Burnup Simulations of the Thorium Cycle in a MSFR using Perturbation Theory

Floris Pauwels

Supervisors:
Danny Lathouwers,
Jan Leen Kloosterman

15 December 2020
Reactor Institute Delft
The Netherlands

TU Delft

Abstract

In this thesis, the goal is to find out if perturbation theory can accurately predict various results like the radiotoxicity of spent fuel from a nuclear reactor, due to changes in the input. This perturbation theory makes use of the adjoint equation by which a change in detector response of a certain system due to a perturbation can be approximated without the need to recalculate the entire system. The system consists of densities of various nuclides in the thorium fuel cycle of a molten salt fast reactor (MSFR). A model is constructed that can calculate a response in the results due to perturbations in the system, both with the approximating perturbation method, and an exact method. Perturbation theory has been shown to work well for a variety of perturbed variables. The accuracy of the approximations depend on the nature and size of the perturbation. However, the simulations contain some crude approximations. For example, the neutron flux is taken as a constant over time and space, while in a real nuclear reactor, the complexity of the flux has a great impact on the workings of the nuclear process. Also, since no accurate cross section data regarding the MSFR is available, the cross section data from the liquid metal reactor (LMR) is used. Both the MSFR and the LMR are fast reactors.

Contents

1 Introduction

Humanity is currently facing the crisis of global warming. To overcome this crisis, the energy production needs to be shifted from energy sources that emit greenhouse gases to sustainable energy like solar and wind energy. It is relatively cheap and easy to generate energy from these techniques. The main problem with renewable energies like solar and wind energies is that they are variable in their power output. When there's no wind, there's no wind energy. When the sun doesn't shine, no solar energy is generated. Batteries can store excess energy for later use, but even though batteries get cheaper every year, for now this is an expensive option. Storing the energy as hydrogen is also possible, but much of the energy is lost due to the storage and conversion of the hydrogen.

Nuclear energy is a reliable source of energy that can be generated without the emission of greenhouse gases. There are however other problems concerned with nuclear energy. For example, the waste of nuclear reactors is radioactive and needs to be stored safely for a long time. Also, the nuclear power plant accidents of Fukushima and Chernobyl are still in people's minds. These accidents, accompanied by the fear of nuclear weapons, make people anxious and restrained with respect to nuclear reactors as a power source. With today's technology however, nuclear reactors can be built that are inherently safe. All in all, there are problems regarding the generation of nuclear energy, but with enough effort, all of these can be overcome.

The atomic nuclei inside the core of a nuclear reactor fission and decay. These processes are stimulated in order to eventually generate useful energy. After neutron absorption, the nucleus can fission, i.e. split into lighter atoms, which releases relatively large amounts of potentially useful energy. The resulting fission products can include a multitude of different isotopes. Because of these and other nuclear processes inside the reactor core, densities of the different nuclides change over time. It is important to understand the composition and behavior of the reactor core, in order to make an operational nuclear power plant.

The goal of this thesis is to research if the perturbation theory can accurately approximate various results like the radiotoxicity of many systems that only differ slightly, without the need of recalculating these systems for every minor change. A simulation is made in python to perform calculations on these systems, both with the approximate perturbation and exact methods. These results are compared to find the accuracy of the perturbation method.

In this thesis, the theory that describes how nuclear reactors operate is explained first. Special attention is given to the behavior of the fuel inside a reactor. After this, the perturbation theory is introduced. This is the theory that makes it possible to approximate the change of a result, due to a perturbation of a known system, without the need to recalculate this system. It is explained how the theory is embedded into the simulation code. Finally, the results of these simulations are discussed. In this discussion, the accuracy of perturbation theory is analyzed.

2 Reactor & Fuel Theory

2.1 Nuclear Reactor Physics

First, the basic workings concerning a nuclear reactor are discussed. Much of the theory below is derived from the book of Duderstadt and Hamilton, 'Nuclear Reactor Analysis'[1]. See this book for more elaborate information on the different aspects concerned with nuclear reactor analysis.

Nuclear Reactors

Nuclear reactors perform many tasks to create safe, economically viable energy. Inside the reactor, many parameters have to be carefully monitored and controlled. These include the fuel concentration, temperature, and the neutron chain reaction. A shield is in place to protect the surroundings from the intense radiation of the reactor core.

A nuclear reactor generates energy, which dissipates in the form of heat. This heat is diverted to create high temperature, high pressure steam. A steam turbine converts the energy from this steam into mechanical energy. A generator uses this mechanical energy to generate electricity.

There are different kinds of reactors. The kinetic energy of the neutrons responsible for the chain reaction varies depending on the reactor type. The atoms inside the reactor core react differently to different neutron speeds. Thermal reactors make use of slow 'thermal' neutrons, while a 'fast reactor' uses highly kinetic neutrons. Different reactor kinds also distinct themselves with different fuel & coolant substances.

The highly energetic fission reactions inside the reactor generate a large variety of mostly radioactive isotopes. Therefore, the nuclear composition of the core changes, making it harder to sustain the fission chain reaction. Most decay products are useless or even harmful to the process. So it is important to keep track of the core composition. If necessary, the nuclear reactor core has to be refueled, cleansed from decay products, or in other way modified to remain critical. In other words, maintaining the core composition is important to keep the fission chain reaction self-sustaining and constant.

Nuclear Reactions

An unstable atom can spontaneously decay due to the quantum mechanical phenomenon of tunneling. There are a variety of different forms of radioactive decay. In alpha-decay, the nucleus emits a ${}^4_2He$ particle. In beta-decay, the proton and neutron can change into each other, by emitting a beta-particle. In neutron emission, single neutrons can be emitted from the nucleus. By definition, nuclear fission occurs when a nucleus splits into at least two particles that are heavier than ${}^4_2He$.

The rate of spontaneous nuclear decay for particle x is given by the formula:

$$-\frac{dn_x(t)}{dt} = \lambda_x n_x(t), \tag{1}$$

with n_x the number or density of 'x' particles, and λ the decay rate corresponding to the nucleus type. So a nucleus always has the same probability of spontaneously decaying.

A more complete function takes into account other particles 'y' that can decay into particle x.

$$\frac{dn_x(t)}{dt} = -\lambda_x n_x(t) + \sum_y \lambda_{y\to x} n_y(t). \tag{2}$$

Another important reaction is the collision between a nucleus and a neutron. Scattering occurs when a neutron is bounced from the nucleus without entering its core, while giving off some kinetic energy. With other collisions, the neutron is absorbed into the nucleus, creating a compound nucleus. This compound nucleus is in an excited state and can decay to the ground state by emitting a gamma ray. This process is called *neutron capture*.

The compound nucleus is the beginning of multiple possible reactions. The often unstable compound nucleus can for example undergo alpha-decay, fission into two lighter nuclides, or radiate excess energy with a high-energy photon. It can also re-emit one or multiple neutrons.

Microscopic Cross Section

Neutron capture is a very important phenomenon in nuclear physics. With an increasing neutron flux, the probability for neutron capture also increases, as would be expected. The probability of a neutron capture also depends on the 'microscopic cross section' of the nuclides. This nuclide specific property determines the susceptibility of absorbing a neutron. The cross sections can vary for different neutron speeds.

The neutron capture cross section σ is measured in barns where 1 barn = $10^{-28}m^2$. 1 barn is roughly the geometrical cross section of a nucleus (i.e. the size of the nucleus). The microscopic cross section can be drastically different from the geometric cross section.

The rate of neutron absorption in nuclides of type x is given as:

$$\frac{dn_x(t)}{dt} = \phi(\boldsymbol{r}, t)(-\sigma^x n_x + \sum_y \sigma^{y\to x} n_y), \tag{3}$$

where the sum of y corresponds to the different nuclides that change into x after a neutron interaction, and $\phi(\boldsymbol{r}, t)$ is the neutron flux density. In a reactor core, fission events have sufficient energy to expel a few solitary neutrons with high speeds from the fissioning nucleus. These expelled neutrons can be the catalyst for new fissions, when these neutrons interact with fissile or fissionable nuclide.

The normal operating condition of a reactor in which nuclear fuel sustains a fission chain reaction is called criticality. A reactor achieves criticality (and is said to be critical) when each fission event releases a sufficient number of neutrons to sustain an ongoing series of reactions.[2] In a real reactor, the neutron flux depends on time and position, but for our purposes, we approximate the flux as constant over time and space.

The neutron cross section σ can be split up into cross sections specific to the subsequent reaction, like fission σ_f, radiative capture σ_γ, scattering σ_s, and so on. These different cross sections all change differently to a change in the neutron energies. For example, if the fission reaction of a nuclide needs lots of energy to occur, the cross section σ_f is smaller for low energy neutrons. The high energy of faster neutrons are in this case a better match for the energy required for a fission reaction.

The Burnup Equation

The change in nuclide density over time is calculated by merging equation 2 and 3 into the following equation:

$$\frac{dn_x(t)}{dt} = -n_x(t)(\lambda_x + \phi(t)\sigma^x) + \sum_y n_y(t)(\lambda_{y\to x} + \phi(t)\sigma^{y\to x}), \tag{4}$$

where the first term on the right is the decrease in 'x' due to spontaneous decay and neutron absorption. The second term is the sum of the different nuclides 'y' that can transform into nuclide 'x' by decay or neutron absorption.

The nuclide densities in the reactor core change due to neutron reactions and radioactive decay. It is important to know these changes in density to accurately predict the behaviour of the reactor. Simulations can be made to estimate the nuclide densities over time.

Formal Solution

The equation for a single nuclide was found in equation 4. Now, a system of equations is given that incorporates this equation for all different nuclides in the form of:

$$\frac{d\mathbf{n}(t)}{dt} = A\mathbf{n}(t). \tag{5}$$

Here, the vector $\mathbf{n}$ contains the densities of the different nuclides that are used in the problem. The matrix A comprises of the change of the densities via decay and neutron absorption corresponding with the nuclides. These give the following, more extensive equations:

$$\frac{d}{dt}\begin{bmatrix} n_1 \\ n_2 \\ \vdots \\ n_j \end{bmatrix} = \begin{bmatrix} -\lambda_1 - \phi\sigma^1 & \lambda_{2\to1} + \phi\sigma^{2\to1} & \dots & \lambda_{j\to1} + \phi\sigma^{j\to1} \\ \lambda_{1\to2} + \phi\sigma^{2\to1} & -\lambda_2 - \phi\sigma^2 & \dots & \lambda_{j\to2} + \phi\sigma^{j\to2} \\ \vdots & \vdots & \ddots & \lambda_{j\to3} + \phi\sigma^{j\to3} \\ \lambda_{1\to4} + \phi\sigma^{1\to4} & \lambda_{2\to4} + \phi\sigma^{2\to4} & \lambda_{3\to4} + \phi\sigma^{3\to4} & -\lambda_j - \phi\sigma^j \end{bmatrix} \begin{bmatrix} n_1 \\ n_2 \\ \vdots \\ n_j \end{bmatrix} \tag{6}$$

We define $\mathbf{n}_0$ as the initial density. This is the density at time t_0, the starting time. We assume the matrix A to be time-independent.

2.2 Thorium Fuel Cycle

Actinides

The nuclides in a reactor core fuel cycle can be divided into actinides and fission products. The actinides are a range of heavy nuclides that are important for nuclear fission. These actinides include uranium, plutonium and thorium. In a thorium fuel cycle, fissile, fissionable, and fertile actinides are used. Below, these different types of nuclides are explained.

Fissile materials are easiest to fission, since low energy neutrons are sufficient to fission fissile isotopes. Fissile isotopes include ${}^{233}_{92}U$, ${}^{235}_{92}U$, ${}^{239}_{94}Pu$, and ${}^{241}_{94}Pu$.

Fissionable materials are a broader class of materials that are able to fission. This includes isotopes that can only fission after the absorption of a fast neutron. For these nuclides, the energy of absorbing a thermal neutron is not sufficient for the critical energy required for fission.

