

Trajectory Periods Folding Method for Modeling of Uranium and Thorium Fuel Transmutations

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Abstract. The Continuous Energy Monte Carlo Burnup Code (MCB) is a general-purpose tool dedicated for simulations of radiation transport and radiation-induced changes in matter in any nuclear system. In the recent years the methodology of the trajectory periods folding was integrated with MCB. In the proposed methodology, the mass flow of direct nuclide-to-nuclide transitions leading to nuclide transmutation chains in every step is interpreted over entire period of interest. In this way, all quantitative information about the transmutation process for the period beyond single calculation step is preserved. In the study we present the background of the method as well as its implementation for the modeling of the Lead Cooled Fast Reactor in the adiabatic equilibrium state.

Keywords: Monte Carlo, MCB, LFR, Transmutations, Nuclear Fuel

1. Introduction

The complexity of burnup process in the nuclear reactors requires application of an integrated calculation system, which allows taking into consideration spatial effects of heterogeneous reactor core model with continuous energy representation of cross sections and the thermo-hydraulic coupling. The integrated Continuous Energy Monte Carlo Burnup Code (MCB) developed in recent years satisfies needed requirements [1,2]. The MCB code is a general-purpose tool dedicated for simulations of radiation transport and radiation-induced changes in matter. The code is developed at the Department of Nuclear Energy, Faculty of Energy and Fuels of the AGH University of Science and Technology, Krakow, Poland. The code integrates A General Monte Carlo N-Particle Transport Code (MCNP) for radiation transport simulations with the Transmutation Trajectory Analysis solver (TTA) for analysis of nuclide density time evolution [3,4]. The TTA approach allows to transfer numerical solution of the burn-up problem described by the Bateman equations into a linear probability problem described by the set of linear chains for each separate burn-up time step.

In typical burnup calculation, transmutation rates calculated in a particular time step are used for the calculation of a new material composition and they are no longer used, yet they are overwritten by the values of the following step. In the proposed methodology of trajectory periods folding, the mass flow of direct nuclide-to-nuclide transitions leading to nuclide transmutation chains in every step is interpreted over entire period of interest [5]. In this way, all quantitative information about the transmutation process for the period beyond single calculation step is preserved. The method builds sets of transmutation trajectories prepared for each computing time step and then combines them in the process of time period folding. Resulted period folded trajectories are interpreted as they would be obtained by the set of parameters from one calculation step. The trajectory periods folding methodology is an alternative to the matrix multiplication method. Both methods allow to obtain

individual nuclide transmutations.

The method was derived from the observation that it is possible to prolong single trajectories evolution by calculating the transition of trajectories obtained from two consecutive steps, where the trajectory from the second step starts with the same nuclide as the trajectory from the first step ends. It turns out that the appropriate sum of trajectories transitions corresponds to the matrix element obtained as a result of matrices multiplication. A set of folded trajectories obtained for the considered period can be used to present various dependencies in the analyzed system. Folded trajectories describe the relation between nuclides through different sequential reactions. The trajectory period folding method can be used in modeling of every nuclear system like e.g. nuclear reactors of each generations fueled with uranium and thorium. In the study we present the background of the method as well as its implementation for the modeling of chosen nuclear system.

2. Numerical Model

For the sake of the MCB modeling, the European Lead-Cooled Fast Reactor (ELFR) [6,7] engineering model was transferred to the 3D computational geometry of the numerical model of MCNP. A core barrel has been filled with fuel assemblies (FA) arranged in a hexagonal pattern. The decision was taken to use the 2-batch active core consists of three fuel zones split into 8 radial and 10 axial burn-up regions for detailed modeling of neutronic and following burn-up effects – see Figure 1. Each of the 427 fuel assemblies contains 169 fuel rods in a triangular lattice. The rods are immersed in liquid lead and bound by a wrapper made of T91 steel. The height of the active column is 1400 mm, the pitch fuel assemblies is 209 mm and the pitch between the fuel rods is 15 mm.

