Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles

A Comparative Study

8.   FISSION PRODUCT TRANSMUTATION

8.1 Introduction

Transmutation of actinides and fission products using thermal reactors has been extensively discussed in the "Status and Assessment Report on Actinide and Fission Product Partitioning and Transmutation" [2].

As regards the transmutation of long-lived fission products to shorter lived or stable nuclides, the report concluded that this is theoretically possible in some cases, but higher fuel enrichments are necessary in LWRs. The transmutation of ^{99}Tc and ^{129}I would take a long time, characterised by a half-time by transmutation of about 44-70 years. Intensive development of target and fuel assembly materials would be required, e.g. in the case of iodine, as well as refined isotopic separation in the case of elements such as Se , Zr, Cs and Sn.

This chapter will again touch upon these aspects and will briefly describe new considerations in relation to the possibility of transmuting fission products and the respective role of the ADS.

8.2 Fission product transmutation

The first phase P&T systems report [2] already discussed the possibilities and limits to transmute fission products. In essence, this study concluded that from a view of reducing radiotoxicity, transmuting fission products is of very little interest. The majority of the fission products has decayed after about 250 years, and their contribution to the radiotoxicity of the spent fuel, which was very high during the first 100 years of storage, has become small. However, some fission products are very mobile in certain geological environments and can thus contribute significantly to the radiological effects of disposal in underground repositories. In addition, the treatment of spent fuel results in releases through gaseous and liquid effluents which also contribute to the long-term radiological effects of nuclear power generation. The fission products that deserve most attention in this respect are ^{129}I, ^{135}Cs, ^{79}Se, ^{99}Tc and ^{126}Sn (see Chapter 2, Section 2.2).

Unlike transuranics, fission products in a transmutation process produce no supplementary neutrons but are purely consumers. As was seen previously in Chapter 2, neutron consumption is the most important parameter, if one wants to assess the potential of transmutation in a given nuclear system.

The rate of transmutation of a nuclide J can be characterised by the time T_J^{transm} needed to incinerate half of an initial mass, which is function of the cross-section $\sigma_{n,\gamma}^J$ (barns) and of the neutron flux ϕ (n/cm^{2}s):

$$T_J^{transm} = \frac{\ln 2}{\sigma_{n,\gamma}^J \phi \times 3.15 \times 10^7} \text{ years}$$

Transmutation of the toxic fission products in nuclear reactors and sub-critical systems may be sensible if rates of nuclear interactions with neutrons are much higher than rates of natural decay, which are defined by decay half-life $T_{1/2}$. That is, transmutation in a neutron flux can be reasonable if $T_{1/2} \gg T_J^{transm}$. Table 8.1 gives some properties of long-lived fission products (LLFP) and Table 8.2 compares natural decays and transmutation rates.

Table 8.1. **Physical parameters of the major LLFP**

Isotope	Period (y)	Decay mode	Thermal power (W/Bq)	Dose (ingestion) (Sv/Bq)	Fraction in an irradiated fuel (g/t)[a]
^{14}C	5.7×10^3	β	1.6×10^{-14}	5.7×10^{-10}	1.3×10^{-1}
^{36}Cl	3.0×10^5	β^-, β^+	4.4×10^{-14}	8.2×10^{-10}	1.6×10^0
^{79}Se	6.5×10^4	β	6.5×10^{-15}	2.3×10^{-9}	4.7×10^0
^{90}Sr	2.9×10^1	β	2.8×10^{-14}	3.9×10^{-8}	5.0×10^2
^{90}Y	7.3×10^{-3}	β	1.5×10^{-13}		1.3×10^1
^{93}Zr	1.5×10^6	β	2.6×10^{-15}	4.2×10^{-10}	9.8×10^1
^{99}Tc	2.1×10^5	β	1.4×10^{-14}	3.4×10^{-10}	8.2×10^2
^{107}Pd	6.5×10^6	β	1.4×10^{-15}	3.7×10^{-11}	2×10^2
^{126}Sn	1×10^5	β	4.2×10^{-14}	5.1×10^{-9}	2.0×10^1
^{126}Sb	3.4×10^{-2}	β	5.0×10^{-13}		6.9×10^{-6}
^{129}I	1.6×10^7	β	1.3×10^{-14}	7.4×10^{-8}	1.7×10^2
^{135}Cs	2.3×10^6	β	9×10^{-15}	1.9×10^{-9}	1.3×10^3
^{137}Cs	3.0×10^1	β	3.2×10^{-14}	1.4×10^{-8}	1.1×10^3
^{137m}Ba	4.9×10^{-6}	β	1.1×10^{-13}		1.7×10^{-4}
^{151}Sm	9.0×10^1	β	3.2×10^{-15}	9.1×10^{-11}	1.6×10^1

(a) UOX from a PWR (3.5%, 33 GWd/t).