Fertile materials are not fissionable by themselves, but they have the potential to change into fissile isotopes after absorbing a neutron. The fertile nuclides are thus converted. Fertile material includes $^{232}_{90}Th$.

Fuel Cycle

$^{232}_{90}Th$, has a half life of more than 10 billion years. This is much longer than the half-lives of the other thorium isotopes. This is why almost all the thorium found in nature is $^{232}_{90}Th$. This nuclide is not fissile, so it can't directly be used for fission. It can however absorb a neutron to eventually create $^{233}_{92}U$. This is the fissile nuclide that is responsible for most of the fission in the thorium fuel cycle.

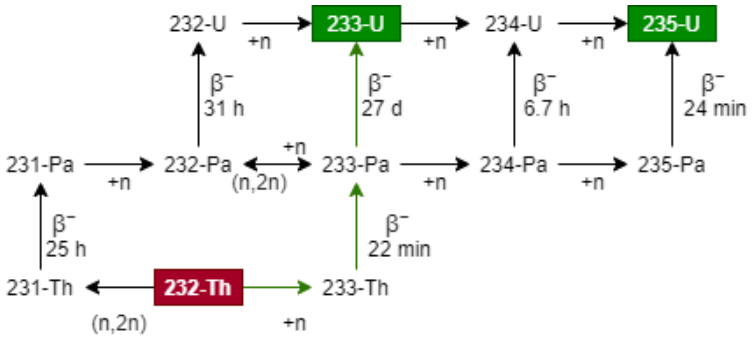

Figure 1: Simplified Thorium Fuel Cycle. $^{232}_{90}Th$ (red) is converted to create the fissile (green) $^{233}_{92}U$ and $^{235}_{92}U$ isotopes.

In this thesis, the simplified thorium fuel cycle from figure 1 will be used. This fuel cycle has three different reactions. The most common reaction is the absorption of a neutron. This is indicated by an arrow to the right, accompanied by +n. An arrow pointing up indicates β^- decay. The corresponding decay rate is also stated for the different decaying nuclides. The arrow to the left indicates an (n, 2n) reaction, where two neutrons are expelled after the absorption of a single neutron. This results in a net expulsion of one neutron.

Fission Yield

During a fission reaction, the nucleus usually splits into two smaller nuclei. These decay products are called the fission products. The fission products can include a variety of different nuclides. Often, these nuclides are radioactive and decay further into a stable nucleus.

As can be seen in figure 2, the resulting fission product of a fission reaction comprises of two atoms with different weight. The distribution of fission products is called the fission yield. This fission yield differs between different fissile isotopes. However, all fissile isotopes have in common that they usually split into two atoms of a notable mass difference.

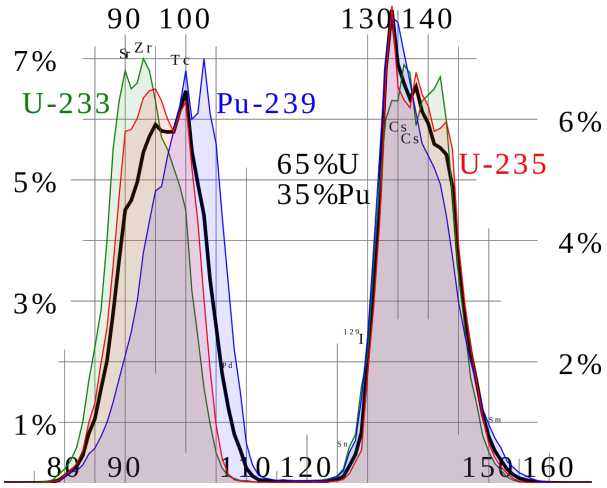

Figure 2: Fission product yields by mass number for fissile isotopes [3].

Poisons

The fission products are often useless, and can even have a negative effect on the fission chain reaction. These products can have large microscopic cross section, thereby absorbing neutrons needed for the fission chain reaction. For example, $^{135}_{54}Xe$ has an enormous thermal neutron cross section of 2.7 million barns. Even with a small concentration of this isotope, the fission chain may change drastically. These nuclides are called poisons, and they create what is called 'negative reactivity'. Poisons can also be inserted into the core to control the neutron density.

2.3 MSFR Fuel Management

In order to keep the fission process critical, neutron depleting poisons and other nuclides are removed from the reactor core. Different nuclides require different methods of extraction, provided they can be extracted at all. The possibility and procedure of extraction also depends on the reactor type.

MSFR Specific Properties

Take for example a 'Molten Salt Fast Reactor', or MSFR. This reactor type is a Generation IV reactor, which means it has significant enhancements over previous nuclear reactor designs. An advancement of a MSFR over most operational reactors is that it can operate close to atmospheric pressure. However, no MSFR reactor is operational as of yet. This reactor type is still in the design phase.

A MSFR makes use of fast neutrons. This means that no neutron moderator is required. A neutron moderator is a medium in the reactor core (e.g. water or graphite) that slows down neutrons to thermal energies. The fission cross section of fissile material is often much larger for these slow thermal neutrons than for high energy neutrons. In short, neutron moderators are used to increase the probability of fission.

To compensate for the absence of a neutron moderator, fast reactors require more fissile material. Also, to make up for the depletion of this fissile material, fertile material can be used for breeding new fissile material. In a Thorium fuel cycle, the fertile ${}^{232}_{90}Th$ isotopes located in the reactor core and blanket are used to create the fissile ${}^{233}_{92}U$.

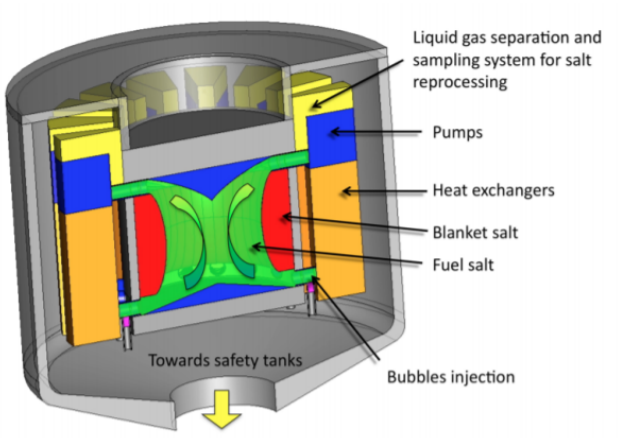

Figure 3: Design of a MSFR[4]. The major parts of the reactor core are mentioned, including the flowing fuel, installments to maintain core criticality, and a safety system.

Unlike other reactor salt, a MSFR has its nuclear fuel mixed in salt to form a high temperature molten composition. Because of this liquid core composition, the fuel can be displaced using pumps. This makes reprocessing the fuel salt possible while the reactor is operational[5]. See figure 3 for the design of a MSFR. The initial fuel composition of a Thorium MSFR core can be found in the Appendix.

Fuel Reprocessing

Fuel from a MSFR can be reprocessed relatively easy, since the liquid fuel can be diverted into reprocessing containers. The fuel reprocessing can also occur within the reactor core. Two of the most promising reprocessing methods in a MSFR are helium bubbling and fluorination[5].

Helium bubbling is a flotation process. This means that helium bubbles are injected at the bottom of the reactor (see figure 3). These bubbles are moving upwards due to the buoyant force. Certain elements bind to the helium bubbles, and will subsequently float to the surface, where these elements can be extracted.

Helium bubbling can be used when the reactor is in operation. The process is continuous, which means that certain particles can be continuously removed from the reactor core.

In addition to helium bubbling, there are other techniques to reprocess the fuel of a MSFR, like fluorination or lanthanide extraction[5]. These methods are implemented outside the reactor core. A portion of the homogeneous fuel can be tapped from the core for reprocessing, after which this fuel is pumped back into the reactor.

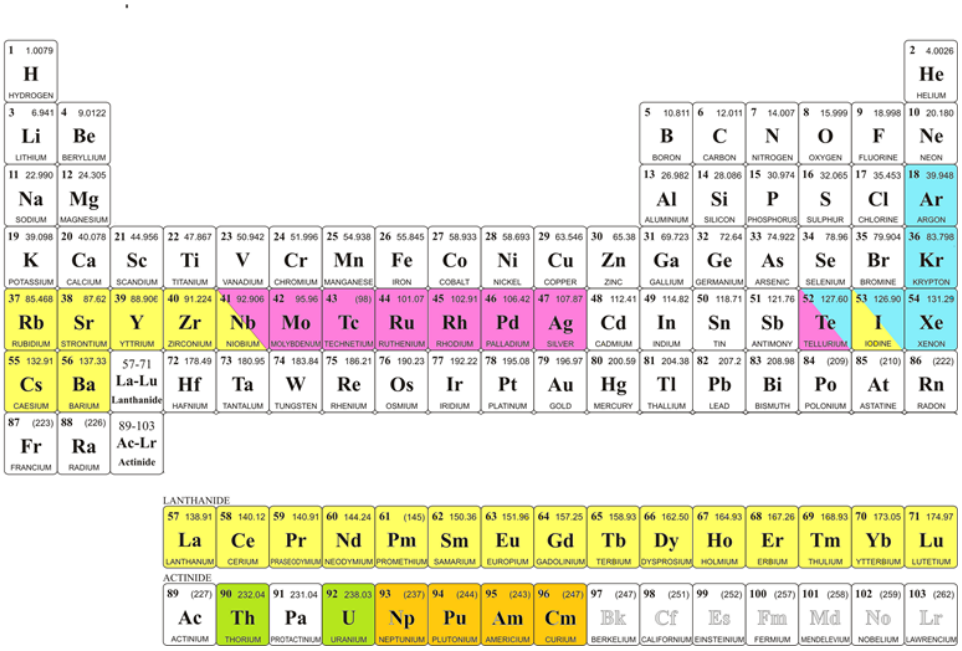

Figure 4: Overview of the elements formed in the fuel salt. Yellow elements remain dissolved in the fuel salt. The noble metals (purple) and gaseous fission products (blue) can be removed from the fuel salt via helium bubbling and/or fluorination. [6]

3 Perturbation Theory

Say, one is interested in the different effects due to many variations of a system. Calculating the burnup equation (eq. 4) for each of these system variations becomes computationally expensive. For these calculations, a more efficient method was devised by Gandini in 1975[7]. This method exploits time-dependent generalized perturbation techniques. For a given quantity, say density, one needs to calculate the forward and adjoint solutions of the burnup equations at unperturbed conditions. From these solutions, the sensitivity of a perturbation in this quantity on different parameters can be found with little computations. This method is called the 'Time-Dependent Generalized Perturbation Theory'.

Perturbation theory is a mathematical technique that is broadly used in many branches of physics. If many complex systems needs to be solved, it is often much easier to approximate these systems via perturbation theory than to find an exact solution. When all systems are variations of each other, it is unnecessary to calculate the whole system for every variation, or 'perturbation'.

In this sub-chapter on 'perturbation theory', first the general workings of perturbation theory in reactor physics are described. Important terms, like the response, and the adjoint equation are discussed. The importance function is introduced. Extra attention is given to the depletion perturbation theory. This theory is focused on systems where nuclides are transmuted over time.

3.1 Response

The response is a quantity that is to be analyzed. This response can be various quantities, like the density of one or multiple nuclides, and measures of radioactivity. The response R can be written as:

$$R = \int_0^{t_F} \mathbf{h}^T(t)\mathbf{n}(t)dt. \tag{7}$$

Here, $\mathbf{n}$(t) again represents the nuclide densities. The vector $\mathbf{h}$(t) represents assigned values, and can be seen as weights[8]. These values differ depending on what quantities are analyzed.

Say for example, one is only interested in the response of the density of a certain nuclide n_i integrated over time. The corresponding value h_i is thus set to 1, while the other values of $\mathbf{h}$ are set to 0. When quantities like the radiotoxicity are calculated, the vector function $\mathbf{h}$(t) becomes more composite, since multiple different nuclides can contribute to this radiotoxicity in varying degrees.

