NUCLEAR FUEL CYCLE MODELING APPROACHES FOR RECYCLING AND TRANSMUTATION OF SPENT NUCLEAR FUEL

THESIS

Presented in Partial Fulfillment of the Requirements for the
Degree Master of Science in the Graduate
School of The Ohio State University

By

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*****

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2008

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ABSTRACT

Policy decisions regarding the direction of nuclear power utilize sophisticated fuel cycle models. Both static and dynamic models are used to determine how a proposed technology will integrate into existing nuclear fuel cycle strategy. Dynamic models in particular are useful because they can better evaluate what-if scenarios. However, some of these models rely on static fuel recipes for loading recycled nuclear fuel. This static approach will lead to considerable uncertainty because it cannot capture the dynamic changes in separated inventory. Thus a better approach for loading recycled fuel is necessary. This thesis presents a better approach to load recycled fuel dynamically in an attempt to best match an available composition to a target composition by employing a novel isotopic reactivity worth value assignment. Once fuel is loaded and consumed in reactors, the resultant spent fuel composition can be determined by utilizing the transmutation approach explained in this thesis. These two approaches allow for modeling of dynamic fuel recycling. This thesis describes these approaches and presents illustrative results that can be incorporated into larger nuclear fuel cycle models.
Acknowledgments

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SPECIAL NOMENCLATURE

LWR- Light Water Reactor
FR- Fast Reactor
ADS- Accelerator Driven System
VISION- Verifiable Fuel Cycle Simulation
U- Uranium
TRU- Transuranics
MA- Minor Actinides
$k_B$- Boltzmann’s constant, 8.6273x$10^{-5}$ eV/K
$\eta$- Neutron reproduction factor
$f$- Neutron utilization factor
$\epsilon$- Neutron (fast) fission factor
$p$- Neutron resonance escape probability
$\gamma$- Average number of free neutron released per fission
$P_{\text{thl}}$- Probability of thermal neutron non-leakage
$P_{\text{fl}}$- Probability of fast neutron non-leakage
$P_{	ext{nll}}$- Probability of neutron non-leakage
$\phi$- Neutron flux (#/cm$^2$/s)
$\Sigma_f$- Macroscopic neutron fission cross section (cm$^{-1}$)
$\Sigma_c$- Macroscopic neutron radiative capture cross section (cm$^{-1}$)
$\sigma_f$- Microscopic neutron fission cross section (cm$^2$)
$\sigma_c$- Microscopic neutron radiative capture cross section (cm$^2$)
$\gamma$- Fission yield
$k$- Neutron multiplication factor ($k_{\text{eff}}$ or $k_\infty$)
$L^2$- Diffusion area (cm$^2$)
$B_g^2$- Geometric Buckling (cm$^2$)
$M^2$- Migration area (cm$^2$)
$\rho$- Reactivity worth value
$M_{\text{U}}$- Uranium group mass fraction
$M_{\text{TRU}}$- Transuranic group mass fraction
$m_{\text{U}}$- Uranium isotopic mass fraction
$m_{\text{TRU}}$- Transuranic isotopic mass fraction
CHAPTER 1

INTRODUCTION

Providing clean, sustainable energy is the single most important problem facing society today. With increasing awareness of the adverse impact of global warming (Robinson et al., 2007), utilities are turning to alternative energy technologies to reduce CO₂ emissions and reduce humanity’s dependence on fossil fuels. Solar, wind, geothermal, and hydroelectric sources are attractive options for domestic use; however, these alternatives are not capable of sustaining industry’s large demand for power. Nuclear power is the only short and midterm solution to this problem (Massachusetts Institute of Technology, 2003) and in response, the US nuclear power industry is currently experiencing a renaissance as plans exist to build as many as 28 new nuclear power plants (104 plants are currently in operation in the U.S.) in the near future (American Nuclear Society, 2008). This resurgence of nuclear power will influence all aspects of the nuclear fuel cycle and this overall impact on fuel cycle is largely unknown.
1.1 Nuclear Fuel Cycle Overview

The nuclear fuel cycle can be divided into three parts: (1) the front-end, (2) the reactors, and (3) the back-end. The front-end of the fuel cycle focuses on the how raw material is processed to produce fuel for reactors. Reactors then use that fuel to generate power. Finally, the back-end deals with the byproducts following fuel irradiation (namely used or spent nuclear fuel) and how they are disposed or recycled. Each of these aspects of the fuel cycle has its own inherent challenges.

1.1.1 Front-end

For example, the front-end process begins with mining and milling raw ore. Most reactors utilize the uranium fuel cycle; however, alternative strategies which rely on the thorium fuel cycle exist. In the uranium fuel cycle, uranium ore is mined. This ore, however, varies in grade from 0.03 to 0.5% uranium (Chochran and Tsoulfanidis, 1999) with some high grade at 2% uranium (Shropshire et al., 2007). One of the major concerns facing the front-end of the fuel cycle is the abundance of uranium resources. While uranium, as shown in Table 1.1 (Mason, 1966), is more abundant in the earth’s crust than silver or gold, the grade of uranium ore may not be sufficient to make mining a specific resource economically competitive. It is for this reason that one of the major concerns facing the nuclear power industry is the amount of available uranium resources at the current market price. As the price of uranium increases, additional sources of uranium will become economically competitive (Shropshire et al., 2007).
<table>
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<tbody>
<tr>
<td>Gold</td>
<td>0.004</td>
</tr>
<tr>
<td>Silver</td>
<td>0.07</td>
</tr>
<tr>
<td>Uranium</td>
<td>1.8</td>
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<td>7</td>
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<td>Lead</td>
<td>13</td>
</tr>
<tr>
<td>Copper</td>
<td>55</td>
</tr>
<tr>
<td>Iron</td>
<td>50,000</td>
</tr>
<tr>
<td>Aluminum</td>
<td>81,300</td>
</tr>
</tbody>
</table>

Table 1.1: Abundance of material in the earth's crust for selected elements (Mason, 1966).

Once the uranium ore has been mined it undergoes a process known as milling where the mineral form of uranium is transformed into yellow cake (U$_3$O$_8$). Numerous mineral forms of uranium exist with some prominent forms being uraninite (UO$_2$) and uranophane (Ca(UO$_2$)$_2$SiO$_3$(OH)$_2$-5H$_2$O) (Chochran and Tsoulfanidis, 1999). The milling process basically consists of crushing the ore and chemically processing (leaching, ion exchange, or solvent extraction) the minerals to extract the U$_3$O$_8$. The byproducts of this process, known as the mill tailings, are of additional environmental and health concern as radioactive uranium decay daughters, such as radium, are naturally present in the acidic byproducts. Like most ore extraction processes, the chemical nature of these byproducts is of concern, but with uranium milling, additional radiological challenges are present.

Once the U$_3$O$_8$ has been extracted it must undergo a conversion and enrichment process. The U$_3$O$_8$ is first converted into uranium hexafluoride (UF$_6$). To do this, U$_3$O$_8$
is baked and reduced to uranium dioxide (UO$_2$) in the presence of hydrogen. The UO$_2$ then undergoes a hydrofluorination process where hydrofluoric acid (HF) reduces the UO$_2$ to uranium tetrafluoride (UF$_4$). UF$_4$ is then introduced to fluorine gas (F$_2$) where it is finally converted into UF$_6$ (Chochran and Tsoulfanidis, 1999).

Once in this form, the UF$_6$ undergoes an enrichment process. Naturally occurring uranium consists of the isotopes shown in Table 1.2 (KAPL and Lockheed-Martin, 2002). $^{235}$U is the primary fissile isotope present in many nuclear fuels and these fuels are nominally enriched to 3-5 wt% $^{235}$U with the remaining uranium consisting of fertile $^{238}$U. For reference, fissile material is that which can easily sustain fission reactions and fertile material is that which can be then converted into fissile material following neutron absorption. Conventional enrichment processes consist of centrifugal enrichment, gaseous diffusion enrichment, or laser isotope separation. All of these processes rely on the fact that $^{235}$U is lighter since it contains 3 fewer neutrons than the more abundant $^{238}$U. Each of these enrichment processes requires several energy intensive stages with their own technological challenges. For example, gaseous diffusion requires large amounts of energy, centrifuge enrichment suffers from mechanical failures, and laser enrichment has yet to be proven as a viable industrial technique.

<table>
<thead>
<tr>
<th>Uranium Isotope</th>
<th>Nominal Abundance (atom %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{234}$U</td>
<td>0.0055</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>0.7200</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>99.2745</td>
</tr>
</tbody>
</table>

Table 1.2: Abundance of uranium isotopes in naturally occurring sources (KAPL and Lockheed-Martin, 2002).
However, once UF₆ is enriched to the proper isotopic ratio, it undergoes another chemical process before being made into fuel. The UF₆ gas is first bubbled through water to produce UO₂F₂ before ammonia is introduced to cause the uranium to precipitate out of solution as ammonium diuranate ((NH₄)₂U₂O₂, also known as ADU). This precipitate is then dried at high temperatures into U₃O₈ before being reduced with hydrogen to UO₂. The UO₂ is then ground, pressed, and sintered into pellets to be used as fuel (Chochran and Tsoulfanidis, 1999).

These pellets are then typically loaded into cylindrical tubes creating fuel rods (or pins). The cylindrical tubes surrounding the pellets are known as fuel cladding and are typically made from zircaloy. These fuel rods are then assembled into fuel assemblies (or bundles) as an array containing typically 9x9 or 8x8 assemblies for a boiling water reactor (BWR) or 15x15, 16x16, or 17x17 assemblies for a pressurized water reactor (PWR). Many of these fuel assemblies are then loaded into the core of a reactor where the assemblies provide fuel for the sustained fission reaction.

1.1.2 Reactors

To initiate a sustained fission reaction, a material that undergoes spontaneous fission is assembled close to fissile material. The neutrons following spontaneous fission then cause fission in the fissile material, which in turn generate more neutrons. Following fission, energy is released generally in the form of kinetic energy of resulting fission particles. The resulting neutrons from fission are of keen interest as their kinetic energy is harnessed as heat, which is eventually used to generate steam to drive a turbine and generate power. To do this, the neutrons experience inelastic collisions with a
moderator material. This interaction is referred to as moderation. For most conventional reactors (BWRs and PWRs), the moderator is water, which also serves as the heat transfer fluid. The moderator serves to slow down the neutrons, which makes (in the case of thermal reactors) additional fission more likely at lower neutron energies (thermal energies). Other reactors (known as fast reactors), however, utilize neutrons that experience little to no moderation.

Following convention, the neutron population is described by a neutron multiplication factor (denoted as $k_{\text{eff}}$). In the case of a sustained fission reaction at a prescribed thermal power level, on average the number of neutrons that are lost to cause fission equal the number of neutrons generated following fission. In this situation, the reactor is called critical ($k_{\text{eff}}=1$). Similarly, supercritical ($k_{\text{eff}}>1$) refers to when more neutrons are generated than lost and subcritical ($k_{\text{eff}}<1$) refers when fewer neutrons are generated than lost. In addition, higher power levels can be achieved by allowing the reactor to become supercritical, thus increasing the number of neutrons (and heat generated), before returning to critical operation ($k_{\text{eff}}=1$) for that higher power level. Conversely, decreasing the power level can be achieved by bringing the reactor subcritical before returning to a critical operation for that lower power level. The fission reaction is then controlled by elements known as control rods. These control rods absorb neutrons thus lowering the neutron population and hindering fission. Only a few control rods need to be present to control (or stop) the fission reaction. Because of this, the cores tend to be symmetrical with one-eighth or one-quarter symmetry for cylindrical geometries.
While utilizing symmetry attempts to ensure uniform conditions throughout the core, the neutron flux experiences strong geometry dependence since neutrons that leave the active core volume are not likely to return. Therefore, each fuel assembly (and each pin) will experience a different neutron flux depending upon its location in the reactor. For this reason, fuel management is of primary importance. The goal behind fuel management is to extract as much energy from each assembly as possible. This parameter is commonly reported as burnup in units of thermal energy produced per amount of material. The burnup parameter varies for each reactor but for perspective 33 gigawatt-days per (metric) tonne of uranium (GWd/MTU) is an appropriate nominal value for current reactors (Salvatores et al., 2003). However, there are means as well as subsequent challenges with increasing the burnup.

In order to increase the burnup and thus the fuel economy, fuel assemblies are shuffled within the reactor during refueling outages. In other words, the fuel assemblies are relocated to another position within the core during refueling. Refueling typically takes place every 18 months and fuel assemblies are likely to experience 3 cycles before they are discharged. When shuffling the fuel, reactor-specific algorithms are used to determine the configurations. Several challenges are present in this part of the fuel cycle and new approaches are constantly being developed (Thomas, 2005).

After irradiation in a reactor, the present approach is to discard the fuel. This approach is known as the once-through approach. Following discharge, the fuel is then placed in fuel storage pools for an extended period of time where it is allowed to undergo radioactive decay and cool. These pools are literally filled with water to facilitate heat
transfer and fuel assemblies are separated sufficiently far apart from one another to prevent a sustained fission reaction. This storage is known as wet storage and after cooling for a sufficient time, the radioactive fuel assemblies are moved to casks for dry storage.

1.1.3 Back-end

At this point, the fuel cycle transitions to the back-end. At present, there is no concrete plan to handle the used nuclear fuel. The intent is to place all the used fuel in a geological repository (i.e., Yucca Mountain) but this is yet to become a reality. However, several options exist where this used nuclear fuel can be recycled. Recycling (also known as reprocessing) is one method that can help to close the nuclear fuel cycle.

Typical used nuclear fuel generated by a commercial power plant is composed primarily of reusable uranium with trace amounts of minor actinides (Np, Am, and Cm), and long-lived fission products, as listed in Table 1.3 (World Nuclear Association, 2007).

<table>
<thead>
<tr>
<th>Composition</th>
<th>Weight Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium Total</td>
<td>95.6%</td>
</tr>
<tr>
<td>$^{232}\text{U}$</td>
<td>0.1-0.3%</td>
</tr>
<tr>
<td>$^{233}\text{U}$</td>
<td>0.1-0.3%</td>
</tr>
<tr>
<td>$^{234}\text{U}$</td>
<td>0.4-0.7%</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>0.5-1.0%</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>93.3-94.5%</td>
</tr>
<tr>
<td>Stable Fission Products</td>
<td>2.9%</td>
</tr>
<tr>
<td>Plutonium</td>
<td>0.9%</td>
</tr>
<tr>
<td>$^{135}\text{Cs}$ and $^{90}\text{Sr}$</td>
<td>0.3%</td>
</tr>
<tr>
<td>$^{129}\text{I}$ and $^{99}\text{Tc}$</td>
<td>0.1%</td>
</tr>
<tr>
<td>Minor Actinides</td>
<td>0.1%</td>
</tr>
<tr>
<td>Other Long-lived Fission Products</td>
<td>0.1%</td>
</tr>
</tbody>
</table>

Table 1.3: Composition of a typical commercial nuclear power plant’s used nuclear fuel (World Nuclear Association 2007).
This used fuel can then be separated and some isotopes can be reused as fuel. Fuel separation first involves taking the used nuclear fuel rods and chopping these rods into manageable segments (1 or 2 centimeter long). Once physically dismantled, a separation process is used to extract the reusable isotopes. The separation process can be categorized as pyroprocessing or aqueous reprocessing with further subsets in each of these categories.

Pyroprocessing is a form of electro-refining where material is first removed from the cladding before being dissolved into a molten halide salt. Electro-refining then extracts various substances on the cathode. A low-carbon steel shaft is used as the cathode for uranium extraction and a cadmium cathode is used for plutonium-uranium extraction. The selective deposition associated with each cathode is governed by the chemistry of the halide salt. This option is attractive as the transuranics commonly accompany the uranium and plutonium. These transuranics are typically highly radioactive and present health risks. However, this risk is a proliferation deterrent and discourages tampering with separated material. In addition, the remaining halide salt consists mostly of fission products that undergo radioactive decay quicker than the transuranics. In this manner, the long-lived fission products are recycled and consumed rather than being left in the waste. From a waste standpoint, this is desirable since the waste would become even less radiotoxic over shorter times (Chochran and Tsoulfanidis, 1999).

Aqueous reprocessing, on the other hand, is a chemical process where fuel is first separated from the cladding by dissolving the used nuclear fuel in a nitric acid solution.
Next, different solvents are used to extract various streams of material. Due to the complexity of actinide chemistry, several solvent extraction processes exist, each with a slight variation to extract the isotopes of interest. Two primary subcategories are the plutonium-uranium extraction process (PUREX) and the uranium extraction process (UREX). Both processes rely on oxidation-reduction reactions and liquid-liquid extraction techniques as the primary mechanisms for extraction (Chochran and Tsoulfanidis, 1999). The major difference between in the PUREX and UREX process involves the separation of plutonium. In the PUREX process, plutonium is extracted by itself. This is a proliferation concern as plutonium could be used for weapon-related purposes. In the UREX process, plutonium is never in a form by itself; other transuranics are always present. This is a strong deterrent for proliferation as weapons development becomes more difficult with the added radioactivity of the transuranics. It is for this reason that UREX processes tend to be more favorable.

Several variations of the UREX process exist. Each of these processes has different product streams depending on the fuel cycle interest. Table 1.4 provides an example of these variations known as the UREX+ processes (Pereira, 2008).
Table 1.4: Product streams for the UREX+ processes consisting of the primary elements (such as uranium (U), technetium (Tc), cesium (Cs), and strontium (Sr)), the transuranic elements (TRU) (such as plutonium (Pu), neptunium (Np), americium (Am), and curium (Cm), the lanthanides (Ln), and fission products (FPs). (Iodine is typically removed prior in the dissolution process)

<table>
<thead>
<tr>
<th></th>
<th></th>
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<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>UREX+1</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>TRU/Ln</td>
<td>FPs</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>UREX+1a</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>TRU</td>
<td>Ln/FPs</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>UREX+1b</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>U/TRU</td>
<td>Ln/FPs</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>UREX+2</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>Pu/Np</td>
<td>Am/Cm/Ln</td>
<td>FPs</td>
<td>-</td>
</tr>
<tr>
<td>UREX+2a</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>U/Pu/Np</td>
<td>Am/Cm/Ln</td>
<td>FPs</td>
<td>-</td>
</tr>
<tr>
<td>UREX+3</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>Pu/Np</td>
<td>Am/Cm</td>
<td>Ln/FPs</td>
<td>-</td>
</tr>
<tr>
<td>UREX+3a</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>U/Pu/Np</td>
<td>Am/Cm</td>
<td>Ln/FPs</td>
<td>-</td>
</tr>
<tr>
<td>UREX+4</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>Pu/Np</td>
<td>Am</td>
<td>Cm</td>
<td>Ln/FPs</td>
</tr>
<tr>
<td>UREX+4a</td>
<td>U</td>
<td>Tc</td>
<td>Cs/Sr</td>
<td>U/Pu/Np</td>
<td>Am</td>
<td>Cm</td>
<td>Ln/FPs</td>
</tr>
</tbody>
</table>

The various streams would then either be discarded as waste or recycled in reactors. The streams to be recycled are those streams containing uranium (U) and the transuranics (TRUs, namely plutonium, neptunium, americium, and curium).

It is envisioned that the TRUs can be recycled in light water reactors (LWRs), fast reactors (FRs), or in particle accelerator driven systems (ADSs). This recycling would consist of using TRUs as fuel while consuming (through transmutation) other less desirable TRUs. The basic concept is to expose TRUs to a neutron flux to induce fission in the fissile isotopes and, through neutron absorption and subsequent decay, reduce the quantity and radiotoxicity of the remaining TRUs, which tend to be predominantly fertile isotopes. This concept will be discussed in more detail later in Chapter 2.

Following separation, a small fraction of waste is present. This waste would primarily consist of technetium, iodine, cesium, strontium, and other fission products as
shown in Table 1.3. This comprises only approximately 3.4% of the fuel while the other 96.6% can be recycled. The remaining waste would then be disposed in a geological repository (i.e., Yucca Mountain, (US Department of Energy, 2002)). Preliminary studies indicate that recycling before disposal could significantly increase geological repository loading (Wigeland et al., 2006), that is more energy extracted for the same repository footprint. Namely, Cs and Sr removal alone can decrease waste by as much as 5.7 times, and combined Cs, Sr, and actinide removal can decrease waste by as much as 225 times (as will be described later).

Ultimately the fuel cycle (even a closed fuel cycle) has an end. Some amount of waste will not be recycled and reused due to economic viability, technological challenges, or because doing so would pose too much of a health threat to workers. In every fuel cycle, some amount of waste will be generated. However, the goal behind a well functioning closed fuel cycle is to minimize the amount of waste in order to reduce the volume, heat, and radiotoxicity of the remaining waste. In order to do this, recycling is an attractive option and is experiencing intense investigation.

1.2 Recycling and Transmutation Concept

The recycling concept relies on re-irradiating separated material. This ensures better fuel economy and waste reduction. Fuel economy is improved by utilizing isotopes that can still serve as fuel. Waste reduction is obtained by transmuting less desirable isotopes. Transmutation is the process of changing one isotope (or element) into another isotopes (or element) through neutron absorption and subsequent decay. This can be accomplished by exposing the material that is to be transmuted to a neutron
source. The source of neutrons may come from a particle accelerator driven system (ADS) or from a nuclear reactor. In a nuclear reactor, there are two dominant classes: 1) thermal reactors and 2) fast reactors. Each of these reactor types can be used for transmutation.

1.2.1 Thermal Neutrons

Thermal reactors are the type of reactors whose neutron spectrum is dominated by neutrons near the thermal energies of the surrounding material. When the energy of neutrons is randomly distributed among the available energy states for a material at a temperature, $T$, the kinetic energy of the neutron corresponding to the most probable speed is given in eq. (1.1):

$$E_T = k_B T$$  \hspace{1cm} (1.1)

where $k_B$ is Boltzmann’s constant ($8.627343 \times 10^{-5}$ eV/K). As an example, free neutrons at room temperature ($T = 293$ K) have an energy of 0.025 eV which equates to speeds of about 2200 m/s. This is commonly referred to as the thermal speed of the neutron. Since neutron interaction (due to the moderator) can be modeled as binary inelastic collisions with nuclei, the neutron spectrum of thermal neutrons is modeled by the Maxwell-Boltzmann kinetic energy distribution function:

$$f_{MB}(\varepsilon) = \frac{2}{\sqrt{\pi (k_B T)^3}} \exp\left(\frac{-\varepsilon}{k_B T}\right)$$  \hspace{1cm} (1.2)

depicted in Fig. 1.1. Neutrons following fission are slowed down by these binary inelastic collisions with a moderator, until they eventually reach thermal equilibrium with
the surrounding material. Neutrons that undergo this process are said to have experienced moderation and are thermalized.

![Maxwell-Boltzmann distribution function for $T = 293$ K.](image)

Figure 1.1: Maxwell-Boltzmann distribution function for $T = 293$ K. The average energy is approximately 0.025 eV which corresponds to free neutrons at a speed of 2200 m/s.

Since the dominant fraction of the neutrons in a thermal reactor exists in this thermal energy range, thermal reactors are typically concerned with fission that occurs around this nominal thermal value. As a result, when studying neutron interactions in a thermal reactor, the energy range corresponding to thermal neutrons is of particular interest. Specifically, the energy range below 1 eV is considered the thermal energy range.
1.2.2 Fast Neutrons

Alternatively, fast reactors are the type of reactor whose neutron spectrum is dominated by epithermal (fast) neutrons that arise following fission but prior to slowing down through scattering in the moderator. This spectrum is dependent on the isotopes experiencing fission and weakly dependent on the energy of the neutron causing fission. Figure 1.2 depicts an adequate approximation of the fission neutron spectrum for $^{235}$U based upon the following empirical expression (Knief, 1992) reported as the energy distribution function:

$$f_{^{235}U}(\varepsilon) = 0.453 \exp(-1.036\varepsilon) \sinh\left(\sqrt{2.29}\varepsilon\right)$$  \hspace{1cm} (1.3)

Figure 1.2: Fission-neutron distribution function of $^{235}$U.
Since the majority of neutrons in fast reactors experience little moderation before they are absorbed by a nucleus, fast reactors are primarily concerned with fission that occurs above 0.1 MeV. This energy range is known as the epithermal or fast energy range.

When describing this energy range, if the spectrum is skewed to the lower energies, closer to 0.1 MeV, the spectrum is called “soft.” If the spectrum is skewed to the higher energies, closer to 10 MeV, the spectrum is called “hard.”

1.2.3 Cross Sections

Neutrons can interact with material in several ways. A neutron may undergo collision (scattering), cause fission, or it may even be captured by a nucleus. If a neutron causes fission or is captured by the nucleus, this is referred to as an absorption event. Fission causes the nucleus to split into residual isotopes and capture results in the nucleus undergoing a transition to de-energize. Typical mechanisms consist of radiative capture \((n, \gamma)\), charge-particle \([ (n, p), (n, d), (n, \alpha), (n, 2\alpha), \ldots ] \) , multiple neutron \([ (n, 2n), (n, 3n), \ldots ] \) , or charge-particle/neutron \([ (n, pn), (n,dn), \ldots ] \) interactions (Knief, 1992). In order to predict these interactions, the concept of a cross section is introduced given that the effective size of the nucleus is proportional to the probability of interaction. As a result, cross sections are given units of area. Typically, cross sections are reported in units of barns \( (1 \text{ barn (b)} = 10^{-24} \text{ cm}^2) \) where this dimension is used to represent the probability of interaction. Or in other words, the cross section represents the ratio of the number of neutrons that cause a particular event to the number of incident neutrons on a target.

