Monte Carlo methods for numerical simulations of the Lead-Cooled Fast Reactor

M Oettingen and P Stanisz

1AGH University of Science and Technology, Faculty of Energy and Fuels, Department of Nuclear Energy, Krakow, Poland

*E-mail: moettin@agh.edu.pl

Abstract. Closed nuclear fuel cycle is one of the most promising options for the efficient use of nuclear energy resources with full recycling of long-lived transuranic elements. However, it can be implemented only in fast breeder reactors of Generation IV. The paper shows our methodology applied in the analysis of the lead-cooled fast reactor equilibrium fuel cycle using the Continuous Energy Monte Carlo Burnup Code – MCB. The implementation of novel modules for nuclear transmutation trajectory folding allows us to trace the life cycle of crucial minor actinides from the beginning of the reactor’s life towards the state of adiabatic equilibrium. Changes in the mass contribution to gateway isotopes $^{242}$Pu, $^{241}$Am and $^{244}$Cm during 124.2 years were considered in the study. Numerical demonstration was performed for the reactor core designed within the European Lead-cooled System (ELSY) project and redefined in the follow-up Lead-cooled European Advanced Demonstration Reactor (LEADER) project.

1. Introduction

Nuclear power generation is strictly related to the production of used nuclear fuel containing notably radioactive transuranic elements. The global spent fuel inventory is growing year by year, causing potential long-term risk for the environment and the mankind. A long-term solution to this problem is the first requirement for further development of nuclear power technologies. The second requirement refers to the security of the nuclear fuel supply. Long-term operation of nuclear reactor systems may be assured only by the implementation of a reasonable strategy for nuclear fuel management. An integrated solution to these problems is not trivial and postulates the introduction of a closed-fuel cycle strategy based on fast breeder reactors. The perfect choice of a novel reactor system fulfilling both requirements is the Lead-Cooled Fast Reactor (LFR), operating in the adiabatic (self-breeder) state [1]. In such a state, the reactor converts depleted or natural uranium into plutonium, while consuming any self-generated Minor Actinides (MA) and transferring only fission products as waste. Therefore, it fulfils both of the aforementioned requirements and may be considered as a promising option to satisfy global energy demand in the long term.

The complexity of the burnup process requires the application of an integrated calculation system, which allows to consider spatial effects of a heterogeneous reactor core model with continuous energy representation of the cross section and thermo-hydraulic coupling. The integrated Continuous Energy Monte Carlo Burnup Code (MCB) [2], developed in the recent years, satisfies these requirements. The implementation of novel modules for nuclear transmutation trajectory folding enables to follow direct transmutation of the crucial MA in the adiabatic process from the beginning of the reactor’s life towards the equilibrium state. Therefore, the main goal of this study is the analysis of nuclear fuel depletion
under the fast neutron spectrum with the investigation of transmutation and decay chains using the novel method of nuclear trajectory folding. The implemented mathematical apparatus allows us to trace nuclear trajectories and identify the buildup contribution of the crucial MA during continuous fuel multireprocessing towards the adiabatic equilibrium state. Changes in the mass of gateway isotopes $^{242}\text{Pu}$, $^{243}\text{Am}$ and $^{244}\text{Cm}$ during 124.2 years were considered in the study. The gateway isotopes control the formation of higher actinides similarly as the transmutation of $^{238}\text{U}$ controls the breeding of $^{239}\text{Pu}$ [3].

2. Numerical tool
The neutron transport and burnup simulations were performed using the Continuous Energy Monte Carlo Burnup Code (MCB) equipped with JEFF3.2 nuclear data libraries. The MCB code is a general-purpose tool dedicated to simulations of radiation transport and radiation-induced changes in the matter. The code was 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 the General Monte Carlo N-Particle Transport Code (MCNP) for radiation transport simulations [4] with the Transmutation Trajectory Analysis solver (TTA) for nuclide density time evolution analysis [5]. It is a multi-purpose numerical tool dedicated to neutron transport and burnup simulations of advanced nuclear systems, including nuclear reactors of all generations. The code was implemented on the supercomputer Prometheus of the Academic Computer Center CYFRONET of the AGH University within PL GRID Infrastructure [6]. Prometheus is ranked 59th in the TOP500 list of the world’s fastest supercomputers (November 2016) and is currently the fastest Polish cluster for scientific calculations.