The goal is to find the change in the response, due to a perturbation. This perturbation can occur in multiple variables. Let's say there is a perturbation in $\mathbf{h}$(t), called $\Delta\mathbf{h}(t)$. The perturbed vector function becomes $\mathbf{h}(t) = \mathbf{h}_0(t) + \Delta\mathbf{h}(t)$, where $\mathbf{h}_0(t)$ is the original vector function for which the system is calculated. Similarly, the perturbed nuclide densities are written as $\mathbf{n}(t) = \mathbf{n}_0(t) + \Delta\mathbf{n}(t)$. From now on, the time dependence is excluded from notation.

A perturbation can also occur in the 'burnup and decay matrix' A (see equation 6). If for example the decay constant or the neutron cross sections are not well known, perturbation theory can be used to analyze the system across the range that these values can take. The range in possible values can be thought of as perturbations. With this technique, it becomes much easier to see the effects on the system for different possible physical values.

The perturbation in the 'burnup and decay matrix' ΔA induces a change in the nuclide densities, so Δn is dependent on ΔA. With this in mind, a response that includes perturbations can be formulated:

$$R = \int_0^{t_F} (\mathbf{h}_0 + \Delta\mathbf{h})^T (\mathbf{n}_0 + \Delta\mathbf{n}) dt \approx R_0 + \int_0^{t_F} \Delta\mathbf{h}\mathbf{n}_0 dt + \int_0^{t_F} \mathbf{h}_0 \Delta\mathbf{n} dt. \tag{8}$$

Here, the integral containing $\Delta\mathbf{h}\mathbf{n}_0$ is called the direct response, and can be calculated easily and directly. The second integral is called the indirect response. Since $\Delta\mathbf{n}$ changes for every change in the system, this integral cannot be calculated easily. $\Delta\mathbf{n}$ has to be recalculated for every new perturbation. We make use of the adjoint equation to find a difference in response, without requiring $\Delta\mathbf{n}$. This method can drastically decrease the computational time, although some approximations are made.

The inner product of two vector functions, say $\mathbf{x}$ and $\mathbf{y}$, is defined as:

$$< \mathbf{x}(t), \mathbf{y}(t) >= \int_0^{t_F} \mathbf{x}^T \mathbf{y} \; dt. \tag{9}$$

With this definition, R can be more concisely written as $\mathrm{R} = < \mathbf{h}, \mathbf{n} >$. This shorthand notation becomes useful in the following text.

3.2 Adjoint Equation

The adjoint of an operator, let's say L, is written as L*. This adjoint operator is defined so that the following equation holds:

$$< \mathbf{x}, L\mathbf{y} >=< L^*\mathbf{x}, \mathbf{y} > . \tag{10}$$

The adjoint of an operator is also sometimes called the hermitian conjugate, and the adjoint of a matrix is called the conjugate transpose.

Equation 5 can be more compactly written as:

$$L\mathbf{n} = 0, \tag{11}$$

where $\mathbf{n}$ is once again the density of the different isotopes, and the operator L is defined as:

$$L = \frac{d}{dt} - A. \tag{12}$$

The adjoint of a matrix can be found by taking the transpose of the matrix, and changing each element into its complex conjugate. The adjoint of an odd-ordered differential operator has an opposite sign from the forward operator.

$$\frac{d^*}{dt} = -\frac{d}{dt}.$$

Since we know the adjoint formulations for both the differential operator as well as the matrix contained in L, we can find L*:

$$L^* = \frac{-d}{dt} - A^T. \tag{13}$$

The matrix A only comprises of real elements, so each element is the same as its complex conjugate. Therefore, the conjugate transpose of matrix A is just its transpose.

Now, with the adjoint operator found in equation 13, the adjoint equation can be constructed:

$$-\frac{d\mathbf{n}^*(t)}{dt} = A^T\mathbf{n}^*(t). \tag{14}$$

This adjoint equation contains the importance function $\mathbf{n}^*$. The values included in $\mathbf{n}^*$ represent the importance of a certain nuclide on the final response, hence the name. To find the values of the importance function at every time, the value at a final time t_F is set to the previously defined vector $\mathbf{h}$ [9]. From these values of $\mathbf{n}^*(t_F)$, together with the adjoint equation (eq. 14), the importance function can be calculated at every time.

3.3 Response Difference

From the compact notation of equation 11, containing only the operator L, and the vector function $\mathbf{n}$, the inner product with the importance function $\mathbf{n}^*$ can be provided:

$$< \mathbf{n}^*, L\mathbf{n} > = < L^*\mathbf{n}^*, \mathbf{n} > = \ 0. \tag{15}$$

From the response equation (eq. 8), it is clear that if there is no perturbation in $\mathbf{h}$, the change in response can be written as:

$$\Delta R =< \mathbf{h}_0, \Delta\mathbf{n} > . \tag{16}$$

In the elaborations below, we attempt to rewrite this equation, so that ΔR no longer depends on $\Delta\mathbf{n}$, in order to simplify any future calculations of ΔR.

If the operator L is perturbed, this can be written as L = L_0 + ΔL, similar to $\mathbf{n}$, and $\mathbf{h}$. ΔL is equal to ΔA, since the perturbation can only occur in the matrix. Now, equation 11 becomes:

$$(L_0 + \Delta A)(\mathbf{n}_0 + \Delta\mathbf{n}) = 0,$$

or equivalently:

$$L_0\mathbf{n}_0 + L_0\Delta\mathbf{n} + \Delta A\mathbf{n}_0 + \Delta A\Delta\mathbf{n} = 0. \tag{17}$$

$L_0\mathbf{n}_0$ is 0 (eq. 11). Also, $\Delta A\Delta\mathbf{n}$ can be approximated as 0, as long as the perturbations are sufficiently small[10]. With this in mind, equation 17 can be abbreviated:

$$L_0\Delta\mathbf{n} = -\Delta A\mathbf{n}_0.$$

Taking the inner product of these terms with the importance function, and using the adjoint definition gives:

$$< L_0^* \mathbf{n}^*, \Delta \mathbf{n} >= - < \mathbf{n}^*, \Delta A \mathbf{n}_0 > . \tag{18}$$

If $\mathbf{n}^*$ is solved for:

$$L_0^* \mathbf{n}^* = \mathbf{h}. \tag{19}$$

Equation 16 and equation 18 can be combined to write the response difference as:

$$\Delta R = - < \mathbf{n}^*, \Delta A \mathbf{n}_0 > . \tag{20}$$

This is the final change in the response for a perturbation in the 'burnup and decay matrix' A. All of these values to calculate ΔR are already known. $\mathbf{n}^*$ doesn't have to be recalculated for every perturbation, unlike Δn. This method drastically decreases computational time when many perturbations from the same general system are modelled.

4 Application into Python

A system is modelled in python to see if perturbation theory can be used to calculate results from a variation in initial values faster. This model makes use of the equations covered in previous sections.

Model Features and Uncertainties

The workings of a MSFR are modelled. The physical values, like the cross section values that are used, can be found in the appendix, chapter B1. It should be noted that these values may not all be accurate for the MSFR. Below, the most important examples are given in which the model differs from a real reactor core.

The neutron flux density is modelled for two values, namely for 10^{14} and $5 \cdot 10^{15}$ neutrons per square centimeter per second. This flux is approximated to be homogeneous over the reactor core. This is clearly not what is happening in an actual reactor core, where the flux can vary greatly in position and time[1].

To accurately simulate the behaviour of a nuclear reactor, thermodynamics, fluid mechanics, and heat transfer have to be taken into consideration. However, these disciplines are not tackled in the simplified simulation of this thesis.

Also, the cross sections and fission yields both depend greatly on the distribution of neutron energies. An accurate system would know this distribution of neutron energies, and could calculate the cross section and fission yields for this distribution. However, accurate data of the energy distribution, and the range of dependencies of the cross sections and fission yields on this distribution are unavailable. Instead, the values of cross sections and fission yields are taken from states with approximately the same conditions.

Furthermore, the applied cross-sections are from the data of a liquid metal reactor. These values are similar to the cross-sections inside a MSFR, but the data undoubtedly contains discrepancies from the intended values. This thesis is therefore not suited for those who want accurate results for the MSFR. The method can however give insight in modelling the workings of a reactor core, and performing perturbation calculations.

Model Workings

In figure 5, a flowchart of the workings of the Python code is given. This code calculates properties like nuclide density and radioactivity in a system over time, both with the forward and the adjoint equation. The system can be perturbed in multiple ways.

First the initial values are declared. These values include the neutron flux, the simulated time range, and also the different physical constants for the important nuclides. For fissionable nuclides, it is also necessary to give the probability that certain fission products will emerge from a fission reaction.

Following this, a burnup and decay matrix is made, corresponding to equation 6. For each nuclide that is simulated, the rate of change is calculated for every reaction that decreases its density. In accordance with this decreasing rate of change, the entry that is responsible for the density change in the original nuclide on the diagonal of figure 6 is decreased. If this nuclide transmutes into another simulated nuclide, the corresponding entry between the original and the new nuclide is added by the rate of change that was subtracted from the original nuclide entry.

Now the system can be perturbed. This can occur both in the initial density, as well as in any variable contained in the burnup and decay matrix.

The forward calculations are performed with the use of the implicit Euler method:

$$\mathbf{n}[t+1] = (I + A * \Delta t) \cdot \mathbf{n}[t]. \tag{21}$$

This equation is derived from equation 5. If the initial density is known, the densities at every instance in time can be calculated with this equation. Here, **n**[t] is the current density, and **n**[t+1] is the density at a timestep Δt later. 'I' is the identity matrix, which just returns the same vector, when its inner product is calculated with a vector. '$A * \Delta t$' calculates the difference between densities in this time interval.

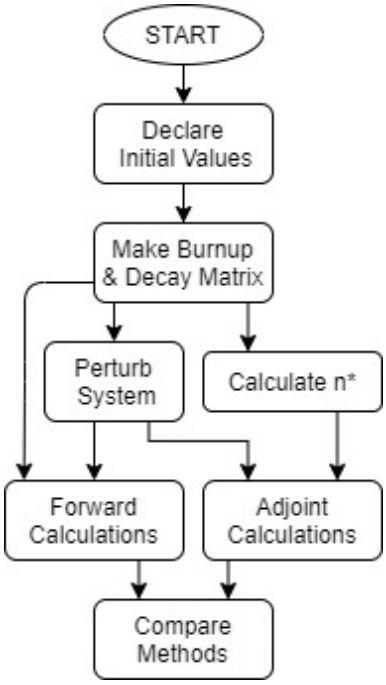

Figure 5: Flowchart of the code that compares the results of the forward with the adjoint calculations.

The importance function n* can be calculated at every instance with the following equation:

$$\mathbf{n}^*[t-1] = (I + A^T * \Delta t) \cdot \mathbf{n}^*[t]. \tag{22}$$

This equation is derived from the adjoint equation (eq. 14). This equation runs backwards in time. So when the values of $\mathbf{n}^*$ at a final time t_F are known, these values can be calculated at every instance. $\mathbf{n}^*(t_F)$ is taken to be **h**, where this vector function **h** depends on what is being calculated.

Now the difference in response can be calculated in two ways. With the forward equation, this response difference is calculated with the following equation:

$$\Delta R = (\mathbf{n}_{pert}(t_F) - \mathbf{n}(t_F)) \cdot \mathbf{h}. \tag{23}$$

This is a computationally expensive method, since $\mathbf{n}_{pert}$ has to be recalculated for every perturbation. For the following equations, the adjoint functions are used. With this method, no new calculations have to be done to find the response difference.

The following equation represents the difference in response due to a perturbation in the burnup and decay matrix δA:

$$\Delta R = \sum_{0}^{t_F} \mathbf{n}^*[t] \cdot \delta A \cdot \mathbf{n}[t] \ \Delta t. \tag{24}$$

This equation is derived from equation 20. Notice that in this equation, the response difference is calculated with a summation, instead of an integral. This is due to the discrete nature of the computer code.

