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The Physics of TRU Transmutation – A Systematic Approach to the Intercomparison of Systems

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In this collaborative effort, a methodology is developed to enable systematic analysis and comparison of diverse nuclear fuel cycle strategies. First, transmutation potential is assessed by considering the neutron balance for destruction of each actinide isotope; a range of thermal and fast reactor systems are considered. In general, a harder neutron energy spectrum results in a more favorable neutron balance. The method is extended to compute equilibrium actinide compositions for a generalized fuel cycle model (open or closed). In this paper, the technique is employed to compare the transmutation performance of 1) PWRs with varying moderator-to-fuel ratio, 2) PWR closed cycle strategies with varying treatment of the minor actinide elements, and 3) fast reactors with either plutonium or transuranic recycle. The method is demonstrated to quickly evaluate the main characteristics of the associated fuel cycles (e.g., isotopic mass flows, neutron balance for critical enrichment) and give some indication of other performance parameters (e.g., reactivity effects).

KEYWORDS: transmutation, nuclear fuel cycle, transuranics

1. INTRODUCTION

The management of transuranics (TRU) is a major challenge for the future of nuclear power development. A variety of strategies have been proposed (see for example Refs. 1 and 2), which span from Plutonium (Pu) management in thermal spectrum reactors with the remaining minor actinides (MA) being sent to the waste, to unseparated TRU (intimate mixture of Pu and MA) management both in thermal and fast neutron spectra reactors. Selective management of MA (e.g., transmutation targets) has also been considered as an option.

For many of the proposed approaches, there is a diverse body of existing international studies; however, these studies tend to provide detailed analysis of specific options, focused on concepts favored by the authors. A consistent comparison of transmutation options requires a synthesis of these previous results, *and* targeted analysis to assess the performance for a common set of criteria.

In order to gain insight into the relative merits, a collaborative study has been initiated to intercompare on physics grounds the characteristics of different systems dedicated to transmutation. A first attempt in this direction was made in Ref. 3, where a novel global physics approach was proposed to compare the neutron balance for diverse transmutation systems. The original analyses showed that fast neutron spectrum systems have an advantage in terms of

neutron balance over thermal neutron systems. In this joint study, a similar approach, as described in Section 2, is employed to intercompare different types of thermal and fast reactors (see Section 4); and the scope is expanded to consider other key transmutation criteria.

The physics approach is also extended here, and in order to intercompare the systems from the fuel cycle point of view the notion of "equilibrium" is applied to solve the standard Bateman equations, but generalising them in order to account for different strategies, e.g. open cycle, selective TRU reprocessing, different type of fuel feeds to the reactors, different burn-ups, etc. The extended E-method for computing equilibrium composition is explained in Section 3; and specific fuel cycle scenarios are compared in Section 5.

2. THE PHYSICS APPROACH - THE D-METHOD

Let us consider an infinitely large homogeneous core (with a given neutron flux Φ) that is fed by several actinides with a given rate S (nuclides/s) and that there is a continuous discharge of part of its fuel inventory for reprocessing and/or storage in a repository. The processes of transmutation of the incoming nuclides (the "fathers") under neutron flux according to their nuclear chains gives rise to "families" which are independent. Hence, one can consider the "transmutation behaviour" of each "family" separately. During its life in a core, the father and his family are producing neutrons up to their "death" (complete disappearance). The reasons for the nuclide destruction can be:

- nuclide fission,
- natural decay,
- fuel cycle procedures, either discharge to storage or fuel losses during reprocessing.

One can calculate how many neutrons each father (together with his family) is able to consume/produce during its irradiation in a core with a chosen fuel cycle. Multiple nuclear reactions lead to different branches of family members: some of these reactions consume neutrons through neutron capture (e.g., (n,γ) reactions), other paths produce more neutrons (e.g., fission, (n,2n) reactions) than consumed, and some (e.g., (n,n) reactions, decay) do not influence the total neutron balance.

The total number of neutrons D_j consumed by the given J-family can serve as an indicator of the capability of a core to achieve destruction of a given J-feed if there is neither "parasitic" neutron consumption nor neutron leakage. In the case of a negative D (i.e., when the J-family produces more neutrons than it consumes), the core fed by J-nuclides produces enough neutrons to destroy the source material at equilibrium if the neutron excess compensates for parasitic captures (e.g. by structures, fission products etc) and for neutron leakage. In the case of a positive D, the neutron consumption in the fuel dominates over neutron production and the core requires a supplementary neutron source to support transmutation.

