Minimization of actinide waste by multirecycling of thoriated fuels in an EPR

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Abstract

This master's thesis explores how to minimize the long-lived actinide waste that is produced in nuclear power plants by performing simulations of thoriated nuclear fuels in existing reactor designs.

An European pressurized water reactor (EPR) assembly fueled with a mixture of thorium and highly enriched uranium (20% and 90% ^{235}U) was simulated. The spent thoriated fuel is less active, and for a much shorter period of time, than uranium or uranium/plutonium fuels and less decay heat is generated from the waste. Nuclear waste from the thorium cycle can therefore be stored in much smaller repositories than conventional fuels, providing an economical as well as an environmental gain. Also, there will be a substantial net production of fissile ^{233}U, and this makes the multi-recycling of uranium possible; hence significantly lowering the costs of fresh enriched uranium to blend with the recycled fuel.

Acknowledgements

Ever since I first saw the terrifying and amazing pictures of the atomic bombs exploding I have been extremely fascinated about nuclear physics, and the power deep inside the nucleus. After the course "Physics and energy resources" I knew that I wanted to do my masters project on nuclear reactor physics. Unfortunately the University of Oslo did not teach reactor physics, so I was sort of on my own.

Luckily I met Sunniva Siem, who became my supervisor, and she understood what I really wanted. She arranged for me to go to Paris where I worked with my other supervisor, Jon Wilson. First and foremost I must thank both of you: Sunniva and Jon! You have both been all you could wish for in a supervisor, and then some more. I look at you both as close friends after these two years that have gone by since I started out as a fresh master's student. Both supervising me not only in the process of doing my masters project, but in life.

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Acronyms and abbrivations

ADS – accelerator driven system. Subcritical reactor dependent on supply of neutrons from an accelerator

BOC – beginning of cycle. Namely, the nuclear fuel cycle

BWR – boiling water reactor

CANDU – CANada Deuterium Uranium; a Canadian PHWR

CR – conversion ratio

EOC – end of cycle. Namely, the nuclear fuel cycle

EPR – European pressurized water reactor. A 3rd generation PWR; the reactor studied in this project

GCR – gas cooled reactor

HEU – highly enriched uranium. Uranium with fissile content of 20% ^{235}U or higher

HLW – high level waste

IPCC – Intergovernmental Panel on Climate Change. Leading body for assessment of climate change, to provide the world with scientific view on the state of climate change and its consequences

LEU – low-enriched uranium

LMFBR – liquid metal fast breeder reactor

LOCA – loss-of-coolant-accident

LOFA – loss-of-flow-accident

LWR – light water reactor

MCNP – Monte Carlo N-particle

MOX – mixed oxides. Fuel made of plutonium and uranium

MSR – molten salt reactor

MURE – MCNP utilities for reactor evolution. Program used for simulations in this thesis

OTC – once through cycle. Fuel is disposed of as waste after irradiation

PWR – pressurized water reactor

PHWR – pressurized heavy water reactor

RBMK – graphite moderated water cooled reactor

SWU – separative work units; enrichment work

Th/HEU – fuel made of a mixture of thorium and highly enriched uranium;

that is thorium and 20% enriched uranium and thorium and 90% enriched uranium

TMSBR – thorium molten salt breeder reactor

TRU – transuranium element

UOX – uranium oxide. Conventional nuclear fuel used in most reactors world wide

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1. Introduction

The growth of energy consumption and human induced climate change will be the major challenges facing mankind this century.

The world's energy consumption has risen almost exponentially for the last 50 years, and a strong growth is still expected in the future, especially from the two highest populated countries in the world, China and India, with approximately 2.5 billion people. At present, coal is the worlds most widely used source for electrical power generation, and it is also the most carbon-intensive. A typical 1 GW coal-fired power plant running for one year will emit 8.7 million tons of CO_2. The growth of energy consumption will therefore result in a large increase in the CO_2 emissions, which, according to the Intergovernmental Panel on Climate Change[1] (IPCC), will increase the global temperature and provoke a climate crisis which will severely impact human populations. A 1GW nuclear power plant, on the other hand, releases negligible quantities of CO_2, and this is one of the reasons why there today is a renewed interest in the expansion of nuclear power for energy generation. Other reasons are its economic viability, and potential for energy security and independence.

Electricity generation from nuclear power is actually projected to increase by at least 50% from 2005 to 2030 world wide, as concerns about rising fossil fuel prices, energy security, and greenhouse gas emissions support building more nuclear plants [1]. There will therefore be a large build out of nuclear power, and as developing countries rapidly increase their installed nuclear capacity easily extractable uranium resources will become depleted. The overall result will likely be much higher uranium price over the course of this century and a legacy of large quantities of nuclear waste [2]. With no reprocessing of the spent fuel, the currently known world uranium resources could be exhausted before the middle of this century.

Thorium fuels may be the answer.

Thorium, element number 90, is a naturally-occurring, slightly radioactive material discovered in 1828 by the Swedish chemist Jons Jacob Berzelius, who named it after

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[1] The IPCC is the leading body for the assessment of climate change, established by the United Nations Environmental Programme (UNEP) and the World Meteorogical Organization (WMO) to provide the world with a

Thor, the Norse god of thunder. It is found in small amounts in most rocks and soils, and the average concentration is about 7.2 ppm, which is about 2 to 4 times more abundant than uranium. Thorium slowly oxides in air, becoming grey and eventually black. Thorium oxide (ThO_2), also called thoria, has one of the highest melting points (3300°C) of all oxides. Although not fissile itself, like uranium or plutonium, thorium may easily be transformed into the fissile uranium isotope ^{233}U by neutron absorption. The main advantage of using thorium as a nuclear fuel is that virtually no transuranic elements (TRUs) such as plutonium, americium, curium, etc are produced, leading to dramatically lower quantities of nuclear waste, as opposed to e.g. conventional uranium fuel (UOX).

There has been great interest in thorium as a fuel for nuclear energy in the past, and in the 1960s and 70s it was shown that thorium could be used practically in any type of existing reactors [3]. However, it was not clear that the thorium fuel cycle could compete economically with the more well-known uranium cycle, and when the uranium prices fell, thorium lost its appeal. Most projects using thorium in their fuel cycles were therefore terminated by the 1980s. Today, almost all of the activity on thorium as a nuclear energy source is found in India where two of their nuclear reactors both are loaded with 500 kg of thorium blanket. The reason for this is that India is dependent on nuclear power to generate electricity, and they have the largest deposits of thorium in the world. They have not signed the Non-Proliferation Treaty (NPT), an act that has restricted India's supply of uranium.

The work presented is very relevant to the Norwegian situation, where huge reserves of thorium are present, alongside Europe's "biggest battery" – the Norwegian hydropower. Norway enjoys cheap and clean hydropower and has developed this resource extensively. Almost all of the electricity is produced from hydro, but there are occasional imports from Europe, and this power is produced from coal. According to the US Geological Survey (USGS 2007), Norway also has one of the major thorium resources in the world. The thorium enriched minerals are mostly found in 3 regions: the Fen Complex in Telemark County, the Permian Oslo Province, and on the Southeast coast of Norway, in the Kragerø and Langesund area. The Fen Complex is considered to be the most promising resource. However, exploration specifically for thorium has never been undertaken, and therefore knowledge of the grade and associated volumes is scarce[2]. The quoted USGS 2007 weight estimates of the thorium resources date from the 1950s-1960s and are uncertain.

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[2] Knowledge of Norwegian thorium enriched minerals and their grades are mainly based on results from uranium exploration. The thorium levels were estimated from analyses of uranium in mineral samples, the correlation between uranium and thorium, and from helicopter and ground gamma surveys.

Conventional nuclear power (with thoriated fuels) could provide the Norwegian people with their base load of electricity, freeing up flexible hydro power to be exported at peak prices, when energy demand and prices on the rest of the European electricity grid is at its highest.

Norway has two research rectors, but does not currently have commercial nuclear power, and there has been a great public opposition to nuclear energy. However, lately there has been a thorium debate in Norway; the Norwegian government appointed a committee to investigate the possibilities of using Norwegian thorium as a nuclear fuel [3]. Thorium, and even nuclear power, got a lot of positive media attention. Furthermore, outside of Norway research into the thorium option is re-starting; Areva announced 24 July this year that they plan to investigate the use of thorium in their pressurized water reactor (the same reactor type already studied in this project) [4].

The accelerator driven system (ADS) is an innovative concept designed with the thorium cycle in mind, where a sub-critical $Th/^{233}U$ core is bombarded by neutrons generated by an external spallation source. Another reactor concept for thorium fuels is the Thorium Molten Salt Breeder Reactor (TMSBR), a critical reactor with a liquid molten salt fuel in a graphite moderated core. However, the construction of a working industrial-scale reactor for either of these concepts is likely to be far into the future (near the end of this century), and thus too late to make much impact on the climate change problem which requires strong action now. These reactors are based on fission, like all of today's operating reactors, but there is also the concept of fusion; the way the sun produces its energy. Fusion is the opposite process of fission; in a fusion reaction two light nuclei, like hydrogen, are fused together, forming a heavier element and in the process releasing energy. The nuclear waste problem for a fusion reactor is smaller than with conventional fission reactors, and there is a great interest in developing a fusion reactor that will have a net production of energy. However, this technology also requires decades of development.

This thesis is about existing reactor concepts, and focuses on what is achievable with current reactor designs. It may be possible to utilize the uranium resources much better with the existing technology by mixing uranium and thorium. There is also the possibility of reduced activity of long lived waste production and less likelihood of nuclear proliferation. Because of the production of ^{233}U from thoriated fuels, the uranium resources can last longer if the fuel is multi-recycled. The dismantling of the nuclear weapon arsenal after the end of the cold war has created large excess stocks of highly enriched uranium (HEU) from the obsolete weapons. Some of these stacks have already been neutralized by down blending it into low enriched uranium (LEU); already

a total of 367 tons of bomb-grade HEU, equivalent to 14´686 nuclear warheads have been eliminated[3] in the Megatons to Megawatts[TM] Program[4] [5]. The program, however, deenriches the uranium, but in terms of waste production, using HEU mixed with thorium is a much better option than down-blending it and using it as conventional UOX fuel. These currently existing stockpiles of HEU (mostly in the US and the former Soviet Union), can be destroyed – without losing its potential energy content – by using it as a neutron source in a reactor with thorium fuel. In this way *the HEU would be neutralized as weapons material, and at the same time its potential energy content would be fully extracted.*

Both chapters 2 and 3 deal with background theory for the project. First, in chapter 2 the basic nuclear physics with respect to the phenomenon of fission is explained. A brief history of fission is given, and the basic theory of fission and neutronics; neutron multiplication and moderation of neutrons, is provided. Then, in chapter 3 the reactor physics is explained; the differences and similarities between the different types of reactors. The pressurized water reactor is of particular interest, since this is the type of reactor studied in this project. Finally, the chapter deals with reactor safety, an important element in reactor physics. The delayed neutrons make it possible to control the fission reaction, and reactivity feedback makes the reactor passively safe.

In chapter 4 the methodology is described; what kind of simulations were performed, and how. A short comparison of the different computer codes developed for research purposes and for the nuclear industry is performed. The Monte Carlo based research code MURE is presented, which is the code used for all simulations in this project. The long-term behavior of a nuclear reactor core is explained, along with how the fuel composition changes with time, and how this influences the reactivity of the system.

In chapter 5 the project is described in detail; especially the multirecycling of the uranium isotopic vector from thorium based fuels. A presentation of the nuclear fuel cycle, with emphasis on the thorium fuel cycle – and how this differs from the uranium fuel cycle in some aspects, is provided. The important distinction between the **independent** and the **dependent** thorium fuel cycle is made. The advantages and disadvantages of the thorium cycle are discussed. It is shown that the reprocessing substantially reduces the volume and most importantly the heat production of the waste.

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[3] As of June 30, 2009.

[4] The Megatons to Megawatts[TM] Program is a government-industry partnership in which bomb-grade uranium from dismantled Russian warheads is being recycled into LEU used to produce fuel for American nuclear power plants.

This will reduce the need of geological repositories, thus massively reducing the costs of the final disposal of the long-lived waste. The costs of the nuclear fuel cycle and how these can be minimized by multirecycling of the fuels are discussed.

All the simulation results are presented in chapter 6. The simulations of the thoriated fuels are evaluated under three different criteria: waste produced, safety, and economy. The difference between the once through cycle and multireprocessing is evaluated. It is shown that reprocessing is absolutely necessary if "cleaner" nuclear waste is to be produced. The reactivity feedbacks, and how they change as uranium is recycled, and the effect of protactinium production from the thorium, which is an important feature of the thorium fuel cycle, are both important results for reactor safety. Whether thoriated fuels are currently economically viable is examined. Proliferation is also an important question. The production of the ^{232}U will make the fuel proliferation resistant since remote fuel handling will be required, although this is also a drawback for the reprocessing. The ^{232}U production is thus both a positive and a negative feature of the thorium cycle. The conclusions and a future outlook of this project can be found in chapter 7.

"If you worry about climate change then there is no other economically or environmentally stable alternative to nuclear power." -Mikko Elo, an MP for Finland´s Social Democrat Party in an interview with BBC [6].

2. Basic theory

2.1 Discovery of fission

Nuclear fission was discovered by Otto Hahn and Fritz Strassmann, in Germany in 1938 [7]. The term 'fission' was given by Otto Frisch, a word he borrowed from biology – binary fission, which means cell division [8]. Enrico Fermi actually discovered fission in 1934, but he did not recognize it [8].

Shortly after the discovery of fission they also discovered the possibilities of the enormous energy release, and in 1942 the "Manhattan Project" was established in Los Alamos, New Mexico in the USA, scientifically lead by Robert Oppenheimer. The aim of the project was to develop the first nuclear weapon in history. Later that year Fermi built the first controlled fission reactor – the "Chicago Pile number 1". This first sustained fission reaction happened in a pile of graphite, with just air cooling – it was really a prototype for the graphite moderated gas cooled reactor [9].

2.2 Principle of fission

Fission is the process where a heavy nucleus is split into two lighter nuclei, and the binding energy of the original nucleus is transformed into kinetic energy. A qualitative picture of what happens when a nucleus fission can be given on the basis of the liquid-drop model of the nucleus, where the nucleus is regarded as an electrically charged drop. E.g. a ^{235}U nucleus (A in Figure 2-1) absorbs a neutron, becoming a $^{236}U^*$ nucleus with excess energy. This excess energy causes violent oscillations, during which a neck between the two lobes develops (B in Figure 2-1). The electrical repulsion of these two lobes stretches the neck farther, and finally two smaller drops are formed that move rapidly apart (C in Figure 2-1) [10]. If neutron absorption results in excitation energy greater than the energy barrier height, fission occurs immediately. This is the case for ^{233}U, ^{235}U, and ^{239}Pu, among others.
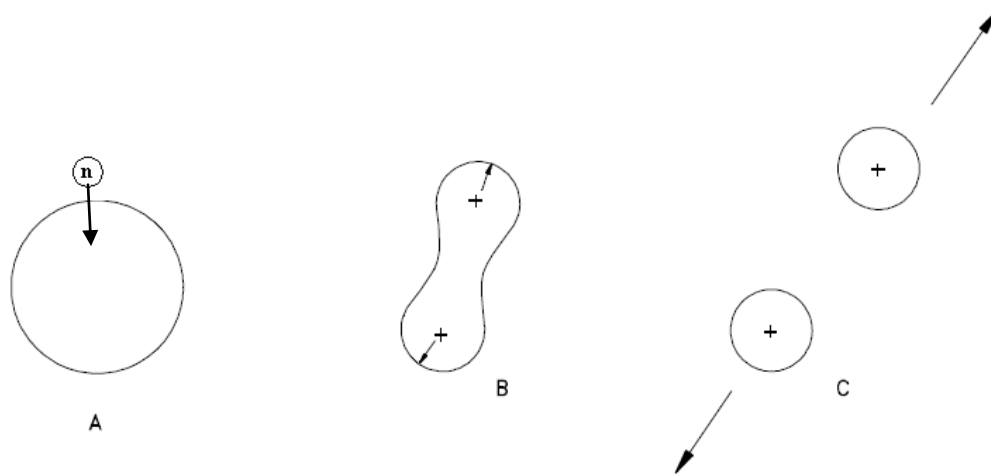
Figure 2-1: The shapes of fission

2.2.1 The curve of binding energy

The weight of the nucleus of i.e. ^{233}U is less than the sum of the masses of the 92 protons and 141 neutrons that make up the core. In general; if the masses of the Z protons and N neutrons that make up the nucleus of element X are added, it is found that the weights of these constituent masses exceed the weight M_X of the nucleus as a whole. The total binding energy of a nucleus is given by the mass difference $Zm_p + Nm_n - M(A,Z)$, where M(A,Z) is the mass of the nucleus as a whole. The binding energy per nucleon is:

$$B(A,Z) = \frac{Zm_p + Nm_n - M(A,Z)}{A}$$

B(A,Z) provides a measure of nuclear stability; the larger it is the more stable the
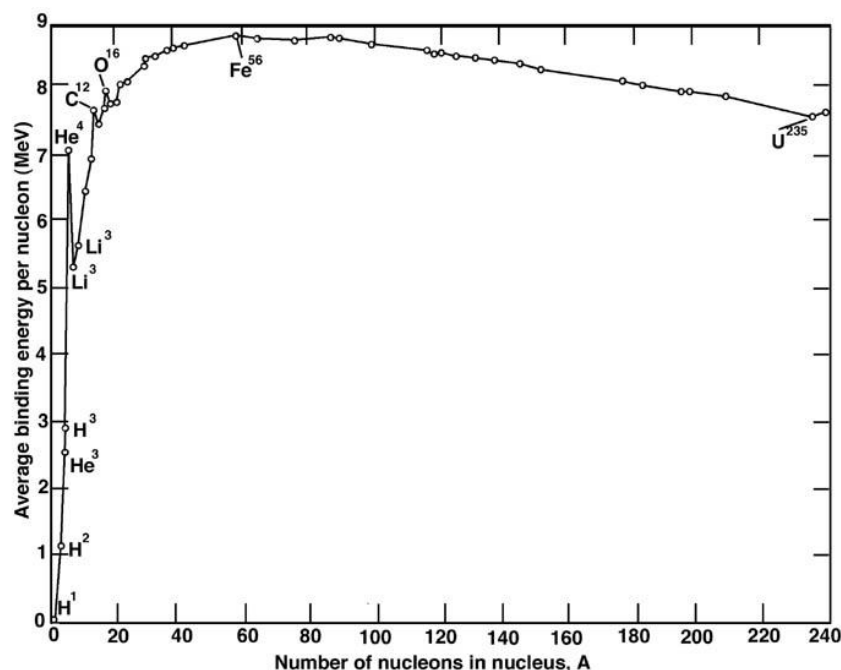
Figure 2-2: Binding energy per nucleon

nucleus will be. Figure 2-2 [11] shows B(A,Z) plotted as a function of the number of nucleons in the nucleus. The binding energy is released when a heavy nucleus fission or two light nuclei undergo fusion.

A typical fission reaction is:

$$n + {}^{235}U \rightarrow {}^{236}U^* \rightarrow {}^{141}Ba + {}^{92}Kr + 3n + \gamma$$
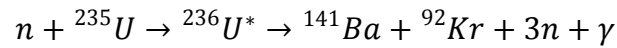
The Q-value of the fission process is defined as:

$$Q \equiv M_{{}^{235}U} + M_n - \left(M_{{}^{141}Ba} + M_{{}^{92}Kr} + M_{3n} \right) = T_f - T_i$$
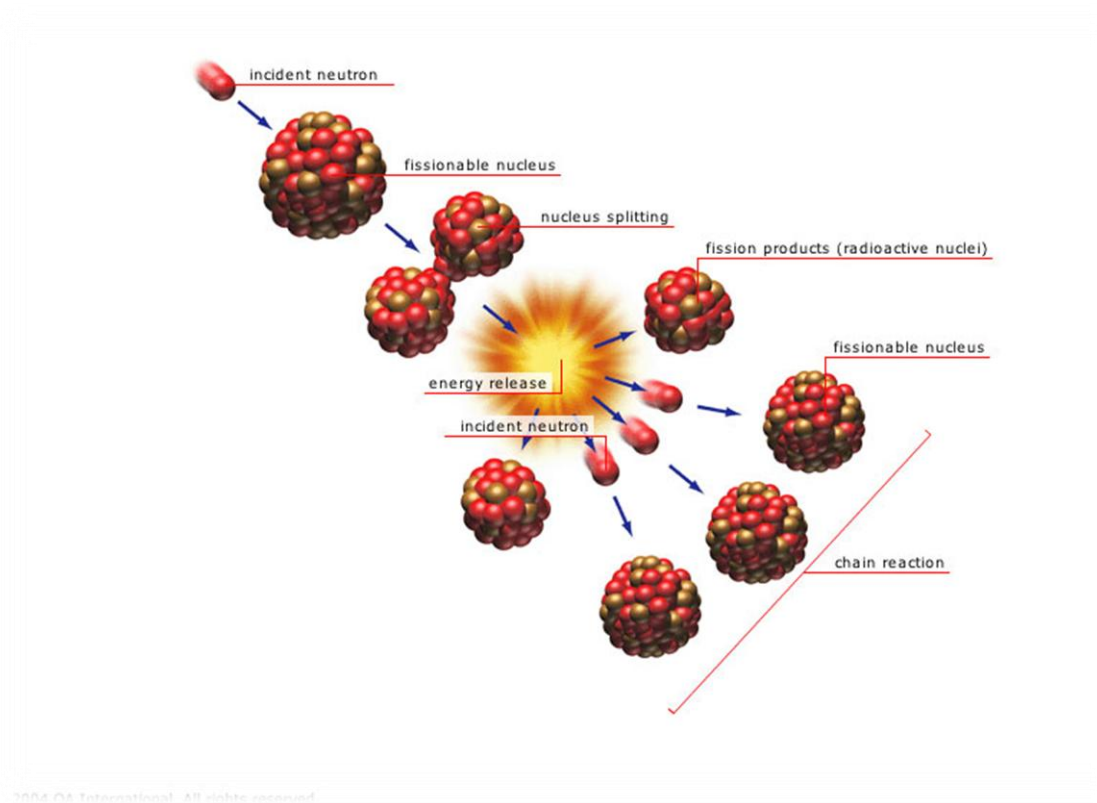
Figure 2-3: The fission process

where Q is the energy released by a reaction, and the gain in kinetic energy, and T_i and T_f is the initial and final kinetic energy of the reaction respectively. Neutron induced fission in e.g. ${}^{233/235}U$ has a Q-value of ~200MeV. As can be seen Figure 2-3 [12] the reaction produces two or three neutrons and two lighter nuclei (fission fragments). The

additional energy released is γ rays and neutrinos, which are not shown in the figure. The energy released appears as kinetic energy of the fission fragments, neutrons, and gamma rays, as well as that from beta particles, gamma rays, and neutrinos emitted as the fission products undergo radioactive decay. This activity of the fission fragments will cause production of heat, but more than 80% of the energy released by fission appears as the kinetic energy of the fission fragments [13] .

Table 2-1: Released and regained energy by fission [14]

|	Released energy (MeV)	Regained energy (MeV)
Kinetic energy of fission fragments	168	168
Activity from fission products:		
β	8	8
γ	7	7
Neutrinos	12	0
Prompt γ	7	7
Kinetic energy of fission neutrons	5	5
Sum	**207**	**195**

The energy of the neutrinos is lost, since they very weakly interact with matter, and simply leak out of the reactor.

Fission neutrons

At the moment when the nucleus actually splits (~10^{-17} seconds after the process starts) most of the neutrons are emitted. These are called the *prompt neutrons*. The two or three neutrons born with each fission will travel in straight lines until making a collision, at which point they scatter or are absorbed. If they scatter, they change direction and energy, and continue along a new straight line. If absorbed, they cease to exist, with their energy dissipated by the collision. The absorption of a neutron will in many cases cause the absorbing nucleus to become radioactive. The neutron can also induce a new fission,

and under the right circumstances a chain reaction of fissions. Neutron lifetime begins with neutron emission from fission, and ends with absorption.

2.2.2 Fission products

The total number of particles that participate in a fission reaction is conserved, but there can be several different fission fragments. The reaction of thermal neutron induced fission is anti symmetric; the fission fragments consist of one light (A ~ 90) and one heavy (A ~ 140) nucleus. Figure 2-4 [15] shows the thermal fission yield for different fissioning nuclei. The mass distributions for the heavy fragments overlap quite well, of while the light fragment distribution is somewhat various for the different parent nuclei initiating the fission; it tends to peak at higher Z the heavier the fissioning nucleus, thus ^{233}U peaks just above A=90 and ^{239}Pu peaks just above A=100. If the liquid-drop model fission was a complete description of the process, it would be expected that the average masses should scale roughly with the mass of the drop. Instead, the observed average
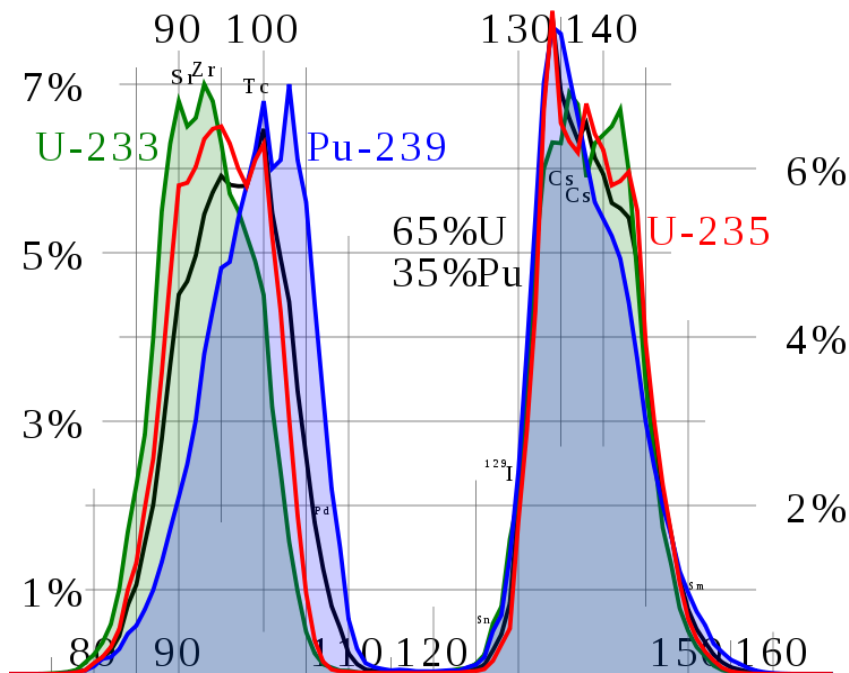
Figure 2-4: Thermal fission yield by mass: ^{233}U (green), ^{235}U (red), ^{239}Pu (blue), and a mixture of uranium and plutonium (black).

mass of the heavy fragment stays nearly constant at about 140, while the average mass of the lighter fragment increases linearly as A increases. Just at the lower edge of the heavy fragment mass distribution is the doubly magic nucleus ^{132}Sn, with its 50 protons and 82 neutrons. This exceptionally stable configuration determines the low edge of the

mass distribution of the heavier fragment. No such effect occurs for the lighter fragments; the light fragment mass distribution is less by shell closures [7].

