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DOSSIER

DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS : RECHERCHES ACTUELLES

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

The physics of transmutation in critical or subcritical reactors

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Abstract The physics of transmutation is presented with application to critical and subcritical reactors. The concept of the 'neutron economy' is used to indicate the most promising approaches. The impact of transmutation on the reduction of the radioactive waste radiotoxicity is underlined in the framework of different strategies for the implementation of transmutation in reactors. The need of experimental validation, in particular in the fields of nuclear data, of the physics of accelerator driven subcritical systems and of fuels dedicated to transmutation, is also mentioned. ***To cite this article: M. Salvatores, C. R. Physique 3 (2002) 999–1012.***

transmutation / neutron economy / nuclear cross sections / sub-critical reactor / radioactive wastes / radiotoxicity

Physique de la transmutation en réacteur critique ou sous-critique

Résumé La physique de la transmutation est présentée avec application aux réacteurs critiques et sous-critiques. Le concept de « économie des neutrons » est utilisé pour dégager les pistes les plus prometteuses. L'impact de la transmutation sur la réduction de la radiotoxicité des déchets radioactifs est indiqué dans le cadre de différentes stratégies d'implémentation de la transmutation en réacteur. La nécessité d'une validation expérimentale, en particulier dans les domaines des données nucléaires, de la physique des réacteurs sous-critiques pilotés par accélérateur et des combustibles dédiés à la transmutation, est aussi évoquée. ***Pour citer cet article : M. Salvatores, C. R. Physique 3 (2002) 999–1012.***

transmutation / économie des neutrons / sections efficaces / réacteur sous-critiques / déchets radioactifs / radiotoxicité

1. Introduction

The present paper gives a summary of the physics principles and methods related to the transmutation of the radioactive nuclear wastes. These principles, when applied to reactor core concepts, define the

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performances of each concept. Some major results of the applied studies are also indicated both for critical or subcritical systems and their associated fuel cycles. Most of the discussions concern minor actinides (and, necessarily, plutonium management). However some indications concerning long-lived fission products are also given.

Finally, needs for experimental validation in the transmutation physics area are discussed briefly.

2. Physics of transmutation

The 'transmutation' concept in a neutron field applies to the physical phenomena that transform a fresh fuel into an irradiated fuel.

The description of such phenomena is obtained by the solution of the set of Bateman equations (see Fig. 1) which allow us to obtain the vector of the nuclei densities $\overline{n}$ at a time $t = t_F$, starting from an initial value $\overline{n}_{t=t_0}$.

Any type of transmutation is function of the neutron cross sections and their spectral dependence. In the transmutation of nuclear wastes, the physics process to be privileged is obviously fission. The competition between the capture and fission processes is then of high relevance.

It is useful to have a close look to the ratios $\alpha = \overline{\sigma}_c/\overline{\sigma}_f$ of the average capture and fission cross section of the different isotopes, see Table 1. This table shows the clear advantage of fast neutron spectra, where α values are the smallest.

For a full understanding of the transmutation potential of different neutron fields a new parameter has been defined [1], the neutron consumption/fission of isotope J, D_J.

The 'neutron consumption/fission' D_J for nucleus J is defined as: "The number of neutrons needed to transform the nucleus and its reaction products into fission products."

To evaluate D_J, a scheme can be set up for the nucleus J and its reaction products (Fig. 2).

From Fig. 2, an algorithm [1] can be obtained:

$$D_J = \sum_{J1_i} P_{J\to J1_i}\left\{R_{J1_i} + \sum_{J2_k} P_{J1_i}\to P_{J2_k} \times \left[R_{J2_k} + \sum_{J3_n} P_{J2_k\to J3_n} \times \cdots\right]\right\},$$

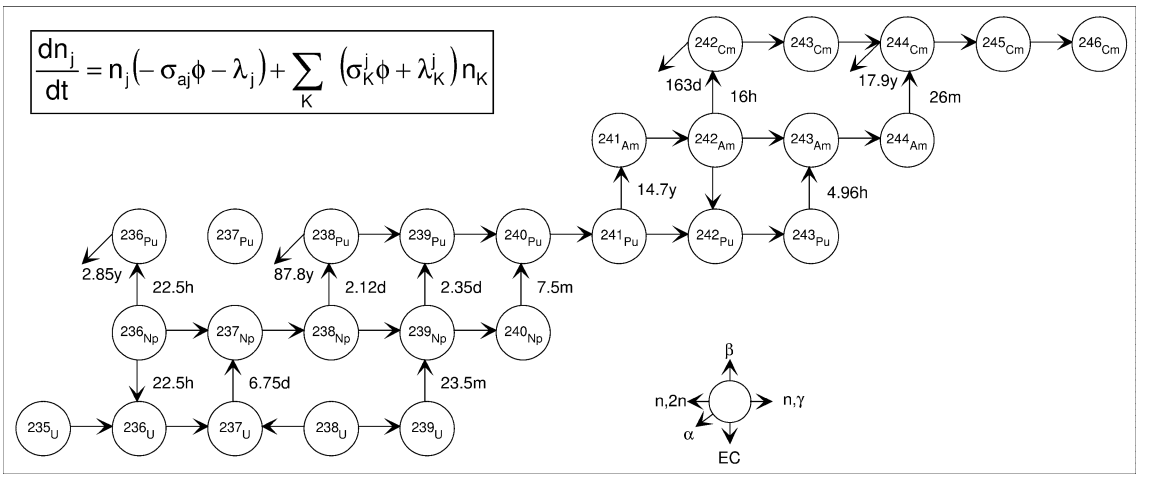


Figure 1. Actinide transmutation chain and nuclei time-evolution equations.