The procedure of the D_j calculation (Ref. 3) can be illustrated by the following scheme:

$$D_j = \sum_{J1i} P_{j\to J1_i} \left\{ R_{J\to J1_i} + \sum_{J2i} P_{J1\to J2_i} x \left[R_{J1\to J12_k} + \sum_{J3n} P_{J2_k\to J3_n}(..) \right] \right\} \qquad (1)$$

where $P_{JNr\to J(N+1)s}$ is the probability of transmutation of the nuclide J_{JNr} (which belongs to the number N of the chain J) into nuclide J(N+1)s (which belongs to the number N+1 of the chain J). All these nuclides are the members of the J-family. $R_{A\to B}$ is the neutron consumption factor showing the number of neutrons consumed during transition A→B. The neutron consumption values for each reaction type are defined as follows:

- neutron capture (n,γ) with **1** neutron being captured,
- neutron capture and subsequent multiplication (n,mn) with (**1-m**) neutrons being produced,
- fission with (**1-ν**) neutrons being produced,
- natural decay with **0** neutron being captured,
- discharge and nuclide loss with **0** neutron being captured.

Two approximations are used to simplify the practical implementation of D-calculations: 1) "effective" one-group cross-sections are obtained from a unit-cell (LWR) or infinite medium (fast system with average composition) model of the given system, and 2) continuous feed-discharge models are employed (even for solid fuels) avoiding possible temporarily transitions of the fuel compositions, taking however into account the main (average) fuel transmutations under neutron flux.

The linearity of the nuclide concentration equation with respect to the fuel feed allows a very simple algorithm of the D-evaluation for cores loaded with different "father" nuclides simultaneously. Taking into account that all feed components (families) are irradiated independently, one gets the total neutron consumption of the core at equilibrium (D_{fuel}) as

$$D_{fuel} = \sum_J \varepsilon_J D_J \tag{2}$$

where ε_J is the fraction of J-family in the feed stream.

The D-factor accounts only for the neutron balance of the actinide "families" as modelled in Eq. 1. However, the global neutron balance of a core must consider the total neutron production (consumption) of the fuel families, the parasitic captures of other core components (C_{par}) and of the accumulated fission products (C_{FP}) and neutron leakage (L_{core}). The general equation for the Neutron Surplus (NS_{core}) then becomes:

$$- D_{fuel} - C_{par} - C_{FP} - L_{core} = NS_{core} \tag{3}$$

with all quantities of Eq. 3 normalized to one disappeared actinide nuclide of the fuel feed. For a critical system, the neutron surplus must equal zero; and the neutron production of the fuel feed (as calculated in Eq. 2) must be sufficient to overcome the other loss terms. Thus, D_{fuel} is a useful parameter for quantifying the transmutation potential of a given isotopic fuel mix.

3. THE "EQUILIBRIUM" E-METHOD

The standard Bateman equations can be generalized to account for the characteristics of specific fuel cycles and solved for the equilibrium concentrations of the actinide nuclides. This method computes the equilibrium composition and associated global neutron production potential of a simplified model of the nuclear reactor core if all "external" conditions (such as the neutron flux level, the fuel feed, the discharge and reprocessing rates) stay unchanged during sufficient time. Under these conditions, the actinide concentrations and their proportions in a core approach a unique time-independent equilibrium. From this "equilibrium" composition, a variety of transmutation parameters can be derived such as the radiotoxicity and decay heat in the discharged fuel (wastes).

If a core is fed by a source S_j of J-nuclides, under neutron flux Φ, the fuel concentrations N_J the J-family are time-dependent and described by the following matrix-type - GFC equation:

$$\frac{\overline{\partial N_J}}{\partial t} = \left(\hat{M}\times\Phi - \hat{\Lambda}_n - \hat{\Lambda}_{dl}\right)\overline{N}_J - \overline{S}_J \tag{4}$$

where $\bar{S}_J$ is the nuclei feed vector with a single non-zero component J, and $\bar{N}_J$ is the vector of the atomic concentrations of the J-family members including its "father". $\hat{M}$ is the matrix related to all nuclear interaction processes which is composed of the one-group absorption cross section on the diagonal and off-diagonal terms representing the transmutation to different isotopes.

