

Definition of Breeding Gain for the Closed Fuel Cycle and Application to a Gas-Cooled Fast Reactor

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Received June 9, 2006

Accepted November 23, 2006

Abstract—The Generation IV gas-cooled fast reactor (GCFR) is intended to have a closed fuel cycle: During irradiation enough fissile material is produced to allow refueling of the same reactor, adding only fertile material. This is the well-known “zero breeding gain” objective. In this paper a theoretical framework is derived to track compositional changes of the fuel during irradiation, cooldown, and reprocessing, in order to calculate the reactivity of the new fuel compared to the original fuel material. Using first-order perturbation theory, the effect of variations of the initial fuel composition on the reprocessed material and breeding gain can be calculated. The theory is applied to the fuel cycle of a 600 MW(thermal) GCFR. The result is that the change of material composition during cooldown has a nonnegligible effect on the breeding gain. A truly closed fuel cycle can be obtained if the reprocessing efficiency is high enough (<1% loss). If this high efficiency cannot be obtained, adding a small amount of minor actinides (Np, Am, Cm) to the new fuel results in a zero breeding gain. Perturbation theory provides a powerful tool to estimate the effects of changing fuel cycle parameters.

I. INTRODUCTION

To estimate the performance of breeder reactors, several performance parameters can be defined. Examples are the breeding ratio (BR), which is the ratio of the amount of fissile material produced (FP) during irradiation to the amount of fissile material destructed (FD) during irradiation: $BR \equiv FP/FD$. 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): $BG \equiv FG/FD$. Since the net gain of fissile material is equal to the total production minus the destruction, BG can be written as: $BG \equiv FG/FD = (FP - FD)/FD = BR - 1$ (e.g., see Ref. 1). The above definition is integral in nature, as it depends on the amounts of nuclear fuel at beginning and end of cycle. The above definitions also require 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 weighting is required. Common weightings 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 (Refs. 1 and 2).

Another definition of BR is given in terms of reaction rates: BR is then the ratio of the production rate of fissile material to the destruction rate. This definition is differential in nature, and can be calculated at any moment of the irradiation history, once the flux spectrum, cross sections, and number densities are known.² In this scheme, BG can be defined as the ratio of the net production rate of fissile material to the destruction rate, but in practice BG is normalized to the total fission rate in the reactor. BG then reflects the change of fissile material per fission. The differential definition also requires a proper reactivity weighting. The differential definition can be extended to include production and destruction by radioactive decay as well. Such a definition is included in the ERANOS simulation software.³

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If a nuclear reactor is required to run in a closed fuel cycle, such as envisaged for the Generation IV gas-cooled fast reactor⁴ (GCFR), a definition of BG is required taking into account irradiation, cooldown, 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 weighting 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. It will be shown that a truly closed fuel cycle is possible if the efficiency of reprocessing is high enough. For less efficient reprocessing, adding a small amount of minor actinides (MAs) to the fuel allows an almost closed fuel cycle. Nuclide perturbation theory provides a powerful tool to estimate the influence of individual isotopes to the overall fuel cycle performance.

II. DEFINITION OF BREEDING GAIN

Let the space-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 Dirac delta function $\delta(t - t_0)$ to obtain R at t_0 . Assume that the weights \vec{w} account for the reactivity of each nuclide, i.e., the contribution of each nuclide to the overall reactivity of the reactor. Then, if we have two nuclide compositions at two times t_1 and t_2 [$\vec{N}_1(r)$ and $\vec{N}_2(r)$], and we have two corresponding sets of weights (\vec{w}_1 and \vec{w}_2), R can be calculated for both mixtures and we can consider the difference as a measure of BG:

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

III. DEFINITION OF \vec{w}

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

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

where L_0 and P_0 are the loss and production operators, respectively.⁵ 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 by⁵

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

where ϕ_0^* is the solution of the adjoint unperturbed system. The small perturbations ΔL and ΔP can be expanded in a Taylor series, retaining only the first term:

$$\Delta L = \frac{\partial L}{\partial \alpha_i} \Delta \alpha_i \quad (6)$$

and

$$\Delta P = \frac{\partial P}{\partial \alpha_i} \Delta \alpha_i, \quad (7)$$

where α_i is any data element appearing in the operators L_0 and P_0 . In our case, we are interested in finding the response to a change in the density of nuclide i . Substituting Eqs. (6) and (7) into Eq. (5) for a small perturbation ΔN_i results in

$$\Delta \lambda = \frac{\left\langle \phi_0^*, \left[\frac{\partial L}{\partial N_i} \Delta N_i - \lambda_0 \frac{\partial P}{\partial N_i} \Delta N_i \right] \phi_0 \right\rangle}{\langle \phi_0^*, P_0 \phi_0 \rangle}. \quad (8)$$

Dividing both sides by ΔN_i results in

$$\frac{\Delta \lambda}{\Delta N_i} = \frac{\left\langle \phi_0^*, \left[\frac{\partial L}{\partial N_i} - \lambda_0 \frac{\partial P}{\partial N_i} \right] \phi_0 \right\rangle}{\langle \phi_0^*, P_0 \phi_0 \rangle}. \quad (9)$$

Now introduce the reactivity ρ_0 , defined as $\rho_0 = 1 - \lambda_0$. Then $\Delta \rho = -\Delta \lambda$, and the reactivity weights \vec{w} are defined by

$$w_i \equiv \frac{\Delta \rho}{\Delta N_i} = \frac{\left\langle \phi_0^*, \left[\lambda_0 \frac{\partial P}{\partial N_i} - \frac{\partial L}{\partial N_i} \right] \phi_0 \right\rangle}{\langle \phi_0^*, P_0 \phi_0 \rangle}. \quad (10)$$

Equation (10) 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 . As an example, consider a reactor described in a one-group, infinite homogeneous diffusion formalism. The operators L_0 and P_0 are given by

$$P_0 = \nu \Sigma_f = \sum_{i=1}^I N_i \nu_i \sigma_{f,i}$$

and

$$L_0 = \Sigma_a = \sum_{i=1}^I N_i \sigma_{a,i} , \quad (11)$$

with the index i running over all isotopes in the system. In the one-group formalism, ϕ_0, ϕ_0^* reduce to single numbers. Taking the derivatives to N_i in Eq. (11) and substituting in Eq. (10) results in

$$w_i = \frac{\phi_0^* (\lambda_0 \nu_i \sigma_{f,i} - \sigma_{a,i}) \phi_0}{\phi_0^* \nu \Sigma_f \phi_0} = \frac{1}{\nu \Sigma_f} (\lambda_0 \nu_i \sigma_{f,i} - \sigma_{a,i}) , \quad (12)$$

which is similar to traditional definitions of reactivity weights as, for instance, found in Ref. 2:

$$w_i = \nu_i \sigma_{f,i} - \sigma_{a,i} . \quad (13)$$

The differences between Eqs. (12) and (13) are the presence of the factors $1/\nu \Sigma_f$ and λ_0 . These factors are not problematic because $\lambda_0 = 1$ in a critical reactor, which is usually assumed in the derivation of Eq. (13), and $1/\nu \Sigma_f$ can be removed by normalizing.

IV. 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 Fig. 1 a schematic is given of the fuel cycle with reprocessing. The fuel from the fuel fabrication is formally described as

$$\vec{N}_{new} = \vec{N}_{repro} + \vec{N}_{feed} , \quad (14)$$

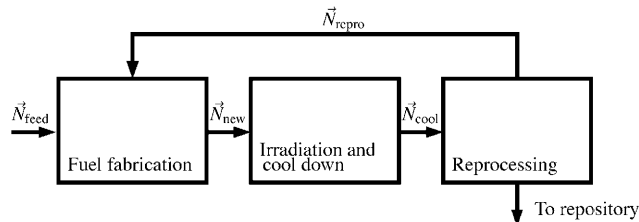


Fig. 1. Material flow in a reprocessing fuel cycle. The symbols are as in the text, i.e., \vec{N}_{feed} is the feed material (e.g., depleted uranium), which is used together with \vec{N}_{repro} to make the new fuel \vec{N}_{new} . After irradiation and cooldown, the material is given by \vec{N}_{cool}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

where

\vec{N}_{new} = composition of the new fuel

\vec{N}_{repro} = reprocessed material

\vec{N}_{feed} = feed material.