Since neutrons (and target nuclei) are energy dependent, these cross sections are also energy dependent. The energy spectrum of nuclear cross sections are divided into
three distinct regions: the thermal region (less than 1 eV), the resonance region (between 1 eV and $10^5$ eV), and the epithermal (fast) region (great than $10^5$ eV). For many isotopes, radiative capture becomes just as likely as fission in the resonance region (low resonance escape probability). Because of this, many neutrons do not survive the slowing down process to become thermal neutron. This is known as resonance capture.

Figures 1.3 and 1.4 illustrate the radiative capture cross section and the fission cross section for $^{235}$U and $^{238}$U, respectively (extracted from the Joint Evaluated Fission and Fusion (JEFF-3.1) database), as a function of energy. Note that the fission cross section for $^{235}$U is greater than the capture cross section. This indicates the fissile nature of $^{235}$U. Also note that only at high neutron energies does fission become more likely than radiative capture for $^{238}$U. It is for these reasons, that $^{235}$U is used as the primary fuel in thermal reactors as $^{238}$U is likely to transmutate into $^{239}$Pu (following neutron capture and subsequent double beta decay) in thermal reactors.
Figure 1.3: Fission ($\sigma_f$, red) and radiative capture ($\sigma_c$, green) cross sections for $^{235}$U at 293 K extracted from the JEFF-3.1 database. Note that $^{235}$U is more likely to fission than experience radiative capture over most of the energy range of interest. This illustrates that $^{235}$U is a fissile isotope.
Figure 1.4: Fission (\(\sigma_f\), red) and radiative capture (\(\sigma_c\), green) cross sections for \(^{238}\)U at 293 K extracted from the JEFF-3.1 database. Note that \(^{238}\)U is more likely to experience radiative capture than experience fission over most of the energy range of interest. This illustrates that \(^{238}\)U is a fertile isotope as it will be transmutated into fissile \(^{239}\)Pu.

With regard to recycling and transmutation, two primary contributors to radiotoxicity present in conventional used nuclear fuel are \(^{241}\)Am and \(^{238}\)Pu. The fission and radiative capture cross sections for these two isotopes can be found Figs. 1.5 and 1.6. In thermal reactors, radiative capture is more likely to occur for these isotopes. With respect to recycling these isotopes as fuel (fissile isotopes), this is not practical in a thermal reactor (unlike in a fast reactor) because the radiative capture cross section
dominates the fission cross section, that is, they are both fertile isotopes. However, thermal reactors can still be used to perform transmutation. Instead of relying on fission to consume these highly radiotoxic isotopes, these isotopes can transition to different isotopes following radiative capture.

Figure 1.5: Fission ($\sigma_f$, red) and radiative capture ($\sigma_c$, green) cross sections for $^{241}$Am at 293 K extracted from the JEFF-3.1 database. Note that radiative capture dominates fission for most of the energy range, except for fast neutron energies. This indicates that fast reactors would fission this isotope more often than thermal reactors.
Figure 1.6: Fission ($\sigma_f$, red) and radiative capture ($\sigma_c$, green) cross sections for $^{238}$Pu at 293 K extracted from the JEFF-3.1 database. Note that radiative capture dominates fission for most of the energy range, except for fast neutron energies. This indicates that fast reactors would fission this isotope more often than thermal reactors.
CHAPTER 2

CURRENT WORK

Considerable amounts of work have gone into analyzing the nuclear fuel cycle based upon the neutron cross section characteristics of various isotopes. In particular, methods for transmutation, radiotoxicity reduction, and waste reductions have been investigated. Presented within this chapter are a series of static transmutation studies that investigate these methods as well as the progress and shortcomings of dynamic transmutation models.

2.1 Transmutation Studies

Several studies have been performed that look at the results of using different (thermal or fast) reactor technologies to perform recycling and transmutation. Since fission becomes more likely for some isotopes at higher neutron energies, recycling in fast reactors tends to dominate these studies; however, transmutation in thermal reactors is also a possibility. In addition, particle accelerator driven systems (ADSs) can generate both fast and thermal neutrons. For this reason, ADSs are also considered for transmutation. To illustrate these concepts, a few static transmutation studies are summarized.
2.1.1 Transmutation Study #1 (Bergelson, et al. 2004)

The first study (Bergelson et al., 2004) of note examines the effects of preliminary transmutation of minor actinides in various reactor technologies. Specifically, thermal pressurized (light) water reactors (PWRs) in the form of the Russian VVER-1000, thermal pressurized heavy water reactors (PHWRs) in the form of the Canadian CANDU reactor, and fast reactors (FRs) similar to the French Superphénix reactor are compared. This study investigated the waste amount and radiotoxicity reduction associated with transmutation.

The transmutation approach used in this study focused on the consumption and production of two primary radiotoxic isotopes, $^{241}$Am and $^{238}$Pu. The $^{241}$Am is present in conventional used nuclear fuel along with $^{244}$Cm and these isotopes (and their progeny, such as $^{237}$Np) dominate the radiotoxicity of used nuclear fuel. As shown in the activation and decay series below, $^{244}$Cm has a relatively short half-life (18.1 years) compared to $^{241}$Am (432.2 years) and as a result $^{241}$Am tends to contribute to the long-term radiotoxicity:

\[
^{237}\text{Np} + n \rightarrow ^{238}\text{Np} \quad (2.12\, \text{days}) \rightarrow ^{238}\text{Pu} \quad (87\, \text{years})
\]

\[
^{241}\text{Am} + n \rightarrow ^{242}\text{Am} \quad (82.7\%, 16\, \text{hours}) \rightarrow ^{242}\text{Cm} \quad (161\, \text{days}) \rightarrow ^{238}\text{Pu}
\]

\[
^{241}\text{Am} + n \rightarrow ^{242}\text{Am} \quad (17.3\%, 16\, \text{hours}) \rightarrow ^{242}\text{Pu}
\]

\[
^{242}\text{Pu} + n \rightarrow ^{243}\text{Pu} \quad (5\, \text{hours}) \rightarrow ^{243}\text{Am}
\]

\[
^{243}\text{Am} + n \rightarrow ^{244}\text{Am} \quad (10\, \text{hours}) \rightarrow ^{244}\text{Cm} \quad (18.1\, \text{years})
\]
Once transmutation begins, the radiotoxicity of the recycled fuel tends to increase as $^{238}\text{Pu}$ is generated from the $^{237}\text{Np}$ and the $^{241}\text{Am}$ that is present. Eventually, however, $^{238}\text{Pu}$ will become consumed (by absorbing a neutron) and the radiotoxicity will decrease.

As a result of the initial increase in radiotoxicity due to $^{238}\text{Pu}$ production, there is an optimal length of time for thermal transmutation where the amount of actinides and radiotoxicity are realistically minimized. In order to investigate this, the study of interest used the composition of used fuel from a PWR (generating 1000 MWe) with a 40 GWd/MTU burnup and subsequent 3 year cooling time to determine the composition of the recycled fuel that would then be used for transmutation. The fuel composition following 10 years of re-irradiation and transmutation was simulated and the composition results (from this study) in various reactors are presented in Table 2.1. From this table, it is visible that the amount of Np, Am, and $^{244}\text{Cm}$ present in the used fuel (without transmutation category) are significantly reduced while other actinides are generated after 10 years of irradiation.

Since other actinides are generated, the radiotoxicity of the fuel is determined to facilitate comparison. Figures 2.1 and 2.2 indicate that after 10 years of transmutation, the radiotoxicity of the actinides in the PWR and PHWR is about the same as before the used fuel experienced transmutation. However, the amount of actinides has been reduced by 3 to 4 times. In other words, without significantly increasing the radiotoxicity of the used nuclear fuel, the amount of actinide waste can be decreased by 3 to 4 times utilizing a single recycle in thermal reactors. Ultimately, this study is significant because it demonstrates the benefits and feasibility of thermal reactor transmutation and recycling;
however, the long term results and feasibility are questionable, as is demonstrated in the next study.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Without Transmutation</th>
<th>Transmutation in a PHWR</th>
<th>Transmutation in a PWR</th>
<th>Transmutation in a FR</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{237}\text{Np}$</td>
<td>13.7</td>
<td>0.246</td>
<td>0.827</td>
<td>2.49</td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>0</td>
<td>0.206</td>
<td>2.26</td>
<td>4.26</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>0</td>
<td>0.0942</td>
<td>0.674</td>
<td>1.23</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>0</td>
<td>0.105</td>
<td>0.202</td>
<td>0.518</td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>0</td>
<td>0.0643</td>
<td>0.304</td>
<td>0.0549</td>
</tr>
<tr>
<td>$^{242}\text{Am}$</td>
<td>0</td>
<td>0.767</td>
<td>0.559</td>
<td>0.414</td>
</tr>
<tr>
<td>$^{241}\text{Am}$</td>
<td>5.90</td>
<td>0.301</td>
<td>0.0248</td>
<td>0.901</td>
</tr>
<tr>
<td>$^{242}\text{Am}$</td>
<td>0.024</td>
<td>$9.29 \times 10^{-5}$</td>
<td>$1.03 \times 10^{-3}$</td>
<td>0.0944</td>
</tr>
<tr>
<td>$^{243}\text{Am}$</td>
<td>2.79</td>
<td>0.561</td>
<td>0.430</td>
<td>0.765</td>
</tr>
<tr>
<td>$^{242}\text{Cm}$</td>
<td>0</td>
<td>$2.15 \times 10^{-3}$</td>
<td>$9.95 \times 10^{-3}$</td>
<td>0.0704</td>
</tr>
<tr>
<td>$^{243}\text{Cm}$</td>
<td>0</td>
<td>$1.50 \times 10^{-4}$</td>
<td>$7.82 \times 10^{-4}$</td>
<td>0.0119</td>
</tr>
<tr>
<td>$^{244}\text{Cm}$</td>
<td>0.800</td>
<td>1.64</td>
<td>0.952</td>
<td>0.971</td>
</tr>
</tbody>
</table>

Table 2.1: Isotopic composition of actinides after 10 years of transmutation (kg). Reproduced from (Bergelson et al., 2004).

Figure 2.1: Radiotoxicity of actinides versus transmutation time in a (1) PHWR, (2) PWR, and (3) FR. The radiotoxicity is reported as the amount of drinking water necessary to dilute the material to allowable ingestion limits according to Russian radiation safety standards and the transmutation time is reported in years. Reproduced from (Bergelson et al., 2004).
2.1.2 Transmutation Study #2 (Cometto, Wydler and Chawla 2004)

The second study (Cometto et al., 2004) of note takes a more detailed approach than only looking at individual reactors. In this study, the pros and cons of various fuel cycle strategies are investigated. Specifically, this study focused on pseudo-steady-state scenarios involving thermal light water reactors (LWRs), advanced fast-spectrum systems (e.g., fast reactors, FRs), and accelerator (fast spectrum) driven systems (ADSs).

Figure 2.3 illustrates the 6 fuel cycle schemes that this study investigated. The first scheme (1) is the current once-through approach utilized by the US. The second (2) scheme recycles only Pu and thus does not classify as a transmutation strategy, but it does reduce the heavy metal inventory and plutonium accumulation in the fuel cycle. The third (3A) scheme is a transmutation scheme that consumes transuranic (TRU) isotopes in critical fast reactors where the fourth scheme (3B) consumes TRU isotopes in sub-critical
ADSs. The fifth scheme (4) utilizes the double strata approach where plutonium and minor actinides are consumed separately. Finally, the last scheme (5) utilizes fast reactors in an idealized close fuel cycle where a uranium blanket is used to breed fuel for the fast reactor.

![Fuel cycle schemes](image)

Figure 2.3: Fuel cycle schemes investigated. Reproduced from (Cometto et al., 2004).

Continuing with this study, the fuel composition for each fuel cycle scheme was reported. Since each scheme utilized a different reactor concept, the overall reactor
composition was reported rather than the composition of a transmutation channel like the Bergelson study (2004). The compositions following fuel fabrication (pre-irradiation) and following irradiation have been reproduced in Table 2.2. In addition, the mass reduction factor of using alternative strategies relative to the current once-through approach for various groups (namely plutonium (Pu), minor actinides (MA), and TRU) were reported as shown in Table 2.3. These reduction factors indicate that significantly less high-level radioactive waste (HLW) is generated in transmutation schemes (3A, 3B, 4, and 5) and more than 400 times the amount of plutonium can be consumed in the plutonium recycle scheme (2).

These benefits are not limited to only waste reduction, but this study also showed decreased radiotoxicity for long term storage as shown in Fig. 2.4. Specifically, after discharged fuel undergoes separation, the remaining HLW is as much as two orders of magnitude less radiotoxic (reported as Sieverts per terra-watt-hour electric produced). In addition, this study showed little difference in radiotoxicity between transmutation schemes. This indicates that waste reduction may be the deciding factor between these recycling and transmutations schemes.
Table 2.2: Fuel composition following fabrication and irradiation reported in weight percent for the fuel cycle schemes of interests. Reproduced from (Cometto et al., 2004).

<table>
<thead>
<tr>
<th>Scheme 1</th>
<th>Scheme 2</th>
<th>Scheme 3A</th>
<th>Scheme 3B</th>
<th>Scheme 4 Double Strata</th>
<th>Scheme 5 FR</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWR-MOX</td>
<td>Pu burner</td>
<td>TRU (FR)</td>
<td>TRU (ADS)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>U 91.90</td>
<td>55.57</td>
<td>66.96</td>
<td>1.21</td>
<td>4.62</td>
<td>76.44</td>
</tr>
<tr>
<td>Pu 8.10</td>
<td>44.43</td>
<td>29.14</td>
<td>82.37</td>
<td>40.42</td>
<td>22.87</td>
</tr>
<tr>
<td>Np  -</td>
<td>-</td>
<td>0.94</td>
<td>3.05</td>
<td>7.89</td>
<td>0.17</td>
</tr>
<tr>
<td>Am  -</td>
<td>-</td>
<td>2.06</td>
<td>8.12</td>
<td>30.19</td>
<td>0.43</td>
</tr>
<tr>
<td>Cm  -</td>
<td>-</td>
<td>0.89</td>
<td>5.25</td>
<td>16.87</td>
<td>0.09</td>
</tr>
</tbody>
</table>

Table 2.3: Mass reduction factors for plutonium (Pu), minor actinides (MA), and transuranics (TRU) relative to the once-through strategy. Reproduced from (Cometto et al., 2004).

<table>
<thead>
<tr>
<th>Scheme 2 Pu burner</th>
<th>Scheme 3A TRU (FR)</th>
<th>Scheme 3B TRU (ADS)</th>
<th>Scheme 4 Double Strata</th>
<th>Scheme 5 FR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu 412.91</td>
<td>342.20</td>
<td>191.76</td>
<td>333.94</td>
<td>176.18</td>
</tr>
<tr>
<td>MA 0.48</td>
<td>322.24</td>
<td>116.42</td>
<td>137.73</td>
<td>728.00</td>
</tr>
<tr>
<td>TRU 4.35</td>
<td>339.90</td>
<td>178.98</td>
<td>288.56</td>
<td>192.27</td>
</tr>
</tbody>
</table>
2.1.3 Transmutation Study #3 (Salvatores 2005)

The third study (Salvatores, 2005) of note conducted a more detailed investigation of how partitioning and transmutation of the various isotopes will affect the nuclear fuel cycle using fast FRs and (fast neutron) ADSs. In addition, this study also examined the possibility of thermal recycling. In general, this study took an even broader approach to the fuel cycle and concerned itself with how different partitioning groups would perform for recycling in the next generation of nuclear power plants (e.g., GEN-IV reactors).
With regard to partitioning and transmutation, this study focused on separating the fuel into Pu, Pu+Np, Pu+Np+Am, and Pu+MA (minor actinides, namely Np+Am+Cm) groups for transmutation. The reason for doing this is to investigate the affects of different separation strategies as the radiotoxicity of various groups behave differently with time as illustrated in Fig. 2.5. Specifically, the radiotoxicity of fission products dominate (over the minor actinides) for the first 100 years after discharge. If fission products were the only isotopes not recycled, the radiotoxicity of this waste would approach that of the reference level (naturally occurring uranium) after about 300 years. For this reason, this study envisioned to recycle the actinides (indefinitely) in multi-pass systems.

When investigating the fuel cycle performance of thermal reactor recycling, this study found that such a practice would cause an increase in decay heat and neutron and gamma ray doses as shown in Table 2.4. This is due to the dominant radiative capture cross section of some actinides in the thermal neutron range (see Figs. 1.5 and 1.6 for examples). This study attributes the reason for this to the Cm (specifically $^{244}\text{Cm}$) that is generated (see previous activation and decay chain in Sec. 2.1.1). This study notes that if thermal recycling is to be used, then partitioning with Pu+Am recycling should be avoided.

Ultimately this study concluded that transmutation in fast reactors is the best choice for sustained nuclear energy. In addition, this study found that no significant penalty resulted from implementing homogeneous recycling. In other words, selective partitioning in fast reactors does not result in significant benefit. Also, since the use of
fast reactors in terms of GEN-IV technologies may not be timely, this study also concluded that ADSs would be effective while FR technology is delayed.

Figure 2.5: Radiotoxicity, reported as Sieverts per tonne of heavy metal (Sv/tHM), of uranium oxide (UOX) used fuel versus time. Irradiation occurs in an UOX-LWR with 3.7% $^{235}$U and 45 GWd/tHM burnup. Reproduced from (Salvatores, 2005).

<table>
<thead>
<tr>
<th>Fuel cycle parameter</th>
<th>Increase factor</th>
<th>Dominant contributing isotopes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Decay heat</td>
<td>~10</td>
<td>$^{244}$Cm ~70%, $^{238}$Pu ~30%</td>
</tr>
<tr>
<td>Neutron source</td>
<td>~5000</td>
<td>$^{252}$Cf ~90% if cooling time=20 years, $^{252}$Cf ~40%, the rest is due to $^{244}$Cm, $^{250}$Cf, $^{246}$Cm, and $^{248}$Cm</td>
</tr>
<tr>
<td>Gamma source</td>
<td>~10</td>
<td>$^{241}$Am, $^{244}$Cm, $^{238}$Pu, $^{252}$Cf, and $^{243}$Cm</td>
</tr>
</tbody>
</table>

Table 2.4: Impact of fuel cycle parameters for multiple recycling in (thermal) LWRs. Reproduced from (Salvatores, 2005).
2.1.4 Transmutation Study #4 (Wigeland, et al. 2006)

The final study (Wigeland et al., 2006) of note deals with the ultimate disposal of used nuclear fuel. In this study, the heating effects on a geological repository (i.e., Yucca Mountain) were investigated. The purpose of a repository is to safely store hazardous radioactive waste for extended periods of time (to permit radioactive decay) in a manner that limits the release of radioactive material to the environment. Releasing radioactive material into water is of primary concern. For this reason, the dry environment of Yucca Mountain has been selected as the site for long-term storage of commercial used nuclear fuel in the US. The repository in Yucca Mountain is located 324 m below the surface and 344 m above the water table (Wigeland et al., 2006). The rock conditions and the emplacement strategy is such that surface water will be able to pass through Yucca Mountain without contacting the repository tunnels if certain waste loading parameters are met.

Specifically, this study mentions that the waste loading parameters are dominated by the “maximum decay heat for each waste package at the time of placement and the maximum average linear heat rate for the array of waste packages in a repository drift [tunnel]” (Wigeland et al., 2006) which is in accord with the Yucca Mountain technical documents (US Department of Energy, 2002). The limiting parameters are that the rock temperature midway between repository tunnels must not exceed 96 °C to permit water to flow downward and not condense and flood the tunnels when it cools. In addition, the tunnel walls must also remain below 200 °C to prevent any alteration to the crystalline structure of the surrounding rock.
Given these constraints, the study then identified the major contributors to decay heat for isotopes present in conventional pressurized water reactor fuel. Figure 2.6 illustrates which isotopes are major contributors to decay heat as a function of time. As also demonstrated by the radiotoxicity of discharged fuel (Fig. 2.5), the major contributor to short-term decay heat is the fission products, which sharply decrease after the first 100 years. The dominant fission products are $^{137}$Cs and $^{90}$Sr and their associated decay products, $^{137m}$Ba and $^{90}$Y. Next, the major long-term contributors are the actinides with $^{238}$Pu, $^{241}$Am, $^{244}$Cm dominating just like in radiotoxicity.

After identifying the problematic isotopes, this study then investigated the increase in repository loading if the problematic isotopes were to be removed by partitioning and transmutation. This study found, as shown in Fig. 2.7, that if 99.9% of the actinides, namely Pu, Am, and Cm, were removed (0.1% remaining) then the repository loading could see an increase of 5.7 times. If both 99.9% of actinides, and Cs and Sr were removed, then the repository loading would increase by 225 times. However, this study made the incorrect assumption that the number of waste packages would stay the same following partitioning, whereas in reality, the number of waste packages would increase in a non-linear fashion as separated waste takes a different form than conventional used fuel waste forms. This study did, however, aid in better understanding the effects of recycling and transmutation on the ultimate disposal of radioactive waste in a geological repository.
Figure 2.6: Primary decay heat contributors present in PWR fuel with burnup of 50 giga-watt-day per metric tonne of initial heavy metal (GWd/MTIHM). Reproduced from (Wigeland et al., 2006).
2.2 Dynamic Fuel Cycle Simulation

All of the previous fuel cycle studies used static nuclear fuel cycle models. In these studies, a time-invariant model was used where a fuel cycle strategy with appropriate parameters was fixed at the start of the model. These parameters did not vary as the fuel cycle responded to the strategy of choice. This is not realistic since feedback that can be associated with the impact of a new fuel cycle strategy was not included. As with any complex system, these feedback parameters are essential. In short, these static studies are helpful at demonstrating a proof of concept, but they cannot evaluate what-if
scenario questions for a constantly changing fuel cycle due to the lack of dynamic response.

Alternatively, a dynamic fuel cycle model would be more appropriate to answer these what-if scenarios. In such a model, the interrelationship of each stage of the fuel cycle is taken into account. For example, deploying technology Y (e.g., fast reactors) may compete with existing technology X (e.g., thermal reactors) if both technologies share a common mode (e.g., electrical power generation). Assume, that technology Y is dependent on technology X (e.g., for recycled fuel). Therefore, when technology X experiences a decline due to technology Y’s deployment, technology Y will respond in like. This is the dynamic approach. In the static approach, even if technology X changed, technology Y would (more or less) generally remain unchanged. The interrelationship and therefore determining the ultimate effectiveness of a fuel cycle strategy rely on the system’s dynamics not solely on the attributes of a fuel cycle strategy.

For the case of the current nuclear fuel cycle, the resource management, economics, and amount of waste are key metrics in determining the effectiveness of the fuel cycle. These metrics are affected by the interrelationship within the fuel cycle. For example, fast reactors may be deployed to consume the amount of used fuel produced by thermal reactors as this used fuel is recycled for fuel in fast reactors. In addition, this affects the economics as the fast reactors would compete with the thermal reactors for shares of the electricity generation market. This in turn could reduce the capacity of thermal reactors and thus reduce the amount of recycled fuel sent to fast reactors. Finally, this would decrease the capacity of fast reactors and an oscillatory response
ensues. Theoretically, given sufficient time, this oscillatory response would settle resulting in a steady-state operation or (cyclic) pseudo-steady-state operation. However, given economics’ susceptibility to perturbations (e.g., war, government incentives, inflation, etc.), future advances in technology, and the large time constants of such a system, a pseudo-steady-state result is not likely; the system will experience a series of transient responses. The only way to capture the transient response of the nuclear fuel cycle is to use a dynamic model where the parameters change with time.

2.2.1 Introduction to the Verifiable Fuel Cycle Simulation (VISION) Model

One such dynamic model is the Verifiable Fuel Cycle Simulation (VISION) model. This model was developed at Idaho National Laboratory (INL) as a US Department of Energy (DOE) Advanced Fuel Cycle Initiative (AFCI) project with the assistance of Argonne National Laboratory (ANL) and in collaboration with universities (including The Ohio State University, North Carolina State University, Massachusetts Institute of Technology, University of Illinois at Urbana-Champaign, University of Wisconsin, University of Texas, Idaho State University, and University of Idaho).

The purpose of this model was to answer what-if scenario questions for the US nuclear fuel cycle regarding reactor fuel performance, reactor technology deployment, and processing (recycling) options from a systems perspective. The model was not intended for process and design modeling of fuel cycle facilities and it was not intended to track individual fuel elements through the nuclear fuel cycle. To clarify, the model is intended to examine interactions between fuel cycle facilities (components) for varying system parameters. For this reason, this model is not a steady-state approximation, but
rather a dynamic approximation that can be used to evaluate the effectiveness of a fuel cycle option for numerous metrics including resource management, economics, and the amount of radioactive waste (Jacobson et al., 2007).