The $\hat{\Lambda}_n$ and $\hat{\Lambda}_{dl}$ terms in Eq. 4 are matrices of the family natural decays and of fuel discharge-losses. The λ_{dl} elements of the matrix $\hat{\Lambda}_{dl}$ are defined by the ratio of nuclides loss (the transfer to a repository or reprocessing technology wastes, nuclear decay during fuel "cooling" time interval τ_{cool}) and it depends also upon the discharge "frequency" τ_{core}. A general expression λ_{dl} for each nuclide and for all fuel cycles is given by:

$$\lambda_{dl} = \frac{1}{\tau_{core}}\{\text{loss} + (1-\text{loss})\,[1-\exp(-\lambda_n\ \tau_{cool})]\} \tag{5}$$

where "loss" could be assessed as the fraction of the nuclide inventory which is lost during reprocessing or send to a repository (i.e. which does not return back to core) and the term $(1-\text{loss})\,[1-\exp(-\lambda_n\ \tau_{cool})]$ describes the isotopic decay during storage and processing. This expression, being applied to each nuclide "individually", allows a different set of fuel cycle assumptions (e.g., reprocessing recovery factor) to be applied to each specific nuclide.

An open (once-through) fuel cycle for a nuclide means that this nuclide, being discharged, is sent to a repository and its loss term is equal to 1. A closed fuel cycle for a given nuclide, means that this nuclide, after cooling and inevitable losses due to natural decay and after reprocessing with the relatively small losses, will return back to the core (for example, loss = 0.001 if 0.10 % of this nuclide is lost during fuel reprocessing and 99.9 % is returning back to the core). Transition (due to the natural decay of a family during cooling time) to other nuclides can be taken into account by a matrix-type representation of the "operator" λ_n in Eq. 5.

In the case of a fuel feed consisting of several fathers, the corresponding set of equations (each with the form of Eq. 4) has a similar matrix structure. However its source vector consists of several non-zero components. The general solution of these equations is time-dependent and of an exponential type. The asymptotic ($t\to\infty$) solution (if all operators are time independent) corresponds to the "equilibrium" case i.e. when $\frac{\partial N}{\partial t} = 0$ in Eq. 4. This equilibrium solution can be presented in the following matrix form, $\bar{N} = \hat{A}^{-1}\,\bar{S}$ where $\bar{S}$ includes all "fathers" nuclei contained in the core feed and $\hat{A} \equiv \hat{M} + \hat{\Lambda}_n + \hat{\Lambda}_{dl}$ as defined for Eq. 4. The linear character of the equation allows evaluation of the nuclear concentrations of all families independently, with a "unit" source for the corresponding feed: $S_J^{unit} = 1$.

For this equilibrium actinide concentration in the core, the neutron balance can be computed for a specified composition consisting of i-components with proportions x_i :

$$-D_{eq}^{fuel} = \bar{\nu} - 1 - \bar{\alpha} \ \text{(neutron/fission)} \tag{6}$$

where

$$\bar{\nu} = \frac{\sum_i x_i\ \nu_i\ \sigma_i^f}{\sum_i x_i\ \sigma_i^f}; \qquad \bar{\alpha} = \frac{\sum_i x_i\ \sigma_i^c}{\sum_i x_i\ \sigma_i^f}$$

It is important to note that the neutron balance computed from the equilibrium composition using Eq. 6 will be identical to that computed for the fuel feed using the D-factor method (Eq. 2); and the consistency of the two methods was verified for identical fuel cycle models. However, the E-

method also yields the equilibrium composition which can be used to derive transmutation performance parameters and derived core parameters as shown in Section 5.

4. NEUTRON BALANCE (D-FACTOR) INTERCOMPARISON

The physics approach in Section 2 has been applied to a large variety of reactors, both with thermal and fast spectra. First we have compared a standard PWR (moderator-to-fuel ratio = 2) with MOX fuel with variation in the LWR moderator-to-fuel ratio, r, and also fast reactors with differing fuels and coolants. The D values for major actinides in Table 1 are for a closed fuel cycle for each individual TRU isotope with repeated recycle (no fuel cycle losses). In the case of LWRs, there is a significant dependence of the D_i on the level of the flux. This is also shown in Table 1, where the standard PWR D_i values are shown for two different flux levels ($1x10^{14}$ and $2.5x10^{14}$ n/cm^2.s). This effect is due to the competition between absorption rates, which depend on the flux level, and decay (independent of the flux level).

