Minor actinide transmutation in a gas-cooled fast reactor

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Abstract

Within the Generation-IV Initiative the gas-cooled fast reactor (GFR) is one of the reactors dedicated to minor actinide (MA) transmutation. This paper summarises the research performed on the GFR600 design in order to assess its MA burning capabilities. For this study various modules of the SCALE programme system were used.

Single cycle parametric studies were performed with cores having different MA content and spatial distribution. It was shown that the addition of MAs to the fuel greatly reduces reactivity loss during burn-up. Moreover, the higher the MA content of the core is, the higher fraction of it is fissioned, however, the more the delayed neutron fraction and the fuel temperature coefficient degrade. Significant reductions can be achieved in the amount of neptunium and americium, while curium isotopes accumulate.

The study of multiple consecutive cycles showed that, by adding only depleted uranium (DU) to the reprocessed actinides in fuel fabrication (pure DU feed strategy, up to 70% of the initially loaded MA can be fissioned during the first 5 cycles. Moreover, the reactor can be made critical during that time if the initial MA content is higher than 3%. By feeding MAs as well (constant MA content strategy) the reactivity shows a steady increase from cycle to cycle, predominantly due to ^{238}Pu breeding from ^{237}Np.

The effects of the isotopic composition of the plutonium and MA were also examined by performing calculations with data specific to the spent fuel of traditional western pressurised water reactors and Russian type VVER440 reactors. Despite the considerably different MA vectors no significant deviation was found in their overall transmutation. However, the Pu composition has a strong effect on the reactivity and the delayed neutron fraction in the first cycles.

Finally, cores having non-uniform MA content were investigated. It was found that though the MA destruction efficiency is significantly higher in the middle of the core than at the edge, moving some of the MA from the outer regions to the centre only results in a minor improvement in their destruction. However, the spectral changes caused by the rearrangement increase the k-effective, which allows higher burn-ups and increased MA destruction. Unfortunately, some of the safety parameters of the reactor degrade.

Introduction

In 2002 the Generation-IV Initiative outlined the most important requirements that future nuclear reactors should meet and embraced six reactor concepts which have the highest potential to do so. One of the designs is the gas-cooled fast reactor (GFR), which is anticipated to be prominent in sustainability by having a closed fuel cycle and a self-breeder core [1]. In such a system all actinides would be recovered from the spent nuclear fuel (SNF) of the reactor during reprocessing and they would be used in fuel fabrication for the same reactor, thus only fission products (FPs) and actinides due to reprocessing losses would be sent to the geological repositories. At the same time, only fertile isotopes would have to be added to the recovered actinides during fuel manufacturing due to the efficient breeding. These would obviously result in a significantly reduced spent fuel output and a better use of fissile materials compared to todays' reactors.

The GFR is also envisioned to play an important role in nuclear waste management. Present reactors are mostly operated in an open fuel cycle, in which the spent fuel is not reprocessed and all actinides end up as waste together with the fission products. This is not only disadvantageous from the waste management, but also from the fuel economics point of view, since, on the one hand, the actinides increase the volume, heat load and radioactivity of the nuclear waste and represent its long-term danger, on the other hand, many fissile isotopes are discharged instead of being reused in thermal reactors. Hence it is desired to shift to a nuclear system where the spent fuel is always reprocessed, all actinides are recycled and only fission products are disposed of (together with the reprocessing losses). The reuse of uranium and plutonium is already possible as mixed-oxide (MOx) fuel in appropriate thermal reactors, but minor actinides (MA) are not usable today since they are not fissile in the present commercial reactors (nor is it daily practice to recover and reuse plutonium from spent MOx fuel). The situation is different in fast reactors, where, due to the increased energy of neutrons, all actinides are more fissionable and the build-up of heavier isotopes via capture is less probable. This leads to the idea of using them as actinide burners in order to effectively destroy actinides gained by reprocessing the spent fuel of other reactors. The main focus is on the elimination of minor actinides (neptunium, americium and curium) as they represent significant heat load and neutron source in the spent fuel and are not thermally fissile.

