The Closed Nuclear Fuel Cycle for the Gas Cooled Fast Reactor

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Abstract

In this study the possibility of an integral fuel cycle has been examined for the GFR600. The GFR600 is a design by the CEA for the Gas Cooled Fast Reactor and this design has been adopted by the partners of The European FP6 GCFR-STREP (GCFR Specific Targeted REsearch Project). The fuel cycle used adds only depleted uranium and minor actinides during reprocessing. Another condition is that the value of $k_{\rm eff}$ will be kept between 1.00 and 1.05. The GFR600 has been investigated for a total operation time of 41.9 years, consisting of five periods of 1300 days burning and four periods of 2200 days fuel reprocessing. The nuclear fuel at the beginning consists of 79% depleted uranium, 16% plutonium and 5% minor actinides. The theoretical density of the fuel is either 85% or 90% and the reprocess loss is either one or two percent. The goal for this research is to find the parameters for which the GFR600 consumes the most minor actinides within the boundaries that has been chosen.

For all the settings under consideration it is possible to sustain the nuclear chain reaction. The amount of minor actinides consumed in the GFR600 is larger than the production of minor actinides in a PWR with an electric

The optimum setting is for a theoretical density of 85% and a reprocess loss η of two percent. This leads to a consumption of minor actinides of about 1541 kg. The amount of minor actinides used in the five periods is 2,853.7 kg which is equal to the amount of minor actinides of 106 annual discharges of the PWR. A bigger inlet of minor actinides does lead to a bigger reactivity swing, which has a negative influence on the safety and stability of the reactor.

Chapter 1

Introduction

More than 400 nuclear power plants are currently operating throughout the world and they provide about fifteen percent of the world's electricity supply. In Europe this is nearly 35%. Nuclear power plants perform safely and reliably and help meet the demand for diversity of the energy market and a reduced dependency on fossil fuel. Although the scientific world has not reached consensus about the relationship between global warming and carbon dioxide, agreements between governments around the world have been made to reduce the amount of carbon dioxide produced, e.g. the Kyoto Protocol. Nuclear plants have the advantage that, unlike fossil fuel plants, they do not produce any carbon dioxide, nitrous oxide or sulphuric components.

It is estimated that the world demand for electricity in the year 2030 will increase fifty percent in comparison to today's demand. In the year 2050 it is estimated to be doubled. Growing concerns for the impact of our society on the environment will make sustainability, low environmental impact and competitive economics for the energy market an interesting option. "If advances are made that fully apply the potential benefits of nuclear energy systems, the next generation of nuclear systems can provide a vital part of a long-term, diversified energy supply." [2]

This has led to a 'roadmap' for a new generation of nuclear power plants, the Generation IV. Generation IV reactors promise to improve sustainability, safety, and economics of the current nuclear reactors. The fast reactors that are included within the different Generation IV designs promise to drastically decrease the amount of waste produced by a nuclear reactor park.

1.1 Generation IV

It is common to divide nuclear power plants into generations. Currently most nuclear power plants belong to generation III or an equally safe (due to enhancements, modernisations and modifications) generation II. The reactors that are being made today are differentiating themselves from all the others on areas of safety. A revolutionary improvement has been set for the generation IV reactors. A few years ago an international committee selected six reactor types for further research. These reactors are: Gas Cooled Fast Reactor (GCFR), Very-High-Temperature Reactor (VHTR), Supercritical-Water-Cooled Reactor (SCWR), Sodium-Cooled Fast Reactor (SFR), Lead-Cooled Fast Reactor (LFR) and the Molten Salt Reactor (MSR). Many universities, research institutes and reactor manufacturers are working together to make these improvements possible.

The goals that has been set by the Generation IV International Forum for these reactors are the following [2]:

Sustainability Generation IV nuclear energy systems will provide sustainable generation that meets clean air objectives and promotes long-term availability of systems and effective fuel utilisation for world wide energy production.

Generation IV nuclear energy systems will minimise and manage their nuclear waste and notably reduce the long-term stewardship burden in the future, thereby improving protection of public health and the environment.

Safety and Reliability Generation IV nuclear energy systems will excel in safety and reliability.

Generation IV nuclear energy systems will have a very low likelihood and degree of core damage.

Generation IV nuclear energy systems will eliminate the need for offsite emergency response.

Economics Generation IV nuclear energy systems will have a clear lifecycle cost advantage over other energy sources.

Generation IV nuclear energy systems will have a level of financial risk comparable to other energy projects.

Proliferation Resistance and Physical Protection Generation IV nuclear energy systems will increase the assurance that they are very unattractive and least desirable route for diversion or theft of weapon-usable materials and provide increased physical protection against acts of terrorism.

1.2 GCFR

The number of neutrons emitted in the fission process per neutron absorbed for nuclides is higher in a fast neutron spectrum than in a thermal spectrum. This higher number of emitted neutrons means that more neutrons are available for conversion of 238 U to 239 Pu. This has created the the idea

that a breeder reactor operating on fast neutrons would use ²³⁸U more efficiently than one operating on thermal neutrons. From this idea came the first fast reactor, Clementine, built at Los Alamos in 1946. The next step was an experimental fast breeder reactor, EBR-I, designed by the Argonne National Laboratory. On 20 December 1951, EBR-I became the world's first nuclear plant of any type to generate electricity.

The original idea for the Gas Cooled Fast Reactor originated in the US in the 1960's at General Atomic. This first idea was followed by initiatives in Germany and Switzerland. Several gas coolants were considered, such as steam, carbon dioxide and helium. In Russia N_2O_4 was considered. Today's primary choice is Helium. The reason that helium Gas Cooled Fast Reactors are still interesting to research is because of the characteristics of the gas. One of the most interesting and promising characteristics of a Gas Cooled Fast Reactor is that such a reactor can sustain the nuclear reaction with a relative high percentage of minor actinides (MA). This could mean that the GCFR has a potential to close the fuel cycle. It is an interesting option to investigate, because minor actinides are highly radiotoxic isotopes with long half-life times. Therefore the amount of radiotoxic waste produced by the nuclear park would decrease by being used again.

1.3 Goals for This Research

In this research the closed fuel cycle for a GCFR is examined. The GCFR is used as a converter and the goal is to find the conditions that lead to the highest consumption of minor actinides. To find out how radiotoxic the waste from the GCFR is relative to the current reactors, it will be compared with a Generation III Pressurized Water Reactor (PWR).

Chapter 2

Theory

2.1 Nuclear Reactor Physics

2.1.1 Nuclear Reactions

Closing the fuel cycle to recycle minor actinides and uranium is possible because of breeding. It is necessary to produce more fissile material than there is fissile material destroyed. In order to achieve breeding, a fertile isotope must be converted via neutron capture into a fissile isotope. The degree of conversion that occurs in a reactor is denoted by the general term conversion ratio CR, which is denoted as

$$CR = \frac{\text{fissile material produced}}{\text{fissile material destroyed}} = \frac{FP}{FD}$$
(2.1)

A reactor is called a breeder if the conversion ratio is greater than unity. If the conversion ratio is greater than unity, he name of the equation changes into the breeding ratio.

The microscopic cross sections (σ) represents the probability of a neutronnucleus reaction. If for example a uniform beam of neutrons with intensity I in cm⁻² s⁻¹ strikes a thin 'film' of atoms (one atomic layer thick) with N atoms/cm². Then the number of interactions C per cm² will be proportional with the atom density N and the intensity I. Defined as the proportionality factor as the (microscopic) cross section.

$$C = \sigma N I \tag{2.2}$$

When a nucleus absorbs a neutron, the nucleus will either transform into a heavier nucleus or fission. This is expressed in the formula (2.3). The chance of absorption of a neutron by a nucleus is equal to the sum of the chance of a nucleus to fission and to capture.

$$\sigma_a = \sigma_f + \sigma_c \tag{2.3}$$

From this theory the reproduction factor can be constructed. The reproduction factor is the number of neutrons produced per fission (ν) times the chance of fission occurring when a neutron has been absorbed. The reproduction factor can be written as

$$\eta = \frac{\nu \sigma_f}{\sigma_f + \sigma_c} = \frac{\nu}{1 + \sigma_c / \sigma_f} = \frac{\nu}{1 + \alpha}$$
(2.4)

With α is defined as

$$\alpha = \frac{\sigma_c}{\sigma_f} \tag{2.5}$$

This reproduction factor is important, because the neutron yield varies quite heavily at different energies and for different isotopes, see figure 2.1 The number of neutrons produced per fission is fairly constant for neutron energies up to about 1 MeV and slowly rises at higher energy. The most influential is α , because it varies considerably with energy and between isotopes. It is this higher reproduction factor at higher energies that is important in the fast reactors in general.