The vector \vec{N}_{repro} is given by

$$\vec{N}_{repro} = \underline{S} \vec{N}_{cool} , \quad (15)$$

where \underline{S} is a diagonal matrix whose elements quantify the recovery efficiency of individual isotopes ($0 \leq S_{ii} < 1$, $S_{ij, i \neq j} = 0$), and \vec{N}_{cool} is the material from the previous cycle after irradiation and cooldown. The vector of feed material \vec{N}_{feed} is given by

$$\begin{aligned} \vec{N}_{feed} &= N_{feed} \cdot \vec{v}_{feed} = [N_{target} - N_{repro}] \vec{v}_{feed} \\ &= [N_{target} - |\underline{S} \vec{N}_{cool}|] \vec{v}_{feed} . \end{aligned} \quad (16)$$

Here $N_{feed} = |\vec{N}_{feed}|$ 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). The target total amount of new fuel is N_{target} . For multirecycling in the same reactor, N_{target} equals $|\vec{N}_{new}|$, 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 $|\vec{N}_{repro}| = |\underline{S} \vec{N}_{cool}|$.

It is important to realize what the free parameters are in this formulation. The composition of the fuel after irradiation and cooldown is determined by the burnup, neutron energy spectrum in the reactor, and the length of the cooldown interval. The composition of the reprocessed material is determined by the reprocessing strategy and efficiency, which are (more or less) free parameters. In this paper the target is to obtain a closed fuel cycle, i.e., all actinides are recycled, and only fertile material is added to make new fuel. In a truly closed fuel cycle, the choice of fertile material to be added is limited: Natural uranium is one option, and depleted uranium (tails from enrichment) is another option. Either one of these options results in a closed fuel cycle. Thus, the parameters to influence the closed fuel cycle are the burnup, cooldown interval length, and reprocessing strategy and efficiency. A third option explored in this paper is use of depleted uranium and a small amount of MAs from future stockpiles. This would be an almost closed fuel cycle.

Given Eq. (14), the R of the new fuel mixture can be written as

$$\begin{aligned} R &= \langle \vec{w}, \vec{N}_{new} \rangle = \langle \vec{w}, \vec{N}_{repro} \rangle + \langle \vec{w}, \vec{N}_{feed} \rangle \\ &= R_{repro} + R_{feed} , \end{aligned} \quad (17)$$

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 Eq. (17) using Eqs. (15) and (16) results in

$$R = \langle \vec{w}, \underline{S}\vec{N}_{cool} \rangle + [N_{target} - |\underline{S}\vec{N}_{cool}|] \langle \vec{w}, \vec{v}_{feed} \rangle . \quad (18)$$

The R calculated for the new material using Eq. (18) can be compared to the R of the initial fuel, and BG can be calculated by

$$BG = \frac{R_2 - R_1}{R_1} = \frac{R_2}{R_1} - 1 = \frac{\langle \vec{w}_2, \vec{N}_2 \rangle}{\langle \vec{w}_1, \vec{N}_1 \rangle} - 1 , \quad (19)$$

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.

V. COMPARISON TO TRADITIONAL DEFINITIONS OF BREEDING GAIN

In Sec. I two categories of BG were described, namely, integral and differential definitions. The definition of BG given in Eqs. (2) and (19) are integral in nature because they use R at two different points in time. A differential definition can be readily obtained from Eq. (1) by taking the time derivative:

$$\frac{dR}{dt} = \frac{d}{dt} \langle \vec{w}, \vec{N} \rangle = \left\langle \vec{w}, \frac{d\vec{N}}{dt} \right\rangle + \left\langle \frac{d\vec{w}}{dt}, \vec{N} \right\rangle . \quad (20)$$

We now use the quasi-static approach, by assuming $\langle d\vec{w}/dt, \vec{N} \rangle$ to be negligible compared to $\langle \vec{w}, d\vec{N}/dt \rangle$. A justification of this approach is given in Sec. VI and the Appendix. Now introduce the standard transmutation equation in matrix form:

$$\frac{d\vec{N}}{dt} = \underline{M}\vec{N} + \vec{Q} , \quad (21)$$

where \underline{M} is the transmutation matrix containing nuclear data, and \vec{Q} is an independent source term. Applying Eq. (21) to Eq. (20) gives

$$\frac{dR}{dt} = \langle \vec{w}, \underline{M}\vec{N} \rangle , \quad (22)$$

where the source term \vec{Q} is set to zero, i.e., no fuel is added or removed during the burnup calculation. Now Eq. (22) can be expanded as

$$\begin{aligned} \frac{dR}{dt} = \int \sum_{i=1}^I w_i \left\{ -N_i(\sigma_{a,i}\phi + \lambda_i) + \sum_j y_{j \rightarrow i} N_j \sigma_{f,j} \phi \right. \\ \left. + \sum_k N_k \sigma_{c,k \rightarrow i} \phi + \sum_l \lambda_{l \rightarrow i} N_l \right\} dp , \end{aligned} \quad (23)$$

where the indices $i, j, k,$ and l run over all nuclides in the system, and p indicates the phase space of the problem. $N, \sigma_x,$ and ϕ are space-time dependent. Equation (23) can be compared to the definition of BG given in Ref. 2:

$$BG = \frac{\int \sum_{i=1}^I w_i (N_{i-1}(r, t) \sigma_{c,i-1}(r, t) - N_i(r, t) \sigma_{a,i}(r, t)) \phi dp}{\int \sum_{i=1}^I \Sigma_{f,i}(r, t) \phi dp} , \quad (24)$$

where the weights \vec{w} are defined by Eq. (13). Equation (24), like Eq. (23), sums weighted production and destruction rates of nuclides, so Eq. (23) can be interpreted as an extension of Eq. (24). Note that Eq. (24) requires the predefinition of the mother/daughter nuclides for capture (the $\sum_{c,i-1} w_i$ term) and is only applicable during irradiation. This is a drawback for calculations on fuel that is to be recycled as a whole, where compositional changes during cooldown can significantly affect the new fuel (e.g., decay of ^{241}Pu to ^{241}Am). Equations (23) and (24) are differential in nature, i.e., they provide an instantaneous BG and do not require solving an entire irradiation history.

VI. NUCLIDE PERTURBATION THEORY

It is desirable 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. To calculate the effect of variations of the initial fuel composition on the composition of the fuel after irradiation and cooldown, the use of first-order nuclide perturbation theory is proposed. The formal derivation of perturbation analysis is given in the Appendix, and an illustration of adjoint calculations is given in Sec. VIII. From the definitions of R and BG in Eqs. (18) and (19), it is clear that variations of the initial fuel composition have an effect on BG through \vec{N}_{cool} . In fact, there are two effects:

1. One effect is the variation in the total amount of reprocessed material. From Eq. (18) this effect is quantified by the $|\underline{S}\vec{N}_{cool}|$ term. As explained in the Appendix, the corresponding final time condition for the inventory adjoint equations equals

$$\vec{N}_0^*(t = t_f) = \underline{S}\vec{I} . \quad (25)$$

2. The second effect is the variation of the reactivity of the reprocessed material. This term is quantified by the $\langle \vec{w}, \underline{S}\vec{N}_{cool} \rangle$ term in Eq. (18). The corresponding final time condition for the reactivity adjoint equation is given by

$$\vec{N}^*(t = t_f) = \underline{S}\vec{w} . \quad (26)$$

TABLE I
One-Group Cross Sections for a Representative GCFR Fuel Mixture in a Closed Fuel Cycle*

	σ_a Unirradiated	σ_c Unirradiated	σ_a Irradiated	σ_c Irradiated	$\Delta\sigma_a$ (%)	$\Delta\sigma_c$ (%)
^{238}U	$3.54\text{e}-1^{\text{a}}$	$3.00\text{e}-1$	$3.57\text{e}-1$	$3.02\text{e}-1$	+0.92	+0.63
^{238}Pu	$1.95\text{e}-0$	$6.19\text{e}-1$	$1.96\text{e}-0$	$6.24\text{e}-1$	+0.71	+0.77
^{239}Pu	$2.51\text{e}-0$	$6.09\text{e}-1$	$2.52\text{e}-0$	$6.13\text{e}-1$	+0.45	+0.78
^{240}Pu	$9.71\text{e}-1$	$5.64\text{e}-1$	$9.78\text{e}-1$	$5.65\text{e}-1$	+0.72	+0.3
^{241}Am	$2.33\text{e}-0$	$2.01\text{e}-0$	$2.35\text{e}-0$	$2.02\text{e}-0$	+0.59	+0.4

*Cross sections are given for fresh and irradiated fuel (6.5% FIMA). The cross sections are almost constant during irradiation.

^aRead as 3.54×10^{-1} .

It should be noted that the dimension (or units) of the adjoint nuclide density depend on the type of response under consideration. In our case, both inventory and reactivity adjoint are dimensionless: The weights \vec{w} are dimensionless by normalization, and the inventory adjoint merely expresses the number of recycled nuclides per nuclide added.