This paper focuses on a detailed assessment of the MA burning capabilities of GFRs using the CEA designed GFR600 concept as a reference. For the work a full three-dimensional model of the reactor was built with the SCALE code system [2] and several burn-up calculations were performed. The effects of the isotopic composition of MA and plutonium were studied and the performance of cores having spatially non-uniform MA content was examined.

The GFR600 reactor concept and its modelling using the SCALE code system

The CEA-designed "effective" GFR600 concept was chosen as a reference gas-cooled fast reactor, featuring 600 MW thermal power, plate type fuel and high pressure helium coolant. The exact details of the reactor are easy to find in most relevant articles (see [3] [4] for example), here only the most important parameters are repeated.

The core consists of 112 fuel assemblies (FAs) arranged in 6 rings around a central piece. Each assembly is hexagonal, has a 120 degrees rotational symmetry and contains 21 fuel plates in 3 compartments, each having 7 parallel plates fixed by the assembly wrapper and the Y-shaped central mechanical restraint. The fuel concept is based on dispersed fuel – the ceramic actinide compound (actinide carbide, 70 V/V% – volume percentage) is embedded in an inert ceramic matrix (SiC, 30 V/V%) forming the CERCER fuel. The structural material is also SiC (cladding, assembly wrapper, etc.), while the

reflector is Zr_3Si_2. In the reference fuel the actinide compound is made up of 84 n/n% (molar percentage) UC (natural uranium) and 16 n/n% PuC (legacy plutonium from spent fuel). These main parameters are summarised in Table 1.

<div align="center">Table 1: Main characteristics of GFR600</div>

Core parameters		Assembly parameters	
Thermal power	600 MW	Number of FAs	112
Power density	103 MW/m^3	Number of plates/FA	21
Core height/diameter	1.95 m/1.95 m	Fuel UPuC/SiC ratio	70/30 V/V%
Coolant temperature	490°C/850°C	Fuel temperature	990°C
Coolant material	Helium	Cladding material	SiC
Fuel parameters		**Natural U composition**	
SiC density	3.21 g/cm^3	^{235}U fraction	0.7 n/n%
UPuC density	12.63 g/cm^3	^{238}U fraction	99.3 n/n%
Porosity	15%		

It was investigated how minor actinides can be destroyed by being added to the fuel. In these burn-up calculations it was supposed that they substitute some of the uranium in the fuel, hence the actinide compound was chosen to be made up by 16 n/n% PuC, X n/n% MA carbide (minor actinide carbide) and (84-X) n/n% UC. To examine the effects of the isotopic composition of the recycled plutonium and minor actinides two sets of data were used in our study corresponding to the spent fuel of western pressurised water reactors and Russian VVER440 reactors, respectively. The former composition is based on the '"Pu-2016" scenario study of CEA and represents twice recycled MOx fuel [4], while the latter is characteristic of 45GWd/MTU burn-up and 5 years of cooling [5]. In this paper these two compositions will be referred to as the PWR and VVER cases, respectively.

In this study the TRITON6 module of SCALE was used to perform automatic Monte-Carlo transport and burn-up calculations with a fully detailed three-dimensional model of the reactor (as no detailed concept was available for the reflector regions and the plenums, these were modelled as homogeneous mixtures). Calculations were made with the two-dimensional method of characteristics NEWT module as well (on a corresponding 2D model of the core featuring homogenised fuel assemblies) to determine the burn-up dependence of the fuel temperature coefficient (FTC). The TSUNAMI sensitivity module was used to determine k-effective sensitivity coefficients and the isotopic breakdown of the FTC. Finally, the ORIGEN-S module was used to calculate several characteristics of the spent fuel.

The effects of adding minor actinides to the fuel

First, the effects of substituting some of the uranium in the fuel with MA were investigated by performing burn-up calculations with cores having uniform initial MA content of different amount and origin. All results discussed in this section correspond to the first cycle.

Figure 1 shows the change of reactivity during burn-up in cores having different initial VVER MA content. Two important effects should be observed; the addition of minor actinides decreases the initial reactivity and the reactivity loss alike. Obviously,

both are advantageous from the reactor operation point of view since a smaller reactivity has to be suppressed in the beginning and introduced during burn-up. Moreover, the decreased reactivity loss enables longer campaigns and higher burn-up values, consequently increasing the MA destruction.

