

Transmutation Trajectory Analysis in the Modelling of LFR Fuel Cycle

M. Oettingen¹, P. Stanis¹, J. Cetnar¹, G. Kępisty¹

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

E-mail contact of main author: moettin@agh.edu.pl

Abstract. The closed nuclear fuel cycle belongs to the most promising options for an efficient usage of the 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 to the analysis of lead-cooled fast reactor equilibrium fuel cycle using the Continuous Energy Monte Carlo Burnup code – MCB. The implementation of novel modules for nuclear transmutation trajectories folding, allows us to trace the life cycle of crucial minor actinides from the beginning of reactor life towards the state of adiabatic equilibrium. The changes in mass contribution to gateway isotopes ²⁴²Pu, ²⁴³Am and ²⁴⁴Cm during 124.2 years have been considered in the study. The numerical demonstration has been 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.

Key Words: Transmutation dynamics, Equilibrium fuel cycle, LFR, Burnup chains trajectory folding

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 continuously growing year by year, causing a potential long-time risk for the environment and humankind. The long-term solution to this problem is the first requirement for the further development of nuclear power technologies. The second requirement refers to the security of the nuclear fuel supply. The long-term operation of nuclear reactor systems may be assured only by the implementation of a reasonable strategy for nuclear fuel management. The integrated solution to those 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. It therefore fulfils both of the aforementioned requirements and may be considered to be a promising option to satisfy global energy demand in the long term.

The complexity of burnup process 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 section and the thermo-hydraulic coupling. The integrated Continuous Energy Monte Carlo Burnup Code (MCB) [2] developed in recent years satisfies needed requirements. The implementation of novel modules for nuclear transmutation trajectories folding enables following direct transmutation of the crucial MA in the adiabatic process from the reactor beginning of life towards equilibrium state. Therefore, the main goal of this study is an analysis of the nuclear fuel depletion under fast neutrons spectrum with investigation of transmutation and decay chains using the novel method of nuclear trajectory folding. The implemented mathematical apparatus allows tracing nuclear

trajectories and identifying buildup contribution of crucial MA during continuous fuel multi-reprocessing towards adiabatic equilibrium state. The changes in mass of gateway isotopes ^{242}Pu , ^{243}Am and ^{244}Cm during 124.2 years have been considered in the study. The gateway isotopes control formation of higher actinides in similar way as transmutation of ^{238}U controls breeding of ^{239}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 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 [4] with the Transmutation Trajectory Analysis solver (TTA) for analysis of nuclide density time evolution [5]. The code is a multipurpose numerical tool dedicated for neutron transport and burnup simulations of advanced nuclear systems, including nuclear reactors of all generations. The code has been implemented on the supercomputer Prometheus of the Academic Computer Center CYFRONET of the AGH University within PL GRID Infrastructure [6]. The Prometheus is ranked 59th in the TOP500 list of world's fastest supercomputers (November 2016) and is currently the fastest polish cluster dedicated for scientific calculations.

3. Method

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

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

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 similar as in the extension process. The *Transition* function allows establishing mathematical relation between 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 at the m^{th} nuclide ($a = 1, m$) can be represented as a one step, see Eq. 2.

$$\frac{N_m(t)}{N_1(t_0)} = \sum_{a=1}^m \frac{N_a(t_1)}{N_1(t_0)} \cdot \frac{N_m(t_2)}{N_a(t_1)} \quad (2)$$

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 *FIG.1*. The reactor was numerically reconstructed as a lead cylinder of height 6100 mm and diameter 3500 mm filled with 427 hexagonal fuel assemblies containing 169 fuel rods having the same equilibrium fuel composition – see Table I. The height of the fuel stack equals 1400 mm, the pitch of fuel assemblies is 209 mm, the fuel pellet diameter is 9 mm and the pitch of 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 a steel wrappers. The inner void in the fuel pellets was introduced to flatten 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 were symmetrically located in the reactor core. The assemblies were modelled as hexagonal wrappers, similar to the shield assemblies but filled with 127 B₄C absorber rods enriched to 90% ¹⁰B. The control rods are located below the active core and the shutdown rods above it.