As indicated in the Appendix, a first-order error is introduced if the change of the weights \vec{w} caused by a change of the composition \vec{N} is neglected. However, this error is small. The weights \vec{w} depend on the microscopic cross sections σ . The composition \vec{N} has its influence by causing a change of the neutron energy spectrum. The reactor under investigation in this paper is a GCFR. In a fast reactor, the dependence of the (groupwise) cross sections to the nuclide composition of the fuel is not very strong. Also, the effect of fission products on the spectrum is smaller than for thermal reactors. To illustrate this, the equivalent one-group cross sections for capture and absorption were calculated for a typical GCFR fuel mixture, for the fresh fuel and after irradiation. As can be seen from Table I, the cross sections do not change very much, and hence it is concluded that a first-order error is present, but small. In Sec. VIII.E the validity of the quasi-static approach is discussed in more detail.

VII. REACTOR MODEL AND CALCULATIONAL TOOLS

The above theory is developed for a Generation IV GCFR (Ref. 4). This type of reactor is envisaged to run in a closed fuel cycle, breeding just enough fissile material to refuel the reactor and only adding fertile material to the reprocessed material. All actinides are recycled indiscriminately in this concept. Within the European Sixth Framework Program, GFR600 is investigated, a 600 MW(thermal) Generation IV GCFR. The theory is applied to the fuel cycle of this reactor to determine BG for several fuel concepts and reprocessing options. Some

key parameters of the reactor are given in Table II. 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 illustrated in Fig. 2. Table III 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 BG is researched.

The weights \vec{w} are obtained using the sensitivity module TSUNAMI-1D of the SCALE 5 code system.⁶ TSUNAMI-1D calculates the sensitivity of k_{eff} to the nuclear data, based on a unit cell calculation on an infinite lattice with fuel, cladding, and moderator, in a slab, cylindrical, or spherical configuration. Axial leakage is taken into account by a buckling correction. TSUNAMI-1D calculates the sensitivity of k_{eff} to the nuclear data in the following way.^{6,7} Starting from Eq. (4) the sensitivity coefficient is defined as

TABLE II
GFR600 Core Parameters

Reference Data for GFR600	
Power [MW(thermal)]	600
Coolant	He
Power density (MW/m ³)	103
Specific power (W/g) ^a	45
$T_{core,in}$ (°C)	480
$T_{core,out}$ (°C)	850
Core height/diameter (m/m)	1.95/1.95
p (MPa)	7.0
Fuel type	Plates
Fuel material	UPuC + MA
Structural material	SiC
Reflector material	Zr ₃ Si ₂
Coolant/structural materials/fuel (vol%)	55/10/35

^aPer gram of heavy metal.

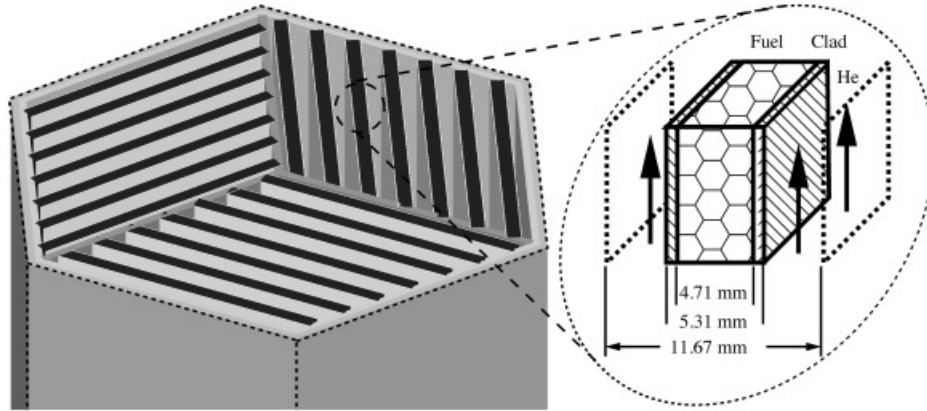


Fig. 2. GFR600 fuel assembly. All parts are made of SiC. The fueled length is 1.95 m, total length of the assemblies is ~ 4 m. The overall volume fractions are 55 vol% helium, 10 vol% SiC for structures and cladding, and 35 vol% fuel/matrix (70% UPuC/30% SiC).

$$\frac{\Sigma_x}{\lambda_0} \frac{\Delta \lambda}{\Delta \Sigma_x} = S_x = \frac{\Sigma_x}{\lambda_0} \frac{\left\langle \phi_0^*, \left[\frac{\partial L}{\partial \Sigma_x} - \lambda_0 \frac{\partial P}{\partial \Sigma_x} \right] \phi_0 \right\rangle}{\left\langle \phi_0^*, P_0 \phi_0 \right\rangle}. \quad (27)$$

Now, $k_{eff} = 1/\lambda_0$, and taking the Gâteaux derivative,

$$\begin{aligned} \frac{d}{d\epsilon} \{ \lambda_0 + \epsilon \Delta \lambda \}_{\epsilon=0} &= \frac{d}{d\epsilon} \left\{ \frac{1}{k_{eff} + \epsilon \Delta k_{eff}} \right\}_{\epsilon=0} \Rightarrow \Delta \lambda \\ &= \frac{-\Delta k_{eff}}{k_{eff}^2}, \end{aligned} \quad (28)$$

we find $\Delta \lambda / \lambda_0 = -\Delta k_{eff} / k_{eff}$. Now substitute into Eq. (27):

$$\begin{aligned} \frac{\Sigma_x}{k_{eff}} \frac{\Delta k_{eff}}{\Delta \Sigma_x} &= -\frac{\Sigma_x}{\lambda_0} \frac{\left\langle \phi_0^*, \left[\frac{\partial L}{\partial \Sigma_x} - \lambda_0 \frac{\partial P}{\partial \Sigma_x} \right] \phi_0 \right\rangle}{\left\langle \phi_0^*, P_0 \phi_0 \right\rangle} \\ &= -\Sigma_x \cdot k_{eff} \cdot \frac{\left\langle \phi_0^*, \left[\frac{\partial L}{\partial \Sigma_x} - \frac{1}{k_{eff}} \frac{\partial P}{\partial \Sigma_x} \right] \phi_0 \right\rangle}{\left\langle \phi_0^*, P_0 \phi_0 \right\rangle}. \end{aligned} \quad (29)$$

Multiplying numerator and denominator by $1/k_{eff}^2$ results in

$$\begin{aligned} S_{k_{eff}, \Sigma_x} &\equiv \frac{dk_{eff}/d\Sigma_x}{k_{eff}/\Sigma_x} \\ &= -\frac{\Sigma_x}{k_{eff}} \frac{\left\langle \phi_0^*, \left[\frac{dL}{d\Sigma_x} - \frac{1}{k_{eff}} \frac{dP}{d\Sigma_x} \right] \phi_0 \right\rangle}{\left\langle \phi_0^*, \frac{P}{k_{eff}^2} \phi_0 \right\rangle}, \end{aligned} \quad (30)$$

TABLE III
GFR600 Fuel Compositions: Reference Fuel
and a Fuel Containing 5% MA

		Reference Fuel		MA Fuel	
Elemental Composition					
U (%)		84		79	
Pu (%)		16		16	
MA (%)		—		5	
Isotopic Composition					
		Atomic Density (b·cm) ⁻¹	at. %	Atomic Density (b·cm) ⁻¹	at. %
²³⁵ U	1.1631e-4 ^a	0.60	1.0938e-4	0.56	
²³⁸ U	1.6291e-2	83.52	1.5320e-2	78.60	
²³⁷ Np	—	—	1.6242e-4	0.83	
²³⁸ Pu	8.3625e-5	0.43	8.3625e-5	0.43	
²³⁹ Pu	1.7395e-3	8.92	1.7395e-3	8.92	
²⁴⁰ Pu	8.0163e-4	4.11	8.0163e-4	4.11	
²⁴¹ Pu	2.2940e-4	1.18	2.2940e-4	1.18	
²⁴² Pu	2.2235e-4	1.14	2.2235e-4	1.14	
²⁴¹ Am	2.1687e-5	0.11	6.0680e-4	3.11	
^{242m} Am	—	—	2.2072e-6	0.01	
²⁴³ Am	—	—	1.5020e-4	0.77	
²⁴² Cm	—	—	1.9219e-7	0.001	
²⁴³ Cm	—	—	7.0178e-7	0.004	
²⁴⁴ Cm	—	—	4.9558e-5	0.254	
²⁴⁵ Cm	—	—	1.2086e-5	0.062	
²⁴⁶ Cm	—	—	9.4528e-7	0.005	

^aRead as 1.1631×10^{-4} .