### 2.2.2 Modeling and Organization of VISION

The VISION model is constructed using a system dynamics software package known as Powersim (Studio 7). This package is similar to other well-known system dynamics software, such as Simulink, VenSIM, and iThinK, where the scripting language consists of block diagrams connected by wires. The individual blocks represent fuel cycle system components. In the case of Powersim, the blocks can be considered as a series of levels (rectangles), valves (circles), constants (square diamonds) and wires (connecting lines) as shown in Fig. 2.8. Levels represent locations of material accumulation, valves control the material flow from the accumulators, and wires are used to represent the flow of material (double walled) or information (single walled). Each of these components is then given functionality in the form of mathematical equations or logic statements. For example, $x$ kg/year of mass $X$ enters component $A$ and after $t$ year (constant time delay) $y$ kg/year of mass $Y$ leaves for component $B$ and $z$ kg/year of mass $Z$ leaves for component $C$. Each facility of the fuel cycle can be constructed in a similar manner to determine the interrelationship between facilities.
VISION, however, is not the first US fuel cycle model to be developed to aid in determining the direction of nuclear power. A precursor to the VISION model was the Dynamic Model of Nuclear Development (DYMOND) model (Moisseytsev, 2001) that was used to better understand the interrelationship of uranium resources, nuclear fuel, reactors, and waste management. This model was used primarily to answer fuel cycle questions for GEN-IV reactor technologies. Some of these questions include (Yacout et al., 2006):
• What alternatives exist to building multiple geological repositories while still supporting an expanding role for nuclear energy?
• How can the principles of reduce, reuse, and recycle best be applied to nuclear power development?
• What types of reactors and fuels will be needed and when will they be needed to minimize long term waste management, while maintaining economic competitiveness?
• What elements of the nuclear fuel cycle are most sensitive to changes in economics, waste policy, energy supply options, and development of new technologies? How do we make the system flexible, robust, and dependable?

In order to answer these questions, the DYMOND model was subdivided into a series of system components which further divided the major fuel cycle categories (front-end, reactors, and back-end). The components of interest are (1) mining and enrichment, (2) fuel fabrication, (3) reactor park, (4) reactor operations, (5) used nuclear fuel storage, (6) recycling and reuse of used nuclear fuel, (7) additional storage requirements, (8) waste packaging and transportation, and (9) repository disposal (Yacout et al., 2006). For the GEN-IV scenarios, these components and these questions were sufficient; however, the DYMOND model lacked the capabilities to answer questions regarding advanced fuel cycle strategies.
For this reason, VISION was developed to correct for these inabilities. The VISION model and approach were based upon the DYMOND model; however, additional details were added. In the VISION model the fuel cycle was divided into modules (which are similar to the system elements used in DYMOND) as depicted in Fig. 2.9. These modules and additional modules corresponding to economics and non-fuel byproducts (co-flow) have also been developed as VISION grew. However, with the growth, the original complexity of some system aspects was overlooked in the initial development. For such a large complicated system, assumption that introduces even a small amount of error may propagate and result in gross system error.
Figure 2.9: Advanced Fuel Cycle Initiative’s (AFCI’s) fuel cycle flow sheet depicting modules of interests in VISION. Reproduced from (Shropshire et al., 2007).
2.2.3 Areas for Improvement within VISION

Even though VISION has seen significant development and is nearing completion, there are some significant challenges with the model that need to be addressed. Specifically, the recycled fuel fabrication module (D2) and the thermal and fast reactor modules (R1 and R2) need additional refinement. The functionality of these modules is limited and an error in these modules will propagate throughout the fuel cycle model. In particular, the recycled fuel fabrication model is intended to load recycled fuel. The recycled fuel is made by combining isotopes from the separation process and fresh uranium. The limiting factor is the amount of recycled material from the separation process, which will contain mostly transuranic isotopes. Since VISION is used to examine interactions and not meant to be used as a design tool, the details of the proposed advanced reactor designs (i.e., geometry, neutron flux, and temperature) are not directly incorporated into the model. Thus, the thermal and fast reactor modules lack dynamic reactor physics calculations. Instead, a wider systems approach is taken but further refinement of these modules is desired.

At present, the recycled fuel fabrication module consists of static fuel recipes that prescribe the composition of recycled fuel. The isotopes for this recipe come from recycled product storage and fresh uranium. In the current version, only those isotopes that are prescribed by the recipe are loaded into the new fuel. This is not realistic as elemental separation (not isotopic separation) is employed. If the recipe calls for x grams of $^{241}\text{Pu}$, then y grams of $^{238}\text{Pu}$ will accompany the x grams of $^{241}\text{Pu}$ since isotopic separation at this level is not performed. The recipes do not account for this. In addition,
the recipe may call for a fixed ratio of TRUs to U. Since the properties of the TRU vary considerably with the isotopic composition, simply matching the TRUs to U ratio is not sufficient to qualify the fuel (e.g., guarantee the fuel will obtain criticality). A dynamic fuel loading approach that accounts for criticality is needed.

Following this dynamic loading, the resulting composition after irradiation also needs to be determined. Currently, static fuel output recipes are utilized to represent the transmutation of fresh (or recycled) fuel recipes following irradiation in a specific reactor technology. Given that the fresh fuel recipe will change due to the dynamic nature of the fuel cycle and recycling, the static used fuel recipes are not accurate. In other words, if the reactor input recipes changes with dynamic fuel loading, then the reactor output recipes will also change. A dynamic reactor response is currently not implemented in VISION.

The coupling of these two problems introduces significant error into the system. For example, currently non-recipe prescribed isotopes are accumulating in certain modules of the fuel cycle. The volume and radiotoxicity of waste is not accurately approximated since these non-recipe prescribed isotopes are not present and since the transmutation in reactors is also not dynamically performed. And finally, this static approach hinders VISION’s robustness as fixed technologies and compositions are used. A more dynamic approach within these modules would perform better.

2.3 Scope of Work

The goal of the research presented here is to remedy deficiencies in VISION by developing approaches that allow for dynamic fuel loading and the subsequent
transmutation. This approach involves dividing the problem into two parts: (1) dynamic fuel loading based upon a novel reactivity worth approach and (2) subsequent transmutation in reactors based upon a recursive matrix exponentiation method. Implementing these approach into fuel cycle models, like VISION, should aid in eliminating the fictitious accumulation of non-recipe prescribed isotopes and more accurately model the resulting transmutation of fuel in reactors.

The specific purpose of the fuel loading approach is to develop a method for determining the fraction of TRU and U that can be loaded into a specific reactor. Since the composition of the used nuclear fuel will vary as technology changes, the composition of the recycled elements will also have to adapt accordingly. The goal of the fuel loading approach is to best approximate a target fuel recipe given that the available ingredients do not match what is prescribed. In particular, an adjustment of the fraction of TRU and U is performed such that the fuel maintains approximately the same beginning of cycle reactivity quality.

Also, the specific purpose of the transmutation approach is to determine the composition of the resulting fuel once the adjusted recycled fuel is irradiated. Since details of the reactor core or other necessary reactor physics parameters are not tracked in VISION, an approach that utilizes a minimum set of input parameters yet still approximates the used fuel composition well is necessary. An approach utilizing neutron cross sections, approximate neutron energy distribution, fission yields, and activation and decay chains is employed for this purpose. In order to execute in a timely manner and without expending excessive computation power, these parameters are utilized in a
recursive matrix exponentiation approach patterned off a well-validated transmutation code (e.g., ORIGEN-S).

These two approaches aid in reducing the error inside VISION by correcting the problem of relying on static fuel recipes. These two approaches also add a level of fuel loading and reactor dynamics to VISION that are not present in any other fuel cycle simulation. With regard to accuracy, this will improve VISION’s ability to approximate the real fuel cycle and aid in decision making by answering what-if scenario questions better. These approaches are also novel and far reaching as the concepts introduced here can be applied to other problems and technologies.
CHAPTER 3

FUEL LOADING APPROACH

Developing an appropriate algorithm for the VISION model to load fuel into the reactor has inherent challenges due to the limitations of the fuel cycle model. For example, with VISION individual fuel assemblies are not tracked. Likewise, core geometry is not a working parameter in the fuel cycle model. In addition, conducting full length reactor physics calculations are too computationally intensive to perform at each time step of a system dynamic model. VISION also tracks the entire fleet of reactors. Tracking individual fuel assemblies and performing detailed reactor physics models is not feasible with common computational power. For these reasons, assumptions must be made to develop simplified approaches that still approximate reality.

With regard to fuel loading, the approach must utilize a minimum set of input parameters. The goal behind all good models is to be able to approximate reality with the minimum number of degrees of freedom (i.e., inputs). For this reason, the fuel loading approach should only rely on the isotopic composition of available material and the known properties of those isotopes. In addition, since geometry and reactor specific parameters (i.e., $k_{eff}$, temperature, and rated power) are not readily available for each reactor in the fleet, an approach that relies solely on a working isotopic composition for a
reactor is best. In short, the fuel loading approach that would be beneficial for VISION and other similar fuel cycle models is one that only relies on: (1) an available isotopic composition of fuel materials, (2) an isotopic composition representing a working fuel recipe, and (3) known properties corresponding to the isotopes present in those compositions.

In addition, the loading approach must also be realistically feasible. In most reactors, transmutation channels will be employed to consume transuranic (TRU) material. These channels will differ from regular fuel channels, which contain mostly uranium (U). Realistically, the goal is to determine the ratio of the transmutation channels to regular fuel channels or, in other words, to determine the mass fraction of TRU and U. Since isotopic separation is not envisioned, only elemental partitioning processes will take place. In many cases, the partitioning can be dichotomized into U and TRU (i.e., Pu, Np, Am, Cm).

The novel fuel loading approach developed here is based upon assigning a dimensionless reactivity worth value to each isotope in the available inventory of fuel material (based upon the isotopic composition of the working fuel recipe) and then balancing this reactivity worth such that fuel behaves similar to the working fuel. Balancing reactivity worth value attempts to determine the fractions of TRU and U such that there is no net quality change for using recycled fuel at the beginning of cycle. The quality or performance of the fuel is based upon its ability to fission and create energy, or in short, how the neutron population will multiply with this fuel.
3.1 Isotopic Reactivity Worth Value

The temporal variation of the neutron population, $n(t)$, can be described by the following first order rate equation shown eq. (3.1):

$$\frac{dn(t)}{dt} = r_1(t)M_1(t) + r_2(t)M_2(t) + r_3(t)M_3(t) + ... = \sum_i r_i(t)M_i(t) \quad (3.1)$$

where $r_i(t)$ represents the neutron production (or consumption) rate constant for species (isotope) $M_i$. In this case, each isotope will either contribute to subtract from the neutron population dependent upon if it absorbs more neutron than results from fission. When the reactor is critical ($k_{\text{eff}}=1$), the neutron population remains constant with respect to time and the right hand side of the rate equation equals zero as those isotopes that absorb neutrons are balanced by those that generate neutrons by fission. Also note that the amount of each species will change with time dependent on the neutron population, thus a system of coupled equations results. Such a rate approach, however, is unconventional in describing the neutron population for a homogenized reactor. Nevertheless, this illustrates the fundamental concept of reactivity worth value (RWV), which is simply a parameter describing the neutron production rate constant of a species.

The RWV introduced here is defined as a dimensionless parameter that describes an isotope’s contribution to the neutron population. If an isotope is a pure neutron absorber, then every neutron that encounters such an isotope would be captured (negative RWV). On the other hand, if an isotope fissions, then it will contribute to the neutron population (positive RWV). However, the neutron population is not solely determined by the fuel and neutrons are also lost due to leakage (escape without return from the active reaction volume) and absorption in structural material. For this reason, the RWV for each isotope is dependent on the specific reactor. However, if the active reaction volume
is assumed to be infinite and contain only one isotope, then the neutron multiplication can be determined using the conventional Four Factor Formula.

### 3.1.1 Four Factor Formula

The Four Factor Formula (FFF) is a simple calculation that can be used to approximate the infinite multiplication factor \( k_{\infty} \). This approach lacks geometry dependence and is therefore not appropriate for design work; however, it is useful in approximating \( k_{\infty} \) given only the composition. A description of the FFF approach follows and can also be found in many nuclear reactor theory texts (Duderstadt and Hamilton, 1976), (US Department of Energy vol. 2, 1993).

For a thermal (represented by the subscript \( t \)) neutron spectrum reactor \( k_{\infty} \) can be approximated by eq. (3.2) which is known as the FFF as.

\[
 k_{\infty,t} = \varepsilon_t p_t f_t \eta_t 
\]

where

- \( \varepsilon_t \equiv \text{fast neutron factor} \), \( \varepsilon_t = \frac{\text{Total \# of fission neutrons produced by all neutrons}}{\text{# of fission neutrons produced by thermal neutrons}} \)
- \( p_t \equiv \text{resonance escape probability} \), \( p_t = \frac{\text{# of thermal neutrons absorbed}}{\text{# of all neutrons absorbed}} \)
- \( f_t \equiv \text{thermal utilization factor} \), \( f_t = \frac{\text{# of thermal neutrons absorbed inducing fission}}{\text{Total \# of thermal neutrons absorbed}} \)
- \( \eta_t \equiv \text{neutron reproduction factor} \), \( \eta_t = \frac{\text{# of fission neutrons produced}}{\text{# of thermal neutrons absorbed for fission}} \)

The fast neutron factor accounts for additional fission outside the thermal spectrum of interest. For \( \varepsilon_t = 1 \) no fast neutrons cause fissions and for \( \varepsilon_t > 1 \) some fast neutrons cause fissions. This factor is fixed by the fuel composition and typical ranges in thermal reactors are between 1.03 and 1.15 (Duderstadt and Hamilton, 1976).
The resonance escape probability, $p_t$, accounts for thermal neutrons that are lost due to resonance capture. Some population of neutrons will always be lost to radiative capture. However, this factor accounts for losses that are preferentially removed as neutrons thermalize. In other words, as neutrons slow down they become absorbed by heavy nuclides because of the capture cross section peaking in the resonance energy range (see Fig. 1.4). For $p_t=1$ no preferential loss of thermal neutrons are encountered and for $p_t<1$ preferential loss of thermal neutrons are encountered. Typical values for this factor are around 0.87 (Duderstadt and Hamilton, 1976).

The thermal utilization factor, $f_t$, is the ratio of the macroscopic absorption cross sections in the fuel to the total macroscopic absorption cross sections. It is used to describe the number of neutrons that are absorbed in the fuel compared to the total number of neutrons absorbed in all material. Using a one-group cross section approach that is dichotomized into only fission and absorption cross sections, the following relationship develops:

$$f_t = \frac{\sum_{i=\text{fuel}}(\Sigma_{f,i} + \Sigma_{c,i})}{\sum_{i=\text{fuel}}(\Sigma_{f,i} + \Sigma_{c,i})} = \frac{\sum_i(N_i\sigma_{f,i} + \Sigma_{c,i})}{\sum_i(N_i\sigma_{f,i} + \Sigma_{c,i})}$$

(3.3)

where

- $\Sigma_{f,i}$ = the macroscopic fission cross section of isotope $i$, [cm$^{-1}$]
- $\Sigma_{c,i}$ = the macroscopic radiative capture cross section of isotope $i$, [cm$^{-1}$]
- $N_i$ = the number density of isotope $i$, [cm$^{-3}$]
- $\sigma_f$ = the microscopic fission cross section of isotope $i$, [cm$^2$]
- $\sigma_a$ = the microscopic absorption cross section of isotope $i$, [cm$^2$]
Conventionally this factor also accounts for the neutron capture in the fuel, moderator, and structural material. However, since the moderator and structural material are not modeled inside VISION, all the material that is tracked can be considered fuel. Also, if we assume that the reactor is infinite and contains only one isotope, then the summations disappear and the factor reduces to eq. (3.4).

\[
f_t = \begin{cases} 
1 & \text{for fuel isotopes } (\sigma_f > 0) \\
0 & \text{for non-fuel isotopes } (\sigma_f = 0)
\end{cases}
\] (3.4)

For \(f_t = 1\), no neutrons are absorbed by material other than fuel (all neutrons cause fission in the fuel); for \(f_t = 0\), no neutrons cause fission in any material; and for \(f_t < 1\), only a fraction of neutrons cause fission in fuel material.

The neutron reproduction factor, \(\eta_t\), is the ratio of the number of neutrons produced in a subsequent generation given the number of neutrons absorbed from the previous generation and is described by:

\[
\eta_t = \nu \frac{\sigma_f}{\sigma_f + \sigma_c}
\] (3.5)

where \(\nu\) is the average number of neutrons per fission.

For a reactor consisting of only one isotope, this factor represents how that isotope will contribute to the neutron population. If \(\eta_t > 1\), then the isotope is fissile and if \(\eta_t < 1\), then the isotope consumes more neutrons than it produces. This is a very useful metric for determining the quality of an isotope.

The FFF is a useful approximation of the infinite multiplication factor for a reactor, but in this problem of fuel recycling, the FFF will be used to determine the RWV of an isotope. To do this, the FFF is applied to an infinite volume containing only one
isotope. Given the previously mentioned simplifications associated with this assumption the FFF can be expressed as:

\[ k_{\infty,t} = \varepsilon_i p_i f_i \eta_i = \varepsilon_i p_i f_i \left( v \frac{\sigma_f}{\sigma_f + \sigma_c} \right) \]  

(3.6)

This equation is strongly dependent on the microscopic fission and radiative capture cross sections. For this calculation, one-group cross sections were used; however, a multi-group approaches could also be used.

3.1.2 One-group Neutron Cross Sections

The one-group cross section is developed based upon the neutron flux energy spectrum of a reactor. Even though each reactor has its own unique spectrum, a typical neutron flux energy spectra for a typical fast and thermal reactor are shown in Fig 3.1 (US Department of Energy vol. 1, 1993). For thermal reactors, it can be seen that the neutron flux has two prominent peaks. The lower energy peak corresponds to those neutrons that have thermalized exhibiting the Maxwellian distribution, as depicted in Fig. 1.1. The higher energy peak corresponds to neutrons released after fission that have not undergone moderations, as depicted in Fig.1.2. For fast reactors, the spectrum is characterized by the (hard) spectrum skewed towards highly energetic neutrons as the name implies (e.g., fast neutrons have high kinetic energy).
Figure 3.1: Typical neutron flux energy spectrum for a thermal reactor and a fast (breeder) reactor. Reproduced from (US Department of Energy vol. 1, 1993).

The neutron cross sections (both for fission and radiative capture) and the neutron flux spectrum are continuous functions of neutron energy. However, for the recycling problem addressed in this thesis, it is more convenient to divide these continuous functions into discrete groups (i.e., the multi-group approach) where the continuous function is approximated in the limit as the number of groups approach infinity. Within these groups, an average cross section and average neutron flux are specified over that discrete energy range. As illustration, let the neutron cross sections and the neutron flux spectrum be divided into ten energy groups:

$$\sigma(E) \approx \{\sigma_1, \sigma_2, \sigma_3, \sigma_4, \sigma_5, \sigma_6, \sigma_7, \sigma_8, \sigma_9, \sigma_{10}\}$$

$$\phi(E) \approx \{\phi_1, \phi_2, \phi_3, \phi_4, \phi_5, \phi_6, \phi_7, \phi_8, \phi_9, \phi_{10}\}$$

(3.7)
where $\sigma_i$ represents the neutron cross section (both fission and radiative capture) and $\phi_i$ represents the neutron flux of the $i^{th}$ group. These group-wise parameters are determined based upon energy bins described by the limits $E_i$. The continuous functions are discretized into groups:

\[
\sigma_i = \frac{1}{E_{i+1} - E_i} \int_{E_i}^{E_{i+1}} \sigma(E) dE \\
\phi_i = \frac{1}{E_{i+1} - E_i} \int_{E_i}^{E_{i+1}} \phi(E) dE
\]  

(3.8)

Next, the multi-group cross sections are collapsed into a one-group cross section weighted by the neutron flux spectrum. This is accomplished by using the neutron flux spectrum to generate weighting parameters, $W_i$, which are then applied to collapse the cross sections into a one-group approximation as:

\[
W_i = \frac{\int_{E_i}^{E_{i+1}} \phi(E) dE}{\int_0^{\infty} \phi(E) dE}
\]

(3.9)

\[
\sigma \approx \sum_i \sigma_i W_i
\]  

(3.10)

where $\sigma$ represents the one-group neutron cross section approximation. Typical one-group cross sections for select isotopes used in VISION are included in Table 3.1 or in Appendix A.2.
### Thermal Reactor

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Radiative Capture Cross Section, $\sigma_{n,\gamma}$ (b)</th>
<th>Fission Cross Section, $\sigma_f$ (b)</th>
<th>Fast Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{234}\text{U}$</td>
<td>17.19</td>
<td>0.4911</td>
<td></td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>6.398</td>
<td>22.69</td>
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<tr>
<td>$^{236}\text{U}$</td>
<td>8.487</td>
<td>0.2166</td>
<td></td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>0.8718</td>
<td>0.1105</td>
<td></td>
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<tr>
<td>$^{237}\text{Np}$</td>
<td>24.23</td>
<td>0.5713</td>
<td></td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>15.26</td>
<td>2.033</td>
<td></td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>26.04</td>
<td>46.45</td>
<td></td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>43.88</td>
<td>0.6222</td>
<td></td>
</tr>
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<td>$^{242}\text{Pu}$</td>
<td>16.72</td>
<td>54.52</td>
<td></td>
</tr>
<tr>
<td>$^{243}\text{Pu}$</td>
<td>25.56</td>
<td>0.5354</td>
<td></td>
</tr>
<tr>
<td>$^{244}\text{Pu}$</td>
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<td>0</td>
<td></td>
</tr>
<tr>
<td>$^{241}\text{Am}$</td>
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<td>$^{242m}\text{Am}$</td>
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<td></td>
</tr>
<tr>
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<td></td>
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<td>$^{247}\text{Cm}$</td>
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<td>$^{248}\text{Cm}$</td>
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<td>$^{250}\text{Cm}$</td>
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<tr>
<td>$^{249}\text{Cf}$</td>
<td>29.13</td>
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<tr>
<td>$^{250}\text{Cf}$</td>
<td>293.4</td>
<td>1.002</td>
<td></td>
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<tr>
<td>$^{251}\text{Cf}$</td>
<td>128.8</td>
<td>285.3</td>
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</tr>
<tr>
<td>$^{252}\text{Cf}$</td>
<td>2.139</td>
<td>5.183</td>
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<table>
<thead>
<tr>
<th>Avg. # of neutrons per Fission, $\nu$</th>
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</thead>
<tbody>
<tr>
<td>2.631</td>
</tr>
<tr>
<td>2.421</td>
</tr>
<tr>
<td>2.734</td>
</tr>
<tr>
<td>2.801</td>
</tr>
<tr>
<td>3.005</td>
</tr>
<tr>
<td>2.833</td>
</tr>
<tr>
<td>2.875</td>
</tr>
<tr>
<td>3.135</td>
</tr>
<tr>
<td>2.934</td>
</tr>
<tr>
<td>3.28</td>
</tr>
<tr>
<td>3.3</td>
</tr>
<tr>
<td>3.277</td>
</tr>
<tr>
<td>3.162</td>
</tr>
<tr>
<td>3.732</td>
</tr>
<tr>
<td>3.746</td>
</tr>
<tr>
<td>3.434</td>
</tr>
<tr>
<td>3.725</td>
</tr>
<tr>
<td>3.832</td>
</tr>
<tr>
<td>3.858</td>
</tr>
<tr>
<td>3.592</td>
</tr>
<tr>
<td>3.796</td>
</tr>
<tr>
<td>3.8</td>
</tr>
<tr>
<td>4.062</td>
</tr>
<tr>
<td>3.97</td>
</tr>
<tr>
<td>4.14</td>
</tr>
<tr>
<td>4.126</td>
</tr>
</tbody>
</table>

Table 3.1: Example one-group cross sections for thermal and fast reactors and the average number of neutrons per fission of selected isotopes that are used within VISION.

### 3.1.3 Neutron Losses

Currently, the development of the FFF has assumed that the active core volume is infinite. In other words, no neutrons leak out of the core volume. This is not the case in reality as the core volume is finite in size and neutrons do indeed leak out. To account
for this loss, the probability of non-leakage (i.e., the probability that neutron will not leak out of the core is included). Following the typical dichotomy of fast and thermal neutrons, the neutron non-leakage probabilities are also separated into a thermal non-leakage fraction and a fast non-leakage fraction. When considering losses, the effective multiplication factor, $k_{eff}$, is a more appropriate description of criticality. An approximate description of $k_{eff}$ is shown in eq. (3.11) and is commonly referred to as the Six Factor Formula (SFF):

$$k_{eff} = k_{e,v}P_{tnl}P_{fnl} = \epsilon_i p_i f_i \eta_i P_{tnl}P_{fnl}$$  \hspace{1cm} (3.11)

where $P_{tnl}$ is the thermal non-leakage fraction and $P_{fnl}$ is the fast non-leakage fraction. $P_{tnl}=1$ implies no thermal neutron losses and $P_{fnl}=1$ implies no fast neutron losses.

The thermal non-leakage fraction as defined in eq. (3.12) is strongly dependent on the geometry of the core and can be described from thermal neutron diffusion theory. In particular it is related to the square of a characteristic diffusion area, $L^2$ (eq. (3.13)) (Lamarsh and Baratta, 2001) and the geometric buckling of a reactor, $B_g^2$ (eq. (3.14)) (Duderstadt and Hamilton, 1976).

$$P_{tnl} = \frac{1}{(1 + L^2 B_g^2)}$$  \hspace{1cm} (3.12)

$L^2$ and $B_g^2$ commonly appear in diffusion theory for a finite geometry. The diffusion length, $L$, is the average distance a neutron will travel away from a source before it is absorbed (Duderstadt and Hamilton, 1976) and the diffusion area is simply this length squared. This term, as shown in eq. (3.13), is also related to the neutron diffusion coefficient, $D$, and the neutron macroscopic cross sections for scattering and absorption, $\Sigma_s$ and $\Sigma_a$.
\[ L^2 = \frac{D}{\Sigma_a + \Sigma_s} \quad (3.13) \]

For most existing reactors, the finite geometry of a reactor can be approximated as a finite right cylinder, which has a closed form solution for the geometric buckling term as (Lamarsh and Baratta, 2001):

\[ B_g^2 = \left( \frac{\nu_0}{\tilde{R}} \right)^2 + \left( \frac{\pi}{\tilde{H}} \right)^2 \quad (3.14) \]

where

\[ \nu_0 = \text{the first zero of the zero-order Bessel function of the first kind} \ (\approx 2.405) \]
\[ \tilde{R} = \text{extrapolated radius} \]
\[ \tilde{H} = \text{extrapolated height} \]

The extrapolated radius and height are simply the radius and height, respectively, plus the extrapolated length a neutron will travel once it leaves the core volume before being absorbed.

