Impact of MA content on breeding gain definition for innovative fast reactor fuel

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Abstract

In the frame of partitioning and transmutation (P&T) strategies, some well-known key parameters, like the conversion ratio (CR), the breeding gain (BG) and the doubling time (DT) impacting on fuel cycle logistics have been revisited for their applicability to critical "burner" fast reactors loaded with a high fraction of minor actinides (MA).

This work focuses on the analysis of the available definitions for the total BG. Two main approaches could be distinguished: the "American approach", establishing a mass balance between the initial and the final reactor states (e.g. begin and end of cycle), and the "European approach", paying additional attention to the reactivity balance between the same states. These approaches have been applied to the analysis of two fast burner reactor concepts developed at KIT, loaded with different MA/Pu ratios (MA/Pu = 0.1 and MA/Pu = 1).

The analysis shows that all the classical definitions could be applied also to high MA content innovative fuels, even though additional attention has to be devoted to their interpretation in order to draw appropriate conclusions and assessments from fuel cycle analysis. Further activities for a more detailed analysis of these key parameters will be the equilibrium cycle condition evaluation (analysing their impact on BG and DT values).

Introduction

In the frame of partitioning and transmutation (P&T) strategies, where innovative fuels and systems are considered to burn transuranics (TRU) mass discharged from conventional thermal reactors, the analysis of the applicability of the existing definitions to characterise the breeding or burning capability of a system could help in order to draw appropriate conclusions and assessments from fuel cycle analyses.

The aim of this work is to present an overview of the existing definitions of the breeding gain (BG), the breeding ratio (BR) and the conversion ratio (CR) and to analyse the impact of their application to critical fast "burner" reactors loaded with a high content of minor actinides (MA).

Low CR critical burner fast reactor concepts loaded with different MA/Pu ratios are investigated at KIT [1] as possible alternatives to accelerator driven systems in a so-called "doublestrata" strategy developed in order to manage spent fuel (SF) inventories of European countries. The interest in this kind of system is to show the high flexibility of fast reactor technology to revert the core characteristics from "breeder" through "iso-generator" to "burner", as it has been shown within the CAPRA international programme in the early 90s [2].

For the fuel cycle logistic, one of the main parameters characterising a fast reactor is the doubling time (DT – linear in case of a single reactor or composite in case of reactor fleet) that is the time needed to produce a sufficient amount of fissile fuel (by conversion of fertile material loaded into the reactor, e.g. in the blanket) to build up a new identical core. The DT is defined on the basis of BG, a parameter that gives a measure of the fuel produced in excess by the system during a so called fuel cycle. For this reason, the BG definitions have been analysed in depth.

For defining the BG, two approaches could be distinguished: the "American approach", which takes into account integral quantities establishing a mass balance between the reactor initial and final states [i.e. beginning of life (BOL) and end of life (EOL)], and the "European approach", more oriented towards "point in time" quantities and the reactivity balance between the same states. Whereas in the early definitions, fuel components have been simply distinguished as "fissile" and "fertile" (e.g. for mixed-oxide fuel ^{239}Pu, ^{241}Pu are fissile and ^{238}U, ^{240}Pu are fertile nuclides), refined definitions take additionally into account the contribution of each isotope to the reactivity of the system (by means of reactivity equivalence coefficients).

Historically the definition of BG, BR/CR and DT have been formulated for fuel containing conventional mixed-oxide (MOX) where only uranium and plutonium are included [e.g. typical for the LWR spent fuel or the fuel self-recycled in fast reactors (FR)]. In advanced fuel cycles, the introduction of variable fractions of MA (in homogeneous and heterogeneous modes) still has to be carefully analysed. Therefore, the applicability of the existing definitions of BG, CR, DT has to be analysed too.

