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RECYCLING OF VVER MINOR ACTINIDES IN A GAS-COOLED FAST REACTOR

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ABSTRACT

The Gas-Cooled Fast Reactor (GCFR) is one of the six Generation IV reactor designs, which has the potential to efficiently consume Minor Actinides (MAs). In this study, the possibility of using the reference GFR600 design as a MA burner was examined, assuming an initial MA composition corresponding to that of VVER440 spent nuclear fuel (SNF). For the calculations the KENO-VI Monte Carlo code and the ORIGEN-S burnup code of the SCALE 5.1 system were used, applying a precise three-dimensional reactor model.

Burnup studies were performed for cores with different initial MA contents. Besides the reactivity swing, the fuel inventory and the delayed neutron fraction were calculated as a function of burnup. Multiple recycling of MAs was also examined, assuming two recycling strategies, furthermore a comparison was made between the results of using MAs and Pu from LWR and VVER spent fuel.

It was concluded that the isotopic composition of the Pu has a strong effect on the reactivity. The loss of reactivity is significantly larger when using Pu from VVER SNF, and the reactivity swing does not turn positive when MAs from VVER SNF are added to the fuel. The MA burning capability of the reactor is to a large extent determined by the composition of the MAs and the Pu. Mainly Am is destroyed when using MAs and Pu from LWR, and Np when using those from VVER spent fuel. However, the delayed neutron fraction shows only a slight dependence on the origin of Pu and MAs.

Key Words: Gas-Cooled Fast Reactor, fuel cycle, three-dimensional burnup calculation, VVER spent fuel recycling

1. INTRODUCTION

The Generation IV International Forum, in order to develop the next generation of nuclear reactors identified six reactor concepts for further studies. Each has its own weaknesses and strengths and all of them are to meet the increased requirements in sustainability, economics, safety and proliferation resistance. One of the designs is the Gas-Cooled Fast Reactor (GCFR), which excels in the field of sustainability, having a self-breeder core (thus minimizing the need of fresh fissile material), full actinide recycling (hence reducing the quantity of discharged nuclear waste), and high coolant outlet temperature allowing high energy conversion efficiency[1]. From the different plans, the CEA designed GFR600 was chosen as a reference[2].

In the European GCFR-Specific Targeted Research Program (STREP) the GFR600 was examined by TU Delft from many points of view using a one-dimensional model of the so-called ”efficient” design[2] of the reactor (a good review of their work is given in [3], for more information see [4] and [5]). Among other issues, the studies focused on the potential of burning some Minor Actinides (MAs) arising from LWRs in order to assess the achievable reduction of MA stockpiles (being a result of advanced reprocessing methods used in the future to decrease the radiotoxicity of spent fuels going to repository). The behavior of cores containing MAs in the fuel (which would be the case due to the full actinide recycling envisioned) was investigated alongside this. For a benchmark calculation, a three-dimensional model in MCNP4B was also built and used by CIRTEN along with MONTEBURNS[4], but few other three-dimensional burnup studies are known of, and none regarding VVER spent fuel.

Hence research was done to see how the GCFR would work with VVER440 spent fuel, of which Pu (used in the fabrication of the startup fuel of the GFR600) and MA isotopic composition is different from that of LWR spent fuel. For the calculations the TRITON6 module of SCALE 5.1 was used with a geometrically precise three-dimensional model of the GFR600 built in KENO-VI. The results showed a significant difference in the change of reactivity in the first cycles between the LWR and the VVER case, and the change of MA elements during burnup was also different.

The structure of the paper is as follows. In Section 2 the KENO-VI model of the GFR600 is presented, followed by details of the calculations in Section 3. In Section 4 and Section 5 the results for the first cycle and for multiple cycles with different recycling strategies are described respectively.

2. The KENO-VI model of the GFR600

In SCALE 5.1 the TRITON6 module was introduced, which couples the traditional burnup module ORIGEN-S with the KENO-VI Monte Carlo criticality module, making it possible to perform precise three-dimensional burnup calculations[6]. In our study this module was used and an exact model of the GFR600 was built in KENO-VI (a KENO3D visualization can be seen in Figure 1). The main parameters and the geometrical description of this reactor concept can be found in many articles (e.g. in [2]), here just a short overview is given (Table I).

The core consists of 112 hexagonal sub-assemblies (S/As) composing 6 annular rings around a central piece, each S/A containing 21 fuel plates (the 6 annular rings will be referred to as the 6 regions of the reactor, region 1 being the outermost ring and region 6 being the innermost one plus the central piece, colored red in Figure 1). Fissile material is in carbide form, the structural material is SiC (cladding of fuel plates, central restraining device of S/As, S/A wrapper), the reflectors are made of Zr_3Si_2. The reference fuel consists of 16% PuC (Pu arising from legacy SNF) and 84% UC (natural U). Other important aspects of the fuel can be seen in Table II. Cores containing MAs were also examined, in those cases the composition of the initial fuel was 16% PuC, $X\%$ MAC and $(84 - X)\%$ UC. The main goal of the research was to investigate how the GCFR would work with Pu and MAs arising from VVER440 SNF, so their isotopic composition is characteristic of that (shown in Table III along with the LWR SNF composition used in earlier studies). No detailed reflector concept has been designed, therefore both axial and radial reflectors are modeled as homogeneous mixtures of structural material, coolant and Zr_3Si_2 in correspondence with earlier studies (the volume fractions of the different components can be seen in Table IV).