3. Method
The TTA approach allows to transfer the numerical solution of the burnup problem described by Bateman equations into a linear probability problem described by a set of linear chains for each separate burnup time step. The obtained transmutation chain trajectories between an arbitrary couple of nuclides can be used to form the transition probability matrix (Equation 1). In the TTA approach, the value of $\mathbf{P}(t_i)$ is represented by two fundamental functions – Transition and Passage [5], describing all chains above the predefined truncation parameter. Both functions control the integrity of the transmutation system and thus – the mass balance. In the study, for the first time, we take advantage of the fact that the obtained linear chains can be used to calculate joint trajectories for many subsequent time steps.

$$\vec{N}(t_i) = \mathbf{P}(t_i) \vec{N}(t_{i-1})$$

(1)

The methodology of trajectory folding is able to find the functions of Transition and Passage describing joint transmutation chains for many subsequent time steps. The folding procedure is controlled by the Passage function similarly as in the extension process. The Transition function allows to establish the mathematical relationship between the transmutation chains for multiple time steps. The process of trajectory extension is recursively repeated for each last nuclide in the previously defined trajectory. The trajectory chain is assumed to fold if the joint value is above the truncation parameter. The joint probability of the transmutation chain trajectories based on a sequence of direct nuclide-to-nuclide transitions for the folded period of time, starting from the first nuclide and ending with the $m$th nuclide ($a=1,m$), can be represented as one step, see Equation 2.

$$\frac{N_m(t)}{N_1(t_0)} = \sum_{a=1}^{m} \frac{N_a(t_1)}{N_1(t_0)} \cdot \frac{N_m(t_2)}{N_a(t_1)}$$

(2)
4. Numerical model
As a reference reactor model, we apply the core design developed initially under the framework of the European Lead-cooled System (ELSY) project [7] and refined in the follow-up Lead-cooled European Advanced Demonstration Reactor (LEADER) project [8]. The numerical model of the investigated LFR is presented in Figure 1. The reactor was numerically reconstructed as a lead cylinder with the height of 6100 mm and the diameter of 3500 mm, filled with 427 hexagonal fuel assemblies containing 169 fuel rods with the same equilibrium fuel composition—see Table I. The height of the fuel stack equals 1400 mm, the pitch of the fuel assemblies is 209 mm, the fuel pellet diameter is 9 mm and the pitch of the fuel rods is 15 mm. The 2-batch active core was divided into three fuel zones split into 8 radial and 10 axial burnup regions. The fuel assemblies are designed according to the New Paradigm for Nuclear Power [9]. They are immersed in liquid lead and separated by steel wrappers. The inner void in the fuel pellets was introduced to flatten the core power distribution. The diameter of the inner void in the fuel pellets equals 4 mm for zone one and 2 mm for zones two and three. The 132 shield assemblies containing 127 Y-stabilized zirconia rods surround the active core. The 12 shutdown and 12 control assemblies are located symmetrically in the reactor core. The assemblies were modelled as hexagonal wrappers, similarly as the shield assemblies, but filled with 127 B$_4$C absorber rods enriched to 90% $^{10}$B. The control rods are located below the active core and the shutdown rods are situated above it.

The beginning-of-cycle core is loaded with two fuel batches with the initial fuel composition of spent LWR fuel—see Table I. The fuel is initially irradiated during a sub-cycle of 900 days, and then the first batch is replaced with fresh equilibrium fuel, while the second one remains in the core for the next sub-cycle of 900 days. After the second sub-cycle, the second batch reaches a 1800 days’ in-pile residence time and is unloaded. Thus, each batch is twice burned and three times cooled before the reprocessing and reloading into the reactor core. In total, five batches are needed to make the system inventory, out of which two are on load, while three are off load at any point in time. The unloaded fuel is cooled for 7.5 years and then reprocessed. The reprocessing consists in mixing of the entire mass from all regions to obtain a homogenized fuel composition, and then replacing the fission products by fertile material containing $^{238}$U. The process is repeated for the subsequent batches [10]. The result is that every batch in the designed fuel cycle presents the capability of self-breeding. The investigated LFR operates on thermal power of 1500 MW$_{th}$ for ten reactor cycles and ten cooling periods, which gives 124.2 years of fuel operation in total (about 50 years of irradiation and 75 years of cooling).

| Isotope | BOL [g] | EOL [g] | Isotope | BOL [g] | EOL [g] | Isotope | BOL [g] | EOL [g] |
|---------|---------|---------|---------|---------|---------|---------|---------|---------|
| U       | 2.14E7  | 2.00E7/ | $^{240}$Pu | 1.30E6  | 1.69E6  | $^{242}$Cm | 0       | 3.77E1  |
| $^{234}$U | 6.42E2  | 5.71E4  | $^{241}$Pu | 2.94E5  | 1.18E5  | $^{243}$Cm | 2.39E2  | 5.35E2  |
| $^{235}$U | 8.64E4  | 2.23E4  | $^{242}$Pu | 3.71E5  | 2.07E5  | $^{244}$Cm | 1.09E4  | 3.06E4  |
| $^{236}$U | 2.14E3  | 3.57E4  | $^{244}$Pu | 0       | 3.19E0  | $^{245}$Cm | 4.14E3  | 1.06E4  |
| $^{238}$U | 2.13E7  | 1.99E7/ | Am      | 3.29E5  | 2.93E5  | $^{246}$Cm | 3.23E2  | 5.63E3  |
| $^{237}$Np | 1.36E4  | 2.65E4  | $^{241}$Am | 2.70E5  | 2.08E5  | $^{247}$Cm | 6.05E0  | 9.73E2  |
| Pu      | 4.82E6  | 4.80E6  | $^{242}$mAm | 9.11E2  | 1.55E4  | $^{248}$Cm | 4.93E-1 | 3.49E2  |
| $^{238}$Pu | 1.13E5  | 1.16E5  | $^{243}$Am | 5.79E4  | 6.96E4  | HM      | 2.66E7  | 2.52E7/ |
| $^{239}$Pu | 2.74E6  | 2.67E6  | Cm      | 1.53E4  | 4.87E4  |         |         |         |

