Parametric Studies on the Moderation Ratio of a 2-zone 1-fluid Molten Salt Reactor

Finding high power density within safety and sustainability constraints

Master Thesis Report

by

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Abstract

In this thesis a numerical model to simulate the burnup and neutronics in a MSR (Molten Salt Reactor) has been used. Both a 1-zone core as well as a 2-zone core for improved breeding have been investigated. Simulation is done with the SCALE code system in combination with a burnup code LOWFAT.

Aim of this study is to find a core that can operate at the highest power density within certain constraints. Different constraints are set, with respect to sustainability, safety and feasibility. This led to constraints for breeding (at least a self-breeder) graphite lifetime (>10 y), temperature feedback (<-1 pcm/K) initial fissile load and radiotoxicity of the TRUs (Trans-Uranic Elements) in the used salt.

First a 1-zone core is compared to a PWR (Pressurised Water Reactor) in terms of radiotoxicity of TRUs. Results show a much cleaner energy production for the thorium fuel cycle used in a MSR compared to the uranium fuel cycle in a PWR. Also different self-breeder 1-zone MSRs are run for four cycles of 50 years, cleaning the salt from FP (Fission Products) in between cycles, to investigate effects on the breeding gain (BG) and radiotoxicity. The buildup of actinides in the salt during the cycles causes the BG to drop over time.

A parametric study dividing a MSR into two zones radially with different graphite to salt volume ratios to improve breeding is done. The 2-zone MSR uses the same salt composition and chemical reprocessing scheme as the 1-zone MSR. Results show that due to a large increase in BG the 2-zone MSR is capable of operating at higher power densities while staying a self-breeder compared to a 1-zone MSR. As a drawback graphite lifetime is shortened, as power production is concentrated mostly in one zone.

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Nomenclature

Abbreviations

$\dot{H}_T$	Equivalent dose rate
λ_n	Decay constant of nuclide n
Φ_{50}	Neutron fluence of energies higher than 50 keV
ρ	Reactivity of the core
B	Geometrical buckling
$D_{T,R}$	Absorbed dose
$E(\tau)$	Committed effective dose
$E_{50,n}$	Committed effective dose after 50 years, due to nuclide n, 0 being the mother nuclide, any other number being a daughter nuclide
$e_{50,n}$	50 year dose equivalent of nuclide n
$E_{50,tot}$	Committed effective dose after 50 years due to the decay of a mother nuclide and all of her daughter products
e_{50}	50 year dose equivalent factor
H_T	Equivalent dose
$H_T(\tau)$	Committed dose over time τ
k_∞	Infinite multiplication factor
k_{eff}	Effective neutron multiplication factor
L	Diffusion length
$N_n(t)$	Number of nuclides n present at time t
P_{NL}	Non-leakage probability
R_a^{23}	Neutron absorption reaction rate of ^{233}U
R_a^{25}	Neutron absorption reaction rate of ^{235}U
R_c^{02}	Neutron capture reaction rate of ^{232}Th
R_c^{13}	Neutron capture reaction rate of ^{233}Pa
R_c^{24}	Neutron capture reaction rate of ^{234}U
$R_d^{13\_s}$	Decay rate of ^{233}Pa in the stockpile
$R_d^{13\_s}$	Decay rate of ^{233}Pa
R_{fe}^{23}	Feed rate of ^{233}U

T	Temperature of the core
w_R	Radiation weighting factor
w_T	Tissue weighting factors
ARE	Aircraft Reactor Experiment
BG	Breeding Gain
BG	Breeding Gain
FBR	Fast Breeder Reactor
FP	Fission Products
GIF	Generation IV International Forum
MSBR	Molten Salt Breeder Reactor
MSR	Molten Salt Reactor
MSRE	Molten Salt Reactor Experiment
PHWR	Pressurized Heavy Water Reactor
PWR	Pressurized Water Reactor
TRU	Trans-Uranic Elements

Chapter 1

Introduction

With oil becoming more scarce and the problem of global warming becoming more important, alternatives to fossil fuels become more interesting. Nuclear energy is a form of energy that can compete with oil and coal on a large scale without producing large amounts of carbon dioxide emissions [3]. This is why now nuclear energy is back in the scope, especially the reactor types chosen by the Generation IV International Forum (GIF). The GIF is an intergovernmental organization which is founded to stimulate the development of new types of nuclear reactors and the collaboration between its members. Six types of reactors have been chosen which all have improvements on sustainability, economics, safety, reliability and proliferation-resistance above conventional generation II and III nuclear reactors.

Research of the Molten Salt Reactor (MSR) dates back to the fifties at the Oak Ridge National Laboratory (ORNL), with the Aircraft Reactor Experiment (ARE). The ARE was the research of a liquid uranium fluoride fuelled reactor with a BeO moderator for use in an airplane [1]. After this experiment, the focus was on civil applications with reactors using the thorium fuel cycle. The Molten Salt Reactor Experiment (MSRE) (1964 - 1969) [6] concerned an 8MWth MSR with a graphite moderator and a liquid salt: LiF-BeF_2-ZrF_4-UF_4. It operated with ^{233}U as a fuel in 1969. The results of the MSRE were very promising in terms of piping corrosion, graphite damage due to the salt and radiation damage of the salt. Most of all it was a working proof of concept for the MSR. The success of this experiment led to the design of the Molten Salt Breeder Reactor (MSBR)[19]. The MSBR was a design of a high breeding power producing reactor. However, the MSBR suffered from drawbacks in terms of safety and fuel reprocessing.

Since the GIF has chosen MSRs to be one of the six generation IV reactors there has been renewed interest in this type of nuclear reactor. With the fuel being carried by a liquid and using thorium to breed fissile uranium, the technology of a MSR is very different of that of conventional solid fueled uranium reactors. The technology has great potential though. Using thorium has advantages over conventional reactors, as much less plutonium and radiotoxic minor actinides like neptunium, americium and curium are produced using the thorium fuel

cycle than when using the uranium fuel cycle. Also thorium does not need to be enriched like uranium for use in reactors, making it cheaper. On the other side, much is still to be researched for MSR's. Different safety, economical and feasibility studies are still to be done.

This thesis will focus on the thermal MSR. An optimization of the MSR core for breeding with a 2 zone configuration, while meeting certain constraints with respect to safety, chemical reprocessing of the salt, initial amount of ^{233}U and durability of the graphite moderator, will be done. The power of the core will be increased within the boundaries of the constraints to find the highest possible power density whilst maintaining a self breeder. Also an analysis of the radiotoxicity of the fuel salt will be performed, comparing different scenarios.

Chapter 2

Theory

This chapter focuses on the physics behind the MSR and the aspects concerned in this thesis. Also the theory behind the methods used to compute several aspects of the MSR and its inventory are explained. First, a description of the thorium fuel cycle is given, followed by a description of the thermal MSR as it is studied for this thesis. Both the theory behind breeding gain (a measure for the breeding performance in a reactor) for a MSR and radiotoxicity is presented, as these are two aspects that will be researched in further depth in this study. Next, the constraints layed upon the design of the 2 zone MSR are discussed, followed by a description of the computational methods used for simulation of the MSR and for the calculation of breeding gain and radiotoxicity.

2.1 The thorium fuel cycle

The fuel used in a MSR is the naturally occurring ^{232}Th isotope. Actually, the term fuel is wrong, because ^{232}Th itself is not fissile. The (not naturally occurring) product ^{233}U of the neutron absorption of ^{232}Th is fissile though. Thorium decays to the fissile nuclide ^{233}U via a protactinium isotope as follows:

$$^{232}_{90}Th + ^{1}_{0}n \longrightarrow ^{233}_{90}Th \xrightarrow{\beta^-} ^{233}_{91}Pa \xrightarrow{\beta^-} ^{233}_{92}U. \tag{2.1}$$

The cross-sections of the three most important isotopes in the thorium fuel cycle are plotted in figures 2.1, 2.2 and 2.3.^{233}U has a thermal capture to fission ratio of around $\frac{1}{10}$ compared to a higher $\frac{1}{6}$ for ^{235}U, making ^{233}U even better suitable as a fuel. ^{233}Th has a half life of just 21.83 minutes, whereas ^{233}Pa has a half life of 27 days. This relatively long half life combined with a high thermal neutron capture cross-section makes ^{233}Pa vulnerable for neutron capture whilst in the core; thus not being able to decay to ^{233}U. It is therefore necessary to keep the

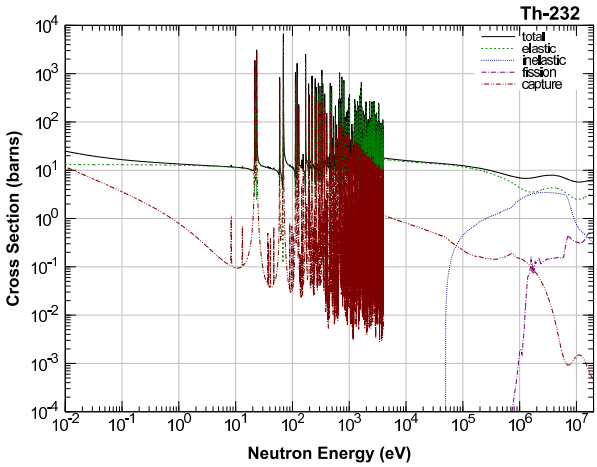

Figure 2.1: Neutron cross-sections of ^{232}Th in the energy range of 10^{-2} to 10^{7} eV.[20]

protactinium outside of the core in order to let it decay to ^{233}U. A MSR suits that need as the molten salt, which acts as carrier for the fuel, can be manipulated, while still operating the reactor, because of its liquid state. Part of the molten salt can be taken out of the core to be set aside long enough to let most of the ^{233}Pa decay to ^{233}U. More about the processes in a MSR is found in section 2.2.

2.1.1 Sustainability aspects

As in a PWR, in a MSR uranium is fissioned for the generation of power. In conventional nuclear reactors ^{235}U is fissioned for the generation of power. This naturally occurring fissile isotope represents only a 0.7% of the uranium found in ore on earth. Naturally occurring thorium consists entirely of ^{232}Th. This means that in theory all thorium found on earth can

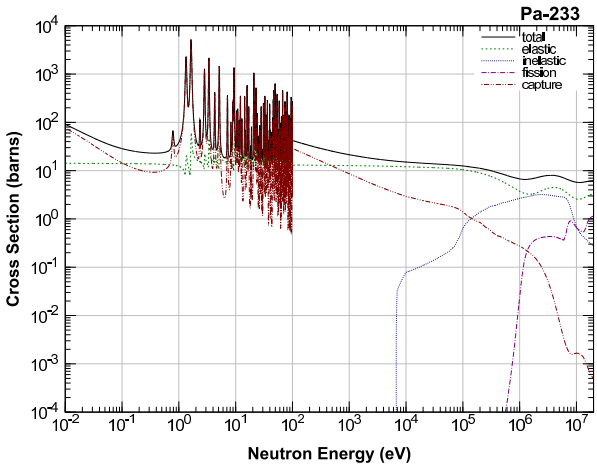

Figure 2.2: Neutron cross-sections of ^{233}Pa in the energy range of 10^{-2} to 10^{7} eV.[20]

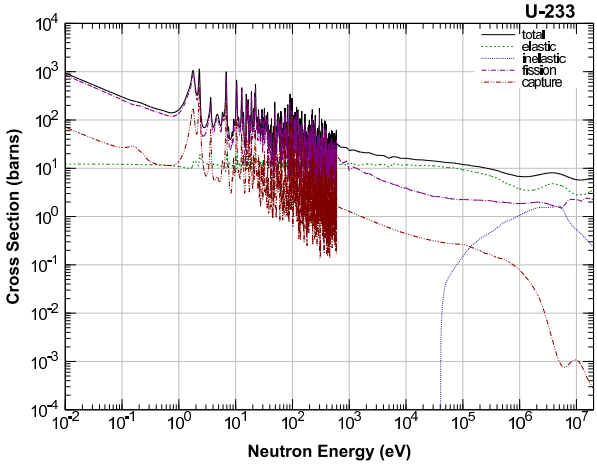

Figure 2.3: Neutron cross-sections of ^{233}U in the energy range of 10^{-2} to 10^{7} eV.[20]

be converted to fissile ^{233}U, whereas of the uranium found on earth only the small 0.7% part of uranium ore consisting of ^{235}U can be fissioned efficiently in thermal reactors. However, using Fast Breeder Reactors (FBRs) ^{238}U can also be bred into fissile Pu for producing energy. FBRs are technologically more comprehensive than MSRs.

As can be seen in Table 2.1 identified uranium ore resources are more abundant than thorium ore resources. This is because, up till now, more research has been done to find uranium than to find thorium. Until now thorium has not been considered a valuable element in the earths crust, and was seen as a rather useless by-product of uranium and other heavy metal mining. Comparing the half-life of uranium ($4.47*10^{9}$ a) with that of thorium ($1.40*10^{10}$ a), one can conclude that it is more likely that thorium will be more abundant in the earths crust. The costs for mining do not include costs for enriching uranium; thus producing thorium for use in a reactor is much cheaper. Taking the above into account the amount of thorium available for mining is enough to feed reactors for thousands of years. Estimates of uranium depletion range from within hundred years to tens of thousands of years in the future, depending on

energy consumption, type of nuclear reactors (including breeding reactors) and the price people are willing to pay for uranium mining. Oceans, for instance, contain many more times the amount of uranium found on land, although it would be much more expensive to extract.

Table 2.1: Identified resources as of January 1 2009 of thorium (l) and uranium (r) minable at <$80/kg(r), data taken from [17]

thorium		
Country	10^3 Tonnes	% of total
Australia	452	20
USA	400	18
India	319	14
Brazil	302	14
Venezuela	300	13
Norway	132	6
Egypt	100	4
Russia	75	3
Greenland	54	2
Canada	44	2
South Africa	18	1
Other	33	1
World total	2,229	100

uranium		
Country	10^3 Tonnes	% of total
Australia	1,612	43
Kazakhstan	476	13
Canada	447	12
South Africa	233	6
Brazil	231	6
Russia	158	4
China	150	4
Jordan	112	3
Uzbekistan	86	2
Niger	73	2
Ukraine	54	1
Mongolia	42	1
USA	39	1
Other	29	1
World total	3,742	100

As for the radiotoxicity of the spent fuel of the thorium fuel cycle, it is known to be much lower, due to the production of less (radiotoxic) actinides than in conventional ^{235}U powered nuclear reactors [18]. Most actinides that are produced in the uranium fuel cycle come from the neutron capture of ^{238}U. This leads to a relatively large amount of radiotoxic plutonium isotopes. The thorium fuel cycle on the other hand produces less plutonium and breeds ^{233}U as a fuel. Of the ^{233}Th a large fraction will eventually breed to the fissile ^{233}U, whereas most ^{238}U will transmute to radiotoxic, non-fissile plutonium isotopes. Also the capture to fission ratio of ^{233}U is lower than that of ^{235}U, which leads to less captures, meaning a lower amount of TRUs as well. Having different mass numbers the isotopes of the different fuel cycles will produce a different composition of minor actinides (with different radiotoxicities) as well. This will be researched in further depth in this thesis.

2.1.2 Proliferation resistance

Being fissile, ^{233}U is suitable for use in nuclear weapons. The USA has tested the use of this isotope in nuclear weapons in operation 'Teapot MET' in 1955 [2]. There is a disadvantage though of using ^{233}U as a weapon material: the thorium fuel cycle has ^{232}U as a byproduct

via the following fast neutron (n,2n) reactions:

$$^{233}_{91}Pa + ^{1}_{0}n \longrightarrow ^{232}_{91}Pa + 2^{1}_{0}n \overset{\beta^-,1.31d}{\longrightarrow} ^{232}_{92}U \tag{2.2}$$

$$^{233}_{92}U + ^{1}_{0}n \longrightarrow ^{232}_{92}U + 2^{1}_{0}n \tag{2.3}$$

$$^{232}_{90}Th + ^{1}_{0}n \longrightarrow ^{231}_{90}Th + 2^{1}_{0}n \overset{\beta^-,1.06d}{\longrightarrow} ^{231}_{91}Pa + ^{1}_{0}n \longrightarrow ^{232}_{91}Pa \overset{\beta^-,1.31d}{\longrightarrow} ^{232}_{92}U \tag{2.4}$$

Here, in the first and third reaction chain, the last step is a β-decay from ^{232}Pa meaning that protactinium extracted from the core into the stockpile (as in a MSR, as will be explained in the next chapter) will also, partly, decay to ^{232}U. The ^{232}U has a half life of 68.9 years and decays in various steps to the stable ^{208}Pb:

$$^{232}_{92}U \overset{\alpha,68.9a}{\longrightarrow} ^{228}_{90}Th \overset{\alpha,1.9a}{\longrightarrow} ^{224}_{88}Ra \overset{\alpha,3.6d}{\longrightarrow} ^{220}_{86}Rn \overset{\alpha,55s}{\longrightarrow} \ldots \tag{2.5}$$

$$\ldots ^{216}_{84}Po \overset{\alpha,0.15s}{\longrightarrow} ^{212}_{82}U \overset{\beta^-,10.6h}{\longrightarrow} ^{212}_{83}U \overset{\alpha,61m}{\longrightarrow} ^{208}_{81}Tl \overset{\beta^-,3m}{\longrightarrow} ^{208}_{82}Pb.$$

The above steps include daughter nuclides that emit very high energy gamma rays, especially ^{208}Tl (2.6 MeV). This radiological hazard makes uranium from a MSR difficult to handle as ^{232}U cannot be chemically separated from other uranium isotopes. Also high energy gamma rays can even damage electrical systems that might be used in bomb triggers. For these reasons MSR's will be more proliferation resistant than conventional reactors, as more expensive and more advanced technological steps need to be taken in the process of producing a weapon than with plutonium.

2.1.3 Drawbacks

Because a MSR uses ^{233}U as fissile material, and because of the fact that ^{233}U is not a naturally occurring isotope, starting a MSR will not be possible without previously produced ^{233}U from other MSR's or fast breeder reactors (FBR). Also India already breeds ^{233}U in their Pressurized Heavy Water Reactors (PHWR) on a large scale. It can be seen in the Table 2.1 that India has vast amounts of thorium, while there is little uranium found. This combined with the fact that India was cut from foreign uranium supply for a long time due to their nuclear weapons program, forced India to invest in techniques for thorium breeder reactors. Another option is to startup the reactor with other fissile material like ^{235}U or Pu from the spent fuel of PWR's. This has been studied with promising results [18], [7]. For the fast deployment of a large fleet of MSR's it is necessary to use one of the above options for the startup of the first reactors of this fleet. These first MSR's do not start breeding excess uranium in their stockpile for up to 10-20 years [16]. The initial amount of ^{233}U should be as small as possible to ensure a deployment of MSR's as fast as possible. Once a desired MSR fleet size is achieved the average MSR can be a self breeder as no excess uranium is needed anymore to start up new reactors. This leads to two types of reactors: one with the highest breeding gain and the other with the highest power output at a breeding gain of zero. Increasing the power density of a MSR will cause the breeding gain to drop, as more protactinium will capture a neutron.

