

Definition of breeding gain for the closed fuel cycle and application to a Gas Cooled Fast Reactor

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Abstract

In this paper a definition is given for the Breeding Gain (BG) of a nuclear reactor, taking into account compositional changes of the fuel during irradiation, cool down and reprocessing. A definition is given for the reactivity weights required to calculate BG . To calculate the effects of changes in the initial fuel composition on BG , first order nuclide perturbation theory is used. The theory is applied to the fuel cycle of GFR600, a 600 MWth Generation IV Gas Cooled Fast Reactor. This reactor should have a closed fuel cycle, with a BG equal to zero, breeding just enough new fuel during irradiation to allow refueling by only adding fertile material. All Heavy Metal is recycled in the closed fuel cycle. The result is that a closed fuel cycle is possible if the reprocessing has low losses ($< 1\%$). For reprocessing options with higher losses, adding a small amount of MA makes BG equal to zero. The equilibrium fuel contains about 80% ^{238}U , 15% Pu, and low amounts of the Minor Actinides.

KEYWORDS: *Gas Cooled Fast Reactor, Breeding Gain, Closed Fuel Cycle, Adjoint Transmutation Theory, Minor Actinide Recycling*

1. Introduction

To estimate the performance of breeder reactors, several performance parameters can be defined. The *Breeding Gain* (BG) is usually defined as the ratio of the net gain of fissile material (FG) to the net destruction of fissile material (FD): $BG \equiv FG/FD$. This definition is integral in nature, as it depends on the amounts of nuclear fuel at beginning and end of cycle. The above definition also requires a choice of which nuclides are considered fissile. Usually ^{235}U , ^{239}Pu and ^{241}Pu are considered fissile. Since the fissile nuclides have different nuclear properties, a weighing is required. Common weighings are +1 for fissile, and 0 for other nuclides, or 0 for ^{238}U , 1 for ^{239}Pu , 1.5 for ^{241}Pu and 0.75 for ^{235}U [1, 2].

If a nuclear reactor is required to run in a closed fuel cycle, such as envisaged for the Generation IV GCFR [3], a definition of BG is required taking into account irradiation, cool down and reprocessing. The concept of 'fissile' versus 'fertile' also needs revision if the fuel is to be recycled in a fast reactor, because in a fast spectrum almost all actinides can be fissioned. In this paper such a new definition of BG is proposed, together with the corresponding weighing scheme for the nuclides. Adjoint nuclide theory is developed to estimate the influence of variations of the initial fuel to the BG of the fuel cycle.

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2. Theoretical framework

Let the space and time dependent nuclide inventory of a reactor be given by $\vec{N}(r, t)$. As a performance parameter of the nuclide mix at time t_0 , define a response parameter R as:

$$R(t_0) = \langle \vec{w}(r, t), \vec{N}(r, t) \rangle \quad (1)$$

where angles indicate scalar products (i.e. integration over space and time, summation over all nuclides). In this paper it is implicitly assumed that \vec{w} contains a Kronecker delta function $\delta(t - t_0)$ to obtain R at t_0 . Assume that the weights \vec{w} account for the reactivity of the nuclides, quantifying how each nuclide contributes to the overall reactivity of the reactor. Assume we have two nuclide vectors $\vec{N}_1(r)$ and $\vec{N}_2(r)$, i.e. nuclide composition at two different times t_1 and t_2 . Now assume we have two sets of corresponding weights: \vec{w}_1 and \vec{w}_2 . Now R can be calculated for both mixtures and we can consider the difference as a measure of Breeding Gain:

$$BG = \frac{R(t_2) - R(t_1)}{R(t_1)} = \frac{\langle \vec{w}_2, \vec{N}_2(r) \rangle - \langle \vec{w}_1, \vec{N}_1(r) \rangle}{\langle \vec{w}_1, \vec{N}_1(r) \rangle} \quad (2)$$

where the weights \vec{w} account for the 'fissibility' of the individual nuclides. To define \vec{w} , consider the change of the eigenvalue of a reactor caused by a small change of the number density of nuclide i , ΔN_i . The reference reactor is described by

