (approx.)		USD 2 360

Data from the World Nuclear Association.

One of the often claimed advantages of the thorium cycle is that it produces less plutonium and other actinides and significantly reduces the radiotoxicity of resulting waste. While a pure Th/²³³U cycle will indeed produce less plutonium and MAs, the long-term radiotoxicity of thorium-based spent nuclear fuels is more accurately described as being comparable to that of uranium-based spent nuclear fuels (see Chapter 8). Furthermore, whether a reduction in radiotoxicity is translated into an actual advantage will depend in practice on whether this reduction leads to a significant change in the likelihood of making a safety case for the disposal and whether this translates into a reduction in disposal cost. Many disposal studies show safety cases driven by long-lived mobile fission products rather than by highly radiotoxic actinides;⁹ this means that the most direct driver of disposal difficulty is more likely to be the amount of power generated, rather than the system it is generated by. Similar arguments can be advanced against waste volume *per se* being the major driver of disposal economics. In any case, the fact that disposal can be delayed for many decades will militate against it being a major factor in any discounted cash financial assessment.

The development of FRs will, for the foreseeable future, be driven by governmental programmes with very limited industrialisation prospects before 2050. In addition, technical and economic challenges must be resolved before an industrially mature deployment of FRs could be considered. This has introduced uncertainties on timing and deployment rates of FRs worldwide. The increasing “need” to transition towards more sustainable nuclear energy systems driven by continuing socio-political concerns essentially focused on spent fuel piling-up will make the medium-term period 2030-2050 a decisive period where nuclear energy may or may not transition towards a worldwide sustainable energy solution. Within this context, the use of thorium as complement to enhance the flexibility of the present fuel cycle is increasingly being considered by some of the main actors in the nuclear energy industry as a possibility (but, it must be noted, without it being the prime option).

In a context where deployment of FRs is uncertain and an increased use of nuclear energy is required, the following options could become drivers for the use of thorium in the medium term:

- the desire to manage spent fuel by recycling uranium and plutonium and/or transuranics in appropriate reactors, i.e. LWRs/HWRs;
- the possibility that, in the case of high nuclear deployment scenarios, increasing pressure occurring on natural uranium exploration and mining may become an issue to consider by 2050;

9. See, for example, *Geological Disposal: Generic Post-closure Safety Assessment*, Radioactive Waste Management Directorate, Nuclear Decommissioning Authority, United Kingdom (2010), www.nda.gov.uk/publication/geological-disposal-generic-post-closure-safety-assessment-december-2010/.

- the development of flexible fuel cycle schemes adapted to local nuclear park conditions though globally seeking for synergies.

The physical and neutronic characteristics of ^{232}Th and ^{233}U could provide the means to achieve longer fuel cycle lengths, higher burn-ups and/or to reduce the burning of ^{235}U or fissile plutonium. Thorium use could be considered in various reactor systems both in a homogeneous as in a heterogeneous manner, i.e.:

- mixing thorium with uranium and/or plutonium as a homogeneous fuel;
- fuel assemblies with a combination of uranium and/or plutonium fuel rods and thorium fuel rods or;
- in a heterogeneous core with fuel assemblies specifically dedicated to thorium introduction without modifying the rest of the usual UOX/MOX assemblies.

The thorium fuel options that may be considered in LWRs vary according to the purpose envisaged, i.e. reduction in natural uranium consumption, optimising ^{233}U breeding for recycling, or its later use in more dedicated reactor types. Probably the most workable approach for thorium use in LWRs is via the heterogeneous option with heterogeneity on the level of the fuel assembly, and where the thorium and ^{233}U are neutronicly coupled to the uranium/plutonium cycle, but with the two element types treated within their respective fuel cycles. As thorium/uranium/plutonium reprocessing schemes are not yet industrially feasible, and bearing in mind the very significant challenges posed by the re-fabrication of recycled fissile materials, any recycling of the thorium/ ^{233}U may need to be initially limited to a small fraction of fuel assemblies.

Schemes with thorium facilitating the recycling of reprocessed uranium in PHWRs, or improving the recyclability of plutonium in LWRs are under study and could be realisable in the medium term. These “heterogeneous” strategies could facilitate rapidly growing third generation nuclear reactor fleets to improve their overall fissile material balance and especially the management of spent fuel through (multi-)recycling of uranium and, especially, plutonium. These types of strategies could become attractive options to consider if fast reactor deployment in synergy with LWRs (which is, in general, currently the first-choice of future fuel cycle options supported by industry), does not occur or is limited.

Provided the deployment of fast breeder reactors using the uranium/plutonium cycle remains the preferred long-term strategy for industry to replace generation II and generation III reactors, any substantial move to dedicate plutonium inventories to breed ^{233}U in thorium fuels would continue to be seen as risky in terms of plutonium economy strategies, since it could compromise the future deployment of fast breeder reactors that require large initial plutonium inventories to start.

2.6 Long term (post-2050): Dedicated breeder systems using Th/ ^{233}U closed fuel cycles

Longer-term options for thorium implementation may investigate increasing the use of thorium by using combinations of reactor types or dedicated breeder reactor systems to establish a full Th/ ^{233}U fuel cycle.

It is important to understand that all of these longer-term options would have to receive fissile material, either plutonium of adequate fissile quality from the existing uranium/plutonium fuel cycle, or ^{233}U bred during a transitioning period, in order to be realisable. The three-stage development programme currently supported by India is today a prime example of the need to introduce thorium fuels progressively in order to match fissile material availability.

An autonomous thorium fuel cycle is only realisable if the ^{233}U is recycled. In particular, advanced fuel cycle options will depend on successful development of

processes associated with spent thorium fuel reprocessing and re-fabrication of irradiated fuels with ^{233}U , including appropriate consideration of radiation protection and non-proliferation issues. These advanced options would also depend on the successful implementation of thorium fuels reprocessing methods at industrial scales (thorium-uranium extraction [THOREX] or alternatives if these are proven) and would also require significant R&D in thorium-based fuel technology and qualification of the fuel.

2.6.1 Generation IV concepts with thorium

Among the six future reactor concepts that the Generation IV International Forum (GIF) considers, MSRs stand out as specifically considering the use of the thorium fuel cycle. The Canadian design supercritical water reactor fuel considers the use of $(\text{Th,Pu})\text{O}_2$ fuel.

MSR concepts implement very innovative fuel management approaches with the use of fuel (thorium or uranium based) in liquid form, which in principle allows the continuous “online” reprocessing of the fuel in order to extract fission products and ^{233}Pa (precursor of ^{233}U and a neutron absorber). Liquid fuel concepts allow, in principle, greater power densities and smaller initial fissile inventories than solid fuel concepts. The online liquid fuel management of MSRs allows for theoretical breeding ratios equal or greater than one.

In its latest *Technological Roadmap Update* (GIF, 2014), GIF has extended the viability study phase of MSR concepts until 2025, reflecting the fact that MSRs concepts are still in need of substantial development before they are deemed technologically feasible. Particularly challenging are the essential steps of on-site or online treatment of liquid fuel, which requires the implementation of pyro-chemical processes, for which the actual feasibility or performances are not yet known. The safety analysis methods in their current form cannot be applied to liquid-fuelled MSRs, due to the innovative form of the fuel (i.e. absence of cladding, molten fuel conditions under normal operation, continuous circulation of fuel in and out of the active core, etc.). The development of methodologies for design and safety evaluations that also address safeguards and non-proliferation aspects of liquid-fuelled MSRs is needed.

In 2011, China announced the start of an ambitious R&D programme on MSRs led by the Chinese Academy of Sciences aiming, in particular, at the construction of a 2 MW thermal spectrum, graphite-moderated pilot thorium molten salt reactor TMSR-LF (“liquid fuel”). In 2014, the TMSR-LF prototype was in a pre-conceptual design phase with limited use of thorium in the foreseen candidate compositions for the molten salt fuel. Large R&D and technology developments are required to exploit the potential benefits of thorium-fuelled molten salt reactors (fast or thermal).

2.6.2 Hybrid reactor concepts

Innovative “hybrid” reactor concepts that combine the characteristics of different future reactor concepts (namely, hybrids of accelerator-driven with molten salt blanket systems, hybrids of fission and fusion reactors, etc.), have been envisaged as potentially making use of thorium. Although these concepts may have interesting theoretical properties, they inevitably reflect the disadvantages, uncertainties and unknowns of the various technologies that enable them. These unknowns are often independent of the fact that these concepts may or may not use thorium and, as such, would first need to be further studied, developed and demonstrated. Consequently, these composite or “hybrid” concepts are very unlikely to provide any credible application for commercial electricity production in this century.

References

- Bromley, B.P. (2014), "High utilization lattices for thorium-based fuels in heavy water reactors", *ANS Nuclear Technology Journal*, April 2014.
- Bromley, B.P. and B. Hyland (2014), "Heterogeneous cores for implementation of thorium-based fuels in heavy water reactors", *ANS Nuclear Technology Journal*.
- DOE (2014), "Nuclear Fuel Cycle Evaluation and Screening – Final Report", INL/EXT-14-31465, US Department of Energy.
- Ganda, F., et al. (2012), "Self-sustaining thorium boiling water reactors", *Sustainability*, Vol. 4, pp. 2472-2497.
- GIF (2014), *Technological Roadmap Update*, Generation IV International Forum, Paris.
- IAEA (2013), *Nuclear Power Reactors in the World*, International Atomic Energy Agency, Vienna.
- NEA (2012), *The Role of Nuclear Energy in a Low-carbon Energy Future*, OECD, Paris.

3. Front end of the thorium fuel cycle

3.1 Availability and recovery

Regardless of the mineral reserves considered, the amounts shown in the literature heavily depend on the degree of confidence in the actual occurrence of the estimated quantity of the desired materials and of the technical and economic feasibility of their recovery. A distinction is often made between “resources” and “reserves”, although there are still no real international standards to define these two categories. According to the model presented by the NEA and the International Atomic Energy Agency (IAEA) joint report on *Uranium: Resources, Production and Demand* (IAEA/NEA, 2014), a mineral “resource” is “a concentration or the presence of material of economic interest in the earth's crust in such quantity and quality, that the prospects for eventual economic extraction are reasonable”. In some cases, the characteristics of a mineral resource are based on evidence, specific sampling and geological knowledge. A mineral “reserve” is the economically mineable part of a deposit under current economic conditions. The classification is based on evaluations and studies, taking into account a set of constraints affecting mining in relation to economic, market, metal, legal, environmental, social and governmental considerations.

Thorium is a relatively common element in the earth’s crust. Based on its longer half-life ($1.41 \cdot 10^{10}$ years) compared to that of uranium ($4.5 \cdot 10^9$ years), it is estimated to be roughly three times more abundant than uranium. This figure, however, does not imply that the exploitable reserves of thorium are two or three times higher than those of uranium, as frequently cited in the literature. Because of its very limited use so far, there has never been a comprehensive survey of the thorium resources in the world, and current estimates of exploitable world resources of thorium are therefore not very accurate.

A number of significant thorium reserves have been identified in recent decades. These reserves are fairly well distributed around the world, though India, Turkey, Brazil, Australia, the United States, Egypt, Norway, Venezuela, and especially the Asian part of former Commonwealth of Independent States (CIS), possess particularly significant shares of the estimated global total. Table 3.1 provides the joint NEA-IAEA 2014 estimates of identified global thorium resources. Some studies, such as the one published by the US Geological Survey (2014), provide much lower estimates of thorium resources (1-2 million tonnes). The IAEA has made an ongoing effort to construct an online inventory database of different types of thorium deposits worldwide (THDEPO, n.d.).

In spite of thorium’s relative abundance, its current market demand is small and little is currently being mined. Only about 100 metric tonnes are recovered annually, entirely as a by-product of mining operations for titanium (AMR, 2012).

A significant amount of the earth’s total thorium content is found in seawater; however, at a concentration below 0.01 parts per billion (ppb), compared to about 3 ppb for uranium, concentrations that are too low for commercial recovery (Herring, 2012).

There are more than 100 thorium-bearing minerals in the crust of the earth, of which about 60 have a thorium concentration larger than 0.1%. Among the most notable of these minerals are thorite, thorianite, bastnasite and monazite.

Historically, thorite was considered the most likely thorium resource if a major thorium industry were to emerge. Although experiments have found chemical separation of thorium from thorite to be feasible, thorite has not been pursued recently for commercial purposes.

Thorianite, an oxide mineral, also has significant thorium content, as it is almost entirely composed of thorium oxide. However, its viability as a source of thorium has not been explored, possibly due to its relatively infrequent occurrence. While thorite and thorianite have not seriously been considered in thorium recovery analyses since the early 1980s, these minerals are often found in conjunction with uranium at major mining sites. Thus, by-product recovery of thorium at uranium mines may be a viable option.

Unlike thorite and thorianite, bastnasite is the object of major commercial mining operations, due to its rare earth element (REE) content (>50%). Its thorium content is not particularly high, usually not much greater than 0.2% (IAEA, 2012b). However, it is mined in massive amounts, particularly in China; at the largest bastnasite mining complex, Bayan Obo, 55 000 tonnes of rare earth oxides (REO) are recovered annually from bastnasite (Long et al., 2012). The largest bastnasite mine in the United States, Mountain Pass, has previously achieved an annual production of about 1 500 tonnes of REE in peak years (van Gosen, 2012). Though closed for some time, Mountain Pass was reopened in 2012. There has not been any large-scale thorium recovery from bastnasite, although chemical techniques for thorium separation have been developed in China for waste reprocessing.

In recent times, the phosphate mineral monazite has been widely regarded as the most likely candidate for thorium by-product recovery, as it contains a higher thorium concentration than bastnasite and still contains a high REE content (>50%) (Zhu and O'neils, 1999). Monazite alluvial deposits comprise a significant amount of total thorium reserves and have an average thorium content of 3-10%. Global production of REO from monazite is considerably lower than that from bastnasite. Less than 10 000 tonnes is recovered annually, most of it as a by-product of ilmenite (titanium ore) production from the beach sands of India (Hedrick, 2004). However, monazite's much higher thorium content (compared to bastnasite) makes thorium recovery viable even though less total ore is produced.

Furthermore, like the active thorium-recovering sites of India, many of the world's monazite deposits are collocated with titanium ore. Since about 7 million metric tonnes of titanium is recovered annually, the potential exists for 2-3 orders of magnitude greater Th production from titanium mining than is currently observed. Thorium by-product recovery from rare earths and currently active titanium mines could potentially be greater than the estimated volumes of uranium annually consumed by the world's entire nuclear reactor fleet.¹

1. Around 7 000 metric tonnes of uranium are used yearly by the currently installed world nuclear capacity (376 GWe). The demand for REEs is estimated to be 200 000 metric tonnes in 2014. For monazite alone, the US Geological Survey Minerals Yearbook for thorium (2004) mentions 5 700 metric tonnes of annual monazite production (with around 0.1 Th/REE content), mostly as a by-product of ilmenite from the beach sands of India. Since about 7 million metric tonnes of titanium is recovered annually, the potential could exist for 2 or 3 orders of magnitude greater thorium production than currently observed.

Table 3.1. Global estimates of identified thorium resources 2014

Region	Country	Total thorium resources, tonnes (<i>in situ</i>)
Europe	Turkey*	374 000
	Norway	87 000
	Greenland (Denmark)	86 000-93 000
	Finland*	60 000
	Russia	55 000
	Sweden	50 000
	France	1 000
	Total	713 000-720 000
Americas	United States**	595 000
	Brazil	632 000
	Venezuela*	300 000
	Canada	172 000
	Peru	20 000
	Uruguay*	3 000
	Argentina	1 300
	Total	1 723 300
Africa	Egypt*	380 000
	South Africa	148 000
	Morocco*	30 000
	Nigeria*	29 000
	Madagascar*	22 000
	Angola*	10 000
	Mozambique	10 000
	Malawi*	9 000
	Kenya*	8 000
	Democratic Republic of the Congo*	2 500
	Others*	1 000
	Total	649 500
Asia	CIS* (excluding Russia)	1 500 000
	- includes Kazakhstan, estimated	(>50 000)
	- includes Russia, Asian part, estimated	(>100 000)
	- Uzbekistan, estimated	(5 000-10 000)
	- others	Unknown
	India	846 500
	China, estimated	>100 000 (including 9 000* Chinese Taipei)
	Iran, the Islamic Republic of*	30 000
	Malaysia	18 000
	Thailand*, estimated	10 000
	Viet Nam*, estimated	5 000-10 000
	*Korea	6 000
	Sri Lanka*, estimated	4 000
	Total	>2 647 500-2 684 500
Australia	595 000	
World total	6 355 300-6 372 300	

CIS = Commonwealth of Independent States.

Identified Th resources may not have the same meaning in terms of classification as identified U resources. Higher range of the estimates wherever given is taken for a region.

* Data not updated. ** Estimate of identified resources (RAR + inferred) of thorium in the United States is based on a recent comprehensive review of published data by the US Geological Survey (Staatz et al., 1979, 1980). Earlier estimates in the *Uranium 2014: Resources, Production and Demand* indicated thorium resources as much as 770 000 tonnes in the United States, which may have included estimates of undiscovered resources (prognosticated and speculative). This higher value cannot be replicated or substantiated, so it is not repeated here.

Source: IAEA/NEA (2014).

The currently installed world nuclear capacity is 376 GWe.² A conventional 1 GWe PWR contains the equivalent of 80 tonnes (metric tonnes) of heavy metal (MTHM) of uranium dioxide fuel in its core, with 25% of this quantity needed each year to reload the core with fresh fuel; i.e. 20 MTHM of fresh fuel, per year, for each PWR. If this fuel were replaced in all reactors by Th-HEUO₂ or Th-PuO₂, it would require over 7 000 tonnes of thorium per year. If this output was supplied by a closed Th/²³³U cycle, the requirement would fall to 400 tonnes of thorium.

Since the current thorium market is very limited, there are few incentives to open new mines with thorium as the primary product. Most likely, near-future thorium recovery will utilise pre-existing mining operations, which currently surface thorium but route it to waste because of the small demand. Titanium and uranium mines appear to represent the most promising options for by-product recovery of thorium. Since most of the thorium processing schemes developed in the last 30 years have been designed for monazite, flowsheets based on these are useful for evaluating thorium recovery flowsheets in general. However, other ores such as bastnasite and thorite may ultimately prove to be useful as well.

Therefore, it should be noted that the overall abundance of thorium is not an issue in whatever nuclear energy system scenario considered for the foreseeable future. Furthermore, the sustained use of thorium inherently demands the recycling of fuel and its combined use with fissile materials coming from the uranium/plutonium fuel cycle in the near to medium term which would result in an even lower thorium demand.

It is concluded that by-product production of thorium from other industrial mining activities can provide more than ample quantities of thorium for the nuclear industry for this century and beyond.

Annex A gives a detailed overview of the different steps needed to recover thorium, beginning at its major mineral forms and proceeding to nuclear-grade purity suitable for fuel fabrication.

3.2 Thoria fuel fabrication

This section reviews experience with the fabrication of thoria-based nuclear fuels based on past or ongoing work performed in Canada (Hamilton, Livingstone and Popov, 2007), Germany, France and Norway (Insulander-Bjork et al., 2015).

An essential distinction must be noted between fresh fuel fabrication (pre-irradiation) and recycle fuel fabrication (post-irradiation, re-fabrication), as these two steps are performed under very different conditions and face entirely different challenges.

3.2.1 Fresh thoria fuel fabrication

Thorium-based fuels (as opposed to thorium blanket elements) necessarily contain more than one component, namely thorium and a fissile element, such as plutonium or uranium. For implementation of homogeneous thoria-based fuel cycles, issues associated with fabrication of thoria (ThO₂) fuel must be addressed. Fabrication of high quality, representative thoria-based fuel pellets for irradiation testing requires preparatory studies of how such ceramics are formed, including the processes of powder comminution, component oxide blending and sintering into a high density ceramic.

The addition of a fissile component to the fuel poses a challenge, as the components should be adequately mixed on a microscopic level and the fissile component evenly distributed so as to avoid areas having undesirably high-fission densities. Ensuring that

2. www.iaea.org/pris/.

the various constituents are mixed sufficiently is of the utmost importance to ensure even power production in the fuel elements. The following four methods of mixing are generally used:

- Mechanical mixing.
- Sol-gel process: A sol is a liquid containing solid particles that are evenly distributed and stably suspended and may be made to gel by the removal of the inter particle repulsive forces. This process has been used at the Atomic Energy of Canada Limited (AECL) to produce thoria UO_2 micro-spheres (Hamilton, Livingstone and Popov, 2007). The idea behind the sol-gel synthesis is to “dissolve” the compound in a liquid in order to recover it as a solid in a controlled manner; it results in small particles, which are easily sinterable. This method has the advantages of reducing dust, reducing the sintering temperature and generally increasing the reactivity of the powder. The fabrication of the fuel used in HTR is based on this technique. The uranyl, Pu or Th nitrate solutions are mixed in the required proportion and the viscosity is adjusted by the addition of trace amounts of organic thickeners to produce a feed solution. This feed solution is dispersed into droplets, which are then collected in a hydroxide bath, where gelation occurs within the original droplets, forming microspheres. Finally, the microspheres are washed, dried and calcined.
- Solution blending/co-precipitation: This method has been used to prepare $\text{ThO}_2\text{-UO}_2$ powders by co-precipitating oxalates from a nitrate solution. With the correct conditions, mixing at the atomic scale is achieved (IAEA, 2012a).
- Solution impregnation: This is a technique, in which green pellets³ of pure thoria are placed in solutions of uranyl nitrate. The uranyl nitrate is adsorbed by the pellet, and the nitrate is removed during the sintering process.

A key challenge in thorium oxide-based fuel manufacture lies with the high-melting point of ThO_2 (365 degrees higher than that of UO_2), which means that sintering kinetics are quite slow unless the inter-particle contact is already high in the green pellet form. This means that the precursor powder must have a high-surface area. Fortunately, additive oxides – including PuO_2 – serve as sintering aids. If successful powder pre-processing techniques are established, sintering temperatures need not be higher than for standard UOX fuels and densities greater than or equal to 96% of theoretical density (TD) are possible.

3.2.2 Fabrication of test thoria fuel at IFE (Norway)

Fuel ceramic fabrication work has been carried out as part of the thoria fuel irradiation campaign conducted at Halden and commissioned by the private Norwegian technology company “Thor Energy” (see also Section 4.1), in partnership with IFE in Norway (Insulander-Bjork et al., 2015). Pellet specimens have been made at IFE from blended ThO_2 and UO_2 powders and from blended ThO_2 and CeO_2 , simulating Th-MOX fabrication⁴. Earlier trials at LANL (Chen et al., 2014) focused on finding optimal procedures for thoria powder conditioning (using a dispersant⁵ and/or heat treatments). Pressing and sintering parameters were also examined and $(\text{Th,Ce})\text{O}_2$ pellets with a density greater than 94% of the TD were produced. The resulting fuel manufacturing experience is applied to ongoing thoria fuel development.

-
3. Pellets after pressing from powder, but before sintering.
 4. Cerium has similar chemical properties similar to those of plutonium making it easier to work with cerium in the early process development stages.
 5. Dispersants are large surfactant molecules which can help to control electrostatic forces which cause particle agglomeration and dust during the milling of such electrically insulating powders.

The ThO₂ pressing and sintering trials at IFE yielded a density of 96% of TD for a mixed (Th,Ce)O₂ ceramic. Ongoing studies (2015) are addressing the need to achieve a good ceramic microstructure for (Th,Pu)O₂ fuel pellets. Fuel manufacturing is adhering strictly to commercial powder metallurgical methods used for UOX and MOX fuel production. This is performed to develop and demonstrate commercially compatible manufacturing techniques for Th-MOX fuel:

- Powder production: Pure thorium oxide powder is usually prepared by calcining thorium oxalate which has been obtained by the addition of oxalic acid to a thorium nitrate solution. The morphology of such oxide particles is quite “blocky”, and powders of this material must be milled in order to increase their bulk surface area (and thus the interfacial contact area between particles). This facilitates their compaction to high density. Co-milling ThO₂ with other oxides may require surfactant “dispersants” to help control electrostatic forces which cause particle agglomeration and dust – this is especially important to avoid when making (Th,Pu)O₂. If dispersants are used, a calcination step is necessary to remove the dispersant, followed by a light milling. If thorium powder is milled separately, intermediate sieving followed by recycling of larger platelets could reduce overall milling times.
- Pellet pressing – green pellet production: Green pellet production is conducted with a double-sided press as typically used for standard UOX fuel production at IFE. A standard die lubricant is used in the press. Organic or other binders are not necessary. Studies are conducted to examine the sensitivity of green pellet density to the pressing regime used to form the fuel pellet.
- Pellet sintering: Thorium oxide pellets should be sintered in an appropriate atmosphere, depending on whether uranium or plutonium is the other fuel constituent. An oxidising atmosphere is ideal for Th-MOX, but if this is not possible (due to the need to protect furnace heating elements from damage), a second heat treatment at 1 000-1 200°C in air will ensure correct oxygen stoichiometry of (Th,Pu)O₂ pellets. Sintering aids such as Nb₂O₅ and MgO have not been needed in these trials. Fuel pellet production is deemed successful if pellet densities are equal or greater than 95% of TD.

References

- AMR (2012), Thorium 2012 (PowerPoint presentation), AMR Mineral Metal Inc. www.amrmineralmetal.com/download/products/THORIUM_AMR_2012.pdf.
- Chen, C.F., et al. (2014), “Processing of ThO₂/CeO₂ ceramic fuel”, *Journal of the American Ceramic Society*, Vol. 97, pp. 3062-3070.
- Hamilton, H., S. Livingstone and B. Popov (2007), “Thorium fuel fabrication and testing at AECL”, International Workshop on Thoria Utilization for Sustainable Development of Nuclear Energy, TU2007, AECL report CW-124950-CONF-001, Beijing.
- Hedrick, J.B. (2004), “Thorium”, United States Geological Survey (USGS).
- Herring, J.S. (2012), “Uranium and Thorium Resources”, *Chapter 18 of Nuclear Energy*, ed. N. Tsoulfanidis, Springer, New York, pp. 462-490.
- IAEA (2012a), *Role of thorium to supplement fuel cycles of future nuclear energy systems*, IAEA Nuclear Energy Series No. NF-T-2.4, International Atomic Energy Agency, Vienna.
- IAEA (2012b), *World Thorium Deposits and Resources*, International Atomic Energy Agency, Vienna.
- IAEA/NEA (2014), *Uranium 2014: Resources, Production and Demand*, OECD, Paris, www.oecd-nea.org/ndd/pubs/2014/7209-uranium-2014.pdf.

- Insulander-Bjork, K., et al. (2015), "Commercial thorium fuel manufacture and irradiation: Testing (Th,Pu)O₂ and (Th,U)O₂ in the Seven-Thirty program", *Annals of Nuclear Energy*, Vol. 75, pp. 79-86.
- Long, K.R., et al. (2012), "The principal rare earth elements deposits of the United States: A summary of domestic deposits and a global perspective", *Non-Renewable Resource Issues*, pp 131-155.
- Staatz, M.H., et al. (1979), *Principal Thorium Resources in the United States*, US Geological Survey Circular 805, pp. 42.
- Staatz, M.H., et al. (1980), *Thorium Resources of Selected Regions in the United States*, US Geological Survey Circular 824, pp. 32.
- THDEPO (n.d.) Integrated Nuclear Fuel Cycle Information System (web page), <https://infcis.iaea.org/THDEPO/> (password protected).
- US Geological Survey (2014), *Mineral Commodities Summaries*, US Geological Survey, Reston.
- Van Gosen, B. (2012), USGS, Personal Communication.
- Zhu, X.K. and R.K. O'nions (1999), "Monazite chemical composition: Some implications for monazite geochronology", *Contributions to Mineralogy and Petrology*, Vol. 137, Issue 4.

4. Thoria fuel testing and qualification

The licensing and commercialisation of thorium fuels is dependent on reliable physical fuel testing and predictive thermo-mechanical behaviour codes being established for the specific type of mixed ceramic (Th, HM)O₂¹ that is slated for commercial reactor use. Such codes need to be benchmarked against measured irradiation performance data sets that include information on thermal conductivity, thermal expansion, radiation-induced swelling rate and fission gas retention. In order for thoria-based fuel to be licensed for use in current reactors, these material properties must be well known for fresh as well as irradiated fuel, and it must be possible to predict fuel behaviour in both normal operation and transient accident scenarios.

Fuel performance data is generally collected from irradiation experiments on instrumented rods containing test-fuel pellets. Fuel temperature and dimensional changes are measured continuously while the thorium fuel pellets are operating “at-power” in the simulated commercial reactor conditions and also during periods when the fuel is undergoing power and temperature changes. Testing aims at monitoring fuel behaviour during all relevant operational scenarios. Although not exhaustive, the following subsections aim to provide information on past or ongoing fuel irradiation tests on thorium oxide-based fuels, focusing on more recent programmes.

4.1 Thoria fuel testing programmes

4.1.1 The Halden Reactor (Th,Pu)O₂ and (Th,U)O₂ test programme

A thorium fuel irradiation experiment is underway in Halden, Norway, which is also host to the OECD/NEA Halden Reactor Project. This is a commercially driven programme where tests are performed on thorium fuel specimens that represent fuel compositions which could be used by a nuclear utility. The irradiation itself is carried out by IFE, which operates the Halden (heavy water) material test research reactor. Instrumented testing rigs have been constructed to house the test fuel. In this experiment, the starting linear heat generation rate is of around 35 kW/m. The irradiation conditions experienced by test fuel pellets closely simulate those of an LWR. The suite of instrumentation used comprises:

- thermocouples that are inserted into the centre of the fuel column to measure fuel temperature;
- extensometers on the cladding and on the fuel column to measure dimensional changes;
- internal rod-pressure transducers to measure FGR.

The extensometers and pressure transducers are based on high precision linear voltage differential transformers. Spatial (dimension and pressure) resolution is very high.

As discussed above, important parameters are known for fresh material. For (Th,U)O₂, some burn-up dependent data are available, although not for material with low Th

1. Where “HM” (heavy metal) is uranium or plutonium.

content. The irradiation campaign aims at expanding the experience base to irradiated $(\text{Th,Pu})\text{O}_2$ and $(\text{Th,U})\text{O}_2$ fuel with low Th content. The resulting comprehensive data set will support the broader goal of undertaking lead test rod (LTR) irradiations for fuel in a commercial reactor and provide a platform for commercial fuel performance code development.

Six rods, each with a length of 30 cm, are mounted parallel to each other in an instrumented irradiation rig. The rig layout is shown schematically in Figure 4.1. The instrumentation facilitates online measurement of fuel centre temperature (TF), cladding elongation, fuel stack elongation and fuel rod pressure (PF). In addition, monitoring inlet and outlet coolant temperature, coolant flow and neutron flux allow power monitoring.

Of the six rods currently (2015) being irradiated, two contain $(\text{Th,Pu})\text{O}_2$ pellets that were fabricated within an earlier European Union project that studied oxide fuels microstructure and composition variations (OMICO). Two rods contain $(\text{Th,U})\text{O}_2$ with 7 wt% Th and two rods contain reference UO_2 fuel that matches the reactivity of the test fuels.

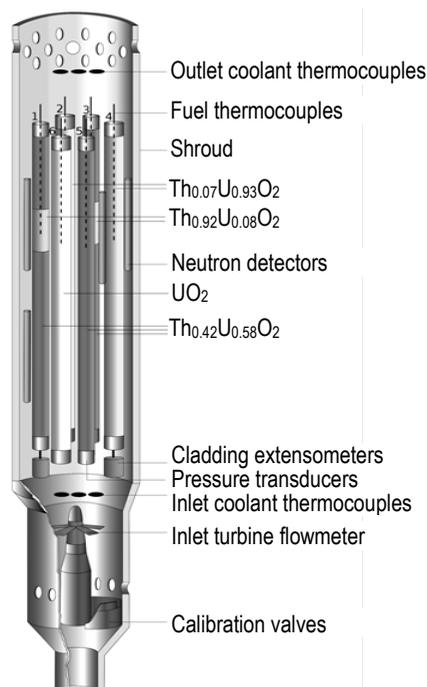
The fuel “doped” with 7 wt% thorium (“U-7Th”) was manufactured at IFE with the view that this material could serve in commercial power reactors and lower the need for burnable absorbers. Due to the relatively small addition of thorium, the co-milling of U and Th oxide powders was not difficult and the final sintered fuel density was 96% TD, with a fairly homogeneous distribution of thorium oxide within the pellet.

As of 2015, the measured fuel temperatures are in accordance with known values for the thermal conductivity of the respective materials. The centreline temperature of the rods containing U-7Th fuel pellets is about 30-40 K lower than that of the pure UO_2 reference rod, indicating that the thorium dopant is improving the thermal conductivity of the fuel pellet material, as expected. No FGR has been observed so far.

The test rig shown in Figure 4.1 will be complemented with a new instrumented rig devoted to producing new irradiation data for $(\text{Th,Pu})\text{O}_2$ fuel fabricated at IFE. Fabrication trials using $(\text{Th,Ce})\text{O}_2$ have been conducted in preparation for $(\text{Th,Pu})\text{O}_2$ fuel pellet production.

The thorium fuel manufacture and irradiation campaign will continue at least until late 2017 and include several activities supporting the irradiation of LTRs – the next required step in a thorium LWR fuel commercialisation programme. Among the suite of PIE analyses foreseen are internal rod gas analysis (providing data on the release of specific fission gases from the fuel ceramic), ceramography (showing how the fuel pellet structure has changed over the burn-up period), advanced microscopy (for examining elemental distributions within the irradiated fuel ceramic and for visualising various radiation induced microstructures).

Figure 4.1. A sketch of the instrumented fuel testing rig being used in the thorium fuel testing campaign at the NEA Halden Material Test Reactor in Norway



Source: Insulander-Bjork et al. (2015).

4.1.2 The Obrigheim (Th,Pu)O₂ PWR test programme

The Obrigheim thorium fuel test irradiation programme started in 2000 under the Fifth Framework Programme of the European Union and continued under the Sixth Framework Programme. It was carried out in Germany at the 350 MW_e PWR Obrigheim power station (KWO).

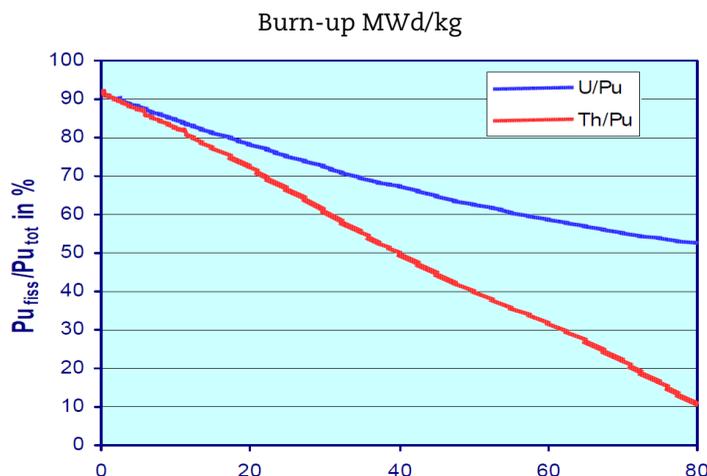
The objective of the programme was the irradiation of (Th,Pu)O₂ fuel up to a significant burn-up under as close as possible standard PWR conditions to gain experience with the thermo-mechanical and general performance behaviour of the fuel. The test irradiation ended in May 2005 and final post-irradiation measurements were performed until 2009.

Another goal of the tests was to demonstrate the capability of degrading, within reasonable irradiation times, high-quality plutonium to a quality much lower than the plutonium from discharged LWR uranium assemblies. This was also considered an option for effectively denaturing plutonium from nuclear warheads (Porsch and Sommer, 2002). The degradation rate of high-quality plutonium in U/Pu and Th/Pu fuel can be seen in Figure 4.2.

The (Th,Pu)O₂ fuel used in the Obrigheim tests was manufactured at the Institute for Transuranium Elements (ITU), Karlsruhe and loaded into the test-segment. The specifications for manufacturing process and pellet design were provided by Framatome ANP (now Areva NP), with plutonium provided by Cogema (now Areva NC).

Based on the experience gained in the framework of the German/Brazilian R&D programme, the powder was prepared using the sol-gel route followed by the standard pelletising process, providing the required homogeneity of plutonium particles and the dimensional stability requested by commercial fuel rod design. The composition of the fuel at the time of pellet manufacturing is shown in Table 4.1.

Figure 4.2. Plutonium quality degradation vs. burn-up in Obrigheim (KWO) irradiation tests



Source: Porsch and Sommer (2002).

Table 4.1. Fuel composition of the (Th,Pu)O₂ fuel irradiated in Obrigheim (KWO)

²³² Th	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴¹ Am
96.705	0.001	2.995	0.271	0.005	0.000	0.022

Source: Porsch and Sommer (2002).

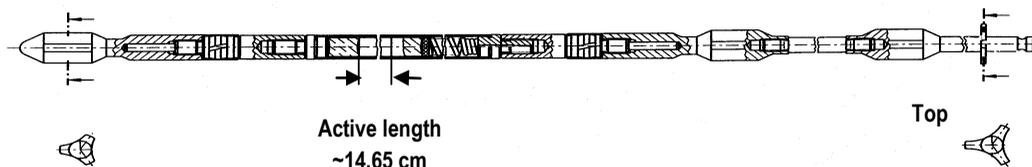
The structural components of the test rod used in the Obrigheim irradiation programme were manufactured by Framatome ANP and supplied to the ITU in Karlsruhe. Fuel manufacturing and completion of the fuel rod including welding of the second-end plug were the responsibility of ITU.

The specification of the fuel and the qualification of the manufacturing process were derived from the experience gained in the Brazilian/German programme and provided by Framatome ANP. The burn-up reached 37 GWd/tHM. Due to the fact that the design criteria for (Th,Pu)O₂ test fuel segmented rod were not complete, the existing data from (Th,U)O₂ (from the German/Brazilian programme, see below) were taken, as the majority of the material is thorium and the thermal-mechanical properties are governed by this matrix.

The test segment is depicted in Figure 4.3. It contained 17 pellets of (Th,Pu)O₂ fuel with a fissile plutonium content of 3.0 w/o. The plutonium was of high quality (see Table 4.1) and appropriate for achieving the second objective of the programme, which was to demonstrate the denaturing and incineration rate of high-fissile quality plutonium.

