Transmutation : Physics and Strategies

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- Much can be said (and has been said) about "criteria".
- However, the simple parameter represented by the activity of the materials stored in a repository (or the "source of potential radiotoxicity") is still widely used. Much debate, not many new suggestions.
- Long-lived fission products : since 1990 (1st OECD Exchange Meeting here in MITO) no much progress towards a clear assessment of the relevance of their associated toxicity.

Transmutation (with neutrons) :

- everything has already been said !

  few clear guidelines :

  - fission should be privileged and neutron economy is essential,
  - the evaluation of the consequences on the physics parameters of the fuel cycle is crucial. Again, it is better to "transmute" by fission than by capture,
  - Np is not a priority : Americium is !
  - Cm : no unique strategy is easily available,
  - when evaluating benefits/drawbacks of transmutation, the role of the strategy chosen for Pu management is dominant.

The Physics of Transmutation

- At equilibrium :

  $\frac{dN_J}{dt} = AN_J + S_J \equiv 0$ (S$_J$: continuous feed of isotope J to the system)

  $\Rightarrow$ equilibrium is readily achieved !

- At each isotope J, one can associate a "neutron consumption/fission"

  D_J ($D_J < 0$ means "production").

  One can define a D_{comb} for a fuel, which is a mixture of isotopes J with fractions ε_J :

$$D_{comb} = \sum_J \varepsilon_J D_J$$

D_J values

ISOTOPE (or fuel type)	Neutron Spectrum and flux level (n/cm^2.s)			
	Fast spectrum	Standard PWR		Thermalized spectrum
Th (with extraction of Pa-233)	-0.39	-0.24	-0.24	-0.27
Th (without extraction of Pa-233)	-0.38	-0.20	1.22	1.14
U-238	-0.62	0.07	0.05	0.01
Pu -238	-1.36	0.17	0.042	-0.13
-239	-1.46	-0.67	-0.79	-1.07
-240	-0.96	0.44	0.085	0.14
-241	-1.24	-0.56	-0.91	-0.86
-242	-0.44	1.76	1.10	1.12
Np -237	-0.59	1.12	0.53	-0.463
Am -241	-0.62	1.12	0.076	-0.54
-243	-0.60	0.82	0.16	0.21
Cm -244	-1.39	-0.15	-0.53	-0.48
-245	-2.51	-1.48	-1.46	-1.37
D_{TRU} (fuel unloaded from a PWR)	-1.17	-0.050	-0.35	-0.54
D_{TRPu} + Np (fuel unloaded from a PWR)	-0.70	1.1	0.3	-0.4
D_{Pu} (fuel unloaded from a PWR)	-1.1	-0.20	-0.40	-0.53

- The system neutron balance can be expressed in terms of a (positive or negative) neutron "surplus" G :

$$G = - D_{comb} - (CM + L)$$

(CM and L are the neutrons/fission lost in parasitic captures or by leakage).

- Since CM + L is generally equal to ≈ 0.3 n/fission (for most systems) one can assess what system can allow "transmutation" (fast reactors or thermal reactors with increased fuel enrichment).

- For a source-driven subcritical system :

$$G = S_{ext} - D_{comb} - (CM + L)$$

In a subcritical system driven by an accelerator which supplies protons of ~ 1.5 GeV to a target producing ~ 40 neutrons/proton (i.e. Max $S_{ext} \simeq 1 \div 1.5$ n/fission),

⇒ The relations among the subcritical K_{eff}, the fraction f of the fission energy <u>used</u> to feed the accelerator, the proton current i and the power P of the subcritical core give for example :

$K_{eff} \simeq 0.95$ $\quad f \simeq 10\ \%$ $\quad P \simeq 300$ MWth $\quad i \simeq 5$ mA
$K_{eff} \simeq 0.9$ $\quad f \simeq 20\ \%$ $\quad P \simeq 1000$ MWth $\quad i \simeq 30$ mA

⇒ The surplus of neutrons has a price (f). However, can help to provide an alternative option to critical fission reactors for transmutation if :

- fuels very exotic (better $K_{eff} << 1$!)
- possible to concentrate all minor actinides in a limited number of dedicated machines (~ $2 \div 5$ % of a total power reactor park).

Transmutation in critical reactors (Homogeneous recycling) :

- Both LWRs and FRs :
  - Reactivity loss over the cycle : reduced
  - Temperature coefficients (and boron effectiveness) : worse
  - Coolant void reactivity effect : less negative (or more positive).
- For LWRs : need of over-enrichment (neutron economy).
  However : Higher moderator-to-fuel ratio is beneficial.
- Maximum allowable MA : 5 % of total HI for FRs - 1 ÷ 2 % for LWRs.