2.2 The thermal MSR

In a MSR the fuel is carried by a liquid, the molten salt. Advantages of a liquid fuelled reactor include:

- Fuel can continuously and easily be moved in and out of the moderated core; i.e. in and out of the neutron flux.
- The salt can easily be chemically reprocessed, to remove unwanted fission products, while still operating the reactor.
- Protactinium can be taken out of the fuel loop in order to decay without neutron capture.
- The desired amount of fuel can continuously be added or removed to the salt loop to maintain criticality.
- The high temperature liquid salt in the reactor is at low pressure, making it relatively safe.

This section gives a description of the (epi)thermal MSR as it was studied in this thesis. Figure 2.4 shows a simplified schematic view of the MSR. In the MSR the fuel salt in the primary loop flows through channels in the reactor core that is built of graphite. The graphite serves as a structural material and is in direct contact with the salt. No cladding is needed as the graphite is non-porous for the salt. The graphite is subject to radiation damage though, which will be discussed later in further detail. The graphite also serves as the moderator for the neutrons in the core. Energy released by fission reactions in the core heat up the fuel salt. Out of the moderated core, where neutron flux dies out quickly, the salt is led through various important steps. The salt is led through a heat exchanger, where heat is exchanged with a secondary salt or water loop. This secondary loop then exchanges heat with a helium or steam loop that powers a gas turbine to generate electrical power. An off-gas bubbling system removes soluble fission products (this is not depicted in Figure 2.4 but takes place directly in the primary loop; no extraction of salt is needed). Further chemical reprocessing of the salt is either done on line, or in a plant nearby. For this, a part of the salt is taken out of the primary loop. Reprocessed salt with desired properties is then continuously added back into the primary loop.

2.2.1 Geometry of the core

As stated above, molten salt flows through a graphite moderator. The core is build up out of graphite hexagonal prisms, which will be referred to as blocks. Through each of the blocks runs a cylindrical channel through which the molten salt flows. Figure 2.5 shows a cross-section of such a hexagonal graphite block. Many prisms are stacked side by side to form a large cylinder with many fuel channels. The result can be seen in Figure 2.6. Note that this

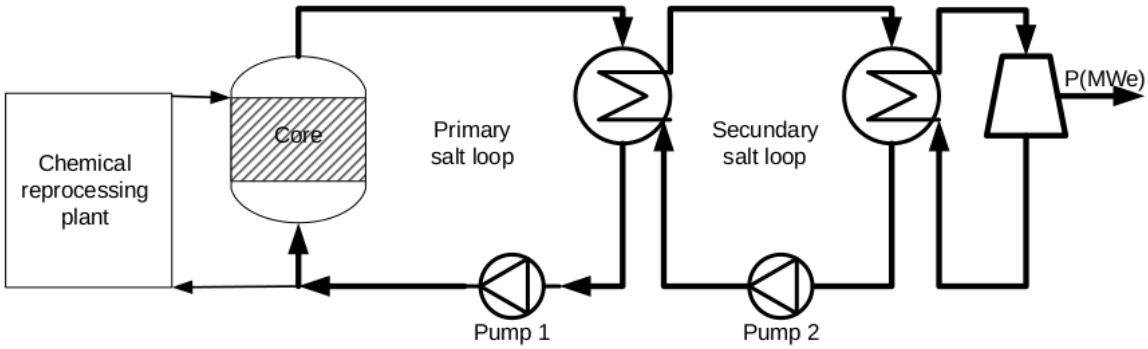

Figure 2.4: A schematic representation of a MSR

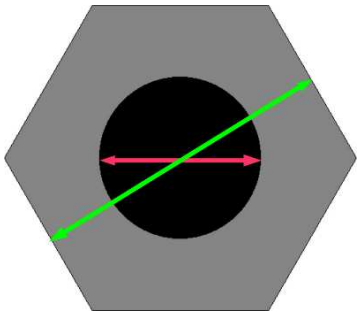

Figure 2.5: A cross-section of a graphite block. The black area represents the fuel channel, through which the fuel salt will flow. The gray area is the graphite moderator. The hexagon size is determined by its block width, the green arrow in the figure. The red arrow shows the diameter of the fuel channel

is a 2 zone MSR core. It can be seen that the core is surrounded by a reflector (zone 3 in the figure). The reflector is built up out of the same blocks, but without the fuel channels running through it. The reflector is added to reduce neutron leakage. A two zone MSR, as will be studied in this thesis, is a MSR with two zones differing in properties. The zones are divided radially as seen in Figure 2.6 and differ only in fuel channel diameter and amount of blocks. By altering the fuel channel diameter the moderation ratio of a zone is changed, and therefore its infinite multiplication number (k_∞) and neutron spectrum. This leads to different neutron fluxes and neutron spectra in the zones. Zones can in this way be optimized for either breeding or burning fuel.

2.2.2 Fuel reprocessing

Due to the liquid state of the fuel, the fuel can be continuously reprocessed. Fission products with large absorption cross-sections have a negative effect on the reactivity of the reactor and should be removed to maintain a critical reactor with as little feeding as possible. Also protactinium needs to be taken out of the core to let it decay to ^{233}U without capturing a neutron, to increase breeding. Desired amounts of bred uranium need to be reinserted into the core to keep it critical. Also thorium is constantly added to the core, to maintain the same

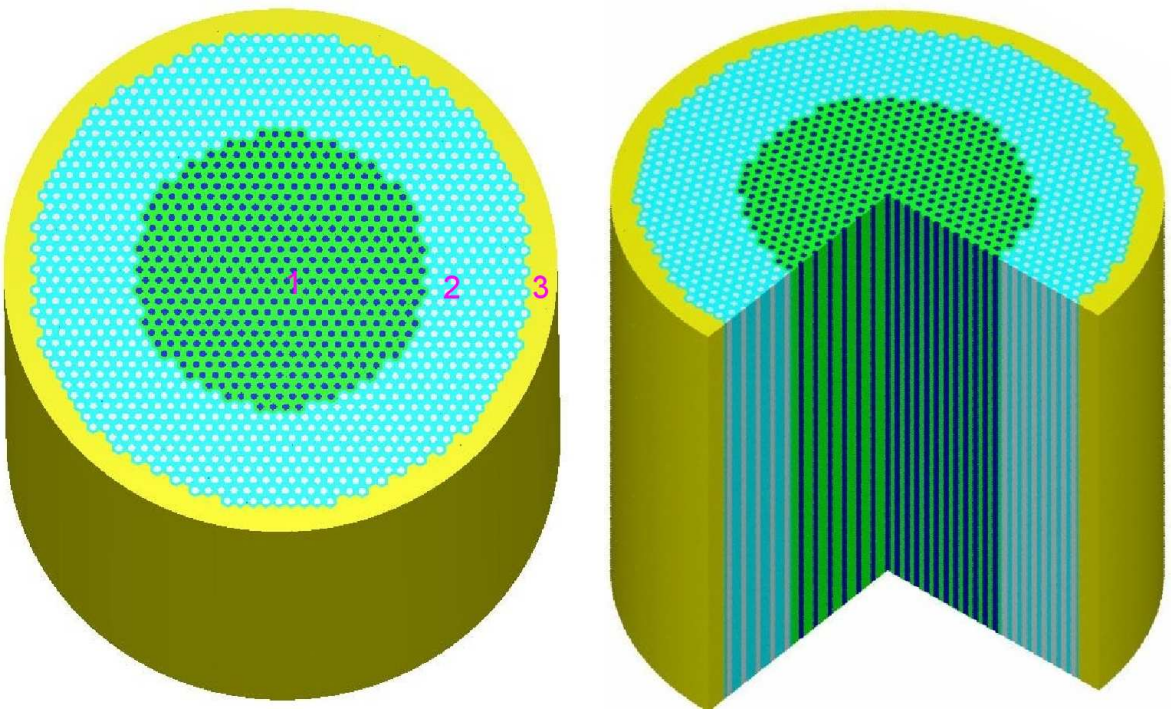

Figure 2.6: The MSR core, with left: the core divided into three zones: 1. The inner zone with a certain channel diameter. 2. The outer zone with certain channel diameter and 3. The graphite reflector. Note that the colors green, cyan and yellow all represent (the same) graphite, whereas blue and white represent the fuel salt. The different colors are only to clarify the different zones. And right: The same core with a slice cut out.

concentration throughout the lifetime of the reactor. All the reprocessing can be divided into these categories:

- Helium bubbling for non soluble fission products
- removal of Np and U and fluorination for direct re-insertion
- Extraction of other fission products
- Extraction and storage of Pa
- Re-insertion of salt with all actinides and bred ^{233}U

There are different methods with different cycle times for removing fission products. First, there is helium bubbling, which quickly removes noble gases and non soluble noble metals. Helium bubbling is used in all theoretical schemes that were studied before, and has been tested with success in the MSRE. This can happen without extracting salt from the loop. Fission products that are soluble can be removed by chemical processes. This can happen either on line or in a chemical plant nearby after extracting salt from the core. From the extracted salt Np and U are directly reinserted into the core. The Pa can be set aside to decay to ^{233}U. After the reprocessing, the salt can be reinserted into the reactor, with the

desired amount of ^{233}U to keep the core critical. The ^{233}U that is not reinserted is kept in a stockpile to be used for re-insertion later on or to fuel new reactors. The MSBR design from the 1960's used an on line reprocessing scheme that processes the whole volume of salt in just ten days. This gave good breeding results, but is not very feasible, because of its complexity. Many studies have focused on slowing down chemical reprocessing in a MSR [11], [13], [**?**], [12], [4]. To keep a reactor technically feasible and affordable, different slow reprocessing schemes (cycle times of months or even years) have been studied. Breeding, although lowered, is also possible when one uses a slow reprocessing scheme. Such a scheme is also used in this study, as can be seen in section 2.5.2.

2.2.3 Salt composition

Due to the good coolant properties of molten salts (low fusion temperatures, high heat capacity and medium heat conductivity) the molten salt in a MSR serves both as a coolant of the core as well as a carrier for the fuel. Also the high temperature at which a MSR operates allows for high efficiency energy conversion. Another advantage is that the pressure of the salt at high temperatures is around the ambient level and does not require a high pressure vessel, like PWRs (Pressurized Water Reactors). Many different salt compositions have been studied, including mixtures with different concentrations of: LiF, NaF, BeF_2, ZrF_4 RbF [**?**], [13], [11]. The molten salt LiF-BeF_2 (67%-33%) gives best results, as it has low parasitic neutron capture. This is the same salt that was chosen for the MSBR. Not only ^{232}Th is added to the fuel to breed ^{233}U but also a start-up amount of ^{233}U is needed to start the reactor with a critical core. This is a drawback of the MSR, as ^{233}U has to come from other breeder reactors, because it is not a naturally occurring isotope. Other fissile isotopes, like ^{235}U or Pu from spent fuel of PWRs can be used to start up the MSR as well. ^{232}Th and ^{233}U are added as $^{232}ThF_4$ and $^{233}UF_4$. In this research the reactor is started up with 12mol%$^{232}ThF_4$ and approximately 0.3 mol% $^{233}UF_4$, as suggested for the MSBR [19]. The startup concentration is dependent on the composition of the core, but lies around 0.3 mol%. In a thermal MSR the fluid generally can be in 2 places, inside and outside of the core. Inside the core the salt streams through a moderator, in this case graphite, making the mixture critical. Once the fluid leaves the core, neutrons are no longer moderated and the neutron flux will die out.

2.2.4 Multi-zone MSR

Fuel channel diameters of a MSR can be changed, resulting in an over or under moderated reactor, depending on narrower or wider fuel channels, respectively. Lowering moderation ratio will make the neutron spectrum less thermal, resulting in a change of the capture and fission cross-sections of the nuclides involved in the thorium fuel-cycle. In this study there will be referred to volume ratio, the ratio between the volume of the graphite and the volume of the salt in the core (or zone in the case of a multi-zone MSR). Volume ratio is easier quantifiable then moderation ratio.

A less moderated zone will induce a larger capture to fission ratio in a thorium-uranium fueled

reactor, resulting in a lower value for k_∞ in that zone, and thus a lower power production (and neutron flux). As for ^{233}Pa, a harder spectrum will lead to a slight decrease in capture cross-section (see Figure 2.2). This combined with the lower power production, will lead to less neutron capture in ^{233}Pa in a less moderated zone, favouring the breeding of ^{233}U in that zone. Having a more moderated zone will cause opposite events to occur (to a certain extent of moderation). Having two zones can be seen as a redistribution of the power in the core, i.e. the over moderated zone (with a relatively higher k_∞) will have a higher power density than the under moderated zone (with a relatively lower k_∞). Manipulating the volume ratio in different parts of the core can therefore alter the breeding properties of a MSR. A 2-zone core like in Figure 2.6 can be made with different volume ratios in zone 1 and 2. Chapter 4 will analyze results from different 2-zone configurations. An optimal configuration for a 2-zone core with high power density or high breeding gain will be searched for. Different safety, sustainability and design constraints will be set for the design and operation of the 2-zone MSR (Section 2.5).

2.2.5 Previous results from multi-zone MSR studies

Studies in optimizing a 1 zone core for breeding have been done by Nagy et al., [16]. This is also the reactor model that is used as a base for the 2-zone MSR studied in this thesis. The 2-zone MSR uses the same outer and reflector dimensions. Also the recommended salt composition and reprocessing scheme were used.

Different studies have been done dividing a MSR into zones of different volume ratios. As stated before, altering the volume ratio alters the power distribution in the reactor core due to different values of k_∞ in the zones. Flattening the flux profile throughout the core will spread neutron flux more evenly and will result in a longer graphite lifetime (see Subsection 2.5.1 on page 21). This is done by placing higher moderated outer zones in the core. By flattening the flux profile in the core, high flux values are closer to average flux values which benefits overall graphite lifetime. Mitachi et al. designed a reactor model with a 3 region core, [14]. The zones were radially increasing in volume ratio, resulting in a flattened flux profile. The result is a 450 MW (7.2 MW/m^3) converter, with a CR (Conversion Ratio, see Section 2.3) of 0.95. Resulting graphite lifetime is 30 years (although another value for maximum neutron fluence was used. Using the value used in this thesis and also for the MSBR, the graphite lifetime is 24 years). A drawback of the design is the low breeding capability. With a CR of 0.95 the reactor does not convert enough thorium into ^{233}U to meet its own needs. Also neutron leakage is larger, due to the increased amount of fissions near the outer surface of the MSR. Another Japanese study concerning a 2-zone core designed for long graphite lifetime obtained similar results [7].

On the other hand, dividing a MSR into different zones can also benefit breeding (compromising on graphite lifetime). A core can be divided into a relatively high volume ratio zone (the fissile zone), and a lower volume ratio outer zone (the fertile zone) shifting the power distribution more to the fissile zone of the core. This is studied by Nuttin et al., [18] and by Mathieu et al., [11]. While keeping the overall salt volume the same, Nuttin divides the hexagon celled core into two zones. One fertile zone and one fissile zone. Two different sce-

narios were studied, one with the fertile zone in the center, and the other one with the fertile zone at the edge. Conclusions were that a fertile zone at the edge will give a higher breeding ratio. This makes sense, as due to leakage the flux in the fertile zone will be lower when placed at the edge of the core. More Pa can capture a neutron without decaying to U, when the fertile zone is in the center. No research was done though to find the optimal value of the volume ratios of inner and outer zones. Geometry of the core used in this paper was as follows: The block width was 26 cm. Volume ratio inner zone was 4.15, corresponding to a channel diameter of 12 cm. volume ratio outer zone was 1.30, corresponding to a channel diameter of 18 cm. The resulting breeding ratio was 1.051, compared to 0.987 when the fertile zone was placed in the center. Temperature feedback was not affected, however as a negative effect of these results, graphite lifetime was reduced by 50 % due to a 50 % higher flux at the center of the core, compared to their 1 zone scenario. Mathieu et al. used a similar reactor model. The block width was 26 cm. Volume ratio inner zone was 5.14, corresponding to a channel diameter of 11 cm. volume ratio outer zone was 1.61, corresponding to a channel diameter of 16.9 cm. Depending on the chemical reprocessing scheme and reactor geometries promising results were achieved in terms of breeding. These results encourage further study in the use of multiple zones to achieve high breeding. This thesis will look for an optimal core configuration for breeding, while meeting certain constraints. A measure of breeding is the Breeding Gain, as will be explained in the next section.

2.3 Breeding gain

When dealing with MSR it is desirable to know the amount of ^{233}U produced, as well as consumed by the reactor. It tells whether the reactor is a breeder, a self breeder or a converter. A breeder produces more uranium than it consumes, a self breeder breeds as much uranium as it consumes and a converter consumes more than it produces. Making a MSR a breeder is important, especially in the first deployment phase of MSR's, as no large stock of ^{233}U will be present yet. Further on in the future, an average nuclear power plant which will be a self breeder, will be enough to sustain (large scale) energy production. A measure to describe the above is the breeding gain. The breeding gain (BG) is usually defined as the ratio of the net gain of fissile material (FG) to the net destruction of fissile material (FD): BG = FG/FD, which is equal to the breeding ratio (BR) (also referred to as Conversion Ratio (CR) if <1) minus 1. It can be calculated by comparing the amount of fissile material in a reactor and its stockpile at the end and beginning of a cycle. This is also known as the integral BG. But as a MSR is a continuous reactor, as chemical reprocessing can take place whilst operating the reactor, i.e. there are no irradiation cycles like in solid fuelled reactors. This means the integral BG definition only holds if one compares the amounts of fissile material at the startup and after shutdown of the reactor. For continuous measurement of the breeding gain another BG definition is necessary. Using reaction rates, a differential BG can be defined [15]. A brief description of the differential breeding gain definition as defined by [15] will be described here. The breeding gain in the core and stockpile at any moment in time can be defined as the ratio of capture and absorption reaction rates at that moment:

$$BG_{core+stock} = \frac{R_c^{02} - R_c^{13}}{R_a^{23}} - 1, \tag{2.6}$$

where R is the reaction rate, subscript c and a refer to capture and absorption. The first digit of the superscript denotes the last digit of the atomic number (the first digit being a 9) and the second digit of the superscript denotes the last digit of the weight number of the isotope, for example: 02 will be ^{232}Th. Not only ^{233}U is fissile, but ^{235}U as well, so it should be added to the Equation as ^{235}U is also being formed during the operation of the reactor by the neutron capture of ^{234}U via ^{233}U.

$$BG_{core+stock} = \frac{R_c^{02} - R_c^{13} + R_c^{24}}{R_a^{23} + R_a^{25}} - 1 \tag{2.7}$$

Subtracting the feed rate from the stockpile, as that amount of ^{233}U is not produced in the core itself, yields for the breeding gain in the core:

$$BG_{core} = \frac{R_c^{02} - R_c^{13} - R_{fe}^{23} + R_c^{24}}{R_a^{23} + R_a^{25}} - 1, \tag{2.8}$$

where the subscript fe stands for feeding. The amount of fissile material in the core will be just enough to maintain criticality in the reactor. The more interesting BG is the BG of the stockpile as it is a measure for ^{233}U not used in the core. If the breeding gain of the stockpile is positive, ^{233}U is produced that can be used to start up or maintain other reactors. For the breeding gain in the stockpile we get:

$$BG_{stock} = \frac{R_d^{13\_s}}{R_{fe}^{23}} - 1, \tag{2.9}$$

where the superscript 13_s stands for the ^{233}Pa in the stockpile and the subscript d refers to decay. Equation 2.9 is only the breeding gain in the stockpile. A similar Equation can be made for the BG in the core:

$$BG_{core} = \frac{R_d^{13} + R_c^{24} + R_{fe}^{23} - R_a^{23} - R_a^{25}}{R_a^{23} + R_a^{25}}, \tag{2.10}$$

where the -1 from Equation 2.8 has vanished by placing $-\left(R_a^{23} + R_a^{25}\right)$ in the numerator. The feeding of fissile material is now added instead of subtracted to make Equation 2.10 a measure for the gain of all fissile material in the stock. The neutron capture of ^{232}Th minus the capture of ^{233}Pa $\left(R_c^{02} - R_c^{13}\right)$ is replaced by the decay of ^{233}Pa in the core $\left(R_d^{13}\right)$, as that is an exact measure of the amount of ^{233}U produced in the core, as some of the protactinium is extracted from the core. The -1 is removed from Equation 2.9 by placing $-R_{fe}^{23}$ in the numerator. Changing the denominator for the stockpile BG of Equation 2.9 to match Equation 2.10, as just enough ^{233}U is fed to keep the core critical, yields:

$$BG_{stock} = \frac{R_d^{13\_s} - R_{fe}^{23}}{R_a^{23} + R_a^{25}}. \tag{2.11}$$

Adding equations 2.10 and 2.11 up will give the total breeding gain again:

$$BG_{total} = BG_{core} + BG_{stock} = \frac{R_d^{13} + R_d^{13\_s} + R_c^{24}}{R_a^{23} + R_a^{25}} - 1. \tag{2.12}$$

In this study the focus is on the BG of the stockpile. Achieving maximum BG or setting BG to zero will thus mean setting Equation 2.11 to zero.