The length of the pellet column of the test rod was about 14.4 cm. Spacers were designed to centre the rod in its insertion position and were responsible for defined thermohydraulic conditions at the probe level. The pellet diameter and the outer fuel rod diameter of 9.5 mm are consistent to the majority of commercial LWRs. The cladding material was zircaloy-4, commonly used cladding for the Obrigheim Power Station.

Figure 4.3. Schematic of the KWO (Obrigheim) test rod



Source: Porsch and Sommer (2002).

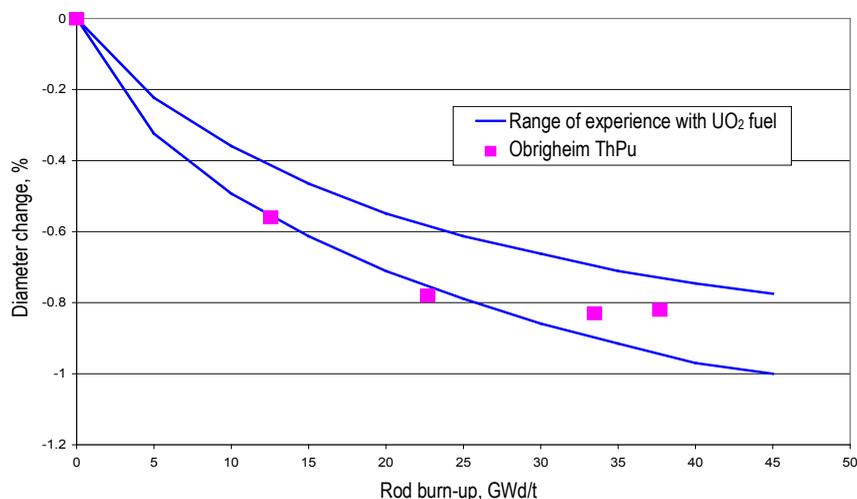
The sample test rod was inserted in the guide tube of a standard U/Pu MOX assembly with an average burn-up of 15.1 MWd/kg after one in-core cycle. This ensured the irradiation of the sample in a “real” PWR plutonium neutron spectrum and the representativeness of the experiment. Achieving PWR conditions was important for code validation and qualification of the fuel for use in commercial LWRs. Due to the limited coolant flow in the guide tube, the sample power had to be limited to 220 W/cm, somewhat higher than the fuel rod average power of typical commercial PWR plants. The accumulated burn-up at discharge of the fuel rod was calculated to be 37.7 MWd/kg. In all cycles, the power of the carrier U/Pu MOX assemblies was measured with an aeroball measurement system. Table 4.2 shows the results derived from this method of measuring the power history.

Table 4.2. (Th,Pu)O₂ sample irradiation history

Cycle		32	33	34	35
Linear heat generation rate q'_{local}	W/cm	199	148	160	142
Cycle length	EFPD	342	372	365	162
Burn-up difference	MWd/kg	12.5	10.1	10.8	4.3

Diameter measurements were performed on the sample rod during each refuelling outage and after discharge of the sample. The characteristics determined for the fuel were very similar to common UO₂ fuel. Figure 4.4 shows that the diameter change of the (Th,Pu)O₂ rod is very similar to that of standard UO₂ rods. This demonstrates that the dimensional behaviour of the (Th,Pu)O₂ fuel is similar to UO₂ up to ~38 MWd/kg.

Figure 4.4. Change of outer diameter of the (Th,Pu)O₂ test fuel rod used in the Obrigheim irradiation tests



Source: Somers, Papaiannou and Sommer (2009).

4.1.3 The German-Brazilian co-operation on thorium utilisation in PWRs

A co-operative R&D programme on “thorium utilisation in PWRs” was carried out in the 1980s by NUCLEBRAS/CDTN (Brazil) and KFA-Jülich (Germany), with the participation of Siemens/Kraftwerk Union AG and NUKEM (Germany).

The original objective of the project was to design, manufacture and irradiate PWR thorium assemblies and to close the thorium fuel cycle by reprocessing the thorium fuel.

By choosing a standard PWR as a reference plant to define the requirements to be fulfilled, a programme was initiated with the following general objectives:

- to analyse and to prove Th-utilisation in PWRs;
- to use standard fuel assembly designs;
- to develop manufacturing processes for Th/U fuel, and later on, Th/Pu fuel suitable for an operation in standard PWRs;
- to study the closing of the Th-fuel cycles by reprocessing and re-fabrication of spent Th FAs.

The programme was planned to run in three phases:

- Phase I (1979-1983): To establish the technological basis for further work on PWR (Th,U)O₂ fuel and demonstrate the principal feasibility of the chosen fuel cycle concept. To develop a (Th,U)O₂ fuel manufacturing technology on a laboratory scale, manufacture fuel for irradiation tests with segmented fuel rods in a research reactor, scope studies of the transferability to the manufacturing process to produce (Th,Pu)O₂ with cerium as a surrogate material.
- Phase II: To demonstrate the feasibility and safety of (Th,U)O₂ fuel, nuclear core design and manufacture process and improve knowledge on spent fuel treatment. To transfer the lab-scale process into a pilot-scale process operated in a manufacturing site, manufacture segmented fuel rods for irradiation and performance testing in a power reactor, transfer the manufacturing process from (Th,U)O₂ to (Th,Pu)O₂ by the use of cerium as simulant material.
- Phase III: To implement an irradiation test programme. To manufacture complete Th-containing pathfinder fuel assemblies for LWRs for commercial operation in power reactors, design a production line for the use of Pu or ²³³U as fissile material.

Phase I was practically completed. Lab-scale production lines in all contributing laboratories were all fully available for test fuel production. First efforts to build a pilot manufacturing line in the Brazilian fuel manufacturing site had been initiated. The feasibility studies on fuel reprocessing and Th/Pu fuel manufacturing indicated no principle problems at first sight.

Within the former D₂O-Th-Programme, (Th,U)O₂ fuel pellets were manufactured in 1964. Since the powder mixing + pressing + sintering approach and the use of vibrational compacted particle fuel were not fully satisfactory, it was emphasised in the German-Brazilian programme to use:

- The chemical ex-gel conversion process resulting in calcined un-sintered (Th,U)O₂ particles where both constituents ThO₂ and UO₂ form a perfect solid solution in combination. This technology was derived from the HTR process.
- The standard pellet pressing and sintering techniques derived from LWR manufacturing.

By the use of this combination of proven HTR and LWR fuel fabrication methods, the manufacturing technology allows maximum benefit to be taken from the existing capability and equipment practiced for standard PWRs and for HTRs fuel manufacturing,

as well as for quality assurance programmes. The combined process could easily fulfil the general fuel manufacturing requirements. However, the needs of the fuel design, fuel in-service performance and the back end of the fuel cycle, needed R&D work for adapting the intersection of both process lines to meet the new requirements.

In the course of the programme, the objectives were adapted to the actual requirements with focus on fuel fabrication, manufacturing and testing, as well as on technology transfer. The programme was determined in the middle of Phase II after it was recognised that the Th/Pu fuel cycle would be the most interesting of the investigated options. Since Brazil was not interested in reprocessing spent fuel at that time, the programme was terminated after the design of a test assembly for the use in the 600 MWe PWR Angra-1.

The results achieved by the programme are summarised in KFA (1979-1988). It contains a comprehensive summary of the status of knowledge at the end of the programme.

4.1.4 Other thoria irradiation programmes

In Canada, the majority of the AECL thoria-based irradiations have been in high-temperature (~300°C), high-pressure (10 MPa) light or heavy water-cooled, heavy water moderated reactors (National Research Experimental [NRX], Nuclear Power Demonstration [NPD] and National Research Universal [NRU]). Other irradiations were performed in the high-temperature, low-pressure, organic-cooled WR-1 reactor, and the low-temperature, low-pressure heavy water moderator in the NRU reactor (Livingstone and Floyd, 2013).

In Germany, in the early 1970s, some (Th,Pu) oxide fuel assemblies were irradiated at the Lingen power plant (BWR). The total plutonium content in the rods was 2.6 w/o with a fissile content of about 86%. The discharge exposure of the fuel rods was in the range of about 20 MWd/kg. Although the Lingen experience was not representative of commercial fuel, it contributed to collecting fundamental data and benchmarking of codes for this fuel material.

In the 1970s and 1980s at the HTGR Atom Versuchs Reaktor (AVR) experimental reactor in Jülich, two sets of irradiation testing of (Th,U)O₂ fuel were performed under simulated PWR conditions with:

- vipac (Th,U)O₂ fuel in the frame of the D₂O-Th-programme;
- (Th,U)O₂-pellets, in order to investigate the irradiation behaviour of the thoria/urania fuel as an “advanced” nuclear fuel manufactured by novel methods.

The test fuel rods including the test fuel were fabricated in accordance with specified processes and proper specifications.

Highly instrumented rodlets had been irradiated simultaneously for a short time. The burn-up achieved a maximum of 10 MWd/kgHM under conditions covering the loads of a PWR. The main objectives of the AVR irradiation programme were to investigate the irradiation behaviour of the vibro-compacted and pellet fuels, the FGR, the thermal conductivity and also to check for any evidence for fuel rod failure mechanisms that are typical for vibro-compacted fuel.

Some important results from these test irradiation programmes are highlighted below.

Fission gas release (FGR)

FGR is dependent on fuel element power (primary factor) and fuel burn-up (secondary factor). During post-irradiation examination of the thorium pellets irradiated at Jülich AVR, none of the fuel rods showed significant diameter changes or fuel swelling. Few data were available to compare the FGR between calculated and measured data for UO₂ fuel and Th-based fuel. Nevertheless, the FGR rate for Th-pellets was between 25 and 61%,

which indicates that U- and Th-based fuels have similar release characteristics, at relatively low burn-up.

Past experiments at AECL confirm non-granular² thoria demonstrates superior performance, with granular³ thoria performing similarly to UO_2 under similar operating conditions. Below ~ 40 kW/m, fuel microstructure plays a minimal role in FGR due to the low-fuel temperature. Figures 4.5 and 4.6 are plots of FGR data from UO_2 (Floyd, 2001) and various thoria-based fuels (Livingstone and Floyd, 2013) plotted against their element power and burn-up.

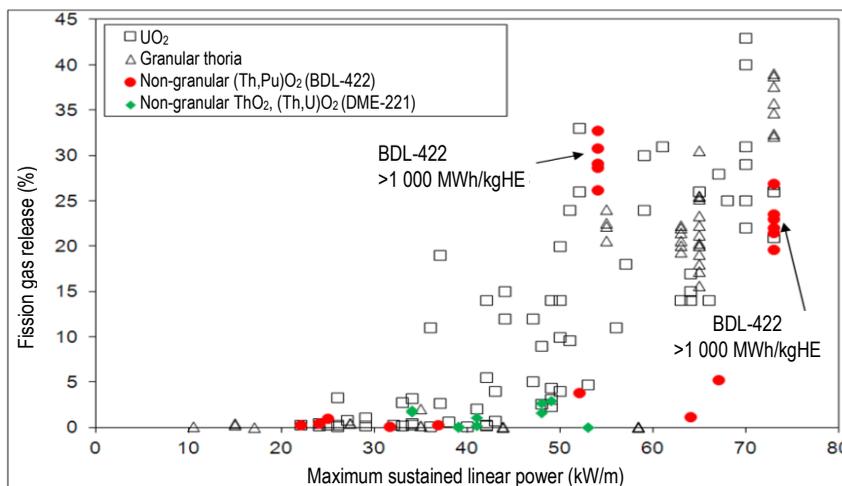
Effect of microstructure

It is expected that thoria-based fuels should experience reduced FGR and operate at lower temperatures compared to UO_2 (operating at the same power). However, this performance is achieved only when the as-fabricated microstructure of the thoria is of high quality as it is noted that fuel with a highly granular structure suffers degradation of its thermal conductivity (IAEA, 2012; Smith et al., 1985).

AECL thoria irradiation experiments confirm improved FGR performance of high-quality non-granular homogeneous thoria fuel over UO_2 of similar powers and burn-ups (Corbett et al., 2013; Floyd, 2001; Karam, Dimayuga and Montin, 2010). Recent tests are represented by the “non-granular” data points in Figures 4.5 and 4.6.

In the case of BDL-422 fuel (1.5 wt% Pu in $[\text{Th},\text{Pu}]\text{O}_2$ [Hastings, 1983]), the FGR was low ($<6\%$) below 42 MWd/kgHE and high ($>19\%$) above 42 MWd/kgHE (Floyd, 2001). For the high burn-up elements in BDL-422, the power and burn-up combination (54-73 kW/m to 42-49 MWd/kgHE) is above that of AECL experience with UO_2 (Manzer, 1996).

Figure 4.5. FGR (%) as a function of maximum sustained linear power (kW/m) for AECL irradiations of thoria-based fuel



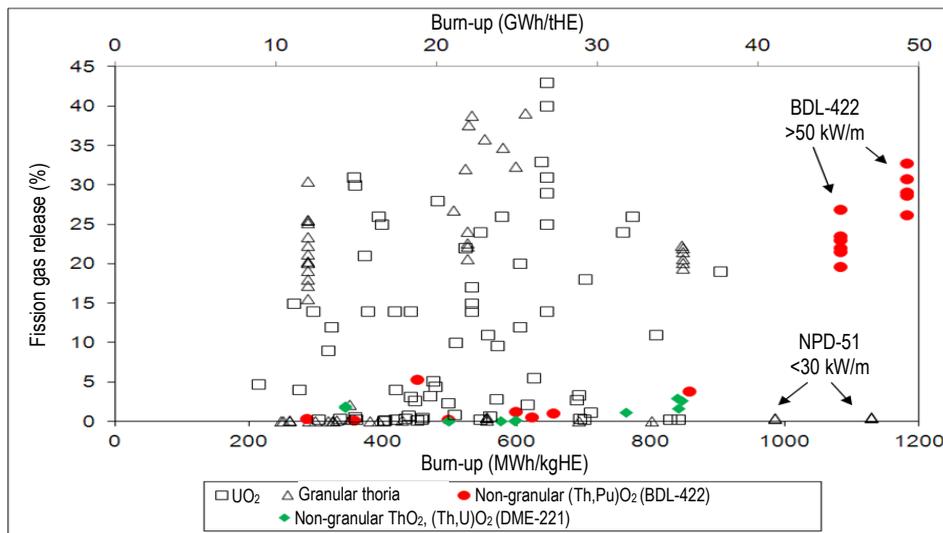
Source : Livingstone and Floyd (2013).

In the case of the Jülich test irradiations, the thermally highly rated vipac fuel restructured rapidly to a solid fuel column, altered by a relatively short irradiation time and a low burn-up. Regarding the microstructure of test fuel pellets irradiated at low

2. i.e. pellets where the uranium and thorium are homogeneously mixed.
3. i.e. pellets where the uranium and thorium are segregated as distinct “grains”.

burn-up, ceramography of the irradiated fuel did not show any irradiation-induced change of the fuel microstructure. The microstructure exhibited a typical crack pattern as for UO_2 fuel.

Figure 4.6. FGR (%) as a function of burn-up for AECL irradiations of thoria-based fuel



Source : Livingstone and Floyd (2013).

4.1.5 Defected thoria fuel

A fuel defect refers to a breach in the fuel sheath that allows heat transport system coolant to enter the element and fission products (and potentially, fuel) to escape from the element. ThO_2 is chemically stable and does not oxidise, even in defected fuel. In comparison, defected UO_2 oxidises to UO_{2+x} , which enhances fuel erosion, degrades thermal conductivity (increased fuel temperature) and enhances FGR.

To date, AECL experience has shown no defect root causes that are unique to thoria-based fuel (Livingstone and Floyd, 2013). Defect root causes in thoria-based fuel experienced at AECL include:

- stress-corrosion cracking from internal gas overpressure or power ramping;
- primary hydriding;
- longitudinal sheath ridging (from low density fuel pellets);
- incomplete endcap-to-sheath closure welds.

All of these defect mechanisms also occur in standard CANDU UO_2 fuel (Manzer, 1996), and are related to manufacturing or operational issues.

Defected thoria fuels irradiated by AECL have consistently exhibited reduced pellet erosion and fission product release (Livingstone and Floyd, 2013) when compared to UO_2 . For example, in one experiment, defected UO_2 (4.5 wt% ^{235}U in total U) and defected $(\text{Th,U})\text{O}_2$ (4 wt% HEU in ThO_2) elements were irradiated under similar conditions. The $(\text{Th,U})\text{O}_2$ exhibited minimal fuel erosion, while the UO_2 eroded under the defect site, as expected. Furthermore, the ^{133}Xe release to the coolant was at least an order of magnitude lower for the defected $(\text{Th,U})\text{O}_2$ element compared to the defected UO_2 element.

4.2 Summary

The technical feasibility of using thorium as a fuel component in current power reactors has been shown in numerous theoretical studies, although the supply of the necessary fissile driver component is often assumed. Thorium dioxide has material properties that make it well suited for use as a fertile fuel matrix in present reactors, especially as compared with uranium dioxide used in current MOX fuels.

Numerous test irradiation programmes of thoria fuels have been carried out in the past and are currently ongoing. Additional tests are foreseen to determine key properties and behaviour of thorium dioxide fuels such as thermal conductivity, swelling and FGR as a function of burn-up. Test results with thoria-based fuel have confirmed expected fuel performance, in terms of centreline temperature and FGR. ThO₂ is more resistant to water corrosion than UO₂ in the case of a defective fuel rod. These results constitute a significant step towards the broader use of thoria fuel ceramics for achieving “near-term” fuel cycle goals.

Thoria-based fuels for LWRs and PHWRs exhibit improved defect performance (in terms of reduced fission product release and reduced erosion) and are a highly prospective technology for consuming or transmuting transuranic nuclides of concern using currently licensed reactor infrastructure.

Thoria-based fuels must be qualified to assure their safe performance in the usual suite of normal/accident scenarios of prime concern to regulators. Processes require further development to manufacture optimal industrial thorium-based fuels. The optimised performance of newly developed fuels will require confirmation with further test programmes.

Before full qualification is achieved, test programmes are anticipated to be performed progressively in the three following phases or levels:

- Level 1 is the irradiation of small numbers of individual fuel rods for the demonstration of the technological and nuclear behaviour of the envisaged thorium-based fuels. This will provide answers to basic questions on characteristics and performance of the fuels. The design of these rods requires the use of existing information on technological and nuclear behaviour and code qualification based on limited irradiation experience and theoretical considerations.
- Level 2 is the insertion of test fuel assemblies predominantly containing thorium-based fuel. This will require qualification of the design code systems at the level of licensing mode nuclear design. Restrictions due to the lack of qualified measurement data and knowledge of technological behaviour may be applicable. This will have to be agreed upon by the respective licensing authority. Generally, no full scope safety analysis is required as long as the number of test assemblies is limited (e.g. ≤ 4). Higher uncertainties may apply in the different domains of analysis. Conservative uncertainties will have to be derived and justified for acceptance by authorities. Test assemblies may already be placed at in-core measurement positions for qualification of the power reconstruction methodologies with the core monitoring systems.
- Level 3 is the introduction of thorium fuel in reload quantities. This will require full qualification of the design codes and thermal-mechanical input data used for technological and nuclear design as well as safety analyses, as there are nuclear data, fuel temperature determination, limits of fuel temperatures, burn-ups and power densities. Power reconstruction with the core monitoring systems has to be sufficiently validated. All safety-related documents shall be extended and licensed. Higher uncertainties may apply until sufficient measurement information is available for complete code qualification with uncertainty analyses. Those conservative uncertainties will have to be derived and justified and may have impact on reactor operation.

All relatively recent programmes have been at Level 1. In Canada, several full-scale PHWR design thorium-based fuel bundles have undergone irradiation, which is a significant progression beyond Level 1 irradiations of small numbers of individual fuel rods.⁴ As has also been mentioned, full core loadings have taken place in the past which would appear to be Level 2 or 3; but these were in the early days of nuclear power and provide little assurance for today's regulatory regimes.

The 2011 NEA report on *Trends towards Sustainability in the Nuclear Fuel Cycle* (NEA, 2011) concludes that “the successful large-scale reactor technology demonstration efforts conducted in the past suggest that there should not be insurmountable technical obstacles preventing the use of thorium fuel and its fuel cycle in existing and evolutionary LWRs. However, the industrial infrastructure, research, design and licensing data are not in place to allow a rapid deployment of thorium fuels in current reactors in the short term”.

The 2014 report by the US NRC on the *Safety and Regulatory Issues of the Thorium Fuel Cycle* (US NRC, 2014), highlights that thorium's “... fundamental nuclear properties have impacts on a number of key areas related to reactor and safety analyses, including steady state and transient performance, fuel handling and management (fresh and irradiated), reactor operations and waste management. The uncertainties on these data and the resulting impact on key safety parameters need to be fully evaluated”. Such evaluation processes, even for existing infrastructures, would be resource- and time-consuming.

References

- Corbett, S., et al. (2013), “DME-221 thorium fuel: Fabrication, irradiation testing and post-irradiation examination”, *Proceedings of the 12th International Conference on CANDU Fuel*, Kingston.
- Floyd, M.R. (2001), “Extended burn-up CANDU fuel performance”, *Proceedings of the 7th International Conference on CANDU Fuel*, Vol. 2, ISBN 0 919784 71 2, Kingston.
- Floyd, M.R. (2011), “Advanced fuel cycle development at Chalk River Laboratories”, *Proceedings of the International Conference on Future of Heavy Water Reactors (HWR-Future)*, Ottawa.
- Hastings, I.J. (1983), “Effect of initial grain size on fission gas release from irradiated UO₂ fuel”, *Journal of the American Ceramic Society*, Vol. 66, No. 9.
- IAEA (2012), *Role of thorium to supplement fuel cycles of future nuclear energy systems*, IAEA Nuclear Energy Series No. NF-T-2.4, International Atomic Energy Agency, Vienna.
- Insulander-Bjork, K., et al. (2015), “Commercial thorium fuel manufacture and irradiation: Testing (Th,Pu)O₂ and (Th,U)O₂ in the Seven-Thirty program”, *Annals of Nuclear Energy*, Vol. 75, pp. 79-86.
- Karam, M., F.C. Dimayuga and J. Montin (2010), “Post-irradiation examination of CANDU fuel bundles fuelled with (Th,Pu)O₂”, *Proceedings of the 11th International Conference on CANDU Fuel*, Niagara Falls.
- KFA (1979-1988), *Program of Research and Development on the Thorium Utilization in PWRs*, Final Report, ISBN 3-89336-019-0, Kernforschungsanlage Juelich G.m.b.H.

4. Thorium-based fuel bundles irradiations in the NRU and NPD reactors. NPD was the first demonstration of a fully licensed, power-producing (25 MWe), CANDU PHWR. Prior to its closure in 1987, it was used for demonstration irradiations of several standard 19-element CANDU fuel assemblies that entirely contained MOX (U,Pu)O₂ (“NPD-40”) and (Th,U)O₂ (“NPD-51”) fuel pellets to burn-ups <40 MWd/kgHE (Livingstone and Floyd, 2013).

- Livingstone, S.J. and M.R. Floyd (2013), "Thoria irradiation and post-irradiation examination experience at AECL", *Proceedings of the 12th International Conference on CANDU Fuel*, Kingston.
- Manzer, A.M. (1996), "CANDU fuel performance", *Proceedings of the Water Channel Reactors Fuels and Fuel Channels: Design, Performance, Research and Development*, Technical Committee Meeting, IAEA-TECDOC-997, Vienna.
- NEA (2011), *Trends towards Sustainability in the Nuclear Fuel Cycle*, OECD, Paris.
- Porsch, D. and D. Sommer (2002), "Thorium fuel in LWRs: An option for an effective reduction of plutonium stock piles", presented at the NEA Workshop on Advanced Reactors with Innovative Fuels (ARWIF-2001), 22-24 October 2001, Chester.
- Smith, A.D., et al. (1985), "In-reactor measurement of operating temperature in (Th,U)O₂ fuel: Effect of pellet microstructure", *Proceedings of the 6th Annual Conference of the Canadian Nuclear Society*, Ottawa.
- Somers, J., D. Papaoiannou and D. Sommer (2009), "Irradiation of thorium-plutonium mixed oxide fuel to 37.7 GWd/tonne in the Obrigheim Pressurised Water Reactor (KWO)", *Proceedings of Global 2009*, French Nuclear Energy Society (SFEN), Paris.
- US NRC (2014), *Safety and Regulatory Issues of the Thorium Fuel Cycle*, NUREG/CR-7176, US Nuclear Regulatory Commission, Washington

5. Thorium fuel cycles in present day reactors

5.1 Thorium utilisation in light water reactors

A brief overview has been given on past LWR thorium-related research in Section 1.2. There is currently no identified preference for the use of thorium fuels in BWRs or PWRs, though comprehensive nuclear design studies have been performed to determine the technical feasibility of implementing a thorium fuel cycle in modern LWRs. These have shown that a reasonable fissile content in the thorium fuels and reload batch sizes comparable to U/Pu use would be adequate for creating equivalent annual cycles.

The safety characteristics of core designs with thorium fuels do not indicate the necessity for adaptations of the reactor shutdown or other safety-related systems from those used for uranium fuel. It is feasible for standard PWRs to operate with thorium fuels without changes to the fuel assembly design. However, more detailed safety evaluations, in particular with three-dimensional transient calculations, would be needed before final conclusions could be drawn and possible limitations could be identified.

A more general view on the *pros* and *cons* of the use of thorium fuels in PWRs derived from the results of past R&D programmes, some of which are mentioned in the previous chapter, are shown in Table 5.1.

Table 5.1. Advantages and disadvantages of thorium PWR fuel compared to UO₂-based fuel

	Pro	Con
Thorium characteristics	High-thermal capture cross-section results in high ²³³ U breeding ratio. Stable element, no oxidation or corrosion.	Requires high amount of initial fissile material for initiation of the thorium cycle. Doppler coefficient is less negative than of ²³⁸ U. (n,2n) process results in high production of ²³² U and its daughter product ²⁰⁸ Tl.
Thorium fuels characteristics	Higher melting temperature and higher thermal conductivity.	
²³³ U	Highest η values in thermal region of all fission nuclides. High content of fissile material at discharge makes recycling attractive.	<i>In situ</i> fission of ²³³ U is smaller than Pu in U/Pu cycle ($\sigma_{f, U233} < \sigma_{f, Pu}$).
Highly enriched uranium (HEU)	Highest thorium content in fuel.	Not available for commercial use. Proliferation concerns.
Low-enriched uranium (LEU) (<20% ²³⁵U)	May, to some extent, avoid proliferation concerns.	Significant reduction of thorium usage compared to HEU. Results in significant plutonium production.
Plutonium	Available from reprocessing. Significant reduction of amount and denaturing of plutonium.	Could compromise plutonium inventories required to deploy future FRs.

Table 5.1. Advantages and disadvantages of thorium PWR fuel compared to UO₂-based fuel (Cont.)

	Pro	Con
²³³ Pa	None.	Significant reduction of conversion ratio at high neutron flux.
²³² U	None.	Intensive gammas by daughter nuclides (²⁰⁸ Tl).
Once-through fuel cycle	Non-proliferation aspect: low solubility of thorium fuel. In Th/Pu fuel high degradation and mass reduction rate of plutonium. No further production of plutonium in Th/HEU and Th/Pu cycles.	High content of fissionable material at discharge. The Th/U cycle exhibits only minor savings in natural uranium – if any – compared to U/Pu cycle.
Closed fuel cycle	Higher conversion factor than U/Pu fuel cycle (10%-15% higher). THOREX process is similar to PUREX process.	Thorium fuel more difficult to dissolve, needs addition of hydrofluoric acid. THOREX process needs to be industrially developed. With no countermeasures, breeding of highly fissionable ²³³ U in high-quality (countermeasures result in reduction of attractiveness for thorium fuel cycle). No experience with reprocessing of Th/Pu fuels.
Manufacturing	Known technology.	Th/U assembly manufacturing might need shielding. Th/U/Pu fuel (recycling option) manufacturing is even more penalised. Th/Pu/U fuel not developed.

5.2 Thorium utilisation in heavy water reactors

The use of thorium as a fuel material can be implemented in PHWRs such as CANDU.¹ These reactors are cooled and moderated using heavy water and are continuously fuelled online at full power. The fuel is placed in horizontal fuel channels. Coolant flows past the fuel bundles in the channels, which are kept at high pressure. The space between channels is filled with heavy water moderator. All structural materials are made of zirconium alloys, which have low neutron absorption cross-sections. These design features give the pressure tube heavy water reactor (PT-HWR) a very high neutron economy, since parasitic absorption of neutrons is minimised.

The pressure tube design and the small, simple fuel bundle design (~50 cm long, ~20 kg mass) facilitate a diversity of ways in which new fuels can be introduced to the reactor.

Low-enriched uranium and reactor-grade plutonium (recycled from spent fuel) have been considered as fissile driver materials for thorium fuels in PHWRs. The fissile component can be added via the following three strategies:

1. Note that CANDU reactor fuel has a different nomenclature to LWR fuels. What is the fuel “element” in an LWR is termed a “bundle”, and the individual “pins” are termed “elements”.

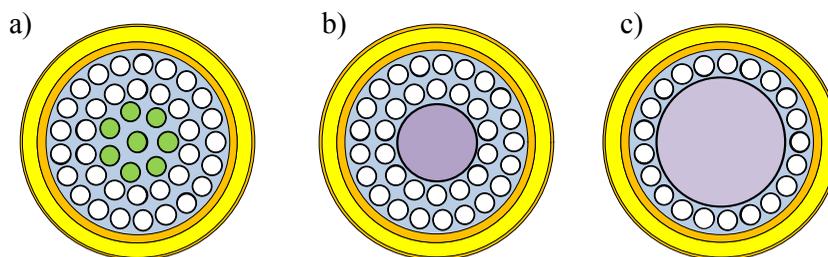
- Heterogeneous bundle: The fertile thorium and the fissile driver are both contained within the fuel bundle, but in separate fuel elements. This strategy has been adopted for the nearest-term deployment option in the advanced fuel CANDU reactor (AFCR).
- Homogeneous bundle: Thorium and the fissile component are mixed homogeneously, (see fuel fabrication in Chapter 3) and are contained in each fuel pin in the bundle.
- Heterogeneous cores: Thorium “blanket” fuel and the fissile driver “seed” fuel are separated into different channels in the reactor. The thorium-based bundles may contain zero, or a small initial amount of fissile fuel (in the form of ^{233}U , ^{235}U and/or Pu). The fissile driver seed fuel bundles could be of any fuel type, but will have a sufficient content of fissile isotopes to sustain reactor criticality and to achieve a desired level of fuel burn-up.

5.2.1 Thorium-based lattice concepts for pressurised heavy water reactors

Several different homogeneous thorium fuel bundle concepts have been developed by Canadian Nuclear Laboratories (CNL; formerly AECL) since the 1960s. Concepts have been developed to use a range of fissile additives (e.g. reactor-grade Pu, weapons-grade Pu, LEU, HEU, ^{233}U) and for a variety of reactor designs, including reactors that are marketed commercially.

Alternative fuel bundle and lattice concepts for thorium-based fuel cycles in PHWRs have been explored recently to achieve maximum resource utilisation. PHWRs are advantageous because of their neutron economy, online refuelling capability and flexibility in using one, two, or more different types of fuels (Griffiths, 1983; IAEA, 2002). Homogeneous or heterogeneous PT-HWR cores can be optimised for power density, burn-up and fissile fuel utilisation, as well as production of new fissile material. In an optimised heterogeneous core, higher fissile content seed driver fuel produces power, while lower fissile content blanket fuel produces ^{233}U . Five different lattice concepts were investigated for potential use in a once-through thorium cycle in a conventional PT-HWR (lattice pitch = 28.6 cm). Lattices involved 43, 35 and 21 element bundles with a central cluster of ThO_2 elements (43-element bundle only) or a zircaloy-4 central displacer tube containing either stagnant D_2O coolant or solid ZrO_2 , to help reduce coolant void reactivity (CVR) (see Figure 5.1). The fuel in the outer elements is a homogeneous mixture of Th and LEU (~5 wt% $^{235}\text{U}/\text{U}$) or reactor-grade Pu (~67 wt% fissile). The content of the LEUO_2 or PuO_2 mixed with ThO_2 was varied to achieve different burn-ups.

Figure 5.1. PHWR high utilisation thorium-based lattice concepts



a) 43-element bundle with 8 ThO_2 elements in central region; b) 35-element bundle with central zircaloy-4 tube filled with ZrO_2 or D_2O coolant; c) 21-element bundle with central zircaloy-4 tube filled with ZrO_2 or D_2O coolant.

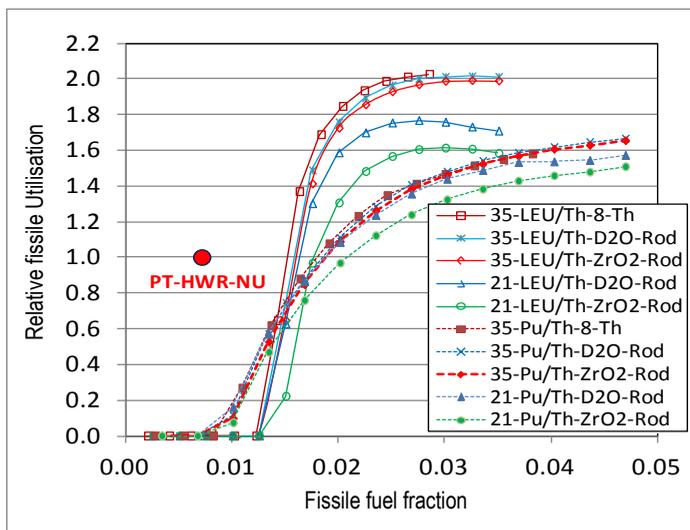
Source: Bromley (2014).

Bundles with ≥ 35 wt% LEUO_2 (≤ 65 wt% ThO_2), or ≥ 3 wt% PuO_2 (≤ 97 wt% ThO_2) are suitable for seed, while bundles with ~ 20 wt% LEUO_2 (~ 80 wt% ThO_2) or 1 wt% to 2 wt% PuO_2 (98 wt% to 99 wt% ThO_2) are suitable for blanket fuel. As stated previously, the fissile

content of the uranium used in the LEUO₂ is 5 wt% ²³⁵U/U. Burn-ups ranging from ~10 MWd/kgHE to 80 MWd/kgHE are possible. As shown in Figure 5.2, the fissile utilisation (energy yield per mass of initial fissile fuel) could be 60% to 100% higher than what is achieved in a PT-HWR using natural uranium fuel (~1 056 MWd/kg-fissile).

Depending on the lattice type, burn-up and fuel composition, the burn-up-averaged CVR ranges from ~+1 mk to +16 mk and is lowest for the 21-element bundle concepts. Leakage may reduce CVR by another 1 mk to 6 mk.

Figure 5.2. Relative fissile utilisation for various PT-HWR thorium-based fuel bundle concepts



Source: Bromley (2014).

Selected cases for each lattice concept with target burn-ups close to ~20 MWd/kgHE and ~45 MWd/kgHE are shown in Figure 5.3. For the same bundle-average fissile content, the 35-element bundles with a central zircaloy-4 tube filled with stagnant D₂O coolant are able to achieve higher burn-ups and fissile utilisation, however, these lattices will also have higher values of CVR (+12 mk to +16 mk, with a reduction of 2 mk to 3 mk due to leakage).

With an assumed linear element rating (LER) limit of ~50 kW/m, the maximum bundle power for the various lattice concepts ranges from ~520 kW to 800 kW. A core with more channels at a smaller lattice pitch (e.g. 24 cm), or higher LER limits for the thorium-based fuels will prevent power de-rating.

Figure 5.3. Low-void thorium bundle concepts for (left) mid burn-up (right) and high burn-up

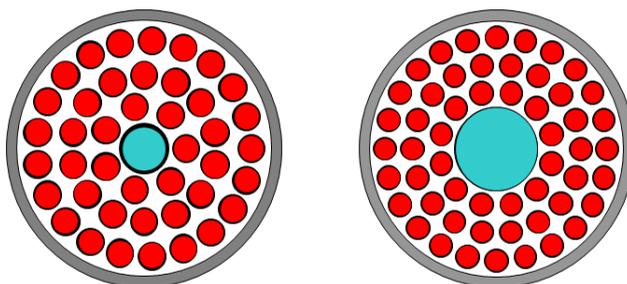


Table 5.2. Performance of high utilisation thorium-based lattice concepts for PHWRs

Lattice concept	Burn-up (MWd/kg-HE)		LEUO ₂ mass fraction in fuel*	Bundle average fissile mass fraction**	Fissile utilisation (MWd/kg-fiss)	FU relative to PT-HWR-NU***
	Low	High				
35-LEU/Th-8-Th (Figure 5.1 a)	Low	23.5	0.400	0.0163	1 447	1.37
	High	45.8	0.550	0.0224	2 044	1.93
35-LEU/Th-D ₂ O-Rod (Figure 5.1 b)	Low	27.5	0.350	0.0175	1 572	1.49
	High	45.0	0.450	0.0225	2 000	1.89
35-LEU/Th-ZrO ₂ -Rod (Figure 5.1 b)	Low	26.1	0.350	0.0175	1 493	1.41
	High	44.1	0.450	0.0225	1 960	1.86
21-LEU/Th-D ₂ O-Rod (Figure 5.1 c)	Low	24.1	0.350	0.0175	1 375	1.30
	High	46.3	0.500	0.0250	1 853	1.75
21-LEU/Th-ZrO ₂ -Rod (Figure 5.1 c)	Low	17.9	0.350	0.0175	1 023	0.97
	High	46.6	0.550	0.0275	1 695	1.61
Lattice concept	Burn-up (MWd/kg-HE)		PuO ₂ mass fraction in fuel*	Bundle average fissile mass fraction**	Fissile utilisation (MWd/kg-fiss)	FU relative to PT-HWR-NU***
	Low	High				
35-Pu/Th-8-Th (Figure 5.1 a)	Low	21.7	0.035	0.0191	1 137	1.08
	High	46.4	0.055	0.0300	1 548	1.47
35-Pu/Th-D ₂ O-Rod (Figure 5.1 b)	Low	23.6	0.030	0.0201	1 172	1.11
	High	47.2	0.045	0.0302	1 564	1.48
35-Pu/Th-ZrO ₂ -Rod (Figure 5.1 b)	Low	23.1	0.030	0.0201	1 149	1.09
	High	46.6	0.045	0.0302	1 544	1.46
21-Pu/Th-D ₂ O-Rod (Figure 5.1 c)	Low	23.0	0.030	0.0201	1 146	1.08
	High	45.8	0.045	0.0302	1 520	1.44
21-Pu/Th-ZrO ₂ -Rod (Figure 5.1 c)	Low	20.6	0.030	0.0201	1 023	0.97
	High	42.2	0.045	0.0302	1 398	1.32

* Fissile content of LEUO₂ is 5 wt% ²³⁵U/U. Fissile content of PuO₂ is 67wt% (²³⁹Pu + ²⁴¹Pu)/Pu.

