Development of a fast burn-up method and investigation of transmutation in Generation IV fast reactors

Ph.D. thesis

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<th>Description</th>
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<tr>
<td>ADS</td>
<td>Accelerator Driven System</td>
</tr>
<tr>
<td>BG</td>
<td>Breeding gain</td>
</tr>
<tr>
<td>BOC</td>
<td>Beginning-of-cycle</td>
</tr>
<tr>
<td>BOL</td>
<td>Beginning-of-life</td>
</tr>
<tr>
<td>BR</td>
<td>Breeding ratio</td>
</tr>
<tr>
<td>CR</td>
<td>Conversion ratio</td>
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<td>CTMC</td>
<td>Continuous-time Markov chain</td>
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<td>DTMC</td>
<td>Discrete-time Markov chain</td>
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<td>EOC</td>
<td>End-of-cycle</td>
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<tr>
<td>EPR</td>
<td>European Pressurized Reactor</td>
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<tr>
<td>EU</td>
<td>Enriched uranium</td>
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<tr>
<td>FP</td>
<td>Fission product</td>
</tr>
<tr>
<td>GFR</td>
<td>Gas-cooled Fast Reactor</td>
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<tr>
<td>HLW</td>
<td>High level waste</td>
</tr>
<tr>
<td>LFR</td>
<td>Lead-cooled Fast Reactor</td>
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<tr>
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<td>Light Water Reactor</td>
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<tr>
<td>MA</td>
<td>Minor actinide</td>
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<tr>
<td>MOX</td>
<td>Mixed oxide</td>
</tr>
<tr>
<td>NMRSE</td>
<td>Normalized root mean square error</td>
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<tr>
<td>PWR</td>
<td>Pressurized Water Reactor</td>
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<tr>
<td>SF</td>
<td>Spent fuel</td>
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<tr>
<td>SFR</td>
<td>Sodium-cooled Fast Reactor</td>
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<tr>
<td>TRU</td>
<td>Transuranium</td>
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<td>TTA</td>
<td>Transmutation trajectory analysis</td>
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<td>UOX</td>
<td>Uranium oxide</td>
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<td>VVER</td>
<td>Water-Water Energetic Reactor</td>
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Chapter 1

Introduction

Generation IV fast reactors are envisaged to operate in closed fuel cycles due to their ability to breed their fuel from $^{238}\text{U}$ or $^{232}\text{Th}$ and burn minor actinides produced by themselves or thermal reactors in the nuclear park [1]. The production of nuclear waste can therefore be limited to fission products and reprocessing losses, while the reduced waste volume and decay heat can contribute to a more economical use of geological repositories [2]. Strategic decisions about the deployment of fast reactors and the transition from open to closed fuel cycle need detailed models, which are capable of modeling the important facilities of the nuclear fuel cycle and the material flows between them. The main challenge of fuel cycle studies that concern multiple recycling of the spent fuel is that the evaluation of different strategies can only be performed by knowing the detailed composition of the final waste, which requires the tracking of a large number of isotopes in the fuel cycle and the accurate determination of the spent fuel composition. In addition, minor actinide recycling results in a wide range of possible fuel compositions, influencing the neutron spectrum and therefore the burn-up process.

Several scenario codes use burn-up tables or burn-up dependent cross-section sets to calculate fuel depletion in the reactors, which may not be flexible enough for such analyses [3]. Despite recent developments in decreasing the computational demand of detailed burn-up calculations (e.g. the time-dependent matrix coefficients method in the ALEPH burn-up code [4], or the $\text{UW}_1$ fast fuel depletion code [5]), these methods are still too time consuming to be integrated into the simulation of the whole fuel cycle. In order to overcome these difficulties, a fast and flexible burn-up scheme called FITXS is presented in the first part of the thesis, which is based on the parametrization of one-group microscopic cross-sections as functions of the detailed fuel composition, capable of providing accurate results in very short computational time even when multiple recycling of the fuel is considered. The FITXS method was
used to develop burn-up models of Generation IV fast reactors and MOX (Mixed OXide) fuel assemblies of Generation III thermal reactors in order to investigate their transmutation capabilities in closed fuel cycle operation.

The analysis of underlying processes in minor actinide burning and fissile material breeding in the reactors motivated the development of stochastic models of the individual nuclide chains based on discrete-time and continuous-time Markov chains. The models are consistent with the Bateman equations, but they describe the transmutation and decay chains of individual atoms as stochastic processes. The continuous-time Markov chain model allows to identify the prevailing processes of minor actinide burning and fissile material breeding with the calculation of time-dependent probabilities of the different transmutation and fission trajectories in the nuclide chains, which are shown to constitute the general solution of the Bateman equations. Based on the Markov chain models, a method was also developed to count labeled transitions in the transmutation chains, which was then used to derive closed formulas for time-integrated and asymptotic fuel cycle performance parameters, such as time-dependent fission probabilities, D-factors and the average neutron balance of the different nuclides.

The structure of the thesis is the following. In Chapter 2 nuclear fuel cycle analyses are overviewed in general, discussing the role of fast reactors in the fuel cycle, and methods to assess transmutation and breeding capabilities. The FITXS method and the developed burn-up models of the reference Generation IV fast reactor cores and MOX fuel assemblies of Generation III thermal reactors are presented Chapter 3, followed by the discussion of fitting results and the verification of the burn-up models. In order to demonstrate the applicability of the FITXS method, the burn-up models were integrated in detailed fuel cycle models containing Generation IV fast reactors and Generation III thermal reactors. The equilibrium closed fuel cycles of the three fast reactors, as well as more complex transition scenarios from open to closed fuel cycles were investigated, which are discussed in detail in Chapter 4 of the thesis. The Markov chain models and related calculations are presented in Chapters 5 and 6, which were then applied to analyze minor actinide burning and fissile material breeding in the three Generation IV fast reactors in Chapter 7. The main conclusions of the thesis are summarized in Chapter 8.
Chapter 2

Overview of nuclear fuel cycle analyses

The sustainability of nuclear energy production largely depends on the structure and organization of the nuclear fuel cycle, with two main issues being the efficient utilization of natural uranium, as well as nuclear waste management. This chapter presents an overview of the main concepts and methods of nuclear fuel cycle analyses. The first and second sections explain the essential role fast reactors enact in the closure of the fuel cycle, while the third and fourth sections describe the procedure of nuclear fuel cycle simulations and additional methods of theoretical considerations and guiding concepts.

2.1 Classification of nuclear fuel cycles

The nuclear fuel cycle incorporates the life cycle of nuclear materials from the mining of natural uranium ores until final disposal. The different possible nuclear fuel cycle schemes are usually classified into three main types based on their organization [6]:

- open fuel cycles;
- partially closed fuel cycles;
- fully closed fuel cycles.

Currently operating thermal reactors mostly use low enriched uranium with 3-5% $^{235}$U content in the once-through open fuel cycle depicted in Figure 2.1, or in the open fuel cycle with single recycling of separated plutonium as MOX fuel depicted in Figure 2.2. However, the isotopic composition of Pu degrades after each recycling, and the two-times-reprocessed plutonium is no longer suitable for use in common
Overview of nuclear fuel cycle analyses

Light Water Reactors (LWRs). The open fuel cycle operation thus results in inefficient uranium utilization with less than 1% use of the theoretical energy content of uranium (i.e. in less than 1% uranium utilization ratio) combined with several hundred thousand years of geological storage needed for the spent fuel waste to reach clearance level (see Section 2.2.2) [7].

Fuel cycle schemes of the second type are closed in terms of plutonium and possibly americium management, but the neptunium and curium content of the spent fuel is transferred to waste. The plutonium content can be recycled in fast reactors, as well as in thermal reactors (see Figures 2.3 and 2.4, respectively), but continuous recycling in the latter case demands the use of special MOX fuel with enriched uranium content. Although plutonium no longer dominates the long-term radiotoxicity of the disposed waste in a partially closed fuel cycle, the multiple recycling of Pu and possibly Am can result in increased amount of Np and Cm in the spent fuel, especially in the case of thermal reactors.

Fast spectrum reactors, in particular Generation IV fast reactors and Accelerator Driven Systems (ADS) are capable of multiple recycling of both plutonium and minor actinides, including neptunium and curium, which allows the full closure of the fuel cycle [8]. Figures 2.5 and 2.6 show two representatives of fully closed fuel cycles: the integral fast reactor system based on critical TRU burners and the double strata system, which burns minor actinides in dedicated ADS transmuters. The uranium utilization ratio can be increased up to 20% in a fully closed fuel cycle [7], while the production of nuclear waste can be limited to fission products and reprocessing losses, and the reduced waste volume can contribute to more economical use of geological repositories [2]. Two physical processes contribute to favorable features of closed fuel cycles, and both of them are feasible in fast neutron spectrum: the breeding of fissile material and the transmutation of minor actinides.

2.2 The role of fast reactors

The closure of the nuclear fuel cycle is possible with fast spectrum reactors due to their ability to breed their fuel from fertile $^{238}$U or $^{232}$Th and burn minor actinides.
2.2. The role of fast reactors

![Diagram of fuel cycle with single Pu recycling](image1)

**Figure 2.2:** Once-through fuel cycle with single Pu recycling

![Diagram of partially closed fuel cycle with Pu multirecycling in fast reactors](image2)

**Figure 2.3:** Partially closed fuel cycle with Pu multirecycling in fast reactors

![Diagram of partially closed fuel cycle with Pu multirecycling in thermal (MOX-EU) reactors](image3)

**Figure 2.4:** Partially closed fuel cycle with Pu multirecycling in thermal (MOX-EU) reactors
Figure 2.5: Fully closed fuel cycle with TRU recycling in fast reactors

Figure 2.6: Double strata fully closed fuel cycle
produced by themselves or by thermal reactors in the nuclear park. The following two subsections give an overview about the physical background of these processes, and explain why fast reactors have advantageous properties compared to thermal reactors in these aspects.

2.2.1 Fissile material breeding

As of 2016, identified natural uranium resources of 5.7 million tons are sufficient for over 135 years of supply at the 2014 level of uranium requirements [9]. Prognosticated and speculative uranium resources are estimated 7.4 million tons, which might prolong this supply period, although uranium demand is expected to continue to grow with increasing electricity demand and the need for clean air electricity generation, especially in developing countries [9]. It was known from the early 1940’s that $^{238}\text{U}$ and $^{232}\text{Th}$ isotopes could capture neutrons below the MeV energy range and thereby convert to fissile $^{239}\text{Pu}$ and $^{233}\text{U}$, respectively. These isotopes are therefore called fertile, and the process is called fissile material breeding or fuel breeding. The conversion of fertile $^{238}\text{U}$ and $^{232}\text{Th}$ to fissile material is the key to exploit the potential of both natural uranium and thorium resources, which can extend the possible lifespan of nuclear energy production to several thousand years.

The breeding properties of a reactor can be described with the conversion ratio, which quantifies the ratio of the produced ($FP$) and destroyed ($FD$) fissile material [8]:

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

Depending on the conversion ratio the following cases can be identified:

- $CR > 1$: The reactor is called a breeder. In this case $CR$ is called the breeding ratio ($BR$);
- $CR \approx 1$: The reactor is an iso-breeder, or self-breeder;
- $CR < 1$: The reactor is a converter, or burner.

The breeding gain ($BG$) is derived from the breeding ratio, describing the net balance in the amount of fissile material:

$$BG = \frac{FP - FD}{FD} = BR - 1.$$  \hfill (2.2)

The above relation shows that a positive $BG$ corresponds to a net production of fissile material in the case of breeder reactors. Iso-breeders have zero breeding gain,
whereas burners (or converters) have negative breeding gain. A more detailed description of BG, including differential and integral forms, as well as the consideration of different fissile isotopes can be found in Section 2.4.1.

The advantage of the fast neutron spectrum in fuel breeding can be understood through examining the neutron economy by a rather crude and simplified but illustrative model of the process. The average number of neutrons produced per neutron absorbed can be expressed with the following formula [8]:

$$\eta(E) = \frac{\nu(E) \sigma_f(E)}{\sigma_a(E)}, \quad (2.3)$$

where $\nu$ is the average number of neutrons produced in one fission, $\sigma_f$ is the microscopic fission cross-section of the main fissile isotope, and $\sigma_a$ is its absorption cross-section. Although the value of $\nu$ does not vary much over intermediate energies of the incident neutrons, the ratio $\sigma_f/\sigma_a$ shows considerable variations, resulting in the energy behavior of $\eta$ depicted in Figure 2.7. In order to reach break-even breeding ($BR = 1$) one additional neutron is needed to convert a fertile atom, besides the one which is needed to induce fission. Let us denote the loss term due to leakage and parasitic absorptions with $L$. The neutron balance in this simplified model can be written as the following [8]:

$$\bar{\eta} - (1 + L) \geq 1, \quad (2.4)$$

where $\bar{\eta}$ is the value of $\eta$ averaged over the neutron spectrum. As the loss term is always positive, a minimum criterion for break-even breeding can be derived from this simple model:

$$\bar{\eta} > 2. \quad (2.5)$$

It can be seen in Figure 2.7, that break-even breeding in thermal neutron spectrum is only possible in the Th-U cycle, whereas in fast spectrum $^{239}$Pu significantly outperforms $^{233}$U and $^{235}$U in terms of neutron economy. However, note that due to its simplicity, the above model can be used as a conceptual guide only, and the assessment of the breeding properties of different reactor designs requires more detailed models of the neutron economy and the breeding process.

### 2.2.2 TRU transmutation

In the last few decades, the issue of waste disposal has gained higher attention besides nuclear safety and started to dominate public opinion concerning nuclear energy production. The reason behind this shift of attention is the potential risk associated with the disposal of nuclear waste in geological repositories for very long
2.2. The role of fast reactors

Figure 2.7: Average neutron yield per neutron absorbed of main fissile isotopes [10]

Time periods. Despite significant advancements regarding storage technologies, it is not possible to guarantee the proper confinement of radiotoxic materials in artificial structures for several thousand years. Sustainable energy production assumes the clearance of such nuclear wastes only when they have reached the radiotoxicity level of raw materials: natural uranium and its equilibrium decay products [11]. The potential threat that a given radioactive waste poses in the final repository can be measured with the radiotoxicity, \( RTOX \) [12]:

\[
RTOX = \sum_i A_i(t) \cdot DCF_i,
\]

where \( i \) represents the quality of radioisotopes in repository, \( A_i \) is the activity of isotope \( i \) [Bq], and \( DCF_i \) is the dose conversion factor or effective dose coefficient [Sv/Bq]. The dose conversion factor describes the risk associated with the intake of unit activity from a given radioisotope (inhalation or ingestion) [13]. In order to determine the necessary storage time to reach clearance level, the radiotoxicity index of the nuclear waste is compared to the that of natural uranium ore. More detailed hazard indices were also developed that take into account environmental processes which determine human radiation exposure after the failure of geological repositories [14]. Figure 2.8 shows the radiotoxicity of spent Light Water Reactor fuel as a function of time passed after disposal [15]. In the case of direct disposal the long-term radiotoxicity of nuclear waste is dominated by plutonium. The recycling of transuranium elements can contribute to reducing the radioactive inventory and its associated radiotoxicity by several orders of magnitude. Another factor that
makes TRU recycling favorable is the associated decay heat at the time and in the first few hundred years of disposal, which majorly affects the repository size [2]. The goals of partitioning (separation from waste) and transmutation (fission in the case of actinides) concerning waste management are therefore the following:

- reduction of the medium-term and long-term risks associated with spent nuclear fuel by decreasing the quantity of Pu and MA isotopes in the waste;
- reduction of the time needed to reach clearance levels by recycling TRU elements.

The advantages of fast spectrum reactors in terms of transmutation are twofold, but both of them originate from the energy dependence of actinide cross-sections, in particular the ratio of fission and absorption reaction rates. The first advantage comes from the fact that the ratio in question is higher at higher neutron energies (especially above 1 MeV), therefore actinides are more likely fissioned than being converted into elements with higher atomic numbers. Figure 2.9 shows the ratio of fission and absorption cross-sections of $^{237}$Np, $^{240}$Pu and $^{241}$Am as a function of neutron energy, where the increase in the ratio of the two cross-sections can be indeed observed in each case. The second advantage can be explained with the higher number of available neutrons in the core, which is favorable because additional neutrons above the ones which maintain criticality are available for transforming fertile TRU isotopes to fissile ones.

2.3 Nuclear fuel cycle simulations

Strategic decisions about the deployment of fast reactors and the transition from open to closed fuel cycles are supported by fuel cycle scenario codes, which are capable of modeling the important facilities of the fuel cycle and tracking material flows between them. The following subsections describe the operation of such codes, and in particular the procedure of calculating the spent fuel composition of the reactors as the most challenging task of fuel cycle simulations.

2.3.1 Scenario codes

The nuclear fuel cycle can be defined as the set of processes to make use of nuclear materials and to transfer them to final state. The purpose of scenario codes is the modeling of the fuel cycle along with its most important facilities and the material flows between them in order to determine natural uranium, conversion, and enrichment demands, fuel fabrication and reprocessing requirements and spent fuel
2.3. Nuclear fuel cycle simulations

**Figure 2.8:** Time evolution of the relative radiotoxicity of spent LWR fuel compared to natural uranium [15]

**Figure 2.9:** Ratio of fission and capture cross-sections of $^{237}\text{Np}$, $^{240}\text{Pu}$ and $^{241}\text{Am}$ as a function of neutron energy [10]
arising [16]. Several scenario codes have been developed and used in the last two decades, such as COSI6 (CEA, France) [17], FAMILY 21 (JAERI, Japan) [18], VISION (Idaho National Laboratory, USA) [19], EVOLCODE (CIEMAT, Spain) [20], and DESAE 2.2 (ROSATOM, Russia) [21], whose capabilities are listed in Table 2.1. More recent developments include CLASS (CRNS, France) [22], CYCLUS (Argonne National Laboratory, USA) [23] and SITON v2.0 (Centre for Energy Research, Hungary) [P4]. The elements of the nuclear fuel cycle that have to be accounted for in the fuel cycle models are the following:

- mining,
- conversion,
- enrichment,
- fuel fabrication,
- reactors,
- reprocessing,
- spent fuel storages,
- HLW storage.

Fuel cycle facilities and material flows are either treated in continuous manner such as in DESAE 2.2 and VISION, or in the form of discrete facilities and material packages, which is applied in COSI6, EVOLCODE and FAMILY 21, as well as in more recent scenario codes including CLASS, CYCLUS and SITON v2.0. The specific attributes of each facility can affect the performance of given fuel cycle strategies. These features include reprocessing losses, spent fuel storage capacities, cooling times, fuel fabrication times and reprocessing capacities – that can be on-demand or manually set – among many others [16]. With strategy parameters, fuel attributes and control parameters as input, scenario codes can estimate long-term fuel cycle material and service requirements, as well as generated waste streams, and economic and non-proliferation issues.

Fuel cycle scenarios can be divided into two categories regarding the organization of their fuel cycle models: equilibrium scenarios where the global infrastructure is fixed, and the material flows are constant and transition scenarios, which model the transition from current open or partially closed fuel cycles to equilibrium. In the former case, so-called equilibrium fuel cycle codes are also used, which determine the equilibrium feed and fuel compositions of the reactors based on iterative methods,
such as EQL3D [24]. Scenario studies have already resulted in a deep understanding of the possibilities of partitioning and transmutation (P&T) to address nuclear waste issues and have indicated the infrastructural requirements for several key technical approaches [3]. While the issues are country specific when addressed in detail, it is believed that there exists a series of generic issues related only to the current situation and the desired end point. Specific examples for these issues include [16]:

- time lag to reach equilibrium, which can take from decades to centuries;
- wide range of transmutation performance for the various technologies involved;
- accumulation of stockpiles of materials during transition phase;
- significant, and possibly prohibitive investments required to reach equilibrium;
- complex interactions with the final waste disposal path;
- etc.

The investigation of both the transition phase and the equilibrium requires accurate models that can follow material flows and determine spent fuel compositions in the reactors, as well as storage requirements and waste compositions.

### 2.3.2 Determination of the spent fuel composition

The evolution of the fuel composition during irradiation can be described with the Bateman equations or nuclide chain equations, which represent balance equations for the atomic densities (or number of atoms) of the different nuclides. In general form, the Bateman equation for nuclide $i$ can be written as the first order ordinary differential equation

$$\frac{dN_i(r)}{dt} = \text{Production rate} - \text{Destruction rate} , \quad (2.7)$$

where $N_i$ is the atomic density of nuclide $i$ [26]. The production and destruction rates can be expressed with the one-group cross-sections and integrated neutron flux, as well as the decay constants and branching ratios in the following space-dependent form:

$$\frac{dN_i(r)}{dt} = \sum_{j \neq i} \left[ \sigma_{ji}(r)\Phi(r) + f_{ji}\lambda_i \right] N_j(r) - \left[ \sigma_{ai}(r)\Phi(r) - \lambda_i \right] N_i(r) , \quad (2.8)$$

where $\sigma_{ij}$ is the microscopic one-group cross-section of the $i \rightarrow j$ reaction, $\sigma_{ai}$ is the absorption cross-section of nuclide $i$, $\Phi$ is the one-group neutron flux, $\lambda_i$ is the decay
<table>
<thead>
<tr>
<th>Attribute</th>
<th>COSI6</th>
<th>DESA2 2.2</th>
<th>EVOLCODE</th>
<th>FAMILY 21</th>
<th>VISION</th>
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<td>Yes (up to 81 isotopes)</td>
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<td>Yes (18 isotopes of U/Pu/MA/FP)</td>
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<td>Stated depletion</td>
<td>Stored depletion</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
</tr>
<tr>
<td>Stored depletion</td>
<td>No</td>
<td>No</td>
<td>Yes (isotopes of U/Pu/MA/200 FP)</td>
<td>No</td>
<td>Yes (isotopes of U/Pu/MA/200 FP)</td>
</tr>
<tr>
<td>Burnup tables</td>
<td>Burnup tables with interpolation</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
</tr>
<tr>
<td>Isotopes tracking</td>
<td>Yes (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
<td>No (isotopes of U/Pu/MA/200 FP)</td>
</tr>
<tr>
<td>Fuel facilities</td>
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<td>N/A</td>
<td>Discrete</td>
<td>Discrete</td>
<td>Discrete</td>
</tr>
<tr>
<td>Isotopes tracking</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes (18 isotopes of U/Pu/MA/FP)</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>LWR, HTR, FR, (SFR, GFR, LFR and ADS + different types of fuels)</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes (3300 isotopes)</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>LWR, HTR, FR, (SFR and GFR), ADS + different types of fuels</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes (up to 81 isotopes)</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>One-tier, two-tier scenarios (+ number of recycling)</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes (isotopes of U/Pu/MA/200 FP)</td>
<td>No</td>
<td>Yes (isotopes of U/Pu/MA/200 FP)</td>
</tr>
<tr>
<td>System Dynamics / Power Sim</td>
<td>Java</td>
<td>Fortran</td>
<td>MS Visual Basic</td>
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<td>Fortran</td>
<td>MS Visual Basic</td>
<td>System Dynamics / Power Sim</td>
<td>Java</td>
</tr>
</tbody>
</table>

Table 2.1: Comparison of scenario code capabilities [3, 22]
constant of nuclide $i$, and $f_{ij}$ is the branching ratio of the $i \rightarrow j$ decay. Equation (2.8) can be also written in the matrix form

$$\frac{dN(r)}{dt} = A(r)N(r),$$

(2.9)

where $A(r)$ is commonly referred to as the transmutation matrix. There are numerous methods available for solving the Bateman equations, such as Runge-Kutta methods, matrix exponential methods including the Padé approximation, Krylov subspace methods [27], uniformatization [28] and the Chebyshev rational approximation [29], as well as the transmutation trajectory analysis (TTA) method [30], whose efficiency and accuracy depend on the properties of the specific transmutation matrix, in particular the size and stiffness of the problem.

The greatest challenge in performing fuel depletion calculations lies in the determination of the one-group cross-sections and neutron flux, which requires detailed transport calculations in order to determine the spatial and energy dependence of the neutron flux. The spatial dependence is often treated with homogenization in order to obtain region-wise cross-sections and fluxes that correctly reproduce the total reaction rates in the selected geometric region. In addition, the changes in the fuel composition affect the spatial distribution and energy dependence of the neutron flux, therefore the one-group cross-sections and neutron flux have to be determined in multiple time steps during the burn-up calculation. Due to the fact that detailed transport calculations are too time-consuming to be integrated into dynamic fuel cycle simulations, several approximations are used in scenario codes to compromise in accuracy and computational time. These approximations can be classified into two main categories:

- burn-up tables (or recipes), which contain the spent fuel composition in tabular form at different discharge burn-up levels for given fresh fuels (used in COSI6, DESAE 2.2, EVOLCODE, FAMILY 21 and VISION);

- parametrized cross-section libraries, including burn-up dependent cross-sections and more detailed parametrizations based on multivariate regression and curve fitting (used in COSI6 and EVOLCODE).

If the fresh fuel composition can vary greatly during the simulation, for example when multiple recycling of the spent fuel is considered, the use of burn-up tables might provide inaccurate results. In these cases cross-section parametrization methods have to be used, which are discussed in the following subsection. It is important to note that different approximations can be used for different reactors in the same fuel cycle simulation. For example, burn-up in thermal reactors which operate in
Overview of nuclear fuel cycle analyses

Once-through fuel cycles can be calculated with burn-up tables, while at the same time burn-up in the fast reactors which utilize the reprocessed plutonium can be calculated using parametrized cross-sections.

2.3.3 Cross-section parametrization

The parametrization of few group cross-sections is a common approach used in scenario codes with the average fuel burn-up, initial uranium enrichment – or initial plutonium content in the case of MOX fuel – being the most common descriptive parameters [25], as well as in reactor dynamics calculations [31, 32].

In general the task of this parametrization can be formulated as finding a relatively simple function based on existing data, that approximates the cross-sections as functions of arbitrary descriptive parameters, \( x_i \) \((i = 1, 2, \ldots, n)\), without the need for time-consuming detailed neutron transport calculation:

\[
\sigma = \sigma(x_1, x_2, \ldots, x_n) .
\] (2.10)

The core geometry, structural materials, temperatures and other thermal hydraulic parameters are usually considered constant in scenario studies, therefore the one-group cross-sections depend solely on the fuel composition. In this case \( \sigma \) can be written as a function of specific atomic densities, \( N_i \) \((i = 1, 2, \ldots, m)\):

\[
\sigma = \sigma(N_1, N_2, \ldots, N_m) .
\] (2.11)

Descriptive parameters such as average burn-up, uranium enrichment and plutonium content fit into this category as well, as they are also functions of the fuel composition. On the other hand, the description of cross-sections as functions of such global parameters might provide inaccurate results if the isotopic composition of the fuel changes greatly, for example when multiple recycling of plutonium and minor actinides is considered.

Vidal et al. [33] have introduced the fractions of individual Pu isotopes and \(^{241}\)Am as smoothing parameters in the CESAR5.3 code (also used in COSI6), which decreased the discrepancies between CESAR results and reference calculations by a factor of 6. Leniau et al. [34] have recently developed a neural network based model implemented in CLASS to account for the changing Pu isotopic composition during burn-up in MOX fuel. Their model uses the proportions of \(^{238-242}\)Pu isotopes, \(^{241}\)Am, \(^{235}\)U and \(^{238}\)U, as well as the irradiation time as input parameters for the neural network, resulting in less than 3% difference in EOC inventories obtained with CLASS (Core Library for Advanced Scenario Simulation) and MCNP based MURE depletion calculations.
2.4 Analysis of breeding and transmutation

The comparison of the transmutation and breeding capabilities of different reactor designs and fuel cycle schemes requires specific fuel cycle performance parameters, which allow the characterization of fissile material production and minor actinide burning potential. In the following subsections some parameters and methods that can assess transmutation capabilities and analyze underlying processes are overviewed, such as the breeding gain and the neutron consumption per fission, or D-factor. In a recent study Oettingen et al. [35] used transmutation trajectory analysis (TTA) – described in the third section – to investigate the build-up of minor actinides with different transmutation trajectories in the closed fuel cycle of the Generation IV Lead-cooled Fast Reactor, which is based on the analytical solution of the Bateman equations for linear chains. These methods motivated the development of the stochastic models of nuclide transmutation chains in Chapters 5 and 6, with the aim to establish a general mathematical framework for the calculation of such quantities and to investigate the time evolution of individual transmutation chains, either in terms of finite irradiation or decay time, or the number of occurred nuclear transitions.

2.4.1 Breeding gain

Basic figures of merit for the characterization of the breeding capabilities of a reactor are the breeding ratio (BR) and breeding gain (BG). These quantities measure the amount of fissile material produced and consumed during irradiation, either in terms of reaction rates (differential definition) or integrated over an irradiation cycle (integral definition) [36]. In a fast neutron spectrum several nuclides are fissile, therefore different weighting schemes are used to account for different nuclear properties. The Baker and Ross formula [37], \( BG^{B&R} \) uses critical mass equivalence weights (\( w_{i}^{CM} \)), which are calculated from the traditional microscopic worths with a normalization which yields \( w_{239Pu}^{CM} = 1 \) and \( w_{238U}^{CM} = 0 \):

\[
BG^{B&R} = \frac{\sum_i w_{i}^{CM}(P_i - A_i)}{\sum_i F_i},
\]

(2.12)

where \( P_i, A_i \) and \( F_i \) represent the production, consumption and fission rates of nuclide \( x_i \). The differential definition (2.12) is used in the ERANOS code [38]. Common previous weightings include +1 for fissile and 0 for other nuclides, \( \eta_i/\eta_{239Pu} \) used by Ott [39], as well as \( \eta_i/\eta_{235U} \) for fissile and 0 for fertile isotopes suggested by Csom [40], where \( \eta_i \) is the neutron yield per neutron absorbed in nuclide \( x_i \).