The final equation is the response difference due to a difference in initial density $\delta \mathbf{n}_0$:

$$\Delta R = \mathbf{n}^*(t=0) \cdot \delta \mathbf{n}_0. \tag{25}$$

It is clear this equation is correct, when the meaning of the importance function $\mathbf{n}^*$ is properly understood. Each entry in the importance function at a certain time describes the fraction of which this entry will contribute to $\mathbf{n}^*$ at a final time t_F, where $\mathbf{n}^*(t_F)$ is determined as $\mathbf{h}$. The final response difference is the fraction of each nuclide that will contribute to the final response difference at the initial time, multiplied by the perturbation in initial density [11].

The results from the forward and adjoint calculations are compared, to detect how accurate the perturbation theory can approximate the correct values.

Obtaining Results

The different modelling results that need to be created are stated and justified. Firstly, it is important to get an understanding of the densities inside the reactor core. These densities are calculated with the relatively simple forward calculations of equation 21. The number of fission reactions per second are counted to create an estimation of the reactor power generation, assuming a 200 MeV gain per fission reaction.

After this, the importance function is plotted over time, to get a better understanding of the effect of the different nuclides on each other. A similar simulation is performed in the CRC Handbook of Nuclear Reactor Calculations[11], discussing the importance of uranium isotopes on the production of certain plutonium isotopes. The results of the handbook are compared to the findings of this thesis. It is interesting for discussion if, and how the two findings differ.

Finally, for the main part of the thesis, the system is perturbed in different ways to calculate the resulting response difference. These values are compared to the results obtained with the more computationally expensive forward calculations. First, the initial density $\mathbf{n}_0$ is perturbed. Hereafter, the values inside the 'burnup and decay matrix' A are perturbed. These values consist of the flux, the extraction coefficient, and the cross sections. It is interesting to evaluate how the system reacts to perturbations in these values, since the values are not always accurately known and can change over time. In the calculations, Δt is made sufficiently small to ensure that the choice of Δt doesn't have an impact on the results.

5 Results & Discussion

In this section, the results found with the python code are displayed and discussed. To get a feeling of the workings of a thorium cycle in the MSFR, properties like the evolution of the reactor composition are analyzed first.

5.1 Reactor Composition over Time

The initial composition of the simulated reactor is taken from table 1 in the appendix. The initial number of Th-232 nuclides is 0.0056 $b^{-1}cm^{-1}$. U-233 has an initial nuclide density of 0.0007 $b^{-1}cm^{-1}$. With these initial values, the densities of the actinides in the reactor core are calculated over time with the forward equation (eq. 21). These densities are plotted over a 1000 day period in figure 6.

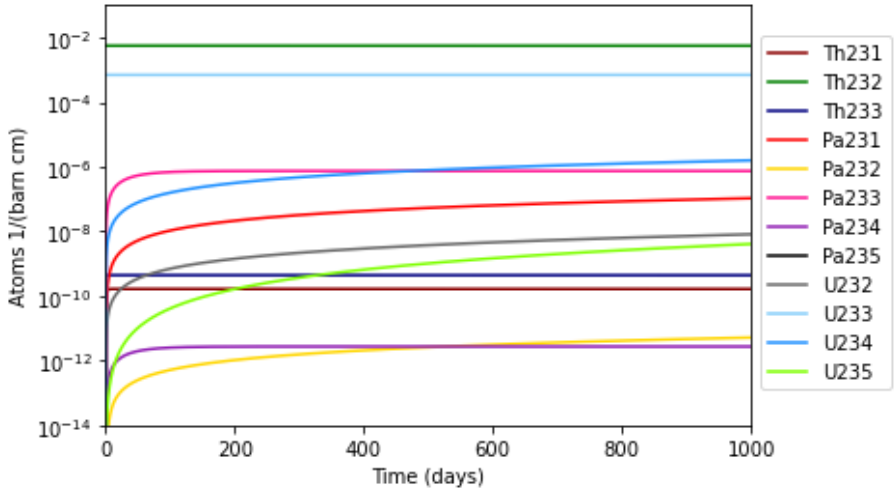

Figure 6: Densities of the important actinides in the thorium cycle over a period of 1000 days.

According to this density data, the initial densities don't change much. The final density of Th-232 is 0.00558 $b^{-1}cm^{-1}$. This is a mere 0.36% reduction over the period of 1000 days. The density of the fissile nuclide U-233 remains almost the same, since it's fission reaction is compensated by its production from other nuclides.

There can be different reasons for the relatively small change in reactor composition over time. It may be that the composition of a real reactor in practice also doesn't change all that much over the course of 1000 days. The relative changes in density are minuscule, compared to the vast amounts of nuclides in a reactor core.

Power Generation

Another reason figure 6 doesn't show significant density changes over time may be due to impractically low cross sections values, or similarly, a low neutron flux. To test this

hypothesis, the power generation of the simulated nuclear reactor is analyzed. The thermal power generation of a MSFR should be about 3GW [4]. By counting the number of fission reactions per second, the power generation can be calculated. A 200 MeV thermal energy gain is assumed per fission event. The resulting power can be seen in figure 7.

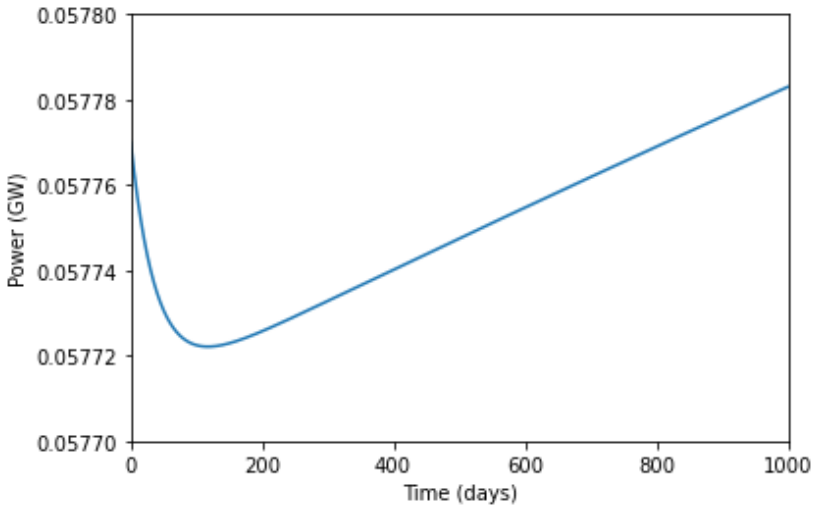

Figure 7: Power generation of the simulated reactor. The initial power decline is due to the time Th232 takes to convert to fissile uranium isotopes.

This figure shows the power generation is about 50 times lower lower than the expected 3GW power generation of a MSFR. In figures 19 and 20 in the appendix, results are displayed from simulation that increase the flux by a factor of 50 which results in the expected power. This power deficit is therefore probably attributed to low fission cross sections or a low flux. Increasing the neutron flux has the same effect as increasing the neutron cross sections. In the following results, the flux is assumed to be 10^{14} neutrons per square centimeter per second. The subsequent power deficit is no problem for the results in the following sections, since the workings and efficiency of the methods can be presented regardless of the rate of the nuclide transmutations.

Actinide Density Analysis

Now, figure 6 is used again to analyze the nuclides that had no initial density. For the analysis of these densities over time, the thorium fuel cycle in figure 1 is kept in mind.

Pa-233 is the most abundant protactinium isotope. This has two causes. Firstly, the half-life of Pa-233 is much higher than that of Pa-232, Pa-234, and Pa-235. Also, when the initially abundant Th232 isotope undergoes neutron capture, it becomes Th-233, which quickly decays into Pa-233. Pa-233 has a relatively sharp density increase until day 100, where the density remains constant. It is at this time that an equilibrium has been reached between the increase and decrease of the nuclide density of Pa-233. The nuclide is mostly produced from the decay of Th-233, and the decrease in density is mostly due to the beta-decay into U-233.

Pa-231 is also a relatively common protactinium isotope. Although this isotope is produced less than Pa-233, Pa-231 gradually builds up over time, instead of reaching an equilibrium. This can be explained with the much higher half-life of Pa-231 relative to the other protactinium isotopes

It is interesting to see that the uranium isotopes also gradually accumulate over time, due to the negligible half-life of these isotopes over the course of 1000 days.

Fission Products Analysis

The densities of some fission products are shown in figure 8. These nuclides are chosen, because they either produce or are produced by the Xenon-135 poison. Tellurium-135 rapidly decays into Iodide-135, which further decays into Xenon-135. This poison can further decay into Cesium-135 or absorb a neutron to become the stable isotope Xenon-136.

Xe-135 is important in reactor physics, because it has a very high neutron cross section. The presence of Xe-135 can therefore influence the fission chain reaction, even in small concentrations.

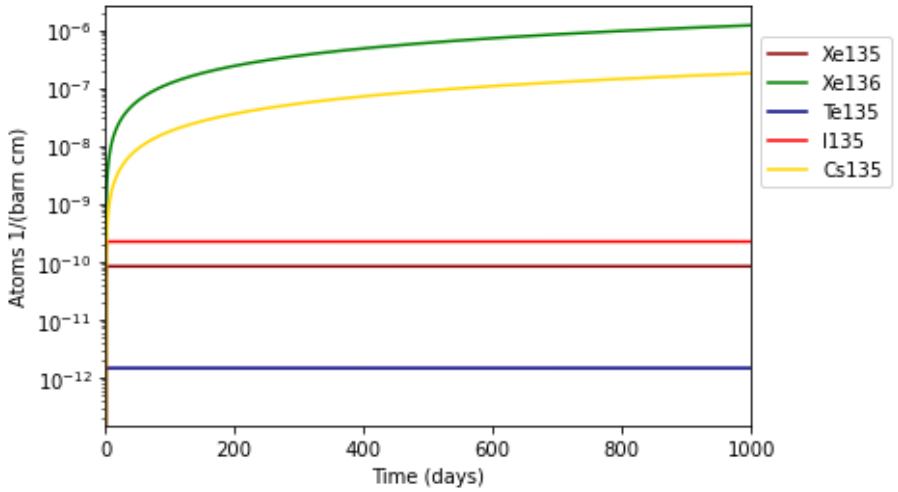

Figure 8: Densities of fission products in the reaction cycle of Xenon-135.

As can be seen from table 4 in the appendix, Xe-135 has a low half-life of 0.38 days. Usually, when nuclides have a half-life this low, they decay before they can capture a neutron. This is not the case for Xe-135, as can be interpreted from figure 8. Some of the Xe-135 nuclides perform beta-decay, and thereby change into Cs-135. A larger portion of the Xe-135 nuclides however undergo neutron capture and become Xe-136. This can be concluded from the higher density of Xe-136 relative to Cs-135 in the figure.

The poison Xe-135 can be extracted from the salt[5]. Therefore, the poisons are no longer taken into consideration in the next sections. However, the extraction methods may also remove useful actinides. The system will be analyzed for different extraction coefficients due to this mechanism in a later section.

5.2 Importance Functions

The importance function $\mathbf{n}^*$ signifies for every nuclide in time by what fraction these nuclides contribute to the final response at time t_F. In specific, say the response of a system is taken as the density of an important nuclide. In this system, $\mathbf{n}^*(t_F)$ is 1 for the entry corresponding with the important nuclide, and 0 for all other entries that correspond with the other nuclides in the system.

From equation 14, the importance function $\mathbf{n}^*$ can be calculated from start to finish, when the values at the final time are expressed. Now, at each time, the meaning of $\mathbf{n}^*$ for each nuclide is how likely it is that this nuclide is transmuted into the important nuclide at time t_F. The likelihood where the important nuclide is transmuted further before t_F is not included in the importance function.