** For example, for the 35-element and 21-element bundles, if the LEUO₂ mass fraction is 0.350, then the ThO₂ mass fraction is 0.650 and the bundle-average fissile mass fraction $\sim 0.050 \times 0.350 = 0.0175$. If the PuO₂ mass fraction is 0.030, then the ThO₂ mass fraction is 0.970 and the bundle-average fissile mass fraction $\sim 0.030 \times 0.67 = 0.0201$. For the 43-element bundle, the mass of the ThO₂ in the central 8 pins has to be taken into account in determining the bundle-average fissile mass fraction. For example, if the LEUO₂ mass fraction is 0.400, then the bundle-average fissile mass fraction $\sim (0.400 \times 0.05) \times 35/43 \sim 0.0163$. If the PuO₂ mass fraction is 0.035, then the bundle-average fissile mass fraction $\sim (0.035 \times 0.67) \times 35/43 \sim 0.0191$.

*** Fissile utilisation for PT-HWR with natural uranium fuel is $\sim 1\ 056$ MWd/kg-fiss (7.5 MWd/kg / 0.0071).

5.2.2 Low coolant void reactivity concepts

The change of fuel material and geometry affects the reactivity coefficients and kinetics of the reactor. Replacing ²³⁸U with ²³²Th as the primary fertile material reduces the delayed neutron fraction, β and it also reduces the neutron resonance absorption. Concepts which use plutonium rather than ²³⁵U as a fissile driver have a further reduction in the delayed neutron fraction. A lower value of β reduces the margin for approaching prompt criticality ($\rho=\beta$), when the growth in neutron multiplication becomes exponential. To compensate for the reduction in margin, the reactivity coefficients, in particular the CVR coefficient, should be decreased. These bundle concepts have been developed with a CVR of $\sim +5$ mk, a reduction of 65% from that of a natural uranium-fuelled HWR. This reduction is greater than the expected decrease in β for thorium-based fuels with Pu as the main fissile driver.

Two bundle concepts have been developed (see Figure 5.3), for a mid-range burn-up of ~ 20 MWd/kgHE and a higher burn-up of ~ 45 MWd/kgHE. These bundle concepts each

have a central larger element containing a neutron-absorbing poison. Hafnium was used in these simulations, but the composition of the centre poison can be adjusted further when a particular bundle concept is chosen for fuel cycle development. The remaining elements are a homogeneous mixture (dioxide) of either; reactor-grade Pu and thorium, reactor-grade Pu, recycled ^{233}U and thorium, or 20 wt% $^{235}\text{U}/\text{U}$ enriched LEU and thorium. While the use of a central poison pin can help reduce the CVR, the trade-off is that the neutron economy and the achievable burn-up and fissile utilisation will be reduced.

For the high-burn-up and recycled ^{233}U cases, the fuel in the bundle used a spatially graded fissile content, with a higher fissile content in the outer rings. This grading helps minimise the amount of poison needed in the central absorbing element, which leads to better utilisation of the fuel. However, this grading also leads to larger radial form factors. In order to decrease the LER, the size of the fuel elements was reduced and the number of fuel elements increased. A maximum LER of 60 kW/m was assumed for this study. This value is greater than the nominal LER for uranium fuel, due to the expected superior fuel performance characteristics of thorium. This change in geometry results in approximately 10% less fuel in the bundle. The low-burn-up cases have 42 fuel elements, with 7, 14 and 21 elements in the inner, intermediate and outer rings, respectively. The high-burn-up cases have a larger centre element, and 12, 18 and 24 fuel elements in the inner, intermediate and outer rings, respectively. The bundle composition is given in Table 5.3.

Table 5.3. Compositions and results for the low void homogeneous thorium concepts

Case	Burn-up (MWd/kg HE)		Number of fuel elements	Bundle average Pu % wt or LEU wt%	Bundle average ^{233}U wt%	Fissile utilisation (MWd/kg-fiss)	Relative fissile utilisation to HWR NU
	Low	High					
Pu-driven thorium	Low	19.4	42	3.5	N/A	830	0.79
	High	45.0	54	4.9	N/A	1 375	1.30
Pu-driven thorium with ^{233}U recycle	Low	19.7	42	0.8	1.4	1 018	0.96
	High	44.0	54	2.1	1.4	1 570	1.49
LEU-driven thorium	Low	20.1	42	12.2	N/A	824	0.78
	High	44.3	54	14.2	N/A	1 560	1.48

The placement of ^{233}U in the bundles in the recycle cases is not in a configuration designed to maximise the breeding of ^{233}U . The requirement for these models was to have approximately the same amount of ^{233}U input and output (or slightly more on output to allow for losses during reprocessing). Thus, the ^{233}U -recycle concept is self-sustaining, and the added Pu or LEU allows the bundle to achieve the desired burn-ups (e.g. ~20 MWd/kgHE, 45 MWd/kgHE).

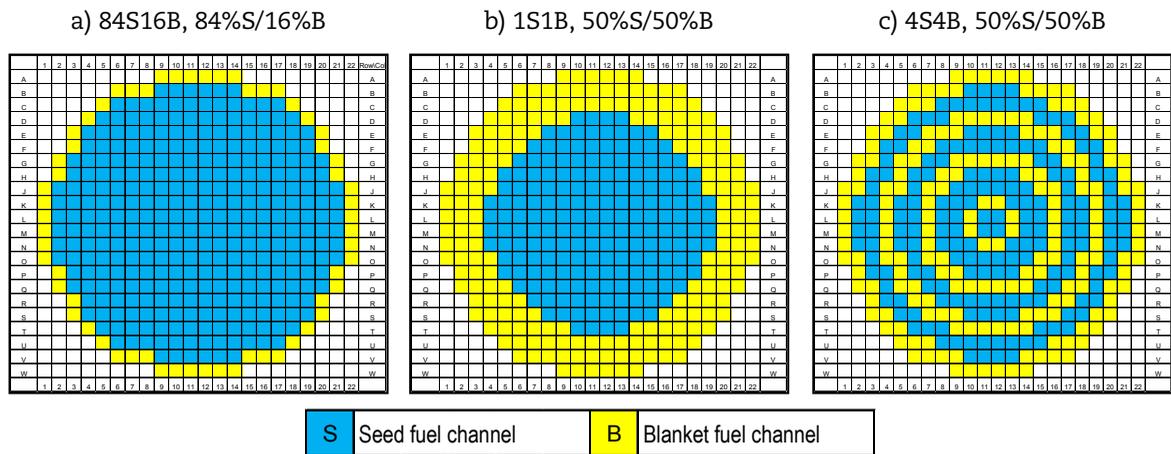
As noted above, the results show that reducing the CVR by the use of a neutron absorbing material in the central pin has a trade-off in fissile utilisation; for the low burn-up fuels, the fissile utilisation of the thorium fuel is lower than that of the typical natural uranium-fuelled HWR. However, this reduction can be mitigated by pushing the fuel to higher burn-ups with a higher initial fissile content. At burn-ups of 45 MWd/kgHE, the fissile utilisation of these thorium options is up to 50% higher than a PHWR operating with natural uranium fuel.

5.2.3 Heterogeneous seed/blanket core concepts for thorium-based fuels

Heterogeneous seed/blanket core concepts (Radkowsky, 1986) for implementation of thorium-based fuels in PHWRs have been explored to achieve maximum resource

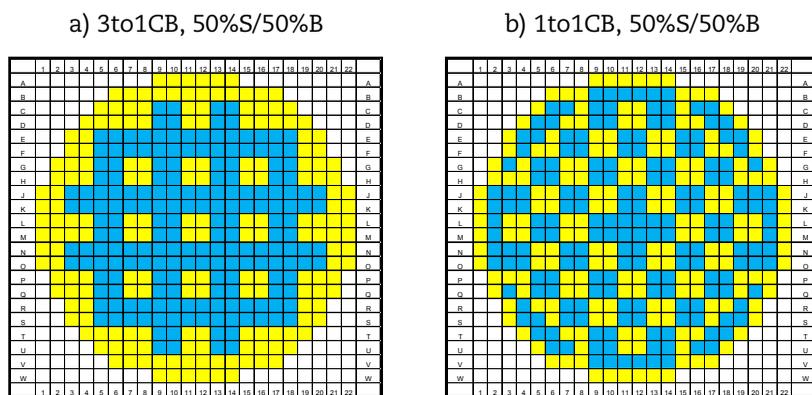
utilisation with an once-through thorium fuel cycle (Milgram, 1982 and 1984). The use of higher-fissile-content seed fuel bundles and lower-fissile content blanket bundles that are physically separated in the core allows more flexibility and control in fuel management to maximise the fissile utilisation and production of ^{233}U . The lattice concept utilised was a 35-element bundle made with a homogeneous mixture of reactor grade PuO_2 (~67 wt% fissile) and ThO_2 , with a central zirconia rod to reduce CVR. Seed fuel was 3 to 4 wt% PuO_2 , while blanket fuel was 1 to 2 wt% PuO_2 . Several annular and checkerboard-type core concepts in a 700-MWe-class PT-HWR (Ovanes et al., 2012) in a once-through thorium fuel cycle were analysed (see Figures 5.4 and 5.5). Cores contained 50% to 84% seed fuel; the balance was blanket fuel.

Figure 5.4. Annular heterogeneous seed/blanket PT-HWR core concepts



Source: Bromley and Hyland (2014).

Figure 5.5. Checkerboard-type heterogeneous seed/blanket PT-HWR core concepts



Source: Bromley and Hyland (2014).

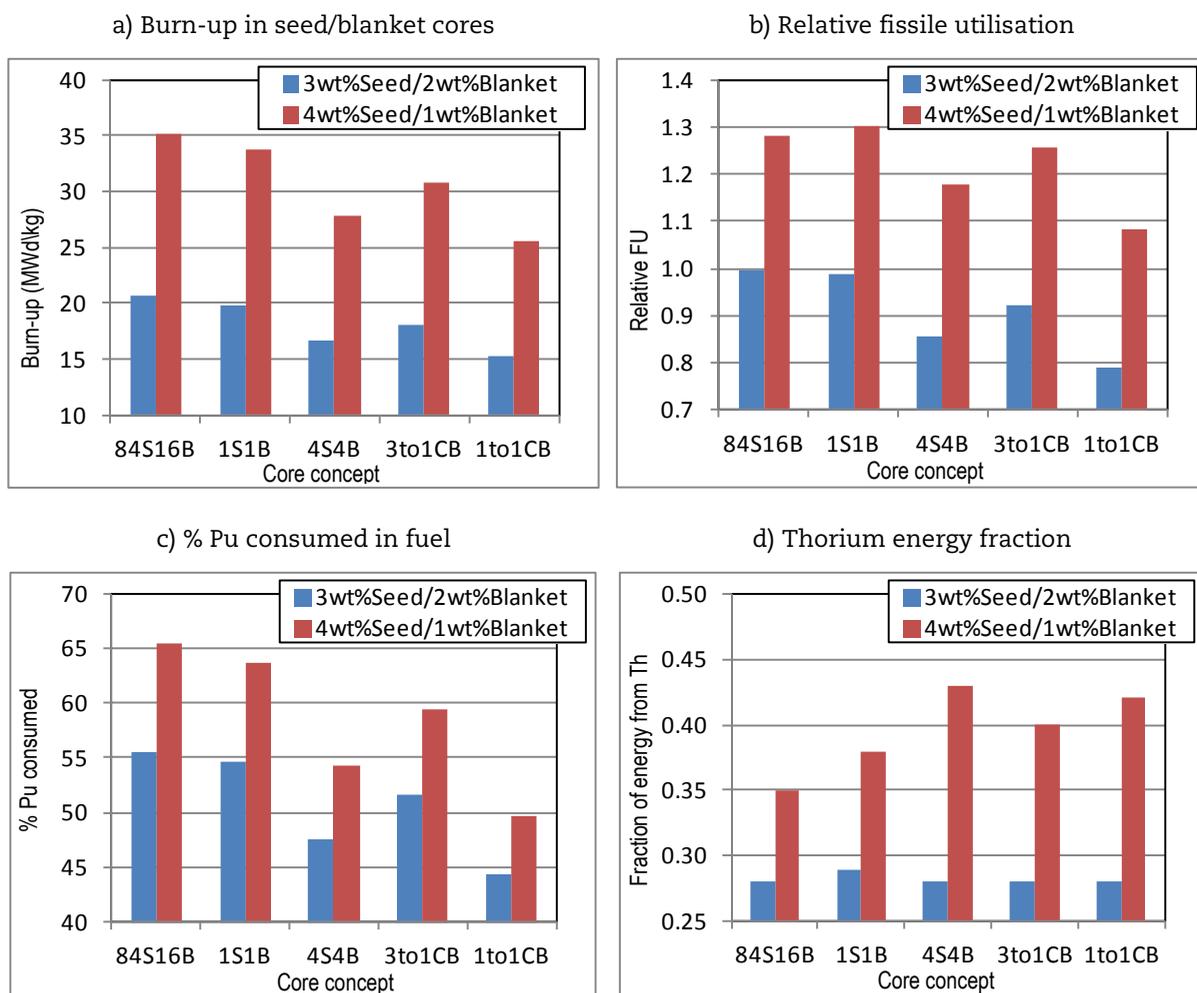
Different combinations of seed/blanket fuel were studied to determine the impact on core-average burn-up, fissile utilisation, power distributions and other performance parameters. Reactor physics calculations were performed with the WIMS-AECL/RFSP code system (Altiparmakov, 2008; Liang et al., 2008; Shen et al., 2010).

Figure 5.6 shows how various core concepts can achieve core-average burn-ups ranging from ~15 MWd/kgHE to 35 MWd/kgHE, the highest being for an annular core with 84% seed. In Figure 5.6, it is observed:

- The fissile utilisation is up to 30% higher than is currently achieved in a PHWR using natural uranium fuel bundles (~1 000 MWd/kg-fissile).
- Up to 67% of the Pu is consumed and up to 43% of the energy is produced from thorium and up to 345 kg/year of fissile U (mainly ²³³U) is produced in the discharged fuel.

Two-region annular cores give higher burn-up and fissile utilisation, while checkerboard cores give a slightly higher conversion ratio (production of fissile nuclei/consumption of fissile nuclei). Core CVR is estimated to range from +7 mk to +10 mk. Cores with nearly 50% blanket fuel must be de-rated to 58% to 74% of full power to avoid exceeding bundle power limits. Utilities may consider such a reduction in operating power unacceptable from a commercial perspective. Use of an alternative fuel bundle design with enhanced heat transfer characteristics to permit higher bundle and channel powers and a smaller lattice pitch (for example, 24 cm instead of 28.6 cm) to permit more channels (538 instead of 380) may help mitigate this operational constraint.

Figure 5.6. Performance of seed/blanket cores in PT-HWR with Pu/Th fuel



Source: Bromley and Hyland (2014).

5.3 Thorium fuel cycle system scenarios

If optimising the use of natural resources is the prime driver, the use of thorium in present day or slightly modified thermal neutron reactors can allow for variable levels of savings on uranium consumption which depend on the combination of reactor fuels, reactor types and recycling options considered. These can range, once equilibrium of the systems is reached, from few tens of percent to about 80% as compared to present use of natural resources in conventional reactors, per unit energy produced. A variety of “generic” scenarios which illustrate how these optimisations in the use of natural resources could in principle be achieved are described.

It must be highlighted that the feasibility of some of the generic scenarios presented here is conditioned by major assumptions, which, for the closed fuel cycles, include the need to have industrialised the thorium fuel recycling and re-fabrication processes. These scenarios are therefore given here as a qualitative illustration of different “families” of combination of thorium fuel cycles and reactors that could be envisaged if the necessary conditions are met. They aim at giving the order of magnitude of uranium savings that the use of thorium in these scenarios could allow, when compared with the present use of the uranium mineral in the conventional once-through uranium/plutonium fuel cycle. Nevertheless, as has been stated before, economy of natural resources may or may not be the prime driver to warrant the deployment of such fuel cycles and other factors may intervene over time that could impair or enhance the interest and feasibility of the scenarios presented here. This would include the possible development and deployment of evolutionary, fast, or future generation reactor systems. The symbiotic fuel cycles presented illustrate the possibilities that the planned interdependence of two or more different types of reactors allows in terms of optimisation in the use of fertile/fissile matter.

The economic impact of transitioning to these innovative fuel cycles is not considered in these examples.

5.3.1 Open fuel cycles in thermal reactors

Near-term thorium fuel design proposals, primarily based on a once-through fuel cycle, without recovery of ^{233}U , may be primarily considered by countries currently operating their reactor fleet with uranium fuel in an open cycle mode. Thorium fuels may then be used with LEU, either homogeneously mixed or in separate pins, assemblies or bundles in the core. Thorium may also be considered by countries wishing to dispose of excess plutonium, in which case thorium and plutonium would likely be used as homogeneously mixed fuel.

Core design studies are used to investigate conditions under which thorium fuel, operating for many cycles in a reactor, may build up enough ^{233}U to offset the initially necessary additional fissile driver (^{235}U or plutonium). CANDU PHWRs, featuring a high neutron economy and having primarily been designed for the ability to use natural uranium, are particularly well suited to achieve such a high conversion of thorium into ^{233}U (IAEA, 2002).

Figure 5.7 shows a generic example of two possible uses of thorium fuel in once-through fuel cycles, which can be applied to either LWRs or HWRs. The top example in this figure shows the use of $(\text{Th}, \text{U}_{\text{LEU}})$ fuel with LEU enriched to 19.75% ^{235}U (instead of typically ~4-5% ^{235}U in present LWRs, or ~1% in HWRs). Either LWR or HWR reactors may use a homogeneously mixed $(\text{Th}, \text{U}_{\text{LEU}})$ fuel, or heterogeneously mixed fuel bundles made of separate U_{LEU} and thorium fuel pins, arranged in order to optimise the system’s conversion factor (Boczar et al., 2002). Even though this option slightly improves the conversion factor of the system, it is of limited interest in terms of optimising the use of natural resources in LWRs. While $(\text{Th}, \text{U}_{\text{LEU}})$ fuel will effectively breed ^{233}U , much less

plutonium is produced than in conventional uranium fuel, resulting, overall, in little savings of uranium for a given supply of energy.

Figure 5.7. Indicative values on the use of thorium-based fuels in LWRs with an open fuel cycle



Comparable use can be made in HWRs fuelled with ~5% UO_2 (LEU at 19.75% ^{235}U) and ~95% thorium.

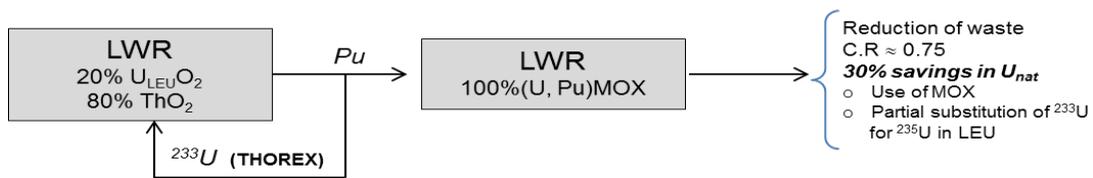
The bottom example in Figure 5.7 consists in burning a thorium-based fuel enriched with plutonium produced in standard uranium-fuelled LWR or HWRs in a two-tiered generating fleet. For LWRs, this approach is believed to achieve approximately 20% savings in uranium, compared to present consumption of LWRs operating in an open fuel cycle. Savings at this level are not higher than those achieved in standard LWRs with a single recycle of plutonium as MOX fuel and reprocessed uranium (saving of ~10% at each step). In terms of natural uranium savings only, this use of thorium would therefore be only moderately attractive, unless the spent thorium/plutonium fuel is reprocessed for retrieving and recycling of ^{233}U in reactors operated with a uranium/thorium fuel cycle at a later stage. This recycle mode would allow for a future transition from a uranium/plutonium fuel cycle into a thorium/uranium fuel cycle in terms of upstream ^{233}U production.

5.3.2 Closed fuel cycles in thermal reactors

The introduction of spent fuel recycle opens up many more fuel cycle options. Most nuclear countries, which currently reprocess spent nuclear fuel or acknowledge recycle as an essential feature of sustainable nuclear power, consider closed thorium fuel cycles as the most efficient path to the use of thorium, through its conversion into ^{233}U . As for uranium, open fuel cycles lead to an utilisation of less than 1% of the overall thorium energy content, whereas more than 80% can be burned as ^{233}U with the implementation of multiple recycle.

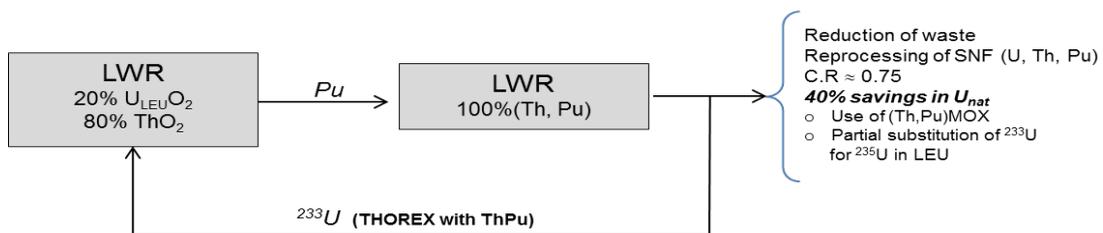
Separating ^{233}U allows fuel to be manufactured with an inherent fissile content analogous to enriched uranium. Separating the thorium also allows it to be recycled back to the reactor. However, the reprocessing of thorium-based fuel cannot be achieved with processes that are currently used for uranium fuel. It calls for a specific process (THOREX) that requires a mixture of hydrofluoric and nitric acids instead of only nitric acids in the case of uranium and plutonium fuels. The implementation of the THOREX process at industrial scale can therefore only be envisaged in the medium to longer term, after demonstration of the process is made at pre-industrial scales.

Figures 5.8 and 5.9 show examples of symbiotic fuel cycles in a two-tiered generating fleet of LWRs or HWRs with recycling of plutonium and ^{233}U from low-enriched uranium fuels and thorium/plutonium fuels, respectively. Both examples require an industrial use of the THOREX process, not presently available.

Figure 5.8. Potential use of thorium-based fuels in LWRs with a closed fuel cycle

Values are approximate and illustrate only an order of magnitude. Comparable fuel cycles may also be considered with HWRs.

The first example (Figure 5.2) consists of (Th,U_{LEU}) and 100% (U,Pu) MOX-fuelled reactors. Plutonium and ²³³U from spent (Th,U_{LEU}) fuel are recycled. This overall recycle scheme could achieve uranium savings of the order of ~30% compared to standard LWRs operating today with an open uranium fuel cycle.

Figure 5.9. Potential use of thorium-based fuels in LWRs with a closed fuel cycle and Th/Pu reprocessing

Values are approximate and illustrate only an order of magnitude. Comparable fuel cycles may also be considered with HWRs.

The second example (Figure 5.9) considers U_{LEU} and (Th,Pu)-fuelled reactors. Plutonium from spent uranium fuel is recycled in (Th,Pu) fuels, whereas ²³³U arising from this use is recycled as a component of U_{LEU} fuel. This overall recycle scheme could achieve uranium savings of the order of ~40% compared to standard LWRs operating with an open uranium fuel cycle.

(Th, ²³³U) fuel is strongly radioactive from nuclides which are linked to the creation of ²³³U (namely ²²⁹Th, ²³¹Pa, ²³²U and daughter products) and fabrication of this fuel, as has been mentioned before, will require remote fuel manufacturing installations which are costly. Technical and economic studies are needed to assess the commercial viability of such use of thorium, as a function of uranium price, recycled ²³³U and plutonium cost (investment and operating cost for fuel reprocessing and re-fabrication, etc.) and the costs of the resulting back end of the fuel cycle. Non-proliferation (safeguards) issues will also need to be addressed.

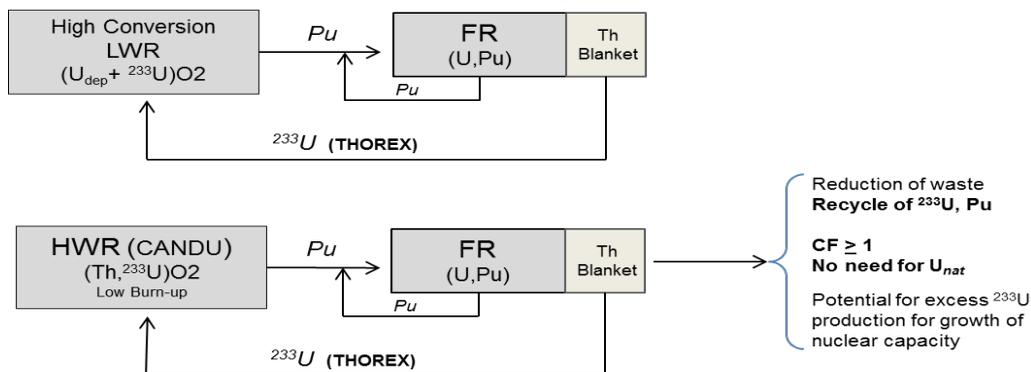
5.3.3 Symbiotic fleets of reactors

An example of symbiotic fleets of thermal and fast reactors

Another example of symbiotic fuel cycles that can be envisaged in two-tiered generating fleets, composed of ²³³U-fuelled light or HWRs and fast neutron reactors breeding plutonium for their own needs, as well as ²³³U from thorium blankets. This approach may achieve a sustainable nuclear production if both types of reactors feature a high-conversion ratio. Figure 5.10 shows two examples of such symbiotic generating fleets with flows of nuclear materials between various reactor types. Both examples require industrial use of the THOREX process.

In the upper example in Figure 5.10, the thermal reactor is a high-conversion LWR using uranium fuel made from recycled ^{233}U and depleted uranium. In principle, this fleet only requires depleted uranium and thorium as fuel, thus achieving an efficient use of both resources. The number of LWRs supported by a fast neutron reactor depends on the breeding ratio of the latter and the conversion factor of the former, which also depends on the fuel discharge burn-up. Increasing the conversion factor calls for reducing the discharge burn-up, as parasitic neutron absorptions in fission products tend to reduce this factor.

Figure 5.10. Use of thorium-based fuels in symbiotic nuclear fuel cycles with LWRs, HWRs and fast reactors



Values given are indicated only.

In the lower example in Figure 5.10, the thermal reactor is a high-conversion HWR operated with a uranium/thorium fuel cycle, thus recycling ^{233}U bred in blankets of fast neutron reactors. These latter reactors need to achieve break-even breeding in plutonium in the core alone (without the blankets). As in the previous fleet, only depleted uranium and thorium are required as fuel, thus allowing this fleet to also achieve an efficient use of both resources. As in the previous example, the number of HWRs supported by a fast neutron reactor will depend on the breeding ratio of the latter and the conversion factor of the former, which also depends on the fuel discharge burn-up. India's nuclear programme has focused on such synergies between advanced HWRs (AHWRs) and fast neutron reactors with advanced fuel cycles based on uranium, MOX and thorium as important components of a self-sufficient energy system. This example is generic as other reactor types such as HTRs or super critical water-cooled reactors may supplement LWRs and HWRs in such symbiotic generating fleets and be supported in the same way by fast neutron breeder reactors.

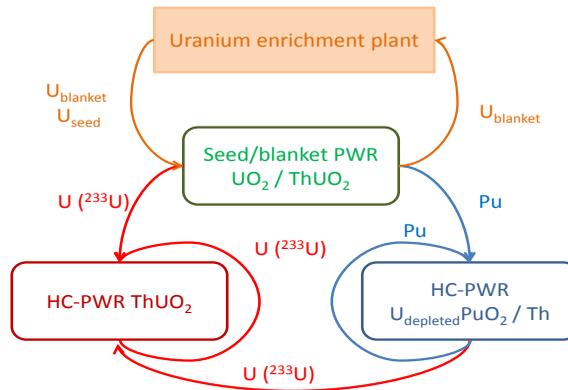
An example of symbiotic fleets of thermal and epithermal reactors

The transition from PWRs with UOX or MOX fuels to a thorium cycle requires either a technological breakthrough in the design of current nuclear reactors, or, with current technology, a very long period of transition. A different approach has been conducted in (Vallet, 2012) in order to take full advantage of the potential of both the uranium and thorium fuel cycles in a symbiotic scenario. It has been shown that annual uranium savings of more than 50% (as compared to the present use of natural uranium) could be expected within a little more than a generation of reactors, via implementation of a two-step scenario using three kinds of symbiotic LWRs.

The first step in Figure 5.11 above involves slightly modified PWRs with assemblies having a “seed/blanket” structure. In these specific PWRs, each assembly has an internal zone of standard UOX fuel pins (enriched to 5 wt% of ^{235}U), and a peripheral zone of mixed (Th,U) O_2 fuel pins with 25 wt% of uranium (enriched to 20 wt% of ^{235}U). The average

fissile content is 5 wt% of ^{235}U in the whole assembly. As these reactors do not require major changes in their design and hence in their neutronic behaviour, the irradiation of thorium in the blanket of these reactors could be initiated at any moment, in order to start building-up a stock of both plutonium and ^{233}U fuels.

Figure 5.11. A two-step scenario using three kinds of symbiotic LWRs



The second step in Figure 5.11 involves the modified PWR mentioned above, but also a high-conversion PWR (HC-PWR). The HC-PWR has a reduced moderation ratio (MR~1) that leads to a high Fissile Inventory Ratio² (FIR~0.8). These modifications to the PWR design have been considered in order to enhance the transmutation of fertile isotopes into fissile ones, while keeping a cycle length of 300 equivalent-full-power-days and a power output equal to 4 250 MW thermal. The plutonium resulting from the reprocessing of UOX or MOX fuels would then feed the HC-PWRs with depleted uranium and MOX fuels, whereas the ^{233}U that has been produced in the blanket pins would feed the HC-PWRs loaded with thorium fuel. Each type of LWR allows for a spatial separation of both uranium and thorium zones in order to ease the fuel reprocessing.

A complete scenario study has been realised based on the French reactor fleet using the COSI code (Meyer and Boucher, 2009). This study includes physical models, which have been validated and describes the variations of the fissile fractions of each fuel in irradiation/cooling/storage times, adjusting the corresponding enrichments in order to maintain the cycle length of each reactor constant; it shows that the transition from the current UOX/MOX cycle in standard PWRs towards a symbiotic reactor fleet would take about 80 years (Vallet, 2012). The length of this transition period illustrates the times needed for such a transition to operate. It has been calculated by taking into account all the fuel cycle steps, rather than only equilibrium inventories for each kind of reactor. After this transition period, the deployed reactor fleet would be composed of 50% of seed/blanket PWRs and 50% of HC-PWRs and the annual uranium savings would be of the order of 50% compared to the open UOX fuel cycle. The study has also demonstrated that the $^{235}\text{U}/\text{U}$ fraction in the discharged blankets is still higher than 5 wt%. Allowing for multi-recycling of this fuel in order to build-up the 20 wt%-enriched UOX of the blankets would, therefore, increase the annual uranium savings up to about 70%.

2. The Fissile Inventory Ratio is the ratio between the mass of fissile isotopes in the discharged fuel and the initial mass of fissile isotopes in the loaded fuel.

5.4 Summary

Technical and economic studies are needed to assess the commercial viability of using thorium, in comparison with fuel cycles using uranium and plutonium only, in different symbiotic generating reactor fleets. This commercial viability depends on: Uranium price, breeding and conversion performance in various reactors, recycled ^{233}U and plutonium costs and back-end costs of the fuel cycle. Non-proliferation issues also require assessment.

If the necessary economic drivers for devoting significant industrial resources to implementing thorium into the nuclear fuel cycle appear (in the form of short-term commercial incentives), then the nearest-term opportunity for establishing a thorium fuel cycle would be the implementation of thoriated fuels in LWRs or HWRs. Although this perspective can be considered a nearer-term one (especially in comparison to other potential uses of thorium), it will take considerable time before the use of thorium fuels in current commercial reactors technologies reaches a level of experience equivalent to current uranium-based fuels (with respect to qualification of the technological and nuclear behaviour).

Multiple practical issues would first need to be resolved for the use of thorium in commercial reactors. Commercial supplies and manufacturing infrastructures do not currently exist. Although the fundamental technology exists, multiple scientific tests related to safety, fuel performance and evaluation of the radioactive source term must be performed and evaluated before a thorium-fuelled reactor could be licensed.

Although significant work has been carried out (and more is planned) at the different levels of thorium utilisation, extensive developments and qualification steps are yet to be accomplished before industrialisation can be considered. Little feedback exists from irradiation of thorium-based fuels in LWRs. This has implications on the status of qualification of codes for nuclear and thermal-mechanical design and on the knowledge of behaviour of thorium-based fuels.

More experimental programmes must be planned to address known technology gaps before thorium-based fuels can be used on an industrial scale. Among these gaps are:

- The qualification of the nuclear data for design, safety analyses, reactor operation, handling, transport and storage.
- The qualification of the design codes for the use with thorium-based fuels. In particular, a qualification is required for the determination of the control rod worth, of reactivity coefficients, the xenon and samarium worth, of the fraction of delayed neutrons, etc.
- The further validity of existing safety analyses. This will be very much dependent on the technological and nuclear behaviour of the fuel and will result in the requirement for repeating safety analyses for the respective plant(s).
- The derivation of uncertainties for application in design, safety analysis and core surveillance functions. A full scope uncertainty analysis requires feedback from critical experiments, with the measurement of the fission rate distributions and from in-reactor measurements. Both require planning as a generic validation programme.
- The qualification of nuclear data and codes for nuclear and thermal-mechanical design requires extension to higher burn-up for nominal and off-nominal operating conditions. Code qualification requires validation based upon irradiation tests followed by post irradiation examinations of high burn-up thorium fuel at different operational conditions.

References

- Altiparmakov, D.V. (2008), "New capabilities of the lattice code WIMS-AECL", *Proceedings of PHYSOR 2008*, Interlaken.
- Boczar, P.G., et al. (2002), "Thorium fuel-cycle studies for CANDU reactors, in thorium fuel utilization: Options and trends", *Proceedings of three IAEA meetings held in Vienna in 1997, 1998 and 1999*; IAEA TECDOC-1319, Vienna.
- Bromley, B.P. (2014), "High utilization lattices for thorium-based fuels in heavy water reactors", *ANS Nuclear Technology Journal*, April 2014.
- Bromley, B.P. and B. Hyland (2014), "Heterogeneous cores for implementation of thorium-based fuels in heavy water reactors", *ANS Nuclear Technology Journal*.
- Griffiths, J. (1983), *Reactor Physics and Economic Aspects of the CANDU Reactor System*, AECL-7615, Chalk River.
- IAEA (2002), *Heavy water reactors: Status and projected development*, IAEA Technical Report Series No. 407, Vienna.
- IAEA (2012), *Role of thorium to supplement fuel cycles of future nuclear energy systems*, IAEA Nuclear Energy Series No. NF-T-2.4, International Atomic Energy Agency, Vienna.
- Liang, T., et al. (2008), "Improvement and qualification of WIMS utilities," *Proceedings of 29th CNS Annual Conference*, Toronto.
- Meyer M. and L. Boucher (2009), "New developments on COSI6, the simulation software for fuel cycle analysis", *Proceedings of GLOBAL 2009*, Paris.
- Milgram, M.S. (1982), *Once-Through Thorium Cycles in CANDU Reactors*, AECL 7516, Chalk River.
- Milgram, M.S. (1984), *Thorium Fuel Cycles in CANDU Reactors: A Review*, AECL 8326, Chalk River.
- Ovanes, M., et al. (2012), "Enhanced CANDU-6: Reactor and fuel cycle options – natural uranium and beyond", *Proceedings of PHYSOR 2012*, Knoxville.
- Radkowsky, A. (1986), "Seed-blanket reactors", *CRC Handbook of Nuclear Reactor Calculations*, Volume 3, (Y. Ronen, editor), CRC Press.
- Shen, W., et al. (2010), "Evolution of computer codes for CANDU analysis", *Proceedings of PHYSOR 2010*, Pittsburgh.
- Vallet, V. (2012), *Recherche de l'économie des ressources naturelles par des études de conception de cœurs de réacteurs à eau légère et à haut facteur de conversion à combustibles mixtes thorium/uranium/plutonium*, PhD.

6. Thorium fuel cycles in molten salt reactor designs

The full benefits of a closed, self-sustaining thorium/²³³U fuel cycle may only be realised in dedicated breeder reactors (generation IV or beyond) which are now still in the design study phase and may not appear before the end of this century. We focus here on molten salt reactors (MSRs), which stand out as a specific class of breeder reactor system due to their very innovative approach to fuel management in liquid form. In recent years, MSRs have been at the centre of a renewed interest in thorium due to some MSR concepts having historically considered thorium as their reference fuel. The current status of MSR and thorium research within the Generation IV International Forum, as well as potentials and current limitations, are highlighted here.

6.1 The MSR concept

MSR concepts are a broad family of fission reactors in which molten salts may serve either as both the primary coolant and fluid fuel of a reactor, or only as the reactor coolant. Within the scope of this report, however, the term MSR will specifically refer to fluid-fuelled systems. Fluid-fuelled reactors benefit from potential advantages over solid-fuelled systems, including:

- a high coefficient of thermal expansion which provides a large negative temperature coefficient of reactivity;
- the possibility of continuous fission-product removal using physical (inert gas sparging) and pyrochemical processes;
- the possibility of fuel composition (fertile/fissile) adjustment without shutting down the reactor;
- the possibility of overcoming the difficulties of solid fuel fabrication/re-fabrication with large amounts of transuranic elements (TRU);
- the potential for better resource utilisation by achieving high fuel burn-ups (with transuranic elements in principle remaining in the fluid fuel of the core to undergo fission or transmutation to a fissile element).

MSRs have long been of particular interest when used with the thorium/²³³U fuel cycle due to the possibility for online continuous removal of neutron absorbers, including noble gases, lanthanides and, in particular, ²³³Pa, precursor nuclide of ²³³U. MSRs also allow the possibility of continuous recycling of ²³³U and TRUs in a closed fuel cycle. This potential, combined with other favourable properties of MSRs, allows, at least in principle, to reach breeding in some MSR concepts.