Table 1. D (neutron consumption/fission) value for different isotopes in different systems

Isotope	MOX-LWR[1] r[2] = 1.4	MOX-LWR[1] r[2] = 2	MOX-LWR[4] r[2] = 2	MOX-LWR[1] r[2] = 4	He-cooled carbide fuel FR[3]	SUPER-PHENIX[3]	Lead-cooled nitride fuel FR[3]	Na-cooled oxide fuel FR[3]	Na-cooled metal fuel FR[3]
U-235	-0.31	-0.38	-0.43	-0.55	-0.84	-0.86	-0.92	-0.95	-1.04
U-238	0.104	0.068	-0.06	-0.007	-0.63	-0.62	-0.71	-0.79	-0.90
Np-237	0.91	0.93	0.75	0.96	-0.51	-0.56	-0.65	-0.73	-0.88
Pu-238	0.014	0.024	-0.16	0.038	-1.25	-1.33	-1.36	-1.41	-1.50
Pu-239	-0.60	-0.64	-0.79	-0.73	-1.44	-1.46	-1.58	-1.61	-1.71
Pu-240	0.65	0.56	0.14	0.38	-0.93	-0.91	-1.02	-1.13	-1.27
Pu-241	-0.26	-0.37	-0.80	-0.58	-1.25	-1.21	-1.26	-1.33	-1.39
Pu-242	1.27	1.22	0.73	1.13	-0.65	-0.48	-0.73	-0.92	-1.13
Am-241	0.92	0.93	0.71	0.95	-0.56	-0.54	-0.65	-0.77	-0.91
Am-242m	-1.55	-1.56	-1.66	-1.56	-2.03	-1.87	-2.08	-2.10	-2.16
Am-243	0.44	0.36	-0.15	0.25	-0.84	-0.65	-0.85	-1.01	-1.15
Cm-242	0.004	0.014	-0.18	0.026	-1.26	-1.34	-1.37	-1.41	-1.51
Cm-244	-0.51	-0.60	-1.12	-0.71	-1.54	-1.44	-1.53	-1.64	-1.71
Cm-245	-2.46	-2.46	-2.44	-2.44	-2.70	-2.69	-2.71	-2.74	-2.77

(1) $\varnothing = 1 \times 10^{14}$ n/cm^2.s
(2) r = moderator-to-fuel ratio
(3) $\varnothing = 1 \times 10^{15}$ n/cm^2.s – Sodium-cooled burner configuration, GFR and LFR conventional concepts
(4) $\varnothing = 2.5 \times 10^{14}$ n/cm^2.s

The results allow comparison of the feasibility of transmutation of the different isotopes in each reactor concept. As an example, in the case of the Am isotopes, the Am-241 transmutation is a neutron-consuming process in any LWR concept, relatively independent from the moderator-to-fuel ratio (r). For a flux value of $1x10^{14}$ n/cm^{2}s, Am-243 is also a neutron-consuming process in a LWR, but a higher r value is preferable because the corresponding D value is positive but smaller than with lower values of r. However, at $\phi=2.5x10^{14}$ n/cm^{2}s, the transmutation of Am-243 becomes a neutron-production process. Am-242 transmutation is a neutron-production process, whatever the spectrum, with a slight advantage for fast neutron spectra.

Significant variations in the D-factor are observed between the fast reactor concepts. In general, a harder neutron spectrum leads to a more favorable neutron balance; thus, the metal-fueled SFR provides the most excess neutrons for every actinide isotope. However, all of the fast reactor systems exhibit a significantly more favorable neutron balance compared to the MOX-