The fission fragments have neutron to proton ratios that are too large, and will therefore undergo beta minus decay. During this process they may emit neutrons – β-delayed neutron emission. For example, ^{138}I beta decays with a half-life of 6.5 seconds to ^{138}Xe. Most of the beta decays populate low excited states in ^{138}Xe, but about 5% the ^{138}I decays populate states in ^{138}Xe at about 6.5 MeV, this energy exceed the neutron separation energy and these states therefore decay by neutron emission to ^{137}Xe [7]. These so-called *delayed neutrons* are emitted seconds to minutes after the fission, and are an addition to the prompt neutrons emitted in the instant of the fission process. The delayed neutrons play an important role in the process of controlling the reactor (more details in chapter 3.4.1).

When a chain reactor is shut down, radioactive decay of the fission products will **continue to produce significant amounts of heat.**

2.3 Neutronics

The neutron is the key to the nuclear reactions in a reactor; it initiates the fission, and converts fertile nuclei to fissile. Because it is a neutral particle, it does not experience any coulomb repulsion, and it can easily be absorbed by the nucleus.

2.3.1 Fissionable materials

There are two classes of materials of interest for a nuclear reactor: fissile materials – that fission when hit by a neutron with the correct, low energy, and fertile materials – that have the ability of capturing a neutron and then decay, one or several times, to an isotope that is fissile. Fertile isotopes may also undergo fission directly, but only if impacted by a high-energy neutron, typically in the MeV range. Both fissile and fertile materials are fissionable materials. Important fissile isotopes are $^{233/235}$U and ^{239}Pu, and fertile isotopes are ^{238}U and ^{232}Th.

For a material to be useful as reactor fuel it must satisfy these criteria:

1. The nucleus must undergo fission with a certain cross section absorbing neutrons
2. It must emit enough fission neutrons, so that at least one neutron will induce another fission.

If the nucleus has properties 1 and 2 it is called a fissile material. In addition, for practical purpose the material must also be available in sufficient amounts.

Uranium is the only naturally occurring element that has a fissile isotope – ^{235}U, with a half-life long enough that it has survived the geological life of the earth. For practical purpose there are also two other fissile materials that can be used as fuel in a nuclear reactor, these are ^{233}U and ^{239}Pu, which are converted from ^{232}Th and ^{238}U respectively:

$$n + {}^{232}Th \rightarrow {}^{233}Th \rightarrow {}^{233}Pa \rightarrow {}^{233}U, \qquad n + {}^{233}U \rightarrow fission$$

$$n + {}^{238}U \rightarrow {}^{239}U \rightarrow {}^{239}Np \rightarrow {}^{239}Pu, \qquad n + {}^{239}Pu \rightarrow fission$$
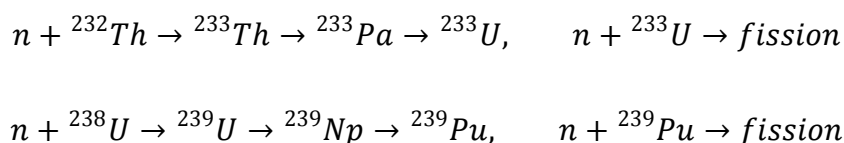
When "burning" nuclear fuel several fissile isotopes, other than ^{233}U and ^{239}Pu, are produced (more details in chapter 4.3). Examples are $^{243/245}$Cm and ^{242}Am. These isotopes will, when they fission, release similar energies as $^{233/235}$U and ^{239}Pu, and therefore add to the total energy production of the reactor. They are, nonetheless, not suitable as nuclear fuel, even though they hold criteria 1 and 2. The reason for this is that the fraction of delayed neutrons emitted of these nuclei when they fission is too small, and as a consequence a reactor fuelled with for instance ^{243}Cm would not be possible to control. More details of reactor control in chapter 3.4.1.

As can be seen from these two equations: When dealing with fertile materials, one neutron is necessary to sustain the chain reaction and more than one neutron is needed to convert fertile to fissile material.

2.3.2 Absorption

To sustain a chain reaction, on average one of the two or more neutrons created by each fission reaction must survive to create a subsequent fission. At the core of neutron interactions is the concept of the cross section, σ, which is a measure of the probability that a certain interaction – i.e. fission – will take place. Each nucleus has a cross-sectional area of σ cm^2. This is the area the incoming neutron "sees"; a picture of how "large" the nucleus is to the neutron. Thus $\sigma = cm^2/nucleus$. This cross-sectional area is in the order of 10^{-24} cm^2, and usually the (microscopic) cross-section is tabulated in barns (b) where $1b = 10^{-24}$ cm^2.

In its simplest form, the absorption reaction – where a neutron enters a nucleus – creates a compound nucleus, which is in an excited state. The probability of the formation of a compound nucleus is given by the absorption cross-section. This excited compound nucleus will most probably either fission or it will emit a gamma and be transmuted to a

heavier element, thus the $\sigma_{absorption}$ is the sum of these two possibilities, and the $\sigma_{fission}$ is really the probability of a compound nucleus formation (the absorption of a neutron) minus the probability of a (n,γ) reaction:

$$\sigma_{fission} = \sigma_{absorption} - \sigma_\gamma$$

The fissile nuclei have cross sections of fission of typically several hundreds of barns; i.e. the cross-section of fission of [233]U when hit by a thermal neutron is 530 barns. This number is orders of magnitude larger than the physical size of the nucleus.

2.3.3 Neutron energy spectrum

The ability to sustain a chain reaction depends a great deal on the energy distribution of the neutrons, because neutron cross sections are strongly energy dependent, which in turn is determined by the composition of non-fissile materials in the core and their effectiveness in slowing down the neutrons from fission toward thermal energies. The neutron energy distribution is determined largely by the competition between scattering and absorption reactions. In a medium for which the average energy loss per collision and the ratio of scattering to absorption cross section are both large, the neutron distribution in energy will be close to thermal equilibrium and is then referred to as a soft or thermal spectrum.

The neutron energy spectrum in a thermal reactor range from 0.001 eV to 10 MeV, and is shown in Figure 2-5 [16].

Fission neutrons are born in the MeV energy range with and average of about 2 MeV
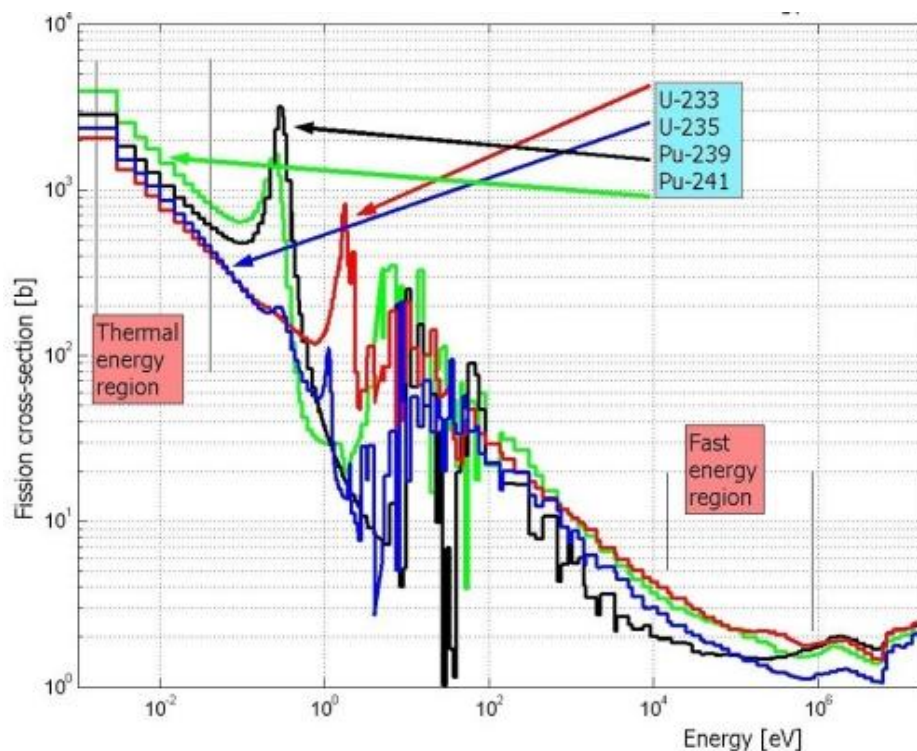

Figure 2-5: Energy dependece of fission cross-section

and an upper limit of 10 MeV. The intermediate energy range is often referred to as the resonance or slowing down region of the energy spectra because of the importance of these two phenomena. Often the terms intermediate and resonance are used interchangeably in describing the energy range between 1.0 eV and 0.1 MeV because as neutrons slow down from fast to thermal energy the large cross sections caused by the resonances in uranium, plutonium, and other heavy elements account for nearly all of the neutron absorption in this energy range. In the thermal and intermediate ranges no fission neutrons are born. The likelihood of a compound nucleus formation greatly increase if the excitation energy brought by the incident neutron corresponds to a quantum state of the resulting nuclei, and the cross-section exhibit resonance peaks at neutron kinetic energies corresponding to those quantum states. In general, the heavier a nucleus is, the more energy states it will have, and these will be more closely packed together.

A **thermal neutron** has a kinetic energy of 0.025 eV, which is the most probable energy at room temperature. From the of the Maxwell-Boltzmann distribution for this temperature (~300K). However, neutrons with energies from 0.001 eV to 1.0 eV are referred to as thermal; they have small enough energies that the thermal motions of the surrounding atoms can significantly affect their scattering properties. The fission cross sections are largest in the thermal energy region, that is E<~1eV.

Concentrating neutrons at either high or low energies and avoiding the range between roughly 1.0 eV and 0.1 MeV most easily achieves a chain reaction.

2.3.4 Moderation of neutrons

The purpose of a moderator is to slow down fast neutrons with relatively few collisions to the thermal energies where the fuel´s ratio of neutron production to absorption exceeds one by a substantial margin. Moderation is the process of the reduction of the initial high kinetic energy of the free neutron. Neutron energy degradation caused by scattering is referred to as neutron slowing down, since along with the reduction of energy comes a reduction in speed.

Moderation

If E is the neutron energy before a collision and E´ the energy after the collision:

$$\frac{E'}{E} = \left(\frac{A-1}{A+1}\right)^2$$
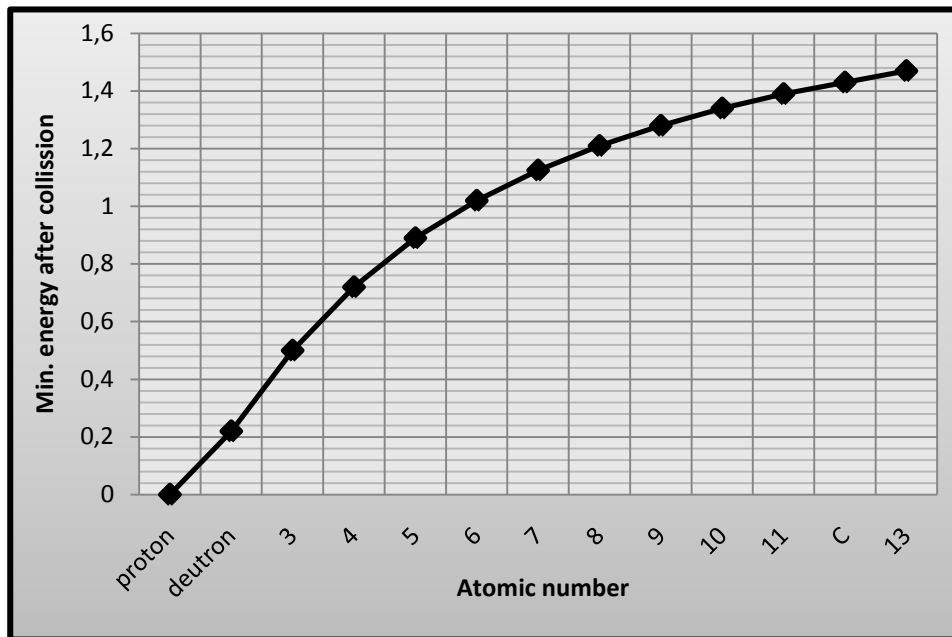
Figure 2-6: Energy after collision as function of atomic number of moderator. The energy before collision is 2 MeV

The largest neutron energy losses result from collisions with light nuclei, which is shown in Figure 2-6 where minimum energy after a collision is plotted as a function of atomic number of moderator. Therefore hydrogen-rich material, like water (H_2O or D_2O), is used as moderators. Another commonly used moderator is solid graphite (^{12}C). A collision of a neutron, which has a mass of 1, with a ^1H nucleus could result in the neutron losing virtually all of its energy in a single head-on collision, although more generally, it is necessary to take into account both glancing and head-on collisions.

A nuclide's ability to slow neutrons down by elastic scattering is called the slowing down decrement, ξ, which is the mean logarithmic reduction of neutron energy per collision. In elastic collisions the neutron loses on average the same logarithmic fraction of its energy, regardless of its initial energy, for ξ depends only on the atomic mass of the scattering nuclide. For A=1, ξ=1.

$$\xi \approx \frac{2}{A + 2/3}$$

Using the definition of ξ it is possible to make a rough estimate of the number n of elastic collisions required to slow a neutron from fission to thermal energies:

$$n = \frac{1}{\xi} ln\left(\frac{E_0}{E_n}\right)$$

Taking fission energy as E_0=2 MeV and thermal energy as E_n=0.025 eV, we have $\ln\left(\frac{E_0}{E_n}\right) = \ln(2.0 \cdot 10^6/0.025) = 18.2$, hence $n = 18.2/\xi$. Thus for hydrogen n~18, for deuterium (A=2) n~25, for carbon (A=12) n~115, and for ^{238}U n~2275. Only with low atomic weight materials is the slowing down decrement large enough to slow neutrons down to thermal energies with relatively few collisions.

Moderator properties

Some nuclei have larger capture cross sections than others, and they remove free neutrons from the flux. Therefore, a further criterion for an efficient moderator is one for which this parameter is small. In addition the moderator must have a large macroscopic scattering cross section; $\Sigma_s = N\sigma_s$, so that scattering is the dominant action/reaction.

The moderating efficiency gives the ratio of the macroscopic cross sections of scattering, $\Sigma_{scatter}$, weighted by ξ divided by that of capture, Σ_γ:

$$\frac{\xi\Sigma_{scatter}}{\Sigma_\gamma(\text{thermal})}$$

If the thermal capture cross section $\Sigma_\gamma(thermal)$ is large, a material cannot be used as a moderator; even though it may be effective in slowing down neutrons to thermal energy, it will then absorb too many neutrons.

Table 2-2: Slowing down properties of common moderators [13]

Moderator	Slowing Down Decrement	Scattering cross section	Capture cross section
	ξ	$\Sigma_{scatter}$	$\Sigma_\gamma(\text{thermal})$
H_2O	0.93	1.38	0.022
D_2O	0.51	0.35	$8.6\cdot10^{-6}$
C	0.158	0.035	0.00028

Neutrons are more rapidly moderated by light water, as ^{1}H has a far higher $\Sigma_{scatter}$ than D_2O and C. However, it also has a far higher Σ_γ(thermal), so that the moderating efficiency is nearly 80 times higher for heavy than for light water. Reactors using a light

water moderator and fueled with natural uranium are not possible; some enrichment of the uranium is required to compensate for the larger thermal capture cross section of the H_2O.

The ideal moderator is of low mass, has a high scattering cross section, and a low capture cross section.

2.3.5 Neutron multiplication

The chain reaction´s neutron multiplication, k, is defined as the ratio of the number of fission neutrons produced to the number of neutrons absorbed:

$$k = \frac{number\ of\ neutrons\ in\ ith + 1\ generation}{number\ of\ neutrons\ in\ ith\ generation}$$

For k greater than, less than, or equal to one, the neutron population will increase, decrease, or remain the same, respectively.

The multiplication can be approximated by

$$k = k_\infty P_{NL}$$

Where P_{NL} is the neutron non-leakage probability and k_∞ is the multiplication that would exist if the reactor´s dimensions were infinitely large, and no neutrons would leak out of the system. The energy dependence of the cross sections dominates the determination of k_∞.

Some fraction of the neutrons from the fission reaction will be captured. η is the number of neutrons produced per neutron absorbed – the "excess" neutrons:

$$\eta(E) = \frac{\nu \Sigma_{fission}\ (E)}{\Sigma_{absorption}\ (E)} = \frac{neutrons\ produced\ per\ fission}{neutrons\ absorbed}$$

Where ν is the average number of neutrons produced per fission. The average value of η must be substantially more than one, since neutrons will be lost by capture in structural, coolant, and other materials and some will simply leak out of the system.

Table 2-3: Fuel properties of common fissile nuclei; in a thermal neutron spectrum

	^{233}U	^{235}U	^{239}Pu
σ_{fission}	530	586	752
σ_γ	47	95	270
ν/η	1.093	1.175	1.370

Table 2-3 shows fuel properties of the most common fissile nuclei, ^{233,235}U and ^{239}Pu, in a thermal neutron spectrum. The characteristics of a fissile nucleus are its fission cross section ($\sigma_{fission}$), and its capture cross section (σ_γ) –the absorption not resulting in fission but a (n,γ) reaction and thus a heavier isotope of that element. Most important is its ratio of capture to fission; defined as $\frac{\nu}{\eta}$, a property that tells how big the probability is that the fuel nuclei will fission. It should be as close to unity as possible. For ^{235}U, and especially ^{239}Pu, more of the neutron captures will result in a (n,γ) reaction, than the wanted (n,fission) reaction. It is obvious that in a thermal spectrum, ^{233}U is the best fissile material, even though the fission cross-section of ^{235}U and especially ^{239}Pu is bigger than that of ^{233}U. Hence, a 1 MW$_t$ reactor needs 1.15 g ^{233}U, 1.24 g ^{235}U, or 1.44 g ^{239}Pu per day [14].

The four factor formula

The four factor formula for k$_\infty$ was developed early in the history of reactor physics, to relate neutron behavior to the thermal hydraulic feedback.

Most of the fission neutrons are born as a result of the absorption of thermal neutrons in the fuel, and they emerge as fast neutrons. If $N_0 = 1000$ such fast neutrons are produced from the fission, some nominal fraction of these neutrons will cause fast fission in the fertile material, resulting in a total number of $\varepsilon N_0 = 1040$ fast neutrons produced from fission, where ε is the fast fission factor. Generally ε > 1, in this specific case (see Figure 2-7) it is 1.04. The 1040 fission neutrons then migrate into the moderator region where they are moderated. However, only some fraction p survive to thermal energies, with the remaining neutrons lost to the resonance capture in the fuel; p (<1) is referred to as the resonance escape probability.
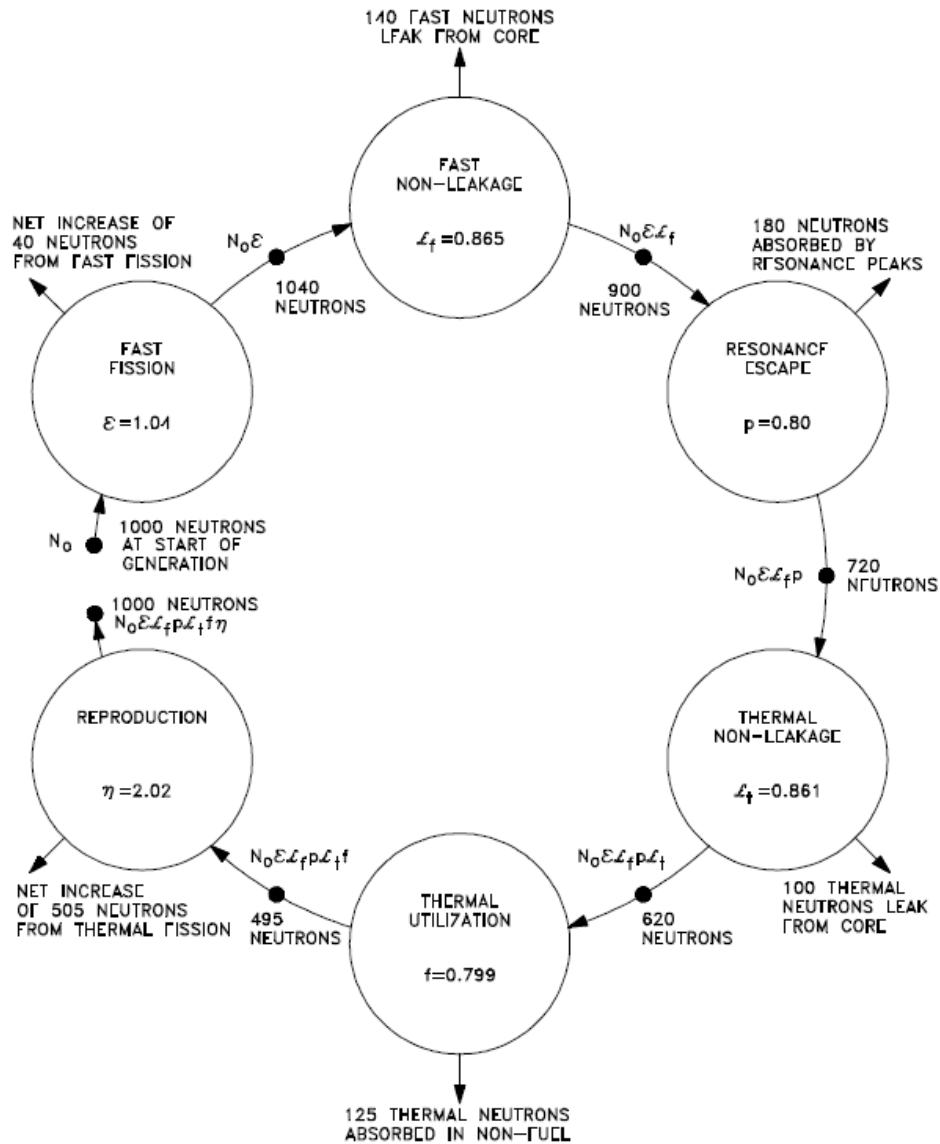
Figure 2-7: Illustration of the neutron life cycle for a thermal reactor with k=1

Thus the fuel absorbs $\varepsilon p f N_0 = 655$ (while the moderator absorbs $\varepsilon p(1-f)N_0 = 177$) neutrons. For each thermal neutron absorbed in the fuel, η_T (>1) fission neutrons result. $\varepsilon p f \eta_T N_0 = 1343$ fission neutrons appear from thermal fission with MeV energies generated from N_0 such fission neutrons of the previous generation. Hence, the infinite neutron multiplication can be written as follows:

$$k_\infty = \varepsilon p f \eta_T$$

In an operating reactor, fuel depletion, which reduces η_T, the presence of control rods or other control poisons, which reduce f, when taken together with P_{NL}, the nonleakage probability, must yield $k = k_\infty P_{NL} = 1$. In Figure 2-7 [17] the k-value is shown;

$k = \varepsilon p f \eta_T P_{NL}$, where P_{NL} is divided into the non leakage probability of first fast neutrons ($\mathcal{L}_f$), and then the non leakage probability of thermal neutrons ($\mathcal{L}_t$).