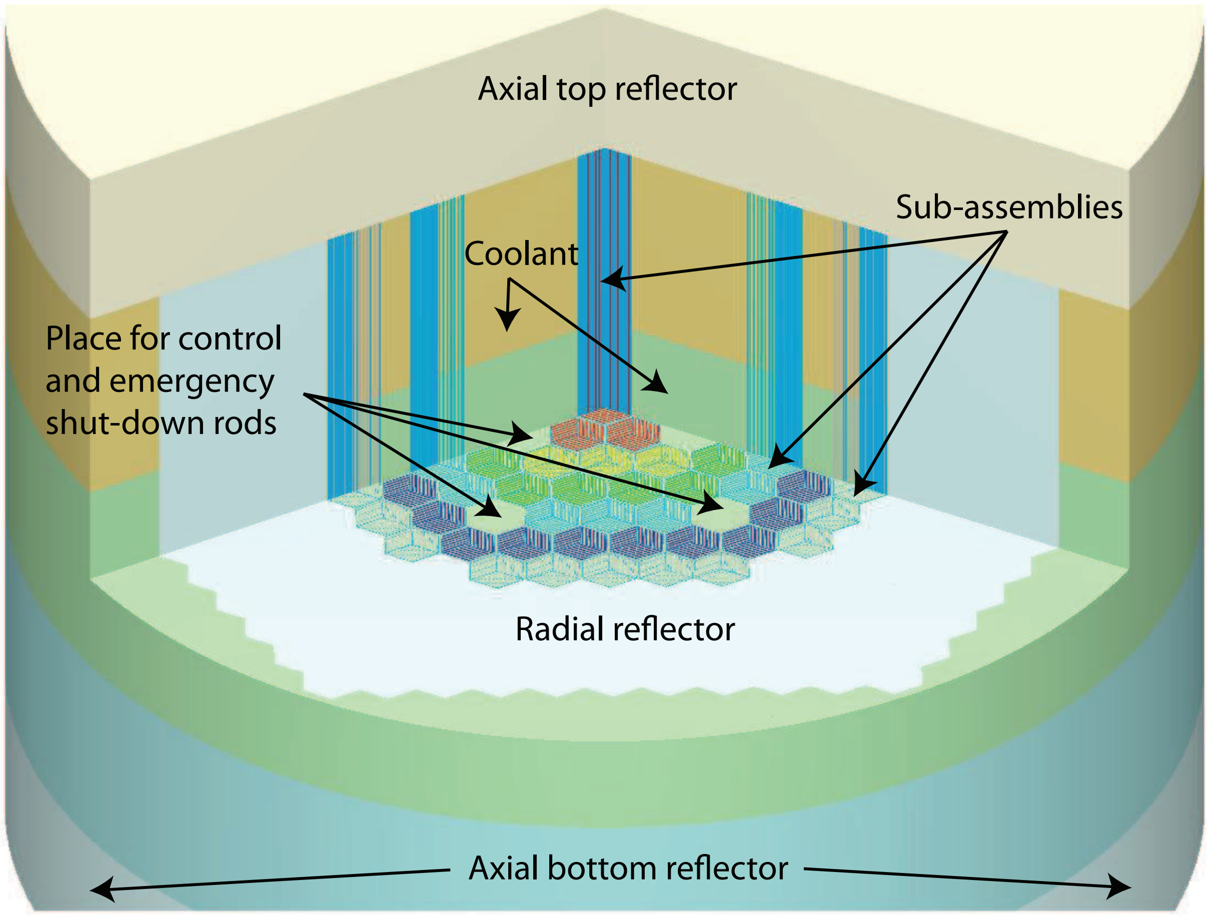

Figure 1. The model of GFR600 built in KENO-VI (the visualization was done by KENO3D)

Table I. The basic characteristics of GFR600

Core parameters		Sub-assembly parameters	
Thermal power	600 MW	Number of S/As in core	112
Power density	103 MW/m^3	Number of plates per S/A	21
Core height/diameter	1.95 m / 1.95 m	Fuel composition (UPuC/SiC)	70/30 V/V%
Coolant inlet/outlet temp.	490 °C / 850 °C	Fuel temperature	990 °C
Coolant material	Helium	Cladding material	SiC

[†] VVER spent fuel composition corresponds to 45GWd/MTU burnup and 5 years of cooling, LWR to twice recycled MOx fuel expected to be accessible from 2016 based on the 'Pu-2016' scenario study of CEA.

Table II. Some characteristics of the reference fuel of the GFR600

Fuel type	SiC vol. fraction	Fuel vol. fraction	SiC density	UPuC density	Fuel porosity
CERCER	30%	70%	3.21 g/cm^3	13.62 g/cm^3	15%

Table III. Isotopic composition of Pu and MA vectors characteristic of LWR and VVER440 spent fuel. †

Plutonium vector			MA vector		
Isotope	Fraction [n/n%]		Isotope	Fraction [n/n%]	
	LWR	VVER		LWR	VVER
Pu-238	2.7	2.71	Np-237	16.86	48.88
Pu-239	56	54.86	Am-241	60.64	31.56
Pu-240	25.9	23.38	Am-242m	0.23	0.11
Pu-241	7.4	12.27	Am-243	15.69	14.65
Pu-242	7.3	6.78	Cm-242	0.02	0.001
Am-241	0.7	0	Cm-243	0.07	0.05
			Cm-244	5.14	4.43
			Cm-245	1.25	0.26
			Cm-246	0.1	0.05

Table IV. The composition of the reflectors (volume fractions).