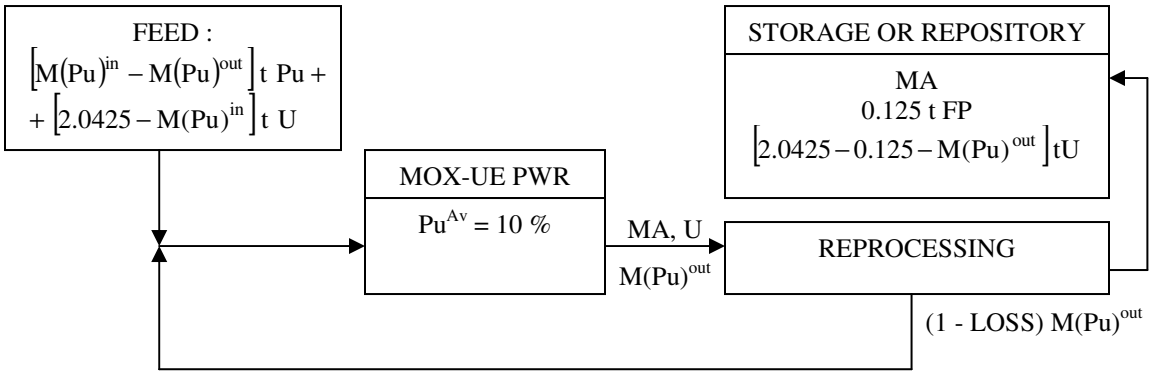
LWR results. Therefore, these results confirm the trends observed in Ref. 3 that fast neutron spectrum systems have a marked advantage in terms of neutron balance over thermal systems.

5. INTERCOMPARISON OF SPECIFIC FUEL CYCLE SYSTEMS

The physics approach described in Section 3 has also been applied to a large variety of fuel cycle strategies. Three example scenarios are given to illustrate the comparison results.

5.1 Pu multirecycling in LWRs

In this case, plutonium is multirecycled in PWRs using the MOX-UE concept[4]. Minor actinides (MA) are separated at the end of each cycle and sent to a repository, as illustrated in Figure 1. It is assumed that Uranium is discharged and not further recycled. The mass of U+Pu loaded in the core is 2.0425 t/TWhe and the mass of fission products built-up is 0.125 t/TWhe. In this strategy, the average Pu content of fresh fuel is fixed at the maximum targeted value of 10 % and U-235 is added to maintain criticality; the average burnup is set at 60 GWd/MT. The feed requirement at equilibrium is chosen such that the fixed Pu content (10 %) is preserved. The U-235 enrichment in the feed is determined by the criticality condition given in Eq. 3 with a total loss of neutrons per fission ($L_{core} + C_{par} + C_{FP}$) estimated at 0.38; thus, a $D_{fuel}^{eq} \simeq 0.38$ is needed.



$M(Pu)^{in}$ = mass (t) of loaded Pu
$M(Pu)^{out}$ = mass (t) of unloaded Pu at the end of the equilibrium cycle
Pu^{Av} = average Pu content in the core
LOSS = 0.1 %

Figure 1 Scheme for Pu multirecycling in a PWR with a burnup equal to 60 GWd/t

The D_{fuel}^{eq} is obtained from Eq. 6, and the x_i are obtained from the solution of the generalized Bateman equation. For example, the corresponding feed at equilibrium is ~ 3 % Pu and 97 % U enriched with ~ 5 % U-235 in the case of a moderator-to-fuel ratio equal to 2. When mixed with the recovered Pu, this results in a 10% Pu content in the fresh MOX fuel. Inventories of all transuranic isotopes can be obtained directly from the solution of the matrix form of Eq. 4 and applying the scheme of Figure 1.

The neutron balance and mass flow results are shown in Table 2 for the same three moderator-to-fuel ratios evaluated in Table 1. By design as described above, the D-factor for the fuel feed is –0.38 and the average plutonium enrichment is 10%. The results show an increase in Pu consumption by a factor of ~3 from 38 kg/TWh to 108 kg/TWh as the moderator-to-fuel ratio is increased. However, the MA build-up roughly doubles from 13.9 to 24.8 kg/TWh as the moderator-to-fuel ratio is increased; the Cm build-up is particularly evident (10.6 kg/TWh) when

the neutron spectrum is overmoderated (r=4). Overall, the ratio of the quantity of Pu burned over that of MA produced is clearly in favor of high moderator-to-fuel ratio. The feed values also show the reduced need for U-235 enrichment in cores with high moderator-to-fuel ratios.