where Σ_x is a nuclide reaction cross section. The sensitivities have an explicit component and an implicit component. The implicit component is due to the effect of perturbations of a reaction cross section on the self-shielding of other cross sections. The implicit component is important, for instance, for ^{238}U self-shielding in water moderated lattices. TSUNAMI-1D treats both implicit and explicit effects and calculates the total sensitivity. For nuclide i , the variation of the total cross section $d\Sigma_{t,i}$ equals

$$d\Sigma_{t,i} = N_i d\sigma_{t,i} + \sigma_{t,i} dN_i, \quad (31)$$

where $d\Sigma_{t,i}$ can be interpreted as either the sensitivity to the nuclear data at constant number density N_i or the sensitivity to the density at constant nuclear data $\sigma_{t,i}$. Substituting the RHS of Eq. (31) into Eq. (30) gives

$$S_{k_{eff}, \Sigma_t} = - \frac{N_i \sigma_{t,i}}{k_{eff}} \frac{\left\langle \phi_0^*, \frac{1}{\sigma_{t,i}} \left[\frac{dL}{dN_i} - \frac{1}{k_{eff}} \frac{dP}{dN_i} \right] \phi_0 \right\rangle}{\left\langle \phi_0^*, \frac{P}{k_{eff}} \phi_0 \right\rangle}. \quad (32)$$

Comparing this to Eq. (10) shows that the weight w_i can be calculated from Eq. (32):

$$w_i = S_{k_{eff}, \Sigma_t} \frac{k_{eff}}{N_i}, \quad (33)$$

allowing for easy evaluation of w_i . TSUNAMI-1D returns the total S_{k_{eff}, Σ_t} ; i.e., the implicit component is taken into account. For GFR600, the calculations have shown that for all actinides, the implicit component is two to three orders of magnitude smaller than the explicit component. The approach of Eqs. (32) and (33) is recommended in the TSUNAMI-1D manual as a way to compare the perturbative calculations with direct calculations; i.e., perform two forward calculations with perturbed initial composition and calculate the sensitivity to $\sigma_{t,i}$ as $\Delta k_{eff} / \Delta N_i$. In our case, we reverse the argument, and take S_{k_{eff}, Σ_t} as a measure of Δk_{eff} brought on by a change ΔN_i .

A burnup calculation code [Like Origen With Forward and Adjoint Transmutation (LOWFAT)] similar to ORIGEN-S but with adjoint capabilities was written to calculate the depletion, cooldown, and reprocessing of the heavy metal 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. All calculations were done over a period of 1300 days irradiation at a constant flux $\phi = 2 \times 10^{15} \text{ n/cm}^2 \cdot \text{s}^{-1}$, leading to a burnup $\approx 6.5\%$ FIMA

(fissions per initial metal atom). This calculational approach is justified as follows: In the closed fuel cycle it is reasonable to assume that the reactor will be restarted with the same Σ_f at beginning of cycle from cycle to cycle. In the closed fuel cycle, the “user” cannot influence Σ_f of the fuel during reprocessing, because the fertile material to be added (mainly ^{238}U) has a negligible fission cross section. Thus, the total fission cross section of the fuel needs to be roughly preserved during irradiation. Since the Generation IV GCFR has no blankets, breeding of new fuel takes place homogeneously in the core, so globally the distribution of fissile material is rather homogeneous during irradiation. Under the assumption that the energy release per fission is comparable for all actinides, this means that a constant power requires constant flux level. Perturbing the amount of fissile nuclides in the reactor will change the power produced by the fuel. As long as the perturbations of Σ_f are not larger than several percent, the change of the power is also only several percent.

Furthermore, the reactivity of the fuel is rather constant during irradiation in the closed fuel cycle. This is due to the roughly constant Σ_f , the absence of blankets, and the fact that the absorption effect of fission products is not very large in a fast reactor. Since the fuel reactivity is rather constant, “zoning” of the fuel to shape the power profile is not very relevant. The investigations reported in this paper concern general scoping studies to close the fuel cycle. Therefore, the simplified calculational approach, based on unit cell calculations with constant flux, is deemed adequate. For more detailed analyses, a fully coupled treatment of the eigenvalue/depletion sensitivity, such as described in Ref. 8, could be beneficial.

After irradiation a cooldown period of 6 yr (2192 days) was assumed. Reprocessing is assumed to take a small amount of time compared to the cooldown period. Reprocessing is assumed to be like PUREX (only U, Pu recycled) or integral, where U, Np, Pu, Am, and Cm are recycled. An example of the corresponding elements S_{ii} of \vec{S} for PUREX with 99% efficiency and integral recycling with 95% efficiency is given in Table IV.

VIII. RESULTS

VIII.A. Comparison with Existing Definitions of \vec{w}

To check the consistency of our definition of \vec{w} with existing definitions, some reactivity weights were calculated for GFR600 and compared to LMFBR values from Ref. 2. The result is given in Table V. The weights in the table are normalized as follows:

$$w_i = \frac{w^+ - w^8}{w^9 - w^8}, \quad (34)$$

TABLE IV

Examples of the Reprocessing Matrix S , for a PUREX-Type Reprocessing with 99% Recovery Efficiency and an Integral Reprocessing with 95% Recovery Efficiency

Nuclide	$S_{ii,PUREX}$	$S_{ii,Integral}$
^{235}U	0.99	0.95
^{238}U	0.99	0.95
^{237}Np	0.0	0.95
^{238}Pu	0.99	0.95
^{239}Pu	0.99	0.95
^{240}Pu	0.99	0.95
^{241}Pu	0.99	0.95
^{242}Pu	0.99	0.95
^{241}Am	0.0	0.95
^{242}Am	0.0	0.95
^{242m}Am	0.0	0.95
^{244}Cm	0.0	0.95

TABLE V

Weights for Various Nuclides from Ref. 2, Calculated for GFR600 Using Eq. (10)*

	From Ref. 2	GFR 600 (Reference)	GFR600 (5% MA)
^{235}U	0.777	0.810	0.799
^{238}U	0.000	0.000	0.000
^{239}Pu	1.000	1.000	1.000
^{240}Pu	0.130	0.144	0.175
^{241}Pu	1.542	1.520	1.457
^{242}Pu	0.032	0.093	0.120

*The GFR600 fuel with MA has a harder spectrum, which is evidenced by the change of weights: The threshold fissioners, like ^{240}Pu , become somewhat more important. Fuel compositions according to Table III.

where

w^+ = weight of a nuclide calculated with Eq. (13)

w^8 = weight of ^{238}U

w^9 = weight of ^{239}Pu .

This definition results in a weight of 0.0 for ^{238}U and 1.0 for ^{239}Pu . The weights calculated for GFR600 using the new definition of Eq. (10) are similar to the ones cited in Ref. 2, giving confidence that our new definition is consistent with existing definitions.

In Table VI we present the weights of various isotopes calculated from Eq. (33). All weights are normalized to ^{239}Pu , i.e., \vec{w} is dimensionless. Note that ^{238}U , ^{237}Np , and ^{241}Am have negative weights ($\sigma_a > \nu\sigma_f$).

TABLE VI

Weights for Various Nuclides Calculated from Eq. (10), Normalized to ^{239}Pu *

	GFR 600 (Reference)	GFR600 (5% MA)
^{235}U	0.794	0.785
^{238}U	-0.079	-0.067
^{237}Np	—	-0.163
^{238}Pu	0.639	0.659
^{239}Pu	1.000	1.000
^{240}Pu	0.077	0.120
^{241}Pu	1.560	1.486
^{242}Pu	0.026	0.063
^{241}Am	-0.368	-0.222
^{242}Am	—	2.140
^{242m}Am	—	2.104
^{244}Cm	—	0.213

*In the somewhat harder spectrum of the fuel with 5% MA, the threshold fissioners like ^{238}U have a higher weight.

Also note that the weight of ^{238}Pu in the GFR600 spectrum is comparable to ^{235}U . The fuel with MA has a harder spectrum, which is evidenced by the change of weights: The threshold fissioner ^{240}Pu becomes somewhat more important.

VIII.B. Illustrations of Adjoint Calculations

Two illustrative examples of adjoint calculations are given in Figs. 3 and 4. Figure 3 shows three adjoints for

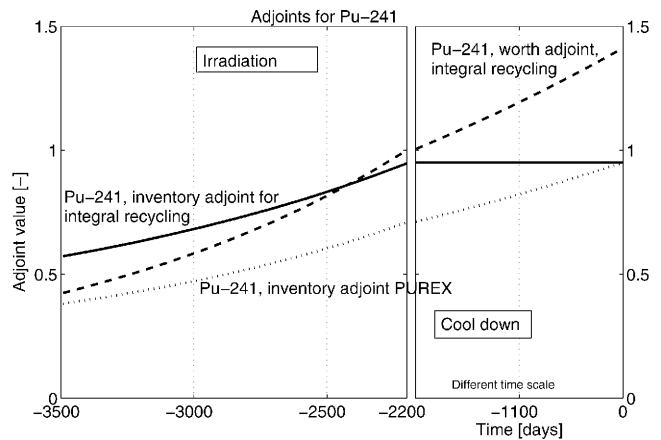


Fig. 3. Three adjoints for ^{241}Pu . The inventory adjoint is a measure of the contribution of ^{241}Pu to the amount of reprocessed material, while the worth adjoint gives the contribution of ^{241}Pu to the reactivity of the reprocessed material. Note the time axis: The cycle length is 1300 days of irradiation and 2192 days of cooldown. Since the adjoint is solved backward in time, we are “looking back” in time.