The fast non-leakage fraction, on the other hand, is derived from neutron slowing down theory (i.e., Fermi age theory). In particular the fast non-leakage fraction, which is valid for a bare uniform reactor, is defined by:

\[ P_{nl} = e^{\left( -\frac{\tau B_g^2}{\tau} \right)} \approx \frac{1}{\left( 1 + \tau B_g^2 \right)} \quad (3.15) \]

where \( \tau [\text{cm}^2] \) is the Fermi age corresponding to the thermalizations.

The slowing down density, resulting from a point source, demonstrates that the age corresponding to thermalization is one-sixth the average (straight line) distance squared from a point where the neutron enters a system with a fast spectrum energy to the point at
which it slows down to thermal energies. For a moderator, the age-to-indium resonance at 1.45 eV is used to approximate $\tau$ (Duderstadt and Hamilton, 1976).

Combining the thermal non-leakage probability and the fast non-leakage probability, a total non-leakage probability, $P_{nl}$, can be approximated. Since $L^2B_g^2$ and $\tau B_g^2$ are both much smaller than one, any term consisting of $L^2\tau B_g^4$ is negligible and an approximate value for $P_{nl}$ can be obtained as:

$$P_{nl} = P_{nl}P_{fnl} = \frac{e^{-\tau B_g^2}}{\left(1 + L^2B_g^2\right)} \approx \frac{1}{\left(1 + L^2B_g^2 + \tau B_g^4\right)} \equiv \frac{1}{\left(1 + M^2B_g^2\right)}$$

In this approximation, $M^2$ is the neutron migration area, which is given in eq. (2.17). $M^2$ can also be interpreted as the $\frac{1}{\sqrt{6}}$ times the root-mean-square distance that a neutron travels from its appearance as a fast fission neutron to its capture as a thermal neutron (Duderstadt and Hamilton, 1976).

$$M^2 = L^2 + \tau$$

Given these parameters and approximations, the non-leakage probability can be calculated using properties of the moderator and the geometry of the reactor. Typical properties for many moderators are reproduced in Table 3.2 and the non-leakage terms are approximately $P_{tnl} \approx 0.95$, $P_{fnl} \approx 0.95$, and $P_{nl} \approx 0.90$ for most thermal reactors.

<table>
<thead>
<tr>
<th>Moderator</th>
<th>Density ($\text{g/cm}^3$)</th>
<th>$D$ (cm)</th>
<th>$\Sigma$ (cm$^{-1}$)</th>
<th>$L$ (cm)</th>
<th>$\tau$ (cm$^2$)</th>
<th>$M$ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{H}_2\text{O}$</td>
<td>1.00</td>
<td>0.16</td>
<td>0.0197</td>
<td>2.85</td>
<td>26</td>
<td>5.84</td>
</tr>
<tr>
<td>$\text{D}_2\text{O}$</td>
<td>1.10</td>
<td>0.87</td>
<td>2.9x10$^{-5}$</td>
<td>170</td>
<td>131</td>
<td>170</td>
</tr>
<tr>
<td>Be</td>
<td>1.85</td>
<td>0.50</td>
<td>1.0x10$^{-3}$</td>
<td>21</td>
<td>102</td>
<td>23</td>
</tr>
<tr>
<td>Graphite</td>
<td>1.60</td>
<td>0.84</td>
<td>2.4x10$^{-4}$</td>
<td>59</td>
<td>368</td>
<td>62</td>
</tr>
</tbody>
</table>

Table 3.2: Neutron diffusion properties for various moderator. Reproduced from (Duderstadt and Hamilton, 1976).
While these parameters can ultimately be used to approximate the neutron losses and thus relate the infinite multiplication factor to the effective multiplication factor, the variations in reactor geometry and operation make this approach unreliable from a fuel cycle standpoint. However, an alternative approach, which is described later, can be used to approximate these and other reactor specific parameters based upon a working fuel composition.

3.1.4 Reactivity Worth Value Definition

The ultimate goal here is not to determine the neutron losses of a reactor, but rather develop a method to load recycled fuel into a reactor. To achieve this goal, a reactivity worth value is first assigned to every isotope of interest by adapting the above guiding approach. In particular, the RWV for each isotope is determined by first assuming an infinite reactor containing only that isotope is present and the infinite multiplication factor, $k_\infty$, derived from the FFF is used. Note in the following equations, that the thermal designation, subscript $t$, has been replaced by a designation, subscript $i$, which represents each isotope. Next, that infinite reactor assumption is corrected for a finite geometry to produce the effective multiplication factor, $k_{eff}$. Since $k_{eff} = 1$ represents a critical reactor, the (positive or negative) deviation from that criticality is represented by $k_{eff} - 1$. This concept is extended to define the RWV for an isotope as:

$$\rho_i \equiv (\epsilon_i p_i P_{nd}) \eta_i f_i - 1 = \frac{\epsilon_i p_i}{1 + M^2 B_g^2} \left( \nu_i \frac{\sigma_{f,i}}{\sigma_{f,i} + \sigma_{a,i}} \right) - 1$$

(3.18)

If no neutrons outside the energy range of interest cause fission events (i.e., $\epsilon_i = 1$) and there is no resonance escape (i.e., $p_i = 1$), then eq. (3.18) reduces to:

$$\rho_i \approx (P_{nd}) \eta_i f_i - 1 = \frac{1}{1 + M^2 B_g^2} \left( \nu_i \frac{\sigma_{f,i}}{\sigma_{f,i} + \sigma_{a,i}} \right) - 1$$

(3.19)
This implies that the RWV for an isotope has a range \(-1 \leq \rho_i \leq \infty\). Equation (3.19) is a valid approximation as \(\varepsilon_i\) and \(p_i\) have competing trends and the product of these two parameters is near unity. Specifically, the coefficient \(P_{nl}\) in eq. (3.19), now referred to as the lumped loss factor, can be determined from a known working composition termed the target composition as is demonstrated next.

3.2 Fuel Loading Approach

The purpose of this loading approach is to determine the mass fraction of transuranic isotopes \(M_{TRU}\) that most closely match a target composition given the available inventory of recyclable material. This is done because in a dynamic fuel cycle with multiple technologies, the exact prescribed recycled fuel recipe would be nearly impossible to meet. In VISION, however, the previously mentioned approach of only using prescribed recipes was used and this approach proved to be unrealistic as non-prescribed isotopes were fictitiously accumulating in the available isotopic inventory. This is not realistic because separating individual isotopes is not reasonable for a recycling facility.

3.2.1 Derivation

Since one of the only real-world controls in determining and loading the composition of recycled (available) fuel is based on elemental separation, the available fuel can most simply be partitioned into a uranium portion and a transuranic portion as shown in eqs. (3.20) and (3.21):

\[
M_U = \frac{\text{Total mass of uranium isotopes}}{\text{Total mass of fuel}} = \frac{\sum_i U_i}{\sum_i U_i + \sum_i TRU_i} \tag{3.20}
\]
where each of these partitions of the available fuel is described by their mass fraction $M_U$ and $M_{TRU}$, respectively. Note that the sum of the mass fractions is unity and serves as one constraint in the fuel loading approach:

$$M_U = 1 - M_{TRU}$$  \hspace{1cm} (3.22)$$

If a different real-world control (other than U and TRU partitions) or a combination of real-world controls (e.g., TRU+U partition and Pu+U partitions) were present, the same procedure would still apply as outlined below. The only difference would be the number of governing equations and resulting closed form solution. For example, in the case of TRU+U partition and Pu+U partition, one reactivity balance equation (which will be demonstrated later) and two partitioned equations relating (1) TRU, Pu, and U (e.g. $M_U + M_{Pu} + M_{TRU} = 1$), and (2) Pu and U (e.g. $\frac{M_{Pu}}{M_{TRU}} = \text{constant}$) would be necessary.

After the (available) fuel is partitioned into a U group and a TRU group, the dichotomized composition of the available inventory of isotopes is also established. This is the relative composition of each partition reported as a fraction of the partitioned amount as:

$$m_{U,i} = \frac{\text{Mass of uranium isotope } i \text{ present}}{\text{Total mass of uranium isotopes}} = \frac{U_i}{\sum_i U_i}$$  \hspace{1cm} (3.23)$$

$$m_{TRU,i} = \frac{\text{Mass of transuranic isotope } i \text{ present}}{\text{Total mass of transuranic isotopes}} = \frac{TRU_i}{\sum_i TRU_i}$$  \hspace{1cm} (3.24)$$
where $m_{Ui}$ is the dichotomized composition for uranium isotope $i$ and $m_{TRUi}$ is the dichotomized composition of transuranic isotope $i$. This is simply the mass fraction of a given isotope in its partitioned group. For example, the amount of $^{238}\text{U}$ divided by the total amount of uranium would be the dichotomize composition of $^{238}\text{U}$. Similarly, the amount of $^{241}\text{Pu}$ divided by the total transuranic amount would represent the dichotomized composition of $^{241}\text{Pu}$. The dichotomized composition of $^{238}\text{U}$ is then identified as $m_{U-238}$ and the dichotomized composition of $^{241}\text{Pu}$ is identified as $m_{Pu-241}$.

Next, the partitioned composition of a fuel composition that works for a given reactor technology is established in the same manner. This working composition is termed the target composition because it represents a desired composition for a specific reactor. From this target composition, reactor specific parameters, such as the lumped loss parameter, can be determined as will be demonstrated later. The goal here, however, is to use this composition as a target for which an adjusted composition, consisting of the isotopes from the available inventory, attempts to match. Now, the target composition is partitioned into a uranium portion ($M_{TRU}^T$) and a transuranic portion ($M_{TRU}^T$) just like the available fuel composition.

Next, a relationship between these terms is established. It is important to emphasize that the purpose here is to determine $M_{TRU}$ (and $M_U$) and that the product of partitioned and dichotomized composition represents the mass fraction of an isotope in the fuel:

$$m_{U, M_U} = \frac{U_i}{\sum_i U_i} \left( \frac{\sum_i U_i}{\sum_i U_i + \sum_i \text{TRU}_i} \right) = \frac{U_i}{\sum_i U_i + \sum_i \text{TRU}_i}$$  \hspace{1cm} (3.25)
Given that the mass of the target fuel and the mass of recycled (available) fuel, eq. (3.25) and eq. (3.26) can be written as:

\[
m_{U_i} M_U = \sum_i M^T_{TRU_i} \frac{U_i}{U^T_i} \tag{3.27}
\]

\[
m_{TRU_i} M_{TRU} = \frac{M^T_{TRU_i} \sum_i TRU_i}{\sum_i TRU_i} \tag{3.28}
\]

where \( U^T_i \) represents the mass of uranium isotope \( i \) present in the target composition and \( TRU^T_i \) represents the mass of transuranic isotope \( i \) present in the target composition.

Also note that if the target and the available compositions are identical (i.e., \( M_{TRU} = M^T_{TRU} \) and \( M_U = M^T_{U} \)), then the dichotomized composition identity is expressed (eqs. (3.23) and (3.24)). However, since these compositions are not likely to be identical, each isotope can be weighted by the RWV such that the reactivity deficit caused by a non-prescribed isotope can be corrected for by a more highly reactive isotope and vice versa. This eliminated the need for isotope matching but rather RWV matching.

Finally, rearranging eqs. (3.27) and (3.28) and weighting these equations with the RWV for each isotope \( i \) (denoted as \( \rho_i \)), a RWV balance equation is obtained:

\[
0 = \sum_U \frac{\rho_U m_U}{M_U} M_U + \sum_{TRU} \frac{\rho_{TRU} m_{TRU}}{M^T_{TRU}} M_{TRU} \tag{3.29}
\]

Equation 3.29 and 3.22 can then be combined to solve for the partition of TRUs that can be loaded into the same reactor as the target recipe, given that this composition’s transuranic isotopic ratios matches the available composition.
In other words, this is the mass fraction of TRUs that can be loaded into the reactor and will behave in a similar manner from a neutron population (energy generation) standpoint.

In short, by determining $M_{TRU}$, the adjusted composition is loaded as fuel. This adjusted composition is determined from the available composition and the target composition. The available composition is the fuel composition that is available to be made into fuel and the target composition is the desired composition. The available and target compositions will differ as some isotopes following reprocessing (recycling) will be more or less abundant than others. It is for this reason that a fuel loading approach is necessary to adjust if the prescribed target recipe is unavailable.

### 3.2.2 Reactor Dependent Losses and Uncertainty

It is necessary to introduce the description of fuel loading before introducing reactor dependent losses because determining such losses can easily be illustrated given the governing RWV balancing equation (eq. (3.29)). If this equation is applied to the target composition, eq. (3.29) would reduce to eq. (3.31) given that $M_{TRU} = M_{TRU}^T$ and $M_U = M_U^T$:

$$0 = \sum_U \rho_U m_U^T + \sum_{TRU} \rho_{TRU} m_{TRU}^T$$

where $m_{TRU}^T$ and $m_{TRU}^T$ represent the target’s dichotomized composition, respectively (i.e., mass fractions, see eq. (3.23) and (3.24)) and as such, the sum of each dichotomized composition is unity:
\[
\sum_{U} m_{U}^{T} = 1 \quad \text{and} \quad \sum_{TRU} m_{TRU}^{T} = 1 \tag{3.32}
\]

Substituting the approximation of RWV, eq. (3.20), into the above equation and rearranging yields an approximation for the neutron losses of a reactor (i.e., neutron non-leakage probability) as:

\[
P_{\text{nl}} = \frac{\sum_{U} m_{U}^{T} + \sum_{TRU} m_{TRU}^{T}}{\sum_{U} \eta_{U} f_{U} m_{U}^{T} + \sum_{TRU} \eta_{TRU} f_{TRU} m_{TRU}^{T}} = \frac{2}{\sum_{U} \eta_{U} f_{U} m_{U}^{T} + \sum_{TRU} \eta_{TRU} f_{TRU} m_{TRU}^{T}} \tag{3.33}
\]

Using eqs. (3.20), (3.23), (3.24), (3.30), and (3.33) with a known target composition and a known available composition, an adjusted composition can now be determined. This adjusted composition will closely match the target composition but contain isotopes that are present in the available composition but are not present in the prescribed recipe of the target composition.

Unfortunately, this approach relies strongly on the accuracy of isotope specific parameters, namely neutron cross sections. Since these one-group cross sections are strongly temperature and neutron energy spectrum dependent, significant error can be introduced based upon inaccurate cross sections. Specifically, the error in the reactivity worth value, \( U_{\rho} \), from only uncertainty in the cross-sections, \( U_{\sigma_f} \) and \( U_{\sigma_c} \), is given by:

\[
\left( \frac{U_{\rho}}{\rho} \right)^2 = \left( \frac{1}{\rho} \frac{d\rho}{d\sigma_f} \right)^2 \left( U_{\sigma_f} \right)^2 + \left( \frac{1}{\rho} \frac{d\rho}{d\sigma_c} \right)^2 \left( U_{\sigma_c} \right)^2 \tag{3.34}
\]

which simplifies to:

\[
\left( \frac{U_{\rho}}{\rho} \right) = \left( \frac{\sigma_f + 2\sigma_c}{\sigma_f (\sigma_f + \sigma_c)} \right) \sqrt{\left( U_{\sigma_f} \right)^2 + \left( U_{\sigma_c} \right)^2} \tag{3.35}
\]
As can easily be seen in eq. (3.35), the uncertainty in the reactivity worth values is even
greater than just the uncertainties in the cross sections. Model dependence on accurate
neutron cross sections, however, is not a new surprise. All reactor physics, activation,
and transmutation calculations rely on accurate neutron cross sections and it is for this
reason that experimental research to improve the accuracy of neutron cross sections will
be ongoing.

3.3 Example of the Methodology

In short, the above approach can be used to approximate a recycled fuel
composition especially for the case when the available composition does not match the
prescribed target composition. This may be better understood graphically as shown in
Fig. 3.2.

![Available Composition Domain](image)

Figure 3.2: The available composition spans a domain which typically does not include the target
composition due to the presence of non-recipe prescribed isotopes

As in the illustration, the available composition can be thought of as spanning a domain
which represents the recycled fuel compositions that can be made. In many cases, the
target composition does not fall within that domain because of the presence (or absence)
of non-recipe prescribed isotopes in the available composition. The goal behind any fuel
loading approach is to find the point in the available composition domain that is closest to
the target composition. To do this, the presented approach relies on assigning reactivity
worth value to each isotope and then balancing that worth such that if one isotope is not
present its reactivity can be compensated for by another isotope. Utilizing this approach,
the amount of transuranic material that can be loaded is then determined.

3.3.1 Procedure

The proposed method is summarized in the following computational steps:

1. Compute the neutron utilization factor, $f$ (eq. (3.4)), and the reproduction
   factor, $\eta$ (eq. (3.5)), for each isotope of interest.
2. Compute the dichotomized compositions of the available composition, $m_U$
   and $m_{TRU}$, and the target compositions, $m^T_u$ and $m^T_{TRU}$ (eqs. (3.23) and
   (3.24)).
3. Compute the neutron losses from the target composition, $P_{nl}$ (eq. (3.33))
4. Compute the RWV of each isotope, $\rho$ (eq. (3.19)).
5. Compute the partitioned uranium, $M^T_U$, and transuranic, $M^T_{TRU}$, portions
   of the target composition (eqs. (3.20) and (3.21)).
6. Compute the partitioned transuranic $M^T_{TRU}$, and uranium, $M_U$, portions of
   the adjusted composition (eqs. (3.30) and (3.22)).

This method is straightforward and surprisingly simple. It only relies on isotopic
parameters and information hidden within the composition of the target fuel. In addition,
this method also provides a method for approximating the infinite multiplication factor of
the resulting adjusted composition using the FFF (eq. (3.2)).
3.3.2 Results

To illustrate the method proposed here, an example using VISION parameters is discussed. The selected parameters are (1) typical target fuel recipes, (2) available compositions, (3) neutron cross sections, and (4) the average number of neutrons per fission (for each isotope). The one-group cross sections and the average number of neutrons per fission (for each isotope) are given in Table 3.1 and in Appendix A.2. From these parameters, the product of the neutron reproduction factors and the utilization factors, $\eta f$, can be determined as shown in Table 3.3.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Thermal Reactor</th>
<th>Fast Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\eta_f$</td>
<td>$\eta_f$</td>
</tr>
<tr>
<td>$^{234}\text{U}$</td>
<td>7.31E-02</td>
<td>9.93E-01</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>1.89E+00</td>
<td>1.89E+00</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td>6.80E-02</td>
<td>4.86E-01</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>3.15E-01</td>
<td>4.24E-01</td>
</tr>
<tr>
<td>$^{237}\text{Np}$</td>
<td>6.92E-02</td>
<td>6.44E-01</td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>3.33E-01</td>
<td>1.79E+00</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>1.84E+00</td>
<td>2.28E+00</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>4.38E-02</td>
<td>1.46E+00</td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>2.25E+00</td>
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<td>6.73E-02</td>
<td>1.43E+00</td>
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<td>1.71E+00</td>
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<td>2.85E+00</td>
<td>3.65E+00</td>
</tr>
<tr>
<td>$^{252}\text{Cf}$</td>
<td>2.92E+00</td>
<td>2.99E+00</td>
</tr>
</tbody>
</table>

Table 3.3: The product of the reproduction factor and the utilization factor for select isotopes based upon one-group cross sections (see Table 3.1) for two given reactor types.

Table 3.3 is also presented graphically in Fig. 3.3. From this figure, transmutation characteristics can be determined. Namely those isotopes whose $\eta_f$ is greater than one are considered useful fuel isotopes for a given reactor type. Note that some isotopes that
are not useful as fuel in one reactor type may prove useful as fuel in another reactor type. This demonstrates that the RWV for each isotope (since RWV is proportional to $\eta f$) is dependent on the reactor type because the neutron interactions are energy dependent. This also demonstrates that recycling isotopes in certain reactor types would be better than others because those certain isotopes can be used as fuel isotopes in one reactor type where as they are activation isotopes in another reactor type.

Figure 3.3: $\eta f$ for select isotopes for thermal and fast reactors. $\eta f$ greater than one indicates that the isotope is a useful fuel for that reactor type.
The rest of the parameters can be determined from VISION recipes. The target thermal composition is the VISION recipe composition that corresponds to a mixed-oxide NpPuAm used in the first pass of a recycling strategy (MOX-NpPuAm cycle 1). The target fast composition is the VISION recipe that corresponds to the 0.50 conversion ratio fast reactor that is fed by a light water reactor used fuel with a 50 GWe/MTU burnup (CR-0.50, UOX-50). The available thermal composition is similar to the target composition except that it lacks Am. This can be used to demonstrate how a change in separation strategies can still be handled by this approach. These compositions are summarized in Table 3.4.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Target Composition (mass fraction)</th>
<th>Available Composition (mass fraction)</th>
<th>Target Composition (mass fraction)</th>
<th>Available Composition (mass fraction)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><strong>Thermal Reactor</strong></td>
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<td><strong>Fast Reactor</strong></td>
<td></td>
</tr>
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<td></td>
<td></td>
<td></td>
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<td>$^{234}$U</td>
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<tr>
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<td>0</td>
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</table>

Table 3.4: Example available and target compositions for thermal and fast reactor recycling strategies adopted from the VISION fuel recipes.

Given the target compositions and the isotope specific parameters in Table 3.1, the neutron losses for a specific reactor can be predicted using eq. (3.33). Using this method, the reactor specific losses and the infinite multiplication factor, $k_\infty$, are approximated. The results are produced in Table 3.5. In this table, the actual values...
represent a known static composition that can be used to compare against these predicted values that were produced using the RWV approach. $k_\infty$ is a gross approximation as it does not consider neutron losses (absorption) in structure materials. In addition, for the fast reactors, the majority of fissions occur at high (fast) energies, and the simplifying assumption that for a fast fission factor, $\varepsilon$, is approximately unity is not valid. This would increase $k_\infty$ such that $P_{nl}k_\infty$ is just slightly greater than the values reported in Table 3.5.

<table>
<thead>
<tr>
<th></th>
<th>Thermal Reactor</th>
<th>Fast Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{nl}$</td>
<td>0.767029</td>
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</tr>
<tr>
<td>$k_\infty$</td>
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<tr>
<td>Actual</td>
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</tr>
<tr>
<td></td>
<td>1.359</td>
<td>1.518</td>
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</table>

Table 3.5: Typical loss factors and infinite multiplication factors for the thermal and fast reactors.

Given these loss factors RWVs can be assigned to each isotope as described by eq. (3.19). The isotopic trend of these RWVs is similar to the trend depicted in Fig. 3.3, however the magnitude differs by the loss factor (with an offset of 1). The RWVs for each isotope in this example can be found in Table 3.6.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Thermal Reactor</th>
<th>Fast Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Reactivity Worth Value, $\rho$</td>
<td>Reactivity Worth Value, $\rho$</td>
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<tr>
<td>$^{234}$U</td>
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<td>-0.409</td>
</tr>
<tr>
<td>$^{242m}$Am</td>
<td>2.556</td>
<td>1.492</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>-0.947</td>
<td>-0.322</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
<td>-0.592</td>
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<td>1.716</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>-0.681</td>
<td>0.228</td>
</tr>
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<td>$^{245}$Cm</td>
<td>3.452</td>
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</tr>
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<td>-0.421</td>
<td>0.897</td>
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<td>-1.000</td>
<td>-1.000</td>
</tr>
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<td>$^{249}$Cf</td>
<td>3.120</td>
<td>2.075</td>
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<td>$^{250}$Cf</td>
<td>-0.982</td>
<td>1.473</td>
</tr>
<tr>
<td>$^{251}$Cf</td>
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<td>2.140</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td>2.955</td>
<td>1.572</td>
</tr>
</tbody>
</table>

Table 3.6: Reactivity worth values for selected isotopes for given reactor types.

After determining the RWVs, the partitioned transuranic portion, $M_{TRU}$, and uranium portion, $M_U$, of the adjusted composition can be determined by using eqs. (3.30) and (3.22), respectively. In addition, the partitioned uranium portion, $M_U^F$, and
transuranic portion, $M^{T}_{TRU}$, of the target composition can also be determined for comparison. These values can be found in Table 3.7.