Figure 2.1: Reproduction factor varying over energy range

2.1.2 Neutron Transport

Studying the distribution of neutrons in a reactor is an important part of reactor physics, because the free neutrons are essential in sustaining the nuclear reaction. The mathematical description of the neutron distribution is based on a neutron balance equation, which is called the neutron transport equation.

$$\frac{1}{v} \frac{\partial \phi(\underline{r}, \underline{E}, \underline{\Omega}, t)}{\partial t} = S(\underline{r}, \underline{E}, \underline{\Omega}, t) \\
+ \int_{0}^{\infty} \int_{4\pi} \Sigma_{s}(\underline{r}, \underline{E}' \to \underline{E}, \underline{\Omega}' \to \underline{\Omega}) \phi(\underline{r}, \underline{E}', \underline{\Omega}', t) d\underline{E}' d\Omega' \\
- \underline{\Omega} \cdot \underline{\nabla} \phi(\underline{r}, \underline{E}, \underline{\Omega}, t) - \Sigma_{t}(\underline{r}, \underline{E}) \phi(\underline{r}, \underline{E}, \underline{\Omega}, t) \quad (2.6)$$

- $\frac{1}{v} \frac{\partial \phi(\underline{r}, \underline{E}, \underline{\Omega}, t)}{\partial t}$ This is derived from the change in the number of neutrons in time ($\phi = nv$ the flux density equals the neutron density times the velocity)
- $S(\underline{r}, \underline{E}, \underline{\Omega}, t)$ This is the source term. Increase of neutrons by the presence of neutron sources, among which fission.
- $\int_0^\infty \int_{4\pi} \Sigma_s(\underline{r}, E' \to E, \underline{\Omega'} \to \underline{\Omega}) \phi(\underline{r}, E', \underline{\Omega'}, t) dE' d\Omega'$ Increase by scattering of neutrons with other energies and directions to the energy range dE and solid angle d Ω .
- $\underline{\Omega} \cdot \underline{\nabla} \phi(\underline{r}, E, \underline{\Omega}, t)$ Term to describe the net outflow of neutrons

 $\Sigma_t(\underline{r}, E)\phi(\underline{r}, E, \underline{\Omega}, t)$ Decrease by neutrons undergoing an interaction

In the case of mono-energetic neutrons, the energy-dependence disappears and the neutron transport equation can be written with some calculations to the (one-group) diffusion equation:

$$\frac{1}{v}\frac{\partial\phi(r,t)}{\partial t} = \underline{\nabla}\cdot D(\underline{r})\underline{\nabla}\phi(\underline{r},t) - \Sigma_a(\underline{r})\phi(\underline{r},t) + S(\underline{r},t)$$
(2.7)

A special form of the diffusion equation is the one-group diffusion equation with the source as a result of fissions, which looks like

$$\frac{1}{v}\frac{\partial\phi}{\partial t} = \underline{\nabla} \cdot D(\underline{r})\underline{\nabla}\phi(\underline{r},t) + v\Sigma_f(\underline{r})\phi(\underline{r},t) - \Sigma_a(\underline{r})\phi(\underline{r},t)$$
(2.8)

Only when the production and the losses exactly balance will the neutron flux be constant. For this situation the time derivative can be set zero. To also have a measure for the production, absorption and leakage of neutrons from the reactor for a non-critical reactor, one can artificially adjust the source term in (2.8). Mathematically this yields an eigenvalue equation with eigenvalue k.

$$k = \frac{\text{total neutron production rate by fissions}}{\text{total neutron loss rate by leakage and absorption}}$$
(2.9)

This factor is called the effective multiplication factor k_{eff} . For more information on these subjects, see 'Nuclear Reactor Physics, lecture notes AP3341' [3].

2.2 Fuel Cycle Physics

2.2.1 Reprocessing

One of the most important reasons for reprocessing fuel is to recover unused uranium and plutonium in the spent fuel elements. Reprocessing to recover uranium and plutonium avoids the wastage of a valuable resource because most of the used fuel (uranium at less than one percent 235 U and a little plutonium) can be recycled as fresh fuel, saving some thirty percent of the natural uranium otherwise required. A secondary reason is to reduce the volume of high-level waste to be disposed of. A positive side effect is that the level of radiotoxicity of fission products in the disposed fuel falls more rapidly after about hundred years than the original used fuel itself. It is also from that point of view better to reprocess the fuel. In recent years interest has grown in separating or partitioning individual radionuclides in order to reduce long-term radiotoxicity in the waste and to change, mostly by fission, the long-lived nuclei into shorter-lived ones. There are several ways for a fuel cycle, but the most important difference is the reprocessing part obviously.

- **OTTO** Technically this is not a cycle per se. OTTO stands for Once Through Then Out meaning that once the nuclear core has been used it is not being used again. After the core has been used it is to be stored. This cycle OTTO is preferred in the US because of the proliferation risk of the other options.
- **PUREX / COEX** Reprocessing is a way of fuel cycle that consists of recycling the uranium and plutonium by separating them from the other spent fuel. All commercial reprocessing plants use the well-proven hydrometallurgical PUREX (Plutonium URanium EXtraction) process. This involves dissolving the fuel elements in concentrated nitric acid. Chemical separation of uranium and plutonium is then undertaken by solvent extraction steps. The Pu and U can be returned to the input side of the fuel cycle; the uranium to the conversion plant prior to re-enrichment and the plutonium straight to fuel fabrication. Alternatively, some small amount of recovered uranium can be left with the plutonium which is sent to the MOX plant. This is known as COEX process. For a schematic view of the nuclear fuel cycle with reprocessing see figure 2.2.

In either case, the remaining liquid after Pu and U are removed is highlevel waste, containing about three procent of the used fuel in the form of fission products and minor actinides (Np, Am, Cm). It is highly radioactive and continues to generate a lot of heat. It is conditioned by calcining and incorporation of the dry material into borosilicate glass, then stored pending disposal. In principle any compact, stable, insoluble solid is satisfactory for disposal. After reprocessing, the recovered uranium may be handled in a fuel fabrication plant (after re-enrichment), but the plutonium must be recycled via a dedicated mixed oxide (MOX) fuel fabrication plant.

- **PUREX 2** Another version of PUREX has the minor actinides (americium, neptunium, curium) being separated in a second aqueous stage and then directed to an accelerator-driven system cycling with pyroprocessing for transmutation. The waste stream then contains mainly fission products.
- **Other options** There are some more options, but they are all in early stage of development and on laboratory scale. These include reprocessing methods such as transmutation, pyro-processing and DUPIC.



Figure 2.2: Schematic drawing of the nuclear fuel cycle with PUREX [5]

2.2.2 Closed Fuel Cycle

Closed fuel cycle is a fuel cycle that besides the more usual recycling of plutonium and uranium other nuclei are being retrieved, for example curium or americium. The Gas Cooled Fast Reactor under consideration has the ability to use fuel on which other nuclear power plants would not work. The closed fuel cycle in combination with a reactor is called the integral cycle. For an overview see 2.3.



Figure 2.3: Overview of integral cycle

Chapter 3

Reactor Model

This chapter describes the used GCFR model and the programs to calculate the required data. First of all the chosen GCFR is described. After that section the specifications used in this research are described and as a last section the used and written programs that model the physics of the chosen GCFR are explained.

3.1 CEA GFR600 GCFR

The European FP6 GCFR-STREP (GCFR Specific Targeted REsearch Project) [8] has started in March 2005 and concerns the research and development of Gas Cooled Fast Reactors. The partners agreed to adopt the CEA GFR600 GCFR design as the reference. This design is also the design used in this research. Stated in the table 3.1 are some of the specifications of the nuclear reactor.

Power (MWth)	600
Coolant	He
Power density (MW/m^3)	103
Specific power (W/gHM)	45
$T_{core,in}$ (^o C)	480
$T_{core,out}$ (^o C)	850
Core H/D (m/m)	1.95/1.95
Pressure (MPa)	7.0
Ratio coolant/	
structures /fuel	55/10/35

Table 3.1: GFR600 core parameter [9]

3.1.1 Coolant

The CEA GFR600 is helium cooled. The use of helium cooling has several advantages:

- **Chemically inert and transparent** Helium gas is not flammable, nor corrosive. This means that concerning maintenance no damage will be done on the materials inside, which are in contact with the gas. Also helium gas is transparent making it easier to see possible damages to the system.
- Neutronically inert Helium gas has a low absorption cross section, which is good for breeding. The helium has low moderation, which results in a harder spectrum. A harder spectrum is beneficial for breeding and for using minor actinides as fuel. Helium also has a small coolant reactivity effect (temperature and voiding). A direct cycle is possible, because helium is a non-radioactive coolant.
- **Single phase** With helium gas as coolant there is no possible flashing or abrupt change of properties, because it. The high outlet temperatures that give an high efficiency.

Helium gas also has some disadvantages, which are also important to take into account. High neutron leakage, lower Doppler coefficient (due to hard spectrum), larger fissile mass, and poor heat transfer properties. High pressure and/or surface roughening are required to improve heat transfer (high pumping power). This is unwanted because high pressure worsens the effects of a depressurisation accident.

3.1.2 Fuel

The fuel itself has a plate configuration, which is designed by the CEA [4], called the CERCER plate SA. The selected fuel concept is based on dispersed fuel, a CERamic actinide compound in an inert CERamic matrix, CERCER. It consists on a hexagonal tube containing the plate bundle; there are three CERCER plate sub-bundle, with a form of regular rhombus (30 degrees of inclination), arranged in order to obtain an hexagon see figure 3.1. There are two cores presented based on these plates CERCER, the one used in this model is the "efficient one" with CERCER (U,Pu)C-SiC30% vol. The fuel is a matrix fuel, with 70% UPuC and 30% SiC by volume.

Another important aspect of the fuel is its theoretical density. The nuclides in the fuel form a lattice. This lattice is not optimal and has a certain porosity. The theoretical density is one minus the porosity fraction.



