Fuel cycle studies on minor actinide burning in gas-cooled fast reactors

Máté Szieberth, Máté Halász, Sándor Fehér, Tibor Reiss
Budapest University of Technology and Economics,
Institute of Nuclear Techniques, Hungary

Abstract

The gas-cooled fast reactor (GFR) is a Generation-IV reactor type considered an alternative fast neutron reactor design aimed to improve the sustainability of nuclear energy by improving uranium utilsation efficiency. Besides fuel breeding, the especially fast neutron spectrum of the GFR also provides excellent opportunities for minor actinide (MA) burning. The development of the design of a 2 400 MW$_{th}$ full-scale and a 70 MW$_{th}$ demonstrator GFR (ALLEGRO) is ongoing in the EURATOM sponsored GoFastR Project.

The paper presents fuel cycle studies performed at the Budapest University of Technology and Economics (BME) for the assessment of the transmutational capabilities of the 2 400 MW$_{th}$ GFR design in the framework of the GoFastR Project.

A new approach was developed for the modelling of the reactor burn-up. Multi-dimensional regression method was used to determine one-group cross-sections as a function of the core composition based on the results of numerous (several thousand) core transport calculations with different isotopic compositions. The set of isotopic compositions for which the core calculations were performed had to cover the space of the possible core compositions with the considered recycling options. With the help of the generated composition-dependent cross-section functions, a fast and flexible burn-up calculation scheme was developed, which can be easily integrated into the fuel cycle simulations. The developed model was verified against detailed burn-up calculations with satisfactory results.

The burn-up model was applied for the analysis of fuel cycle scenarios including a mixed fleet of GFRs and conventional LWRs. The homogeneous recycling of minor actinides into the GFRs was considered and their MA consumption was investigated. The results confirm the expectation that GRFs can also be applied for burning of MAs produced in other reactors. Calculations were performed with different MA ratios in the GFR core and different ration GFRs in the reactor fleet in order to find an optimum.

The present work focuses on the analysis of GFRs, but the developed methodology for burn-up modelling can be applied to other reactor types, as well.

Introduction

Fast reactors have the unique ability to be sustainable by not only being able to generate their own fuel, but through being able to burn minor actinides to reduce the quantity and radiotoxicity of nuclear wastes. The latter ability enables fast reactors to not only burn the minor actinides produced by themselves but, in addition, the minor actinides arising from legacy wastes and thermal reactors in the nuclear park. The Generation-IV International Forum [1] has identified six systems which merit development to achieve the goals of sustainability, proliferation resistance, economics and improved safety. Of these six systems, three are fast reactors, the gas-cooled fast reactor (GFR) being one of these. The sustainability goal has been developed further within Europe through the establishment of the Sustainable Nuclear Energy Technology Platform (SNE-TP). As well as setting-out a vision for the development of sustainable nuclear energy within Europe, the SNE-TP has devised a Strategic Research Agenda (SRA) [2] that identifies the priorities for research through which this vision can be realised. In this context, sodium-cooled fast reactors have been identified as the near-term technology that would allow rapid deployment of fast reactors. The SRA also identifies the possibility that gas-cooled and the lead-cooled fast reactors could be deployed in the longer-term. Both of these technologies will be capable of operating at higher temperatures than the use of a liquid sodium coolant will allow. As such, high efficiency electricity generation and a wider range of non-electrical applications becomes possible, such as the generation of high quality process heat and the efficient mass production of hydrogen. In addition, the harder neutron spectrum improves the transmutation capabilities, allowing minor actinides to be destroyed more effectively.

The EURATOM sponsored GoFastR Project [3] [4] concentrates on the gas-cooled fast reactor (GFR). The design goals for GFR are ambitious, aiming for a core outlet temperature of around 850°C, a compact core with a power density of about 100 MW/m^3, a low enough plutonium inventory to allow wide deployment, a self-sustaining core in terms of plutonium consumption, and a proliferation resistant core by not using specific plutonium breeding elements.