Table 2. Plutonium multirecycling in MOX-UE PWRs

	Closed cycle for Pu - Average Pu content 10 %		
	r = 1.4	r = 2	r = 4
Pu/U+Pu in Feed	2.00 %	3.03 %	5.67 %
U5/U in Feed	5.90 %	4.90 %	2.53 %
D_{fuel}	-0.380	-0.380	-0.380
D_{fuel}^{void}	-0.416	-0.316	+0.016
$M(Pu)^{in}$ (kg/TWhe)	217	226	252
$M(Pu)^{out}$ (kg/TWhe)	179	170	144
ΔM(Pu) (kg/TWhe)	38	56	108
$Pu^{(in)}$ (%)	10.6 %	11.1 %	12.3 %
$Pu^{(out)}$ (%)	8.78 %	8.30 %	7.06 %
$MA^{(in)}$ (kg/TWhe)	0	0	0
$MA^{(out)}$ (kg/TWhe)	13.9	17.1	24.8
$Np^{(out)}$ (kg/TWhe)	1.50	1.38	0.901
$Am^{(out)}$ (kg/TWhe)	8.40	9.89	13.4
$Cm^{(out)}$ (kg/TWhe)	3.97	5.87	10.6
ΔM(Pu)/ΔM(MA)	2.73	3.27	4.35

r = moderator-to-fuel ratio

To provide an indicator of the impact of different strategies on the void reactivity coefficient, neutron balance factors were also generated using one-group cross-sections corresponding to the coolant-voided condition. The expected trends are observed with a slight decrease in the neutron surplus (from –0.38 to –0.32) for the standard PWR lattice corresponding to the 10% plutonium loading limit. However, for the undermoderated lattice (r=1.4) the more negative D-factor in the voided case is a clear indicator of a potential positive void reactivity coefficient.

5.2 Pu and MA multirecycling in LWRs

A similar fuel cycle approach to that shown in Fig. 1 was applied for multirecycling of the minor actinide species in addition to the Pu in a MOX-UE PWR with moderator-to-fuel ratio of two. Successive cases were analyzed with Pu+Np, Pu+Np+Am, and Pu+Np+Am+Cm recycle. In each case, the average content of Pu+MA in the core was set at 10 % (7.7 % Pu and 2.3 % MA when all MA are recycled : 0.3 % Np, 0.6 % Am and 1.4 % Cm). The results are shown in Table 3.

To counterbalance the "poison" effect of MA, a higher percentage of U-235 is needed with Np and Am recycle; the enrichment steadily increases from 4.9% with Pu recycle (Table 2) to 6.45% (Pu+Np) to 9.0% (Pu+Np+Am). A reverse trend is observed (down to 6.45% U-235 enrichment) when the Cm is also recycled. However, *note that the MA inventory is dominated by Cm when all MA elements are recycled;* and this fuel would be very difficult to handle. The Pu consumption is drastically reduced from 56 kg/TWh for Pu recycle to 43 with Np recycle to ~20 with Pu+MA recycle, and 0 with Pu+Np+Am. This behaviour is attributed to the transmutation of some of the MA isotopes (especially Am-241 and Np-237) into Pu isotopes. The minor actinide production is reduced from 17 kg/TWh (Table 2) to 10 with Np recycle and 4 in the Am recycle case. However, only with full MA recycle can the MA inventory be stabilized, with the in-core destruction balanced by the Pu-241 decay source during recycle. However, as indicated previously the high Cm content of this case will severely complicate fuel handling.

Another important indicator of the feasibility of the different recycling options is given by the Pu vector at equilibrium, as shown in Table 4. An inspection of the results obtained with the E-method, indicates a strong increase of the Pu-238 content when any MA are recycled; and the fuel cycle consequences of associated increases in decay heat must be investigated.