The decreased initial reactivity is due to the replacement of uranium with stronger absorbing minor actinides. The decreased reactivity loss during burn-up is caused by the more robust production of fissionable isotopes, mainly ^{238}Pu and ^{242m}Am from the higher amount of ^{237}Np and ^{241}Am present in the core at the beginning of burn-up (BOB). Obviously, both these effects decrease the reactivity loss. Two major differences arise between the PWR and the VVER cases: both the initial reactivity and the reactivity loss during burn-up are higher in the VVER case. The former is caused by the higher initial ^{241}Pu content of the VVER plutonium, while the latter is explained by the more rapidly decreasing ^{241}Pu and the slower increasing ^{242m}Am amount [6].

Unfortunately, the safety of the reactor is negatively affected by the presence of MA. As can be seen in Figure 2, the delayed neutron fraction (DNF) decreases with the increasing MA content, though its decline during burn-up is almost entirely independent of that. The majority of delayed neutrons is produced by plutonium (^{239}Pu and ^{241}Pu primarily) and uranium (^{238}U mainly), moreover, the decreased number of delayed neutrons produced by the decreasing amount of uranium present in the fuel is basically compensated by the minor actinide isotopes (mostly by ^{237}Np). The true cause of the decrease in the DNF is that the contribution of the unchanged amount of plutonium in the fuel decreases, since the MAs make the spectrum harder, increasing the prompt, but leaving the delayed neutron yield unaffected.

Figure 1: The reactivity during burn-up in cores having different initial MA content
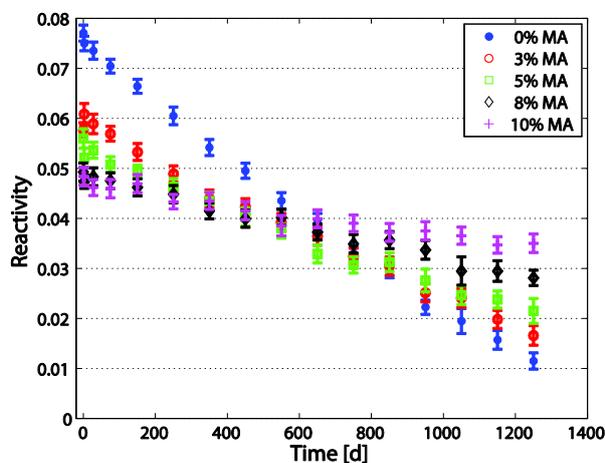
**Figure 2: The DNF during burn-up in cores having different initial MA content**
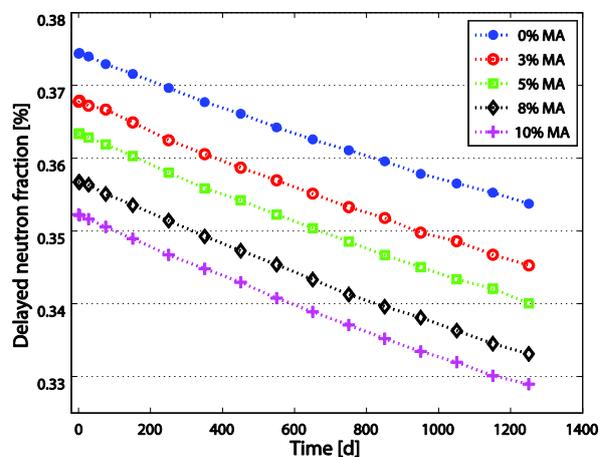
The delayed neutron fraction is higher in the VVER case (mainly due to the higher amount of ^{241}Pu and ^{237}Np) but also has a more rapid decrease during burn-up (as ^{241}Pu is being consumed more rapidly). The DNF at BOB is 0.360% and 0.325% in cores having 0% and 10% uniform initial PWR MA contents respectively, while the decrease during the first irradiation is 0.015% with both cores, compared to the more than 0.02% decrease observed in the VVER case. This difference, however, disappears in later cycles.