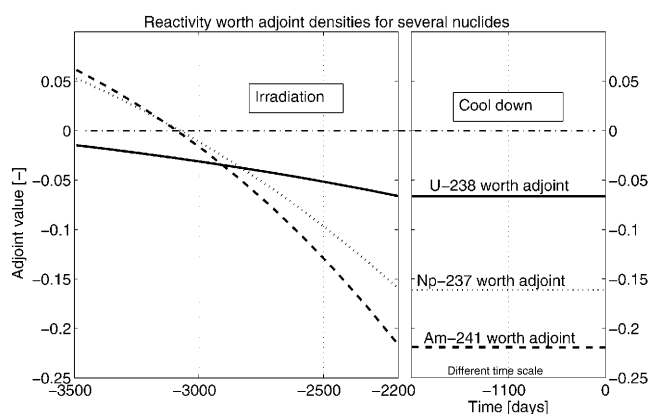


Fig. 4. Worth adjoints for the absorbing nuclides ^{238}U , ^{237}Np , and ^{241}Am , i.e., the contribution of these nuclides (by themselves or daughter isotopes) to the reactivity of the reprocessed fuel. Notice that ^{237}Np and ^{241}Am have a positive contribution if added at the beginning of the irradiation because they transmute to fissile isotopes. The time axis corresponds to a cycle of 1300 days of irradiation and 2192 days of cooldown.

^{241}Pu . Consider the solid line, being the “inventory adjoint,” i.e., the adjoint quantifying changes of the amount of reprocessed material due to ^{241}Pu in an integral fuel cycle. It is the solution of the adjoint transmutation equation using the final time condition of Eq. (25) with \mathcal{S} for integral recycling. In the integral recycling scheme, reprocessed material due to ^{241}Pu is the plutonium itself, plus any daughter products of ^{241}Pu that are also recycled. For example, in the integral reprocessing scheme, the daughter product ^{241}Am is also reprocessed material due to ^{241}Pu , while in the PUREX reprocessing scheme it is not.

During cooldown (the right part of the graph in Fig. 3), the inventory adjoint for ^{241}Pu is flat: ^{241}Pu and the daughter product ^{241}Am are recycled. The final time value is 0.95, because it is assumed that the reprocessing efficiency is 95%. Thus, adding ^{241}Pu during the cooldown period will always lead to more reprocessed material, either as ^{241}Pu or ^{241}Am . In the PUREX reprocessing, on the other hand, only U and Pu are recycled. If ^{241}Pu were to be added at the beginning of the cooldown period, some of it would decay before reprocessing. Since the daughter product is not recycled in this scheme, the inventory adjoint for ^{241}Pu becomes time dependent, as given by the dotted line in Fig. 3, which gives the inventory adjoint for ^{241}Pu in a PUREX fuel cycle.

For both reprocessing schemes, the inventory adjoint is time dependent during irradiation: If ^{241}Pu is added at the beginning of the irradiation period, the probability of fission during the irradiation is quite large. Thus, ^{241}Pu added to the fresh fuel will have a small contribution to the total amount of reprocessed material, and hence it will have a low inventory adjoint at the beginning of the irradiation interval.

In Fig. 3 the “worth adjoint” of ^{241}Pu is given by the dashed line. It is the solution of the adjoint transmutation equation with the final time condition of Eq. (26). This line represents the contribution to the reactivity of the reprocessed material by ^{241}Pu in the integral reprocessing scheme. During irradiation and cooldown this adjoint is time dependent, because in both cases ^{241}Pu has a probability of not surviving until reprocessing, which implies that it does not contribute to the reactivity of the new fuel. The slope of the worth adjoint for ^{241}Pu differs from the slope of the PUREX inventory adjoint because we are in fact solving two different systems: For the PUREX inventory adjoint the final time condition is given in Table IV, while for the worth adjoint the final time condition is given by $\mathcal{S}\bar{w}$.

In Fig. 4 the worth adjoints for some absorbers are illustrated, namely, ^{238}U , ^{237}Np , and ^{241}Am . These lines correspond to the adjoint transmutation equation with final time condition Eq. (26). The worth adjoint represent the contribution to the reactivity of the reprocessed material due to the isotopes in question. Since these nuclides all have long half-lives, the worth adjoints during cooldown are constant. During irradiation the worth adjoints are time dependent. Both ^{237}Np and ^{241}Am have positive worth adjoint values at the beginning of the irradiation. This may seem strange, because these nuclides have negative weights. During irradiation ^{237}Np transmutes to ^{238}Pu , and ^{241}Am transmutes to ^{242m}Am . These products of transmutation have positive weights. Thus, if ^{237}Np and ^{241}Am are added at the beginning of the irradiation, they have enough time to capture a neutron, after which their transmutation products contribute positively to the reactivity of the new fuel made from the reprocessed material. If ^{237}Np and ^{241}Am were to be added at a later stage of the irradiation, the probability of transmutation would be smaller, and their overall contribution to the reprocessed fuel would be negative. The behavior of the worth adjoint for ^{238}U has a similar behavior, but is never positive. This means that under the chosen irradiation conditions, the transmutation to ^{239}Pu does not have enough time to take place on a large enough scale. Here one immediately sees the influence of the final fluence (burnup) on the fuel cycle, because if the final fluence would be larger, more ^{238}U would transmute to ^{239}Pu , and the reactivity worth of the reprocessed material would be larger.

VIII.C. Breeding Gain Calculations on the GFR600 Fuel Cycle

One cycle of irradiation, cooldown, and reprocessing was calculated 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. Refer to Table VII: 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 yr of cooldown, and R_{repro} is

TABLE VII
Evolution of R (cm^{-3}) for the Reference Fuel
and a Fuel with MA*

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

*All values are 10^{-3} unless stated otherwise.

^a R_{ini} = R of initial fuel mixture.

^b R_{irrad} = R after irradiation.

^c R_{cool} = R after 2192 days of cooldown.

^d R_{repro} = R of reprocessed material.

^e R_{feed} = reactivity of the feed material.

^f R_{new} = R of new fuel.

the reactivity of the reprocessed material. The new fuel is composed of reprocessed and feed material [see Eq. (17)]. The R of the feed material is given by R_{feed} and the R of the new fuel is $R_{new} = R_{repro} + R_{feed}$. The R_{feed} is dominated by the weight of ^{238}U , which is negative. With the reference fuel, BG over the irradiation ($R_{irrad}/R_{ini} - 1$) is close to zero, but during cooldown 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 cooldown, resulting in an overall BG close to zero (-3.56%). In conclusion, adding a small amount of MA to the fuel results in a positive BG over the irradiation. 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:

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 it 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 VIII, where the symbols are as before. Table VIII gives the values of R for the reference calculation (same as in Table VII), after which the ΔR introduced by the perturbations are given. For example, adding ^{235}U has a positive ΔR_{repro} : The reactivity of the reprocessed material is higher. Also, ^{235}U has a positive ΔR_{feed} : More ^{235}U means a smaller

TABLE VIII

Results of Two Perturbations on the Evolution of R^*

	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	$2.39\text{e}-6^a$	$2.39\text{e}-7$	^{238}U	$-1.14\text{e}-5$	$4.92\text{e}-5$
^{239}Pu	$5.18\text{e}-5$	$3.86\text{e}-6$			
^{240}Pu	$6.71\text{e}-6$	$2.34\text{e}-6$			
^{241}Pu	$5.08\text{e}-6$	$4.55\text{e}-7$			
$\sum R'_{new}$	$6.60\text{e}-5$	$6.90\text{e}-6$	$\sum R'_{new}$	$-1.14\text{e}-5$	$4.92\text{e}-5$
Forward			Forward		
R_{ini}	1.27		R_{ini}	1.11	
R_{irrad}	1.36		R_{irrad}	1.28	
R_{cool}	1.29		R_{cool}	1.21	
R_{repro}	1.28		R_{repro}	1.20	
R_{feed}	0.09		R_{feed}	0.048	
R_{new}	1.19		R_{new}	1.15	
BG	-6.42%		BG	+3.71%	

*The reference situation is a 5% MA fuel. Two perturbations are applied and their effects calculated. Two forward calculations were done to check the result. All values are 10^{-3} unless stated otherwise.