2.4 Radiotoxicity

Probably the biggest complaint against the use of nuclear reactors is their long-lived radioactive waste. Radioactive waste produces ionizing radiation which is harmful to the human health. Because of the very long half-life of certain isotopes, nuclear waste has to be kept in a safe containment for thousands of years. Radioactive nuclear waste can be divided into three groups, namely fission products, actinides and neutron-activated material like cladding or containment vessels. Fission products (fission products) are elements with atomic number less than Uranium, formed by the fission of actinides. Fission products are usually β^--emitters instead of the usually α-emitting actinides. Examples are iodine, cesium and technetium. Although these are long-lived, the fission products are not as radiotoxic as the actinides, as the dose per decay for α emitters is much higher, after intake. Actinides are elements with an atomic number between 89 and 103. The only actinides present in nature are thorium and uranium. Due to their long half lives, they can still be found in the earths crust and oceans. Transuranics are actinides with an atomic number larger than that of uranium (92) and can only be formed artificially. Transuranics in a reactor are formed by the neutron capture and α-decay of other actinides. As transuranics are more radiotoxic and more abundant in the spent fuel of nuclear reactors than long-lived radiotoxic fission products, the main focus should be to reduce the production of transuranic elements.

In this thesis the total so called committed effective dose during 50 years after the ingestion of all the spent fuel is taken as a measure for the radiotoxicity of that spent fuel. The background of how that is calculated is shown below. Radiotoxicity dose is measured in Sieverts (Sv). All different types of ionizing radiation can be treated with this same unit. The committed effective dose following after the ingestion or inhalation of a radionuclide depends on several factors:

- Type and energy of the particles emitted
- The different organs that are irradiated (way of intake)
- The average time a radionuclide spends in the body

The type and energy of particles emitted contribute to the so called equivalent dose H_T in Sv (=J kg^{-1}) by a specific particle and/or energy dependent dimensionless weighting factor w_R:

$$H_T = \sum_R w_R D_{T,R}. \tag{2.13}$$

This is the sum for radiation consisting of different components, where $D_{T,R}$ is the absorbed dose in Gy (=J kg^{-1}). The subscripts T and R denote the radiated tissue and the radiation that the tissue is exposed to. The weighting factor for different types of radiation is shown in table 2.2. The committed equivalent dose $H_T(\tau)$is then the dose committed over a certain time τ. This is simply the integral over a time τ of a certain equivalent dose rate $\dot{H}_T$:

$$H_T(\tau) = \int_{t_0}^{t_0+\tau} \dot{H}_T dt \tag{2.14}$$

Table 2.2: Weighting factors for the different types of radiation, for the calculation of the equivalent dose [21].

Type of radiation		w_R
X and γ rays, electrons, positrons and muons		1
Neutrons, energy	<10 keV	5
	10 - 100 keV	10
	100 keV - 2 MeV	20
	2 - 20 MeV	10
	>20 MeV	5
Protons		2
α particles, fission fragments and non-relativistic heavy nuclei		20

Finally, a weighting factor is needed for each tissue and organ, to calculate the total committed effective dose received due to a certain time and radiation of different organs, as some organs (like gonads or bone marrow) are more vulnerable to radiation than others (like skin or bone surface). Just as for different particles, a dimensionless weighting factor is introduced for organs. The committed effective dose $E(\tau)$ is the sum over all irradiated organs radiated for a time τ

$$E(\tau) = \sum_T w_T H_T(\tau) \tag{2.15}$$

Were w_T are the tissue weighting factors, as defined in table A.1 of the Appendix. Committed effective doses are usually calculated over a time of 50 years, and are dependent on the way of intake (ingestion or inhalation) and the chemical composition of the isotope involved. The most affected organs are determined by these factors. For easy calculation of the committed effective dose after 50 years for every radionuclide there is a so called dose equivalent factor (e_{50}) that corresponds to either an inhalation or an ingestion of a certain amount of that isotope to calculate the committed effective dose. Dose equivalents are measured in Sv/Bq, and are thus a measure for how much every decay contributes to the effective committed dose. Dose equivalent factors used are according to ICRP 68.

Dose equivalent factors, half life, type of decay and most important daughter nuclides for the most abundant actinides and longest living fission products are shown in table 2.3. Note that most radiotoxic actinides listed in table 2.3 are α-emitters and are thus often more toxic when inhaled (see table 2.2), whereas all the long-lived fission products are β-emitters. The amount and composition of the fission products of both thermal ^{233}U and ^{235}U fission will roughly be the same, as the fission product yield of both nuclides do not differ much from each other, see figure 2.7, and energy per fission is also roughly the same. The amount of fission products will largely depend on the energy produced, whereas actinides produced in the core can differ a lot in both cases, as it will be shown in this study. When referred to radiotoxicity in this thesis, the 50 year committed effective dose for ingestion by members of the public is meant. Because a lot of daughter nuclides listed in table 2.3 are radionuclides themselves, radiotoxicity of a specific actinide will be calculated for the actinide itself and for all the daughter nuclides involved. This can lead to the fact that the radiotoxicity of a certain actinide will increase in time, as the concentration of daughter nuclides (with potentially higher dose equivalents and different half-lifes) can build up over time. The radiotoxicity of

Table 2.3: Most important transuranic nuclides and long-lived fission products that contribute to the (long term) radiotoxicity of spent fuel in nuclear reactors. *This is the combined radiotoxicity of ^{126}Sn and ^{126}Sb, as the decay of both isotopes can be treated as 1 decay, because the half life of ^{126}Sb is much smaller than that of ^{126}Sn. ^{126}Sn is the only long-lived fission product that decays in more than one step to another stable nuclide

Isotope	Half life	Dose equivalent factor[Sv Bq^{-}1]	Decay	Important daughter nuclides
^{237}Np	2.14 e6 a	1.1×10^{-7}	α	^{233}Pa, ^{233}U
^{238}Pu	87.7 a	2.3×10^{-7}	α	^{234}U, ^{230}Th, ^{226}Ra, ^{222}Rn
^{239}Pu	2.41 e4 a	2.5×10^{-7}	α	^{235}U, ^{231}Th, ^{232}U
^{240}Pu	6.54 e3 a	2.5×10^{-7}	α	^{236}U, ^{232}Th
^{241}Pu	14.4 a	4.8×10^{-9}	β^{-}	^{241}**Am**, ^{237}**Np**
^{242}Pu	3.76 e5 a	2.4×10^{-7}	α	^{238}U
^{241}Am	4.32 e2 a	2.0×10^{-7}	α	^{237}**Np**
^{243}Am	7.38 e3 a	2.0×10^{-7}	α	^{239}Np, ^{239}**Pu**
^{242}Cm	163 d	1.2×10^{-8}	α	^{238}**Pu**
^{244}Cm	18.1 a	1.2×10^{-7}	α	^{240}**Pu**
^{79}Se	6.5 e4	2.9×10^{-9}	β^{-}	-
^{93}Zr	1.53 e6	1.1×10^{-9}	β^{-}	-
^{99}Tc	2.13 e5	6.4×10^{-10}	β^{-}	-
^{107}Pd	6.5 e6	3.7×10^{-11}	β^{-}	-
^{126}Sn	1.0 e5	7.1×10^{-9} *	β^{-}	^{126}Sb (β^{-}, $t_{\frac{1}{2}}$=12.46d)
^{129}I	1.57 e7	1.1×10^{-7}	β^{-}	-
^{135}Cs	2.3 e6	2.0×10^{-9}	β^{-}	-

a certain isotope at any time is calculated as follows:

$$E_{50,0}(t) = e_{50,0}\lambda_0 N_0(0)\, e^{-\lambda_0 t}, \tag{2.16}$$

Where λ is the decay constant of the isotope and $N(t)$ is the amount of nuclides present at time t. The index 0 is used to note that this is the mother isotope. If daughter products are also radioactive, one needs to perform the same calculation for all the daughter products as well. Actinides decay in multiple steps to a stable isotope, usually lead, producing many daughter products. Radiotoxicity due to a daughter nuclide is than calculated by:

$$E_{50,n}(t) = e_{50,n}\lambda_n N_n(t), \tag{2.17}$$

with index $n = 1, 2, ..m$ for all the subsequent daughter products and $\frac{dN_n}{dt}$ being a function of both N_{n-1} and N_n:

$$\frac{dN_n}{dt} = \lambda_{n-1}N_{n-1}(t) - \lambda_n N_n(t) \tag{2.18}$$

The total radiotoxicity of a nuclide due to itself and all of its daughter nuclides is then

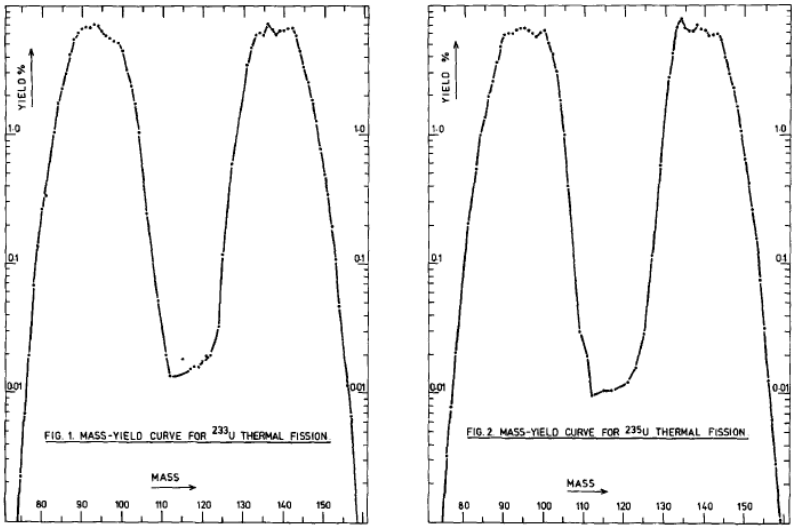

Figure 2.7: Fission product yield for the thermal fission of both ^{233}U (left) and ^{235}U (right)

calculated as follows:

$$E_{50,tot}(t) = \sum_{i=0}^{m} E_{50,i}(t) \tag{2.19}$$

When solving differential equation 2.18 for a mother and a daughter and using equation 2.19 to calculate radiotoxicity due to both isotopes, one obtains:

$$E_{50,tot}(t) = e_{50,0}\lambda_0 N_0(t) + e_{50,1}\lambda_1 \frac{\lambda_0 N_0(0)}{\lambda_1 - \lambda_0}\left(e^{-\lambda_0 t} - e^{-\lambda_1 t}\right). \tag{2.20}$$

Actinides have multiple radiotoxic daughter nuclides (see Table 2.3). When including more daughter nuclides, Equation 2.19 will become very long and comprehensive. In this study, radiotoxicity is therefore calculated with the use of a data library containing radiotoxicity values for an actinide and all its daughter nuclides at different times (see Section 2.6).

Note that the radiotoxicity calculated in this way is in a sense only a measure of how much harm potentially can be done to humans at a certain moment in time. The actual doses that people receive is much smaller, and will only occur if nuclear waste is spread into the environment. For a dose to be received, radiotoxic waste must first leak from its containment, after which it will spread and dilute in nature, in water or air, depending on its chemical composition, the way in which it has leaked and the type of storage of the radiotoxic waste. Radionuclides can then be either directly ingested or inhaled, or else taken in via plants or animals that people consume. In every step above, the committed dose will be reduced, as the radionuclides are more and more diluted each step. Thus, different disaster scenarios should be investigated to calculate real committed effective doses. Only that is a reasonable way to estimate average committed effective doses to members of the public. Many different scenarios have to be considered, with all of them having their own probability of actually happening. Also, spent fuel is not treated in any way in this study to give a clear view of the

radiotoxicity of it. In practice, spent fuel can be treated by plants to produce reusable MOX, or it can be fed into fast reactors to transmute the actinides into less radiotoxic isotopes.

2.5 constraints

If there were to be no constraints on the design and operation of a MSR, high power and breeding could be achieved. However the desire to keep the reactor technically feasible, safe, durable and and affordable leads to certain constraints that have to be met. Constraints that limit the performance and design of a MSR include: moderator and vessel lifetime (material sciences), chemical processing (chemistry, economics, feasibility), feedback coefficients (safety), fissile startup amount (feasibility), and radiotoxicity of the used salt (sustainability). There could be many more constraints that restrict the design and operation of a MSR, but those are beyond the scope of this thesis.

2.5.1 Graphite lifetime

If graphite undergoes high neutron fluence, it can become damaged and unsuitable for use in a MSR. Graphite is not only the moderator, but also a structural material of the core and is in direct contact with the salt. This makes replacing the damaged graphite a necessity and a comprehensive, time consuming task for which the reactor would have to be shut down. Graphite shrinks and expands if it is irradiated and this leads to structural defects in the graphite. An upper limit on the amount of radiation is used to determine the lifespan of the graphite moderator. For neutrons with an energy higher than 50 keV and fluences higher than $3 \times 10^{22}\text{n/cm}^2$ the damage becomes too severe. This fluence can also be written in terms of displacements per atom or dpa via 2 conversion factors:

$$1dpa = \frac{0.5}{7.62 \times 10^{20}} \times \Phi_{50}.$$

Here Φ_{50} is the neutron fluence of energies larger than 50 keV. 0.5 is the conversion factor from Φ_{50} to Equivalent DIDO Nickel Dose (EDN) and $\frac{1}{7.62\times10^{20}}$ is the conversion factor from EDN to dpa [8]. The resulting upper limit in terms of dpa is then 20 dpa. This upper limit for the neutron fluence of $3 \times 10^{22}\text{n/cm}^2$ was used in the MSBR and has been widely used afterwards, hence it will also be used in this thesis. For a 1-zone MSR one could say that the graphite blocks can be 'shuffled' once in a while so that every block will have undergone the same neutron fluence in the end as neutron fluence will be higher near the axis of the cylinder shaped core than on the sides. This results in all of the blocks being irradiated with the same average neutron fluence and all will thus have the same average lifetime, whereas otherwise blocks near the center will have shorter lifetimes due to the higher flux there.

For a 2-zone MSR this however is not (entirely) possible as the two zones contain blocks with different channel sizes. Within each zone the blocks can be shuffled, with only minor lifetime gain as the higher moderated fissile zone will have a higher average neutron fluence and cannot be interchanged with blocks from the fertile zone. Graphite lifetime in a 2-zone MSR that is optimized for high breeding gain will therefore always be shorter compared to a 1-zone MSR with the same power density. On the other hand, the flux in the inner zone will be more thermal, meaning a smaller part of the total flux will be of an energy higher than the damaging 50 keV. Graphite lifetime can be increased using multiple zones as well by

placing a less moderated zone in the center and a more moderated zone at the edges to create a more flat flux profile [7], [14]. This however decreases breeding capability of the MSR. The MSBR suggests a graphite replacement every 5 years. This is very short. Different other studies have shown that graphite lifetimes of up to 30 years are also possible with certain core configurations and reprocessing schemes. Minimal graphite lifetime that is set as a constraint in this study will be 10 years. This will be the peak lifetime (i.e. the graphite lifetime at the center of the inner zone of a 2-zone MSR).

2.5.2 Chemical reprocessing

Chemical reprocessing is expensive and energy consuming, therefore it should be minimized. Above that, there is a doubt if some chemical reprocessing schemes are even technically feasible. The reprocessing scheme as suggested for the MSBR treats the entire volume of the salt in 10 days. Such a reprocessing scheme gives very good results in terms of breeding and reactor operation, but is not considered to be practically feasible, because of its complexity. The slow reprocessing scheme as suggested in [15] is feasible though, and will, in certain reactor configurations, produce desirable results for breeding. This reprocessing scheme is therefore used in this research. In table 2.4 the cycle time of the different elements removed from the core is shown, as a result of this reprocessing scheme. The elements with a cycle time of 50 s and 2.4h are removed by the gas system and the elements with a 1 year cycle time are removed off line by various chemical processes, depending on the chemical properties of the elements. Pa is removed from the core to decay with a cycle time of 3 months. All transuranic elements (TRUs) are kept in the core continuously. A slow reprocessing scheme also favors

Table 2.4: Reprocessing scheme used in this study

Elements	Cycle time
Kr, Xe	50s
Zn, Ga, Ge, As, Se, Zr, Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te, I	2.4h
Zr, Ni, Fe, Cr, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er	1y
Pa	90d

the proliferation resistance of a MSR as more (n,2n) reactions with ^{233}Pa are possible, leading to a higher concentration of ^{232}U in the salt.