3. The power reactor core

Coal is the fossil fuel that has been most widely used for the production of electricity; with the chemical reaction

$$C + O_2 \rightarrow CO_2$$
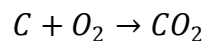
this results in the release of ~4 eV per atom combusted. The nuclear reaction releases approximately 200 MeV per uranium nucleus that fissions [13]. In other words: the energy per atom from a nuclear reaction is 50 million times more than that of a chemical reaction. If 1.053 grams of ^{235}U is "burned" in a nuclear power plant (all ^{235}U nuclei fission), the thermal energy released will be 1 MWd, which is enough energy for an average Norwegian family for one year. According to Statistisk Sentrabyrå (Statistics Norway) an average Norwegian household consumes roughly 22 000 kWh of energy in one year (2006), which is approximately 1 MWd [18]. If one were to get the same energy from burning coal, 4 tons of coal is needed [19]! (5% or more of the coal burned becomes ash that must be removed and stored in a landfill or elsewhere [13].)
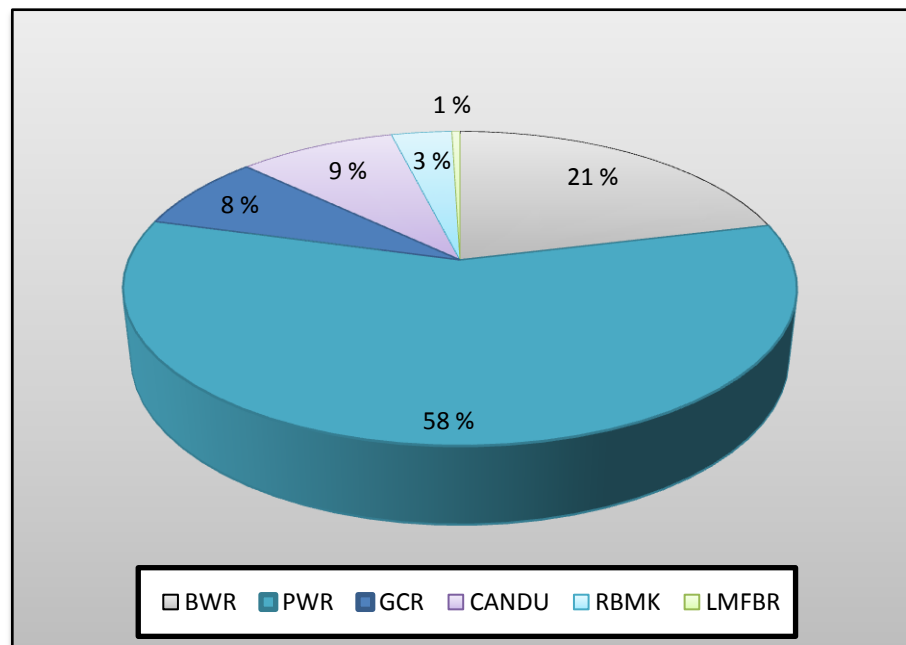

Figure 3-1: Operating reactors worldwide

A nuclear power plant is in principle a thermal power plant, where the energy is produced by fission reactions in the fuel. Each fission will on an average release about

2.5 neutrons($\bar{v} = 2.5$). To get a constant energy yield from the reactor, the multiplication factor, k, must be one – then the reactor is said to be critical. In other words: when a reactor is critical it is really balanced; the neutron population is constant. Criticality must be maintained over the range of required power levels and over the life of the core as fuel is depleted. The design must also allow the thermal energy produced from fission to be transferred out of the core without overheating any of its constituents. Neutrons that do not induce fission are mostly absorbed by (n,γ) reactions in the fuel, or other places in the reactor.

There are 436 central station nuclear power reactors operating in the world today (2009), and these reactors produce 370 221 MW of electrical power [20]. As seen in Figure 3-1 [9] 79% of these are Light Water Reactors (LWR); 58% Pressurized Water Reactors (PWR) and 21% Boiling Water Reactors (BWR). The remaining 21% are Gas Cooled Reactors (GCR), Pressurized Heavy Water Reactors (CANDU), Graphite Moderated Water Cooled Reactors (RBMK), and Liquid Metal Fast Breeder Reactors (LMFBR). 5 of the 436 reactors in operation are in long term shutdown, however, 45 new reactors are under construction [20]. According to the Energy Information Administration of the U.S. Government, nuclear power is responsible for producing 15.2 % of the world's total electricity consumption (2005) [1].

There are two main categories of reactors: Thermal and fast – according to the energy of the neutrons initiating the fission reactions. The thermal reactors make use of thermalized, slow neutrons, while fast reactors operate in the fast energy range. In general, most power reactors are cylindrical in shape with coolant flowing through channels extending the axial length of the core. In all cases, heat from fission is produced within the fuel and conducted to the coolant. The heat is removed from the core coolant. All reactors consist of the same essential elements: the *fuel*, or fissile material; a *moderator* to thermalize the neutrons (not present in a fast reactor); a *reflector* surrounding the *core* (fuel elements plus moderator) to reduce neutron leakage and thereby reduce the critical size of a reactor; a *containment vessel* to prevent the escape of radioactive fission products; *shielding* to prevent neutrons and γ rays from causing biological harm to operating personnel; a *coolant* to remove heat from the core; a *control system* allowing the operator to control the power level and to keep it constant during normal operation; and various *emergency systems* designed to prevent runaway operation in the event of a failure of the control or coolant systems.

A large power reactor contains many thousands of cylindrical fuel elements – often referred to as fuel rods or fuel pins. These fuel elements are grouped together to form fuel assemblies, and the assemblies are grouped together to form the reactor core – the heart of a nuclear power plant. The assemblies will also have holes – guide tubes, where control rods can be inserted. Control rods consist of strong neutron absorbers – such as

boron, cadmium, or hafnium. Their insertion controls the reactor multiplication during power operations, and they shut down the chain reaction when fully inserted. The reactivity of a reactor must be possible to control, so that the flux, hence the effect, may be changed when needed. This is achieved by varying the fuel's position, by adding neutron absorbing matter to the coolant/moderator, or by neutron absorbing control rods.

3.1 Thermal reactors

The thermal reactors (LWR, CANDU, GCR, and RBMK) [14] dominate by far the energy production by nuclear fission; all of today's successful reactor systems are thermal reactors, using slow or thermal neutrons to maintain the fission chain reaction in the fuel (mostly ^{235}U as fissile). Thermal reactors use a neutron moderator to sow down the fast neutrons from fission. The moderator is often also the coolant, most commonly water under high pressure to increase the boiling point.

The LWR has become the most popular reactor type, and there are two types; PWR and BWR. Both reactors run on low enriched uranium, of 2-5%, and they have an efficiency of about 30% - that is, the thermal energy is three times the electrical energy. Light water reactors use ordinary water both as moderator and to remove heat. More details on the PWR in chapter 3.3.

3.2 Fast reactors

The fast reactor employ a fast neutron spectrum, and materials must therefore be chosen so that neutrons are moderated as little as possible. The coolant must be a material that is transparent to neutrons – like sodium.

The advantage of the fast reactor is that it can produce more fissile material than it consumes; it breeds its own fuel, and is therefore often referred to as a fast breeder. The primary fissile nuclide for a fast breeder is ^{239}Pu, and the primary fertile nuclide is ^{238}U.

In the core of a fast reactor there is a high fissile concentration, typically around 20%. This active core is surrounded by fertile material; where the breeding takes place. Assemblies containing only UO_2, or a lower Pu content, are placed in a blanket around the core for breeding of fissile plutonium. Otherwise, design practice follows established lines, with fuel assemblies of clad pins arranged together in the core, interspersed with control rods. The reactor is largely un-pressurised since sodium does not boil at the temperatures experienced, and is contained within steel concrete shields.

A typical LMFBR core is about 1 meter high and 2 meters in diameter. LMFBRs tend to have a positive void coefficient (see chapter 3.4.2). The reason is that reduction of the sodium density hardens the neutron spectrum, which results in a lower capture-to-fission ratio in the fuel and reduces the number of neutrons absorbed in the large ^{23}Na resonance in the keV energy range [9].

Fast reactors have the potential to increase the energy available from a given quantity of uranium by a factor of fifty or more, and can utilise the existing stocks of depleted uranium, which would otherwise have no value [21]. However, they are still currently at the prototype or demonstration stage, and will become commercial only if uranium or other energy prices substantially increase.

3.3 The Pressurized Water Reactor

The PWR was first developed in the United States based on experience from the naval reactor program. It is a thermal reactor where neutrons are moderated by ordinary water which also serves the purpose of coolant. As mentioned in chapter 2.3.4, light water´s large thermal absorption cross section precludes the possibility of achieving criticality with natural uranium fuel in a LWR, which is possible in the CANDU reactor, where neutrons are moderated by heavy water. Some enrichment, typically 2-5% is therefore required.

3.3.1 The reactor core

A fuel element is made up by small pellets, which are ~1 cm in diameter and 1 cm high. These uranium oxide (UOX) pellets are encapsulated in cladding made of Zirkaloy, which is an alloy of zirconium, tin, iron, chrome, and possibly nickel. The cladding offers structural support and it prevents fission product leakage into the coolant. The pellets are packed up in fuel rods, and these again are mounted in the fuel assembly. Figure 3-2 shows, from left to right, a pellet, a fuel pin, and the assembly. A typical fuel assembly may consist of an 17×17 array of fuel rods. The assembly is approximately $20cm \times 20cm \times 4m$ high. About 190 to 240 fuel assemblies containing 90 000 to 125 000 kg of UO_2 constitute a typical PWR core, which is about 3.5 m in diameter and 3.5 to 4.0 m high.

The schematics of a PWR is shown in Figure 3-3 [22]: Coolant flows in an open lattice structure; it typically enters the pressure vessel near the top, flows downward between the vessel and the core, is distributed at the lower core plate, flows upward through the core, and exits the vessel at the top. The core is contained in a vessel pressurized to 1520

bar to prevent coolant boiling at operating temperatures in the range of 316 °C [13]. Water exiting the core (pink) circulates through heat exchangers, called steam generators, before being pumped back to the core inlet. The steam generator operates at a lower pressure such that feed water entering it boils (blue), thus supplying steam to the turbine, where its energy is liberated, before it cools in the third loop (turquoise). This third loop is normally water from a lake or a river.
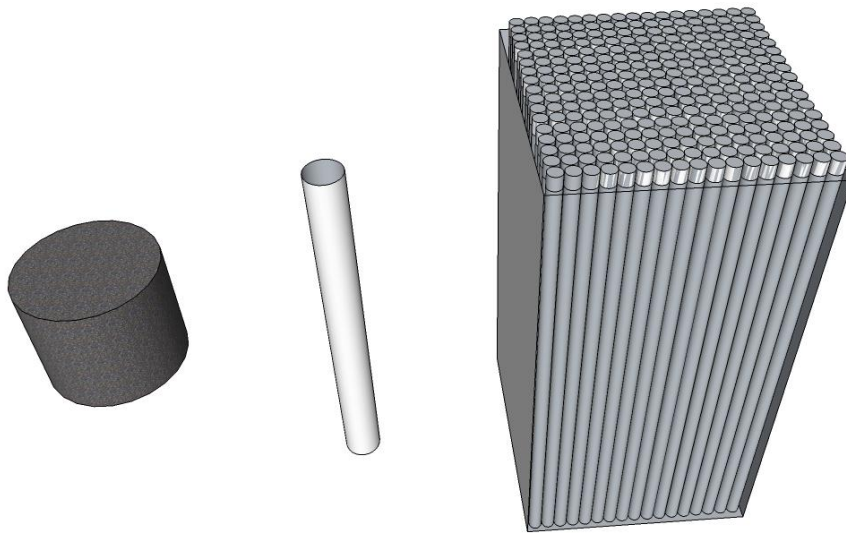
Figure 3-2: Pellet, fuel pin, and assembly

Control rods of the PWR are employed in clusters, and are inserted from the top of the reactor.

Not all of the assemblies that make up the core are identical. They may differ in fuel enrichment in order to flatten the power across the core, or they may have been placed in the core during different refueling operations. The reactor is shut down at regular intervals, ranging from 1 to 2 years. During shutdown, typically lasting a number of weeks, 20-30% of the assemblies containing fuel from which the fissile material is most depleted are removed and replaced by fresh assemblies. With the core consisting of "batches" of more or less depleted fuel, less neutron poison is needed in the coolant since the most depleted batches will work as a neutron poison itself; by stealing more neutrons than the fresh fuel. The most depleted fuel will be placed in the center of the core where the neutron flux is more intense, while the freshest fuel will be placed in the periphery of the core, thus leaking more of its neutrons.
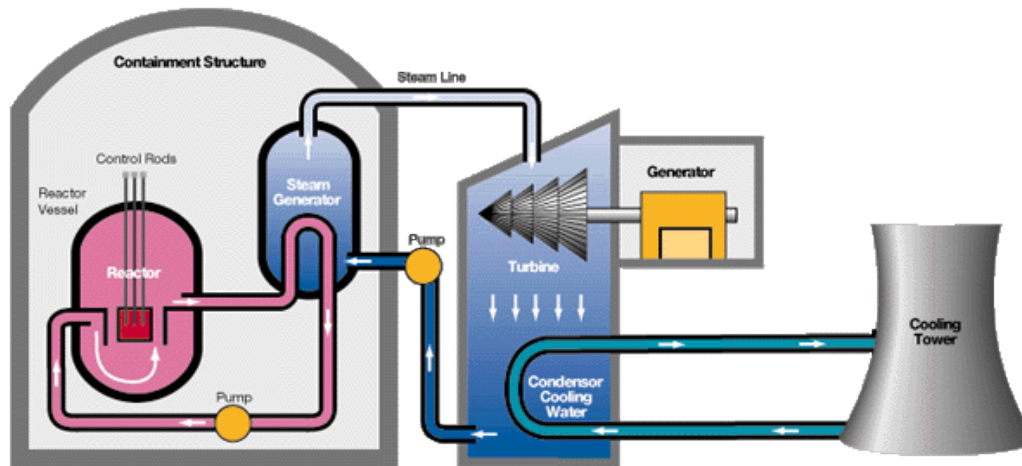
Figure 3-3: Schematics of a PWR

3.3.2 Long-term reactivity control

Long-term reactivity control of a PWR is provided by adding boric acid to the coolant; the content of this neutron poison is then adjusted to keep the core at criticality. Boron concentration is then reduced with fuel burn up to compensate fuel reactivity loss, like buildup of the fission products ^{135}Xe and ^{149}Sm, which are both extreme neutron absorbers, and fuel depletion. In most thermal reactors burnable poisons placed in the fuel or elsewhere also serve to compensate for fuel burn up. The boron, however, makes a positive contribution to the moderator temperature coefficient of reactivity, and therefore maximum concentration is limited.

Soluble poisons are used to compensate fuel-depletion reactivity in PWRs but not in BWRs, because of the possibility that they will plate out on boiling surfaces, as the coolant in the BWR boils.

3.3.3 The European Pressurized water Reactor

The reactor that has been studied in this project is the European Pressurized water Reactor (EPR). The EPR is a Generation III+ PWR, developed by Framatome ANP, a subsidiary of French AREVA and Siemens. This reactor generates about 1600+ MW of electric power – which is higher than that of the most recent plants – and features

enhanced safety, and simplified operations and maintenance. It also has a projected service life of 60 years, compared with a 40-year service life for existing power reactors[5] [23]. The EPR is the most modern reactor design, currently being built in France and Finland. It is an evolutionary product, but not a revolutionary one, because it is based on proven pressurized water technology; which is currently the most widely-used technology worldwide.

The EPR uses all the different types of fuel currently burned in PWRs: fuel containing slightly enriched uranium (up to 5%) and recycled fuel based either on reprocessed, re-enriched uranium or on mixed plutonium and uranium (MOX). Contrary to older PWRs, *this reactor can be loaded entirely with MOX fuel [23]*. Due to its flexibility with respect to different fuel types it is reasonable to believe that a mixture of thorium and ^{235}U can be employed as well, and that a full core of thoriated fuel is un-problematic.

The core is made up of 241 fuel assemblies, with an average of 265 fuel pins per assembly. To achieve long burn-ups, burnable gadolinium poison is added to the fuel.

Main design features of the EPR

- The reactor containment building has two walls: an inner pressurized concrete housing covered internally with a leak tight metallic liner and an outer reinforced concrete shell both 1.3 meters thick.
- The containment houses the reactor coolant system: the reactor vessel, four steam generators, a pressurizer and four reactor coolant pumps. *The larger volumes of the main components, as compared to previous designs, give additional benefits in terms of operating and safety margins.*
- The nuclear island, covering the reactor building, the fuel building, the four safeguard buildings, the nuclear auxiliary building and the waste building, stands on a single thick reinforced concrete basement. *This provides protection against a major earthquake.*
- EPR is designed to achieve 92% availability averaged over the entire 60 years of its design lifetime, with long irradiation cycles, shorter refueling outages and in operation maintenance.
- Optimized core design and higher overall efficiency with savings on uranium consumption. *Costs are therefore reduced for the entire fuel cycle.*

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[5] This does not mean that the existing reactors will have to be shut down after forty years. On the other hand, no-one can predict which upgrades will be required at that time, or say how safety regulations will be possibly hardened by the Authority.

Key nuclear safety aspects

- Increased protections against accidents include core meltdown and their radiological consequences. Within the containment there is a special area where, *in the very unlikely event of core meltdown, any of the molten fuel would be collected, retained and cooled.*
- *Robust against external hazards including commercial plane impact and severe earthquake.*
- Enhanced radiological protection of the public and operating and maintenance personnel.
- Fourfold redundant safety systems, and their supporting systems, with independent and physically separated "trains" or subsystems to minimize the consequences of internal and external hazards. The different trains of the safety systems are located in four different buildings with strict physical separation. *Each train is capable of performing the entire safety function independently.*
- The EPR approach is based mainly on the "defense in depth" deterministic approach backed up by probabilistic risk assessment studies of all faults and hazards.

The very first EPR is currently being built in Olkiluoto in Finland. Construction work commenced in February 2005 with the planned start of electricity generation in 2011. When finished, the Olkiluoto 3 will be the largest reactor in the world. There is also an EPR under construction in Flamanville in France.

3.4 Elements of reactor safety

The fundamental objective of reactor safety is to ensure that radio nuclides emitting ionizing radiation are not released to create a health hazard to the general public or operating personnel [9].

The first level of reactor safety is to design it to *prevent* the occurrence of any event that could result in damage to the fuel or other reactor systems. Negative reactivity coefficients (temperature, void) lead to naturally stable operating conditions. Second level is *protective*; the protective systems are designed to halt or bring under control any transients resulting from operator error or component failure. The third level is the *mitigation systems,* which limit the consequences of accidents if they do occur. Such systems are emergency core cooling; emergency secondary coolant feed water, emergency electrical power systems, systems for removing fission products that have been released into the reactor hall, and a reinforced containment building that can withstand high overpressure.

3.4.1 Delayed neutrons; controlling the chain reaction

The neutron density as a function of time is given by:

$$n(t) = n_0 e^{(k-1)\frac{t}{l}}$$

Where t is the time in seconds, and l is the neutron lifetime. If $k = 1$ the neutron density is constant, and the reactor is critic, or balanced. If $k > 1$ the neutron density will increase exponentially [13]. The reactor period, T, is defined as $T = l/(k - 1)$, and the neutron density can be written as

$$n(t) = n_0 e^{\frac{t}{T}}.$$

In a prompt critic LWR, that is with k=1 for the fission neutrons not taking into account the delayed neutrons, the average neutron lifetime is l=10^{-4} s [14]. If this reactor has an increase of only 0.001, then k=1.001, and the reactor period $T = \frac{10^{-4}}{10^{-3}} = 0.1s$. This means that in just one second the neutron density will increase by a factor e^{10}=22026.

Fortunately, the delayed neutrons, mentioned in chapter 2.2.2, change the reactor period dramatically:

Table 3-1: Comparison of a LWR with and without the delayed neutrons

	Without delayed neutrons	**With delayed neutrons**
Neutron lifetime l	0.0001 seconds	0.084 seconds
Reactor period T	0.1 seconds	84.1 seconds
Increase in power in one second	1 MW ➔ 22000 MW	1 MW ➔1.012 MW
The time to double	0.07 seconds	58 seconds

Less than 1% of the total number of neutrons will be delayed; still, they give a stable reactor period which is long and easy to control.

Table 3-2 gives an overview of the delayed neutrons in % of the total number of fission neutrons. The fraction of delayed neutrons from fission of ^{233}U is almost 1/3 of that from fission of ^{235}U. Because of this, a critical reactor with thorium based fuels is closer to being prompt critical than one that is fueled with ^{235}U. This means that the margin for control is smaller than for normal UOX.

Table 3-2: Delayed neutron fraction [14]

	% from thermal fission
^{233}U	0.26
^{235}U	0.65
^{239}Pu	0.21

3.4.2 Reactivity feedback

The reactivity, ρ, measures the change in neutron multiplication in the reactor core:

$$\rho = \frac{k-1}{k}$$

The reactivity is affected by many factors, including coolant/moderator temperature and density, fuel temperature and density, and structural temperature and density. The net reactivity in a reactor is the sum of all these contributions, and this total coefficient of reactivity has to be negative.

Temperature coefficient

The temperature coefficient of reactivity is a measure of how the reactor responds to increased temperature. A positive number denotes a trend of increasing power production as temperature rise, whereas a negative number denotes a trend of decreased power production as temperature rise, consequently, a negative feedback makes the reactor stable against changes in temperature.

$$\alpha_T \equiv \frac{d\rho}{dT}$$

Because the temperature changes originate in the fuel, and then are transferred to the moderator, it is common to define one temperature coefficient for the fuel and one for the moderator.

A negative fuel temperature coefficient is generally considered to be even more important than a negative moderator temperature coefficient because fuel temperature immediately increases following an increase in reactor power, while the time for heat to be transferred to the moderator is measured in seconds. In the event of a large positive reactivity insertion, the moderator temperature cannot turn the power rise for several seconds, whereas the fuel temperature coefficient starts adding negative reactivity immediately.

Fuel temperature coefficient:

Also called the "prompt" temperature coefficient because an increase in reactor power causes an immediate change in fuel temperature. Another name applied to the fuel temperature coefficient of reactivity is fuel Doppler reactivity coefficient. This name is applied because in LWRs, where there is typically a low enrichment, the fuel temperature coefficient of reactivity is mostly a result of the Doppler Effect, also called Doppler broadening. The phenomenon of the Doppler Effect is caused by an apparent broadening of the resonances due to the constant thermal motion of nuclei. Raising the temperature causes the nuclei to vibrate more rapidly, effectively broadening the energy range of neutrons that may be resonantly absorbed in the fuel. This means a broadening of the resonance capture cross sections of the fertile material, hence a decrease in the resonance escape probability p, and less reactivity [14].

Two nuclides present in large amounts in the fuel of some reactors with large resonant peaks that dominate the Doppler Effect are ^{238}U and ^{240}Pu in conventional UOX fuel. The Doppler Effect is even bigger for ^{232}Th, if thoriated fuels are being used.

Moderator temperature coefficient:

Increase in temperature in a liquid-moderated thermal reactor will make the moderator/coolant density decrease, and contribution to the moderator coefficient, derive primarily from these density changes, with changes in the thermal neutron energy spectrum playing a secondary role. A decrease in moderator density decreases the effectiveness by which neutrons are slowed down through the resonance region. Therefore the resonance absorption increases, causing the resonance escape probability to decrease, and the moderator temperature coefficient to be negative.

Void coefficient

If the coolant is a liquid, increasing temperatures can cause small gas bubbles – voids – to form, displacing the coolant. Voids may also form if the coolant is lost from the reactor (a loss-of-coolant accident). If the coolant acts as a neutron absorber due to i.e. boric acid, then displacing it will give a positive reactivity, but if it also acts as a neutron moderator then the displacement also gives a negative reactivity. The void coefficient of reactivity represents how the reactor responds to the formation of such bubbles.

A negative void coefficient means that the reactivity decreases as the void content inside the reactor increases – but it also means that the reactivity increases if the void content inside the reactor is reduced. If a reactor is designed to operate with no voids at all (like the PWR), a large negative void coefficient may serve as a safety system. A loss of coolant in such a reactor decreases the thermal output, but of course heat that is generated is no longer removed, so the temperature could rise (if all other safety systems simultaneously failed).

3.4.3 Accident scenarios

Loss of flow and Loss of coolant

A Loss-of-flow accident (LOFA) would be caused by a failure of one or more pumps in the primary coolant systems. The worst case scenario in a reactor is a Loss-of-coolant accident (LOCA). This happens if the reactor vessel is destroyed, or that the main pressure pipe breaks. Both the LOFA and the LOCA would result in *increased temperature and decreased density of the coolant*, and with a LOCA, *possibly uncovering of the core*. The negative coolant temperature reactivity coefficient of PWRs (and BWRs), which would provide for an immediate power reduction, is an important feature in the early stages of such accidents. However, the residual heat from a 1000 MW$_e$ reactor is around 225MW$_t$ [14]. Even after the chain reaction is stopped, the core needs cooling; which is why the reactor will have an emergency core cooling system.

Reactivity insertion

Uncontrolled control rod withdrawal is the most common type of initiator for a reactivity insertion accident, but also cold water into the primary coolant system would cause a positive reactivity insertion in reactors with a negative coolant reactivity coefficient. If the coolant flow in the secondary system is increased, there would be increased heat

removal from the primary coolant. In reactors with a negative coolant reactivity coefficient this would result in a positive reactivity insertion. Because of this there are also limits on the allowed magnitude of negative coolant reactivity coefficients.

4. Method

4.1 Simulations

Over the last 30 years, the development of computers has had a dramatic effect on the possibilities of doing advanced simulations. Full core three dimensional simulations are now possible. Reactor physics rely more and more on simulations, and less on reactor experiments – that are both expensive and difficult.

There are mainly two types of computer codes for simulations/calculations of a nuclear reactor: **Deterministic** codes and **Monte Carlo** (probability based) codes.

A deterministic code, such as *Apollo* or *Eranos*, both French industrial codes, solves the diffusion equation, also called the transport equation:

$$\vec{\nabla} \cdot D\vec{\nabla}\varphi(\vec{r}) + \nu\Sigma_f \varphi(\vec{r}) - \Sigma_a \varphi(\vec{r}) = 0$$

This equation describes how the neutrons lose their energy as function of spacial distribution. Practically solving it for realistic geometries/systems is extremely difficult, but is possible with some assumptions and approximations. The diffusion equation has a positive flux solution within a reactor only if it is exactly critical. Otherwise, the neutron population will vary with time. Therefore the challenge with the deterministic method is to solve the diffusion equation to find the neutron flux, φ, as a function of space and energy.