The initial core design of the 2 400 MW thermal power GFR was provided by the CEA at the beginning of the project [5]. The core is composed of carbide fuel element cladded with ceramic composite SiC/SiC fibre material. The ceramic cladding is needed in order to reach high-operation temperature and withstand as high as 1 200°C cladding temperature during transients.

At the Institute of Nuclear Techniques of the Budapest University of Technology and Economics (BME) fuel cycle studies were performed to investigate the fuel breeding and minor actinide burning capabilities of the GFR2400. Fuel cycle studies concerning transmutation options pose two main challenges:

- The evaluation of the different transmutation options can be performed based on the detailed composition of the final waste, which requires the tracking of a wide range of isotopes in the fuel cycle and the determination of the accurate composition of the spent fuel.

- Minor actinide recycling options in transmutation fuel cycles also results in a wide range of possible isotopic compositions of the core, influencing the neutron spectrum and therefore the burn-up process.

Most scenario codes contain only cross-section sets at a few burn-up steps, which are not flexible enough for such analysis. On the other hand, detailed burn-up calculations are too time-consuming to be inserted in the simulation of the complete fuel cycle. Development of a quick and flexible burn-up model has been started at the BME in order to cope with these problems.

Calculational method

Mathematical principles of the burn-up model

The evolution of the isotopic composition in the core during the burn-up can be described by the well-known Bateman differential equation system, which represents a balance equation for the number density of each isotope:

$$\frac{dN_i}{dt} = \sum_{j \neq i} \left(\sigma_{j \to i} \Phi + f_{j \to i} \lambda_j \right) N_j - \left(\sigma_i \Phi + \lambda_i \right) N_i \, , \tag{1}$$

where N_i is the number density if isotope i in the core, $\sigma_{j \to i}$ is the microscopic cross-section of the reaction leading from isotope j to isotope i, σ_i is the total cross-section of the reactions consuming isotope i, $f_{j \to i}$ is the branching ratio of the decay form of isotope j leading to isotope i, λ_j is the decay constant of isotope j. Φ is the average one group neutron flux in the core, which can be determined from the power:

$$\Phi = \frac{P}{\sum_{i=1}^{n} E_{f,i} \sigma_{f,i} N_i + E_\gamma \sum_{i=1}^{n} \sigma_{\gamma,i} N_i} \, , \tag{2}$$

where P is the thermal power of the reactor, $E_{f,i}$ is the energy released in a fission of fissile isotope i (~200 MeV), $\sigma_{f,i}$ is the fission cross-section of isotope i, E_γ is the average energy released in the core in an (n,γ) reaction (~5 MeV) while $\sigma_{\gamma,i}$ is the (n,γ) cross-section of isotope i. Obviously, the above one-group cross-sections have to be generated with a proper weighting in order to reproduce the average reaction rates in the core. Combining the number densities into a single isotope vector N, the differential equation system (1) can be written in a matrix form:

$$\frac{d\overline{\mathbf{N}}}{dt} = \overline{\overline{\mathbf{A}}}_k \overline{\mathbf{N}} \, , \tag{3}$$

where the matrix A_k is composed of coefficients which are determined by the cross-sections and decay constants. Index k distinguishes between the coefficient matrices at different time steps of the burn-up, since the change in the isotopic composition results in the change of the spectrum and therefore also the one-group collapsed cross-sections. This is why the time consuming core calculations need to be repeated during the burn-up calculations or burn-up dependent cross-section sets have to be used. In order to develop a quick and flexible burn-up calculation method, we chose to describe the cross-section as a function of the isotopic composition based on fitting to the results of numerous core calculations.