2.5.3 Feedback coefficients

Feedback coefficients are a measure for how the reactivity in the core changes after a change in temperature in the core. Negative (overall) temperature coefficients are desired to keep the reactor intrinsically safe. The temperature coefficients in a MSR can be divided into two main groups: the feedback coefficient for the graphite and the feedback coefficient for the

salt. The total temperature feedback coefficient is the sum of the underlying coefficients:

$$\left(\frac{\partial \rho}{\partial T}\right)_{total} = \left(\frac{\partial \rho}{\partial T}\right)_{graphite} + \left(\frac{\partial \rho}{\partial T}\right)_{salt}, \tag{2.21}$$

where T is the temperature in °C or K and ρ is the reactivity:

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

The feedback coefficient of the graphite is usually positive. This is due to a slight hardening of the thermal neutron spectrum when temperature is increased. The average number of fission neutrons produced per neutron absorbed in the fuel increases for neutrons of 0.2 eV and higher, for a mixture of ^{232}Th, ^{233}Pa and ^{233}U. This is because the fission cross-section of ^{233}U increases faster around that energy when neutron energy is increased. This is also the energy range where the hardening occurs. More fission neutrons per absorption leads to an increase of reactivity. With the feedback of graphite being positive, a negative temperature feedback coefficient for the salt is required for a safe reactor operation. The coefficient for the salt can be split into two coefficients, as the feedback effect in the salt is due to both the Doppler broadening and change in density of the salt:

$$\left(\frac{\partial \rho}{\partial T}\right)_{salt} = \left(\frac{\partial \rho}{\partial T}\right)_{Doppler} + \left(\frac{\partial \rho}{\partial T}\right)_{density}. \tag{2.22}$$

Density effects are small. When temperature rises, the density of the salt decreases, while the graphite does not significantly expand. The neutron spectrum then becomes more thermalized, as a part of the salt is moved out of the core. This results in a positive feedback coefficient for an over moderated core and a negative feedback coefficient for an under moderated core. The Doppler effect is the temperature feedback effect that is due to the broadening of cross section resonances with increasing temperature. This is the dominant effect in the salt. Thorium has its first neutron capture cross section resonance around 20 eV and this produces a negative temperature feedback effect. The total effect of the salt is negative as well. Having a negative overall feedback coefficient means an intrinsically safe reactor. Feedback coefficients are usually in the order of pcm/K. For this study the maximum safe temperature feedback coefficient is chosen to be -1 pcm/K.

2.5.4 Initial load

The MSR studied in this thesis uses ^{233}U as a fuel and is started up using that fissile isotope as well. Initial load is not accounted for in the definition for breeding gain and after the lifetime of the MSR the fissile inventory can be reused, so in effect it is not consumed. However, as ^{233}U is not naturally occurring, the reactor should be started up with as little ^{233}U as possible, see section 2.1.3. Looking at results from previous studies on salt composition, the thorium concentration should be low. With a lower initial load of thorium, the reactor needs less fissile material to become critical, as thorium will be the largest neutron absorber at the startup of the reactor. A lower thorium concentration on the other hand will compromise on breeding

as thorium is the fertile component in the salt. 12 mol% of thorium gives good results for breeding, graphite lifetime and initial fissile load [16]. The initial ^{233}U concentration should be around 0.3 mol% as sugested for the MSBR. However the optimal amount needed to startup the MSR studied here is searched for. With a 12 mol% startup amount of thorium the amount of ^{233}U will end up to be lower than 0.3 mol% for the studied core configurations, as will be seen in Chapter 4. Better-moderated zones, with a relatively high volume ratio, will have a higher overall capture to fission ratio, leading to a decreased initial concentration of ^{233}U. Also the higher volume ratio means less salt as a whole.

2.5.5 Radiotoxicity of the used salt

With radiotoxic waste being the largest drawback of todays nuclear reactors, keeping the radiotoxicity of the spent fuel, in this case used salt, as low as possible is an important aspect when designing reactors. Actinides, and especially transuranic elements have the largest contribution to the total radiotoxicity of the used salt. Altering the volume ratios of the MSR core, alters the energy distribution of the neutron flux and the average absorption cross-sections of different actinides, altering, on its turn, the concentration of different TRUs over time. Also the average power density in the core will have its effect on TRU concentrations in the used salt. Increasing the power density will increase the overall neutron flux in the core, leading to more neutron absorptions by different actinides. Then there is less chance for the actinides to decay. A hard constraint will not be set on this aspect of the MSR, however it can of course influence the decision for a certain core configuration.

2.6 Computation

For this study different aspects of the MSR were studied. These were all calculated with the (output of) the same model that simulates the MSR. A description of this model and the methods used to calculate different aspects are explained below.

2.6.1 MSR

Calculation of the operation of the core is done with the SCALE code system [22], in combination with an in-house developed and modified burnup code, LOWFAT. A PERL script that executes different modules of the SCALE code system and LOWFAT, as well as reads input and produces output is used. First the script reads all the desired input data and translates this into different settings as the geometry of the core, different element removal constants, calculation time, desired output etc.. The 1-zone core is calculated as an infinite lattice of graphite blocks with a cylindrical fuel channel running lengthwise through them. A XSDRN input file for a unit cell is created, a circle (fuel channel) with an annulus (graphite) around it and a white boundary to simulate an infinite lattice. For the 2-zone core a precise KENOVI input file is created, that builds up an exact core, as in Figure 2.6. A slice of $\frac{1}{6}$ of the core is then taken out with mirror boundaries placed at the sliced edges, to shorten the calculation time. This can be done because of symmetry (see Figure 2.6). XSDRN is a 1-dimensional deterministic neutron transport code, whereas KENO is 3-dimensional and uses Monte Carlo. Because of this difference the 2-zone core needs much more calculation time (about 15 times longer).

Once the core is built up, the initial load of ^{233}U is searched for. CSAS/KENO (for the 2-zone MSR) and CSAS/XSDRN (for the 1-zone MSR) of the SCALE code system then checks for criticality until the right value of k_{eff} (for the 2-zone core) or k_{∞} (for the 1-zone core) is found with the corresponding initial load of fissile material. When the initial fissile load is found, a burnup calculation is done for the first timestep with a guessed feed rate of fissile material. The burnup calculation is done with the modified LOWFAT code that uses ORIGEN-S format cross-sections for every zone separately, prepared by CSAS and COUPLE. The LOWFAT code also takes into account the removal of nuclides from the core to simulate chemical reprocessing. Also the $^{233}UF_4$ feed rate is taken into account as well as the inventory and decay of nuclides in the stockpile. A mixing constant is used to simulate the mixing between two zones. The code assumes that the salt is irradiated half of the time, to simulate the out of core time, as no flow is modelled in the code. Next a CSAS/KENO calculation is done to check k_{eff}. If it is satisfied (see Table 2.5), the timestep is accepted and a new timestep follows, otherwise a new feed rate guess is done until it satisfies the right value of k_{eff}. Once a right value is found, the code moves on to the next timestep, until the desired time of operation is achieved. During every timestep the flux (in a zone) is assumed to be constant and is adjusted for every new timestep. Table 2.5 shows different settings used for the simulation of the 1-zone and the 2-zone core.

Table 2.5: Computational settings

Setting	1-zone MSR	2-zone MSR
Timestep (s)	2592000	2592000
Energy groups	238	238
Accepted multiplication factor	1.03	1.005
Deviation of multiplication factor		0.0009

It can be seen that the multiplication factor that is accepted for a timestep is much higher for the 1-zone case than for the 2-zone case. This is because for the 1-zone case an infinite core is calculated, whereas the 2-zone core is finite. The corresponding value of k_{eff} for a 1-zone core is then calculated after finding the non leakage probability of the core:

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

where the P_{NL} is the non leakage probability and is calculated as follows:

$$P_{NL} = \frac{1}{1 + L^2 B^2}.$$

B is here the geometrical buckling of the cylindrical shaped core and L the diffusion length. For the 1-zone core with different volume ratios this is found to be no smaller than 0.99. However this might change as the inventory of the reactor changes due to burnup. Also as no flow is modelled in the code, a fraction of the delayed neutron precursors will exit the core and will therefore not contribute to the multiplication factor. The fraction of delayed neutrons is 0.0027 for ^{233}U. Depending on the velocity of the flow through the core a certain part of the precursors will emit neutrons outside of the core. As only long term criticality control is modelled (feeding ^{233}U), there is chosen for values of $k_{eff} > 1$. For a working MSR short term reactivity control like control rods will be needed.

Every timestep output is generated with which analysis of different aspects of the core can be done. Analysis is important as it gives a measure of performance of the reactor. Constraints can be met when feedback coefficients, graphite lifetime, breeding gain and radiotoxicity are calculated. The method of calculation for these aspects will be discussed below.

2.6.2 Feedback coefficients

In order to calculate feedback coefficients one needs to perform calculations for k_{eff} with very low variance, as reactivity changes are only very small (in the order of pcm/K). This takes a lot of time when using a Monte Carlo type calculation like KENO. Therefore feedback coefficients are only calculated at the end of every run (i.e. at the end of a 50 year reactor operation). To make sure this is not a large over or underestimation, the feedback for 1 configuration is also calculated at different time intervals. In order to calculate feedback coefficients, KENO is run three times, with very low variance (in the order of pcm) to calculate k_{eff}. Once

as a reference, once with the graphite temperature increased and once with the whole core temperature (i.e. salt and graphite) increased. The change in k_{eff} and thus the reactivity ρ from the reference is than a measure for the temperature feedback of the core and the graphite. The increase in temperature is chosen to be 100K to shorten calculation time, as the variance can be higher than for larger temperature increases. Calculation time is chosen to be 500 hours for every SCALE-6 run, which corresponds to a deviation in k_{eff} of around 9 pcm (per 100K). The temperature feedback is chosen to be linear for this 100K, to be able to give a value per K.

2.6.3 Graphite lifetime

Only the average spatial neutron fluxes of the energy groups in each zone are output by the script. This is enough to give an estimate of the average as well as the peak graphite lifetime. The average graphite lifetime for a 2 zone configuration is calculated by taking the sum of all the neutron fluxes higher than 50 keV in the inner zone at the end of a calculation and dividing the maximum neutron fluence (3×10^{22}n/cm^2) by that number. Neutron fluxes in the outer zone will not be the limiting factor, as they are lower anyway. For the peak lifetime, the core is treated like a 2 zone slab reactor. It is assumed that the neutron flux is cosine shaped in the inner zone. A cosine with its equilibrium at the level of the average neutron flux of the outer zone and its average at the level of the average flux of the inner zone is used to reconstruct the flux profile in the inner zone. Figure 2.8 demonstrates this. There are

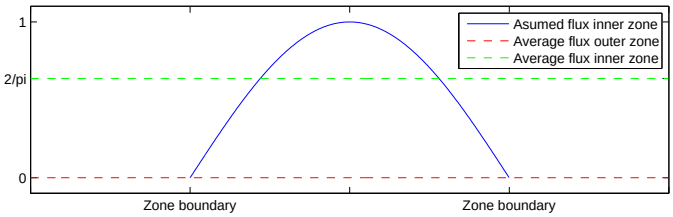

Figure 2.8: Method used for the calculation of the maximum neutron flux in the inner zone. Here the maximum is at 1, the average neutron flux in the inner zone is at $\frac{2}{\pi}$ (average value of a cosine) and the average neutron flux in the outer zone at 0.

some assumptions made in this estimation of the graphite lifetime:

- The flux at the end is taken to be the flux throughout the whole life of the MSR.
- The flux in the inner zone has a perfect cosine distribution, with it's equilibrium at the average flux of the outer zone and its average value at the average value of the inner zone.

The thermal neutron flux at the end of the life of the MSR will be lower than the average neutron flux, as the ^{233}U concentration is higher, because fission products and actinides with high capture cross-sections will build up over time. To keep the core critical, the amount of ^{233}U has to be increased. Increasing the amount of ^{233}U will cause the overall flux to drop if

one desires the same power. But also due to the fission products and actinides the neutron flux will harden over time, making it more damaging to the graphite. The flux profile in the inner zone will not be a perfect cosine with an equilibrium value at the average of the outer zone flux. Rather, the equilibrium will lie higher, resulting in a lower peak value (overestimate). The flux shape will be more bell shaped as the flux at the zone boundary is nonzero, resulting in a higher peak value, because the peak of a bell will lie further away from its average value than the peak of a cosine (underestimate). Taking all assumptions into account, this will provide an estimate of the graphite lifetime that will lie close to the true graphite lifetime.

2.6.4 Breeding gain

The breeding gain is calculated using the equations in section 2.3. Equation 2.11 is used as it gives a measure for the breeding gain in the stock. All other bred uranium will be needed in the core and is therefore not considered as excess bred fissile material. Every timestep breeding gain is calculated for that period. After the whole calculation (i.e. 50 years) an average value of the breeding gain of all the cycles is then calculated to be the overall breeding gain. For the 1 zone case where there is searched for the power density to produce a self-breeder, total average breeding gain in the stockpile is calculated for a higher and a lower power density than the previous cycle, resulting in a negative and a positive breeding gain (providing the boundaries are wide enough). The power density is now set between the two values, and depending on the result it will be set at either the high boundary or the low boundary. This is repeated until the power density is found that results in a breeding gain of 0 (within a set error).

2.6.5 Radiotoxicity

Actinides and fission products When solving differential Equation 2.18 and the number of daughter nuclides m is high, the calculation can become very long and complex. Radiotoxicities of actinides in this study are therefore calculated using a data library containing radiotoxicity values (in Sv/mole) for actinides at different moments in time, up to half a million years in the future also used in the BIFTOX program [9]. Values for radiotoxicities of the different actinides used are per mole and include the radiotoxicities due to the decay of all the daughter nuclides of the specific nuclides.

For the long-lived fission products no comprehensive database is needed, because all except ^{126}Sn decay directly to a stable isotope. ^{126}Sn decays in just one step, via ^{126}Sb, which decays much faster. On a large timescale, the fast decay lets us treat the two steps as if it were one. The radiotoxicity of long-lived fission products is then easily calculated with Equation 2.16. One must then take for the dose equivalent of ^{126}Sn the sum of both the dose equivalent of ^{126}Sn and ^{126}Sb. As can be seen in table 2.4 most fission products are removed, including almost all long-lived fission products: ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn and ^{129}I. ^{135}Cs is not removed. The removed fission products are however kept track of to calculate radiotoxicity after reactor operation on the total amount of long-lived fission products produced during a

whole cycle. The decay of the long-lived fission products between removal from the salt and the end of a cycle is neglected, as their half-lives are thousands of times larger than the time concerned.

Uranium ore In order to view the radiotoxicity of spent fuel in perspective, one needs to define the radiotoxicity of uranium ore. The value of radiotoxicity for uranium ore is defined here as the radiotoxicity due to the amount of uranium ore mined for the production of 1 GWthy (1 year @ 1 GigaWatt of thermal energy) of energy in a generation II Pressurized Water Reactor (PWR). The average uranium burnup in a uranium fuelled generation II PWR is around 40-50 GWd/TU (TU = metric tons of uranium) [5], meaning 8.14 tons of Uranium is needed for 1GWy. In generation III reactors a burnup of 200GWd/TU is achievable. Using the BIFTOX data the radiotoxicity of 11.1 tons of uranium is then: 9.9×10^4Sv. ^{234}U has a large contribution to this value, although only small traces are found in uranium ore (0.0054%). The half-life of ^{234} on the other hand is thousands of times smaller (2.44×10^5a, compared to 4.47×10^9a for ^{238}U and 7.04×10^8a for ^{235}U), leading to a much higher activity per mole.

Chapter 3

One Zone MSR

Before trying to optimize a MSR using two zones, an analysis of a more simple form of a MSR is made. This chapter focuses on the radiotoxicity of the actinides and fission products in the used salt of a MSR as well as the breeding gain of a one zone MSR. For the radiotoxicity calculations a 1-zone MSR after 50 years will be compared to the nuclear power plant in Borssele, a PWR. The MSR studied here will have the same specifications as proposed in [16], as it gives good results for breeding as well as graphite lifetime and feedback coefficients. Specifications can be seen in Table 3.1. The volume ratio is defined by the volume of the graphite in the core divided by the volume of the salt in the core. As for the chemical reprocessing, the extraction scheme in Table 2.4 was used. With these dimensions the reactor

Table 3.1: Specifications of the 1-zone MSR

Core height	500 cm
Core radius	250 cm
Fuel channel diameter	9 cm
Volume ratio	2.1
Salt temperature	1000K
Startup salt composition	LiF-BeF_2 (67%-33%) with 12 mol% $^{232}ThF_4$ and approx. 0.25 mol% $^{233}UF_4$
Graphite density	1.84 g/cm^3
Graphite temperature	1050 K

core has a total volume of 98 m^3. The amount of salt is determined by the volume ratio. A volume ratio of 2.1 corresponds to a salt volume of 32 m^3 in the core. Half of the time the salt mixture is outside the core, so the total amount of fuel salt will be double the amount of salt in the core at any time.

First the MSR is operated for 50 years with a constant power density of 5MW/m^3, amounting to a total thermal power of 490 MW. After these 50 years the salt composition is compared to the spent fuel of a PWR.

Next, the reactor is run for four cycles of 50 years, reusing the fuel salt every cycle. In between cycles, only fission products are removed (all actinides stay in the salt). The salt is assumed to be processed for 1 year before starting up the reactor again with the cleaned salt. A MSR does not actually need cycles, as it can be run continuously by feeding ^{233}U and by reprocessing the salt. On the other hand, graphite lifetime is limited, see Section 2.5.1. This means that the graphite in a MSR should be replaced once in a while, preventing the MSR to run continuously. Also, other maintenance on the reactor will be necessary, requiring a reactor shutdown.

Finally, a 1-zone MSR is run for the same cycles as for the case with a set power density, but now the power is adjusted to produce a self-breeder. If the power is increased, the neutron flux is increased. Keeping the same reprocessing scheme results in more protactinium capturing a neutron, making it unable to decay to ^{233}U in the core, requiring a higher feed rate. Also less protactinium can be extracted from the core, decreasing the amount of protactinium in the stockpile. Less ^{233}U will be bred in that way and the breeding gain will decrease.

3.1 MSR vs a PWR

In Figure 3.1 the radiotoxicity per GWy of thermal energy due to the most radiotoxic actinides present in the salt of a MSR is plotted against the time after reactor shutdown. From [10]

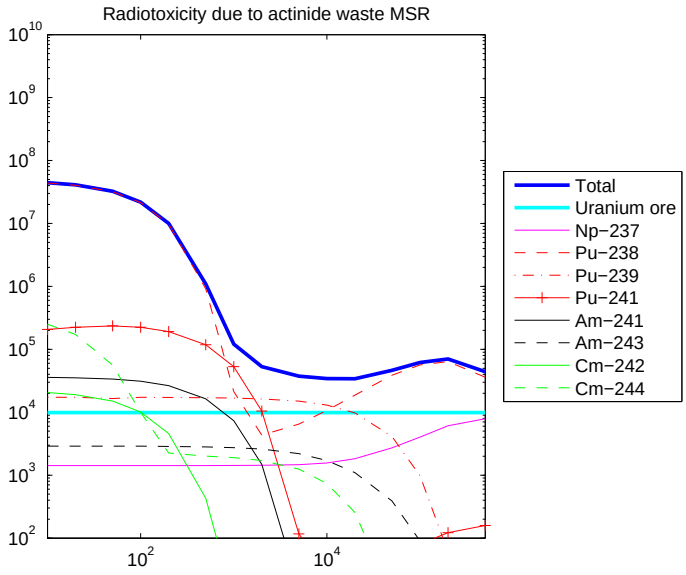

Figure 3.1: Radiotoxicity as a function of time due to the different actinides in the fuel salt of a MSR compared to the radiotoxicity of an amount of uranium ore needed to achieve the same energy in a generation II reactor. Note that this is Radiotoxicity [Sv] per energy produced [GWth*y], so the actual radiotoxicity after 50 years is higher as an energy of 24.5 GWth*y is produced every 50 years.

the transuranic composition of the spent fuel of the Borssele power plant is obtained. A plot is made (see Figure 3.2) to see the contribution of the most abundant TRUs to the total radiotoxicity of the TRUs in spent fuel from the Borssele power plant. As a comparison to the radiotoxicity of uranium ore needed for 1 GWth*y is also plotted. It is clear that the radiotoxicity due to TRUs of this PWR is much higher than that of the MSR studied. Both radiotoxicity plots are characteristic for the different fuel cycles. For the PWR ^{241}Pu, ^{239}Pu and ^{238}Pu are the most dominant actinides, giving the graph its characteristic shape. These are mainly the products of the neutron capture of ^{238}U. The shape of the graph of the MSR on the other hand is mainly formed by ^{238}Pu. As fission product yields are roughly the same (see Figure 2.7), one can say that a MSR has a much cleaner waste composition. Also, comparing the curves of Figure 3.2 and Figure 3.1 one can see that the initial peak of the MSR drops more rapidly than the one of the PWR, as the half-life of the most radiotoxic isotope of the MSR is much shorter than that of a PWR. It should be noted that the radiotoxicity levels for both the spent fuel of a PWR as well as of the used salt of an MSR are per GWy of thermal energy. MSRs which operate at higher temperatures will have a higher efficiency of thermal to electrical energy conversion than PWRs.