^aRead as 2.39×10^{-6} .

amount of feed material is added to make the new fuel, and since the weight of the feed material is negative, adding less of it has a positive effect. For the other isotopes similar arguments apply.

The perturbation of ^{238}U (right column of Table VIII) has a negative effect on R_{repro} but a positive effect on R_{feed} . In the two bottom cells of each column of Table VIII, 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 cooldown 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: $+3.71\%$. The equations for a perturbation in ^{238}U are written out as follows:

$$\begin{aligned} \Delta R_{repro} &= N^*(0)\Delta N(0) \\ &= -1.4849 \times 10^{-2}\Delta N(0), \end{aligned} \quad (35)$$

and for ΔR_{feed}

$$\begin{aligned}\Delta R_{feed} &= N^*(0) \langle \vec{w}, v_{feed} \rangle \Delta N(0) \\ &= (0.9669)(6.6423 \times 10^{-2}) \Delta N(0) \\ &= 6.4225 \times 10^{-2} \Delta N(0),\end{aligned}\quad (36)$$

where $\Delta N(0)$ is the perturbation in the initial concentration of ^{238}U . A perturbation in ^{238}U has a much stronger positive effect through the amount of feed material (R_{feed}) than the negative effect on the reprocessed material (R_{repro}): Under the given circumstances, adding extra ^{238}U gives a better new fuel (but the reactivity of the initial fuel is lower).

VIII.D. Long-Term Behavior

To investigate the long-term behavior, a series of 15 irradiation, cooldown, and reprocessing cycles was calculated. Since the cycle length is some 9.5 yr, 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 strategy 2 is given by the fact that R_{repro} is lower if S_{ii} is smaller (i.e., reactivity is lost because of large reprocessing losses), leading to the necessity to increase R_{feed} to make the overall BG closer to zero. In Figs. 5 and 6 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 [Eq. (19)] during irradiation is about +15%. The 95% efficient strategy (strategy 2), which adds some MA to the new fuel, has a BG between +15 and +25% during irradiation to offset the losses of reprocessing. In Table IX the fuel composition for the 10th cycle is given. In both fuel cycles the MA loading of the fuel remains limited. Of the two presented strategies, strategy 1 (refueling with reprocessed material and depleted uranium 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. Strategy 2 requires an external source of MA, but no Pu. Note that the amount of feed material is the same as the burnup reached, so for 6.5% FIMA, the amount of feed material is 6.5% of the core inventory. Adding material with 10% MA means that the overall amount of added MA equals 0.65% of

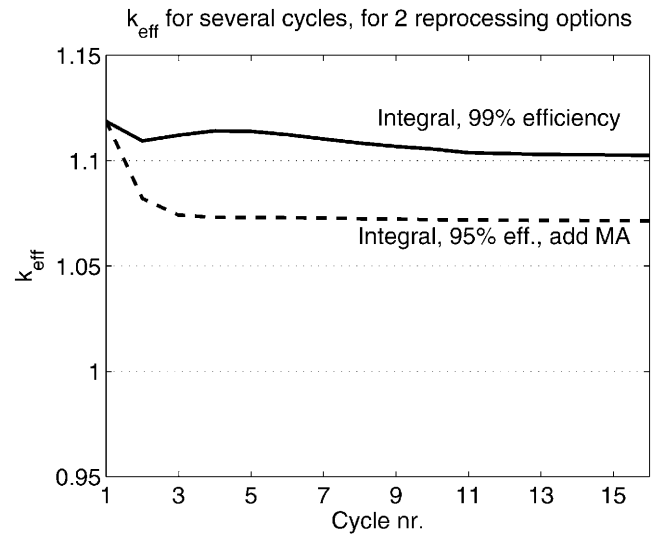


Fig. 5. Evolution of k_{eff} of the fresh fuel during multiple recycles for the two fuel cycle schemes indicated in the text. Both schemes converge to a situation where the k_{eff} is constant from cycle to cycle.

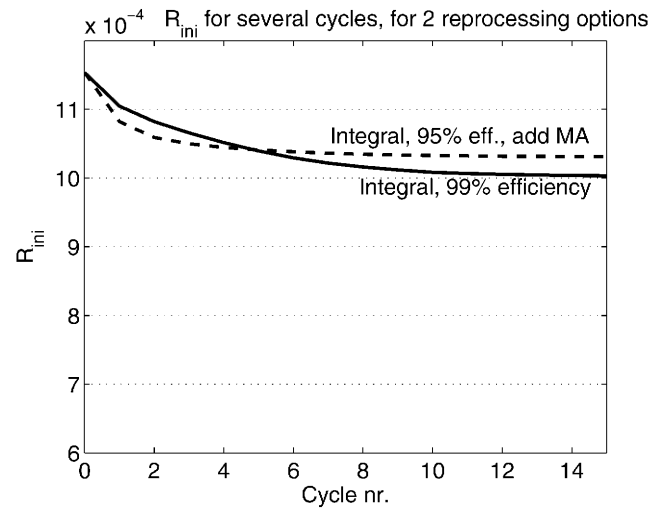


Fig. 6. Evolution of R during multiple recycles for the two fuel cycle schemes indicated in the text. Both schemes converge to a situation where the R_{new} is constant from cycle to cycle. Since R_{new} is the same from cycle to cycle, BG = 0 is approached.

the total core inventory (≈ 105 kg, the core inventory being some 16 tons of heavy metal).

We can thus draw the conclusion that a truly closed fuel cycle is possible if the reprocessing losses are small enough (strategy 1). 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.

TABLE IX

Fuel Composition at the Start of the Tenth Irradiation Cycle for Two Different Reprocessing Strategies*

Isotope	Strategy 1	Strategy 2
U	82.512%	80.78%
²³⁴ U	0.201%	0.331%
²³⁵ U	0.072%	0.091%
²³⁶ U	0.105%	0.086%
²³⁸ U	81.933%	80.272%
Np	0.119%	0.588%
²³⁷ Np	0.119%	0.588%
Pu	16.513%	15.355%
²³⁸ Pu	0.457%	1.173%
²³⁹ Pu	9.258%	8.410%
²⁴⁰ Pu	5.607%	4.648%
²⁴¹ Pu	0.489%	0.384%
²⁴² Pu	0.702%	0.740%
Am	0.838%	2.709%
²⁴¹ Am	0.577%	1.952%
^{242(m)} Am ^a	0.038%	0.106%
²⁴³ Am	0.223%	0.651%
Cm	0.219%	0.568%
²⁴³ Cm	0.003%	0.007%
²⁴⁴ Cm	0.153%	0.419%
²⁴⁵ Cm	0.033%	0.095%
²⁴⁶ Cm	0.023%	0.038%
²⁴⁷ Cm	0.005%	0.007%
²⁴⁸ Cm	0.002%	0.002%

^aThis is ²⁴²Am and ^{242m}Am summed.

*Strategy 1 assumes 99% efficient reprocessing, after which depleted uranium is added. Strategy 2 assumes 95% efficient reprocessing, after which depleted uranium and 10% MA are added. All numbers are in at. %.

VIII.E. Checking the Validity of the Quasi-Static Approximation

With the 15 cycles the validity of Eq. (19) and of the quasi-static approach in Secs. V and VI and the Appendix can be checked. To this end, the BG was calculated for both strategies using two slightly different definitions. The first definition is

$$BG = \frac{\langle \vec{w}_i, \vec{N}_{i+1} \rangle}{\langle \vec{w}_i, \vec{N}_i \rangle} - 1, \quad (37)$$

where i is the cycle number: The same set of weights \vec{w}_i is used to calculate the reactivity of the fuel of batch i and the new fuel of batch $i + 1$. This is the quasi-static

calculation: The weights are kept constant while the nuclide vectors are allowed to vary.

The second definition calculates BG using weights \vec{w}_i of the fuel in batch i , and weights \vec{w}_{i+1} for the new fuel in batch $i + 1$:

$$BG = \frac{\langle \vec{w}_{i+1}, \vec{N}_{i+1} \rangle}{\langle \vec{w}_i, \vec{N}_i \rangle} - 1. \quad (38)$$

If the weights \vec{w} change considerably between cycle i and cycle $i + 1$, the two definitions would result in different values of BG. If both BG definitions give comparable results, it shows that \vec{w} does not change too much from cycle to cycle and that the quasi-static approach is valid. The result is given in Figs. 7 and 8. The BGs from one cycle to the next are all close to zero. It is concluded that, since the BGs calculated with the two equations given above are comparable, the weights \vec{w} do not change much between the cycles and the quasi-static approach is valid for the closed fuel cycle with integral recycling. An explanation is needed for the BG calculated for the first cycles, which are all much lower than the BGs of the later cycles. Please refer to the fifth column of Table III and to Table IX. In strategy 1, with high reprocessing efficiency and adding depleted uranium, the conversion of ²³⁸U to ²³⁹Pu gradually increases because of an increasing ²³⁸U content, while the plutonium content remains more or less stable. Strategy 2, with its high reprocessing losses and adding extra MA, has a higher content of americium and ²³⁷Np, both of which contribute positively to the reactivity of the new fuel, which is beneficial to obtaining a zero BG in the later cycles.