The 15 most important actinide isotopes were considered for the description of the isotopic composition: [234-236,238]U, [237,239]Np, [238-242]Pu, [241,243]Am, [242,244]Cm. Due to the large number of variables, fitting of the cross-sections is feasible only with the Moore-Penrose pseudo inverse [6]. A limitation of this method is that it is only applicable for fitting of functions ($f(x)$) which are linear in their coefficients:

$$f(\overline{x}) = \sum_j a_j f_j(\overline{x}) \tag{4}$$

where $f_j(x)$ can be any function of x. Polynomials meet this condition and since preliminary investigations had shown that accuracy of the fit does not improve with the insertion of third order terms, the following polynomial was chosen:

$$\sigma(\overline{N}) = a_0 + \sum_{j=1}^{n} a_j N_j + \sum_{j=1}^{n}\sum_{k=j}^{n} a_{j,k} N_j N_k \tag{5}$$

(5) can be written in a vector form:

$$\sigma(\overline{N}) = \overline{N}'\overline{a} , \tag{6}$$

where the $n+n^2$ element of the extended isotope vector $\overline{N}'$ contains, besides the isotope vector $\overline{N}$ the cross-products in the last term of (5). If the rows of a matrix $\overline{\overline{N}}'$ contain the above extended isotope vectors $\overline{N}'$ of the different compositions and vector $\overline{\sigma}_C$ contains the corresponding calculated cross-sections, then the vector $\mathbf{a}$ of the fitting parameters a_j and $a_{j,k}$, can be determined by a least-square-fit based on the theorem of the Moore-Penrose pseudo inverse:

$$\overline{a} = \overline{\overline{N}}'^{+} \overline{\sigma}_C , \tag{7}$$

where the pseudo inverse $\overline{\overline{N}}'^{+}$ can be determined by definition as:

$$\overline{\overline{N}}'^{+} = \left(\overline{\overline{N}}'\overline{\overline{N}}'^{*}\right)^{-1} \overline{\overline{N}}'^{*} \tag{8}$$

By substituting (5) in (1) the differential equation system, (3) takes the following form:

$$\frac{d\overline{N}}{dt} = \overline{\overline{A}}(\overline{N})\overline{N} , \tag{9}$$

where matrix $\overline{\overline{A}}(\overline{N})$ is a function of isotope vector $\overline{N}$, since it contains the coefficients determined by the cross-sections depending on the isotopic composition. This is not a linear differential equation system any more, but its numerical integration is still fast enough to be integrated into a fuel cycle simulation.

Core calculations

In order to perform the least-square-fit in (7) the one group cross-sections have to be calculated for numerous different compositions with detailed core calculations. Due to the large number of fitted parameters practically several thousand calculations have to be performed, therefore a simplified core model had to be set up, which could provide the homogenized one-group cross-sections for the core.

In the case of the GFR-2400 the codes of the SCALE code system [7] were used for this purpose. First the elementary cell of the GFR-2400 was modelled, which consists of the ceramic fuel pin with the surrounding He coolant. A one-dimensional cylindrical model was built, where the height of the core was taken into account with buckling correction. The T-XSEC sequence produced the resonance self-shielded 238 groups cross-section set based on ENDF/B-VII data. The XSDRN-PM one-dimensional discrete ordinates code was applied to generate the cell weighted homogenised cross-sections for the elementary cell. The hexagonal fuel assembly was also modelled in a one-dimensional cylindrical geometry containing the homogenised fuel region, the ceramic assembly wall and the gap between the assemblies. Other core components (axial and radial reflector, rod followers and gas plenum) were homogenised in infinite homogenous medium approximation. The 3D model of the core was built from the homogenised fuel assemblies and other core components, as can be seen in Figure 2. Full core calculations

were performed by the KENO-VI multi-group Monte Carlo criticality code in 238 groups. The correctness of the homogenization techniques applied was checked against detailed Monte Carlo calculations.

More than 7 000 core calculations were performed with the above simplified core model assuming different isotopic compositions for the fuel. The actinide composition of the fuel was randomly sampled for the different calculations taking into account the following constraints:

- Pu number density in the fuel changed between ~14-22% of the total actinide number density. The ratio of the Pu content of the inner and outer core was kept at 0.8 which is also the case for the initial loading.

- MA number density in the fuel changed between 0-10% of the total actinide number density. The MA content of the inner and outer core was the same, allowing ±5% random variation.

- The rest of the actinide content of the fuel was U.