MSR development is still at a conceptual study stage, with most investigations of these concepts based today on numerical modelling (with the exception of China, where a large effort of thorium MSR prototype development started recently). Experimental studies were conducted in the past at ORNL in the 1950s and 1960s, which provided an experimental basis for the feasibility of such reactors. In 1958, a water-based liquid fuel was used in a 5 MWt homogeneous reactor experiment (called HRE-2), demonstrating the auto-stability of homogeneous reactors. From 1966 to 1969, the Molten Salt Reactor Experiment (MSRE) operated an 8 MWt experimental graphite-moderated MSR,

demonstrating that using a molten fluoride salt at 650°C for four years was possible. However, the MSRE only tested fissile isotopes (^{233}U , ^{235}U and Pu) and not thorium for breeding. Later on, ORNL studied a power reactor called molten salt breeder reactor (MSBR) that was never built. This 1 GWe design was a thermal reactor with graphite moderated core that required a heavy chemical fuel salt treatment with removal time of approximately 30 days for soluble fission products; a drawback that could potentially be eliminated by using a fast spectrum instead.

In general, many MSR concepts have been proposed considering different fuel options (uranium, plutonium and thorium-based) and diverse salt compositions (chlorides, fluorides). These research projects have had as a common goal to define an optimum fuel salt composition, which at the same time is able to satisfy a broad range of different constraints in terms of:

- neutronic properties (neutron moderation, breeding ratio, fissile inventory);
- operating conditions (melting temperature, radiation stability, transport properties);
- actinide and fission products solubility in the molten salt (homogeneity of the core composition);
- materials compatibility and salt chemistry control;
- online or in-batch processing feasibility;
- resulting waste.

Within the MSR System Steering Committee (SSC) of the Generation IV International Forum (GIF/MSR),¹ simulation studies and conceptual design activities are ongoing to verify that fast spectrum MSR systems satisfy the goals of generation IV reactors in terms of sustainability (closed fuel cycle, breeder system), non-proliferation (integrated fuel cycle, multi-recycling of actinides), safety (no reactivity reserve, strongly negative feedback coefficient) and waste management (actinide burning capabilities).

Two fast spectrum MSR concepts studied under the GIF/MSR are presented in this chapter: the molten salt fast reactor (MSFR) concept (initially developed at CNRS, France) and the molten salt actinide recycler and transmuted (MOSART) concept (under development by Russia).

The reference MSFR concept is a 3 GWt reactor a homogeneous liquid salt fuel 3 GWt reactor concept (Mathieu et al., 2009; Brovchenko, 2013; Merle-Lucotte, 2013). In Russia, MSR activities focus on the development of the 2.4 GWt MOSART system (Ignatiev et al., 2014), initially studied as an efficient burner of TRU waste from spent UOX and MOX LWR fuel without any uranium or thorium support. The MOSART system may also be fuelled more generally with different compositions of plutonium and MA trifluorides with or without thorium support.

These two concepts, MSFR and MOSART, illustrate the existing interest in fast spectrum MSR concepts potentially based on the thorium fuel cycle that are seen as a possible long-term alternative to solid-fuelled fast neutron reactors.

In China, under support of the Chinese Academy of Sciences, the Centre for Thorium Molten Salt Reactor System at the Shanghai Institute of Applied Physics was founded in 2011. Since then, the TMSR Centre has undertaken an ambitious project to develop a liquid-fuelled MSR prototype (called TMSR-LF1). TMSR-LF1 is currently at a pre-conceptual development stage and will employ sufficient thorium in its fuel salt

1. Participating countries and organisations in the GIF/MSR are EURATOM, France, Russia (full members) and China, Japan, Korea and the United States (active observers).

composition to enable small-scale demonstration of fuel salt processing technologies in a reactor environment, while relying on a low-enrichment uranium fuel cycle to maintain criticality.

In its last *Technology Roadmap Update for Generation-IV Nuclear Energy Systems (2014)* (GIF, 2014), the GIF/MSR System Steering Committee has extended the viability study phase of MSR concepts at least until 2025. In particular, the MSR viability study phase will need to focus on:

- materials compatibility;
- salt chemistry control;
- tritium confinement;
- confirmation of bubbling efficiency for gaseous fission products;
- heat exchanger viability;
- validation of processing flow sheets at laboratory scale;
- definition of safety analysis methodology and specification of accident scenarios.

6.1.1 Fast spectrum MSR fuel composition options

MSRs may be in principle operated with various fissile and fertile elements in the fuel composition. Molten fluorides salt mixtures containing LiF, NaF or BeF₂ are considered as candidate solvents for transuranic burning/breeding respectively, with and without U and Th support. The status and characteristics of the MOSART and MSFR designs operated in the Th/U cycle (see Table 6.1) are described in further detail in (Delpech et al., 2009).

Note that for any MSR system, the minimum permissible temperature in the fuel salt circuit is determined not only by the fuel salt melting point, but also by the solubility of TRU trifluorides in the solvent. The maximum temperature of the fuel salt in MSR primary circuits, made of special Ni-based alloys, is mainly limited by thermo-mechanical and tellurium corrosion issues.

6.1.2 The MSFR breeder concept

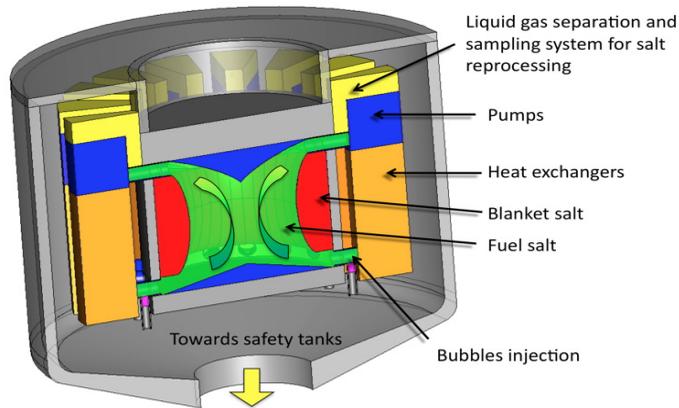
The MSFR is a homogeneous 3 GWt liquid salt fuel reactor concept. The current MSFR reference concept is illustrated in Figure 6.1. It should not be considered as a finalised nor optimised reactor but as a working example for interdisciplinary studies within the GIF/MSR. The calculated neutron spectrum for MSFR is shown in Figure 6.2. The fuel salt considered in the simulations is a molten binary fluoride salt with 77.5% of lithium fluoride; the other 22.5% are a mix of heavy nuclei fluorides. This proportion, set throughout the reactor operation, leads to a fast neutron spectrum in the core.

The total fuel salt volume in the fuel circuit of the MSFR is of approximately 18 m³, operated at a maximum fuel salt temperature of 750°C (Heuer et al., 2014; Serp et al., 2014). The total fuel salt volume is distributed half in the core and half in the external part of the fuel circuit. Fuel salt flows from the bottom to the top of the core cavity (note the absence of solid matter in core). After exiting the core, the fuel salt is fed into 16 groups of pumps and heat exchangers located around the core, travelling once around the fuel circuit in 3-4 seconds (Brovchenko, 2013). The MSFR concept is assessed as being efficient with ²³³U, LEU and/or (Pu+MA) as fissile seeds in the composition of its thorium-based fuel salt (see Section 6.3). A burner concept that does not use thorium in the fuel salt but may use it in a blanket salt can also be envisaged. In all cases, pyro-processing of the salt is required at rates that depend on the scenario studied (e.g. increasing number/size of nuclear reactors on a site, burning plutonium, burning MAs, etc.).

The main proposed safety features of the MSFR arise from the fuel being in liquid form, without any moderator or construction materials other than the salt components

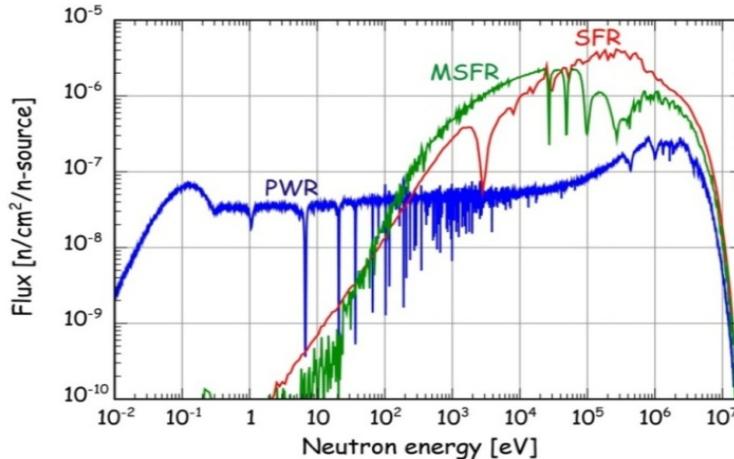
inside the core. The fuel circuit is connected to a salt draining system, which can be used for a planned shutdown or (in case of an incident/accident) leading to an excessive increase of the core temperature. In such situations, the fuel salt geometry can be passively reconfigured by gravity draining the fuel salt into tanks located under the reactor, where passive cooling and adequate reactivity margin can be obtained. The liquid fuel salt thermal dilation provides a thermal feedback coefficient of about -5 pcm/K, allowing power tuning by heat extraction. Because of a negative void feedback coefficient, draining of the liquid fuel salt in geometrically subcritical tanks facilitates long-term shutdown with passive cooling for decay heat removal.

Figure 6.1. The 3 GWt MSFR conceptual design



Fluoride-based fuel salt is in green, fertile blanket salt is red.

Figure 6.2. Calculated neutron spectrum of the reference MSFR (green curve)



For comparison, a typical sodium-cooled fast neutron reactor spectrum (SFR, in red) and a typical PWR thermal spectrum (in blue) are given.

6.1.3 The fast spectrum MOSART concept

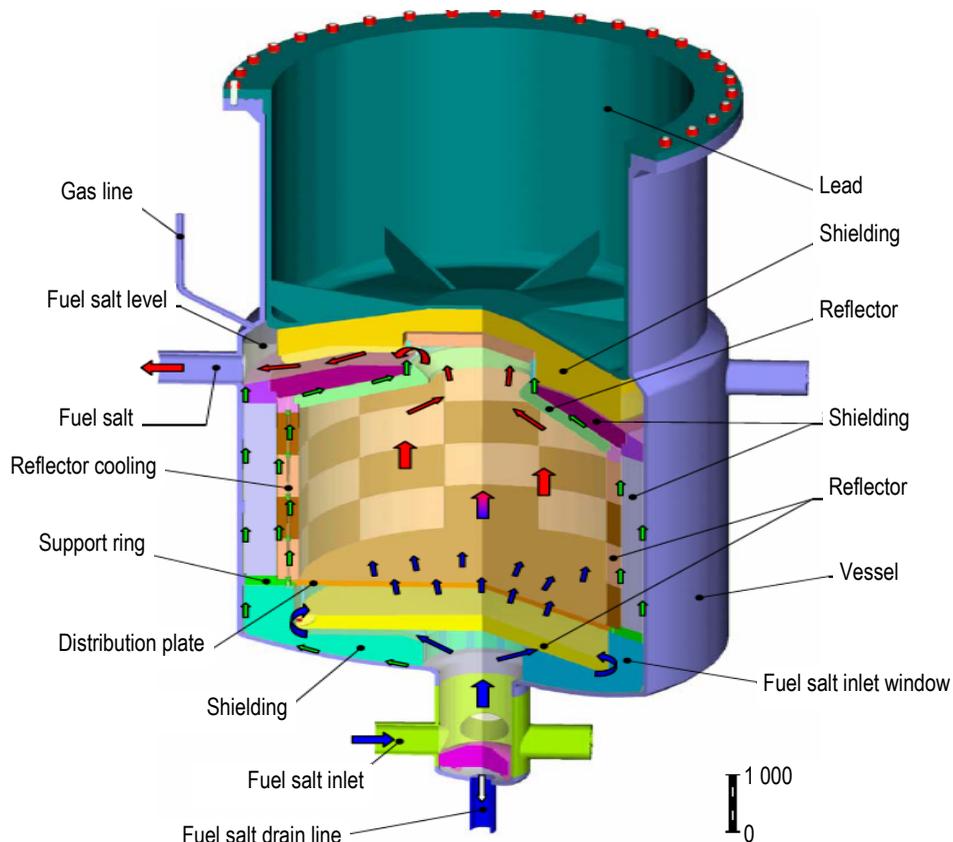
The primary requirements for the MOSART concept are to allow for an efficient transmutation and recycling of TRUs from UOX or MOX PWR spent fuel in an MSR system. MOSART has been designed so that the necessary fissile concentration in the fuel salt and the geometry allows for a 2.4 GW thermal power system. Figure 6.3 shows the

preliminary concept design used for the feasibility assessment and the evaluation of the neutronic and thermal hydraulic characteristics.

As starting load and composition evolution for MOSART, the following compositions of TRU from used LWR fuel were investigated:

- UOX spent fuel after one year of cooling with MAS to TRU ratio MA/TRU ≈ 0.1 (^{238}Pu -3.18%; ^{239}Pu -43.93%; ^{240}Pu -21.27%; ^{241}Pu -13.52%; ^{242}Pu -7.88%; ^{237}Np -6.42%; ^{241}Am -0.55%; ^{243}Am -2.33%).
- MOX spent fuel after one year of cooling with MA/TRU ratio ≈ 0.2 . (^{238}Pu -2.77%; ^{239}Pu -48.36%; ^{240}Pu -19.97%; ^{241}Pu -8.30%; ^{242}Pu -6.25%; ^{237}Np -6.51%; ^{241}Am -5.56%; ^{243}Am -1.69%).

Figure 6.3. The 2.4 GWt MOSART reference core without graphite moderator is a cylinder 3.4 m in diameter and 3.6 m in height



In MOSART, the fuel salt inlet and outlet pipes are assumed to be 1 m in diameter. Radial, bottom and top reflectors are attached to the reactor vessel. This leaves an annulus filled with fuel salt surrounding the core to cool reflector and reactor vessel. The molten salt flow rate is 10 000 kg/s. At nominal conditions, the fuel salt enters the core at 600°C and transports 2.4 GWt to the secondary salt in the primary heat exchanger. The fluoride fuel salt mixture is circulated through the reactor core by four pumps. Pumps circulate salt through heat exchangers and return it to a common plenum at the bottom of the reactor vessel. The total fuel salt volume in the fuel circuit for the reference MOSART design is of around 50 m³, with around 18 m³ of this total being in the external part of the reactor vessel.

The container material for the fuel circuit is a special Ni-Mo alloy, HN80MTY, with 1% in mass of Al, developed in Russia. It does not undergo tellurium intergranular cracking in the LiF-BeF₂-ThF₄ fuel salt with addition up to 2 mole % UF₄ at [U(IV)]/[U(III)] ratios below 100. HN80MTY alloy can be recommended for further consideration as the main container material for the fuel circuit with operating temperature up to 750°C required for MOSART and MSFR designs.

The MOSART concept is being studied in different configurations, which consider different core dimensions and different compositions of the fuel salt and/or salt blanket that allow for different modes of utilisation. A detailed description of MOSART can be found in Ignatiev (2014). Of particular significance are the applications of MOSART that consider the use of thorium; in particular, the use of the MOSART concept as a transmuting system cycle initially fed with TRU loading from LWR SNF for producing the necessary fissile ²³³U quantities for an uranium-thorium fuel cycle.

MOSART converter mode (two fluid system with thorium salt blanket)

Surrounding MOSART's core by a thorium-containing salt blanket with molar composition 75LiF-5BeF₂-20ThF₄ or 78LiF₂-22ThF₄ (50-60 cm thickness) makes it possible for MOSART to be used in a converter mode by recovering ²³³U (80-100 kg/year) from the blanket through a volatility process and recycling it into the fuel stream of this two-fluid system, while returning the thorium-bearing salt to the blanket. The use of ²³³U in the fuel facilitates a decrease in the equilibrium TRU concentration and thus expands the range of possible fuel compositions to MA-bearing ones. A two-fluid 2.4 GWt MOSART design can operate in such a converter mode and transform approximately 700 kg of TRUs from LWR SNF to 100 kg of ²³³U per year. The total conversion ratio of the converter system and the burning efficiency of the transmuter can be essentially increased by forcing the specific power of the core; i.e. by reducing the core dimensions.

It is possible to significantly increase the rate of ²³³U production from the thorium blanket by further decreasing the core's radius. However, the limiting factors are:

- solubility of TRU trifluorides in the fuel salt;
- heat removal from primary circuit in the heat exchanger;
- lifetime of structural materials (in particular the dividing wall between the core and the blanket, reflector, distribution plate, etc.).

The optimal result from the point of maximum ²³³U production reached is of approximately 300 kg/year in a modified 2.4 GWt MOSART core using (75LiF-5BeF₂-20ThF₄). This requires the replacement of the dividing wall between the core and the inner blanket around every six years, due to metallic material resistance to neutron fluxes at high temperatures.

MOSART self-sustainable mode

The use of MOSART core with reduced dimensions shows potential for reaching a conversion ratio of unity with a strategy of gradual increase of thorium concentration in the fuel salt. Single fluid 2.4 GWt Li,Be/F MOSART core (radius: 1.4 m, height: 2.8 m) containing as initial loading 2 mole % of ThF₄ and 1.2 mole % of TRUF₃ with a rare earth removal time 1 equivalent full power years after 12 years can (in principle) operate without TRUF₃ addition and continue operating only on thorium support (see Figure 6.4).

The maximum concentration of TRUF₃ during this transition does not exceed 1.7 mole %. At equilibrium, the molar fraction of ThF₄ in the fuel salt is near 6 mole % and is sufficient to provide the system with CR=1 for up to 50 years of reactor operation. The reactivity temperature coefficient of the homogeneous core is negative; its value on equilibrium is 6.7 pcm/K. Use of a thorium blanket facilitates a reduction in the transition time to self-sustainable mode to approximately four equivalent full power years; this makes the system more complicated from technical point of view, however.

Figure 6.4. Transition to equilibrium of ThF_4 (1), TRUF_3 (2), UF_4 (3) in single fluid 2.4 GWt MOSART with Li,Be,Th/F core (CR=1 gradual increase of thorium)

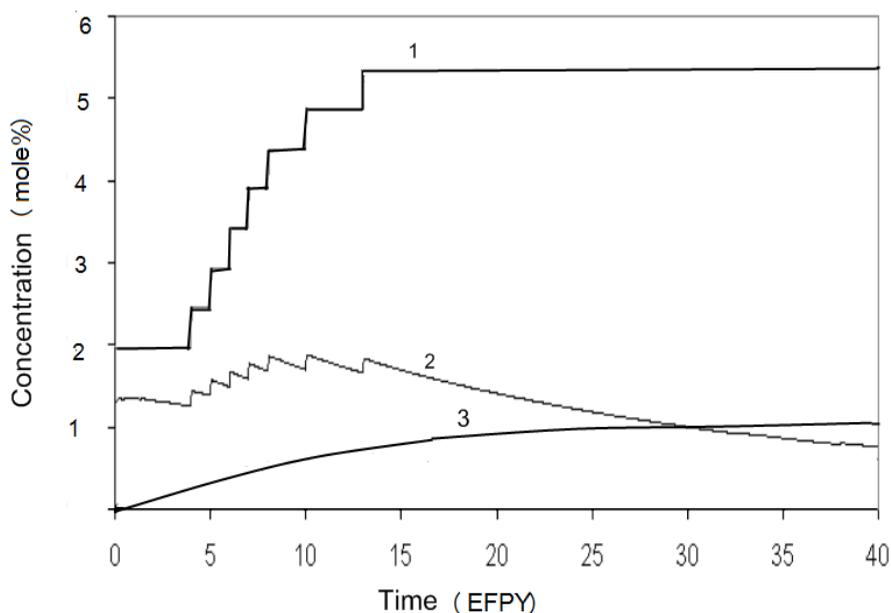


Table 6.1. Main characteristics of the MOSART and MSFR designs studied within GIF/MSR

Fuel circuit	MOSART	MSFR
Fuel salt, mole %	LiF-BeF ₂ +1TRUF ₃ LiF-BeF ₂ +5ThF ₄ +1UF ₄	78.6LiF-12.9ThF ₄ -3.5UF ₄ -5TRUF ₃ 77.5LiF-6.6ThF ₄ -12.3UF ₄ -3.6TRUF ₃
Temperature, °C	620-720	650-750
Core radius/height, m	1.4/2.8	1.13/2.26
Core specific power, W/cm ³	130	270
Container material	Ni-Mo alloy HN80MTY	Ni-W alloy EM 721
Removal time for soluble fission products, years	1-3	1-3

6.2 MSFR fuel salt processing

The reprocessing times required and methods foreseen for fission product removal and actinide recycling for MSFR and Th-U MOSART systems are similar and summarised in Table 6.2.

The fuel salt undergoes two types of treatment: online by neutral gas bubbling in the fuel circuit and remote mini-batch on-site reprocessing. The purpose of these fuel salt treatments is to recycle actinides and remove most of the fission products without reactor shutdown and consequently requiring a small fissile inventory outside the core as compared to current LWRs.

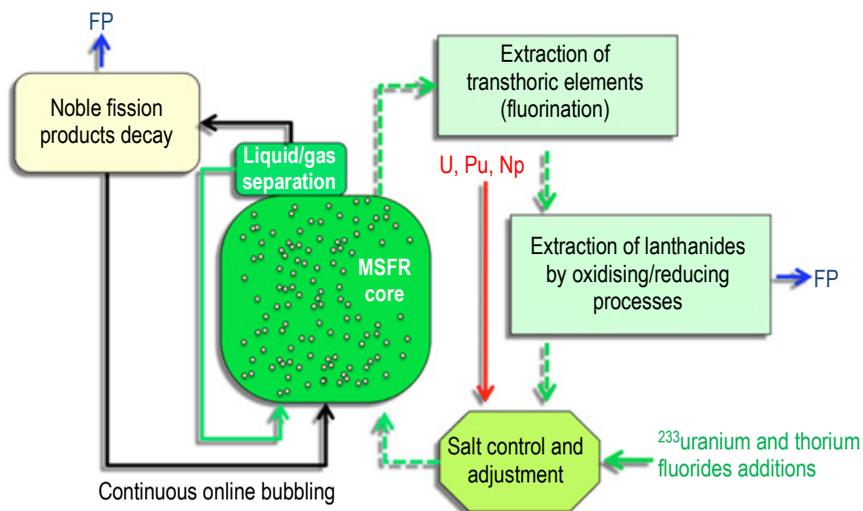
Table 6.2. Fuel salt processing time needed and methods used for fission product removal and actinides recycling in MSFR and Th-U MOSART

Element	Time	Method
Kr, Xe	50 seconds	Sparging with He
Zn, Ga, Ge, As, Se, Nb, Mo, Ru, Rh, Pd, Ag, Tc, Cd, In, Sn, Sb, Te	2-4 hours	Partial plating on surfaces, removal to off-gas system, limited filtering
U	10-15 days	Fluorination
Zr, ²³³ Pa	1-3 years	Reductive extraction
Ni, Fe, Cr	1-3 years	
Pu, Am, Cm, Np	1-3 years	
Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er	1-3 years	
Sm, Eu	1-3 years	
Sr, Ba, Rb, Cs	5-10 years	

The fuel salt treatment is schematically presented in Figure 6.5. It consists of two main streams. The first one is a continuous gas bubbling system in the fuel circuit, extracting noble gases and part of the noble metals (fission products) present in the fuel salt. The gaseous stream is sent to a delay storage where most of the Kr and Xe decay into Rb and Cs, preventing their accumulation in the fuel salt. The remaining gas is then recycled.

The second stream is a semi-continuous batch pyro-chemical processing unit, with a fuel salt removal rate of about ten litres per day, in order to recycle actinides and limit the lanthanide and Zr concentration in the core. The sampled salt returns to the reactor after purification and after addition of ²³³U and Th, as required to adjust the fuel composition in fissile or fertile nuclides. This is also an opportunity to tune the redox potential of the fuel salt by controlling the U⁴⁺ to U³⁺ ratio.

Figure 6.5. Schematic presentation of the fuel salt treatment with two main streams



The effect of the batch pyro processing rate is shown in Figure 6.6. It should be noted that, with the reactor configuration used for the calculation (under-breeding core), a regeneration rate is reached for the processing of a full load in 4 000 days (Heuer et al., 2014).

6.3 Breeder MSFR starting modes

To deploy thorium fuel cycles in breeder MSFRs, the following exercise scenarios have been studied (Heuer et al., 2014):

- MSFR directly started with ^{233}U as initial fissile material, assuming that this ^{233}U has been produced already (a major assumption), for example in fertile blankets of other reactors (LWRs or FRs).
- Using the plutonium or using the mix of TRU elements produced in used fuel of generation II or III reactors.
- Starting MSFRs with LEU (enriched up to 20%) as initial fissile material.

Figures 6.7 and 6.8 present comparisons of fuel composition evolutions of a “3-GWt reference MSFR” reactor started with ^{233}U , TRU, MOX-Th or enriched ^{235}U and TRU. Figure 6.9 shows production of ^{233}U for different starting modes of a breeder MSFR.

An exercise deployment scenario of MSFRs for the French case is given in Annex C.

Figure 6.6. Effect of the batch processing rate on the breeding ratio in the core and in the whole MSFR system (core and fertile blanket)

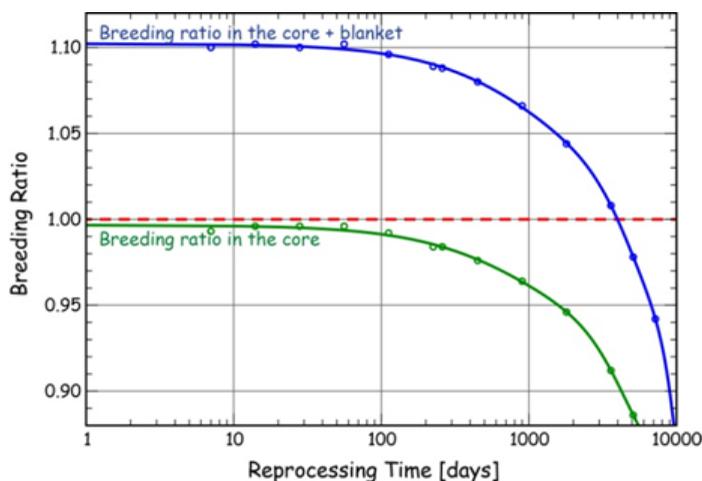
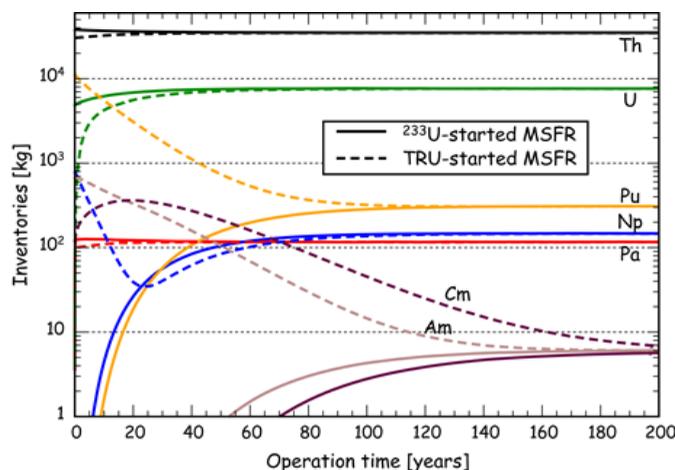


Figure 6.7. Time evolution up to equilibrium of the heavy nuclei inventory for the ^{233}U -started MSFR (solid lines) and for the TRU-started MSFR (dashed lines)



Operation time is given in equivalent full power years.

Figure 6.8. Time evolution up to equilibrium of the heavy nuclei inventory for the optimised MSFR configuration started with enriched uranium and TRU elements

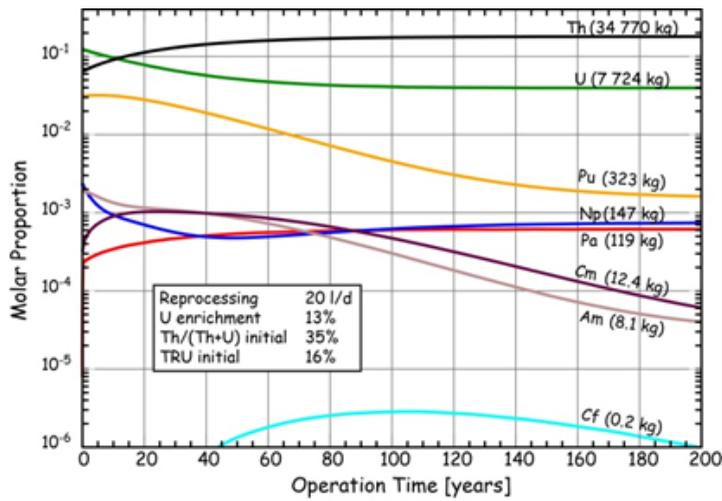
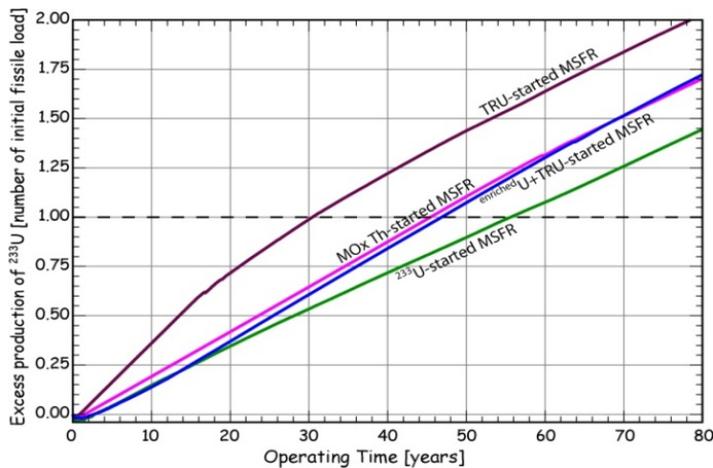


Figure 6.9. Excess production of ²³³U for the different starting modes of the MSFR during reactor operation in number of initial fissile loads produced



6.4 Safety approach and risk analysis for MSFRs

The MSFR is at its early stages of conceptual development and aims at adopting an inherent safety-by-design approach. However, the unique characteristics of liquid-fuelled MSRs warrant a full revision of their design and safety analyses; for instance:

- The safety principle of defence-in-depth and multiple barriers must be readapted, since conventional barriers (such as clad, primary circuit and containment in LWRs) are no longer applicable.
- Diversity and independence of MSFR deterministic reactivity control mechanisms must be demonstrated (no control or shutdown rods, no burnable poisons so far in the present concepts).
- New safety criteria must be developed to evaluate reactor response during normal, incidental and accidental conditions, given the liquid form of the MSFR fuel, which would not be an acceptable situation in LWRs.

- Evaluation of severe accident scenarios necessitates detailed investigation of the interactions between the fuel salt and groundwater and on the determination of the source term.
- Evaluation of the risk posed by the residual decay heat and the radioactive inventory existing in the reprocessing unit is also necessary.

A novel methodology for the design and safety evaluation of the MSFR is required. As is expected, any MSFR safety methodology must rely on current accepted safety principles, such as the principle of the defence-in-depth, the use of multiple barriers and the three basic safety functions, which are: reactivity control, fuel cooling and radioactive products confinement. In addition, due to the limited amount of operational experience and some of its novel features, any new methodology will need to be proven as robust and comprehensive and integrate both deterministic and probabilistic approaches. In order to fulfil these objectives, a MSFR design and safety analysis methodology is currently being developed according to the following steps:

- systemic modelling of all reactor components by using a model-based risk analysis tool;
- identification the safety functions, to be identified from the components functional criteria;
- identification of reactor abnormal events (failure modes and dangerous phenomena);
- risk evaluation: evaluation of the probability of occurrence and consequences.

The consideration of the Integrated Safety Analysis Methodology (ISAM) developed by the GIF Risk and Safety Working Group (RSWG) has already identified strengths and limitations of the MSFR concept. Calculation tools presently available do not allow for the same level of analysis for all of the accidents considered. A first qualitative and quantitative analysis has led to a better understanding of MSFR safety characteristics at this very preliminary stage. The next steps for this safety evaluation will take place under the framework of the Horizon 2020 EC project SAMOFAR (Safety Assessment of Molten Salt Fast Reactors), which will start in the second half of 2015.

6.5 Limiting factors of fast spectrum MSRs

Several limiting factors have been identified in the development of the MSFR reactor concept at the pre-conceptual stage. Among these factors the following are highlighted:

- Material resistance to high temperatures. A first temperature limit is given by the fuel salt melting point (565°C) to which a safety margin should be added to avoid local solidification (50°C for instance) and an additional 100 to 150°C for carrying the heat to heat exchangers without excessive pressure drop. This leads to temperatures of about 750°C at the outlet of the core, where the fuel enters the gas-salt separation device and the pump. Those devices may be maintained at 700°C by cooling; i.e. the same temperature at which the heat exchanger plates are during the heat transfer with an intermediate coolant salt at about 650°C. Based on current understanding, it appears that known alloys can withstand this temperature for extended periods of time; but this could be limited, as with solid fuel cladding, unless regular replacement of the material is performed.
- Material resistance to neutron fluxes at high temperatures. Unless operation at low power density is chosen, preliminary calculations of the maximum displacement per atom (dpa) of core walls lead to 7.5 dpa/year for a power density of 330 W/cc. This is less than expected for other solid fuel fast reactors because of the neutron spectrum change due to inelastic scattering on fluorine nuclei, and by the absence of solid material in the core.

6.6 Summary

Within the GIF/MSR, experimental research on basic data has been supported by Euratom and Rosatom to confirm the theoretical advantages of the MSFR and MOSART concepts. No major impediments have at this stage been identified; but almost all of the associated technology remains to be tested, and demonstration experiments have yet to be conducted to assess the feasibility and potential advantages of fast-spectrum MSR based on the thorium fuel cycle.

The strong international co-operation between the countries and organisations involved within GIF/MSR (China, Euratom, France, Korea, Japan, Russia, and the United States) has shown to be effective in tackling prominent R&D issues on MSR design. However, a continued effort is required by all parties to converge on a basic reactor design around 2020.

The GIF/MSR system has extended the viability study phase of fast spectrum MSR concepts at least until 2025. After this viability phase, a performance study phase is expected post-2025, where the decision to support further development of MSR systems within GIF will need to be supported by adequate data. For innovative reactors such as liquid-fuelled MSRs, an approved licensing and regulation basis, yet to be developed, is required to support their technical feasibility and commercial viability.

References

- Brovchenko, M., et al. (2013), "Design related studies for the preliminary safety assessment of the molten salt fast reactor", *Nuclear Science and Engineering*, Vol. 175, pp. 329-339.
- Delpech S., et al. (2009), "Reactor physics and reprocessing scheme for innovative molten salt reactor system", *Journal of Fluorine Chemistry*, Vol. 130, Issue 1, pp. 11-17.
- GIF (2014), *Technological Roadmap Update*, Generation IV International Forum, Paris.
- Heuer, D., et al. (2014), "Towards the thorium fuel cycle with molten salt fast reactors", *Annals of Nuclear Energy*, Vol. 64, pp. 421-429.
- Ignatiev V., et al. (2014), "Molten salt actinide recycler and transforming system without and with Th-U support: Fuel cycle flexibility and key materials properties", *Annals of Nuclear Energy*, Vol. 64: pp. 408-420.
- Mathieu L., et al. (2009), "Possible configurations for the thorium molten salt reactor and advantages of the fast non-moderated version", *Nuclear Science and Engineering*, Vol. 161, pp. 78-89.
- Merle-Lucotte, E., et al. (2013), "Recommendations for a demonstrator of molten salt fast reactor", *Actes de l'International Conference on Fast Reactors and Related Fuel Cycles: Safe Technologies and Sustainable Scenarios (FR13)*, Paris.
- Serp, J., et al. (2014), "The molten salt reactor (MSR) in generation IV: Overview and perspectives", *Progress in Nuclear Energy*, pp. 1-12.

7. Spent fuel reprocessing

Scenarios using once-through processes will have a limited effect on the overall long-term utilisation of fissile material. The real benefit of thorium's fertile potential will be achieved by implementing a fully closed fuel cycle, in other words with reprocessing of irradiated fuel and recycling of ^{233}U .

R&D experiments show that the treatment of thorium-based fuels is technically more complex than that for uranium-based fuels, mainly due to the complexity of the dissolution step. Thorium is much more difficult to dissolve than uranium, either in the form of metal or oxide. Historically, ORNL in the United States developed a hydrometallurgical process called THOREX for thorium-based fuel reprocessing, derived from the plutonium-uranium redox extraction (PUREX) process currently used for the reprocessing of uranium-based fuels.

This section provides an overview of the current state of knowledge on the treatment of thorium oxide fuel in the thorium cycle and exemplified by the process envisaged in India as well as the THOREX liquid-liquid extraction process. Two types of thorium-based fuels will be discussed ($\text{Th},^{233}\text{U}\text{O}_2$ and $(\text{Th},\text{Pu})\text{O}_2$, though $(\text{Th},\text{Pu},^{233}\text{U})\text{O}_2$ would also be possible.

7.1 THOREX process

THOREX is a liquid-liquid extraction process similar to the PUREX process; it was first described in 1955 by A.T. Gresky (Gresky, 1956) of ORNL. Two variants, "Interim-23" (Flanary et al., 1964) and "Acidic THOREX" (Gresky, 1956), were the most widely implemented processes at laboratory or pilot scale in the United States, in France and in India (Merz, 1982; Lung, 1997; IAEA, 2000; Srinivasan et al., 1973) (see Table 7.1).