Figure 3 shows the fuel temperature coefficient during burn-up calculated with the NEWT model of the reactor for cores having different initial PWR MA content (the VVER case is basically identical to the PWR). Just like the delayed neutron fraction, the Doppler-coefficient also decreases in absolute value with the addition of minor actinides. However, unlike the DNF, the change of the FTC during burn-up depends on the initial MA content

of the core – at low MA contents the FTC decreases in absolute value, while at high contents it stays almost constant. Using the k-effective sensitivity coefficients calculated by the TSUNAMI module and standard perturbation techniques, the isotopic decomposition of the FTC was estimated for 4 cores (beginning and end of burn-up for a MA free core and one with 10% initial PWR MA content). As shown in Figure 4, ^{238}U is clearly dominant and spectrum effects can also be seen. In the MA free core the approximately 785 kg decrease in the ^{238}U content together with a 70 kg increase in the MA amount correspond to a decrease of -0.21 pcm/K in the FTC during burn-up, while in the core having 10% initial PWR MA content the 610 kg decrease of ^{238}U along with the 314 kg decrease of MAs basically leave the FTC unchanged. Consequently, at low MA contents, the ^{238}U decrease and the spectral hardening due to the MA accumulation both decrease the FTC, while at high MA contents the spectral softening due to the MA consumption counterbalances the effect of the decreasing ^{238}U.

Figure 3: The FTC during burn-up in cores having different MA content
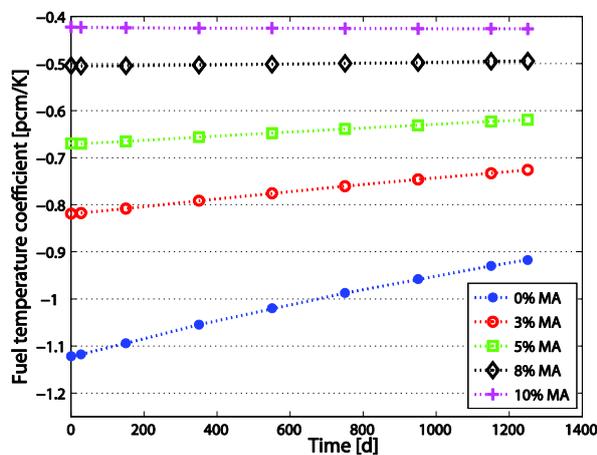
Figure 4: The isotopic contributions to the FTC in cores having different MA content
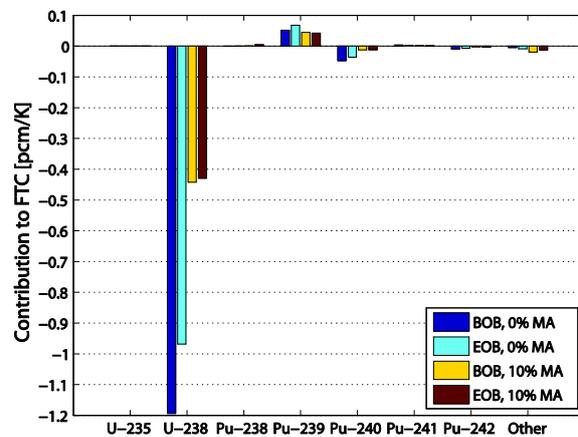
As the GFR is planned to be operated in a closed fuel cycle, its spent fuel will have to be reprocessed after every irradiation cycle and the recovered actinides will have to be incorporated in the new fuel of the reactor. It is important to know what conditions are to be expected during these procedures, hence it was analysed what effect the addition of minor actinides to the fuel has on the overall radioactivity, the thermal heat and the neutron source of the spent fuel.

The minor actinides have the most significant effect on the neutron source. Immediately after irradiation for 1 300 days the total neutron source of the spent fuel from the full core having 3% initial VVER MA content is approximately 5·10^{11} n/s, which is roughly 8 times the value calculated for the MA free core (6.5·10^{10} n/s), moreover, this difference only increases during the cooling period to over a factor of 13. With the further addition of MA the neutron source basically rises linearly to approximately 1.4·10^{12} n/s at 10% initial VVER MA content. Mainly Cm isotopes are responsible for the increase, as the more curium is present at the beginning of burn-up, the more is left at the end, consequently resulting in stronger neutron emission.