The beginning-of-cycle core is loaded with two fuel batches having initial fuel composition of spent LWR fuel – see Table I. The fuel is initially irradiated during the 900 day long sub-cycle, and then the first batch is replaced with the fresh equilibrium fuel while the second remains in the core for the next 900 day sub-cycle. After the second sub-cycle the second batch reaches an 1800 day in-pile residence time and is unloaded. Thus, each batch is twice burnt and three-times cooled before 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 assumes mixing the entire mass from all regions to obtain a homogenized fuel composition, and then replacing the fission products by fertile material containing ²³⁸U. The process is repeated for the following batches [10]. The result is that every batch in designed fuel cycle presents capability of self-breeding. The investigate LFR operates on thermal power 1500 MW_{th} for ten reactor cycles and ten cooling periods, which gives in total 124.2 years of fuel operation (about 50 years of irradiation and 75 years of cooling).

TABLE I: INITIAL AND FINAL BATCH FUEL COMPOSITION

Isotope	BOL [g]	EOL [g]	Isotope	BOL [g]	EOL [g]	Isotope	BOL [g]	EOL [g]
U	2.14E7	2.00E7/ 2.15E7*	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/ 2.14E7*	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	^{242m} Am	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		2.67E7*
BOL: Beginning of Life (fresh fuel); EOL: End of Life (after 124.2 years); HM: Heavy Metal; *before and after admixing of depleted uranium.							

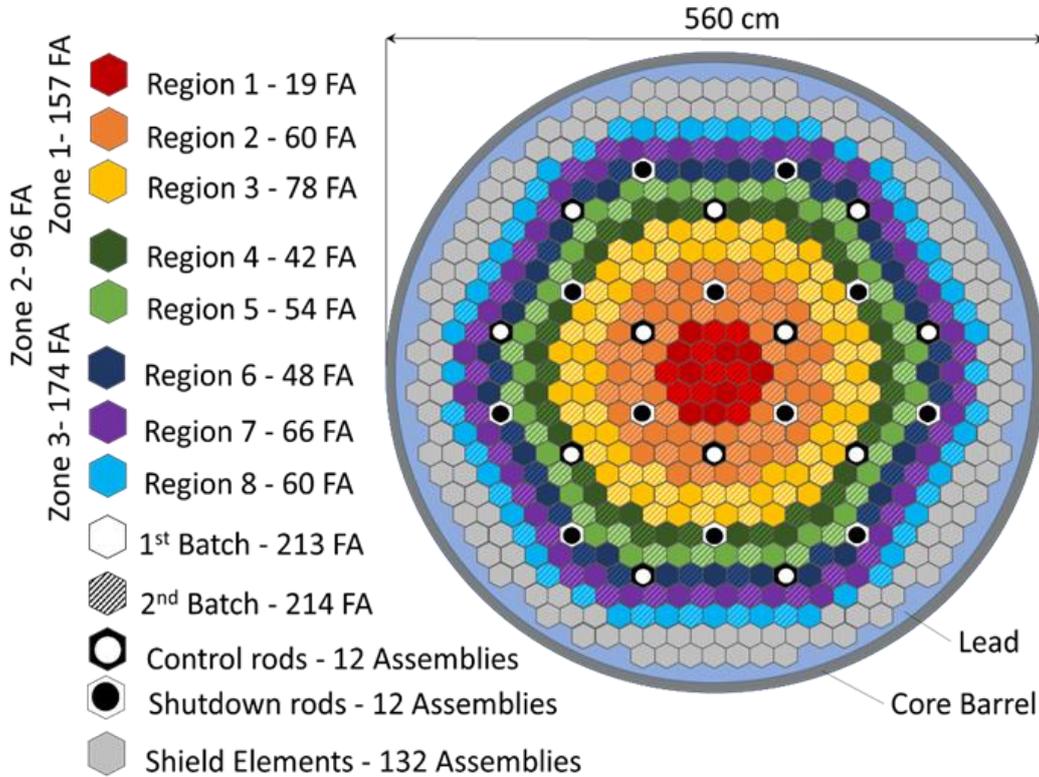


FIG. 1. Reactor core.

5. Mass evolution

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

5.1 Plutonium - 242

The source production of ^{242}Pu is shown in FIG. 2. The ^{242}Pu originates mainly from the initial fuel vector but its production from ^{238}U and ^{240}Pu is remarkable. The source destruction of ^{242}Pu is presented in FIG. 3. The main destruction channel of ^{242}Pu is its fission in the fast neutron spectrum, which contribution is increasing with time. However, ^{242}Pu also transmutes to the higher actinides, especially to gateway isotopes ^{243}Am and ^{244}Cm . It is also worth mentioning relatively high contribution of $(n,2n)$ and $(n,3n)$ reactions on ^{242}Pu following to

production of ^{240}Pu . The 7.5 years long off core cooling does not influence ^{242}Pu destruction because of its long half live time of 373300 years. To sum up, the batch mass of ^{242}Pu is continuously decreasing cycle to cycle over considered period of fuel operation from initial 371 kg to 207 kg (by 56%).