A new breeding gain definition, \( BG^{VR} \) that can be applied to arbitrary fuels
Overview of nuclear fuel cycle analyses and closed fuel cycles was given by van Rooijen et al. [41] based on perturbation theory. If $w_i$'s are defined as microscopic reactivity weights, then the reactivity worth of a nuclide mixture can be used as a performance parameter for $BG$. With the inner product, $\langle ., . \rangle$ indicating the integration over space and summation over all nuclides, the breeding gain between two fuel compositions at times $t_1$ and $t_2$ (such as the beginning and end of an irradiation cycle or the beginnings of two consecutive cycles including fuel management), $N_1$ and $N_2$ with corresponding reactivity weight sets $w_1$ and $w_2$ can be expressed as

$$BG_{VR} = \frac{\langle w_2, N_2 \rangle - \langle w_1, N_1 \rangle}{\langle w_1, N_1 \rangle}.$$  

(2.13)

The authors in [41] used the TSUNAMI-1D sensitivity module of the SCALE 5 code system to calculate the $w_i$ weighting factors in unit cell geometry, and their results were shown to be consistent with the Baker and Ross weighting scheme [41].

### 2.4.2 Static and dynamic D-factor

The D-factor, defined by Salvatores et al. [42, 43] describes the average number of neutrons consumed by a given nuclide and its daughter products until one of them is finally fissioned and the specific actinide transmutation chain ends. If the neutron consumption of an isotope is negative (i.e. it has an average neutron production per fission which is positive), that means the isotope is either fissile or fertile with positive integral contribution to the neutron economy. On the other hand, a positive D-factor means that there is a neutron cost which is needed to fission an atom of the given isotope. To evaluate the neutron consumption/fission $D_J$ for nuclide $x_J$, a scheme was set up by Salvatores to iteratively add up the contribution of the specific reactions of the $n$th generation reaction products weighted with the probability of the transitions, $P_{Jn \rightarrow J(n+1)}$ [42]:

$$D_J = \sum_{J_{1i} \rightarrow J_1} P_{J \rightarrow J_{1i}} \left\{ R_{J \rightarrow J_{1i}} + \sum_{J_{2k} \rightarrow J} P_{J_{1i} \rightarrow J_{2k}} [R_{J_{1i} \rightarrow J_{2k}} + \ldots] \right\} ,$$

(2.14)

where $Jn$ denotes the $n$th nuclide generation and $R_{Jn \rightarrow J(n+1)}$ is the neutron loss (or gain) for the specific reaction which results in the appearance of nuclide $x_{J(n+1)}$:

$$R_{Jn \rightarrow J(n+1)} = \begin{cases} 1 & \text{for capture}, \\ 0 & \text{for radiative decay}, \\ 1 - \bar{\nu} & \text{for fission}, \\ -1 & \text{for (n,2n) reactions}, \ldots \end{cases}$$

(2.15)
The evaluation of $D_J$ is not trivial, because – due to the possible presence of recurring nuclides – the number of possible transmutation trajectories is infinite, and even in the case of automatic evaluation, a probability threshold has to be applied (see Krepel and Losa [44]). It follows from the definition, that the D-factor is an asymptotic quantity by nature (thus valid for infinite irradiation time due to infinite recycling of the initial atoms), because the calculation accounts for transmutation chains which ultimately end with fission. Krepel and Losa therefore refer to the above definition as static D-factor, and this terminology was also applied in the thesis.

In order to describe the dynamics of the evolution towards the static D-factor, Krepel and Losa have defined the dynamic D-factor ($D_{dyn}$) [44]. The definition is based on the fact that the solution of the Bateman equations for fractions of the fuel describes the daughter products of the initial fuel fraction. The initial atoms of a given nuclide therefore also evolve according to the Bateman equations, and the average neutron consumption per fission until irradiation time $t$ can be expressed with the integral of the respective reaction rates weighted with their neutron consumption:

$$D_{dyn}(t) = \frac{\int_0^t \sum_i (1 - \nu_i) R_i^f(t) dt + \int_0^t \sum_i R_i^c(t) dt - \int_0^t \sum_i R_i^{(n,2n)}(t) dt}{\int_0^t \sum_i R_i^f(t) dt}, \quad (2.16)$$

where $R_i^l$ is the respective reaction rate for fission $f$, capture $c$ and $(n,2n)$ reaction. From the definition it follows that the dynamic D-factor converges to the static D-factor in the $t \to \infty$ limit:

$$D_{dyn}(t \to \infty) = D_{static} . \quad (2.17)$$

Krepel and Losa showed that the convergence of the dynamic D-factor takes place on timescales of several tens of EFPY (Effective Full Power Years), which justifies the use of the finite-time-integrated neutron consumption per fission.

### 2.4.3 Transmutation trajectory analysis

In general nuclear transmutation and radioactive decay problems are described with the first-order differential equations called the nuclide chain differential equations, or Bateman equations (note that in some sources the Bateman equations refer to the solution given by Bateman). Linear decay chains are governed with the following form of the equations:

$$\frac{dN_i}{dt} = -\lambda_i N_i,$$
\[
\frac{dN_i}{dt} = \lambda_{i-1}N_{i-1} - \lambda_i N_i, \quad i = 2, \ldots, n, \tag{2.18}
\]

where \(N_i\) and \(\lambda_i\) denote the concentration and decay constant of the \(i\)th nuclide in the chain. The analytical solution of the (2.18) differential equation system was given by Bateman \[45\]. If the concentrations of all daughters are zero, then the solution can be written in the following form:

\[
N_n(t) = \frac{N_1(0)}{\lambda_n} \sum_{i=1}^{n} \lambda_i \alpha_i e^{-\lambda_i t}, \tag{2.19}
\]

where the \(\alpha_i\) factors are given as

\[
\alpha_i = \prod_{j \neq i} \frac{\lambda_j}{\lambda_j - \lambda_i}. \tag{2.20}
\]

In the case of nuclear transmutation problems and multiple decay modes with branching the transmutation chain can be broken up into a set of independent linear chains taking into account the branching ratios, and the analytical solution can be applied for each of these chains by substituting the microscopic reaction rates and decay rates into the equations \[46\]. This method is called the linear chain method which is used in several burn-up codes, such as CINDER \[47\] and MCB \[48\]. Cetnar \[30\] pointed out that in nuclear transmutation problems the application of the (2.19) solution may face numerical convergence problems in the presence of similar \(\lambda_i\) coefficients. Moreover, if one or more recurring nuclides are present in the linear chain then the (2.19) solution cannot be applied due to infinities in the \(\alpha_i\) factors. In order to overcome these difficulties, Cetnar derived the general solution of the Bateman equations for linear chains, which is also valid in the case when one or more nuclides are included more than once in the chain (or in general when equal coefficients occur in the equations). In particular, Cetnar removed the infinities by introducing shifts in the \(\lambda_k\) coefficients as \(\lambda_k, \lambda_k + \Delta, \ldots, \lambda_k + (m_k - 1)\Delta\), where \(m_k\) is the number of equal \(\lambda_k\) coefficients in the chain. After expanding the exponential terms into Taylor-series and taking the \(\Delta \to 0\) limit, Cetnar obtained the general solution for \(N_1(0) = 1\) and \(N_i(0) = 0, i = 2, \ldots, n\) as the following \[30, 49\]:

\[
A_n(t) = \sum_{i=1}^{n} \lambda_i \alpha_i e^{-\lambda_i t} \cdot \sum_{m=0}^{\mu_i} \frac{(\lambda_i t)^m}{m!} \cdot \Omega_{i,\mu_i-m}, \tag{2.21}
\]

where \(\mu_i = m_i - 1\), \(A_n(t)\) is the activity of the \(n\)th nuclide and

\[
\alpha_i = \prod_{j \neq i} \left(\frac{\lambda_j}{\lambda_j - \lambda_i}\right)^{m_j}, \tag{2.22}
\]
and the $\Omega_{i,j}$ terms can be expressed as

$$\Omega_{i,j} = \sum_{h_1=0}^{j} \cdots \sum_{h_{k-1}=0}^{i} \sum_{h_{k+1}=0}^{j} \cdots \sum_{h_n=0}^{j} \prod_{i=1}^{n} \left( \frac{h_k + \mu_k}{h_k} \right) \left( \frac{\lambda_i}{\lambda_i - \lambda_k} \right)^{h_k} \delta \left( j, \sum_{l=1}^{n} h_l \right).$$

(2.23)

In the case of nuclear transmutation problems and multiple decay modes with branching, the concentration of the $n$th trajectory nuclide in the linear chain can be obtained by taking into account the $b_k$ ($k = 1, 2, \ldots, n - 1$) branching ratios:

$$N_n(t) = N_1(0) \frac{B}{\lambda_n} A_n(t),$$

(2.24)

where the branching factor $B$ can be expressed in the following form:

$$B = \prod_{k=1}^{n-1} b_k.$$  

(2.25)

The probabilistic interpretation of the linear chain method is discussed in the works of Raykin and Shlyakhter [46] and Cetnar [30], and it was used by Oettingen et al. [35] to analyze the build-up of minor actinides in different specific transmutation trajectories. It will be shown that for the case when unit concentration is assumed for the starting nuclide, the (2.24) solution corresponds to the time-dependent transmutation trajectory probability in the continuous-time Markov chain model of the nuclide transmutation chains, which is presented in Chapter 5.
Chapter 3

The FITXS method

It was discussed in Section 2.3.3 that the parametrization of the cross-sections as functions of a few descriptive parameters (such as the burn-up, initial uranium enrichment or initial plutonium content) might provide inaccurate results, if the isotopic composition of the fuel changes greatly, for example when multiple recycling of plutonium and minor actinides is considered. The main principle of the FITXS method is to fit the one-group cross-sections as functions of the detailed fuel composition, taking into account every actinide isotope – including a wide selection of MA isotopes – which has significant influence on the neutron spectrum. This allows the models to calculate spent fuel compositions with high accuracy for a wide range of initial compositions, while the very short computational time allows the method to be integrated into a dynamic fuel cycle simulation. However, due to the large number of fitting parameters, appropriate cross-section databases with numerous (few thousand) data points are needed in order to perform the least-squares fittings of the cross-sections and the $k_{\text{eff}}$ with satisfactory results. The application of the FITXS method can therefore be divided into three main steps [P1]:

1. *selection of the fitting parameters*: the chosen parameters should thoroughly describe the neutron spectrum, i.e. the one-group cross-sections, and the multiplication factor;

2. *preparation of the cross-section database*: detailed transport calculations have to be performed for numerous different isotopic compositions in order to determine corresponding cross-section sets;

3. *cross-section parametrization*: non-linear fittings are performed on the prepared database in order to parametrize the cross-sections as functions of the selected atomic densities.
The FITXS method was used to develop burn-up models for three Generation IV fast reactors and MOX fuel assemblies of two Generation III thermal reactors, whose core configurations are described in the first section. The above three steps of application are presented in detail from the second to the fourth sections of the chapter, whereas the accuracy and limitations of the method, including the verification of the burn-up models are discussed in the fifth section.

3.1 Reference core designs

The primary aim of the development of the FITXS method was to allow the fast simulation of closed fuel cycles containing Generation IV fast reactors. For this purpose the method was used to develop burn-up models for three Generation IV fast reactor types: the Gas-cooled Fast Reactor (GFR), the Lead-cooled Fast Reactor (LFR), and the Sodium-cooled Fast Reactor (SFR). In order to be able to analyze minor actinide burning and plutonium management in more complex fuel cycle scenarios containing both fast and thermal reactors, the burn-up models of MOX fuel assemblies of two Generation III Light Water Reactors, the European Pressurized Reactor (EPR) and the VVER-1200 were also developed using the FITXS scheme. Three-dimensional models of the fast reactor cores and MOX fuel assemblies were created in the KENO-VI transport module of the SCALE 6.0 code [50, 51], which were then used in the selection of fitting parameters and the preparation of cross-section databases for cross-section parametrization. The reference cores of both the fast and thermal reactors that were selected for the analyses are described in the following sections.

3.1.1 Generation IV fast reactors

The following three reference cores were selected for the development of Generation IV fast reactor burn-up models:

- the 2400 MWth reference design GFR2400 for the GFR [P2],
- the 1500 MWth ELSY (European Lead-cooled SYstem) core for the LFR [52],
- the 3600 MWth ESFR working horse concept for the SFR [53].

The three-dimensional core models were created in the KENO-VI transport module of the SCALE 6.0 code according to the core layouts depicted in Figure 3.1. Main relevant parameters of the reference cores can be found in Table 3.1.
The reference core for the GFR is the GFR2400 reactor, an industrial scale design with 2400 MW thermal power. The design goals for GFR2400 aim for a core outlet temperature of around 850°C, a compact core with 100 MWth/m³ power density, a low enough plutonium content to allow wide deployment, and a self-sustaining core in terms of plutonium consumption without the need for fertile blanket in order to reduce proliferation risks [54]. The previous design of the GFR in the original GIF roadmap was a 600 MWth concept, which was modified because it could not meet the break-even breeding requirement. The original concept of the GFR2400 with carbide fuel pins and silicon-carbide ceramic cladding was designed by the French Atomic Energy Commission (CEA) and further developed in the framework of the GoFastR project of the Euratom Seventh Framework Programme [55, 56]. Due to the high porosity of the SiC fiber, an additional W/Re and Re liner was built in the fuel pins to withhold gaseous fission products. Although the rhenium liners cause significant neutron penalty during normal operation, these metals have favourable effect in accidental situations with spectrum thermalisation, for example in the case of steam or water ingress in the core. The reference core design is composed of inner and outer fuel regions with 252 and 264 assemblies, respectively, as well as 13 Diverse Shutdown Devices (DSD) and 18 Control and Shutdown Devices (CSD). The inner core assemblies have lower and the outer core assemblies have higher Pu content in order to flatten the flux distribution and power profile of the core.

The European design of the LFR is the ELSY (European Lead-cooled SYstem), which is a 1500 MWth pool-type reactor cooled by pure lead, that was developed in the framework of the EU-FP6-ELSY project financed by the Euratom FP6 programme [57]. During the development of the ELSY core two types of fuel assemblies were examined: the first is a hexagonal assembly with steel assembly wrapper, and the second is a wrapperless square assembly, both of which contain UO₂-PuO₂ or nitride based fuel pins with steel cladding [52]. In the present analyses the core design with hexagonal MOX fuel assembly was studied. The hexagonal core consists of three regions with different Pu contents (inner, middle and outer core with 163, 102 and 168 fuel assemblies) in order to flatten the power distribution. The total actinide content of the core is 50 tons, and the fuel cycle operation is envisaged in a three-batch cycle with 3×547.5 EFPD (Effective Full Power Day) length, therefore one-third of the core is loaded with fresh fuel at the end of each cycle.

The reference design of the Generation IV SFR was developed in the framework of the CP-ESFR (Collaborative Project for a European Sodium-cooled Fast Reactor) project financed by the Euratom 7th Framework Programme [58]. The French CEA initially developed two core concepts for the ESFR (European Sodium-cooled Fast Reactor): one with oxide fuel and one with carbide fuel. As the carbide core is a
3.1. Reference core designs

Figure 3.1: Core layouts in the KENO-VI models of the reference Generation IV fast reactor core configurations (light, medium and dark blue: fuel assemblies, yellow: control rod assemblies, gray: radial reflector) [P3]

converter with $CR < 1$, therefore the iso-breeder oxide core was selected as subject of the analyses. The reference SFR core investigated in this thesis is therefore the 3600 MWth MOX fueled design specified by the OECD/NEA Working Group on Reactor Systems (WPRS) [53]. The core contains 225 inner fuel assemblies and 228 outer fuel assemblies with different Pu content, as well as 18+9 CSD and DSD assemblies. The French CEA and the EDF and AREVA companies also started a new research project in 2010 aimed at the development of a demonstrator SFR called ASTRID (Advanced Sodium Technological Reactor for Industrial Demonstration), which is also a converter core due its smaller size [59].

3.1.2 Generation III thermal reactors

In order to investigate the recycling of excess plutonium produced by fast reactors as MOX fuel in thermal reactors, burn-up models were developed for MOX fuel assemblies of two Generation III Light Water Reactors: the European Pressurized Reactor (EPR) and the VVER-1200. The EPR is a 4500 MWth Generation III+ Pressurized Water Reactor (PWR) design, which combines the safety and environmental features of the most recent French and German PWRs with improved neutronic and thermal efficiency. Specific features of the EPR will also allow the reactor to accommodate fuel management with higher MOX fuel assembly ratio than its predecessors [60]. The VVER-1200 (AES-2006) is the most recent Generation III+ PWR design of the Russian Rosatom company, a further developed version of the VVER-1000 with increased thermal power (3200 MWth) and additional passive safety systems [61].
Table 3.1: Main parameters of the reference Generation IV fast reactor core configurations [54, 52, 53]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>GFR2400</th>
<th>ELSY</th>
<th>ESFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal power</td>
<td>2400 MW</td>
<td>1500 MW</td>
<td>3600 MW</td>
</tr>
<tr>
<td>Fuel material</td>
<td>(U,Pu)C</td>
<td>(U,Pu)O₂</td>
<td>(U,Pu)O₂</td>
</tr>
<tr>
<td>Cladding material</td>
<td>SiC</td>
<td>T91 steel</td>
<td>ODS steel</td>
</tr>
<tr>
<td>Coolant</td>
<td>He</td>
<td>liquid Pb</td>
<td>liquid Na</td>
</tr>
<tr>
<td>Avg. coolant temp.</td>
<td>665°C</td>
<td>440°C</td>
<td>470°C</td>
</tr>
<tr>
<td>Active core volume</td>
<td>24 m³</td>
<td>21 m³</td>
<td>18 m³</td>
</tr>
<tr>
<td>Actinide mass</td>
<td>67.7 t</td>
<td>50 t</td>
<td>71.4 t</td>
</tr>
<tr>
<td>Fuel assembly type</td>
<td>hexagonal</td>
<td>hexagonal</td>
<td>hexagonal</td>
</tr>
<tr>
<td>Nr. of fuel assemblies</td>
<td>252+264</td>
<td>163+102+168</td>
<td>225+228</td>
</tr>
<tr>
<td>Nr. of fuel pins in FA</td>
<td>217</td>
<td>169</td>
<td>271</td>
</tr>
<tr>
<td>Active height</td>
<td>165 cm</td>
<td>120 cm</td>
<td>101 cm</td>
</tr>
<tr>
<td>Fuel assembly pitch</td>
<td>17.83 cm</td>
<td>21.6 cm</td>
<td>21.22 cm</td>
</tr>
<tr>
<td>Fuel pin lattice pitch</td>
<td>1.157 cm</td>
<td>1.55 cm</td>
<td>1.19 cm</td>
</tr>
<tr>
<td>Average burn-up</td>
<td>50 MWd/kgHM</td>
<td>60 MWd/kgHM</td>
<td>100 MWd/kgHM</td>
</tr>
<tr>
<td>Fuel management</td>
<td>3×481 EFPD</td>
<td>3×547.5 EFPD</td>
<td>5×410 EFPD</td>
</tr>
</tbody>
</table>

The AES-2006 design shares a number of common elements with the VVER-1000, including similar fuel assemblies (TVS-2 and TVS-AES-2006), which both contain top and bottom nozzles and spacer grids. The main difference in the assembly structures is the active height which was increased by 20 cm in the case of the AES-2006, in contrast with the length of the bottom nozzle, which was shortened in the new design. The increased number (121) of control rod bundles permit a high percent of MOX fuel loading in the VVER-1200 as well [62]. Main relevant core parameters of the two reference Light Water Reactors are listed in Table 3.2, and the fuel assembly layouts are depicted in Figure 3.2.

3.2 Selection of the fitting parameters

It is a common assumption that the core geometry, structural materials, temperatures and other thermal hydraulic parameters do not change during the fuel cycle simulation, therefore the fuel composition was considered as the only focus of the cross-section parametrization (except for the boric acid concentration in the case
3.2. Selection of the fitting parameters

Figure 3.2: Assembly layouts in the KENO-VI models of the reference Generation III thermal reactor MOX fuel assemblies (dark blue: fuel pins, light blue: moderator)

of thermal reactors). The spectral effects of reactivity control were also neglected, which is a usual approximation in fuel cycle calculations, and it is not expected to have significant effects in the fuel cycle performance of the reactors. The one-group microscopic cross-sections are weighted with the neutron spectrum, therefore those actinides and fission products can best describe the fitted cross-sections which have the greatest influence on the spectrum. This can be estimated by their contribution to the total reaction rates, i.e. isotopes with the highest absorption and scattering reaction rates have the most significant effect on the one-group cross-sections. On the other hand, the number of data points needed for an accurate fit increases rapidly with the number of fitting parameters, therefore a compromise had to be made regarding the number of descriptive isotopes. Having considered the desired accuracy of the parametrization, those actinides were chosen to describe the fuel composition which had taken up 99.9% of the reaction rates in preliminary core calculations and fuel cycle studies [P10, P11]. Following this principle, actinide isotopes which are listed in Table 3.3 as well as the overall quantity of fission products were selected for the fast reactor and thermal reactor burn-up models.

Fission products which were considered in the transport calculations were selected with a similar approach based on the results of assembly-wise SCALE 6.0 TRITON [50] depletion calculations for each reactor (see Tables 3.4 and 3.5). In contrast with the actinide isotopes, the consideration of the large number of FPs was possible because their contribution was only described with their overall quantity in the fitting procedure. Exceptions extend to the most important reactor poisons $^{135}\text{Xe}$ and $^{149}\text{Sm}$, which were additional fitting parameters in the MOX fuel assembly burn-up models due to their high thermal absorption cross-sections.
Table 3.2: Main parameters of the reference Generation III thermal reactor core configurations [60, 61]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>EPR</th>
<th>VVER-1200</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal power</td>
<td>4500 MW</td>
<td>3200 MW</td>
</tr>
<tr>
<td>Fuel material</td>
<td>UO₂ or MOX</td>
<td>UO₂ or MOX</td>
</tr>
<tr>
<td>Cladding material</td>
<td>M5 steel</td>
<td>Zr-1%Nb</td>
</tr>
<tr>
<td>Coolant</td>
<td>H₂O</td>
<td>H₂O</td>
</tr>
<tr>
<td>Avg. coolant temp.</td>
<td>314°C</td>
<td>329°C</td>
</tr>
<tr>
<td>Active core volume</td>
<td>14.1 m³</td>
<td>8.7 m³</td>
</tr>
<tr>
<td>Actinide mass</td>
<td>127.1 t</td>
<td>78.4 t</td>
</tr>
<tr>
<td>Fuel assembly type</td>
<td>17×17 square</td>
<td>hexagonal</td>
</tr>
<tr>
<td>Nr. of fuel assemblies</td>
<td>241</td>
<td>163</td>
</tr>
<tr>
<td>Nr. of fuel pins in FA</td>
<td>265</td>
<td>312</td>
</tr>
<tr>
<td>Active height</td>
<td>420 cm</td>
<td>373 cm</td>
</tr>
<tr>
<td>Fuel assembly pitch</td>
<td>17.95 cm</td>
<td>23.6 cm</td>
</tr>
<tr>
<td>Fuel pin lattice pitch</td>
<td>1.26 cm</td>
<td>1.275 cm</td>
</tr>
<tr>
<td>Average burn-up</td>
<td>50 MWd/kgHM</td>
<td>50 MWd/kgHM</td>
</tr>
<tr>
<td>Fuel management</td>
<td>4×353 EFPD</td>
<td>4×365 EFPD</td>
</tr>
</tbody>
</table>

3.3 Preparation of the cross-section databases

The fitting of the one-group cross-sections as functions of the detailed fuel composition can be performed based on an appropriate cross-section database, which contains numerous (few thousand) data points with different fuel compositions and corresponding cross-section sets. The transport calculations needed for the cross-section databases of both the Generation IV fast reactor burn-up models and the burn-up models of Generation III thermal reactor MOX fuel assemblies were performed with the SCALE 6.0 code, but the difference in neutron spectrum required different methodologies for fast and thermal reactors. In particular, the short migration length in thermal reactors allowed the transport calculations to be performed for an individual fuel assembly with reflective boundary conditions, but a more rigorous treatment of fission products was needed due to their higher absorption at thermal energies. The following subsections give a detailed description of the applied calculation schemes and the preparation of the cross-section databases in the case of Generation IV fast reactors and Generation III thermal reactor MOX fuel assemblies.
### Table 3.3: Isotopes selected as fitting parameters for fast and thermal neutron spectrum based on preliminary GFR2400 and EPR MOX calculations

| Component | GFR2400 | | | EPR MOX | | | |
|-----------|---------|---------|--------|---------|---------|--------|
|           | Parameter | Rel. reaction rate (%) | | Parameter | Rel. reaction rate (%) | | |
| U         |          |                     |        |          |                     |        |
|           | $^{234}\text{U}$ | 0.43 |        | $^{234}\text{U}$ | 0.02 |        |
|           | $^{235}\text{U}$ | 0.43 |        | $^{235}\text{U}$ | 0.75 |        |
|           | $^{236}\text{U}$ | 0.35 |        | $^{236}\text{U}$ | 0.03 |        |
|           | $^{238}\text{U}$ | 43.05 |        | $^{238}\text{U}$ | 23.29 |        |
| Pu        | $^{238}\text{Pu}$ | 2.26 |        | $^{238}\text{Pu}$ | 0.86 |        |
|           | $^{239}\text{Pu}$ | 35.99 |        | $^{239}\text{Pu}$ | 35.83 |        |
|           | $^{240}\text{Pu}$ | 7.80 |        | $^{240}\text{Pu}$ | 15.04 |        |
|           | $^{241}\text{Pu}$ | 2.54 |        | $^{241}\text{Pu}$ | 12.47 |        |
|           | $^{242}\text{Pu}$ | 0.73 |        | $^{242}\text{Pu}$ | 2.72 |        |
| MA        | $^{237}\text{Np}$ | 1.59 |        | $^{237}\text{Np}$ | 0.07 |        |
|           | $^{239}\text{Np}$ | 0.04 |        | $^{239}\text{Np}$ | 0.01 |        |
|           | $^{241}\text{Am}$ | 2.62 |        | $^{241}\text{Am}$ | 1.59 |        |
|           | $^{242m}\text{Am}$ | 0.34 |        | $^{242m}\text{Am}$ | 0.08 |        |
|           | $^{243}\text{Am}$ | 0.85 |        | $^{243}\text{Am}$ | 1.16 |        |
|           | $^{244}\text{Cm}$ | 0.34 |        | $^{242}\text{Cm}$ | 0.01 |        |
|           | $^{244}\text{Cm}$ | 0.21 |        | $^{244}\text{Cm}$ | 0.22 |        |
| FP        | Total FP | 0.33 |        | Total FP | 5.84 |        |
|           | $^{135}\text{Xe}$ | |        | $^{135}\text{Xe}$ | |        |
|           | $^{149}\text{Sm}$ | |        | $^{149}\text{Sm}$ | |        |
| Total     | 99.9 | | | 99.9 | | |
| Other     | B | | | - | | |
Table 3.4: The most important fission products considered in the fast reactor core calculations and their contributions to the total FP reaction rates

<table>
<thead>
<tr>
<th>Fission product</th>
<th>Rel. reaction rate (%)</th>
<th>GFR2400</th>
<th>ELSY</th>
<th>ESFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{101}$Ru</td>
<td>9.37</td>
<td>101Ru</td>
<td>11.30</td>
<td>105Pd</td>
</tr>
<tr>
<td>$^{105}$Pd</td>
<td>8.87</td>
<td>105Pd</td>
<td>11.13</td>
<td>105Pd</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>7.96</td>
<td>103Rh</td>
<td>8.05</td>
<td>103Rh</td>
</tr>
<tr>
<td>$^{103}$Rh</td>
<td>7.28</td>
<td>107Pd</td>
<td>7.68</td>
<td>107Pd</td>
</tr>
<tr>
<td>$^{133}$Cs</td>
<td>6.08</td>
<td>133Cs</td>
<td>7.24</td>
<td>133Cs</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>5.85</td>
<td>149Sm</td>
<td>5.90</td>
<td>149Sm</td>
</tr>
<tr>
<td>$^{149}$Sm</td>
<td>4.74</td>
<td>151Sm</td>
<td>4.99</td>
<td>151Sm</td>
</tr>
<tr>
<td>$^{118}$Sn</td>
<td>0.01</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>99.9</td>
<td>Total</td>
<td>99.9</td>
<td>Total</td>
</tr>
</tbody>
</table>

Table 3.5: The most important fission products considered in MOX thermal reactor calculations and their contributions to the total FP reaction rates

<table>
<thead>
<tr>
<th>Fission product</th>
<th>Rel. reaction rate (%)</th>
<th>EPR</th>
<th>VVER-1200</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{135}$Xe</td>
<td>19.51</td>
<td></td>
<td>16.44</td>
</tr>
<tr>
<td>$^{103}$Rh</td>
<td>9.94</td>
<td></td>
<td>10.28</td>
</tr>
<tr>
<td>$^{131}$Xe</td>
<td>6.47</td>
<td></td>
<td>9.09</td>
</tr>
<tr>
<td>$^{149}$Sm</td>
<td>6.45</td>
<td></td>
<td>6.75</td>
</tr>
<tr>
<td>$^{133}$I</td>
<td>6.45</td>
<td></td>
<td>6.38</td>
</tr>
<tr>
<td>$^{152}$Sm</td>
<td>5.03</td>
<td></td>
<td>5.07</td>
</tr>
<tr>
<td>$^{147}$Pm</td>
<td>4.94</td>
<td></td>
<td>5.05</td>
</tr>
<tr>
<td>$^{112}$Cd</td>
<td>0.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>99.9</td>
<td></td>
<td>Total</td>
</tr>
</tbody>
</table>
3.3. Preparation of the cross-section databases

3.3.1 Fast reactors

The three-dimensional models of the three fast reactors depicted in Figure 3.1 were created in the KENO-VI transport module of the SCALE 6.0 code. In order to decrease the computational time needed for the core calculations, a SCALE 6.0 sequence with two-step homogenization was used in the preparation of the cross-section databases (see Figure 3.3), and the core geometries were built from the homogenized fuel assemblies of the different fuel regions. In the first homogenization step the hexagonal unit cell was homogenized in 1D cylindrical geometry using white boundary conditions. The height of the core was taken into account with buckling correction in the resonance treatment for the proper consideration of axial leakage. The fuel assemblies consisted of the homogenized unit cells, the assembly wrapper and the coolant between the assemblies, and they were also homogenized in 1D cylindrical geometry. Other core components and structural elements (axial and radial reflectors, rod followers and gas plenums) were modeled with smeared densities and treated in infinite homogeneous medium approximation. The three-dimensional core models were built from the homogenized assemblies and other core components, and the full core calculations were performed using the KENO-VI criticality code. The actinide reaction rates and the average fluxes in the different fuel regions were recorded from the calculations, as well as the $k_{\text{eff}}$ for the different fuel compositions. With the two-step homogenization the time demand of one full core calculation was about 40 minutes on one 2 GHz CPU with 1 GByte memory.