Figure 3.1: Subassembly of the 600 MWth plate SA with efficient CERCER [4]

Theoretical density (%)	Reprocessing loss $(\%)$
85	1
	2
90	1
	2

Table 3.2: Settings for GFR600

3.2 Specifications of Model

In order to bound this research on the GCFR, some limitations and boundary conditions has been used. Because the GCFR is meant to be used as a converter the average k_{eff} is chosen to only vary between the values 1.00 and 1.05. Two different densities for the fuel has been chosen and for each of these densities two options for the reprocessing. This creates four different closed fuel cycles for the GCFR.

The fuel is burned for a period of 1300 days. After the burnup the fission products are removed from the fuel at reprocessing. Reprocessing is a chemical process and can not be done without some losses, so either one or two percent of the heavy metals are also leaving the integral cycle. This percentage of loss of fuel has been given the variable η , not to be confused

Isotope	Percentage
U-235	0.25
U-238	99.75

Table 3.3: Depleted uranium vector

with the reproduction factor. The losses will be completed with a varying composition, consisting of minor actinides and/or depleted uranium. See figure 3.2



Figure 3.2: Specific overview of reactor model used

3.2.1 Fuel

The composition of the core consists of silicium cladding and fuelcarbide. The fuelcarbide for the first cycle consists of a mixture of uraniumcarbide, plutoniumcarbide and a small percentage of minor actinides (MA). The fuel carbide is 70% and the the sicilicium carbide of 30%. The fuel that is being used for the first cycle consists of 79% uranium, 16% plutonium and 5% minor actinides. The uranium used for the fuel is depleted uranium, which is the uranium left after the enrichment of natural uranium. It is a mixture of two uranium isotopes, ²³⁵U and ²³⁸U, see table 3.3. The minor actinides vector used for the fuel and for reprocessing is given as standard by the CEA. It originates from LWRs and other reactors in France and are obtained by reprocessing the fuel cores of the reactors. The specific content of the minor actinides can be seen in the table 3.4. Also the plutonium used in the fuel consists of several plutonium isotopes, see table 3.5.

Isotope	Percentage
Np-237	16.86
Am-241	60.62
Am-242m	0.24
Am-243	15.70
Cm-242	0.02
Cm-243	0.06
Cm-244	5.14
Cm-245	1.26
Cm-246	0.10

Table 3.4: Minor actinides vector

Isotope	Percentage
Pu-238	2.7
Pu-239	56.0
Pu-240	25.9
Pu-241	7.4
Pu-242	7.3
Am-241	0.7

 Table 3.5: Plutonium vector

3.2.2 Reprocessing

After 1300 days the reactor is shut down and the fuel is taken out of the reactor to be reprocessed. The fuel first needs to cool down, as the nuclides in the fuel are still producing a lot of heat. After the fuel has cooled down it is being reprocessed. After 2200 days the fuel is being placed in the GCFR to begin a new cycle.

The burnup of the fuel is about five percent of the total fuel for 1300 days burning, so about five percent of the fuel is fissioned. When the fuel is reprocessed these fission products are separated from the actinides. Not all actinides are separated from the fission products. This results in a loss of fuel η . This loss has been chosen to be one or two percent of the actinides left in the fuel after the burnup.

To create the same amount of material as at the beginning of the cycle, the actinides are complemented with either depleted uranium or minor actinides, the same vectors that has been used to construct the fuel (tables 3.4 or 3.3).

3.2.3 Radiotoxicity

There are two products which leave the cycle with fuel reprocessing: shortlived fission products and the long-lived heavy metals. Only the heavy metals are taken into account. An amount of the same nuclides will decay and produce daughters, which in their turn could also decay. This results that for an amount of the same nuclides will have a different radiotoxicity level at different times and therefore every nuclide needs to be calculated with its own radiotoxicity data.

The data used to do these calculations come from a ECN report written by dr. ir. J.L. Kloosterman [7]. In this report the radiotoxicity of many isotopes are stated in Sv/mole for ten to one million years. Not for all isotopes is radiotoxicity data available.

To compare the found radiotoxicity results with a generation III thermal reactor, the net production of radiotoxicity is taken per energy produced in the reactor. So for our GFR600 model the energy produced is 275 MW electric and it is assumed that the total core is discharged after the five cycles. The material that need to be stored are four times a small loss at reprocessing and the whole core after the fifth cycle. For the composition of the core after the fifth cycle, the composition of the loss vector after the fourth cycle and 2200 days of decay is taken, corrected to account for the whole fuel. To take the net production, the total amount of minor actinides added and plutonium used is subtracted from the total waste produced. A reactor that is burning for hundred percent of the time, that means five times 1300 days times 24 hours, gives $1.56 \ 10^5$ hours, which leaves the amount of electric energy produced at 42.9 Tera Watt-hour. With this energy, the net radiotoxicity of the waste can be calculated per energy produced (in Sv/TW-h) and is compared with a Generation III thermal reactor.

Only the total amount of material that has left the integral cycle after the fifth cycle is used to analyse the radiotoxicity. This approach leads to a small error in the toxicity calculations, because the time difference between the first reprocessing loss and the last reprocessing loss is about 38 years and during that time the a small part of the isotopes would have decayed. But because this is only for the first 38 years this only contributes to the first hundred years and is not important after thousand years. For the radioactivity of the core after the fifth cycle it is assumed that the composition is almost the same as after decay of loss after the fourth cycle.

The chosen generation III reactor is a Presurized Water Reactor (PWR), with a core mass of 101,000 kg, a thermal power of 3230 MW and an efficiency of 33%. Each 12 months 30,400 kg is discharged, about 1/3 of the core. For the calculation of the radiotoxicity a calculation done by Dr. Ir. J.L. Kloosterman has been used. I would like to refer for the data to appendix B

3.3 **Program Description**

For this research a program written by W.F.G. van Rooijen has been used. This Perl script, called Burn1d, uses Scale 4.4 modules to calculate average cross-sections in the unit cells and the reactor parameters of the GFR600. For the calculations of the change in nuclide densities during burnup and decay the depletion code ORIGEN-S has been used. Burn1d uses nuclide densities of the original fuel to calculate the nuclide densities and a value for the k_{eff} at specified times during the burnup of the GFR600. For more information on this subject, I would like to refer to the thesis of Dr. Ir. W.F.G. van Rooijen, 'Improving fuel cycle design and safety characteristics of a Gas Cooled Fast Reactor' [9].

There are two steps in calculating the properties of the GFR600 during the integral cycle. A cycle starts with a burnup and ends with addition of material to the fuel. First part is to put the densities for the original composition of the fuel in the burn1d script. The burn1d script gives as output the nuclide densities and k_{eff} at specified times during the 1300 days of burning. The vector of nuclide densities at 1300 days is taken and used to calculate the composition of the fuel after a decay of 2200 days with Scale 4.4.

For the second and last part, the burn1d script has been altered to calculate the k_{eff} as a function of the input needed after reprocessing the nuclear fuel. The program now reads two vectors with the nuclide densities: one at the beginning of cycle 1 and one at the end of decay for the same cycle. Because with reprocessing only the actinides are retrieved to be used again as fuel, the nuclides of 90 or higher are taken from these vector. This only affects the vector at the end of decay, because this vector consists of fission products. Because the fission products are ignored, there is a difference in densities between the first and the second vector. This loss of material is complemented to maintain the same amount of material for the fuel. The percentage minor actinides is guessed and the rest of the material is completed with depleted uranium. The burn1d then calculates the densities and the k_{eff} at the specified times. The time average of k_{eff} is compared with the boundary given for k_{eff} . If the time average k_{eff} is higher than the upper boundary than the percentage of MA is decreased. If the average k_{eff} is lower than 1.045 than the percentage MA is increased. Between 1.045 and 1.05 for an average of $k_{\rm eff}$ the right value for minor actinides is found. When the right value for the average k_{eff} is found, the densities at the end of burnup for cycle 2 will decay for 2200 days. Then the script starts over again till all four cycles are found.

Chapter 4

Results & Discussion

This chapter is divided in sections of the different parameters. These sections are divided in subsections by the different calculated parts of the integral cycle. First the results for k_{eff} are shown and discussed in the subsection 'Calculation of k_{eff} '. In the subsection 'Additions to the Fuel' results are shown on the addition of minor actinides and depleted uranium. In the last subsection the waste stream is shown and the radiotoxicity of the waste is shown. In the last section of this chapter, the results on the radiotoxicity for the GFR600 are compared to the PWR.

4.1 85 Percent Theoretical Density and One Percent Loss

4.1.1 Calculation of k_{eff}

The first graph 4.1 shows the k_{eff} of the reactor fuel for each cycle over time. The first cycle shows k_{eff} for the burnup of the original fuel with theoretical density of 85%. The k_{eff} of the first cycle is much lower than the k_{eff} of the second cycle. The k_{eff} of the second cycle is lower than the k_{eff} of the third cycle. The k_{eff} of the third, fourth and fifth cycle are close together.

The k_{eff} over time of the first cycle differs from the other cycles. The k_{eff} of the first cycle has a declining slope, while the k_{eff} of the other cycles are rising. If the leakage of neutrons is constant over time, it follows from formula (2.9), that there are three options for the second to the fifth cycle. The absorbers are decreasing, the fissile material is increasing or both effects are occurring. What happens is that minor actinides are first absorbers and can become fissile.

The small dip in k_{eff} at the very beginning of each cycle is a result of an increase of absorbers, mainly short-lived fission products like ²³⁵Xe. For the last two cycles figure 4.1 shows that lines are declining, meaning that the amount of fissile material in comparison to the absorbers is decreasing.