BOL: Beginning of Life (fresh fuel); EOL: End of Life (after 124.2 years); HM: Heavy Metal; *before and after admixing of depleted uranium.
5. Mass evolution

The neutron capture on $^{238}\text{U}$ initiates the formation of transuranic isotopes in the following series of nuclear transmutation and radiative decays. Some of the newly-built isotopes playing the key role in the formation of MA were called the gateway isotopes. The gateway isotopes present a relatively high probability of nuclear transmutation to higher actinides in the fast neutron spectrum. Therefore, from the formal point of view, the gateway isotopes are of central importance in a given transmutation chain, so that its partitioning during the recycle results in a greatly reduced buildup of its transmutation daughters in the fuel cycle [3]. In this section, we present the mass evolution of three main gateway isotopes: $^{242}\text{Pu}$ as a gateway to americium, $^{243}\text{Am}$ as a gateway to curium, and $^{244}\text{Cm}$ as a gateway to berkelium and californium.

5.1. Plutonium – 242

The source production of $^{242}\text{Pu}$ is shown in Figure 2. $^{242}\text{Pu}$ originates mainly from the initial fuel vector; however, its production from $^{238}\text{U}$ and $^{240}\text{Pu}$ is remarkable. The source destruction of $^{242}\text{Pu}$ is presented in Figure 3. The main destruction channel of $^{242}\text{Pu}$ is its fission in the fast neutron spectrum, whose contribution increases with time. However, $^{242}\text{Pu}$ also transmutes to higher actinides, especially to the gateway isotopes $^{243}\text{Am}$ and $^{244}\text{Cm}$. It is also worth mentioning a relatively high contribution of (n,2n) and (n,3n) reactions on $^{242}\text{Pu}$, leading to the production of $^{240}\text{Pu}$. The 7.5 years long off-core cooling period does not influence the destruction of $^{242}\text{Pu}$ because of its long half-life time of 373 300 years. To sum up, the batch mass of $^{242}\text{Pu}$ is continuously decreasing cycle-to-cycle over the considered period of fuel operation from the initial 371 kg to 207 kg (by 56%).

![Figure 1. Reactor core.](image-url)
5.2. Americium – 243

$^{243}\text{Am}$ is considered as the gateway isotope determining the production of curium as well as heavier actinides and is a component of the initial fuel vector. It is mainly formed through beta decay of $^{243}\text{Pu}$.
with half-life time of 4.96 hours, which in practice is determined by $^{242}$Pu – see Figure 4. Only small amounts of $^{243}$Am are produced from $^{238}$U and $^{240}$Pu. The production from $^{241}$Am and $^{242m}$Am equals just a few grams and, in comparison with other production channels, is negligible. Figure 5 shows the source destruction of $^{243}$Am. The main destruction channel of $^{243}$Am is fission; however, some $^{243}$Am transmutes to $^{240}$Pu through $^{244}$Am/$^{244m}$Am and $^{244}$Cm. $^{243}$Am decays to $^{239}$Np with half-life time of 7370 years and thus the destruction of this isotope is determined rather by the neutron-induced in-core transmutation than the off-core radiative decay. The mass of $^{243}$Am at EOL equals 69.6 kg, which is about 17% higher compared to the initial mass of 57.9 kg. The main reason for the increase of $^{243}$Am is its production from $^{242}$Pu, which reaches the peak value at about 45 years of fuel operation.