$$[L_0 - \lambda_0 P_0] \phi_0 = 0, \quad (3)$$

with L_0, P_0 the loss and production operators ([4]). Throughout this paper, reference values are denoted by the subscript '0', perturbed values have no subscript: $L = L_0 + \Delta L$, etc. Thus the perturbed reactor is described by:

$$[L - \lambda P] \phi = 0 \quad (4)$$

Using first-order perturbation theory, the change in eigenvalue $\Delta \lambda$ is given by ([4]):

$$\Delta \lambda = \frac{\langle \phi_0^*, [\Delta L - \lambda_0 \Delta P] \phi_0 \rangle}{\langle \phi_0^*, P_0 \phi_0 \rangle} \quad (5)$$

Dividing by ΔN_i and taking the limit for infinitesimal perturbations results in:

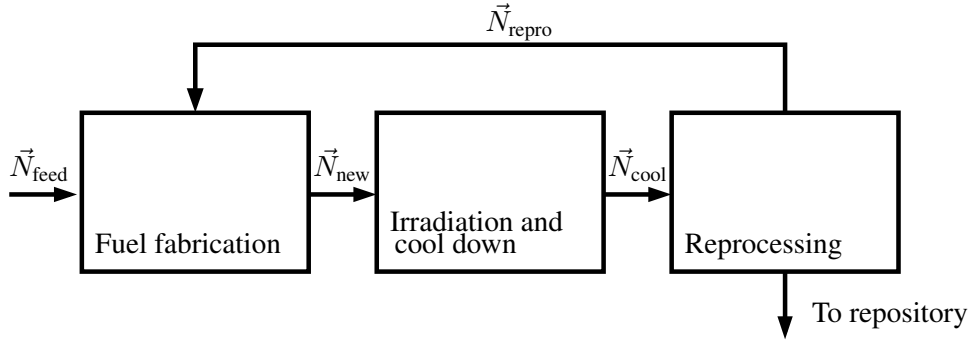
$$\lim_{\Delta N_i \rightarrow 0} \frac{\Delta \lambda}{\Delta N_i} = \frac{d\lambda}{dN_i} = \frac{\langle \phi_0^*, [\frac{dL}{dN_i} - \lambda_0 \frac{dP}{dN_i}] \phi_0 \rangle}{\langle \phi_0^*, P_0 \phi_0 \rangle} \quad (6)$$

Now introduce the reactivity ρ , defined as $\rho = 1 - \lambda$. Then $d\rho/dN_i = -d\lambda/dN_i$, and the reactivity weights \vec{w} are defined by:

$$w_i \equiv \frac{d\rho}{dN_i} = \frac{\langle \phi_0^*, [\lambda_0 \frac{dP}{dN_i} - \frac{dL}{dN_i}] \phi_0 \rangle}{\langle \phi_0^*, P_0 \phi_0 \rangle} \quad (7)$$

Equation (7) is comparable to the expressions used in sensitivity and uncertainty theory, and gives a measure of the change of the reactivity of the reactor caused by density changes of an individual nuclide i .

Figure 1: Material flow in a reprocessing fuel cycle. The symbols are as in the text, i.e. \vec{N}_{feed} is the feed material (e.g. depleted uranium), which is used together with \vec{N}_{repro} to make the new fuel \vec{N}_{new} . After irradiation and cool down the material is given by \vec{N}_{cool} .



3. Reprocessing formalism

In a fuel cycle with reprocessing, a new fuel can be made using the reprocessed material, to which feed material can be added. In a closed fuel cycle, material has to be added to the fuel to offset the losses of burnup and reprocessing. In figure 1 a schematic is given of the fuel cycle with reprocessing. The fuel from the fuel fabrication is formally described as:

$$\vec{N}_{\text{new}} = \vec{N}_{\text{repro}} + \vec{N}_{\text{feed}} \quad (8)$$

with \vec{N}_{new} the composition of the new fuel, \vec{N}_{repro} the reprocessed material and \vec{N}_{feed} the feed material. The vector \vec{N}_{repro} is given by