The nuclear industry uses deterministic codes; both the Apollo and the Eranos are codes designed for a PWR. For research purposes, however, with new types of fuel, or reactor designs which have never been constructed, the knowledge of the flux distribution is poor; and the results of deterministic codes may not be the most precise. It is also difficult to obtain information about the industrial codes, since they are normally not available, or they have limited availability. Hence, for research Monte Carlo based codes, that statistically track particles through the geometry (e.g. a neutron), are the best suited and most accurate since they do not use assumptions or approximations. The simulations for this project were carried out with the Monte Carlo based research code MURE (more details in chapter 4.1.2). Other (semi) open codes that are competitors to

MURE are *Origen*, *Cinder*, and *Monteburns*. These can be found in the database of the Nuclear Energy Agency (NEA).

The advantage of the deterministic codes is without doubt speed. Simulating an assembly with a Monte Carlo based code (e.g. MURE) may take days; this is due to the statistical nature of the method, the calculated quantities have statistical uncertainties which decrease only slowly with the number of neutron histories sampled. This normally is no problem for calculating quantities averaged over large regions in space and energy; however, when results with a high resolution are required, often very long computational times are necessary to achieve sufficiently small statistical uncertainties. The same calculation may take only minutes with a deterministic code. However, the deterministic codes are not suited for simulation of innovative systems and fuel cycles.

An example of problems that may occur with a deterministic code, but is no problem with MCNP/MURE, is energy self-shielding (resonance) – the outer portion of the fuel shields the interior from the neutrons: The neutron flux is lower in the fuel than it is in the moderator, which is caused by the fact that some of the neutrons entering the fuel from the moderator are absorbed near the surface of the fuel – they do not survive to contribute to the flux in its interior [24].

4.1.1 The Monte Carlo method

The Monte Carlo method is generally attributed to the scientists working on the development of the atomic bomb in Los Alamos during the 1940s.

Monte Carlo can be used to duplicate theoretically a statistical process – such as the interaction of nuclear particles with material – and is particularly useful for complex problems that cannot be modeled by computer codes that use deterministic methods. The statistical sampling process is based on the selection of random numbers – analogous to throwing dice in a gambling casino – hence the name "Monte Carlo". In particle transport, the Monte Carlo technique consists of actually following each of many particles from a source throughout its life to its death in some terminal category (absorption, escape, etc.). Numbers between 0 and 1 are selected randomly to determine what (if any) and where interaction takes place, based on the rules (physics) and probabilities (transport data) governing the processes and materials involved. The method obtains answers by simulating individual particles and recording some aspects (tallies) of their average behavior. The average behavior of particles in the physical system is then inferred from the average behavior of the simulated particles.

Monte Carlo "solves" a transport problem by simulating particle histories, while deterministic methods solve the transport equation for the average particle behavior. *The Monte Carlo method is well suited to solving complicated three-dimensional, time-dependent problems.*

MCNP

MCNP is a general-purpose Monte Carlo N-Particle code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport. The neutron energy regime is from 10^{-5} eV to 20 MeV for all isotopes and up to 150 MeV for some isotopes, the photon energy regime is from 1 keV to 200 GeV, and the electron energy regime is from 1 keV to 1 GeV. The capability to calculate k_{eff} eigenvalues for fissile systems is also a standard feature.

MCNP was originally developed by the Monte Carlo Group, currently the Diagnostic Applications Group, (Group X-5) in the Applied Physics Division (X Division) at the Los Alamos National Laboratory. The code represents over 500 person-years of sustained effort. It is comprised of about 425 subroutines written in Fortran 90 and C – to be as system independent as possible to enhance its portability.

Nuclear data

The user creates an input file that is subsequently read by MCNP, which contains geometry specification, description of materials and selection of cross-section evaluations, etc.

MCNP uses continuous-energy nuclear and atomic data libraries. Evaluated data are processed into a format appropriate for MCNP by codes such as NJOY, which is a cross section modifier. It modifies the measured cross-sections for different temperatures, since the cross-sections are only measured for one temperature. This is true for most nuclei, except some, like ^{235}U and ^{239}Pu where measurements are done for several temperatures, since these measurements are really difficult to perform.

Nuclear data tables exist for neutron interactions, neutron-induced photons, photon interactions, neutron activation, and thermal particle scattering. Over 836 neutron interaction tables are available for approximately 100 different isotopes and elements. More neutron interaction tables are constantly being added as new and revised evaluations become available. Cross sections for nearly 2000 activation reactions involving over 400 target nuclei in ground and excited states are part of the MCNP data package. For neutrons, all reactions given in a particular cross-section evaluation are accounted for.

Geometry

The geometry of MCNP treats a 3-dimensional configuration of user-defined materials in geometric cells. The cells are defined by the intersections, unions, and complements of the regions bounded by the surfaces. Particles are tracked through this geometry.

Cells are defined on cell cards. Each cell is described by a cell number, material number, and material density, followed by a list of operators and signed surfaces that bound the cell. Each surface divides all space into two regions, one with positive sense with respect to the surface and the other with negative sense. The intersection operator in MCNP is simply the blank space between two surface numbers on the cell card.

Example:

```
1 0 1 -2 -3 6
```
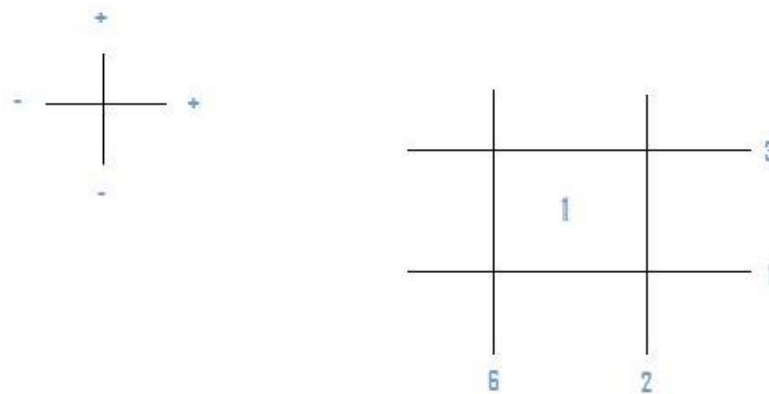

Figure 4-1: MCNP geometry

In Figure 4-1, cell 1 is a void and is formed by the intersection of the region above (positive sense) surface 1 with the region to the left (negative sense) of surface 2, intersected with the region below (negative sense) surface 3, and finally intersected with the region to the right (positive sense) of surface 6.

4.1.2 MCNP Utilities for Reactor Evolution – MURE

All simulations for this project were carried out with the evolution code MURE based around MCNP. MURE perform nuclear reactor time-evolution; the code automatically performs consecutive MCNP calculations to determine reaction rates and hence deduce core material evolution over time at a constant reactor power.

About MURE

MURE is a precision research code and has been developed jointly at the Institut de Physique Nucléaire d´Orsay (IPN) and the Laboratoire de Physique Subatomique et Cosmologie (LPSC) of Grenoble. It consists of a powerful ensemble of utilities for reactor simulations, written in C++; there are currently more than 25 000 lines of code, representing about 15 person-years of development.

The program gives a realistic modeling of reactors. It is possible to do fuel time evolution, safety studies, and sensitivity analysis. Calculations of fissile inventories, fluxes, average cross sections, waste produced, etc, everything as a function of time, can be performed. It allows the construction of advanced reactors and innovative systems.

From error analyses of MURE it has been determined that typical errors in the inventories of various isotopes at the end of cycle are in the order of 2%. For certain nuclei, produced only by reactions at an energy threshold, the errors can be larger (~5%) since the production of these nuclei are highly sensitive to changes in the shape of the neutron spectrum [2].

MURE consists of four major parts:

Part 1

Interfaces with MCNP (input geometries, materials, neutron sources etc.). Makes it "easy" to generate MCNP input files. Ability to create large lattice´s of similar components.

Part 2

Construction of the network of connection between nuclides via reactions and decays. Builds a specific "nuclear tree" from an initial material composition (list of nuclei): The tree of each "evolving" nucleus is created by following the links between neighbors via radioactive decay and/or reactions until a self-consistent set of nuclei is extracted.

Part 3

Aims at simulating the evolution of the fuel within a given reactor over a time period of up to several years, by successive steps of MCNP calculation and numerical integration of Bateman's equations. Each time MCNP is called, the reactor fuel composition will have changed due to the fission/capture/decay processes occurring inside.

Part 4

Extra modules:

- Interface to NJOY, to process cross-sections at the wanted temperature.
- Graphical interface for data visualization (MureGui).

All graphs that show results from this project were made using MureGui.

Geometry generation, nuclear data management, and evolution calculations

Modification of an existing geometry in MCNP requires more or less a complete rewriting of the input file. With MURE, however, it is much easier. Main objects define a system; these are Shapes, Cells, Materials, MCNPSource and Tallies. When a Shape is defined, it can be translated, rotated or placed in another Shape.

Cells associate a Shape with a Material. Materials are defined by giving the density and adding nuclei (identified by their proton number, Z, and their atomic number, A) with a proportion:

```
Material *H2O=new Material(1.0);    //density in g/cm³
H2O->SetTemperature(600);           //temperature K
H2O->AddNucleus(1,1,2);
H2O->AddNucleus(8,16,1);
```

Here the material is defined and given the name H2O. It is given the density of 1.0 g/cm^3. Then the temperature is given of 600 Kelvin. The material consists of protons – Z=1 and A=1, with the proportion 2 – and of oxygen – Z=8 and A=16, with the proportion 1.

Nuclear data management is based on the NJOY code, which can compute thermal effects such as thermal scattering in moderators or Doppler Effect. MURE allows thus to have as much different temperatures taken into account as needed (which is useful for precise temperature coefficients calculations for example).

Fuel evolution is based on a coupling between a static MCNP run and the resolution of nuclei evolution equations. Before an evolution, a full tree of nuclei is built according to nuclear data (decays and available nuclear cross-sections), the nuclei tree is built once and for all at the first MCNP run; initial compositions of all materials are entered by the user and these will evolve automatically. The MCNP input files with the composition at

a given time t_i is built and a MCNP run is performed. The production of a nuclide N_i after a specified time, when a specified number of atoms of the parent nuclide are initially present, is described by the Bateman equations. The assumption is that at t=0, the parent substance alone is present, which is the case in the fuel of the reactor, and the parent is either ^{232}Th, ^{235}U or ^{238}U.[6] dN_i/dt is the total production of nuclide N_i by reactions and decays, and the destruction by reactions and decays. For example the net production of ^{233}U in a thorium fueled reactor:

- Production

  Decay: $^{233}Pa \xrightarrow{\beta^-} {}^{233}U$

  Reaction: $^{232}U(n,\gamma)^{233}U$

- Destruction

  Decay: $^{233}U \xrightarrow{\alpha} {}^{229}Th$

  Reaction: $^{233}U(n,2n)^{232}U$

The Bateman equations turn into a complicated system of coupled differential equations, since the fuel evolves during irradiation, constantly changing the neutron flux and fuel inventories. In short; *dN/dt is the rate of change dependent on the amount of all the precursors.*

The Bateman equations are solved by a standard 4th order Runge-Kutta method using fluxes and cross-sections of MCNP run over a given Δt_i time interval. Then, a new MCNP file with the composition at $t_{i+1}=t_i+\Delta t_i$ is performed, and so on. The tree is simplified by means of a few physical criteria, such as a minimal half-life for decays and an integral cross-section threshold for nuclear reactions. All the necessary tallies for calculating mean neutron fluxes and cross-sections in evolving cells are automatically built.
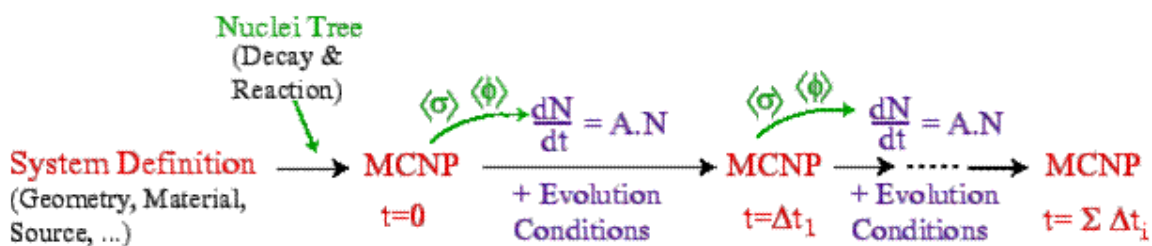

Figure 4-2: General scheme of an evolution calculation in MURE

[6] If the fuel is reprocessed the parent may also be ^{232}U, ^{234}U, or ^{236}U.

Figure 4-2 [25] shows the system of coupled differential equations that make up the nuclei tree. This system describes creation and destruction, by reactions and decay, of around 500 nuclei, with huge variations in time scales; from hours to millennia.

4.2 Creation of geometry and initialization of burn-up calculations

4.2.1 The assembly

The first step was to build the assembly that was to be simulated. A 17×17 lattice of an EPR assembly was modeled using MURE, see Figure 4-3. The assembly consists of 265 fuel pins (yellow pins) of 0.475 cm outer radius with a M5 Zirkaloy cladding of 0.57 mm thickness, and 24 guide tubes (purple pins) for insertion of control rods. The guide tubes are filled with water, and there is burnable gadolinium in the fuel. There are mirrors on the sides of the assembly, so neutrons bounce off and are scattered back. An infinite lattice in the x-y plane was simulated with the use of mirrored surfaces on the assembly sides, which scatter neutrons back into the assembly. In a finite core there will be radial and axial leaks. In these simulations axial leaks were allowed, but radially it was an infinite lattice, and not a finite one. Simulations of just one assembly, instead of a full core, works very well for a thermal reactor. Neutrons born in the assembly of a LWR normally stays in that assembly, and have a small chance of actually getting out of it. This is because the mean free path for a neutron in a thermal reactor is about 6 cm, so the neutrons do not move far before they are captured. In a fast reactor, on the other hand, a lot of neutrons come from other assemblies. Full core simulations are for these reasons not necessary for a thermal neutron spectrum, since more or less all neutrons end their life even in the same or neighbouring fuel pin. In other words: one thing missing from the simulations are radial leaks, which are assumed to be 2% of the total. So, therefore this is compensated by demanding that the k_{inf} ~1.02.[7]

This is an average assembly, and the fuel pins have the same, homogeneous mixture of fuel in all the 241 assemblies that make up the core. In a real core, however, the assemblies would differ depending on where they were placed (explained in chapter 3.3.1); if they were in the periphery of the core they would e.g. have higher enrichment than the average one.

---

[7] With the assumption that the leaks of a 900 MW PWR is normally around 3%; which gives a k_{eff} of ~1.03.

The fuel is a homogenous mixture of UO_2 and thorium; all fuel pins are identical which
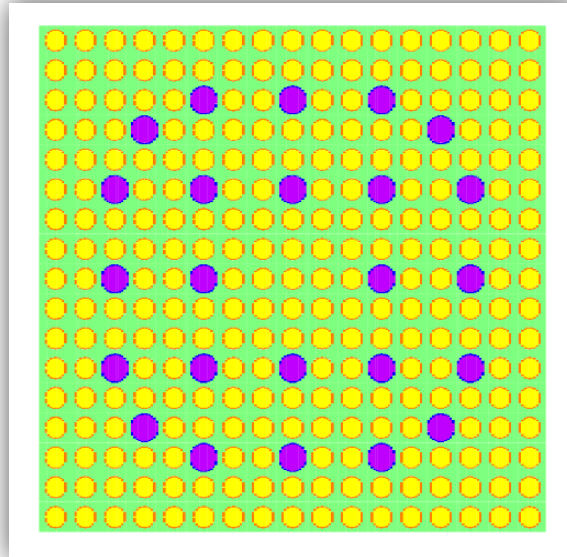

Figure 4-3: An average assembly of the EPR

make them easy, thus cheap, to produce.

4.2.2 Fuel composition/ the right k_{eff}

The next step was to find the correct fuel compositions, and the amount of neutron poisons. The correct fuel enrichment was found by varying the degree of UO_2 in the Th/UOX mixture, with different degrees of enrichment – from 10% to 90%. For each case the k_{eff} (multiplication), was plotted as a function of the UOX content in the fuel (not enrichment). Figure 4-4 shows the result for the 90% enriched case. It was decided to focus on a 20% enriched mixture, which is the proliferation limit, in addition to a 90% enriched case. From Figure 4-4 it was found that there is need for ~4.5% UOX that is 90% enriched, and similarly it can be found for Th/UOX 20% enriched that ~22.5% UOX is needed. This means that a total of 4.05% ^{235}U is needed for a mixture of thorium and 90% enriched UOX, and a total of 4.5% ^{235}U for the mixture of thorium and 20% enriched UOX, to reach a k_{eff} of exactly 1. This difference has to do with ratios of cross-sections for the different materials involved (^{235}U, ^{238}U, Th). However, during the course of a fuel evolution, where the core is to be held at criticality for as much as three years, more fissile material is needed to – compensate to fuel depletion (chapter 4.3). The correct amount of 20% and 90% enriched UOX was found in an iterative way. The calculations of the type shown in Figure 4-4 were taken as the starting point for the first iteration.

90 % UOX enrichment
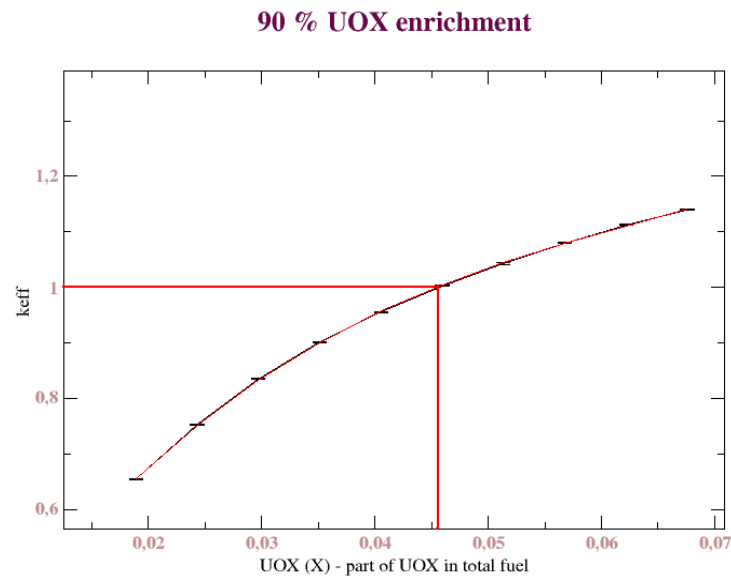
Figure 4-4: k_{eff} as function of UOX content in fuel mixture.
90% enriched UOX

4.3 Long-term core behavior

Long-term changes in the properties of a nuclear reactor over its lifetime are determined by the changes in composition of the fuel. The economics of nuclear power is strongly affected by the efficiency of fuel utilization to produce power, which in turn is affected by the long-term changes associated with fuel burn up. Fuel burn up is measured as *fission energy released per unit mass of fuel*. The fission energy release in gigawatt-days divided by the total mass (in units of 1000 kg or 1 tonne) of fuel nuclei (fissile plus fertile) in the initial loading is referred to as *gigawatt-days per tonne* (GWd/t). Burn-up time is the time at which the reactor can no longer be maintained critical with the control rods withdrawn as fully as allowed by safety considerations.

4.3.1 The burn-up simualtions

k_{eff} of the simulated assembly was kept at 1.02 for each step in the calculation by control of the amount of the boron poison added to the water moderator (more details in 4.3.3).

The value of k_{eff} of 1.02 was chosen for all evolutions, to takes into account leakage of neutrons from the outer edges of the reactor core.

Around 60 MCNP calculation steps at 3-week intervals were performed for a total burn up at the end of cycle (EOC) ~40 GWd/t, burn up time being 1080 days (~3 years). The spent fuel was allowed to cool for ten years before reprocessing and fabrication of recycled fuel. All fuels were in this way compared under the same constant power of a total burn-up of 40 GWd/t.

All simulations were performed for the one average assembly, and then, to get the results for the entire core, multiplied by 241 (the number of assemblies in an EPR core).

Figure 4-6 shows what happens during the three year burn up for some important materials. This specific burn up is for a mixture of thorium and 90% enriched uranium. The principal fissile ^{235}U is being consumed, while several other isotopes are produced, the most significant ones being the fission products. The production of fissile ^{233}U can be seen, but it does not increase in a linear fashion, because the more that gets produced, the greater the rate of fission. There is very little production of plutonium, whereas in conventional UOX there is a considerable production of this material. This is shown in Figure 4-5, which is the burn up result for conventional UOX fuel, 4.5% enriched. The destruction of ^{235}U, and production of fission products are comparable with the thoriated example. Nonetheless, much more plutonium is produced; both fissile ^{239}Pu (still less than ^{233}U in the thorium case) and all other plutonium isotopes ("Pu (rest)"). In addition, the production of TRUs is much bigger in the UOX case than in the Th/UOX case; where it is very close to zero. The detailed results of these burn-up simulations are shown in Table 6-1 and Table 6-3 in the Results-chapter.
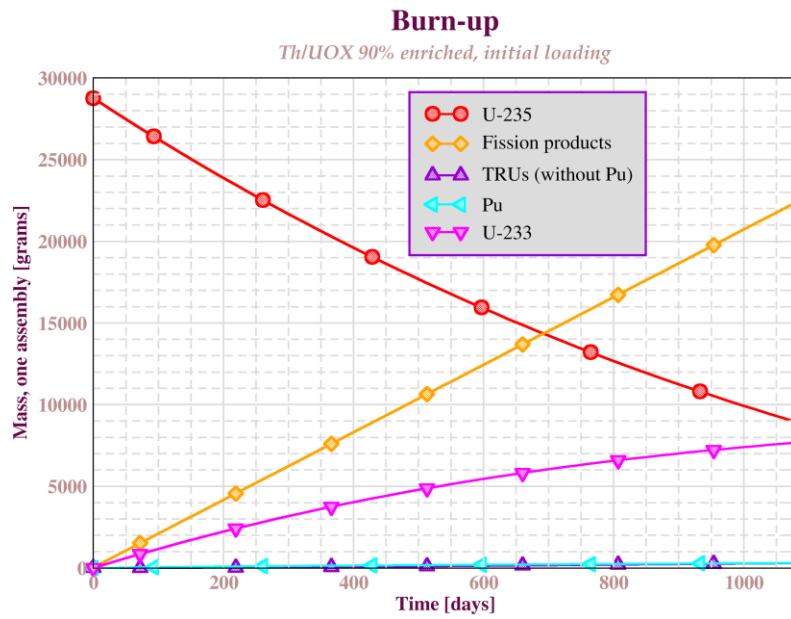
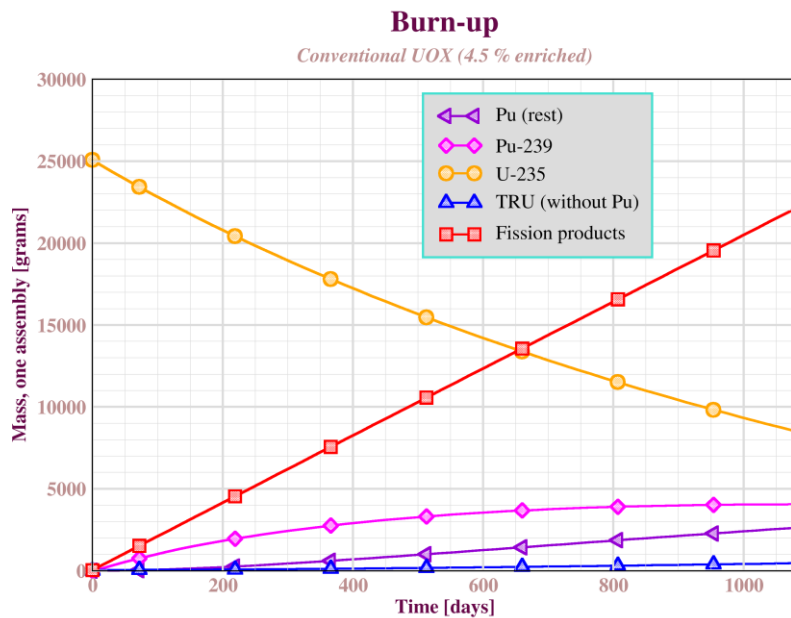*Figure 4-6: Burn-up of the most important materials, for the initial loading of the Th/UOX 90% enriched case.*

Figure 4-5: Burn-up of most important materials, for the conventional UOX fuel, 4.5% enriched.

4.3.2 Changes in fuel composition

Isotopic concentrations change with time as the fissile material is depleted. Fuel nuclei are transmuted by neutron capture and subsequent decay. Some of the fissile nuclei also undergo neutron transmutation via radiative capture followed by decay or
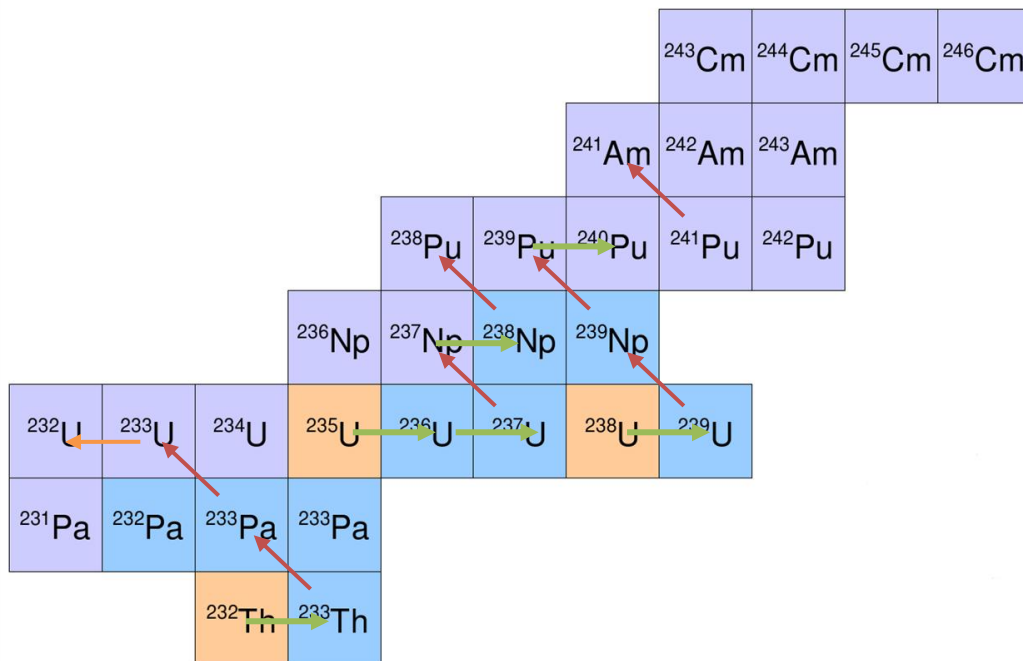
Figure 4-7: Section of the chart of nuclides; the waste precursors.
Green arrow is a (n,γ) reaction, red arrow is β⁻ decay, and the orange
arrow is a (n,2n) reaction

further transmutation. For a thorium-fuelled reactor, a number of uranium isotopes are produced: uranium-232, 3, 4, 5, 6, and 8. ^{237}U has a half-life of 6.75 days and therefore is quickly transformed into ^{237}Np.