Table 1. Fusion, caption cross sections, and the ratio for different elements. Average cross section (barn): $\overline{\sigma} = \int \sigma(E)\phi(E)\,\mathrm{d}E \,/\, \int \phi(E)\,\mathrm{d}E$

Isotope	PWR spectrum			Fast neutron spectrum		
	σ_f	σ_c	α	σ_f	σ_c	α
Np-237	*0.52*	*33*	**63**	*0.32*	*1.7*	**5.3**
Np-238	134	13.6	0.1	3.6	0.2	0.05
Pu-238	2.4	27.7	12	1.1	0.58	0.53
Pu-239	102	58.7	0.58	1.86	0.56	0.3
Pu-240	0.53	210.2	396.6	0.36	0.57	1.6
Pu-241	102.2	40.9	0.40	2.49	0.47	0.19
Pu-242	0.44	28.8	65.5	0.24	0.44	1.8
Am-241	*1.1*	*110*	**100**	*0.27*	*2.0*	**7.4**
Am-242	159	301	1.9	3.2	0.6	0.19
Am-242m	595	137	0.23	3.3	0.6	0.18
Am-243	*0.44*	*49*	**111**	*0.21*	*1.8*	**8.6**
Cm-242	1.14	4.5	3.9	0.58	1.0	1.7
Cm-243	88	14	0.16	7.2	1.0	0.14
Cm-244	1.0	16	16	0.42	0.6	1.4
Cm-245	116	17	0.15	5.1	0.9	0.18
U-235	38.8	8.7	0.22	1.98	0.57	0.29
U-238	0.103	0.86	8.3	0.04	0.30	7.5

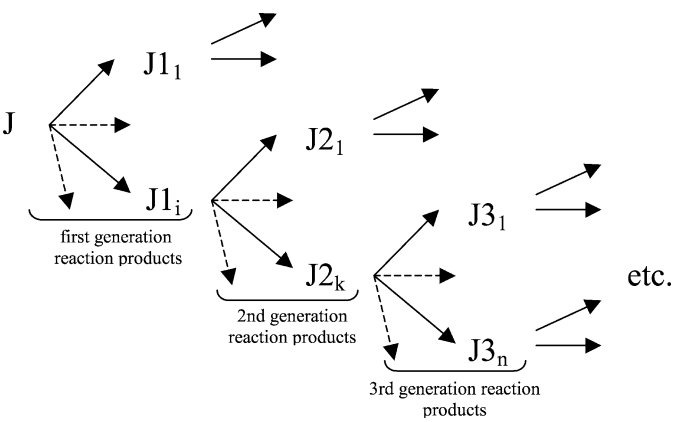


Figure 2. Scheme for evaluating D_J.

where $P_{JN_m \to J(N+1)_s}$ are probabilities (functions of neutron cross sections) to transform JN_m into $J(N+1)_s$ and R_x, neutron loss (or gain) due to the appearance of 'x':

$$R_x = \begin{cases} 1 & \text{for a transmutation by neutron capture,} \\ 0 & \text{for radioactive decay,} \\ 1-\nu & \text{for fission,} \\ -1 & \text{for (n, 2n) reactions,} \\ \text{etc.} & \end{cases}$$

Table 2. Typical values of the neutron consumption per fission (D) for fast and thermal systems. $D \geqslant 0$ implies a source of neutrons is required, whereas $D < 0$ implies excess neutron self-production

Isotope (or fuel type)	Fast spectrum	Standard PWR*
^{238}U	−0.62	0.07
^{238}Pu	−1.36	0.17
^{239}Pu	−1.46	−0.67
^{240}Pu	−0.96	0.44
^{241}Pu	−1.24	−0.56
^{242}Pu	−0.44	1.76
^{237}Np	−0.59	1.12
^{241}Am	−0.62	1.12
^{243}Am	−0.60	0.82
^{244}Cm	−1.39	−0.15
^{245}Cm	−2.51	−1.48
D_{TRU}*	−1.17	−0.05
D_{Pu}*	−1.10	−0.20

* Value for fuel as unloaded from UOX PWR.

Positive D means 'consumption' and negative D means 'production'. Typical values of neutron consumption/fission for fast and thermal neutron systems and for different isotopes (or mixture of isotopes) are given in Table 2.

In Table 2, D_{TRU} is given by:

$$D_{\mathrm{TRU}} = \sum_{\mathrm{J}} \varepsilon_{\mathrm{J}}^{\mathrm{TRU}} D_{\mathrm{J}}^{\mathrm{TRU}}$$

where $\varepsilon_{\mathrm{J}}^{\mathrm{TRU}}$ are the fractions of the different transuranium isotopes present in the irradiated fuel unloaded by a standard PWR and $D_{\mathrm{J}}^{\mathrm{TRU}}$ the corresponding D values – D_{Pu} is given by

$$D_{\mathrm{Pu}} = \sum_{\mathrm{J}} \varepsilon_{\mathrm{J}}^{\mathrm{Pu}} D_{\mathrm{J}}^{\mathrm{Pu}}$$

where $\varepsilon_{\mathrm{J}}^{\mathrm{Pu}}$ and $D_{\mathrm{J}}^{\mathrm{Pu}}$ are the corresponding fractions and D values for the Pu isotopes of the same irradiated fuel.