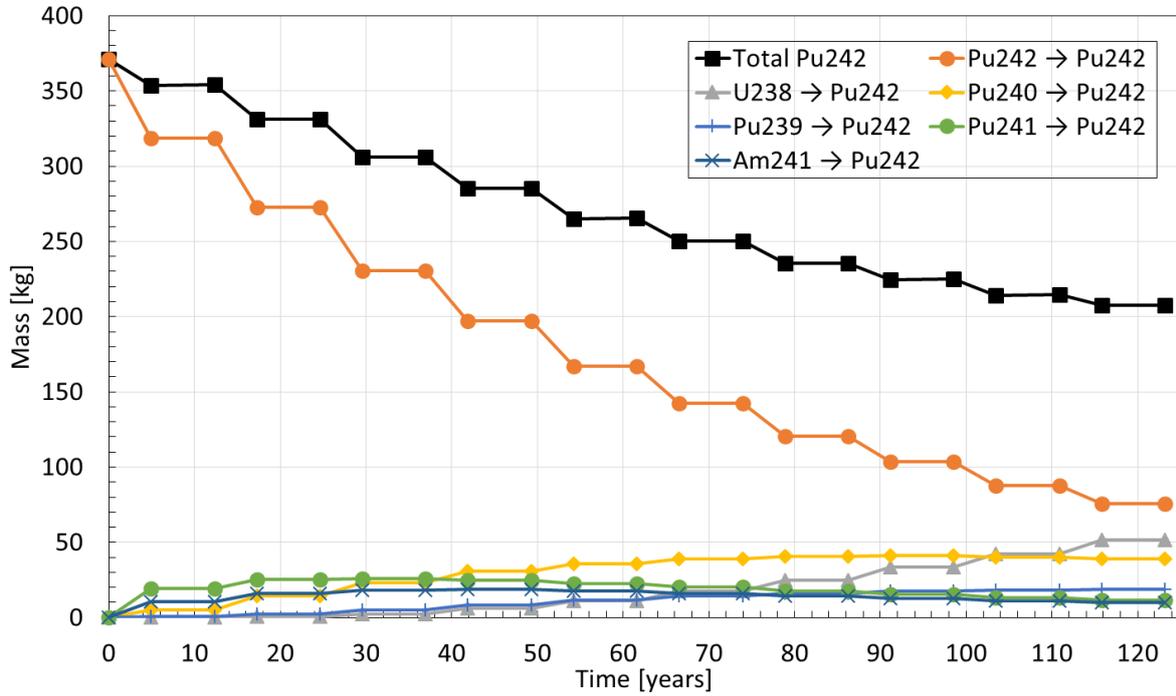


FIG. 2. Batch mass flow of ^{242}Pu with source production from the initial fuel.