Table 7.1. Summary of spent thorium (metal and oxide) fuel reprocessing campaigns

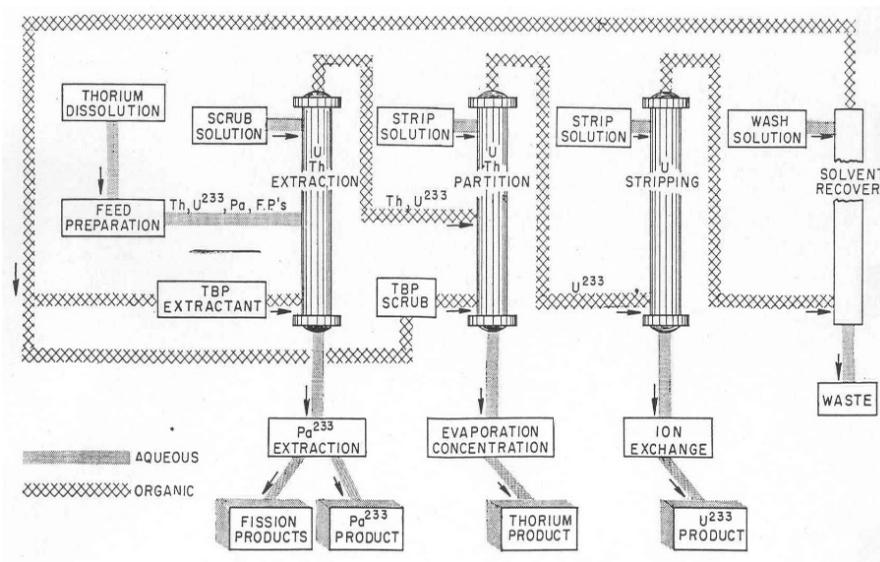
Site	Years	Th (t)	^{233}U (kg)	^{232}U (ppm)	Flowsheet
Oak Ridge National Laboratory, United States	1954 and 1958	5	8	10-40	Interim 23
	1955 to 1958	30	47	10-40	THOREX
	Total	35	55		
Savannah River Plant, United States	1964 to 1965	14	107	225	Interim 23
	1965	9	19	38	Interim 23
	1966 and 1968	193	412	6-9	THOREX
	Total	216	538		
Hanford, United States	1965	4	–	–	Interim 23
	1966	250	270	6-10	Acid THOREX
	1970	400	589	6-10	Acid THOREX
	Total	654	859		
Nuclear Fuel Services, United States	1969	17	103*	125	Interim 23
Atelier Pilote de Marcoule, France	1962 to 1969	2.8	>2	–	Interim 23
Bhabha Atomic Research Centre and Indira Gandhi Centre for Atomic Research, India	>1970	–	–	<1 200	Interim 23

* Mixture of ^{233}U - ^{235}U ($^{233}\text{U} + ^{235}\text{U} = 1\ 019\ \text{kg}$), Th added as salting-out agent (15.8 GWd/t).

7.1.1 THOREX process flowsheet

The THOREX process flowsheet is shown in Figure 7.1. It is similar to the PUREX process and uses the same liquid-liquid extraction solvent: tributyl phosphate (TBP) diluted in *n*-dodecane (30-42 vol%). As thorium oxide is insoluble in nitric acid, the dissolution step requires a very high nitric acid concentration (13-15 M) and the addition of hydrofluoric acid. The acidity of the solution resulting from dissolution step is thus about 8-9 M nitric. To improve the decontamination factors, the feed solution for the extraction cycles must be acid-deficient, with a deficiency of about 0.15 M. The dissolution solution must be adjusted by dilution and/or steam stripping, a delicate operation because the final acidity must be well controlled. As the thorium partition coefficients are too low at these weak acidities, aluminium nitrate is added as a salting-out agent.

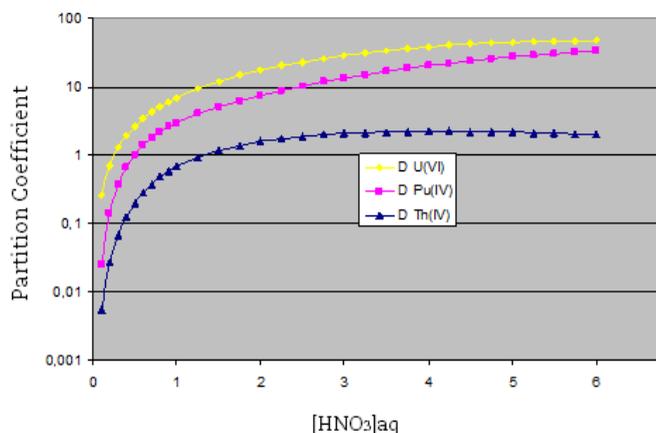
Figure 7.1. THOREX process flowsheet



Source: IAEA (2005).

Finally, since the thorium oxidation state cannot be modified, U-Th separation is based only on the difference in the affinity (separation factor ~10) of these two elements for TBP (see Figure 7.2.).

Figure 7.2. Partition coefficients for U(VI), Pu(IV) and Th(IV), 30% TBP at 25°C (PAREX)

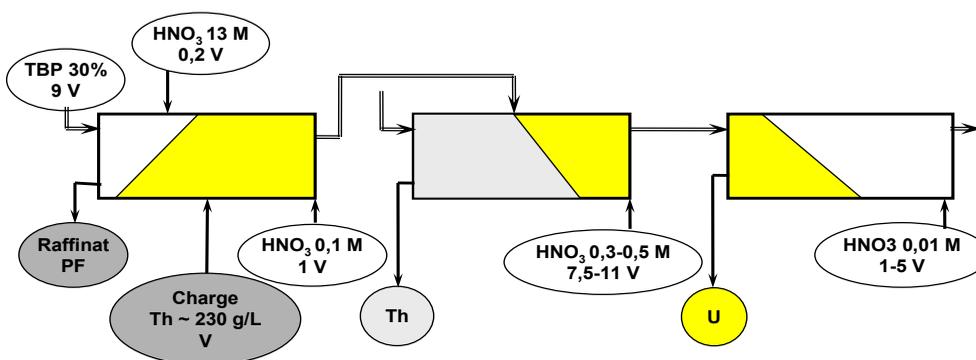


Source: Montuir et al.(2011).

7.1.2 The acid THOREX process

The only difference between the acid THOREX process (Rainey and Moore, 1962) and the original THOREX process is that the salting-out nitrates (aluminium nitrate) in the extraction zone are replaced by nitric acid to reduce the salt concentration in the raffinate without significantly affecting the decontamination factors with respect to the fission products according to Rainey and Moore (1962). The flowsheet for this variant (as implemented at Hanford) is shown in Figure 7.3.

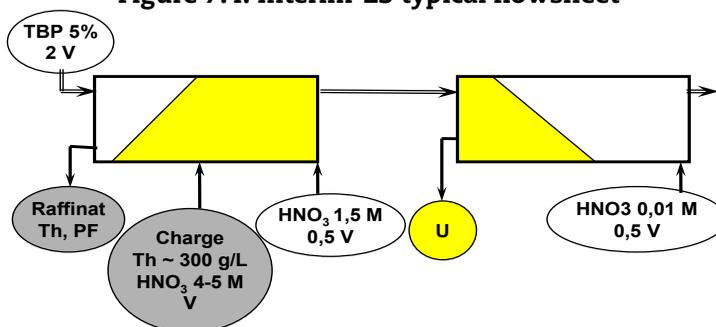
Figure 7.3. Acid THOREX flowsheet implemented at Hanford



7.1.3 The Interim-23 process

The aim of the Interim-23 process (Flanary et al., 1964) is to separate only uranium, while leaving the thorium in the extraction raffinates with the fission products. Selective extraction of uranium uses the same extractant molecule, TBP, but at an appreciably lower concentration: 3-5 vol%. The lower TBP concentration not only slightly improves the separation factor, but especially limits the extraction of thorium by saturating the solvent with uranium, which also improves the decontamination factors with respect to the fission products. A typical flowsheet for this variant is shown in Figure 7.4.

Figure 7.4. Interim-23 typical flowsheet

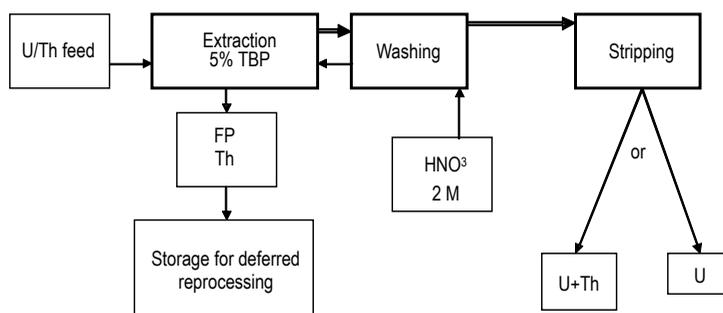


A total of ~2 metric tonnes of ^{233}U were processed in the ORNL Radiochemical Pilot Plant during the period 1943-1988. Most of this was processed using the Interim-23 process, using 5% di-secondary-butyl phenyl phosphonate in diethylbenzene as the uranium extractant. Another ~1 metric tonne of uranium contained in the initial core ($\text{Th}/^{235}\text{U}$) from the Consolidated Edison Indian Point-1 reactor was processed using the Interim-23 process at the West Valley reprocessing plant in 1968, and the mixed $^{235}\text{-}^{233}\text{U}$ product solution was transported to ORNL for solidification and eventual disposal. The Interim-23 process uses a high acidity feed solution which is more compatible with dissolver solution and an acid-deficient scrub solution to enable high decontamination factors from fission product elements.

A total of 2.8 metric tonnes of thorium metal were reprocessed in *Atelier Pilote de Marcoule* in France; the fuel was irradiated in the G1 and G2 reactors at Marcoule with a burn-up of 335 and 900 MWd/t and cooled for about seven years. In order to obtain the desired decontamination factor relative to thorium, a second purification cycle was performed with 30% TBP after inter-cycle uranium concentration. The overall decontamination factor was better than 6×10^7 , comparable to values typically observed in the PUREX process.

The flowsheet often selected in India (Srinivasan et al., 1973) (see Figure 7.5) is based on the Interim-23 process, with the solvent flow rate adjusted depending on the objective (with or without co-conversion of thorium with uranium) to co-extract a fraction of thorium with the uranium. The option of deferred thorium processing after uranium separation is justified by the abundance of thorium in India, but also by the presence of ^{228}Th (half-life 1.9 years), a daughter product of ^{232}U (always found with ^{233}U), one of whose decay products is ^{208}Tl (a 2.6 MeV gamma emitter). Deferred recycling (>20 years) would simplify the reprocessing operations by avoiding the need for heavy biological shielding to recycle the thorium, especially in view of fabricating impregnated low-density ThO_2 pellets. However, recycle of the ^{233}U will always require remotely operated, shielded facilities because of the presence of ^{232}U daughters.

Figure 7.5. Flowsheet adopted in India (Interim-23)



Source: Srinivasan et al. (1973).

7.2 Problem areas and prospects of the THOREX process

7.2.1 Dissolution

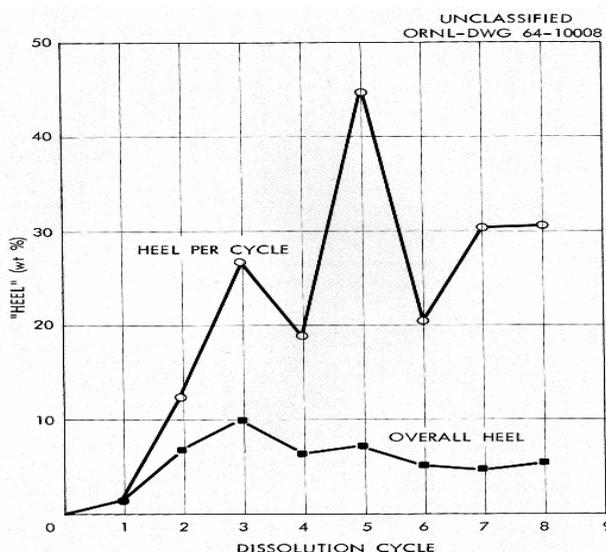
Thoria (ThO_2), like PuO_2 , is practically inert with respect to nitric acid. Although the plutonium oxidation state can be modified to accelerate the dissolution of PuO_2 (e.g. Ag(II)-mediated electrolytic dissolution), this is not possible for thorium. The dissolution of thorium oxide (or thorium metal) fuel requires a mixture of nitric acid and hydrofluoric acid, causing equipment corrosion problems. To limit corrosion, the aggressiveness of the mixture is buffered by adding aluminium nitrate. The dissolving solution typically chosen for this fuel is 13-15 M HNO_3 , 0.03-0.1 M HF, ~ 0.1 M $\text{Al}(\text{NO}_3)_3$, for a final thorium concentration of 400 g/L. Despite the aggressiveness of the mixture, the dissolution kinetics is relatively slow compared with UOX fuel. At the boiling point, the dissolution time is around 24 h (sometimes 30-40 h) for virtually complete (>99%) dissolution. In addition, un-dissolved “blue thoria” is often observed and is capable of sequestering fissile ^{233}U . The dissolution kinetics varies with the fabrication process, as shown by Goode and Flanary (ORNL report, 1965). This is practically the only study found concerning variable burn-up ranging up to very high values (98 000 MWd/t). In this study, the dissolution kinetics of ThO_2 fuel with 4-4.5% UO_2 (enriched to 95% ^{235}U) prepared according to three fabrication processes were compared (see Table 7.2). The dissolving solution for this fuel was 13 M HNO_3 , 0.05 M NaF and the desired Th concentration 0.5 M to 1 M (for a final acidity of 9-10 M).

Table 7.2. Fuel samples used in the study

Type of oxide	Burn-up (MWd/t)	Fabrication process	Density (% theoretical)
Arc-fused	3 000	Vibro-compaction	83-86
Sol-gel	Un-irradiated	Vibro-compaction	83-86
Sol-gel	4 000	Vibro-compaction	83-86
Sol-gel	13 000	Vibro-compaction	86-87
Sol-gel	14 000	Vibro-compaction	86-87
Sol-gel	16 000	Vibro-compaction	86-87
Sol-gel	17 000	Vibro-compaction	86-87
Sintered	50 000	Pelletisation	95
Sintered	77 000	Pelletisation	95
Sintered	98 000	Pelletisation	95

Source: Goode and Flanary (1965).

For the vibro-compacted samples, 77% were dissolved after 1 h, 84% after 2 h, 96% after 6 h and 99.8% after 24 h. As expected, the dissolution of sintered pellets was slower (especially at the beginning of the reaction due to the smaller attack surface area), but complete dissolution (99.9%) was nevertheless obtained in 24 h. In this study, the burn-up appeared to have little impact on the dissolution kinetics, unlike the observations for UOX or MOX fuel. By comparison, total dissolution of the un-irradiated sample required nearly 75 h. To shorten the dissolution time to 5 h while maintaining near 90% dissolution, the authors recommended cyclic dissolution in which the insolubles are recovered after each step and reintroduced in the following dissolution step. Figure 7.6 shows the results of a test on irradiated, vibro-compacted sol-gel samples. After eight dissolution cycles, the total residue was well below 10% (~6-7%) of the total fuel mass. Although this dissolution procedure is attractive because of the shorter dissolution time, it requires prior mechanical or chemical separation of the oxide from the cladding (the SULFEX process could be used and the cladding can be dissolved in boiling 6 M sulphuric acid) and theoretically leads to an accumulation of residues that must be recycled (some of which are probably completely insoluble) although no such accumulation was observed during this test.

Figure 7.6. Cyclic dissolution test results

Source: Goode and Flanary (1965).

A more recent Indian study (Vijayan et al., 2000) on un-irradiated sintered pellets showed the effect of doping the fuel with MgO (which also allowed the use of a lower sintering temperature). A reduction in the dissolution time by a factor of ten was observed with 1.5% MgO (optimum value in the 0.5-2.5% range). Moreover, in the context of recycling fabrication scrap, a second Indian study (Mallik, Malav and Kamath, 2000) in a pressurised reaction vessel with microwave heating showed a beneficial effect of pressure on the dissolution time (15.5 M HNO₃, 0.05 M NaF):

10-12 h	at	90 psi (6.2×10^5 Pa)
12.5-13.5 h	at	60 psi (4.14×10^5 Pa)
13-13.5 h	at	40 psi (2.76×10^5 Pa)
>20 h	at	20 psi (1.38×10^5 Pa)

The dissolution of thorium-based fuel is thus difficult to implement at an industrial scale due to the extended dissolution times and the necessary presence of hydrofluoric acid, which must be buffered with aluminium nitrate to mitigate equipment corrosion. The influence of the burn-up and the impact of the conversion/fabrication process and microstructure in particular, will require further studies to control and optimise the industrial implementation of this operation.

Another important point that must be considered for the dissolution, as well as for all steps of the process, is the necessity to control the vapour transfer of ²⁰⁸Tl, a decay daughter of ²²⁰Rn (~1 minute half-life), which is a gas in the decay chain from ²³²U and ²²⁸Th. The ²⁰⁸Tl has a 2.6 MeV gamma emission and is formed in the radon gas phase as very finely divided, highly radioactive particles. Therefore, the off-gas system design must include a ten-minute hold-up for ²²⁰Rn decay prior to entering high-efficiency particulate air filters so that gas-phase transport of the highly radioactive decay daughters of ²²⁰Rn can be prevented.

7.2.2 Separation by liquid-liquid extraction

The main difficulties in the treatment of Th/U fuel by extraction with TBP are:

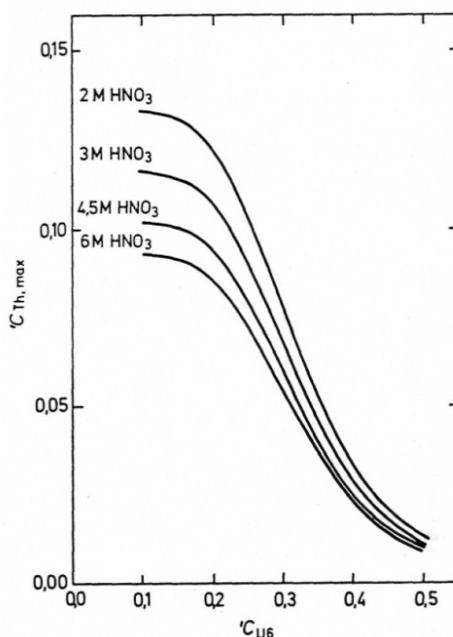
- the risk of formation of a third phase due to the low solubility of the Th(IV)-TBP complex in aliphatic solvents;
- the formation of di-butyl phosphoric acid (HDBP), a product of TBP degradation by radiolysis and catalysed hydrolysis in the presence of macroscopic thorium concentrations with the formation of strong Th(IV)-DBP complexes.

7.2.3 Risk of third phase formation

To avoid the risk of forming a third phase (Nakashima and Kolarik, 1983), the Th(IV) concentration in 30% TBP + n-dodecane must remain below 35 g/L (<0.15 M).

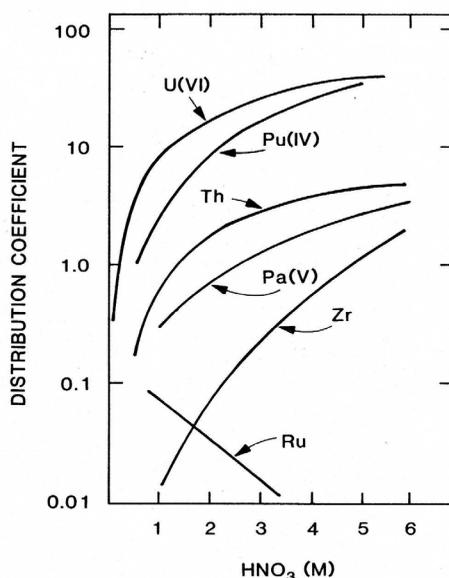
Thorium is the major element in the fuel; it is therefore impossible, as in the PUREX process, to benefit from a solvent saturation effect to improve the decontamination factors, especially with respect to Pa, Zr and Ru (Figure 7.8 [Schulz, 1989]). This limitation led the designers of the THOREX process to reduce the acidity of the feed in order to form less-extractable hydrolysed species of these fission products (Gresky, 1956). Adjusting the feed acidity implies delicate operations with the formation of significant amounts of precipitates compared with the PUREX process and would require a clarification step prior to the enhanced extraction operations.

Figure 7.7. Limit of third-phase formation Th(IV)org vs. U(VI)org, 30% TBP + n-dodecane at 25°C



Source: Nakashima and Kolarik (1983).

Figure 7.8. Partition coefficients of Th(IV), U(VI), Pu(IV), Pa(V), Zr(IV) and Ru(III), 30% TBP



Source: Schulz (1989).

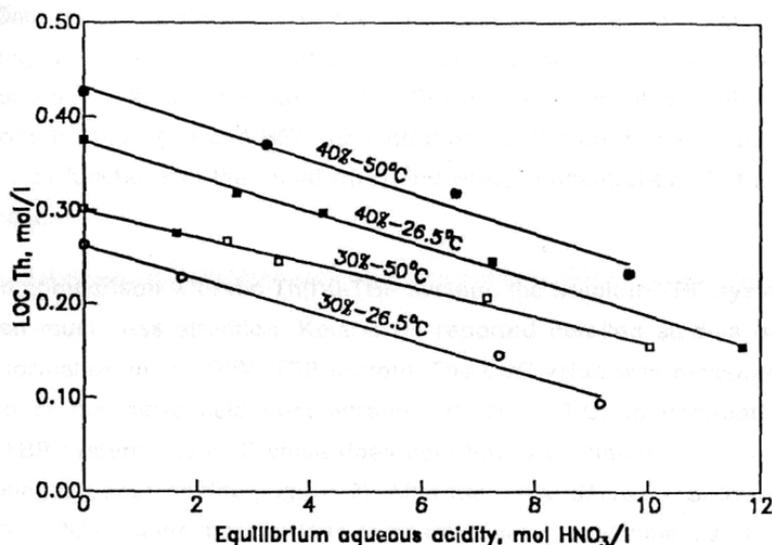
The third-phase formation limit can be pushed back (but not eliminated) by the use of a more branched diluent, such as Amsco, HTP or odourless kerosene, the addition of modifiers (heavy alcohols, benzene, toluene, etc.), or the application of higher temperatures (see Figure 7.9 [Rao and Kolarik, 1996]). Although raising the temperature might be tempting because of its simplicity, the drawback this option is that it increases the TBP degradation kinetics leading to the formation of HDBP (dibutyl phosphate) with a negative impact on the process as described below.

Replacing TBP by a tri-alkyl phosphate with longer alkyl chains such as tri-amyl phosphate (TAP), tri-isoamyl phosphate (TiAP) or tris(2-ethylhexyl) phosphate (T2EHP) provides substantial gains with improved U/Th separation factors for the most branched extractant molecules. TiAP, which belongs to the same family as TBP, was investigated and used in Russia to replace TBP in the Pu cycle of the PUREX process. Similarly, replacing the aliphatic diluent by an aromatic (polar) diluent such as Decalin (decahydronaphthalene) would eliminate this constraint.

It must be highlighted that:

- As only uranium is extracted in the Interim-23 process, the constraint due to the third-phase formation limit is practically non-existent.
- When reprocessing (Th,U,Pu)O₂ fuel, since the partition coefficients for Pu(IV) are relatively close to those for U(VI) (see Figure 7.2), the plutonium will tend to follow the uranium (with a minor adjustment of the flowsheet parameters). It will then be possible to separate them by reductive plutonium stripping, as in the PUREX process, or to co-manage them as in the COEX™ process.
- In the THOREX process, technetium is co-extracted with thorium in the solvent and is found in the Th product stream. It could probably be eliminated by an additional high acidity wash comparable to that of the first cycle of the PUREX process implemented at La Hague, but this would generate very large effluent volumes due to the high solvent flow rate compared with the feed rate (Figure 7.3, $Q_{\text{solvent}}/Q_{\text{feed}} = 9$, compared with only 3 in the PUREX process).

Figure 7.9. Limit of third-phase formation Th(IV), 30% TBP and 40% Amsco at 26.5°C and 50°C



Source: Rao and Kolarik, 1996.

7.2.4 Problems arising from the presence of HDBP (TBP degradation product)

The presence of HDBP, the main TBP hydrolysis and radiolysis degradation product, is unavoidable. It forms strong complexes with all the tetravalent actinides and is thus responsible for their retention in the solvent during tetravalent actinide stripping (as in the PUREX process). Residual thorium contamination of the uranium product is partly related to this phenomenon, hence the need for an additional purification cycle (as in the Interim-23 flowsheet at Atelier Pilote de Marcoule or purification on ion exchange resins [Anantharaman et al., 2000]). In the PUREX process this difficulty was taken into account

by implementing a “plutonium barrier”, based on the use of U(IV) to reduce Pu(IV) to Pu(III), and as a competitor for complexation by HDBP. In the case of thorium, U(IV) cannot reduce Th(IV) and may be less effective. However, a thorium barrier can be designed, as proposed in patent (Lecomte et al., 2003) using mono-vacant heteropolyanions (e.g. $P_2W_{17}O_{61}^{10-}$ or $SiW_{11}O_{39}^{8-}$), which are powerful aqueous complexing agents for the tetravalent actinides.

Moreover, under certain conditions, these Th-DBP complexes tend to form precipitates in the same way as the Th-MBP complex (mono-butyl di-hydrogen phosphate is a degradation product of HDBP itself). These precipitates are potential sources of organic phase airlift plugging as observed in India during thorium fuel reprocessing campaigns using the Interim-23 process (Lakshmanan et al., 2000). These problems associated with the presence of TBP degradation products will probably be exacerbated at higher burn-up due to the increased concentrations of ^{232}U , ^{228}Th and ^{229}Th .

7.2.5 Di-secondary-butyl phenyl phosphonate in diethylbenzene as the solvent

The Interim-23 process is used to extract uranium (^{233}U) and leave thorium with the fission product components of the spent fuel. The Interim-23 process is more efficient when using di-secondary-butyl phenyl phosphonate in diethylbenzene as the solvent which enables a better separation factor for uranium from thorium than when using TBP. Uranium losses to the raffinate stream are typically <0.01% and the decontamination factor obtained from fission products is ~105.

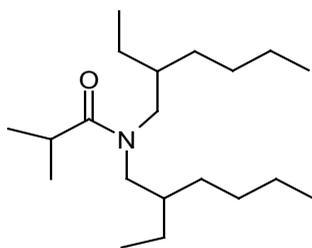
7.2.6 N,N dialkylamides as alternatives to TBP

Dialkylamides (or monoamides) have been investigated since the 1980s as potential alternatives to TBP for reprocessing spent uranium-based fuels. Their main advantages with respect to TBP are the following (Musikas, 1994; Miguiditchian et al., 2008; Patak et al., 1999; Patak and Manchanda, 2000; Patak, Prabhu and Manchanda, 2003):

- fully incinerable molecules composed only of carbon, hydrogen, oxygen and nitrogen (CHON concept), generating no saline effluent;
- safe degradation products;
- excellent selectivity with respect to fission products (even better than TBP);
- actinide(VI)/actinide(IV) selectivity easily tailored by branching alkyl chains;
- inexpensive easily synthesised molecules.

Di(2-ethylhexyl)isobutyramide (DEHiBA, Figure 7.10) was selected by the CEA for the first cycle of the GANEX process (Miguiditchian et al., 2008) designed for selective uranium extraction prior to co-management of the other actinides. In India, the same molecule is currently being studied (Patak et al., 1999; Patak and Manchanda, 2000; Patak, Prabhu and Manchanda, 2003) as a substitute for 5% TBP in the Interim-23 process.

Figure 7.10. Di(2-ethylhexyl)isobutyramide (DEHiBA)



7.2.7 Conversion to oxides

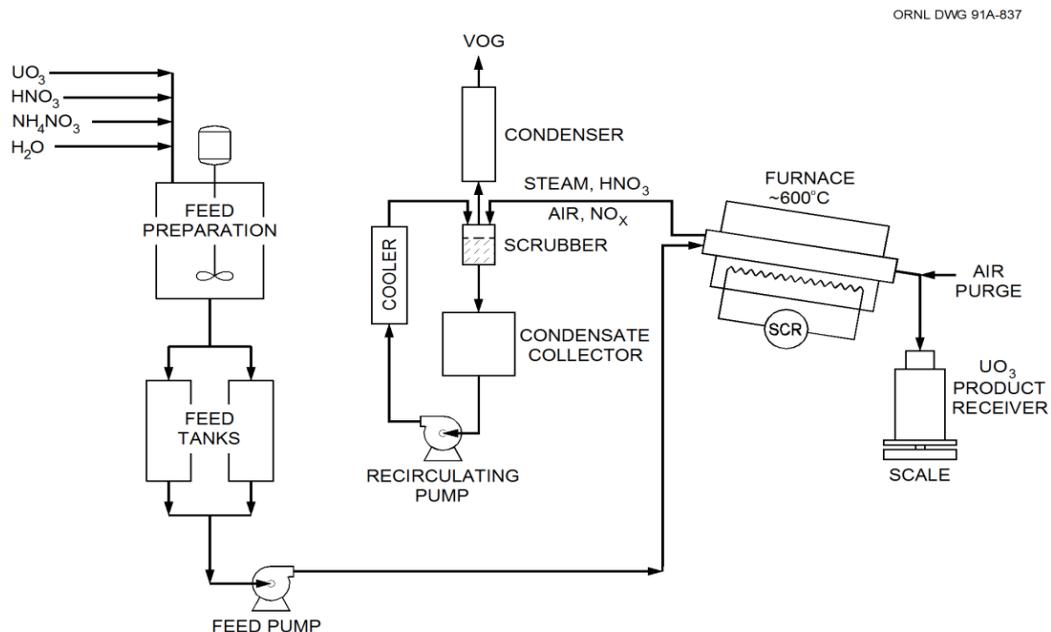
The main disadvantage of the thorium cycle is related to the unavoidable presence of ^{232}U , which accompanies ^{233}U . One of its decay products, ^{208}Tl , is a 2.6 MeV gamma emitter that requires substantial changes in the downstream fuel fabrication process because it cannot be handled in gloveboxes, as is the case for UPuO_2 MOX fuel. Various conversion routes are being investigated in India to streamline the fabrication process (Anantharaman et al., 2000; Rao et al., 2000; Kutty et al., 2009; Kutty et al., 2008a; Kutty et al., 2008b):

- oxalate co-precipitation, followed by granulation as either a dry process or by paste/extrusion to mitigate problems arising from contaminating particulates during press compaction and sintering;
- sol-gel, with the production of either porous microspheres to obtain pellets by press compaction and sintering, or dense microspheres prior to vibropac fuel fabrication, impregnation of partially sintered ThO_2 pellets.

7.2.8 Modified direct denitration (MDD) process

MDD is a modified thermal denitration process developed at ORNL in the early 1980s (Haas, Arthur and Stines, 1981). The MDD process is capable of converting uranium nitrate to oxide or conversion of mixed U-Pu or Th-U nitrates to mixed oxides. The process utilises decomposition of ammonium-metal double nitrates (Notz and Haas, 1981) to avoid the sticky mastic phase obtained during simple thermal denitration, and to obtain an active, free-flowing agglomeration of UO_3 -based powder suitable for use in cold-pressed fuel pellets. The MDD process uses a simple rotary kiln as the reactor and has been operated successfully in up to 16-cm rotary kilns for UOX production. The process has also been operated in glovebox contained equipment for the production of 100 g/hour of mixed U-Pu oxide. The process can likely be adapted to operation in fully shielded facilities for ^{233}U conversions.

Figure 7.11. Diagram of modified direct denitration process



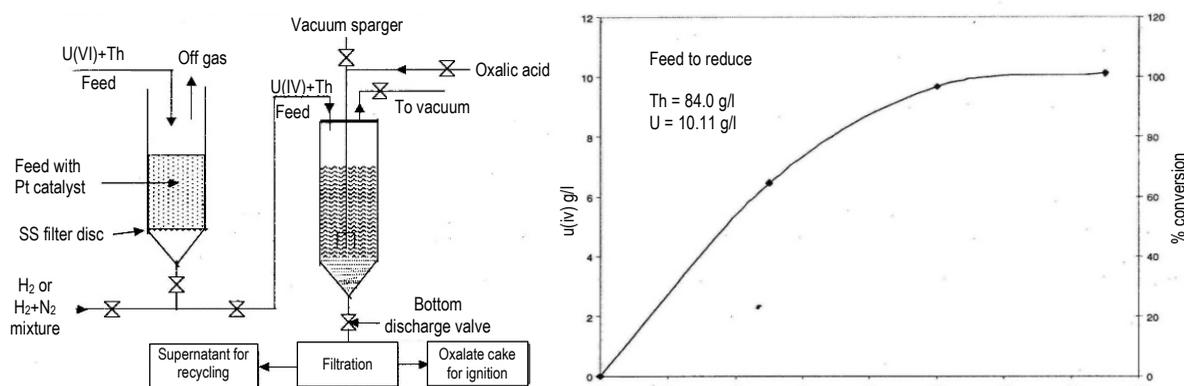
Source: Haas, Arthur and Stines (1981).

7.2.9 Oxalate co-conversion

In this case, the output solution from the separation process is a Th(IV)-U(VI) mixture containing ~10% U. The final ^{233}U concentration (1-10%) is adjusted by dilution with thorium nitrate. The total heavy metal content is increased to ~100 g/L by concentration in an evaporator. Oxalate co-precipitation requires prior reduction of U(VI) to U(IV), for which several routes, including the electrochemical route, can be considered. Two options are under consideration in India:

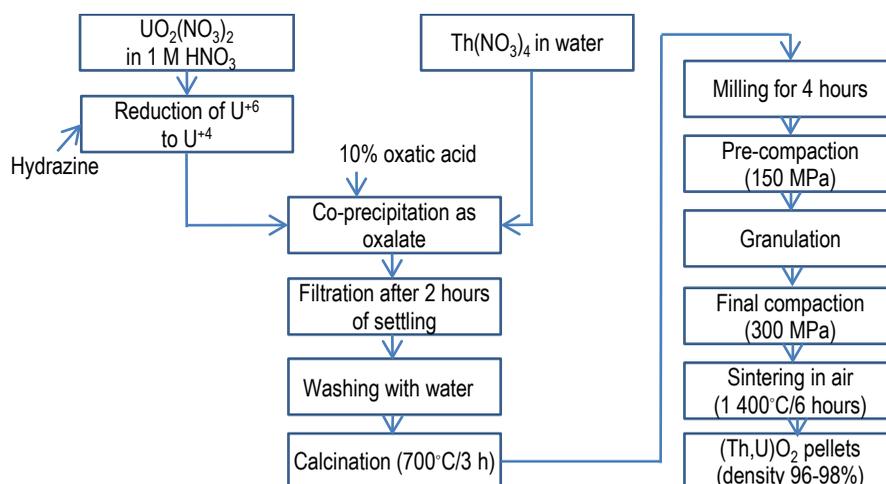
- reduction by hydrogen, either pure or diluted to 8% in argon or nitrogen on PtO_2 catalyst (Figure 7.12);
- reduction by hydrazine on PtO_2 catalyst (see Figure 7.13) with >96% efficiency in 0.1 M hydrazine and 0.4 M HNO_3 .

Figure 7.12. Schematic diagram of reduction/precipitation unit and reduction kinetics



Source: Rao et al. (2000).

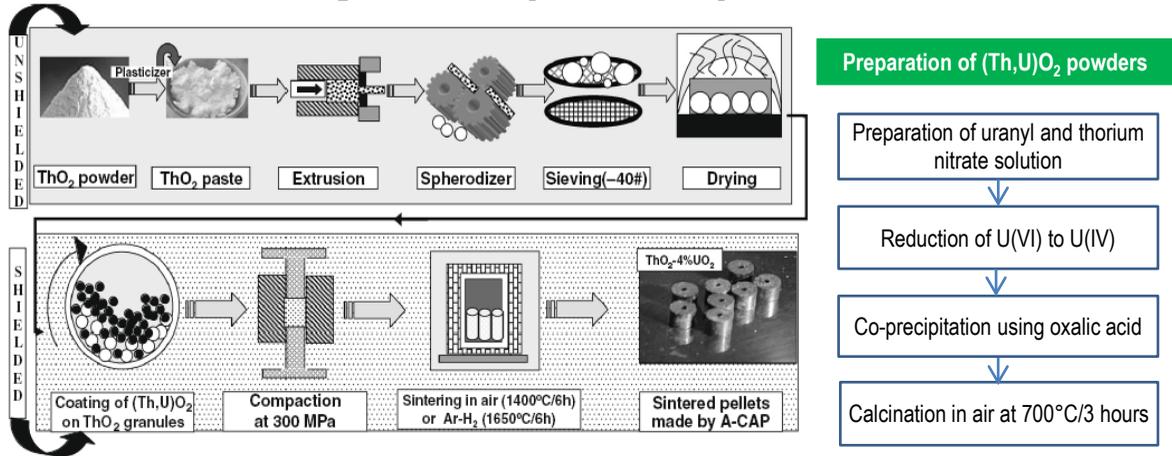
Figure 7.13. Fabrication process: Oxalate co-conversion followed by mechanical granulation



Source: Kutty et al. (2009).

In the CAP process developed in India (Kutty et al., 2008a), ThO_2 microspheres are produced by extrusion, then encapsulated in $(\text{Th,U})\text{O}_2$ powder produced by oxalate co-conversion (Figure 7.14).

Figure 7.14. CAP process developed in India

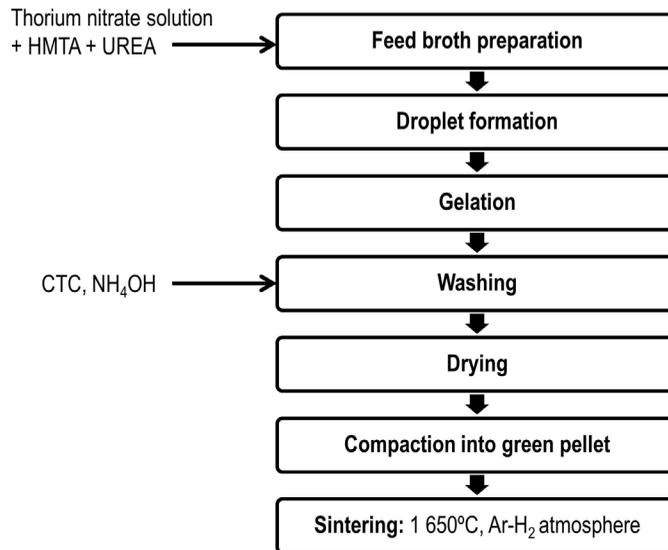


Source: Kutty et al. (2008a).

7.2.10 Sol-gel

The sol-gel route is studied in India for both the Sol-Gel Microsphere Pelletisation (SGMP) and vibropac SGM processes. In both cases, the sol-gel conversion process is the same, using an internal sol-gel (see Figure 7.15).

Figure 7.15. Flowsheet for SGMP pellet fabrication



Source: Anantharaman et al. (2000).

The internal sol-gel co-conversion route is identical to the process investigated by the CEA for uranium-based fuel or transmutation targets (Grandjean et al., 2007).