The thermal power of the spent fuel also increases with the MA content of the fuel, although to a lesser extent. The heat produced by the spent fuel from the full core having 10% and no initial VVER MA content at discharge (i.e. immediately after the first irradiation) is 4.7 MW and 2.7 MW, respectively. This modest increase is due to the fact that a significant portion of the thermal power can be attributed to the fission products, which build up very similarly in the different cores if the burn-up is the same. The expected heat load during fuel manufacturing from the actinide mix recovered from the

spent fuel of cores having 10% and 0% initial VVER MA content after 5 years of cooling is 27.9 W/kg and 3.3 W/kg, respectively (31.6 W/kg and 3.3 W/kg in the PWR case).

Finally, the addition of minor actinides also results in a more radioactive spent fuel, however, this increase is very modest, as the short-term radioactivity is dominated by the fission products (just like the thermal power). The expected values from the reprocessed actinides during fuel manufacturing are 72 GBq/g and 43GBq/g for cores having 10% and no initial VVER MA content (64GBq/g and 33 GBq/g for PWR), and mostly Pu isotopes are responsible for this activity [6].

Transmutation capabilities of GFR

The main goal of our investigation was to assess the minor actinide transmutational capabilities of the GFR. As the reactor is planned to be operated in a closed fuel cycle in which the spent fuel is always reprocessed and all the recovered actinides (including the minor actinides) are incorporated into new fuel assemblies, it was envisioned that the extra minor actinides that are sought to be destroyed would simply be added to the recovered (or the initial) actinides and no specific target assemblies would be embedded into the design.

Spatially uniform transmutation

Figure 5 shows the change of the individual minor actinide elements during burn-up for the VVER case. Mainly americium and neptunium are destroyed, predominantly due to the transmutation of ^{241}Am and ^{237}Np into fissionable ^{242m}Am and ^{238}Pu, respectively. The same is true in the PWR case, the destruction rate of Am and Np is basically the same in the two cases, approximately 20% and 25% of their initially loaded amount is fissioned (directly or after transmutation) during the 1300 days of operation.

The amount of the destroyed minor actinides heavily depends on their initial amount. At 5% initial MA content 111 kg and 138 kg, at 10% 284 kg and 314 kg of MA are destroyed in the VVER and PWR cases, respectively. Moreover, the efficiency of the destruction also increases with the MA content, the higher it gets, the bigger fraction of the initially loaded MA is destroyed during burn-up (approximately 10% at 3% and 20% at 10% initial MA content).

Unfortunately, the curium isotopes accumulate (mainly due to the production of ^{242}Cm and ^{244}Cm from ^{241}Am and ^{243}Am, respectively), though their production rate decreases with the initial MA content and the burn-up as well. Nevertheless, the more MA are added to the fuel, the more Cm is present at the end of the cycle. Even with multiple recycling its amount can only be kept constant.

Multiple cycles were also investigated with two different refuelling strategies. It was supposed that each burn-up period of 1300 days is followed by a cooling period of 5 years, after which the spent fuel of the whole core is instantly reprocessed and actinides are recovered with 99.9% efficiency, except for curium (99.3%). The total actinide content of the fuel was kept the same at the beginning of each cycle and the loss during burn-up and reprocessing was compensated either by adding purely depleted uranium (pure DU strategy) or a mix of DU and minor actinides in a way that the MA content of the core was the same at the beginning of each cycle (constant MA content strategy).

Figure 6 shows the change of reactivity for the pure DU feed strategy for the VVER case. The most important result is that if the initial MA content of the fuel is high enough, the criticality of the core can be ensured in the first five cycles by only adding depleted uranium in fuel manufacturing. During this time up to 50-70% of the initially loaded MA can be fissioned, depending on the MA content of the first core, and though the reduction is only due to the Am and Np isotopes, the amount of Cm only has a slight increase or stays approximately constant as it is being fissioned along with its mother isotopes. The

case is essentially the same with PWR spent fuel, the small differences present in the first cycle disappear later.