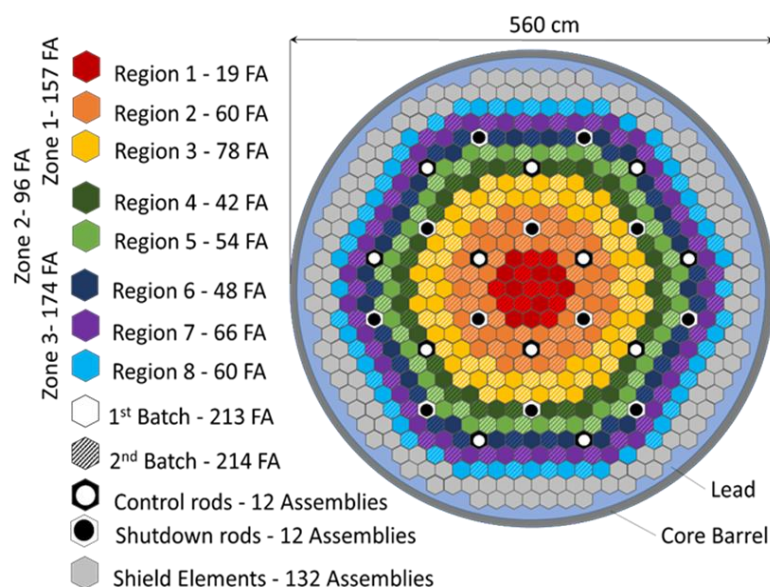


Figure 1. Horizontal cross-section of the numerical model.

In the two-batch refueling scheme of a 900-day (approx. 2.5 year) irradiation period cycles with 7.5-year cooling period was proposed. After cooling, fuel is reprocessed assuming mixing

the entire mass from all regions forming fuel batch to obtain a homogenized fuel composition, and then replacing the fission products by the fertile material containing ^{238}U [8,9]. The fuel composition obtained in this way is subjected to the adiabatic changes. The process is repeated for the following batches. The subsequent fuel compositions obtained by applying this schema are considered as a consecutive composition for the ELFR core operating under a two-batch reloading scheme. The result is that every batch is burnt twice and cooled thrice before reprocessing and reloading it into the core. Five batches make up the system inventory. Two of them are on load, while three are off load. As Beginning of Life (BOL) core must resort to physically available fuel material, the vector compositions of Light Water Reactor (LWR) spent fuel are used, which are shown in Table 1. As the cycle length is affected by breeding gain, we can adjust the desired plutonium enrichment to 18.15%. Minor Actinide (MA) composition was characteristic for LWR waste. In the previous studies [6,10], the solution of the extended equilibrium state was used to find MA contribution, where mass fraction was established at 1.347%.

Table 1. Initial fuel vector.

Nuclide	Vector fraction in heavy metal [wt.%]	Nuclide fraction in heavy metal [wt.%]
Uranium vector		
U233	80.50%	-
U234		0.002%
U235		0.325%
U236		0.008%
U238		80.17%
Plutonium vector		
Pu238	18.15%	0.423%
Pu239		10.32%
Pu240		4.9%
Pu241		110.8%
Pu242		1.396%
Pu244		-
MA vector (Np+Am+Cm)		
Np237	1.35%	0.051%
Am241		1.016%
Am242m		0.003%
Am243		0.218%
Cm242		-
Cm243		0.0009%
Cm244		0.041%
Cm245		0.016%
Cm246		0.0012%
Cm247		0.000023%
Cm248		0.000002%