<table>
<thead>
<tr>
<th></th>
<th>Thermal Reactor</th>
<th>Fast Reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M^{T}_{U}$</td>
<td>82.59%</td>
<td>70.54%</td>
</tr>
<tr>
<td>$M^{T}_{TRU}$</td>
<td>17.41%</td>
<td>29.46%</td>
</tr>
<tr>
<td>$M_{U}$</td>
<td>Predicted 84.38%</td>
<td>Predicted 68.54%</td>
</tr>
<tr>
<td></td>
<td>Actual 86.89%</td>
<td></td>
</tr>
<tr>
<td>$M_{TRU}$</td>
<td>Predicted 15.62%</td>
<td>Predicted 31.46%</td>
</tr>
<tr>
<td></td>
<td>Actual 13.11%</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.7: Partitioned uranium and transuranic portions in target fuel ($M^{T}_{U}$ and $M^{T}_{TRU}$) and adjusted fuel compositions ($M_{U}$ and $M_{TRU}$).

These values represent the amount of transuranic and uranium materials that can be loaded into a reactor as the adjusted composition. The adjusted composition is what closely matches the target composition given that the isotopes in the available composition are those used for fuel. In other words, the available isotopes are loaded as recycled fuel based on two portions, a uranium portion and a transuranic portion. In this example for thermal reactors, the transuranic portion constitutes 15.62% and the uranium portion 84.38%. The isotopes in the uranium portion are the uranium isotopes in the available composition and the isotopes in the transuranic portion are the transuranic isotopes in the available composition (weighted to these percentages).

Weighting the available composition (Table 3.4) to the transuranic and uranium percentages yields (Table 3.6) the adjusted composition, which is used as the recycled fuel. For this example, the adjusted composition is expressed in Table 3.8.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Predicted</th>
<th>Actual</th>
<th>Predicted</th>
<th>Actual</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Uranium</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{234}$U</td>
<td>1.69E-04</td>
<td>1.74E-04</td>
<td>1.30E-04</td>
<td>0</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>6.93E-03</td>
<td>7.14E-03</td>
<td>5.78E-03</td>
<td>1.36E-03</td>
</tr>
<tr>
<td>$^{236}$U</td>
<td>5.16E-03</td>
<td>5.31E-03</td>
<td>2.78E-03</td>
<td>0</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>8.32E-01</td>
<td>8.56E-01</td>
<td>6.77E-01</td>
<td>6.90E-01</td>
</tr>
<tr>
<td><strong>Transuranic</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>7.86E-03</td>
<td>6.60E-03</td>
<td>1.20E-02</td>
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<tr>
<td>$^{238}$Pu</td>
<td>3.90E-03</td>
<td>3.28E-03</td>
<td>3.10E-03</td>
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</tr>
<tr>
<td>$^{239}$Pu</td>
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<td>1.67E-01</td>
<td>1.64E-01</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
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<td>7.38E-02</td>
<td>7.26E-02</td>
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<tr>
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</tr>
<tr>
<td>$^{242}$Pu</td>
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<td>1.54E-02</td>
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<td>3.02E-02</td>
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<td>1.01E-05</td>
<td>1.00E-05</td>
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<td>2.57E-03</td>
<td>2.56E-03</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
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<td>0</td>
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<td>2.43E-08</td>
</tr>
<tr>
<td>$^{243}$Cm</td>
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<td>0</td>
<td>3.78E-06</td>
<td>3.76E-06</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
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<td>2.29E-04</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
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<td>3.35E-05</td>
<td>3.34E-05</td>
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<tr>
<td>$^{246}$Cm</td>
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<td>3.13E-06</td>
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<tr>
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</tr>
<tr>
<td>$^{251}$Cf</td>
<td>0</td>
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<td>1.01E-12</td>
<td>0</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td>0</td>
<td>0</td>
<td>2.05E-16</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 3.8: Adjusted fuel composition based upon the RWV approach for select isotope of given reactor types.
3.3.3 Summary

The approach presented in Section 3.2 to determine the homogenized composition of recycled fuel is novel. It relies strictly on an isotopic target composition, an isotopic available composition, and isotope specific parameters. This is a new approach to adjust the recycled fuel to best match a target composition based upon equalizing isotopes by assigning reactivity worth value to each isotope. Next, this reactivity worth value is balanced, so that the presence (or absence) of non-recipe prescribed isotopes in the available composition is corrected for by other isotopes. Since isotopic separation of the used fuel is not realistic, this balance occurs on a uranium and transuranic partitioned basis.

The proposed approach is unique, because it requires no additional reactor specific information. All necessary parameters are extracted from the homogenized target fuel composition. This approach can benefit fuel cycle models (such as VISION) because it liberates such models from having to track reactor specific parameters through time for dynamic simulations. This makes the data management behind large fuel cycle models more manageable.

In addition, this approach has several other attractive attributes for application in fuel cycle models. One such attribute is that it does not require an a priori assumption of the effective neutron multiplication factor, $k_{eff}$. In order to account for neutron losses in a particular reactor while loading the recycled fuel, it is conventionally necessary to know this parameter. By approximating the neutron losses based upon this reactivity worth value approach, such a priori knowledge is not necessary. Also, it preserves the enrichment of the uranium channels in the reactor (i.e., the $^{235}$U enrichment in the
uranium portion is maintained). Since the fuel in the reactor will (as it is envisioned) be
dichotomized into uranium and transuranic channels, the relative compositions within
each channel cannot change since isotopic separation is not realistic for spent fuel. This
approach maintains the relative compositions within each channel only by adjusting the
dichotomized proportions (and thus the amount of transuranic material loaded into a
reactor).

The benefits of this approach include computational simplicity as it provides a
way to approximate reality for modeling purposes. This approach is in no way intended
for design optimization of fuel loading, but it does provide a geometry independent way
to approximate how much transuranic material can realistically be loaded into a reactor.
In addition, it allows for dynamic adjustment of the static fuel input recipes used within
fuel cycle models.
CHAPTER 4

TRANSMUTATION APPROACH

After recycled fuel is loaded into a reactor, it will be consumed to produce power and electricity. The heat released from nuclear fission and subsequent radioactive decay is used (via primary or secondary heat exchangers) to produce steam, which drives turbines connected to generators to produce electricity. However, the process by which the material in the reactor undergoes fission and decay is of importance from a fuel cycle standpoint. If the reactor is treated like a simple plant with fuel coming in and waste going out, it is natural to be concerned about the waste stream. In particular, a known composition enters the plant and a different composition leaves the plant as waste. In a closed fuel cycle, this waste is recycled and therefore knowing the composition of the discharged fuel is necessary. Also, if a variable recycled fuel composition is used in a dynamic model, a variable discharged composition is necessary instead of relying on static discharge recipes. Determining the composition of discharged fuel requires performing detailed transmutation calculations in a timely manner.

Transmutation algorithms exist and have been integrated into various burnup and depletion codes. One of the most commonly used codes is ORIGEN. This code was developed at Oak Ridge National Laboratory to compute the composition and
radioactivity of fission and activation products in fuel and structural materials. ORIGEN has seen many development stages and is integrated into many computation suites. In particular, a PC-based graphic user interface version known as ORIGEN-ARP and a version known as ORIGEN-S, which is integrated into the SCALE, have been developed. In general, ORIGEN relies on reactor specific parameters (temperature, geometry, flux, and cross sections) in various sub-modules to determine conditions in a reactor. For individual reactors over moderate time scales, ORIGEN executes in a timely manner. However, when attempting to implement ORIGEN into larger fuel cycle models with hundreds of reactors over several decades, the computational resources are not sufficient to perform full burnup and depletion calculations using ORIGEN.

For a fuel cycle model such as VISION, the fuel composition is of primary concern rather than the radioactivity and activation of adjacent structural materials. For VISION, it is desired that the discharged fuel composition be based solely upon the fresh fuel composition. However, knowing just the fresh fuel composition is not enough to approximate the discharged composition. Since that fresh fuel composition can be consumed at different rates and under different reactor power constraints, the length of time the fuel is irradiated and the neutron flux, which is related to the power, is also necessary. In addition, isotope specific parameters such as neutron cross sections and the average number of neutrons per fission are necessary. The neutron cross section, however, is not reactor independent as it relies on temperature and the neutron spectrum of the reactor as discussed in Section 3.1.3.

This chapter outlines an approach based upon the ORIGEN-S algorithm (Hermann and Westfall, 1998) that can be used within fuel cycle models such as
VISION. Hermann and Westfall (1998) are credited for developing the basis of this approach but changes have been made to meet the problem requirements of fuel cycle simulations. The required input parameters for this transmutation approach utilize a minimum set of input parameters and this approach approximates the appropriate composition of a discharged fuel from a fresh fuel composition. This differs from the ORIGEN-S algorithm since only a sub-set of isotopes is considered, specifically those isotopes tracked in the VISION model. In addition, the flux spectrum adjustment modules included in ORIGEN-S are not utilized in this transmutation approximation. Additional approximations have been made to estimate reactor specific parameters as discussed in this chapter.

4.1 Transmutation Governing Equation

Transmutation is the temporal change of each isotope as each isotope undergoes a transition in the presence of neutrons. Specifically, neutron induced fission and activation are considered. During fission, heavier isotopes fission into lighter isotopes and during activation, isotopes absorb a neutron and become a slightly heavier isotope. Now after both fission and activation, the residuals of that reaction tend to be unstable and undergo radioactive decay into yet another isotope. From these processes, the generation and destruction of each isotope can be described by a rate equation:

\[
\frac{dN_i}{dt} = \sum_j \gamma_{f,j} N_j \phi + \sigma_{e,j-1} N_{i-1} \phi + \lambda_i N_i' - \sigma_{f,j} N_i \phi - \sigma_{e,j} N_i \phi - \lambda_i N_i
\] (4.1)

where \( N_i \) is the number of isotopes \( i \), and \( \phi \) is the average neutron flux. Each term of this governing equation has its own significance and represents either a production or destruction term for a particular isotope.
The first term (eq. (4.2)), represents the rate of production of isotope \( i \) from the fission of all other isotopes \( j \):

\[
\sum_j \gamma_{ij} \sigma_{f,j} N_j \phi
\]  

(4.2)

where \( \gamma_{ij} \) represents the fission yield from isotope \( j \) to isotope \( i \), \( \sigma_{f,j} \) represents the one-group neutron fission cross section of isotope \( j \), and \( N_j \) represents the number of \( j \) isotopes present. Here, several isotopes can fission to produce the residual fragment that is isotope \( i \). This term is the dominant source for the production of lighter isotopes.

The second term (eq. (4.3)) represents the rate of production of isotope \( i \) from the radiative capture of a neutron from an isotope whose nucleus contains the same number of protons but one less neutron (\( i-1 \)):

\[
\sigma_{c,i-1} N_{i-1} \phi
\]  

(4.3)

where \( \sigma_{c,i-1} \) represents the one-group radiative capture cross section of isotope \( i-1 \), and \( N_{i-1} \) represents the number of \( i-1 \) isotopes present. This term is the dominant source of high-\( Z \) isotopes that are not initially present in the fuel.

The third term (eq. (4.4)) represents the rate of production of isotope \( i \) from the radioactive decay of other isotopes:

\[
\lambda_i N_i'
\]  

(4.4)

where \( \lambda_i \) is the decay constant \( \left( \lambda = \ln 2 / T_{1/2} \right) \) of isotope \( i' \) and \( N_i' \) represents the number of \( i' \) isotopes. This parameter is representative of the complicated decay chains. These chains, however, have been simplified as only alpha and beta decay have been considered. Additional decay modes exists (i.e., internal transition, electron capture, and positron emission); however, only these two modes are considered in the current study.
In addition, when an isotope undergoes alpha decay, a helium nucleus \(^{(4}\text{He})\) is produced since an alpha particle is simply a high energy helium nucleus.

The fourth term (eq. (4.5)) represents the rate of destruction of isotope \(i\) from the fission of that isotope:

\[
\sigma_{f,i} N_i \phi
\]  

(4.5)

where \(\sigma_{f,i}\) represents the one-group neutron fission cross section of isotope \(i\), \(N_i\) represents the number of \(i\) isotopes present, and \(\phi\) represents the average neutron flux. This term is the dominant destruction mode of fissile isotopes such as \(^{235}\text{U}\).

The fifth term (eq. (4.6)) represents the rate of destruction of isotope \(i\) from the radiative capture of neutrons:

\[
\sigma_{c,i} N_i \phi
\]  

(4.6)

where \(\sigma_{c,i}\) represents the one-group radiative capture cross section of isotope \(i\), \(N_i\) represents the number of \(i\) isotopes present, and \(\phi\) represents the average neutron flux. This term is the dominant destruction mode of fertile material as it become activated and then decays into fissile material.

The final term (eq. (4.7)) represents the rate of destruction of isotope \(i\) from the radioactive decay of that isotope:

\[
\lambda_i N_i
\]  

(4.7)

where \(\lambda_i\) is the decay constant of isotope \(i\). This term is a prominent mode of destruction for all unstable nuclides since this mode occurs without the presence of neutrons.

In addition, other reactions such as \((n,2n)\), \((n,3n)\), \((n,4n)\), \((z,n1)\), \((z,n2)\), \((z,n3)\), etc. can be incorporated, but the contributions from these reactions tend to be small except in the case of fast reactors where the \((n,\_n)\) reactions become more important. If additional
reactions are to be included, a production and destruction term for that reaction is simply added to the right hand side of the governing equation, eq. (4.1). The difficulty, however, in including some of these additional reactions is developing one-group neutron cross sections for these reactions for each reactor type.

There are two conservation equations that can be extracted from eq. (4.1). By integrating over all isotopes, the number of isotopes that undergo a fission event is half the number of isotopes that are produced from fission events since each fission event produces only two residuals. In addition, the number of isotopes that are generated by the decay is equal to the number of isotopes that are lost by decay. Also, from this governing equation it is clear to see that the total number of nuclides will increase in the presence of a neutron flux. At least two nuclei are generated for each fission event and alpha decay generates an additional helium nucleus. Also, the average Z-number will tend to decrease resulting from fission and radioactive decay.

Equation 4.1 is then written for all isotopes of interest resulting in a set of coupled first order nonlinear ordinary differential equations. This set of equations can then be easily solved and applied over the time period of interest. A difficulty arises when the solution process has to be conducted in a timely manner such as between time steps in a fuel cycle model. Also, with a large set of isotopes, the number of differential equations and thus the computational effort increases. To deal with these limitations, a less computationally intensive approach that applies a matrix exponentiation solutions with a recursive algorithm, has been developed.
4.2 Transmutation Approach

The species rate equation, eq. (4.1), can be written in an alternative manner, pivoting around the isotope of interests with off diagonal terms representing the transformation of one isotope into another as:

$$\frac{dN_i}{dt} = \sigma_{c,i-1} N_{i-1} \phi - \left( (\sigma_{f,j} + \sigma_{c,j}) \phi + \lambda_i \right) N_i + \lambda_i' N_i' + \sum_j \gamma_{ij}\sigma_{f,j} N_j \phi$$  (4.8)

This equation can then easily be written into matrix notation as:

$$\frac{dN}{dt} = \left[ \begin{bmatrix} \sigma_{c,i-1} \phi \\ -\sigma_{f,j} - \sigma_{c,j} - \lambda_i \\ \vdots \\ \gamma_{ij} \sigma_{f,j} N_j \phi \end{bmatrix} \right] \left[ \begin{bmatrix} N_{i-1} \\ N_i \\ \vdots \\ N_{j-1} \\ N_j \\ \vdots \\ N_{j=n} \end{bmatrix} \right]$$  (4.9)

Similarly the system of equations can be represented as:

$$\frac{dN_1}{dt} = \left[ \begin{bmatrix} a_{11}, a_{12}, \cdots, a_{1n} \end{bmatrix} \right] \left[ \begin{bmatrix} N_1 \\ N_2 \\ \vdots \\ N_n \end{bmatrix} \right]$$  (4.10)

or as:

$$\dot{\mathbf{N}} = \mathbf{A} \mathbf{N}$$  (4.11)

where \( \mathbf{N} \) is a vector with elements representing the number of nuclides of a specific isotope, \( \dot{\mathbf{N}} \) is a vector with elements representing the time rate of change of the number of nuclides of a specific isotope, and \( \mathbf{A} \) represents the square transition matrix of
elements $a_{ij}$, which are the coefficients elements in eq. (4.9). The transition elements simply define the transition from isotope $j$ to isotope $i$ (column to row) and the diagonal element represents the destruction of isotope $i$.

4.2.1 Recursive Matrix Exponentiation

The solution of this first-order matrix ordinary differential equation takes the well known form:

$$N(t) = \exp(At)N_0$$

(4.12)

where $t$ is just a scalar representing the time lapse (i.e., irradiation or decay time) of interest, and $N_0$ is the initial composition at time $t=0$. The matrix exponential form can then be approximated through a Taylor expansion as:

$$\exp(At) \approx I + At + \frac{(At)^2}{2} + \ldots + \sum_{m=0}^{\infty} \frac{(At)^m}{m!}$$

(4.13)

While the form of this solution is straightforward, the computational power required to track numerous isotopes while the transition matrix is summed and raised to several exponents is expensive. For this reason, a recursive matrix exponentiation approach is developed. To illustrate this approach, the solution, eq. (4.12), can alternatively be written as:

$$N(t) = \begin{bmatrix} I + At + \frac{(At)^2}{2} + \ldots \end{bmatrix} N_0 = \left[ \sum_{m=0}^{\infty} \frac{(At)^m}{m!} \right] N_0$$

(4.14)

Focusing on a single isotope (or row) and expanding the matrix multiplication, the solution for the $i^{th}$ isotope can be written as:

$$N_i(t) = N_{i0} + t \sum_j a_{ij}N_{j0} + \frac{t^2}{2} \sum_k \left[ a_{ik}t \sum_j a_{kj}N_{j0} \right] + \frac{t^3}{3} \sum_m \left[ \sum_k a_{lm}t \sum_j a_{kj}N_{j0} \right] + \ldots$$

(4.15)
where the indices on the summation range from 1 to the size of the transition matrix $A$ containing the elements $a_{xy}$.

From this form of the solution, a recursive element can be seen. In particular, once a summation is calculated the result is later used in the next summation. Recursion is a handy programming tool as it reduces the number of terms that must be stored in memory. In particular, only 3 vectors must be stored in memory: (1) the initial composition, (2) a recursive vector and (3) the solution vector. One isotope (or row) of the vectors is expressed in eqs. (4.16)-(4.18):

$$C_i^0 = N_{0_i} \quad (4.16)$$

$$C_i^{n+1}(t) = \frac{t}{n+1} \sum_j a_{ij} C_j^n(t) \quad (4.17)$$

$$N_i(t) = \sum_{n=0}^{\infty} C_i^n(t) \quad (4.18)$$

or in vector notation as in eq. (4.19)-(4.21):

$$\mathbf{C}_i^0 = \mathbf{N}_{0_i} \quad (4.19)$$

$$\mathbf{C}_i^{n+1}(t) = \frac{t}{n+1} \mathbf{A} \mathbf{C}_i^n(t) \quad (4.20)$$

$$\mathbf{N}(t) = \sum_{n=0}^{\infty} \mathbf{C}_i^n(t) \quad (4.21)$$

This recursive matrix exponentiation approach is considerably less computationally intensive and can easily be performed on demand. However, it is not computationally possible to evaluate this infinite series so that a partial summation (using $S_k$ terms) is used as an approximation:

$$\sum_{n=0}^{\infty} \mathbf{C}_i^n(t) = \lim_{S_k \to \infty} \sum_{n=0}^{S_k} \mathbf{C}_i^n(t) \quad (4.22)$$
Determining the number of terms in the partial summation is no trivial task since isotopic concentrations of a few million nuclides (~10^{-18} kg) are significant. Accounting for these small amounts is influenced by both the partial summation and the computational precision of the computer. The computational precision of the computer is fixed and the error associated with the partial sum depends on the elements of the transition matrix and the time scale of interest since it is assumed that flux parameters are constant over the time scale of interest as will be discussed later. This algorithm, similar to the ORIGEN-S algorithm (Hermann and Westfall, 1998), depends on the norm of the transition matrix (Lapidus and Luss, 1967) to determine the machine precision and number of terms in the partial sum:

\[
[A] \equiv \min \left\{ \max \left( \sum |a_{ij}| \right), \max \left( \sum |a_{ji}| \right) \right\}
\] (4.23)

In other words, the norm of the transition matrix is the lesser of (1) the maximum column of value of the summation of absolute values of the row elements or (2) the maximum row of value of the summation of absolute values of the column elements.
4.2.2 Machine Precision

A very rapidly changing isotope will have large transition elements. Those isotopes with short half-lives or large neutron cross sections will not be present long before they are transformed into another isotope. The transition element corresponding to the destruction of an isotope are the diagonal elements, -$d$, which is simply the negative summation of the decay constant and the product of the absorption (fission plus radiative capture) cross section and the average flux, as shown in eq. (4.8), for that isotope. This value is sometimes referred to as the effective half-life when expressed as:

$$ T_{1/2, eff} = \frac{\ln 2}{d} $$ (4.24)

It is sufficient to say that an isotope has essentially vanished when 0.1% of the initial amount remains:

$$ \frac{N_i(t)}{N_0} = 0.001 = e^{-(\sigma_f + \sigma_s)\phi \lambda t} = e^{-dt} $$ (4.25)

This is equivalent to saying that the effective half-life is 10% of the time interval, $t$, or that 10 effective half-lives have passed in that time interval:

$$ \left( \frac{1}{2} \right)^{10} = \frac{1}{1024} \approx 0.001 $$ (4.26)

Also, applying the conservation equation that each fission will produce only two residual isotopes, the summation of the absolute value of each column will not exceed $2d$:

$$ 2d \leq \sum_j |a_{ij}| $$ (4.27)
For a completely conservative transition matrix (i.e., when all daughter nuclides are contained in the transition matrix and all possible isotopes are accounted for) this summation will always equal $2d$. This can be related to the matrix norm (eq. (4.23)) by:

$$[A] \leq 2d$$

(4.28)

or by multiplying through by $-t$ and utilizing the relation in eq. (4.25):

$$2dt = -2 \ln (e^{-dt}) = -2 \ln (0.001)$$

(4.29)

and thus combining eq. (3.28) and (3.29):

$$[A]^t \leq -2 \ln (0.001) \approx 13.8155$$

(4.30)

Next, by relating the matrix norm definition back to the matrix exponentiation function (eq. (4.13)) it is clear that the maximum term in the Taylor expansion summation is:

$$\frac{([A]^t)^m}{m!}$$

(4.31)

where $m$ is the largest integer that does not exceed $[A]^t$. Given the equality of eq. (4.30), $m$ is 13 and the maximum term in the Taylor expansion summation is:

$$\frac{([A]^t)^{13}}{13!} \approx \frac{13.8155^{13}}{13!} \approx 1.0727 \times 10^5 - 10^5$$

(4.32)

Most computers rely on 32-bit word size and can store 16 significant figures using double-precision. This means that numerical accuracy can be maintained to 11 significant figures past the decimal point at values of $10^5$ (e.g., $5+11=16$). In most cases, however, it is sufficient to have 5 significant figures after the decimal at values of $10^{-6}$ since the maximum term is $\sim 10^5$ (e.g., $5+6+5=16$). This places a numerical accuracy limit on this transmutation approach and it is the same limit as that used in ORIGEN.

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However, if more precise computers are used (i.e., greater than 32-bit word size) the accuracy could be improved.

4.2.3 Number of Partial Sum Terms

The number of partial sum terms used in ORIGEN is variable. This as well is related to the matrix norm and the time interval of interest is the integer value of:

\[ \int \left( \frac{7}{2} |A| t + 5 \right) \]

This expression was empirically determined; however, it has been shown to be sufficient to limit the summation error to less than 0.1% for most real world transmutation calculations (Hermann and Westfall, 1998). Specifically, given the maximum value of \(|A|t\) (eq. (4.30)), the partial sum contains 53 terms,

\[ \int \left( \frac{7}{2} (13.8155) + 5 \right) = 53 \]

with the last term (eq. (4.31)) not exceeding \(6.4 \times 10^{-10}\),

\[ \frac{(13.8155)^{53}}{53!} = 6.4 \times 10^{-10} \]

This is sufficient for most applications where 5 significant figures after the decimal place of values of \(10^{-6}\) are used. Therefore, this ultimately serves as the final calculation of precision where the accuracy is thus limited by the number of terms in the summation for a specified machine precision. In this case, calculated results are reliable to \(\sim 10^{-10}\) even though numerical accuracy permits smaller values.

4.2.4 Average Neutron Flux

During long irradiation times, the neutron flux will change. Furthermore, the neutron flux is energy dependent. Figure 3.1 in Chapter 3 shows the neutron flux
spectrum for typical thermal and fast reactors. The average neutron flux refers to the energy averaged values of this spectrum. The neutron spectrum is also influenced by several factors. Temperature variation can shift the spectrum as binary collisions between neutrons and nuclides can no longer be accurately modeled with stationary nuclide assumptions (refer to Section 1.2). This temperature shift is known as Doppler broadening and strongly affects the energy dependent neutron cross sections of each isotope differently. This in turn alters the neutron flux spectrum as energy-dependent reactions become more or less favorable. In addition, during transmutation, the neutron flux (spectrum) will change as isotopes undergo changes. Different isotopes have different cross sections, which are also influenced by the temperature, and thus influence the neutron flux. In short, the composition influences the neutron flux and the neutron flux in turn influences the composition.