The authors (Anantharaman et al., 2000) emphasise that although this option is attractive (the production of microspheres would eliminate the problem of highly radioactive particulates), it generates large volumes of aqueous effluents that are not easy to manage due to the presence of ammonium nitrate. In addition, the microspheres

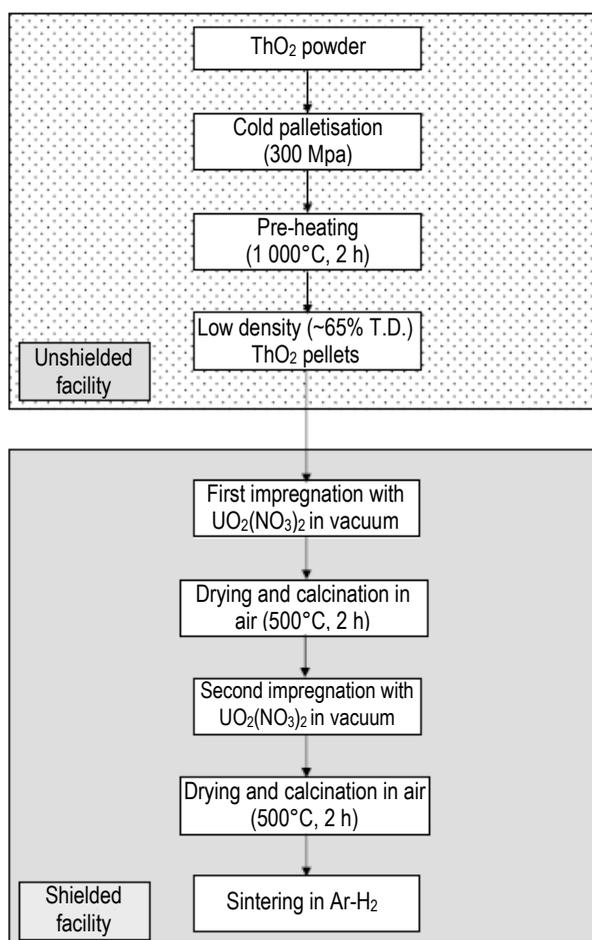
require multi-coatings of carbon and silicon carbide by means of the continuous vapour deposition process, which has not been demonstrated at industrial scale.

7.2.11 Impregnation

To minimise and simplify remote operations behind heavy biological shielding, the study focuses on impregnating ThO_2 pellets that are partially sintered (2 hours at $1\,000^\circ\text{C}$) and of low density ($\sim 65\%$ of the theoretical density). Impregnation is performed with a uranyl nitrate solution (1.5 M) either in a vacuum or by microwave heating of the pellets (Anantharaman et al., 2000; Kutty et al., 2008b) (Figure 7.16).

Currently the main limitation of this process is the maximum feasible uranium concentration, about 2-2.5% in a single impregnation step and 4% with two consecutive impregnation steps.

Figure 7.16. Pellet impregnation process



Source: Anantharaman et al. (2000).

7.3 Fluoride volatility

As an alternative to aqueous dissolution, the use of fluoride volatility to separate uranium from thorium in the initial stage of a recycling process has been investigated. Fluoride volatility reprocessing belongs to the more general halide volatility family of

reprocessing methods. Halide volatility methods employ the high chemical reactivity of halogen gases to convert nearly all the species in irradiated fuel to halide compounds. The relative volatilities of the halide compounds can then be exploited to separate them by distillation or other methods. The boiling points of uranium hexafluoride and thorium tetrafluoride differ significantly, with the boiling point of ThF_4 higher than that of UF_6 . Therefore, it is feasible to apply fluoride volatility to irradiated thorium dioxide fuel to separate uranium from thorium.

7.3.1 Removal of uranium from thorium/uranium SIMFUEL

To investigate the prospects of fluoride volatility to separate uranium from thorium fuel, a laboratory scale apparatus was used at AECL to expose small samples of thorium/UOX ($\text{ThO}_2\text{-UO}_2$) and simulated irradiated thorium/UOX fuel (SIMFUEL) to fluorine gas at elevated temperatures (Hamilton, 2012). The effectiveness of the uranium removal was determined by analysis of the non-volatile residue. Further purification of the volatile and non-volatile fractions is not considered here. It is worth noting that considerable work has been performed in the design of a system for the purification of the volatile fraction arising from the application of the fluoride volatility process to uranium dioxide-based fuel and that effort can be utilised for the thorium dioxide system as well (Shatalov et al., 2001).

Three types of thorium/UOX pellets were used. Initial experiments to investigate the effects of temperature, time and particle size were conducted on thorium dioxide samples containing 1.5 wt% UO_2 . Optimised conditions were applied to two different SIMFUELS;¹ one based on the irradiation of pure thoria (low burn-up SIMFUEL) and the other on a high burn-up (60 GWd/tHE) SIMFUEL with a high (13 wt%) initial loading of UO_2 . The chemical composition of irradiated fuel is too complex to duplicate exactly. For the fabrication of SIMFUEL, the most abundant fission products, actinides and rare-earths are either represented directly or as a group with non-radioactive elements. The elements simulated are: Ag, Am, Ba, Ce, Cm, Dy, Er, Eu, Gd, Ho, La, Mo, Nb, Nd, Np, Pa, Pd, Pm, Pr, Pu, Rh, Ru, Sm, Sn, Sr, Tb, Tc, Th, U, Y and Zr.

All samples were made by dry powder milling, followed by compaction into pellets and sintering. Microscopy and x-ray diffraction results confirm that the uranium is in solid solution with the thorium dioxide. Microprobe analysis showed that the SIMFUEL contained micro-structural features found in irradiated fuel (Kleykamp et al., 1985), including metal particles (mostly ruthenium) and a second oxide phase containing barium, strontium, zirconium and uranium.

A horizontal tube furnace containing a 13 mm diameter Monel tube was connected to a 4% fluorine-in-argon gas supply via a 3 mm Monel line. A gas flow of 30 mL/min was used for all experiments discussed here. Gas exiting the furnace tube was directed to the gas scrubbers via another 3 mm Monel line. The gas scrubber consisted of a dry bed of sodium hydroxide beads in the first stage, with the gas bubbled through a sodium hydroxide solution in the second stage. Gas flow was controlled with a needle valve. Alumina (Al_2O_3) boats were used to hold the samples for all experiments.

Using the thorium dioxide with 1.5 wt% UO_2 , the variables of temperature, time and particle size were investigated. As expected in a gas-solid reaction, the extent of uranium removal increased as the sample particle size decreased. This is a surface area effect, but since the surface area was not known, the results are discussed as a function of particle size.

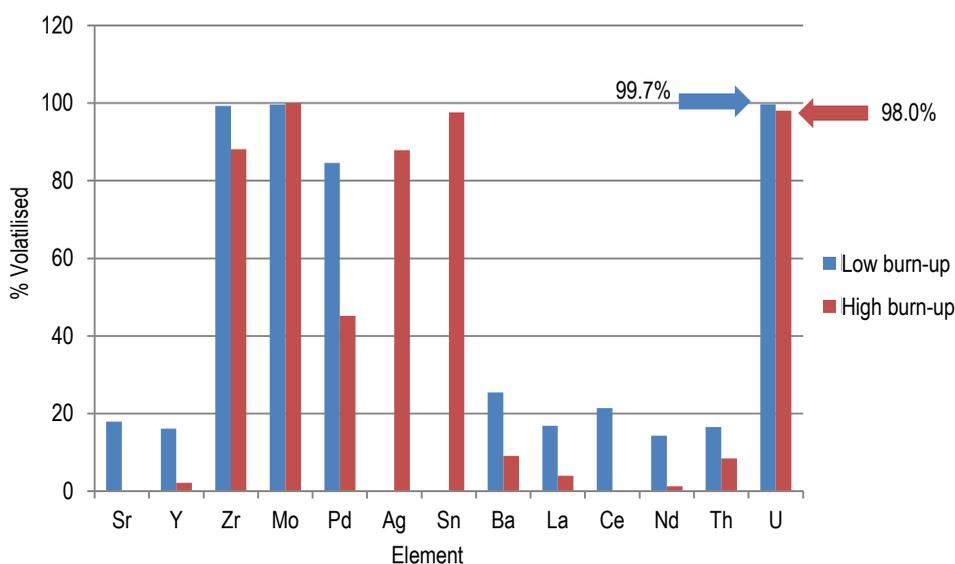
1. SIMFUEL = SIMulated burn-up FUEL (an unirradiated surrogate for irradiated fuel that contains non-volatile simulated fission products).

Based on the results from previous work (Shimada, Okumura and Higashi, 1973), it was anticipated that the ThF_4 product of the reaction between ThO_2 and F_2 was impeding the penetration of the F_2 gas and preventing a complete conversion of the ThO_2 to ThF_4 . Consequently, this prevented the complete release of the uranium (as UF_6). To overcome this, a technique of intermediate crushing was investigated. Three iterations of removal, crushing and F_2 exposure at two-hour intervals resulted in the best uranium removal for the conditions examined here, greater than 98%.

Soak temperatures of 400°C, 550°C and 700°C were examined. Experiments were performed with intermediate crushing steps after two and four hours of F_2 gas exposure at maximum temperature. The maximum uranium removal corresponded with the maximum temperature of 700°C used in these tests.

The two SIMFUEL formulations, corresponding to low and high burn-up, were exposed to the same experimental conditions, a total of six hours of F_2 exposure at 700°C with intermediate crushing steps after two and four hours. The results are presented in Figure 7.17. There is no Ag or Sn additive in the low burn-up SIMFUEL. The results demonstrate similar behaviour regarding the volatility of the various additives. Results for ruthenium and rhodium are not included because analysis of the starting material did not produce representative results for these two elements. It is suspected that the dissolution technique used did not dissolve the noble metal particles, resulting in their anomalously low concentration. The same analytical technique was used on the non-volatile residue following fluoride exposure; the results from this analysis for ruthenium and rhodium could not be considered valid for the same reason.

Figure 7.17. Extent of element volatility in low and high burn-up SIMFUEL



Source: Hamilton (2012).

The most effective uranium separation was achieved at 700°C, the highest temperature used in this study, suggesting higher temperatures should be investigated. It is expected that the use of higher temperatures will also increase the extent of fluorine reaction with structural materials and that the optimum temperature will be one that is a compromise between these opposing effects.

The ThF_4 reaction layer formed at the surface of the oxide particles impedes further reaction. Exposure of fresh oxide surface and re-exposure to fluorine has been shown to facilitate uranium removal in excess of 99%. The effect of particle size was investigated,

but some experimental results had significantly large uncertainties due to process variations. The limited information available suggests that, with a particle size $>45\ \mu\text{m}$, complete uranium removal was not possible after two hours at 700°C . This suggests a smaller particle size is required if complete uranium removal is to be achieved in a single step without comminution of the intermediate product.

These “proof of principle” tests suggest that fluoride volatility could achieve an effective separation of uranium and thorium. It is suggested that an operation combining the processes of comminution and fluoride volatility could achieve uranium and thorium separation in a single, dry step.

7.4 Summary

Thorium fuel is generally reprocessed by a form of the THOREX liquid-liquid extraction process, which is similar to the PUREX process. This process (and its variants) has been successfully used at pilot scale, notably in the United States to reprocess about 900 tonnes of thorium fuel (~ 1.5 tonnes ^{233}U) and also in France. A pilot scale demonstration also took place at AECL’s Whiteshell Laboratories in the 1970s and 1980s. This was known as the Thorium Fuel Reprocessing Experiment (TFRE).

However, extrapolating the THOREX process to industrial scale will require further development of the dissolution, liquid-liquid extraction and conversion steps. The dissolution of thorium metal or oxide is more complex than for uranium and requires the addition of hydrofluoric acid, whose corrosiveness with respect to the process equipment must be buffered by adding aluminium nitrate. The influence of the burn-up and the impact of the conversion and fabrication process and fuel microstructure also require further investigation. The main difficulties in the purification step by liquid-liquid extraction are related to the low solubility of the Th-TBP complex and to the interaction between the main TBP degradation product, HDBP and thorium. It might be advantageous to modify the solvent formulation by replacing TBP or the diluent.

Currently, no developed alternative to variants of the THOREX exists for reprocessing thorium-based fuels although other extractants have been investigated and fluoride volatility has been investigated as discussed in Section 7.3. THOREX itself has yet to reach the maturity of the commercial PUREX process.

Finally, a major challenge associated to thorium reprocessing is related to the unavoidable presence of ^{232}U , which accompanies ^{233}U . One of its decay products, ^{208}Tl , is a 2.6 MeV gamma emitter that will require substantial changes in the downstream fuel fabrication process because it cannot be handled in gloveboxes, as is the case for (U,Pu) O_2 MOX fuel. In these conditions, the process of conversion to oxide and the recycle fuel fabrication process would need to be developed and demonstrated to be remotely operated within a fully shielded facility. A related challenge is the handling and storage of excess thorium, which will contain ^{228}Th and its highly radioactive daughters for about 20 years.

References

- Anantharaman, K., et al. (2000), “Thorium-based fuel reprocessing and re-fabrication technologies and strategies”, *Proceedings Annual Conference of Indian Nuclear Society INSAC 2000*, Vol. 2, pp. 107-122.
- Flanary, J.R., et al. (1964), *Chemical Development of the 25-TBP Process (Interim-23)*, Report ORNL-1993.
- Goode, J.H. and J.R. Flanary (1965), “Dissolution of irradiated, stainless-steels-cald ThO_2 - UO_2 in fluoride-catalysed nitric acid solutions: hot-cell studies on pelletized, arc-fused and sol-gel-derived oxides”, Report ORNL-3725.

- Grandjean, S., et al. (2007), "Synthesis of mixed actinide compounds by hydro-metallurgical co-conversion methods", *Proceedings of Global 2007*, Boise.
- Gresky, A.T. (1956), "Solvent extraction separation of ^{233}U and thorium from fission products by means of tributyl phosphate", *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy*, Geneva, Vol. 9, pp. 505-510.
- Haas, P.A., R.D. Arthur and W.B. Stines (1981), *Development of Thermal Denitration to Prepare Uranium Oxide and Mixed Oxides for Nuclear Fuel Fabrication*, ORNL-9735.
- Hamilton, H. (2012), "Removal of uranium from thorium-uranium SIMFUEL by fluoride volatility", *Proceedings of the 12th Information Exchange Meeting on Partitioning and Transmutation*, Prague.
- IAEA (2000), *Thorium Based Fuel Options for the Generation of Electricity: Developments in the 1990s*, IAEA TECDOC-1155, International Atomic Energy Agency, Vienna.
- IAEA (2005), *Thorium Fuel Cycle-Potential Benefits and Challenges*, IAEA-TECDOC-1450, International Atomic Energy Agency, Vienna.
- Kleykamp, H., et al. (1985), "Composition and structure of fission product precipitates in irradiated oxide fuels: Correlation with phase studies in the Mo Ru Rh Pd and BaO- UO_2 - ZrO_2 - MoO_2 Systems", *Journal of Nuclear Materials*, Vol. 130, pp. 426-433.
- Kutty, T.R.G., et al. (2008a), "Characterization of (Th,U) O_2 pellets made by advanced CAP process", *Journal of Nuclear Materials*, Vol. 384, pp. 303-310.
- Kutty, T.R.G., et al. (2008b), "Characterization of (Th,U) O_2 fuels pellets made by impregnation technique", *Journal of Nuclear Materials*, Vol. 374, pp. 9-19.
- Kutty, T.R.G., et al. (2009), "Characterization of ThO_2 - UO_2 pellets made by co-precipitation process", *Journal of Nuclear Materials*, Vol. 389, Issue 3, pp. 351-358.
- Lakshmanan, K., et al. (2000), "Recovery of ^{233}U from irradiated J rods – operating experience", *Proceedings of Indian Nuclear Society Annual Conference INSAC 2000*, Vol. I, pp. 166-167.
- Lecomte, M., et al. (2006), *Procédé de séparation de l'uranium (VI) d'actinides (IV) et/ou (VI) et ses utilisations*, CEA-AREVA EP1685573.
- Lung, M. (1997), "A present review of the thorium nuclear fuel cycles", European Report EUR 17771 EN, European Commission, Luxembourg.
- Mallik, G.K., R.K. Malav and H.S. Kamath (2000), "Impact of microwave heating technique on dissolution of sintered thoria pellets", *Proceedings Annual Conference of Indian Nuclear Society INSAC 2000*, Vol. 1, p. 150.
- Merz, E. (1982), "Reprocessing of $^{232}\text{Th}/^{233}\text{U}$ fuels", *Gmelin Handbook*, Thorium supplementary A4.
- Miguirditchian, M., et al. (2008), "Development of the GANEX process for the reprocessing of Gen-IV spent nuclear fuels", *Proceedings of ATALANTE 2008*, Montpellier.
- Montuir M., et al. (2011), "Parex simulation code: an efficient tool to model and simulate solvent extraction operations, Récents progrès en génie des procédés", Numéro 101, ISSN 1775-335X, Ed SFGP, Paris.
- Musikas, C. (1994), *Method of extracting uranium (VI) and/or plutonium (IV) present in an aqueous acid solution by means of a mixture of N,N-dialkyl amides, for use in the reprocessing of irradiated nuclear fuels*, CEA EP0381579.
- Nakashima, T. and Z. Kolarik (1983), "The formation of a third phase in the simultaneous extraction of actinide (IV) and uranyl nitrates by tributyl phosphate in dodecane", *Solvent extraction and ion exchange*, Vol. 1(3), pp. 497-513.

- Notz, K.J. and P.A. Haas (1981), *Properties and Thermal Decomposition of the Double Salts of Uranyl Nitrate-Ammonium Nitrate*, ORNL/TM-7820, Oak Ridge National Laboratory, Oak Ridge.
- Patak, P.N., D.R. Prabhu and V.K. Manchanda (2003), "The role of oxidants in Pu-Th separation studies employing Di-2-ethylhexyl isobutyramide (D2EHIBA) as extractant", *Radiochim, Acta*, Vol. 91, pp. 141-146.
- Patak, P.N., et al. (1999), "Separation studies of uranium and thorium using Di-2-ethylhexyl isobutyramide (D2EHIBA)", *Separation Science and Technology*, Vol. 34(1), pp. 2601-2614.
- Patak, P.N. and V.K. Manchanda (2000), "Separation of uranium from irradiated thorium employing branched chain amides as extractants", *Proceedings of Indian Nuclear Society Annual Conference INSAC 2000*, Vol. I, pp. 151-153.
- Rainey, R.H. and J.G. Moore (1962), "Laboratory development of the acid THOREX process for recovery of consolidated EDISON thorium reactor fuel", Report ORNL-3155.
- Rao, P.R.V. and Z. Kolarik (1996), "A review of third phase formation in extraction of actinides by neutral organophosphorus extractants", *Solvent extraction and ion exchange*, Vol. 14(6), pp. 955-993.
- Rao, K.S., et al. (2000), "Co-processing and catalytic reduction and remote controlled oxalate precipitation – a new route for ²³³U/Th MOX", *Proceedings of Indian Nuclear Society Annual Conference INSAC 2000*, Vol. I, pp. 157-159.
- Schulz, W.W. (1989), *Science and Technology of Tributyl Phosphate*, Vol. III, CRC Press, pp. 225-247.
- Shatalov, V.V., et al. (2001), "Gas-fluoride technology for processing spent oxide fuel", *Atomic Energy*, Vol. 90, No. 3.
- Shimada, S., I. Okumura and K. Higashi (1973), "Experimental study on fluoride volatility process for thorium fuels", *Journal of Nuclear Science and Technology*, Vol. 10 (11), p. 689-695.
- Srinivasan, N., et al. (1973), *Laboratory Studies on Acid Thorax Process*, BARC report n°681.
- Vijayan, K., et al. (2000), "Studies on the dissolution of sintered thoria pellets in HNO₃+Al (NO₃)₃ mixtures containing HF and NaF", *Proceedings Annual Conference of Indian Nuclear Society INSAC 2000*, Vol. 1, pp. 148-149.

8. Waste management issues

The use of thorium as nuclear fuel is often associated with advantages in the radiotoxicity of the resulting waste as compared to conventional uranium fuels. It must not be overlooked, however, that the implementation of thorium fuels with the view of developing a self-sustainable thorium fuel cycle will require the use of mixed fuel forms (thorium-LEU or thorium-plutonium fuels) during very long transition phases before a full Th/²³³U cycle can be achieved. For these mixed fuel forms, a comparison in terms of advantages or disadvantages over current UO₂ fuels will strongly depend on the mixed fuel form considered and on fuel management and recycling strategies.

Thorium-based fuels (in the form of ThO₂) have slightly more flexible characteristics than those of uranium fuels with regard to interim storage of spent fuel, because of the more chemically inert character of thorium oxide as compared with UOX (substantially, all uranium fuels are in the form of oxide). This particularity results in the maximum acceptable temperature in dry storage conditions of spent UO₂ fuel being lower than that of ThO₂ fuels. If the fuel matrix is accidentally exposed to the atmosphere, UO₂ can oxidise to U₃O₈ leading to a volume expansion with subsequent risk of fuel cladding failure. This oxidation risk does not apply to ThO₂ fuel, as it is already in its maximum oxidation state. Nevertheless, the temperature limits in the storage phase are imposed by those of the fuel cladding (in particular for zirconium alloys).

The benefits of improved chemical stability of ThO₂ compared to UO₂ may have advantages for the final disposal of spent fuels in geological sites (if this option is chosen). When exposed to groundwater under oxidising conditions, uranium can be converted into the uranyl cations (UO₂)²⁺ and other derivatives that are soluble in water. This can lead to premature degradation of the fuel matrix and an increased release of radioactive materials, which might lead to difficulties in the repository safety case. However, thorium and uranium behave similarly in reducing environments.

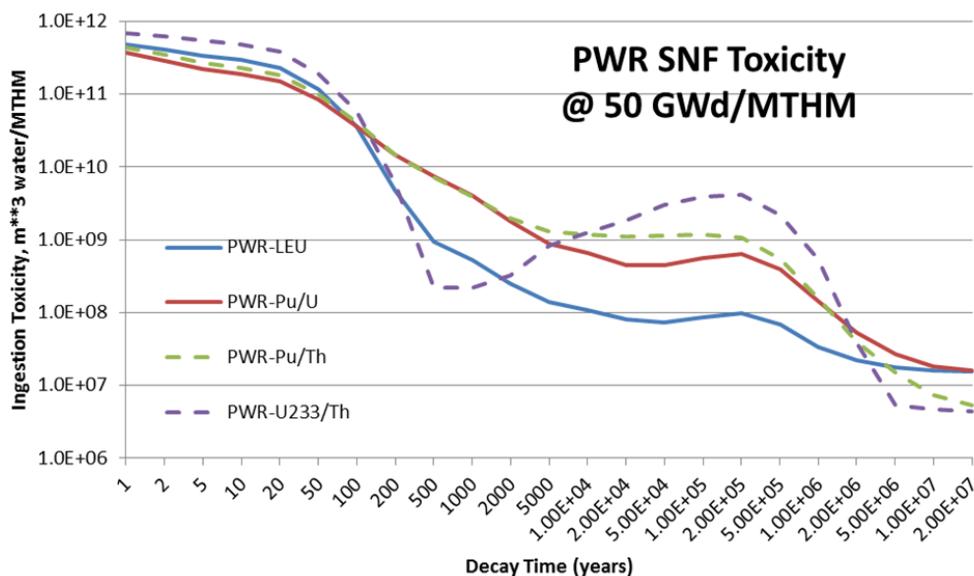
One of the often claimed advantages of the thorium cycle is that it produces less plutonium and other actinides which significantly reduce the radiotoxicity of waste. While a pure Th/²³³U cycle will indeed produce a much reduced amount of plutonium and MAs than conventional UO₂ fuels, this is not the case for thorium-plutonium mixed fuel forms, and is less clear for thorium-LEU fuels. Furthermore, decay products from ²³³U drive radiotoxicity to be higher than that of LEU or U/Pu for the period between about ten thousand years and one million years: this is mainly due to the presence of ²³⁴U (mainly produced through neutron capture on ²³³U) and its decay product, ²²⁶Ra. Figure 8.1 provides a comparison of spent fuel radio-toxicity for similar uranium and thorium-based fuels, for the same discharge burn-up. The relative differences between radiotoxicities resulting from the use of both cycles vary greatly depending on recycling strategies and recycling efficiencies considered and must therefore be interpreted with care.

The long-term radiotoxicity of thorium-based spent nuclear fuels is therefore more accurately described as being comparable to that of uranium-based spent nuclear fuels.

Whether a reduction in radiotoxicity is translated into an actual advantage will in practice depend on whether this reduction leads to a significant change in the likelihood of making a safety case for the disposal. Many disposal studies show safety cases driven by long-lived mobile fission products rather than by highly radiotoxic actinides; this

means that the most direct driver of disposal difficulty is more likely to be the amount of power generated, rather than the system it is generated by. Similar arguments can be advanced against waste volume per se being the major driver of disposal economics, but a potential advantage of thorium fuel would result from the possibility of achieving higher burn-up compared to uranium (as for a given amount of energy released in a reactor, the mass of spent fuel to manage is, by definition, inversely proportional to the burn-up).

Figure 8.1. Comparison of spent fuel ingestion toxicity¹ for different PWR fuel types at the same discharge burn-up



1. Ingestion toxicity, or ingested toxic potential, is a method of representing the hazard posed by radioactive waste. The International Commission for Radiological Protection (ICRP) has given recommended figures for the dose which would result from the ingestion of each radionuclide in an amount of radioactive waste. The method considers that the waste is dissolved in water and calculates how much water would be needed to dilute the waste to such an extent that someone could use the water for all his or her liquid intake in a year without exceeding the maximum permitted public dose of 1 millisievert. This is termed the ingested toxic potential of the waste.

Source: Croff and Krahn (2014).

8.1 Fission products

For a given amount of thermal energy produced in a reactor core, the amount of fission products is essentially the same in thorium or uranium fuels since, on a first approximation, a fission event releases approximately 200 MeV, regardless of the nuclide that fissions, and gives birth to two fission products.

The relative proportion of each of the radioisotopes is slightly different according to the origin of the fission (²³⁵U, ²³⁹Pu, ²⁴¹Pu, ²³³U) and consequently the evolution in time of the global radiotoxic inventory (GRI) of these products is slightly different in the two cycles. However, this is of minor importance as most of the radioactivity from fission products disappears after a few centuries, and there is likely to be little difference in the amounts of long-lived mobile fission products that tend to drive repository safety cases. Consequently, there is almost no difference between thorium and uranium-based fuels with regard to the fission product part of the nuclear waste.

8.2 Actinides

The difference between the two fuel cycles may come from the amount of MAs generated in each case. In the case of the uranium cycle, significant amounts of three long-lived transuranic elements (other than plutonium) are generated: ^{237}Np (with half-life of 2.14 million years), americium (mainly ^{241}Am , with a half-life of 432 years and ^{243}Am , with a half-life of 7 380 years) and curium (mainly ^{245}Cm , with a half-life of 8 530 years). These nuclides are particularly radiotoxic alpha emitters and the main contributors to GRI of ultimate radioactive waste beyond a few hundred years (with the exception of plutonium, which may be recycled depending on the specific fuel cycle). These nuclei are poorly soluble in water and have very low mobility in geological media (provided these media are under reducing conditions): this practically excludes the possibility of their migrating to the biosphere, at least over timescales during which they could deliver significant radiological doses to living species. The partitioning of some or all of these elements and their subsequent transmutation in nuclear reactors is an option that is studied today with the view of further minimising this risk. Research has been conducted in this direction for many years in several countries (such as France, Japan and the United States) suggesting that these operations could be carried out industrially. However, this would require the implementation of complex processes and technologies with important additional costs.

In the pure Th/ ^{233}U fuel cycle, almost none of the above mentioned MAs are produced. Americium and curium mainly come from plutonium (via neutron capture and various radioactive decays), while ^{237}Np mainly comes from ^{235}U (via the formation of ^{237}U , which decays to ^{237}Np).

Irradiated Th/ ^{233}U fuels produce, however, other long-lived radionuclides such as ^{231}Pa (half-life of ~33 000 years), which results from the beta-decay of ^{231}Th (half-life of ~25 hours) which in turn is formed by (n,2n) reactions of ^{232}Th . The long half-life of ^{231}Pa makes it a significant contributor to long-term GRI. In Th/ ^{233}U fuel, MAs associated with the uranium-plutonium cycle are also found, although in much smaller amounts: in a Th/ ^{233}U fuel with a burn-up of ~60 GWd/t, there is approximately 30 times less ^{237}Np (half-life of ~2 million years), than in a uranium-plutonium fuel. This ratio is about 60 for ^{238}Pu (half-life 88 years) (Lung, 1997).

A disadvantage of the thorium-uranium fuel cycle arising from reprocessing spent fuel (for recovery and recycle of ^{233}U), is the requirement to avoid off-gas radon transport of the highly radioactive radon daughters through high-efficiency particulate air (HEPA) filters to the environment. Retention of around ten minutes of the off-gas must be incorporated into the design of the off-gas system to allow for decay of ^{220}Rn prior to HEPA filtration.

References

- Croff, A. and S. Krahn (2014), "Comparative assessment of thorium fuel cycle radiotoxicity", *Proceedings of ANS Winter Meeting 2014*, Anaheim, California.
- Lung, M. (1997), "A present review of the thorium nuclear fuel cycles", European Report EUR 17771 EN.

9. Non-proliferation issues

The non-proliferation of nuclear weapons from the use of nuclear energy has always been considered as an essential element to be taken into account in the development of any new technology or process implemented in this field. This issue was specifically addressed in an extensive study conducted between 1978 and 1980 on the nuclear fuel cycle at large (INFCE, 1980). A general conclusion was that the technical obstacles to military use of thorium cycles with uranium enriched to less than 20% are similar to those of the uranium/plutonium cycle. However, it should be underlined that this was an exercise involving nearly fifty countries and more than 500 experts of varying provenance. The study's deliberations¹ were based on a vast amount of technical data mixed with some contributions affected by commercial interests, as well as political or diplomatic considerations. Thus, it is not sufficient to refer only to the study's general conclusions to compare more precisely the non-proliferation relative merits (or demerits) of the uranium and thorium cycles.

For such a comparison, there is a need to consider the physical characteristics that need to be taken into account in assessing the difficulty of using fissile materials for atomic bombs. There are basically four:

- The critical mass of a bare homogeneous sphere of fissile material, that is without neutron reflector. This determines the amount of material required to be manufactured and also the weight and size of the explosive device. Note that in reality, the amount of material needed to make a bomb is less than the critical mass, given a number of measures such as the introduction of neutron reflectors or compression devices.
- Spontaneous neutron emission, which should be as low as possible for reasons related to the functioning of a "rudimentary" nuclear bomb. In effect, excessive spontaneous emission of neutrons requires the design of very complex devices to guard against the premature triggering of the chain reaction by these neutrons before the fissile mass reaches the maximum critical configuration (maximum multiplication factor). Too highly neutron-emitting fissile materials can make it practically impossible to design an "effective" and reliable bomb.
- The heat generated by the intrinsic radiation emitted by the fissile material itself. Excessive heat can complicate the process for making a bomb and may even jeopardise its operation unless sophisticated specific provisions are implemented to deal with this effect.
- The radiation dose (particularly gamma) from the fissile material. For the obvious reasons of radiation protection of personnel handling these materials, but also because of potential damage to electronic components.

1. The final document runs to 20 000 pages.

9.1 Critical mass

Values associated with each of these characteristics for common fissile materials are given in Table 9.1 (those that correspond to less common materials such as ^{237}Np are not considered here).

Table 9.1. Physical characteristics of the main fissile material vis-à-vis non-proliferation (ease of use for the manufacture of nuclear weapons)

Fissile material	Critical mass (kg)	Neutron emission (neut./s.kg)	Heat emission (W/kg)	Radiation dose
Pure ^{233}U	16	1.23	0.281	Depends on ^{232}U content (see the text)
Pure ^{235}U	48	0.364	0.00006	Very low
Plutonium PWR-type	13	470 000	20	Low or average
Plutonium "weapon grade"	11	60 000	2	Low

The critical mass of ^{233}U and plutonium are fairly similar, while that of ^{235}U is comparatively higher. Depending on the design, it takes between 5 and 15 kg of ^{233}U to make an atomic bomb, which is not very different from the required mass of plutonium. However, the most important difference between uranium (either ^{233}U or ^{235}U) and plutonium comes from the neutron emission, mainly due to the isotope ^{240}Pu . ^{240}Pu may be present significant proportions depending on the origin of the plutonium, depending upon the extent to which the fuel was irradiated (fuel burn-up). This explains why the spontaneous emission of neutrons is much more important for PWR-sourced plutonium than for "weapons grade" plutonium produced in fuels having a very short irradiation time. Such a phenomenon does not exist for ^{233}U , as it is almost always mixed with ^{234}U (in varying proportions depending on its origin), which is not a neutron source. Therefore, with ^{233}U , it is possible – due to this very low emission of neutrons – to produce a much simpler type of bomb than with plutonium. This simplification in the case of ^{233}U can be mitigated by the high-alpha activity of ^{232}U . Nuclear reactions of the type (alpha, n) may occur on light elements that may be present in trace amounts in the fissile material, which causes unwanted emission of neutrons. However, this process produces far fewer neutrons than the ^{240}Pu , and its effects can be minimised by reducing the levels of contamination in light elements. Finally, with regard to heat, it is quite low in all four cases, and this does not pose a major problem, even with plutonium, in which it appears much higher than uranium.

In summary, it is possible to make an atomic bomb with ^{233}U , although it contains a certain proportion of ^{234}U (which is typically 10 to 20% for conventional reactors).

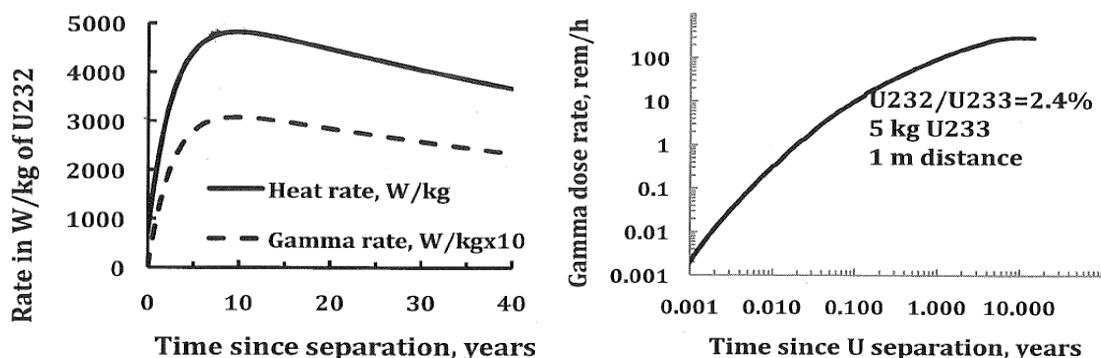
9.2 Self-protection from ^{232}U

The presence of ^{232}U in uranium (particularly in the case of thorium-based fuel cycles) is often cited as providing self-protection against proliferation. This is due primarily to the decay products of ^{232}U . The main source of gamma emission comes from the isotope ^{208}Tl which, upon decay, emits a 2.6 MeV gamma ray. The gamma fields from this material are very high after some decay time and require significant shielding and remote handling for any reprocessing activity. The degree of proliferation resistance this gamma field provides is dependent on the motives and the facilities available to the proliferators.

Higher radiation dose rate from materials requires either the financial capacity to build or otherwise acquire shielded facilities, or conversely accept large radiation dose to those handling or processing the materials. As a result, the proliferation resistance provided by the presence of ^{232}U in spent thorium-based fuels is heavily dependent upon the threat scenario. This underscores the limited usefulness of a simple quantitative assessment based on material characteristics and points to the need for a more comprehensive approach.

In general, the diversion and processing of spent nuclear power reactor fuel is not the preferred method to obtain material for a nuclear explosive device. It is likely that separated uranium from spent fuel would be used in a single explosive device rather than in creating a stockpiled arsenal of weapons. It is generally assumed that for thorium-based fuels, the uranium, after being separated, would be used without much delay, such that a substantial amount of ^{232}U will not have had significant time to decay. The presence of ^{232}U increases the dose rate (and energy release rate) particularly at times of approximately ten years after separation (depicted in Figure 9.1). It is the opinion of experts (Sleaford et al., 2010), that the dose rate from freshly separated uranium will likely be a nuisance rather than a deterrent to a determined adversary, who has gone to the trouble to divert and reprocess used nuclear fuel for his goal. In the interest of performing a conservative proliferation resistance assessment, it is recommended that any material attractiveness values (e.g. the figure of merit proposed in [Bathke et al., 2009]) be computed assuming no ^{232}U decay. Subsequently, calculations can be performed assuming elapsed time after separation in order to account for an increased radiation dose rate from the decay products of ^{232}U .

Figure 9.1. Gamma and energy release rates from ^{232}U *



* 1 rem = 0.01Sv.

Source: Moir (2010).

9.3 Safeguard considerations

From the standpoint of safeguards (i.e. the means to prevent unauthorised use of ^{233}U), the radiation emitted by the daughter products of ^{232}U become an asset to the extent that they assist in detecting ^{233}U that could be diverted for non-civilian purposes. Another way to reduce the risk of proliferation with the thorium cycle is to design systems in which the ^{233}U produced in the reactor is diluted “at source” by ^{238}U . This can easily be achieved by mixing thorium initially with natural or depleted uranium. However, this option would lead to increased production of plutonium and also raise proliferation concerns (because plutonium can easily be chemically separated). Another option would be the isotopic dilution of the ^{233}U at the stage of processing to thorium fuels, by blending it with natural or depleted uranium. Such an option would reduce much of the interest in ^{233}U recycling.

References

- Bathke, C.G., et al. (2009), "The attractiveness of materials in advanced nuclear fuel cycles for various proliferation and theft scenarios", *Proceedings of Global 2009*, Paris.
- INFCE (1980), *Advanced Fuel Cycle and Reactor Concepts*, Vol. 8, STI/PUB/534, International Nuclear Fuel Cycle Evaluation, International Atomic Energy Agency, Vienna.
- Moir, R. (2010), *²³²U Non-proliferation Features*, Lawrence Livermore National Laboratory, Report LLNL-TR-438648.
- Sleaford, B.W., et al. (2010), "Nuclear material attractiveness: An assessment of material from PHWR's in a closed thorium fuel cycle", *Proceedings of the European Nuclear Conference*, Barcelona.

10. Economic aspects of thorium fuel cycles

Given that the thorium cycle has not yet been deployed on an industrial scale, there is no accurate data on the costs associated with different stages of this cycle. The only evidence available on this subject comes from assessments based on limited experience from the manufacture of thorium fuel for different reactors. Nevertheless, it is possible to infer general trends by estimating costs relative to conventional uranium/plutonium cycles.