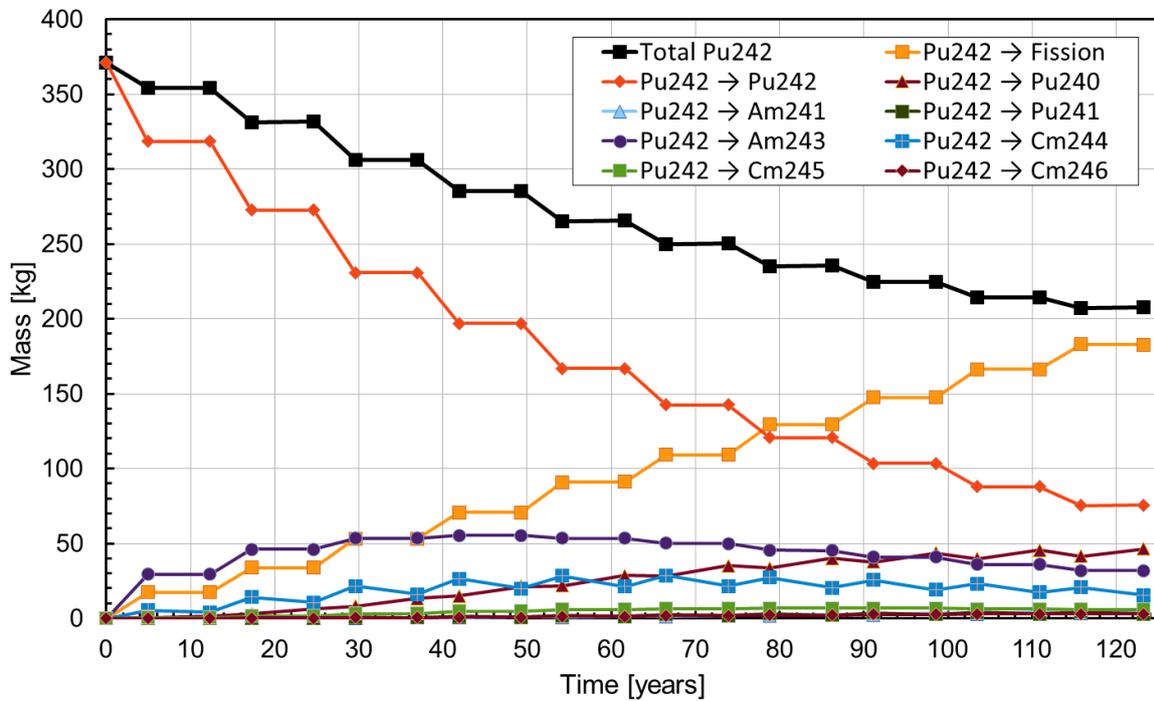


FIG. 3. Batch mass flow of ^{242}Pu with source destruction from the initial fuel.

5.2 Americium - 243

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

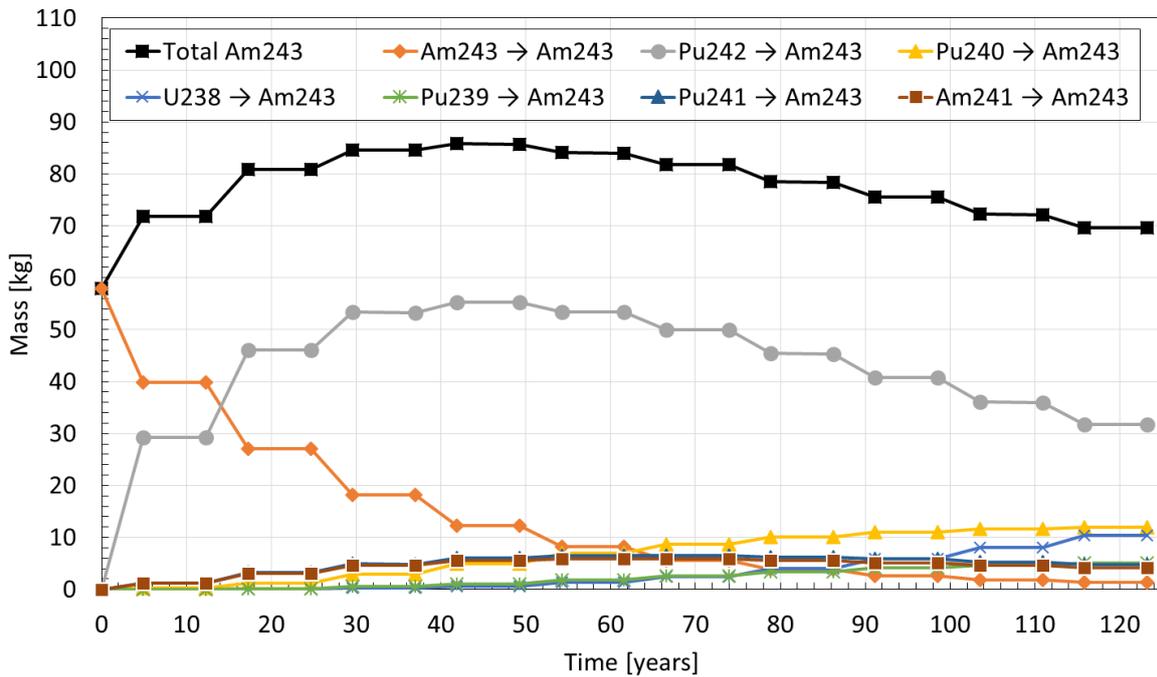


FIG. 4. Batch mass flow of ^{243}Am with source production of the initial fuel.