An accurate neutronic assessment of fast reactors loaded with a high content of MA (~50%), shows that the computation of BG, BR, DT could be performed using basically the same formalisms as adopted in the past for MOX fuel, but one should be careful when analysing results in order to avoid misinterpretations. All these aspects will be discussed in detail on the basis of the results obtained by their application to critical burner concepts developed at KIT. Two burners with the same CR (~0.8) and, respectively, loaded with 10% of MA (MA/Pu = 0.1) and 50% of MA (MA/Pu = 1) have been chosen for this comparison.

Overview of breeding gain definitions

For the breeding gain definitions, two main approaches could be distinguished: an integral approach based on the mass balance between initial (e.g. BOL) and final states (e.g. EOL) and a punctual or "point in time" approach based on the reactivity balance between the same states (where reactivity equivalence coefficients are applied to take into account the contribution of each isotope to the system's reactivity). In general, two conceptual definitions for the BG could be found in literature. The first one, "standard" Eq. (1), considers the surplus of the fissile isotopes

produced by the systems over the total destruction rate (the variant is called "British" when the fission rate is considered for the normalisation [3]). The second one, called "French", considers the surplus of the fissile isotopes produced but just scaled on the ^{239}Pu equivalent [Eq. (2)] [4]:

$$BG_{stand.} = \frac{surplus - production - rate - of - fissile}{destruction - rate - of - fissile} \quad (1)$$

$$BG_{French} = \frac{(production - minus - absorption - rate) - of - ^{239}Puequivalent}{fission - rate} \quad (2)$$

They differ only in the way how to calculate the surplus of fissile material (normalised or not on ^{239}Pu equivalent). The French definition seems more convenient when different fuels (with different Pu vectors) generated by thermal reactors are compared.

The approach based on mass balance is unambiguous once the fuel has been defined (fissile vs. fertile isotopes); meanwhile the approach based on reactivity balance is affected by the fuel cycle logistic considered. As indicated in [5], the reprocessing scheme and the equilibrium vs. non-equilibrium operation regimes could influence the breeding capability of a system and they have to be taken into account for the BG and DT evaluation. Therefore, only the quantities obtained in the equilibrium operation regime (which is the property of the reactor alone), give the proper basis to compare and characterise the breeding capabilities, otherwise the value obtained is strongly influenced by the composition of feed fuel from the stockpile, and it becomes a figure of merit for the fresh fuel composition [5,6]. At the same time, the asymptotic BG (or equilibrium BG) has not a clear definition too [7]. In fact, it could be defined at least by four different relations (considering the reactor-park definition, the detailed single reactor calculation, the integrated continuous model for a single reactor, the isotopic breeding worth) that lead to different results. Of course these differences are not negligible in the transition scenarios from a thermal reactor to a FR fleet (where the doubling time is a crucial parameter for the strategy to adopt).

All these considerations have been underlined by the analysis of the critical burner concepts developed at KIT [1] and modelled by means of the ERANOS neutronic code [8]. In particular, mass vs. reactivity approaches are considered as well as the decay contribution of the isotopes (e.g. ^{239}Np, $t_{1/2}$ = 2.3565 days, ^{242g}Am, $t_{1/2}$ = 16.02 hours and ^{242}Cm, $t_{1/2}$ = 162.8 days.

Another figure of merit associated to the BG is the breeding ratio (BR), see Eq. (3). It gives a measure of the system conversion capability. Historically it is called BR when it exceeds 1 (net fissile production) and CR when it is less than 1 (net fissile concentration reduced). This equation is respected by the mass balance approach but it could result in misinterpretation if it is considered to be valid always (see below the comparison among burners).

$$BG_{stand.} = 1 + BG_{stand.} \quad (3)$$

The above concepts could be made explicit by adopting both the reactivity equivalence coefficients ("European approach") and the masses ("American approach"), as will be shown in the following sections.

The integral approach oriented to mass balance

The integral approach makes a mass balance between two states: BOL and EOL, but also over a single irradiation cycle (beginning of cycle, BOC, and end of cycle, EOC).