Material	Axial top reflector	Axial bottom and radial reflector
He coolant	40%	20%
SiC structure	10%	10%
Zr_3Si_2 reflector	50%	70%

For all calculations the 238 group ENDF/B-V library was used. Resonance self-shielding was done with BONAMI and NITAWL, using the Symmslabcell ‡ option with fuel/cladding/coolant ratios corresponding to the 35%/10%/55% volume ratios of the S/A design, and the fuel width being equal to the width of the fuel inside the plates. The temperatures of the fuel, the cladding

‡This option assumes an infinite array of fuel, cladding and coolant slabs.

and the reflectors are 990 °C, 665 °C and 565 °C respectively. Burnup modeling was done based on the planned burnup of the reference fuel: in each cycle a burnup period of 1300 efpd (effective full power day) was followed by 5 years of cooldown, when the fuel was allowed to decay before being reprocessed.

3. Analytical calculations

For a better understanding of the change of reactivity during burnup, the worths of the individual isotopes have to be investigated. In Section 3.1 the method used is explained, while in Section 3.2 the calculation of the important delayed neutron fraction in the reactor is described.

3.1. Worth calculations

Traditionally the reactivity weights $w_i = \overline{\nu_i \sigma_{f,i}} - \overline{\sigma_{a,i}}$ [7] are used to measure the influence of individual isotopes on the reactivity. These spectrum averaged values can be computed for each "i" isotope in each "k" region of the reactor at any "t" time $(w_{i,k}(t) = (\nu\sigma_f)_{i,k}(t) - (\sigma_a)_{i,k}(t))$ using the cross sections and fluxes calculated by KENO-VI in the 3-group structure required by the depletion module ORIGEN-S. From that we easily gain the appropriate one group values, e.g.:

$$(\sigma_a)_{i,k}(t) = \frac{(\sigma_a)_{i,k,therm}(t)\Phi_{k,therm}(t) + (\sigma_a)_{i,k,res}(t)\Phi_{k,res}(t) + (\sigma_a)_{i,k,fast}(t)\Phi_{k,fast}(t)}{\Phi_{k,therm}(t) + \Phi_{k,res}(t) + \Phi_{k,fast}(t)}. \quad (1)$$

The region dependent $w_{i,k}(t)$ values, which will be referred to as microscopic worths, show whether an isotope at a specific region contributes positively or negatively to the neutron number. The average behavior of an isotope is characterized by its average microscopic worth, defined as:

$$w_i(t) = \sum_{k=1}^{6} w_{i,k}(t) \frac{m_{i,k}(t)}{m_{i,Total}(t)}, \quad (2)$$

where m_i is the mass of the given isotope in the given region of the reactor or in the whole reactor. To take into account the quantity of the individual isotopes in the reactor, their average macroscopic worth is introduced (referred to as worth from now on):

$$W_i(t) = \sum_{k=1}^{6} W_{i,k}(t) \frac{V_k}{V_{core}} = \sum_{k=1}^{6} w_{i,k}(t) N_{i,k}(t) \frac{V_k}{V_{core}}, \quad (3)$$

where $N_{i,k}(t)$ is the number density of isotope "i" in region "k", V_k is the volume of the fuel in region "k" and V_{core} is the volume of the core.

The isotopes in the fuel can be divided into 3 groups: Fission Products (FPs), Heavy Metal isotopes of positive average microscopic worth (HMp isotopes) and Heavy Metal isotopes of negative average microscopic worth (HMn isotopes). Summing up the worths of all isotopes in the 3 groups we gain the fuel fissionability $(Fiss_{fuel}(t) = W_{FP}(t) + W_{HMp}(t) + W_{HMn}(t))$, and adding it to the fissionability of the cladding and the coolant we get to the fissionability of the core, which is a good measure of the reactivity[4]:

$$Fiss(t) = Fiss_{fuel}(t) + Fiss_{cladding}(t) + Fiss_{coolant}(t). \quad (4)$$

By analyzing the worths of different groups and the individual isotopes we will get a clear picture of what is happening in the reactor and why the reactivity is changing the way it is.

3.2. Delayed neutron calculations

The safety of a reactor is greatly influenced by the effective delayed neutron fraction. For such an estimation usually adjoint and forward solutions of the transport equation are both needed. Determining the fundamental delayed neutron fraction (the simple fraction of the number of delayed and total neutrons produced) is simpler, as it only requires the forward transport solution provided by KENO, and it also provides a powerful tool to analyze the safety of the reactor:

$$\beta(t) = \frac{\int\int \nu_D(E)\Sigma_f(\underline{r}, E, t)\Phi(\underline{r}, E, t)\mathrm{d}r^3 dE}{\int\int \nu(E)\Sigma_f(\underline{r}, E, t)\Phi(\underline{r}, E, t)\mathrm{d}r^3 dE} =$$

$$= \frac{\sum_i \sum_{k=1}^{6} \sum_j (\nu_D)_{i,j} N_{i,k}(t)(\sigma_f)_{i,k,j}(t)\Phi_{k,j}(t)}{\sum_i \sum_{k=1}^{6} \sum_j (\nu)_{i,j} N_{i,k}(t)(\sigma_f)_{i,k,j}(t)\Phi_{k,j}(t)} = \sum_i \beta_i(t), \quad (5)$$

where $(\nu_D)_{i,j}$ is the average number of delayed, while $(\nu)_{i,j}$ is the average total number of neutrons produced by the fission of isotope ”i”, induced by a neutron with an energy in group ”j”. Furthermore $\beta_i(t)$ is the fraction of delayed neutrons produced by isotope ”i” [§].