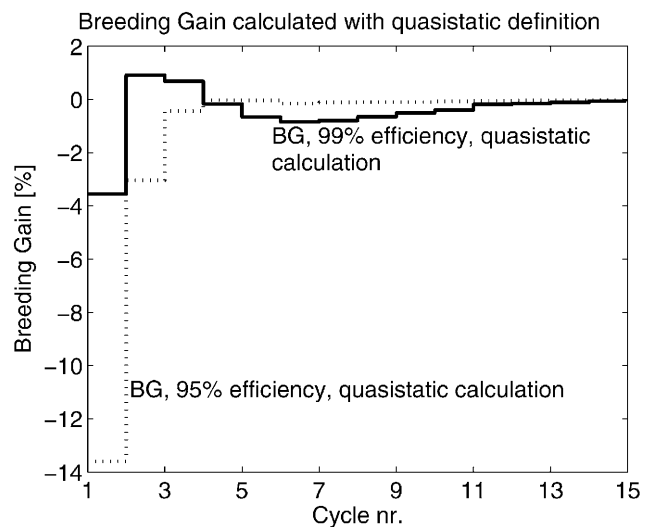


Fig. 7. BG calculated with the quasi-static approximation [Eq. (37)] for the two fuel cycle schemes presented in the text. The low values in the earlier cycles are explained in the text.

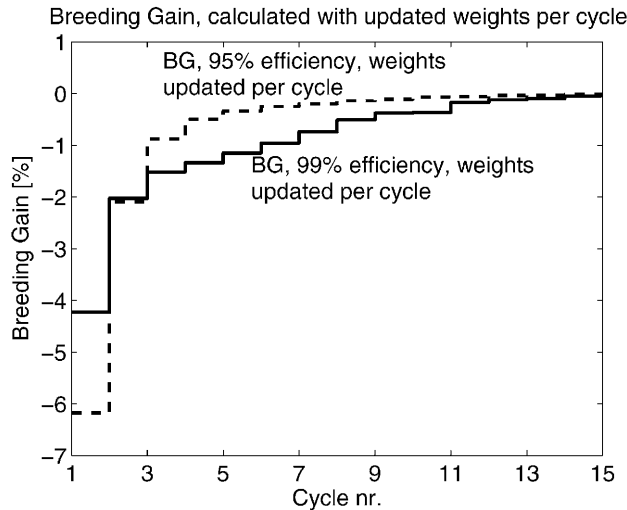


Fig. 8. BG calculated using new weights for each cycle, i.e., Eq. (38) for the two fuel cycle schemes presented in the text. The low values in the earlier cycles are explained in the text.

IX. DISCUSSION AND CONCLUSION

A mathematical framework was derived to describe the change of a nuclear fuel as a function of irradiation, cooldown, and reprocessing. A new definition of BG is proposed, taking into account irradiation, cooldown, and reprocessing. Corresponding reactivity weights \vec{w} are defined. An adjoint formalism is derived to estimate the effect of initial changes of the fuel mixture on the reprocessed fuel, regarding both the total amount of reprocessed material and its reactivity weight. This theory is applied to an integral fuel cycle for a GCFR running on uranium, plutonium, and MA. It is found that a fuel with uranium, plutonium, and 5% MA yields a positive BG during irradiation, and a BG close to zero over the entire cycle of irradiation, cooldown, 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 fuel contains mostly ^{238}U ($\approx 81\%$) and a small portion of Pu ($\approx 15\%$), with the rest comprised of Np, Am, and Cm.

The cell calculations cannot fully capture all effects of changing the nuclide composition before or during irradiation. For example, perturbing the fuel composition can cause power peaks or change the burnup reac-

tivity swing. A fully coupled eigenvalue and nuclide depletion perturbation could capture these effects in more detail. However, the Generation IV GCFR, which has no blankets and a self-breeding target, has special properties. The reactivity of the homogenous fuel stays more or less constant during irradiation, and thereby the spatial flux profile is approximately constant during the irradiation. Zoning becomes somewhat arbitrary. Because of this, the decoupled nuclide perturbation approach is especially suited for the Generation IV GCFR.

APPENDIX

A.I. NUCLIDE PERTURBATION THEORY

In this Appendix nuclide perturbation theory will be developed, following Ref. 9. The transmutation equation is given by

$$\frac{\partial \vec{N}_0}{\partial t} = \underline{M}_0 \vec{N}_0 + \vec{Q}_0, \quad (\text{A.1})$$

with the initial condition

$$\vec{N}_0(t=0) = \vec{N}_i. \quad (\text{A.2})$$

The general response function R is defined as

$$R_0 = \langle \vec{h}, \vec{N}_0 \rangle, \quad (\text{A.3})$$

where \vec{h} is a response selection vector [for instance, \vec{h} can contain $\sigma_a \phi$ at position i to give the time integrated absorption rate of nuclide i , or it can contain a Dirac delta $\delta(t - t_0)$ at position i to give the density of nuclide i at time t_0]; the transmutation matrix \underline{M}_0 contains the nuclear cross sections σ_x , the decay constants λ_x , and the flux ϕ ; \underline{M} is space-time dependent through the cross sections and flux; and \vec{Q}_0 is an independent source term (e.g., adding and extracting material from a tank during reprocessing). This system is an example of a much broader class of functionals and responses for which a perturbation development is possible (see, for instance, Ref. 10) if R is functional of the data and the variable governed by the functional (in this case \vec{N}), but \vec{h} may not be dependent on \vec{N} . In our definition of R in Eq. (1), \vec{w} takes the place of \vec{h} . As seen from Eq. (10), \vec{w} depends on \vec{N} through ϕ , ϕ^* and the normalization factor in the denominator. Neglecting the dependence of \vec{w} on \vec{N} introduces a first-order error, but one that may be small for most variations under consideration. Assume a reference calculation $R_0 = \langle \vec{w}_0, \vec{N}_0 \rangle$ and a small perturbation giving R :

$$\begin{aligned} R &= \langle \vec{w}_0 + \Delta \vec{w}, \vec{N}_0 + \Delta \vec{N} \rangle \\ &= \langle \vec{w}_0, \vec{N}_0 \rangle + \langle \vec{w}_0, \Delta \vec{N} \rangle \\ &\quad + \langle \Delta \vec{w}, \vec{N}_0 \rangle + \langle \Delta \vec{w}, \Delta \vec{N} \rangle. \end{aligned} \quad (\text{A.4})$$

In the first-order approximation, the term $\langle \Delta \vec{w}, \Delta \vec{N} \rangle$ is neglected. Neglecting the term $\langle \Delta \vec{w}, \vec{N}_0 \rangle$ compared to $\langle \vec{w}_0, \Delta \vec{N} \rangle$ is in fact comparable to the quasi-static approximation used in burnup calculations, where it is assumed that the nuclide density \vec{N} changes (much) quicker as a function of the neutron fluence ϕt than the equivalent one-group cross sections σ_x appearing in \underline{M}_0 . The validity of the quasi-static approximation is checked in Sec. VIII.E. To fully treat the variation in R due to $\Delta \vec{N}$, generalized perturbation theory must be used, such as illustrated in Ref. 8. For the current study, a simplified approach is deemed instructive and sufficient.