Starting from the CR definition (the ratio of fissile material produced to fissile material destroyed), that is applicable to thermal and fast reactors; it is possible to define the BG as follows. According to [7,9], the definition of the CR (or BR), applied to an equilibrium fuel cycle, could be expressed as in Eq. (4), where FP indicates the fissile material produced per cycle and FD the fissile material destroyed per cycle, respectively, where FP has the following contributions: FD, FEOC (fissile inventory in the core and blankets at the EOC) and FBOC (fissile inventory in the core and blankets at BOC).

$$BR = \frac{FP}{FD} = \frac{FD + FEOC - FBOC}{FD} \tag{4}$$

From this formulation, it is clear that the BR is averaged over a fuel cycle, where all the fissile material produced (expressed in kg or in atoms/barn*cm [7]), including the fissile material destroyed in the cycle itself, is taken into account. The BG has been defined [Eq. (5)] to evaluate the net fissile amount produced during the cycle (i.e. the fissile material available for loading another reactor). It is, indeed, the ratio of the fissile material gained per cycle (FG) over the fissile material destroyed in the cycle (FD). By this approach, the well known relation [Eq. (3)] between BG and BR is preserved.

$$BG = \frac{FEOC - FBOC}{FD} = \frac{FG}{FD} \tag{5}$$

Based on this approach, the fast critical burner CR has been evaluated. In particular, ^{238}U, ^{238}Pu and ^{240}Pu are the fertile isotopes considered and ^{235}U, ^{239}Pu and ^{241}Pu are the fissile isotopes considered. In detail, the formulation adopted [Eq. (6)], considering the capture (TCA) and the absorption (TAB) rates calculated by the ERANOS code over the cycle, takes into account also the contribution of the decay (rates calculated on the basis of the Bateman equation).

$$CR \cong \frac{TCA_{U238} + TCA_{Pu238} + TCA_{Pu240}}{TAB_{U235} + TAB_{Pu239} + TAB_{Pu241}} \tag{6}$$

Applying Eq. (6) to the two burner concepts presented in the section following the next one, a CR~0.8 has been obtained for MA/Pu = 0.1 and MA/Pu = 1 cases. This value (<1) confirms that the system is a burner reactor. The expected BG in both cases is ~(-0.2) but the values provided by ERANOS are not always in agreement with that expected value (see following paragraphs).

In order to take into account different isotopes reactivity properties, an additional step (introduced in Ref. [10]) is to consider a breeding ratio based on ^{239}Pu equivalent fuel, where, for instance, the contribution of each isotope to the system reactivity, instead of simply lumping all the fissile isotopes together, is taken into account to evaluate the critical mass of the system [10,11]. This additional approach leads to the BG formulation based on the reactivity balance.

The punctual or point in time approach oriented to reactivity balance

In order to take into account the contribution of each isotope to the reactivity of the system, a set of weighting coefficients (called ω-values) has been defined. They represent the weights in terms of ^{239}Pu equivalent reactivity of each isotope contained in the fuel (historically defined for MOX fuel) evaluated by comparing the system neutronic balance at criticality (fixing the total fuel mass and the material buckling, B_m^2) with a systems loaded only with ^{239}Pu [4,11]. A reactivity scale, where ^{239}Pu and ^{238}U have, respectively, $\omega = 1$ and $\omega = 0$, is obtained on the basis of Eq. (7) [4,11]:

$$\omega_i^n = \frac{\left(\nu\sigma_f - \sigma_a\right)_i^n - \left(\nu\sigma_f - \sigma_a\right)_8^n}{\left(\nu\sigma_f - \sigma_a\right)_9^n - \left(\nu\sigma_f - \sigma_a\right)_8^n} = \frac{\sigma_{i,n}^+ - \sigma_{8,n}^+}{\sigma_{9,n}^+ - \sigma_{8,n}^+} \tag{7}$$

The extension to fuel containing MA could be performed and the contribution of each isotope (in particular Cm isotopes) could be taken into account. One of the key points is that these weights enable to determine a doubling time of critical mass material independent of the specific plutonium isotopic composition [7]. Different weights can be defined in order to take into account the different contributions to the reactivity originating from fuel containing isotopes due to transition and equilibrium FR loading, as it will finally be the case in a closed FR fuel cycle. Some examples may be found in [7,12].