Actinide Importance on Fissile Isotopes

For the following simulation, the importance function at time t_F, $\mathbf{n}^*(t_F)$ is chosen to be 1 for U-233 and U-235, and 0 for the other nuclides. The reason for this choice is that these uranium nuclides are important fissile nuclides. U-232 also has a fission cross section comparable to that of U-233 and U-235 in this configuration, as can be seen in table 2. However, U-232 is often not considered as a fissile isotope, since its fission cross section is much lower than that of U-233 and U-235 in thermal-neutron reactors [12]. The described importance functions are plotted in figure 9, and the importance functions with U-232 included as a response can be found in figure 22 in the appendix. The method for analyzing these plots is practically the same. The importance functions with the neutron flux adjusted to the 3GW reactor power can also found in the appendix in figure 21.

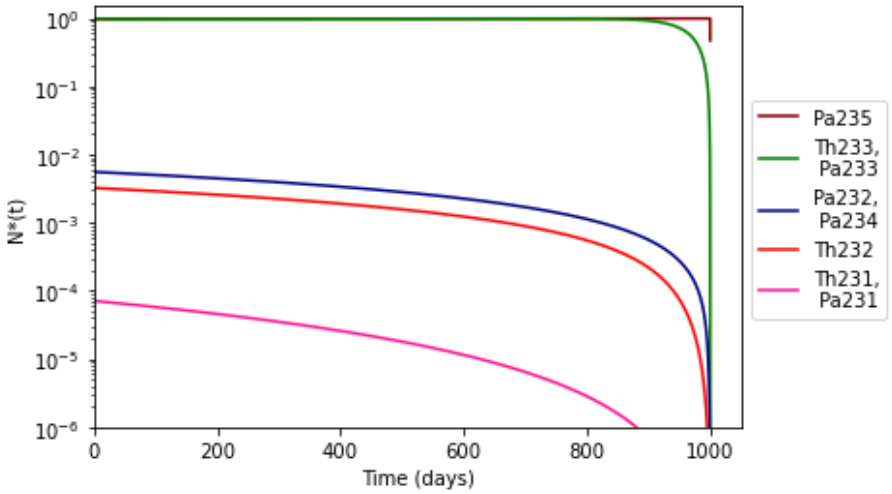

Figure 9: Importance functions of thorium and protactinium nuclides on the production of fissile uranium isotopes U-233 and U-235.

Now, the results of figure 9 are explained in order to get a better understanding of the workings of the importance function $\mathbf{n}^*$. First, let's look at Pa-235. This nuclide only needs to undergo beta-decay to transmute to U-235. This beta-decay occurs rapidly, since the half-life of Pa-235 is only 24 minutes. The importance function for this nuclide is almost 1 near the end of the simulation, since virtually all Pa-235 nuclide at this time are transmuted into U-235 at time t_F. The reason the importance function is close to 1 for the entire simulation, is because the U-235 nuclide doesn't fission or in other ways transmutes significantly. If there would be an initial amount of Pa-235 nuclides, most of these nuclides would decay to U-235 within a day and remain this important nuclide. This is in contrast to figure 21 in the appendix, where the fission rate is much higher. When the important nuclide can transmute further, nuclides that rapidly transmute to the important nuclide show a discernible slope.

The importance functions between Th-233 and Pa-233 are almost identical. This is because the half-life of Th-233 is just 22 minutes. after which the nuclide undergoes beta-decay into Pa-233. Pa-233 has a half-life of 27 days before it transmutes into U-233. This explains the descent of the importance function near the end of the simulation.

The importance functions of Pa-232 and Pa-234 also exhibit significant similarities. These nuclides most likely decay into U-232 and U-234 respectively. These uranium nuclides have a similar capture cross section. This neutron capture reaction results in the important U-233 and U-235 nuclides.

From figure 6, it can be seen that Th-232 most likely doesn't transmute at all in case of a low flux. This is in contrast with figure 21, where the importance function of Th-232 is much higher. If Th-232 undergoes neutron capture, the nuclide transmutes into Th-233. This nuclide is almost certainly transmuted into U-233 at time t_f, as explained before.

Th-231 and Pa-231 also have almost the same importance function. This is because Th-231 quickly decays into Pa-231. The reason this importance function is so low, is because neutron capture reaction is needed twice, in order to transmute into U-233. This reaction has a low probability of occurring, let alone twice.

Comparison with Williams Handbook

In figure 10, the importance function is analyzed for the uranium fuel cycle in a pressurized water reactor, in which the entry of $\mathbf{n}^*$ at t_F is 1 for the nuclides Pu-239 and Pu-240 and 0 for the other nuclides.

The same calculations are performed on page 157 in the "CRC Handbook of Nuclear Reactor Calculations"[11], albeit with differently used physical constants. The resulting plot is included in the appendix as figure 23.

These plots contain similarities, but there are also some notable differences. The U-240 importance functions show no large discrepancy. However, the U-239 importance function is a factor 10 larger in figure 10 than in the Williams book. This discrepancy may be contained in differently used values, but also in one or multiple missing reactions in the simulations.

Also, in the Williams book, the U-237 importance is higher, while the U-238 importance is lower, even though the shapes are roughly similar. This shows again that simulations can depend very much on the used constants.

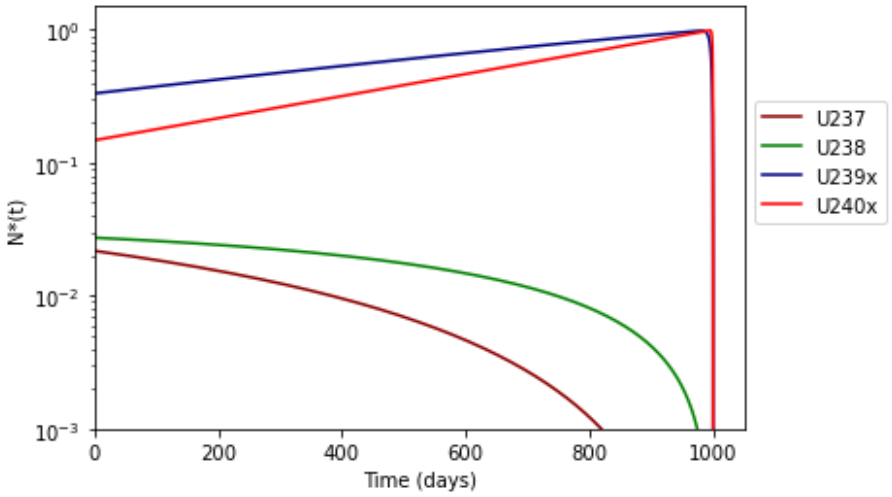

Figure 10: Importance functions of uranium nuclides on the production of fissile plutonium isotopes Pu-239 and Pu-240 in a PWR. The cross section values for U-239 and U-240 are missing, hence the x.

5.3 Accuracy of Perturbation Methods

Now, the perturbation method is used on a variety of perturbations.The initial densities, neutron flux, extraction coefficients, and cross sections are all perturbed. For each of these variables, the relative response change is calculated for both the adjoint method and the forward method, in order to find the accuracy of the adjoint method for differently sized perturbations.

The differences in response due to the perturbations are analyzed and discussed. This response is chosen to be the radiotoxicity of the nuclides at t_F, after a decay period of 10 years. See table 5 for the used values of the radiotoxic actinides. The available values contained in this table only include the toxicity of actinides, so the additional toxicity of fission products is not taken into account.

There are two reasons for the choice of radiotoxicity as the response. Firstly, it is important to know how high the radiotoxicity of spent fuel is, in order to safely store this fuel over long periods of time. Secondly, radiotoxicity is a representative variable of other response choices. Radiotoxicity depends differently on many actinides, both directly and indirectly. This translates into the variety of values in the vector function h corresponding with the radiotoxicity values. It can be estimated that other response choices with similarly complex h vectors give similar results.

5.3.1 Perturbed Initial Density

The initial density for Th-232 is perturbed. The resulting relative radiotoxicity response is plotted as a function of the initial density in figure 11, where the unperturbed system has a density of 0.0056 atoms per barn centimeter. The flux from these simulations is adjusted to the 3GW power output of a MSFR. See figure 24 for the results obtained with a significantly lower neutron flux.

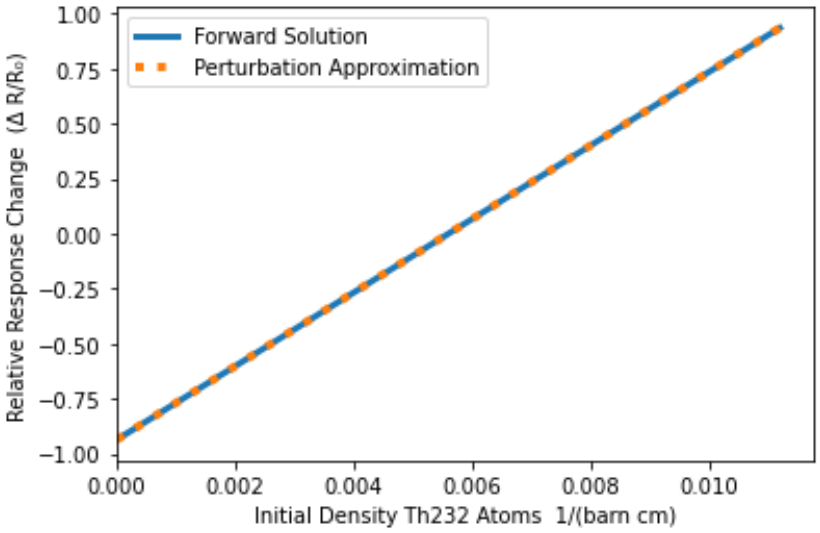

Figure 11: The relative radiotoxicity response as a function of the initial density in Th232 atoms, calculated with the perturbation and forward methods. The unperturbed system has a density of 0.0056 atoms $b^{-1}cm^{-1}$, and the neutron flux is $5 * 10^{15}$ $cm^{-2}s^{-1}$

From the figure, it can be seen that the perturbation approximation is the same as the forward solution. This is an interesting and useful result. The reason that this approximation is exact, can be explained by the linearity of the result. A certain perturbation in the density of a nuclide always changes the same amount of radiotoxicity, independent of the unperturbed composition. The perturbation approximation is a linear approximation, so when the exact solution is also linear, these methods provide the same result.

A disclaimer to this ideal result has to be made. It is approximated that no other variables change due to a perturbation in the initial density of Th-232. This is clearly an inaccurate approximation. As an example, in a real reactor the flux would undoubtedly change if the initial density of Th-232 would be doubled, as is done in the perturbation simulations.

5.3.2 Perturbed Flux

Now, the neutron flux is perturbed. The relative change in response as a function of this perturbation can be seen in figure 12.

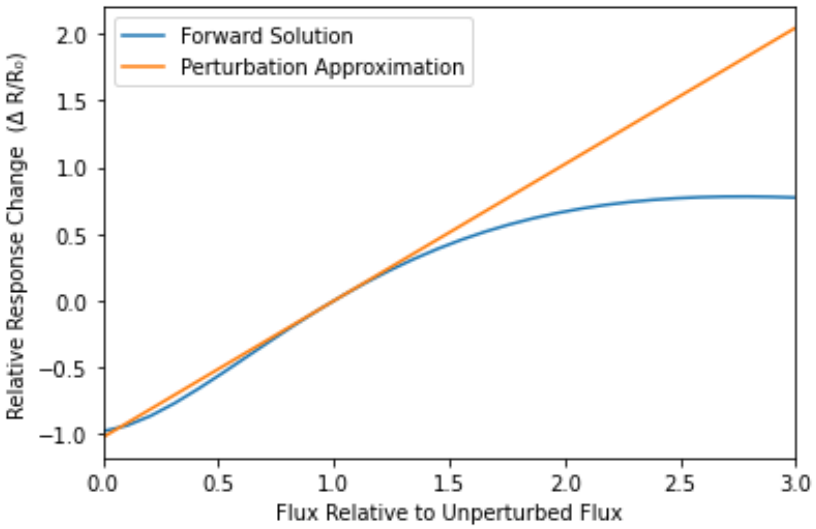

Figure 12: The relative radiotoxicity response as a function of the perturbed flux, relative to the unperturbed flux with a value of $5 * 10^{15}\ cm^{-2}s^{-1}$.