3. Results and discussions

Figure 2 presents an example transmutation chain system obtained using the trajectory chain folding method. The considered chain system presents the transition channels from ^{238}U to ^{244}Cm after 124.2 years. A similar chain system may be obtained for an arbitrary couple of isotopes in the nuclear fuel. The chain system represents the batch mass as an integrated function of the evolving fuel vector for the entire operation and cooling period, including multi-cycle reloading, described in Section 2. Thus, the obtained results represent the cumulated trajectories for one large period of 124.2 years – not just for the last reactor cycle or off-core cooling. Figure 3. depicts the 12 tracked trajectories governing the transmutation of ^{238}U to ^{244}Cm . The difference in each individual transmutation in one channel depends on the transition through meta-stable americium isotopes i.e. $^{242}\text{Am}/^{242m}\text{Am}$ and $^{244}\text{Am}/^{244m}\text{Am}$ isotopes. Results show that the highest contribution from ^{238}U to ^{244}Cm comes through blue trajectories (A, B) - 63.72%. The contribution of remaining trajectories equals, red (C, D) 0.61%, green (E, F, G, H) 13.27% and orange (I, J, K, L) 22.40%, respectively.

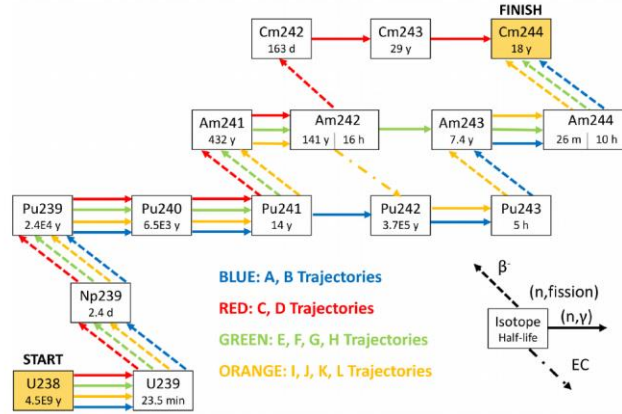
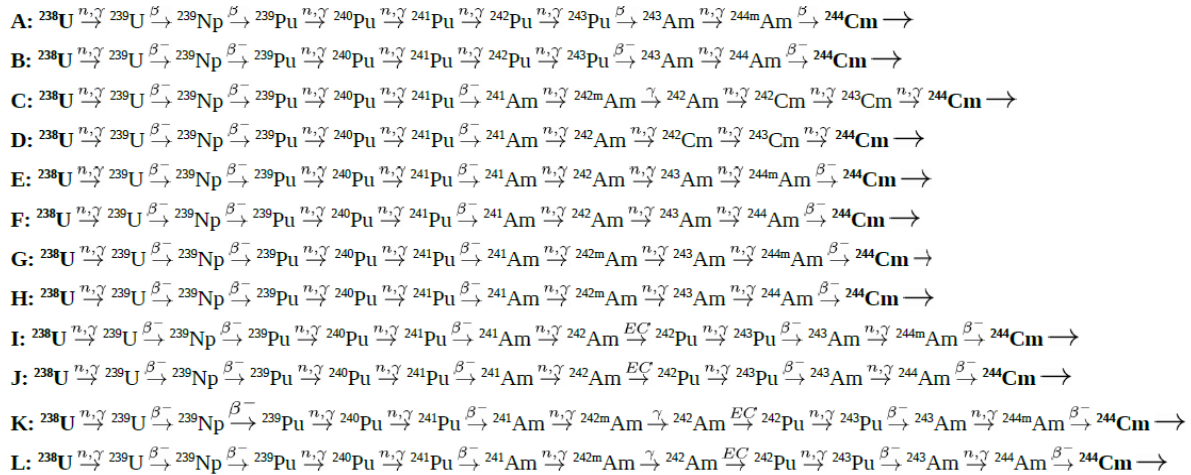


Figure 2. Transmutation chain system for ^{238}U - ^{244}Cm .



*EC: Electron capture

Figure 3. Trajectories governing the transmutation of ^{238}U to ^{244}Cm .