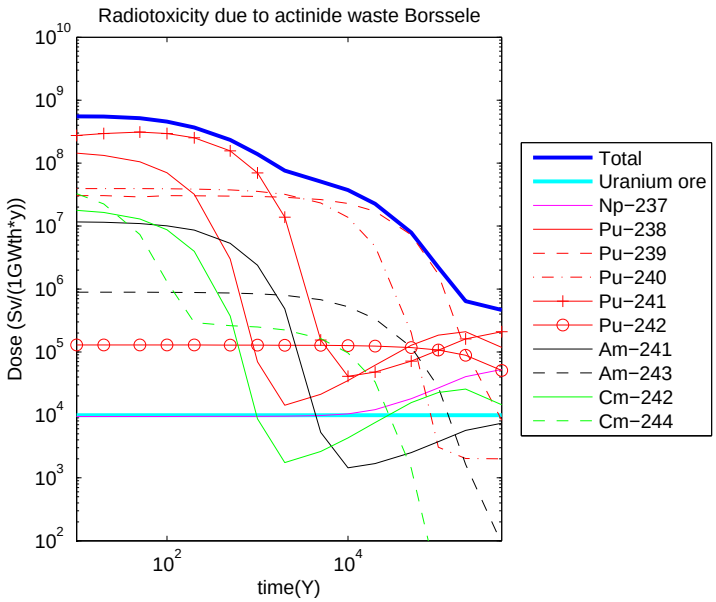

Figure 3.2: Radiotoxicity as a function of time due to the presence of the different actinides in the spent fuel of the nuclear power plant in Borssele, compared to uranium ore of the same amount.

3.2 Cycled MSR

Because of the continuous reprocessing of the salt of a MSR and the continuous feeding of ^{233}U and thorium, it is possible to operate a MSR for extended amounts of time. It is therefore interesting to investigate the case where a MSR is run for a longer period. One way to do

that is to run the MSR for a certain time, removing fission products in between these 'cycles'. After every cycle intensive maintenance and replacements can be done in the reactor. Here a cycle runs for 50 years. Shutdown time between the cycles is chosen to be 1 full year, after which all fission products are removed from the salt. All the actinides stay present in the mixture after every cycle. Four of these consecutive cycles are simulated for two cases:

- A constant power density of 5MW/m^3(same as the case in Section 3.1)
- A self-breeder reactor with corresponding power density.

The results of these two cases are discussed in the following subsections.

3.2.1 Constant power density

Here the effects, such as BG, radiotoxicity of the used salt, feedback coefficients and graphite lifetime of a cycled MSR with a constant power density of 5 MW/m^3 are analyzed.

Breeding gain

From [16] it is known that the power density of 5 MW/m^3 gives a positive breeding gain when operated for 50 years. The breeding gain in the next three cycles is shown in Table 3.2. It shows that the breeding gain is the highest in the second cycle and decreases in the following cycles, resulting in a breeder in the first two cycles and a converter in the last two cycles.

Table 3.2: Breeding gains for 4 consecutive 50 year cycles at 5 MW/m^3

cycle	BG
1	0.0022
2	0.0090
3	-0.0010
4	-0.0080

When comparing the different cycles there are two main differences in the reactor operation:

- The reactor is started up with a different composition of actinides each time. After every cycle the salt is cleaned from fission products, but all actinides stay in the salt.
- The first 50 years the reactor is started up with fresh salt, containing (apart from the salt) only ^{232}Th and ^{233}U.

Most abundant actinides, other than ^{232}Th, ^{233}Pa and ^{233}U, that are present in the salt are shown in Table 3.3. Actinides play an increasing role in the neutronics of the reactor, as they build up and are not cleaned from the salt after every cycle. The table shows that ^{234}U and

Table 3.3: Thermal macroscopic capture cross-sections of the most abundant actinides present in the salt (except ^{232}Th, ^{233}Pa and ^{233}U). Note that ^{235}U and ^{239}Pu are fissile isotopes.

	$\Sigma_c(10^{-3}\text{cm}^{-1})$			
Isotope	1	2	3	4
^{234}U	2.25	2.82	3.09	3.25
^{235}U	0.84	1.17	1.31	1.40
^{236}U	0.02	0.04	0.05	0.06
^{237}Np	0.08	0.21	0.38	0.45
^{238}Pu	0.17	0.85	1.52	2.02
^{239}Pu	0.01	0.04	0.08	0.11

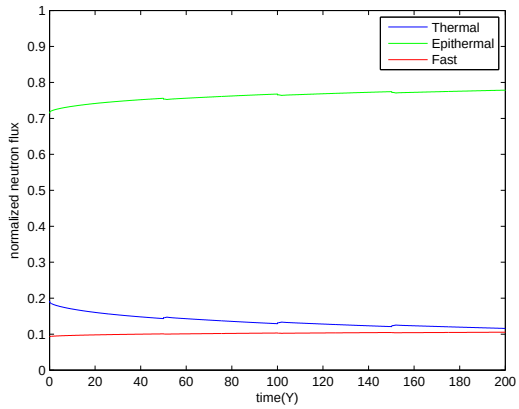

Figure 3.3: Thermal, epithermal and fast neutron fluxes in the core as a fraction of the total neutron flux.

^{238}Pu have the largest macroscopic capture cross-sections. An equilibrium is not yet formed after 4 cycles, but the concentrations of the different TRUs are flattening. The actinides (and fission products with large neutron capture cross-sections) have a hardening effect on the neutron spectrum in the core. This can be seen in Figure 3.3. The sawtooth effect is due to the fact that all fission products are removed between cycles. This causes a stepwise softening of the neutron spectrum. Both the table and the figure show that an equilibrium of actinides and neutron spectrum is being formed. Thermal neutron capture has a negative effect on criticality, which is compensated by increasing the amount of ^{233}U added to the core and decreasing the flux to keep the reactor critical, while operating at the same power density.

The hardened spectrum has two effects on the BG. As the spectrum hardens, less protactinium will be formed in the core, as the average capture cross-section of ^{232}Th will decrease as Figure 2.1 shows. The protactinium on the other hand will capture less neutrons as well due to the hardening spectrum. This latter effect is smaller, as Figures 2.1 and 2.2 show. As less protactinium is being formed in the core, less will be extracted, lowering the amount of protactinium in the stockpile and ultimately the BG in the stock. The above effects result in a decreasing BG throughout the cycles. However, the second cycle shows a higher BG than the first, which is due to the second difference between the cycles. The second difference

between the cycles has its effect on BG as well. As the reactor is started up for the first time, there is no protactinium in the core nor in the stockpile. An equilibrium concentration of protactinium in the core is reached after about three months. During these months, relatively large amounts of ^{233}U are fed into the reactor, as the core does not yet breed the maximum amount of ^{233}U. The same holds for the stockpile, were the equilibrium concentration of protactinium is reached after approximately 10 months. This is mainly determined by the the removal rate from the core. After the protactinium reaches its equilibrium concentration in both the core and the stockpile, the reactor as a whole produces as much uranium as possible at that moment. Cycle 2, 3, and 4 do not suffer this negative effect on BG. Although the majority of the protactinium has decayed in the year between cycles, no ^{233}U will need to be fed during the first year and a half of the second third and fourth cycles. This is because all the protactinium in the core has decayed to ^{233}U in the year between two cycles, resulting in a relatively high concentration of fissile material in the core. Also the salt is cleaned from all fission products, decreasing the capture to fission ratio. These two facts explain the positive jump in breeding gain from cycle 1 to cycle 2.

Radiotoxicity of transuranic elements

In Figure 3.4 the radiotoxicity of the TRUs from used salt of the 5 MW/m^3 cycled MSR is shown after each cycle of 50 years. As with the 50 year case compared with the PWR, by far the most radiotoxic long-lived TRU directly after reactor shutdown is ^{238}Pu. It stays the most dominant TRU throughout almost all of the next 500,000 years for all four cycles. It can be seen that radiotoxicity is increased after every cycle, and there seems to be forming an equilibrium as cycles 3 and 4 hardly differ from each other, confirming the trend in Figure 3.3. Also the TRUs with higher mass number have an increasing fraction in the total radiotoxicity. This is the result of the fact that the total actinide composition shifts to isotopes of higher mass number through (multiple) neutron captures in the salt due to the increasing neutron fluence throughout the 4 cycles.

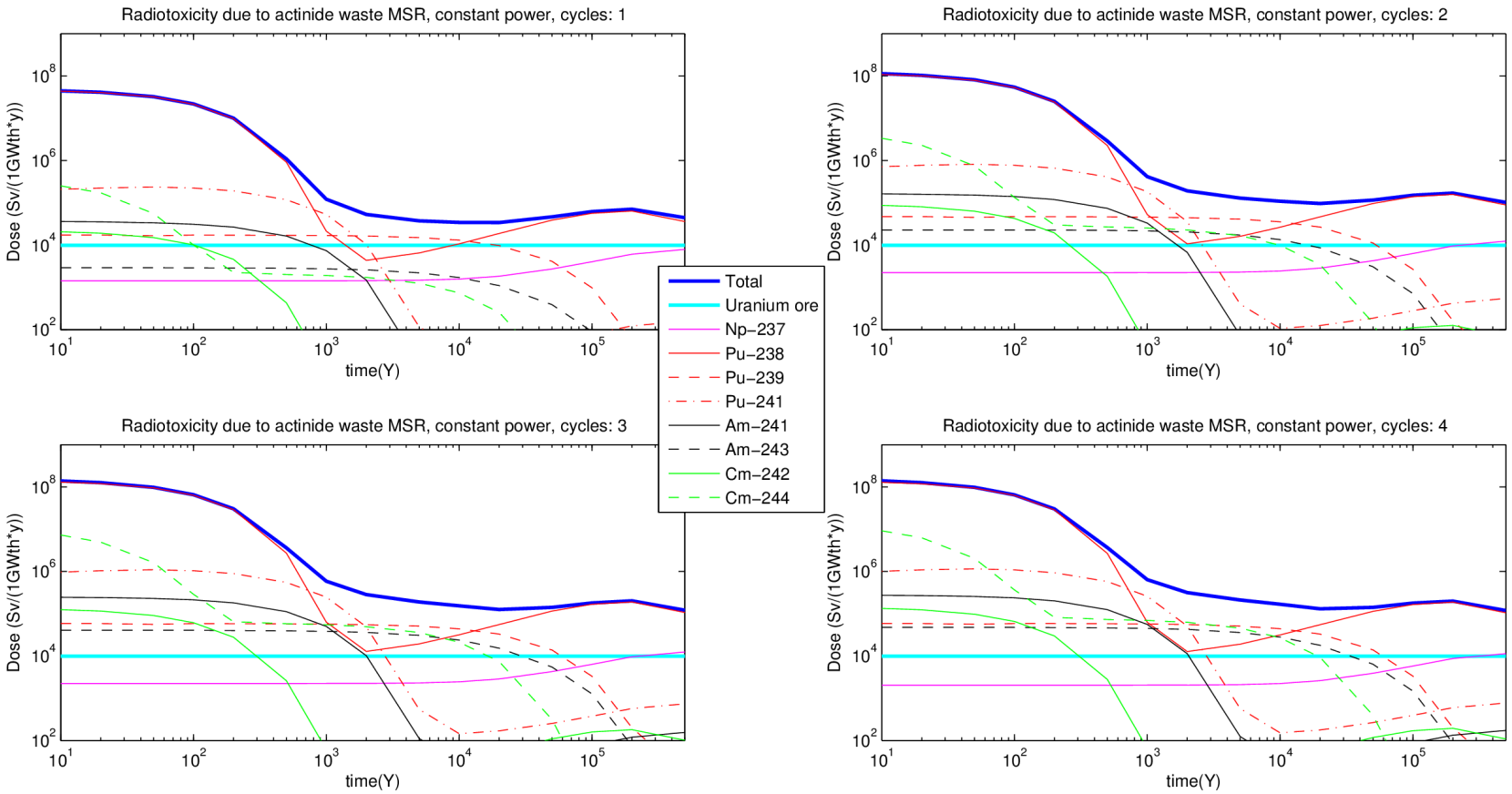

Figure 3.4: Radiotoxicity as a function of time due to the most abundant TRUs in the fuel salt of a MSR compared to the radiotoxicity of the uranium ore needed for the same energy in a PWR. Each cycle represents 50 years of reactor operation. Note that this is Radiotoxicity [Sv] per energy produced [GWth*y], so the actual total radiotoxicity after a certain amount of cycles will be larger, as in 50 years an energy of 24.5 GWth*y is produced.

Radiotoxicity of long-lived fission products

Figure 3.5 shows the radiotoxicity of long-lived fission products after reactor shutdown for the different cycles. This includes all the long-lived fission products that have been removed by chemical processing during the reactor operation, and the fission products that have been removed after reactor operation to start the next cycle with a fresh salt. Figure 3.5 shows that the radiotoxicity of fission products normalized for 1 GWthy remains constant. Only small variations that are not visible on the logarithmic scale of the graph occur. The reason for these constant values is that the amount of fission products scales with the power of the reactor. The isotopes concerned are formed by fission and the neutron capture or decay of other fission products. This means that the produced amount of fission products will remain constant when normalized for energy and that the composition is only dependent on fission yields. Although the composition of fissile material changes (^{235}U and ^{239}Pu concentrations increase with operation time), the composition of the fission products will remain constant as fission yields do not differ much. Note that the radiotoxicity values are for all the long-lived fission products produced up till that time, also in previous cycles. Even though the salt is cleaned after every cycle, also the fission products of previous cycles are accounted for in the calculation of radiotoxicity. The radiotoxicity of fission products plays a decreasing role throughout the four cycles. One year after the reactor shutdown the share of radiotoxicity due to long-lived fission products of the total radiotoxicity (of long-lived fission products and TRUs) is: 0.018%, 0.006%, 0.004% and 0.002% respectively. This is a very small share. This is due to the fact that the amount of TRUs per unit energy increases throughout the cycles. A larger fraction of non-fissile actinides builds up throughout the cycles. This means that the energy production in this particular cycled MSR will become less clean over time. Although the long-lived fission products only play a small part in radiotoxicity after reactor shutdown, their halflives are large compared to the TRUs (see Table 2.3), and will play an increasing role in the total radiotoxicity of the salt, as can also be seen from Figure 3.5. However, the radiotoxicity of the long-lived fission products allways remains below the level of the amount of uranium ore needed per unit energy.

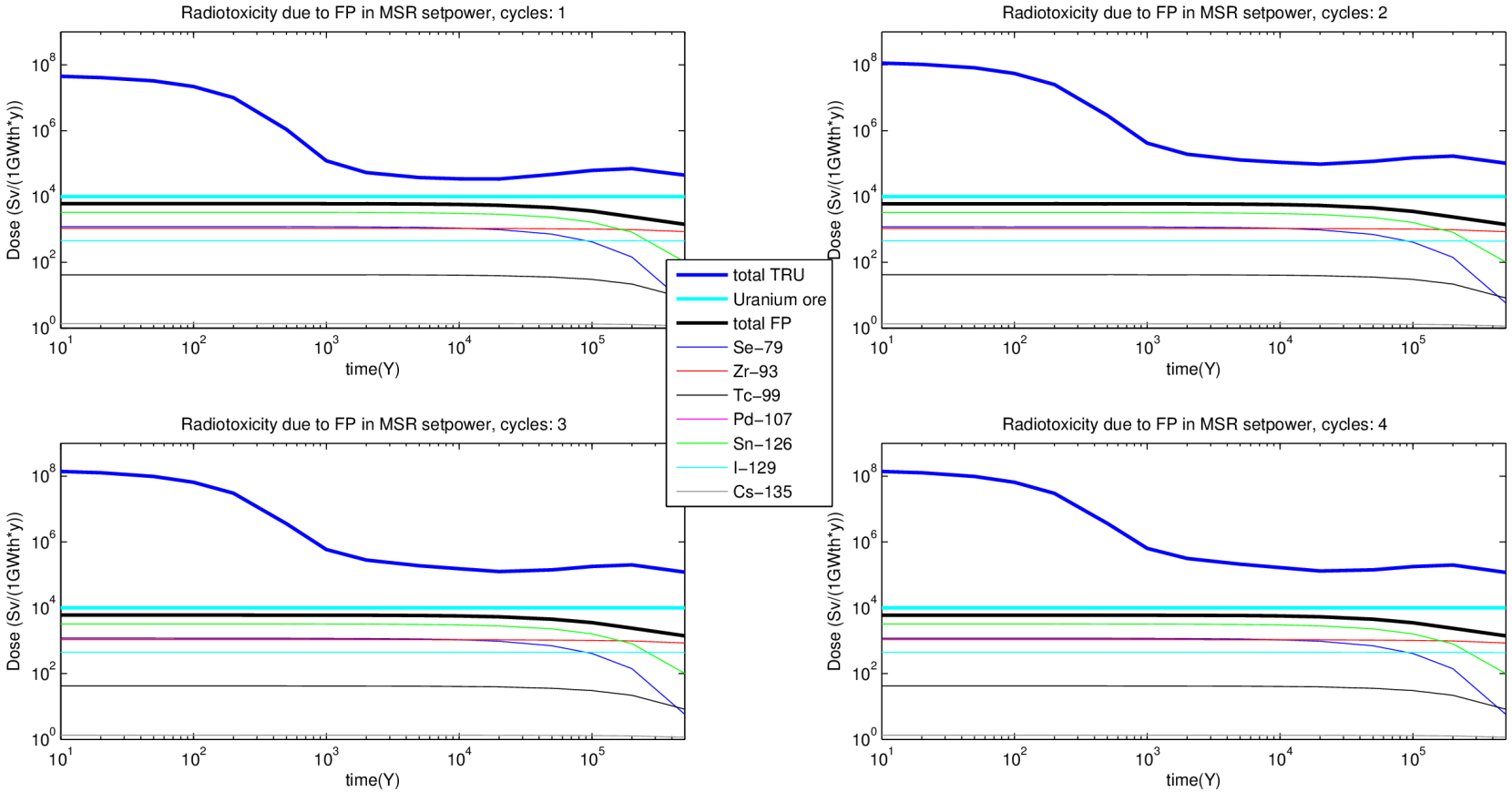

Figure 3.5: Radiotoxicity of long-lived fission products per GWth*y of energy produced, as a function of time for the four cycles of 50 year reactor operation.