The initial condition for Eq. (A.1) can be removed by adding the initial condition of Eq. (A.2) to Eq. (A.1) as a delta function:

$$\frac{\partial \vec{N}_0}{\partial t} = \underline{M}_0 \vec{N}_0 + [\vec{N}_i \delta(t) + \vec{Q}_0] , \quad (\text{A.5})$$

with initial condition

$$\vec{N}_0(t=0) = 0 . \quad (\text{A.6})$$

The initial condition is treated as an extra term to source \vec{Q}_0 . Now assume that we have some perturbation in the independent source: $\vec{Q} = \vec{Q}_0 + \Delta \vec{Q}$, resulting in perturbed densities \vec{N} . The perturbed system is given by

$$\frac{\partial \vec{N}}{\partial t} = \underline{M}_0 \vec{N} + (\vec{Q}_0 + \Delta \vec{Q}) , \quad (\text{A.7})$$

and the corresponding perturbed response

$$R = \langle \vec{h}, \vec{N} \rangle . \quad (\text{A.8})$$

Using the general properties of adjoint equations, the adjoint to Eq. (A.1) is defined as

$$\frac{-\partial \vec{N}_0^*}{\partial t} = \underline{M}_0^* \vec{N}_0^* + \vec{Q}_0^* . \quad (\text{A.9})$$

It can be shown, e.g., as in Ref. 10, that in order to obtain a meaningful interpretation of the adjoint equations, the adjoint boundary conditions and source must be chosen as

$$\vec{N}_0^*(t=t_f) = 0 \quad (\text{A.10})$$

and

$$\vec{Q}_0^* = \frac{\partial R}{\partial \vec{N}} = \vec{h} . \quad (\text{A.11})$$

Now form the scalar products of Eq. (A.7) with \vec{N}_0^* , form the scalar products of Eq. (A.9) with \vec{N} , insert Eq. (A.11) and subtract:

$$\left\langle \vec{N}_0^*, \frac{\partial \vec{N}}{\partial t} \right\rangle = \langle \vec{N}_0^*, \underline{M}_0 \vec{N} \rangle + \langle \vec{N}_0^*, (\vec{Q}_0 + \Delta \vec{Q}) \rangle ,$$

$$\left\langle \vec{N}, \frac{-\partial \vec{N}_0^*}{\partial t} \right\rangle = \langle \vec{N}, \underline{M}_0^* \vec{N}_0^* \rangle + \langle \vec{N}, \vec{Q}_0^* \rangle ,$$

and

$$\begin{aligned} & \left\langle \vec{N}_0^*, \frac{\partial \vec{N}}{\partial t} \right\rangle + \left\langle \vec{N}, \frac{\partial \vec{N}_0^*}{\partial t} \right\rangle \\ &= \langle \vec{N}_0^*, (\vec{Q}_0 + \Delta \vec{Q}) \rangle - \langle \vec{N}, \vec{h} \rangle . \end{aligned} \quad (\text{A.12})$$

Here the adjoint property $\langle \vec{N}_0^*, \underline{M}_0 \vec{N} \rangle = \langle \vec{N}, \underline{M}_0^* \vec{N}_0^* \rangle$ is used. The left side can be written as $(d/dt)(\vec{N}(t)\vec{N}_0^*(t))$ and integrated as indicated by the \langle, \rangle operator:

$$\begin{aligned} \int_0^{t_f} \frac{d}{dt} (\vec{N}(t)\vec{N}_0^*(t)) dt &= \vec{N}(t_f)\vec{N}_0^*(t_f) \\ &- \vec{N}(0)\vec{N}_0^*(0) = 0 , \end{aligned} \quad (\text{A.13})$$

because of the boundary conditions $\vec{N}(t=0) = 0$ and $\vec{N}_0^*(t=t_f) = 0$. What remains is

$$\begin{aligned} \langle \vec{N}_0^*, (\vec{Q}_0 + \Delta \vec{Q}) \rangle &= \langle \vec{N}, \vec{h} \rangle \\ &= \langle \vec{N}_0, \vec{h} \rangle + \langle \Delta \vec{N}, \vec{h} \rangle = R_0 + \Delta R , \end{aligned}$$

and thus

$$\Delta R = \langle \vec{N}_0^*, \Delta \vec{Q} \rangle . \quad (\text{A.14})$$

This is an exact relation between a perturbation of the source \vec{Q} and the resulting perturbation of the response R . Until now, \vec{h} and $\Delta \vec{Q}$ were completely arbitrary. Remember that according to Eq. (A.5) a perturbation in the initial condition can be written as a perturbation to the source term:

$$\Delta \vec{Q} = \Delta \vec{N}_0 \delta(t) . \quad (\text{A.15})$$

Then, from Eq. (A.14), the resulting change in response at the final time t_f is given by

$$\Delta R = [\vec{N}_0^*(t=0) \Delta \vec{N}_0]_x , \quad (\text{A.16})$$

where the $[]_x$ indicates that the integration is over the entire phase space except t . From Eq. (A.16) the change in a final time response caused by a perturbation of the initial conditions can be readily calculated.

Equation (A.14) gives ΔR for a perturbation of the source \vec{Q} and/or the initial condition. Another possible perturbation is a perturbation of the nuclear data appearing in the transmutation matrix \underline{M}_0 ; \underline{M} will lead to a perturbed solution \vec{N} , but the initial conditions $\vec{N}_0(t=0)$ are not affected. Inserting $\underline{M} = \underline{M}_0 + \Delta \underline{M}$, $\vec{N} = \vec{N}_0 + \Delta \vec{N}$ into Eq. (A.1) gives

$$\frac{\partial}{\partial t} (\vec{N}_0 + \Delta\vec{N}) = (\underline{M}_0 + \Delta\underline{M})(\vec{N}_0 + \Delta\vec{N}) + \vec{Q}_0, \quad (\text{A.17})$$

which is expanded to

$$\begin{aligned} \frac{\partial \vec{N}_0}{\partial t} + \frac{\partial \Delta\vec{N}}{\partial t} &= \underline{M}_0 \vec{N}_0 + \vec{Q}_0 + \vec{N}_0 \Delta\underline{M} \\ &+ \Delta\vec{N} \underline{M}_0 + \Delta\vec{N} \Delta\underline{M}, \end{aligned} \quad (\text{A.18})$$

from which Eq. (A.1) can be subtracted to give

$$\frac{\partial \Delta\vec{N}}{\partial t} = \Delta\underline{M} \vec{N}_0 + \underline{M}_0 \Delta\vec{N} + \Delta\underline{M} \Delta\vec{N}. \quad (\text{A.19})$$

Now forming the same scalar products as in Eq. (A.12) leads to a left side $(d/dt)(\vec{N}_0^*(t)\Delta\vec{N}(t))$, which can be integrated to give zero because of the boundary conditions $\vec{N}_0^*(t = t_f) = 0$ and $\Delta\vec{N}(t = 0) = 0$. Neglecting the second-order term $\Delta\underline{M}\Delta\vec{N}$, ΔR becomes

$$\Delta R = \langle \vec{N}_0^*, \Delta\underline{M} \vec{N}_0 \rangle. \quad (\text{A.20})$$

Taking together the effects of a change in source \vec{Q} , initial condition $\vec{N}_0(0)$, and \underline{M} gives

$$\begin{aligned} \Delta R &= [\vec{N}_0^*(t = 0) \Delta\vec{N}_0]_x \\ &+ \int_0^{t_f} \vec{N}_0^* [\Delta\vec{Q} + \Delta\underline{M} \vec{N}_0]_x dt. \end{aligned} \quad (\text{A.21})$$

In the scope of this work, we are dealing with the final time response R as defined by Eq. (17), which can be written as a sum of two final time responses involving \vec{N}_{cool} [Eq. (18)]. Thus, from Eq. (A.21) only the first term on the right side remains for each response, and we find that we have to solve two adjoint calculations to correctly estimate the perturbation of R caused by perturbations to the initial nuclide inventory. First, the perturbation to R_{repro} , the total reactivity of the reprocessed material, must be calculated. Since R_{repro} is given by

$$R_{repro} = \langle \vec{w}, \underline{S} \vec{N}_{cool} \rangle, \quad (\text{A.22})$$

the corresponding adjoint problem to evaluate ΔR_{repro} is defined by the final condition:

$$\vec{N}_0^*(t = t_f) = \underline{S} \vec{w}. \quad (\text{A.23})$$

Second, the amount of feed material \vec{N}_{feed} is perturbed, leading to a perturbation in R_{feed} . Since R_{feed} is given by

$$R_{feed} \propto -N_{feed} = -|\underline{S} \vec{N}|, \quad (\text{A.24})$$

the corresponding adjoint problem is defined by the final condition:

$$\vec{N}_0^*(t = t_f) = \underline{S} \vec{I}. \quad (\text{A.25})$$

Note that it is assumed that \vec{S} and \vec{v}_{feed} , the reprocessing efficiency and vector of feed material, respectively, do not change because of the perturbation. Also note that in these calculations the flux ϕ , and not the

power, is kept constant, so the final burnup also changes as a function of composition.

ACKNOWLEDGMENTS

The authors would like to acknowledge the support of the European Commission. The GCFR Specific Targeted Research Project (STREP) is carried out under contract 012773 (FI6O) within the EURATOM Sixth Framework Programme (<http://www.cordis.lu/fp6/>), effective from March 1, 2005 to February 28, 2009. More information about the GCFR STREP is available from the project website at <http://www.gcfr.org>.

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