$$\vec{N}_{\text{repro}} = \vec{S} \vec{N}_{\text{cool}} \quad (9)$$

with \vec{S} the vector quantifying the recovery efficiency of individual isotopes ($0 \leq S_i < 1$). \vec{N}_{cool} is the material from the previous cycle after irradiation and cool down. The vector of feed material \vec{N}_{feed} is given by

$$\vec{N}_{\text{feed}} = N_{\text{feed}} \cdot \vec{v}_{\text{feed}} = [N_{\text{target}} - N_{\text{repro}}] \vec{v}_{\text{feed}} = [N_{\text{target}} - \langle \vec{S}, \vec{N}_{\text{old}} \rangle] \vec{v}_{\text{feed}} \quad (10)$$

Here $N_{\text{feed}} = |\vec{N}_{\text{feed}}|$ is the total amount of feed material. The isotopic composition of the feed material is given by the unit vector \vec{v}_{feed} (80% ^{238}U and 20% ^{240}Pu , for instance). N_{target} is the target total amount of new fuel. For multi-recycling in the same reactor, N_{target} equals $|\vec{N}_{\text{new}}|$, i.e. the total amount of fuel at beginning of irradiation in the reactor. The amount of feed material to be added thus equals the target amount of new fuel, minus the amount of material recovered from the previous irradiation N_{repro} . The amount of reprocessed material is given by $|\vec{N}_{\text{repro}}| = \langle \vec{S}, \vec{N}_{\text{old}} \rangle$.

Given equation (8), the R of the new fuel mixture can be written as:

$$R = \langle \vec{w}, \vec{N}_{\text{new}} \rangle = \langle \vec{w}, \vec{N}_{\text{repro}} \rangle + \langle \vec{w}, \vec{N}_{\text{feed}} \rangle = R_{\text{repro}} + R_{\text{feed}} \quad (11)$$

i.e. R of the new fuel is the sum of the R of the reprocessed material and the R of the feed material. Expanding (11) using (9) and (10) results in:

$$R = \langle \vec{w} \vec{S}, \vec{N}_{\text{old}} \rangle + [N_{\text{target}} - \langle \vec{S}, \vec{N}_{\text{old}} \rangle] \langle \vec{w}, \vec{v}_{\text{feed}} \rangle \quad (12)$$

The R calculated for the new material using (12) can be compared to the R of the initial fuel, and BG can be calculated by

$$BG = \frac{R_2 - R_1}{R_1} = \frac{R_2}{R_1} - 1 = \frac{\langle \vec{w}_2, \vec{N}_2 \rangle}{\langle \vec{w}_1, \vec{N}_1 \rangle} - 1 \quad (13)$$

where the index '1' indicates the fresh fuel at the start of the previous irradiation and '2' indicates the new fuel for the current irradiation. The set of weights \vec{w}_2 for the new fuel can be calculated once \vec{N}_{new} is known. Note here that small variations in the initial fuel composition in the previous cycle have an influence on R (and thus BG) through the resulting variation in \vec{N}_{old} .

4. Nuclide perturbation theory

It is desired to estimate the effect of variations of the initial fuel composition on the total BG of the fuel cycle, for instance to optimize fuel design. From the definitions of R and BG in (12) and (13) it is clear that variations of the initial fuel composition have an effect on BG through \vec{N}_{old} . In fact, there are two effects:

1. One effect is the variation in the total amount of reprocessed material. From equation (12) this effect is quantified by the $\langle \vec{S}, \vec{N}_{\text{old}} \rangle$ term.
2. The second effect is the variation of the reactivity of the reprocessed material. This term is quantified by the $\langle \vec{w} \vec{S}, \vec{N}_{\text{old}} \rangle$ term in equation (12).