Due to the high-dimensional fitting domain, purely deterministic sampling methods such as orthogonal sampling or Latin hypercube sampling would have required
an impracticable number of data points [63, 64]. The different compositions were therefore obtained by random sampling of the actinide mass fractions – although combined deterministic and Monte Carlo sampling might be a future improvement [34] – taking into account the following constraints:

- Pu fraction in the fuel was uniformly sampled between 10-25% of the total actinide mass.
- MA fraction varied between 0-10% of the total actinide mass.
- The rest of the actinide content was U.
- The Pu content of the different core regions were kept at constant ratio corresponding to the initial loading. The MA fraction was the same in all regions.
- Fission products were considered with an average fission yield vector, and their total quantity was calculated from the sampled burn-up which varied between the specified limits.

The use of an average, characteristic fission product composition was possible due to the fast neutron spectrum of the reactors. Since most fission products have high absorption mainly at thermal energies, the variation of the FP composition during burn-up has small effect on the actinide one-group cross-sections. Isotopic compositions of the actinide elements on the other hand were also randomly sampled between the beginning-of-life (BOL) composition and equilibrium compositions estimated by preliminary studies (see Table 3.6) [P10]. The aim of the random sampling was to cover a wide range of possible fuel compositions that can occur in the fuel cycle simulations, therefore unrealistic compositions were not only unnecessary in the cross-section database, but they would have decreased the accuracy of the fitted polynomials for realistic fuel compositions. To eliminate these points from the database, preliminary fits of the $k_{eff}$ were performed based on the results of few hundred core calculations, and rejection method was used to sample compositions with estimated $k_{eff}$ ranging from 0.90 to 1.15.

The FITXS models of the reactors calculate the evolution of the average fuel composition in the cores, therefore separate handling of the different core regions is not possible during the fuel cycle simulation. On the other hand the fitting is performed on core homogenized cross-sections which are calculated using region-wise fluxes and atomic densities. In order to preserve total reaction rates for the full cores, a homogenization correction factor was introduced ($h$), that can be expressed as a weighted sum of atomic density ratios and flux ratios between the different core
3.3. Preparation of the cross-section databases

<table>
<thead>
<tr>
<th>Component</th>
<th>Isotope</th>
<th>Mass percent in component</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Minimum</td>
</tr>
<tr>
<td>U</td>
<td>234U</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>235U</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>236U</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>238U</td>
<td>99</td>
</tr>
<tr>
<td>Pu</td>
<td>238Pu</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>239Pu</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>240Pu</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>241Pu</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>242Pu</td>
<td>1</td>
</tr>
<tr>
<td>MA</td>
<td>237Np</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>239Np</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>241Am</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>242mAm</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>243Am</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>244Cm</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>245Cm</td>
<td>0</td>
</tr>
</tbody>
</table>

\[ ^{239}\text{Np fraction was compared to the total actinide mass} \]

The total reaction rate can be calculated using the correction factors as follows:

\[ R = N\sigma h\Phi V, \]  \hspace{1cm} (3.1)

where \( N \) denotes the average atomic density of the nuclide in the core, \( \sigma \) denotes the core homogenized one-group microscopic cross-section, \( h \) denotes the homogenization factor and \( V \) is the total fuel volume. The general form for \( n \) regions is the following:

\[ \sigma = \frac{\sum_{k=1}^{n} N_k\sigma_k\Phi_k V_k}{\sum_{k=1}^{n} N_k\Phi_k V_k}, \]  \hspace{1cm} (3.2)

\[ N = \frac{\sum_{k=1}^{n} N_k V_k}{V}, \]  \hspace{1cm} (3.3)

\[ \Phi = \frac{\sum_{k=1}^{n} \Phi_k V_k}{V}, \]  \hspace{1cm} (3.4)
3.3.2 Thermal MOX fuel assemblies

Calculating the average one-group cross-sections in infinite lattice geometry is a good approximation for central fuel assemblies of thermal reactors, since the radial reflector and distant fuel assemblies have low or negligible effect on the spectrum due to the short migration length. This greatly decreases the computational cost of one data point, because the transport calculations can be performed on an individual fuel assembly using reflective boundary conditions. The three-dimensional SCALE models of the MOX fuel assemblies were built in accordance with the fuel assembly layouts in Figures 3.2. The axial structures of the assemblies were also considered including bottom and top nozzles. Different types of fuel pins could have been handled using the same methodology as the fast reactor core regions in the previous subsection, but they tend to have small effect on the evolution of the average fuel composition from the fuel cycle point of view, therefore average fuel pin compositions were used in the preparation of the cross-section databases. The computational time needed for one transport calculation was low enough (around 30 minutes on one 2 GHz CPU with 1 GByte memory) so that no cell homogenization was needed in the case of the MOX fuel assemblies (see Figure 3.4).
The different fuel compositions were also randomly sampled with some additional constraints due to the thermal neutron spectrum. In the fast reactor core calculations fission products were considered with an average fission yield vector because the variation of individual FP fractions had small effect on the hard neutron spectrum. A more rigorous treatment of the fission products was needed in the case of thermal reactors, therefore the mass fractions of the selected fission product isotopes were fitted as fourth-order polynomial functions of the burn-up (see Figure 3.5). Since the most important reactor poisons \(^{135}\text{Xe}\) and \(^{149}\text{Sm}\) are also fitting parameters, their quantities were randomly sampled between specific limits with uniform distribution, and their nuclide chains are included explicitly in the burn-up calculation. Boric acid concentration was chosen as an additional parameter as well because of its significant thermal absorption. Assuming a linear decrease from the beginning to the end of each cycle, the boric acid concentration used in the given transport calculation was also calculated from the sampled burn-up. The different fuel (and moderator) compositions for the MOX fuel assembly cross-section databases were therefore obtained using the following constraints:

- Pu fraction in the fuel was uniformly sampled between 5-11% of the total actinide mass.
- MA fraction varied between 0-1% of the total actinide mass.
- The rest of the actinide content was U.
- The mass fractions of individual FP isotopes were calculated from the sampled burn-up (0-50 MWd/kg) based on the fitted fourth-order polynomials.
- \(^{135}\text{Xe}\) and \(^{149}\text{Sm}\) concentrations were randomly sampled between appropriate upper and lower limits.
• Boric acid concentration was calculated from the sampled burn-up assuming a linear decrease throughout the cycles.

Isotopic compositions of elements were also randomly sampled to cover a wide range of possible MOX fuel compositions that can occur in the fuel cycle simulations. The upper and lower limits for the isotopic fractions used in the transport calculations can be found in Table 3.7.

### 3.4 Cross-section fitting procedure

The large number of data points and fitting parameters suggests that it is reasonable to choose a fitting method that directly provides the global minimum of the squared residuals. This limits the set of possible fitted functions to those which are linear in their coefficients. Polynomials meet this condition, and since preliminary investigations showed that higher-order terms did not improve the accuracy of the fittings, therefore the following second-order polynomial was chosen to describe the one-group cross-sections and the $k_{\text{eff}}$ as functions of the fuel composition:

$$
\sigma(N_1, N_2, \ldots, N_n) = a_0 + \sum_{i=1}^{n} a_i N_i + \sum_{i=1}^{n} \sum_{j=i}^{n} a_{ij} N_i N_j,
$$  \hspace{1cm} (3.9)

which consists of a constant parameter, the first and second order terms of the atomic densities and their cross-products. If we apply this equation for all fuel compositions
### Table 3.7: Limits for the random sampling of the EPR isotopic compositions

<table>
<thead>
<tr>
<th>Component</th>
<th>Isotope</th>
<th>Mass percent in component</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Minimum</td>
</tr>
<tr>
<td>U</td>
<td>$^{234}$U</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>$^{235}$U</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>$^{236}$U</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>$^{238}$U</td>
<td>99</td>
</tr>
<tr>
<td>Pu</td>
<td>$^{238}$Pu</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>$^{239}$Pu</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>$^{240}$Pu</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>$^{241}$Pu</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>$^{242}$Pu</td>
<td>0</td>
</tr>
<tr>
<td>MA</td>
<td>$^{237}$Np</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>$^{239}$Np*</td>
<td>5·10$^{-3}$</td>
</tr>
<tr>
<td></td>
<td>$^{241}$Am</td>
<td>5</td>
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<tr>
<td></td>
<td>$^{242m}$Am</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>$^{243}$Am</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>$^{244}$Cm</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>$^{245}$Cm</td>
<td>0</td>
</tr>
<tr>
<td>FP</td>
<td>$^{135}$Xe*</td>
<td>3·10$^{-5}$</td>
</tr>
<tr>
<td></td>
<td>$^{149}$Sm*</td>
<td>7·10$^{-5}$</td>
</tr>
</tbody>
</table>

*Compared to the total actinide mass
The FITXS method included in the core calculations, we get an overdetermined linear equation system concerning the fitted coefficients for each cross-section and the multiplication factor. Written in matrix form for \( m \) fuel compositions in the cross-section database:

\[
M \mathbf{a} = \begin{bmatrix}
1 & N_1^{(1)} & N_2^{(1)} & \ldots & N_{n-1}^{(1)} & N_n^{(1)} \\
1 & N_1^{(2)} & N_2^{(2)} & \ldots & N_{n-1}^{(2)} & N_n^{(2)} \\
\vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\
1 & N_1^{(m)} & N_2^{(m)} & \ldots & N_{n-1}^{(m)} & N_n^{(m)}
\end{bmatrix}
\begin{bmatrix}
a_0 \\
a_1 \\
a_2 \\
\vdots \\
a_{n-1} \\
a_n
\end{bmatrix} = \mathbf{\sigma},
\]

where the \( m \) rows of the matrix \( M \) contain 1 as the constant parameter, the atomic densities from the corresponding core calculation as first order terms, as well as their squares and cross-products as second order terms. The least-squares solution of the (3.10) equation system (\( \| \mathbf{\sigma} - M \mathbf{a} \|^2 = \text{min.} \)) can be obtained by solving the Gaussian normal equation

\[
(M^T M) \mathbf{a} = M^T \mathbf{\sigma}.
\]

Provided that the column vectors of \( M \) are linearly independent (which is true with great certainty due to the random sampling of the fuel compositions), the normal equation has a unique solution, which is given by the pseudoinverse [65]:

\[
\mathbf{a} = M^+ \mathbf{\sigma},
\]

where

\[
M^+ = (M^T M)^{-1} M^T.
\]

By substituting the fitted functions in the Bateman equations, the transmutation matrix can be expressed as a function of the fuel composition:

\[
\frac{dN}{dt} = A(N) N.
\]

Equation (3.14) is a nonlinear differential equation system, but its numerical integration is fast enough to be integrated into dynamic fuel cycle simulations. In the fuel cycle studies described in the next chapter a predictor-corrector scheme was applied for this purpose based on the matrix exponential solution of linear ODEs (see Section 4.1.2).

### 3.5 Fitting results and verification

The verification of the accuracy and applicability of the FITXS method and the developed burn-up models was twofold: first, the accuracy of the fitted cross-sections
and $k_{\text{eff}}$ was checked, and second, the results of the burn-up models were compared with the results of SCALE 6.0-based burn-up calculations. The fitting results and the verification of the burn-up models are discussed in the following two subsections.

### 3.5.1 Accuracy of the fitted functions

The large number of fitted parameters is an obvious challenge for any fitting algorithm, therefore the accuracy of the fitted functions needed to be carefully checked. The results of the fittings were compared with the calculated cross-sections of the prepared database. The errors of the fittings were measured with the normalized root mean square error (NRMSE):

$$\text{NRMSE} = \frac{1}{\bar{y}} \sqrt{\frac{\sum_{i=1}^{n} [f(x_i) - y_i]^2}{n}}$$

(3.15)

where $f(x_i)$ is the value of the fitted function at $x_i$, $y_i$ is the reference value at $x_i$, $n$ is the number of data points used for the fitting, and $\bar{y}$ is the average of the values.

The fitted functions showed good accuracy at describing the one-group cross-sections with typical errors well below 1% for the important (n,f) and (n,\gamma) reactions, and generally in the order of magnitude of the statistical uncertainties of the Monte Carlo transport calculations. Higher errors occurred in the case of (n,2n) and (n,3n) cross-sections, because the high threshold energies of these reactions lead to high statistical uncertainties in the Monte Carlo determination of reaction rates. Leniau et al. [34] have also experienced higher errors for these reactions when training their mean cross-section predictors. However, the very low cross-sections of these reactions also indicate that these errors have limited effect on the spent fuel composition. The average relative errors for the different reaction types and the $k_{\text{eff}}$, as well as the flux ratios are listed in Tables 3.8 and 3.9, whereas typical fitting results are shown in Figures 3.6-3.8. The results also show higher fitting errors in the case of MOX fuel assemblies (see Table 3.9), which is due to the fact that one-group cross-sections are more sensitive to variations of the fuel composition in thermal neutron spectrum.

Comparison of the fitting errors of individual cross-sections with their Monte Carlo statistical uncertainties recorded from the transport calculations (see Figures 3.9 and 3.10) shows that major fraction of the errors can be accounted for the Monte Carlo simulation statistics, and that the low errors are not due to overfitting. Histograms of the relative errors (see Figures 3.11 and 3.12) also reveal that the errors have normal distributions with mean values close to zero, which means that no global under- or overpredictions of the cross-sections occur. The magnitude of the relative errors are acceptable for cross-section parametrization used in scenario
Figure 3.6: Relative errors for the fitting of the $k_{\text{eff}}$ for the GFR2400 (left) and the $k_{\infty}$ for the EPR (right)

Figure 3.7: Relative errors for the fitting of the $^{239}\text{Pu} (n,f)$ cross-section for the ELSY (left) and the $^{238}\text{U} (n,\gamma)$ cross-section for the VVER-1200 (right)
3.5. Fitting results and verification

Figure 3.8: Relative errors for the fitting of the $^{244}$Cm $(n,2n)$ cross-section for the ESFR (left) and the $^{243}$Am, $(n,3n)$ cross-section for the GFR2400 (right)

In order to verify the accuracy of the FITXS burn-up models, two hundred depletion calculations were performed for one burn-up cycle of the fast reactors and the MOX fuel assemblies using cross-sections calculated at each time step with the SCALE 6.0 code, as well as with the fitted cross-sections. The initial fuel compositions of the reactors were also selected with random sampling, and rejection method was used to obtain compositions with initial $k_{\text{eff}}$ ranging from 1.0 and 1.05 in the case of fast reactors, and $k_{\infty}$ ranging from 1.05 to 1.15 in the case of thermal MOX fuel assemblies. In the case of fast reactors the FITXS method showed very good agreement between the results of SCALE 6.0-based and FITXS-based burn-up calculations, with typical relative errors well below 0.1% for actinides which have higher than $10^{-5}$ relative end-of-cycle mass in the fuel (see Figure 3.13 and Table A.1 in Appendix A). The
Figure 3.9: Comparison of the relative fitting errors and statistical uncertainties of the cross-sections (GFR24/00)

Figure 3.10: Comparison of the relative fitting errors and statistical uncertainties of the cross-sections (EPR)
### 3.5. Fitting results and verification

#### Figure 3.11: Histograms of the relative errors of the fittings of the $k_{\text{eff}}$ and $f_1$ ($GFR2400$)

- $\mu = -2.59 \cdot 10^{-7}$, $\sigma = 4.0 \cdot 10^{-4}$
- $\mu = -9.6 \cdot 10^{-6}$, $\sigma = 3.1 \cdot 10^{-3}$

#### Figure 3.12: Histograms of the relative errors of the fittings of the $^{238}\text{U} (n,\gamma)$ and $^{239}\text{Pu} (n,f)$ cross-sections ($GFR2400$)

- $\mu = -8.5 \cdot 10^{-8}$, $\sigma = 3.2 \cdot 10^{-4}$
- $\mu = -6.4 \cdot 10^{-8}$, $\sigma = 2.8 \cdot 10^{-4}$
Table 3.8: Average relative fitting errors for fast reactors

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Relative error (%)</th>
<th>GFR2400</th>
<th>ELSY</th>
<th>ESFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n,f)</td>
<td>0.062</td>
<td>0.052</td>
<td>0.056</td>
<td></td>
</tr>
<tr>
<td>(n,γ)</td>
<td>0.159</td>
<td>0.046</td>
<td>0.053</td>
<td></td>
</tr>
<tr>
<td>(n,2n)</td>
<td>0.622</td>
<td>0.725</td>
<td>0.701</td>
<td></td>
</tr>
<tr>
<td>(n,3n)</td>
<td>7.086</td>
<td>7.937</td>
<td>7.985</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Relative error (%)</th>
<th>GFR2400</th>
<th>ELSY</th>
<th>ESFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>k_{eff}</td>
<td>0.040</td>
<td>0.045</td>
<td>0.047</td>
<td></td>
</tr>
<tr>
<td>Φ_1/Φ_{average}</td>
<td>0.31</td>
<td>0.68</td>
<td>0.83</td>
<td></td>
</tr>
<tr>
<td>Φ_2/Φ_{average}</td>
<td>0.38</td>
<td>0.51</td>
<td>0.85</td>
<td></td>
</tr>
<tr>
<td>Φ_3/Φ_{average}</td>
<td>-</td>
<td>0.85</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.9: Average relative fitting errors for thermal MOX fuel assemblies

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Relative error (%)</th>
<th>EPR</th>
<th>VVER-1200</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n,f)</td>
<td>0.119</td>
<td>0.073</td>
<td></td>
</tr>
<tr>
<td>(n,γ)</td>
<td>0.250</td>
<td>0.151</td>
<td></td>
</tr>
<tr>
<td>(n,2n)</td>
<td>0.565</td>
<td>0.735</td>
<td></td>
</tr>
<tr>
<td>(n,3n)</td>
<td>7.755</td>
<td>8.229</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Relative error (%)</th>
<th>EPR</th>
<th>VVER-1200</th>
</tr>
</thead>
<tbody>
<tr>
<td>k_{∞}</td>
<td>0.141</td>
<td>0.427</td>
<td></td>
</tr>
</tbody>
</table>
Figure 3.13: Average relative difference between GFR2400 burn-up results using SCALE 6.0 and FITXS calculated cross-sections for two hundred different initial fuel compositions

Comparisons for MOX fuel assemblies showed higher errors in accordance with the higher fitting errors (see Figure 3.14 and Table A.2 in Appendix A), although the differences are comparable to those of Leniau et al. [34], and are still considered acceptable in scenario studies. In general the results for actinide isotopes with low mass fractions are less accurate due to the fact that these are mainly produced with (n,2n) and (n,3n) reactions, which have higher fitting errors caused by the large statistical uncertainties.

The overall good agreement between the results of SCALE 6.0-based and FITXS-based burn-up calculations for important actinide isotopes encourages that the FITXS method can be successfully applied to determine the spent fuel composition in fuel cycle simulations with low computational time. The average CPU times needed for the performed burn-up calculations are summarized in Table 3.10. Depending on the time demand of the SCALE 6.0 calculations and the number of points in the database, the preparation of a FITXS burn-up model returns in a few hundred simulated burn-up cycles.
Figure 3.14: Average relative difference between EPR MOX burn-up results using SCALE 6.0 and FITXS calculated cross-sections for two hundred different initial fuel compositions.

Table 3.10: Average computational costs of burn-up calculations with SCALE 6.0 and FITXS calculated cross-sections on a 2 GHz CPU with 2 GByte RAM.

<table>
<thead>
<tr>
<th>Case</th>
<th>CPU time</th>
<th>SCALE</th>
<th>FITXS</th>
</tr>
</thead>
<tbody>
<tr>
<td>GFR2400</td>
<td>1.2 h</td>
<td>0.09 s</td>
<td></td>
</tr>
<tr>
<td>ELSY</td>
<td>1.5 h</td>
<td></td>
<td>0.10 s</td>
</tr>
<tr>
<td>ESFR</td>
<td>1.2 h</td>
<td></td>
<td>0.09 s</td>
</tr>
<tr>
<td>EPR MOX</td>
<td>1.1 h</td>
<td>0.12 s</td>
<td></td>
</tr>
<tr>
<td>VVER-1200</td>
<td>1.1 h</td>
<td>0.12 s</td>
<td></td>
</tr>
</tbody>
</table>
Chapter 4

Fuel cycle studies

In order to demonstrate the applicability of the FITXS method in nuclear fuel cycle simulations, the developed burn-up models were integrated into fuel cycle models containing Generation IV fast reactors, as well as MOX fueled Generation III thermal reactors and conventional Light Water Reactors. The equilibrium closed fuel cycles of the three Generation IV fast reactors were analyzed first, then a more complex scenario was investigated regarding the transition from a conventional LWR fleet to a mixed fleet of fast reactors and MOX fueled thermal reactors.

The simulation program developed for use in the equilibrium closed fuel cycle and transition scenario studies is described in the first section, while the specifications and results of the closed fuel cycle and transition scenario analyses are presented in the second and third sections, respectively. The FITXS burn-up models have also been implemented in the SITON v2.0 code developed in the Hungarian Academy of Sciences Centre for Energy Research (MTA EK), results of which can be found in Brolly et al. [P4] and Halász et al. [P3].

4.1 The simulation program

In order to perform fuel cycle analyses in more complex fuel cycle schemes, I have developed an object-oriented simulation program called JOSSETE (ObJect Oriented Simulation Program for ScEnario STudiEs) in C++ programming language. The simulation program follows the quantities of the important actinide isotopes in the fuel cycle and the reactors, as well as the total mass of fission products. The specifications of the simulation program according to those in Section 2.3.1 are listed in Table 4.1. The program models fuel cycle facilities and material flows in discrete form, and advances time according to discrete events, which represent the operation of the facilities. Front-end facilities cover the enrichment and fuel fabrication (there-
fore mining and conversion are not modeled), whereas back-end facilities cover the interim storage, reprocessing plants and different storages (including final disposal). Lag times are modeled in fuel fabrication plants and interim storages, thus excluding reprocessing times. Fuel depletion in the reactors can be calculated either with burn-up tables, or with the use of cross-section models (precalculated cross-section sets, the FITXS method or a SCALE 6.0 model). Waste radiotoxicity and the isotopic composition of fission product waste is not calculated during the simulation.

Fuel cycle facilities and models in the simulation program were implemented as classes (used defined types with specific data and functions, or methods), and inheritance was used to create the derived classes of specific facilities and calculation methods. The main components and structure of the simulation program are described in the following subsections, including the methods used for burn-up calculation and fresh fuel loading.

### 4.1.1 Facilities and models

There are two main types of components that can be used to build fuel cycle models in the simulation program: facilities and models. Instances of each facility type and model can have connections to other fuel cycle components, which connections can be set anywhere in the simulation input file after the given facilities or models were created. The implemented facilities are derived from the abstract Facility class:

- Enrichment plant;
- Reprocessing facility;
- Fuel fabrication plant;
- Reactor;
- Storage;
  - Interim storage;
  - Mixing storage;
  - Infinite storage.

Models are implemented as derived classes from the abstract base class Model, and include the following:

- Burn-up model;
  - Burn-up table model;
### Table 4.1: Simulation program capabilities

<table>
<thead>
<tr>
<th>Attribute</th>
<th>JOSSETE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Language</td>
<td>C++</td>
</tr>
<tr>
<td>Facilities</td>
<td>Discrete</td>
</tr>
<tr>
<td>Fuel</td>
<td>Discrete</td>
</tr>
<tr>
<td>Simultaneous advanced technologies</td>
<td>LWR, FR (SFR, GFR, LFR)</td>
</tr>
<tr>
<td>Isotopes tracking</td>
<td>Yes (isotopes of U/Pu/MA), only total FP</td>
</tr>
<tr>
<td>Burn-up calculation</td>
<td>Burn-up tables, FITXS method or direct coupling with SCALE 6.0</td>
</tr>
<tr>
<td>Front-end facilities</td>
<td>Enrichment, fabrication</td>
</tr>
<tr>
<td>Reprocessing plants</td>
<td>Yes</td>
</tr>
<tr>
<td>Reprocessing capacity</td>
<td>Automatic/manual</td>
</tr>
<tr>
<td>Reprocessing order</td>
<td>First-in-first-out or homogeneous</td>
</tr>
<tr>
<td>Waste radiotoxicity</td>
<td>No</td>
</tr>
</tbody>
</table>
Fuel cycle studies

- Bateman equation solver;
- Transmutation trajectory solver;

- Cross-section model;
- FITXS model;
- SCALE model;
  * Detailed SCALE model;
  * Homogenized SCALE model;

- Fuel loading model.

Material is transferred between facilities in the form of material packages, which also constitute the base class from which fuel batches in the reactors are derived. Miscellaneous classes are listed in the following:

- Material package;
  - Fuel batch;
- Calendar;
- Facility manager;
- Scenario editor;
- Inventory log;
- Input reader;
- Output processor;
- Transmutation trajectory generator;
- Transmutation trajectory calculator;
- Matrix exponential calculator;
- Command line reader.

The detailed operation of the different facilities and the process of simulation are described in the following subsections.
4.1.2 Burn-up calculation

The simulation program follows the quantities of important actinides (in practice 30-50 nuclides) in the fuel cycle and the reactors, as well as the total mass of fission products. Depletion calculations in the reactors can be performed with three different methods, which are implemented in derived classes of the Burnup model base class:

- Burn-up table model;
- Bateman equation solver;
- Transmutation trajectory solver.

The burn-up tables are tabular databases that provide spent fuel compositions at different burn-up levels of the corresponding fresh fuel composition. In the simulation program burn-up tables are applied for LWRs with UOX fuel, for which spent fuel compositions were calculated with the SCALE 6.0 code in infinite fuel assembly lattice geometry. The actual spent fuel composition is determined with linear interpolation based on the discharge burn-up and the data points in the burn-up table.

The Bateman equation solver calculates fuel depletion based on the matrix form of the nuclide chain equations:

\[
\frac{dN}{dt} = A(N(t)) N(t),
\]  

(4.1)

where the dependence of the transmutation matrix \( A \) on the fuel composition, \( A(N(t)) \) is handled by a Cross-section model, either in the form of parametrized cross-sections with the FITXS method, or a SCALE transport model (Detailed SCALE model or Homogenized SCALE model), which creates the inputs for a full core or infinite fuel assembly lattice calculation with the SCALE 6.0 code, runs the transport calculation and processes its output to obtain the one-group cross-sections. Once the cross-sections and neutron flux are calculated, the Bateman equations are solved with a predictor-corrector scheme based on the matrix exponential solution of linear ordinary differential equations. In the predictor step the transmutation matrix \( A \) is considered constant and the predictor fuel composition is calculated with the following formula:

\[
\tilde{N}(t + \Delta t) = e^{\tilde{A} t} N(t).
\]  

(4.2)

The predictor composition determines another transmutation matrix at \( t + \Delta t \), \( \tilde{A} = A(\tilde{N}(t + \Delta t)) \), and the corrector step is performed with the average of the two
transmutation matrices:

\[ N(t + \Delta t) = e^{(A + \tilde{A}) \Delta t} N(t) . \quad (4.3) \]

The matrix exponentials are calculated by the Matrix exponential calculator class, in which the Padé approximation with scaling and squaring is implemented along with the Krylov subspace method. The above predictor-corrector scheme was compared with the Runge-Kutta-Fehlberg method with satisfactory results (see Halász et al. [P12]).