Heterogeneous recycling :

- Targets at the periphery of the core for minimum perturbation to power distributions.

⇒
- Performances (mass inventory)
- Consequences on the physics characteristics of the fuel cycle

Mass inventories after 1 recycle

	FR of EFR type (1500 MWe) Homogeneous recycling (content : 2.5 %)	FR of EFR type Heterogeneous recycling in radial blankets (content : 40 %)	PWR-MOX with $V_m/V_F = 3$ (content : 1 %) BU = 4.7 GWd/t	PWR-UOX Heterogeneous recycling in the core BU = 42 GWd/t
Np				
Consumption (Kg/TWhe)	10	13	11	15
Consumption rate (%)	60	60	45	38
Fission rate (%)	27	24	9	3
Am				
Consumption (Kg/TWhe)	9	14	10	8
Consumption rate (%)	45	60	42	70
Fission rate (%)	18	22	6	13

Consequences on physics parameters of fuel cycle

Type of recycling		HOMOGENEOUS			HETEROGENEOUS
Actinide content		**2.5 % Np**	**2.5 % Am**	**2.5 % Cm**	**40 % Am + MgO**
FABRICATION	Power	× 1	+ 71 %	× 24	× 14
	Activity	× 1	+ 3 %	× 2	× 19
	Gamma source	× 1	× 4	× 12	× 55
	γ-dose at 1 m	× 4	× 76	× 470	× 1680
	Neutron source	× 1	+ 40 %	× 1700	× 7
End of irradiation	Decay heat	+ 2 %	+ 1 %	- 4 %	÷ 3
	Activity	+ 0.3 %	+ 1 %	- 2 %	÷ 5
	Gamma source	- 0.7 %	- 1 %	- 3 %	÷ 3
	γ-dose at 1 m	+ 6 %	+ 2 %	- 1 %	÷ 2
	Neutron source	- 5 %	× 4	× 6	/
End of irradiation + 5 years	Decay heat	+ 80 %	× 3	× 6	× 32
	Activity	+ 5 %	+ 13 %	+ 30 %	× 5
	Gamma source	+ 1 %	+ 5 %	+ 7 %	+ 43 %
	γ-dose at 1 m	- 1 %	- 2 %	- 2 %	÷ 4
	Neutron source	- 6 %	× 4	× 8	/

Am transmutation in a FR

- Heterogeneous mode

- Multirecycling ? Large amounts of Cm are produced

- Cm : what strategy ?

  - Temporary storage → decay to Pu (60 ÷ 100 y !)
  - Recycle as for Am - Higher mass MA production
  - Reduce its production :

→ fission Am to 90 ÷ 95 %
long irradiation in a high flux (thermalised) at the periphery of a FR.
However, high DPA (> 200 DPA NRT ?)
Also : optimisation to avoid too small initial Am loadings

Application to a reactor park

- Whatever the strategy, Pu recycling reduces potential radiotoxicity by factor ~ 2 ÷ 3 (in particular if FRs are used) : this is a first step in the right direction !

- If Am is recycled, in an equilibrium park :

PWR-UOX	PWR-MOX	FR
70 %	10 %	20 %

the potential source of radiotoxicity is reduced by factors f :

At t (years)	⇒	10^2	10^3	10^4	10^5	10^6
f	⇒	71	75	84	140	80
(theoretical f	⇒	510	480	630	700	570)

if Cm is not put to wastes - Losses : Pu 0.1 % Am, Cm, Np 1 %
if Cm is put to wastes, f are reduced :

f	⇒	10	11	7	94	32

Conclusions

- Data and methods for transmutation :
  ⇒ Need of improvements (reduction of uncertainties).
  However, present uncertainties allow reasonable evaluations.

- Physics analysis (at equilibrium) allows to point out major features and to intercompare different systems/strategies.

- Accelerator-driven systems : a few extra neutrons available, but at a cost. Can help if dedicated (i.e. MA-fuelled) reactors are envisaged in a "double-strata" - type of approach.

- Fast reactors can do most of the job. Limited consequences on the fuel cycle. Transmutation in PWRs : less "attractive".

- Major issue : target development for (once-through) Am irradiation.
  ⇒ Role of SUPERPHENIX : irradiations foreseen.