The new 'D' concept helps to understand if transmutation is feasible in a particular type of reactor.

In fact, each reactor type is characterized by its neutron 'energy spectrum' and by its 'neutron economy balance', that we have defined as follows:

$$G = S_{\mathrm{ext}} - D_{\mathrm{FUEL}} - (L + CM),$$

where G is the neutron surplus (if $G > 0$); S_{ext} is a potential external neutron source (e.g. in a source-driven system), expressed in neutrons/fission; i is a component of the nuclear fuel with ε_i fraction; $D_{\mathrm{FUEL}} = \sum \varepsilon_i D_i$; $L + CM$: neutrons lost (per fission) due to leakage and 'parasitic' captures (structural materials, etc.).

If $G \geqslant 0$, transmutation is feasible in that particular system, and some examples are given in Table 3.

The advantages of fast neutron spectra, already shown by the data of Tables 1 and 2, are evident, if transmutation is foreseen. The hardest spectra are the most suitable, if, as we have indicated, fission is to

Table 3. *G* values for different type of fuels in different types of reactors

Type of fuel	PWR [a]	PWR [b]	'Well' thermalised spectrum[c]	FNR [d]
ϕ (n·cm^{-2}·s^{-1})	10^{14}	10^{15}	10^{15}	10^{15}
Minor actinide fuel	−1.09	−0.80	−0.20	+0.52
Pu + MA fuel	−0.14	+0.08	+0.55	+1.12

[a] Standard flux level in a PWR.
[b] Hypothetical flux level.
[c] Corresponding to a D_2O or graphite-moderated neutron spectrum with a hypothetical high flux level.
[d] Fast neutron reactor: standard flux level.

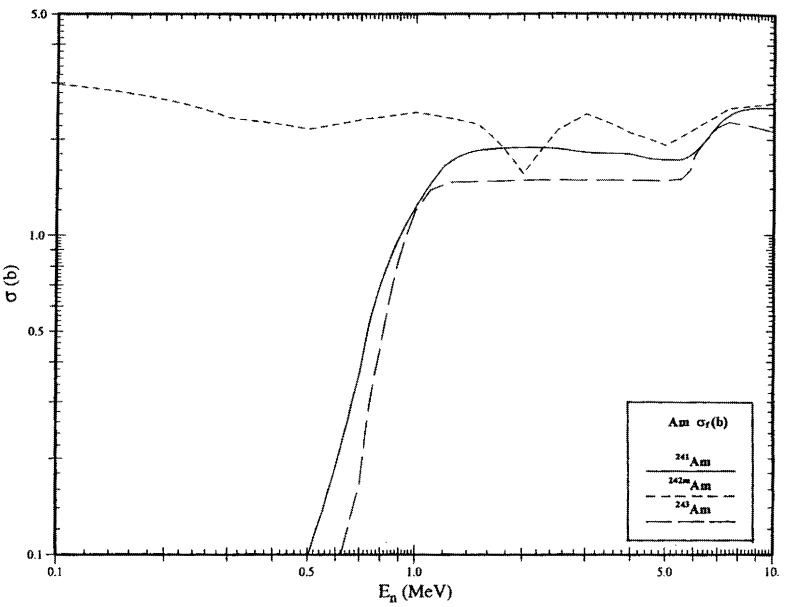


Figure 3. The fission cross sections of americium isotopes in the mega-electron-volt region.

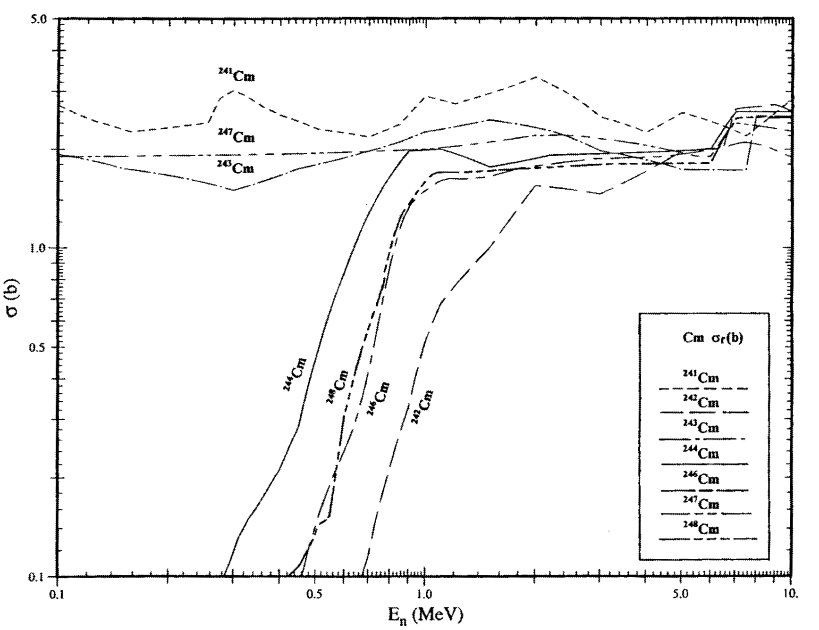


Figure 4. The fission cross sections of curium isotopes in the mega-electron-volt region.

be privileged. Inspection of Figs. 3 and 4 makes this point very clear once more. These figures show the fission cross sections of the Am and Cm isotopes, respectively, most of them being of the threshold type.