Figure 4.1: Change of $k_{\rm eff}$ over time while burning of every cycle for 85% TD and 1% loss

The figure 4.2 shows the time average of $k_{\rm eff}$ over one cycle. Interesting thing to notice is that the boundary for $k_{\rm eff}$ is reached at the third cycle, the first two cycles are beneath the chosen boundary. The upper boundary is approached very precisely for the third to the fifth cycle.



Figure 4.2: Time average $k_{\rm eff}$ of every cycle for 85% TD and 1% loss

4.1.2 Additions to the Fuel

In this subsection the addition of minor actinides or depleted uranium in kilograms to the fresh fuel is shown for each cycle. The total addition per cycle is about 950 kg. This varies a little per cycle, about 10 kg. This difference arises because the loss of material after a burnup is known in a number of nuclides and this number is being complemented. Due to the difference in mass/mole of the two different vectors, the amount in kilograms varies. The amount added is a result of a loss of fission products of about five percent of the total nuclear fuel and one percent loss of the remaining 95 procent from reprocessing.

For the first figure, figure 4.3, the total added material is 3,816.5 kg, the total added minor actinides is 1,631.6 kg, which leaves the total added depleted uranium on 2,184.9 kg. For the first cycle only minor actinides are added, this is because the the boundary is not reached for k_{eff} .

If the additions are compared to the results for $k_{\rm eff}$ (figure 4.1), it can be concluded that an addition of minor actinides leads to a increase of $k_{\rm eff}$. This effect is not linear as can be seen for the cycle number five. The addition of minor actinides, which is more than for cycle four and about half of cycle three, does not lead to the same increase of $k_{\rm eff}$. The effects of repeated addition of minor actinides leads to a smaller increase of $k_{\rm eff}$.



Figure 4.3: The addition of mass in kg for a theoretical density of 85% and a loss of 1%

Number	Isotope	Number	Isotope
1	U-238	9	Cm-244
2	Pu-239	10	Pu-241
3	Pu-240	11	U-234
4	Am-241	12	Am-242m
5	Pu-238	13	U-235
6	Am-243	14	Cm-245
7	Pu-242	15	U-236
8	Np-237		

Table 4.1: The numbering of the different isotopes, which are used in the barplots of the reprocessing losses

4.1.3 Waste Stream Analysis

Out of all isotopes for which information exists, only 15 are chosen to be plotted. The chosen isotopes are listed in table 4.1. Each bar stands for each isotope in the table and is divided into four parts. Each part is the fraction of loss of material for that nuclide. So all parts with the same colour for all the nuclides add up to 100%. This is done for all the four cycles with reprocessing. Three plots of the same results are shown with different scales. In the appendix A tables for each parameter is included with the amounts of loss in kilogram per isotope per reprocessing cycle and their totals.

In the next three figures 4.4, 4.5 and 4.6, the irretrievable nuclides from the reprocessing that exit the integral cycle are shown. The four losses, as a result of four fuel reprocessing, add up to 400% with each loss per reprocess normalised at 100%. Therefore it is visible how the quantity of an isotope changes for different cycles. Because the loss vector is taken to be 1% for every heavy metal isotope that is present in the fuel after burnup, also something about the content of the nuclear fuel at the end of the cycle is known.



Figure 4.4: The losses per nuclide for storage are shown for a theoretical density 85% and reproceeding loss of 1%. The full y-axis is shown.



Figure 4.5: The losses per nuclide for storage are shown for a theoretical density 85% and reproceesing loss of 1%. The scale of the y-axis stops at 40%

Something that is very obvious is that 238 U has the biggest quantity present in the fuel. As can be noticed immediately, the total of 238 U at the end of cycle four is less than the beginning condition. This was 79%, while the figure shows an percentage of around 75%.

In the figure 4.5 and figure 4.6 it can be seen that some isotopes are increasing in the waste stream and others are decreasing. Some of the more radiotoxic isotopes are quite large. The most radiotoxic isotope is 244 Cm



Figure 4.6: The losses per nuclide for storage are shown for a theoretical density 85% and reproceesing loss of 1%. The scale of the y-axis stops at 2.5%

(number 9 in table 4.1). This isotope stays the most radiotoxic for the first 100 years, after the first 100 years ²⁴¹Am becomes the most radiotoxic. Hundred years is not much compared to the lifespan of a typical heavy metal isotope, so the radiotoxicity decreases fast.

The second most radiotoxic isotope is 241 Am. This isotope is added with the addition of MA-vector, defined in table 3.4. The isotope 241 Am in the first cycle starts with a 3% content in the mixture of the nuclear fuel. After the first burn-up that percentage is only 2,6%, indicating that 241 Am is being destroyed faster than created. This is something that can be seen with some simple calculations. However, because 241 Am is added to the fuel (for the first cycle that is around 570 kg, corresponding to 3.5%) the amount of 241 Am is growing in the reprocessing losses.

After the ²⁴¹Am the plutonium isotopes have the biggest influence on the radiotoxicity of the waste. In the order of biggest to smallest Sv/mole during time ²³⁸Pu, ²⁴⁰Pu and ²⁴¹Pu. In contrast with the americium isotope, plutonium, although a big influence in radiotoxicity, is a vital element for the nuclear reaction. Therefore, it is not desirable to decrease this amount.

An estimation for the amount of minor actinides consumed can be made with the data of the waste stream in the appendix A. The amount of minor actinides at the end of burnup can be calculated with this table. Because the inlet of minor actinides is also known an estimation of the consumed minor actinides can be made. No waste stream exists for the fifth cycle, therefore the ratio of burnup of the minor actinides over the amount at the beginning from the previous cycle is taken to calculate the difference for the last cycle. This is about 20 percent, so 20 percent of the original amount of minor actinides are consumed during burnup. An amount of 1435 kg of minor actinides is consumed for a theoretical density of 85% and 1% reprocess loss.



Figure 4.7: The radiotoxicity of the waste produced by loss at reprocessing per Tera Watt-hour produced. For the theoretical density 85% and loss of 1%

Figure 4.7 shows the net radiotoxicity of the fuel per Tera Watt-hour that has been produced by the GFR600. The net toxicity at ten years is 2.8 10^8 Sv/TW-h and decreases to zero in in 100 years. Then the net toxicity drops even lower to lower than - 2.4 10^8 Sv/TW-h at 500 years. From this lowest point the net radiotoxicity slowly rises to zero net radiotoxicity. More will be explained in the subsection 4.6.

4.2 85 Percent Theoretical Density and Two Percent Loss

4.2.1 Calculation of k_{eff}

The first cycle of the k_{eff} during burnup for a theoretical density of 85% and a loss of 2%, figure 4.8, is precisely the same as for a theoretical density of 85% and a loss of 1%. This is because the initial composition of the nuclear fuel is the same for both settings. There are no big differences in these two figures. The one thing that seems important to notice is that the first figure 4.1 for the last three periods the k_{eff} slightly decreases compared to the same last three periods of figure 4.8. That is a result of a larger addition of minor actinides for the loss of two percent as can be seen on the figures in 4.3 and 4.10. For the loss of 2% as well as for the loss of 1%, the averages of k_{eff} for the last three periods are very close to the boundary.

An interesting difference is that for two percent loss the average k_{eff} for the second cycle is smaller in figure 4.9 than the average k_{eff} in figure 4.2. For 2 percent that average is 1.04177, for one percent that average is 1.04241. This is unexpected because for two percent the amount of minor actinides added is larger. The value for 2 percent start a bit lower and ends higher than the k_{eff} for one percent. This can be explained by the fact that minor actides are absorbers, so for more MA added the value of k_{eff} starts lower. At the end an addition of MA gives a rise as absorbers capture neutrons and then become fissile, which can be seen by a stronger rise in k_{eff} over time. This stronger reactivity swing is not a positive property in view of safety and stability for a reactor.

4.2.2 Additions to the Fuel

For the reprocess loss of 2%, figure 4.10, the minor actinides total is 2.053,7 kg. The total depleted uranium added is 2.361,4 kg and therefore the total of added material to the nuclear fuel is 4.415,1 kg. The ratio of minor actinides added for the 2% loss is bigger than that for the 1% loss, but the difference in ratio is not significant enough to call it an improvement. The improvement of using more minor actinides as nuclear fuel is in the fact that there is about 150 kg extra material needed for completing the nuclear fuel per period. This will also give a bigger outlet of radioactive material that leaves the integral cycle.



Figure 4.8: Change of $k_{\rm eff}$ over time while burning of every cycle for 85% TD and 2% loss



Figure 4.9: Time average $k_{\rm eff}$ of every cycle for 85% TD and 2% loss



Figure 4.10: The addition of mass in kg for a theoretical density of 85% and a loss of 2%

4.2.3 Waste Stream Analysis



Figure 4.11: The losses per nuclide for storage are shown for a theoretical density 85% and reproceeding loss of 2%. The full y-axis is shown.



Figure 4.12: The losses per nuclide for storage are shown for a theoretical density 85% and reproceesing loss of 2%. The scale of the y-axis stops at 40%

The waste loss for the fuel with a theoretical density and a loss of 2%, is twice as big as with 1%. The composition of the loss vector is about the same, that is of course, due to the fact that only about 6% for 1% loss and about 7% for 2% loss is needed to complement the fuel each period, which



Figure 4.13: The losses per nuclide for storage are shown for a theoretical density 85% and reproceesing loss of 1%. The scale of the y-axis stops at 2.5%

is not a really big influence on the composition of the total nuclear fuel. As seen in the reprocessing addition figure 4.10, the amount of minor actinides added is a bit bigger. This results in some slight changes, for example in the 241 Am and in the amount of 243 Am and 237 Np.