To calculate the effect of variations of the initial fuel composition on the composition of the fuel after irradiation and cool down, we have to solve the adjoint transmutation equations. The forward transmutation equation describing the evolution of a mixture of nuclides (in or out of pile) is given by:

$$\frac{\partial \vec{N}_0}{\partial t} = \underline{M}_0 \vec{N}_0 + \vec{Q}_0 \quad (14)$$

with the initial condition

$$\vec{N}_0(t = 0) = \vec{N}_i. \quad (15)$$

The transmutation matrix \underline{M}_0 contains the nuclear cross sections σ_x , decay constants λ_x , and the flux ϕ . \underline{M} is space-time dependent through the cross sections and flux. \vec{Q}_0 is an independent source term (e.g. adding and extracting material from a tank during reprocessing). Using equation (14) the evolution of the fuel mixture during irradiation can be calculated. Consider a reponse $R = \langle \vec{h}, \vec{N}_0 \rangle$. If, for example, the vector \vec{h} contains $\sigma_a \phi \delta(t - t_0)$ on location i , R represents the absorption rate of neutrons by nuclide i at time t_0 . If the initial condition \vec{N}_i changes, R will also change. To calculate this effect, the adjoint transmutation equation needs to be solved. This equation is given by:

$$-\frac{\partial \vec{N}_0^*}{\partial t} = \underline{M}_0^* \vec{N}_0^* + \vec{Q}_0^* \quad (16)$$

It can be shown, e.g. as in [5], that in order to obtain a meaningful interpretation of the adjoint equations, the adjoint boundary conditions and source must be chosen as:

$$\vec{N}_0^*(t = t_f) = 0 \quad (17)$$

$$\vec{Q}_0^* = \frac{\partial R}{\partial \vec{N}} = \vec{h} \quad (18)$$

Once the adjoint equation (16) is solved, the perturbation of the response ΔR due to changes ΔN_0 of the initial conditions are calculated from:

$$\Delta R = [\vec{N}_0^*(t = 0) \Delta \vec{N}_0]_x \quad (19)$$

where the $[\]_x$ indicates that the integration is over the entire phase space except t . Using equation (19), one can calculate the effect of a variation of the initial nuclide composition to the final composition. The adjoint transmutation equation 'propagates' the effects of perturbations of nuclide densities to the final composition.

5. Reactor model and calculational tools

The above theory is developed for a Generation IV Gas Cooled Fast Reactor (GCFR) [3]. This type of reactor is envisaged to run in a closed fuel cycle, breeding just enough fissile material to refuel the reactor, only adding fertile material to the reprocessed material. All actinides are recycled indiscriminately in this concept. Within the European 6th Framework Program, GFR600 is investigated, a 600 MWth Generation IV GCFR. The theory is applied to the fuel cycle of this reactor to determine BG for several fuel concepts and reprocessing options. All calculations are based on unit cell calculations. Some key parameters of the reactor are given in table 1. The fuel for this reactor is a plate fuel, using a fuel composed of a mix of (U, Pu, MA)C (70 vol%) and SiC matrix (30 vol%), clad by thin layers of SiC. The GFR600 fuel assembly is shown in figure 2. Table 2 gives the composition of the reference fuel and a fuel containing 5% MA (Np, Am, Cm). Because an integral fuel cycle is envisaged for GFR600 the influence of adding MA on the breeding gain is researched.

The weights \vec{w} are obtained using the sensitivity module TSUNAMI-1D of the SCALE 5 code system [6], which calculates the sensitivity of k_{eff} to the number densities in the reactor. A burnup calculation code (LOWFAT: Like Origen With Forward and Adjoint Transmutation) similar to ORIGEN-S but with adjoint capabilities was written to calculate the depletion, cool down and reprocessing of the HM mixture. This code uses problem dependent nuclear cross sections in the ORIGEN-S format, prepared by the CSAS and COUPLE modules of the SCALE 5 system. A special numerical recipe for stiff ODEs (DVIDE [7]) is used to solve the systems (14) and (16).

5.1 Breeding Gain calculations on the GFR600 fuel cycle

One fuel cycle was calculated, with 1300 days of irradiation at constant flux $\phi = 2.10^{15}$ n/cm².s¹, (burnup ≈ 6.5 % FIMA), cool down (2192 days = 6 years) and reprocessing for the GFR600 reference fuel, and for a fuel containing 5% MA. The reprocessing scheme is integral with 99% efficiency, adding depleted uranium to make the new fuel.