3. Application of the generic physics features of transmutation

In general, the physical analysis performed according to the guidelines indicated above shows that:

1. Transmutation rate of fissionable nuclides (MA, Pu, etc.) does not depend either on neutron flux level or on neutron spectrum, but is a direct function of fission rate (or power). This means that if one fixes the power then all transmuter types (fast, thermal, superthermal with or without elevated flux) have the same transmutation rate. This results from the fact that, for ACTINIDES, TRANSMUTATION means FISSION.

   Fast spectrum systems give the best condition for transmutation due to an excellent neutron economy, i.e. in the fast spectrum, actinides can be transmuted in practically any combination. In a thermal neutron spectrum, extra enrichment (e.g. of U-235) is needed, to compensate a worse neutron economy.
2. Long-Lived Fission Products (LLFP)-transmutation demands a significant neutron surplus because LLFP-TRANSMUTATION means CAPTURE and the rate of LLFP-transmutation is proportional to neutron surplus production rate.

A fast spectrum produces many more 'extra' neutrons/fission (which may be available for LLFP-transmutation). It means that, for example, the LLFP-transmutation rate potential of fast spectrum ADS is higher by a factor of 5–10 than the potential of thermal spectrum ADS.

In practice, transmutation in standard critical reactors has been studied using two hypotheses on the form of the fuels which should eventually contain the Minor Actinides (MAs):

(a) the so-called 'homogeneous recycling' of MA, when MA are homogeneously mixed in standard fuel (e.g. PuO_2–UO_2 oxides);
(b) the 'heterogeneous recycling', when MA targets are considered, separated from standard fuel.

Both LWRs and FRs allow the homogeneous recycling of Np and Am. However, some general features have to be mentioned about core performances:

- The reactivity loss over the cycle is reduced (which is a favourable feature), since most of the original MA are transformed in MA with better fission cross sections.
- The temperature coefficients (and boron effectiveness) become worse (from a safety point of view). In fact the replacement of U-238 by MA, gives harder spectra and then, lower Doppler coefficients.
- The coolant void reactivity effects both for LWRs and FRs, become less negative (or more positive). Again, this is an effect related to the harder neutron spectra due to replacement of U-238 by MA.
- For LWRs, there is the need of over-enrichment (which is due to the tight neutron economy), as it was already mentioned.
- Reduction of the effective fraction of delayed neutrons (see Section 4.3).
- Finally, one can say in general that the maximum allowable fraction of MA in the fuel is $\sim 5\%$ of total heavy isotopes for FRs, and 1–2% for LWRs.

For the case of heterogeneous recycling, it has been found that an option is represented by targets irradiated at the periphery of the core, to have a minimum perturbation of power distributions and reactivity coefficients. This option has been particularly investigated in the case of Am transmutation in targets put at the periphery of the core, both of fast and thermal reactors.

4. Homogeneous and heterogeneous recycling, and their consequences on the fuel cycle

4.1. Homogeneous recycling

The IFR concept [2] is still today the most outstanding example of an 'inherently transmuting' concept in the so-called 'homogeneous' recycling mode. The IFR concept can be seen as an energy producing system capable to recycle Pu and minor actinides (MA), to reach equilibrium, both stabilizing the Pu and MA mass

Table 4. Am and Cm homogeneous mode transmutation; reduction of radiotoxicity (with respect to open cycle)

Time after disposal (years)	10^2	10^3	10^4
Open cycle	1	1	1
Pu recycling			
(100% Am and Cm to wastes, 0.1% Pu losses)	3	2.5	4.5
Pu + Am recycling (homogeneous mode)			
Theoretical maximum reduction (Am = 0, 100% Cm to wastes)	20	23	15
Actual recycling (with 1% Am losses)	7	10	6
Pu + Am + Cm recycling (homogeneous mode)			
Theoretical maximum reduction (Am, Cm = 0)	490	400	390
Actual recycling (with 1% Am, Cm losses)	90	100	120
Actual recycling (with 0.1% Am, Cm losses)	270	335	310

flows, and sending to the wastes only a very small fraction of the radiotoxic isotopes. This fraction is of the order of 0.1% or less, according to the announced performances of the pyrochemical process involved, which has of course still to be demonstrated at large scale in the framework of the transmutation application.

The appealing aspects of the IFR concept or other similar concepts in the frame of transmutation are:

– The concept is mainly designed to produce energy, making an optimised use of resources and using a robust reactor and fuel cycle layout. In particular the 'integral' characteristic of the fuel cycle is of particular relevance.
– The fuel cycle does not imply the separation of Pu and MA.
– The concept can accommodate in principle several options in terms of reactor size and fuel, reactor coolant, waste-forms, etc.

In general, the homogeneous recycling has equivalent performances for whatever the type of fuel in the fast reactor. In fact, if the losses at reprocessing are assumed to be of the order of 0.1%, the homogeneous recycling allows to reach a reduction of the potential radiotoxicity with respect to the open cycle scenario of a factor of 200 and more, and this over all the time scale ($10^2 \rightarrow 10^6$ years), ([3] and Table 4). This reduction is such that the radiotoxicity in a deep geological storage becomes comparable to that of the initial uranium ore, after less than a thousand years. However, the consequences on the fuel cycle have to be taken into account, and their impact evaluated. With respect to this last point, it has to be mentioned that recent studies performed at CEA-France, allow to envisage also a multirecycle of both Pu and MA in PWRs. However, even if a specific core assembly design can in principle allow acceptable core performances, the impact on the fuel cycle (e.g. at fuel fabrication) is much stronger than for the homogeneous recycle in a FR, and probably not acceptable.