4. Results for the first cycle

First the effects of mixing VVER MAs into the fuel was examined during a time period of 1300 efpd. Cores having 0%, 3%, 5%, 8% and 10% initial MA content were investigated. The change of reactivity can be seen in Figure 2.

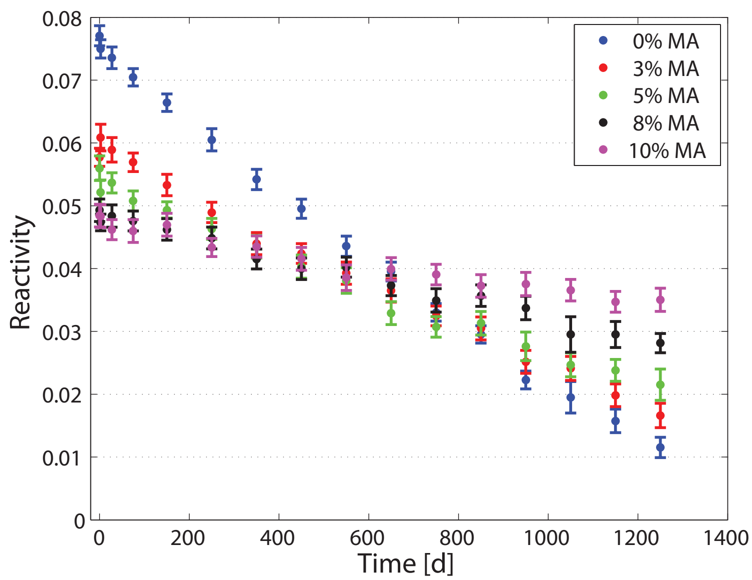

Figure 2. The reactivity change of cores with different initial VVER MA content during the first burnup period

[§]Note, that the V_k multiplication factors are missing due to the fact, that KENO-VI uses the KMART6 module to print fluxes, and the default normalizing factor is 1[6].

Similarly to the LWR case[8], the initial reactivity and the reactivity loss during burnup decreases as the MA content of the fuel increases (although at initial MA contents higher than 10% the initial reactivity stops decreasing and starts increasing). Two significant differences emerge between the use of MAs and Pu from LWR and VVER spent fuel:

- In the VVER case, the loss of reactivity is higher and the reactivity swing does not turn positive. This is caused by the more rapidly decreasing Pu, and the slower increasing Am-242m worth (both belong to HMp isotopes), as can be seen in Figure 3. The background of the smaller decrease of Pu worth in the LWR case is the smaller decrease of Pu-241 worth, while the more rapid growth of Am-242m worth can be explained by the higher Am-241 content of LWR spent fuel, from which highly fissile Am-242m is produced (Am-242m has the highest microscopic worth in the reactor).