Because of this change in composition, the average neutron flux will change during long irradiation times. In a commercial power plant, however, the electrical power is usually maintained at a fixed value and not the neutron flux. The power of a nuclear power plant does not experience significant fluctuations from a fuel cycle standpoint. Power plants are typically rated at a fixed electricity maximum capacity (e.g., 1000 MWe rated plant) and most nuclear power plants operate at greater than 90% capacity (e.g., 900 MWe for a 1000 MWe rated plant). The electrical power generation is related to the thermal power generation through the plant’s thermal efficiency (e.g., around 33%). Another useful parameter is the rating of the power plant at a specific thermal power density. Typical values for the thermal power densities of fast and thermal reactor can be found in Table 4.1.
Each fission event releases a known amount of energy based upon the reaction that takes place. This energy eventually (ignoring second order energy losses such as that carried away by neutrinos) is expressed as heat and is used to generate electricity. For example, the average amount of energy released (and eventually transferred to heat) is approximately 200 MeV/fission for a multitude of isotopes. A better approximation can be obtained by determining the specific energy released for each neutron interaction (i.e., sum of all Q values). This amount of energy can be used to relate the average neutron flux to the thermal power density:

\[
P = \frac{200 \text{MeV}}{\text{fission}} \times \frac{1.602 \times 10^{-19} \text{MW s}}{\text{MeV}} \times \frac{1}{\text{fission neutron}} \phi \sum_i N_i \sigma_{f,i} \tag{4.36}
\]

\[
P = 3.2 \times 10^{-17} \sum_i N_i \sigma_{f,i} \phi \tag{4.37}
\]

where \(P\) is the thermal power density in unit of MW/cm³, \(N_i\) is the atom density of isotope \(i\) in unit of atoms/cm³, \(\sigma_{f,i}\) is the microscopic neutron fission cross section of isotope \(i\) in unit of cm², and \(\phi\) is the energy average neutron flux in unit of neutrons/cm²/s. The summation is commonly referred to as the total macroscopic fission cross section given in unit of cm⁻¹ as:

\[
\Sigma_f = \sum_i N_i \sigma_{f,i} \tag{4.38}
\]
The macroscopic fission cross section will change over time as the isotopic composition changes. Therefore, given that the power is held constant during the operating time of interest, the energy averaged flux at a specific time can be determined by rearranging eq. (4.37) as:

\[ \phi(t) = \frac{3.125 \times 10^{16} P}{\Sigma_f(t)} \]  

Since the fission cross section will change smoothly with time, an additional approximation of the time-energy averaged neutron flux, \( \bar{\phi} \), over a time interval \( \Delta t \) around \( t \) can be made using a Taylor series expansion:

\[ \phi(t + \Delta t) = 3.125 \times 10^{16} P \left[ \frac{1}{\Sigma_f(t)} + \frac{\Delta t \dot{\Sigma}_f(t)}{\Sigma_f(t)^2} + \frac{\Delta t^2}{2} \left( \frac{2 \ddot{\Sigma}_f(t) - \Sigma_f(t) \dot{\Sigma}_f(t)}{\Sigma_f(t)^3} \right) \right] \]  

The time-energy averaged value can then be determined by integrating over the interval \( 0 < \Delta t' < \Delta t \) then dividing by \( \Delta t \):

\[ \bar{\phi}(t, \Delta t) = \frac{1}{\Delta t} \int_0^{\Delta t} \phi(t + \Delta t')d\Delta t' \]  

which reduces to:

\[ \bar{\phi}(t, \Delta t) = \phi(t) \left[ 1 - \frac{\Delta t \dot{\Sigma}_f(t)}{2 \Sigma_f(t)} + \frac{\Delta t^2}{6} \frac{2 \ddot{\Sigma}_f(t) - \Sigma_f(t) \dot{\Sigma}_f(t)}{\Sigma_f(t)^2} + \ldots \right] \]  

where

\[ \dot{\Sigma}_f(t) = \sum_{i} \sigma_{f,i} (A^N(t)) \]  

and

\[ \ddot{\Sigma}_f(t) = \sum_{i} \sigma_{f,i} (A^{3N}(t)) \]
This latter approximation (eq. (4.42)) can be rather time consuming to compute but is more accurate for longer time intervals. However, if the time intervals, $\Delta t$, are sufficiently small, then $\bar{\phi}(t, \Delta t) \approx \phi(t)$ is valid since the flux typically changes slowly.

### 4.3 Methodology Example

The approach outlined in Section 4.2 can be integrated into a fuel cycle model, such as VISION, to perform transmutation calculations without the use of computationally intensive reactor physics models. The following section will use the VISION model as an example of how this approach can be applied. This approach is a simplified form of full scale burnup and depletion calculations that performs calculations similar to ORIGEN but without detailed reactor specific parameters. As a result, this approximation is a more gross approximation of conventional burnup and depletion calculations; however, its application is better suited for large fuel cycle models.

VISION only tracks a small subset of isotopes as they flow through various aspects of the fuel cycle. Specifically, VISION tracks 60 isotope groups which are listed in Table 4.2. The purpose of the transmutation approach is to determine the composition of the recycled fuel after irradiation using the aforementioned approach.
Table 4.2: List of isotopes groups present in VISION. Isotopes in grey represent fictitious groups that are not properly accounted for using the method in VISION. FP_OTHER represents other fission products not listed. Act_OTHER represent other actinides that are not listed. C_OTHER and 14C represent carbon and activated carbon present in the fuel cladding that are also not accounted for properly using the above fuel method.

<table>
<thead>
<tr>
<th>4He</th>
<th>210Pb</th>
<th>227Ac</th>
<th>236U</th>
<th>241Pu</th>
<th>242Cm</th>
<th>248Cm</th>
<th>H</th>
<th>134Cs</th>
<th>Sr OTHER</th>
</tr>
</thead>
<tbody>
<tr>
<td>226Ra</td>
<td>228Th</td>
<td>231Pa</td>
<td>238U</td>
<td>242Pu</td>
<td>241Cm</td>
<td>250Cm</td>
<td>81Kr</td>
<td>135Cs</td>
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<td>232U</td>
<td>237Np</td>
<td>244Pu</td>
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<td>249Cf</td>
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<td>Tc OTHER</td>
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<tr>
<td>209Pb</td>
<td>230Th</td>
<td>233U</td>
<td>238Pu</td>
<td>241Am</td>
<td>245Cm</td>
<td>250Cf</td>
<td>86Sr</td>
<td>14C</td>
<td>Cs OTHER</td>
</tr>
<tr>
<td>207Pb</td>
<td>232Th</td>
<td>234U</td>
<td>239Pu</td>
<td>242mAm</td>
<td>251Cf</td>
<td>99Tc</td>
<td>129I</td>
<td>Kr OTHER</td>
<td>Act OTHER</td>
</tr>
<tr>
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<td>209Bi</td>
<td>235U</td>
<td>240Pu</td>
<td>243Am</td>
<td>247Cm</td>
<td>252Cf</td>
<td>129I</td>
<td>Kr OTHER</td>
<td>Act OTHER</td>
</tr>
</tbody>
</table>

4.3.1 Procedures

To demonstrate the approach outlined in Section 4.2, the method can be summarized in the following algorithm:

1. Divide the total irradiation time into reasonable time steps (i.e., time intervals) where the neutron flux can be treated as constant for that step (e.g., typically between 1 and 100 days as used in ORIGEN).

2. Gather necessary parameters:
   a. Initial composition of that time step, \( N_i(0) \)
   b. Reactor type (e.g., thermal or fast reactor)
   c. One-group fission and radiative capture cross sections, \( \sigma_{f,i} \) and \( \sigma_{c,i} \)
   d. Fission yield of each fissile isotope, \( \gamma_{ji} \)

3. Determine the average neutron flux based upon the power density for that time interval, \( \phi \) or \( \bar{\phi} \) (eqs. (4.40) or (4.42))

4. Populate the transition matrix, \( A \) (eqs. (4.8)-(4.10))
5. Perform the recursive calculation to determine the new composition at the end of that time step, \( N_i(t) \) (eqs. (4.18)-(4.20))

6. Increment to the next time step and repeat procedures 1-5 using the new composition until the total irradiation time has elapsed

While the algorithm straightforward, gathering the necessary data is challenging. The following sections are devoted to detailing how data was collected and the necessary simplifying assumptions that were made to handle the subset of only 60 isotopes.

### 4.3.2 One-Group Cross Sections

The one-group cross sections are the most difficult to generate. The cross sections recorded in Table 3.1 of Chapter 3 are currently used in VISION. Neutron cross sections are reactor (and thus composition) dependent as described in Section 3.1.3. For the purposes of approximating these cross sections, the fission and radiative capture cross sections associated with each target fuel recipe, as described in Section 3.3, are used. Neutron cross sections from various databases (ENDF-IV library and the JEFF-3.1 library) were extracted (References in Appendix A.1); however, these results were not convincing as spurious gross approximations of the neutron spectrum were necessary to achieve consistent results. For the purposes of fuel cycle modeling, not only does the composition of target fuel recipes need to be provided by the user, but also the associated one-group cross sections.

Various nuclear codes can be used to compute one-group cross sections by the user. Namely, well known NJOY-99 coupled with MCNP codes can be used to adjust for temperature and flux dependent cross sections for a specific reactor design. These
calculations, however, are extremely computationally intensive (tens of CPU-hrs) and could not possibly be used within a dynamic fuel cycle simulation at each time step for each reactor type. These codes, however, can be used outside of the fuel cycle simulation (as was already performed for VISION) to determine the composition of static fuel recipes and approximate neutron cross sections for the dynamic cases. Because of the complexity and the associated unknown reactor parameters, one-group neutron cross sections were not self-generated. Instead, the pre-existing cross sections inside VISION were used. The values can be found in Appendix A.2.

4.3.3 Average Number of Neutrons per Fission

In the case of the average number of neutrons generated per fission, \( \nu \), again the values predetermined inside VISION can be used. This value is weakly dependent on energy and in many cases, a single energy independent value is recorded. However, this property can also easily be extracted from the ENDF-IV database file format (MT=1452). A MATLAB script to do that can be found in Appendix A.3 and A.4. This script compresses this factor into 10 energy groups, which is then typically collapsed further into one-group. Upon doing this, nearly identical values were obtained to the parameters used inside VISION (the minor discrepancies are credited to the use of different sources and databases). As a result, the values currently used in VISION for \( \nu \) (recorded in Appendix A.2) are used.

4.3.4 Fission Yield

The fission yield represents, \( \gamma_{ij} \), the fractional yield of an isotope from the fission of another isotope (eq. (4.2)). Each fissionable isotope, \( i \), has a fractional value that represents the generation of isotope \( j \). For example, imagine that isotope A will fission
into isotopes B and C 75% of the time and then into isotopes B and D the other 25% of
the time. The fission yield of A to B (i.e., $\gamma_{AB}$) is 1.0. Similarly the fission yield of A to
C (i.e., $\gamma_{AC}$) is 0.75 and of A to D (i.e., $\gamma_{AD}$) is 0.25. For each isotope A, the fission yield
into a set of all other isotopes is known and recorded in various databases.

Conveniently $\gamma_{ij}$ can be extracted from these databases. The database references
can be found in Appendix A.1 organized by ZAID (Z=atomic number, A= Atomic
number, ID=identification, e.g., $^{235}$U’s ZAID is 92235). The ZAID number is a
conventional identification code for isotopes used in many nuclear simulations. These
databases are formatted in the ENDF-IV format and are separated by material cards (as is
convention). The material card, MT, corresponding to $\gamma_{ij}$ is MT=8454. Data extraction
scripts written using MATLAB can be found in Appendix A.5-A.6.

Typically, only heavy isotopes fission and the resulting residuals are significantly
lighter isotopes. Upon extracting data, the fission yield matrix as recorded in Appendix
A.7 was generated. This matrix represents the fission of a heavier (row, $i$) isotope into a
lighter (column, $j$) isotope. All other isotopes tracked within VISION have fission yield
values of zero (or nonexistent data as is the case for, $^{209}$Bi, $^{230}$Th, $^{227}$Ac, $^{244}$Pu, $^{247}$Cm,
$^{250}$Cm, $^{250}$Cf, and $^{252}$Cf). This matrix is then used to populate the transition matrix.

4.3.5 Decay Chains

The decay chains are complicated to generate as well. Each radioactive isotope
has a half-life, $T_{1/2}$, which can be used to calculate the decay constant, $\lambda$, of each isotope
(eq. (4.4)). The half-lives and decay constant used in VISION can be found in Appendix
A.8. Following serial decay (i.e., sequence of decays) an effective half-life and the
effective decay constant are usually assigned:
\[ T_{1/2,\text{eff}} = \left( T_{1/2_A} + T_{1/2_B} + \ldots + T_{1/2_N} \right)^{-1} = \left( \sum_i T_{1/2_i} \right)^{-1} \] (4.45)

\[ \lambda_{\text{eff}} = \lambda_A + \lambda_B + \ldots + \lambda_N = \sum_i \lambda_i \] (4.46)

where the subscripts represent the species in the serial decay. However, since VISION only tracks a limited set of isotopes, the effective half-life is approximated by the half-life of the parent nuclide. This is valid because the majority of serial isotopes are short lived in comparison to the parent nuclide with the exception of $^{210}$Pb and $^{242m}$Am. The activity of these exceptions is slightly over-approximated using this simplified approach. If more isotopes were tracked, then this assumption could be removed as the longer lived daughters would also be tracked.

The decay chains are recorded in Appendix A.9. These chains also incorporate the appropriate branching ratio where the effective decay constant is simply multiplied by this branching ratio. Upon assembling the decay constant data, a (spares) decay chain matrix as recorded in Appendix A.10 was generated. This matrix represents the decay of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate decay constant. Note that each alpha decay generates a $^4$He nuclide. Also, note that this sparse matrix can be replaced be a vector of elements to save memory.

**4.3.5 Radiative Capture Chains**

When an isotope undergoes radiative capture it will transmute into another isotope. Specifically, the isotope’s atomic number will increase by 1 and a gamma ray will be released ($n, \gamma$). For example:

\[ ^{235}_{92}U + ^{1}_n \rightarrow ^{236}_{92}U + \gamma \]
In many cases following radiative capture, additional radioactive decay takes place when the resulting nucleus is unstable. To better represent these transitions, a radiative capture chain matrix, similar to the decay chain matrix, has been developed. This matrix is used to populate the transmutation matrix elements corresponding to radiative capture transitions.

Detailed radiative capture chains for the isotopes tracked in VISION can be found in Appendix A.11. These chains assume negligible time for the transition mentioned. This is valid because the majority of daughter isotopes following radiative capture are short-lived with the exception of the $^{232}$Th, $^{248}$Cm, and $^{252}$Cf chains. The activity of these exceptions is slightly over-approximated using this simplified approach. If more isotopes were tracked, then this assumption could be removed as the longer lived daughters would also be tracked. Next, the radiative capture transition matrix is populated by values 0 or 1, which represents the radiative capture of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate fraction. For example, a value of 1 means the isotope undergoes the transition mentioned from the isotope in row $i$ to isotope in column $j$. This capture chain matrix for the select isotopes in VISION can be found in Appendix A.12. Note that this sparse matrix can be replaced by a vector of elements to save memory.

This is the final piece of information that must be gathered to populate the transition matrix. To summarize, the following information must be assembled: (1) the composition to be irradiated, (2) the power density of the reactor, (3) the irradiation time, (4) the one-group fission and radiative capture cross sections, (5) the average number of neutrons per fission, (6) the fission yield, (7) the isotopic decay chains, and (8) the isotopic radiative capture chains. The majority of this information (4-8) can be collected
in a database and then be used to populate the transition matrix which is the key to determining the composition following irradiation.

4.3.6 Results

After assembling all the necessary information, the composition following irradiation can be determined using the approach described in this chapter. This approach is most useful in large fuel cycle models such as VISION, where dynamic approximations of irradiated compositions need to be determined without relying on computationally intensive reactor physics models. To best illustrate the use of this approach, an illustrative calculation is performed using the static fuel recipes within VISION. Specifically, the static fresh fuel compositions for fast and thermal reactors are used as input compositions and the irradiated compositions using this approach are compared against the static spent fuel composition used by VISION.

One major discrepancy is expected, however. VISION categorizes the composition into non-fundamental groups, namely: $^{14}$C, C$_{\text{Other}}$, Kr$_{\text{Other}}$, Sr$_{\text{Other}}$, Tc$_{\text{Other}}$, I$_{\text{Other}}$, Cs$_{\text{Other}}$, FP$_{\text{Other}}$, and Act$_{\text{Other}}$. All of these groups (with the exception of $^{14}$C which is associated with the cladding instead of the fuel) contain numerous isotopes and as a result they cannot be accurately modeled with isotope specific parameters. If these non-fundamental groups, however, were further divided into their constituent isotope and hence tracked within VISION, these parameters could be better approximated using this method.

Both illustrative examples for fast and thermal reactors are discussed next. In these examples, the fresh composition is the same target composition mentioned in Section 3.3.2. The thermal reactor composition is the VISION recipe that corresponds to
a mixed-oxide NpPuAm fuel used in the first pass of a recycling strategy (MOX-NpPuAm cycle 1). The fast reactor composition is the VISION recipe that corresponds to the 0.50 conversion ratio fast reactor that is fed by a light water reactor used fuel with a 50 GWd/MTU burnup (CR-0.50, UOX-50). These fresh fuel recipes (reported as mass fractions) have been produced below in Table 4.3.
Table 4.3: Fresh fuel compositions (mass fraction) for illustrative example. The thermal reactor is a mixed-oxide fuel containing Np, Pu, and Am. The fast reactor has a conversion ratio of 0.5 that is fed by a UOX fuel with 50 GWd/MTU burnup.
These fresh fuel recipes then undergo irradiation and the resulting composition is determined using the approach outlined in this section. Both reactor types experience 3 years (1095 days) of continuous irradiation at the power densities described in Table 4.1. This total irradiation time has been divided into several smaller time intervals such that the neutron flux is considered constant over a time interval. Time steps of 10 days (or less in the case of the last time step) are used except where the flux varies by more \( \pm 1\% \) between sequential time steps. In these cases, time steps of 1 day are used instead in order to capture the large change in flux (i.e. \( >1\% \) change). This is essential because during startup, the flux changes more rapidly and, in order to capture this change, smaller time steps are used. This does increase execution time; however, larger time steps could easily be used. The compositions upon discharge have been produced in Tables 4.4 and 4.5 along with the predetermined static spent fuel recipes currently used in VISION.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Predicted Composition</th>
<th>VISION Recipe</th>
<th>Percent Difference</th>
<th>Isotope</th>
<th>Predicted Composition</th>
<th>VISION Recipe</th>
<th>Percent Difference</th>
</tr>
</thead>
<tbody>
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<td>$^4\text{He}$</td>
<td>$4.45E-06$</td>
<td>$6.52E-05$</td>
<td>27.86%</td>
<td>$^{242}\text{Cm}$</td>
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<td>$5.83E-04$</td>
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<tr>
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<td>$7.18E-14$</td>
<td>4.52%</td>
<td>$^{243}\text{Cm}$</td>
<td>$3.10E-07$</td>
<td>$3.26E-05$</td>
<td>53.49%</td>
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<td>$1.02E-19$</td>
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<td>$^{230}\text{Th}$</td>
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<td>2.66%</td>
<td>$^{250}\text{Cf}$</td>
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<td>$9.86E-11$</td>
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<td>$3.96E-11$</td>
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</tr>
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<td>$6.03E-10$</td>
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<td>$\text{C Other}$</td>
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<td>$0$</td>
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</tr>
<tr>
<td>$^{233}\text{U}$</td>
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<td>$1.36E-08$</td>
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<td>$^{81}\text{Kr}$</td>
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<td>$7.41E-11$</td>
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</tr>
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<td>4.79%</td>
<td>$^{85}\text{Kr}$</td>
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</tr>
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<td>$\text{K Other}$</td>
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<td>12.83%</td>
</tr>
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<td>$7.88E-01$</td>
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<td>$2.89E-04$</td>
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<td>2.17%</td>
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<td>$\text{Tc Other}$</td>
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<td>6.42%</td>
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<td>$5.67E-02$</td>
<td>15.39%</td>
<td>$^{129}\text{I}$</td>
<td>$7.14E-11$</td>
<td>$3.53E-04$</td>
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<td>$3.72E-02$</td>
<td>1.36%</td>
<td>$\text{I Other}$</td>
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<td>$1.73E-02$</td>
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<td>$^{134}\text{Cs}$</td>
<td>$4.23E-08$</td>
<td>$2.06E-04$</td>
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<td>$1.24E-02$</td>
<td>1.24%</td>
<td>$^{135}\text{Cs}$</td>
<td>$1.31E-06$</td>
<td>$9.43E-04$</td>
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</tr>
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<td>$2.25E-07$</td>
<td>119.31%</td>
<td>$^{137}\text{Cs}$</td>
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</tr>
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<td>$\text{Cs Other}$</td>
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<td>$^{243}\text{Am}$</td>
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<td>$\text{Act Other}$</td>
<td>$0$</td>
<td>$7.03E-05$</td>
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</tr>
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Table 4.4: Spent fuel compositions (mass fraction) for the thermal reactor illustrative example. The thermal reactor is a mixed-oxide fuel containing Np, Pu, and Am.
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<th>Isotope</th>
<th>Predicted Composition</th>
<th>VISION Recipe</th>
<th>Percent Difference</th>
<th>Isotope</th>
<th>Predicted Composition</th>
<th>VISION Recipe</th>
<th>Percent Difference</th>
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<td>6.54E-05</td>
<td>21.86%</td>
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<td>5.98E-06</td>
<td>4.83E-04</td>
<td>57.52%</td>
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<td>1.78E-14</td>
<td>4.52%</td>
<td>$^{241}\text{Cm}$</td>
<td>2.32E-06</td>
<td>3.36E-05</td>
<td>25.92%</td>
</tr>
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<td>9.96E-22</td>
<td>1.33%</td>
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<td>2.74E-03</td>
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<td>2.45E-19</td>
<td>5.27%</td>
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<td>5.74E-04</td>
<td>4.06E-04</td>
<td>4.42%</td>
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<td>$^{207}\text{Pb}$</td>
<td>1.98E-17</td>
<td>5.72E-17</td>
<td>2.83%</td>
<td>$^{246}\text{Cm}$</td>
<td>7.63E-05</td>
<td>4.20E-05</td>
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<td>4.68E-10</td>
<td>52.00%</td>
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<td>3.76E-06</td>
<td>1.59E-06</td>
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<td>8.07E-18</td>
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<td>$^{248}\text{Cm}$</td>
<td>3.17E-07</td>
<td>1.02E-07</td>
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</tr>
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<td>$^{228}\text{Th}$</td>
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<td>49.66%</td>
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<td>29.79%</td>
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<td>3.45E-09</td>
<td>63.9%</td>
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<td>4.61E-11</td>
<td>1.76E-11</td>
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<td>33.25%</td>
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<td>3.69E-12</td>
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<td>$^3\text{H}$</td>
<td>0</td>
<td>3.32E-07</td>
<td>0.00%</td>
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<td>7.10E-11</td>
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<td>$^{14}\text{C}$</td>
<td>0</td>
<td>0</td>
<td>0.00%</td>
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<td>2.21E-07</td>
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<td>0</td>
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<td>2.68E-09</td>
<td>1.47E-08</td>
<td>9.42%</td>
<td>$^{81}\text{Kr}$</td>
<td>1.27E-11</td>
<td>5.62E-11</td>
<td>62.9%</td>
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<td>1.07E-04</td>
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<td>9.95%</td>
<td>$^{85}\text{Kr}$</td>
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<td>2.74E-04</td>
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<td>4.35%</td>
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<td>76.54%</td>
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<td>17.46%</td>
<td>FP Other</td>
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<td>1.62%</td>
<td>Act Other</td>
<td>0</td>
<td>4.83E-04</td>
<td>0.00%</td>
</tr>
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</table>

Table 4.5: Spent fuel compositions (mass fraction) for the fast reactor illustrative example. The fast reactor has a conversion ratio of 0.5 that is fed by a UOX fuel with 50 GWd/MTU burnup.
Due to the gross approximations in the neutron cross sections, the power density of the core and the irradiation time, and the small sub-set of isotopes, significant differences between the predicted composition and the composition used in VISION are observed. In addition, since only the relative magnitudes are important (and not the absolute difference), the percent difference is reported based upon a logarithmic scale:

\[ \%\text{Difference} = \left| \frac{\log_{10}(\text{Predicted Composition}) - \log_{10}(\text{VISION Composition})}{\log_{10}(\text{VISION Composition})} \right| \] (4.47)

Based upon this figure of merit, only a few isotopes have major discrepancies. These isotopes include \(^{4}\text{He},^{244}\text{Pu},^{242}\text{Cm},^{99}\text{Tc},^{129}\text{I}, and^{134}\text{Cs}. First, \(^{4}\text{He}\) is under approximated because additional alpha decays that are not accounted for by the limited set of isotopes takes place. These decays are present in other actinides and in the decay chains of the fission products. Again, if more isotopes were accounted for, a more detailed approximation could have been made. Both \(^{244}\text{Pu}\) and \(^{242}\text{Cm}\) are under approximated because of the simplified decay chains and the approximations of the cross sections. In both of these cases, these isotopes arise after a series of complicated activation and decays. A small error in the neutron cross sections will propagate through and compound the error in the activation and decay chains. In the case of these two isotopes, the error is the largest at the end of the chain.

In addition, this error propagates to the fission products. Namely, \(^{99}\text{Tc},^{129}\text{I}, and^{134}\text{Cs}\) are all under approximated because (1) the fission cross sections of the parent nuclides introduce error and (2) the under approximation of fissile nuclides also introduce error. These errors are further exacerbated by how these isotopes are then transformed into other fission products. Also, other actinide (and fission products) fission (or decay)
into these isotopes and better approximations cannot be made without further refining the non-fundamental groups.