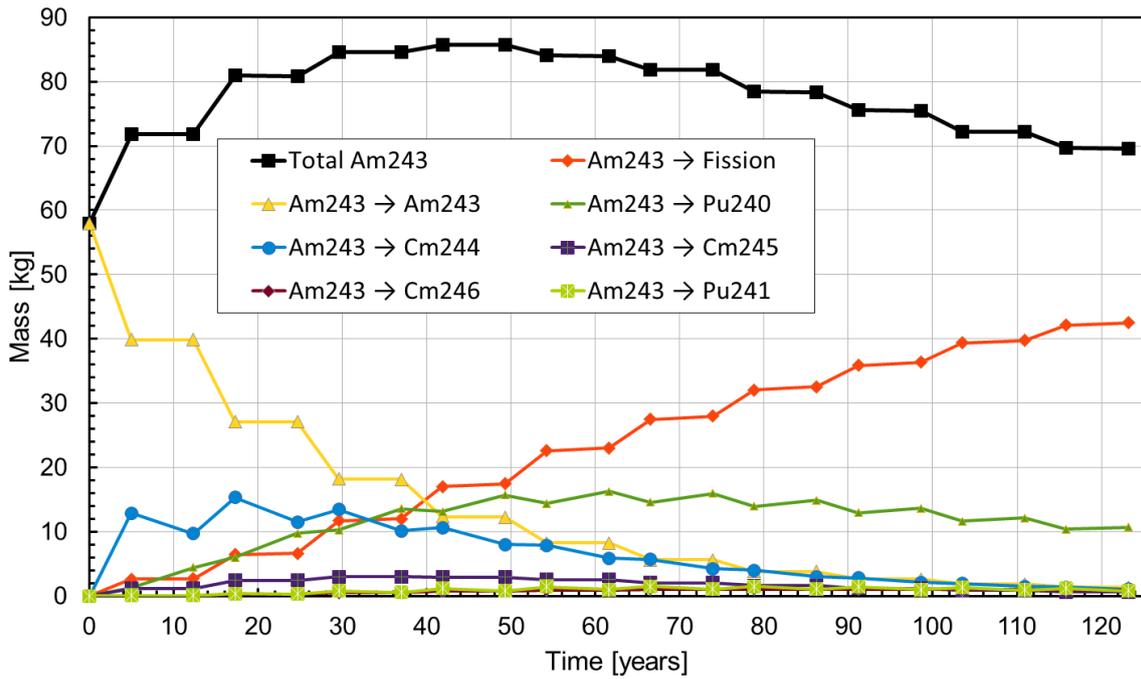


FIG. 5. Batch mass flow of ^{243}Am with source destruction from the initial fuel.

5.3 Curium - 244

^{244}Cm is the component of initial fuel vector and last transmutation gateway to trans-actinoid 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. FIG. 6. shows source production of ^{244}Cm . It is mainly produced from ^{242}Pu through ^{243}Pu , ^{243}Am and $^{244}\text{Am}/^{244\text{m}}\text{Am}$. 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. FIG. 7. presents 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 .

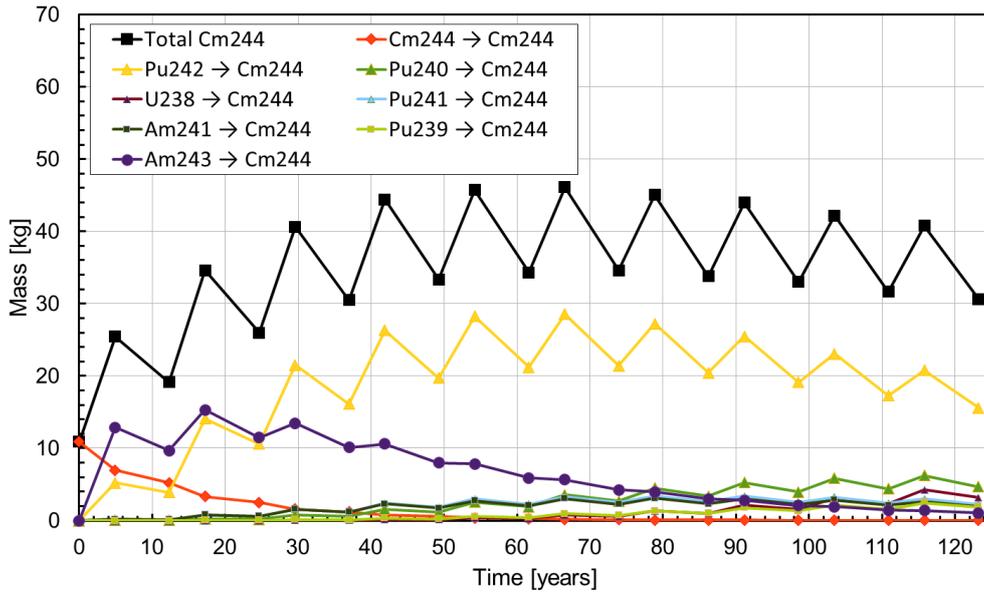


FIG. 6. Batch mass flow of ^{244}Cm with source production of the initial fuel.