Figure 2: GFR600 fuel assembly. The light coloured wrapper and central mechanical restraint are made of SiC. The darker fuel slabs contain the fuel mixture clad with SiC. Each assembly contains 21 fuel plates. The fueled length is 1.95 m, the overall length of the assembly about 4 m. The overall volume fractions are 55 vol% helium, 10 vol% SiC for structures and cladding and 35 vol% fuel/matrix (70 % UPuC/30 % SiC).

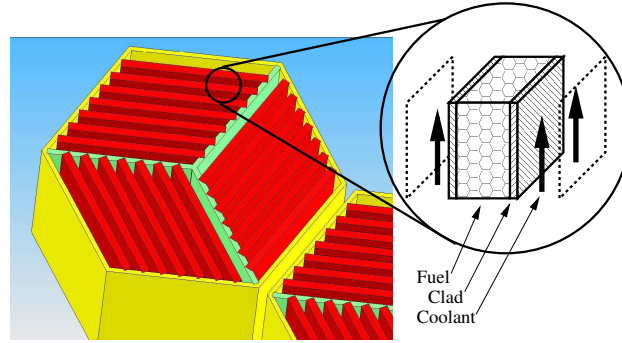


Table 1: GFR600 core parameters.

<i>Reference data for GFR600</i>	
Power [MWth]	600
Coolant	He
Power density [MW/m ³]	103
Specific power [W/gHM]	45
T _{core,in} [°C]	480
T _{core,out} [°C]	850
Core H/D [m]	1.95/1.95
p [MPa]	7.0
Fuel type	plates
Fuel material	UPuC + MA
Struct. material	SiC
Refl. material	Zr ₃ Si ₂

Refer to table 3: R_{ini} is R for the fresh fuel, R_{irrad} is at the end of the irradiation (i.e. after 1300 days), R_{cool} is after 6 years of cool down, and R_{repro} is the reactivity of the reprocessed material. The new fuel is composed of reprocessed and feed material (see eq. (11)). R of the feed material is given by R_{feed} and the R of the new fuel is $R_{new} = R_{repro} + R_{feed}$. R_{feed} is dominated by the weight of ²³⁸U, which is negative. With the reference fuel, BG over the irradiation ($R_{irrad}/R_{ini} - 1$) is close to zero, but during cool down reactivity is lost, resulting in an overall BG from the initial fuel to the new fuel of -20.2%. For the MA fuel BG is positive during the irradiation (+ 12%), effectively offsetting the loss during cool down, resulting in an overall BG close to zero (-3.56%). Concluding: adding a small amount of MA to the fuel results in a positive BG over the irradiation, and over the entire cycle, BG is close to zero.

For the MA fuel adjoints were calculated corresponding to 99% efficient integral recycling to determine the influence of initial variations on BG . To get BG closer to zero, two perturbations were considered, namely:

Table 2: GFR600 fuel compositions: reference fuel, and a fuel containing 5% MA.

<i>Isotopic composition</i>		
	Reference fuel	MA fuel
	at. %	at. %
^{235}U	0.60	0.56
^{238}U	83.52	78.60
^{237}Np	-	0.83
^{238}Pu	0.43	0.43
^{239}Pu	8.92	8.92
^{240}Pu	4.11	4.11
^{241}Pu	1.18	1.18
^{242}Pu	1.14	1.14
^{241}Am	0.11	3.11
$^{242\text{m}}\text{Am}$	-	0.01
^{243}Am	-	0.77
^{242}Cm	-	0.001
^{243}Cm	-	0.004
^{244}Cm	-	0.254
^{245}Cm	-	0.062
^{246}Cm	-	0.005

Table 3: Evolution of R [m^{-1}] for the reference fuel and a fuel with MA. All values are 10^{-3} unless stated otherwise. $R_{\text{ini}} = R$ of initial fuel mixture, R_{irrad} is after irradiation, R_{cool} is after 2192 days of cool down, R_{repro} is the R of the reprocessed material. The reactivity of the feed material is given by R_{feed} , and the new fuel is described by R_{new} .