10.1 Fuel cycle part of the total cost

To measure the economic differences between the thorium/uranium and uranium/plutonium cycles, it is first of all important to distinguish the proportion of the overall cost of nuclear electricity that the fuel cycle (including the cost of the management of residual waste) represents. In general, this contribution is relatively minor, but not negligible. The estimates found in the literature on this subject are quite disparate, as they depend on many parameters, such as the types of fuel cycles considered, calculation methods used, assumptions made on the cost of goods and services and economic models applied in the calculations, especially in terms of discounting rates. Examples include a recent paper (National Nuclear Laboratory, 2012), in which the fuel cycle represents only 11.6% of the total cost per kWh and an article (Gras, 2010) that indicates a 25% share for the whole fuel cycle, with a 3/4 contribution for the front end and 1/4 for the back end. Meanwhile, the NEA, in its 2010 publication on the estimated cost of electricity, gives an average value of 16% for the 11 OECD countries considered in the study, and a discount rate of 5% (this share falls to 9.5% for a 10% discount rate). Nevertheless, the cost of the fuel cycle is on average about 15% to 20% of the cost per kWh, for nuclear electricity.

For a more accurate comparison between the thorium/uranium and the uranium/plutonium fuel cycles, it is necessary to know the cost breakdown between different stages of the nuclear fuel cycle. The most realistic option for the thorium cycle is a closed cycle in which the ^{233}U produced by thorium is recovered from the spent fuel through its reprocessing and then re-used to make new fuels. The French case, where a closed uranium/plutonium fuel cycle is implemented at a largest industrial scale, is taken as a reference. The following data have been published in an official report issued by the French government in 1997 (Ministère de l'économie et des finances, 1997):

Uranium:	24.6%
Uranium conversion:	3.3%
Uranium enrichment:	21.3%
Fuel fabrication:	16.4%
Interim storage and reprocessing of spent fuel and recycling:	26.2%
Final disposal of ultimate waste:	8.2%

Regarding raw material, the comparison of uranium with thorium cannot be based on a market price, since one does not exist. In fact, there is already a stock of about 25 000 tonnes of thorium in the world.¹ This would constitute a sustainable supply of fuel for several tens of nuclear power plants, before it became necessary to extract additional thorium. These stocks would likely be traded at very low prices, as they are mainly considered a storage burden for industry or organisations owning them. Beyond the exhaustion of current stocks of thorium, it would be necessary to seek new resources; but as thorium would be extracted together with other marketable materials (such as rare-earths, for example), the price would probably be much lower than that of uranium, especially as exploitable deposits are mostly opencast, which facilitates the recovery of minerals.

The enrichment step is not necessary for a thorium cycle, unless the fissile material used initially in this cycle is enriched uranium (LEU with up to 20% enrichment), which is not considered to be the best option. In such a case, it would be necessary to feed the cycle with significantly larger amounts of natural uranium and separative work units (SWU) than for the standard uranium cycle for the same energy output. The amount of this surplus largely depends on, for example, reactor type, burn-up and fuel management. For other thorium-based cycles using recycled fissile material (plutonium or ²³³U), the cost of these materials depends on the back end of the cycle, which will be examined later in this chapter after considering the manufacturing stage of fresh fuel.

10.1.1 Fuel fabrication

When considering the fabrication of thorium-based fuels, it is necessary to distinguish the type of fissile material associated with the thorium:

- If LEU is used as fissile material, two different kinds of nuclear materials (thorium and LEU) must be managed. This is expected to lead to additional costs compared to the manufacture of standard enriched uranium fuels. This is particularly the case for HTR fuel, where the fertile material (thorium) is packaged in coated particles that are different from those particles containing LEU.
- If plutonium is used, processes should not be very different from those currently used in the manufacture of MOX fuel; therefore, the costs should be comparable.
- If the fissile material is ²³³U, the presence of the daughter products of ²³²U will require shielded operations. This would certainly generate significant additional costs, which are difficult to estimate, since there is no detailed study available on this subject and even less experience on an industrial scale. The only assumption that can help is based on the ratio between the cost for the production of MOX fuel (operations that are mostly performed semi-automatically in gloveboxes) and standard uranium fuel. This ratio is usually quoted to be between three and five, to which should be added another factor to take account of the need for remote operation for the fabrication of Th/²³³U fuels. As this remote operation would extend to fuel finishing and storage, this factor will be considerable.

10.1.2 Reprocessing

The reprocessing step of thorium fuels (whatever the fissile material mixed with it) can be compared to that of uranium fuel, except for the chemical part of the process, which is significantly more complicated for thorium-based fuels than for uranium-based fuels, particularly for the dissolution of materials. Again, it is not possible to provide estimates

1. Of which 9 400 tonnes are recorded in France by ANDRA in its national inventory of waste and radioactive materials released in 2012.

for the additional costs that may result from these complications of the process; but, it should be noted that the chemical part is less than one-third of the overall cost of treatment. The rest of the facilities of a reprocessing plant should not present major differences between the two types of fuel to be reprocessed, notably:

- Before the fuel dissolution, i.e. reception of spent fuel containers, unloading of spent fuels, interim storage in pools, mechanical dismantling or shearing.
- after the central part of the chemical treatment, i.e. processing and packaging of recyclable materials once chemically separated, storage of fission product, vitrification of the ultimate waste (the bulk of fission products and MAs), packaging of other radioactive waste (fuel structure in particular), treatment of secondary aqueous effluents, treatment of solid and gaseous process waste, intermediate storage facilities for all waste as well as all ancillary facilities and common infrastructure of the site.

10.1.3 Disposal

The final step in the back end of a closed nuclear cycle is the final disposal of medium and high-level long-lived waste. For this, the solution adopted almost universally is that of a deep geological repository. There is no reason to suggest a significant difference between uranium and thorium cycles in this final stage, especially as it is assumed here that only residual waste from reprocessing are disposed of after separation of all recyclable materials.

References

- Gras, J.M. (2010), “Le cycle du combustible nucléaire”, *Les techniques de l'ingénieur- Génie Nucléaire*.
- Ministère de l'économie et des finances (1997), *Les coûts de référence de la production électrique*, DGEMP.
- National Nuclear Laboratory (2012), *Nuclear Fuel Cycle Science and Engineering*, Woodhead publishing, Cambridge, p.114.

11. Conclusions

If the use of nuclear energy is to expand and become a sustainable source of energy, the major challenges of improving the utilisation of mineral resources while reducing ultimate waste streams will need to be addressed. In a post-Fukushima context, where deployment of fast neutron reactors is uncertain and the realisation of geological repositories has been delayed in some countries, continuing socio-political concerns are focused on the accumulation of spent fuel and its final disposal. In the absence of fast neutron reactors, the issue of plutonium management will have to be dealt with in the medium to long term, at least for current and future separated plutonium.

The use of thorium in the nuclear fuel cycle as a complement to the uranium/plutonium cycle shows potential for improving the medium-term flexibility of nuclear energy and its long-term sustainability. More specifically, options for thorium's introduction into the nuclear fuel cycle should be kept open and continue to be investigated, including:

- using thorium as a means of burning plutonium (and possibly other higher actinides) as an option for plutonium management;
- the possibility of reaching higher conversion factors in thermal or epithermal neutron spectra, using thorium-based fuels, in evolutionary generation III+ systems, with the aim of recycling the fissile material from used fuels;
- examining the promising physicochemical characteristics of thorium dioxide, which would offer improved performance of thoria-based fuels over current fuel designs.

The development of new fuels or new reactor concepts is a time- and resource-consuming process likely to span several decades. Any industrial application of thorium as a nuclear fuel would continue to require the input of fissile material from the existing uranium/plutonium cycle until the required amounts of ^{233}U could be produced to ultimately make the thorium cycle self-sustaining.

If a thorium fuel cycle is pursued, an important factor governing the rate at which ^{233}U could be produced from the introduction of thorium/plutonium or thorium/uranium/plutonium cycles would be plutonium availability. The limitations imposed by fissile plutonium availability result in rather long transition periods between thorium/plutonium and thorium/ ^{233}U systems, which are likely to be of the order of many decades.

The development of a fully self-sustaining thorium/ ^{233}U cycle would also require the development of industrial scale reprocessing capabilities to recover ^{233}U from spent fuel, along with fuel fabrication facilities to prepare the material for re-use. In particular, impediments to closing the thorium fuel cycle arise from the following issues:

- To date, the THOREX process has been demonstrated in pilot-plant facilities, but is yet to reach the maturity of the commercial PUREX process. Other extractants and alternative processes (e.g. fluoride volatility) are also being investigated, but are still at a conceptual stage.
- A major challenge associated with the recycling of ^{233}U is the presence of radioactive ^{232}U . Remotely operated and fully shielded recycled fuel fabrication processes will be

required, for which there are currently no proven equipment or processes at the industrial scale.

- A related challenge is the handling and storage of excess thorium, which will contain ^{228}Th and its highly radioactive daughters for about 20 years.

Ultimately, thorium technologies require significant further development. Thorium fuel R&D initiatives are currently being funded by some countries concerned with long-term nuclear energy sustainability (as is the case of Canada). However, given their cost and the lack of clear economic incentives for nuclear power plant operators to pursue this route, industrial development activities for thorium remain somewhat limited at present.

If thorium fuel cycles are pursued, it is to be expected that short- to medium-term development of thorium fuels would be carried out in a step-wise fashion and in synergy with the existing uranium/plutonium fuel cycle.

In the longer term, the potential introduction of advanced reactor systems may present an opportunity to realise the full benefits of a closed thorium/ ^{233}U fuel cycle in dedicated breeder reactors (generation IV or beyond) that are presently in the design study phase.

Molten salt reactors in particular may offer the prospect of using thorium fuels with online recovery and re-use of the ^{233}U while recycling long-lived actinides and ensuring minimal losses to the final waste stream. It must, however, be recognised that the development, licensing and construction of such novel systems is a long-term undertaking.

Annex A: Thorium recovery

A.1 Introduction

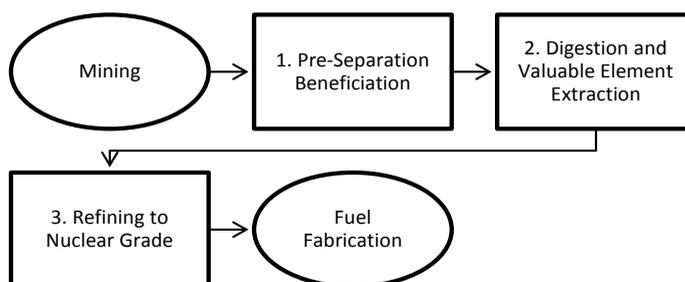
This section is primarily concerned with the steps needed to recover thorium, beginning at its major mineral forms and proceeding to nuclear-grade purity suitable for fuel fabrication.

If recovered, thorium will most likely initially be a by-product (or co-product, depending on cost sharing analysis) of ilmenite (titanium ore) mining, uranium mining, iron ore mining, and/or rare earth ore mining. Beach sand mining requires less waste rock removal, ore crushing and sizing energy than the other options, but the chemical processes used to isolate thorium are relatively similar between cases. Since bastnasite and monazite are both actively recovered minerals, even with thorium's currently small market, they will be the primary focus of this assessment.

Both bastnasite and monazite are almost always obtained in conjunction with other minerals and must be isolated before being processed further. The steps used to achieve this isolation are similar for both minerals. The initial step involves crushing the raw ore. Then, gravitational, flotation, electromagnetic, and/or electrostatic methods or, more often, a combination of these techniques, are used to achieve mineral purities greater than 90%. The exact sequence depends on the mix of minerals encountered in the ore deposit, the primary products desired and any co-products or by-products desired. Once bastnasite or monazite is isolated from the other minerals, the particle size must be further reduced for the subsequent acidic or caustic attack (known as digestion) to be effective. The impacts of these steps are attributable to producing the primary product (e.g. REEs, titanium) and not to thorium production.

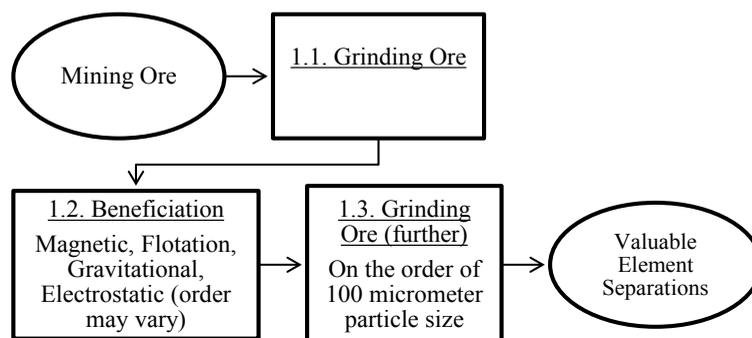
To obtain a reactor-grade thorium product, thorium-bearing ores that may contain multiple valuable elements are subjected to multiple processing steps (see Figure A.1). First, the thorium-bearing minerals must be isolated from other ore constituents, a step which is typically accomplished by mechanical, electrostatic, and magnetic processes. Second, these minerals are attacked by aggressive chemicals to dissolve the valuable elements and other constituents. The valuable elements are separated from the other constituents and each other using chemical processes such as solvent extraction or ion exchange. Finally, the separated thorium stream is purified to yield high-purity reactor-grade thorium suitable for use in nuclear fuel. Each of these steps is described in more detail in the following sections.

Figure A.1. Overview of thorium recovery process



A.2 Pre-separation beneficiation

Figure A.2. Pre-separation beneficiation processes to concentrate minerals of interest



A.2.1 Initial grinding

The minerals are ground to facilitate the subsequent separation of different minerals. The particle size must be sufficiently small to be compatible with typical heavy metal concentrators, which separate the desired heavy minerals (e.g. ilmenite, zircon, rutile, monazite, bastnasite) from lighter undesired minerals, like quartz.

A.2.2 Beneficiation of bastnasite or monazite from other minerals

Bastnasite beneficiation schemes have been developed as a primary product (e.g. Mountain Pass, Nevada, United States) and as a secondary product of iron ore production (e.g. Bayan Obo, Mongolia). Even with its rare earth content, monazite is rarely the primarily sought mineral where it is produced. In the case of beach sand mining, ilmenite is often the primary product, followed by zircon and rutile. At least some monazite is generally present at all major bastnasite deposits; however, the reverse is not necessarily true.

A.2.2.1 Bastnasite beneficiation

▪ A.2.2.1.1 Magnetic separation

At Bayan Obo, the presence of highly magnetic iron ores, like magnetite and hematite, make electromagnetic separations a logical first step to remove them after particle sizing. Bastnasite's low magnetic attraction makes this step a very clean and effective separation.

A.2.2.1.2 Flotation

The number of flotation steps that attract unwanted content into a surface layer that can be skimmed depends on the content of the raw ore, but unwanted gangue minerals like calcite and quartz are generally removed by exploiting bastnasite's hydrophilicity (attraction to water). Surfactants such as fatty acids or dicarboxylic acids are added to enhance the attraction of unwanted minerals to the surface layer.

▪ A.2.2.1.3 Gravitational separation

Bastnasite is mostly isolated after flotation, but monazite has very similar hydrophilic characteristics to bastnasite and must be separated from the bastnasite. Due to monazite's exceptionally high specific gravity, density-based separations are effective. Fairly high-purity streams of both minerals are produced as a result of this process. In principle, subsequent processing of monazite could be viable if the REE content in the ore

is sufficiently high. However, monazite's considerably higher specific radioactivity relative to bastnasite is often a deterrent to subsequent monazite processing.

A.2.2.2 Monazite beneficiation

▪ A.2.2.2.1 Gravitational separation

With a specific gravity often in excess of five, monazite is usually among the densest, if not the most dense, mineral obtained during monazite mining operations. As a result, gravity-based separations are effective in isolating the mineral.

▪ A.2.2.2.2 Magnetic separation

Ilmenite (frequently the primary ore of interest where monazite is found) is highly magnetic and can be readily removed from the monazite. Monazite has a rather moderate susceptibility to magnetism and remains with the less magnetic minerals.

▪ A.2.2.2.3 Electrostatic separation

Unlike most of its accompanying minerals, monazite has a low electrostatic attraction. Thus, most remaining minerals are removed by this means, and very pure monazite can be obtained. The order of the monazite separation steps may vary by facility, based on the predominance of the various materials; for instance, electrostatic separations may precede the magnetic separations.

A.2.3 Further size reduction of particles

For subsequent chemical processing to be effective, the particle size must be reduced to the order of a hundred microns, by grinding and/or crushing. In some cases, the initial crushing may have been sufficient, and this step would be unnecessary.

A.3 Digestion and valuable element extraction processes

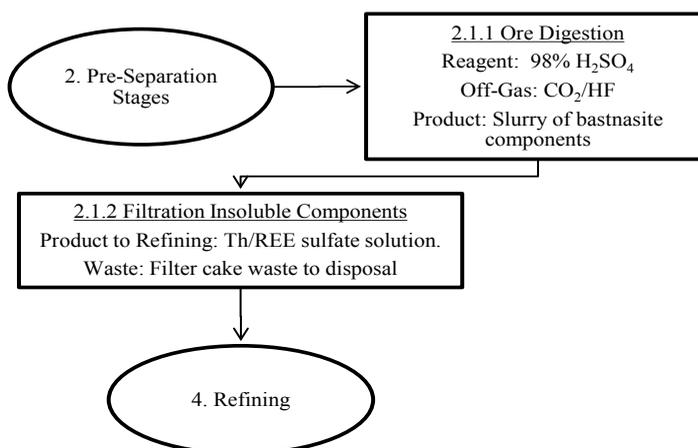
Once the purified thorium mineral has been sized to an appropriate range, it must be solubilised to achieve REE and thorium recovery. Both bastnasite and monazite have very refractory crystal structures, which require a highly concentrated, potent chemical agent to open the lattice and extract the valuable content. Either a strong acid or a base can be used for this, and variations exist within both routes. In the case of bastnasite, the prospect of readily removing fluoride and carbonate anions make the acidic process preferable. Both variants have been used extensively for monazite. Neither the acid nor the alkali digestion process is selective for thorium extraction, so further separation is necessary.

This section of the report addresses bastnasite and monazite processing up to the point where solids containing significant concentrations of thorium are recovered by filtration. These solids can undergo subsequent refining to recover a nuclear-grade thorium product. In bastnasite processing, the REEs remain with the thorium until the refining stages. In monazite processing, most of the rare earths are separated from the thorium prior to refining. This is because thorium is a much more notable impurity in monazite (>5%) than in bastnasite (<0.5%), and more care is taken to keep this thorium out of the REE refining systems.

The following sections describe acidic processes for recovering thorium from bastnasite and monazite and a process using strong bases for recovering thorium from monazite.

A.3.1 Acidic bastnasite digestion processes

Figure A.3. Sulphuric acid digestion process for extraction of thorium from bastnasite



A.3.1.1 Ore digestion

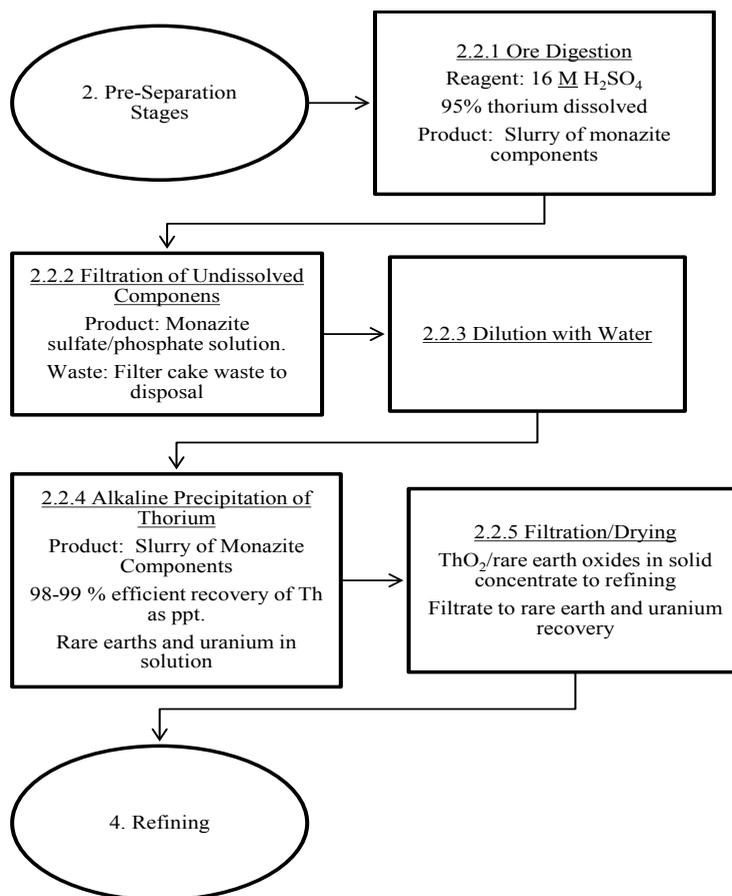
The initial strong acid attack accomplishes two main objectives. The first is to break down bastnasite's tightly bound mineral structure and release its content to an aqueous state for subsequent processing. The second is that bastnasite is a carbonate mineral with substantial fluoride content. The addition of a strong acid converts these into carbon dioxide and hydrogen fluoride gas, respectively, which is liberated from the system. While the efficiency of this separation makes the acidic process convenient, potential corrosion and safety concerns arise from the release of hydrogen fluoride. Sulphuric acid is frequently identified as the acid of choice, though hydrochloric acid has also been used.

A.3.1.2 Filtration of non-extracted components

Any ore components which remain undissolved after the digestion process must be removed from the system. Since thorium and the REEs are in the aqueous phase of the slurry resulting from digestion at this point, a solid-liquid separation is required. The solids resulting from the separation (assumed to be filtration here) are disposed of as waste. The thorium/REE solution is then sent to thorium refining.

A.3.2 Monazite acid digestion processes

Figure A.4. Sulphuric acid digestion process for extraction of thorium from monazite



A.3.2.1 Ore digestion

In this step, a strong acid is used to break monazite's sturdy mineral structure and dissolve the desired components into the aqueous phase. Monazite particles are added to a solution of concentrated strong acid. A solution consisting of 93% sulphuric acid can, with sufficient digestion time, achieve 95% recovery of monazite particles up to 150 μm . This puts most of the contents of the monazite in a soluble sulphate form. Nitric acid or hydrochloric acid could also be used, resulting in nitrates and chlorides, respectively. At this degree of acidification, virtually all of the phosphate ions present in monazite become phosphoric acid.

A.3.2.2 Filtration of non-extracted components

The purpose of this step is to remove undissolved monazite constituents and other solid ore components such as silica and zircon. This is a physical, rather than a chemical, stage. The digested solution is typically diluted and allowed sufficient time to permit the solids from the aqueous monazite to settle. Some sources indicate that a flocculating agent may be added in this step to speed settling of the solids.

A.3.2.3 Dilution

The thorium-bearing sulphate-phosphate solution is diluted with water by a factor of 6-7 in preparation for subsequent steps.

A.3.2.4 Precipitation of thorium concentrate

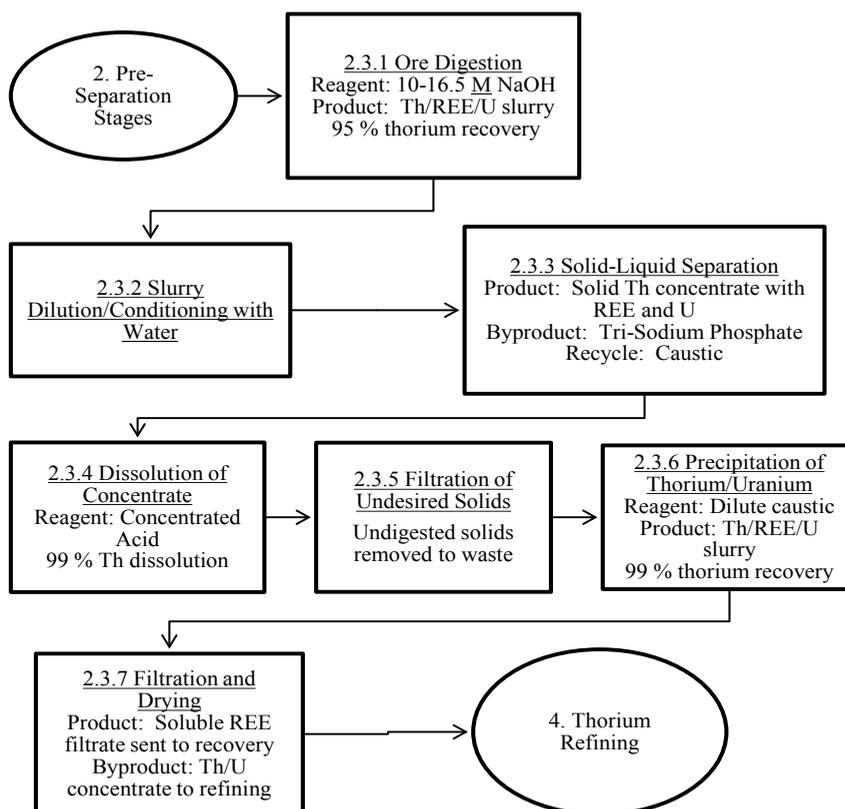
Depending on the process, more than one precipitation reaction could be used. The goal here is to add an agent that results in the formation of a thorium-rich precipitate. Ammonium hydroxide can be used to lower the pH and shift the phosphoric acid solution back to phosphate ions, resulting in a thorium phosphate precipitate. Another option is adding sodium hydroxide in sufficiently large amounts, which shifts the thorium, REEs, and uranium to a solid hydroxide form.

A.3.2.5 Filtration of thorium concentrates

Once the thorium has been precipitated, physical processes can again be applied to separate the aqueous product containing most of the REEs from the solid phase which contains the thorium. This is not a perfect separation and rare earths can constitute up to 30% of the concentrate. However, subsequent refining to nuclear grade removes these impurities.

A.3.3 Caustic monazite digestion processes

Figure A.5. Caustic (alkali) digestion process for extraction of thorium from monazite



A.3.3.1 Ore digestion

Much like the acidic version, a strong caustic chemical is used to break monazite's mineral structure and dissolve components to an aqueous phase. Monazite particles are dissolved in a solution of concentrated strong base, usually sodium (assumed here) or potassium hydroxide; monazite particles at 44 µm have been extracted with 45% sodium hydroxide, and with sufficient digestion time can extract 95% of monazite's thorium content, i.e. roughly the same as the acidic process. In contrast to the acidic process, this version results in the thorium, uranium, and REEs being present in the solid phase as hydroxides and oxides, while the phosphates from the monazite end up in the aqueous phase as sodium or potassium phosphate. Separating the phosphates at this early stage is one of the major advantages of the caustic process, since it simplifies subsequent precipitation schemes. It also enables the phosphate content to be recovered as a by-product such as tri-sodium phosphate.

A.3.3.2 Slurry dilution/conditioning

The hydroxide slurry is diluted with water to reduce the sodium hydroxide concentration to about 30%. Sufficient time (about 1 hour) at 110 degrees Celsius allows the hydroxides to form a precipitate that is suitable for subsequent solid-liquid separation.

A.3.3.3 Solid-liquid separation

Physical processes such as filtration are used to separate the aqueous phosphate solution from the solid thorium/uranium/REE hydroxide concentrate. The phosphate solution may be subsequently refined as a by-product.

A.3.3.4 Dissolution of the concentrate

The hydroxide concentrate is dissolved in a strong acid. Some processes involve the addition of concentrated hydrochloric acid, while others use sulphuric acid. Sufficient time is allowed to dissolve at least 99% of the hydroxides.

A.3.3.5 Filtration of undesired solids

Physical processes such as filtration are used to separate the thorium/uranium/REE solution from undesired solids which are managed as waste. This step is analogous to step A.2.2.2 for the acid process.

A.3.3.6 Precipitation of thorium/uranium

This step is analogous to A.2.2.4 of the acid separation process. Generally, dilute sodium hydroxide is added to precipitate thorium and uranium as hydroxides. This method has the limitation that extra measures must be taken to remove the uranium later in the refining process. However, washing the precipitate with a mixture of sodium carbonate, sodium hydroxide and hydrogen peroxide can remove as much as 80% of this uranium.

A.3.3.7 Filtration and drying

Filtration separates the thorium-rich solid phase from the REE-rich aqueous phase. The solid phase, which still contains residual uranium and REE hydroxides, is dried and prepared for the refining process.

A.4 Refining thorium concentrates to nuclear-grade material

Depending on the mining scenario, all processes described up to this point may already occur in efforts to recover REEs. Even when obtained as a by-product of iron ore, bastnasite's REE content is almost always recovered, meaning that beneficiation and extraction impacts would not be attributable to nuclear applications. The same is not necessarily the case for monazite, which is generally a by-product of ilmenite. Though the impacts of physical beneficiation would still be covered, many ilmenite operations simply discard monazite-bearing material as waste.

The impacts of the steps described in this section would generally be attributable to the nuclear fuel cycle, since their primary purpose is to produce a purified thorium product for nuclear applications from a material that would otherwise be managed as a waste. Since schemes to produce a thorium product from bastnasite have not been formally considered, the processes discussed are based on experiences with bastnasite that remove thorium as a waste. In contrast, thorium has been refined from monazite by a number of entities around in the world and on a commercial scale in India.

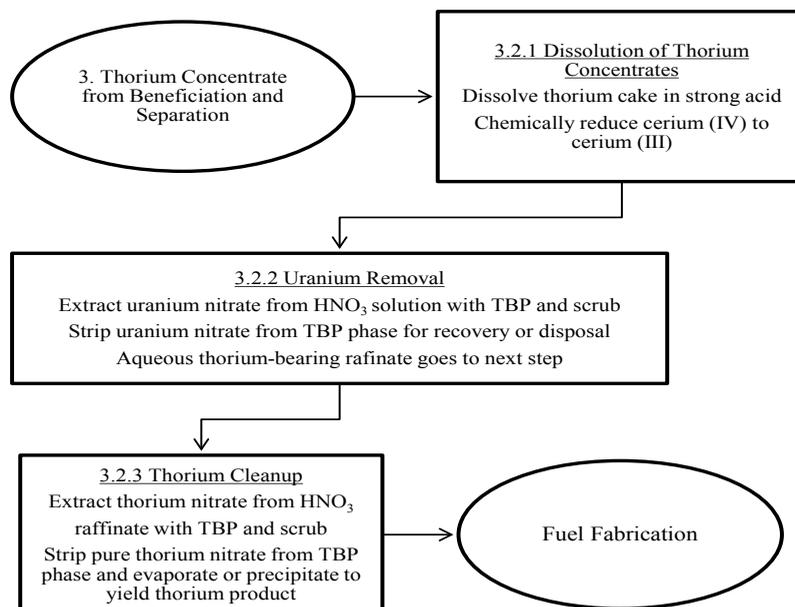
A.4.1 Solvent extraction following bastnasite processing

The concentration of thorium in bastnasite is low enough that it is often neglected in mass flow diagrams altogether, as subsequent separation of individual REEs is generally sufficient to remove most thorium impurities. However, a few Chinese laboratories have developed processes for removing the impurities in REEs simultaneously via solvent extraction. Not all of the documentation concerning these processes has been translated, but those that are translated provide a basis for the thorium by-product recovery methods discussed below.

The incoming stream to this solvent extraction system would likely be a REE/thorium sulphate solution (see Section A.3.1). Generally, the uranium content of bastnasite is negligible and would not impact the requirements of the solvent extraction system. A primary amine, N-1923, can be used to recover well over 99% of the thorium from bastnasite feed, although it is also accompanied by a nontrivial amount of cerium. Other designs using different amines or esters have been developed, employing a sequence of solvent extraction steps to recover bastnasite's cerium, fluorine, and thorium content. In these latter cases, the objective was to remove undesired impurities, and the resultant purity of the thorium streams is not clear.

A.4.2 Solvent extraction following monazite processing

Figure A.6. Refining monazite thorium to nuclear grade



A.4.2.1 Dissolution of thorium concentrates and cerium reduction

This step achieves two objectives simultaneously. The first is to dissolve the thorium concentrate with a strong acid. As thorium nitrate has historically been the preferred compound for storage and use and TBP-nitrate systems are well-known, nitric acid is the most likely candidate. Furthermore, cerium, a rare earth present in significant quantities in monazite, has much of the same chemical behaviour as thorium in the tetravalent state. Since the only other available valence states for cerium are less than four, a reducing agent must be used. Hydrogen peroxide is the typical choice.

A.4.2.2 Uranium removal

Uranium mostly follows thorium up to this point in processing and must be separated from the final thorium product. The extent and nature of this step depends on two factors. First, the separation method used (see Section A.2) directly impacts how much uranium is present in the refining steps. The acidic digestion process removes considerably more uranium than the caustic process, so feed from the caustic process might require a greater number of uranium extraction stages. Second, the solvent choice could potentially eliminate the need for this step entirely. The solvent with the most nuclear-related experience is TBP. Its familiarity and availability are strong advantages, but it is more selective for uranium than thorium, thus necessitating a series of stages that keeps most of the thorium in the raffinate.

Research with other solvent choices like esters and amines is ongoing and should a viable solvent be found that is selective for thorium, the need for these stages could be eliminated. Recent experiments suggest that some amines can be configured to be slightly selective for thorium. However, a dual extraction scheme in which uranium is first separated and then thorium is purified is still generally used.

A.4.2.3 Thorium clean-up

Once the uranium has been removed, a series of solvent extraction and scrubbing stages is used to obtain a thorium-rich product stream containing only trace amounts of other elements. The choice of solvent should be selective for thorium versus rare earths and other elements. TBP is generally used for this purpose, since U levels are negligible after the first solvent extraction system. The extraction steps are used in conjunction with scrubbers that return rare earth impurities to the aqueous phase.

A number of solvent/extractant systems have been proposed as alternatives. One study explored the use of aliquat 336, a long-chain amine, in kerosene. This species achieved 96% extraction for thorium, 93% for cerium (this experiment did not pre-reduce the cerium), 53% for uranium, and much less for rare earths in a single stage. Other promising amines include primene JM-T and primene ⁸¹R, which have been shown to achieve over 90% extraction for thorium while less than 10% for uranium.

Once purified, the thorium in the product stream must be removed from the organic phase. A strong acid is usually used for this purpose. If thorium nitrate is the desired product, nitric acid is the ideal choice. However, in recent times, thorium oxalate may be a more desirable product since it is safer to store and transport. Subsequent treatment would depend on the fuel type. For instance, for oxide fuels, calcination of thorium nitrate/oxalate to thorium oxide would be required prior to fuel fabrication.

A.5 Summary of predicted health impacts

A.5.1 Introduction

The following section intends to briefly identify the categories of health impacts that might result from the various stages of a thorium refining facility. This list is not intended to be comprehensive but rather a starting point for considerations in future efforts to evaluate health impacts.

A.5.2 Projected health impacts for pre-separation beneficiation

These impacts are expected to be absorbed by extant mining industries unless a new mine were opened for the primary purpose of obtaining thorium, which is unlikely. The grinding and crushing of ores would release ^{220}Rn , the analogue of the U-decay-chain's ^{222}Rn . ^{220}Rn has a much shorter half-life than ^{222}Rn (56 seconds vs. 3.8 days), so it would only pose a hazard to those in the immediate vicinity. The radon hazard could be heightened as bastnasite or monazite is concentrated from other, less radioactive ores. This is more of a concern in the case of monazite, since its thorium content is much higher than that of bastnasite.

A.5.3 Projected health impacts for digestion and separation processes

The use of concentrated strong acids and strong bases represents a significant chemical hazard regardless of processing scheme. The digestion process also produces radioactive waste streams. In any scheme, undigested solids with a moderate specific activity must be disposed of. The bulk of the radioactivity is carried by ^{228}Ra , which chemically tends to follow the REEs. This results in a waste stream with a high specific activity during REE processing, but this would not be attributable to the nuclear fuel cycle. In the case of the caustic monazite process, another low-activity waste stream would result from refining phosphate into a usable by-product. As in previous stages, radon inhalation would continue to be a hazard. As discussed at the outset of Section A.4, some of these impacts would likely be attributable to primary product (Ti, REE, iron, etc.) production while the incremental impacts would be attributable to thorium production with the allocation depending on the specific flowsheet being used.

Another waste from recovery of thorium or uranium is the undigested solid tailings that contain around 5% of the thorium or uranium that was in the feed material, plus some of the decay products from the thorium or uranium. In the case of uranium, the activity and toxicity of the tailings decline slowly because the rate-controlling radionuclide is ^{226}Ra which has a 1 600-year half-life. Beyond 10 000 years, the activity and radiotoxicity of uranium tailings have dropped by about 20-fold (for 5% residual uranium) and then remain constant for a time controlled by the 4.5-billion-year half-life of ^{238}U (Li et al., 2004). However, in the case of thorium tailings, the rate-controlling radionuclide is ^{228}Ra , with a half-life of only 5.6 years. As a result, the activity and radiotoxicity drop by 20-fold within a century and then remain constant for a time controlled by the 14-billion-year half-life of ^{232}Th . The long-term activity and toxicity of thorium tailings per tonne of thorium recovered is about 25% of that for uranium tailings from each tonne of uranium recovered.

A.5.4 Projected health impacts for refining

In addition to the presence of strong acids and bases, extractants for solvent extraction systems are typically toxic and exposure would need to be limited. Any raffinate streams leaving the system would represent radioactive waste, albeit of a low specific activity. As in previous stages, radon inhalation would continue to be a hazard.

References

- Li, D. et al. (2004), "Separation of thorium (IV) and extracting rare earth from sulphuric and phosphoric acid solutions by solvent extraction method", *Journal of Alloys and Compounds*, Vol. 374, Issues 1-2, July 2004, pp.431-433.

Annex B: A transition scenario study of light water reactors to thorium-fuelled heavy water reactors

Scenario studies have been performed by AECL, Canada, to investigate future fuel cycles in the global context. These fuel cycle scenarios envision changes in fuel types (e.g. transition to thorium-based fuels), and, eventually, reactor types that could be employed globally to generate electricity. Each of these scenarios start with the current LEU-fuelled LWRs that are currently operating around the world and transition to new fuel/reactor types (in particular, HWRs), when the current fleet is decommissioned. These scenarios span the period of the present day up to the year 2200.

Transitions from a once-through, LEU fuel cycle with LWRs to mixed thorium/uranium fuel cycles are analysed and developed based on various fuel cycle evaluation criteria such as uranium utilisation and used fuel characteristics. Fresh and used fuel compositions, fuel residence time in the reactor and fuel burn-up for each fuel type are derived from reactor physics calculations. These data were then used in a fuel cycle simulation to compute various fuel cycle evaluation criteria such as natural uranium mined, and used fuel characteristics such as gamma and neutron emissions, and heat rate.