![Figure 4](image1.png)

**Figure 4.** Batch mass flow of $^{243}$Am with source production of the initial fuel.

![Figure 5](image2.png)

**Figure 5.** Batch mass flow of $^{243}$Am with source destruction of the initial fuel.
5.3. Curium – 244

$^{244}$Cm is a component of the initial fuel vector and the last transmutation gateway to trans-amercurium isotopes. It determines the 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 they undergo spontaneous fission. Figure 6 shows the source production of $^{244}$Cm. It is mainly produced from $^{242}$Pu through $^{243}$Pu, $^{243}$Am and $^{244}$Am/$^{244m}$Am. Thus, the production of $^{244}$Cm is significantly influenced by the 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, it is determined by $^{242}$Pu, because the residual $^{243}$Am has already transmuted to $^{244}$Cm. $^{244}$Cm decays with half-life time of 18.1 years in spontaneous fission and alpha decay to $^{240}$Pu, thus its mass significantly decreases during the off-core cooling periods. Figure 7 presents the main source destruction of $^{244}$Cm. $^{244}$Cm mainly undergoes fission but also decays to $^{240}$Pu. The mass of $^{244}$Cm has increased almost three times from 10.9 kg to 30.6 kg due to its production from $^{242}$Pu.

![Figure 6. Batch mass flow of $^{244}$Cm with source production of the initial fuel.](image1)

![Figure 7. Batch mass flow of $^{244}$Cm with source destruction of the initial fuel.](image2)
6. Transmutation chain system

Figure 8 presents an example transmutation chain system obtained using the trajectory chain folding method. The considered chain system presents the transition channels from $^{238}\text{U}$ to $^{244}\text{Cm}$ after 124.2 years. A similar chain system may be obtained for an arbitrary couple of isotopes. 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. 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. This approach allows us to represent the multi-cycle problem of the fuel vector evolution as a one-step problem characterized by the final transmutation chain system. Figure 8 depicts the main four transmutation channels, each consisting of two or four trajectories, which in total gives 12 trajectories governing the transmutation of $^{238}\text{U}$ passing to $^{244}\text{Cm}$. The difference in each individual transmutation in one channel depends on the transition through $^{242}\text{Am}$/$^{242m}\text{Am}$ and $^{244}\text{Am}$/$^{244m}\text{Am}$ isotopes. The transmutation might follow either by the metastable state or the ground state of two americium isotopes, which increases the number of transmutation trajectories.

Figure 8. Trajectories for the chain transition from $^{238}\text{U}$ to $^{244}\text{Cm}$ at EOL.

7. Conclusions

In the study, we present the new methodology for the numerical analysis of LFR towards the adiabatic equilibrium state. The focal point is the implementation and practical application of the novel modules for nuclear transmutation trajectory folding to the MCB code. It allows us to trace the life cycle of gateway $^{242}\text{Pu}$, $^{243}\text{Am}$, $^{244}\text{Cm}$ and other isotopes. In addition, an example folding of trajectories governing the transmutation of $^{238}\text{U}$ to $^{244}\text{Cm}$ is presented. In general, the study shows that:

- the standard TTA Bateman solution for the subsequent time steps can be used to represent the integrated isotope mass evolution for the entire irradiation period including multi-cycle reloading actions,
- the identification of the key trajectories leading to the formation of transuranic elements is crucial for the understanding of the formation mechanism of notably radioactive isotopes, which in turn may help to optimize handling of unloaded fuel before its reprocessing or final disposal,
• the developed numerical method of nuclear transmutation trajectory folding may be used for the analysis of any critical or subcritical nuclear system during an arbitrary number of subsequent irradiation or cooling periods,
• the main transmutation pathways during a multi-cycle reloading scheme pass through the identified gateway isotopes,
• $^{242}$Pu present in the initial fuel vector is far from the equilibrium level and thus it is responsible for the peak in the concentrations of other gateway isotopes.

Acknowledgements

The research was partially supported by PL Grid Infrastructure available at the Academic Computer Centre CYFRONET AGH. In addition, partial financial support of this study under the grant agreement 16.16.210.476 by the Polish Ministry of Science and Higher Education is kindly acknowledged.

References

[1] Stanisz P, Oettingen M, Cetnar J 2016 Nucl Eng Des 301 341
[2] Oettingen M, Cetnar, Mirowski T 2015 Computer Science 16 329
[3] Bays S, Piet S, Pope M, Youinou G, Dumontier A, Hawn D 2009 INL/EXT-09-16857, Idaho Falls
[4] X5-Team 2003 LA-UR-03-1987, Los Alamos National Laboratory
[5] Cetnar J, 2006 Ann Nucl Energy 33 640
[6] PL-grid infrastructure [online], [access: 15-10-2019]. Available in World Wide Web: http://www.plgrid.pl/en
[7] European lead-cooled system (ELSY) [online], [access: 15-10-2019]. Available in World Wide Web: http://cordis.europa.eu/project/rcn/80053_en.html
[8] Lead-cooled european advanced demonstration reactor (LEADER) [online], [access: 15-10-2019]. Available in World Wide Web: http://cordis.europa.eu/project/rcn/96603_en.html
[9] Artioli C, Grasso G, Petrovich C 2010 Ann Nucl Energy 37 915
[10] Stannisz P, Cetnar J, Domanska G 2015 Nukleonika 60 581