The constant MA content strategy aims at maximising the MA destruction by constantly feeding them into the reactor, since it was shown that the amount of destroyed MAs increases with the MA content of the fuel. There is a steady increase in the reactivity from cycle to cycle due to the continuous production of fissionable material from the MA. In each cycle 50 to 320 kg of MA can be fissioned, depending on the MA content of the fuel, which corresponds to an annual minor actinide destruction of 14 to 90 kg (which equals the MA output of 3 LWRs at best). Americium and neptunium make up the majority of the destroyed actinides, while curium isotopes slowly build up in the consecutive cycles (the amount is approximately 50 kg higher at the end of the 5th cooling period than at the beginning of the first burn-up period in the case of 10% initial MA content).

As was demonstrated in Figure 2, the delayed neutron fraction decreases during burn-up, this decline, however, reduces with the cycle number. In the pure DU feed strategy the DNF becomes fairly constant (its value ranges between 0.31% and 0.33%, depending on the MA content of the initial fuel and the burn-up), as the decrease experienced during burn-up is compensated by the increased delayed neutron production of the added uranium in the next cycle. In the constant MA content strategy there is no sign of such a stop, however, the 0.2% decrease of the DNF in the first cycle reduces to around 0.05% in the fifth and the delayed neutron fraction stays above 0.29% at all time.

Figure 5: The change of MA mass in cores having different initial MA content
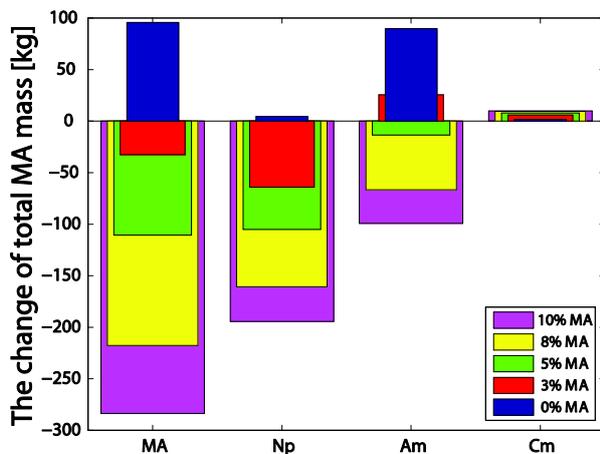
Figure 6: The reactivity during the first cycles in cores having different initial MA content
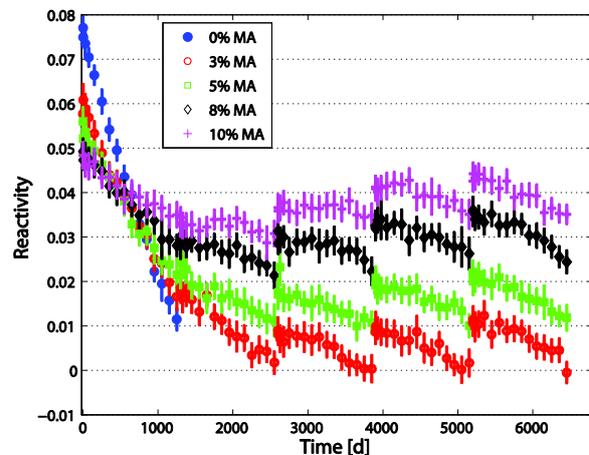
### Spatially non-uniform transmutation

The three-dimensional KENO-VI model made it possible to investigate the spatial dependence of the isotope inventory. As can be expected, the closer we are to the middle of the core (hence the harder the spectrum and the higher the flux is), the more efficient the destruction of MA is. The average destruction rate of 20% experienced in the cores having 10% initial MA content ranges between 16% and 26% (corresponding to the outermost and central regions, respectively). This led to the idea of investigating cores with non-uniform MA content in order to maximise their destruction.

A very simple core modification was done so that the total MA amount was not changed, but the MA content increased stepwise towards the center. It was supposed that the composition of the actinide compound in the fuel in region "r" is 16% PuC, X_r%

MA carbide and $(84 - X_r)\%$ UC, furthermore $X_r = X_1 + \frac{\Delta X}{5}(r - 1)$, where ΔX is the difference between the MA content of the central and the outermost region. The MA content of the outermost regions (i.e. X_1) is fixed by the constraint that the average MA content of the core is X%.