From this figure, it can be seen that the perturbation method remains reasonably accurate for any decrease in the flux. The methods diverge significantly when the change in flux is over 40% greater than the initial flux. The following explanation accounts for this discrepancy.

The reason radiotoxicity increases with an increase in flux comes from the fact that the initial Th-232 nuclides have a low radiotoxicity. When the flux increases, these nuclides transmute more often into more radiotoxic nuclides. Eventually the forward solution reaches an equilibrium, where the radiotoxicity doesn't further increase with an increase in the flux. This is due to the further transmutation and fission of highly radiotoxic nuclides into higher actinides and fission products that are not simulated. Since these nuclides are not simulated, they do not contribute to the radiotoxicity in the simulation. So even though the radiotoxicity may very well increase further with an increase in flux, this can't be concluded from figure 12, since the radiotoxicity of fission products and higher actinides is not taken into account.

The perturbation approximations estimate a higher radiotoxicity for an increasing flux. This is because these approximations are linear, with the tangent and initial point set at the unperturbed flux (1.0 on the x-axis). In conclusion, when the flux is increased, both methods become increasingly erroneous in calculating the radiotoxicity.

5.3.3 Perturbed Extraction Coefficient

Along with the extraction of poisons, a fraction of the actinides may be subtracted from the system. The extraction of these actinides is modelled similar to that of the half-life of a nuclide, a time after which half of the nuclides are removed. Instead of transmuting into another nuclide, this extraction process removes the extracted nuclides from the system altogether.

In figure 13 the relative response alteration as a function of the different extractions rates is plotted.

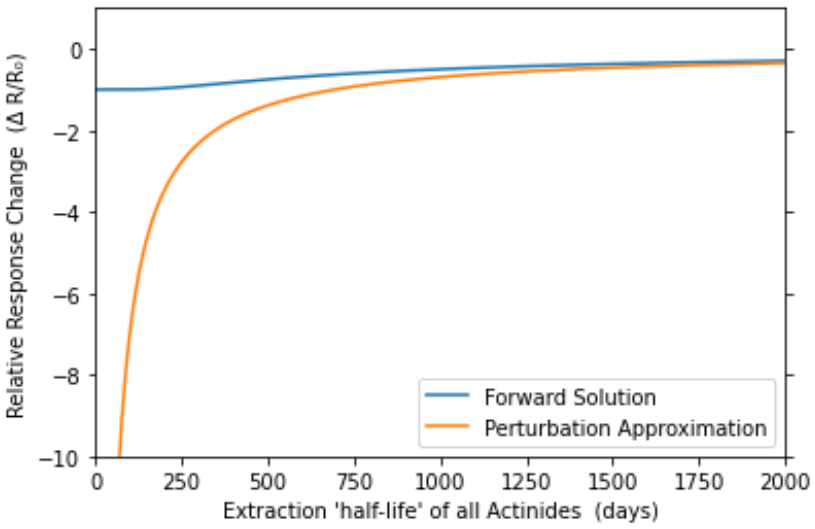

Figure 13: The relative radiotoxicity response as a function of the extraction 'half-life' of the actinides. Each nuclide is modelled to have the same extraction rate. The unperturbed system has no extraction, which means an extraction 'half-life' of infinity.

From this figure it can be seen that the forward solution and perturbation approximation increasingly diverge when the actinides have a shorter extraction 'half-life'. There are a couple things to note on this result.

An infinite extraction 'half-life' is the same as no extraction at all. In this limit the two methods should produce the exact same answer, since there is no perturbation in this situation. This is why the two methods converge for larger extraction 'half-lifes'.

The change in response cannot decrease further than -1, since this value means a 100% decrease in radiotoxicity, and thus no radiotoxicity at all. Less radiotoxicity than no radiotoxicity has no real physical meaning. The forward solution automatically contains this fact. The perturbation approximation however does not. The reason for this is that the response difference of the perturbation approximation is linearly dependent on the extraction strength. An extraction 'half-life' of 0 means an infinite extraction strength, since the actinide are extracted instantaneously. This is why the perturbation approximation can erroneously have a relative response change lower than -1.

Values lower than an extraction 'half-life' even close to 1000 days, at which the methods start to diverge significantly, are not realistic anyway. For extraction values like these, the core would lose its actinides too quickly to keep up a stable nuclear fission reaction.

5.3.4 Uncertainty in Cross Sections

Finally, the neutron capture & fission cross sections are perturbed. It is interesting to see how a change in cross sections would change the response. A change in cross sections can occur due to a change in flux, or due to a measurement error in the cross section data. There might be an uncertainty in the cross sections, which could entail that a system would need to be calculated for multiple cross sections. The relative response change as a function of this perturbation can be seen in figure 14.

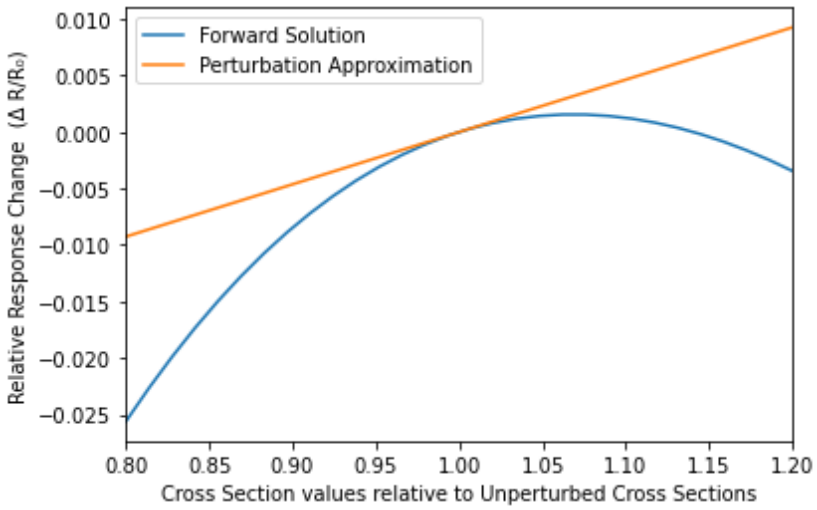

Figure 14: The relative radiotoxicity response as a function of the change in the capture & fission cross sections. Each cross section is modelled to have the same perturbation, which acts as a multiplier relative to its original cross section.

From this figure it can be seen that the responses calculated from the different methods diverge quickly, both when the cross sections are increased as well as decreased. This is due to a steep curve in the forward solution. The perturbation approximation is a linear approximation, so approximating values from a curve like this can be erroneous even when the perturbations are relatively minor.

In figure 14, both the capture and fission cross sections were perturbed with the same factor. The perturbation approximation is also simulated with the capture and fission cross sections perturbed separately. The results from these simulations can be found in figures 15 and 16. The radiotoxicity increases with an increase in capture cross sections, since Th-232 transmutes into more radiotoxic nuclides when this capture cross section is increased. The radiotoxicity decreases in this simulation when the fission cross sections are increased, since the fission products are not simulated. When the fission cross section increases, the density of radiotoxic fissile nuclides decreases, without any simulated fission product to compensate for this decrease in radiotoxicity. Fission products may very well be radiotoxic, so this result is just an inaccuracy in the simulation.

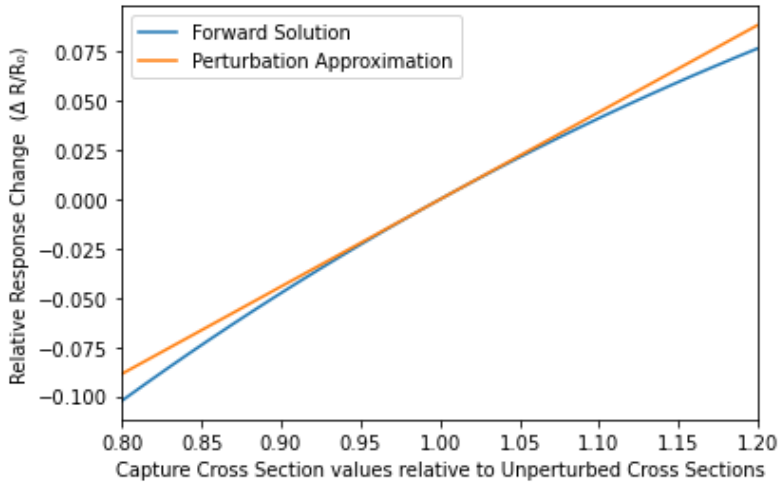

Figure 15: The relative radiotoxicity response as a function of the change in the capture cross sections. Each capture cross section is modelled to have the same perturbation relative to its original cross section.

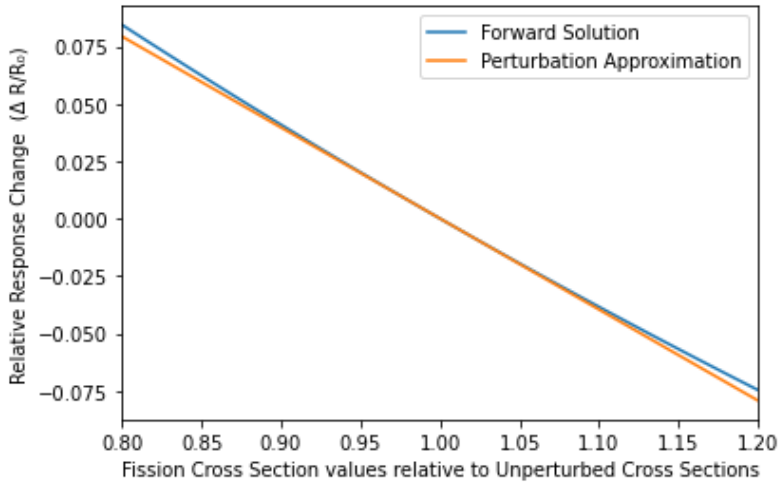

Figure 16: The relative radiotoxicity response as a function of the change in the fission cross sections. Each fission cross section is modelled to have the same perturbation relative to its original cross section.

It is interesting to see that the perturbation approximations can accurately estimate the response due to a perturbation in the capture and fission cross section seperately. When these two effects are combined, the approximation becomes much more erroneous, due to the opposite effects of these capture and fission cross section perturbations on the response.

6 Conclusions

In this thesis, the perturbation theory has shown to work. Perturbations are introduced in the initial densities, neutron flux, actinide extraction rate, and the capture & fission cross sections.

The response resulting from a perturbation in the initial nuclide densities can be precisely calculated from the perturbation theory. This is due to the linearity of the result. Perturbation theory results in a linear approximation, so if the results are linear, the exact results and the approximations coincide.

The perturbation method can also be useful to calculate the response difference as a result of a perturbation in the neutron flux, as long as the perturbed flux isn't significantly higher than the unperturbed flux.

The perturbation method also works for systems where fractions of the actinides are filtered out of the system over time, as long as the extraction doesn't occur too fast. This method is not recommended for systems where at least half of a certain nuclide is removed in less than 1000 days.

Perturbations in cross sections do not give accurate results for a response difference. This is due to the intertwining of a perturbation in both the capture and the fission cross sections simultaneously, which both have an opposite effect on the response. A perturbation in these cross sections separately therefore do give accurate response results.

In order to use these methods for useful simulations, additional work is needed to make the models more accurate.

A more comprehensive neutron flux model is needed for more accurate results. For example, instead of a constant neutron flux, the model should simulate a flux that depends both on position and time, in accordance with the processes and reactions that occur in the reactor. Also, the neutrons have a distribution of energies. This is important, not only for the flux, but also for the cross sections that depend on this energy distribution.

In many of the simulations, the only simulated nuclides were actinides. Many fission products, including the poison xenon-135, can have a significant effect on the workings of a reactor. A more realistic system that keeps track of the buildup and behavior of fission products is therefore needed for more accurate results.

Finally, thermodynamics, fluid mechanics, and heat transfer have to be taken into account for accurate simulations, since these disciplines are of paramount importance in the workings of a reactor core.

A References

[1] J. Duderstadt, L. Hamilton, *Nuclear Reactor Analysis.* Wiley, ISBN: 978-0-471-22363-4, January 1976.