The Transmutation trajectory solver calculates fuel depletion for each time step with constant cross-sections based on the general analytic solution of the Bateman equations and the TTA method, in particular it utilizes the results of Section 5.3.2. The class creates instances of the Transmutation trajectory generator and Transmutation trajectory calculator classes. The former generates possible transmutation trajectories up to a user-defined number of nuclear transitions and the latter calculates the probabilities of these trajectories. Trajectories above a threshold probability level are used to approximate the transition probabilities for the specific time period (i.e. the elements of the matrix exponential). Although actinide transmutation problems are small-scaled and therefore the TTA method is slower in the case of irradiation, the method is much faster in the case of decay calculations, especially if the trajectories are generated in advance and their coefficients are calculated, which is applied in the simulation program to calculate the decay of material packages in the facilities.

4.1.3 Fresh fuel loading

Several dynamic fuel cycle codes, such as COSI6 and CLASS use fuel loading models to predict the initial excess reactivity needed to reach given burn-up. Equivalence models are commonly used, where the plutonium fraction is determined for the fuel to have the same fissile content as a known fresh fuel. For fast reactors the fertile and fissile materials might be evaluated by a comparison with a theoretical fuel containing only \(^{238}\text{U}\) and \(^{239}\text{Pu}\) with equivalence weights based on the Baker & Ross formula [37]. Mouginot et al. [66] have recently developed a polynomial model which fits quadratic polynomials on the Pu composition to calculate the initial Pu content based on the results of several thousand detailed burn-up calculations. Another novel approach was presented by Leniau et al. [34] using neural networks to estimate the initial loading as well.

In FITXS the fitting of the multiplication factor as a function of the detailed fuel composition also allows the fuel cycle model to calculate \(k_{\text{eff}}\) or \(k_{\infty}\) without the need
for detailed transport calculations and the low computational cost of the burn-up calculation makes it possible to iteratively set the initial fissile content of the fresh fuel, based on the beginning-of-cycle and end-of-cycle $k_{\text{eff}}$ calculated for a given fresh fuel composition. The Fuel loading model class in the simulation program uses the secant method [67] to iteratively set the plutonium content of the fresh fuels to maintain criticality throughout the irradiation in each cycle. Fuel fabrication plants are connected to the fuel loading models, which handle the material requests from storages and create fresh fuel batches for the reactors. MOX fuel assemblies are assumed to be mixed with other assemblies in different reactors, therefore it is not possible to estimate the excess reactivity of full cores, and the Pu content of the fresh MOX assemblies are calculated to maintain the same $k_\infty$ for the fabricated assemblies.

According to these criteria, the fuel loading models can determine the fresh fuel composition with five settings:

- Fixed component fractions;
- Set BOC $k_{\text{eff}}$;
- Set EOC $k_{\text{eff}}$;
- Change between BOC and EOC settings depending on the slope of $k_{\text{eff}}$ change during irradiation (use BOC if $k_{\text{eff}}$ increases, and EOC if $k_{\text{eff}}$ decreases);
- Set the same $k_\infty$ for the fabricated fresh fuel assemblies.

The minor actinide content of the fuels can be fixed values, or a maximum MA fraction can be set for the fresh fuel, up to which all the available MA content from the corresponding storages is used. The fabricated fuel batches contain the information about their fabricated masses and their actual burn-up during and after each irradiation period.

### 4.1.4 Simulation program control

The simulation program is controlled by five classes sorted as miscellaneous in Section 4.1.1:

1. Command line reader: The program is executed from the command line with an option parameter which selects the type of simulation and an input file that specifies the fuel cycle model or the details of other calculations. There are three possible option parameters with the following functions:
Fuel cycle studies

- Simulation of the fuel cycle specified in the input file;
- Burn-up calculation;
- Transport calculation;

2. Input reader: The input processor reads the user-specified input file and uses an instance of the Scenario editor class to build the fuel cycle model and set the specified parameters of facilities and models;

3. Scenario editor: The scenario editor creates instances of the different facilities and models specified in the input file, and provides the list of these entities to an instance of the Facility manager class;

4. Facility manager: The facility manager is the class responsible for running the fuel cycle simulation by advancing time to the next event. Time is handled by the singleton class of Calendar, and the time of the next event is determined by calling the NextEvent() method of each facility. This is followed by calling the Operate() method of the facilities that operate at the time of the next event;

5. Output processor: Each facility and model is responsible for storing time series of appropriate data. Besides, inventory logs can be created in the input, which can ask and store the inventory inside single or multiple facilities at specified time steps. The output processor calls the WriteOutput() methods of each facility and model to append their related data to the output of the simulation.

The simulation is started and ended at input-specified times, and each facility can also possess starting dates and shutdown dates.

4.2 Equilibrium closed fuel cycle studies

In order to investigate the transmutation and breeding properties of the reference Generation IV fast reactors, their equilibrium closed fuel cycle operation was analyzed using the developed simulation program and the FITXS burn-up models. The following subsections present the fuel cycle model and recycling strategies used for the simulations, as well as the results of the analyses.

4.2.1 Recycling strategies

The Generation IV fast reactor burn-up models were integrated into the closed fuel cycle model depicted in Figure 4.1. The spent fuel composition of the LWRs was
considered with constant composition corresponding to 50 MWd/kg burn-up and 5 years cooling. The reprocessed Pu content of the spent LWR fuel was used for the start-up of the fast reactors and as fissile material feed when needed in order to set the excess reactivity of the fast reactor cores. The spent fuel of the fast reactors was reprocessed after 5 years cooling time, and its actinide content was partially or entirely recycled in the fast reactors. Reprocessing losses and fabrication times were considered to be zero in order to obtain the maximal fuel utilization capabilities of the reactors. Three different recycling strategies were investigated in order to assess the fissile material breeding and minor actinide burning capabilities of the reactors:

- Case 1: U and Pu multirecycling;
- Case 2: U, Pu and MA multirecycling;
- Case 3: Fixed MA content in the fresh fuel.

The fresh fuel compositions of the reactors were determined in each burn-up cycle with iteration based on the beginning-of-cycle and end-of-cycle $k_{\text{eff}}$. The whole transition from initial state to equilibrium was simulated, and the concentrations of actinide isotopes up to $^{248}\text{Cm}$ were followed in the calculations, although the effect of the inclusion of higher elements on the equilibrium fuel compositions of the reactors is subject to further analysis. The results of the equilibrium closed fuel cycle analyses obtained with the three recycling options are discussed in the following subsection.

4.2.2 Equilibrium closed fuel cycle results

All the investigated scenarios were simulated from the initial state until the equilibrium state was reached in the system. Due to the multiple recycling of spent fuel the equilibrium state was reached on a much longer timescale (around hundred years) than in once-through fuel cycles, but the overall Pu and MA content of the core, as well as core performance parameters such as the breeding gain typically approached their equilibrium value within the expected operational lifetimes of the reactors.

The first examined option, Case 1 assumed only U and Pu recycling into the fast reactors in order to provide a reference case for further investigations. Figure 4.2 shows that the Pu feed from LWR spent fuel decreases quickly and in the equilibrium less than 2% Pu feed is needed in the three fast reactors compared to the total Pu content of the fresh fuel, indicating that the reactors are close to break-even breeding. This Pu need, however, is due to the decay of fissile $^{241}\text{Pu}$ during the 5 years cooling of the spent fuel, as each core has a slightly positive breeding gain, therefore the
reactors would act as slight breeders if there was no cooling before reprocessing. With 2 years cooling time assumed for the spent fuel (which would be possible with advanced pyroprocessing technology), this low external Pu feed already diminishes in the equilibrium. The initial Pu contents of the cores increase due to the piling up of $^{240}\text{Pu}$ and the decrease of $^{241}\text{Pu}$ which is typical for fast reactors (see Figure 4.3).

The next investigated scenario, Case 2 was the multiple recycling of all actinides into the fast reactors without adding MAs from spent LWR fuel. The most important question in this case is whether an equilibrium can be reached and at what MA concentration. Figure 4.4 shows that the equilibrium is reached slightly above 1% MA concentration at end-of-cycle in each fast reactor core. The higher MA content even slightly improves breeding in the cores, which is already enough for break-even breeding, as in the equilibrium no Pu feed from spent LWR fuel is needed (see Figure 4.4). Isotopic compositions also reach an equilibrium which means that all TRU isotopes are consumed by fission and no Cm accumulation occurs due to the multirecycling of MAs (see Figure 4.5). These results prove that each investigated reactor can be applied as a TRU burner. A detailed analysis of the GFR equilibrium closed fuel cycle operation was also performed by Krépel et al. with the EQL3D procedure developed at the Paul Scherrer Institute (PSI) based on a similar GFR2400 design [24, 68]. Despite differences in the reactor design and in
4.2. Equilibrium closed fuel cycle studies

**Figure 4.2:** Pu feed from LWR spent fuel (left) and EOC MA content (right) of the fast reactor cores with U and Pu multirecycling (Case 1)

**Figure 4.3:** BOC Pu content of the fast reactor cores (left) and Pu composition of the GFR2400 core (right) with U and Pu multirecycling (Case 1)
the fuel cycle schemes, the main conclusions of these studies are confirmed by PSI results, namely that in the closed fuel cycle the GFR2400 acts as iso-breeder due to the combined effect of slight breeding and $^{241}$Pu decay during cooling time with approximately 1% MA content in the equilibrium. The detailed equilibrium compositions obtained for the GFR2400 with the FITXS method and EQL3D procedure were also compared with satisfactory results [P2].

The equilibrium MA concentration of 1% in the U, Pu and MA recycling case suggests that higher MA content in the fresh fuel also allows additional feed of MAs and the three fast reactors can be turned into net MA burners. In order to verify this statement simulations were performed with different fixed MA fractions in the fresh fuels varying from 0.5% to 3% (Case 3). As expected with 0.5% MA ratio the need for LWR MAs diminishes, since excess MAs are produced in the cores. In the case of 1.5% or higher MA content a significant MA feed from LWR spent fuel stabilizes (see for example Figure 4.6). The LWR feed needed for the 3% MA load can consume the minor actinides produced by LWRs of several times the fast reactors’ thermal power (see Tables 4.2-4.4). These results confirm that a symbiotic nuclear energy system can be set up where the LWRs produce the Pu required for the start-up of the fast reactors with the utilization of enriched uranium, while the fast reactors burn the MAs produced by the LWRs. The MA output of the whole system therefore reduces to partitioning losses. The increased MA content improves breeding properties due to fissile and fertile MA isotopes, which decreases the equilibrium Pu contents of the cores, as it can be seen in Figure 4.6. This effect was
4.2. Equilibrium closed fuel cycle studies

Figure 4.5: BOC Am (left) and Cm (right) composition of the GFR2400 core with U, Pu and MA multirecycling (Case 2)

also observed by Coquelet et al. [69] and Meyer et al. [70] in the SFR. Although the higher MA content provides better performance in terms of minor actinide burning and fuel utilization, difficulties concerning fuel fabrication and spent fuel management, as well as safety related parameters (such as the decreased delayed neutron fraction [P8]) may limit the MA content at a low fraction [71].

Table 4.2: Equilibrium fuel utilization and MA burning parameters of the GFR2400 with different recycling strategies

<table>
<thead>
<tr>
<th>Case</th>
<th>BOC Pu content (%)</th>
<th>Pu balance [kg/TWhe]</th>
<th>Breeding gain</th>
<th>MA balance [kg/TWhe]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U, Pu recyc.</td>
<td>17.35</td>
<td>-0.91</td>
<td>0.0335</td>
<td>6.51</td>
</tr>
<tr>
<td>U, Pu, MA recyc.</td>
<td>17.08</td>
<td>3.50</td>
<td>0.0562</td>
<td>0.00</td>
</tr>
<tr>
<td>0.5% MA content</td>
<td>17.27</td>
<td>1.16</td>
<td>0.0442</td>
<td>3.57</td>
</tr>
<tr>
<td>1.5% MA content</td>
<td>17.02</td>
<td>4.90</td>
<td>0.0597</td>
<td>-2.32</td>
</tr>
<tr>
<td>2% MA content</td>
<td>16.94</td>
<td>6.58</td>
<td>0.0669</td>
<td>-5.11</td>
</tr>
<tr>
<td>3% MA content</td>
<td>16.84</td>
<td>9.46</td>
<td>0.0804</td>
<td>-10.52</td>
</tr>
</tbody>
</table>
Table 4.3: Equilibrium fuel utilization and MA burning parameters of the ELSY with different recycling strategies

<table>
<thead>
<tr>
<th>Case</th>
<th>BOC Pu content (%)</th>
<th>Pu balance [kg/TWhe]</th>
<th>Breeding gain [kg/TWhe]</th>
<th>MA balance [kg/TWhe]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U, Pu recyc.</td>
<td>18.03</td>
<td>-1.48</td>
<td>0.0407</td>
<td>4.98</td>
</tr>
<tr>
<td>U, Pu, MA recyc.</td>
<td>17.95</td>
<td>0.54</td>
<td>0.0601</td>
<td>0.00</td>
</tr>
<tr>
<td>0.5% MA content</td>
<td>18.11</td>
<td>-0.73</td>
<td>0.0485</td>
<td>2.93</td>
</tr>
<tr>
<td>1.5% MA content</td>
<td>17.93</td>
<td>1.20</td>
<td>0.0625</td>
<td>-1.36</td>
</tr>
<tr>
<td>2% MA content</td>
<td>17.92</td>
<td>2.08</td>
<td>0.0662</td>
<td>-3.32</td>
</tr>
<tr>
<td>3% MA content</td>
<td>17.89</td>
<td>3.85</td>
<td>0.0769</td>
<td>-7.14</td>
</tr>
</tbody>
</table>

Table 4.4: Equilibrium fuel utilization and MA burning parameters of the ESFR with different recycling strategies

<table>
<thead>
<tr>
<th>Case</th>
<th>BOC Pu content (%)</th>
<th>Pu balance [kg/TWhe]</th>
<th>Breeding gain [kg/TWhe]</th>
<th>MA balance [kg/TWhe]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U, Pu recyc.</td>
<td>17.82</td>
<td>-2.77</td>
<td>0.0225</td>
<td>6.28</td>
</tr>
<tr>
<td>U, Pu, MA recyc.</td>
<td>17.63</td>
<td>1.85</td>
<td>0.0324</td>
<td>0.00</td>
</tr>
<tr>
<td>0.5% MA content</td>
<td>17.79</td>
<td>-1.06</td>
<td>0.0288</td>
<td>3.28</td>
</tr>
<tr>
<td>1.5% MA content</td>
<td>17.47</td>
<td>2.20</td>
<td>0.0407</td>
<td>-2.95</td>
</tr>
<tr>
<td>2% MA content</td>
<td>17.24</td>
<td>5.29</td>
<td>0.0495</td>
<td>-5.90</td>
</tr>
<tr>
<td>3% MA content</td>
<td>16.77</td>
<td>10.35</td>
<td>0.0656</td>
<td>-11.84</td>
</tr>
</tbody>
</table>

Figure 4.6: MA feed from LWR spent fuel (left) and BOC Pu content (right) of the GFR2400 core with different fixed MA contents of the fresh fuel (Case 3)
4.3 Transition scenario studies

A scenario describing the transition from a conventional LWR fleet to a mixed fleet of fast and MOX fueled thermal reactors was also simulated in order to demonstrate the applicability of the FITXS burn-up models in more complex fuel cycle schemes [P6, P5]. Different fast reactor and thermal reactor power ratios were investigated in terms of TRU management, in particular the stabilization and reduction of Pu and MA stocks. The specifications and results of the scenario studies are presented in the following subsections.

4.3.1 Scenario specifications

The burn-up models of the ELSY and the VVER-1200 MOX fuel assembly were used in the fuel cycle scheme depicted in Figure 4.7. The VVER-440 reactors were modeled using burn-up tables based on assembly-wise SCALE 6.0 burn-up calculations. The initial plutonium content of the ELSY was provided from the reprocessed VVER-440 spent fuel, and the MA content of the reprocessed VVER-440 fuel assemblies was also recycled in the LFR in order to reduce TRU inventories which were produced prior its commissioning. It was seen in the closed fuel cycle studies that minor actinide feed from spent LWR fuel improves the breeding capabilities of the otherwise iso-breeder fast reactors, which results in the accumulation of excess Pu. In order to stabilize the Pu inventory, VVER-1200 MOX fuel assemblies were assumed to be fabricated using the Pu from spent LFR fuel. It was assumed that the MOX fuel assemblies were loaded into a core mainly composed of UOX assemblies, but the UOX fuel assemblies were not modeled in this study. The total inventories therefore correspond to the actinide content of the VVER-440 spent fuel, the ELSY core, the VVER-1200 MOX fuel assemblies and the corresponding fuel cycle facilities and storages. Two years fabrication time was considered for each fuel type, with 5 years cooling time assumed for spent LWR fuels, 2 years cooling time for the spent ELSY fuel and 0.1% partitioning losses for all actinides. The aim of the study was to find power ratios of the fast reactors and the VVER-1200 MOX fuel assemblies which enable the consumption of MA from the spent VVER-440 fuel without the accumulation of excess Pu, and after that it results in a sustainable equilibrium state with stabilized Pu and MA inventories.

4.3.2 Transition scenario results

As a reference scenario I have investigated the case when the power ratio of the VVER-1200 MOX fuel assemblies was zero, i.e. when no Pu of the spent LFR fuel was recycled in thermal reactors. Results showed that 60 years operation of three
Figure 4.7: Fuel cycle model of the investigated transition scenario
VVER-440 reactors could provide enough Pu for the start-up of the LFR core (see Figure 4.8). Figs. 4.9 and 4.10 show that the LFR is capable of burning the MA content of the spent VVER-440 fuel, as the MA content of storages is almost reduced to zero (the MA inventory therefore corresponds to the MA content of the ELSY core in the equilibrium), and the MA fraction in the fresh fuel does not exceed the 3% limit for homogeneous MA recycling. As expected, the MA feed in the otherwise iso-breeder fast reactor increases the breeding capabilities of the core, which results in a significant increase in the total Pu inventory in the cycle, shown in Fig. 4.9. The simultaneous reduction of TRU inventories is therefore not possible in this case, and the prevention of Pu accumulation requires the operation of Pu burner reactors in the fuel cycle. In our study this was achieved by the utilization of excess Pu as MOX fuel in VVER-1200 fuel assemblies.

The next investigated case was aimed at the stabilization of the Pu inventory, which could be performed with a time-dependent power ratio of MOX fuel assemblies corresponding to the changing MA content of the LFR fresh fuel due to the continuous decrease of MA stocks. After all the MA stocks from spent VVER-440 fuel were burned in the LFR, an equilibrium state was reached with constant VVER-1200 MOX power ratio. The power histogram of the resulting scenario is depicted in Fig. 4.11. The total Pu and MA inventories in the fuel cycle (see Fig. 4.12) confirm that MA burning and the stabilization of the Pu inventory is indeed possible with the mixed fleet of fast and MOX fueled thermal reactors. Fig. 4.13 shows that since Pu from spent MOX fuel was recycled in the LFR before being used again in thermal reactors, the deterioration of its fissile quality was low enough that the
Figure 4.9: Pu inventory (left) and MA inventory (right) in the reference scenario

Figure 4.10: MA content of the ELSY fresh fuel in the reference scenario
Pu content of the fresh MOX fuel assemblies did not exceed 10% throughout the scenario including equilibrium state.

The last investigated scenario was simulated in order to show that higher MOX power ratio can even result in an overall TRU inventory reduction with a decrease in Pu inventory limited by the Pu content of the fast reactor core (see Fig. 4.15). On the other hand, Fig. 4.15 also shows a significant decrease in Pu stocks, which can reduce storage costs and proliferation risks. The equilibrium Pu and MA contents of the fresh LFR fuel were the same as in the previous case, which determined the breeding properties, therefore the same MOX power ratio was needed for the sustainable state with higher ratio in the intermediate state when MAs were burned from the spent VVER-440 fuel (see Fig. 4.14). It was therefore confirmed that a mixed fleet of fast reactors and MOX fueled thermal reactors can be used to set the Pu and MA burning capabilities of the whole system by changing the power ratio of fast and thermal reactors in the nuclear park.

The above presented transition scenario analysis was also performed for the GFR2400 and the ESFR. The results of the analyses are summarized in Table 4.5, which show that each of the three investigated Generation IV fast reactors are able to burn MA stocks of the VVER-440 fleet which produced the Pu needed for their start-up, and fresh fuel limits – such as 3% MA fraction – can be met throughout the scenarios. Differences in the results are due to different fuel cycle specifications of the reference cores (cycle lengths, actinide content, number of batches), for example the higher MOX power ratio needed for Pu inventory reduction in the case of the ELSY is due to the lower thermal power and Pu content of the core.
Figure 4.12: Pu inventory (left) and MA inventory (right) in the Pu inventory stabilization scenario

Figure 4.13: Pu content and Pu composition of the VVER-1200 fresh MOX fuel in the Pu inventory stabilization scenario
4.3. Transition scenario studies

**Figure 4.14:** Power histogram of the TRU inventory reduction scenario

**Figure 4.15:** Pu inventory (left) and MA inventory (right) in the TRU inventory reduction scenario
Table 4.5: Results of the VVER-440–FR–VVER-1200 MOX transition scenario studies

<table>
<thead>
<tr>
<th>Reactor</th>
<th>Case</th>
<th>MOX power fraction (%)</th>
<th>Inventory after 200 years (t)</th>
<th>Pu stabilization</th>
<th>TRU reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Total Pu</td>
<td>Total MA</td>
<td>Equilibrium Pu stabiliziation</td>
</tr>
<tr>
<td>GFR2400</td>
<td>Pu stock</td>
<td>1.9</td>
<td>3.7</td>
<td>11</td>
<td>18.9</td>
</tr>
<tr>
<td></td>
<td>Pu stabiliziation</td>
<td>4.2</td>
<td>6.4</td>
<td>12.0</td>
<td>18.9</td>
</tr>
<tr>
<td></td>
<td>TRU reduction</td>
<td>9.6</td>
<td>15.6</td>
<td>32.4</td>
<td>23.8</td>
</tr>
<tr>
<td>ELSY</td>
<td>Pu stock</td>
<td>1.9</td>
<td>3.7</td>
<td>11</td>
<td>18.9</td>
</tr>
<tr>
<td></td>
<td>Pu stabiliziation</td>
<td>4.2</td>
<td>6.4</td>
<td>12.0</td>
<td>18.9</td>
</tr>
<tr>
<td></td>
<td>TRU reduction</td>
<td>9.6</td>
<td>15.6</td>
<td>32.4</td>
<td>23.8</td>
</tr>
<tr>
<td>CFR2400</td>
<td>Pu stock</td>
<td>1.9</td>
<td>3.7</td>
<td>11</td>
<td>18.9</td>
</tr>
<tr>
<td></td>
<td>Pu stabiliziation</td>
<td>4.2</td>
<td>6.4</td>
<td>12.0</td>
<td>18.9</td>
</tr>
<tr>
<td></td>
<td>TRU reduction</td>
<td>9.6</td>
<td>15.6</td>
<td>32.4</td>
<td>23.8</td>
</tr>
</tbody>
</table>

Table 4.5: Results of the VVER-440–FR–VVER-1200 MOX transition scenario studies
Chapter 5

Markov chain models of nuclear transmutation

This chapter presents stochastic models of the nuclide transmutation chains based on discrete-time and continuous-time Markov chains, which were developed in order to establish a general mathematical framework for the calculation of quantities described in Section 2.4, as well as to provide details about the time evolution of individual transmutation chains and a profound understanding of underlying processes in minor actinide burning and fissile material breeding. The content of both this chapter and Chapter 6 are based on (Halász & Szieberth, 2018) [P7].

The developed models are consistent with the Bateman equations, but they describe the transmutation and decay chains of individual atoms as stochastic processes. The continuous-time Markov chain model of the nuclide chains also allows to identify the prevalent transmutation trajectories with the calculation of time-dependent trajectory probabilities. It is shown that transmutation trajectory probabilities in fact constitute the general solution of the Bateman equations. In addition, the Markov chain models of the actinide transmutation chains make it possible to obtain closed formulas for finite-time integrated and asymptotic fuel cycle performance parameters, which are detailed in Chapter 6.

In order to disambiguate the terminology which is used in these chapters, the applied definitions are listed in the following (see Figure 5.1):

- nuclide: a species of atoms or nuclei characterized by a specific atomic and mass number;
- atom or nucleus: a particular atom or nucleus of a given nuclide;
- transmutation network or decay scheme: the set of nuclides and possible transformations, which can be represented by a directed graph with nuclides as
vertices and nuclear reactions or radioactive decays as edges;

- transmutation or decay chain: the stochastic process describing the transformations of one or more atoms within a transmutation network or decay scheme;

- linear chain: a transmutation or decay chain within a linear (branchless and open) path inside the transmutation network or decay scheme, with possible recurring nuclides;

- transmutation trajectory: an ordered set of nuclides representing a series of transformations (nuclear reactions or radioactive decays) an atom can go through.

The developed mathematical models are presented in detail in the first section of this chapter, whereas the method to count labeled (or weighted) transitions in the transmutation chains and the calculation of transmutation trajectory probabilities are presented in the second and third sections, respectively.

\[ X_0 \quad X_1 \quad X_2 \quad X_3 \quad X_4 \]
\[ x_1 \]
\[ x_2 \rightarrow x_4 \]
\[ x_1 \rightarrow x_2 \rightarrow x_3 \]
\[ x_4 \rightarrow x_5 \rightarrow x_6 \]
\[ x_2 \rightarrow x_4 \rightarrow x_5 \rightarrow x_7 \]
\[ x_3 \rightarrow x_4 \rightarrow x_6 \rightarrow x_4 \rightarrow x_5 \]
\[ \vdots \]

Figure 5.1: Examples of (a) transmutation network, (b) linear paths in the transmutation network, (c) transmutation trajectories. Nuclides are denoted with \( x_i \) (\( i = 1, 2, \ldots, m \)), and the nuclide state of the initial atom after \( n \) transitions is denoted with \( X_n \) (\( n \in \mathbb{N}_0 = \{0, 1, 2, \ldots\} \)) [P7]
5.1 Mathematical models of the nuclide transmutation chains

In order to provide a general framework for methods described in Section 2.4, as well as to investigate the time evolution of individual transmutation chains (either in terms of finite irradiation or decay time, or number of nuclear transitions), detailed mathematical models of the nuclide chains are required. Markov chains are straightforward choices due to the following reasons:

- nuclear reactions and radioactive decays are stochastic by nature;
- the probabilities of the possible transitions depend only on present conditions.

Two approaches were used in order to describe different parameters related to the transmutation and breeding capabilities of the reactors: (1) a discrete-time Markov chain where time corresponds to the number of occurred transitions and (2) a continuous-time Markov chain, in which the timescales of the transitions were also taken into account. The models are presented in the following subsections along with the methodology to count labeled transitions in the transmutation chains, as well as the calculation of transmutation trajectory probabilities.

5.1.1 Discrete-time Markov chain model

An elementary model of the nuclide transmutation chains can be provided if we are not interested in the timescales of the process, and the evolution of the chains is only described with the number of occurred transitions (decays and nuclear reactions). Let us introduce the following notations:

- $S = \{x_i\}_{i=1,2,...,m}$ is the set of nuclides included in the transmutation chains;
- $X_n$ is a random variable with possible values in $S$ describing the nuclide state of the initial atom after $n$ transitions ($n \in \mathbb{N}_0 = \{0, 1, 2, \ldots \}$);
- $p_{ij}^{(n)}$ is the probability that an $x_i$ atom transforms into $x_j$ in $n$ transitions;
- $\omega = (x_{i_0}, x_{i_1}, x_{i_2}, \ldots)$ is one possible realization of the nuclide chains, or a sample path ($\omega \in S^{\mathbb{N}_0}$).

A transmutation chain, $X = \{X_n : n \in \mathbb{N}_0\}$ which starts from an $x_i$ atom corresponds to a discrete-time Markov chain (DTMC) on state space $S$ with an initial distribution concentrated in $x_i$ and transition probabilities $p_{ij}^{(n)}$. The transition probabilities can
be written in forms of stochastic matrices, in which all the elements are nonnegative and elements in one row sum up to 1:

\[
[P^{(n)}]_{ij} = p_{ij}^{(n)} = \mathbb{P}(X_n = x_j | X_0 = x_i).
\] (5.1)

In a quasi-static approximation, where the one-group cross-sections and neutron flux are considered constant, the \( P^{(n)} \) transition matrices can be expressed with the one-step transition matrix \( P^{(1)} \) [72]:

\[
P^{(n)} = (P^{(1)})^n = P^n, \quad P^{(0)} = I,
\] (5.2)

where we introduced \( P = P^{(1)} \), the transition probability matrix for one transition and I is the identity matrix. The elements of this matrix are determined by the relative reaction rates and decay rates, and can be expressed with the nuclear properties in the following form:

\[
[P]_{ij} = p_{ij} = \frac{\sigma_{ij} \Phi + \lambda_i f_{ij}}{\sigma_i^a \Phi + \lambda_i},
\] (5.3)

where \( \sigma_{ij} \) is the microscopic one-group cross-section of the \( x_i \rightarrow x_j \) reaction, \( \sigma_i^a \) is the absorption cross-section of nuclide \( x_i \), \( \Phi \) is the one-group neutron flux, \( \lambda_i \) is the decay constant of \( x_i \), and \( f_{ij} \) is the branching ratio of the \( x_i \rightarrow x_j \) decay. In this case \( \mathbb{X} \) is a homogeneous DTMC with initial distribution \( \rho(0) = \delta_{i} \) (where \( [\delta_{i}]_{j} = \delta_{ij} \), and \( \delta_{ij} \) is the Kronecker delta) and one-step transition probability matrix \( P \).