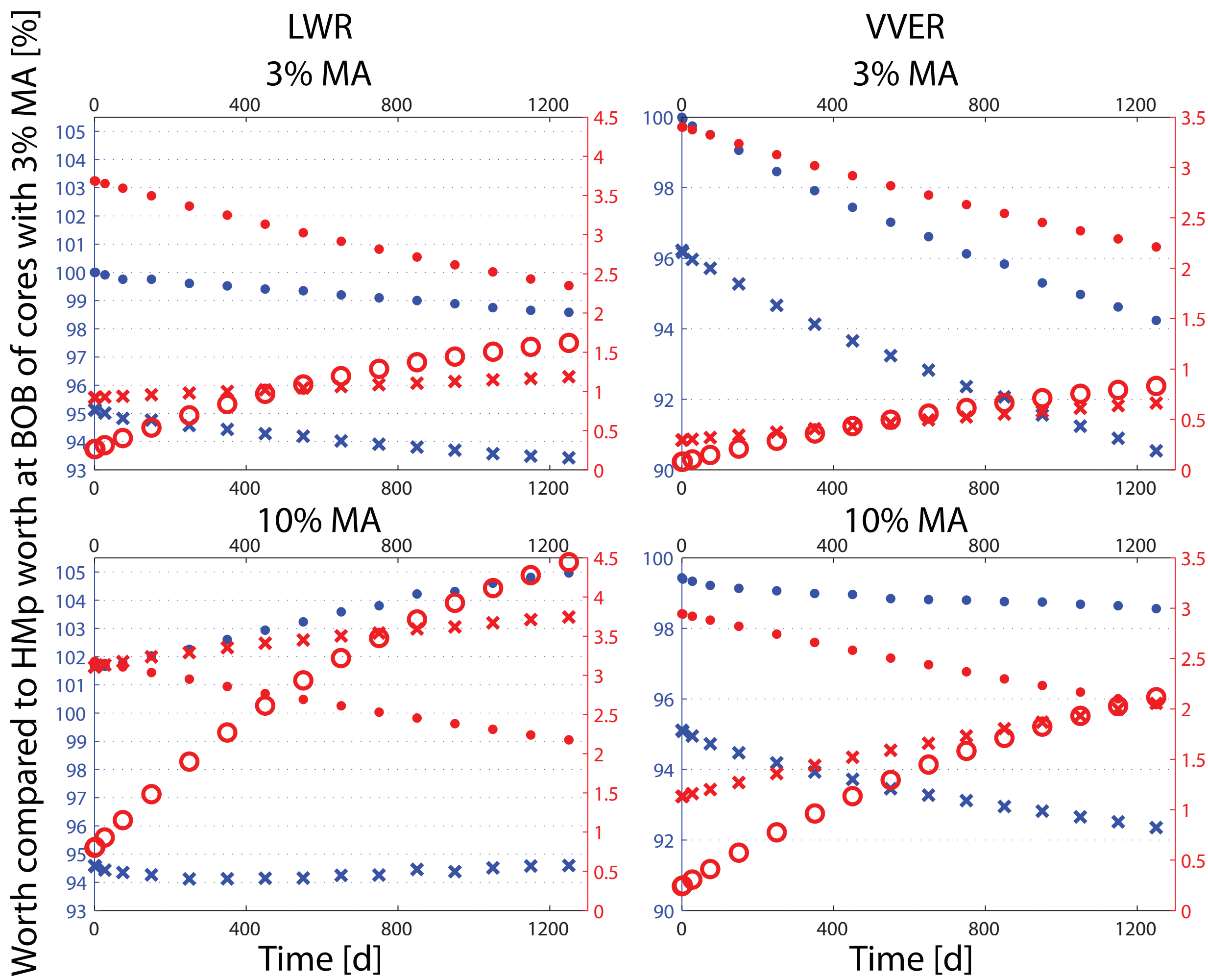

Figure 3. Breakdown of HMp worth of cores with different initial MA content and origin. The components of HMp worth are: Total (blue .), Pu (blue x), U-235 (red .), Cm (red x), Am-242m (red o). Blue axes on the left belong to the blue, red axes on the right to the red graphs.

- Secondly, the VVER case has a higher reactivity at the beginning of burnup (BOB) than the LWR, this is again a result of the higher Pu-241 content of VVER spent fuel. Note that the higher Pu-241 worth is mainly the result of the higher quantity, as its microscopic worth is virtually the same in the VVER and the LWR case. The result of the decreased reactivity loss during burnup and the higher initial reactivity is that though it is possible to lengthen the burnup period in the VVER case as well, at any given initial MA content the achievable burnup period is shorter than in the LWR case.

In Figure 4 we can see the change of the delayed neutron fraction of cores with different initial MA content during burnup and the spatial distribution of the delayed neutron production. The delayed neutron fraction decreases with the increasing MA loading of the fuel, and also during burnup, the latter being almost identical for all MA contents. Comparing the results to the ones obtained in the LWR case we see that the initial delayed neutron fraction is higher in the VVER case, mostly due to the higher Pu-241 and to a smaller extent the higher Np-237 content of the spent fuel (see in Figure 5). The delayed neutron fraction in the VVER case also has a more rapid decrease during burnup, mainly caused by the more rapid decline of Pu-241 content. Still, for the same burnup the VVER case always has a higher delayed neutron fraction than the LWR, hence it is more attractive from the safety point of view.

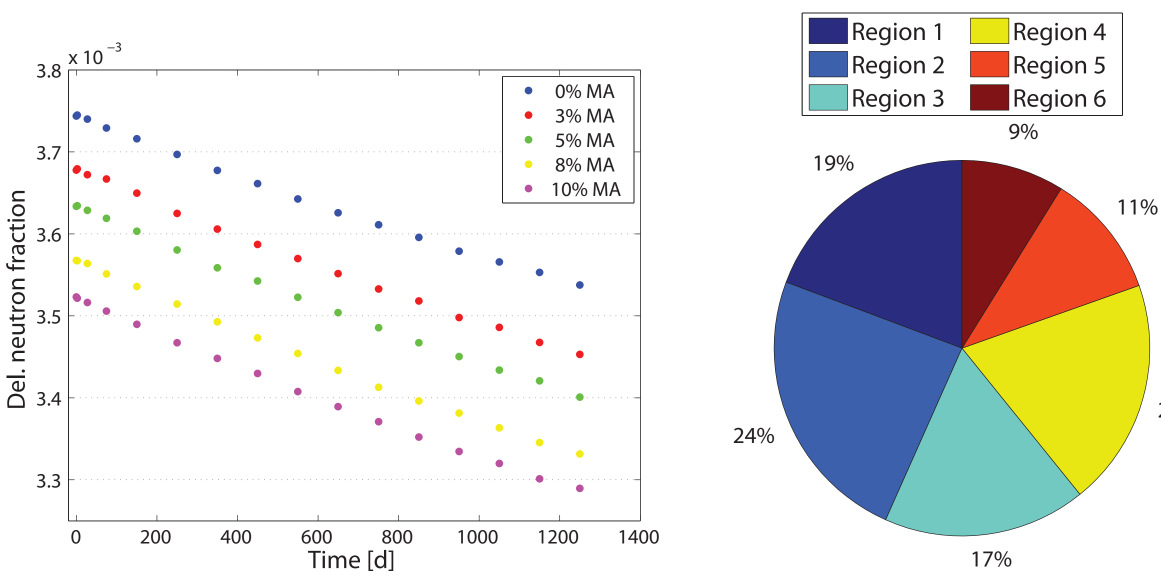

Figure 4. The delayed neutron fraction of cores with different initial VVER MA content during burnup and the spatial dependence of the delayed neutron production in a core with 10% initial VVER MA content at BOB

The contribution of the different regions of the reactor to the delayed neutron production (as can be seen in Figure 4) is slightly different from their volume fractions (which are 27% / 27% / 16% / 16% / 8% / 6%). The inner regions of the core have a higher contribution, the outer ones a lower one than expected from their volume fractions. However, the spatial dependence of the delayed neutron production is very similar for cores of different initial MA content, and it does not change during burnup either.