In Table 5, we give as an example the impact on the fuel fabrication when MA are recycled in a standard fast reactor and in the CORAIL-based PWR concept [4].

The neutron source increase (with respect to a standard MOX) at fuel fabrication is very relevant and probably not tolerable in the case of the CORAIL assembly (loaded with MA). The cause for that increase is the very large Cm and higher mass isotopes (like Bk-249 or Cf-252) production in a thermal spectrum (due to the very high capture cross sections at thermal energies), these isotopes giving a very large production of neutrons via spontaneous fission.

4.2. Heterogeneous recycling

This option has been explored, mainly in Europe and in particular at CEA in France [7] and at JNC in Japan [8], to perform the transmutation of MA in the form of targets to be loaded in critical cores of a 'standard' type. The mode of recycling has been called 'heterogeneous', the potential advantage being to

Table 5. Impact on fuel fabrication of MA recycling

	Reference[a] EPR-MOX 12% Pu	CORAIL Pu recycling only	CORAIL Pu + MA recycling	Standard fast reactor (European Fast Reactor, EFR) Pu + MA recycling
Pu/MA content (%)	12/0	7.8/0	10.3/2.1	20.2/1.2
Activity	1	0.6	1.1	0.1
α-heat[b]	1	0.7	6.7	0.5
β-heat[b]	1	0.6	1.5	0.2
γ-heat[b]	1	0.7	9.1	1.5
Neutron source	1	0.8	370	30

[a] Reference case: MOX fuel with 12% Pu (tot) content as fabricated for an EPR (European Pressurized Reactor, full MOX core loading).

[b] The α-heat is the dominating component of total ($\alpha + \beta + \gamma$) heat.

Table 6. Am and Cm heterogeneous mode transmutation; reduction of radiotoxicity (with respect to open cycle)

Time after disposal (years)	10^2	10^3	10^4
Open cycle	1	1	1
Pu recycling (100% Am and Cm to wastes, 0.1% Pu losses)	3	2.5	4.5
Pu recycling and Am targets irradiation			
Theoretical maximum reduction (Am = 0, 100% Cm to wastes)	20	23	15
With cumulative fission rate = 90%	12	17	10
= 95%	16	20	13
Pu recycling and (Am + Cm) targets irradiation			
Theoretical maximum reduction (Am, Cm = 0)	490	400	390
With cumulative fission rate = 90%	40	45	30
= 95%	72	45	30

concentrate in a specific fuel cycle the handling of a reduced inventory of MA (separated from plutonium). The major obstacles to that approach are:

- the very high irradiation time needed to fission a significant (> 90–95%) amount of MA (which implies very high damage rates);
- the need to separate Am and Cm from Pu and to keep them (Am and Cm) together, in order to reach high values (~ 30) for the radio-toxicity reduction;
- the need to load the MA targets in a very large fraction ($\sim$ 30–50%) of the reactor park, – possibly made of fast reactors, which provide high neutron fluxes, which can be easily tailored in energy to increase fission rates;
- consequences on the power distributions and their evolution with time. In fact, local variations of the power can be significant, creating high gradients which can evolve strongly with time.

In terms of potential radiotoxicity reduction, Table 6 illustrates clearly the first two points and Table 4, allows the comparison with the performance of the homogeneous recycling.

The most relevant point is represented by the fact that, for the heterogeneous recycling, the limiting factor is the fission rate value which can be reached under realistic conditions and, that for the homogeneous recycling the limiting factor is the separation chemistry performance.

Table 7. Influence of Cm for target fabrication

Material in the target	100% Am	90% Am + 10% Cm	80% Am + 20% Cm
Heat	1[a]	×2.3	×3.6
γ dose at 1 m	1	×1.5	×2
Neutronic source	1	×120	×240

[a] Reference value.

Table 8. Delayed neutron fractions β for selected nuclei

Nuclide	β
^{238}U	0.0172
^{237}Np	0.00388
^{238}Pu	0.00137
^{239}Pu	0.00214
^{240}Pu	0.00304
^{241}Pu	0.00535
^{242}Pu	0.00664
^{241}Am	0.00127
^{243}Am	0.00233
^{242}Cm	0.000377