Note that the nucleus undergoes one of the possible decays or reactions in each transition, therefore \( p_{ii} = 0 \), except when no reactions or decays lead out from the nuclide \( x_i \). If only the actinide transmutation chains are studied then fission products can be considered collectively as an absorbing state \( x_f \) (thus \( S = A \cup \{x_f\} \), where \( A \) is the set of actinides), such that \( p_{fi} = 0 \), if \( i \neq f \) and \( p_{ff} = 1 \). A similar decomposition can be applied in the case of decay chains which end in a stable nuclide.

### 5.1.2 Continuous-time Markov chain model

In reality the different transitions do not occur in discrete time steps, but the lifetime of a nucleus in its current state is a random variable with exponential distribution [73] due to the memorylessness property of radioactive decay and nuclear reactions. The corresponding process is therefore a continuous-time Markov chain (CTMC). The following notations are applied in the continuous-time description of the nuclide transmutation chains:

- \( S = \{x_i\}_{i=1,2,\ldots,m} \) is the set of nuclides included in the transmutation chains;
- \( X_t \) is a random variable with possible values in \( S \) describing the nuclide state of the initial atom after \( t \) irradiation or decay time \((t \geq 0, t \in \mathbb{R})\);
• \( p_{ij}(t) \) is the probability that an \( x_i \) atom transforms into \( x_j \) under \( t \) irradiation or decay time;
• \((\omega, \tau) = ((x_{i_0}, \tau_0), (x_{i_1}, \tau_1), (x_{i_2}, \tau_2), ...), \) where \( \tau_k \) is the time spent in state \( x_{i_k}, \)
is one possible realization of the nuclide chains \((\omega, \tau) \in (S \times \mathbb{R})^{N_0} \).

The \( X_t(\omega, \tau) \) sample paths in the continuous-time model are therefore described with the visited \( \{x_{i_k}\} \) nuclide states, as well as the exponentially distributed \( \{\tau_k\} \) dwell times. We can define the transition probability matrix \( P(t) \), similar to the discrete-time case:

\[
[P(t)]_{ij} = p_{ij}(t) = \mathbb{P}(X_t = x_j | X_0 = x_i),
\]

which is in fact the transpose of the transition probability matrix defined by Raykin and Shlyakhter [46]. If the Markov chain is time-homogeneous, then the transition probabilities can be calculated from the transition rate matrix \( Q \), which is the infinitesimal generator of the stochastic semigroup of the \( P(t) \) transition probability matrices [74]:

\[
P(t) = e^{Qt}, \quad Q = P'(0) = \lim_{h \downarrow 0} \frac{P(h) - P(0)}{h} = \lim_{h \downarrow 0} \frac{P(h) - I}{h}.
\]

The elements of the \( Q \) transition rate matrix are

\[
[Q]_{ij} = \begin{cases} 
  q_{ij}, & \text{if } i \neq j; \\
  -q_i, & \text{if } i = j.
\end{cases}
\]

where the \( q_{ij} \) transition rates equal the transition probabilities over unit time. In the case of the nuclide transmutation chains, the transition rates correspond to the appropriate microscopic reaction rates and decay rates:

\[
q_{ij} = \sigma_{ij} \Phi + \lambda_i f_{ij},
\]

\[
q_i = \sum_{j \neq i} \sigma_{ij} \Phi + \lambda_i f_{ij} = \sigma_i^a \Phi + \lambda_i.
\]

With the above notations, a transmutation chain, \( X = \{X_t : t \geq 0\} \) which starts from an \( x_i \) atom is a homogeneous CTMC with an initial distribution concentrated in \( x_i \), transition probabilities \( P(t) \) and transition rate matrix \( Q \).

The discrete-time and continuous-time models are consistent due to the following fact. If the \( Q \)-matrix of a homogeneous CTMC is stable and conservative (the diagonal elements of \( Q \) are finite and \( \sum_{j=1}^{m} q_{ij} = 0, \ i = 1, \ldots, m \)), then the sample
paths of the chain are right-continuous step functions. In this case the so-called jump chain or embedded chain can be constructed, which is a discrete-time Markov chain describing the visited states with the same initial distribution and the following one-step transition probabilities [74]:

\[
[P]_{ij} = \begin{cases} 
\delta_{ij}, & \text{if } q_i = 0; \\
0, & \text{if } q_i > 0 \text{ and } i = j; \\
q_{ij}/q_i, & \text{if } q_i > 0 \text{ and } i \neq j.
\end{cases}
\] (5.9)

Substitution of the \(q_{ij}\) and \(q_i\) transition rates shows that the above defined one-step transition probability matrix is the same as (5.3), in fact the discrete-time model is the embedded chain itself. The time evolution of the nuclide chain can therefore be interpreted as the following. Given an initial atom \(X_0 = x_i\) the chain spends \(\tau_0\) time in the nuclide state \(x_i\), where \(\tau_0\) is an exponentially distributed random variable with parameter \(q_i\) and expected value \(1/q_i\). The next nuclide state (the daughter nuclide) is chosen according to the transition probabilities \(q_{ij}/q_i\), and the chain evolves in similar manner independent of its past history.

The standard nuclide chain differential equations can be constructed from the CTMC model in terms of expected values. The time evolution of a nuclide mixture \(N(t)\) can be expressed with the transition probability matrix, as well as the infinitesimal generator:

\[
N^\top(t) = N^\top(0)P(t) = N^\top(0)e^{Qt},
\] (5.10)

or equivalently

\[
N(t) = P^\top(t)N(0) = e^{Q^\top t}N(0),
\] (5.11)

as the expectation of the number of atoms transformed from \(x_i\) to \(x_j\) under time \(t\) is given by the product \(p_{ij}(t)N_i(0)\), and the sum of every possible transition in the fuel mixture weighted with the number of initial atoms yields formula (5.10) or (5.11). Derivation with respect to time results in the well-known Bateman equation system:

\[
\frac{dN}{dt} = Q^\top e^{Q^\top t}N(0) = Q^\top N(t),
\] (5.12)

where the transpose of the transition rate matrix, \(Q^\top\) is often referred to as the transmutation matrix. In the more commonly written form of Eq. (5.12), the time derivative of the quantity of isotope \(x_i\) can be expressed as

\[
\frac{dN_i}{dt} = \sum_{j \neq i} (\sigma_{ji} \Phi + f_{ji} \lambda_j)N_j(t) - (\sigma^a_i \Phi + \lambda_i)N_i(t).
\] (5.13)
The continuous-time Markov chain model of the nuclide transmutation chains is therefore consistent with the Bateman equations, but it describes individual transmutation chains as stochastic processes instead of the average time evolution of the fuel composition. Although in most cases we will deal with transmutation chains started from a specific nuclide, describing the latter is also possible by setting an "average" initial distribution for the chain, \( \rho_i(0) = P(X_0 = x_i) = N_i/N (i = 1, \ldots, m) \) which corresponds to the transmutation chain of an atom which is chosen randomly from the fuel with uniform distribution. The time-dependent probability distribution of \( X_t \) can be written in this case as the following:

\[
\rho^\top(t) = \rho^\top(0)P(t) = \rho^\top(0)e^{Qt}. \tag{5.14}
\]

Multiplying (5.14) with the total number of atoms \( N \) also results in Eq. (5.10) and (5.11). The above description with an average initial distribution will have significance in the question of time reversal in Section 6.1.5.

Note that there was no explicit spatial dependence denoted in the microscopic one-group cross-sections and neutron flux. However, as the fuel mixture is the ensemble of the individual nuclide chains, the transition rate matrix can be made space-dependent, \( Q = Q(r) \), with which space-dependent expected values can be calculated, and the Bateman equations can also be derived in space-dependent form. The question of how to treat temporal inhomogeneity is discussed in Section 5.2.3.

### 5.2 Counting labeled transitions

The static and dynamic D-factors described in the previous section measure the average time-integrated neutron balance of different nuclides. These quantities, as well as the average number of different reactions correspond to counting labeled (or weighted) transitions during the evolution of the nuclide chains. The presented Markov chain models allow the calculation of these quantities as functions of either the number of occurred transitions or the irradiation or decay time. As it will be shown in the following subsections, the memorylessness property (Markov property) of the transmutation chains makes it possible to derive closed formulas for the expected values which implicitly take into account every possible transmutation trajectory. A different method of counting labeled transitions (as well as their factorial moments) for CTMCs was developed by Minin and Suchard [75] for Markov models of evolution, which requires the reversibility of the chain, usually satisfied in evolutionary models, but generally not true for nuclide transmutation chains.
5.2.1 Counting labeled transitions in discrete time

Let us label certain \( x_i \rightarrow x_j \) transitions with \( \mu_{ij} \) weights, which we aim to sum up for a given overall number of transitions, every time an \( x_i \rightarrow x_j \) transition occurs. The sum, denoted with \( g(n) \) is also a random variable, which can be expressed after \( n \) transitions as

\[
g(n) = \sum_{k=1}^{n} \sum_{i=1}^{m} \sum_{j=1}^{m} \mu_{ij} I_{ij}(k),
\]

where \( I_{ij}(k) \) is an indicator variable with value equal to 1, if an \( x_i \rightarrow x_j \) transition occurs at step \( k \), and 0 otherwise. For example if \( \mu_{ij} = 1 \) for a given \( i \) and \( j \), and 0 for every other transitions, then \( g(n) \) is the total number of \( x_i \rightarrow x_j \) reactions or decays in \( n \) steps. Let \( g_i(n) \) denote the value of \( g(n) \) provided that the initial nuclide is \( x_i \):

\[
g_i(n) = (g(n) \mid X_0 = x_i).
\]

The expected value of \( g_i(n) \) can be calculated with the weighted sums regarding all the possible \( \omega \) trajectories starting from the \( x_i \) nuclide [76]:

\[
\hat{g}_i(n) = \mathbb{E}(g(n) \mid X_0 = x_i) = \sum_{\omega \in S^0} [g(n)](\omega) P(\omega \mid X_0 = x_i),
\]

where \([g(n)](\omega)\) is the value of \( g(n) \) which corresponds to trajectory \( \omega \). The evaluation of these expectations with an explicit calculation of trajectory probabilities is only possible for a limited number of transitions due to the large number of possible trajectories, including those with recurring nuclides e.g. due to consecutive \((n, \gamma)\) and \((n,2n)\) reactions. The law of total expectation (or tower rule) however makes it possible to obtain an equation system for the \( \hat{g}_i(n) \) expected values without the need for an explicit calculation of \( P(\omega)'s \). Let us expand the expected value with respect to the first transition, which is a complete set of events. The \( g_i(n) \) sum can be written in the following form:

\[
g_i(n) = \begin{cases} 
\mu_{i1} + g_1(n - 1), & \text{if } X_1 = x_1; \\
\mu_{i2} + g_2(n - 1), & \text{if } X_1 = x_2; \\
\vdots \\
\mu_{im} + g_m(n - 1), & \text{if } X_1 = x_m,
\end{cases}
\]

where we used the Markov property when we obtained \( g_i(n - 1) \), because \( g_i(n) \) depends only on the number of the remaining transitions. The tower rule gives us the following equation system for calculating the \( \hat{g}_i(n) \) expected values:

\[
\hat{g}_i(n) = \sum_{j \neq i} [\mu_{ij} + \hat{g}_j(n - 1)] p_{ij} = \sum_{j \neq i} \mu_{ij} p_{ij} + \sum_{j \neq i} \hat{g}_j(n - 1) p_{ij}.
\]
5.2. Counting labeled transitions

The right hand side of Equation (5.19) depends only on \((n - 1)\), therefore \(\hat{g}_i(n)\) can be expressed recursively, and also calculated in knowledge of \(\hat{g}_i(0)\)'s. Let us introduce the following vectors:

\[
\hat{g}(n) = \begin{bmatrix}
\hat{g}_1(n) \\
\hat{g}_2(n) \\
\vdots \\
\hat{g}_m(n)
\end{bmatrix}, \quad u = \begin{bmatrix}
\sum_{j \neq 1} \mu_{1j} p_{1j} \\
\sum_{j \neq 2} \mu_{2j} p_{2j} \\
\vdots \\
\sum_{j \neq m} \mu_{mj} p_{mj}
\end{bmatrix}.
\] (5.20)

Equation (5.19) can be rewritten using matrix notations (using that \(p_{ii} = 0\)):

\[
\hat{g}(n) = P\hat{g}(n - 1) + u,
\] (5.21)

with the initial condition

\[
\hat{g}(0) = 0.
\] (5.22)

Substituting \(\hat{g}(n - 1)\) consecutively in Eq. (5.21) a closed form can also be obtained for \(\hat{g}(n)\):

\[
\hat{g}(n) = P^n \hat{g}(0) + \sum_{k=0}^{n-1} P^k u.
\] (5.23)

If we are only interested in the actinide transmutation chains, and consider fission as an absorbing state, then \(n\rightarrow\infty\) corresponds to the case when the initial nucleus is ultimately fissioned with probability one. It is obvious that \(\hat{g}_f(n)\equiv0\) therefore we can introduce the \(P_A\) matrix by deleting the row and column corresponding to state \(f\), and for \(i \neq f\) the same solution will be given by the reduced equation system. Since every actinide has some non-zero fission cross-section, the elimination of fission in \(P_A\) implies that \(\sum_j [P_A]_{ij} < 1\) for every \(i\). The diagonal of \(P_A\) is composed of zeros, therefore it is also true that \(\sum_{j \neq i} [P_A]_{ij} < 1\). It follows from Gersgorin’s theorem [77], that

- the eigenvalues of \(P_A\) are consequently lower than 1;
- the \((I - P_A)\) matrix, where I denotes the identity matrix, is invertible.

The solution for \(\hat{g}(n)\) can thus be written in compact form (also reducing the two vectors for actinides only):

\[
\hat{g}(n) = (I - P_A)^{-1}(I - P_A^n) u,
\] (5.24)

and the \(P_A^n\) series converges to 0. Applying the \(n\rightarrow\infty\) limit on Eq. (5.24) yields

\[
\hat{g}(\infty) = \lim_{n\rightarrow\infty} \hat{g}(n) = (I - P_A)^{-1} u.
\] (5.25)
5.2.2 Counting labeled transitions in continuous time

Let us introduce the $\mu_{ij}$ weights again, which label certain reactions. The time is now a continuous parameter, therefore the sum - denoted here with $h(t)$ - has to be calculated by integrating over time from 0 to $t$:

$$h(t) = \int_0^t \sum_{i=1}^m \sum_{j=1}^m \mu_{ij} I_{ij}(s) ds .$$  \hspace{1cm} (5.26)

The value of the $I_{ij}(s)$ indicator variable is 1, if an $x_i \rightarrow x_j$ transition occurs in the time interval $(s, s + ds)$, and 0 otherwise. We can define $h_i(t)$ similarly as in the discrete case:

$$h_i(t) = (h(t) \mid X_0 = x_i) .$$  \hspace{1cm} (5.27)

The tower rule can be applied again for the calculation of the $\hat{h}_i(t)$ expected values, but in the continuous-time chain we have continuum possible sample paths, because the times of the transitions can change continuously. In general the expected value of $h_i(t)$ can be calculated with the following integral [76]:

$$\hat{h}_i(t) = \mathbb{E}(h(t) \mid X_0 = x_i) = \int_{(S \times \mathbb{R})^0} [h(t)](\omega, \tau) d\mathbb{P}((\omega, \tau) \mid X_0 = x_i) .$$  \hspace{1cm} (5.28)

Let us consider a complete set of events by the end state of the first transition, as well as the time of the transition. For a given $s$ time of transition the $h_i(t)$ sum can be expressed as

$$h_i(t) = \begin{cases} 
\mu_{i1} + h_1(t - s), & \text{if } X_s = x_1; \\
\mu_{i2} + h_2(t - s), & \text{if } X_s = x_2; \\
& \vdots \\
\mu_{im} + h_m(t - s), & \text{if } X_s = x_m . 
\end{cases}$$  \hspace{1cm} (5.29)

The probability of the first transition occurring in the time interval $(s, s + ds)$ is determined by the exponential distribution of the lifetime of the $x_i$ nucleus:

$$\mathbb{P}(\tau_0 \in (s, s + ds) \mid X_0 = x_i) = (\sigma_i^a \Phi + \lambda_i)e^{-(\sigma_i^a \Phi + \lambda_i)s}ds = q_i e^{-q_i s}ds ,$$  \hspace{1cm} (5.30)

while the probability of the transition resulting in nuclide $x_j$ is $q_{ij}/q_i$. Integrating over the possible sample paths, and explicitly accounting for the sum with respect to the first daughter nuclide and the integration over the $s$ time of the first transition, the tower rule yields the form

$$\hat{h}_i(t) = \int_0^t \sum_{j \neq i} q_{ij} e^{-q_i s} [\mu_{ij} + \hat{h}_j(t - s)] ds .$$  \hspace{1cm} (5.31)
The integral on the right hand side of the equation can be transformed using the substitution $s' = t - s$:

$$
\hat{h}_i(t) = - \int_t^0 \sum_{j \neq i} q_{ij} e^{-q_{j}(t-s')} [\mu_{ij} + \hat{h}_j(s')] ds' =
$$

$$
e^{-q_i t} \int_0^t \sum_{j \neq i} q_{ij} e^{q_j s'} [\mu_{ij} + \hat{h}_j(s')] ds' =
$$

$$
(5.32)
$$

Let us derive both sides with respect to $t$, which gives us the following differential equation system concerning $h_i(t)$, $i = 1, 2, \ldots, m$:

$$
\frac{d\hat{h}_i(t)}{dt} = - q_i e^{-q_i t} \int_0^t \sum_{j \neq i} q_{ij} e^{q_j s'} [\mu_{ij} + \hat{h}_j(s')] ds' +
$$

$$
+ e^{-q_i t} \sum_{j \neq i} q_{ij} e^{q_i t} [\mu_{ij} + \hat{h}_j(t)] =
$$

$$= - q_i \hat{h}_i(t) + \sum_{j \neq i} q_{ij} \hat{h}_j(t) + \sum_{j \neq i} q_{ij} \mu_{ij}.
$$

(5.33)

Introducing the following vectors:

$$
\hat{h}(t) = \begin{bmatrix}
\hat{h}_1(t) \\
\hat{h}_2(t) \\
\vdots \\
\hat{h}_m(t)
\end{bmatrix},
$$

$$
\nu = \begin{bmatrix}
\sum_{j \neq 1} \mu_{1j} q_{1j} \\
\sum_{j \neq 2} \mu_{2j} q_{2j} \\
\vdots \\
\sum_{j \neq m} \mu_{mj} q_{mj}
\end{bmatrix},
$$

(5.34)

the (5.33) differential equation system can be written in the matrix form

$$
\frac{d\hat{h}(t)}{dt} = Q\hat{h}(t) + \nu,
$$

(5.35)

with the initial condition

$$
\hat{h}(0) = 0.
$$

(5.36)

Eq. (5.35) is a first-order linear differential equation system, whose total solution can be obtained as the sum of the homogeneous solution and an inhomogeneous particular solution. Let us introduce the $Q_A$ matrix analogously to the discrete-time case, i.e. by removing the absorbing fission state from $Q$. In the modified transition rate matrix $\sum_{j \neq i} ||[Q_A]_{ij}| < ||[Q_A]_{ii}|$, therefore $Q_A$ is strictly diagonally dominant. Applying Gersgorin’s theorem again, it can be stated that the real part of the eigenvalues of $Q_A$ are negative, Re($\alpha_k$) < 0. Since $\alpha_k \neq 0$, therefore $Q_A$ is invertible and the solution of the reduced initial value problem can be given as

$$
\hat{h}(t) = (e^{Q_A t} - I)Q_A^{-1}\nu.
$$

(5.37)
It also follows from $\text{Re}(\alpha_k) < 0$ that the matrix exponential in (5.37) converges to zero as $t \to \infty$ and the solution for $\hat{h}(\infty)$ yields

$$\hat{h}(\infty) = -Q_A^{-1}v.$$ (5.38)

Comparing (5.38) with (5.25) after multiplying each equation in (5.25) with $\sigma_i^a \Phi + \lambda_i$ it can be seen that the continuous-time and discrete-time Markov chain models give the same result in the $n \to \infty$ and $t \to \infty$ limit (which was expected as the discrete-time model is the embedded chain of the continuous-time model):

$$\hat{g}(\infty) = \hat{h}(\infty).$$ (5.39)

### 5.2.3 Time intervals with different transition rates

The calculations of the previous subsections are based on the assumption that the microscopic one-group cross-sections and neutron flux are constant and therefore the Markov-chains are time-homogeneous. On the other hand, the formulas derived for the continuous-time model are valid for arbitrarily short time intervals, therefore it is possible to count labeled transitions throughout multiple time steps with different cross-section sets and neutron flux, as well as subsequent irradiation and cooling periods, which is the case for example in a closed fuel cycle. Let us denote the $Q$-matrix of the $k$th time step restricted to actinides with $Q_A^{(k)}$, the length of the time step with $t_k$, and the sum of the lengths of the first $k$ time steps with $s_k$ (where $s_0 = 0$). Until the beginning of the $k$th time step, the initial atom of $x_i$ transforms into another $x_j$ atom with probability $p_{ij}(s_k-1)$, therefore the expected value of the contribution of the $k$th time step to the sum $\hat{h}(s_k)$ can be expressed as the product of the $\hat{h}_j^{(k)}(t_k)$ expected values and the transition probabilities:

$$\hat{h}(s_k) = \hat{h}(s_k-1) + P_A(s_k-1)\hat{h}_j^{(k)}(t_k),$$ (5.40)

where the $\hat{h}_j^{(k)}(t_k)$ expected values are calculated with formula (5.37):

$$\hat{h}_j^{(k)}(t_k) = (e^{Q_A^{(k)}t} - 1)[Q_A^{(k)}]^{-1}v^{(k)},$$ (5.41)

and the transition probabilities are determined by the $Q$-matrices:

$$P_A(s_k-1) = \prod_{l=1}^{k-1} e^{Q_A^{(k)}t_k}.$$ (5.42)

From Eq. (5.40) and (5.42) the overall sum of the contributions of $n$ time steps, $\hat{h}(s_n)$ can be expressed with the following formula:

$$\hat{h}(s_n) = \sum_{k=1}^{n} \left( \prod_{l=1}^{k-1} e^{Q_A^{(k)}t_k} \right) \hat{h}_j^{(k)}(t_k),$$ (5.43)
where $\hat{h}^{(k)}(t_k)$ is given by (5.41). The above formula can be used to account for the overall history of the nuclide chain (in the same reactor or different reactors), including burn-up with time steps of different cross-section sets, as well as decay during refueling and in interim storage.

5.3 Transmutation trajectory probabilities

The probabilistic interpretation of the Bateman equations and their solution was discussed in detail in the works of Raykin and Shlyakhter [46] and also indicated by Cetnar [30]. It will be shown in the following subsections that the (2.19) and (2.21) solutions are in fact equal to the transmutation trajectory probabilities, which can be derived from the continuous-time Markov chain model of the nuclide transmutation chains. The derivation also reveals which step of the solution of Bateman [45] cannot be applied in the case of recurring nuclides or equal microscopic destruction rates.

5.3.1 Trajectory probabilities in discrete time

Although counting labeled transitions with the evaluation of every transmutation trajectory is only possible for a limited number of transitions, the calculation of the trajectory probabilities itself is simple in the discrete-time model. Let us consider a transmutation trajectory composed of $n$ transitions, $\gamma$:

$$\gamma = (x_{i_0}, x_{i_1}, x_{i_2}, \ldots, x_{i_n}). \quad (5.44)$$

In each transition the probability of the nuclide in state $x_{i_k}$ transforming into nuclide $x_{i_{k+1}}$ is $p_{i_ki_{k+1}}$, and the trajectory probability $p_\gamma$ can be calculated as the sum of sample path probabilities for paths which begin with $\gamma$:

$$p_\gamma = \mathbb{P}\{\omega : \omega_k = x_{i_k}, k = 0, \ldots, n \mid X_0 = x_{i_0}\} = \prod_{k=0}^{n-1} p_{i_ki_{k+1}} =$$

$$= \prod_{k=0}^{n-1} \frac{\sigma_{x_{i_k+1}} \Phi + \lambda_{i_k} f_{i_{k+1}}}{\sigma_{i_k} \Phi + \lambda_{i_k}}. \quad (5.45)$$

The above result is only valid in the discrete-time Markov chain model, as in continuous time nuclear reactions and radioactive decays introduce specific time delays which also influence the trajectory probabilities. It will be shown, however, that if the trajectory ends with fission in the case of the actinide chains (or a stable nuclide in the case of decay chains), then the probability in the discrete-time model equals the probability in the continuous-time model in the $t \to \infty$ limit.
5.3.2 Trajectory probabilities in continuous time

A sample path $X_i(\omega, \tau)$ in the CTMC model can be described with the $\{\omega_k\}$ visited states and the $\{\tau_k\}$ corresponding dwell times. In general the probability of the transmutation trajectory $\gamma$ at time $t$ can be written as the sum of sample path probabilities for paths which visit the $(x_{i_0}, x_{i_1}, x_{i_2}, \ldots, x_{i_n})$ states until time $t$ with no additional transitions. Let us define the sum of the first $n$ dwell times, $T_n$:

$$T_n = \tau_0 + \tau_1 + \tau_2 + \cdots + \tau_{n-1}. \quad (5.46)$$

The probability of the transmutation trajectory, $P_\gamma(t)$ can therefore be expressed with the sample path probabilities in the following form:

$$P_\gamma(t) = \mathbb{P} \left( \{ (\omega, \tau) : \omega_k = x_{ik}, k = 1, \ldots, n, T_n \leq t, T_{n+1} > t \} | X_0 = x_{i_0} \right). \quad (5.47)$$

The above probability can be written as the product of the appropriate one-step transition probabilities and the probability that the $(x_{i_0}, x_{i_1}, x_{i_2}, \ldots, x_{i_n})$ states and only those are visited until time $t$:

$$P_\gamma(t) = \frac{q_{i_0i_1}}{q_{i_0}} \frac{q_{i_1i_2}}{q_{i_1}} \cdots \frac{q_{i_{n-1}i_n}}{q_{i_{n-1}}} \mathbb{P} \left( \{ T_n \leq t, T_{n+1} > t \} | \{ \omega_k = x_{ik}, k = 0, \ldots, n \} \right). \quad (5.48)$$

It is apparent that $T_{n+1} \leq t$ implies $T_n \leq t$, therefore the expression on the right hand side can be transformed into simpler form using the inclusion/exclusion principle (for simplicity the condition $\{ \omega_k = x_{ik}, k = 0, \ldots, n \}$ is not denoted from here on):

$$\mathbb{P}(\{ T_n \leq t, T_{n+1} > t \}) = \mathbb{P}(\{ T_n \leq t \}) + \mathbb{P}(\{ T_{n+1} > t \}) -$$

$$- \mathbb{P}(\{ T_n \leq t \} \cup \{ T_{n+1} > t \}) = \mathbb{P}(\{ T_n \leq t \}) + 1 - \mathbb{P}(\{ T_{n+1} \leq t \}) - 1 = \mathbb{P}(\{ T_n < t \}) - \mathbb{P}(\{ T_{n+1} < t \}) = F_{T_n}(t) - F_{T_{n+1}}(t), \quad (5.49)$$

where $F_{T_n}$ and $F_{T_{n+1}}$ are the cumulative distribution functions of $T_n$ and $T_{n+1}$, respectively, and it was used that $\{ T_n = t \}$ and $\{ T_{n+1} = t \}$ are events of zero probability. The dwell times are independent random variables with exponential distribution, therefore it is most practical to calculate the distribution of their sums using their characteristic functions. The characteristic function of the $k$th dwell time, $\psi_k$ is the following [78]:

$$\psi_k(s) = \frac{1}{1 - \frac{is}{q_{ik}}} = \frac{-iq_{ik}}{-iq_{ik} - s}. \quad (5.50)$$

Since the $\tau_k$ dwell times are independent, the characteristic functions of the sums equal the product of the individual $\psi_k$ characteristic functions [78]:

$$\Psi_{T_n}(s) = \prod_{k=0}^{n-1} \frac{-iq_{ik}}{-iq_{ik} - s}, \quad (5.51)$$
5.3. Transmutation trajectory probabilities

\[ \Psi_{T_{n+1}}(s) = \Psi_{T_n}(s) \cdot \frac{-i q_n}{-i q_n - s} = \prod_{k=0}^{n-1} \frac{-i q_{i_k}}{-i q_{i_k} - s} \cdot \frac{-i q_n}{-i q_n - s}. \quad (5.52) \]