It was found that the rearrangement of minor actinides has a significant effect on reactivity. As can be seen in Figure 7, the more MA are moved to the centre of the core, the higher the reactivity gets. The main underlying effect is the increased fuel fissionability due to spectral changes. The actinides become more fissionable in the inner regions of the reactor (where the MA content increases, hardening the spectrum) and more absorbing in the outer regions (where the MA content decreases, softening the spectrum) - the net effect being an increased fissionability in all heavy metal isotope's case. At the same time the absorber MA isotopes become less absorbent on average [6].

Figure 7: The reactivity during burn-up in cores having different degree of non-uniformity with 5% average initial MA content
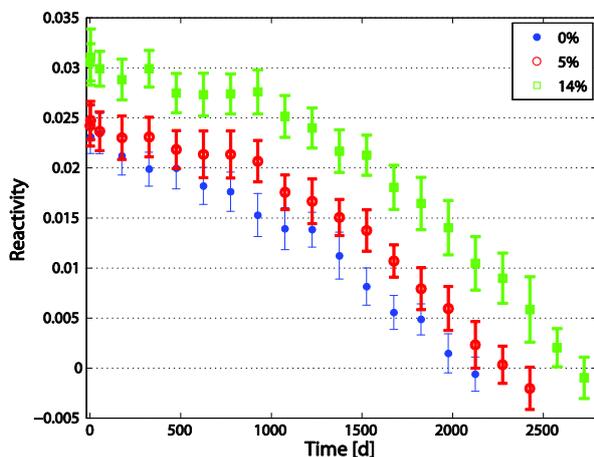
Figure 8: The mass of MAs during burn-up in cores having different degrees of non-uniformity with 5% average initial MA content
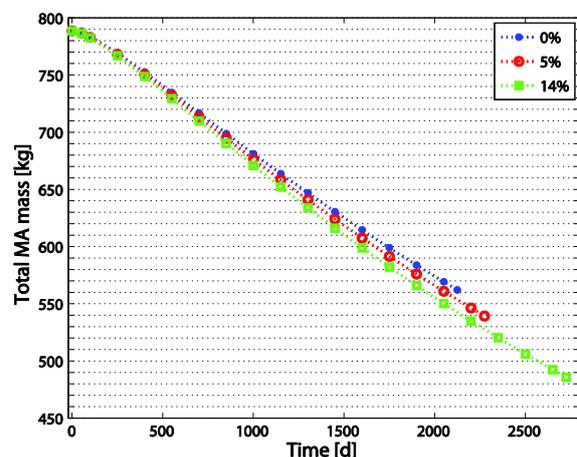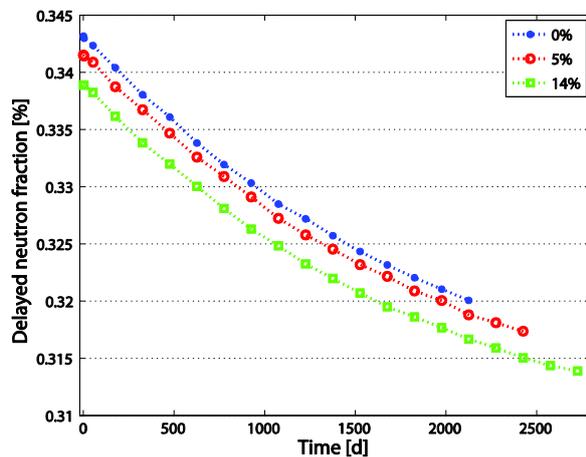

As can be seen in Figure 8, the direct increase in the amount of destroyed actinides gained by the rearrangement is quite modest (the core having maximum non-uniformity can burn 20 kg more MA than the uniform one in case of the maximum burn-up reachable in the uniform core). This is mainly due to the fact that, though the MA destruction efficiency is higher in the middle of the core, the volume of the central regions is small compared to the outer ones. However, the higher reactivity values enable longer burn-up periods, consequently increasing the MA transmutational capability of the reactor (an extra 70 kg of MAs can be burned in a core having 5% initial average MA content by introducing maximum non-uniformity and operating for an extra 600 days).