Figure 4-7 shows the waste precursors. The long-lived waste is produced from the reactor fuel; through successive neutron-captures and β⁻decays, the long-lived, unwanted isotopes of neptunium, plutonium, americium and curium build up (the TRUs). If the fuel in the reactor consists mostly of ^{238}U (e.g. conventional UOX fuel) there will be a large production of TRUs. Even though the ^{239}Pu produced from ^{238}U may fission and contribute to the total energy production, the plutonium isotope will also capture neutrons and be transformed into other plutonium isotopes or heavier elements. If the fuel is mostly ^{232}Th and small amounts of HEU, there will not be much of the heavy ^{238}U to produce the unwanted elements. The ^{233}U produced from thorium will in most cases fission; it has a very small cross section for neutron capture.

^{233}U is the best fissile nucleus because it has the smallest ratio of capture to fission cross-sections, of only ~0.11.

^{238}U is the main waste precursor, and responsible for the large production of different plutonium isotopes and other long-lived, radiotoxic, transuranic elements in reactor fuel.

If the fuel is mixed oxides (MOX) of uranium and plutonium, the production of TRUs will be even bigger. It can easily be seen from Figure 4-7 that the more heavy isotopes in the fuel at the beginning of the fuel cycle, the greater quantities of TRUs are produced.

4.3.3 Reactivity effects

There are three categories of evolutionary effects:

1. buildup and decay of fission products
2. fuel depletion
3. buildup of actinides resulting from neutron capture in fissile and fertile materials

The multiplication factor decreases with time as the fuel is depleted and fission products accumulate. Fuel depletion causes changes in the (macroscopic) fission and absorption cross sections. The control rods or other neutron poisons must be present at the beginning of core life; these are then extracted to maintain criticality as power is produced. In this project the control rods have been completely withdrawn from the core, (and boric acid was added to the water as neutron poison).

Fissile production and destruction

Fission of fuel nuclei produces two negative reactivity effects; the number of fuel nuclei is reduced and fission products are created. Many of the resulting fission products have measurable thermal absorption cross sections, the ones with the largest capture cross section being ^{135}Xe and ^{149}Sm. These, of course, steal neutrons from the flux; so that more initial fissile material is needed to maintain the chain reaction. At shutdown these isotopes will rapidly decay, increasing the reactivity.

The transmutation of a fertile isotope into a fissile isotope has of course a positive reactivity effect; the buildup of ^{233}U early in life of a thorium-fueled reactor produces a large positive reactivity effect which may be greater than the negative reactivity effect of ^{235}U/^{233}U depletion and fission product buildup. The ^{233}U concentration will saturate at a

value determined by the balance between the ^{232}Th transmutation rate and the ^{233}U depletion rate, at which point the continued depletion of ^{235}U/^{233}U and buildup of fission products produce a negative reactivity effect that accrues over the lifetime of the fuel in the reactor. For a uranium-fueled reactor the equivalents will be the buildup of ^{239}Pu early in life, which saturates according to the balance between the ^{238}U transmutation
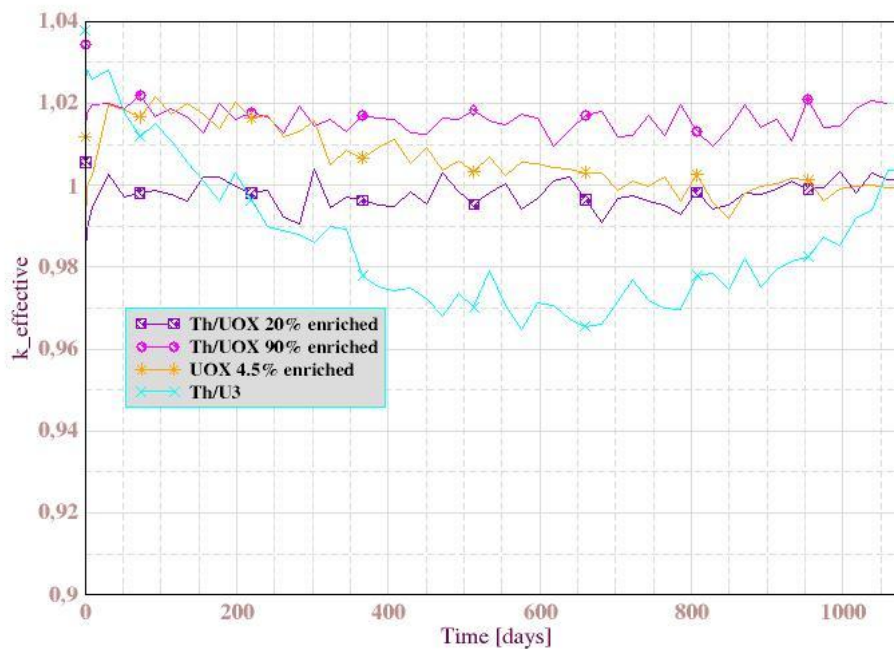
Figure 4-8: k_{eff} as a function of burn-up time.

rate and the ^{239}Pu depletion rate. In Figure 4-6 and Figure 4-5 the buildup and (almost) saturation of ^{233}U and ^{239}Pu can be seen in the Th/UOX 90% enriched fuel case and the UOX 4.5% enriched fuel case respectively. As seen in these figures there is a bigger net production of fissile ^{233}U in a thoriated fuel, than fissile ^{239}Pu in a UOX fuel. The large production of the ^{233}U isotope really holds up the recycling of the uranium from spent thoriated fuels. However, there is also an important safety issue when it comes to the production of ^{233}U: since the half-life of ^{233}Pa, the precursor of the ^{233}U, is 27 days, there will be a buildup of more and more fissile material over the first months after the reactor is shut down. More details of this protactinium-effect in chapter 6.2.2.

Effect of soluble boron poison

In PWRs dissolving a soluble neutron absorber in the coolant and varying the concentration with time compensate for much of the excess reactivity that has to be present at the beginning of core life to achieve a burn up time of 2-3 years. Adjustment of the concentration of the boron in the coolant is used to compensate for fuel-depletion reactivity effects.
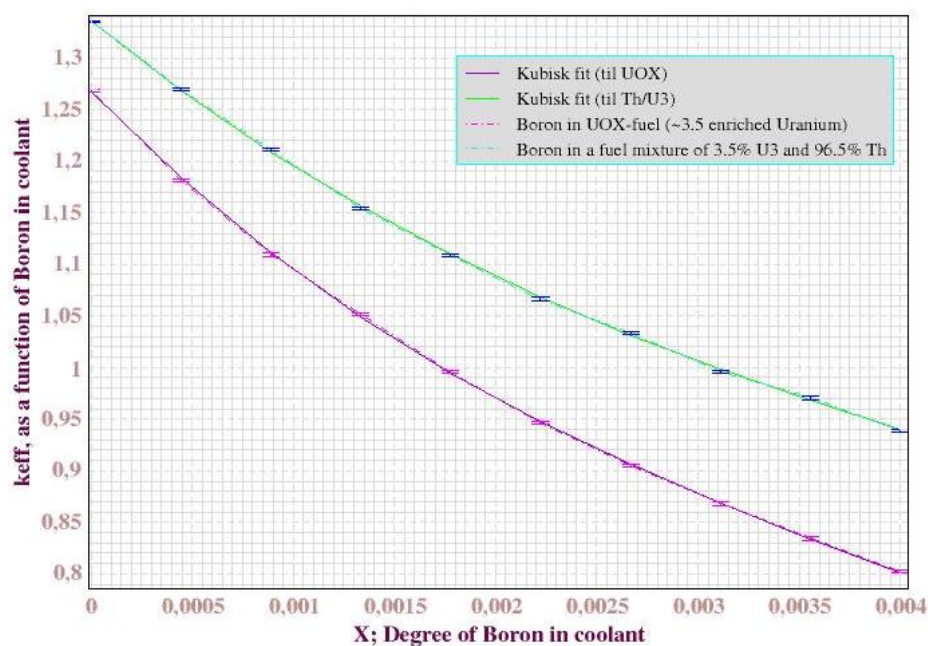
Figure 4-9: Effect of boric acid in coolant; conventional UOX (3.5% enriched), and ^{233}U/Th (3.5% ^{233}U).

Adding neutron poisons to the coolant has the advantage that it will be uniformly spread over the entire reactor core.

Maybe the biggest challenge for this project was to determine the soluble boron concentration for control of the reactivity. The reason for this difficulty is that the fuel composition changes all the time during the cycle, as fissile nuclei fission and fertile nuclei are transmuted into fissile nuclei, and fuel is depleted. The boric acid was removed linearly, and the starting and ending point of the k-value had to be the same. The correct concentration was found by an iterative procedure. As the thorium content in the fuel is brought up, the multiplication will decrease for the first half of the burn-up period, and then rise in the second half. A more precise boron reduction method would therefore be e.g. a quadratic function, rather than a linear. However, the choice of linear boron reduction removal was made to simplify the problem. A more sophisticated

method for determining the boron concentration should of course be developed, but this was outside the scope of this one year masters project. This would not change the results to great extent, but it is a feature of the MURE code that should be made.

Figure 4-9 shows the multiplication plotted as a function of the degree of boron in the coolant. This was the basis for how much initial boron to put in the water. From this figure it is quite clear that ^{233}U is a better fissile material than ^{235}U, since the multiplication of the thorium-based fuel is less affected than that of the uranium-based one, because there are more free neutrons from each fission of ^{233}U than ^{235}U. Also, the effect of boron seems to be bigger for UOX, which is more affected when the concentration of boron is increased. More boron is needed to get the same effect for the thoriated fuel.

5. The nuclear fuel cycle

By converting fertile isotopes into fissile isotopes, one can increase the recoverable energy content from the world's uranium and thorium resources. ^{238}U and ^{232}Th can be converted into ^{239}Pu and ^{241}Pu, and ^{233}U, respectively, which all have large fission cross sections for thermal neutrons and substantial fission cross sections for fast neutrons [9]. The conversion of ^{232}Th starts with the thorium isotope absorbing a neutron, and then decaying by two subsequent β$^-$ into ^{233}U. The half-life of the ^{233}Th into ^{233}Pa is 22.3 minutes, and then the protactinium decays into ^{233}U in 27.0 days.

The rate of fertile-to-fissile isotopes depends on the number of neutrons in excess of those needed to maintain the chain fission reaction that are available; η. The fertile-to-fissile conversion characteristics depend on the fuel cycle and the neutron energy spectrum. For a thermal neutron spectrum, ^{233}U has the largest value of η of the fissile nuclei. Therefore, the best possibility for fertile-to-fissile conversion in a thermal spectrum is with the ^{232}Th-^{233}U fuel cycle.

A quantity that is often used to evaluate fuel performance is the conversion ratio (CR), defined as the ratio of fissile material created to fissile material destroyed. When the conversion ratio is greater than unity, it is conventional to speak of a breeding ratio; producing more fissile material than consumed.

$$CR = 1 + \frac{M_f - M_i}{M_{fissioned}}$$

If a reactor breeds its own fuel, like the LMFBR, the fuel cycle is said to be closed. If all the spent fuel is considered waste – an once through cycle (OTC) – the fuel cycle is said to be open.

5.1 The thorium fuel cycle

The nuclear fuel cycle involve all steps from mining of the uranium, to deposition of the waste. It is conventional to speak of the front end of the fuel cycle as everything involving manufacturing the fuel, and the back end as fuel reprocessing and waste management/storage. The front end of the fuel cycle is shown on the left side of Figure 5-1 [26], and on the right side is the back end.
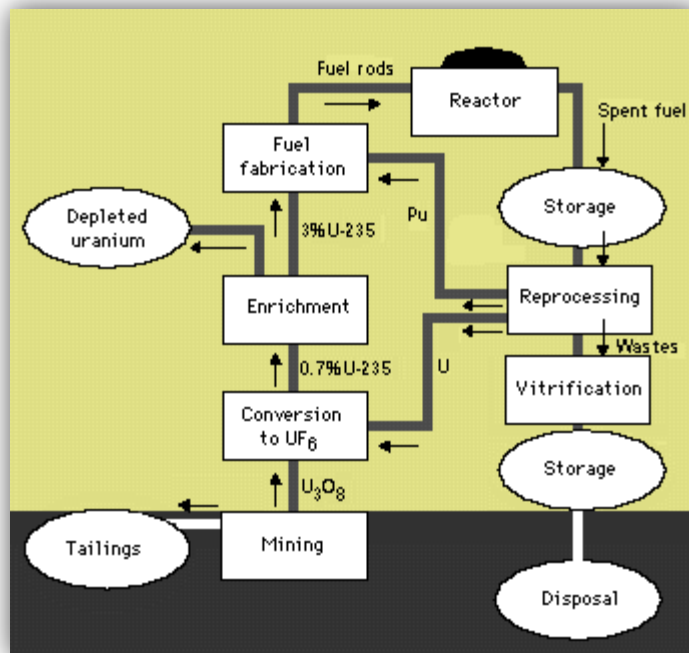
Figure 5-1: The nuclear fuel cycle

5.1.1 The independent thorium cycle (closed fuel cycle)

In the closed thorium fuel cycle the conversion ratio is greater than one and more ^{233}U is produced from the thorium than what is consumed during the chain reaction. This means that once the reaction is started, there will be no need to add any extra fissile material. Since ^{233}U does not exist in nature, the thorium cycle (dependent or independent) has to be started either using an external neutron source to produce the initial ^{233}U, such as neutrons produced by spallation in an ADS, or by adding a fissile material as an internal source in the fuel (^{235}U/^{239}Pu). If an external neutron source is used for the conversion of thorium into ^{233}U, then no mining of uranium is needed. If ^{235}U or ^{239}Pu is used as neutron source, only very little mining is necessary – just to get the fuel cycle started at the initial loading.

The neutron budget in the thermal energy range is very tight; from the average of 2.5 neutrons produced per fission, a maximum of 0.3 neutrons can be absorbed in the fuel or other places in the reactor if breeding is to be achieved.

Reactor concepts that are proposed with the thorium cycle in mind are the ADS and the TMSBR:

The thorium molten salt breeder reactor

The TMSBR is a thermal breeder operating on the ^{233}U/Th cycle. The fuel, fertile material, and coolant are mixed together in one homogeneous fluid, which is composed of various fluoride salts – much research has focused on lithium and beryllium additions to the salt mixture. The reactor core consists of an assembly of graphite moderator elements arranged to allow the flow of the molten salt mixture at some 700°C and at low pressure. When the fluid passes trough the core, the system becomes critical and the fission energy is absorbed directly in the fluid. The heated fluid then passes through a heat exchanger and returns to the reactor. [24]

The ^{233}Pa isotope is removed from the circulating fluid and stored, because it has a large absorption cross-section for thermal neutrons (about 41 b). The ^{233}U is eventually separated from the decaying ^{233}Pa, a portion is returned to the reactor fluid, and the excess is used for fuel in other TMSBRs. The fission products dissolve in the salt and are removed continuously in an online reprocessing loop and replaced with fertile ^{232}Th. Actinides remain in the reactor until they fission or are converted to higher actinides which do so, therefore of this there is no accumulation of fission products in the fuel necessitating periodic fuel changes, and the reactor can operate for long periods between shutdowns. [24]

The molten salt reactor concept was studied in depth in the 1960s, but is now being revived because of the availability of advanced technology for the materials and components. There is now renewed interest in the concept and it is one of the six Generation IV designs selected for further development.

The accelerator driven system

The ADS is a subcritical reactor, which means that there is not enough fissile material in the fuel to sustain a chain reaction; an extra supply of neutrons from outside is therefore necessary. These are high-energy neutrons, which are produced through the spallation of high-energy protons from an accelerator striking heavy target nuclei (lead, lead-bismuth or other material). In such a subcritical nuclear reactor the neutrons produced by spallation would be used to cause fission in the fuel, assisted by further neutrons arising from that fission.

The core of an AD is mainly composed of thorium, located near the bottom of a 25 metre high tank. It is filled with some 8000 tonnes of molten lead or lead-bismuth at high temperature – the primary coolant, which circulates by convection around the core. An accelerator supplies a beam of high-energy protons down a beam pipe to the

spallation target – the lead or lead-bismuth – inside the core, and the neutrons produced enter the fuel and transmute the thorium into ^{233}U. The neutrons also cause fission in the uranium, plutonium and possibly other TRUs present. Since the ADS is subcritical when running, some see this as an enhanced safety feature.

The other role of ADS is the destruction of heavy isotopes, particularly actinides, but also longer-lived fission products such as ^{99}Tc and ^{129}I. The European MYRRHA-project, started in 1997 in Belgium to develop an ADS prototype, which is scheduled to be finished in 2018, is such a project – destruction of long-lived waste.

The breeder technology, however, is many years into the future: First, more *research and development* is required to even build a *demonstrator*. Then a *prototype* of the reactor must be built, before the *industrialization* begins. After all this there can be *wide-scale deployment*. Each of these five phases last at least 10 years, and a TMSBR, for example, is probably at least 50 years into the future. Just the construction of a nuclear power plant including licensing and environmental assessments takes between 7 and 10 years.

5.1.2 The dependent thorium cycle

Since the neutron budget is so tight, and breeding is difficult to achieve with today's technology, the dependent thorium cycle – with existing technology – is a better option for the near future.

If the fuel cycle has a conversion ratio which is less than one, and less fissile ^{233}U is produced than is consumed, extra fissile material is needed to keep the reactor going. We do not have the neutron budget, and we have to bring in extra neutrons from outside. This thorium cycle is therefore **dependent** of extra supply of fissile material from the uranium cycle. There are two sources of neutrons from nature; ^{235}U from natural uranium, or ^{239}Pu produced from other reactors using UOX. Therefore, mining of uranium is still necessary.

The major advantage of the dependent thorium cycle is that no new technology has to be invented [2].

5.2 Front end

The front end of the (dependent) thorium fuel cycle begins with mining of uranium and thorium. The next step is enriching the uranium, (for this project to 20 and 90% ^{235}U).

Uranium has to be enriched in order to increase the ratio of fissile to fertile material; to achieve a chain reaction. The enrichment process takes advantage of the difference in weight of the two isotopes in natural uranium – with atomic masses of 235 and 238. The process has been dominated by the gaseous diffusion method; where UF_6 gas under high pressure is lead over porous membranes. The gas molecules with the lightest isotopes will diffuse through the membrane faster (all the molecules have the same kinetic energy, and their velocity will be proportional to the mass squared). To gain an enrichment of just 3% the gas has to pass through around 1000 membranes, therefore the process is very energy demanding. Actually 5 to 10% of a reactor's produced energy is consumed by enrichment [14]. Other, and more economical enrichment processes are taking over, like the centrifugal method.

The enriched uranium, mostly in the form of UO_2, is transported to the fuel factories where the fuel elements are manufactured, and the transported to the nuclear power plant to be placed in the reactor.

5.3 Back end

Reactors operating on the thorium cycle will initially have fuel which contains ^{232}Th and ^{233}U or ^{235}U. Because there is a substantial production of ^{233}U in such fuels, recycling of the uranium vector is attractive. Recycling uranium implies recycling all of the uranium isotopes; there is no separation of various isotopes in the reprocessing step[8], and therefore the recycled fuel will contain the various isotopes produced in the transmutation-decay processes of uranium: The fissile ^{233}U and ^{235}U, and other isotopes that are created during irradiation in the reactor. Some of these are just parasitic absorbers, like ^{234}U, which have large absorption resonance cross-sections, and will tend to be enriched when uranium is recycled over and over. ^{236}U is produced by neutron capture on ^{235}U (and by electron capture in ^{236}Np), and is another neutron absorber with a significant capture resonance cross-section. Reprocessed uranium is made difficult to handle by the decay product ^{208}Tl, which emits a 2.6-MeV gamma with $t_{1/2}$=3.1 min. This radioisotope is produced by a series of alpha decays of ^{232}U, which is produced by a (n, 2n) on ^{233}U.

When the spent fuel is taken out of the reactor it has to cool, normally more than 100 days (because of the activity of ^{237}U that has a half-life of ~6 days). For this project the

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[8] This would really be a new enrichment step – only more expensive than enriching natural uranium, due to cost of the reprocessing and the necessity of remote handling.

fuel cooled for 10 years before any uranium recycling was performed. After interim storage under water for cooling, the choice has to be made between three options: final disposal of the spent fuel elements, long-term intermediate storage with the aim of later reprocessing, and short term interim storage and reprocessing. If the fuel is to be reprocessed it will, after the cooling period, be transported to a reprocessing plant, like Sellafield (Windscale), Cap de la Hague or Marcoule. Reprocessing is started after about six months to several years of cooling time, with the aim of *recovering the fissile and fertile material.* Reprocessing of U-Th mixtures comprises separation of uranium, (plutonium), thorium and the fission products, including the other actinides. The uranium must be separated from fission products and other actinides, if it shall be useful as fissile material again. At the reprocessing plant the fuel elements are taken apart and the fuel is chopped if necessary. In general, the fuel is dissolved in HNO_3, or another suitable acid, and the unfissioned uranium and plutonium is recovered and can be brought back into fuel fabrication [14]. More aggressive chemicals must be used in the process of recycling the thorium based fuels, because of the stability of thorium dioxide, thus increasing corrosion problems in the reprocessing plant [3]. With respect to activity of the fuel, remote control of the operations is necessary.

The actinides produced in neutron induced decay of the fuel isotopes, and the fission products, are the major contributors to the radioactive waste produced in nuclear reactors. Short-term radio toxicity of the spent fuel is dominated by fission products, which account for almost the entire radioactivity of spent fuel at reactor shutdown, but because of their relatively short half-lives, this radioactivity level decay rather quickly (see Figure 6-1). Only isotopes with long half-lives produced during reactor operation present the truly long-term challenges for waste disposal. The actinides constitute quite a small part of the total radioactivity at reactor shutdown but become relatively more important with time because of the longer half-lives of ^{239}Pu and ^{240}Pu (conventional UOX fuel), and ^{233}U for the Th-cycle, and dominate the radioactivity of spent fuel after about 100 years. Therefore, long-term potential radio toxicity of spent fuel arises principally from the presence of transuranic actinides (Pu and the so-called minor actinides Np, Am, Cm, etc.) all originating with the neutron capture in ^{238}U. *These elements are all radioactive, and may produce significant amounts of heat, for hundreds of thousands of years.*

5.4 Costs

The major expense is the capital cost of building the reactor. However, this will be the same whether or not the reactor will be run on conventional or thoriated fuels. Building a 900 MW nuclear reactor costs typically $1.3 \cdot 10^9$ euros [27].

5.4.1 Fuel

The costs at the front end of the fuel cycle, whether it is the conventional OTC UOX cycle, or a Th/U cycle are the price of the materials – the mining of uranium and thorium, and the enriching of the uranium. Then there is the price of the initial fuel fabrication.

The cost of enriching the natural uranium is measured in separative work units (SWU). The SWU is a unit which is a function of the amount of uranium processed and the degree to which it is enriched. It is a measure of the quantity of separative work (indicative of energy used in enrichment) when feed, tails and product quantities are expressed in kilograms.

The number of SWUs provided by an enrichment facility is directly related to the amount of energy that the facility consumes. Modern gaseous diffusion plants typically require 2 400 to 2 500 kWh of electricity per SWU.

In addition to the SWUs provided by an enrichment facility, the other important parameter that must be considered is the mass of natural uranium (feed) that is needed in order to yield a desired mass of enriched uranium. As with the number of SWUs, the amount of feed material required will also depend on the level of enrichment desired and upon the amount of ^{235}U that ends up in the depleted uranium. However, unlike the number of SWUs required during enrichment which increases with decreasing levels of ^{235}U in the depleted stream, the amount of raw uranium needed will decrease with decreasing levels of ^{235}U that end up in the tails.

Example

In the production of enriched uranium for use in conventional UOX fuel in a LWR it is typical for the enriched stream to contain 3.6% ^{235}U (as compared to 0.72% in natural uranium) while the depleted stream contains 0.2% to 0.3% ^{235}U. In order to produce one kilogram of this enriched uranium it would require approximately 8 kilograms of raw uranium and 4.5 SWU if the tails stream was allowed to have 0.3% ^{235}U. On the other hand, if the depleted stream had only 0.2% ^{235}U, then it would require just 6.7 kilograms of raw uranium, but nearly 5.7 SWU of enrichment. Because the amount of raw uranium required and the number of SWUs required during enrichment change in opposite directions, if raw uranium is cheap and enrichment is relatively more expensive, then the operators will typically choose to allow more ^{235}U to be left in the tails stream whereas if raw uranium is relatively more expensive and enrichment is less so, then the opposite would be chosen.