In general a nuclide state can be visited multiple times, which means that \(-i q_{i_k}\) may not be a simple pole of the characteristic function, but a pole of order \(r_{i_k}\), where \(r_{i_k}\) is the multiplicity of the nuclide state \(x_{i_k}\) in the transmutation trajectory (in this case the Laplace transform of the Bateman equations cannot be decomposed into partial fractions of first order polynomials only as in the solution of Bateman [45]). The product can be rewritten such that the \(k\) index runs over the nuclide states which are present in the transmutation path. Let \(I'\) and \(I\) denote the set of unique state indices in the trajectory not including and including the final nuclide. The characteristic functions of \(T_n\) and \(T_{n+1}\) can therefore be written as

\[ \Psi_{T_n}(s) = \prod_{k \in I'} \left( \frac{-i q_k}{-i q_k - s} \right)^{r_k}, \quad (5.53) \]

\[ \Psi_{T_{n+1}}(s) = \prod_{k \in I} \left( \frac{-i q_k}{-i q_k - s} \right)^{r_k}. \quad (5.54) \]

The cumulative distribution functions, \(F_{T_n}\) and \(F_{T_{n+1}}\) can be calculated with the inversion formula:

\[ F_{T_n}(t) = \frac{1}{2\pi} \int_{0}^{t} \int_{-\infty}^{\infty} e^{-ist} \Psi_{T_n}(s) ds \, d\tau = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{e^{-ist} - 1}{-i s} \Psi_{T_n}(s) ds, \quad (5.55) \]

\[ F_{T_{n+1}}(t) = \frac{1}{2\pi} \int_{0}^{t} \int_{-\infty}^{\infty} e^{-ist} \Psi_{T_{n+1}}(s) ds \, d\tau = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{e^{-ist} - 1}{-i s} \Psi_{T_{n+1}}(s) ds. \quad (5.56) \]

Substituting the previous expressions into (5.49), the probability \(P(\{T_n \leq t, T_{n+1} > t\})\) can be written as

\[ P(\{T_n \leq t, T_{n+1} > t\}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{e^{-ist} - 1}{-i s} \left( \Psi_{T_n}(s) - \Psi_{T_{n+1}}(s) \right) ds = \]

\[ = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{e^{-ist} - 1}{-i s} \Psi_{T_n}(s) \left( 1 - \frac{-i q_n}{-i q_n - s} \right) ds. \quad (5.57) \]

After performing the substraction on the right hand side of the integral, the probability can be finally expressed with the following integrals:

\[ P(\{T_n \leq t, T_{n+1} > t\}) = \frac{1}{q_n} \frac{1}{2\pi} \int_{-\infty}^{\infty} (e^{-ist} - 1) \Psi_{T_{n+1}}(s) ds = \]

\[ = \frac{1}{q_n} \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-ist} \Psi_{T_{n+1}}(s) ds - \frac{1}{q_n} \frac{1}{2\pi} \int_{-\infty}^{\infty} \Psi_{T_{n+1}}(s) ds. \quad (5.58) \]
The integrals can be evaluated by applying the residue theorem [79]. Let us first consider the second integral, and extend the integral to a closed curve consisting of the real axis and a semicircle with a radius which tends to infinity. As $\Psi_{T_{n+1}}(s)$ is a rational function, whose denominator is an at least two orders higher polynomial function of $s$ than the numerator, the integral on the semicircle vanishes in both the upper and lower complex half-planes. Since $\Psi_{T_{n+1}}(s)$ has poles only in the lower complex half-plane, it is straightforward to choose the upper half-plane for the integration:

$$\lim_{R \to \infty} \left( \int_{-R}^{R} \Psi_{T_{n+1}}(s) \, ds + \int_{0}^{\pi} \Psi_{T_{n+1}}(s(R, \varphi)) \, Rd\varphi \right) = \int_{-\infty}^{\infty} \Psi_{T_{n+1}}(s) \, ds .$$  

(5.59)

The l.h.s. of the above expression is an integral of a function which has no poles inside the closed curve, therefore the improper integral on the real axis is also zero:

$$\int_{-\infty}^{\infty} \Psi_{T_{n+1}}(s) \, ds = 0 .$$  

(5.60)

In the case of the first integral containing the $e^{-ist}$ factor, the semicircle has to be in the lower complex half-plane, because $e^{-ist}$ diverges as $\text{Im}(s) \to +\infty$ and vanishes as $\text{Im}(s) \to -\infty$. As the exponential does not introduce any more singular points, the poles of $e^{-ist}\Psi_{T_{n+1}}(s)$ lie on the lower half of the imaginary axis, $\{-iq_k : k \in I\}$. The improper integral on the real axis therefore equals $-2\pi i$ times the sum of the residues in these poles (due to clockwise integration):

$$\int_{-\infty}^{\infty} e^{-ist}\Psi_{T_{n+1}}(s) \, ds = -2\pi i \sum_{k \in I} \text{Res}(e^{-ist}\Psi_{T_{n+1}}(s), -iq_k) .$$  

(5.61)

For poles of order $r_k$ the residue can be calculated with the following formula [79]:

$$\text{Res}(e^{-ist}\Psi_{T_{n+1}}(s), -iq_k) = \frac{1}{(r_k - 1)!} \lim_{s \to -iq_k} \frac{d^{r_k-1}}{ds^{r_k-1}}(s + iq_k)^{r_k} e^{-ist}\Psi_{T_{n+1}}(s) .$$  

(5.62)

Substituting back the characteristic function of $T_{n+1}$, and applying the general Leibniz-rule (product rule), the residue can be written as

$$\frac{(-iq_k)^{r_k}}{(r_k - 1)!} \lim_{s \to -iq_k} \sum_{p=0}^{r_k-1} \binom{r_k - 1}{p} (-it)^p e^{-ist} \frac{d^{r_k-1-p}}{ds^{r_k-1-p}} \prod_{l \neq k} \frac{(-iq_l)^{r_l}}{(-iq_l - s)^{r_l}} .$$  

(5.63)

With the use of the Leibniz-rule for products of multiple factors, the derivative on the right hand side can be expressed as

$$\sum_{h_1 + \ldots + h_{k-1} + \ldots + h_m = r_k - 1 - p} \binom{r_k - 1 - p}{h_1, \ldots, h_{k-1}, h_{k+1}, \ldots, h_m} \prod_{l \notin I} \frac{(r_l - 1 + h_l)!}{(r_l - 1)!} \frac{(-iq_l)^{r_l}}{(-iq_l - s)^{r_l + h_l}} ,$$  

(5.64)
5.3. Transmutation trajectory probabilities

where \( m_\gamma \) is the number of unique nuclides in the transmutation trajectory. The multinomial coefficients which multiply the products can be simplified by using the relations

\[
\begin{pmatrix} r_k - 1 - p \\ h_1, \ldots, h_{k-1}, h_{k+1}, \ldots, h_m \end{pmatrix} = \frac{(r_k - 1 - p)!}{h_1! \cdots h_{k-1}! h_{k+1}! \cdots h_m!} \tag{5.65}
\]

and

\[
\frac{(r_l - 1 + h_l)!}{(r_l - 1)!} = \binom{r_l - 1 + h_l}{h_l} h_l! \tag{5.66}
\]

therefore the derivative of the product can be written in the simpler form:

\[
(r_k - 1 - p)! \left[ \prod_{l \in I} \frac{(-iq_l)^{r_l}}{(-iq_l - s)^{r_l}} \right] \sum_{h_1 + \cdots + h_{k-1} + \cdots + h_m = r_k - 1 - p} \left[ \prod_{l \in I} \frac{(r_l - 1 + h_l)}{h_l} \right] \frac{1}{(-iq_l - s)^{h_l}} \tag{5.67}
\]

By substituting back this expression into Eq. (5.62), and simplifying the residues with \((r_k - 1)!\) and \((r_k - 1 - p)!\) and constant expressions, the probability \( \mathbb{P}(\{T_n \leq t, T_{n+1} > t\}) \) can be finally expressed as

\[
\mathbb{P}(\{T_n \leq t, T_{n+1} > t\}) = \frac{1}{q_n} \sum_{k \in I} q_k \alpha_k e^{-q_n t} \sum_{p=0}^{r_k - 1} \frac{(q_k t)^p}{p!} \Omega_{k,r_k-1-p}, \tag{5.68}
\]

where

\[
\alpha_k = \prod_{l \in I, l \neq k} \left( \frac{q_l}{q_l - q_k} \right)^{r_l}, \tag{5.69}
\]

and

\[
\Omega_{k,j} = \sum_{h_1=0}^{j} \cdots \sum_{h_{k-1}=0}^{j} \sum_{h_{k+1}=0}^{j} \cdots \sum_{h_m=0}^{j} \prod_{l \in I, l \neq k} \left( \frac{r_l - 1 + h_l}{h_l} \right) \left( \frac{q_k}{q_k - q_l} \right)^{h_l} \delta(j, \sum_{l \in I} h_l). \tag{5.70}
\]

The total path probability is the product of the appropriate one-step transition probabilities and the probability that at time \( t \) the nucleus is in the state \( x_{i_n} \):

\[
P_\gamma(t) = \frac{q_{i_{01}} q_{i_{12}} \cdots q_{i_{n-1,n}}}{q_{i_0} q_{i_1} \cdots q_{i_{n-1}}} \cdot \frac{1}{q_{i_n}} \sum_{k \in I} q_k \alpha_k e^{-q_n t} \sum_{p=0}^{r_k - 1} \frac{(q_k t)^p}{p!} \Omega_{k,r_k-1-p}. \tag{5.71}
\]

which is equivalent to the solution (2.21) of Cetnar when \( N_1(0) = 1 \). For trajectories without recurring nuclides or equal microscopic destruction rates, the (5.71) transmutation trajectory probability simplifies to

\[
P_\gamma(t) = \frac{q_{i_{01}} q_{i_{12}} \cdots q_{i_{n-1,n}}}{q_{i_0} q_{i_1} \cdots q_{i_{n-1}}} \cdot \frac{1}{q_{i_n}} \sum_{k \in I} q_k \alpha_k e^{-q_n t}. \tag{5.72}
\]
The exponential terms decrease faster than the powers of \( t \) increase, therefore in the \( t \to \infty \) limit the probability of trajectories which do not end in the absorbing fission state converges to zero:

\[
\lim_{t \to \infty} P_r(t) = 0, \quad \text{if} \ x_i \neq x_f .
\]  
(5.73)

For trajectories that end with fission in the case of the actinide transmutation chains or a stable nuclide in the case of decay chains, \( \gamma_f, q_i = 0 \) and the time-dependent factor simplifies to \( \mathbb{P}(\{T_n \leq t\}) \), therefore the formula has to be modified as

\[
\mathbb{P}(\{T_n \leq t\}) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{e^{-ist} - 1}{-is} \Psi_{T_n}(s) ds .
\]  
(5.74)

The residue theorem can be applied in this case as well, but the integrand has a removable singularity on the real axis, which has to be eliminated first. Let us define the following \( \Phi(s) \) function in order to remove the singularity:

\[
\Phi(s) = \begin{cases} 
\frac{e^{-ist} - 1}{-s} \Psi_{T_n}(s), & \text{if } s \neq 0; \\
0, & \text{if } s = 0 ,
\end{cases}
\]  
(5.75)

as both the left and right limits of the integrand are zero in \( s = 0 \). The probability \( \mathbb{P}(\{T_n \leq t\}) \) can be rewritten using the \( \Phi(s) \) function:

\[
\mathbb{P}(\{T_n \leq t\}) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \Phi(s) ds = -\sum_{k \in I'} \text{Res}(\Phi(s), -iq_k) .
\]  
(5.76)

The integrand has poles of order \( r_k \) in \( \{-iq_k : k \in I'\} \), in which the residues can be expressed in the following form:

\[
\text{Res}(\Phi(s), -iq_k) = \text{Res}\left(\frac{e^{-ist} - 1}{-s} \Psi_{T_n}(s), -iq_k\right) - \text{Res}\left(\frac{1}{-s} \Psi_{T_n}(s), -iq_k\right) = \\
= \frac{1}{(r_k - 1)!} \lim_{s \to -iq_k} \frac{d^{r_k - 1}}{ds^{r_k - 1}} (s + i q_k)^{r_k} e^{-ist} \frac{1}{-s} \Psi_{T_n}(s) + \text{Res}\left(\frac{1}{-s} \Psi_{T_n}(s), -iq_k\right) .
\]  
(5.77)

It can be shown that the sum of the residues of \( (1/s) \Psi_{T_n}(s) \) in \( \{-iq_k : k \in I'\} \) equals \(-1\). Since the function \( (1/s) \Psi_{T_n}(s) \) vanishes in the \( s \to \infty \) limit, therefore its integral on a circle with radius \( R \) which tends to infinity is zero. The function has poles of order \( r_k \) in \( \{-iq_k : k \in I'\} \) and a simple pole in \( s = 0 \) with the following residue

\[
\text{Res}\left(\frac{1}{-s} \Psi_{T_n}(s), 0\right) = \lim_{s \to 0} s \frac{1}{-s} \Psi_{T_n}(s) = \lim_{s \to 0} \Psi_{T_n}(s) = 1 .
\]  
(5.78)
As the integral on the circle with radius \( R \to \infty \) is zero, therefore the sum of all residues has to be zero, from which
\[
\sum_{k \in I'} \text{Res} \left( \frac{1}{s} \Psi_{T_{n}}(s), -iq_{k} \right) = -\text{Res} \left( \frac{1}{s} \Psi_{T_{n}}(s), 0 \right) = -1. \tag{5.79}
\]
Evaluation of the remaining residues therefore leads to the following result concerning \( \mathbb{P}(\{T_{n} \leq t\}) \):
\[
\mathbb{P}(\{T_{n} \leq t\}) = 1 - \sum_{k \in I'} q_{k} \alpha_{k}' e^{-q_{k}t} \sum_{p=0}^{r_{k}-1} \frac{(q_{k}t)^{p}}{p!} \Omega_{k,r_{k}-1-p}, \tag{5.80}
\]
where the modified \( \alpha_{k}' \) factors can be expressed as:
\[
\alpha_{k}' = \frac{1}{q_{k}} \prod_{l \in I'} \left( \frac{q_{l}}{q_{l} - q_{k}} \right)^{r_{l}}. \tag{5.81}
\]
Finally, the trajectory probability can be written in the following form when the trajectory ends with fission or with a stable nuclide in the case of decay chains:
\[
P_{ij}(t) = \frac{q_{0i_{1}} q_{i_{1}i_{2}} \ldots q_{i_{n-1}i_{n}}}{q_{0i}} \cdot \left( 1 - \sum_{k \in I'} q_{k} \alpha_{k}' e^{-q_{k}t} \sum_{p=0}^{r_{k}-1} \frac{(q_{k}t)^{p}}{p!} \Omega_{k,r_{k}-1-p} \right). \tag{5.82}
\]
In the case when there are no recurring nuclides or equal microscopic destruction rates, the (5.82) fission trajectory probability simplifies to the following:
\[
P_{\gamma j}(t) = \frac{q_{0i_{1}} q_{i_{1}i_{2}} \ldots q_{i_{n-1}i_{n}}}{q_{0i}} \cdot \left( 1 - \sum_{k \in I'} q_{k} \alpha_{k}' e^{-q_{k}t} \right). \tag{5.83}
\]
With the application of these results, the most prevalent transmutation trajectories, including those which lead to fission can be identified for finite irradiation time (e.g. one burn-up cycle) or asymptotically for infinite recycling of the initial atoms. If the asymptotic probabilities of different fission trajectories are investigated, then only the one-step transition probabilities determine the result, as the time delays introduced by absorption and decays have no effect in the \( t \to \infty \) limit:
\[
\lim_{t \to \infty} P_{\gamma j}(t) = \lim_{t \to \infty} \frac{q_{0i_{1}} q_{i_{1}i_{2}} \ldots q_{i_{n-1}i_{n}}}{q_{0i}} \cdot \mathbb{P}(\{T_{n} \leq t\}) = \prod_{k=0}^{n-1} \frac{q_{ki_{k+1}}}{q_{ik}} = \prod_{k=0}^{n-1} p_{ki_{k+1}} = p_{\gamma_{f}}. \tag{5.84}
\]
If the fission trajectory probabilities are divided with the total fission probability, then it is in addition possible to analyze which trajectories are responsible for the build-up of fission probabilities. In general the time-dependent transition probability \( p_{ij}(t) = [P(t)]_{ij} \) can be expressed as the infinite sum of transmutation trajectory probabilities from \( x_{i} \) to \( x_{j} \):
\[
p_{ij}(t) = [P(t)]_{ij} = \sum_{\gamma_{i \to j}} P_{\gamma_{i \to j}}(t), \tag{5.85}
\]
where \( \gamma_{i \to j} \)'s are transmutation trajectories which start in \( x_{i} \) and end in \( x_{j} \).
Chapter 6

Description of the actinide transmutation chains

The comparison of the transmutation and breeding capabilities of different reactor designs and fuel cycle schemes requires specific fuel cycle performance parameters such as those presented in Section 2.4, which allow the characterization of fissile material production and minor actinide burning potential. In the analysis of actinide transmutation – whose purpose is the conversion of actinide isotopes to fission products –, fission products can be considered collectively as an absorbing fission state, in which case the discrete-time and continuous-time Markov chain models of the nuclide transmutation chains can be used to obtain closed formulas for the calculation of both finite-time-integrated and asymptotic fuel cycle performance parameters. These include static and dynamic D-factors, average neutron productions and time-dependent fission probabilities, which can be calculated by counting labeled transitions in the actinide transmutation chains with specific weights.

The fact that the developed Markov chain models describe the transmutation chains of individual atoms as stochastic processes can contribute to a more detailed understanding of underlying processes in fissile material breeding and minor actinide burning, including transmutation trajectory-wise contributions to the breeding gain and the neutron balance. The stochastic description of the nuclide chains also allows the calculation of the average time until fission and the time-dependent distribution of fissioned daughter nuclides. The time behavior of the actinide transmutation chains, or fission dynamics is described in the first section of this chapter, whereas parameters related to the neutron economy and breeding capabilities are described in the second and third sections, respectively.
6.1 Fission dynamics

The stochastic description of the nuclide chains allows insight into the behavior of individual transmutation chains, and makes it possible to identify the prevalent processes and typical timescales of the transmutation of specific actinides. Average quantities such as fission probabilities and the average time until fission are derived in the following subsections along with the number of different reactions and decays until fission and the distribution of fissioned daughter nuclides.

6.1.1 Fission probabilities

The continuous-time Markov chain model of the nuclide transmutation chains makes it possible to describe the probability of one initial atom of a given nuclide or its daughters to be fissioned with respect to irradiation time, \( p_{i\text{f}}(t) \), which equals the fraction of the initial atoms of nuclide \( x_i \) that are converted to fission products. Note that this fraction does not equal the relative decrease in \( N_i \) as in the fuel mixture \( x_i \) atoms can also be transformed into other nuclides, and atoms of other nuclides can transform into \( x_i \) atoms as well. On the other hand, the time derivative of the fission probabilities describes the transmutation rates of the different actinide isotopes in a given neutron field. The time-dependent fission probabilities can on the one hand be expressed with the transition probability matrix:

\[
p_{i\text{f}}(t) = [P(t)]_{i\text{f}} = [e^{Qt}]_{i\text{f}}.
\] (6.1)

A different method for calculating the fission probabilities is to count the number of fissions until time \( t \) in the nuclide transmutation chains of nuclide \( x_i \) according to Section 5.2.2, denoted with \( h_{i\text{f}}(t) \). Let us label the fission of every nuclide with \( \mu_{i\text{f}} = 1 \), and other nuclear reactions, as well as radioactive decays with \( \mu_{ij} = 0 \), \( j \neq f \). With this weighting scheme, every transmutation trajectory which ends in fission evaluates as 1, and all others with 0, therefore \( h_{i\text{f}}(t) \) is an indicator variable, whose expectation is equal to the probability of fission. According to Eq. (5.37), the fission probabilities can therefore be calculated with the following formula:

\[
p_{i\text{f}}(t) = \hat{h}_{i\text{f}}(t) = (e^{Qt} - I)Q^{-1}v_f,
\] (6.2)

where the \( v_f \) vector is composed of the microscopic fission reaction rates:

\[
v_f = \begin{bmatrix} q_{1\text{f}} \\ q_{2\text{f}} \\ \vdots \\ q_{m\text{f}} \end{bmatrix} = \begin{bmatrix} \sigma_{1\text{f}} \Phi \\ \sigma_{2\text{f}} \Phi \\ \vdots \\ \sigma_{m\text{f}} \Phi \end{bmatrix}.
\] (6.3)
The advantage of using Eq. (6.2) instead of Eq. (6.1) is that it can be modified to calculate the isotopic distribution of fissioned daughter products (see Section 6.1.4). From Eq. (6.2) it follows that if every actinide isotope has non-zero fission cross-section, then the $t \to \infty$ case results in $p_{if}(\infty) = 1$, for every $x_i \in A$. The time-dependent transmutation rates of the initial atoms can be given as the time derivative of the (6.2) fission probabilities:

$$\frac{dp_{if}}{dt} = e^{Q_A t} G_{if} = P_A(t) G_{if}. \tag{6.4}$$

Note that for each $x_i$, the time-dependent fission rate $p'_{if}(t)$ equals $\sum_j p_{ij}(t)p'_{jf}(0)$, which can be interpreted as the initial $x_i$ atom transforming into $x_j$ under irradiation time $t$ with probability $p_{ij}(t)$, with $x_j$ having its initial fission rate $\sigma_{jf} \Phi$.

### 6.1.2 Average time until fission

Another figure of merit that describes the transmutation rate of the initial $x_i$ atoms to fission products is the average time until fission, $T_{f,i}$, which is related to the effective fission half-life first defined by Mukaiyama and Gunji [80] in order to compare transmutation rates in different reactors. The average time until fission translates as the mean time until absorption in the CTMC and can be calculated with the aid of potential theory. In the model of the actinide transmutation chains, the state space is decomposed to the set of actinides $A$ and the absorbing fission state $x_f$, $S = A \cup \{x_f\}$. The time until fission is therefore the first exit time from the subset $A$, a so-called Markov killing time for the restricted process [81]:

$$T_{f,i} = \inf\{t : X_t = x_f \mid X_0 = x_i\}. \tag{6.5}$$

Let $P_A(t)$ denote the transition probability matrix restricted to actinides (which is substochastic, i.e. the elements in one row sum up to less than one), and $Q_A$ the restricted Q-matrix. It follows from the results of the previous subsection that $P(\{T_{f,i} < \infty\}) = 1$, therefore the special case of Kac’s moment formula [81] can be applied to calculate the $n$th moments of the $T_{f,i}$ time until fission:

$$E(T_{f,i}^n) = n! \delta_i^\top G_A^n 1, \tag{6.6}$$

where $G_A$ is the Green’s operator or potential kernel:

$$G_A = \int_0^\infty P_A(t)dt = -Q_A^{-1}, \tag{6.7}$$

and $1$ is a vector whose elements are ones. Specifically, the vector composed of the average time until fission for the different nuclides can be expressed from (6.6) as
the following:

\[ \hat{T}_f = -Q_A^{-1} \frac{1}{1}. \]  

(6.8)

Based on the above averages it can be determined whether the transformation of a
given nuclide to other nuclides accelerates or delays its conversion to fission prod-
ucts, as if there was only fission and no radioactive decay, as well as other nuclear
reactions, then the expected lifetime of an atom of nuclide \( x_i \) would be \( 1/q_{if} \). A
modified form of Eq. (6.6) also allows the calculation of the average time until fis-
sion conditioned on the fissioned daughter nuclide \( x_j \). The following relations can
be written based on Proposition 1 of Fitzsimmons and Pitman [81]:

\[ P(X_{T_f,i} = x_j) = \delta_i^\top G_A q_{if} \delta_j = q_{if}[Q_A^{-1}]_{ij}, \]

(6.9)

\[ \mathbb{E}(1\{X_{T_f,i} = x_j\}T_{f,i}) = \delta_i^\top G_A^2 q_{if} \delta_j = q_{if}[Q_A^{-2}]_{ij}. \]

(6.10)

The conditional expectation \( \hat{T}_{f,i,j} := \mathbb{E}(T_{f,i} \mid X_{T_i} = x_j) \) therefore evaluates as

\[ \hat{T}_{f,i,j} := \mathbb{E}(T_{f,i} \mid X_{T_f,i} = x_j) = \frac{\mathbb{E}(1\{X_{T_f,i} = x_j\}T_{f,i})}{P(X_{T_f,i} = x_j)} = \frac{[Q_A^{-2}]_{ij}}{[Q_A^{-1}]_{ij}}. \]

(6.11)

Both the fission probabilities and the average time until fission are in close relation
with the average number of reactions and decays in the chains, which is discussed
in the following subsection.

6.1.3 Number of reactions and decays until fission

The time-dependent fission probabilities and the average time until fission are on
the one hand determined by the fission cross-sections, but on the other hand they
are also strongly influenced by the number of absorptions which are needed to fission
the initial atom of a given nuclide, as well as the number and half-lives of radioactive
decays. In general, the more absorptions and decays need to occur before fission the
higher will be the average fission time, as each transition introduces a characteristic
time delay in the process. If the \( \mu_{ij} \) weights are defined as 1 for a specific reaction
type or decay mode, then it is possible to calculate its average number of occurrence
either for a finite irradiation time or asymptotically. As the average fission times
are also asymptotic quantities (integrated over infinite irradiation time), therefore
the average number of reactions and decays are also expressed in the \( t \to \infty \) limit.
The \( \vec{v} \) vectors are defined in this case as the following:

\[
\vec{v}^{(R)} = \begin{bmatrix} \sigma_1^{(R)} \Phi \\ \sigma_2^{(R)} \Phi \\ \vdots \\ \sigma_m^{(R)} \Phi \end{bmatrix}, \quad \vec{v}^{(D)} = \begin{bmatrix} f_1^{(D)} \lambda_1 \\ f_2^{(D)} \lambda_2 \\ \vdots \\ f_m^{(D)} \lambda_m \end{bmatrix},
\]  

(6.12)

where \( \sigma_i^{(R)} \) denotes the microscopic cross-section of the reaction type \( (R) \) for nuclide \( x_i \) and \( f_i^{(D)} \) denotes the branching ratio of the decay mode \( (D) \) for nuclide \( x_i \). The asymptotic values of the average number of the different reactions and decays, \( \kappa^{(R)} \) or \( \kappa^{(D)} \) can therefore be expressed as

\[
\kappa^{(R)} = -Q_A^{-1} \vec{v}^{(R)}, \quad \kappa^{(D)} = -Q_A^{-1} \vec{v}^{(D)},
\]  

(6.13)

where each element of the \( \kappa \) vectors, \( \kappa_i^{(R)} \) and \( \kappa_i^{(D)} \) are the respective average values if the initial atom is an atom of nuclide \( x_i \). The average number of absorptions can also be used to describe the fissile inefficiency of the main fissile nuclides defined by Krepel and Losa [44], as an ideal fissile material would have only one absorption inducing fission. The presented averages, however, are only valid asymptotically, as for finite time trajectories with lower number of required reactions and decays tend to manifest with greater probability.

### 6.1.4 Isotopic distribution of fissions

If the fission of a specific nuclide \( x_j \) is labeled with weight 1, then it is possible to count the average number of fissions as daughter \( x_j \) until given irradiation time (which equals the probability of fission as \( x_j \)), and normalization with the fission probabilities calculated in Section 6.1.1 yields the isotopic distribution of fissions of the initial atoms. In order to calculate the probability of fission of every nuclide \( x_i \) as every possible \( x_j \), Eq. (6.2) can be modified and transformed into a matrix equation. Let us define the \( V_f \) matrix, which is a diagonal matrix with the \( \sigma_{if} \Phi \) fission rates in its diagonal elements:

\[
[V_f]_{ij} = \begin{cases} 
\sigma_{if} \Phi, & \text{if } j = i; \\
0, & \text{if } j \neq i.
\end{cases}
\]  

(6.14)

The columns of \( V_f \) correspond to the calculation of fission probabilities restricted to the \( x_j \in A \) daughter nuclides, therefore the matrix consisting of the isotope-wise fission probabilities can be expressed with the following equation:

\[
P_f(t) = (e^{Q_A t} - I)Q_A^{-1}V_f,
\]  

(6.15)
where the \((i,j)\) element of \(P_f(t)\) equals the probability of fission of the initial \(x_i\) atom as daughter \(x_j\) under irradiation time \(t\). The \(t \to \infty\) limit of the above formula shows that the \([P_f(t)]_{ij}\) elements converge to the (6.9) asymptotic conditional fission probabilities. The time-dependent fission distribution of \(x_i\) can be expressed by normalization with the fission probability, i.e. the distribution is determined by the \([P_f(t)]_{ij}/p_f(t)\) ratios.