The amount of minor actinides consumed for this setting is approximately 1541 kg. This is only a bit higher than the amount of MA consumed for 85% theoretical density and one percent loss.

The net radiotoxicity for a theoretical density of 85% and a reprocess loss of 2% is shown in figure 4.14. The radiotoxicity for this setting is a bit higher compared to the net radiotoxicity of 85% TD and 1% η , about 3.2 10⁸ Sv/TW-h at ten years. The shape of this figure is the same as the shape of the figure 4.7. The zero net radiotoxicity line is also reached at 100 years and the lowest point at 500 years. The lowest point is lower than for 85% TD and one percent loss. This can be explained by the fact that more minor actinides used to run the GCFR for these parameters. So the effects are a bigger for a larger inlet of minor actinides.



Figure 4.14: The radiotoxicity of the waste produced by loss at reprocessing per Tera Watt-hour produced. For the theoretical density 85% and loss of 2%

4.3 90 Percent Theoretical Density and One Percent Loss

For this project also a theoretical density of 90% is investigated. An increase of theoretical density leads to a bigger value for $k_{\rm eff}$, because more material is packed in the same volume, which leads to less neutron leakage and therefore a more efficient use of the neutrons.

4.3.1 Calculation of k_{eff}

The value for $k_{\rm eff}$ for the first cycle of figure 4.15 for 90% TD is higher than the value for $k_{\rm eff}$ in of figure 4.1 for the 85% density. This is a result of the different density used. The value for $k_{\rm eff}$ is about 0.02 higher and decreases the same over time. In the third cycle $k_{\rm eff}$ does not decrease over time. The fourth and fifth cycle have a comparable line for $k_{\rm eff}$, with the exception that at the end of the cycles the value of $k_{\rm eff}$ falls a little because of loss of fissile material.



Figure 4.15: Change of $k_{\rm eff}$ over time while burning of every cycle for 90% TD and 1% loss

For the theoretical density of 85%, only minor actinides are added for the first cycle. For this theoretical density the addition of minor actinides only consists of one third of the total. This means that the boundary for $k_{\rm eff}$ for the second cycle has been reached. This can be seen in figure 4.16. Only the time average of $k_{\rm eff}$ for the first cycle is beneath the boundary.



Figure 4.16: Time average k_{eff} of every cycle for 90% TD and 1% loss

4.3.2 Additions to the Fuel

The jump of k_{eff} between the first and the second cycle is not as large as for the density of 85%. This is because, as shown in figure 4.17, the amount of added minor actinides is only about one third of the total addition. This also results in a decrease of k_{eff} over time. The value of k_{eff} for the second cycle in figure 4.1 does not decrease. The reason for this is that there are less fertile isotopes. The number of fissile material is decreasing. The while more MA are added for the third cycle.

For a theoretical density of 90% more material is present in the fuel. This also shows in the figure 4.17. The difference between addition between 85% and 90% is not large, about 7.5 kg. The average addition for 90% is around 960 kg. The figure 4.17 looks the same as for figure 4.3. Only the first cycle shows a big difference, the rest of the cycles are comparable. The total amount of MA added for this setting is 921.8 kg. The total amount of depleted uranium is 2,924.7 kg and the total addition of material is 3,846.5 kg. The amount of minor actinides is 709.8 kg less than for the same reprocessing loss and 85% theoretical density.

4.3.3 Waste Stream Analysis

Because more 238 U is added, the total value is over 300%, see figure 4.18. This also changes the amount of minor actinides in the figures.



Figure 4.17: The addition of mass in kg for a theoretical density of 90% and a loss of 1%



Figure 4.18: The losses per nuclide for storage are shown for a theoretical density 90% and reproceeding loss of 1%. The full y-axis is shown.



Figure 4.19: The losses per nuclide for storage are shown for a theoretical density 90% and reproceesing loss of 1%. The scale of the y-axis stops at 40%

The amount of minor actinides consumed for 90% theoretical density and one percent loss is 951 kg. This is about 500 to 600 kg less than for a theoretical density of 85%. Obviously this is a result of the higher value for $k_{\rm eff}$ for a higher theoretical density. Therefore less MA are added to the cycle and less MA are consumed.



Figure 4.20: The losses per nuclide for storage are shown for a theoretical density 90% and reproceesing loss of 1%. The scale of the y-axis stops at 2%



Figure 4.21: The radiotoxicity of the waste produced by loss at reprocessing per Tera Watt-hour produced. For the theoretical density 90% and loss of 1%

The net radiotoxicity for 90% theoretical density in figure 4.21 also looks the same as the other radiotoxicity figures. The net radiotoxicity at ten years is a bit lower than for 85% TD and 1% η , see 4.7. The value for the net radiotoxicity is about 2.6 10⁸ Sv/TW-h. Because the inlet of minor actinides is smaller, also the radiotoxicity varies less.

4.4 90 Percent Theoretical Density and Two Percent Loss

4.4.1 Calculation of k_{eff}

The figures belonging to a theoretical density of 90% and a loss of 2%, figures 4.22 and 4.23, look almost completely the same as figures for theoretical density 90% and loss of 1%. From this I conclude that the influence of the percentage of loss during reprocessing can be accurately compensated by changing the composition of the addition, except for the a bit bigger reactivity swing.



Figure 4.22: The change of $k_{\rm eff}$ over time while burning of every cycle for 90% TD and 2% loss

Also for this setting the upper boundary for $k_{\rm eff}$ is reached after the first cycle.

4.4.2 Additions to the Fuel

The difference in reprocess addition for 90% and 2% loss is about 16 kg extra per cycle, compared to TD 85% and 2% loss. The total added minor actinides is 1,355.5 kg. The total addition of depleted uranium is 3,126 kg and the total added material is 4,481.5 kg. The amount of MA added relative to the total amount is bigger for 2% loss than for 1% loss. This can be seen if the addition figures 4.17 and 4.24 are compared. For the second cycle of 2% loss the addition of MA is less than for 1% loss. This can be explained by the fact that more kilogram MA is added in the first cycle for 2% loss.



Figure 4.23: Time average $k_{\rm eff}$ of every cycle for 90% TD and 2% loss

This results for the second cycle in a higher k_{eff} , so that at reprocessing less MA have to be added. Compared to 85% TD and 2% loss, the amount of MA added is smaller for 90% TD 2% loss. This is explained by the fact that a higher TD leads to a bigger k_{eff} . Because the first addition of material is different, it has an influence in the rest of the additions, which can be seen when comparing the figures for 1% and 2%. These two figures differ more from each other than those from 85% do.



Figure 4.24: The addition of mass in kg for a theoretical density of 90% and a loss of 2%

4.4.3 Waste Stream Analysis

The reprocess losses for different isotopes leaving the cycles for a theoretical density of 90% and a loss of 2%.



Figure 4.25: The losses per nuclide for storage are shown for a theoretical density 90% and reproceeding loss of 2%. The full y-axis is shown.



Figure 4.26: The losses per nuclide for storage are shown for a theoretical density 90% and reproceesing loss of 2%. The scale of the y-axis stops at 40%

The total consumption of minor actinides for 90% theoretical density and 2% loss is approximately 1373 kg. This is much more than for a theoretical



Figure 4.27: The losses per nuclide for storage are shown for a theoretical density 90% and reproceesing loss of 2%. The scale of the y-axis stops at 2%

density of 90% and 1% loss. This is still less than 85% TD and 2% loss, but the difference is not as big as for 90% TD and 1% loss.



Figure 4.28: The radiotoxicity of the waste produced by loss at reprocessing per Tera Watt-hour produced. For the theoretical density 90% and loss of 2%

Also with this figure 4.28, the net radiotoxicity has the same form as the other radiotoxicity figures. The graph is shifted a little bit, because of the higher net radiotoxicity. The net radiotoxicity is higher than the net radiotoxicity of 85% TD and a reprocess loss of 1%, although the amount of minor actinides added is smaller. This is probably caused by the higher reprocess loss, so more material is leaving the integral cycle.

4.5 Comparison of Results

In this subsection the differences for the different parameters are presented. The differences between one and two percent loss. In comparison to one percent loss:

- Twice as much material is to be stored for two percent loss. Around 600 kg more material is added in total for two percent loss, about 3.800 kg for one percent loss and 4.400 kg for two percent loss. This also leads to a larger addition of minor actinides.
- This larger addition of minor actinides leads slightly different values for k_{eff} . For two percent loss the k_{eff} for the last three cycles at the end of the cycle is a little higher.
- The net radiotoxicity is higher for two percent loss.
- The consumption of minor actinides is bigger for 2% loss than for 1% loss. The addition of minor actinides is bigger for 2% and therefore more minor actinides can be consumed.

• The net radiotoxicity for 2% is higher than for 1% because of the bigger losses at reprocessing.