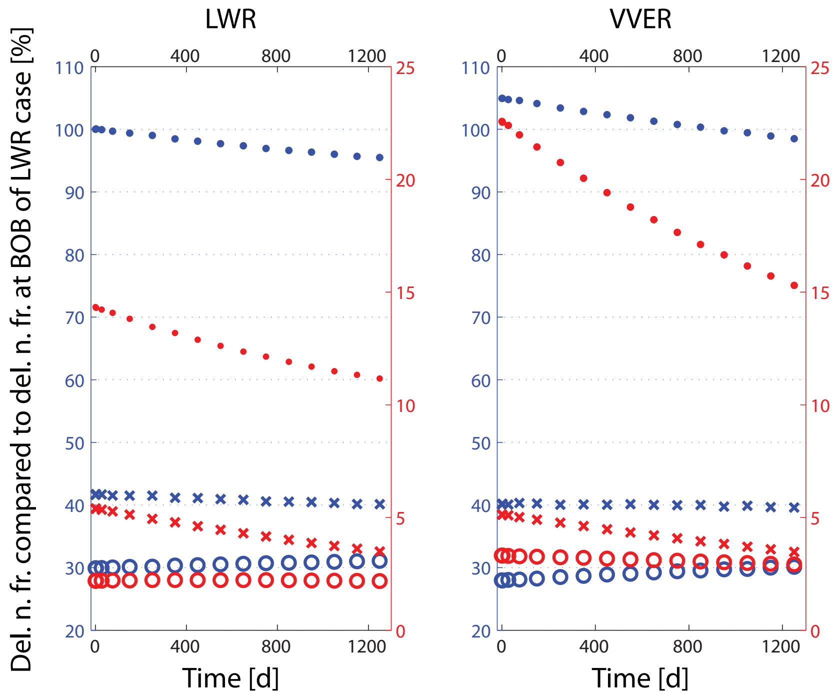

Figure 5. Breakdown of delayed neutron fractions of cores with 3% initial MA content of different origins. The contributors to the delayed neutron fractions are: Total (blue .), U-238 (blue x), Pu-239 (blue o), Pu-241 (red .), U-235 (red x), MAs (red o). Blue axes belong to the blue, red to the red graphs.

Finally, we investigate the change in the isotope inventory. The change of the MA elements is shown in Figure 6. In the LWR case the MA content decreased primarily due to the Am-241 transmutation into highly fissile Am-242m, while in the VVER case the dominant part of the reduction in the MA content is caused by Np, with Am decreasing less (in correspondence with the higher Np and lower Am content of the VVER spent fuel).

It is worth mentioning that the higher the initial MA content of the fuel is, the higher the reduction in the amount of MAs gets: at 5% initial MA content 138 kg and 111 kg, at 10% 314 kg and 284 kg of MAs are destroyed in the LWR and VVER cases respectively. The efficiency of the destruction of MAs also increases with the MA loading of the fuel, meaning that a bigger fraction of the initially loaded MAs is fissioned. It can also be concluded that Np and Am stockpiles can be much more efficiently reduced than Cm inventories. We will see in Section 5 that even with multiple burnup cycles only a slight reduction of Cm can be achieved.

The detailed three-dimensional model of the reactor makes it possible to investigate the spatial dependence of the isotope inventory as well. Spatial results showed that, as can be expected, the closer we are to the center of the core, hence the harder the spectrum is, the more efficiently we can fission MAs. At 10% initial MA content the roughly 20% reduction in the MA quantity ranges from 16% to 26% from the outermost to the innermost region. Based on this effect and the spatial dependence data of the delayed neutron production, it becomes possible to optimize the

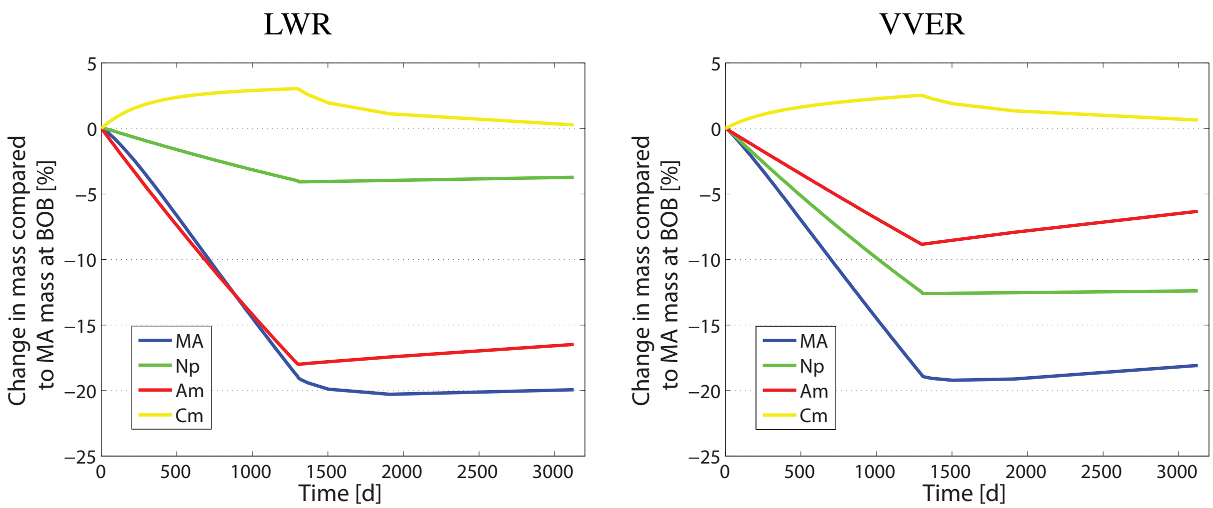

Figure 6. The change of the individual MA elements compared to the total MA mass at BOB with a core of 10% initial MA content

initial MA content of the different regions in order to increase the efficiency of MA destruction without further degrading the safety parameters of the reactor.