The reactivity coefficients depend on the reactor zone considered (due to their dependence on the neutron spectra); therefore, the total BG is obtained by the sum of the contributions from each reactor zone (e.g. different fuel zones and/or blankets). The “standard” definition [Eq. (1)] could be expressed as in Eq. (8) [3].

$$BG_{stand.} = \frac{\sum_n \sum_i \omega_i^n \left(C_{i-1}^n - A_i^n\right)}{\sum_n \sum_i \omega_i^n A_i^n} \tag{8}$$

The "French" formulation [in Eq. (9)] differs from the "standard" one only for the "cut" adopted in the burn-up chain, affecting not so significantly the total BG value for conventional MOX fuel:

$$BG_{French} = \frac{\sum_n \sum_i \left(C_i^n \omega_{i+1}^n - A_i^n \omega_i^n\right)}{\sum_n \sum_i F_i^n} = \frac{\sum_n \sum_i \left(C_i^n \left(\omega_{i+1}^n - \omega_i^n\right) - F_i^n \omega_i^n\right)}{\sum_n \sum_i F_i^n} \tag{9}$$

In both these formulations, "A_i^n", "F_i^n", "C_i^n", "ω_i^n" indicate, respectively, the absorption (capture plus fission), the fission, and the capture rates, and the reactivity equivalent coefficients of the nuclide "i" in region "n".

These expressions do not consider the contribution of the decay (they have been formulated originally for conventional MOX fuel where this contribution is not so significant at least for reasonably short cooling, reprocessing and re-fabrication times). A revised formulation of the "French" BG [Eq. (10)] has been implemented in the ERANOS code [8]: a conversion rate (C_i^n including decay) and a disappearance rate (D_i^n including decay) are adopted instead of the capture and the absorption rates. This modification has a significant impact when fuels containing short-lived nuclides ($^{239}Np \rightarrow {}^{239}Pu$, $^{241}Pu \rightarrow {}^{241}Am$, $^{242g}Am^* \rightarrow {}^{242}Cm$ and ^{242}Pu) are taken into account, as it will be shown for the fast burner reactors case:

$$BG_{ERANOS} = \frac{\sum_n \sum_i \left(C_i^n \omega_{c(i)}^n - D_i^n \omega_i^n\right)}{\sum_n \sum_i F_i^n} \tag{10}$$

The characterisation of the fast critical burners

In order to show the high flexibility of fast reactor technology, several 1 000 MWth fast critical MA burner cores cooled by sodium were investigated at KIT [1]. These cores (with different CR, ~0.5 and ~0.8, oxide and metal) were modelled by means of the ERANOS code [8] on the basis of previous studies at Argonne National Laboratory [13]. The JEF-2.2 nuclear library was adopted [14].

For the analysis of the BG, two cores with CR~0.8 and loaded with $(U\text{-}TRU)O_2$ fuels have been selected (main parameters listed in Table 1). Respectively, two different MA to Pu ratios (MA/Pu) have been considered, respectively MA/Pu ~0.1 and ~1 (isotopic compositions listed in Table 2). These two values are representative of discharged LWR fuels (MA/Pu = 0.1) and of TRU fuels from multi-recycling Pu fuels (MA/Pu = 1) [13]. Further details about the models are given in Refs. [1,15]. Table 3, obtained by applying the burn-up chain presented in Figure 1, shows that the core with MA/Pu = 0.1 has a TRU consumption rate of 13.23 kg/TWh (mainly due to the reduction of ^{239}Pu and of ^{241}Pu) whereas the core with MA/Pu = 1 has a rate of 26.65 kg/TWh (mainly due to the reduction of ^{241}Am and of ^{243}Am). This indicates that from the point of view of the mass balance both reactors are good TRU burners [1,15].