5.4.2 Reprocessing

At the back end are the costs of reprocessing of the highly active, spent fuel, and management of waste. Thoriated fuels have to be handled remote because of the strong gamma emitter ^{208}Tl, which results from decay of the ^{232}U isotope. This makes reprocessing of thoriated fuels more expensive than reprocessing of UOX fuels, since the UOX fuel in principle can be handled in a glove-box.

5.4.3 Radioactive waste disposal

Disposal of the radioactive waste would presumably be done by placing the spent fuel assemblies (no reprocessing) in suitable containers and burying these containers in some stable geological setting. Historically, stable rock formations have been considered for this purpose.

Reprocessing of the irradiated fuel substantially reduces the volume and the activity – thus the decay heat – of the waste. The waste, in liquid form, can then be calcinated – that is, dried at high temperature; mixed with *frit*, the substance from which glass is made; and then vitrified – that is, made into glass. There are also other methods, but they all *solidify the waste, effectively immobilizing the radioactive particles;* something that cannot be done as easily with unprocessed spent fuel. The glass or ceramic complexes are finally placed in canisters and deposited in stable geological formations [24]. Since both the physical volume of the waste and the heat generated from it, is reduced after reprocessing, substantially smaller repositories are needed for the final storage of the canisters. Reprocessing therefore result in money saved on the final storage.

5.5 Multirecycling of thoriated fuels in the EPR

The OTC, where spent fuel is disposed of as high-level waste (HLW), is the cheapest fuel cycle in the short term. Also, there is a policy against reprocessing, motivated by proliferation concerns. However, the potential energy content of the residual fissile material (Pu and U) is lost in the OTC. Countries like the USA and Sweden do not reprocess due to the proliferation concerns, while e.g. France and Great Britain do reprocess. The potential energy content of the fissile and fertile isotopes remaining in spent reactor fuel constitutes a substantial fraction of the potential energy content of the initial fuel loading, providing an incentive to recover the uranium and plutonium isotopes for reuse as rector fuel. Only about 1% of the energy content of the uranium used to produce the fuel is extracted (via fission) in a typical LWR fuel cycle. The OTC

also gives the largest possible volume of HLW, which must be stored in geological repositories for hundreds of thousands to millions of years. About 3% of the energy content of the mined uranium is stored as tails from the original uranium fuel production process, and about 96% remains in the discharged spent fuel in the form of uranium, plutonium (TRUs).

With fuel recycling it is possible both to reduce the mass of HLW that must be stored in geological repositories and the time of high radio toxicity of that HLW, reducing the requirements for both the number of repositories and the duration of secured storage. With continued reprocessing and recycling of spent fuel, there is the possibility of
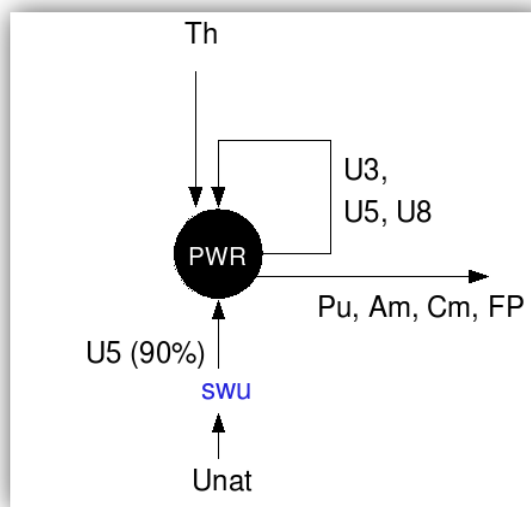
*Figure 5-2: The basic
principle of the project*

recovering much of this remaining energy.

The idea for this project was to make use of the dependent thorium cycle in an EPR, with ^{235}U as a neutron source. By mixing thorium and highly enriched uranium (HEU), and then multirecycling the uranium vector, very "clean" waste is produced. The principle is shown in Figure 5-2: what goes into the reactor is natural uranium that has to be enriched, up to 90% ^{235}U, and thorium. Then the plutonium, americium, curium, and other TRUs are taken out as waste, together with the fission products, while all the uranium is recycled, and refabricated into fuel with more fresh thorium and enriched ^{235}U. The aim is to strongly reduce the production of TRUs, like plutonium, curium, americium, and neptunium, and this is possible by reducing the main waste precursor, ^{238}U. If this is achievable the waste from this cycle will be less active, hence producing less decay heat, and the cost of the waste storage diminish considerably. Elimination of ^{238}U necessitates high enrichment of ^{235}U, and addition of thorium.

During the reactor operation the thorium will be converted into fissile ^{233}U, and recycled uranium will consist of a lot of both ^{233}U and ^{235}U. However, the entire uranium vector will have to be reprocessed, and this will also consist of $^{232/4/6/8}$U. The waste will be substantially less active than that from conventional UOX fuel, and it will also be active for a much shorter period of time (see details in Chapter 6.1.2). This means that the cost of storing waste fuel will be less, because it can be stored in smaller geological repositories.

The continued recycling of spent fuel would lead, after long exposure, to equilibrium distributions of the uranium isotopes in the recycled fuel.

5.5.1 Fuel compositions

Two main fuel compositions were studied:

    I.   **Thorium and 20% enriched uranium**

    II.   **Thorium and 90% enriched uranium**

Enriching uranium to 20% is the highest enrichment defined as reactor grade uranium. Therefore the first fuel composition to be chosen is thorium and 20% enriched uranium, since this is the highest enrichment that is "unproblematic". With higher enrichment than 20% the uranium is classified as weapons grade material. However, higher enrichment is interesting to study, since the ^{238}U content goes down as enrichment goes up (see Figure 5-4). Therefore the second fuel composition studied is a mixture of thorium and 90% enriched uranium; to really minimize the waste precursor of the heavy uranium isotope. Another reason for burning HEU in reactors is that the already existing stockpiles of weapons uranium is destroyed as weapons material, and still its potential energy content is fully exploited – as opposed to deenriching the weapons uranium. Higher enrichment than 90% has not been studied, because it would not change the results to a great extent, whilst the costs of enriching the uranium would be too large (do not gain in enriching higher).

As reference to the two main cases, the OTC for other types of fuel was studied as well:

    1.   "Normal" UOX, 4.5% enriched

    2.   Thorium and (reactor grade) plutonium

    3.   Uranium and (reactor grade) plutonium, "normal" MOX

    4.   Thorium and ^{233}U

The reference cases represent the other possible fuel mixtures for a PWR.[9]

Reactor grade plutonium is that plutonium vector that would be reprocessed from UOX fuel after a normal burn up of 2-3 years. The isotopic composition of reactor grade plutonium is showed in Table 5-1.

The plutonium vector is composed of the plutonium isotopes 238, 239, 240, 241, and 242. There is also some ^{241}Am, that originates from decay of the ^{241}Pu isotope. Weapons grade plutonium on the other hand, consists mostly of ^{239}Pu, and this is produced from a reactor with large ^{238}U content and a short burn up time.

Only parts of the core is filled with MOX fuel in today's operating reactors. This is because the delayed neutron fraction of the plutonium is smaller than that of ^{235}U, and therefore a full MOX core is considered less safe in the existing PWRs. The EPR, however, is designed for a 100% MOX core, and therefore this is also taken as the MOX reference case [23]. It is therefore also a reasonable assumption that a full EPR core of Th/HEU is safe; after all the delayed neutron fraction of ^{239}Pu is smaller than that of ^{233}U. (See Table 3-2.)

Figure 5-3: The different fuel mixtures, initial loading of reactor

All but the ^{235}U/Th fuel compositions have been OTCs; the 20 and 90% cases were multi recycled. That is, after the burn-up time of 1080 days the fuel is taken out and all of the uranium is reprocessed. The multi recycling was continued until the inventory reached
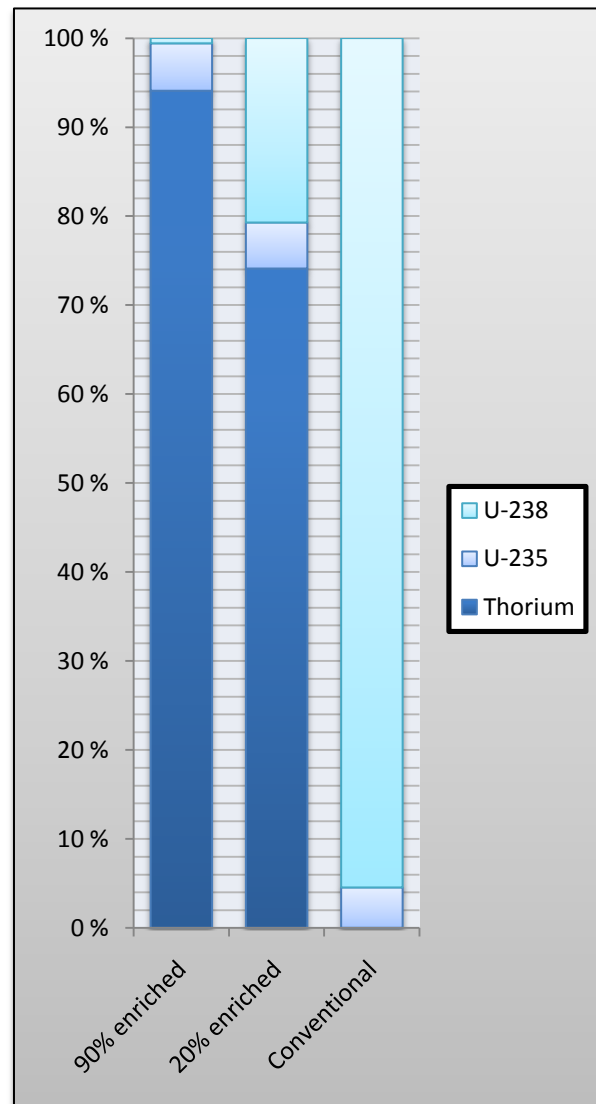
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[9] The mixture of thorium and pure ^{233}U is, however, just a hypothetical fuel case. It is studied because it shows the limit of how good the result may possibly be.

equilibrium, and there was no difference in how much fresh, ^{235}U that has to be added to the new fuel.

Table 5-1: Reactor grade plutonium

	Pu				^{241}Am
238	**239**	**240**	**241**	**242**	
0,027009	0,59236	0,259340	0,074262	0,073072	0,007021

The fuel mixtures of Th/UOX 90% enriched, and Th/UOX 20% enriched, as well as the reference UOX case, is shown in Figure 5-3. *It must be emphasized that at all times the fissile content in the fuel never exceeds 6%!* As can be seen in the figure, the degree of enrichment is given for that part of the fuel which actually is uranium. For conventional fuel it is necessary with 4.5% ^{235}U, with 20% (initial) enrichment there will be a total of 5.17% ^{235}U in the fuel, and for the 90% (initial) enriched case 5.33% of the fuel will be ^{235}U. This is for the initial fuel loading with thoriated fuels, and for the recycled cases the ^{235}U content is lower, since there is also a substantial amount of ^{233}U. The total summary of all different the fuel mixtures are shown in Table 5-2.

Figure 5-4 shows how the fuel content changes when ^{238}U is replaced with HEU and thorium. From Figure 4-7 – the waste precursors, it is clear that with smaller ^{238}U content, fewer long lived isotopes will be produced.

Table 5-2: Summary, fuel mixtures, initial loading

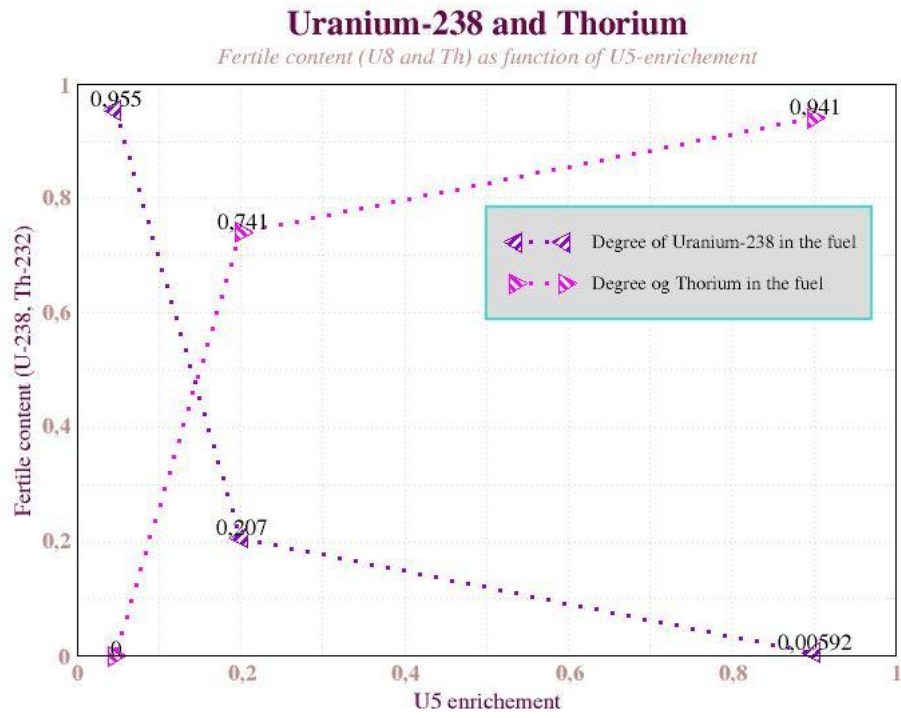|	Th/UOX, 90%	Th/UOX, 20%	UOX	MOX	Th/Pu	Th/^{233}U
^{233}U	-	-	-	-	-	0,042
^{235}U	0,052	0,052	0,045	0,003	-	-
^{238}U	0,006	0,207	0,955	0,907	-	-
Th	0,942	0,741	-	-	0,103	0,958
Pu & ^{241}Am	-	-	-	0,090	0,897	0

Figure 5-4: Replacing ^{238}U with thorium

6. Results

The simulations of the thoriated fuels used in the EPR were evaluated under different criteria:

Waste produced

How much is the HLW reduced? What kind of waste is produced, what is its isotopic composition and how does this influence waste storage requirements?

Safety

How is the safety affected? What are the reactivity feedback coefficients and how do these change if the uranium is recycled? How does the protactinium effect impact reactor safety?

Economics

Will the fuel cycle cost more, and is it possible that this fuel cycle will be economically viable?

There is also a brief discussion of proliferation issues at the end of this chapter.

6.1 Waste produced

After a burn-up two major groups of materials are produced – fission products and TRUs. The quantity of the fission products produced is the same for all types of fuel, and because most of the fission products have short half-lives they are not important with respect to final storage of waste. The decay heat from the fission products for the different fuel types are shown in Figure 6-1; they clearly reach the level of activity of natural uranium very quickly. The contributions from the fission products and the actinides have thus been separated, since only the actinides contribute on the long time scale of final waste storage, of 10^4-10^6 years.

All graphs showing the results for the waste show the decay heat produced, and not the radiotoxicity – a related measure. This is due to the economical focus; that storage of waste is a significant cost, and the more heat generated by the waste, the more space

needed for storage. In other words; less heat from the waste means that the waste can be stored closer together, and more waste can be stored in smaller repositories.

Heat from the Fission Products
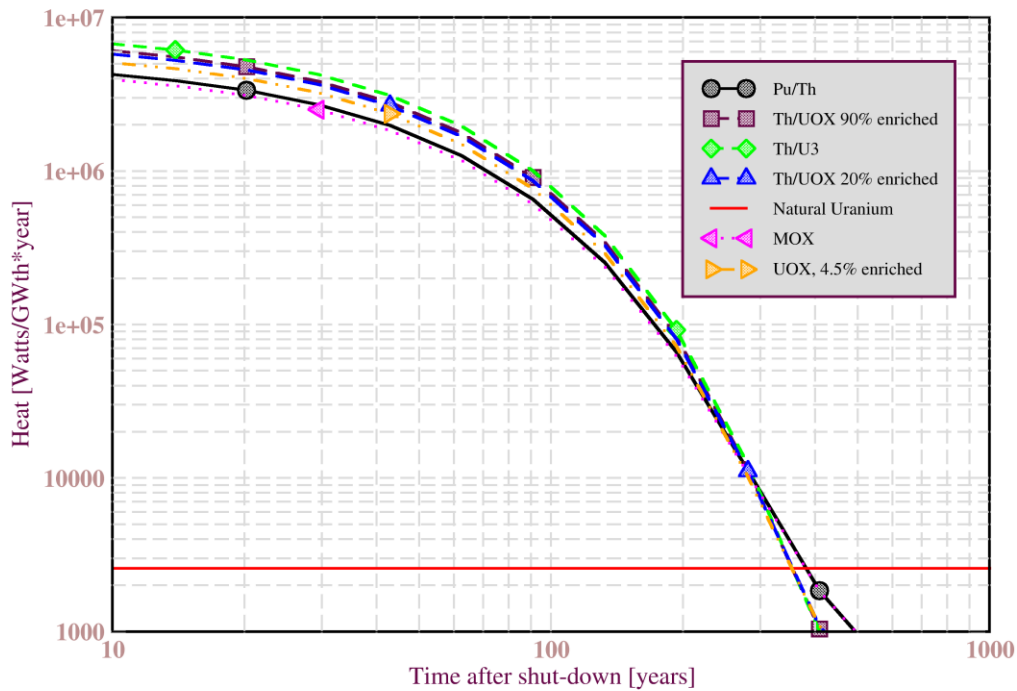
Figure 6-1: decay heat from the fission products, all fuel cycles studied are shown

Table 6-1 and Table 6-2 give an overview of some of the most important isotopes produced and destroyed in the initial loading of thorium and 90 and 20% enriched uranium. The materials of interest are the different uranium isotopes, plutonium, americium, curium, and neptunium. The different uranium isotopes that are produced in the Th/HEU fuels are interesting with respect to reprocessing. Uranium recycling is performed for both fuel cases, but there is a substantially larger fissile content (^{233}U and ^{235}U) in the uranium from the 90% enriched case than the 20% case, where much of the uranium is the ^{238}U isotope. ^{237}U is not shown in the tables because of its short half-life (6.75 days), and it will have decayed into ^{237}Np at that time reprocessing is performed.

Table 6-3, which shows the inventory for the UOX fuel, does not show the uranium isotopes lighter than ^{235}U, since these are practically not produced in the uranium fuel cycle. The initial and final amounts of ^{235}U are practically the same for the Th/HEU fuels and the UOX fuel. Since there is no production of ^{233}U in the UOX case, uranium recycling is not performed, and the amount of fissile ^{235}U is lost in the uranium fuel cycle – and disposed of as waste.

Table 6-1: Inventory of the first run of the Th/UOX 90% enriched

Isotope	Assembly [gram]			Core [kg]
	$M_{initial}$	M_{final}	ΔM	M_{final}
^{232}U	-	29	29	7
^{233}U	-	7 113	7 113	1 714
^{233}U & ^{233}Pa	-	7 669	7 669	1850
^{234}U	-	858	858	210
^{235}U	28 753	8 956	-19 797	2160
^{236}U	-	3 490	3 490	840
^{238}U	3 236	2 688	-548	650
Th	509 912	495 15	-14 758	119330
^{239}Pu	-	126	126	30
Pu, rest	-	182	182	44
Cm	-	1.76	1.76	0.4
Am	-	5.10	5.10	1.2
Np	-	385	385	90

Table 6-2: Inventory of the first run of the Th/UOX 20% enriched

Isotope	Assembly [gram]			Core [kg]
	$M_{initial}$	M_{final}	ΔM	M_{final}
^{232}U	-	21	21	5
^{233}U	-	6 095	6 095	1 469
^{233}U & ^{233}Pa	-	6 517	6 517	1 571
^{234}U	-	654	654	156
^{235}U	28 461	10 149	-18 312	2 446
^{236}U	-	3 257	3 257	785
^{238}U	115 300	109 850	-5 450	26 474
Th	402 814	391 009	-11 805	94 233
^{239}Pu	-	1 430	1 430	345
Pu, rest	-	990	990	239
Cm	-	11	11	2.7
Am	-	36	36	8.7
Np	-	295	295	70.6

Plutonium and the other TRUs are responsible for most of the activity on the long-term scale. It is therefore striking to see the big difference in the amounts produced of these materials in the UOX fuel and the Th/HEU fuels – especially the Th/UOX 90% enriched: No doubt, the smallest quantities of TRUs are produced from this fuel mixture. For example, the UOX fuel produces 30 times more ^{239}Pu.

Table 6-3: Inventory of the UOX fuel, 4.5% enriched

	Assembly [gram]			Core [kg]
Isotope	$M_{initial}$	M_{final}	ΔM	M_{final}
^{235}U	25 069	8 518	-16 551	2 053
^{236}U	-	2 897	2 897	698
^{238}U	538 828	523 206	-15 622	126 093
^{239}Pu	-	4 052	4 052	976
Pu, rest	-	2622	2622	632
Cm	-	28	28	7
Am	-	84	84	20
Np	-	346	346	83

6.1.1 Once through cycle

Figure 6-2 shows the total decay heat from the waste from all the fuels studied, (minus

Total Decay Heat

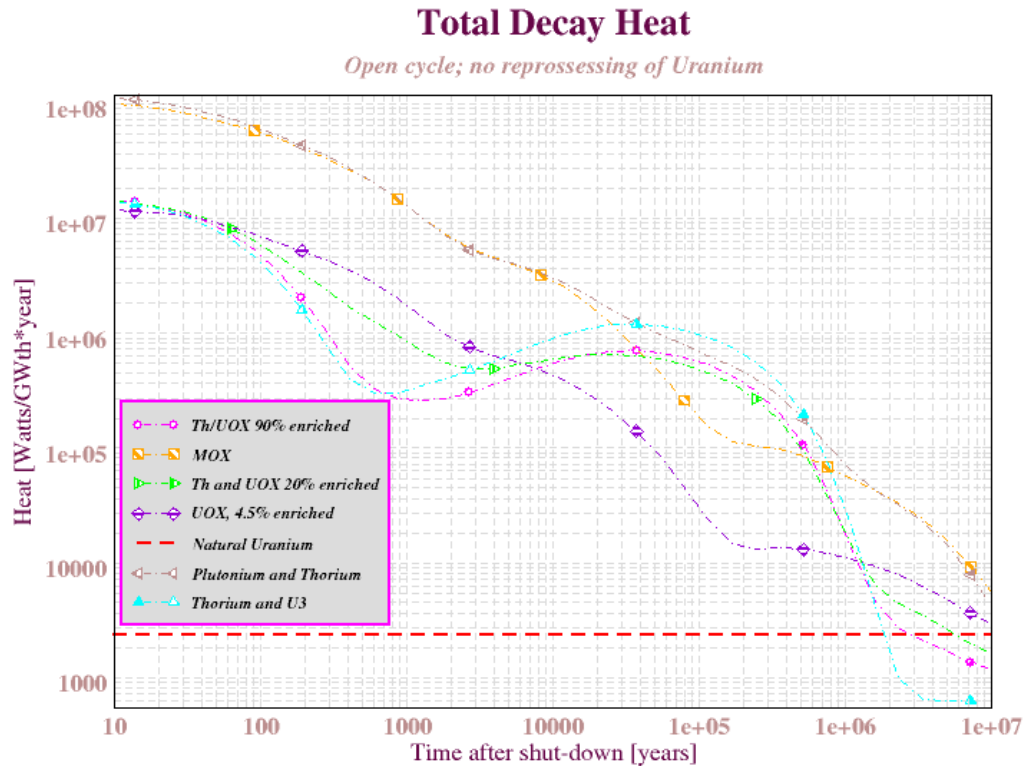*Open cycle; no reprossessing of Uranium*

Figure 6-2: Total decay heat from the TRUs, no reprocessing of uranium

the fission products), without reprocessing of uranium. The red baseline, in all the waste heat figures, is showing the activity from that amount of natural uranium needed to produce the 4.5% enriched conventional UOX fuel.