B.1 Scenario descriptions

The scenario studies presented here span from the beginning of 2010 to the end of 2199. It is assumed that some LWRs in the initial LWR fleet will be retired and replaced with HWRs beginning in the year 2030. Retirement of the initial fleet of LWRs will proceed linearly until the year 2060, at which time all LWRs are expected to be decommissioned. For all scenarios studied, the yearly demand for nuclear electricity was taken from the world scenario in *Transition towards a Sustainable Nuclear Fuel Cycle* (NEA, 2013). The reprocessing of used UO_2 and thoria fuel is limited to 100 and 200 kt/year, respectively, and begins in the years 2020 and 2035, respectively.

The following parameters (Stillman, 2004) are assumed for the LWRs:

- burn-up = 51 GWthd/tHM;
- fresh fuel ^{235}U enrichment = 4.3%;
- nominal electrical power = 0.92 GWe;
- thermal efficiency = 34%;
- load factor = 0.9;
- fuel residence time = 5 years;
- reactor lifetime = 40 years;
- construction and licensing time = 7 years.

The following parameters are assumed in the scenarios for the HWRs:

- reactor power = 0.725 GWe;
- thermal efficiency = 33%;

- load factor = 0.9;
- reactor lifetime = 30 years;
- construction and licensing time = 7 years;
- fuel composition/residence time: variable depending on type.

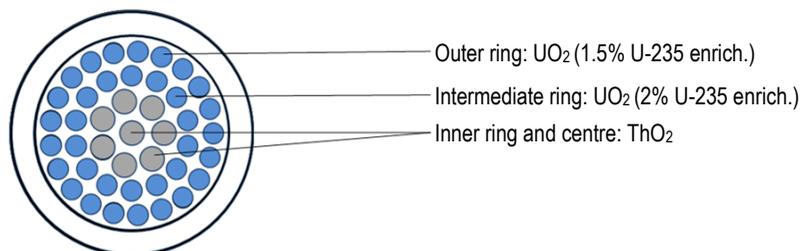
B.1.2 Fuel types

The following are descriptions of the three fuel bundle concepts used in the HWR simulations. All three are 43-element fuel bundle concepts similar to those described in Section 5.2 of this report.

B.1.2.1 LEU+Th fuel

The LEU+Th fuel is made up of two inner rings of ThO₂, and two outer rings of UO₂ with enrichments of 1.5% and 2% ²³⁵U in the outer and intermediate rings respectively. This bundle is shown in Figure B.1. This fuel achieves a burn-up of 23 GWthd/tHM, with an associated residence time of 730 days.

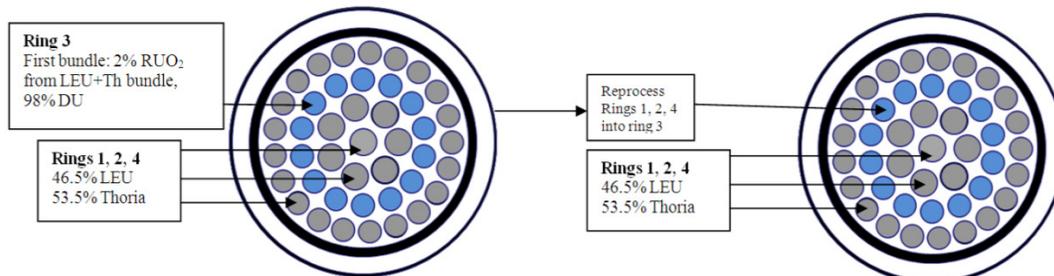
Figure B.1. LEU+Th fuel bundle concept



B.1.2.2 RU-233+LEU+Th fuel

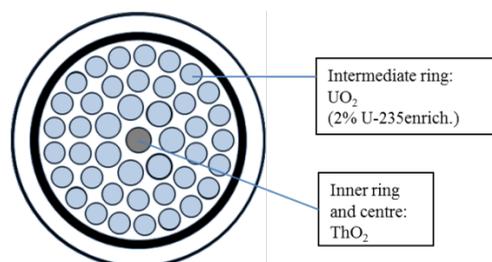
In this concept, fuel in rings 1, 2, and 4 contain a mixture of 46.5 wt% low-enriched (2% ²³⁵U/U) uranium dioxide and 53.5 wt% thoria (ThO₂). Fuel in ring 3 contains UO₂ only, with the uranium being recycled from previously discharged RU-233+LEU+Th fuel. For the initial fuel bundles, the recycled uranium in ring three is to come from the recycled LEU+Th fuel. It is mixed with depleted uranium as 2% RUO₂, 98% depleted UO₂ in order to achieve the desired burn-up of ~20 MWd/kgHE.

Figure B.2. RU-233+LEU+Th fuel bundle



B.1.2.3 RU-233+Pu+Th fuel

This fuel bundle, shown in Figure B.3, also uses recycled uranium from the LEU+Th fuel bundles, mixed homogeneously with PuO₂ (reactor grade Pu, assumed to come from reprocessed LWR fuel) and ThO₂. The centre pin is a mixture of ZrO₂ and Dy₂O₃. This central absorber is used to reduce the magnitude of the coolant void reactivity.

Figure B.3. RU-233+Pu+Th fuel bundle

This fuel achieves a burn-up of 21 GWthd/tHM, with a residence time in reactor of two years. The recycled uranium in this case is assumed to come from Pu+Th fuel bundles. The geometry of the Pu+Th bundle is identical, with fuel composition 3.3% PuO₂ and 96.7% ThO₂ in the three outer rings, and an absorbing centre rod. The discharge burn-up is 22.5 GWthd/tHM, with a residence time in reactor of two years.

B.2 Results

The results of four scenarios are presented here:

- “LWR”, the reference case, corresponds to a once-through fuel cycle utilising LEU fuelled LWRs throughout the duration of the scenario.
- LWR → LEU+Th scenario: in the LWR → LEU+Th scenario, 100% of nuclear power is generated by LEU fuelled LWRs until the year 2030, after which any retiring LWRs are replaced by LEU+Th fuelled HWRs. All used fuel is sent to wet storage for five years, after which it is placed in dry storage for the remainder of the scenario.
- LWR → RU-233+LEU+Th scenario: in the LWR → RU-233+LEU+Th scenario, 100% of nuclear power is generated by LEU fuelled LWRs until the year 2030, after which any retiring LWRs will be replaced by HWRs that are fuelled with RU-233+LEU+Th if there is sufficient RU-233 available to do so. Otherwise, the HWRs will be fuelled with LEU+Th. After the year 2030, any additional reactors that are built are HWRs. The LWR used fuel is sent to wet storage for five years, after which it is placed in dry storage for the remainder of the scenario. Used LEU+Th and RU-233+LEU+Th fuel are both sent to wet storage for five years, after which they are reprocessed for RU-233.
- LWR → RU-233+Pu+Th scenario: in the LWR → RU-233+Pu+Th scenario, 100% of nuclear power is generated by LEU LWRs until the year 2030, after which any retiring LWRs will be replaced by HWRs that are fuelled with RU-233+Pu+Th. If there is insufficient RU-233, then the HWRs will be fuelled with Pu+Th to breed more. After the year 2030, any additional reactors that are built are either RU-233+Pu+Th fuelled HWRs or LEU-fuelled LWRs, depending on the availability of reprocessed Pu.

B.2.1 VISION model

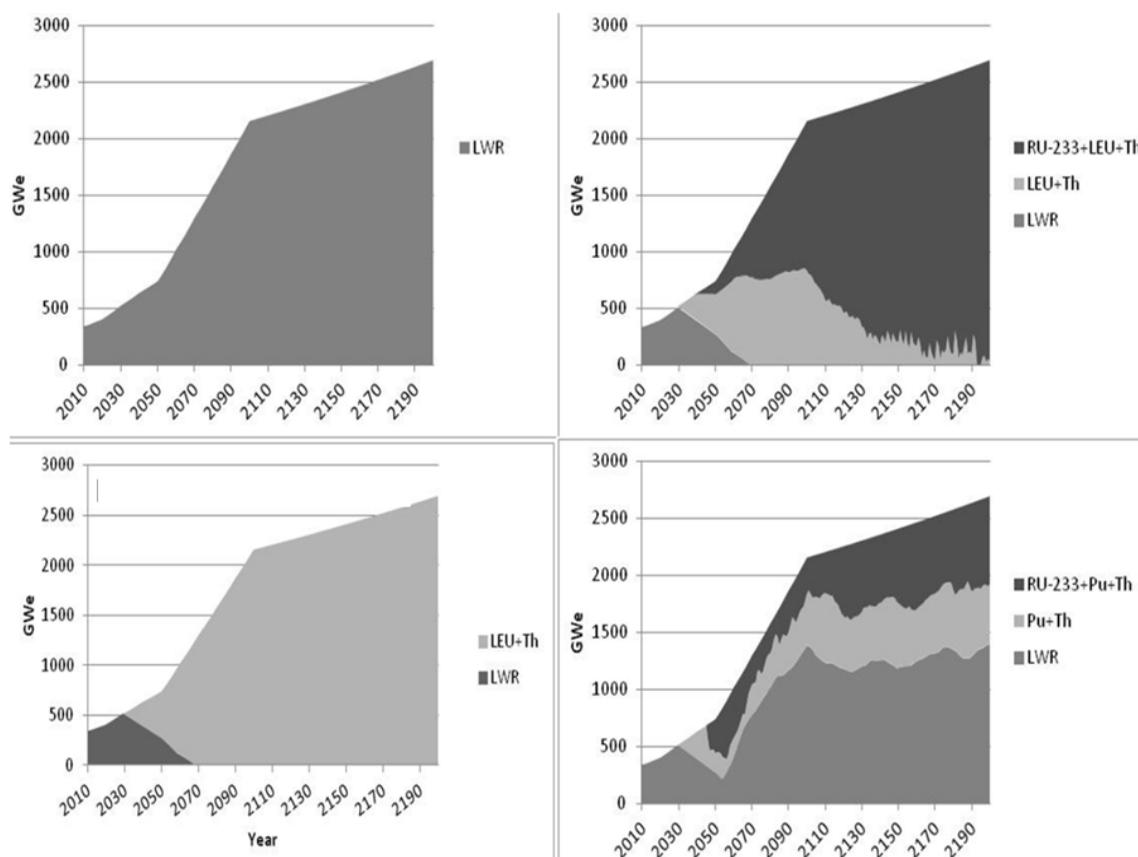
The scenarios described above were implemented using the VISION model version 4 (Jacobson et al., 2009). It models the interaction of the various components of the fuel cycle, including fuel fabrication, nuclear power plants, used fuel storage, used fuel reprocessing, and long-term disposal. The scenarios were simulated to compare their fuel consumption, used fuel, and proliferation resistance characteristics. A start-up time of ten years was added to the beginning of each simulation to allow enough time for reactors to be built to satisfy increased demands for electricity early in the scenario.

B.2.2 Reactor fleet

The scenarios are represented graphically by plotting the power generated from each reactor/fuel type over time, as is shown in Figure B.4.

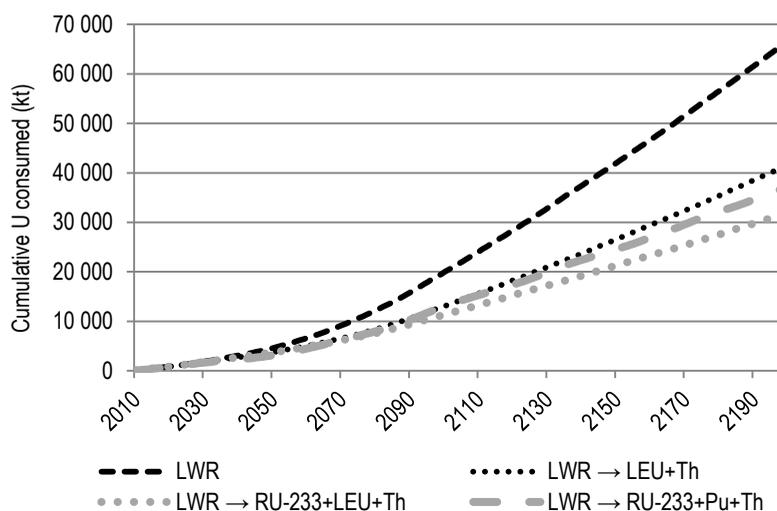
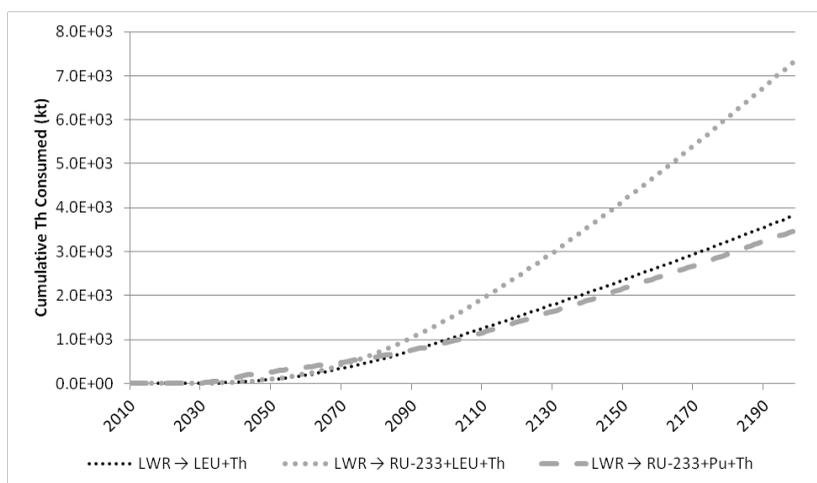
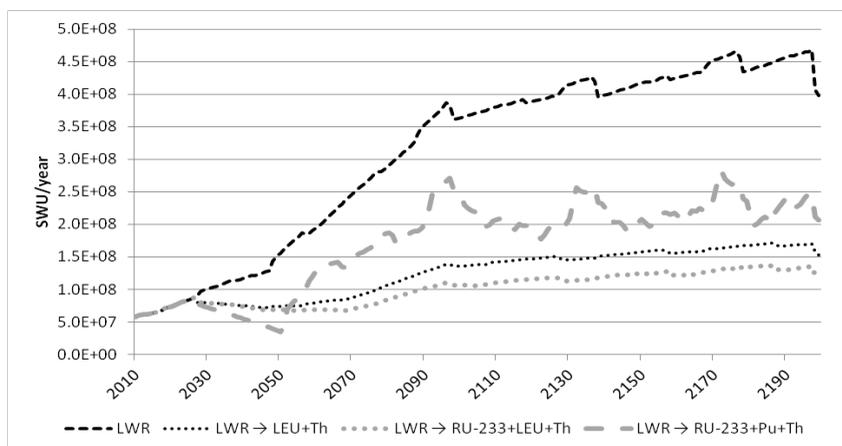
In the “LWR → RU-233+LEU+Th” scenario, approximately 74% of the total electricity was generated from ^{233}U +LEU+Th fuel, with brief periods where 100% of the electricity was generated from this fuel. In the “LWR → RU-233+Pu+Th” scenario, approximately 20% and 24% of the total electricity was generated from Pu+Th and RU-233+Pu+Th fuel, respectively.

Figure B.4. Power generated in the LWR reference case and three different scenarios considered



B.2.3 Natural resource consumption

The cumulative uranium and thorium consumption are shown in Figures B.5 and B.6, respectively. The LWR→LEU+Th, LWR→RU-233+LEU+Th, and LWR→RU-233+Pu+Th scenarios consume 37%, 52%, and 45% less natural uranium, respectively, than the LWR scenario. The thorium consumption in the scenarios is always less than the natural uranium consumption and is shown in Figure B.5. Introducing any of these thorium-based fuel options into the fuel cycle affects the enrichment requirements, as shown in Figure B.7.

Figure B.5. The cumulative natural uranium consumption in each scenario**Figure B.6. Cumulative thorium consumption in each scenario in which thorium fuel is used****Figure B.7. Uranium enrichment separative work unit requirements per year**

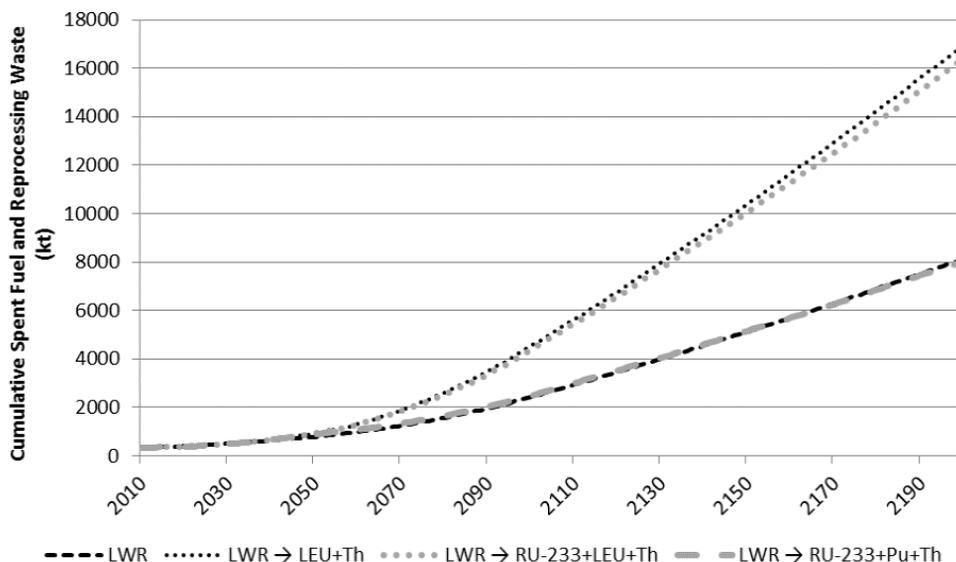
B.2.4 Used fuel characteristics

This section compares the used fuel and waste products resulting from each scenario. The total mass, gamma and neutron emissions and long-term decay power of used fuel and waste produced in each scenario are compared. Gamma and neutron emissions from used fuel have implications for water radiolysis in used fuel storage, on shielding requirements for used fuel and on worker dose (and shielding requirements) for handling, transportation, storage and disposal facilities and possible reprocessing facilities. Decay power on a mass basis has more impact on handling and storage facilities. Decay power per unit energy has more impact on sizing disposal facilities (disposal cost per unit energy).

B.2.4.1 Mass of used fuel

The mass of used fuel, including reprocessing waste, is shown in Figure B.8. The transition to thorium-fuelled HWRs resulted in increased used fuel mass due to the decrease in burn-up for all but the LWR → RU-233+Pu+Th scenario, which resulted in approximately the same mass of used fuel.

Figure B.8. Used fuel in wet and dry storage and reprocessing waste in each scenario



B.2.4.2 Decay power of used fuel

The long-term decay power of the cumulative used fuel and high-level waste generated during the 190-year scenarios is shown in Figure B.9. The ratio of the decay power in the thorium scenarios to the LWR scenario is shown in Figure B.10. All but one of the thorium fuel cycle scenarios resulted in increased decay power of used fuel over the first ten years after discharge from the reactor, the exception being the LWR→LEU+Th scenario. At 10 000 years the used fuel in the plutonium-based thorium fuel scenarios both have lower decay power than the LWR scenario, whereas the LEU-based thorium fuel scenarios have higher decay power. The higher decay power of used fuel at 10 000 years in the LEU-based thorium fuel scenarios is due to the decay products of ^{239}Pu , ^{240}Pu , and ^{233}U , the quantity of which is higher at the end of these scenarios. At 100 000 years the decay power of used fuel in all of the thorium fuel scenarios is over three times higher than the LWR scenario, which is due to the dominance of the neptunium decay series (^{233}U) in that time frame (Bhatti et al., 2013).

Figure B.9. Long-term decay power of all (cumulated) used fuel and reprocessing waste at the end of each 190-year scenario

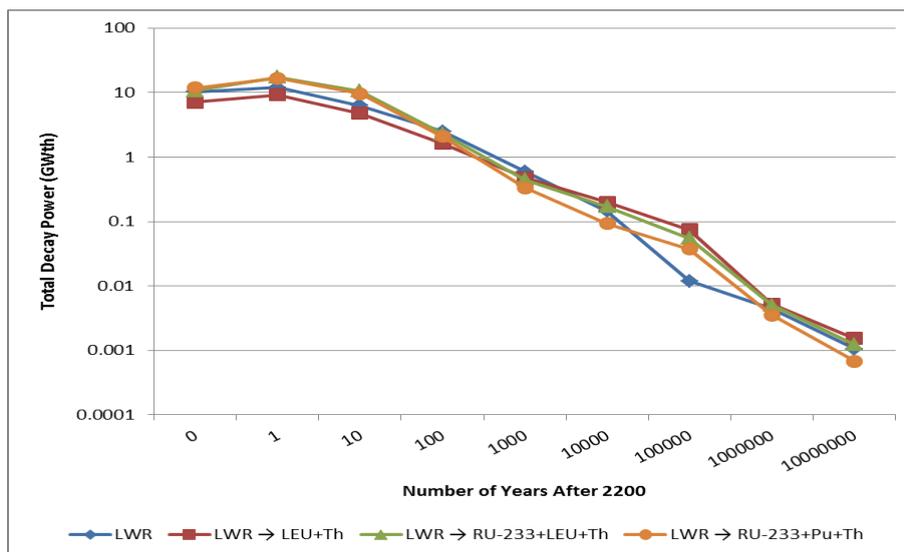
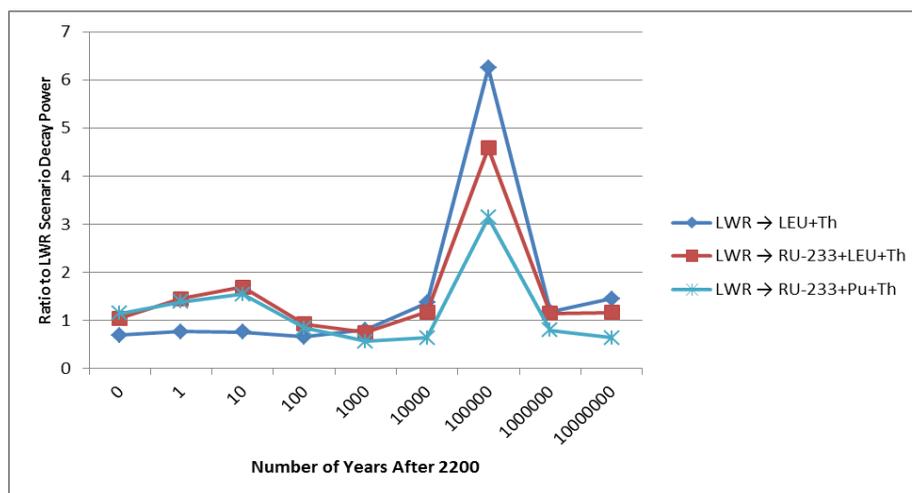


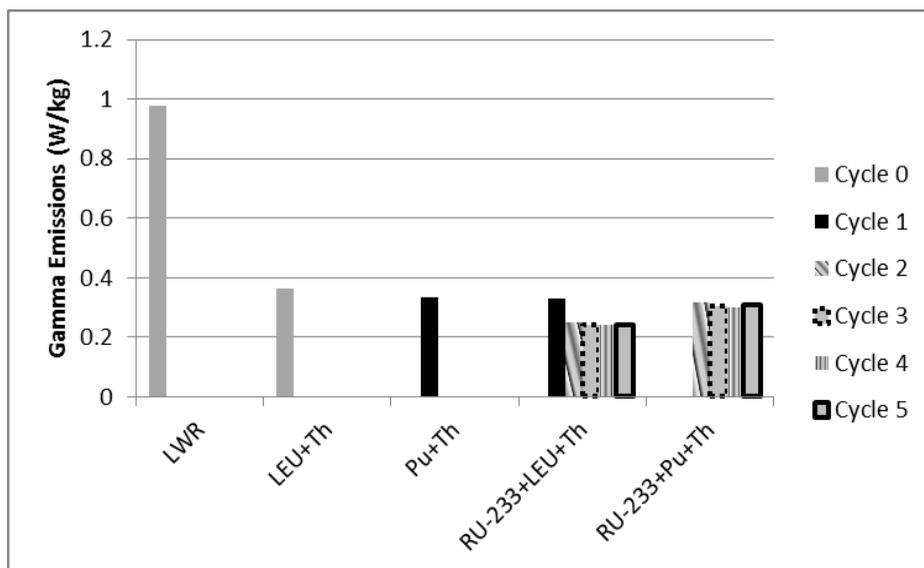
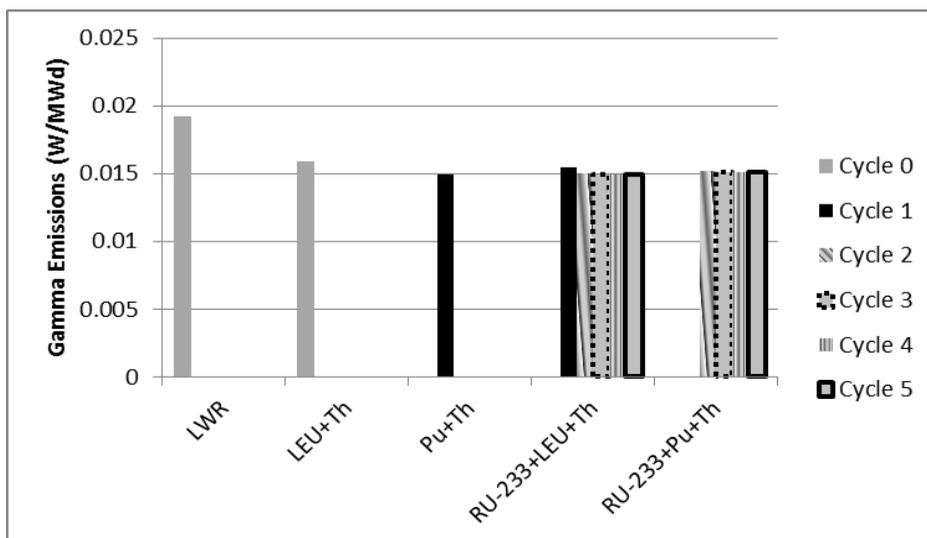
Figure B.10. Ratio of the long-term decay power of all used fuel and reprocessing waste at the end of each thorium fuel scenario to the LWR scenario



B.2.4.3 Gamma emissions of used fuel

An important factor when determining safety requirements for handling fuel after it has been discharged and stored for five years is the gamma radiation produced.

The gamma emissions per mass of spent fuel and per energy produced, after five years of cooling are shown in Figures B.11 and B.12, respectively. All of the thorium-based fuels have less than half of the gamma emission per mass than that from LWR fuel. However, the gamma emissions per energy produced for the thorium fuels are within 20% of that from LWR fuel. It should be noted that the gamma emissions of used LEU-based thorium fuel in wet storage are higher than that of the LWR used fuel, whereas the gamma emissions per energy produced after five years storage are lower. This is because the gamma emissions of used LEU-based thorium fuel at discharge are higher than the LWR used fuel. The gamma emissions of used LEU-based thorium fuels then decrease more rapidly than LWR used fuel prior to leaving wet storage.

Figure B.11. Gamma emissions per mass of used fuel after discharge and five years' decay**Figure B.12. Ratio of gamma emissions to thermal energy produced in used fuel after five years' decay**

References

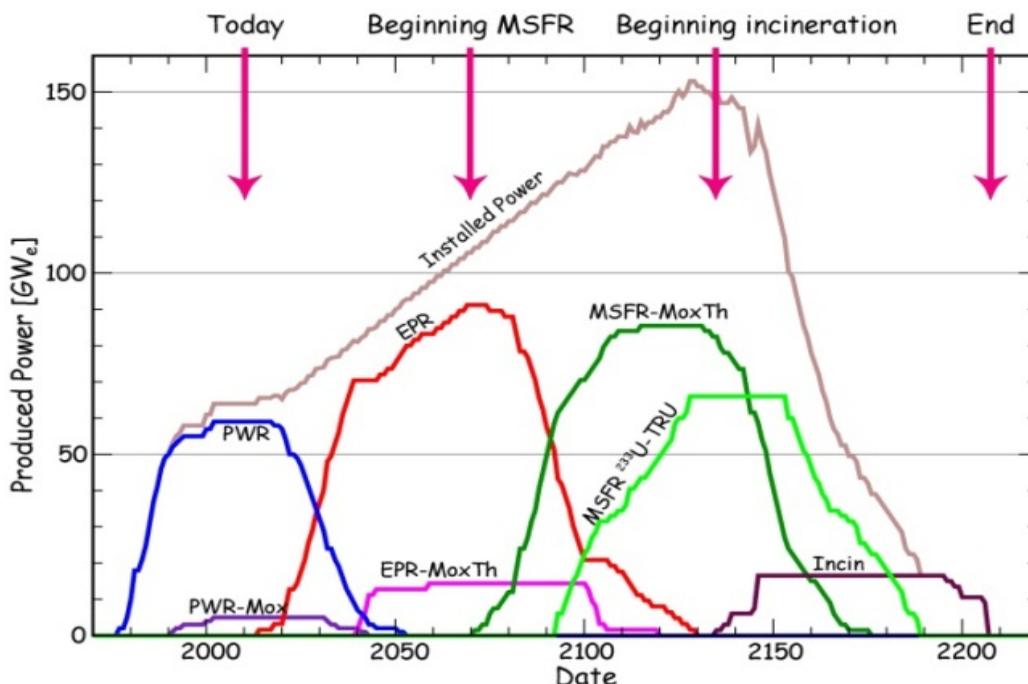
- NEA (2013), *Transition towards a Sustainable Nuclear Fuel Cycle*, OECD, Paris.
- Stillman, J.A. (2004), "Homogeneous Recycling Strategies in LWRs for Plutonium, Neptunium, and Americium Management", ANL-AFCI-124, Argonne National Laboratory.
- Jacobson, J.J., et al. (2009), "VISION: Verifiable fuel cycle simulation model", *Proc. of Global 2009*, pp. 2517-2527, Paris.
- Bhatti, Z., B. Hyland and G.W.R Edwards (2013), "Minor actinide transmutation in thorium and uranium matrices in heavy water moderated reactors", *Proc. of Global 2013*, Salt Lake City.

Annex C: A transition scenario study of light water reactors to molten salt fast reactors

Given the absence of naturally available ^{233}U , a standing question is whether a park of MSFRs could ever be deployed at national, European or worldwide scales. In the exercise below, the deployment and end-of-game capacities of MSFRs has been studied for the French case (Heuer, 2014). The exercise studies a doubling of the produced total installed nuclear power as a test of the deployment capacities of such reactors. This study has led to an estimation of the amount of heavy nuclei produced by such deployment and evaluated the complexity of the management of these heavy nuclei stockpiles as well as their radiotoxicities.

The exercise scenario displayed in Figure C.1 assumes that generation IV reactors are deployed after 2070 only. The deployment scenario starts with generation III light water reactors (EPRs) being deployed. By 2040, some generation III reactors are fuelled with Pu-UOX in a thorium matrix, both to reduce MA production and to prepare a transition to a thorium fuel cycle in MSFRs. Generation III reactors are then progressively replaced with MSFRs fuelled with Th-Pu MOX coming from generation III reactors. The deployment is finally completed with MSFRs directly started with a mix of ^{233}U produced in the existing MSFRs and the remaining stockpiles of plutonium irradiated in LWRs.

Figure C.1. Nuclear power deployment exercise based on PWRs, EPRs and MSFRs for a scenario which doubles the installed French nuclear capacity



The scenario ends with the introduction of burners with a view to optimise the end-of-game and further reducing the final TRU inventories after MSFR shutdown. Note that the end-of-game situation would not be different if it occurred after hundreds of years of operation; it depends only on the installed power.

The evolution of the radioactive element stockpiles other than the fission products during the scenario is shown in Figure C.2. The final stockpiles (Figure C.2) that will have to be managed as the scenario ends are the following:

- depleted uranium at 0.1%: 803 700 tonnes;
- uranium from reprocessing (minimised by the scenario management): 3 250 tonnes;
- irradiated thorium: 5 100 tonnes;
- irradiated UOX fuel: 5 tonnes of Pu standing for 450 tonnes of irradiated UOX (represented by its Pu content labelled “Pu-UOX” in Figure C.2);
- irradiated MOX fuel: 0.76 tonnes standing for 12.4 tonnes of irradiated MOX (represented by its Pu content labelled “Pu+MA MOX” in Figure C.2);
- MAs separated from the Pu when the latter is used as MOX fuel in LWRs and vitrified: 612 tonnes (labelled “MA from UOX”);
- final burner inventories: 106 tonnes.

The evolution of the radiotoxicity corresponding to the final radioactive stockpiles of this scenario, including the fission products, is displayed in Figure C.3 where it appears that the short-term radiotoxicity (a few dozen years) is dominated by the fission products, while the long-term radiotoxicity (103 to 106 years) is mainly due to the vitrified MAs produced in LWRs and not re-used in MOX fuel.

Figure C.2. Evolution of the actinide stockpiles during the scenario considered

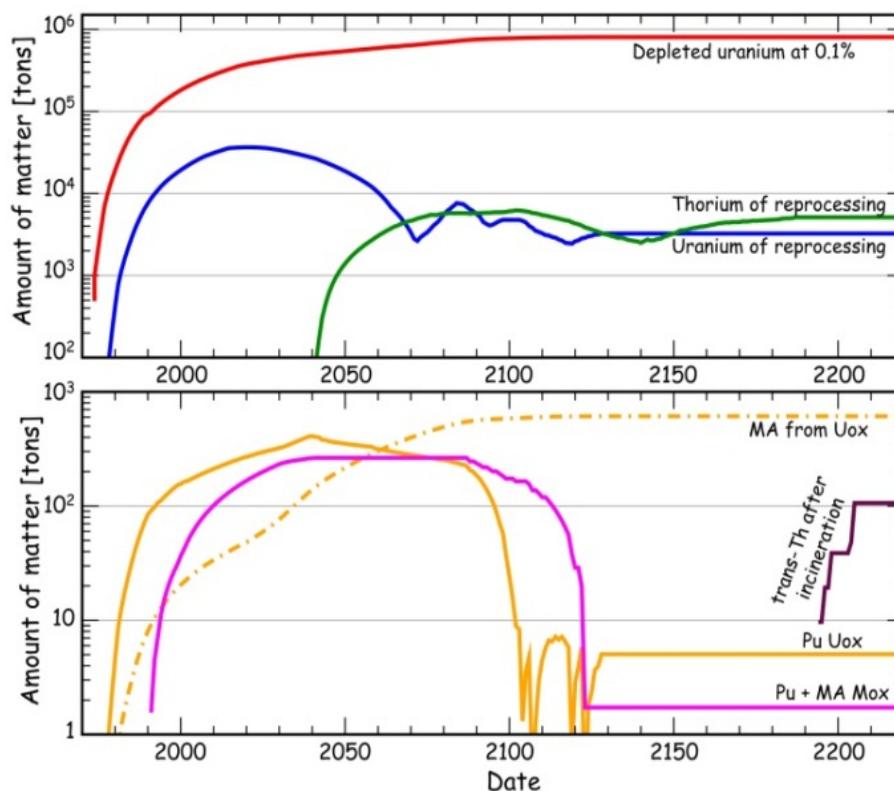
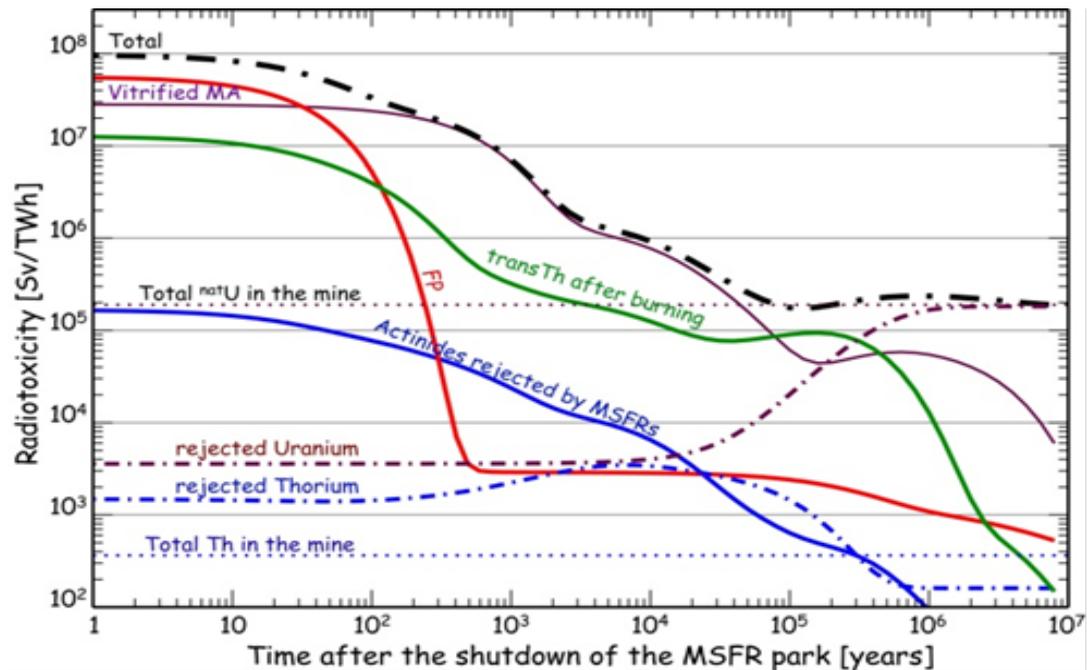


Figure C.3. Time evolution of the various contributions to the radiotoxicity of the final radioactive stockpiles in the French transition exercise towards MSFRs



References

Heuer, D., et al. (2014), "Towards the thorium fuel cycle with molten salt fast reactors", *Annals of Nuclear Energy*, Vol. 64, pp. 421-429.



Introduction of Thorium in the Nuclear Fuel Cycle

Since the beginning of the nuclear era, significant scientific attention has been given to thorium's potential as a nuclear fuel. Although the thorium fuel cycle has never been fully developed, the opportunities and challenges that might arise from the use of thorium in the nuclear fuel cycle are still being studied in many countries and in the context of diverse international programmes around the world. This report provides a scientific assessment of thorium's potential role in nuclear energy both in the short to longer term, addressing diverse options, potential drivers and current impediments to be considered if thorium fuel cycles are to be pursued.