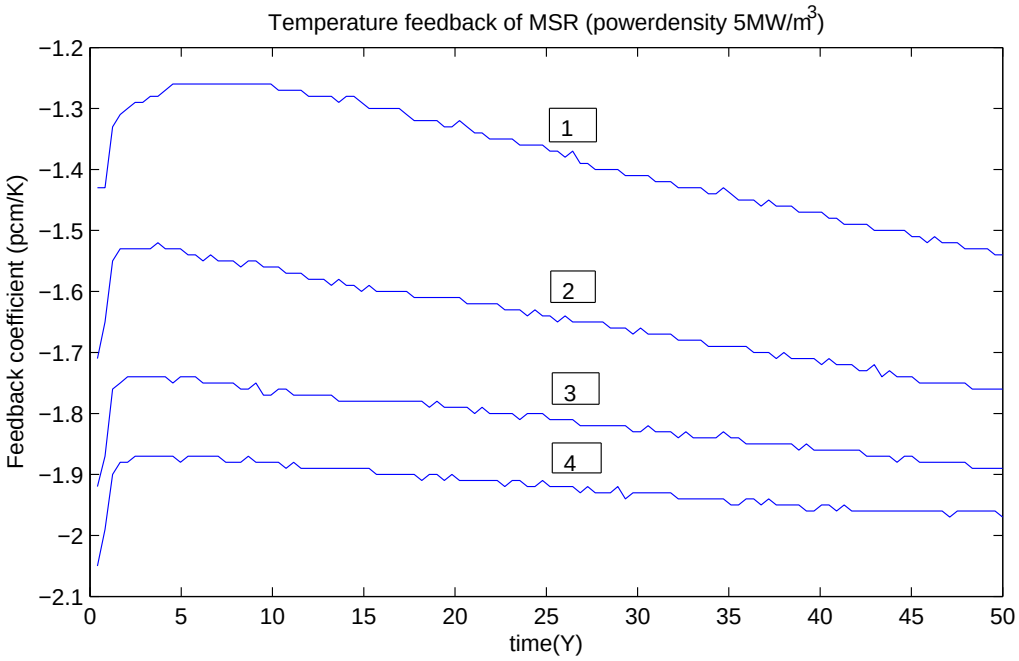

Figure 3.6: Temperature feedback for every cycle, for a MSR with a set power density of 5 MW/m^3

Feedback coefficients

From [15] we know the MSR operates under the safe chosen boundary of -1.5 pcm/K of total temperature feedback coefficient. What happens in the next cycles, where the different inventory can influence the temperature feedback? The result can be seen in Figure 3.6. First of all it shows that the end of life feedback coefficient is not representative for the whole lifetime of 1 cycle, as the feedback varies and has its lowest values at the start and end of every cycle. Negative feedback is stronger after each cycle. Feedback of the salt is not affected a lot during the 4 cycles and stays around -3 pcm/K. Graphite feedback effects on the other hand vary from 1.7 in the first cycle to 0.9 pcm/K in the fourth cycle. Hardening of the spectrum causes this change in graphite temperature feedback. The hardened spectrum causes the thermal peak to shift to higher energies moving it further away from the resonance (discussed in Section 2.5.3) as graphite temperature is increased.

Graphite lifetime

The graphite lifetime, due to high energy neutron fluence, is shown for every cycle in Table 3.4. It is considered that every cycle is started with 'fresh' graphite. It is clear from the table that the graphite lifetime slightly increases as cycles go up. This is a result of the total flux being decreased to maintain the same power density, whilst increasing the ^{233}U concentration to compensate for the thermal neutron loss in. During a cycle the flux hardens, as a result of more thermal neutron capture by the increasing fraction of fission products and TRUs. At the end of a cycle, the contribution of the average flux to the graphite lifetime is therefore

Table 3.4: The graphite lifetime at peak power and average power in years for every cycle

	Lifetime (y)	
Cycle#	peak power	average power
1	21.2	34.0
2	21.9	34.8
3	22.7	35.9
4	23.1	36.7

larger. Lifetimes for a cycled MSR as studied here are not long enough to last a whole cycle, even if reshuffled once in a while. However graphite lifetimes are large when compared to other studies, like the MSBR.

3.2.2 Self-breeder

Interesting is the case were the MSR breeds as much ^{233}U as it consumes, a self-breeder. A self-breeder can achieve the highest power density without decreasing the global stock of ^{233}U. By increasing the power of the reactor, the BG in the stock is set back to zero. A higher power density implies a higher flux in the core. This leads to more protactinium to capture a neutron, lowering breeding gain. An iterative script (see Section 2.6) is used that calculates the core power to reach an average BG of zero over a period of 50 years. The breeding gain will not constantly be zero, because the power is kept constant during a 50 year cycle, but over a 50 year time span the BG in the stock will amount to zero, i.e. after 50 years the total amount of fissile uranium in the stock will equal the total amount of fissile uranium in the stock at the startup of the reactor. The same aspects as studied for the set power density case above (except for BG, which will be 0 for all cycles), will be analyzed for the self-breeder case as well. Three cases are considered: a volume ratio of 2.1 and channel diameter of 9 cm, a volume ratio of 2.4 with a channel diameter of 7 cm and a volume ratio of 2.8 with a channel diameter of 5 cm. For simplicity, these three cases will from here on be referred to as MSR 2.1 MSR 2.4 and MSR 2.8. The results are shown in Table 3.5. For the first cycles, the power densities, graphite lifetimes and temperature feedback coefficients were already calculated in [16]. The three cases from [16] are chosen because they gave good results with respect to those aspects in the first cycle. What happens in the next cycles is discussed below.

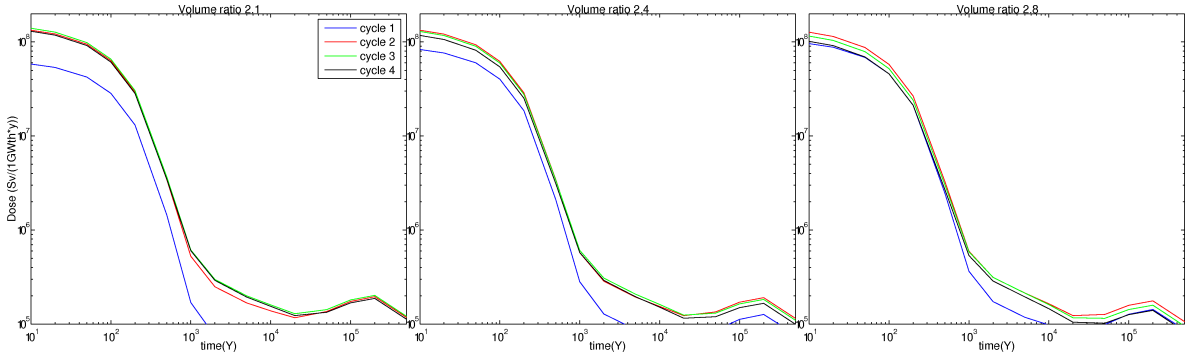

Figure 3.7: The total radiotoxicity of TRUs, normalized to 1 GWthy of energy, for the different self-breeder reactor configurations and the four cycles

Table 3.5: 3 cases of self-breeder reactors: MSR 2.1, MSR 2.4 and MSR 2.8. The power density corresponds to a self-breeder in that particular cycle. Also graphite lifetime and temperature feedback for all 4 cycles of the 3 cases are shown. The last column shows the total energy production after all the four cycles.

MSR (Volume ratio)	Channel diameter (cm)	Power density (MW/m^3)				Graphite lifetime peak-power/average-power (years)				Temperature feedback (pcm/K)				Total energy (GWthy)
		1	2	3	4	1	2	3	4	1	2	3	4	
2.1	9	5.8	6.44	4.47	3.12	18.3/29.4	17.2/27.4	25.6/40.6	37.3/59.2	-1.26	-1.56	-1.81	-2.03	97
2.4	7	7.04	6.30	4.40	3.28	15.4/24.6	17.8/28.2	26.1/41.3	35.3/55.8	-1.21	-1.59	-1.80	-1.90	103
2.8	5	7.84	6.68	4.92	3.95	14.1/22.4	17.0/26.9	23.4/37.0	29.4/46.4	-1.25	-1.64	-1.83	-1.91	115

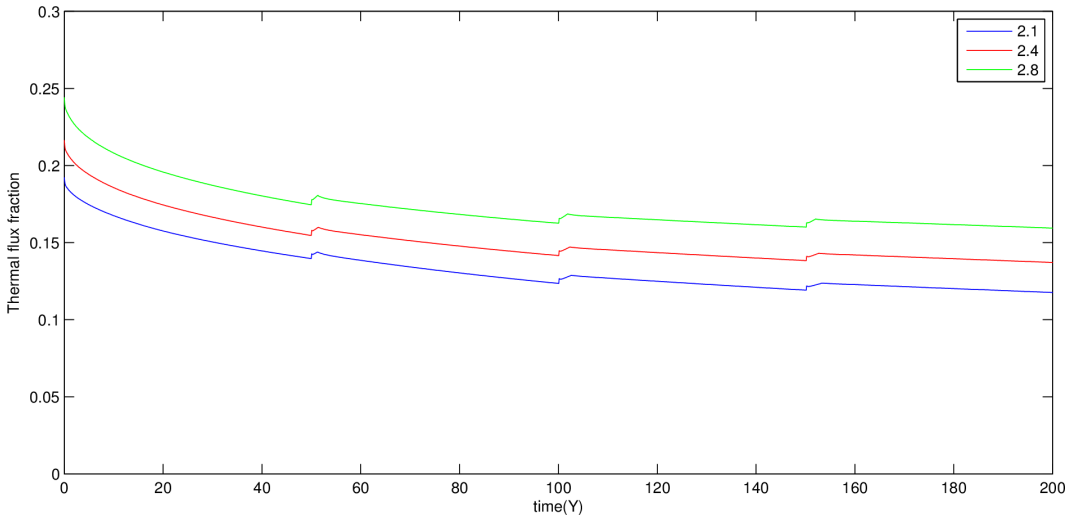

Figure 3.8: Thermal flux as a fraction of total neutron flux for the three self-breeder cases.

Power Density

Table 3.5 shows the different power densities of the three MSRs. Note that the power densities in the salt differ even more from each other, as both the volume ratio and the power densities of the whole core are varied to obtain a self-breeding MSR in each case. It is shown that the power density of MSR 2.1 follows the same pattern as the breeding gain for a set power density in the four consecutive cycles. The power density goes up after the first 50 years, but is set to a lower value in cycles 3 and 4. On the other hand, power density needs to decrease after each cycle for MSR 2.4 and MSR 2.8. Figure 3.8 shows the fraction of thermal flux over the four cycle lifespan of the 3 different reactors. It is clear that the thermal flux increases with the volume ratio. In the higher volume ratio cases the amount of graphite is larger and the channel diameter is smaller, leading to a better moderated flux. This thermalised flux leads to a larger fission cross-section of ^{233}U, meaning less ^{233}U needs to be fed into the core to maintain criticality and produce the same power as for a less moderated core. In this way a core with a higher volume ratio will operate at a higher power density for the self-breeder case. The higher power density will then produce more actinides and fission products with high capture cross-sections. To keep the reactor critical and to compensate for the hardening of the flux, more uranium will need to be fed into the core. This on its turn lowers the breeding gain, which is compensated by lowering power density to satisfy the self-breeder condition. These effects are more dominant than the effect of the removal of fission products and the formed ^{233}U in the core after the year of cooldown period. This causes the higher volume ratio MSRs to drop sharply in power after the first cycle. The total energy produced after four cycles is showed in the last column of Table 3.5. MSR 2.8 has produced the highest amount of energy and MSR 2.1 the lowest. Due to the sharp drop in power density of MSR 2.8 and the rise in power density of MSR 2.1, the relative difference between these energies is much smaller than the relative difference between the power densities in the first cycle for these two cases.

Radiotoxicity of transuranic elements

Figure 3.7 shows the total radiotoxicity due to TRUs present in the used salt after every cycle, for the three considered self-breeder cases. First of all the shape of the plots is the same as for the case with a set power density of 5 MW/m^3, implying that the radiotoxicity is mainly caused by ^{238}Pu. Next it is shown that the three cases behave quite differently. Although MSR 2.8, with highest initial power density starts producing the highest dose per unit energy in the first cycle, it ends up producing the lowest dose per unit energy after four cycles. MSR 2.8 is therefore the cleanest reactor per unit energy over a timespan of 200 years. The total dose will be slightly higher though, as the self-breeder concerned has a higher total energy production over all the cycles (see the last column of Table 3.5). The higher and more thermal flux in the higher volume ratio cases combined with the higher power density in the salt will result in the TRUs building up faster, resulting in a higher radiotoxicity after the first cycle. In this way, an equilibrium concentration of TRUs is reached earlier than for the lower volume ratio cases.

Radiotoxicity of fission products

Radiotoxicity of the fission products is the same compared to the MSR with a set power density of 5 MW/m^3 studied above, as fission products are proportional to the produced energy and therefore always equal if normalized to produced energy. Only small variations are seen, due to the increasing presence of fissile isotopes other than ^{233}U, which have slightly different fission yields.

Temperature feedback

As seen with the case of a MSR with set power density the temperature feedback tends to decrease after every cycle, due to the same effect as discussed in Section 3.2.1. Further more it is clear from Table 3.5 that the feedback coefficients are more or less the same. This is because the volume ratios and channel diameters that are chosen here are the same as in [16], where they all were specifically chosen to operate with a temperature feedback coefficient of -1.5 pcm/K (at the end of cycle 1). Configurations with higher graphite amounts give higher temperature feedback, which was compensated with a better core homogenization, using smaller fuel channel diameters.

Graphite lifetime

As Table 3.5 shows the graphite lifetime increases for every consecutive cycle as reactor power is decreased. The higher thermal flux fraction of the better moderated cases has little effect on the graphite lifetime, as the increase in thermal flux comes mainly from a decrease in epithermal flux and less from a decrease in the neutron damaging fast neutron flux. The effect can be seen in cycle two, where MSR 2.8 has a significantly higher power density than MSR 2.1 but where the graphite lifetimes of the two cases are almost equal. The graphite lifetimes of the first cycles were already calculated in [16] and were known to lie above the reasonable 10 years, they stay above this value in the next cycles as shown in the table. In

the last cycles MSR 2.1 and MSR 2.4 do not even need graphite replacements if the blocks are shuffled once in a while to reduce the peak dose.

Chapter 4

Two Zone MSR

As discussed in Section 2.2.4, a MSR has different geometrical properties that can be modified. Channel diameter, graphite block size, core diameter, core height etc. In this chapter the MSR will be divided radially into two zones (see Figure 2.6). The outer dimensions of the reactor are kept constant. One zone will be optimized for fission of ^{233}U (the fissile zone). The other zone will be optimized for breeding ^{233}U (the fertile zone), i.e. neutron capture of ^{232}Th with as little neutron capture of ^{233}Pa as possible. As concluded by Nuttin et al. the fertile zone is best situated at the edge of the core if one desires to increase breeding gain. This makes sense as neutron flux will be higher in the fertile zone when placed at the center than when placed at the edges, because of the neutron leakage at the edges of the core. The zone with a higher volume ratio will be the fissile zone, as the neutron energy spectrum will be more thermal, leading to less neutron capture by thorium, allowing for a higher flux. The zone with a lower volume ratio will be the fertile zone. This zone will have a less thermalized neutron spectrum, leading to thorium capturing more neutrons, breeding more ^{233}U. The less thermalized neutron spectrum will decrease k_∞ in that zone, leading to a lower flux. The lower flux in the outer zone will cause less ^{233}Pa to capture a neutron before decaying, favoring breeding. The size of the zones and the volume ratio in both zones will be altered to find optimal results for the breeding gain.

The goal of this study is to design a core with high breeding capabilities. High breeding makes it possible to increase the power density as much as possible, while maintaining at least a self breeder reactor and staying within the constraints set in Section 2.5. For the one zone case, power density can be as high as 9.52 MW/m^3 ([16]). This is within the constraints as set in this study. The corresponding peak graphite lifetime is 13.1 years. The different volume ratios of the two zones are achieved by altering the channel sizes. Also the hexagonal block size can be altered. Previous studies [15],[16] have shown that altering volume ratio with constant channel diameter and altering channel diameter with constant volume ratio both have effects on breeding gain, temperature feedback and graphite lifetime of the 1-zone MSR. For the 2-zone MSR this will be translated in having different block sizes and different volume ratios, as block size needs to be consistent over both zones. The amount of salt in the

core (both inner and outer zone) is effected directly by the change in volume ratios, also the total amount of graphite in the core and the volume ratio of both zones is effected directly. These effects on their turn influence the temperature feedback coefficient and initial load of the core. Once in operation (with different power densities), not only changes in breeding gain will occur, but also in graphite lifetime and radiotoxicity of the salt.

In Table 4.1 the dimensions and the specifications of the reactor can be seen. The MSR is simulated with an inner zone volume ratio of 3-5 and an outer zone volume ratio of 0.75-3, based on the results of previous studies [18], [11], [16]. Simulation of the different MSR configurations is done for a cycle length of 50 years, as in the 1-zone case. Also the core will have the same outer dimensions as the 1-zone MSR. A hexagon block width (see Figure 2.5) of 20 cm is used. This makes it easy to shift the boundary between the fissile and the fertile zone. The reactor will have exactly 25 hexagon blocks on its diameter, no rounding is needed. Choosing different block sizes will give different results, as the heterogeneity is changed, even with the same volume ratio. The effect of this is studied and discussed later on, in section 4.0.8. The average power density of the reactor will be increased until an optimal core configuration is found that operates within the constraints set in Section 2.5 and is that breeds at least enough ^{233}U to be a self-breeder. Due to time it was not possible to perform all power density increases needed to find the optimal core configuration, as the calculation of one core configuration in operation for 50 years takes up to 2 weeks. This is due to the long time it takes for the 3D Monte Carlo calculations to complete. Standard deviation used in the Monte Carlo Calculation was $< 1 \times 10^{-3}$, which corresponds to a calculation time of 5-9 minutes for every calculation of k_{eff}. For the calculation of the temperature feedback coefficients, a more exact value of k_{eff} is needed. Here the Monte Carlo calculation was set to run for 500 minutes, resulting in a standard deviation of $< 1 \times 10^{-4}$.