Table 3. Pu+MA multirecycling in MOX-UE PWRs

	Closed cycle for Pu+Np Average Pu+Np content ~10 % MOX-UE with r = 2	Closed cycle for Pu+Np+Am Average Pu+Np+Am content ~10 % MOX-UE with r = 2	Closed cycle for Pu+MA Average Pu+MA content 10 % MOX-UE with r = 2
Pu/(U+Pu+MA) in Feed	2.31%	1.64%	1.10 %
MA/(U+Pu+MA) in Feed	0.17%	0.22%	0.16 %
U5/U in Feed	6.45%	9.0 %	6.45 %
D_{fuel}	-0.380	-0.380	-0.380
D_{fuel}^{void}	-0.343	-0.397	-0.299
$M(Pu)^{in}$ (kg/TWhe)	211	190	163
$M(Pu)^{out}$ (kg/TWhe)	168	189	142
ΔM(Pu) (kg/TWhe)	43	1	21
$Pu^{(in)}$ (%)	10.3%	9.3%	8.0 %
$Pu^{(out)}$ (%)	8.2%	9.3%	6.9 %
$MA^{(in)}$ (kg/TWhe)	10.5	26.2	47.7
$MA^{(out)}$ (kg/TWhe)	20.4	30.4	44.5
$Np^{(in)}$ (kg/TWhe)	10.5	10.1	7.2
$Np^{(out)}$ (kg/TWhe)	7.1	7.7	5.6
$Am^{(in)}$ (kg/TWhe)	0.0	16.2	13.1
$Am^{(out)}$ (kg/TWhe)	8.4	14.2	11.8
$Cm^{(in)}$ (kg/TWhe)	0.0	0.0	27.3
$Cm^{(out)}$ (kg/TWhe)	4.9	8.5	27.1
ΔM(Pu)/ΔM(MA)	4.34	0.24	-6.56

r = moderator-to-fuel ratio

Table 4. Plutonium Vectors at Equilibrium (%) in the MOX-UE cases with r=2

Pu isotope	Closed cycle for Pu	Closed cycle for Pu+Np	Closed cycle for Pu+Np+Am	Closed cycle for Pu+MA
Pu-238	7.5	12.8	16.6	15.1
Pu-239	34.3	32.7	33.9	35.6
Pu-240	24.9	22.2	22.0	22.5
Pu-241	16.1	14.1	13.6	13.6
Pu-242	17.2	14.5	13.9	13.1

With regard to the void reactivity effect, both the Np and Np+Am recycle cases indicate a larger neutron surplus compared to the Pu recycle case (see Table 2). In particular, the Pu+Np+Am recycle case clearly indicates a positive void reactivity coefficient. For the complete recycle of Pu+MA, it appears that the decrease of the Pu content and the relevant content of Cm, counterbalance the effect of Am-241 (increase of the positive void coefficient) with the net result that the void reactivity coefficient is similar to the Pu recycle case.

5.3 Pu and Pu+MA recycling in fast reactors

The following cases have been investigated in the case of fast reactors:

- Pu and Pu+MA multirecycling in a sodium-cooled oxide fuelled burner reactor, as considered in Section 4. The feed is a mixture of Pu and natural U.
- Pu and Pu+MA multirecycling in the sodium-cooled SUPERPHENIX-type reactor self-sustaining regime with a Pu (or Pu+MA) recycle. The feed is natural U.

The results are summarized in Table 5. Significant Pu burning (53 kg/TWhe) is obtained in the burner case together with a limited MA production (6.75 kg/TWhe). These features can be compared with the LWR cases (Table 2). In the case of standard MOX-UE, a comparable Pu consumption (56 kg/TWhe) induces a much higher (factor ~ 3) MA production, in particular Cm. The higher Pu consumption of a MOX-UE with r=4 (108 kg/TWhe), induces an even higher MA production, and Cm production is increased by more than a factor 10 compared to the Pu-Burner fast reactor case.

Table 5. Pu and Pu+MA multirecycling in Fast Reactors

	Oxide Na-cooled burner Average Pu content : 35 %	Oxide Na-cooled burner Average Pu+MA content : 35 %	SUPERPHENIX-type Self-sustaining Pu regime Average Pu content : 16.1 %	SUPERPHENIX-type Self-sustaining Pu+MA regime Average Pu+MA content : 16.9 %
Pu/(Pu+U+MA) in Feed	54.3 %	42.1 %	0	0
MA/(Pu+U+MA) in Feed	0%	6.0%	0	0
D_{fuel}	-1.162	-1.089	-0.796	-0.801
D_{voided}	-1.252	-1.190	-0.914	-0.919
$M(Pu)^{in}$ (kg/TWhe)	286	247	127	124
$M(Pu)^{out}$ (kg/TWhe)	233	206	127	124
$\Delta M(Pu)$ (kg/TWhe)	53	41	0	0
$Pu^{(in)}$ (%)	36,2%	31,3%	16,1%	15,7%
$Pu^{(out)}$ (%)	29,5%	26,2%	16,1%	15,7%
$MA^{(in)}$ (kg/TWhe)	0	35,8	0	4,69
$MA^{(out)}$ (kg/TWhe)	6,75	29,9	1,36	4,69
$Np^{(in)}$ (kg/TWhe)	0	7,19	0	0,939
$Np^{(out)}$ (kg/TWhe)	0,354	4,27	0,389	0,939
$Am^{(in)}$ (kg/TWhe)	0	18,4	0	2,77
$Am^{(out)}$ (kg/TWhe)	5,47	16,0	0,875	2,77
$Cm^{(in)}$ (kg/TWhe)	0	10,1	0	0,979
$Cm^{(out)}$ (kg/TWhe)	0,930	9,69	0,097	0,979
$\Delta M(Pu)/\Delta M(MA)$	7.85	-6.96	0	0