It is also interesting to notice the difference between the different theoretical densities. The differences for the two chosen theoretical densities are the following:

- The value for k_{eff} for the first cycle is about 0.02 higher for a theoretical density of 90%.
- The higher value for k_{eff} of the first cycle results in reaching the chosen boundary for the time average of k_{eff} for the second cycle for a theoretical density of 90% instead of for the third cycle.
- The amount of waste to be stored is a little higher, because the nuclear core consists of more material for a theoretical density of 90%.
- Because of the higher value for the k_{eff} , the amount of minor actinides added is smaller for a theoretical density of 90%.
- The consumption of minor actinides during burnup is bigger for a theoretical density of 85%. This is because more minor actinides are added during reprocess.

4.6 Radiotoxicity Comparison with a PWR

To compare the result for the GFR600 with a PWR, the OTTO cycle is used for the PWR. That means that all the material needed for the PWR is not used before and the uranium used is obtained by enrichment of natural uranium. This creates a reference line for the radiotoxicity of the PWR, which shows the value of the radiotoxicity of the material used in its original state. So the production of radiotoxicity per energy procuced is compared. This is the reason why the radiotoxicity for the GFR600 can become negative.

In the figure 4.29, the radiotoxicity per Tera Watt-hour of an modern generation III PWR is shown. The value at ten years is approximately 2.2 10^8 Sv/TW-h and slowly decreases over time. The minor actinides created in the PWR is very small: about 0.1% of the total nuclear fuel. This value for the PWR is about the same as the results for the GFR600. Though the net radiotoxicity for the GFR600 decreases more rapidly than the radiotoxicity of the PWR. The net radiotoxicity for the GCFR is zero in about 100 years, for the PWR it only decreases about a factor 2 in that time.

The amount of minor actinides produced in one year by this PWR is about 27 kg. For a theoretical density of 85% and a loss of 1% the total of added minor actinides is 1.631,6 kg and for 2% that is 2.053,7 kg. That results in 60 and 76 annual discharges respectively of this PWR. Even for



Figure 4.29: The radiotoxicity of the waste produced per Tera Watt-hour produced for a modern PWR

the 90% TD the number of annual discharges needed is very high, 34 and 50 for a loss of respectively 1% and 2%. What is left out in this calculation is the begin amount of minor actinides used, which is 30 discharges, 800 kg. Compared to the total number of years that the GCFR is in business, 41.9 years, means that the number of PWRs needed to annually discharge their minor actinides to run one GCFR continuously varies from approximately 1.5 to 2.5 per year.

Chapter 5

Conclusion

In this project the possibility of an integral fuel cycle has been examined for the GFR600, with the conditions that only depleted uranium and minor actinides could are and the value of k_{eff} was bounded between 1.00 and 1.05. This has been investigated for a total of 41.9 years, consisting five periods of 1300 days burning and 2200 days of fuel reprocessing. The nuclear fuel at the very beginning consists of 79% depleted uranium, 16% plutonium and 5% minor actinides. With either a theoretical density of 85% or 90% and with a reprocess loss of either one percent or two percent.

The conclusions found are the following:

- It is possible to have an integral fuel cycle for a Gas Cooled Fast Reactor, for a theoretical density of 85% and 90% with a reprocess loss of one or two percent, with the conditions as stated above. The value for $k_{\rm eff}$ is for all of these four situations above critical, this means that the nuclear reaction is sustained during the burning of the fuel.
- Generally speaking addition of minor actinides results in an increase of k_{eff} , addition of depleted uranium in a decrease of k_{eff} . The effect of an increase k_{eff} by use of minor actinides becomes less strong with repeated addition of minor actinides. A bigger input of minor actinides leads to a bigger reactivity swing, which has a negative influence on the safety and stability of the reactor. The amount of minor actinides used in the GCFR is very large. The number of discharges for the PWR used in the research is varying from 64 to 106 annual discharges. The GCFR in various settings uses 1.5 to 2.5 annual discharges of the PWR per year. An equilibrium of additions has not been reached in the 5 periods under consideration.
- The net radiotoxicity of the waste produced by the GFR600 for all settings has has a value at ten years of around 3 10⁸ Sv/TW-h and decreases to zero in about 100 years. For all the settings the net radiotoxicity has a negative lowest point around 500 years. After that

time the net radiotoxicity slowly rises to zero. In comparison with the PWR this value is around the same value, but a bit higher. The net radiotoxicity of the GFR600 decrease a lot faster than the radiotoxicity of the PWR.

• The optimum setting found is a theoretical density of 85% and a reprocess loss of two percent. The amount of minor actinides consumed for these parameters is 1541 kg. It has a time average value for $k_{\rm eff}$ that reaches the upper boundary in the third period and uses a maximum of minor actinides of a total of 2.853,7 kg. The bigger loss however leads to an amount of radiotoxic waste that is about twice as much as for the loss of one percent, 1190 kg in total instead of 595 kg. The level of the net radiotoxicity is almost the same as for other settings and also decreases in around 100 years.

A recommendation for future research is to investigate the possibilities of a closed nuclear fuel cycle for other Gas Cooled Fast Reactors. Another recommendation is to calculate the radiotoxicity more precisely and for different definitions.

Chapter 6

Nomenclature

CR	Conversion ratio, page 4
<i>C</i>	Number of interactions per cm^2 [cm ⁻²], page 4
σ	$\dots \dots $
N	Atom density, page 4
Ι	Intensity $[\mathrm{cm}^{-2}s^{-1}]$, page 4
σ_a	\dots Absorption cross section [barn ⁻¹], page 4
σ_f	
σ_c	Capture cross section $[barn^{-1}]$, page 4
η	Reproduction factor, page 5
ν	\dots Number of neutrons produced per fission, page 5
α	Capture to fission ratio, page 5
<i>v</i>	\dots Velocity [m/s], page 6
ϕ	\dots Angle-dependent flux density $[m^2/s]$, page 6
<i>S</i>	Source strength, page 6
<u>r</u>	Displacement [m], page 6
<i>E</i>	Energy [J], page 6
<u>Ω</u>	Solid-angle [radians], page 6
<i>v</i>	\dots Velocity [m/s], page 6
$\Sigma_s \dots \dots$. Macroscopic differential scattering cross section, page 6
$\Sigma_t \dots \dots$	\dots Macroscopic differential total cross section, page 6
Σ_f	\dots Macroscopic differential fission cross section, page 6
Σ_a	Macroscopic differential absorption cross section, page 6
$\mathbf{k}_{eff} \dots \dots \dots$	Effective multiplication factor, page 6
η	Loss at reprocessing, page 13

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Appendix A

Waste Stream Analysis for GFR600

In this appendix, the amounts of loss at reprocessing is stated in kilograms.

A.1 Theoretical Density of 85 Percent

Table A.1:	The l	loss for	each	nuclid	e in	kg foi	theoretical	density	85%	and
loss of 1%										

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
902270	1.38547e-015	4.94325e-015	1.11480e-014	1.93039e-014	3.67807e-014
902280	1.67626e-007	4.47806e-007	5.29643 e-007	4.61293 e-007	1.60636e-006
902290	6.73293 e-010	2.21241e-009	3.50473e-009	4.35114e-009	1.07415e-008
902300	1.11174e-006	4.25167e-006	8.84545e-006	1.39395e-005	2.81483e-005
902310	2.33829e-012	1.66151e-012	1.27752e-012	1.07958e-012	6.35693e-012
902320	1.59938e-008	2.75547 e-008	3.47232e-008	3.99754e-008	1.18247e-007
902340	1.68534e-009	1.58616e-009	1.56575e-009	1.59623e-009	6.43350e-009
912310	6.58421 e-009	1.81804e-008	3.89871e-008	6.65294 e-008	1.30281e-007
912320	2.90447e-016	7.44221e-016	8.19828e-016	6.57164 e-016	2.51166e-015
912330	3.50198e-008	7.26086e-008	7.87704e-008	6.54461e-008	2.51845e-007
912340	2.53797e-014	2.38862e-014	2.35788e-014	2.40378e-014	9.68827e-014
912341	5.68204 e-014	5.34767e-014	5.27885e-014	5.38159e-014	2.16901e-013
922320	7.92860e-006	2.10924e-005	2.47978e-005	2.15085e-005	7.53275e-005
922330	3.76378e-006	1.40797e-005	2.84529e-005	4.42500e-005	9.05465e-005
922340	0.09356	0.25260	0.454608	0.653912	1.454691
922350	0.57507	0.40862	0.314189	0.265509	1.563395
922360	0.06795	0.10206	0.124715	0.141838	0.436567