Table 4.1: Specifications of the 2-zone MSR

Core diameter	500 cm
Reflector diameter	550 cm
Reactor height	500 cm
Zone boundary (radius)	170cm, 190cm, 210cm
Inner zone volume ratios (channel diameter)	3(10.5cm), 4(9.4cm), 5(8.6cm)
Outer zone volume ratios (channel diameter)	0.75(15.9cm), 1.5(13.3cm), 2.25(11.6cm), 3(10.5cm)
Hexagonal block width	20cm
Power density	7MW/m^3, 9MW/m^3, 11MW/m^3
Startup salt composition	LiF-BeF_2 (67%-33%) with 12 mol% $^{232}ThF_4$ and approx 0.15-0.3 mol% $^{233}UF_4$
Graphite density	1.84 g/cm^3
Fuel temperature	1000 K

All the results obtained from the parametric study can bee seen from pages 50-53. Table 4.2 shows the different MSR configurations with corresponding salt volumes and temperature feedback. Figures 4.1, 4.2, 4.3 show the BG, graphite lifetime and radiotoxicity of TRUs present in the salt after shutdown respectively, for the different configurations at the three different power densities. The following subsections will discuss the different results arising

from the different reactor configurations. First, properties that do not depend on power density are discussed, like temperature feedback and initial load of ^{233}U. Then an analysis is made of the aspects that depend on power density, namely breeding gain, graphite lifetime and radiotoxicity of the TRUs. As will become clear in this chapter, the different constraints on the core yield different ideal core configurations. The next step is then to find a configuration that gives the best overall results. This will be discussed in Chapter 5, the discussion and conclusion.

Table 4.2: Power density independent properties of the different MSR configurations. The two bold printed numbers indicate the best reactor configurations for the temperature feedback and initial load constraints. [1]The temperature feedback coefficient for this core configuration is weaker than the lower limit -1 pcm/K

Inner zone volume ratio	Outer zone volume ratio	Salt volume inner zone (m^3)	Salt volume outer zone (m^3)	Total salt volume (m^3)	Temperature feedback (pcm/K)	Initial load ^{233}U (kg)
Zone boundary at 170 cm						
3	0,75	11.0	31.5	42.5	**-2.9**	1949
	1.5	11.0	22.0	33.0	-2.8	1473
	2.25	11.0	16.9	27.9	-2.9	1222
	3	11.0	13.8	24.8	-2.7	1071
4	0,75	8.8	31.5	40.3	-1.8	1730
	1.5	8.8	22.0	30.8	-1.8	1298
	2.25	8.8	16.9	25.7	-1.6	1068
	3	8.8	13.8	22.6	-1.8	928
5	0,75	7.3	31.5	38.8	-1.3	1616
	1.5	7.3	22.0	28.3	-1.0	1202
	2.25	7.3	16.9	24.2	-1.2	975
	3	7.3	13.8	21.1	-1.2	840
Zone boundary at 190 cm						
3	0,75	13.6	25.5	39.1	-2.9	1754
	1.5	13.6	17.9	31.5	-2.7	1389
	2.25	13.6	13.7	27.3	-2.6	1189
	3	13.6	11.2	24.8	-2.5	1071
4	0,75	10.8	25.5	36.3	-1.4	1527
	*1.5	10.8	17.9	28.7	-1.8	1191
	2.25	10.8	13.7	24.5	-1.6	1004
	3	10.8	11.2	22	-1.5	898
5	0,75	9.0	25.5	34.5	-1.1	1406
	1.5	9.0	17.9	26.9	-1.1	1084
	2.25	9.0	13.7	22.7	-1.0	902
	3	9.0	11.2	20.2	-1.1	799
Zone boundary at 210 cm						
3	0,75	17.2	17.2	34.4	-2.5	1520
	1.5	17.2	12.1	29.3	-2.6	1281
	2.25	17.2	9.3	26.5	-2.8	1151
	3	17.2	7.5	24.7	-2.3	1067
4	0,75	13.8	17.2	31.0	-1.5	1280
	1.5	13.8	12.1	25.9	-1.6	1064
	2.25	13.8	9.3	23.1	-1.7	941
	3	13.8	7.5	21.3	-1.3	862
5	0,75	11.5	17.2	28.7	-0.8[1]	1149
	1.5	11.5	12.1	23.6	-0.9[1]	937
	2.25	11.5	9.3	20.8	-0.8[1]	822
	3	11.5	7.5	19	-0.7[1]	**746**

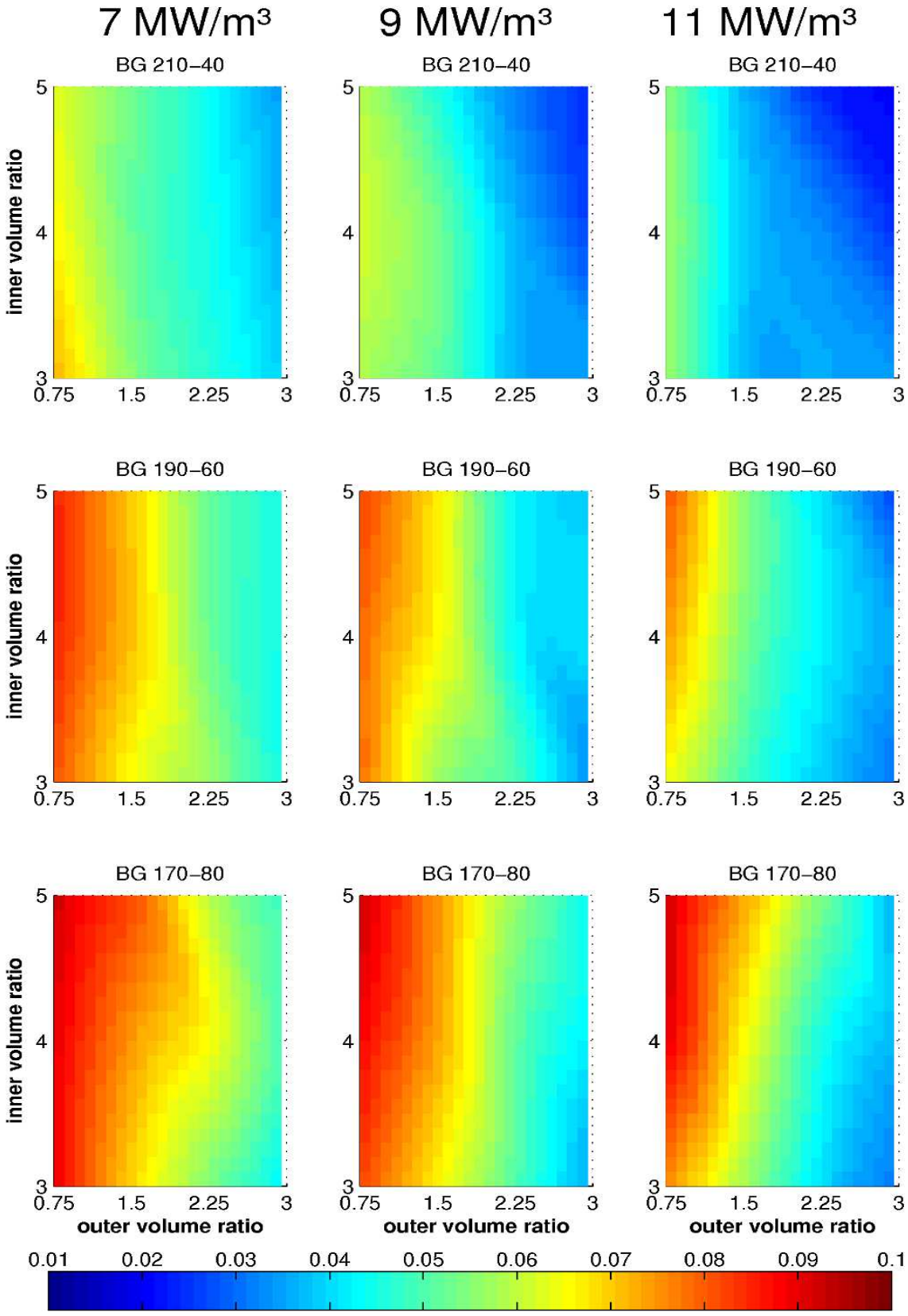

Figure 4.1: Average breeding gain during the 50 years of operation for all the different MSR configurations, operated at three different power densities (7, 9 and 11 MW/m^3). The numbers above each plot (170-80, 190-60, 210-40) indicate the zone boundary and the remaining outer zone thickness. Values between data points are interpolated. Exact values of the data points used can be found in Table A.2 in the Appendix. The color bar at the bottom shows the breeding gain for the corresponding colors.

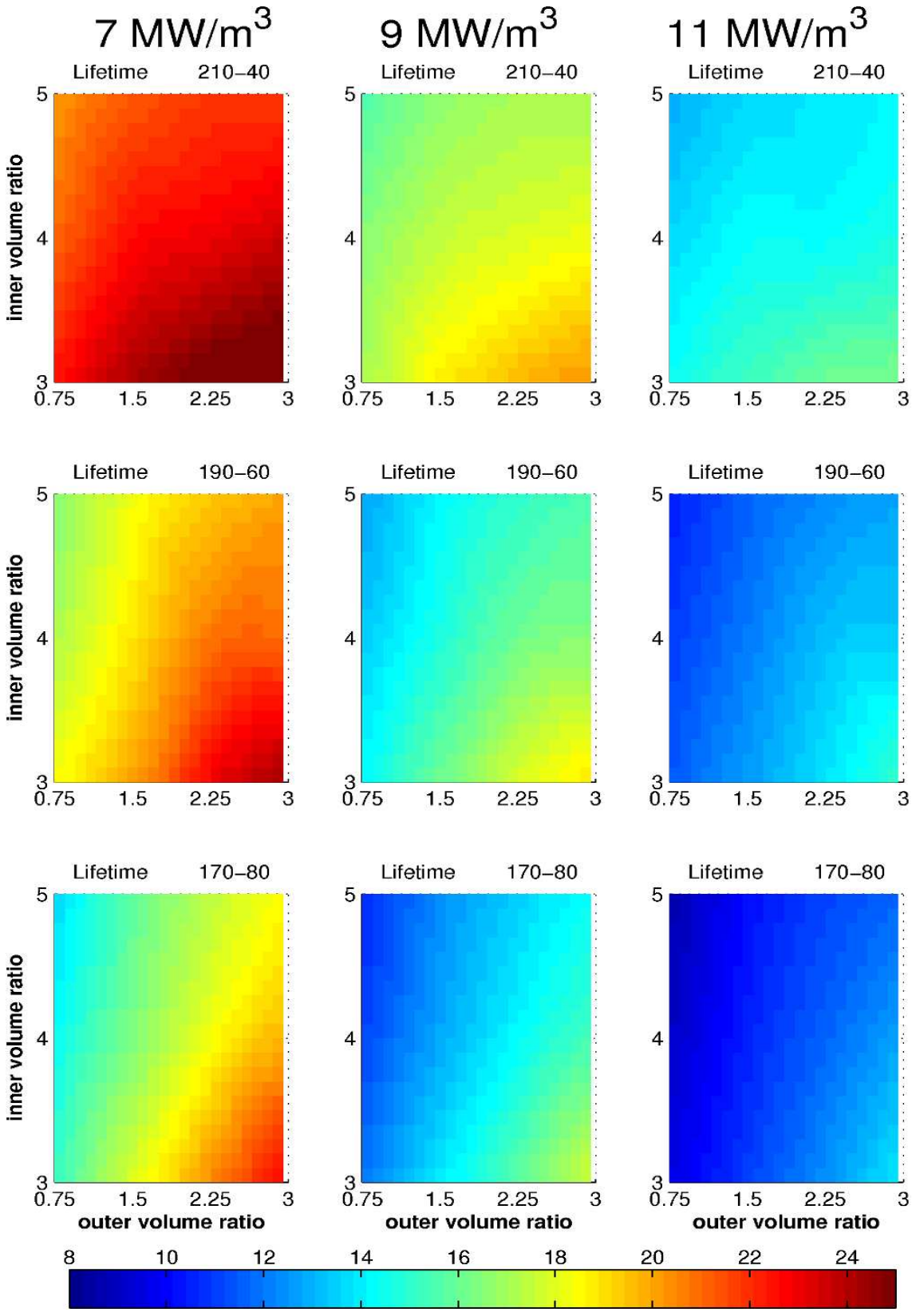

Figure 4.2: Graphite lifetime at the center of the core for all the different MSR configurations, operated at three different power densities (7, 9 and 11 MW/m^3). The numbers above each plot (170-80, 190-60, 210-40) indicate the zone boundary and the remaining outer zone thickness. Values between data points are interpolated. Exact values of the data points used can be found in Table A.2 in the Appendix. The color bar at the bottom shows the graphite lifetime for the corresponding colors.

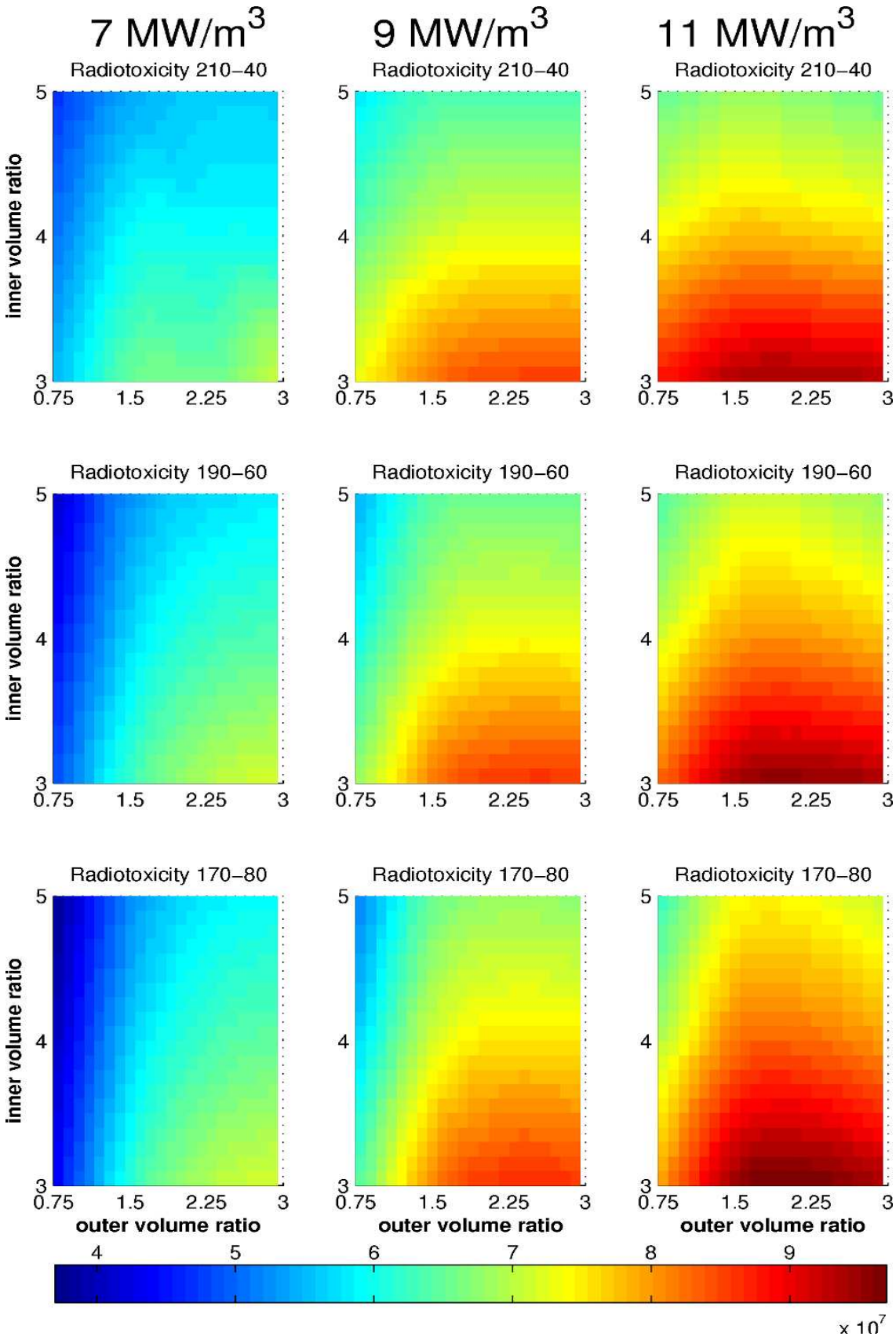

Figure 4.3: The total radiotoxicity of the TRUs present in the salt directly after 50 years of operation for al the different core compositions and power densities(7, 9 and 11 MW/m^3). The numbers above each plot (170-80, 190-60, 210-40) indicate the zone boundary and the remaining outer zone thickness. Values between data points are interpolated. The color bar at the bottom shows the radiotoxicity for the corresponding colors (in 10^7 Sv/GWthy).

4.0.3 Temperature feedback

Total temperature feedback coefficients can be seen in Table 4.2. All configurations yield feedback results within the constraints, except for the 210 cm zone boundary MSR with inner zone volume ratios of 5 (marked with [1] in the table). Temperature feedback coefficients are not calculated for the different power densities, as they are independent of the power of the reactor (although different power densities do have a different burnup, which over time will change the composition of the salt and therefore influence the temperature feedback). From the tables it is clear that the inner zone volume ratio has the biggest effect on temperature feedback.

The inner zone is the zone where most fissions take place, and will therefore have the largest effect on k_{eff}. A lower inner zone volume ratio results in a stronger (i.e. safer) temperature feedback coefficient than a higher volume ratio inner zone. Both feedback due to salt as well as graphite are increased, when the inner zone volume ratio is increased. Though the feedback due to the salt has a stronger effect to the increase. The positive feedback due to the density effects of the salt will be stronger for configurations with higher volume ratio. An increase in salt volume due to a temperature increase will have a larger effect, because the amount of salt is already smaller.

4.0.4 Initial load

The initial load of ^{233}U is affected by both the total salt volume as well as the concentration of ^{233}U needed to start the reactor with a critical salt composition. The high volume ratio, small salt volume cores need the smallest initial load to become critical.

The higher the overall volume ratio, the more thermal the overall neutron flux is, reducing the average capture cross-section of ^{232}Th, which increases the multiplication factor as the fission cross-section of ^{233}U is not strongly affected. A lower concentration of ^{233}U is then needed to start the reactor with a critical load. A larger the total salt volume means a larger initial load of all the salt components. These two effects enhance each other, as larger volume ratios imply smaller salt volumes. This results in the initial load differing more than a factor of 2.5 between the different core configurations. The smallest initial load is therefore found for the core with the highest overall volume ratio. However, this is not the safest core as the temperature feedback is (too) weak.

4.0.5 Breeding Gain

Figure 4.1 on page 51 shows the BG for the different MSR configurations operating with different power densities. As expected, the breeding gain increases with inner zone volume ratio and decreases with outer zone volume ratio. The larger the difference between inner and outer zone, the larger the BG is. Breeding gain is also larger for configurations with larger outer zone dimensions. Al power densities yield a positive breeding gain for each core

configuration.