Fast critical burners: Comparison of BG and CR approaches

For the two fast burner reactors previously described, a BG, of the order of ~ (-0.2) according to Eq. (3), was expected. For the case of MA/Pu = 0.1 the BG value provided by the application of the Eq. (10) is in agreement with the expected value (BG ~-0.262). On the contrary, for the case of MA/Pu = 1 the BG value obtained is equal to 0.691. To explain this positive value, several definitions have been applied and the reactivity loss for each isotope has been evaluated too.

* Ground state.

Table 1: Main design parameters for the two burner cores considered [1,15]

Parameters	Fast reactor burner	
Fuel type	$(U\text{-}TRU)O_2$	
MA/Pu	~0.1	~1
Conversion ratio	0.73	0.75
Cycle length (EFPD)	353	353
Average TRU content (%)	27.1	41.2
Power (GWth)	1.0	1.0
Average discharge burn-up (MWd/kgHM)	149	117
Reactivity loss (%$\Delta\rho$) over the 1st cycle	-4.5	-0.6

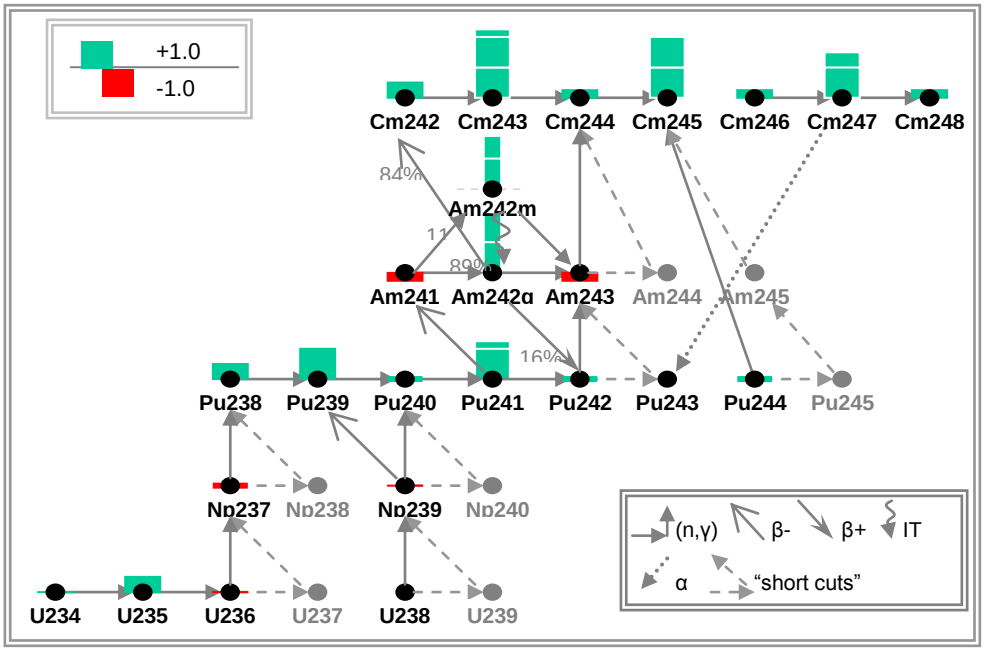
Table 2: Isotopic compositions (wt.%) for MA/Pu = 0.1 and MA/Pu = 1 fuels [13]

Isotope	MA/Pu = 0.1	MA/Pu = 1
	Content (wt.%)	Content (wt.%)
^{237}Np	4.8	7.3
^{238}Pu	2.3	2.0
^{239}Pu	47.9	18.2
^{240}Pu	22.5	13.4
^{241}Pu	10.6	5.8
^{242}Pu	6.5	10.6
^{241}Am	3.4	18.8
^{242m}Am	0.0	0.1
^{243}Am	1.5	15.9
^{243}Cm	0.0	0.1
^{244}Cm	0.5	7.0
^{245}Cm	0.0	0.8
Pu	89.8	50.0
MA	10.2	50.0