- Isotopic composition of the heavy metal elements was also randomly distributed, but limits were set to every isotope considering the isotopic composition of the initial charged fuel and its change during burn-up.

The actinide reaction rates and the average flux in the inner and outer core and the k_{eff} were recorded from the calculations. The reaction rates are defined as:

$$R_i^j = \sigma_i^j \Phi^j N_i^j V^j ,$$ (10)

where index j refers to the inner or outer core region. Since the simplified burn-up model is a point model where the outer and the inner regions are not handled separately and cross-sections, number densities and flux are recorded only for the complete core, a weighted average cross-section needs to be calculated. The following weighting scheme was chosen:

Figure 1: KENO-VI model of the GFR-2400 core
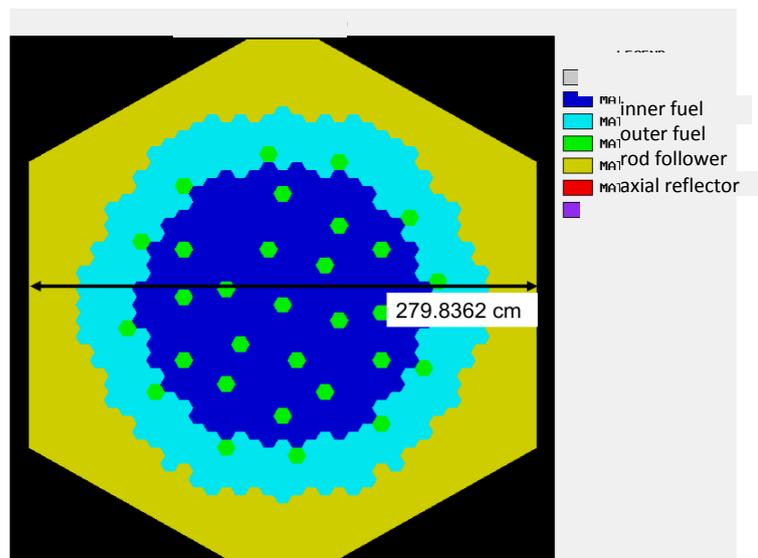
$$\overline{\sigma}_i = \frac{\sum\limits_{j} R_i^j}{\sum\limits_{j} \Phi^j N_i^j V^j} \tag{11}$$

which implies the use of a weighted average number density and average flux:

$$\overline{N}_i = \frac{\sum\limits_{j} \Phi^j N_i^j V^j}{\sum\limits_{j} \Phi^j V^j} \tag{12a}$$

$$\overline{\Phi} = \frac{\sum\limits_{j} \Phi^j V^j}{\sum\limits_{j} V^j} \tag{12b}$$

in order to preserve the total reaction rate:

$$\sum\limits_{j} R_i^j = \overline{\sigma}_i \, \overline{\Phi} \overline{N}_i \, V_{core} \tag{13}$$

Fitting according to the above-mentioned Moore-Penrose method was performed for the weighted cross-sections in (11). Examples for the results of the fitting process can be seen in Figure 2. The results show that the fitted functions can reproduce the calculated cross-sections with a few percent of maximum deviation.

Figure 2: Results of the fitting compared to the calculated points as a function of ^{239}Pu nuclei density: a) ^{239}Pu fission cross-section, b) k_{eff}, c) flux ratio of the inner and outer core region F