### 6.1.5 Time reversal from fission

The distribution of the fissioned daughter nuclides can be determined from another approach as well, namely by calculating the initial distribution of the transmutation chain reversed from the time of fission. It is well known that time reversal preserves the Markov-property, but in general it does not preserve temporal homogeneity. Ikeda [82] and Chung [83, 84] showed, however, that time reversal from the lifetime of a process, such as the time of fission in the actinide transmutation chains results in a homogeneous Markov process with stationary transition rates. Let us define the reverse process \(\tilde{X}_t = \{\tilde{X}_t : t \geq 0\}\) for an arbitrary initial distribution \(\tilde{\rho}(0)\) (that can either be concentrated in some \(x_i\) for a chain started in a specific nuclide, \(\rho(0) = \delta_i\) or an average initial distribution with \(\rho_i(0) = N_i/N\)) as follows. We extend the state space of the actinides, \(A\) with an artificial absorbing state \(\Delta\) and let

\[
\tilde{X}_t = \begin{cases} 
X_{T_f - t} & \text{if } 0 < t \leq T_f < \infty; \\
\Delta & \text{otherwise},
\end{cases}
\]

(6.16)

where \(T_f\) is the time of fission. In this case the elements of the reverse transition probability matrix \(\tilde{P}(t)\) and reverse transition rate matrix \(\tilde{Q}\) can be written in the following form (see Chung et al. [83, 84]):

\[
\tilde{p}_{ji}(t) = \frac{p_{ij}(t)g_i}{g_j}, \quad \tilde{q}_{ji} = \tilde{p}_{ji}'(0) = \frac{q_{ij}g_i}{g_j},
\]

(6.17)

where \(g_i\) is the potential measure defined as follows (\(G_A\) is the potential kernel introduced in Section 6.1.2):

\[
g_i = \mathbb{E} \left( \int_0^\infty 1\{X_t = x_i\} \, dt \right) = \sum_{j=1}^m \rho_j(0)[G_A]_{ji}. \quad (6.18)
\]

The initial distribution of \(\tilde{X}_t\) can be written as

\[
\tilde{\rho}_i(0) = g_i q_{if},
\]

(6.19)

which is equal to the asymptotic fission distribution. In particular if the forward chain was started in \(x_i\), then \(\tilde{\rho}_i(0)\) is equal to the (6.9) conditional fission probability.
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The distribution of $\tilde{X}_t$ describes the probability of the initial atom being a specific nuclide at time $t$ before its fission, and can be calculated with the reverse Q-matrix:

$$\tilde{\rho}^\top(t) = \tilde{\rho}^\top(0) \tilde{P}(t) = \tilde{\rho}^\top(0) e^{\tilde{Q}t}. \quad (6.20)$$

Since the process reversed from fission is also a homogeneous Markov chain, therefore reverse trajectory probabilities can be calculated with formulas (5.71) and (5.82) with the substitution of the reverse transition rates. The latter formula has to be used if the reverse trajectory ends with the absorbing state $\Delta$, which means that the starting nuclide of the reverse trajectory $\tilde{\gamma}$ was created in less than $t$ irradiation time.

It should be noted that although time reversal from a fixed irradiation or decay time does not result in stationary transition rates, the reversed transition probabilities can be used to describe the build-up of specific actinides or fission products from other nuclides. Given an atom of $x_j$ at some fixed time $t_0$, the probability that it was created from an $x_i$ atom in irradiation time $t < t_0$ can be calculated with Bayes’ theorem [78]:

$$\mathbb{P}(X_{t_0-t} = x_i|X_{t_0} = x_j) = \frac{\mathbb{P}(X_{t_0} = x_j|X_{t_0-t} = x_i) \cdot \mathbb{P}(X_{t_0-t} = x_i)}{\mathbb{P}(X_{t_0} = x_j)} = \frac{\rho_{ij}(t) \rho_i(t_0-t)}{\rho_j(t_0)} , \quad (6.21)$$

if $\rho_j(t_0) > 0$ and 0 otherwise. Since the expression on the right hand side contains the fraction $\rho_i(t_0-t)/\rho_j(t_0)$ the reverse transition probabilities depend on $t_0$ besides $t$, therefore a stationary reverse Q-matrix cannot be defined in this case.

### 6.2 Neutron balance

It was discussed in the works of Salvatores et al. [42, 43] and Krepel and Losa [44] that the transmutation and breeding capabilities of different systems are in close relation with the neutron balance of the fuel. On the other hand, transition averaged or time-integrated parameters related to the neutron economy correspond to counting transitions labeled with their specific neutron balance, for which closed form solutions were given based on the developed Markov chain models.

#### 6.2.1 Static D-factor

The calculation scheme presented in [42] is analogous to counting labeled transitions in a discrete-time Markov chain where neutron induced reactions are labeled with the $R_{Jn \rightarrow J(n+1)}$ weights. Let us introduce the $R_{ij}$ notation, where the $(i, j)$ index
pair represents the transition between the $x_i$ and $x_j$ nuclides, as well as the following $\underline{u}_D$ vector:

$$\underline{u}_D = \begin{bmatrix} \sum_{j \neq 1} R_{1j} p_{ij}^{(R)} \\ \sum_{j \neq 2} R_{2j} p_{2j}^{(R)} \\ \vdots \\ \sum_{j \neq m} R_{mj} p_{mj}^{(R)} \end{bmatrix}, \quad (6.22)$$

where the $(R)$ superscript in $p_{ij}^{(R)}$ represents probability of transition by nuclear reaction, excluding radioactive decay:

$$p_{ij} = p_{ij}^{(R)} + p_{ij}^{(D)}. \quad (6.23)$$

In general this separation of the transition probabilities is necessary, because some nuclear reactions can cause the same transition as specific radioactive decays e.g. $(n,p)$ reaction and $\beta^+$-decay. The D-factor corresponds to the case when the given atom or one of its daughters is ultimately fissioned. If only the actinide transmutation chains are considered, then fission is an absorbing state, and the $n \to \infty$ case readily describes these circumstances, as no actinide neutron consumption occurs after a chain ends with fission. The isotope-wise D-factors can be therefore calculated simultaneously with the following closed formula based on Eq. (5.25), implicitly taking into account every possible transmutation trajectory, including those with recurring nuclides:

$$D = (I - P_A)^{-1} \underline{u}_D, \quad (6.24)$$

where $D$ is the vector whose elements are the D-factors of the different nuclides. It was shown that the discrete-time and continuous-time Markov chain models are asymptotically equivalent, thus the D-factor can also be calculated in continuous-time, which yields the same result in the $t \to \infty$ limit:

$$D = -Q_A^{-1} \underline{u}_D, \quad (6.25)$$

where the $\underline{v}_D$ vector is defined as

$$\underline{v}_D = \begin{bmatrix} \sum_{j \neq 1} R_{1j} q_{ij}^{(R)} \\ \sum_{j \neq 2} R_{2j} q_{2j}^{(R)} \\ \vdots \\ \sum_{j \neq m} R_{mj} q_{mj}^{(R)} \end{bmatrix}, \quad (6.26)$$

The advantage of the derived formulas is that no probability threshold is needed to evaluate the isotope-wise D-factors, as Eq. (6.24) and (6.25) take into account every possible transmutation trajectory. In the case of the discrete-time model the (5.24)
Description of the actinide transmutation chains

formula can be used to investigate how the neutron consumption changes with the number of occurred transitions, while both the discrete-time and the continuous-time Markov chain models can be used to analyze the contribution of different fission trajectories to the D-factors.

6.2.2 Average neutron production

It was shown that the static D-factor measures the integrated average neutron balance of a given nuclide for infinite irradiation time. This fact suggests that it would be advantageous to define a quantity which measures the neutron balance for a finite time interval (e.g. for one or several irradiation cycles). In order to later establish a direct connection with the microscopic reactivity worth, the \( n_i(t) \) average neutron production was defined, which is the expected value of the number of neutrons produced by the transmutation chain of one atom of nuclide \( x_i \) during given irradiation time \( t \). The calculation of \( n_i(t) \) as a function of the time spent in the neutron field demands the continuous-time treatment, as specific nuclear reactions and radioactive decays occur on different timescales. Let us label the transitions between \( x_i \) and \( x_j \) by nuclear reactions with \( \nu_{ij} - 1 \) weights where \( \nu_{ij} \) is defined as

\[
\nu_{ij} = \begin{cases} 
0 & \text{for capture;} \\
\bar{\nu}_i & \text{for fission;} \\
2 & \text{for (n,2n) reactions;} \\
\text{etc.}
\end{cases}
\]

(6.27)

In order to obtain the time-integrated neutron productions, we need to define the \( v \) vectors in (5.37):

\[
v_i = \left[ \sum_{j \neq 1} (\nu_{ij} - 1)q_{1j}^{(R)} \right] \left[ \sum_{j \neq 2} (\nu_{2j} - 1)q_{2j}^{(R)} \right] \ldots \left[ \sum_{j \neq m} (\nu_{mj} - 1)q_{mj}^{(R)} \right],
\]

(6.28)

where the \((R)\) superscript in \( q_{ij}^{(R)} \) again represents a transition by nuclear reaction. Simple transformation shows that \( v_i \) is proportional to the traditional microscopic reactivity worth, which describes the neutron balance of nuclide \( x_i \) at \( t = 0 \):

\[
v_i = \sum_{j \neq i} (\nu_{ij} - 1)\sigma_{ij} \Phi = \left( \sum_{j \neq i} \nu_{ij} \sigma_{ij} - \sum_{j \neq i} \sigma_{ij} \right) \Phi = \left( \sum_{j \neq i} \nu_{ij} \sigma_{ij} - \sigma_i \right) \Phi = w_i \Phi,
\]

(6.29)

where \( w_i \) is the microscopic worth of nuclide \( x_i \). Let \( n_i(t) \) denote the expected value of the neutron production if the initial nucleus is \( x_i \), and \( \bar{n}(t) \) the vector composed
of \(n_i(t)\)’s. The average time-integrated neutron productions can be calculated with the following formula:

\[ n(t) = (e^{QA_t} - 1)Q^{-1}_A w \Phi. \]  

(6.30)

The physical meaning of the derivative of \(n_i(t)\) can be understood as the following. From (5.35) it is known that \(dn/dt\) can be expressed as

\[ \frac{dn}{dt} = Q_A n(t) + w \Phi = Q_A[(e^{QA_t} - 1)Q^{-1}_A w \Phi] + w \Phi = e^{QA_t} w \Phi. \]  

(6.31)

The above equation system evaluates for each individual nuclide \(x_i\) as

\[ \frac{dn_i}{dt} = \sum_j [e^{QA_t}]_{ij} w_j \Phi = \sum_j p_{ij}(t) w_j \Phi, \]  

(6.32)

where \(p_{ij}(t)\) is the probability that the initial \(x_i\) atom is transformed to \(x_j\) under irradiation time \(t\). Eq. (6.32) therefore describes the time evolution of the average neutron balance of one atom of nuclide \(x_i\). Accordingly, the macroscopic reactivity worth of the initial \(x_i\) atoms at time \(t\) can be calculated from the number of atoms and the average neutron balance:

\[ W_i = \sum_j [e^{QA_t}]_{ij} w_j N_i(0). \]  

(6.33)

Comparison of Eq. (6.30) in the \(t \to \infty\) limit shows that the asymptotic neutron productions are equal to \((-1)\) times the static D-factors (due to the fact that \(v = -v_D\) and \(p_{ij}(\infty) = 1\)).

### 6.2.3 Dynamic D-factor

The dynamic D-factor was defined by Krepel and Losa [44] based on time integrals of the respective reaction rates of the daughter products weighted with their neutron consumption. The sum of the integrals are equal to \((-1)\) times the integrated neutron productions, while the time integral of the fission rates in the denominator of Eq. (2.16) correspond to the fission probabilities calculated in Section 6.1.1. The dynamic D-factor of nuclide \(x_i\) can therefore be expressed with the above quantities in the following form:

\[ D_{dyn,i}(t) = \frac{-n_i(t)}{p_{ij}(t)}. \]  

(6.34)

The convergence to the static D-factor is also satisfied in formula (6.34), because in the \(t \to \infty\) limit the \(n_i(t)\) neutron productions converge to \((-1)\) times the static D-factors, and the \(p_{ij}(t)\) fission probabilities converge to 1. The \(t \to 0^+\) limit can
be evaluated with L’Hospital rule as both the numerator and the denominator in Eq. (6.34) converge to zero:

\[
\lim_{t \downarrow 0} -n_i(t) \quad \frac{p_i(t)}{p_i(t)} = -\lim_{t \downarrow 0} n'_i(t) = -\frac{w_i \Phi}{q_i f} = -\frac{w_i}{\sigma_{if}},
\]

(6.35)

which is equal to the limit calculated in [44], encouraging that the presented formula correctly reproduces the dynamic D-factors.

6.2.4 Neutron balance in closed fuel cycle equilibrium

Additional conclusions can be drawn about the relation between the time-dependent and asymptotic neutron productions in closed fuel cycle equilibrium conditions. In a simplified fuel cycle scheme, the connection between the equilibrium beginning-of-cycle fuel composition, \( N_0 \), and the equilibrium feed vector, \( N_F \), can be expressed with the following relation according to the fuel evolution during burn-up (B) and cooling in interim storage (C):

\[
e^{(Q_B^t)\top t_B} e^{(Q_C^t)\top t_C} N_0 + N_F = N_0,
\]

(6.36)

as the initial fuel composition is the same at the beginning of each equilibrium cycle (of course the above relation is only valid with the equilibrium cross-sections). The beginning-of-cycle fuel vector can therefore be expressed with the feed as follows:

\[
N_0 = (I - e^{(Q_B^t)\top t_B} e^{(Q_C^t)\top t_C})^{-1} N_F.
\]

(6.37)

In these subsequent irradiation and cooling periods, the neutron production integrated for \( k \) cycles can be calculated with Eq. (5.43), taking into account that the neutrons are only produced and consumed during burn-up (neglecting spontaneous fission):

\[
n_k(t_B + t_C) = \sum_{l=1}^{k} \left( e^{Q_B^t l t_B} e^{Q_C^t l t_C} \right)^{(l-1)} n(t_B).
\]

(6.38)

The asymptotic neutron productions can be given in the \( k \to \infty \) limit:

\[
n(\infty) = (I - e^{Q_B^t l t_B} e^{Q_C^t l t_C})^{-1} n(t_B).
\]

(6.39)

Let us denote the Euclidean scalar product with \( \langle ., . \rangle \), with which the total neutron balance of the fuel during one equilibrium cycle can be written as the following:

\[
n_{N_0}(t_B) = \langle N_0, n(t_B) \rangle.
\]

(6.40)
Using the equation between the equilibrium beginning-of-cycle fuel composition and the feed vector, the following relationship can be derived:

\[
n_{N_0}(t_B) = \langle (I - e^{(Q_0^{*})^T t_c} e^{(Q_B^*)^T t_B})^{-1} N_F, u(t_B) \rangle =
\]

\[
= \langle N_F, (I - e^{Q_B^{*}t_B} e^{Q_A^T t_c})^{-1} u(t_B) \rangle = \langle N_F, u(\infty) \rangle = n_{N_F}(\infty). \quad (6.41)
\]

Therefore it can be concluded that the neutron balance of the equilibrium fuel mixture integrated over one burn-up cycle equals the asymptotic neutron production (or (-1) times the D-factor) of the equilibrium feed vector. The same relationship holds for similar quantities which change only during the irradiation periods, such as the number of fissions, or produced energy excluding decay heat during interim storage. Although the equilibrium fuel composition depends on the actual fuel cycle scheme and available material stockpiles, the above result provides a guiding concept how specific parameters of the equilibrium fuel mixture can be influenced by changing the composition of the feed.

### 6.3 Breeding and fuel utilization

The following subsections aim to describe fuel utilization related parameters of different reactors and fuel cycle schemes in terms of the developed mathematical models. In the first subsection the contribution of individual nuclides, as well as specific transmutation trajectories to the breeding gain is analyzed, while in the second subsection the average energy productions of the transmutation chains are described throughout their overall irradiation history as a figure of merit for fuel utilization in different fuel cycles.

#### 6.3.1 Breeding gain

In quasi-static approximation \( w_1 = w_2 =: w \), and the (2.13) breeding gain can be written for one irradiation cycle without fuel management as the following:

\[
BG = \frac{\langle w, N_2 - N_1 \rangle}{\langle w, N_1 \rangle} = \frac{\langle w, (e^{Q^T t_c} - I) N_1 \rangle}{\langle w, N_1 \rangle} = \frac{\langle (e^{Q t} - I) w, N_1 \rangle}{\langle w, N_1 \rangle}. \quad (6.42)
\]

The contributions of different nuclides can therefore be expressed in terms of the average worth change due to transformation to other nuclides:

\[
BG_i = \frac{1}{\langle w, N_1 \rangle} \left( \sum_{j=0}^{m} p_{ij}(t) w_j - w_i \right) N_{1,i}. \quad (6.43)
\]
Since the transition probabilities \( p_{ij}(t) \) can be expressed as infinite sums of possible transmutation trajectory probabilities from \( x_i \) to \( x_j \), therefore the contribution of specific transmutation trajectories to the breeding gain can also be measured based on the continuous-time Markov chain model. The contribution of a transmutation trajectory \( \gamma_{i \rightarrow j} \) which starts in \( x_i \) and ends in \( x_j \), \( BG_{\gamma_{i \rightarrow j}} \) can be expressed as follows:

\[
BG_{\gamma_{i \rightarrow j}} = \frac{1}{\langle w, N_1 \rangle} P_{\gamma_{i \rightarrow j}}(t)(w_j - w_i)N_{1,i}.
\]  

(6.44)

Another approach can be used as well to calculate isotope-wise contributions to the breeding gain based on counting transitions in the specific transmutation chains labeled with the resulting changes in microscopic reactivity weight. Let us define the vector \( v_{\Delta w} \) accordingly:

\[
v_{\Delta w} = \begin{bmatrix}
\sum_{j \neq 1} (w_j - w_1)q_{1j} \\
\sum_{j \neq 2} (w_j - w_2)q_{2j} \\
\vdots \\
\sum_{j \neq m} (w_j - w_m)q_{mj}
\end{bmatrix} = Qw.
\]

(6.45)

The vector composed of the average reactivity weight changes for transmutation chains which start from the different nuclides can therefore be written as

\[
\Delta w = (e^{Qt} - I)Q^{-1}v_{\Delta w} = (e^{Qt} - I)Q^{-1}Qw = (e^{Qt} - I)w.
\]

(6.46)

Simple substitution shows that normalization with the initial reactivity worth \( \langle w, N_1 \rangle \) and multiplication with the number of initial \( x_i \) atoms \( N_{1,i} \) indeed results in the same contribution to the breeding gain for nuclide \( x_i \) as (6.43), and the sum of these contributions equals (6.42). Both the above and previous descriptions of isotope-wise contributions can be extended for time intervals with different cross-section sets following Section 5.2.3, as well as closed fuel cycles with cooling in interim storage and refueling with external feed (in this case the contribution of the feed has to be calculated separately).

For an infinite homogeneous reactor described in diffusion formalism the reactivity weights are identical to the traditional microscopic reactivity worths [41]. In this approximation the breeding gain for irradiation cycle can also be expressed as a function of time-integrated neutron productions:

\[
BG = \frac{\langle (e^{Qt} - I)w, N_1 \rangle}{\langle w, N_1 \rangle} = \frac{\langle Q(e^{Qt} - I)Q^{-1}w\Phi, N_1 \rangle}{\langle QQ^{-1}w\Phi, N_1 \rangle} = \frac{\langle Qn(t), N_1 \rangle}{\langle Qn(\infty), N_1 \rangle}.
\]

(6.47)

The above formula shows that with the applied approximations the individual contributions of different nuclides to the breeding gain are proportional to \( \sum_{j=1}^{m} q_{j} n_j(t) \), which already depends on the properties of multiple nuclides.
6.3.2 Fuel utilization

The efficiency of uranium utilization is usually measured with the uranium utilization ratio, which equals the energy produced from unit mass of uranium in the specific fuel cycle compared to its theoretical energy content. In this subsection another interpretation of fuel utilization is given in terms of the continuous-time Markov chain model which corresponds to the average energy production of individual atoms of the different nuclides based on the results of Section 5.2.3. Let us denote the energy release in the fission and radiative capture of nuclide \( x_i \) with \( \epsilon_f^i \) and \( \epsilon_c^i \), respectively. The vector composed of the average time-integrated energy productions of different nuclides, \( \mathbf{E}(t) \) can be expressed for \( n \) time intervals with Eq. (5.43):

\[
\mathbf{E}(s_n) = \sum_{k=1}^{n} \left( \prod_{l=1}^{k-1} e^{Q_A^{(k)}t_l} \right) \mathbf{E}^{(k)}(t_k), \quad (6.48)
\]

with the energy production in the different time intervals (e.g. burn-up in different reactors, decay during refueling or in interim storage) being the following:

\[
\mathbf{E}^{(k)}(t_k) = (e^{Q_A^{(k)}t} - I)[Q_A^{(k)}]^{-1} \mathbf{E}_E^{(k)}, \quad (6.49)
\]

where the vector \( \mathbf{E}_E^{(k)} \) can be expressed as

\[
\mathbf{E}_E^{(k)} = \begin{bmatrix}
\epsilon_1 f \sigma_1^f + \epsilon_1 c \sigma_1^c \\ 
\epsilon_2 f \sigma_2^f + \epsilon_2 c \sigma_2^c \\
\vdots \\
\epsilon_m f \sigma_m^f + \epsilon_m c \sigma_m^c 
\end{bmatrix} \Phi. \quad (6.50)
\]

With the above formulas, the average time-integrated energy productions of different nuclides as well as an initial fuel mixture can be measured in arbitrary fuel cycle schemes, providing a figure of merit for the comparison of fuel utilization properties. The total energy production of an initial fuel mixture \( \mathbf{N}(0) \) can be calculated with the following scalar product:

\[
E_{\mathbf{N}(0)}(s_n) = \langle \mathbf{N}(0), \mathbf{E}(s_n) \rangle. \quad (6.51)
\]

Since the energy release during cooling in interim storage is not taken into account, therefore the previous conclusion concerning the neutron balance of the fuel in equilibrium closed fuel cycle conditions is also true for the energy production, i.e. the energy produced in the fuel in one equilibrium burn-up cycle equals the asymptotic energy production of the feed vector. Note that partitioning losses can be also taken into account in this scheme as well in the form of time-independent diagonal matrices with the partitioning efficiencies in the diagonal.
Chapter 7

Minor actinide transmutation in Generation IV fast reactors

The developed mathematical models of the actinide transmutation chains were used to analyze minor actinide transmutation in the three investigated Generation IV fast reactors, in particular to explain why minor actinide feed from spent LWR fuel results in improved breeding in the reference cores. The first two sections give an overview of underlying processes in minor actinide burning in the closed fuel cycles of the reactors based on the analysis of the neutron balance and time behavior of specific transmutation chains. The effect of minor actinide feed on the breeding gain is investigated in the third section of the chapter.

7.1 Minor actinide burning

Minor actinide elements produced in the highest amount in LWRs are neptunium, americium and curium, in particular $^{237}$Np, $^{241}$Am, $^{243}$Am and $^{244}$Cm. Although the radiotoxicity inventory of the spent fuel is dominated by neptunium after a million years, neptunium radiotoxicity always remains below that of natural uranium [85, 86]. $^{244}$Cm has high midterm contribution, but its relative radiotoxicity becomes negligible after few hundred years due its short half-life. Besides Pu isotopes, the main contributors to the radiotoxicity inventory are $^{241}$Am and $^{243}$Am, whose transmutation is therefore of particular interest. The transmutation of Np is partly motivated by its fertile nature, which is verified by the results of the next section, as well as its high transfer rate through human food chains [14]. Curium transmutation is desired in the case of Am multirecycling, which results in the build-up of Cm isotopes with higher mass numbers. The processes that are responsible for the conversion of these particular nuclides to fission products are analyzed in the second subsection.
7.1. Minor actinide burning

7.1.1 Neutron balance

The microscopic reactivity worths of the listed minor actinide isotopes are negative both in thermal and fast neutron spectrum (with different orders of magnitudes due to the decrease of microscopic cross-sections at higher neutron energies), with the exception of $^{244}\text{Cm}$, whose microscopic worth is positive in fast reactors (see Table 7.1). This means that initially $^{241}\text{Am}$, $^{237}\text{Np}$ and $^{243}\text{Am}$ are absorbers both in thermal and fast reactors. On the other hand, their asymptotic neutron balance can be described as well using the static D-factor, which however shows that their transmutation to fission products has positive integral contribution (negative D-factor) to the neutron economy in fast reactors (see Figures 7.1 and 7.2). A negative D-factor is only possible if at some point of the irradiation the bulk of the initial atoms has positive worth, meaning that the original material turns from net absorber to net fissile material. This statement can be confirmed by calculating the average neutron productions and their time derivatives, of which the latter describe the average neutron balance after given irradiation time, and changes sign in these cases. Figure 7.3 shows that the time derivative of the neutron production (which is proportional to the expected value of the microscopic worth) is always positive for $^{244}\text{Cm}$, and indeed changes from negative to positive sign after 5 to 10 years irradiation time in the case of $^{237}\text{Np}$, $^{241}\text{Am}$ and $^{243}\text{Am}$. 

Figure 7.1: Static D-factors of major actinides evaluated in different reactors
Table 7.1: Microscopic worths of actinide isotopes in the EPR and in the three investigated Generation IV fast reactors

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Microscopic worth [barn]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>EPR</td>
</tr>
<tr>
<td>$^{234}$U</td>
<td>-19.45</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>44.51</td>
</tr>
<tr>
<td>$^{236}$U</td>
<td>-8.05</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>-0.71</td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>-30.37</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>-20.94</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>135.73</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>-227.05</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>156.15</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>-33.25</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>-101.65</td>
</tr>
<tr>
<td>$^{242m}$Am</td>
<td>1080.22</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>-49.57</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
<td>-2.64</td>
</tr>
<tr>
<td>$^{243}$Cm</td>
<td>172.12</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>-15.89</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>251.57</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>-1.68</td>
</tr>
</tbody>
</table>
7.1. Minor actinide burning

**Figure 7.2:** Static D-factors of minor actinides evaluated in different reactors

**Figure 7.3:** Average neutron productions of MA isotopes as a function of irradiation time (left) and their time derivatives (right) (GFR2400)
7.1.2 Fission dynamics

The continuous-time Markov chain model of the nuclide chains allows to identify the main processes which are responsible for the conversion of specific minor actinide isotopes to fission products, as well as the timescales of transmutation. Asymptotic quantities such as the average time until fission and the average number of reactions and decays until fission provide global information about relevant processes that can be interpreted in equilibrium closed fuel cycle conditions, when the actinide content of the fuel is recycled until eventually being fissioned. Time-dependent transmutation trajectory probabilities on the other hand show which trajectories contribute to minor actinide burning both on finite timescales and in the case of multiple recycling.

Table 7.2 shows the average time until fission for different actinides in the three investigated Generation IV fast reactors, as well as how much the average time until fission would be if there was only fission and no other nuclear reactions or radioactive decay (which is equal to 1/q_f). Comparison of the cases with and without other reaction types and decay shows that in the case of fissile nuclides such as 235U, 239Pu, 241Pu and 242mAm these transformations increase the average time until fission. On the other hand, the average time until fission in the case of fertile nuclides, including the important minor actinides 237Np, 241Am, 243Am and 244Cm is decreased due to transformation to other nuclei via nuclear reactions and radioactive decay. The most significant decrease can be observed in the case of 236U, 238U and 242Cm with almost one order of magnitude.

Figures 7.4-7.7 show the distribution of fissioned daughter nuclides of 237Np, 241Am, 243Am and 244Cm in the GFR2400, averaged until 50 MWd/kg discharge burn-up, as well as their asymptotic distributions. As expected, on short timescales most fissions occur as the initial nuclide, or after one neutron capture and possibly an additional radioactive decay of the short-lived capture product. An exception with more complex transformations already at short timescales is the fission of 241Am as 238Pu. If the neutron capture of 241Am results in the ground state of 242Am, then the 242Am→242Cm→238Pu decay can happen due to the short half-lives of the β^- decay of 242Am (16.02 h) and α-decay of 242Cm (162.8 d). In general on longer timescales and in the asymptotic distribution more nuclides have significant contributions, as transmutation trajectories with higher number of nuclear reactions and radioactive decays tend to manifest in greater fraction. For example, the fission of an initial 237Np atom occurs mainly as 237Np or 238Pu until the discharge burn-up, while for long irradiation times fission as 239Pu and even 240Pu has considerable probability. Time-dependent fission trajectory probabilities depicted in Figures 7.8-7.11 are
consistent with the time behavior of fission distributions, namely that trajectories with few transitions dominate the fission probability for short irradiation times and trajectories with several transitions appear in the case of multiple recycling. The following observations can be concluded concerning the transmutation of the enlisted four MA isotopes from the fission distributions and fission trajectory probabilities:

- The conversion of MA isotopes to fission products during single recycling mainly occurs directly or after one neutron capture;

- In closed fuel cycles with multiple recycling, $^{237}\text{Np}$ and $^{241}\text{Am}$ mainly fissions as $^{238}\text{Pu}$ and $^{239}\text{Pu}$, while $^{243}\text{Am}$ and $^{244}\text{Cm}$ transmutation occurs in highest fraction as $^{244}\text{Cm}$ and $^{245}\text{Cm}$, as well as Pu isotopes with higher mass numbers, $^{240}\text{Pu}$ and $^{241}\text{Pu}$;

- Fission trajectories with higher number of transitions are realized in general in the case of multiple recycling, but the direct conversion to fission products is also significant on longer timescales as well.

### 7.2 Fissile material breeding

The following subsections aim to analyze fissile material breeding in closed fuel cycle equilibrium conditions in the reference Generation IV fast reactors. Based on the continuous-time Markov-chain models of transmutation, the contributions of
Table 7.2: Average time until fission of different actinides in the three investigated Generation IV fast reactors

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$T_{f,i}$ [years]</th>
<th>$1/q_{if}$ [years]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>GFR2400</td>
<td>ELSY</td>
</tr>
<tr>
<td>$^{234}\text{U}$</td>
<td>34.93</td>
<td>60.05</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>19.56</td>
<td>9.81</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td>52.10</td>
<td>169.11</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>70.64</td>
<td>394.84</td>
</tr>
<tr>
<td>$^{237}\text{Np}$</td>
<td>23.83</td>
<td>55.53</td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>17.49</td>
<td>16.55</td>
</tr>
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7.2. Fissile material breeding

**Figure 7.5:** Average fission distribution of $^{241}$Am until 50 MWd/kg discharge burn-up of the GFR2400 fuel (left), and in the case of infinite recycling (right).