5. Results for multiple cycles

Multiple cycles were also investigated. Each cycle consisted of a burnup period of 1300 efpd and a decay period of 5 years. It was assumed that, due to future advanced aqueous reprocessing methods[9], the fuel can be reprocessed with high efficiency, and the gained actinides can all be used in the fabrication of the new fuel. Refueling was done either with purely depleted uranium (DU), or with a mix of DU and MAs, so that the MA content of the core at the beginning of each cycle was the same.

The change of reactivity in the first five burnup periods for cores with different initial MA contents is shown in Figure 7 for both strategies (the decay periods are discarded from the plots). The most noticeable result is that even with the lowest initial MA content the reactor can be operated during all 5 cycles, using only DU for refueling. It can also be observed that the rapid drop of reactivity is only present in the first cycles, in the later ones it has a much smaller decrease or even an increase.

The delayed neutron fraction shows a similar decline in each burnup period, however the decrease slows down with time (see in Figure 8). When using only DU for refueling, it basically comes to a halt in the fourth and fifth cycles, as the uranium feed always compensates for the loss of delayed neutrons due to the previous burnup period. When keeping the MA content constant we do not see such a definite stabilization, but the decrease during burnup slows down significantly.

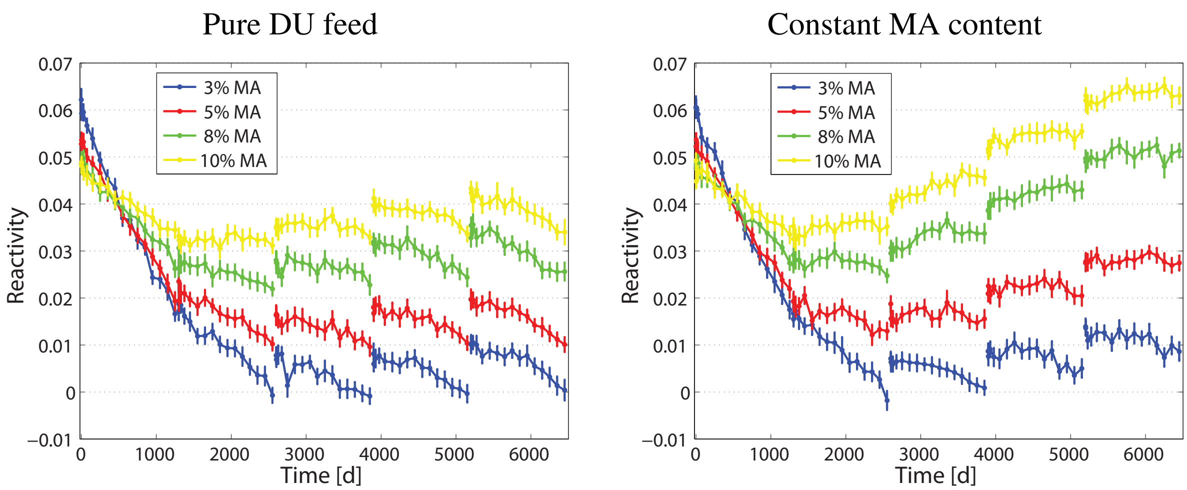

Figure 7. The reactivity of cores with different initial VVER MA content in the first five burnup periods

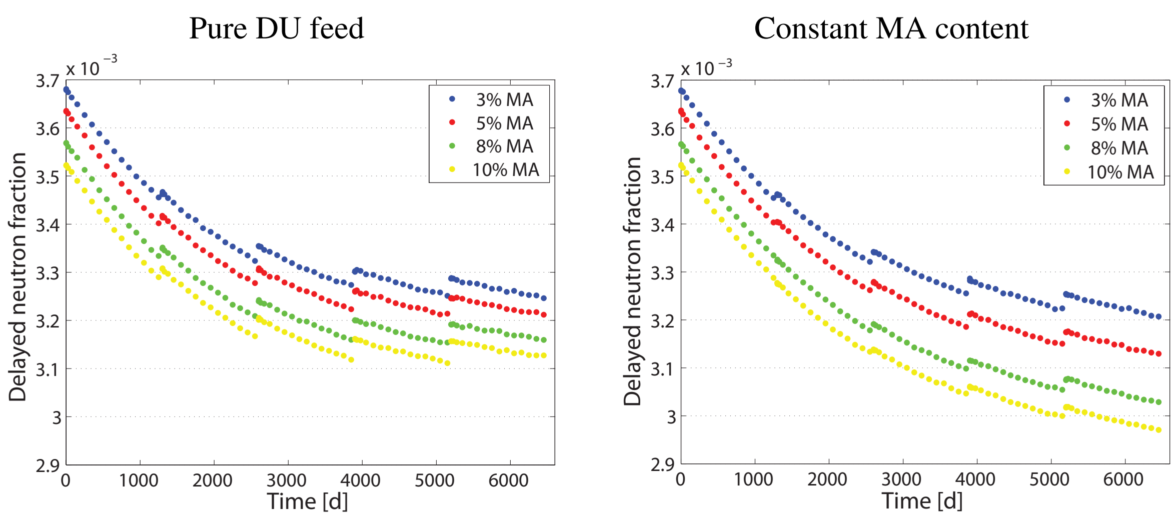

Figure 8. The delayed neutron fractions of cores with different initial VVER MA content in the first five cycles