[2] *United States Nuclear Regulatory Commission https://www.nrc.gov/reading-rm/basic-ref/glossary/criticality.html.* Retrieved: December 2020.

[3] *Wikipedia https://en.wikipedia.org/wiki/Fission_product_yield.* Retrieved: October 2020.

[4] J. L. Kloosterman, *Activities in Euratom projects SAMOFAR and SAMOSAFER https://msrworkshop.ornl.gov/wp-content/uploads/2020/11/27_Kloosterman_MSR_-workshop_20201.pdf.*

[5] D. Journée, *Helium bubbling in a Molten Salt Fast Reactor.* Master of Science Thesis, Delft University of Technology, Januari 2014.

[6] J. L. Kloosterman, *personal communication, October 5, 2020.*

[7] A. Gandini, *Time-Dependent Generalized Perturbation Methods for Burn-up Analysis.* RT/FI (75) 4, Comitato Mazionale per l'Energia Nucleare, Roma, 1975.

[8] A. Gandini, M Salvatores, L. Tondinelli, *New Developments in Generalized Perturbation Methods in the Nuclide Field.* Nuclear Science and Engineering, 62:2, 339-345, DOI: 10.13182/NSE77-A26970, 18 May 1976.

[9] J. M. Kallfelz, G. B. Bruna, G. Palmiotti, M. Salvatores, *Burnup Calculations with Time-Dependent Generalized Perturbation Theory.* 1977.

[10] D. Lathouwers, *personal communication, November 11, 2020.*

[11] Y. Ronen, *CRC handbook of nuclear reactors calculations.* Vol III. United States: CRC Press, Inc., 1986.

[12] J. L. Kloosterman, *Actinide Data for Transmutation Studies.* Energy Research Foundation February 1995 ECN-I–94-056.

[13] *https://wwwndc.jaea.go.jp/cgi-bin/FPYfig.* Japan Atomic Energy Agency, Nuclear Data Center, Retrieved: October 2020.

[14] J. L. Kloosterman, *Program BIFTOX, A Tool to Calculate the Radiotoxicity of a Nuclide Mixture.* ECN nuc-rfa-96-14 1996.

B Appendix

B.1 Used Data

Initial Composition

A reference design of the MSFR has an initial core composition of LiF-ThF4-UF4 (77.5-20-2.5 mol%)[4]. Fuel salt density 4.1 g/cm^3. Average weight of fuel salt is 89.562 g/mol.

Table 1: Initial Composition MSFR in various units

Particles	Comp. (mol)	mass (u or g/mol)	Comp. (mass)	g/cm^3	mol/cm^3	N/(barn cm)
LiF	77.5 %	6.94+19.00=25.94	22.47 %	0.921	0.0355	0.0214
ThF4	20 %	232.04+4(19.00)=308.04	68.88 %	2.824	0.0092	0.0056
UF4	2.5 %	233.04+4(19.00)=309.04	8.64 %	0.354	0.0011	0.0007
Total	100%	89.44	100%	4.1	0.0458	0.0277

Actinides

The data from the used actinides can be seen in table 2. This data includes the decay rate, and the neutron cross sections for the different reactions. The decay values correspond with the half-life of the nuclides in days.

Table 2: Actinide Decay & One-Group Cross Sections LMR-MOX [12]

Nuclides	Decay (days)	σ_c (*barn*)	σ_f (*barn*)	σ_{2n} (*mbarn*)
Th231	1.063	/	/	/
Th232	/	0.396	0.011	2.234
Th233	0.016	/	/	/
Pa231	/	2.980	0.258	1.479
Pa232	1.312	/	/	/
Pa233	26.975	1.056	/	0.612
Pa234	0.279	/	/	/
Pa235	0.017	/	/	/
U232	/	0.662	2.264	1.117
U233	/	0.261	2.774	1.137
U234	/	0.602	0.349	0.222
U235	/	0.535	1.906	1.490

*The missing values are negligibly small.

Fission Yield

The fission yield data from the fissionable actinides is retrieved from the website of the Nuclear Data Center of Japan [13]. A figure of the fission yields of this data can be seen below. With lower neutron energy, the fission yield exhibits a more distinct 'camel hump'.

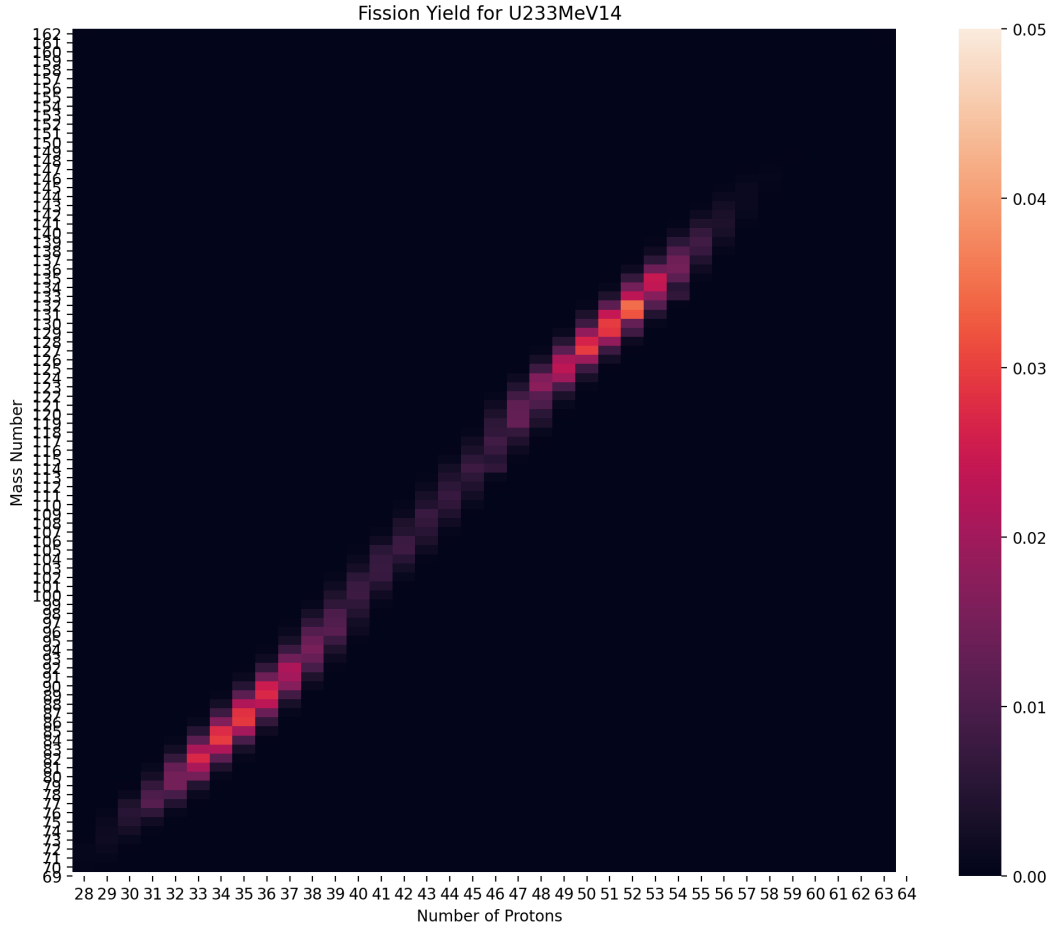

Figure 17: Fission yields of U233 with fast neutrons (14 MeV).
Decay Product: proton number on x-axis , mass number on y-axis.
Created with data from the Nuclear Data Center of Japan [13].

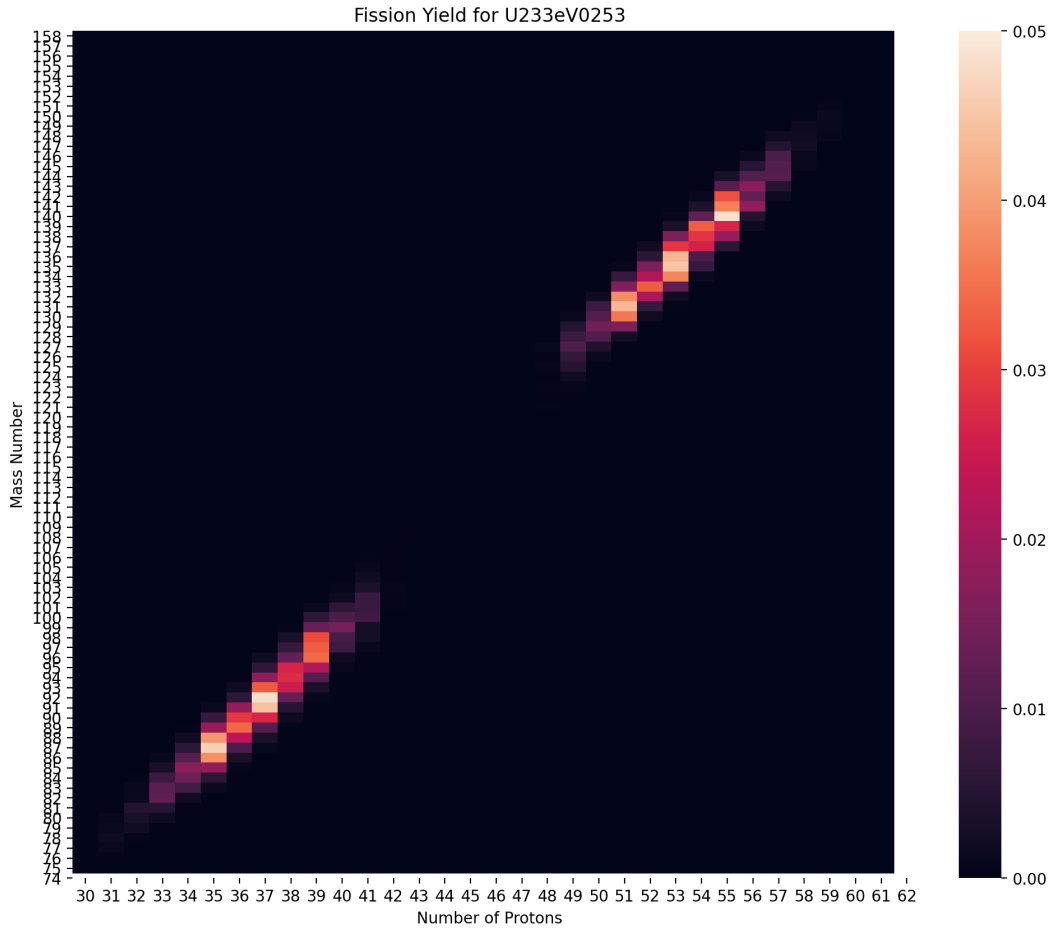

Figure 18: Fission yields of U233 with slow neutrons (0.253 eV).
Decay Product: proton number on x-axis , mass number on y-axis.
Created with data from the Nuclear Data Center of Japan [13].

The yields for the used poisons for different fission products are put in table 3 below. The numbers are in parts per thousand of total fission.

Table 3: Fission yields as used in the simulation.

Poisons\Yields (10^{-3})	Th232	Pa231	U232	U233	U234	U235
54 Xenon 135	1.39	6.57	17.3	18.2	18.4	14.6
54 Xenon 136	6.53	21.6	45.6	28.5	34.4	21.8
52 Tellurium 135	24.5	19.3	6.79	5.20	5.84	10.5
53 Iodide 135	22.8	40.2	28.3	26.5	29.0	31.5
55 Cesium 135	0	0	0	1.63	1.45	0

Fission Product Data

In table 4 the half-life of each used fission product is listed.

Table 4: Simulated poisons, and corresponding half-life.

Poisons	$\beta^- Decay$ (days)
54 Xenon 135	0.380
54 Xenon 136	stable
52 Tellurium 135	≈ 0
53 Iodide 135	0.274
55 Cesium 135	approx. stable
Lanthanides	Varying

Some of the poisons can be extracted from the reactor during operations. Others remain in the fuel salt until the reactor fuel gets recycled. In figure 4 the possibility of removal during operation of the elements can be seen.