The heat production is almost the same for the Th/HEU fuels as the UOX fuel for the first 100 years after it is taken out of the reactor. It is about a factor of 10 less than the MOX and the Th/Pu. The two plutonium fuels are thus the real producers of large quantities of long-lived actinides; of course because of their initial plutonium content, which leads to a large content of plutonium and heavier TRUs. However, if the uranium

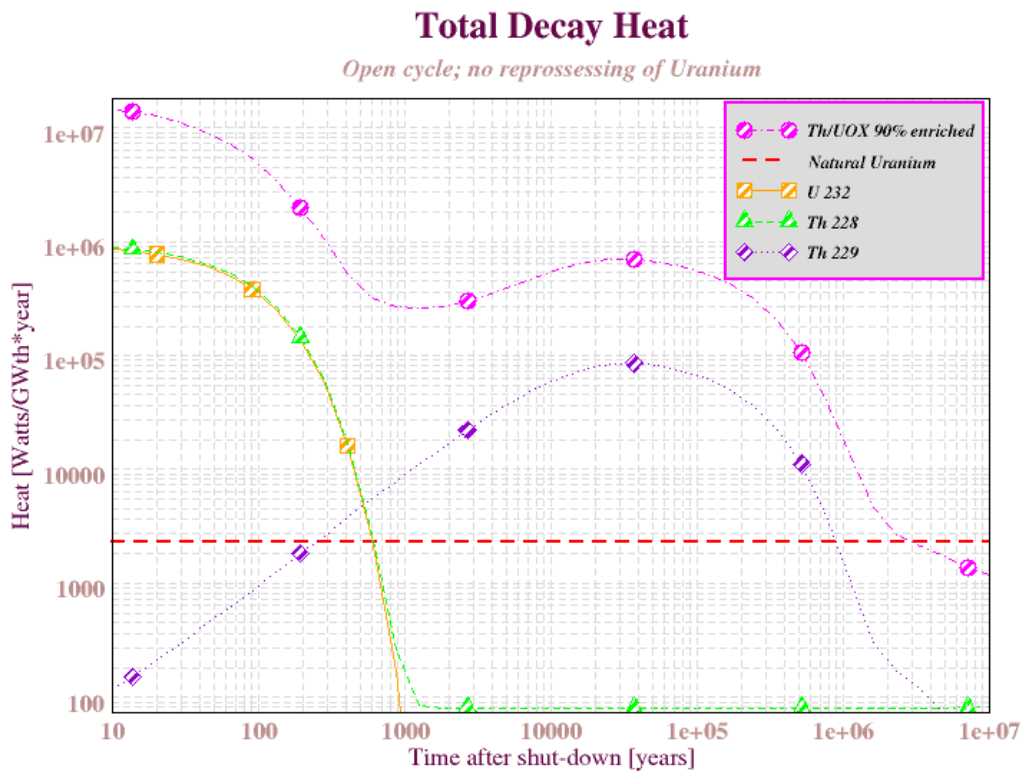is not recycled from the Th/HEU fuels, the decay heat from this will be even

Total Decay Heat

Open cycle; no reprossessing of Uranium

Figure 6-3: Total decay heat from the TRUs, Th/UOX 90% enriched no uranium reprocessing; details

larger than that from normal MOX-fuel or plutonium and thorium, after 100 000 years. The two "bumps" of the Th/HEU fuels are caused by decay of the two uranium isotopes ^{232}U and ^{233}U, and all of their radioactive daughter products. This is shown in more details in Figure 6-3. ^{232}U has a half-life of 68.9 years, and this isotope together with its daughters dominate the activity for the first 300 years. In the figure both the heat from the ^{232}U and her daughter, ^{228}Th following the exact same pattern as the mother nuclide due to the short half-life of the thorium isotope (compared to the mother), is shown. After this time, the ^{233}U, with a half-life of 159 200 years, with its daughter products start to contribute. Only the daughter of ^{233}U, ^{229}Th, is shown in the figure. The two uranium isotopes both have very active, short-lived daughter products – 8 and 9 before ending in stable ^{208}Pb and ^{209}B, therefore their activity follows the same pattern as their uranium mother; as is clearly shown with the ^{232}U and the ^{228}Th.

Because of the production of ^{232}U and ^{233}U, there is no point in using thoriated fuels if uranium-recycling is not performed.

6.1.2 Recycling of the uranium

Figure 6-4 shows the waste heat from all the different fuel types, when the uranium is recycled for the Th/HEU-cases. The difference from Figure 6-2 is evident: Since the uranium is reprocessed there are no uranium daughters, like ^{228}Th and ^{229}Th. 10 years after the spent fuel has been taken out of the reactor, it produces only one seventh of the heat that the conventional UOX fuel produces. After 1000 years the heat production is as little as 20 times less than the UOX. The 90% enriched case reaches the same heat production as natural uranium after 60 000 years, while it takes 10 000 000 years for the UOX fuel to get down to the same level of activity. The 20% enriched case is unfortunately not as good as the 90% enriched fuel: After 10 years of cooling it produces twice as much heat as the 90% case, but this is still substantially less than the UOX. The reason is that there is a greater production of plutonium, due to the larger content of ^{238}U in the fuel (see Table 6-1and Table 6-2).

The best possible result would be achieved from a clean mixture of thorium and pure ^{233}U. With this hypothetical fuel there would be almost zero production of plutonium, and heavier actinides, like americium. The ^{233}U/Th fuel mixture would start off, after the 10 year cooling period, producing 100 times less heat than the Th/90% enriched, and 1000 times less than the UOX. After just 200 years this fuel would reach the same activity as natural uranium, assuming the uranium is recycled. However, if the uranium is not recycled the Th/^{233}U fuel would be the greatest heat emitter after 150 000 (see Figure 6-2).

In Figure 6-5 the details of the waste heat from the Th/UOX 90% enriched case, uranium recycled, is shown. There are mainly two materials generating heat; these are plutonium and americium. The first "bump" is caused by decay of ^{238}Pu, originating from a neutron capture on ^{235}U:

$$^{235}U(n,\gamma)^{236}U(n,\gamma)^{237}U \xrightarrow{\beta^-} {}^{237}Np(n,\gamma)^{238}Np \xrightarrow{\beta^-} {}^{238}Pu$$
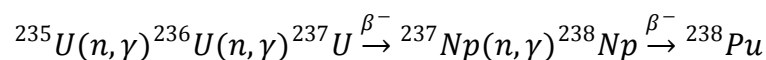
It is thus impossible to get the production of ^{238}Pu down, since ^{235}U is needed to get the chain reaction started. For the recycled fuels there will be smaller amounts of ^{235}U, but there will be certain amounts of ^{236}U (see Table 6-4). The second "bump" is from the decay of ^{239}Pu, which is produced from captures on the very small amount of ^{238}U originally in the fuel. As it turns out, plutonium is still the "problem", but it is minimized (see Table 6-1) since the ^{238}U is exchanged with thorium (see   Figure 5-4). Even though the production of americium is very small, only 1.2 kg in the entire core, it contributes to the waste heat production, between 100 and 1000 years after discharge. Curium and neptunium is of no interest concerning heat produced of the waste, nor does it matter in volume.
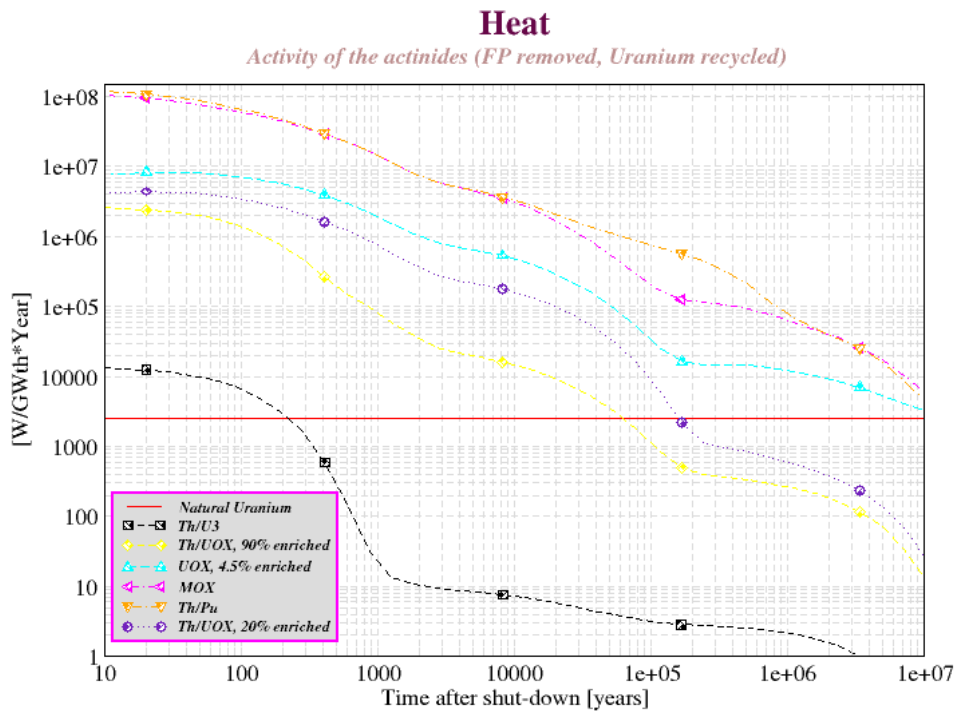
Figure 6-4: Heat generated from the waste when the U-vector is recycled
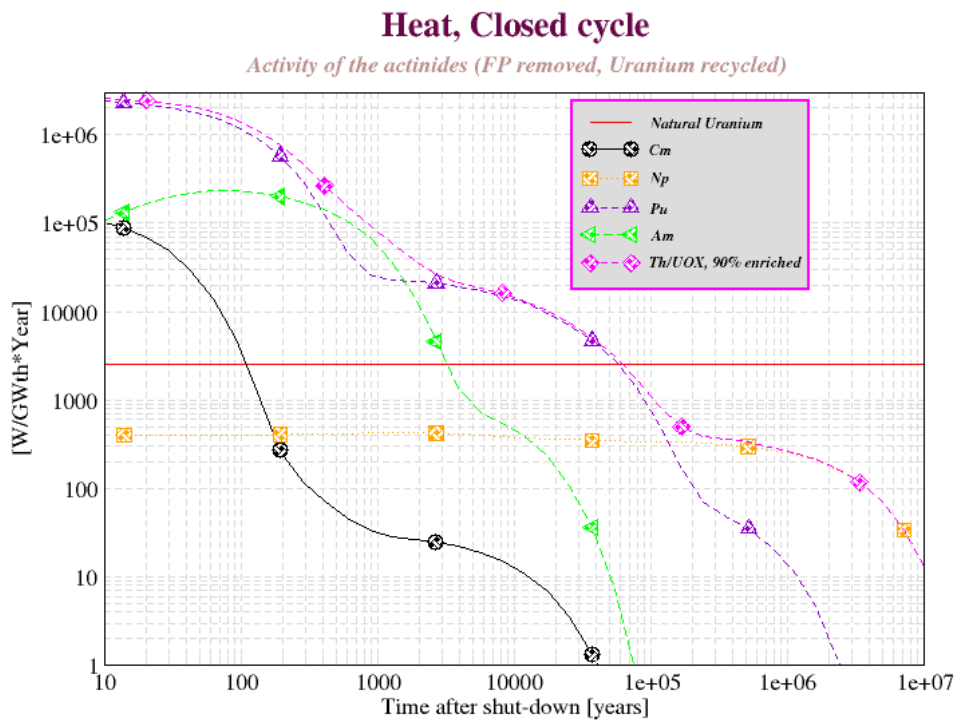

Figure 6-5: Details of the waste heat from the Th/UOX 90% erniched fuel case, initial loading, recycling of the uranium

Multi recycling

Table 6-4: Th/UOX 90% enriched, uranium inventory, EOC (whole core, tons)

Recycle #	^{232}U	^{233}U & ^{233}Pa	^{234}U	^{235}U	^{236}U	^{238}U
0	0.007	1.848	0.207	2.158	0.841	0.648
1	0.011	2.316	0.453	1.518	1.287	0.772
2	0.013	2.474	0.632	1.833	1.693	0.966
3	0.014	2.521	0.770	1.794	2.040	1.089
4	0.014	2.542	0.872	1.876	2.365	1.209
5	0.015	2.551	0.949	1.910	2.667	1.310
6	0.015	2.562	1.010	1.983	2.971	1.413
7	0.015	2.565	1.053	2.067	3.251	1.505

Table 6-5: Th/UOX 20% enriched, uranium inventory, EOC (whole core, tons)

Recycle #	^{232}U	^{233}U & ^{233}Pa	^{234}U	^{235}U	^{236}U	^{238}U
0	0.005	1.571	0.158	2.446	0.785	26.474
1	0.008	1.912	0.330	2.149	1.278	36.776
2	0.008	1.907	0.447	2.489	1.727	49.041
3	0.008	1.774	0.517	2.733	2.159	60.891
4	0.007	1.609	0.551	2.821	2.573	69.891
5	0.006	1.390	0.554	2.958	2.984	81.687
6	0.005	1.127	0.529	2.972	3.385	93.253
7	0.004	0.815	0.475	2.961	3.738	105.163

When the uranium is multirecyled, there is a gradually build-up of unwanted uranium isotopes. A consequence of this is that fresh, enriched uranium has to be added to the fuel, and for each cycle also more ^{238}U will be part of the fuel. As can be seen in Table 6-4 and Table 6-5, the uranium vector that is recycled is shifted towards heavier uranium isotopes. After the initial fuel loading of Th/UOX 90% enriched uranium 0.648 tons of ^{238}U is taken out of the fuel and recycled, after the 7th recycle more than twice that amount is recycled – 1.505 tons of ^{238}U. Since there is gradually more of the heavy uranium isotopes in the fuel, there is also a larger and larger production of plutonium (see Table 6-6 and table 6-7). This, of course, makes the waste more active. The spent fuel from the initial fuel loading, with uranium recycled, is the very best result. Then the spent fuel gradually gets more active, thus producing more heat. However, all in all the result of the multirecycling is good: After the 7th recycle the waste is about twice as active as after the initial loading, which is still much better than the waste from the OTC UOX. This goes for both the 20 and the 90% fuel cases. Nor does it take longer for the waste to reach the level of activity of natural uranium.

Table 6-6: Th/UOX 90% enriched, plutonium inventory, EOC (whole core, tons)

Recycle #	0	1	2	3	4	5	6	7
^{239}Pu	0.03	0.038	0.049	0.054	0.059	0.064	0.068	0.072
Pu rest	0.044	0.074	0.087	0.098	0.107	0.116	0.121	0.127

Table 6-7: Th/UOX 20% enriched, plutonium inventory, EOC (whole core, tons)

Recycle #	0	1	2	3	4	5	6	7
^{239}Pu	0.345	0.448	0.565	0.668	0.743	0.829	0.909	0.982
Pu rest	0.24	0.304	0.347	0.387	0.425	0.463	0.509	0.559

6.2 Safety

The thorium cycle differs from the UOX cycle in safety aspects in some important ways:

1. Delayed neutron fraction, which is similar to that of plutonium based fuels (see Table 3-2)
2. ^{233}Pa reactivity effects, caused by the relatively long half-life of ^{233}Pa, compared to the U/Pu cycle with ^{239}Np half-life of days

The delayed neutron fraction has not been studied, but it is slightly larger than for ^{239}Pu. Since the EPR is designed for 100% MOX core, it is believed that it should also be safe for 100% thorium fuels.

6.2.1 Reactivity feedback

In this section the effects of uranium multirecycling on the main safety feedback coefficients were investigated. The ^{233}U content of the fuel increases with each recycle and it is possible that this could impact the safety.

Fuel temperature coefficient of reactivity

The temperature coefficient of reactivity was defined as $\alpha_T \equiv \frac{d\rho}{dT}$ in chapter 3.4. It is essential that α_T is negative, for passive reactor safety, i.e. any increase in the fuel temperature decreases the reactivity. It is also interesting to see the change in the temperature coefficient as the fuel is recycled, whether it gets better or worse – that is, more or less safe.

The fuel temperature coefficient was found by varying the temperature for the different compositions of fuel, with their respective amounts of boron poison. The multiplication was plotted as a function of fuel temperature, and the slope of this plot is α_T.

The initial temperature coefficient of the fuel for thorium and 90% enriched uranium is $-3.2 \cdot 10^{-5}$. As the fuel is recycled the temperature coefficent stays at $\sim -3.2 \cdot 10^{-5}$, even though it fluctuates. Table 6-8 summarizes α_T for all fuel loadings, and the conclusion that can be drawn from this table is that it does not get better or worse as the uranium is recycled and the isotopic composition is changed. The result is about the same for the 20% enriched case: Table 6-9 summarizes the fuel temperature coefficients of these fuels, and α_T stays at $\sim -3.5 \cdot 10^{-5}$. Both fuels are thus passively safe with respect to fuel temperature reactivity feedback.

The fuel temperature coefficient of reactivity of the Th/HEU does not degrade with multirecycling.

Table 6-8: Fuel temperature coefficient of reactivity, 90% enriched uranium

Recycle #	0	1	2	3	4	5	6	7
α_T	$-3.2 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$	$3.0 \cdot 10^{-5}$	$3.2 \cdot 10^{-5}$	$3.2 \cdot 10^{-5}$	$3.4 \cdot 10^{-5}$	$3.2 \cdot 10^{-5}$
Ppm of boron	2012	2470	2819	2775	2775	2774	2775	2775

Table 6-9: Fuel temperature coefficient of reactivity, 20% enriched uranium

Recycle #	0	1	2	3	4	5	6	7
α_T	$-3.9 \cdot 10^{-5}$	$3.8 \cdot 10^{-5}$	$3.6 \cdot 10^{-5}$	$3.8 \cdot 10^{-5}$	$3.4 \cdot 10^{-5}$	$3.4 \cdot 10^{-5}$	$3.0 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$
Ppm of boron	1878	2300	2527	2711	2727	2782	2790	2800

Void coefficient of reactivity

If voids form in the coolant caused by overheating, or if coolant is lost from the core, the average coolant density will decrease. If this happens the reactivity should also decrease, because when the moderator/coolant loses its density it also loses its ability to slow the fission neutrons: The void coefficient of reactivity should be negative. However, if too much anti-reactivity, i.e. neutron poisons, such as boron, is present in the coolant, this may not be the case.

The void coefficient of reactivity was found by varying the density of the water, for all the different fuel compositions, whith their respective amounts of boron. The multiplication was then plottet as a function of water density, and the results are shown in Figure 6-6 and Figure 6-7.

There is a clear difference between the void coefficient for the 90% enriched case (Figure 6-6) and the 20% case (Figure 6-7). The 90% case is more affected by increase in neutron poison, and is thus less safe than the 20% case. However, both figures show that in the case of a LOCA the reactivity would eventually decrease, effectively stopping the chain reaction. There is only one fuel mixture which is not even safe in the case of a LOCA, and that is the Th/UOX 90% enriched 2nd recycle: If the density decreases, the multiplication increases, and it will not go down to a level below the original.
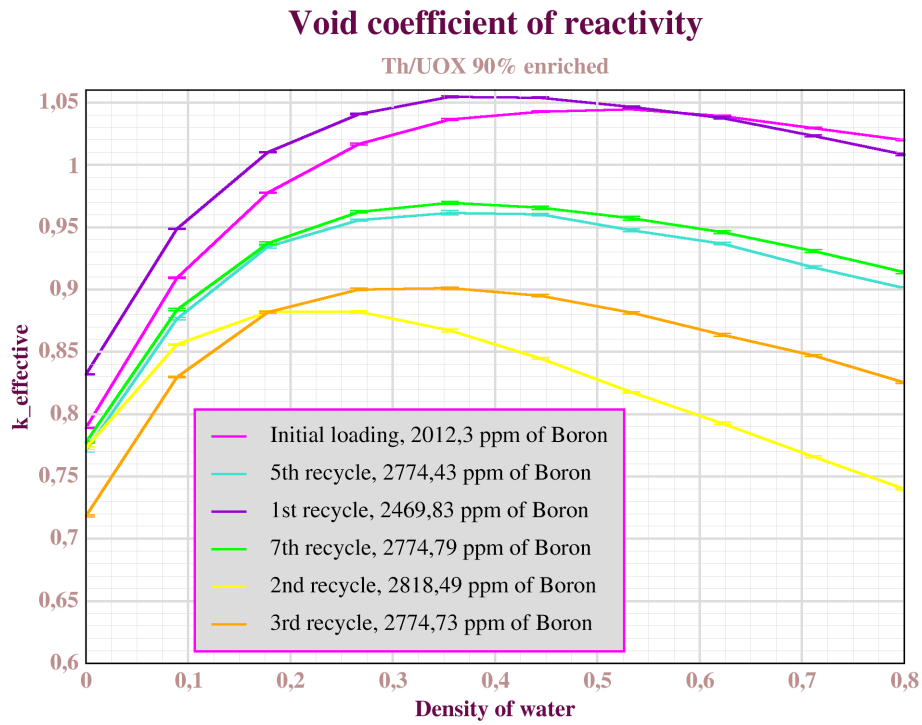
Figure 6-6: Void coefficient of reactivity, Th/UOX 90% enriched, all fuel loadings
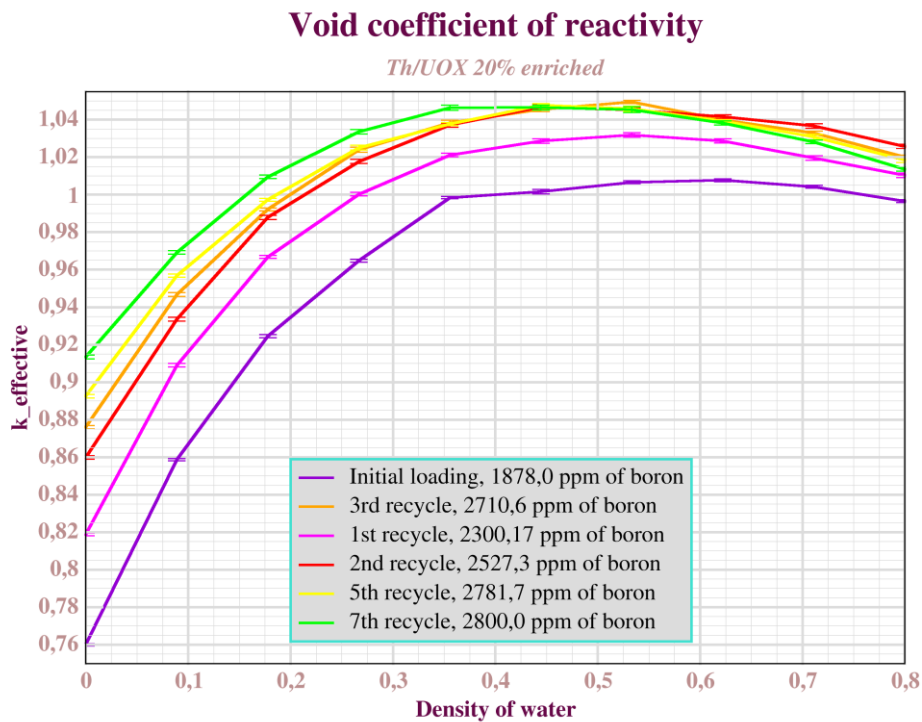

Figure 6-7: Void coefficient of reactivity, Th/UOX 20% enriched, all fuel loadings

This is not acceptable for safe operation, but fortunately this problem can be solved by operating the reactor in batch mode (see Chapter 3.3.1), and using on average 1/3 the amount of boron. This is shown in Figure 6-8, where the initial fuel loading of the reactor, and the 3rd recycle, for both the 20% enriched case and the 90% enriched case are run with 1/3 that amount of boron that was initially needed to keep the core at criticality for 1080 days. In this figure the multiplication does not increase when the moderator density is decreased: The void coefficient of reactivity is thus safe for both thorium fuels.
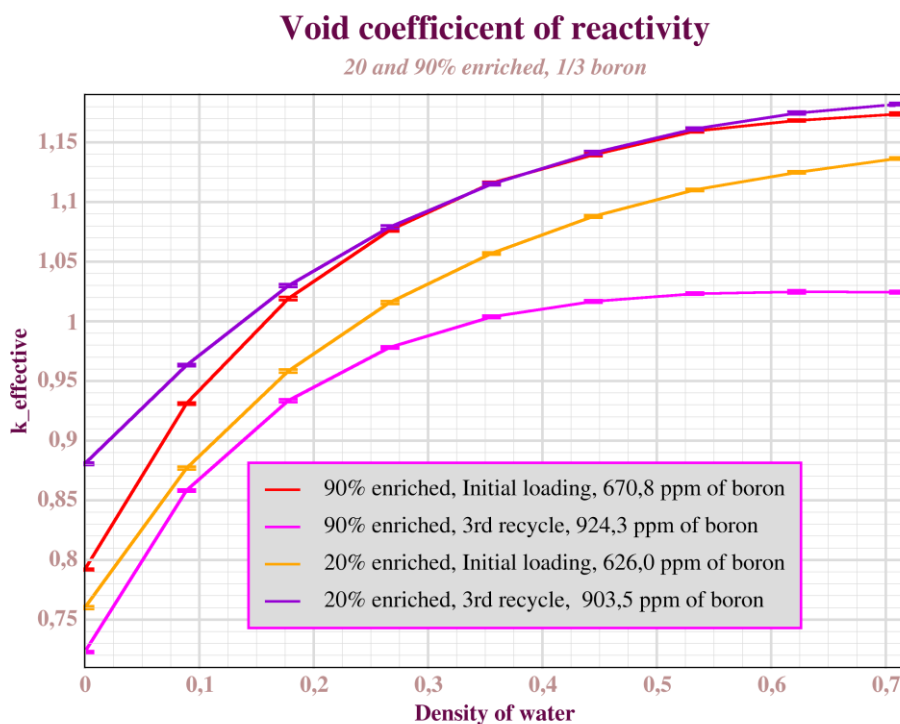

Figure 6-8: Void coefficient of reactivity, Th/UOX 20% and 90% enriched, Batch mode, i.e. 1/3 the amount of boron poison

The conclusions that can be drawn from this are that more boron is needed when the uranium is recycled; since a bigger part of the recycled uranium will be ^{233}U. The higher the concentration of boron in the moderator, the more dangerous that the density of the water decreases; as boron absorbs neutrons. Also, the Th/UOX 90% enriched fuel needs more boron, and has thus smaller margins with respect to the void coefficient of reactivity than the 20% enriched case.

Simulation of accident scenarios such as LOCA using coupled neutronics/thermalhydraulics and point kinetics computer codes could be performed as a continuation of this project. The trajectory of the reactor power in the first few seconds

to minutes of the simulated accidental/incidental scenarios needs to be compared for both thoriated fuels and standard UOX fuels to see if there are significant differences in core behavior.

6.2.2 Effect of protactinium

The objective of a thorium cycle is to produce as much ^{233}U as possible. ^{233}U is produced by beta minus decay of ^{233}Pa, which has a half-life of 27 days. This means that when the reactor is shut down, there will always be a certain amount of protactinium that has not yet decayed into fissile uranium. The issue with the protactinium effect is that for more than one month after the reactor is shut down more and more fissile material will build up inside the reactor core. When all the control rods are inserted to shut down the chain reaction, the total anti reactivity produced by the rods must be larger than the reactivity that builds up over the next month. *The reactor core must not go critical after all the control rods are inserted*! This protactinium effect is an issue with all reactors employing the thorium cycle, whether or not they are critical when running.