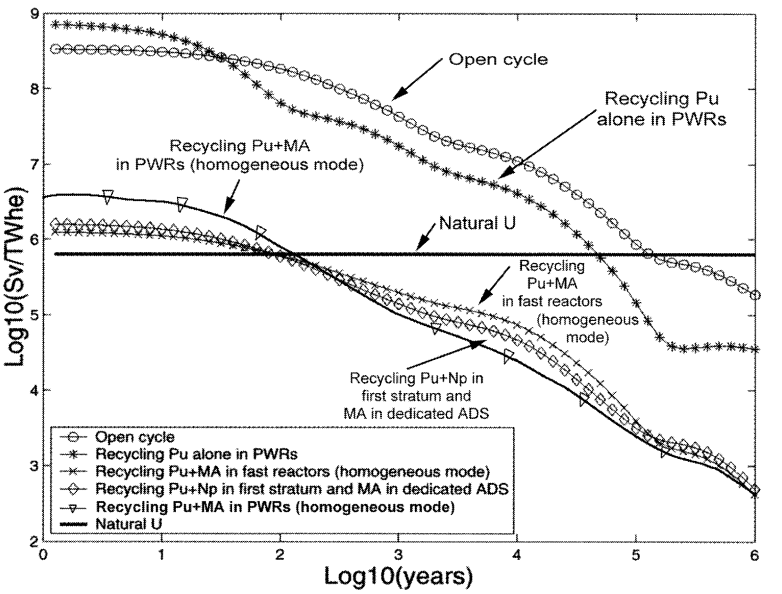


Figure 5. Radiotoxicity evolution with time for different scenarios of partitioning and transmutation.

Finally, the presence of Cm in the targets will have a strong impact on the target fabrication as it is shown in Table 7.

4.3. Dedicated systems

A possible approach to keep the MA fuel cycle and the transmutation technology separated from the electricity production, is the one which calls for the use of 'dedicated' cores, where the fuel is heavily loaded with MA, the rest being, plutonium (the ratio Pu/(Pu+MA) being $\leqslant 0.5$). Work performed at JAERI in Japan [9] and in France [10], has shown that critical 'dedicated' cores can have difficulties, related to the safety parameters degradation. In particular these cores can present a very low delayed neutron fraction ($< 0.2\%\ \Delta k/k$), due to the low delayed neutron fraction of Am, Cm, Np, see Table 8, and a reduced Doppler effect (due to the absence of a fertile like U-238). These characteristics have indirectly helped to promote the Accelerator-Driven Sub-critical Systems (ADS) and the so-called 'double strata' fuel cycle concept [9,10].

In fact, ADS systems offer the feature of the subcriticality ($K_{\text{eff}} \simeq 0.95$–$0.99$), to overcome some of the drawbacks due to low β_{eff} and low Doppler effect. These issues are treated more extensively, e.g., in [11–13].

As for as radiotoxicity, the same reduction is obtained with a reactor park where homogeneous recycling is performed (in fast reactors or in thermal reactors) or with a reactor park of the double strata type, if the same performance of the chemical separations (e.g. recovery factors at all reprocessing steps and

installations, of the order of 99.9% for Pu and $\leqslant$ 99.5% for MA) is assumed. The reduction is such that, at equilibrium, the potential radiotoxicity of the wastes sent to a repository is reduced to the level of the radiotoxicity of the initial uranium ore, after less than a thousand years (see Fig. 5 and [14]).

In the case of the homogeneous recycling, all the reactors can be loaded with MA. In Pu case of the double strata power park, the MA are loaded in a very limited number of dedicated reactors (e.g. corresponding to $\sim$ 5% of the overall power pork energy production, see [10]).

5. The Long-Lived Fission Product (LLFP) transmutation

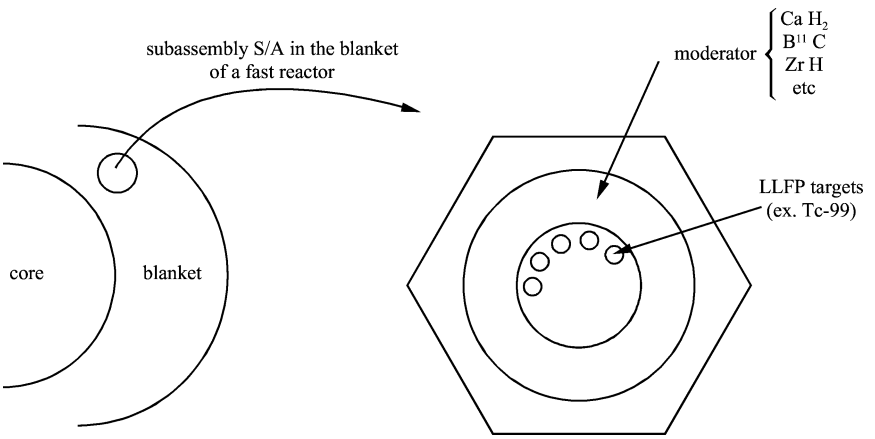
The LLFP transmutation has been associated essentially to a large neutron surplus availability (in units of neutrons/fission). It can easily be calculated the neutron consumption/fission (D parameter), necessary to transmute Tc + I + Cs (elements) or the same D when only the long-lived isotopes (Tc-99, I-129, Cs-135) are supposed to be transmuted (i.e. after isotopic separations) [15]:

$$D_{(\mathrm{Tc+Cs+I})} = 0.15 \text{ n/fission}, \qquad D_{(\mathrm{Tc\text{-}99+Cs\text{-}135+I129})} = 0.08 \text{ n/fission}.$$

This (very large) number of neutrons per fission can be obtained in fast neutron spectra, both in critical or subcritical systems, if a large number of these systems are deployed.

In practice, a possible technique could be to use the high neutron flux ($> 10^{15}$ n/(cm^2·s)) leaking out from a fast reactor core to transmute, e.g., Tc-99 targets, in a moderated (e.g. by CaH_2 or B_4^{11} C) subassembly at the periphery of the core see Figs. 6 and 7. This is a technique with comparable results in terms of transmutation to the 'Adiabatic Resonance Crossing' technique, proposed by Rubbia [16].

