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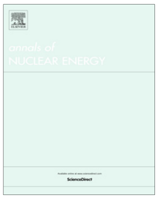

Markov chain models of nuclear transmutation: Part I – Theory

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ABSTRACT

This paper presents mathematical models of the nuclide transmutation and decay chains based on discrete-time and continuous-time Markov chains. The models describe the transmutation chains of individual atoms as stochastic processes, and can provide details about the underlying processes in minor actinide burning and fissile material breeding. In terms of expected values, the continuous-time Markov chain model is consistent with the Bateman equations, and it can be used to derive 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. It is shown that transmutation trajectory probabilities in fact constitute the general solution of the Bateman equations. A method was developed to count labeled transitions in the nuclide chains, and the models of the actinide transmutation chains were used to obtain closed formulas for the calculation of finite-time integrated and asymptotic fuel cycle performance parameters, such as time-dependent fission probabilities and the distribution of fissioned daughter nuclides, the average time until fission, static and dynamic D-factors and the average neutron balance of different nuclides. As a demonstration of application, a detailed analysis of transmutation and breeding in Generation IV fast reactors will be presented in Part II of the article.

1. Introduction

The analysis and comparison of the breeding and transmutation capabilities of different reactor designs requires specific parameters and methods capable of measuring fuel cycle performance. In order to compare the transmutation potential of different systems, the average neutron consumption per fission (D-factor) was defined by Salvatores et al. (1994), which enumerates the number of neutrons consumed (or produced) by an atom of a given nuclide until the initial atom or one of its daughters is eventually fissioned, and the transmutation chain ends. D-factors are asymptotic quantities in the sense that they are averaged over infinite irradiation time or infinite recycling of the fuel, when the initial atom is fissioned with probability one. Krepel and Losa (2016) also introduced the dynamic D-factor, with which the evolution towards the static D-factor can be studied, and more detailed conclusions can be drawn regarding the neutron economy of different reactor designs. These quantities measure the average neutron balance of transmutation chains which start from a specific nuclide, in fact the static D-factors can be calculated with weighted sums over possible transmutation trajectories. The fact that in transmutation networks and branching decay schemes the transmutation chains can be broken down into a set of independent linear chains was proved by Shlyakhter (1983) and Raykin and Shlyakhter (1989), who gave the solution of the Bateman equations in terms of transition probabilities and depletion functions introduced by Siewers (1976). A general procedure that resolves the non-linear chain into linear chains called transmutation trajectory analysis (TTA) was developed by Cetnar (2006), who also provided the general solution of the Bateman equations for linear chains with multiple recurring nuclides. In a recent study (Oettingen et al., 2017) used the TTA method 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.

This paper presents mathematical models of the nuclide transmutation chains based on discrete-time and continuous-time Markov chains, which were developed in order to provide details about the time evolution of individual transmutation and decay chains and a profound understanding of underlying processes in minor actinide burning and fissile material breeding. In particular, the models of the actinide transmutation chains were used to derive closed formulas for finite-time integrated and asymptotic fuel cycle performance parameters, such as fission probabilities and the average time until fission, static and dynamic D-factors, as well as average isotope-wise neutron productions. The continuous-time

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Markov chain model of the nuclide chains also allows to identify prevailing processes in minor actinide burning and fissile material breeding with the calculation of time-dependent transmutation trajectory probabilities.

The structure of the article is therefore as follows. In the second section the above mentioned methods related to the analysis of transmutation capabilities are overviewed. The mathematical models of the nuclide transmutation chains are introduced in the third section, along with the methodology which was used to count labeled transitions in specific nuclide chains and the calculation of transmutation trajectory probabilities. In the fourth section several performance parameters are calculated in order to describe the dynamics of actinide transmutation chains, as well as their neutron balance. As a demonstration of application, a detailed analysis of transmutation and breeding in Generation IV fast reactors will be presented in Part II of the article.

2. Related methods

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 methods that can assess transmutation capabilities and analyze underlying processes are overviewed. In order to disambiguate the terminology which is used in this paper, the applied definitions are listed in the following (see Fig. 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.

Throughout the paper, vectors are denoted with underlined letters and symbols, and matrices are denoted with non-italic, capital letters.

2.1. Static and dynamic D-factor

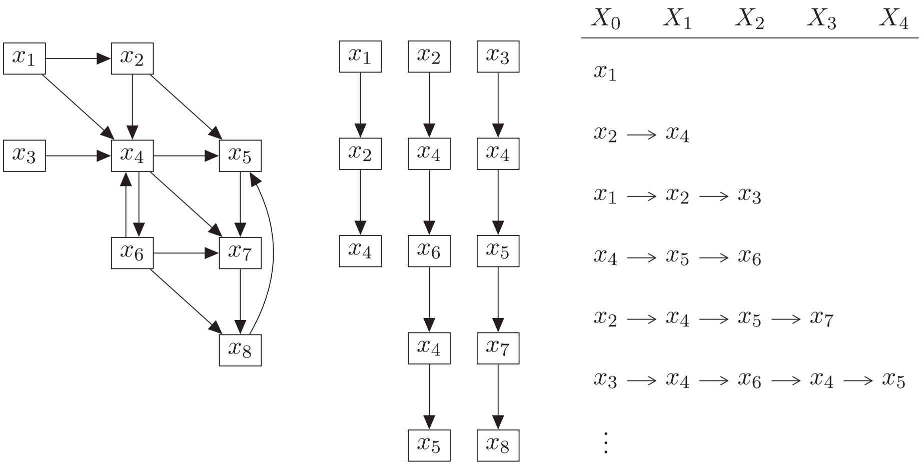
The D-factor, defined by Salvatores et al. (1994) and Salvatores (2002) describes the average number of neutrons consumed by a given nuclide and its daughter products until one of them is finally fissioned and the specific actinide transmutation chain ends. If the neutron consumption of an isotope is 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 nth generation reaction products weighted with the probability of the transitions, $P_{Jn\to J(n+1)}$ (Salvatores et al., 1994):

$$D_J = \sum_{J1_i} P_{J\to J1_i}\left\{R_{J\to J1_i} + \sum_{J2_k} P_{J1_i\to J2_k}\left[R_{J1_i\to J2_k} + \ldots\right]\right\}, \tag{1}$$

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

$$R_{Jn\to J(n+1)} = \begin{cases} 1 \quad \text{for capture,} \\ 0 \quad \text{for radiactive decay,} \\ 1-\bar{\nu} \quad \text{for fission,} \\ -1 \quad \text{for (n, 2n) reactions,} \\ \text{etc.} \end{cases} \tag{2}$$

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, 2016). It follows from the definition, that the D-factor is an



(a) (b) (c)

Fig. 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 ($n \in \mathbb{N}_0 = \{0, 1, 2, \ldots\}$).