^{233}U is a better fissile material ^{235}U, in the sense that only small quantities are necessary to maintain a chain reaction. When checking if the EPR is safe for the reactivity build-up after it is shut down, this fact must be taken into account, since the fissile material in the used fuel will be a composition of ^{233}U and ^{235}U. From Table 6-1 it is seen that there will at shut-down initially be 8 956 grams of ^{235}U, 7 113 grams of ^{233}U, and 556 grams of ^{233}Pa; that is 16 069 grams of fissile material. In one assembly there will be a total of 518 958 grams of spent fuel.

$$\frac{16\,069\ g\ fissile}{518\,957\ g\ total\ fuel} \approx 3.1\%\ fissile$$

At shut-down the spent fuel will consist of 3.1% fissile material, but after all the protactinium has decayed there will be a total 16 625 grams of fissile material, or ~3.2% of the spent fuel. The 16 625 grams of fissile consist of 46% ^{233}U, and 54% ^{235}U. 3.1% fissile corresponds to a k_{eff} of 1.0176 if the fissile is ^{233}U, and 0.92 if it is ^{235}U. These values were found by plotting the multiplication as a function of the fissile content – being either ^{233}U or ^{235}U – in the fuel.

The corresponding values for 3.2% fissile in the core are 1.0342 for ^{233}U and 0.9307 for ^{235}U.

$$0.46(1.0342 - 1.0176) + 0.54(0.9307 - 0.92) = 0.0134$$

The increase in the k_{eff} will then be of **0.013**.

Water in the guide tubes was replaced by boron carbide – to simulate insertion of control rods. When all these control rods are inserted, 89 times 24 in total, the decrease in k_{eff} will be of **0.148**.

The total change in k_{eff} will be – **0.1345**.

In other words: there is no chance that the core will go critical even when all the ^{233}Pa produced during the burn-up decays into ^{233}U. Only ~10% of the control rods need to be inserted to make sure the reactor will not reach criticality again, so there is a sufficiently large shutdown margin. In other words: *The reactor is perfectly safe with respect to the effect of ^{233}Pa decaying into ^{233}U.*

6.3 Economics

Commercial reactors are operating with recycled plutonium in Western Europe, but uranium is not being recycled significantly because of the currently low cost of fresh uranium, which does not contain neutron-absorbing ^{236}U that decreases the reactivity of recycled uranium. With the thorium cycle, it has been shown here; uranium recycling is an absolute necessity. But it is not only a necessity; it is also a possibility to recover perfectly good fissile material, due to net production of ^{233}U and the remaining ^{235}U. Despite the recycled uranium containing both neutron absorbing ^{234}U and ^{236}U.

For conventional UOX fuel, OTC, the amount of natural uranium and SWUs needed to fuel the EPR will be the same each time the reactor is fuelled. For the Th/HEU cases however, this changes dramatically: For the initial loading of the reactor, more natural uranium and a correspondingly larger amount of SWUs is needed to get the initial fissile inventory. However, once recycling is started, much less natural uranium is needed for the remainder of the reactor life. Figure 6-9 and Figure 6-10 shows graphically how much natural uranium and SWUs needed every fuelling, and how this changes as uranium is recycled. A k_{eff} value which is a bit too high in one fuelling will lead to the need of less new fissile material in the proceeding fuelling, which means that less natural uranium and less SWUs are needed to obtain the needed fissile inventory. In the same way a k_{eff} value which is too low will lead to the need of more fissile material in the next fuelling. As already emphasized in chapter 4.3.3: If the boron concentration could be exact at all times, it would be easier to get the absolute correct k_{eff} value, but for a one year masters project, some approximations had to be made, i.e. linear boron removal. However, the overall results are correct even though there are fluctuations; such as seen in Figure 6-9 and Figure 6-10.
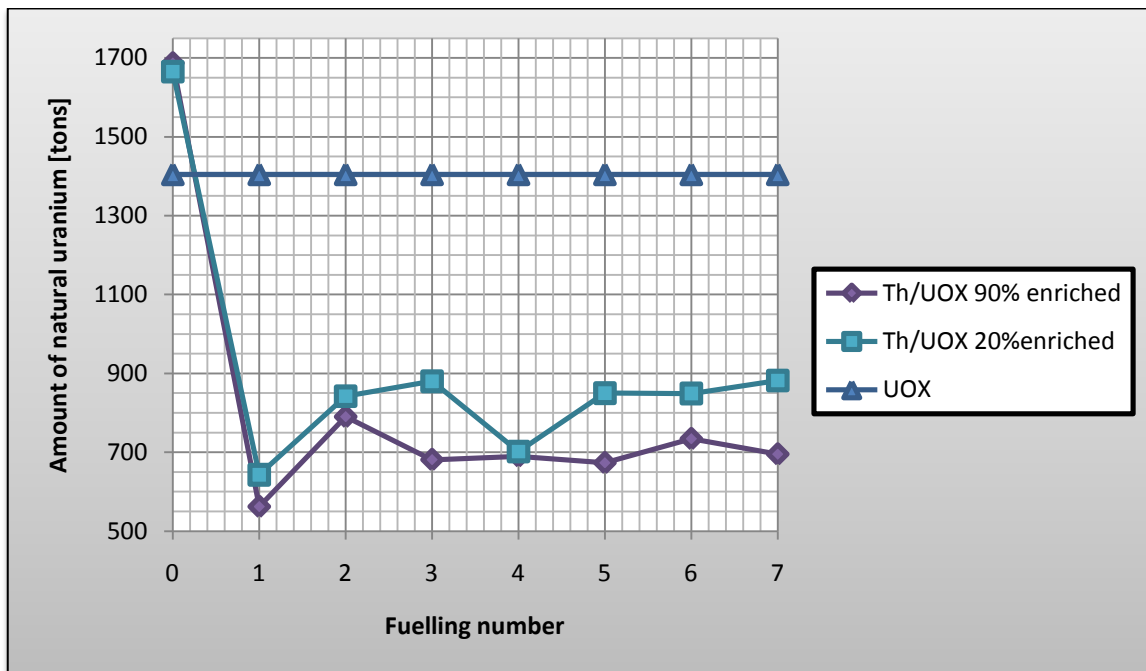
Figure 6-9: Tons of natural uranium needed for a given enrichment and fuelling number
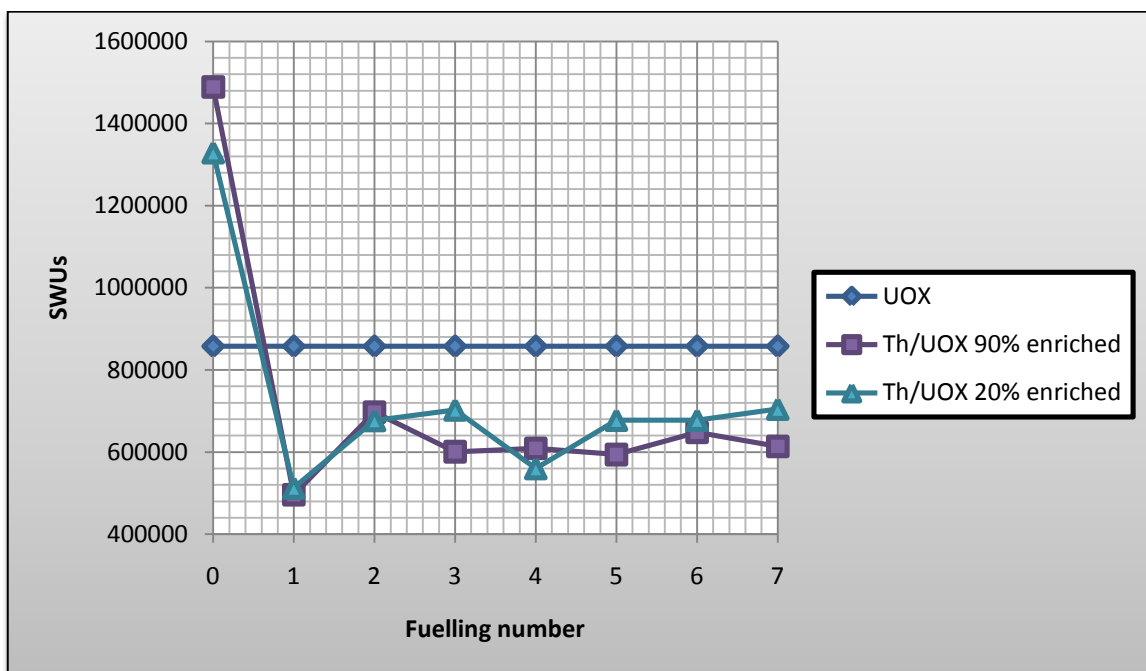

Figure 6-10: Enrichment work (SWUs) needed to get the amount of uranium needed to fuel the EPR

The results for the entire 60 years lifetime of the EPR are summarized in Table 6-10. The Th/UOX 90% enriched fuel uses less natural uranium – almost only half of what the

UOX cycle needs – and less SWUs. So not only does the UOX fuel produce large amounts of TRUs, it is also the biggest consumer of both natural uranium and SWUs.

Table 6-10: Estimate of total need (60 years service) of natural uranium and SWUs, uranium and thorium cycle compared

	Natural uranium [tons]	**SWUs**
OTC UOX	28 110	17 164 000
Th/UOX 20% enriched	18 000	14 398 000
Th/UOX 90% enriched	15 200	13 311 000

A summary of the materials needed, material regained, SWUs and fuel fabricated is shown in Table 6-11 and Table 6-12. That is, everything that will have an impct on the total cost.

The cost of the fuel cycle (thoriated or UOX) is a function of natural uranium needed, enrichment work (SWUs), fuel fabrication, and reprocessing. A rough estimate of the cost of the fuel cycle in cents per kWh of electricity as a function of the price of natural uranium can thus be made. This fuel cycle cost is linearly dependent on the uranium price and the slope of the line represents the efficiency with which uranium resources are used. (Since reprocessing is expensive, the longest burn ups are always favored.) The steeper the slope, the more dependent on the uranium price. Figure 6-11 shows the price per kWh produced as a function of the price of natural uranium. The three different options in the graph is OTC UOX, and the 1st recycle of the Th/UOX 20% and 90% enriched. Table 6-13 summarizes the background for the plot. The price assumptions used in the model are as follows: $160/SWU for *enrichment* of natural uranium, $1400/kg for thoriated fuels spent fuel *reprocessing* costs, $250/kg for normal UOX and $1100/kg for thoriated fuels, *fuel fabrication* costs.[10] The Th/HEU cost assumptions are made on the basis that everything, reprocessing and fuel fabrication, has to be remotely handled, and these prices are therefore higher than that of UOX. The Th/UOX 90% enriched fuel is the most independent of the three options shown in this figure.

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[10] These price assumptions are without the price of thorium, the reason being that it is impossible to predict what the actual price of thorium will be, and it will in any case be a small expence.

Table 6-11: Th/UOX 90% enriched, the costs (BOC)

Recycle #	Thorium (ton)	$U_{natural}$ (ton)	$U_{reprocessed}$ (ton)	SWU	Fuel fabricated (ton)
0	122.89	1 686.63	0	1 489 000	131
1	122.19	562.19	5.7	496 000	131
2	120.70	790	6.35	697 000	131
3	120.04	681.06	7.60	601 000	131
4	119.34	690.47	8.23	609 000	131
5	118.79	672.53	8.88	594 000	131
6	118.17	734.23	9.40	648 000	131
7	117.65	695.87	9.95	614 000	131

Table 6-12: Th/UOX 20% enriched, the costs (BOC)

Recycle #	Thorium (ton)	$U_{natural}$ (ton)	$U_{reprocessed}$ (ton)	SWU	Fuel fabricated (ton)
0	97.08	1 664.72	0	1 329 000	132
1	85.97	643.29	31.4	513 000	132
2	72.79	842	42.45	677 300	133
3	60.39	880.21	55.62	703 000	133
4	51.08	702.14	68.02	560 400	134
5	39.05	849.79	77.45	678 000	134
6	27.43	848.93	90.92	678 000	135
7	15.53	882.79	101.27	705 000	135

Table 6-13: Fuel cycle cost initiators

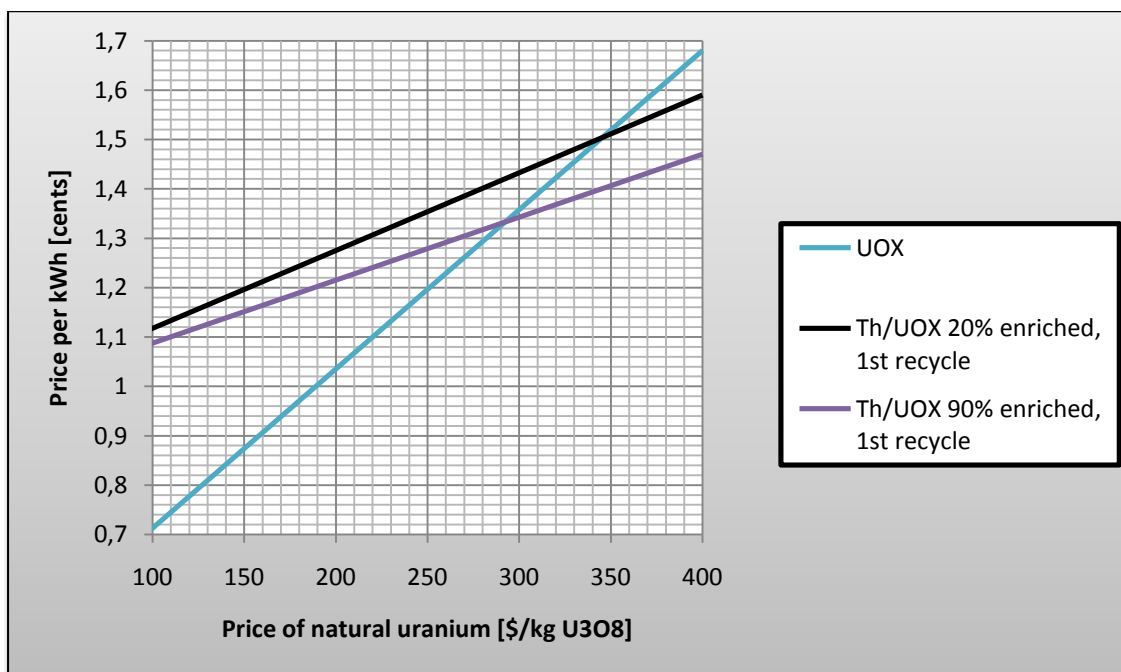|	SWU	U_{nat} [kg]	Fuel produced [kg]	Reprocessd fuel [kg]
UOX 4.5%	858 000	1 405 000	136 000	0
Th/UOX 20%	513 000	643 000	132 000	132 000
Th/UOX 90%	496 000	562 000	131 000	132 000

Figure 6-11: Fuel cycle cost

From the model two "break even" points are found: ~290 \$/kg of natural uranium, at which time the recycling of Th/UOX 90% enriched fuels will be economically viable, and ~340 \$/kg of natural uranium, at which time recycling of the Th/UOX 20% enriched will be. At the first break even point the price per kWh will be ~1.32 cents, and at the second one the price will be ~1.5 cents. The current price of natural uranium is 110 \$/kg of U_3O_8, which gives a price of 0.74 cents/kWh produced by the UOX fuel OTC, 1.1 cents/kWh from the Th/UOX 90% enriched, and 1.14 cents/kWh from the Th/UOX

20% enriched. The thorium cycle is not economically viable today, but the uranium prices will rise, and at 290 and 340 $/kg of natural uranium the reprocessing of thoriated fuels will cost less than UOX fuel.

When uranium is reprocessed from the spent Th/HEU-fuel there will be, as seen earlier in this chapter, substantially less decay heat from the spent fuel that is actually discharged as waste. Less decay heat means less costs of storage, which is <u>not</u> taken into account in this price assumption. It has been shown that the waste heat from the initial loading of Th/UOX 90% enriched will be ~1/7 of that from conventional UOX, which implies that the thoriated fuel could be stored in a correspondingly smaller repository.

As mentioned in the introduction Norway has the biggest battery in Europe in the form of hydro. This hydro power could be exported to Europe at very high prices in the morning and evening, when the electricity is most costly, and Norway could probably make a profit if the electricity base load was covered by nuclear power. The electricity consumption goes up twice a day: in the morning and in the afternoon/evening. The price of the electricity follows the consumption, and it is a factor of two difference from lowest to highest, this is clearly shown in Figure 6-12 [28]. There is thus possible for Norway to export its hydro power at peak prices! This profit made from the sale of hydro power is neither taken into account in the fuel cycle cost estimate of Figure 6-11. This would make it even more interesting to look into the possibilities of the thorium fuel cycle!
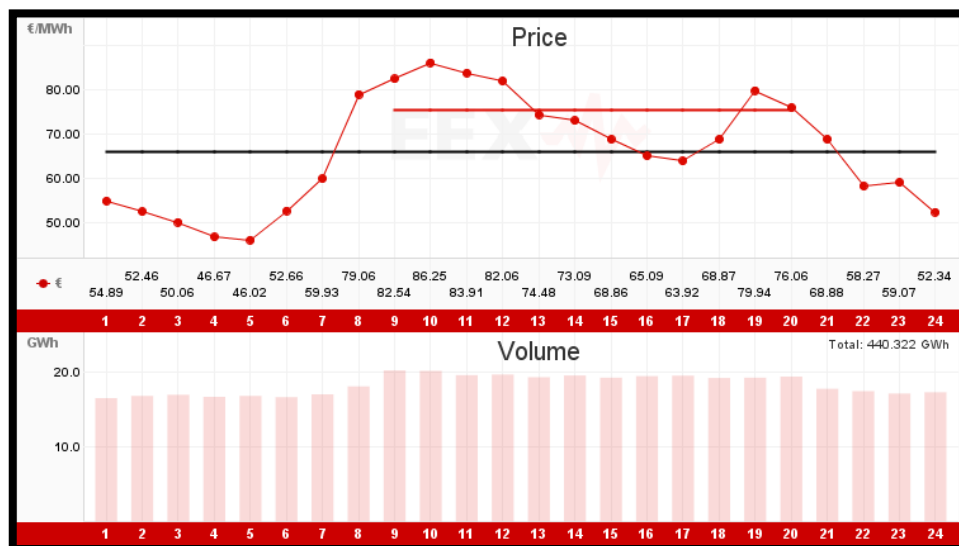

Figure 6-12: European Energy Exchange Spot Electricity prices, 13 February 2008

6.4 Proliferation

An important condition for the manufacture of a nuclear weapon is that the fissile elements have low intensity gamma decay since the presence of high gamma activity would require very thick lead or lead-glass protections behind which operators would have to work. ^{235}U obtained from isotopic separation plants has a low intensity gamma emission, just as the plutonium retrieved from reactor spent fuel. In general, the production of ^{233}U entails also the production of ^{232}U, and successive alpha decays of ^{232}U lead to ^{208}Tl which decays with the emission of a very deeply penetrating 2.6 MeV gamma ray. It is therefore practically impossible to manufacture a ^{233}U based weapon in the presence of ^{232}U contamination. This advantage regarding proliferation has, as mentioned earlier, its counterbalance in the more complex reactor fuel handling and manufacture. The entire process has to be automated, or has to be executed behind heavy shielding. There is, however, a potential proliferation issue with the initial loading because of the possibility of chemically separating the uranium from the thorium before the irradiation in the reactor starts. Chemically separating uranium from thorium, as opposed to enriching uranium, is an "easy" process. Therefore heavy supervision by IAEA would be necessary, but this supervision would only have to be done once; at the initial loading of the reactor. Chemically separating the uranium from a recycled thoriated fuel would give uranium which is useless as a bomb material, mainly for two reasons:

1. ^{232}U is produced by an (n,2n) reaction on ^{233}U, and ^{232}U lead to emission of high energetic gamma radiation. This makes the handling of the fuel much more difficult. Also illegal transportation of such material would be more difficult to do, and easier to discover.
2. The recycled uranium consists of $^{232/3/4/5/6/8}$U (see Table 6-4 and Table 6-5), and it is therefore not possible to use in a nuclear weapon, since only ^{233}U and ^{235}U are fissile isotopes, and the other uranium isotopes are polluting the uranium as weapons material.

At no stage in the fuel cycle will fuel be fabricated with a fissile content greater than 6%, so the proposed development is quite proliferation resistant.

7. Conclusions and Future outlook

The major challenge facing mankind this century is twofold: (i) getting enough energy and (ii) this energy must be produced without emissions of CO_2. The exploding energy consumption all over the world has led to a human induced climate change, and millions of people are on the run – "climate refugees". Nuclear power should not compete with other "green" options, like solar, wind, or hydro power, but it can not be neglected if we are to meet these challenges – as it is absolutely vital in solving this energy/climate crisis. Nuclear energy provides a steady supply of electricity at low cost, and is an important contributor in the energy mix of the 21st century, which puts the emphasis on sustainable development. The EPR is a 3rd generation reactor under construction today; it is an evolutionary reactor that represents a new generation of PWRs. The EPR can guarantee a safe, inexpensive electricity supply, without adding to the greenhouse effect.

It has been shown in this thesis that thorium can be used as fuel in the EPR if there is an extra supply of neutrons; namely HEU of 20% and 90% enrichment. The entire fuel cycle has been studied; from fuel fabrication to the time where the waste reaches the level of activity of natural uranium. The waste generated from the thoriated fuels is significantly less active – thus producing less decay heat – than the other fuel types studied; conventional UOX fuel, MOX fuel and Pu/Th fuel. It will also reach the level of activity of natural uranium faster than these other fuels. Since it produces less heat it can be stored in correspondingly smaller repositories than e.g. UOX waste. *The actinide waste can be minimized if the thorium cycle is employed using HEU, this, however, requires recycling of the uranium.* If uranium is recycled again and again there will be a gradual build-up of more of the heavier uranium isotopes in the uranium vector, and therefore more plutonium is produced. The waste will eventually (after 7 recycles) be twice as active as it was after the initial fuel loading, which is still only about one quarter of that of the UOX waste. The thoriated fuels are therefore much "cleaner" than other fuel types.

The thoriated fuels will over the 60 year lifetime of the EPR only consume half of the amount of natural uranium that conventional UOX fuel will need. The amount of enrichment work will also be substantially reduced. However, there will be increased cost of reprocessing and fuel fabrication due to activity the of ^{232}U, produced in small amounts in all thorium based fuels. The total fuel price will therefore be somewhat higher with today's uranium prices than the fuel price of the UOX open cycle. If the

price of final storage is taken into account it is, however, quite possible that the thorium cycle could be economically viable even with the current price of natural uranium. Furthermore, there is today an extensive development of nuclear power, and easily accessible uranium resources will be depleted which will lead to higher uranium prices; at which time the thorium fuel cycle will almost certainly be economically viable. Clearly, the economic viability of the proposed technology needs to be compared to other means of generating electricity. The thorium fuel cycle may become economically very attractive compared to other power sources if externalities are taken into account: (i) A correct CO_2 price must be established to embed the external environmental costs of generating electricity from fossil fuels such as coal. (ii) The upfront cost of the disposal of nuclear waste must be properly accounted for. Nuclear power covering the Norwegian base load of electricity would free the flexible hydro power to be exported at peak prices. With the attractive features of the thorium cycle, and the prospect of making a profit, the Norwegian people may see the positive sides of nuclear power. Hopefully the interest for developing commercial thorium based nuclear power in Norway will increase.

The recycled uranium will be proliferation resistant since the activity caused by ^{232}U makes it much more difficult to misuse the uranium, e.g. for terrorists to make a nuclear weapon. It will also be easier to discover someone illegally transporting it, due to the strong gammas, which are easily detectable.

Despite all of the attractive features of the thorium based fuel cycle, the development has always run into difficulties. The activity of ^{232}U results in technical challenges in reprocessing fuels. Development of the thorium cycle commercially also has to compete with the uranium cycle which is well known and working. Nevertheless, the thorium fuel cycle holds considerable potential in the long-term, and it is a *significant factor in the long-term sustainability of nuclear energy.*

One of the advantages of solid reactor fuel is that it can be arranged in a heterogeneous manner, with different fuel rods having different enrichments in fissile materials. This allows the possibility to favorize certain nuclear reactions by changing the shape of the neutron spectrum in different spatial regions of the core. The possible advantage of using heterogeneous assemblies for thoriated fuels needs to be quantified for various configurations. It is vitally important to find geometric configurations which optimize the production of fissile ^{233}U during the cycle, which would improve the conversion ratio, and get closer to breeding. In a longer perspective one could also look at optimizing the geometry of the assemblies/core; e.g. a different pin size could be more suitable for thorium fuels.

The intrinsic safety of light water reactor cores using thoriated fuels was investigated and compared to that of more conventional fuels: It was shown that the studied thorium fuels are safe with respect to fuel temperature feedback, void feedback, and the effect of ^{233}Pa decaying into fissile ^{233}U after the reactor is shut down. However, the main focus of this thesis was not safety, and dedicated safety simulations of accident scenarios were not performed. Full core simualtions of e.g. a LOCA must naturally be studied before there can be a commercial implementation of the thorium fuel cycle.

It should be noted that even advanced reactor simulations are only as good as the nuclear cross section data on which they rely. Fortunately, many important nuclear reaction cross sections in the actinide region have been accurately measured. However, for the thorium cycle, nuclear data uncertainties are somewhat larger. More precise measurements are needed, particularly for nuclear reaction cross sections for the fissile nucleus ^{233}U and neighboring nuclei.

The thorium nuclear fuel cycle makes possible a more sustainable nuclear power, using less natural resources, and the possibility of utilising a very abundant resource which has been of so little interest that it has never been quantified properly. This thesis has shown a feasible and realistic option, which is not dependent on huge leaps in technological progress and/or the successful construction/demonstration of radically innovative designs, and could very soon be implemented. *The thorium fuel cycle in the EPR could lead to a significant economy of uranium resources and large reduction in volumes of radioactive waste produced. It could also destroy the already existing stockpiles of weapons uranium.*

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