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
922380	116.07718	109.24634	107.84059	109.9394	443.1035
922400	4.33684 e-017	2.68364 e-017	2.73073e-017	3.11064 e-017	1.28618e-016
932360	7.82626e-006	2.00534 e-005	2.20906e-005	1.77076e-005	6.76779e-005
932370	1.031866	2.13929	2.32075	1.92811	7.420029
932380	2.42122e-008	5.13752e-008	6.53682 e-008	6.41072 e-008	2.05062e-007
932390	8.88462e-007	1.78611e-006	1.93461e-006	1.63694 e-006	6.24613 e-006
932400	3.27655e-018	2.02753e-018	2.06311e-018	2.35014 e-018	9.71734e-018
942360	1.72597e-006	4.51372e-006	5.17443e-006	4.40839e-006	1.58225e-005
942370	1.36903e-020	3.00700e-020	4.08240e-020	4.43080e-020	1.28892e-019
942380	1.42619	2.587851	3.426898	3.703499	11.14444
942390	14.13019	13.57796	13.19940	13.09229	53.99986
942400	6.80337	6.961943	7.118336	7.227269	28.11092
942410	1.00448	0.656500	0.521217	0.479530	2.661729
942420	1.883261	2.002479	2.093707	2.107734	8.087182
942430	2.84214e-014	7.65957 e-014	1.35320e-013	1.95713e-013	4.36051e-013
942440	2.07878e-006	1.28635e-006	1.30892e-006	1.49103e-006	6.16510e-006
952410	3.93761	7.37383	7.47708	5.767198	24.5557
952420	1.71718e-006	3.64364e-006	4.63606e-006	4.546630e-006	1.45435e-005
952421	0.1330841	0.282387	0.35930	0.352369	1.12714
952430	1.0318588	2.074385	2.24686	1.901147	7.25425
962410	1.86463e-028	3.65009e-028	3.67598e-028	2.91154e-028	1.21022e-027
962420	0.000359506	0.000756674	0.000957358	0.000935343	0.00300888
962430	0.01009	0.017529	0.019379	0.017354	0.06435
962440	0.444417	0.890400	1.011284	0.920490	3.26659
962450	0.091898	0.195973	0.227763	0.210367	0.726002
962460	0.014532	0.034957	0.050095	0.059198	0.158783
962470	0.000824172	0.002221	0.003924065	0.005675	0.012644
962480	3.27795e-005	0.000134	0.0003283727	0.0006314	0.00112691
962500	1.60069e-012	7.322541e-012	2.04159e-011	4.6168e-011	7.55078e-011
972490	2.24507 e-009	8.609797e-009	2.24611e-008	4.7564e-008	8.08804e-008
982490	4.52652e-007	2.443512e-006	7.57331e-006	1.7874 e-005	2.83444e-005
982500	1.62908e-008	1.039383e-007	3.58019e-007	9.3863e-007	1.41687e-006
982510	3.81589e-010	3.666205e-009	1.38061e-008	3.8741e-008	5.65958e-008
982520	8.00580e-013	1.048255e-011	4.13605e-011	1.2167 e-010	1.74321e-010
982540	1.17817e-028	1.520159e-027	5.92414e-027	1.8180e-026	2.57428e-026
Total	148.758	148.808	148.811	148.774	595.151

Table A.2: The loss for each nuclide in kg for theoretical density 85% and loss of 2%

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
902270	2.77094e-015	9.762e-015	2.23738e-014	3.94337e-014	7.43404e-014
				Continued	d on next page

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
902280	3.35252e-007	9.83352e-007	1.21278e-006	1.08494e-006	3.61633e-006
902290	1.34659e-009	4.70894 e-009	7.62921e-009	9.56207 e-009	2.32468e-008
902300	2.2235e-006	8.59598e-006	1.82132e-005	2.91109e-005	5.81436e-005
902310	4.6766e-012	3.3047 e-012	2.54009e-012	2.1605e-012	1.26819e-011
902320	3.19876e-008	5.43408e-008	6.76969e-008	7.73103e-008	2.31336e-007
902340	3.37069e-009	3.14208e-009	3.08337e-009	3.14457 e-009	1.27407e-008
912310	1.31684e-008	3.59278e-008	7.82432e-008	1.35892e-007	2.63232e-007
912320	5.80896e-016	1.65483e-015	1.9129e-015	1.57618e-015	5.72481e-015
912330	7.00398e-008	1.58755e-007	1.79124e-007	1.52869e-007	5.60788e-007
912340	5.07596e-014	4.73169e-014	4.64328e-014	4.73544 e-014	1.91864e-013
912341	1.13641e-013	1.05933e-013	1.03954e-013	1.06017 e-013	4.29545e-013
922320	1.58572e-005	4.63344 e-005	5.68064 e-005	5.06119e-005	0.00016961
922330	7.52756e-006	2.94505e-005	6.0558e-005	9.50039e-005	0.00019254
922340	0.187125	0.516602	0.946187	1.37862	3.02854
922350	1.15014	0.812744	0.624698	0.531344	3.11893
922360	0.135902	0.201199	0.243145	0.274455	0.854702
922380	232.154	216.409	212.365	216.58	877.509
922400	8.67369e-017	5.15401e-017	5.14765e-017	5.90176e-017	2.48771e-016
932360	1.56525e-005	4.45902e-005	5.15441e-005	4.2471e-005	0.000154258
932370	2.06373	4.67744	5.27738	4.5037	16.5223
932380	4.84245e-008	1.08525e-007	1.42157 e-007	1.42827 e-007	4.41934e-007
932390	1.77692e-006	3.88427e-006	4.35839e-006	3.76297 e-006	1.37826e-005
932400	6.55311e-018	3.89394 e-018	3.88913e-018	4.45888e-018	1.87951e-017
942360	3.45195e-006	9.92909e-006	1.18729e-005	1.03946e-005	3.56486e-005
942370	2.73807e-020	6.30328e-020	8.80823e-020	9.75757e-020	2.76071e-019
942380	2.85239	5.39855	7.33273	8.08173	23.6654
942390	28.2604	26.8628	25.8616	25.4886	106.473
942400	13.6068	13.8247	14.0619	14.2051	55.6984
942410	2.00896	1.29715	1.01525	0.923839	5.2452
942420	3.76652	4.01512	4.22462	4.27858	16.2848
942430	5.68429e-014	1.57995e-013	2.82429e-013	4.11539e-013	9.08807e-013
942440	4.15758e-006	2.47048e-006	2.46744e-006	2.82891e-006	1.19244e-005
952410	7.87523	16.0632	16.9613	13.4578	54.3575
952420	3.43437e-006	7.69686e-006	1.00821 e-005	1.01296e-005	3.1343e-005
952421	0.266168	0.596517	0.781377	0.78506	2.42912
952430	2.06372	4.51119	5.06183	4.37031	16.007
962410	3.72927e-028	7.90897e-028	8.24222e-028	6.73574 e-028	2.66162e-027
962420	0.000719011	0.00159909	0.00208274	0.00208474	0.00648557

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
962430	0.0201856	0.0371213	0.042083	0.0385091	0.137899
962440	0.888836	1.91595	2.23723	2.06996	7.11198
962450	0.183797	0.424574	0.508565	0.477123	1.59406
962460	0.0290656	0.074233	0.108439	0.129161	0.340899
962470	0.00164834	0.00458159	0.00818996	0.0119339	0.0263538
962480	$6.55591 \mathrm{e}{-005}$	0.000269315	0.000659914	0.00127399	0.00226878
962500	3.20138e-012	1.42112e-011	3.90531e-011	8.86586e-011	1.45124e-010
972490	4.49016e-009	1.68819e-008	4.3654 e-008	9.27828e-008	1.57809e-007
982490	9.05306e-007	4.79136e-006	1.47316e-005	3.48136e-005	5.52419e-005
982500	3.25817e-008	2.00389e-007	6.78264 e-007	1.77898e-006	2.69021e-006
982510	7.63179e-010	7.0249e-009	2.58514 e-008	7.25252e-008	1.06165e-007
982520	1.60116e-012	1.9967 e-011	7.6543 e-011	2.25094 e-010	3.23205e-010
982540	2.35636e-028	2.85871e-027	1.07188e-026	3.29172e-026	4.67303e-026
Total	297.515	297.644	297.664	297.589	1190.41

A.2 Theoretical Density of 90 Percent

Table A.3: The loss for each nuclide in kg for theoretical density 90% and loss of 1%

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
902270	1.48089e-015	5.1925e-015	1.00529e-014	1.61886e-014	3.29148e-014
902280	1.75931e-007	2.677 e-007	3.5291e-007	3.18269e-007	1.11481e-006
902290	6.7534 e-010	1.57824 e-009	2.53046e-009	3.23949e-009	8.02353e-009
902300	1.16822e-006	4.05629e-006	7.62332e-006	1.15035e-005	2.43513e-005
902310	2.5413e-012	1.82898e-012	1.37701e-012	1.12527 e-012	6.87256e-012
902320	1.51124e-008	2.73754e-008	3.50578e-008	4.04154 e008	1.17961e-007
902340	1.79095e-009	1.77578e-009	1.74366e-009	1.76408e-009	7.07447e-009
912310	6.72273e-009	1.8202e-008	3.35679e-008	5.31834e-008	1.11676e-007
912320	3.20681e-016	4.24184e-016	5.53031e-016	4.58418e-016	1.75631e-015
912330	3.86223e-008	4.50461e-008	5.68806e-008	4.83003e-008	1.88849e-007
912340	2.69701e-014	2.67417e-014	2.62579e-014	2.65655e-014	1.06535e-013
912341	6.03808e-014	5.98695e-014	5.87864e-014	5.9475 e-014	2.38512e-013
922320	8.32044e-006	1.2569e-005	1.65382e-005	1.4842e-005	5.22696e-005
922330	4.08752e-006	1.14269e-005	2.25173e-005	3.44558e-005	7.24876e-005
922340	0.0981644	0.227229	0.381814	0.533122	1.24033
922350	0.624996	0.449811	0.338655	0.276744	1.69021
922360	0.0685455	0.108566	0.134174	0.152541	0.463826
922380	123.351	122.306	120.093	121.500	487.25
922400	4.01919e-017	3.12039e-017	2.96629e-017	3.19505e-017	1.33009e-016
932360	8.64092e-006	1.14298e-005	1.49017e-005	1.23523e-005	4.73247e-005
932370	1.13801	1.32723	1.67586	1.42301	5.56411
932380	2.70383e-008	3.92549e-008	4.99913e-008	4.94295e-008	1.65714e-007
932390	9.70837 e-007	1.14446e-006	1.43747e-006	1.24999e-006	4.80275e-006
932400	3.08803e-018	2.39747e-018	2.27906e-018	2.45483e-018	1.02194e-017
942360	1.73589e-006	2.54696e-006	3.32285e-006	2.91964 e006	1.05253e-005
942370	1.36445e-020	2.28666e-020	3.01487e-020	3.26289e-020	9.92887e-020
942380	1.49355	2.07746	2.68467	2.88301	9.1387
942390	14.9502	14.6694	14.4061	14.3374	58.3631
942400	7.18304	7.28217	7.39989	7.49139	29.3565
942410	1.07053	0.703971	0.559439	0.511797	2.84574
942420	1.99515	2.00478	2.03155	2.00416	8.03563
942430	2.85004 e-014	6.31494 e-014	1.08024 e-013	1.54044 e-013	3.53718e-013
942440	2.03495e-006	1.57988e-006	1.50186e-006	1.61768e-006	6.73436e-006
952410	4.34129	4.71469	5.56533	4.40885	19.0302