^{244}Cm is the component of initial fuel vector and last transmutation gateway to trans-amerium isotopes. It determines production of notably radioactive higher curium (e.g. ^{246}Cm , ^{248}Cm) and californium isotopes (e.g. ^{250}Cf , ^{251}Cf , ^{252}Cf). The latter are a significant source of neutrons because undergo spontaneous fission. Figure 4 shows source production of ^{244}Cm . It is mainly produced from ^{242}Pu through ^{243}Pu , ^{243}Am and $^{244}\text{Am}/^{244m}\text{Am}$. Thus, the production of ^{244}Cm is significantly influence by remaining gateway isotopes: ^{242}Pu and ^{243}Am . The initial mass flow of ^{244}Cm is governed by ^{243}Am . However, after about 25 years its mass is determined by ^{242}Pu because the residual ^{243}Am already transmuted to ^{244}Cm . ^{244}Cm decay with half-life time of 18.1 years in spontaneous fission and alpha decay to ^{240}Pu , thus its mass significantly decreases during off core cooling periods. ^{244}Cm mainly undergoes fission but also decays to ^{240}Pu . The mass of ^{244}Cm has increased almost three times from 10.9kg to 30.6kg due to its production from ^{242}Pu .

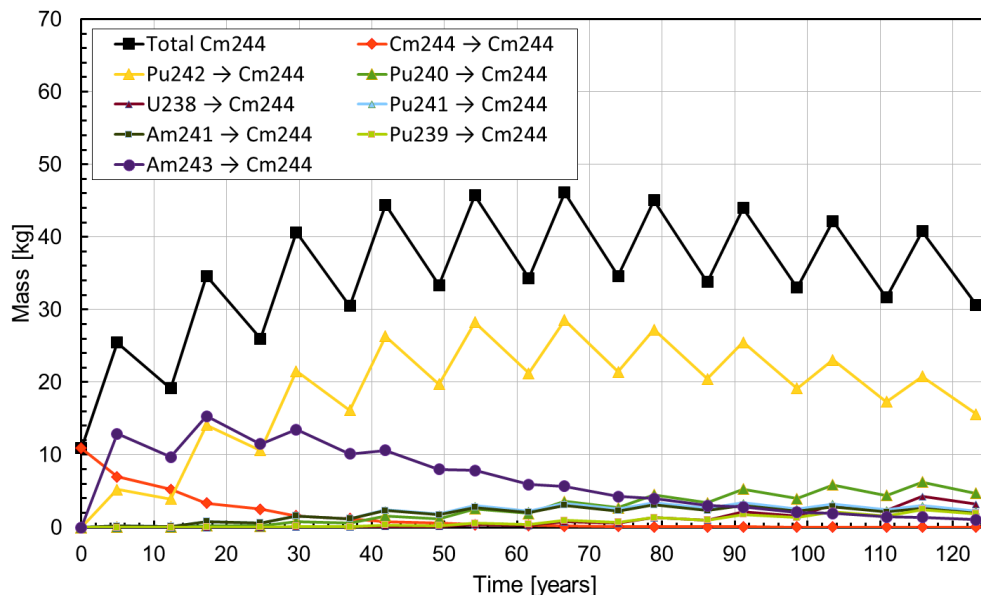


Figure 4. Batch mass flow of ^{244}Cm with source production of the initial fuel.

4. Conclusions

Thus, in the study we presented the new methodology of the trajectory periods folding for modeling of uranium and thorium fuel cycle. The new method allows us to trace the life cycle of chosen isotopes in any nuclear system. The method was applied to modeling of the ELFR in the adiabatic equilibrium state. The example folding of trajectories governing transmutation of ^{238}U to ^{244}Cm has been presented. In general, the study shows that:

- the standard solution for the time evolution of nuclide concentrations governed by a set of first-order differential equation may be used to represent the isotope mass evolution as an integrated function for the entire irradiation period including multi-cycling reloading,

- TTA and period folded trajectory method can indicate trajectories with the highest transition participating to the MA inventory,
- nuclear reactions belonging to chosen trajectories can be quantitatively characterized to allow for the identification of the most efficient one in production of crucial nuclides.

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Conflict of Interest

The authors declare that there are no conflicts of interest regarding this paper.

Author Contributions

The authors equally contributed to all steps of the paper preparation.

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