However, the need for transmuting LLFP is very questionable both in terms of practical impact on the geological storage (the heat production is essentially related to Sr-90 and Cs-137, which are not candidates for transmutations, as it is shown in [15]) and in terms of feasibility, in particular if isotopic separation is envisaged. In fact the transmutation of Cs-135 seems to be out of question, and the transmutation of I-129 would need the development of an appropriate support matrix for the targets to be irradiated, and no satisfactory proposal has been made up to now.



Transmutation in a 'moderated' S/A:
(1) the neutron surplus available leaking out of the core, is used in the blanket/reflector;
(2) these neutrons are slowed down to the required energy ('spectrum tailoring') using moderator (not-absorbing) materials in the specific S/A;
(3) higher integral values of the reaction rate are expected in the thermalized spectrum in comparison with the standard fast spectrum.

Figure 6. An example: reaction rate of Tc-99.

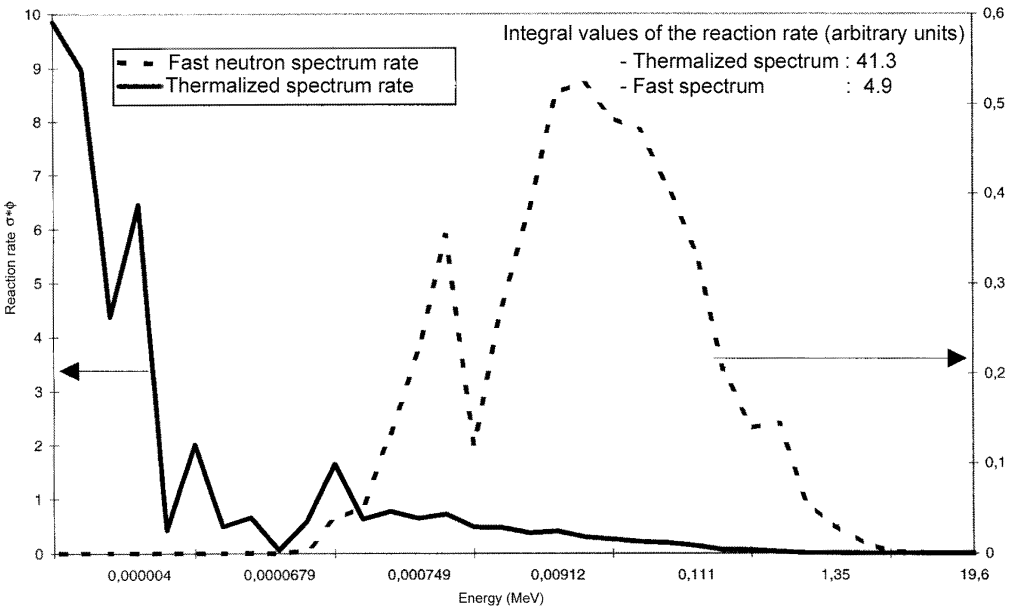


Figure 7. Leakage with Slowing Down (LSD) concept for LLFP transmutation.

6. Experimental validation of transmutation

The major needs for validation of the different transmutation concepts come mainly from the appropriate fuels development [17]. Very few irradiation experiments have been performed up to now, the most extensive being probably the SUPERFACT experiment, performed more than 15 years ago in the PHENIX reactor [18].

As far as physics, there are two areas of validation of major relevance:

– MA neutron cross sections;
– neutronics of the ADS.

In fact, nuclear data uncertainties can play a role in the performance assessment of the different transmutation concepts. In [19], it was shown using perturbation techniques, that even relatively large uncertainties on MA nuclear data do not have a significant impact on a relevant parameter like the potential radiotoxicity source and its evolution in time. On the contrary, similar perturbation techniques applied to dedicated core performances, have shown ([20] and Table 9, taken from that same reference) that the present status of nuclear data is sufficient for preconceptual design studies. More detailed design studies of dedicated cores and of fuel cycles where large amounts of MA would be present, will certainly need much improved nuclear data, in particular in the energy region 100 eV–10 MeV. In this respect, small sample irradiation experiments of single isotopes, are the most meaningful to validate differential data and to reduce uncertainties, as it has been demonstrated by the PROFIL experiments performed in PHENIX [21]. A new series of PROFIL experiments is foreseen [20], with a dedicated irradiation in PHENIX of MA samples.

As far as differential measurements, some improvements of the present situation can be expected if the NTOF facility [23] could be devoted to measurements of capture and fission cross sections of selected isotopes of Am and Cm. The demonstration of the feasibility of such measurements should be a priority for that installation.

Accurate integral measurements of most MA fission rates can be performed in the MASURCA facility in Cadarache, which offers unique features in terms of spectra tailoring and measurement accuracy.

Table 9. An example of the impact of uncertainties for transmutation from [20]. Decomposition of $\Delta K/K$ uncertainty for a dedicated core, using perturbation theory (fuel: (Pu + MA)N particles surrounded by TiN layers; ratio Pu/MA = 0.6). Major contributions to uncertainty (in percentage)

Isotope	Capture[a] (%)	Fission[a] (%)	Total isotope (%)
Pu-238	0.17	0.97	0.99
Am-241	0.99	0.62	1.17
Am-242m	0.01	0.53	0.53
Am-243	0.54	0.26	0.60
Cm-244	0.15	0.46	0.48
Cm-245	0.03	1.14	1.14
Total	1.16	1.78	2.13

[a] Uncertainties on σ vary between 5 and 30% according to the energy range, type of cross section and isotope.