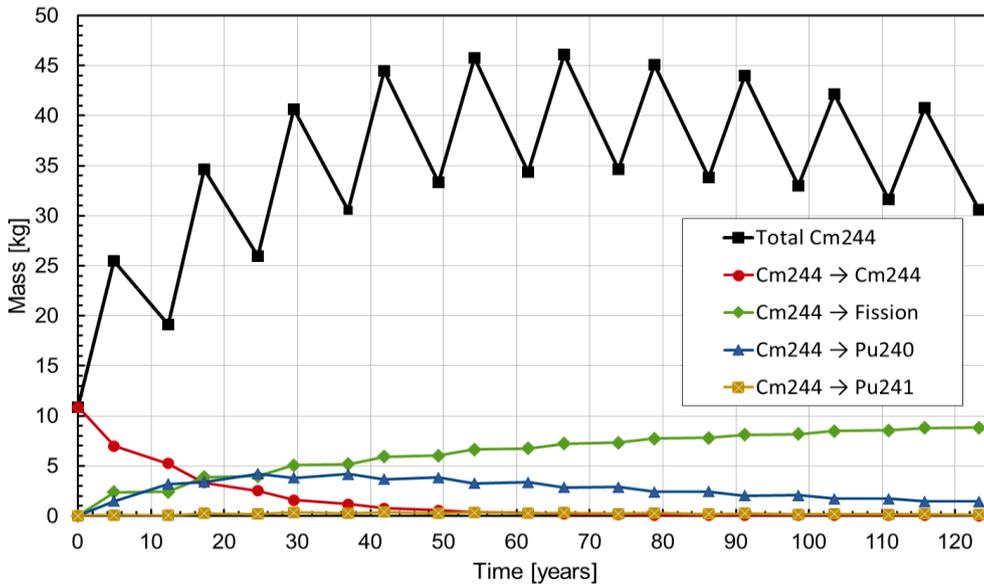


FIG. 7. Batch mass flow of ^{244}Cm with source destruction of the initial fuel.

6. Transmutation chain system

FIG. 8. presents the 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. The similar chain system may be obtained for the arbitrary couple of isotopes. The chains system represents the batch mass as an integrated function for the evolving fuel vector for the entire operation and cooling period including multi-cycle reloading. Thus, the obtained results represents the cumulated trajectories for one large period of 124.2 years – not just for 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. FIG. 8. depicts main four transmutation channels each consisting of two or four trajectories, which in total gives 12 trajectories governing transmutation of ^{238}U passing to ^{244}Cm . The difference in each individual transmutation in one channels depends on the transition through $^{242}\text{Am}/^{242\text{m}}\text{Am}$ and

$^{244}\text{Am}/^{244\text{m}}\text{Am}$ isotopes. The transmutation might follow either by metastable state or ground state of two americium isotopes, which increases the number of transmutation trajectories.