Table 3: U and TRU consumption (kg/TWh) per cycle for the MA/Pu = 0.1 and MA/Pu = 1 cores [1,15]

Isotope	MA/Pu = 0.1	MA/Pu = 1
	kg/TWh	kg/TWh
^{235}U	-0.31	-0.27
^{238}U	-29.44	-22.59
^{238}Pu	1.13	2.32
^{239}Pu	-8.61	-0.72
^{240}Pu	1.94	-2.01
^{241}Pu	-7.07	-6.78
^{242}Pu	0.53	-0.18
^{241}Am	-0.27	-14.83
^{242m}Am	0.01	1.24
^{243}Am	0.15	-10.29
^{237}Np	-2.75	-5.35
^{242}Cm	0.83	5.24
^{243}Cm	0.02	-0.06
^{244}Cm	0.85	4.21
^{245}Cm	0.02	0.55
Total TRU	-13.23	-26.65

Figure 1: MA/Pu = 1; ω-values comparison: burn-up chain implemented in the ERANOS code



Due to the fact that there are no significant differences between the "standard" and the "French" BG definitions, only the "French" formulation has been compared with the ERANOS one in order to evaluate the contribution of the decay to the BG.

As indicated in Tables 4 and 5 (respectively showing results for the total value and the isotopic contributions in a chosen reactor zone), the impact of the decay on the BG evaluation becomes important when a high fraction of MA is loaded into the fuel. Analysing Tables 4 and 5, it is clear that the contribution of the short-lived nuclides to the total BG evaluation is significant in both cases (e.g. the decay of ^{239}Np into ^{239}Pu; if it is neglected the ^{239}Pu positive contribution to the reactivity of the system is lost). In addition, the MA fraction could affect significantly the BG value. In fact, for MA/Pu = 0.1 the French and ERANOS formulations give negative BG that differ only for the contribution of the decay of the ^{239}Np (the ERANOS one is in reasonably good agreement with the value obtained by the mass balance approach). For MA/Pu = 1 the two values are strongly positive (see Table 4). A burner (as indicated in Table 3) with a positive BG seems at first sight to provide contradictory indications.

The positive BG for the MA/Pu = 1 case is explained on the basis of the isotopic burning capability of the system. In fact, looking at Table 3, the MA/Pu = 1 reactor burns high quantities of ^{241}Am and ^{243}Am (both isotopes give a "negative reactivity" contribution, $\omega < 0$, to the system, see Table 5). These isotopes produce (by capture and decay) daughters that give a "positive reactivity" contribution, $\omega > 0$, to the system. One example is the ^{243}Am ($\omega < 0$) that is transmuted to ^{244}Cm ($\omega > 0$). ^{243}Am is loaded into the fuel with a fraction comparable to ^{239}Pu (16% vs. 18%) and it is burned for 10.29 kg/TWh (see Table 3). At BOL it gives a negative contribution to reactivity otherwise at EOC it provides by its transmutation products a positive contribution. The processes followed by the isotopes are indicated in Figure 1, where the positive ω-values are represented in green and the negative ones in red (a column of more than 2 boxes stacked on top of each other meaning an ω-value > 2.0, see e.g. ^{243}Cm). In the same figure, the burn-up chain implemented in ERANOS is represented too. The global effect of the MA transmutation in the MA/Pu = 1 burner core is the very small reactivity loss at EOL respect to the MA/Pu = 0.1 case (see Table 1).