Other problematic groups include $^{250}\text{Cm}$, $^3\text{H}$, $^{14}\text{C}$, C$_{\text{Other}}$, and Act$_{\text{Other}}$. $^{250}\text{Cm}$ is not properly accounted for because VISION does not track the isotopes that generate it. For example, $^{250}\text{Cm}$ results from the radiative capture of $^{249}\text{Cm}$ and from the decay of $^{254}\text{Cf}$, both of which are not tracked within VISION. $^3\text{H}$ results from the interactions in the cladding and moderator as well as from $^6\text{Li}$, which is sometimes present. $^{14}\text{C}$ and C$_{\text{Other}}$ also result from interaction within the cladding. Since cladding and moderator material are not tracked within VISION, these isotopes are not accurately predicted.

Similar approximation errors occurred during the first version of the ORIGEN code. It was determined that the initial subset of isotopes used within ORIGEN was not sufficient to characterize the fuel. In response, the library of isotopes grew. Also, it was determined that the accuracy of the cross sections needed to be refined. For this reason, years of development went into modules to dynamically approximate and adjust neutron cross sections for a vast library of isotopes. In this case, the accuracy of the prediction can only be improved by including a larger set of isotopes and better refined cross sections. However, for the limited set of isotopes and the gross approximations of the neutron cross sections, the irradiation time, and the power density of the reactor, this approach predicts the spent fuel composition remarkably well with the exception of the previously mentioned problematic groups.
CHAPTER 5

SUMMARY, CONCLUSIONS, AND FUTURE WORK

A method for determining the amount of recycled material that can be loaded into a reactor (Ch. 3) and an approach for determining the composition of that recycled material after irradiation in that reactor (Ch. 4) have been presented. These approaches are expected to be useful in nuclear fuel cycle simulations. With the time-dependent nature of the nuclear fuel cycle, dynamic models constitute one set of tools that can aid in the decision making process governing nuclear power policy. Specifically what-if-scenarios can be addressed using fuel cycle models. Fuel material is at the heart of all fuel cycle models. How this material transitions from natural ore to fuel and eventually to waste is of utmost importance.

Along the way, this material can be recycled. Following reprocessing of fuel material, certain isotopes (e.g., TRU isotopes such as $^{241}$Am, $^{237}$Np, $^{244}$Cm, and $^{239}$Pu) can be consumed in reactors. Some of these isotopes are extremely radiotoxic and can therefore be transmuted into less radiotoxic isotopes in reactors. Other isotopes still have fuel quality and can be recycled. The difficulty lies in determining how to load these isotopes into a reactor. The goal of recycling is to consume as much used fuel material as possible without significantly affecting the operation of the reactor. To accomplish this, it is envisioned that reactors will have designated transmutation channels where
recycled fuel will be placed. Other channels will contain fresh fuel. Another way of
approaching this problem is to homogenize the reactor and assume that a fraction of the
reactor contains that recycled (transuranic) inventory.

Since certain reactors can handle more recycled material than others and since the
composition of available recycled material constantly changes, a dynamic approach to
load fuel in fuel cycle models is necessary. This approach must accommodate the ever
changing composition and the changing reactor technologies. An approach to do that (for
fuel cycle models) has been developed (in Ch. 3). This approach relies on a novel
concept of assigning reactivity worth value to each isotope so recycled fuel can be better
loaded dynamically. The reactivity worth value is reactor dependent and uses a static
recipe as a target to guide the dynamic adjustment. This approach is favorable because it
relies solely on a target composition and isotope specific parameters. Unlike other
techniques, it does not require a priori information, such as $k_{eff}$. Ultimately, this approach
determines the fraction of transuranic material that can be loaded into a reactor for a
varying transuranic inventory.

After loading a fuel that has a constantly varying composition, it was then
necessary to determine the resulting composition following irradiation; a dynamic output
must follow a dynamic input. To do this, a transmutation and depletion approach was
established that is suited for fuel cycle simulations (as explained in Ch. 4). Such a
calculation is computationally conservative and is versatile enough to be used in a
multitude of reactor technologies. However, the limited isotopic inventory and the
accuracy of one-group neutron cross sections affect the accuracy of this approach. It is
useful, however, only for approximating the transuranic composition of fuel following irradiation.

These approaches have been established to fulfill the need of fuel cycle simulations to more realistically model how recycled material is to be reused for energy and to determine the resulting waste from the recycling approach. Specifically, the fuel cycle model VISION was used as a target application. The approach was illustrated for application in VISION; however, other fuel cycle models can utilize the same approach for better dynamic modeling of recycled fuel.

These approaches are preferable because they approximate reality well with a minimum set of inputs. All models are inherently limited as they will never predict reality exactly; however, the approximation can be sufficient and the error can be tolerated. These approaches do just that. While they may not replicate the exact composition of recycled fuel, given the limited inputs and computational constraints they approximate reality fairly well based upon the comparison to appropriate static fuel compositions. In addition, these approaches utilize theoretical foundations rather than empirical relationships to produce results. The combination of all these attributes makes these approaches useful. They are of particular use when evaluating what-if-scenarios which can help guide energy policy.

5.1 Applications

The approaches developed in this thesis have been specifically designed for use in fuel cycle simulations. In particular, the approaches have been refined for use in the VISION model. As mentioned in Chapter 1, the VISION model has been developed using the systems dynamic software Powersim. This software has a rudimentary
programming language, but can also execute Visual Basic (VB) scripts. This functionality allows VISION to apply both the fuel loading approach and the transmutation approach. The fuel loading approach can be developed inside Powersim using the rudimentary systems programming language and the transmutation approach can then be developed in VB where more sophisticated scripts can be executed.

To demonstrate how the fuel loading approach can be developed inside Powersim, the fuel loading approach has been programmed in the form of a sub-model using the Powersim program. The block diagram shown in Fig. 5.1 is the Powersim script that will actually perform the fuel loading calculations. Each block in this script has its own functionality which is described in Ch. 2. In addition, the same calculations have been programmed using MATLAB and a script is given in Appendix A.13.
The transmutation approach can also be applied within the VISION model in the form of VB scripts. A MATLAB script that illustrates how to perform these calculations is given in Appendix A.14. MATLAB has been the language of choice in this thesis and several utilities exist to convert MATLAB scripts into VB scripts. In addition, MATLAB’s language is easily interpreted, which better aids the readability for application purposes.
The approaches described here can easily be integrated into many fuel cycle models and are of key importance to VISION. These approaches suit the needs of the VISION model and help to better refine the fuel loading and subsequent transmutation involved in recycling fuel material. Specifically, these approaches address the issue inside VISION where non-recipe prescribed isotopes fictitiously accumulate in various aspects of the fuel cycle model. This approach overcomes this problem by determining the amount of transuranic material (containing all recipe and non-recipe prescribed isotopes) that can be loaded into a reactor without adversely affecting fuel performance. Then following this dynamic loading calculation, dynamic transmutation is performed. This best aids VISION in providing more versatility as dynamic transmutation (and possibly user defined fuels) can be handled. Applying these methods in VISION is ultimately beneficial in refining the fuel cycle model further.

5.2 Future Work

Applying the ideas described in this thesis to VISION is not the only future work that can be performed. Several other tasks can be performed to better refine these approaches. Specifically, including more isotopes and refinement of reactor specific parameters could improve the accuracy of both the fuel loading and the transmutation approach.

The fuel loading approach would benefit from using more isotopes because the absorbing material could also be considered. The cladding and moderator should also be included to better approximate the neutron losses and \( k_{\text{eff}} \) of the reactor. By incorporating additional isotopes, their loading effect can be examined. The fuel loading approach would also benefit from refinement of reactor specific parameters. By
considering the energy dependence of the neutron cross sections, a better approximation of the reactivity worth can be made for each isotope. Developing one-group cross sections is challenging and there is inherent error. The neutron flux distribution is affected by the composition and will shift depending on the radiative capture and fission cross sections. Therefore, considering more isotopes and the effect of the neutron flux on varying neutron cross section, would all serve to improve this approach.

The transmutation approach would also benefit by using more isotopes. The activation and decay chains used presently are incomplete. Some isotopes are over predicted and others are under predicted. This is due to not tracking certain isotopes in the decay chain. For example, in some situations it is assumed that the decay of one isotope will transition directly to another isotope skipping the intermediate isotopes. These intermediate isotopes generally have short enough half-lives that the delay is not significant; however, the activation of these isotopes is significant as it initiates a different decay chain. The result of this approximation is an over prediction of the concentration of some isotopes and under prediction of others. The transmutation approach would also benefit from better reactor specific parameters. The one-group neutron cross sections are the primary source of error. As previously mentioned, the one-group neutron cross sections change as the neutron flux spectrum changes. The neutron flux spectrum is then altered by the ever changing composition. While the transmutation approach accounts for the change in the neutron flux, it does not consider the shift in the energy spectrum and thus the change in the one-group neutron cross sections.

Tracking more isotopes and refining the neutron cross sections to include energy dependence are two simple improvements that can be made to these approaches. In
addition, determining subroutines to approximate the neutron flux spectrum based solely on composition would provide additional functionality for fuel cycle simulations. These approaches by themselves drastically improve the functionality and accuracy of fuel cycle simulations through the use of these dynamic approaches. Static fuel cycle models are useful to demonstrate technology, but how those technologies integrate into the entire nuclear fuel cycle requires dynamic refinement.


APPENDIX A: SUPPLEMENTAL MATERIAL
Appendix A.1: Isotope Reference Library

List of isotopic database references organized by ZAID number. (Z=atomic number, A=Atomic number, ID=identification, e.g., $^{235}\text{U}$’s ZAID is 92235)

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<td>01003</td>
<td>pjef22/T-3:16</td>
<td>1-T - 3 NEA RCOM-JUN82 SCIENTIFIC CO-ORDINATION GROUP</td>
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90229
jeff31a/TH229:18 90-TH-229 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning
jeff31a/TH229:102 90-TH-229 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning

90230
jeff31a/TH230:18 90-TH-230 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning
jeff31a/TH230:102 90-TH-230 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning

83209
mcnp/Bi209:102 Bi-209 $T=300$ K from ENDF/B-6.0 mat8325

91231
jeff31a/PA231:18 91-PA-231 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning
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92232
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92233
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92235
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jeff31a/U235:102 92-U -235 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning

92236
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jeff31a/U236:102 92-U -236 UKAEA EVAL-JUL03 Forrest, Kopecky, Sublet, Koning
## Appendix A.2: Cross Section and Other Isotopic Data

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<th>Fast $\sigma_f$ (b)</th>
<th>Thermal $\sigma_c$ (b)</th>
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Table 6.1: One-group cross section data for fast and thermal reactors.
Appendix A.3: v Data Extractor

function [NU10]=NUEXTR(fn)

clear MT temp linenum s EN NU
format long e

fid=fopen(fn);
s=0;
EN=[];
NU=[];
fgetl(fid);  %Ignore first line of *.endf file
while feof(fid)~=1
    clear temp
    temp=fgetl(fid);
    MT=str2double(temp(72:75));
    linenum=str2double(temp(78:80));
    if s>25 & temp(24)==' '  
        break
    end
    if MT==1452 & linenum>2
        %EN 1
        s=s+1;
        if temp(9)=='+'
            EN(s,1)=str2double(temp(2:8))*10^str2double(temp(10:11));
        elseif temp(9)=='-'  
            EN(s,1)=str2double(temp(2:8))*10^(-1*str2double(temp(10:11)));
        elseif temp(10)=='+'
            EN(s,1)=str2double(temp(2:9))*10^str2double(temp(11));
        elseif temp(10)=='-'  
            EN(s,1)=str2double(temp(2:9))*10^(-1*str2double(temp(11)));
        end
        %NU 1
        if temp(20)=='+'
            NU(s,1)=str2double(temp(13:19))*10^str2double(temp(21:22));
        elseif temp(20)=='-'  
            NU(s,1)=str2double(temp(13:19))*10^(-1*str2double(temp(21:22)));
        elseif temp(21)=='+'
            NU(s,1)=str2double(temp(13:20))*10^str2double(temp(22));
        elseif temp(21)=='-'  
            NU(s,1)=str2double(temp(13:20))*10^(-1*str2double(temp(22)));
        end
        %EN 2
        s=s+1;
        if temp(31)=='+'
            EN(s,1)=str2double(temp(24:30))*10^str2double(temp(32:33));
        elseif temp(31)=='-'  
            EN(s,1)=str2double(temp(24:30))*10^(-1*str2double(temp(32:33)));

end

128
if temp(32)=='+'
    EN(s,1)=str2double(temp(24:31))*10^str2double(temp(33));
elseif temp(32)=='-'
    EN(s,1)=str2double(temp(24:31))*10^... (-1*str2double(temp(33)));
end

%NU 2
if temp(42)=='+'
    NU(s,1)=str2double(temp(35:41))*10^str2double(temp(43:44));
elseif temp(42)=='-'
    NU(s,1)=str2double(temp(35:41))*10^... (-1*str2double(temp(43:44)));
elseif temp(43)=='+'
    NU(s,1)=str2double(temp(35:42))*10^str2double(temp(44));
elseif temp(43)=='-'
    NU(s,1)=str2double(temp(35:42))*10^... (-1*str2double(temp(44)));
end

%EN 3
s=s+1;
if temp(53)=='+'
    EN(s,1)=str2double(temp(46:52))*10^str2double(temp(54:55));
elseif temp(53)=='-'
    EN(s,1)=str2double(temp(46:52))*10^... (-1*str2double(temp(54:55)));
elseif temp(54)=='+'
    EN(s,1)=str2double(temp(46:53))*10^str2double(temp(55));
elseif temp(54)=='-'
    EN(s,1)=str2double(temp(46:53))*10^... (-1*str2double(temp(55)));
end

%NU 3
if temp(64)=='+'
    NU(s,1)=str2double(temp(57:63))*10^str2double(temp(65:66));
elseif temp(64)=='-'
    NU(s,1)=str2double(temp(57:63))*10^... (-1*str2double(temp(65:66)));
elseif temp(65)=='+'
    NU(s,1)=str2double(temp(57:64))*10^str2double(temp(66));
elseif temp(65)=='-'
    NU(s,1)=str2double(temp(57:64))*10^... (-1*str2double(temp(66)));
end

fclose(fid);

if ~isempty(EN) & ~isempty(NU)
    temp1=find(EN>10^-9);
    temp2=find(EN>10^-7);
    temp3=find(EN>10^-6);
    t1=temp1(1);
    t2=temp2(1);
t3 = temp3(1);
if ~isempty(NU(1:t1-1))
    tnu1 = mean(NU(1:t1-1));
end
if ~isempty(NU(t1:t2-1))
    tnu2 = mean(NU(t1:t2-1));
end
if ~isempty(NU(t2:t3-1))
    tnu3 = mean(NU(t2:t3-1));
end

%% Resonance
temp1 = find(EN > 10^-5);
temp2 = find(EN > 10^-4);
temp3 = find(EN > 10^-3);
temp4 = find(EN > 10^-2);
r1 = temp1(1);
r2 = temp2(1);
r3 = temp3(1);
r4 = temp4(1);
if ~isempty(NU(t3:r1-1))
    rnu1 = mean(NU(t3:r1-1));
end
if ~isempty(NU(r1:r2-1))
    rnu2 = mean(NU(r1:r2-1));
end
if ~isempty(NU(r2:r3-1))
    rnu3 = mean(NU(r2:r3-1));
end
if ~isempty(NU(r3:r4-1))
    rnu4 = mean(NU(r3:r4-1));
end

%% Fast
temp1 = find(EN > 10^-1);
temp2 = find(EN > 10^0);
temp3 = find(EN > 10^1);
f1 = temp1(1);
f2 = temp2(1);
f3 = temp3(1);
if ~isempty(NU(r4:f1-1))
    fnu1 = mean(NU(r4:f1-1));
end
if ~isempty(NU(f1:f2-1))
    fnu2 = mean(NU(f1:f2-1));
end
if ~isempty(NU(f2:f3-1))
    fnu3 = mean(NU(f2:f3-1));
end

NU10 = [tnu1, tnu2, tnu3, rnu1, rnu2, rnu3, rnu4, fnu1, fnu2, fnu3];
else
    NU10 = zeros(1, 10);
end
Appendix A.4: ν Data Extractor
clc;
clear all

fid0=fopen('Isotope Reference Library.txt');
temp0=textscan(fid0,'%d');
ZA=temp0{1}; %ZA # for VISION isotopes
fclose(fid0);
n=length(ZA);
MATNU=zeros(n,10);

fid1=fopen('Reference Library nu.txt');
while feof(fid1)~=1
    temp1=fgetl(fid1);
    zaf=str2num(temp1(1:5)); %ZA # for Fission Parent
    fn=temp1(7:25);
    disp(fn)
    [NU10]=NUEXTR(fn);
    k=find(zaf==ZA);
    MATNU(k,:)=NU10;
end
fclose(fid1);
xlswrite('Transmutation.xls',MATNU,'Nu','B2:K61')
**Isotope Reference Library.txt**

(Contains the ZAID number for residual isotopes of interest)

89227
91231
92232
92233
92234
92235
92236
92238
93237
94238
94239
94240
94241
94242
94244
95241
95242
95243
96242
96243
96244
96245
96246
96247
96248
96250
98249
98250
98251
98252
01003
06014
06000
36081
36085
36000
38090
38000
43099
43000
53129
53000
55134
55135
55137
55000
00001
00002

**Reference Library nu.txt**

(Contains the filename of fissionable isotopes of interest)

88226 n-088_Ra_226.endf
90228 n-090_Th_228.endf
90229 n-090_Th_229.endf
90230 n-090_Th_230.endf
90232 n-090_Th_232.endf
91231 n-091_Pa_231.endf
92232 n-092_U_232.endf
92233 n-092_U_233.endf
92234 n-092_U_234.endf
92235 n-092_U_235.endf
92236 n-092_U_236.endf
92238 n-092_U_238.endf
93237 n-093_Np_237.endf
94238 n-094_Pu_238.endf
94239 n-094_Pu_239.endf
94240 n-094_Pu_240.endf
94241 n-094_Pu_241.endf
94242 n-094_Pu_242.endf
94244 n-094_Pu_244.endf
95241 n-095_Am_241.endf
95242 n-095_Am_242m1.endf
95243 n-095_Am_243.endf
96246 n-096_Cm_242.endf
96247 n-096_Cm_243.endf
96248 n-096_Cm_244.endf
96250 n-096_Cm_245.endf
98249 n-096_Cm_246.endf
98250 n-096_Cm_247.endf
98251 n-096_Cm_248.endf
98252 n-096_Cm_250.endf
Appendix A.5: γ Data Extractor

The following MATLAB function script is used to extract the fission yield data from a specific file of the name (fn) and return the ZAID vector of the fission product (ZAFP) along with the corresponding fission yield (FY).

```matlab
function [ZAFP,FY]=FYETR(str)
%This function extracts the ZAFP and the FY from an *.endf file

clear MT temp linenum s ZAFP FY
format long e

fid=fopen(str);
s=0;
fgetl(fid); %Ignore first line of *.endf file
while feof(fid)~=1
    clear temp
    temp=fgetl(fid);
    MT=str2double(temp(72:75));
    linenum=str2double(temp(78:80));
    if s>25 & temp(24))=='
        break
    end
    if MT==8454 & linenum>2
        if mod(linenum,2)==1
            %ZAFP 1
            s=s+1;
            ZAFP(s,1)=str2double(temp(2:9))*10^4;
            %Yield 1
            if temp(31)=='+
                FY(s,1)=str2double(temp(24:30))*10^str2double(temp(32:33));
            elseif temp(31)=='-
                FY(s,1)=str2double(temp(24:30))*10^...(-1*str2double(temp(32:33)));
            elseif temp(32)=='+
                FY(s,1)=str2double(temp(24:30))*10^...(-1*str2double(temp(32:33)));
            elseif temp(32)=='-
                FY(s,1)=str2double(temp(24:31))*10^...(-1*str2double(temp(33)));
            end
            %ZAFP 2
            s=s+1;
            ZAFP(s,1)=str2double(temp(46:53))*10^4;
        else
            %Yield 2
            if temp(2)=='
                if temp(9)=='+
```

133
FY(s,1)=str2double(temp(2:8))*10^str2double(temp(10:11));
    elseif temp(9)=='-
FY(s,1)=str2double(temp(2:8))*10^(-1*str2double(temp(10:11)));
    elseif temp(10))=='+
FY(s,1)=str2double(temp(2:9))*10^str2double(temp(11));
    elseif temp(10)=='-
FY(s,1)=str2double(temp(2:9))*10^(-1*str2double(temp(11))));
end
end
end
%ZAFP  3
s=s+1;
ZAFP(s,1)=str2double(temp(24:31))*10^4;
%Yield 3
if temp(53)=='+'
FY(s,1)=str2double(temp(46:52))*10^str2double(temp(54:55));
    elseif temp(53)=='-
FY(s,1)=str2double(temp(46:52))*10^(-1*str2double(temp(54:55)));
    elseif temp(54)=='+'
FY(s,1)=str2double(temp(46:53))*10^str2double(temp(55));
    elseif temp(54)=='-
FY(s,1)=str2double(temp(46:53))*10^(-1*str2double(temp(55))));
end
end
end
fclose(fid);
while length(ZAFP)>length(FY) length{FY})=][; end
Appendix A.6 $\gamma$ Data Extractor

The following MATLAB function script is used to compile a matrix of the fission yield data for isotopes with listed ZAID numbers given in the file “Isotope Reference Library.txt” from a specific set of *.endf files recorded in the “Reference Library FY.txt”. This compiles a matrix of fission yield representing the variable $\gamma_{ij}$ and write that matrix to an Excel spreadsheet entitile “Transmutation.xls” in the workbook “FissionYield”.

```
clc;
clear all

fid0=fopen('Isotope Reference Library.txt');
temp0=textscan(fid0,'%d');
ZA=temp0{1}; %ZA # for VISION isotopes
fclose(fid0);
n=length(ZA);
MAT=zeros(n,n);

fid1=fopen('Reference Library FY.txt');
while feof(fid1)~=1
    temp1=fgetl(fid1);
zaf=str2num(temp1(1:5)); %ZA # for Fission Parent
    fn=temp1(7:25);
    disp(fn)
    [ZAFP,FY]=FYEXTR(fn);
    k=find(zaf==ZA);
    for i=1:n
        for j=1:length(ZAFP)
            if ZA(i)==ZAFP(j)
                MAT(k,i)=FY(j);
                FY(j)=0;
            elseif ZA(i)==06000 & floor(ZAFP(j)/1000)*1000==06000
                MAT(k,i)=MAT(k,i)+FY(j);
                FY(j)=0;
            elseif ZA(i)==36000 & floor(ZAFP(j)/1000)*1000==36000
                MAT(k,i)=MAT(k,i)+FY(j);
                FY(j)=0;
            elseif ZA(i)==38000 & floor(ZAFP(j)/1000)*1000==38000
                MAT(k,i)=MAT(k,i)+FY(j);
                FY(j)=0;
```

135
elseif ZA(i)==43000 & floor(ZAFP(j)/1000)*1000==43000
    MAT(k,i)=MAT(k,i)+FY(j);
    FY(j)=0;
elseif ZA(i)==53000 & floor(ZAFP(j)/1000)*1000==53000
    MAT(k,i)=MAT(k,i)+FY(j);
    FY(j)=0;
elseif ZA(i)==55000 & floor(ZAFP(j)/1000)*1000==55000
    MAT(k,i)=MAT(k,i)+FY(j);
    FY(j)=0;
elseif ZA(i)==00002 & (floor(ZAFP(j)/1000)>=89 & floor(ZAFP(j)/1000)<=103)
    MAT(k,i)=MAT(k,i)+FY(j);
    FY(j)=0;
elseif ZA(i)==00001
    MAT(k,i)=MAT(k,i)+FY(j);
    FY(j)=0;
end
end
end
fclose(fid1);
xlswrite('Transmutation.xls',MAT,'FissionYield','B2:BI61')
Isotope Reference Library.txt
(Contains the ZAID number for residual isotopes of interest)

89227
91231
92232
92233
92234
92235
92236
92238
93237
94238
94239
94240
94241
94242
94244
95241
95242
95243
96242
96243
96244
96245
96246
96247
96248
96250
98249
98250
98251
98252
01003
06014
06000
36081
36085
36000
38090
38000
43099
43000
53129
53000
55134
55135
55137
55000
00001
00002

Reference Library FY.txt
(Contains the filename of fissionable isotopes of interest)

90229 nfy-090_Th_229.endf
90232 nfy-090_Th_232.endf
91231 nfy-091_Pa_231.endf
92232 nfy-092_U_232.endf
92233 nfy-092_U_233.endf
92234 nfy-092_U_234.endf
92235 nfy-092_U_235.endf
92236 nfy-092_U_236.endf
92238 nfy-092_U_238.endf
93237 nfy-093_Np_237.endf
94238 nfy-094_Pu_238.endf
94239 nfy-094_Pu_239.endf
94240 nfy-094_Pu_240.endf
94241 nfy-094_Pu_241.endf
94242 nfy-094_Pu_242.endf
95241 nfy-095_Am_241.endf
95242 nfy-095_Am_242.endf
95243 nfy-095_Am_243.endf
96242 nfy-096_Cm_242.endf
96243 nfy-096_Cm_243.endf
96244 nfy-096_Cm_244.endf
96245 nfy-096_Cm_245.endf
96246 nfy-096_Cm_246.endf
96248 nfy-096_Cm_248.endf
98249 nfy-098_Cf_249.endf
98250 nfy-098_Cf_250.endf
98251 nfy-098_Cf_251.endf
Table A.7: Fission Yield Data