**Figure 7.6:** Average fission distribution of $^{243}$Am until 50 MWd/kg discharge burn-up of the GFR2400 fuel (left), and in the case of infinite recycling (right).
**Figure 7.7:** Average fission distribution of $^{244}$Cm until 50 MWd/kg discharge burn-up of the GFR2400 fuel (left), and in the case of infinite recycling (right).

**Figure 7.8:** Time-dependent fission trajectory probabilities of $^{237}$Np in the case of multiple recycling (GFR2400).
7.2. Fissile material breeding

Figure 7.9: Time-dependent fission trajectory probabilities of $^{241}$Am in the case of multiple recycling (GFR2400)

Figure 7.10: Time-dependent fission trajectory probabilities of $^{243}$Am in the case of multiple recycling (GFR2400)
different nuclides and transmutation trajectories to the breeding gain were calculated for the equilibrium BOC fuel compositions determined in the closed fuel studies. In the first subsection these contributions are analyzed in the case of U, Pu and MA multirecycling, whereas in the second subsection the effect of minor actinide feed from spent LWR fuel in the fresh fuel is investigated.

7.2.1 Prevalent processes

The results of the closed fuel cycle studies showed that the three reference Generation IV fast reactors are iso-breeders in the equilibrium due to slightly positive breeding gain and $^{241}$Pu decay during interim storage. In order to provide a deeper understanding of underlying processes in fissile material breeding in fast reactors, the contributions of individual nuclides and transmutation trajectories were calculated for one irradiation cycle without fuel management for the equilibrium BOC fuel compositions of the three reactors in the U, Pu and MA multirecycling case.

Figures 7.12 and 7.13 show the nuclide-wise contributions of different major and minor actinides to $BG$ based on Eq. 6.43. As expected, the highest absolute contributions can be accounted for $^{238}$U and $^{239}$Pu, with minor share of $^{240}$Pu, $^{241}$Pu and $^{241}$Am. Average values for minor actinides are one order of magnitude lower. Long-lived actinide isotopes with negative microscopic worth have positive contributions to $BG$, because the quantity of these actinide isotopes has to decrease during the irradiation in the equilibrium. This is due to the fact that there their decay during
cooling is negligible, and the feed is non-negative. The calculated values are very similar for the three Generation IV fast reactors, with somewhat higher variation in $^{241}$Am due to the different BOC americium contents, because the different cycle lengths result in different cooling times.

The continuous-time Markov chain model of the nuclide transmutation chains also allows the calculation of trajectory-wise contributions to the breeding gain during one burn-up cycle. Based on the results of Tables B.1 and B.2 in Appendix B, the following processes can be identified as the most significant positive contributors to the breeding gain in the three Generation IV fast reactors:

- the production of $^{239}$Pu from $^{238}$U and $^{241}$Pu from $^{240}$Pu;
- the production of $^{242m}$Am, $^{238}$Pu and $^{242}$Cm from $^{241}$Am;
- the direct and indirect (via conversion to $^{239}$Pu) fission of $^{238}$U;
- the production of $^{238}$Pu from $^{237}$Np and $^{242}$Cm;
- the production of $^{239}$Pu from $^{238}$Pu, $^{244}$Cm from $^{243}$Am, as well as $^{245}$Cm from $^{244}$Cm.

The greatest negative contributions to the breeding gain in the three investigated fast reactors can be accounted to the following processes:

- the direct fission of $^{239}$Pu and $^{241}$Pu;
- the conversion of $^{239}$Pu to $^{240}$Pu and $^{241}$Pu to $^{242}$Pu;
- the decay of $^{241}$Pu into $^{241}$Am;
- the direct fission of $^{242m}$Am, $^{238}$Pu, $^{240}$Pu, $^{235}$U and $^{245}$Cm.

The above results show that nuclides with large positive microscopic worths tend to have negative contributions on the breeding gain, while nuclides with negative (or positive, but low) microscopic worths have positive contributions. As expected, the set of fissile and fertile nuclides intersect due to the hard neutron spectrum of the investigated fast reactor cores.

### 7.2.2 Effects of minor actinide feed

The results of the closed fuel cycle studies showed that additional minor actinide feed from LWR spent fuel increased the breeding gain of the otherwise iso-breeder Generation IV fast reactors, which was also observed by Coquelet et al. [69] and
Figure 7.12: Contributions of major actinides to the breeding gain

Figure 7.13: Contributions of minor actinides to the breeding gain
7.2. Fissile material breeding

Meyer et al. [70] in the SFR. The improved breeding in the three fast reactors is analyzed in this section by the comparison of main breeding gain contributions in the case of U, Pu and MA multirecycling and fixed 3% MA content of the fresh fuel.

The equilibrium compositions determined in the closed fuel cycle studies were used for the analyses of the breeding gain, integrated for one irradiation cycle without fuel management. The equilibrium BOC composition in the U, Pu and MA multirecycling case is taken as the reference composition $\tilde{N}_0$, therefore the BOC composition in the 3% MA case, $\tilde{T}$ can be seen as a perturbation $\Delta N_0$ of the reference composition (see Table C.1 in Appendix C):

$$\tilde{N}_0 = N_0 + \Delta N_0.$$  \hspace{1cm} (7.1)

The respective breeding gains can be expressed with the corresponding BOC compositions and microscopic worths. In the case of the GFR2400, these evaluate as the following:

$$BG_{\text{ref}} = \frac{\langle (e_qt - I) w, N_0 \rangle}{\langle w, N_0 \rangle} = 0.0562,$$  \hspace{1cm} (7.2)

$$BG_{3\text{MA}} = \frac{\langle (e_\tilde{q}t - I) \tilde{w}, \tilde{N}_0 \rangle}{\langle \tilde{w}, \tilde{N}_0 \rangle} = 0.0804.$$  \hspace{1cm} (7.3)

In order to explore the causes of the increase in breeding gain, the contributions of individual nuclides and transmutation trajectories were calculated based on Eq. 6.43 and 6.44 (see Tables C.2-C.4). The values show that the highest increase in $BG$ in the GFR2400 is due to the increased $^{237}$Np content from spent Light Water Reactor fuel, as well as $^{241}$Am, $^{243}$Am and $^{244}$Cm. The other reason behind the improved breeding is the decrease of the equilibrium Pu content needed to ensure enough excess reactivity at BOC, specifically $^{239}$Pu and $^{241}$Pu due to the presence of fissile and fertile minor actinide isotopes. The improvement is partially counterweighted by the decreased production of $^{239}$Pu from $^{238}$U and $^{241}$Pu from $^{240}$Pu, as well as the increased fission of $^{238}$Pu and $^{245}$Cm.

A different evaluation of nuclide-wise contributions to the change in $BG$ can be performed based on sensitivity analysis [87]. The sensitivity coefficients in general can be calculated with the Fréchet-derivative [88] of the breeding gain with respect to the BOC fuel composition at $\tilde{N}_0$. As a first approximation whose correctness was checked based on the obtained results, the derivative is expressed in terms of the quasi-static approximation, where the change in microscopic one-group cross-sections and the neutron flux is considered negligible compared to the effect of the change in composition:
\[
\frac{dBG[N_0]}{w[N_0]} = \frac{1}{\langle w, N_0 \rangle^2} \left( \langle (e^{Qt} - I)w, \Delta N_0 \rangle \cdot \langle w, N_0 \rangle - \right.
\]
\[
- \langle w, \Delta N_0 \rangle \cdot \langle (e^{Qt} - I)w, N_0 \rangle = \frac{\langle (e^{Qt} - I)w, \Delta N_0 \rangle - BG \cdot \langle w, \Delta N_0 \rangle}{\langle w, N_0 \rangle}. \tag{7.4}
\]

The vector composed of the \( S_i \) sensitivity coefficients, \( \mathbf{S} \) can therefore be written in the following form:
\[
\mathbf{S} = \frac{1}{\langle w, N_0 \rangle} \left[ (e^{Qt} - (1 + BG)I) \mathbf{w} \right], \tag{7.5}
\]
and the change in \( BG \) can be expressed with the sensitivity coefficients, as well as the change in BOC fuel composition:
\[
\Delta BG = \langle \mathbf{S}, \Delta N_0 \rangle. \tag{7.6}
\]

Figures 7.14 and 7.15 show the sensitivity coefficients calculated for the U, Pu and MA multirecycling case, as well as nuclide-wise contributions to the change in \( BG \) in the three fast reactors. The results confirm that the main reasons behind the improved breeding are the increased \(^{237}\text{Np}, \ ^{241}\text{Am}, \ ^{243}\text{Am}\) and \(^{244}\text{Cm}\) content and the decreased \(^{239}\text{Pu}\) and \(^{241}\text{Pu}\) content, while the effect is somewhat moderated by the increased \(^{238}\text{Pu}\) and \(^{245}\text{Cm}\) content, as well as the decrease of \(^{238}\text{U}\). In the case of the GFR2400, the estimated change in \( BG \) based on the sensitivity calculation is 0.0332, while the difference between \( BG_{\text{ref}} \) and \( BG_{3\text{MA}} \) is 0.0242, which means that the quasi-static linear approach overestimates the increase. If \( BG_{3\text{MA}} \) was calculated with cross-sections from the U, Pu and MA multirecycling case, then the difference would be a somewhat higher value, 0.0302. The effect of the change in neutron spectrum and one-group cross-sections is therefore responsible for \( \Delta BG = 0.0060 \), less than one fourth of the total change.
7.2. Fissile material breeding

Figure 7.14: Nuclide-wise sensitivity coefficients to BG in the U, Pu, and MA multirecyling case
Figure 7.15: Nuclide-wise contributions to the change in BG between the U, Pu and MA multirecycling case and with 3% MA content in the fresh fuel based on sensitivity coefficients.
Chapter 8

Summary

Generation IV fast reactors are envisaged to operate in closed fuel cycles due to their ability to breed their fuel from fertile feed and burn minor actinides produced by themselves or thermal reactors in the nuclear park, therefore the production of nuclear waste can be limited to fission products and reprocessing losses. Strategic decisions about the deployment of fast reactors and the transition from open to closed fuel cycles are supported by fuel cycle scenario codes, which are capable of modeling the important facilities of the fuel cycle and tracking material flows between them. The simulation of the fuel cycle involves the calculation of fuel depletion in the reactors, for which most scenario codes use burn-up tables or parametrized few group cross-sections.

The first part of the thesis presented a fast burn-up scheme called FITXS in Chapter 3, which was used for the analyses of closed fuel cycles containing Generation IV fast reactors and Generation III thermal reactors. Based on the fitting of one-group cross-sections as functions of the detailed fuel composition, the developed scheme can provide accurate results even if the isotopic composition changes greatly, for example when multiple recycling of Pu and MAs is considered. The FITXS scheme was used to develop burn-up models for the Generation IV GFR, LFR and SFR, as well as MOX fuel assemblies of the Generation III EPR and VVER-1200. Three-dimensional core models and fuel assembly models of the reactors were created in the KENO-VI module of the SCALE 6.0 code, which were then used to prepare cross-section databases with numerous different fuel compositions in order to perform the least-squares fitting of the one-group cross-sections and the $k_{\text{eff}}$. The results of the fittings showed that the chosen second-order polynomials of the atomic densities could describe the cross-sections of important $(\text{n},\text{f})$ and $(\text{n},\gamma)$ reactions with average relative errors well below 1%, and the relative errors were generally in the order of the statistical uncertainties of the Monte Carlo transport
calculations used for the preparation of the cross-section databases. The burn-up models were verified with burn-up calculations using cross-sections calculated with the SCALE 6.0 code, as well as the fitted cross-sections. The results of the verification showed very good agreement between SCALE 6.0-based and FITXS-based burn-up calculations in the case of fast reactors, with typical relative errors below 0.1% for actinides which have higher than $10^{-5}$ relative mass fractions in the fuel. In the case of thermal reactors the results showed higher discrepancies, but the observed relative errors are still acceptable in scenario studies. The high accuracy of the fitted cross-sections and the good agreement between SCALE 6.0-based and FITXS-based burn-up results encourage that the FITXS method can be successfully applied to determine the spent fuel compositions of the reactors for a wide range of initial compositions in low computational time.

The burn-up models of the Generation IV fast reactors were integrated in equilibrium closed fuel cycle models and more complex transition scenarios. A fuel cycle simulation program called JOSSETE was developed for the analyses, which is presented in Chapter 4, along with the specifications and results of the equilibrium closed fuel cycle studies and transition scenario studies. The equilibrium closed fuel cycle operation of the three fast reactors was investigated by taking into account the whole transition from initial state to equilibrium, while the fresh fuel compositions of the reactors were set with iteration in each burn-up cycle based on the fitted $k_{\text{eff}}$, such that the fissile content of the fresh fuel provided enough excess reactivity to ensure criticality throughout the irradiation. The results of the analyses are consistent with related literature, namely that the three investigated fast reactors are iso-breeders in the equilibrium due to slight breeding and the decay of $^{241}\text{Pu}$ in interim storage, with approximately 1% minor actinide content, and that additional minor actinide feed results in improved breeding in the cores.

A more complex fuel cycle describing the transition from a VVER-440 fleet to a mixed fleet of Generation IV fast reactors and MOX fueled VVER-1200 reactors was also simulated, and different scenarios were investigated concerning the stabilization and reduction of TRU inventories. It was shown that the three Generation IV fast reactors are able to burn the minor actinide stocks which accumulated in the spent fuel of the VVER-440 fleet which produced the Pu for their start-up. Power ratios of the fast reactors and MOX fueled VVER-1200 reactors were determined for the stabilization of the Pu inventory in the fuel cycle, as well as for an overall TRU inventory reduction. The fresh fuel compositions were also determined with iteration based on the calculated $k_{\text{eff}}$, and it was shown that fresh fuel limits can be met throughout the scenarios if the reprocessed Pu from spent MOX fuel is always recycled in the fast reactors first. The results showed that the Pu and MA balance of
the mixed fleet can be set by changing the power ratios of fast and thermal reactors in the nuclear park.

The analysis of the breeding and transmutation capabilities of the reactors motivated the development of stochastic models of the nuclide transmutation chains based on discrete-time and continuous-time Markov chains, which were presented in Chapter 5. The developed models are consistent with the Bateman equations, but they describe the transmutation and decay chains of individual atoms as stochastic processes, either in terms of finite irradiation of decay time, or the number of occurred nuclear transitions. The continuous-time Markov chain model allows to identify the prevailing processes of minor actinide burning and fissile material breeding with the calculation of time-dependent probabilities of the different transmutation and fission trajectories in the nuclide chains. It was shown that the transmutation trajectory probabilities constitute the solution of the Bateman equations, and that the time-dependent trajectory probability is in fact the general solution of the Bateman equations for linear chains if unit concentration is assumed for the starting nuclide.

Based on the Markov chain models, a method was developed to count labeled transitions in the transmutation chains, which was then used to derive closed formulas for finite-time-integrated and asymptotic fuel cycle performance parameters in Chapter 6. Closed formulas were derived for time-dependent fission probabilities, D-factors and average neutron productions, whose time derivatives were shown to be proportional to the expected neutron balance of the different nuclides after given irradiation time. The stochastic description of the actinide transmutation chains also allowed the calculation of the average time until fission and the distribution of fissioned daughter nuclides. Based on the derived results, it was shown in a simplified closed fuel cycle scheme, that the average neutron production of the equilibrium fuel integrated for one burn-up cycle equals the asymptotic neutron production of the equilibrium feed vector.

The results of the equilibrium closed fuel cycle studies and the Markov chain models of the actinide transmutation chains were used to analyze minor actinide burning in the three Generation IV fast reactors, in particular they were used to investigate the effect of minor actinide feed on the breeding gain of the reactors. Based on the calculated D-factors and time-integrated neutron productions, it was shown that every actinide isotope has positive asymptotic contribution to the neutron economy, due to the fact that at some point of the irradiation the bulk of the initial atoms turns from net absorber to net fissile material. The average time until fission for different actinides in the three investigated Generation IV fast reactors was calculated, as well as how much the average time until fission would be if there
was only fission and no other nuclear reactions or radioactive decay. Comparison of the cases with and without other reaction types and decay shows that in the case of fissile nuclides these transformations increase the average time until fission, while in the case of fertile nuclides – including the important minor actinides $^{237}\text{Np}$, $^{241}\text{Am}$, $^{243}\text{Am}$ and $^{244}\text{Cm}$ –, the average time until fission is decreased due to transformation to other nuclei via nuclear reactions and radioactive decay. The most significant decrease was observed in the case of $^{236}\text{U}$, $^{238}\text{U}$ and $^{242}\text{Cm}$ with almost one order of magnitude. The prevalent processes of minor actinide burning in the reactors were identified by calculating the contributions of different fission trajectories to the fission probabilities, as well as the distribution of the fissioned daughter nuclides. The analyses showed that the conversion of MA isotopes to fission products during single recycling mainly occurs directly, or after one neutron capture, whereas fission trajectories with higher number of transitions are realized in general in the case of multiple recycling. Finally, the contributions of different nuclides and transmutation trajectories to the breeding gain in the three investigated fast reactors were calculated, and the effects of minor actinide feed were analyzed based on sensitivity coefficients and the Markov chain models. The results of the analyses showed that the main reasons behind the improved breeding in the otherwise iso-breeder fast reactors are the increased $^{237}\text{Np}$, $^{241}\text{Am}$ $^{243}\text{Am}$ and $^{244}\text{Cm}$ content and the decreased $^{239}\text{Pu}$ and $^{241}\text{Pu}$ content, while the effect is somewhat moderated by the increased $^{238}\text{Pu}$ and $^{245}\text{Cm}$ content, as well as the decrease of $^{238}\text{U}$. The effect of the change in neutron spectrum and the one-group cross-sections was responsible for less than one fourth of the total change in the GFR2400.

The main results of the thesis can be summarized in the following statements:

1. I have developed a fast burn-up scheme called FITXS based on the fitting of microscopic one-group cross-sections and the $k_{\text{eff}}$ as functions of the detailed fuel composition, including a wide selection of minor actinide isotopes. I have developed burn-up models with the FITXS scheme for the Generation IV Gas-cooled Fast Reactor, Lead-cooled Fast Reactor and Sodium-cooled Fast Reactor, as well as for MOX fuel assemblies of the Generation III European Pressurized Reactor and VVER-1200, which can determine the spent fuel compositions of the reactors for a wide range of fresh fuel compositions. I have verified the accuracy of the burn-up models using the SCALE 6.0 code [P1, P2, P3].

2. I have developed a fuel cycle simulation program called JOSSETE, with which I have demonstrated the applicability of the FITXS method in fuel cycle simulations and scenario studies by investigating the closed fuel cycles of the three
Generation IV fast reactors, taking into account the whole transition from initial state to equilibrium, while the fitting of the $k_{eff}$ allowed to determine the fresh fuel compositions of the reactors with iteration. Consistently with previous studies in related literature, the results show that the three investigated fast reactors are iso-breeder in the equilibrium due to slight breeding and $^{241}$Pu decay in interim storage, with approximately 1% minor actinide content. Additional minor actinide feed results in improved breeding in the cores [P1, P3, P4].

3. I have shown in scenarios describing the transition from conventional LWRs to a mixed fleet of Generation IV fast reactors and MOX fueled Generation III thermal reactors, that all of the three investigated fast reactors can burn minor actinide stocks that were accumulated in the spent fuels of the conventional LWRs which produced the plutonium for their start-up. I have determined the power ratios of the fast and MOX fueled thermal reactor fleet needed to stabilize or reduce the plutonium inventory, and shown that fuel composition limits can be met throughout the scenarios if the reprocessed plutonium from spent MOX fuel is recycled in the fast reactors first. A higher power ratio of MOX fueled thermal reactors can counterbalance the improved breeding in the fast reactors due to minor actinide feed in the burner phase, after which a lower thermal reactor power ratio is needed to reach an equilibrium state in the fuel cycle [P5, P6].

4. I have developed the stochastic models of the nuclide transmutation chains based on discrete-time and continuous-time Markov chains. I have shown that the continuous-time Markov chain model can be used to derive both the Bateman equations and time-dependent transmutation trajectory probabilities in the nuclide chains, including decay chains which end in a stable nuclide and actinide transmutation chains which end with fission. I have shown that the transmutation trajectory probability is the general solution of the Bateman equations for linear chains if unit concentration is assumed for the starting nuclide. Transmutation trajectory probabilities in the actinide transmutation chains can be used to identify the prevalent processes in minor actinide burning and fissile material breeding [P7].

5. I have developed a method to count the expected values of labeled transitions in the transmutation chains using the Markov chain models, with which I have derived closed formulas for the calculation of finite-time-integrated and asymptotic fuel cycle performance parameters, such as fission probabilities, average neutron balances, D-factors, the average time until fission and the distribution of fissioned daughter nuclides. Based on the derived formulas I have shown in a simplified closed fuel cycle scheme that the neutron production of the equilibrium fuel integrated for
one burn-up cycle equals the asymptotic neutron production of the feed vector \([P7, P8]\).

6. I have investigated the effect of minor actinide feed from spent LWR fuel on the breeding properties of the three Generation IV fast reactors. I have calculated nuclide-wise contributions to the increase in breeding gain due to minor actinide feed based on sensitivity coefficients, and mapped transmutation trajectories with the highest absolute contributions to the breeding gain based on the Markov chain models. I have shown that the improved breeding in the three fast reactors is mainly due to the production of \(^{238}\text{Pu}\) from \(^{237}\text{Np}\) and the decreased \(^{239}\text{Pu}\) and \(^{241}\text{Pu}\) content of the fresh fuel. The improvement is somewhat moderated by the decreased production of \(^{239}\text{Pu}\) from \(^{238}\text{U}\) and \(^{241}\text{Pu}\) from \(^{240}\text{Pu}\), as well as the increased \(^{238}\text{Pu}\) and \(^{245}\text{Cm}\) content. The spectral effects of the increased minor actinide content are much smaller compared to these changes in production and consumption rates \([P1, P7]\).
Acknowledgements

First I would like to express my gratitude to my supervisor, Máté Szieberth for his guidance and advice during my Ph.D. studies, as well as throughout my graduate and undergraduate years.

I would also like to express my appreciation and gratefulness to my parents and my sister for their support throughout my university studies, as well as my girlfriend, Boglárka Babcsány for her love and encouragement.

Last but not least, I am grateful to my high school physics teacher, György Vastagh for endearing physics to me with his devotion and deep commitment.

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List of publications


Appendix A

Verification of the GFR2400 and EPR burn-up models
Table A.1: Average relative errors of two hundred burn-up calculations using cross-sections calculated with the SCALE 6.0 code and the FITXS method for the GFR2400

<table>
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<td>1.82E-02</td>
<td>0.424</td>
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<td>$^{248}\text{Cm}$</td>
<td>2.33E-04</td>
<td>0.888</td>
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</table>
Table A.2: Average relative errors of two hundred burn-up calculations using cross-sections calculated with the SCALE 6.0 code and the FITXS method for the EPR

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Average EOC mass [kg]</th>
<th>Avg. relative error (%)</th>
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<tbody>
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</tr>
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<td>0.174</td>
</tr>
<tr>
<td>$^{235}$U</td>
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<td>0.185</td>
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<td>$^{236}$U</td>
<td>2.22E+00</td>
<td>0.086</td>
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<td>1.065</td>
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<tr>
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<tr>
<td>$^{248}$Cm</td>
<td>1.50E-07</td>
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Appendix B

Trajectory contributions to the breeding gain
Table B.1: Trajectory-wise contributions to the breeding gain, Part 1

<table>
<thead>
<tr>
<th>Transmutation trajectory</th>
<th>$BG_\gamma$</th>
<th>GFR2400</th>
<th>ELSY</th>
<th>ESFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{U} \rightarrow ^{239}\text{U} \rightarrow ^{239}\text{Np} \rightarrow ^{239}\text{Pu}$</td>
<td>0.29569</td>
<td>0.33487</td>
<td>0.31297</td>
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<tr>
<td>$^{239}\text{Pu} \rightarrow \text{FP}$</td>
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<td>-0.23338</td>
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<tr>
<td>$^{239}\text{Pu} \rightarrow ^{240}\text{Pu}$</td>
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<tr>
<td>$^{240}\text{Pu} \rightarrow ^{241}\text{Pu}$</td>
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<td>$^{241}\text{Pu} \rightarrow \text{FP}$</td>
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<td>-0.03595</td>
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<td>$^{241}\text{Pu} \rightarrow ^{241}\text{Am}$</td>
<td>-0.00925</td>
<td>-0.01348</td>
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<tr>
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<td>0.00701</td>
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<td>$^{238}\text{U} \rightarrow \text{FP}$</td>
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Table B.2: Trajectory-wise contributions to the breeding gain, Part 2

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<th>GFR2400</th>
<th>ELSY</th>
<th>ESFR</th>
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Appendix C

Effects of minor actinide feed in the GFR2400
**Table C.1:** Equilibrium BOC fuel compositions in the U, Pu and MA multirecycling case and with 3% MA content in the fresh fuel (GFR2400)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$N_i$ Ref.</th>
<th>$N_i$ 3% MA</th>
<th>Δ$N_i$</th>
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<tr>
<td>$^{235}$U</td>
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<td>2.967E+26</td>
<td>1.466E+26</td>
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<tr>
<td>$^{236}$U</td>
<td>2.527E+26</td>
<td>3.653E+26</td>
<td>1.126E+26</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>1.368E+29</td>
<td>1.333E+29</td>
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<td>$^{236}$Np</td>
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<td>$^{237}$Np</td>
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<td>1.613E+27</td>
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<tr>
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<td>8.180E+23</td>
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<td>4.630E+21</td>
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<tr>
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<tr>
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<td>9.920E+21</td>
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<tr>
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<tr>
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<tr>
<td>$^{243}$Am</td>
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<tr>
<td>$^{242}$Cm</td>
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<tr>
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<td>1.059E+24</td>
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<tr>
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</table>
**Table C.2: Nuclide-wise contributions to the breeding gain (GFR2400)**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$BG_i$ (Ref.)</th>
<th>$BG_i$ (3% MA)</th>
<th>$\Delta BG_i$</th>
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<td>$^{247}$Cm</td>
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<td>$^{248}$Cm</td>
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<td><strong>Total</strong></td>
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Table C.3: Trajectory-wise contributions to $BG$ in the U, Pu and MA multirecycling case and with 3% MA content in the fresh fuel (GFR2400), Part 1

<table>
<thead>
<tr>
<th>Transmutation trajectory</th>
<th>$BG_{\gamma}$</th>
<th>$3%$ MA</th>
<th>$\Delta BG_{\gamma}$</th>
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<tbody>
<tr>
<td></td>
<td>Ref.</td>
<td></td>
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<td>$^{238}U \rightarrow ^{239}U \rightarrow ^{239}Np \rightarrow ^{239}Pu$</td>
<td>0.29569</td>
<td>0.28438</td>
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</tr>
<tr>
<td>$^{239}Pu \rightarrow FP$</td>
<td>-0.21239</td>
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</tr>
<tr>
<td>$^{239}Pu \rightarrow ^{240}Pu$</td>
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</tr>
<tr>
<td>$^{240}Pu \rightarrow ^{241}Pu$</td>
<td>0.05034</td>
<td>0.04555</td>
<td>-0.00478</td>
</tr>
<tr>
<td>$^{241}Pu \rightarrow FP$</td>
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<td>$^{241}Pu \rightarrow ^{241}Am$</td>
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<tr>
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<td>0.00703</td>
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<tr>
<td>$^{241}Am \rightarrow ^{242Am} \rightarrow ^{242Cm} \rightarrow ^{238}Pu$</td>
<td>0.00579</td>
<td>0.00632</td>
<td>0.00053</td>
</tr>
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<td>$^{242m}Am \rightarrow FP$</td>
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<td>-0.00050</td>
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<td>$^{241}Pu \rightarrow ^{242Pu}$</td>
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<tr>
<td>$^{241}Am \rightarrow ^{242Am} \rightarrow ^{242Cm}$</td>
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<tr>
<td>$^{240}Pu \rightarrow FP$</td>
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<td>$^{244}Cm \rightarrow ^{245}Cm$</td>
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<tr>
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<tr>
<td>$^{234}U \rightarrow ^{235}U$</td>
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<tr>
<td>$^{243}Am \rightarrow ^{244Am} \rightarrow ^{244Cm}$</td>
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<tr>
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<tr>
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**Table C.4:** Trajectory-wise contributions to $BG$ in the U, Pu and MA multirecycling case and with 3% MA content in the fresh fuel (GFR2400), Part 2

<table>
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<th>Transmutation trajectory</th>
<th>$BG_{\gamma}$</th>
<th>3% MA</th>
<th>$\Delta BG_{\gamma}$</th>
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</thead>
<tbody>
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<td>0.00111</td>
<td>0.00008</td>
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<tr>
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</tr>
<tr>
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</tr>
<tr>
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<tr>
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Bibliography