Unfortunately, the safety parameters of the reactor all degrade with the non-uniformity of the MA content. As shown in Figure 9, the delayed neutron fraction further decreases, mainly due to the decreased contribution of plutonium and uranium isotopes, since in the central regions the higher MA content results in a harder, while in the outer ones the lower MA content in a softer neutron spectrum, causing increased and decreased prompt neutron production in the regions respectively – the net effect being a decreased delayed neutron fraction.

The fuel temperature coefficient also degrades with the growing non-uniformity of the MA content. The FTC in the core with maximal non-uniformity is approximately 5-6% smaller in absolute value than in the uniform core at any time (see Figure 10). Moreover

the radial power peaking factor increases from a characteristic value of 1.3 in the case of the uniform core to 1.45 in the case of maximal non-uniformity.

Figure 9: The DNF during burn-up in cores having different degrees of non-uniformity with 5% average initial MA content

Figure 10: The FTC during burn-up in cores having different degree of non-uniformity with %% average initial MA content
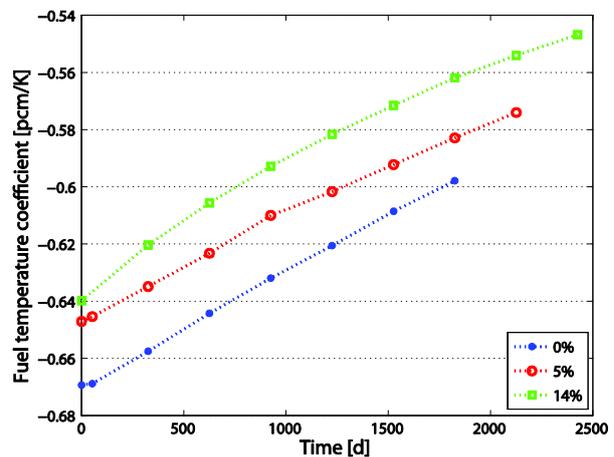
As can be seen in Figure 7, the direct improvement in the MA burning capability of the reactor due to the rearrangement of the MA content is quite modest, at the same time the safety features degrade when MAs are moved towards the centre of the core. The opposite is true when minor actinides are moved from the centre to the outer regions of the core, i.e. the radial power peak factor decreases, while the delayed neutron fraction and the fuel temperature coefficient increase. Hence it is possible to increase the safety parameters somewhat with the non-uniform MA loading without seriously degrading the MA burning capability.

Conclusions

This paper presents that replacing a small amount of uranium with minor actinides in the GFR600 reactor design reduces the initial reactivity and the reactivity loss during burn-up alike. Both facts are advantageous from the reactor control point of view and the latter makes it possible to lengthen campaigns and to reach higher burn-ups. Moreover, the reactor is a very efficient minor actinide burner. Depending on the initial minor actinide content of the fuel and its origin 50 to 300 kg of MA can be destroyed during 1 300 days of operation, which corresponds to the annual MA output of about 3 LWRs of the same power. For MA burning purposes the highest initial MA content is desirable, however, practical limitations are to be expected due to the negative effects on the delayed neutron fraction and the fuel temperature coefficient. The minor actinides also make reprocessing and fuel fabrication more challenging via the increased neutron source and heat load of the spent fuel.

While the MA burning capabilities depend only slightly on the isotopic composition of the MA vector, that of the used plutonium has a strong effect on reactivity and delayed neutron fraction. However, the differences are only present in the first cycles, hence it was concluded that the actual composition of the spent fuel used to fuel the reactor has no significant effect on its long-term performance.

Results for multiple irradiations were also presented. If the initial MA content of the fuel is high enough, the reactor can be made critical during the first 5 cycles by adding only depleted uranium to the reprocessed actinides during fuel fabrication. Meanwhile up to 70% of the initially loaded MA can be fissioned. Though the majority of the fissioned MA consists of neptunium and americium, curium accumulation is limited.

Results for cores having non-uniform MA content were also presented. The k-effective gradually increases when minor actinides are moved closer to the centre, mainly due to the fact that the absorber MA isotopes (^{237}Np, ^{241}Am and ^{243}Am) become more fissionable. This may be exploited to lengthen burn-up periods and increase minor actinide destruction. At the same time, however, safety parameters degrade somewhat.

The interested reader is referred to [6] for a more detailed description of the work presented here.

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