<table>
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<tr>
<th>Residual Isotopes</th>
<th>Other</th>
<th>(^{134}\text{Cs}</th>
<th>(^{135}\text{Cs}</th>
<th>(^{137}\text{Cs}</th>
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</tr>
<tr>
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<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
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<tr>
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<td>\text{Other}</td>
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<td>\text{Other}</td>
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<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
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<tr>
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<td>\text{Other}</td>
<td>\text{Other}</td>
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<td>\text{Other}</td>
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<tr>
<td>(^{239}\text{Pu}</td>
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<tr>
<td>(^{240}\text{Pu}</td>
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<td>\text{Other}</td>
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<td>\text{Other}</td>
<td>\text{Other}</td>
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<tr>
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<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
<td>\text{Other}</td>
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<tr>
<td>(^{90}\text{Sr}</td>
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<td>\text{Other}</td>
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<tr>
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<td>\text{Other}</td>
</tr>
</tbody>
</table>

Fissioned Isotopes, \(i\)

Table 6.2: Fission yield, \(q_{fi}\), data for fissionable isotopes. Data for \(^{209}\text{Bi}, \text{Th}, \text{Ac}, \text{Pu}, \text{Cm}, \text{Cf}, \text{Cf, and Cf nonexistent.}

Appendix A.7: Fission Yield Data

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### Appendix A.8: Decay Constants and Half-lives

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay Constant, ( \lambda ) (day(^{-1} ))</th>
<th>Half-life, ( T_{1/2} ) (day)</th>
<th>Isotope</th>
<th>Decay Constant, ( \lambda ) (day(^{-1} ))</th>
<th>Half-life, ( T_{1/2} ) (day)</th>
</tr>
</thead>
<tbody>
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<td>(^{4}\text{He})</td>
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<td>(^{242}\text{Cm})</td>
<td>2.74E-03</td>
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<td>(^{226}\text{Ra})</td>
<td>1.19E-06</td>
<td>5.84E+05</td>
<td>(^{243}\text{Cm})</td>
<td>6.66E-05</td>
<td>1.04E+04</td>
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<tr>
<td>(^{228}\text{Ra})</td>
<td>3.30E-04</td>
<td>2.10E+03</td>
<td>(^{244}\text{Cm})</td>
<td>1.05E-04</td>
<td>6.61E+03</td>
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<tr>
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<td>2.23E-07</td>
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<tr>
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<td>( \infty )</td>
<td>(^{246}\text{Cm})</td>
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<td>1.73E+06</td>
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<tr>
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<td>(^{247}\text{Cm})</td>
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<tr>
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<td>8.12E+03</td>
<td>(^{248}\text{Cm})</td>
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<td>1.24E+08</td>
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<tr>
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<td>6.98E+02</td>
<td>(^{250}\text{Cm})</td>
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<tr>
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<td>1.28E+05</td>
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<tr>
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<td>(^{250}\text{Cf})</td>
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<td>(^{3}\text{H})</td>
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<td>2.57E+11</td>
</tr>
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<td>(^{90}\text{Sr})</td>
<td>6.60E-05</td>
<td>1.05E+04</td>
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<td>1.64E+12</td>
<td>(^{80}\text{Sr})</td>
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</tr>
<tr>
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<td>7.67E+08</td>
<td>(^{89}\text{Tc})</td>
<td>8.92E-09</td>
<td>7.77E+07</td>
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<td>3.21E+04</td>
<td>(^{90}\text{Tc})</td>
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<td>8.81E+06</td>
<td>(^{129}\text{I})</td>
<td>1.21E-10</td>
<td>5.73E+09</td>
</tr>
<tr>
<td>(^{240}\text{Pu})</td>
<td>2.89E-07</td>
<td>2.40E+06</td>
<td>(^{151}\text{I})</td>
<td>0</td>
<td>( \infty )</td>
</tr>
<tr>
<td>(^{241}\text{Pu})</td>
<td>1.32E-04</td>
<td>5.26E+03</td>
<td>(^{134}\text{Cs})</td>
<td>9.20E-04</td>
<td>7.54E+02</td>
</tr>
<tr>
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<td>5.00E-09</td>
<td>1.39E+08</td>
<td>(^{135}\text{Cs})</td>
<td>8.26E-10</td>
<td>8.40E+08</td>
</tr>
<tr>
<td>(^{243}\text{Pu})</td>
<td>2.30E-11</td>
<td>3.01E+10</td>
<td>(^{137}\text{Cs})</td>
<td>6.32E-05</td>
<td>1.10E+04</td>
</tr>
<tr>
<td>(^{241}\text{Am})</td>
<td>4.40E-06</td>
<td>1.58E+05</td>
<td>(^{139}\text{Cs})</td>
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</tr>
<tr>
<td>(^{242}\text{Am})</td>
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<td>(^{137}\text{Pr})</td>
<td>0</td>
<td>( \infty )</td>
</tr>
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<td>2.69E+06</td>
<td>(^{137}\text{Pr})</td>
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</tbody>
</table>

Table 6.3: Decay constant and half-lives for select isotopes used in VISION. Stable isotopes have a zero decay constant and an infinite half-life.
Appendix A.9: Decay Chains

\[ ^{226}Ra \rightarrow ^{222}Rn \rightarrow ^{218}Po \rightarrow ^{214}Pb \rightarrow ^{214}Bi \rightarrow ^{214}Po \rightarrow ^{210}Pb \]

\[ ^{228}Ra \rightarrow ^{228}Ac \rightarrow ^{228}Th \]

\[ ^{210}Pb \rightarrow ^{210}Bi \rightarrow ^{210}Po \rightarrow ^{206}Pb \]

\[ ^{228}Th \rightarrow ^{224}Ra \rightarrow ^{220}Rn \rightarrow ^{216}Po \rightarrow ^{212}Pb \rightarrow ^{212}Bi \rightarrow ^{212}Po \rightarrow ^{208}Pb \]

\[ ^{229}Th \rightarrow ^{225}Ra \rightarrow ^{225}Ac \rightarrow ^{221}Fr \rightarrow ^{217}At \rightarrow ^{213}Bi \rightarrow ^{213}Po \rightarrow ^{209}Pb \]

\[ ^{227}Ac \rightarrow ^{223}Ra \rightarrow ^{219}Rn \rightarrow ^{215}Po \rightarrow ^{211}Pb \]

\[ ^{211}Pb \rightarrow ^{211}Bi \rightarrow ^{207}Tl \rightarrow ^{207}Pb \]

\[ ^{231}Pa \rightarrow ^{227}Ac \]

\[ ^{232}U \rightarrow ^{228}Th \]

\[ ^{233}U \rightarrow ^{229}Th \]

\[ ^{234}U \rightarrow ^{230}Th \]

\[ ^{235}U \rightarrow ^{231}Th \rightarrow ^{231}Pa \]

\[ ^{236}U \rightarrow ^{232}Th \]

\[ ^{238}U \rightarrow ^{234}Th \rightarrow ^{234}Pa \rightarrow ^{234}U \]

\[ ^{237}Np \rightarrow ^{233}Th \rightarrow ^{233}Pa \rightarrow ^{233}U \]

\[ ^{238}Pu \rightarrow ^{234}U \]

\[ ^{239}Pu \rightarrow ^{235}U \]

\[ ^{240}Pu \rightarrow ^{236}U \]

\[ ^{241}Pu \rightarrow ^{239}Am \]

\[ ^{241}Pu \rightarrow ^{237}U \rightarrow ^{237}Np \]

\[ ^{242}Pu \rightarrow ^{238}U \]

\[ ^{244}Pu \rightarrow ^{240}U \rightarrow ^{240}Np \rightarrow ^{240}Pu \]

\[ ^{241}Am \rightarrow ^{237}Np \]
\[\begin{align*}
242_{\text{Am}} & \rightarrow \text{IT}, 99.54\% \rightarrow 242_{\text{Am}} \\
242_{\text{Am}} & \rightarrow \text{EC}, 17.30\% \rightarrow 242_{\text{Pu}} \\
242_{\text{Am}} & \rightarrow \beta, 82.70\% \rightarrow 242_{\text{Cm}} \\
242_{\text{Am}} & \rightarrow \alpha, 0.46\% \rightarrow 238_{\text{Np}} \rightarrow 238_{\text{Pu}} \\
242_{\text{Am}} & \rightarrow \alpha \rightarrow 239_{\text{Np}} \rightarrow 239_{\text{Pu}} \\
242_{\text{Am}} & \rightarrow \alpha \rightarrow 238_{\text{Pu}} \\
242_{\text{Am}} & \rightarrow \alpha, 99.71\% \rightarrow 239_{\text{Pu}} \\
243_{\text{Am}} & \rightarrow \alpha, 99.29\% \rightarrow 243_{\text{Am}} \\
243_{\text{Am}} & \rightarrow \alpha \rightarrow 240_{\text{Pu}} \\
243_{\text{Am}} & \rightarrow \alpha \rightarrow 241_{\text{Pu}} \\
243_{\text{Am}} & \rightarrow \alpha \rightarrow 242_{\text{Pu}} \\
243_{\text{Am}} & \rightarrow \alpha \rightarrow 243_{\text{Pu}} \rightarrow 243_{\text{Am}} \\
243_{\text{Cm}} & \rightarrow \alpha \rightarrow 244_{\text{Pu}} \\
250_{\text{Cm}} & \rightarrow \text{SF}, 80\% \rightarrow R_1 + R_2 \\
250_{\text{Cm}} & \rightarrow \alpha, 11\% \rightarrow 246_{\text{Pu}} \rightarrow 246_{\text{Am}} \rightarrow 246_{\text{Cm}} \\
250_{\text{Cm}} & \rightarrow \alpha \rightarrow 247_{\text{Pu}} \rightarrow 247_{\text{Cm}} \\
249_{\text{Cf}} & \rightarrow \alpha \rightarrow 245_{\text{Cm}} \\
250_{\text{Cf}} & \rightarrow \alpha \rightarrow 246_{\text{Cm}} \\
251_{\text{Cf}} & \rightarrow \alpha \rightarrow 247_{\text{Cm}} \\
252_{\text{Cf}} & \rightarrow \alpha \rightarrow 248_{\text{Cm}}
\end{align*}\]
Appendix A.10: Decay Matrix

Figure 6.1: Sparse decay matrix for serial decay. This matrix represents the decay of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate decay constant. Since the nonzero values of the matrix follow the primary diagonal, the matrix has been divided into 3 tables.
Table 6.4: Sparse decay matrix for serial decay. This matrix represents the decay of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate decay constant. (Table continues, 1 of 3)
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<th>$^{242}\text{Pu}$</th>
<th>$^{244}\text{Pu}$</th>
<th>$^{241}\text{Am}$</th>
<th>$^{242}\text{Am}$</th>
<th>$^{243}\text{Am}$</th>
<th>$^{244}\text{Cm}$</th>
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</table>

Table 6.5: Continued: Sparse decay matrix for serial decay. This matrix represents the decay of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate decay constant. (Table continues, 2 of 3)
Table 6.6: Continued: Sparse decay matrix for serial decay. This matrix represents the decay of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate decay constant. (Table continues, 3 of 3)
Appendix A.11: Radiative Capture Chains

\[ ^4\text{He} \rightarrow ^5\text{He} \rightarrow ^4\text{He} + ^1\text{n} \]
\[ ^{226}\text{Ra} \rightarrow ^{227}\text{Ra} \rightarrow ^{227}\text{Ac} \]
\[ ^{228}\text{Ra} \rightarrow ^{229}\text{Ra} \rightarrow ^{229}\text{Ac} \rightarrow ^{229}\text{Th} \]
\[ ^{206}\text{Pb} \rightarrow ^{207}\text{Pb} \]
\[ ^{208}\text{Pb} \rightarrow ^{209}\text{Pb} \]
\[ ^{210}\text{Pb} \rightarrow ^{211}\text{Pb} \rightarrow ^{211}\text{Bi} \rightarrow ^{207}\text{Tl} \rightarrow ^{207}\text{Pb} \]
\[ ^{228}\text{Th} \rightarrow ^{229}\text{Th} \]
\[ ^{229}\text{Th} \rightarrow ^{230}\text{Th} \]
\[ ^{230}\text{Th} \rightarrow ^{231}\text{Th} \rightarrow ^{231}\text{Pa} \]
\[ ^{232}\text{Th} \rightarrow ^{233}\text{Th} \rightarrow ^{233}\text{Pa} \rightarrow ^{233}\text{U} \]
\[ ^{209}\text{Bi} \rightarrow ^{210}\text{Bi} \rightarrow ^{210}\text{FP}_{\text{other}} \]
\[ ^{227}\text{Ac} \rightarrow ^{227}\text{Ac} \rightarrow ^{228}\text{Th} \]
\[ ^{231}\text{Pa} \rightarrow ^{232}\text{Pa} \rightarrow ^{232}\text{U} \]
\[ ^{232}\text{U} \rightarrow ^{233}\text{U} \]
\[ ^{233}\text{U} \rightarrow ^{234}\text{U} \]
\[ ^{234}\text{U} \rightarrow ^{235}\text{U} \]
\[ ^{235}\text{U} \rightarrow ^{236}\text{U} \]
\[ ^{236}\text{U} \rightarrow ^{237}\text{U} \rightarrow ^{237}\text{Np} \]
\[ ^{238}\text{U} \rightarrow ^{239}\text{U} \rightarrow ^{239}\text{Np} \rightarrow ^{239}\text{Pu} \]
\[ ^{237}\text{Np} \rightarrow ^{238}\text{Np} \rightarrow ^{238}\text{Pu} \]
\[ ^{238}\text{Pu} \rightarrow ^{239}\text{Pu} \]
\[ ^{239}\text{Pu} \rightarrow ^{240}\text{Pu} \]
\[ ^{240}\text{Pu} \rightarrow ^{241}\text{Pu} \]
\[ ^{241}\text{Pu} \rightarrow ^{242}\text{Pu} \]
\[ ^{242}\text{Pu} \rightarrow ^{243}\text{Pu} \rightarrow ^{243}\text{Am} \]
\[ ^{244}\text{Pu} \rightarrow ^{245}\text{Pu} \rightarrow ^{245}\text{Am} \rightarrow ^{245}\text{Cm} \]
\[ ^{244}\text{Am} \rightarrow ^{242m}\text{Am} \]
$^{242}_{\text{Am}} \rightarrow ^{243}_{\text{Am}}$

$^{243}_{\text{Am}} \rightarrow ^{244}_{\beta} \rightarrow ^{244}_{\text{Cm}}$

$^{242}_{\text{Cm}} \rightarrow ^{243}_{\text{Cm}}$

$^{243}_{\text{Cm}} \rightarrow ^{244}_{\text{Cm}}$

$^{244}_{\text{Cm}} \rightarrow ^{245}_{\text{Cm}}$

$^{245}_{\text{Cm}} \rightarrow ^{246}_{\text{Cm}}$

$^{246}_{\text{Cm}} \rightarrow ^{247}_{\text{Cm}}$

$^{247}_{\text{Cm}} \rightarrow ^{248}_{\text{Cm}}$

$^{248}_{\text{Cm}} \rightarrow ^{249}_{\beta} \rightarrow ^{249}_{\text{Bk}} \rightarrow ^{249}_{\text{Cf}}$

$^{250}_{\text{Cm}} \rightarrow ^{251}_{\beta} \rightarrow ^{251}_{\text{Bk}} \rightarrow ^{251}_{\text{Cf}}$

$^{249}_{\text{Cf}} \rightarrow ^{250}_{\text{Cf}}$

$^{250}_{\text{Cf}} \rightarrow ^{251}_{\text{Cf}}$

$^{251}_{\text{Cf}} \rightarrow ^{252}_{\text{Cf}}$

$^{252}_{\text{Cf}} \rightarrow ^{253}_{\beta} \rightarrow ^{253}_{\text{Es}} \rightarrow ^{249}_{\beta} \rightarrow ^{249}_{\text{Cf}}$

$^{3}_{H} \rightarrow ^{4}_{H} \rightarrow ^{3}_{H} + ^{1}_{n}$

$^{134}_{Cs} \rightarrow ^{135}_{Cs}$
Figure 6.2: Sparse transition matrix for radiative capture. This matrix represents the transition of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate logic parameter. Since the nonzero values of the matrix follow the primary diagonal, the matrix has been divided into 3 tables.
Table 6.7: Sparse transition matrix for radiative capture. This matrix represents the transition of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate logic parameter. (Table continues, 1 of 3)
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Table 6.8: Continued: Sparse transition matrix for radiative capture. This matrix represents the transition of one isotope (row, $i$) into other isotopes (column, $j$) by the appropriate logic parameter. (Table continues, 2 of 3)
Table 6.9: Continued: Sparse transition matrix for radiative capture. This matrix represents the transition of one isotope (row, \(i\)) into other isotopes (column, \(j\)) by the appropriate logic parameter. (Table continues, 3 of 3)
Appendix A.13: Fuel Loading MATLAB Script

```matlab
% Reactivity Worth Approach [Output=Adjusted]
clc; % Clear command window
clear all; % Clear variables from memory

% Initiations
n=26;  %# of isotopes
nU=4;  %# of U isotopes
nTRU=22; %# of TRU isotopes

% Data-----------------------------------------------
templ=xlsread('U to TRU control.xls'); % Load user input data
Target=templ(1:n,1);  % Target composition
Available=templ(1:n,2); % Available fuel composition
XSc=templ(1:n,3);     % One group capture cross sections
XSf=templ(1:n,4);     % One group fission cross sections
v=templ(1:n,5);       % nu-average number of neutrons per fission

eta=v.*XSf./(XSf+XSc); % Neutron reproduction factor
f=(XSf+XSc)./(XSf+XSc); % Neutron utilization factor

% Target Composition Calculations-------------------
for i=1:nU
    Target_d(i,1)=Target(i)/sum(Target(1:nU));
end
for j=nU+1:n
    Target_d(j,1)=Target(j)/sum(Target(nU+1:n));
end
P_T=2/sum(Target_d.*eta.*f); % Neutron loss factor in target fuel
MT_U=sum(Target(1:nU))/sum(Target(1:n));
MT_TRU=sum(Target(nU+1:n))/sum(Target(1:n));

% Available Composition Calculations---------------
for i=1:nU
    Available_d(i,1)=Available(i)/sum(Available(1:nU));
end
for j=nU+1:n
    Available_d(j,1)=Available(j)/sum(Available(nU+1:n));
end
P_Av=2/sum(Available_d.*eta.*f); % Isotope reactivity worth

rho=P_T*eta.*f-1; % Adjusted Composition Calculations-----------------------------
for i=1:nU
    mAv_rho(i)=Available_d(i)*rho(i)/MT_U;
end
for j=nU+1:n
    mAv_rho(j)=Available_d(j)*rho(j)/MT_TRU;
end
```
M_TRU = \frac{\text{sum}(mAv_{\text{rho}}(1:nU))}{(\text{sum}(mAv_{\text{rho}}(1:nU)) - \text{sum}(mAv_{\text{rho}}(nU+1:n)))}
M_U = 1 - M_{\text{TRU}}

\text{for } i=1:nU
\quad \text{Adjusted}(i,1) = \text{Available}_d(i) \times M_U;
\text{end}
\text{for } j=nU+1:n
\quad \text{Adjusted}(j,1) = \text{Available}_d(j) \times M_{\text{TRU}};
\text{end}
\text{for } i=1:nU
\quad \text{Adjusted}_d(i,1) = \frac{\text{Adjusted}(i)}{\text{sum}(\text{Adjusted}(1:nU))};
\text{end}
\text{for } j=nU+1:n
\quad \text{Adjusted}_d(j,1) = \frac{\text{Adjusted}(j)}{\text{sum}(\text{Adjusted}(nU+1:n))};
\text{end}
\text{P}_{\text{Ad}} = \frac{2}{\text{sum}(\text{Adjusted}_d \times \eta \times f)}; \quad \% \text{Neutron loss factor in adjusted fuel}
\quad \% \text{disp}(\text{Adjusted}) \quad \% \text{Adjusted composition}
Appendix A.14: Transmutation Approach

function Comp_o=TRNS(Comp_i,pwr,tt,s);

%TRNS Transmutation.

% This function determines the composition of a fuel in a homogenized (space
% independent) reactor.

% Input Parameters:
% Comp_i = the initial composition (mass fraction)
% pwr = the power density (MWth/cm^3)
% tt = total transmutation time (days)
% s = fast or thermal switch (1=thermal, 2=fast, 3=neither)

% LOAD DATA-------------------------------------------------------------
% FY=xlsread('Transmutation.xls','FissionYield','B2:BI61'); %Fission Yield
% XS=xlsread('Transmutation.xls','Data','E2:H61'); %X-sections
% nu=xlsread('Transmutation.xls','Data','D2:D61'); %Avg. neutrons per fission
% ZA=xlsread('Transmutation.xls','Data','A2:A61'); %1000*Z+A
% M=xlsread('Transmutation.xls','Data','C2:C61'); %Isotopic Weight
% lambda=xlsread('Transmutation.xls','Data','B2:B61'); %days^-1
% DC=xlsread('Transmutation.xls','Decay Chain','B2:BI61'); %Decay chain
% CC=xlsread('Transmutation.xls','Capture Chain','B2:BI61'); %Capture chain

FY=getappdata(0,'FY');
XS=getappdata(0,'XS');
u=1,1,1,1,1,1,lambda;
M=getappdata(0,'M');
lambda=getappdata(0,'lambda');
DC=getappdata(0,'DC');
CC=getappdata(0,'CC');
%--------------------------------------------------------------

n=60; %Number of isotopes
btocm2=10^-24; %Conversion factor: cm^2/barn
Av=6.02E23; %Avogadro’s #: atoms/mol

%Cross Sections
if s==1 %Thermal
    FXS=XS(:,1);
    CXS=XS(:,3);
elseif s==2 %Fast
    FXS=XS(:,2);
$$\text{CXS} = \text{XS};$$

\begin{verbatim}
end

% Initial State
N_t = Comp_i / M; % Fresh fuel composition (mols)
sigma_t = sum((Av*btocm2)*(FXS)'*N_t); % Macroscopic fission XS cm^-1
phi_t = 86400*(3.125E16*pwr/sigma_t); % Average neutron flux n/cm/day
phi_t0 = phi_t;
N = N_t;
A = zeros(n,n);
sigma_tdot = sum((Av*btocm2)*(FXS)'*(A*N_t));
sigma_t2dot = sum((Av*btocm2)*(FXS)'*(A^2*N_t));
ts = tt; % Transmutation time

while ts > 0
  % Smaller Timesteps---------------------------------------------------
  if phi_t < 0.99*phi_t0 | phi_t > 1.01*phi_t0
    t = 1; % Smaller timestep if the flux varies by 20%
  elseif ts > 10
    t = 10;
  elseif ts <= 10
    t = ts;
  end

  % Average Flux over Timestep------------------------------------------
  phi = phi_t*(1-t/2*sigma_tdot/sigma_t +
                t^2/6*(2*sigma_tdot^2-sigma_t*sigma_t2dot)/sigma_t^2);

  % Transition Matrix---------------------------------------------------
  A = zeros(n,n);
  temp1 = zeros(n,1);
  temp2 = zeros(n,1);
  for i = 1:n
    % Sink
    A(i,i) = -lambda(i) - phi*btocm2*(CXS(i)+FXS(i));
    % Fission
    temp1(:,i) = phi*btocm2*FY(:,i).*FXS;
    % Capture
    temp2(:,i) = phi*btocm2*CC(:,i).*CXS;
  end
  A = A + temp1' + temp2' + DC';

  % Recursive Matrix Exponentiation------------------------------------
  m = ceil(7/2*min([max(sum(abs(A),2)), max(sum(abs(A),1)' )] ) + 5);
  clear C
  C = zeros(n,1);
  for l = 0:m
    C = C + C_n(t,1,A,N_t);
  end
  N = C;

  % Next Timestep-----------------------------------------------------
\end{verbatim}

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\[
\begin{align*}
\text{ts} &= \text{ts} - t; \\
N_t &= N; \\

% Flux Correction ---------------------------------------------------------
\text{phi}_t0 &= \text{phi}_t; \quad \% \text{Previous average neutron flux n/cm}^2/\text{day} \\
\text{sigma}_t &= \text{sum}((\text{Av}*\text{btocm2})*(\text{FXS})'*N_t); \\
\text{sigma}_t\text{dot} &= \text{sum}((\text{Av}*\text{btocm2})*(\text{FXS})^T*(A*N_t)); \\
\text{sigma}_t\text{2dot} &= \text{sum}((\text{Av}*\text{btocm2})*(\text{FXS})'^*(A^2*N_t)); \\
\text{phi}_t &= 86400*(3.125E16*pwr/sigma_t); \quad \% \text{New average neutron flux} \\
\text{Comp}_o &= N.*M/\text{sum}(N.*M); \\

% ------------------------------------------------------------------------
\text{function Cn=C_n(t,n,A,N);} \\
% Recursive matrix exponentiation \\
\text{if n==0;} \\
\text{Cn=N;} \\
\text{else} \\
\text{Cn=t/(n)*A*C_n(t,n-1,A,N);} \\
\text{end}
\end{align*}
\]