	GFR 600 (ref)	GFR600 (5% MA)
R_{ini}	1.02	1.15
R_{irrad}	1.01	1.29
R_{cool}	0.932	1.22
R_{repro}	0.923	1.21
R_{feed}	-0.109	-0.097
R_{new}	0.813	1.11
BG	-20.2 %	-3.56 %

1. Increase of 5% of ^{235}U , ^{239}Pu , ^{240}Pu and ^{241}Pu
2. Increase of 5% of ^{238}U

The reason for this choice is that is possible to increase the reactivity of the reprocessed fuel by starting with a larger fissile content (option 1), or by increasing the total amount of reprocessed material, thereby reducing the amount of (negative reactivity from) feed material (option 2). The results are given in table 4, where the symbols are as before. The table gives the values of R for the reference calculation (same as table 3), after which the ΔR introduced by the perturbations are given. E.g. adding ^{235}U has a positive ΔR_{repro} : the reactivity of the

Table 4: Results of 2 perturbations on the evolution of R . The reference situation is a 5% MA fuel. 2 perturbations are applied and their effects calculated. 2 forward calculations were done to check the result. All values are 10^{-3} unless stated otherwise.

			R_{ini}	1.15	
			R_{irrad}	1.29	
			R_{cool}	1.22	
			R_{repro}	1.21	
			R_{feed}	-0.097	
			R_{new}	1.11	
			BG	-3.56%	
ΔN	ΔR_{repro}	ΔR_{feed}	ΔN	ΔR_{repro}	ΔR_{feed}
^{235}U	2.39e-6	2.39e-7	^{238}U	-1.14e-5	4.92e-5
^{239}Pu	5.18e-5	3.86e-6			
^{240}Pu	6.71e-6	2.34e-6			
^{241}Pu	5.08e-6	4.55e-7			
\sum	6.60e-5	6.90e-6	\sum	-1.14e-5	4.92e-5
R'_{new}		1.19	R'_{new}		1.15
<i>Forward</i>			<i>Forward</i>		
		R_{ini}			1.11
		R_{irrad}			1.28
		R_{cool}			1.21
		R_{repro}			1.20
		R_{feed}			0.048
		R_{new}			1.15
		BG			+ 3.71 %

reprocessed material is higher. ^{235}U also has a positive ΔR_{feed} : more ^{235}U means a smaller amount of feed material is added to make the new fuel. The feed material is mostly ^{238}U , and its weight is negative. Therefore, adding less of it has a positive effect. For the other isotopes similar arguments apply.

The perturbation of ^{238}U (right column) has a negative effect on R_{repro} but a positive effect on R_{feed} . In the two bottom cells of each column of table 4 the result of a forward calculation with the perturbed initial vectors is given. These forward calculations yield the same R_{new} . The forward calculations give all values of R during the irradiation and cool down with the perturbed nuclide inventory. Perturbing the fissile content yields a higher R_{new} but the initial R is also higher: the initial mixture is more reactive. The extra initial reactivity is so large, that BG is in fact worse: from -3.56 % to -6.42%. The perturbation of ^{238}U yields a lower initial R_{ini} , and a better BG : +3.71%.

5.2 Long term behaviour

To investigate the long-term behaviour a series of 15 irradiation, cool down and reprocessing cycles was calculated. Since the cycle length is some 9.5 years, 15 cycles is longer than reactor lifetime, but it serves to illustrate the long-term behavior. Two reprocessing strategies are reported here:

1. Integral reprocessing with 99% efficiency, after which depleted uranium is added.

2. Integral reprocessing at 95% efficiency, after which a mix of 90% depleted uranium and 10% MA (same vector as before) is added.

The choice for the second strategy is given by the fact that R_{repro} is lower if \vec{S} is smaller (i.e. reactivity is lost due to large reprocessing loss), leading to the necessity to increase R_{feed} to make the overall BG closer to zero. In figure 3 the evolution of the k_{eff} of the fresh fuel and the reactivity weight R_{new} are illustrated. Both fuel cycle strategies converge to a situation where k_{eff} and R_{new} of the fresh fuel are more or less equal from cycle to cycle, so we approach the required situation of a zero BG over the entire fuel cycle.