Table 4: Total BG evaluated by the French (no decay) and the ERANOS formulations

MA/Pu = 0.1		MA/Pu = 1	
BG treatment	**BG**	**BG treatment**	**BG**
French	-0.98220	French	0.82255
ERANOS	-0.26183	ERANOS	0.69127

Table 5: Contribution to the total BG of each isotope for one (zone 1) of the fuel zones composing the core

Isotope	MA/Pu = 0.1			MA/Pu = 1		
	Omega	BG treatment		Omega	BG treatment	
		French	ERANOS		French	ERANOS
^{235}U	0.852	-0.00078	-0.00078	0.684	-0.00066	-0.00066
^{238}U	0.000	-0.03332	-0.03332	0.000	-0.00602	-0.00602
^{237}Np	-0.373	0.00652	0.00652	-0.132	0.01329	0.01329
^{239}Np	-0.455	0.00005	0.10456	-0.094	0.00002	0.06942
^{238}Pu	0.524	-0.00046	-0.00062	0.645	-0.00234	-0.00272
^{239}Pu	1.000	-0.09702	-0.09704	1.000	-0.06964	-0.06965
^{240}Pu	0.058	0.02205	0.02205	0.183	0.01043	0.01042
^{241}Pu	1.604	-0.04189	-0.05583	1.305	-0.03275	-0.04494
^{242}Pu	0.040	-0.00148	-0.00148	0.127	-0.00277	-0.00277
^{241}Am	-0.442	0.03331	0.03335	-0.218	0.21588	0.21607
^{242g}Am	2.405	-0.00001	-0.01208	1.963	-0.00005	-0.07197
^{242m}Am	2.361	-0.00031	-0.00032	1.905	-0.00290	-0.00297
^{243}Am	-0.424	0.00147	0.00147	-0.200	0.01954	0.01955
^{242}Cm	0.227	0.00019	-0.00059	0.409	0.00066	-0.01176
^{243}Cm	2.634	-0.00003	-0.00003	2.273	-0.00126	-0.00142
^{244}Cm	0.125	0.00026	0.00025	0.279	0.01122	0.00868
^{245}Cm	2.612	-0.00002	-0.00002	2.138	-0.00922	-0.01034
Total_zone1		-0.11145	-0.03391		0.14343	0.11222

All the results furthermore presented have been determined on the basis of a reference burn-up chain originally implemented in the ERANOS code for this study (see Figure 1). A more refined burn-up chain [that neglects the contribution of the isotopes loaded into the fuel with very low fractions (e.g. ^{244}Pu) and takes into account the decay contribution of all the MA isotopes] has been applied. The results obtained differ more or less appreciably but they enable to reach the same conclusion: for MA/Pu = 0.1 the BG is equal to -0.28967 instead of -0.26183 and for MA/Pu = 1, it is equal to 0.52348 instead of 0.69127.

Conclusions

The detailed investigation of parameters like BG, BR and DT could help to avoid erroneous conclusions in the framework of innovative fuel cycle analyses. With this work a preliminary revision of the definitions has been performed in order to understand in detail the parameters necessary to characterise a fast burner reactor.

The definitions historically proposed may be applied also to innovative fuels (like 50% of MA content) even though additional attention to treat them properly has to be used. In particular, the BG defined on the basis of the reactivity balance ("European approach') indicates that a burner with CR ~0.8 and high content of MA can have a positive BG, if it is calculated by means of the reactivity balance approach, since that system transmutes isotopes like ^{241}Am and ^{243}Am that have negative contributions to the reactivity ($\omega < 0$) into isotopes with positive omegas.

The two approaches presented here ("American" and "European") are apparently consistent only when limited quantities of MA are present in the core (see MA/Pu = 0.1 case), but when high contents of MA are considered, the use of the usual correlation between BG and BR (i.e. BG = BR-1) is no longer applicable. In fact, for high MA content, the BR evaluated by the mass balance approach and BG by the reactivity balance approach cannot be correlated by this simple relation as shown in the paper in particular for the MA/Pu = 1 case.

Further activities for a more detailed analysis of these figures of merit will be the equilibrium cycle analysis and the evaluation of a revised reactivity scale of the ω-values for innovative fuels. For this last point, as indicated in Ref. [5], an explicit multi-group first order perturbation formulation will be applied and the decay contribution will be taken into account explicitly for the ω-values evaluation.

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