a)   b)   c)
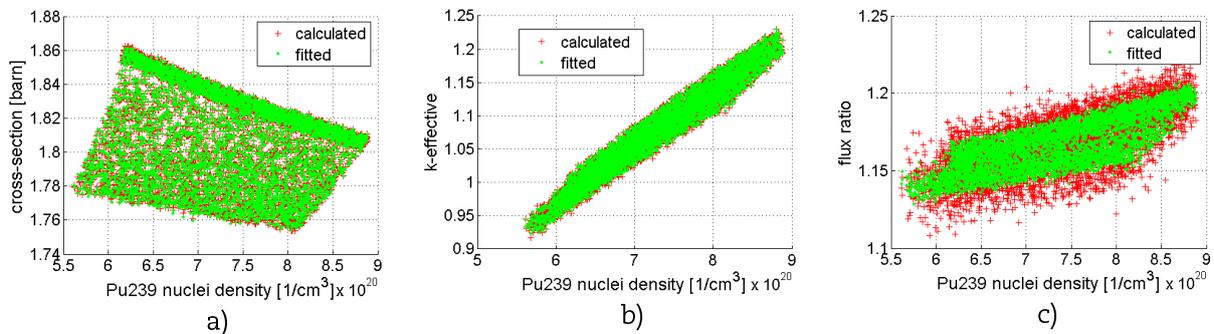
Burn-up model

The fitted cross-sections were applied in a burn-up model developed in MATLAB for the numerical solution of (9). Since in this burn-up model the flux and the number densities are calculated only for the complete core and not for the inner and outer region, the weighted average number density N_i cannot be calculated according to (12a). Instead, the fact is utilised that the ratio of the number density in the inner core and in the outer core is kept constant for all isotopes. The ratio of the flux in the inner and outer core changes with composition, but this dependence can be determined by applying the same fitting process as in the case of the cross-sections (see Figure 2.). If one defines number density ratio C and the flux ratio F:

$$C_i = \frac{N_i^{in}}{N_i^{in} + N_i^{out}} \tag{14}$$

$$F = \frac{\Phi^{in}}{\Phi^{out}} \tag{15}$$

then (13a) can be written as:

$$\overline{N}_i = \left(N_i^{in} + N_i^{out}\right)\frac{CFV^{in} + CV^{in} - V^{in}}{FV_{in} + V_{out}} \tag{16}$$

The full core calculations also determined the k_{eff} for the different composition. This gives the opportunity to also fit the k_{eff} as a function of the composition (see Figure 2.), which has the great advantage that in the fuel cycle model the fissile loading for the required excess reactivity can be determined.

The above-described burn-up model was used to build the simplified fuel cycle model shown in Figure 3. The model was developed in MATLAB and contained the GFR burn-up model and followed the material flows between reactors and storages. The fuel cycle contains two types of reactors: the GFR and conventional light water reactors (LWR). LWRs operate in once-through cycle: they are fed by 3.6% enriched U and the fuel is discharged after 33 MWday/kgU burn-up. The spent fuel was considered with the composition in Table I. No recycling into LWRs was considered but the spent fuel was moved to partitioning and the Pu and MA fractions were recycled into the GFR. GFR was fed with depleted U (as fertile material) produced during the enrichment of U for LWR fuel, and Pu and MA from the reprocessed LWR fuel. The reactor operates in three-batch cycle: one third of the core is discharged and replaced with fresh fuel in every cycle (481 EFPD) and each fuel element spends 3 cycles in the core. The Pu content of the charged fuel was calculated with iteration in order to set the excess reactivity of the beginning of cycle (BOC). The multiplication factor at the end of the cycle k_{eff}^{EOC} was expected to be 1.005, therefore the multiplication factor for the beginning of the next cycle $k_{eff}^{BOC'}$ was set based on the multiplication factor at the beginning of the actual cycle the following way:

$$k_{eff}^{BOC'} = k_{eff}^{BOC} - k_{eff}^{EOC} + 1.005 \tag{17}$$

Table 1: Considered LWR spent fuel composition after 33 MWday/kgU

MA isotope	Ratio	Pu isotope	Ratio
Np-237	0.489	Pu-238	0.027
Am-241	0.316	Pu-239	0.548
Am-242m	0.00105	Pu-240	0.234
Am-243	0.146	Pu-241	0.123
Cm-242	$9.6 \cdot 10^{-6}$	Pu-242	0.0678
Cm-243	0.000488		
Cm-244	0.0444		
Cm-245	0.0026	Component	Ratio
Cm-246	0.000474	Total Pu	0.00905
Cm-247	0.000010	Total MA	0.000673

MA were considered to be loaded homogenously into the core and different options were investigated concerning the MA content of the charged fuel. Fuel discharged form GFR was partitioned after 5 years of cooling and sent to the corresponding storage (U, Pu

or MA). The U, Pu and the MA need of the GFR was taken from the storage and depleted U or Pu and MA from LWR spent fuel was used only when the amount in the storage was not enough. The developed tool is capable of following the above fuel cycles for a long-term (hundreds of cycles) in a reasonable CPU time (a few hours), which is important to reach the equilibrium in the system.