For the 99% efficient reprocessing, BG (equation (13)) during irradiation is about +15%. The 95% efficient strategy, which adds some MA to the new fuel, has a BG between +15% and +25% during irradiation, offsetting the losses of reprocessing. In both cases, the equilibrium fuel composition has $\approx 81\%$ $^{238}_{38}\text{U}$, $\approx 9\%$ ^{239}Pu , $\approx 5\%$ ^{240}Pu , while all other isotope fractions are lower than 1%. The 95% strategy, which adds extra MA during reprocessing, has a larger fraction of ^{238}Pu than the 99% strategy. This is due to the addition of ^{237}Np , which is a precursor for ^{238}Pu . Of the two presented strategies, the first strategy (refueling with reprocessed material and DU only) would be considered as a Generation IV fuel cycle: enough new fissile material is bred during the cycle to allow refueling with a fertile material only.

We can thus draw the conclusion that a truly closed fuel cycle is possible if the reprocessing losses are small enough (the 99% strategy). If the reprocessing scheme is less efficient in recovering the actinides, BG can be made closer to zero by adding depleted uranium with a small amount (10 %) of MA. Transmutation of the MA will then offset the reactivity losses during reprocessing.

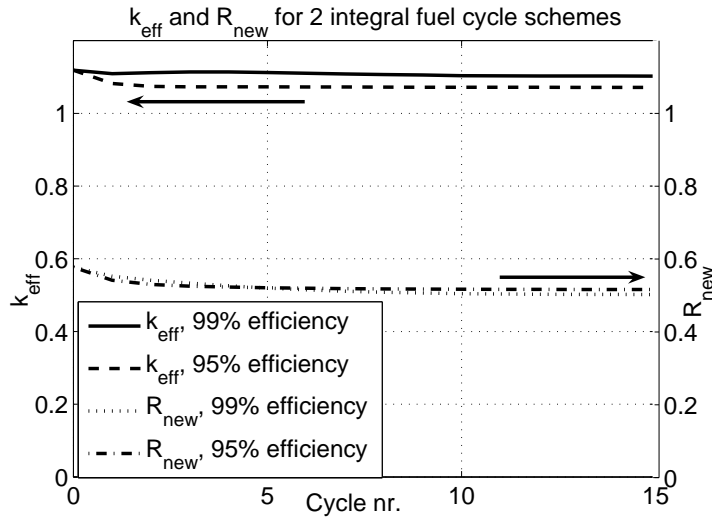
6. Discussion and conclusion

A definition of Breeding Gain was derived taking into account the change of composition of a nuclear fuel during irradiation, cool down and reprocessing. This theory is applied to an integral fuel cycle for a Gas Cooled Fast Reactor running on uranium, plutonium and possibly MA. It is found that the proposed reference fuel has $BG = 0$ during irradiation, but over the entire cycle of irradiation, cool down and reprocessing BG is -20.2 %. A fuel with uranium, plutonium and 5% MA (Np, Am, Cm) yields a positive BG during irradiation, and a BG close to zero over the entire cycle of irradiation, cool down and reprocessing.

A series of 15 consecutive fuel cycles was calculated. The result is that a BG close to zero can be obtained if an integral reprocessing scheme is adopted. If the reprocessing efficiency is high enough (99%), it is possible to obtain $BG = 0$ by adding depleted uranium only. If the reprocessing efficiency is somewhat lower (95%) it is possible to add a mix of depleted uranium and a small amount (10%) of MA. Transmutation of the MA will then give an overall BG close to zero. The evolution of the fuel composition over the 15 cycles seems to indicate that an equilibrium contains mostly ^{238}U ($\approx 81\%$), a small portion of Pu ($\approx 15\%$), with the rest Np, Am, and Cm.

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Figure 3: Evolution of k_{eff} and R during multiple recycles for two fuel cycle schemes: 99% efficiency refers to an integral fuel cycle with 99% reprocessing efficiency, adding depleted uranium to make the new fuel. 95% efficiency refers to the scheme with 95% efficient reprocessing, after which a mix of depleted uranium and MA is added. Both schemes converge to a situation where the k_{eff} and the R_{new} are constant from cycle to cycle. Since R_{new} is the same from cycle to cycle, $BG = 0$.



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