Radiotoxicity values

Table 5: Radiotoxicity of radiotoxic actinides in the thorium fuel cycle. The units are both in sievert per mole, and sievert per nuclide density in a $9m^3$ reactor [14]

Nuclide	Radiotoxicity (Sv/mole)	Radiotoxicity (Sv/(barn cm^2))
Th232	$7.24*10^{-1}$	$1.08*10^{7}$
Pa231	$4.20*10^{5}$	$6.28*10^{12}$
Pa233	$4.20*10^{3}$	$6.28*10^{10}$
U232	$8.08*10^{7}$	$1.21*10^{15}$
U233	$4.20*10^{3}$	$6.28*10^{10}$
U234	$2.64*10^{3}$	$3.95*10^{10}$
U235	$8.74*10^{-1}$	$1.31*10^{7}$

The conversion factor from 1/mole to 1/(barn cm^2) is $1.495 * 10^7$ mole/(barn cm^2)

B.2 Additional Results

Flux Adjusted to Power

In figure 19, the densities of the actinides are shown over time, when the neutron flux is made 50 times higher than the estimated flux.

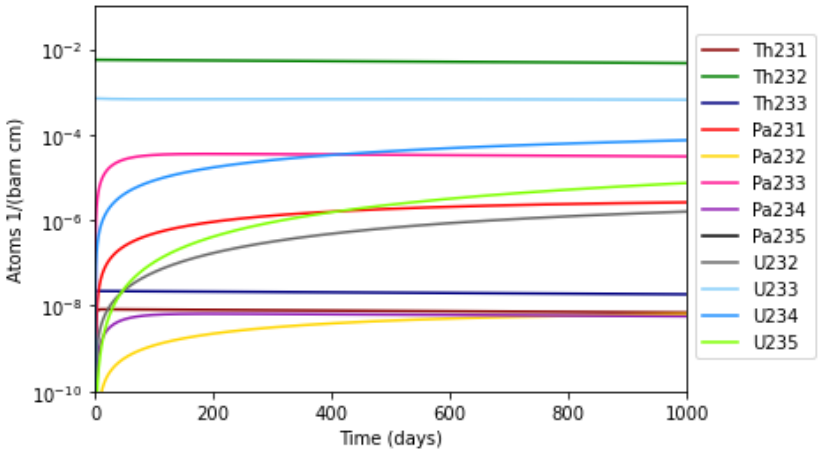

Figure 19: Densities of actinides with an increase in the neutron flux, to get the 3GW reactor power.

Notice a stark increase in the non-initial actinide densities with respect to the original densities in figure 6. The corresponding power generation is plotted in the figure below.

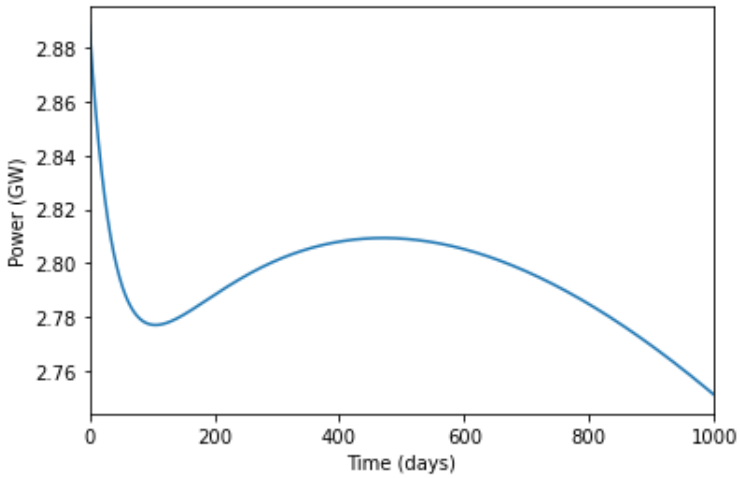

Figure 20: Power generation of simulated reactor with a artificially high flux. The power decreases after around 500 days due to the depletion of the actinides.

Now the power is close to the expected 3GW. Notice how the power in figure 20

decreases after 500 days, unlike the power generation of figure 7. This is due to the depletion of the actinides in the reactor core.

Additional Importance Functions

In figure 21, the same importance functions as in figure 9 are plotted. In this plot, the flux is increased 50 times to adjust for the 3GW reactor power. There are noticable differences between the two plots. For example, the importance function of Pa-235 now has a discernible slope.
In figure 22, U-232 is included as a response to the simulation with the lower flux.

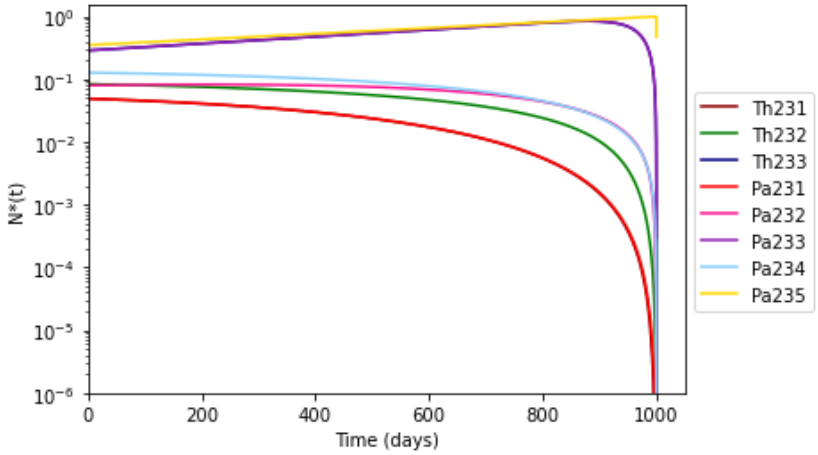

Figure 21: Importance functions of thorium and protactinium on the production of fissile uranium isotopes U-233 and U-235 with the flux adjusted to the reactor power.

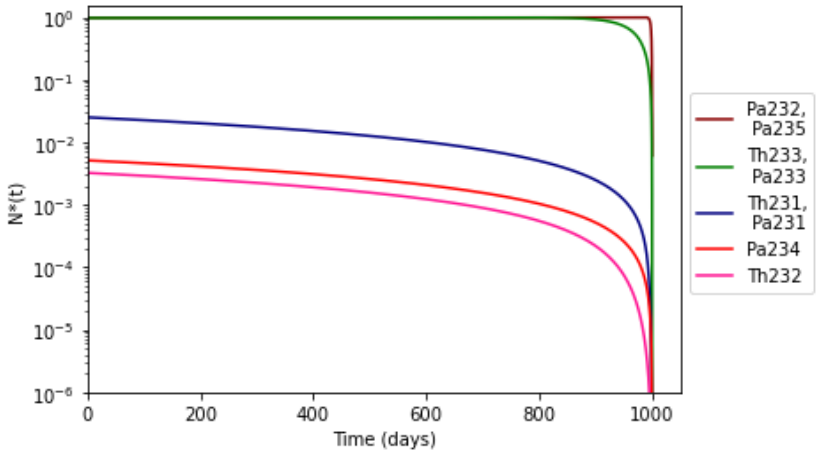

Figure 22: Importance functions of thorium and protactinium nuclides on the production of fissile uranium isotopes U-232, U-233 and U-235.

In figure 23, the importance functions of uranium isotopes are shown, with final adjoint values of plutonium-239 and plutonium-240 being 1. The nuclear data of a pressurized water reactor is used.

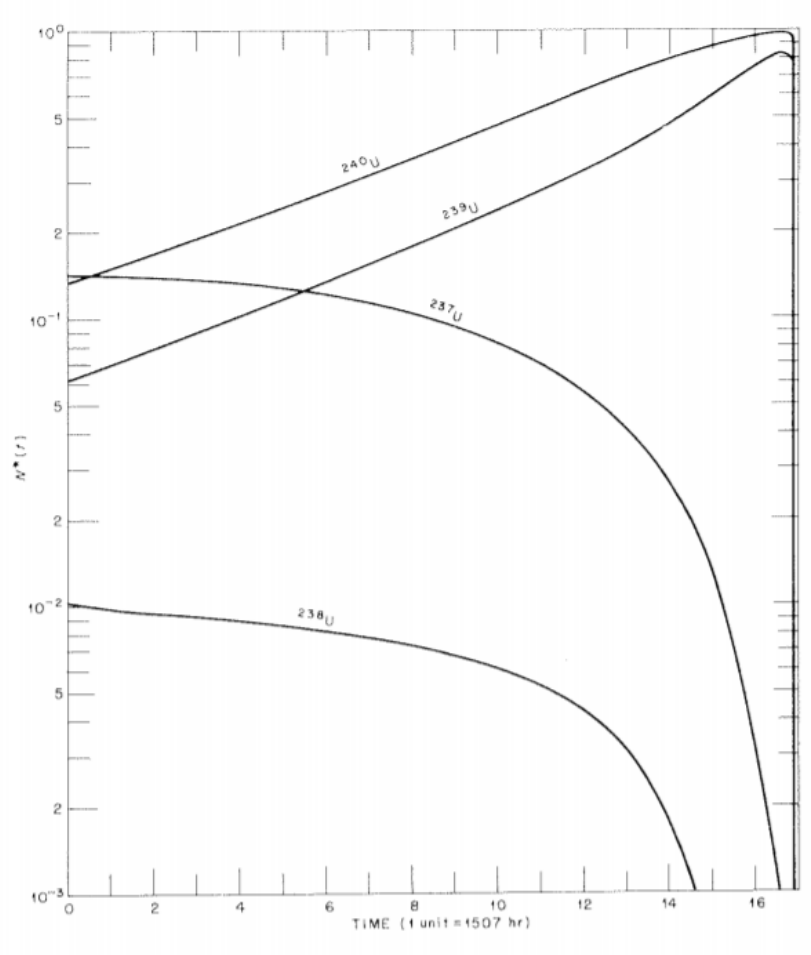

Figure 23: Uranium importance functions for plutonium production responses from the handbook of Williams[11].

Additional Perturbation Results

See figure 24 for the relative radiotoxicity response as a function of the initial density in Th232 atoms, where the flux is not adjusted to the power output of a MSFR. Although the approximation method is still exact, the relative response rate is significantly lower, due to a lower conversion rate of Th-232 into radiotoxic actinides.

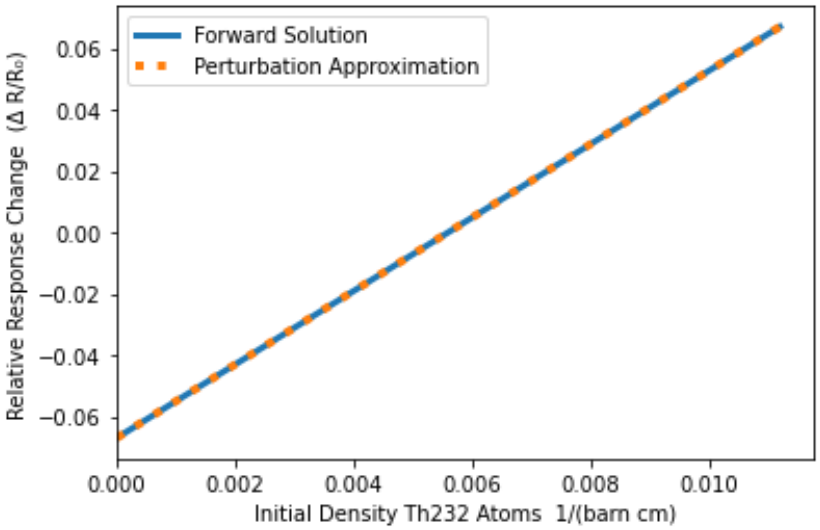

Figure 24: The relative radiotoxicity response as a function of the initial density in Th232 atoms. The unperturbed system has a density of 0.0056 atoms $b^{-1}cm^{-1}$, and the neutron flux is 10^{14} $cm^{-2}s^{-1}$