In order to compare the investigated cases fuel utilisation efficiency was defined in the following way. The total thermal power generated in the system is the thermal power of the GFRs plus the thermal power of the LWRs producing the required Pu amount for the GFR load. The fuel utilisation efficiency is the ratio of the total thermal power to the theoretical energy content of the natural U (1 000 MWday/kg) consumed to produce enriched U fuel for the LWR. The fuel utilisation efficiency of an LWR with the above parameters operating in once-though cycle is 0.44%.

Figure 3: The considered fuel cycle model, Pu and MA from LWR is fed to GFR only when the required amount is not present in the corresponding storage
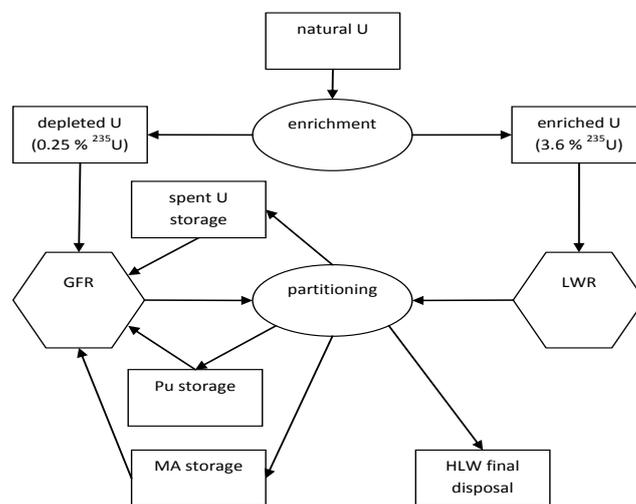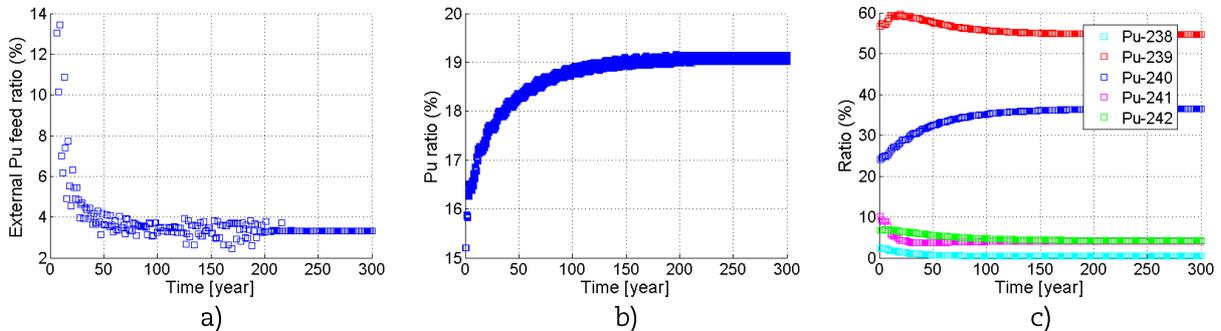

Results

All simulations were continued until an equilibrium was reached in the system. Although the time needed to reach an equilibrium was too long to be realistic in a fuel cycle simulation (several hundreds of years), but the investigation of the equilibrium state of the system is important for its evaluation.