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
952420	1.91762e-006	2.78405e-006	3.5455e-006	3.50565e-006	1.17528e-005
952421	0.148618	0.215767	0.274781	0.271693	0.910859
952430	1.12753	1.32917	1.66948	1.45173	5.57791
962410	1.83174e-028	2.21551e-028	2.62815e-028	2.11781e-028	8.79322e-028
962420	0.000400429	0.000576012	0.00073214	0.000721221	0.0024298
962430	0.00986396	0.0119534	0.0143747	0.0130712	0.0492633
962440	0.471581	0.589421	0.755321	0.702846	2.51917
962450	0.099451	0.126196	0.166583	0.157047	0.549276
962460	0.0154363	0.0253555	0.038053	0.0452182	0.124063
962470	0.000819825	0.00181652	0.00310734	0.00443113	0.0101748
962480	3.11233e-005	0.000123165	0.000283872	0.000526904	0.000965064
962500	1.35897 e-012	7.39473e-012	1.90571e-011	4.04034e-011	6.82142 e-011
972490	2.15154e-009	9.15392e-009	2.20842e-008	4.42889e-008	7.76785e-008
982490	4.16963 e-007	2.48385e-006	7.29016e-006	1.63349e-005	2.65259e-005
982500	1.37592e-008	1.07884e-007	3.52106e-007	8.62752 e-007	1.3365e-006
982510	3.08574e-010	3.80334e-009	1.37872e-008	3.59079e-008	5.3807 e-008
982520	6.25832 e-013	1.09912e-011	4.22491e-011	1.14605e-010	1.68471e-010
982540	8.30061e-029	1.58912 e-027	6.03546e-027	1.68788e-026	2.45864e-026
Total	158.188	158.172	158.194	158.169	632.723

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
902270	2.96179e-015	1.01986e-014	2.07332e-014	3.32695e-014	6.71632e-014
902280	3.51862e-007	6.26996e-007	7.58173e-007	8.21817e-007	2.55885e-006
902290	1.35068e-009	3.44953e-009	5.3838e-009	7.26526e-009	1.74493e-008
902300	2.33644e-006	8.31951e-006	1.60639e-005	2.47441e-005	5.1464 e-005
902310	5.0826e-012	3.62975e-012	2.74239e-012	2.23586e-012	1.36906e-011
902320	3.02247 e-008	5.40684 e-008	6.88473 e-008	7.85762e-008	2.31717e-007
902340	3.5819e-009	3.50328e-009	3.4625e-009	3.44095e-009	1.39886e-008
912310	1.34455e-008	3.57866e-008	6.9165 e-008	1.09272e-007	2.27669e-007
912320	6.41363e-016	1.02566e-015	1.18648e-015	1.2585e-015	4.11201e-015
912330	7.72447e-008	1.09036e-007	1.23844e-007	1.30295e-007	4.40419e-007
912340	5.39402e-014	5.27563e-014	5.21422e-014	5.18177e-014	2.10656e-013
912341	1.20762e-013	1.18111e-013	1.16736e-013	1.1601e-013	4.71619e-013
922320	1.66409e-005	2.94643 e-005	3.54978e-005	3.84053 e-005	0.000120008
922330	8.17504e-006	2.43819e-005	4.71647 e-005	7.51471e-005	0.000154869
922340	0.196329	0.476615	0.811506	1.16686	2.65131
922350	1.24999	0.892685	0.674451	0.549878	3.36701
922360	0.137091	0.214368	0.263712	0.296539	0.911709
922380	246.701	241.286	238.478	236.994	963.459
922400	8.03838e-017	6.19074 e-017	6.11269e-017	6.08581 e-017	2.64276e-016
932360	1.72818e-005	2.7637 e-005	3.19703e-005	3.3911e-005	0.0001108
932370	2.27603	3.2126	3.64877	3.8387	12.9761
932380	5.40766e-008	8.89177e-008	1.10416e-007	1.20939e-007	3.7435e-007
932390	1.94167e-006	2.7313e-006	3.1053e-006	3.27869e-006	1.1057 e-005
932400	6.17606e-018	4.75648e-018	4.69651e-018	4.67586e-018	2.03049e-017
942360	3.47179e-006	5.99076e-006	7.10443e-006	7.62137e-006	2.41884e-005
942370	2.7289e-020	4.87809e-020	6.39017 e-020	7.45698e-020	2.14541e-019
942380	2.98711	4.56046	5.84257	6.74881	20.1389
942390	29.9003	29.0655	28.4581	28.0476	115.471
942400	14.3661	14.5158	14.6900	14.8644	58.4363
942410	2.14107	1.39572	1.10559	0.995253	5.63763
942420	3.9903	4.05101	4.10064	4.12949	16.2714
942430	5.70009e-014	1.37447 e-013	2.36877e-013	3.46568e-013	7.77893e-013
942440	4.06989e-006	3.13442e-006	3.0949e-006	3.08129e-006	1.33805e-005
952410	8.68257	11.2783	11.9889	11.9549	43.9047
952420	3.83524e-006	6.30624 e-006	7.83097e-006	8.57729e-006	2.65497 e-005
952421	0.297236	0.488742	0.60691	0.664751	2.05764

Table A.4: The loss for each nuclide in kg for theoretical density 90% and loss of 2%

Isotope	First loss	Second loss	Third loss	Fourth loss	Total loss
952430	2.25506	3.17213	3.60648	3.80787	12.8415
962410	3.66349e-028	5.19661 e-028	5.56957 e-028	5.53858e-028	1.99682e-027
962420	0.000800857	0.00130635	0.00161647	0.00176708	0.00549075
962430	0.0197279	0.0276865	0.0316037	0.0329362	0.111954
962440	0.943162	1.3941	1.65337	1.7985	5.78913
962450	0.198902	0.300981	0.365039	0.406476	1.2714
962460	0.0308726	0.0577803	0.0834149	0.107423	0.279491
962470	0.00163965	0.00395371	0.00681386	0.00996914	0.0223764
962480	6.22467 e-005	0.000254603	0.000604432	0.00112102	0.0020423
962500	2.71795e-012	1.4814e-011	4.04483e-011	8.06106e-011	1.38591e-010
972490	4.30309e-009	1.86919e-008	4.73567e-008	9.10218e-008	1.61374e-007
982490	8.33925e-007	5.02936e-006	1.53627 e-005	3.38553e-005	5.50813e-005
982500	2.75185e-008	2.15395e-007	7.39139e-007	1.73235e-006	2.7144e-006
982510	6.17147 e-010	7.46779e-009	2.85442e-008	7.04637 e-008	1.07093e-007
982520	1.25166e-012	2.13017e-011	8.67042e-011	2.19823e-010	3.29081e-010
982540	1.66012e-028	3.02693e-027	1.23261e-026	3.10579e-026	4.65769e-026
Total	316.376	316.396	316.418	316.417	1265.61

Appendix B

Waste Stream Analysis for PWR

The data used for the waste stream is the content of the PWR after one

year for 1 ton nuclear fuel. The amounts are given in moles. Table B.1: The loss for each nuclide in kg for theoretical density 85% and loss of 2%

Isotope	Amount	
902300	6.201e-06	
902320	1.078e-06	
912310	1.655e-06	
912330	7.705e-08	
922320	1.414e-06	
922330	9.482e-06	
922340	8.073e-01	
922350	$4.371e{+}01$	
922360	2.212e + 01	
922370	5.086e-02	
922380	3.916e + 03	
932370	2.304e + 00	
932380	5.621 e- 03	
932390	2.977e-01	
942380	7.554e-01	
942390	2.257e + 01	
942400	8.517e + 00	
942410	6.538e + 00	
942420	2.517e + 00	

Isotope	Amount	
942430	2.889e-05	
942440	7.512e-05	
952410	1.773e-01	
952421	3.693e-03	
952420	1.708e-04	
952430	5.366e-01	
962420	5.874 e-02	
962430	1.455e-03	
962440	1.501e-01	
962450	5.584 e-03	
962460	4.700e-04	
962470	4.955e-06	
962480	2.365e-07	
972490	2.274e-09	
982490	2.820e-10	
982500	9.535e-10	
982510	3.299e-10	
982520	1.591e-10	
982530	2.279e-13	
992530	1.770e-13	