The change of MAs during the first five cycles can be seen in Figure 9. When adding only DU to the fuel, the amount of MAs constantly decreases, and we can achieve approximately 70% reduction in five cycles in a core of 10% initial VVER MA content, mainly exclusively due to the transmutation of Np-237 and Am-241. Cm stockpiles can not be decreased, though the Cm level can be kept constant. When we keep the MA content of the core constant, a much higher amount of MAs can be destroyed due to their continuous feed, but we experience a steady increase of Cm. In the core with 10% initial VVER MA content, 320 kg of MAs can be destroyed in each cycle on the average, corresponding to an annual amount of 90 kg. That equals the MA output of approximately 3-4 VVER440 reactors (that being 25 kg per year).

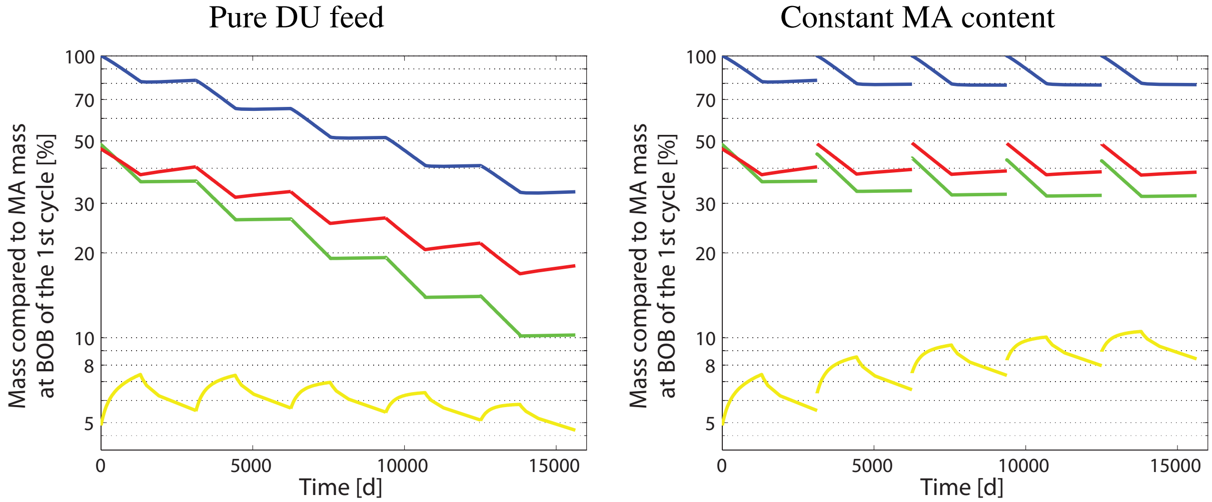

Figure 9. Mass of the individual MA elements compared to the total MA mass at BOB of the 1st cycle in a core with 10% initial VVER MA content. The shown elements are: all MAs (blue line), Am (red line), Np (green line) and Cm (yellow line). The y-axes are logarithmic.

6. Conclusions

In this paper an overview of the results of studying possible fuel cycles of the GFR600 was given. A geometrically precise model of the reactor was used and three-dimensional burnup calculations were performed. The main focus was on the consequences of mixing MAs and Pu originating from VVER440 legacy spent fuel to the fuel of the GFR600, but burnup calculations with LWR spent fuel were also done for comparison. The difference of the isotopic composition of VVER spent fuel from that of the LWR results in a higher initial reactivity, a more significant reactivity loss in the first burnup periods due to the faster decrease of Pu-241, and lower achievable burnups. The addition of MAs to the fuel decreases the reactivity loss in the VVER case as well, and though the swing does not turn positive in the first burnup period, the reactor can be operated for multiple cycles using only DU for refueling. The delayed neutron fraction is significantly higher in the VVER case than in the LWR at BOB (approximately 10% higher with 10% initial MA content), but also has a more rapid decrease during burnup. After several cycles the differences are considerably reduced, though the VVER case still has a slightly higher delayed neutron fraction after five cycles than the LWR. It was also found that the isotopic composition of the MA vector has a significant effect on the efficiency of destroying individual MA isotopes. Am-241 can be destroyed more easily in the LWR case, while Np-237 can be in the VVER case. Unfortunately, Cm stockpiles can not be efficiently diminished by the method presented, however extending the burnup periods from 1300 efpd and using only DU for refueling leads to a reduction of Cm as well.

Future work may include determining the effects of burnup and initial MA content of the fuel on the various reactivity coefficients, or the neutron source and heat load during reprocessing and fuel manufacturing. Furthermore it may be worth optimizing the MA content of the regions in order to maximize the MA consumption without degrading the safety parameters.

Presently the 600 MW thermal power reactor is no longer considered as the prime candidate of GCFR, instead a new design with 2400 MW power is proposed, using pin-type fuel. Therefore the above research should be extended to the new design in the future.

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