The first investigated option assumed only Pu recycling into the GFR in order to provide a reference case for further investigations. The simulation shows that Pu feed from LWRs quickly decreases and in equilibrium it is only 3.3% of the total Pu feed to GFR. This shows that the GFR is close to self-breeding. On the other hand, the production of this amount of Pu still need more than 30% of LWR thermal power ratio in the reactor park, which results in a relatively low fuel utilisation efficiency (~1.4%), which is only three times higher than in the LWR once-through cycle. The initial 16% Pu content of the core increases to 19% due to the pile-up of ^{240}Pu and the decrease of the fissile ^{241}Pu, which is typical of fast reactors (see Figure 4.).

Figure 4a-c: Results for the case without MA recycling: a) external feed of Pu, b) Pu content of the core, c) isotopic composition of Pu. Increase of the Pu concentration is due to the accumulation of ^{240}Pu

a)   b)   c)

The next case was the recycling of Pu and MA into GFR but without adding MA from LWRs. An important question in this case is whether an equilibrium can be reached and at what MA concentration. The results show that an equilibrium is reached at 2.36% of MA concentration in the core. Isotopic compositions also reach an equilibrium, which means that all MA is consumed by fission and no Cm accumulation occurs due to MA recycling (see Figure 5.). This proves that the GFR can be applied as a MA burner. The MA content of the core does not significantly deteriorate the conversion capabilities of the reactor. The slightly higher need for external Pu feed is due to the increased amount of ^{238}Pu in the equilibrium Pu composition, which is produced from ^{237}Np.

Figure 5a-c: Results for the case with MA self-recycling: a) MA composition, b) Am composition, c) Cm composition

a)   b)   c)
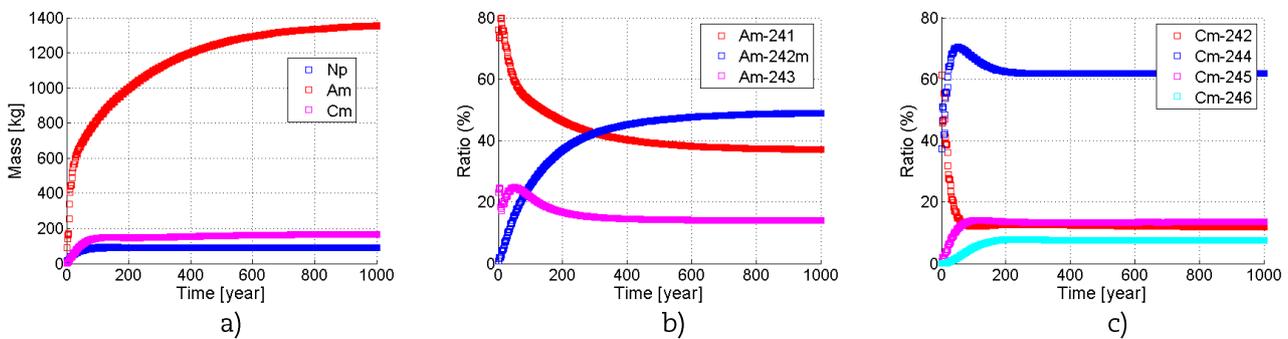
All isotopes reach an equilibrium, which means that GFR is a net MA burner.

The MA equilibrium concentration of 2.36% suggests that a higher MA concentration in the core also allows external feed of MA and the GFR turns into a net MA burner. In order to check this, statement simulations were performed with the assumption of a fixed ratio of MA in the charged fuel varying from 0.5% to 3%. As was expected with 0.5-2% of MA ratio, the need for external MA feed diminishes as an equilibrium is approached while with 2.5% and 3% MA a significant external MA load stabilises (see Figure 6.). The external feed to 3% MA load to the GFR is produced by more LWR than the required Pu external feed (which requires ~35% of the thermal power to be produced by LWRs in the system, see Table 2.). This means that a symbiotic nuclear energy system can be set up where the LWRs produces the Pu required for the operation of the GFR by the utilisation of enriched U, while the GFRs burn the MA produced by the LWRs. In this way the MA output of the system reduces the losses during the partitioning process. It is

also worth noting that in the short-term the increased MA feed improves the fuel utilisation. In the 3% MA feed case there is a time period where the external Pu feed decreases to zero which increases the fuel utilisation efficiency to almost 80%. But as the isotopic composition reaches an equilibrium, the Pu feed becomes proportional to the MA feed.