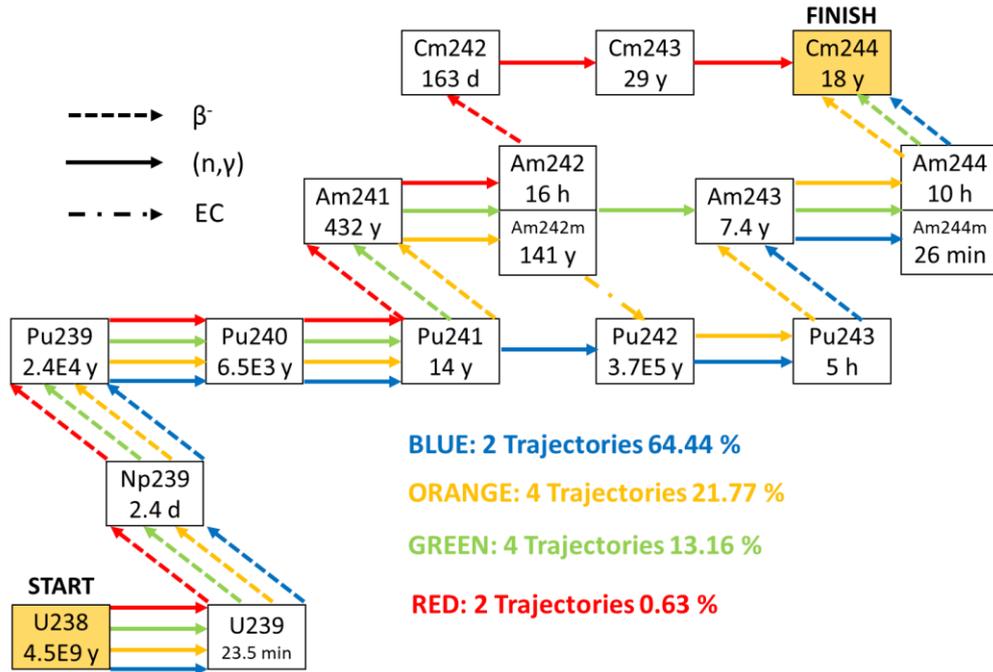


FIG. 8. Trajectories for chain transition from ^{238}U to ^{244}Cm at EOL.

7. Conclusions

In the study we presented the new methodology for the numerical analysis of LFR towards adiabatic equilibrium state. The focal point is the implementation and practical application of the novel modules for nuclear transmutation trajectories folding to the MCB code. It allows us to trace the life cycle of gateway ^{242}Pu , ^{243}Am , ^{244}Cm and other isotopes. In addition, the example folding of trajectories governing transmutation of ^{238}U to ^{244}Cm has been 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 key trajectories following to formation of transuranic elements is the crucial issues to understand 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 trajectories folding may be used for analysis of any critical or subcritical nuclear system during the arbitrary number of subsequent irradiation or cooling periods,
- main transmutation pathways during multi-cycle reloading scheme passes through the identified gateway isotopes,
- that ^{242}Pu present in the initial fuel vector is far from an equilibrium level and thus it is responsible for peak in concentrations of other gateway isotopes.

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References

- [1] STANISZ, P., OETTINGEN, M., CETNAR, J., “Monte Carlo modeling of Lead-Cooled Fast Reactor in adiabatic equilibrium state”, *NUCL ENG DES* **301** (2016) 341–352.
- [2] OETTINGEN, M., CETNAR, J., MIROWSKI, T., “The MCB code for numerical modeling of fourth generation nuclear reactors”, *COMPUTER SCIENCE* **16** (2015) 329-350.
- [3] BAYS, S., et. al., “Transmutation Dynamics: Impacts of Multi-Recycling on Fuel Cycle Performances”, INL/EXT-09-16857, Idaho Falls (2009).
- [4] X5-TEAM, “MCNP – A General Monte Carlo N-Particle Transport Code, Version 5, Volume I: Overview and Theory”, LA-UR-03-1987, Los Alamos National Laboratory (2003).
- [5] CETNAR, J., “General solution of Bateman equations for nuclear transmutations”, *ANN NUCL ENERGY* **33** (2006) 640-645.
- [6] PL-GRID INFRASTRUCTURE [online], [access: 08-01-2017]. Available in World Wide Web: <http://www.plgrid.pl/en>
- [7] EUROPEAN LEAD-COOLED SYSTEM (ELSY) [online], [access: 08-01-2017]. Available in World Wide Web: http://cordis.europa.eu/project/rcn/80053_en.html
- [8] LEAD-COOLED EUROPEAN ADVANCED DEMONSTRATION REACTOR (LEADER) [online], [access: 08-01-2017]. Available in World Wide Web: http://cordis.europa.eu/project/rcn/96603_en.html
- [9] ARTIOLI, C., GRASSO, G., PETROVICH, C., “A new paradigm for core design aimed at the sustainability of nuclear energy: The solution of the extended equilibrium state”, *ANN NUCL ENERGY* **37** (2010) 915-922.
- [10] STANISZ, P., CETNAR, J., DOMAŃSKA, G., “Modeling minor actinide multiple recycling in a lead-cooled fast reactor to demonstrate a fuel cycle without long-lived nuclear waste”, *NUKLEONIKA* **60** (2015) 581-590.