The under moderated outer zone, has a harder spectrum, which in this range of volume ratios, leads to an increase of the average capture cross-section of ^{232}Th, while ^{233}U is not strongly affected. This effect lowers k_∞ of the outer zone, leading to a lower neutron flux in the outer zone as the power production shifts to the over moderated inner zone. This combination of high ^{232}Th capture cross-section and resulting low neutron flux in the outer zone, and the lower ^{232}Th capture cross-section and resulting higher neutron flux in the inner zone, makes the outer zone fertile and the inner zone fissile. Although the neutron flux is lower in the outer zone, the outer zone contains up to 4 times as much salt as the inner zone (see Table 4.2), leaving most of the thorium in the outer zone. Having a larger outer zone concentrates the power production to a even smaller inner zone, leaving a larger part of the core for breeding. As the power density increases, the BG drops due to the higher overall neutron flux in the core, letting ^{233}Pa capture more neutrons before decaying to ^{233}U, as the removal constant of the protactinium remains constant. Also the higher power density causes more neutron capturing actinides to be formed, which are not removed during reprocessing. To keep the core critical, more ^{233}U is fed into the salt. This also has a negative effect on breeding gain.

4.0.6 Graphite lifetime

Figure 4.2 on page 52 shows the graphite lifetime of the center of the core for the different MSR configurations. Most core configurations and power densities yield graphite lifetimes that are within the constraints set in section 2.5. Only the MSR with a zone boundary at 170 cm and a power density of 11 MW/m^3 corresponds to graphite lifetimes of less than 10 years when the outer zone has a volume ratio of 0.75.

Graphite lifetime is affected by both the inner and outer zone volume ratios. The higher the inner zone volume ratio the shorter the graphite lifetime as the more thermalized neutron flux will induce more fissions, increasing the fraction of the total power production in the inner zone. Also an inner zone with a higher volume ratio needs less ^{233}U to become critical, due to the lower capture cross-section of ^{232}Th. Then, a higher overall neutron flux is needed to produce the same power. Increasing the volume ratio of the outer zone will elongate the graphite lifetime as the power production shifts more to the outer zone as well. Increasing the overall average power density of the core decreases the graphite lifetime as overall flux is higher. On the other hand, a higher power density produces more neutron capturing actinides, reducing the multiplication factor. More ^{233}U is then fed into the salt and to keep the amount of fissions constant, overall flux is decreased with a positive effect on graphite lifetime. This effect is however much smaller than the negative effect due to the increase in power density. The smaller the radius of the zone boundary is the shorter the graphite lifetime, as the fissile zone is smaller, concentrating the majority of the power production in a smaller area.

If the power density of the core was raised again (e.g. 13 MW/^{3}), many more core configurations would not comply with the constraint of 10 years minimum graphite lifetime. Many high volume ratio inner zone and low volume ratio outer zone configurations lie already very close to the 10 year limit for the 11 MW/m^3 case.

4.0.7 Radiotoxicity

The total radiotoxicity of the TRUs present in the salt directly after the 50 years of operation for all the different core compositions and power densities can be seen in Figure 4.3. The dominant TRU is ^{238}U for all the compositions, giving the time dependent curve of the radiotoxicity the characteristic shape for MSRs. The initial radiotoxicity then gives a good measure for the radiotoxicity at later times. First of all the figure shows that the radiotoxicity of the used salt due to TRUs is increased with power density. This was also observed with the 1-zone core configuration at different power densities (for the first cycle). A higher power density results in a higher fluence over the total 50 years, increasing the amount of heavier TRUs that need several neutron capturing steps to be formed.

4.0.8 Effects of block width

Heterogeneity plays an important role in reactor design. 'Lumping' fuel together lets fast neutrons travel further through a moderator on average before encountering a fissile nuclide increasing the resonance escape probability. Also high energy fission neutrons in the fissile material travel further on average before encountering moderation, increasing fast fission factor ϵ. Changing the block size, whilst keeping the volume ratio constant, changes the heterogeneity of the core, smaller blocks will yield a more homogenized core, while larger blocks lead to a more heterogeneous core. One core configuration is chosen were the block width is altered. It is marked with a * in Table 4.2 and is chosen because its parameters lie in the middle of the studied values. The power density is chosen to be 11 MW/m^3. The block widths are varied according to Table 4.3. Due to the 'integer' nature of the blocks, the zone

Table 4.3: Results of 5 different block width configurations. Power density was 11 MW/m^3. Breeding gain is average over 50 years of reactor operation. Inner zone volume ratio is 4, outer zone volume ratio is 1.5. The zone boundary lies at approximately 190 cm from the center of the core.

Block width (cm)	Inner zone channel diameter (cm)	Outer zone channel diameter (cm)	Total salt volume (m^3)	Temperature feedback (pcm/K)	Initial load (kg)	BG	Graphite lifetime (years)
10	4.7	6.6	27.9	-3.0	1234	0.050	14.5
15	7.0	10.0	27.0	-2.4	1145	0.051	13.7
20	9.4	13.3	28.7	-1.8	1191	0.053	12.1
25	11.7	16.6	32.1	-1.2	1326	0.059	10.2
30	14.1	19.9	27.7	-0.7	1133	0.050	11.3

boundary will not always lie at exactly 190 cm from the core, as with the 20 cm block width. For that reason also the total salt volume in the core is printed in the table, as it will vary slightly. for the 12.5 cm block size case, the rounding of 190 cm to full blocks has the largest deviation. This results in a significantly smaller inner zone. Results such as temperature

feedback coefficient, initial load, breeding gain and graphite lifetime are also printed in the table.

When comparing these results with Table 4.2, larger block widths correspond to a higher inner zone moderation. Neutron spectrum is more thermalized for larger block widths. The 12.5 cm case shows the most thermal neutron flux in both inner and outer zones. As the outer zone is under moderated, the softer neutron flux has less effect on the capture cross-section of ^{232}Th than in the inner zone. The more thermalized neutron spectra correspond to a lower initial concentration of ^{233}U and a higher BG. On the other hand however, it shifts the power production more to the inner zone, leading to a shorter graphite lifetime and an increase of the temperature coefficient. Depending on the constraints a different block width could be chosen, this will be discussed in the next chapter.

Chapter 5

Discussion and conclusion

First an analysis of a 1-zone MSR has been made, comparing it with a PWR (the Borssele nuclear power plant) for radiotoxicity of TRUs present in the used salt or spent fuel rods. It was found that the MSR concerned (490 MWth), which operates on the thorium fuel cycle produces a different actinide composition, that favours a reduction in radiotoxicity, both in amount as well as lifetime. Directly after reactor shutdown the radiotoxicity per unit energy due to TRUs of a PWR is ten times more than the radiotoxicity of TRUs of a MSR (see figures 3.1 and 3.2). After a thousand years, the difference is a thousandfold. This is because the dominant TRUs, ^{238}Pu for a MSR and ^{239}Pu, ^{240}Pu and ^{241}Pu for a PWR, have different half-lifes, daughter nuclides and dose equivalent values. These all work in favour of the MSR. As for the fission products produced, the amounts do not differ much for both PWRs and MSRs, as fission yields of ^{233}U and ^{235}U are very similar. Taking this into account, makes MSRs much cleaner energy producers. Although 2 of the dominant TRUs (^{239}Pu, ^{241}Pu) of the PWR are fissile and could therefore be considered as reusable waste as they can be recycled into MOX for use in other reactors. Even when not considering these isotopes, radiotoxicity due to the energy production of a MSR is still much cleaner.

Next a simulation of a 1-zone MSR was run, where four 50 year cycles of reactor operation were simulated consecutively. In between cycles salt was cleaned from fission products (Z<90) and cooled down for a year before being reused. This was done for four different reactors: a 5 MW/m^3 which was also compared to a PWR and was a breeder after the first 50 years and 3 different self-breeders, with different volume ratios and power densities (see Table 3.5). For the self-breeders, the power density was adjusted after every cycle to obtain a BG of 0. It was found that for the low power density cases the breeding gain (or power density, in the case of the self-breeders) is increased after the first cycle as fission products are removed and the majority of the ^{233}Pa in the core has decayed to ^{233}U. This abundance of ^{233}U in the core leads to a period of 1.5 years during which no feeding of fissile material into the core was needed, improving breeding gain. For the two self-breeder cases that operate at higher power density initially, power density decreases in all the consecutive cycles. This is due to the higher power density leading to more neutron capturing actinides in the salt and a lower

concentration of ^{233}Pa.

In order to find a 2-zone MSR core configuration which can operate with the highest power density, whilst breeding enough ^{233}U during its lifetime to meet its demand in fissile material (i.e. a self-breeder) first constraints were set. Different safety and sustainability aspects were considered, as can be seen in Table 5.1. Different results yield different ideal core

Table 5.1: Different aspects of a MSR that were translated into constraints.

Aspect	Constraint
Durability of the graphite	Minimum graphite lifetime of 10 years
Passive safety	Maximal temperature feedback coefficient of -1pcm/K
Minimizing radiotoxic waste	Keeping the radiotoxicity due to TRUs as low as possible (no hard constraint)
Initial load	Choosing a 12 mole% $^{232}ThF_4$ concentration
Feasibility	A relatively slow chemical reprocessing scheme (see Table 2.4)

configurations for the range of volume ratios that was run. When comparing the results of the 2-zone core with those of the 1-zone core different advantages and disadvantages can be seen. First of all, the 2-zone core has a much higher breeding gain for all the configurations and power densities tested in this study. Graphite lifetime on the other hand is shortened, as a result of the power production that is shifted more towards the inner zone, leading to a higher neutron flux in that zone. As for radiotoxicity of TRUs, initial load and temperature feedback coefficients, performance compared to the 1-zone case of these factors is dependent of the configuration of the core and the power density that was used. Due to a different neutron spectrum, some 7MW/m^3 2-zone configurations have a cleaner burnup than the 1-zone 5MW/m^3 core.

Due to time it was not possible to increase power density to high enough levels to obtain self-breeder cores within the constraints. Choosing the best core configuration is then setting priorities among the different constraints set in Section 2.5. When looking at breeding gain, it depends largely on the volume ratio of the outer zone. Graphite lifetime on the other hand depends on both outer and inner zone volume ratios. Configurations with the highest breeding gain can operate at the highest power density without becoming a converter. However increasing power density has a negative effect on graphite lifetime, as neutron fluxes are higher. Graphite lifetime is then the first limiting factor if power density is increased beyond 11MW/m^3, as most configurations have strong enough temperature feedback coefficients. A self-breeder with highest power density in the range of configurations tested in this study will then probably be the configuration with the largest inner zone with lowest volume ratio, and smallest outer zone with lowest volume ratio. This is the core with a zone boundary at 210 cm, an inner zone volume ratio of 3 and an outer zone volume ratio of 0.75. Choosing a small outer zone benefits initial load of fissile material, graphite lifetime and to a smaller extent the radiotoxicity of TRUs, as results have shown.

However further study is needed to determine the ideal core configuration exactly. Only one

graphite hexagon block size was tested for all the different configurations. For one configuration the heterogeinity of the core is studied by changing this block size, while keeping the same volume ratio. Larger block sizes correspond to more moderated cores. However due to rounding, it was not possible to place the zone boundary at the same distance for all the different block sizes tested. Further research should be done to fully understand the influence of block sizes on the aspects studied in this thesis. This might yield other ideal core configurations. Further research can also be done by using separate fluids for inner and outer zones. In this way Pa that is formed in the outer zone will not be led through the high flux inner zone, possibly increasing breeding substantially. With two zones, serveral aspects that are not dealt with in the model that was used for this study could be of importance. Velocity of the molten salt in wide and narrow fuel channels will be different. Narrower channels will have a faster flow. This will have an effect on both the temperature in the different zones, as well as the fraction of delayed neutrons in the different zones. Also, in a 2-zone core as studied in this thesis the majority of the power production is concentrated in the inner zone. This will lead to a temperature difference between the zones, which might influence neutronics. The model used in this study uses a uniform temperature distribution and does not include flow of the salt. Short term reactor behaviour, like temperature feedback could be strongly influenced by these two aspects. Therefore further research could be done to incorperate these aspects in a model. However the model used in this study already needs a long calculation time.

When choosing the desired core configuration one could also think of a highest possible breeding MSR, giving two basic MSR types:

- A reactor with a high breeding gain, and a low initial load for the fast deployment of a reactor fleet.
- A reactor with the highest possible power density.

Both of course operating within material and safety constraints. At the early stage of MSRs the first option will be more desirable, as ^{233}U is still scarce and a large fleet should be deployed as fast as possible. This will lead to a different ideal core configuration in the range that is studied in this thesis. A small inner zone with high volume ratio and a medium volume ratio outer zone operating with low power density can be chosen to achieve high breeding with a relatively low fissile inventory. At a later stage, it will become more important to increase the power of the MSRs as much as possible, as the total fleet will not be growing as hard as in the beginning and to provide as much energy as possible with the lowest costs of operation.

Bibliography

[1] The Aircraft Reactor Experiment. *Nuclear Science and Engineering*, 2:797, 1957.

[2] Nuclear Weapons Archive. Operation teapot. http://nuclearweaponarchive.org/Usa/Tests/Teapot.html, 1997.

[3] World Nuclear Association. Energy analysis of power systems. http://www.world-nuclear.org/info/inf11.html, 2011.

[4] S. Delpech et al. Reactor physic and reprocessing scheme for innovative molten salt reactor system. *Journal of fluorine chemistry*, 130:11–17, 2008.

[5] James J. Dunderstadt and Louis J. Hamilton. *Nuclear Reactor Analysis*. Wiley, 1976.

[6] P.N. Haubenreich et al. Experience with the Molten Salt Reactor Experiment. *Nucl. App. and Tech.*, 8:118–136, 1970.

[7] Y. Honma et al. Optimization of flux distribution in a molten-salt reactor with a 2-region core for plutonium burning. 2008.

[8] B. Kelly et al. IAEA-TECDOC–1154 Irradiation Damage in Graphite due to Fast Neutrons in Fission and Fusion Systems, 2000.

[9] J.L. Kloosterman. NUC-RFA–96-14 Program BIFTOX, A Tool to Calculate the Radiotoxicity of a Nuclide Mixture, 1996.

[10] J.L. Kloosterman et al. ECN-RX–94-116 Transmutation of Americium in Thermal Nuclear Reactors, 1994.

[11] L. Mathieu et al. Thorium Molten Salt Reactor: from high breeding to simplified reprocessing. In *Proceedings of the International Conference Global*, 2003.

[12] L. Mathieu et al. The thorium molten salt reactor: Moving on from the MSBR. *Progress in Nuclear Energy*, 48:664–679, 2006.

[13] E Merle-Lucotte et al. Influence of the processing and salt composition on the thorium molten salt reactor. *Nuclear Technology*, 163:358–365, 2008.

[14] K. Mitachi and T. Yamamoto. Three-region core design for 200-MW(electric) molten-salt reactor with thorium-uranium fuel. *Nuclear Technology*, 158:348–357, 2006.

[15] K. Nagy et al. Definition of breeding gain for molten salt reactors. In *PHYSOR 2010 - Advances in Reactor Physics to Power Nuclear Renaissance*, 2010.

[16] K. Nagy et al. New Breeding Gain Definitions and their Application to the Optimization of a Molten Salt Reactor Design. *Annals of Nuclear Energy*, 38:601–609, 2011.

[17] OECD NEA. Uranium 2009, Resources, Production and Demand, 2009.

[18] A. Nuttin et al. Potential of thorium molten salt reactors: Detailed calculations and concept evolutions in view of a large nuclear energy production. *Progress in Nuclear Energy*, 46:77–99, 2005.

[19] R.C. Robertson et al. Conceptual design study of a single-fluid Molten Salt Breeder Reactor. *Oak Ridge National Laboratory*, 1970.

[20] K. Shibata et al. JENDL-4.0: A New Library for Nuclear Science and Engineering. In *to be submitted to J. Nucl. Sci. Technol.*, 2010.

[21] James E. Turner. *Atoms, Radiation, and Radiation Protection.* Wiley-VCH, 2004.

[22] ORNL/TM-2005/39 version 5. *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licencing Evaluations, vols I-III.* Oak Ridge National Laboratory, 2005.

Appendix A

Appendix

Table A.1: Weighting factor for the different organs in a human body, for the calculation of the committed effective dose [21].

Tissue or Organ	w_T
Gonads	0.20
Bone marrow	0.12
Colon	0.12
Lung	0.12
Stomach	0.12
Bladder	0.05
Breast	0.05
Liver	0.05
Esophagus	0.05
Thyroid	0.05
Skin	0.01
Bone surface	0.01
Remainder	0.05

Table A.2: Exact values of breeding gain and graphite lifetime for the different core configurations and power densities of the 2 zone MSR. The three values represent the result for 7/9/11 MW/m^3 respectively

hline Inner zone volume ratio	Outer zone volume ratio	Breeding Gain	Graphite Lifetime (y)
	Zone boundary at 170 cm		
3	0,75	0.087/0.081/0.075	15.3/12.0/9.6
	1.5	0.068/0.065/0.057	18.0/14.0/11.1
	2.25	0.054/0.049/0.040	20.5/15.9/12.5
	3	0.041/0.034/0.031	22.9/17.9/14.0
4	0.75	0.089/0.088/0.089	14.4/11.2/9.1
	1.5	0.074/0.073/0.064	16.5/12.9/10.6
	2.25	0.064/0.051/0.044	18.4/14.3/11.7
	3	0.048/0.041/0.035	19.9/15.5/12.8
5	0,75	0.092/0.091/0.091	13.7/10.7/8.7
	1.5	0.081/0.074/0.073	15.9/12.4/10.2
	2.25	0.058/0.056/0.053	17.4/13.5/11.1
	3	0.047/0.042/0.033	18.6/14.5/11.8
	Zone boundary at 190 cm		
3	0,75	0.080/0.077/0.065	18.4/14.3/11.5
	1.5	0.064/0.057/0.048	20.1/16.1/12.7
	2.25	0.055/0.048/0.038	22.9/17.9/13.9
	3	0.044/0.033/0.030	24.1/18.8/15.1
4	0.75	0.084/0.078/0.074	17.2/13.4/11.0
	1.5	0.070/0.065/0.053	19.0/14.8/12.1
	2.25	0.052/0.042/0.042	20.8/16.1/13.2
	3	0.044/0.040/0.034	21.0/16.4/13.4
5	0,75	0.084/0.082/0.080	16.5/12.8/10.5
	1.5	0.069/0.065/0.055	18.3/14.2/11.6
	2.25	0.052/0.042/0.039	19.3/15.0/12.3
	3	0.044/0.038/0.023	20.0/15.5/12.7
	Zone boundary at 210 cm		
3	0,75	0.071/0.058/0.056	22.5/17.1/14.4
	1.5	0.054/0.051/0.036	24.2/18.7/15.3
	2.25	-/0.036/0.036	-/19.6/16.0
	3	0.037/0.035/0.034	25.9/20.2/16.2
4	0.75	0.065/0.061/0.056	21.1/16.5/13.6
	1.5	0.051/0.052/0.038	22.6/17.5/14.3
	2.25	-/0.036/0.032	-/17.9/14.3
	3	-/0.025/0.024	-/18.2/14.7
5	0,75	0.063/0.058/0.054	20.2/15.6/12.9
	1.5	0.052/0.044/0.035	21.2/16.6/13.6
	2.25	0.043/0.031/0.023	21.8/17.0/13.9
	3	-/0.024/0.020	-/17.0/14.2