Figure 6a-c: Comparison of results for fuel cycle simulations with 2, 2.5 and 3 % MA feed: a) external MA feed, b) external Pu feed, c) fuel utilization efficiency (moving average)
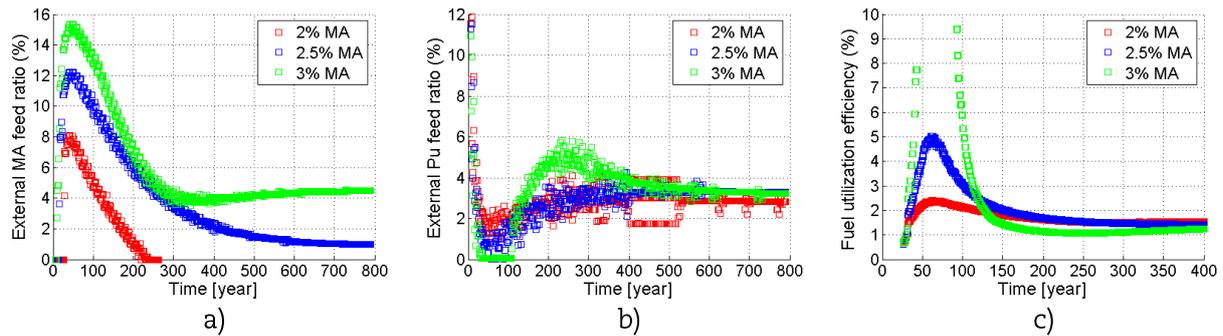

a)   b)   c)

Table 2: Fuel utilisation and MA burning parameters of different scenarios in equilibrium (%)

Case	Fuel utilisation	External Pu feed ratio	External MA feed ratio	LWR power ratio producing	
				Pu feed	MA feed
Pu recycling	1.39	3.30	-	31.85	-
MA self-recycling	1.35	3.38	-	32.89	-
2.5% MA feed	1.37	3.31	0.97	32.37	28.97
3% MA feed	1.27	3.68	3.75	34.93	65.43

Conclusions

A quick and flexible burn-up model was successfully developed based on the fitting of cross-sections and other parameters as a function of the isotopic composition. A model was integrated into a nuclear fuel cycle scheme containing conventional LWRs and GFRs. The fuel utilisation and MA burning capabilities of such a system were investigated.

It was found that the GFR is close to self-breeding and only about 3% of its Pu load needs to be fed from LWR spent fuel. In the case of MA recycling into the GFR, the MA content in the GFR core reaches an equilibrium at about 2.36%. If more MA is fed into the GFR, an optimal value can be found where the GFR consumes both the Pu and the MA production of the LWR park.

Acknowledgements

References

[1] OECD/NEA (2010), The Generation-IV International Forum, http://www.gen-4.org.

[2] Sustainable Nuclear Energy Technology Platform, Strategic Research Agenda, May 2009, http://www.snetp.eu/www/snetp/images/stories/DocsAboutSNETP/SRA2009.pdf .

[3] European GFR public website, http://www.gofastr.org/.

[4] Richard Stainsby *et al.* (2011), The Generation-IV Gas-cooled Fast Reactor, *Proc. of International Congress on Advances in NPPs 2011*, 2-6 May 2011, Nice, France.

[5] P. Richard, Y. Péneliau, M. Zabiégo (2010), Reference GFR 2400 MWth core definition at start of GOFASTR, *Technical report*, CEA/DEN/CAD/DER/SESI/LC4G DO2 26/03/10 Version 0, Commissariat à l'Energie Atomique, Cadarache, France.

[6] R. Penrose (1955), A generalized inverse for matrices, *Proceedings of the Cambridge Philosophical Society* 51: 406–413 (1955).

[7] SCALE (2009), A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation, ORNL/TM-2005/39 Version 6 Vols. I-III, Oak Ridge National Laboratory, US.