As far as the neutronics of an ADS, a series of experiments has been launched in 1995 to validate the neutronics of an ADS at the MASURCA facility in Cadarache [23]. A wide variety of core configurations, subcriticality levels and external source types are investigated [24].

The next step will to be to validate the core and external source coupling 'at power', in particular to understand and validate the transition between a source dominated to a feed-back-dominated regime in the kinetic behaviour of an ADS [13].

7. Conclusions

New theoretical developments have allowed us to fully understand the physics phenomena related to transmutation, but there are still challenging issues, if a practical implementation of transmutation is envisaged.

Studies based on the neutron economy concept indicate that fast spectrum systems are to be preferred. The use of integral fuel cycles and homogeneous recycling of Pu and MA kept together in the fuel of a relatively standard critical fast reactors, seems to be most promising strategy. The use of ADS can be envisaged if one would rely on the multirecycling of Pu in LWRs and on the management of MA in dedicated systems in a separated stratum of the fuel cycle.

Validation experiments, often of a multidisciplinary nature, will play an essential role, in particular in the ADS field.

The future characterization, fabrication and irradiation of appropriate fuels materials for transmutation is also a most crucial area for research and will help to focus physics programs, in particular nuclear data validation with integral or, in few cases, with new differential measurements.

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Discussion

Question de Y. Le Bars

J'ai été très intéressé par la présentation de politique scientifique pour répondre par la transmutation à la question des déchets, et pour les réacteurs nouveaux. Deux questions :

1. Pierre Toulhoat a montré les radio nucléides qui font la toxicité d'un stockage ; ce ne sont pas toujours ceux pris en compte dans la transmutation. Comment la transmutation peut-elle aider le stockage ?
2. Quel est l'inventaire total résultant de la transmutation ? En particulier la consommation d'énergie pour alimenter les réacteurs hybrides (faisceau d'électrons) n'entraîne-t-elle pas la production de produits de fission ?

Réponse de M. Salvatores

1. P. Toulhoat a présenté le cas d'une évolution « normale » d'un stockage. En cas d'évolution « anormale » (par exemple intrusion), la source potentielle de radiotoxicité est dominée par les actinides. Le recyclage du Pu et la transmutation des actinides « mineurs », peuvent réduire le terme « source » d'un facteur 200–300 sur toute l'échelle des temps, si les procédés de séparation au retraitement permettent d'obtenir des facteurs de décontamination de l'ordre de 99.9%. Cela évidemment n'élimine pas le besoin d'un stockage géologique, mais d'une part assoupli les contraintes et d'autre part peut influencer de façon

positive la perception du stockage de la part du public. Public dont les préoccupations légitimes sont à la base de la loi Bataille.

2. Les réacteurs (critiques ou éventuellement sous-critiques) dédiés à la transmutation, produisent de l'énergie exactement comme les réacteurs standard ! Dans le cas d'un hybride, une petite (environ 10%) fraction de cette énergie est utilisée pour alimenter le faisceau de protons. En ce qui concerne la production de produits de fission, elle est naturellement associée à la fission, donc à la production d'énergie mentionnée précédemment, . . . À parité d'énergie produite, un parc de réacteurs « classiques », ou un parc qui comprend aussi des réacteurs dédiés à la transmutation, produisent essentiellement la même quantité de produits de fission.

Question de A. Birkhoffer

Vous avez mentionné comme point critique le faible coefficient Döppler pour les rapides. En fait on a la vérification expérimentale des coefficients Döppler dans les années 70 aux États-Unis pour du combustible assez enrichi de SNR300.

Réponse de M. Salvatores

Un réacteur avec un combustible à base d'actinides mineurs (Am, Cm, . . .) et Pu, a une faible fraction effective de neutrons retardés (3–4 fois plus faible qu'un réacteur rapide standard) et un coefficient Doppler presque égal à zéro (à cause de l'absence d'U-238). Ces caractéristiques très défavorables pour la sureté et le pilotage du réacteur en mode critique, peuvent devenir moins cruciales, si le même réacteur est opéré en mode sous-critique.

Commentaire de P.-H. Rebut

Je voudrais ajouter que les systèmes de réacteurs sous-critiques où les neutrons manquants sont apportés à l'aide d'une source de spallation et d'un accélérateur (ADS) peuvent être améliorés vis-à-vis des schémas présentés en utilisant deux étages d'amplification. Une équipe russe à Sarov travaille sur ce système en proposant un réacteur fait en deux parties : une première partie est un cœur de neptunium qui voit les neutrons de spallation. Ce cœur a un seuil de fission de 0,8 MeV et est très peu sensible aux neutrons d'énergie inférieure. A l'extérieur de ce cœur existe un modérateur qui abaisse l'énergie des neutrons au-dessous de ce seuil. Au-delà de ce modérateur on a un réacteur sous-critique plus conventionnel. Un tel système permet de découpler partiellement les criticités du cœur et du réacteur environnant et, d'après les calculs, on devrait gagner sur l'amplification global un facteur 10 qui pourrait être utilisé soit pour décroître la puissance de l'accélérateur soit pour l'éloigner des valeurs critiques. Je pense que de tels systèmes devraient être aussi étudiés en France comme solution envisageable pour la seconde moitié du siècle.