In the case of a Pu+MA burner, the fast reactor considered can burn 41 kg/TWhe of Pu and 5.9 kg/TWhe of MA. In the corresponding case, a MOX-UE with r=2 (Table 3), the Pu consumption is drastically reduced (21 kg/TWhe) and the MA consumption is only 3.2 kg/TWhe. The Cm inventory is higher by a factor of ~ 3 in the MOX-UE with respect to the fast burner case.

An important distinction of the second set of fast reactor cases (self-sustaining) is the ability to operate at equilibrium with a natural uranium only feed. This is possible because of the large neutron surplus –0.8 to –1.2 in a fast spectrum, avoiding the need for fissile make-up. Furthermore, the build-up of minor actinides is quite small, roughly an order of magnitude less than the thermal reactor cases in Table 2.

To provide an indicator of the impact of different strategies on the void reactivity coefficient, neutron balance factors were also generated using one-group cross-sections corresponding to the coolant-voided condition. All the fast reactor cases exhibit a significant increase in the neutron surplus (-0.1) for the voided condition. However, it is important to note that the D-factor only accounts for the spectral component of the coolant void effect which is known to be largely positive for any fast reactor configuration; the leakage effect, which is not modeled in the D-factor computation is an important negative reactivity feedback effect for fast reactor coolant voiding. Thus, the D-factor changes are only indicative of composition and cross section changes and do not capture all the coolant voiding phenomena.

5. Conclusions

A consistent physics approach has been developed that enables comparison of diverse reactor technologies and specific fuel cycle and TRU management strategies. This method is useful to highlight major features at equilibrium conditions. It is important to note that the accuracy of this methodology is limited by the precision of the one-group cross section data that embodies the basic input. This comparison technique is not a replacement for modern fuel cycle analysis techniques; and best results will be obtained with fuel cycle data generated for the specific reactor and fuel cycle cases being evaluated.

The method can easily be used to intercompare a large number of significant parameters. In this paper, we have compared mass inventories, but the comparison can be easily extended to other fuel cycle parameters, like decay heat, neutron sources, radiotoxicity etc. In fact, the outcome of the generalized Bateman equations can be used directly to evaluate these parameters.

Indications on some relevant reactivity effects can also be obtained, with investigations of neutron balance equation. Moreover, the method offers the possibility to investigate quickly the behaviour of each isotope of interest for the fuel cycle as a function of neutron spectrum and neutron flux level.

References

1) M. Salvatores, “Waste Transmutation Schemes in Different Nuclear Power Deployment Scenarios,” *Proceedings of GLOBAL ’03 Conference on Advanced Nuclear Energy and Fuel Cycle Systems*, New Orleans, LA, Nov. 16-20, 2003.
2) R. N. Hill, T. A. Taiwo, J. A. Stillman, D. J. Graziano, D. R. Bennett, H. Trellue, M. Todosow, W. G. Halsey, and A. Baxter, “Multiple Tier Fuel Cycle Studies for Waste Transmutation,” *Proceedings of 10th International Conference on Nuclear Engineering (ICONE 10)*, Arlington, VA, April 14-18, 2002.
3) M. Salvatores, I. Slessarev, and M. Uematsu, *Nuclear Science and Engineering*, **114**, 1 (1994).
4) G. Youinou, F. Varaine, and A. Vasile, “Plutonium and Americium Multi-recycling in EPR Using Slightly Over-Moderated U-235 Enriched MOX Fuel Assemblies” *Proceedings of GLOBAL ’03 Conference on Advanced Nuclear Energy and Fuel Cycle Systems*, New Orleans, LA, Nov. 16-20, 2003.