For the "transmutable" fission products in Table 8.2, the capture cross-sections are high enough for transmutation to be much faster than natural decay. For most of the transmutable fission products, the thermal spectrum is as good as or better than the fast spectrum (exception ^{107}Pd). For three isotopes, ^{90}Sr, ^{137}Cs and ^{151}Sm, natural decay is or as fast as or much faster than transmutation and it is more reasonable to put them into interim storage to decay. As for "questionable" isotopes (^{79}Se, ^{126}Sn and ^{94}Nb), they are rather long-lived, however, owing to small cross-sections, their transmutation will be slow. However, the yield of these isotopes is limited indicating that total toxicity is rather modest.

In Section 2.4.1 of Chapter 2, we introduced the overall neutron excess parameter -D which is defined as the total number of neutrons which have to be spent to incinerate a radionuclide including its daughter-products. Table 8.3 gives values of D for separated isotopes and Table 8.4 for elements.

Table 8.2. **Parameters of LLFP to be eventually transmuted in a fast**
(E_n (neutron energy) = 0.2 MeV) and thermal (E_n = 1 eV) spectra with standard flux levels:
$\Phi = 10^{15}$ (n/cm^2·s) and $\Phi = 10^{14}$ (n/cm^2·s) respectively

Isotopes, J	$\sigma_{n,\gamma}^{J}$ (barn)		$T_{1/2}$ (year)	T^{transm} (year)		Recommendation from neutronic viewpoint
	Fast spectrum	Thermal spectrum		Fast spectrum	Thermal spectrum	
^{79}Se	0.03	0.1	6.5×10^4	7.3×10^2	2.2×10^3	Questionable
^{90}Sr	0.01	0.14	29	2.2×10^3	1.6×10^3	Non-transmutable
^{93}Zr	0.03	0.28	1.5×10^6	730	790	Transmutable
^{94}Nb	0.04	2.2	2.0×10^4	5.5×10^2	1×10^2	Questionable or transmutable
^{99}Tc	0.2	4.3	2.1×10^5	110	51	Transmutable
^{107}Pd	0.5	0.3	6.5×10^6	44	730	Transmutable
^{126}Sn	0.005	0.05	1×10^5	4.4×10^3	4.4×10^3	Questionable
^{129}I	0.14	4.3	1.6×10^7	160	51	Transmutable
^{135}Cs	0.07	1.3	2.3×10^6	310	170	Transmutable
^{137}Cs	0.01	0.02	30	2.2×10^3	1.1×10^4	Non-transmutable
^{151}Sm	0.7	700	89	31	0.3	Non-transmutable or questionable

Table 8.3. **Overall neutron excess parameter (-D) of "transmutable" and "questionable"**
isotopes together with their yields (Y_J) per fission in LWR (UOX) after
5 years of cooling time. Time interval between reprocessing steps: 3 years
(removable nuclides: all fission products except all isotopes of Zr, Tc, Pd, I, Cs, Sn, Nb, Se)

Transmutable and questionable isotopes (J)	-D (neutron/transmutation)	Y_J nuclei/fission in NP
^{93}Zr	-2.01	0.050
^{99}Tc	-1.01	0.055
^{107}Pd	-2.04	0.015
^{129}I	-1.008	0.009
^{135}Cs	-1.002	0.017
^{126}Sn	~-2	0.0012
^{94}Nb	-0.985	6.3×10^{-7}
^{79}Se	~-2	0.0004

Table 8.4. **Overall neutron excess parameter (-D) of "transmutable" nuclides (including all isotopes) together with its yields (Y_J) per fission in LWR (UOX) after 5 years of cooling time. Time interval between fission products reprocessing: 3 years (removable nuclides: all fission products excluding all isotopes of Zr, Tc, Pd, I, Cs)**

Transmutable nuclides, J	-D (neutron/transmutation)	Y_J (nuclei/fission in NP)
all Zr	-2.03	0.26
all Tc	-1.01	0.055
all Pd	-3.22	0.095
all I	-1.01	0.011
all Cs	-0.58	0.13

In fact, it is helpful to use two types of neutron consumption definitions, depending on the choice of units, -D* (neutron/transmutation) and -D (neutron/fission). D can be obtained as the product of D* and of the yield of a nuclide per fission, Y. If a LLFP transmuter is fed constantly with a group of nuclides, then the D value of this group is the sum of Y × D* of the group components.

For example, taking into account LLFP yield, one can calculate the total neutron consumption needed to incinerate all "transmutable" and "questionable" long lived isotopes of fission products:

$$D(^{93}Zr, ^{99}Tc, ^{107}Pd, ^{129}I, ^{135}Cs, ^{126}Sn, ^{94}Nb, ^{79}Se) = \sum_J D_J^* Y_J \cong 0.22 \, (\text{neutron/fission in NP})$$

This value defines the total neutron consumption for incineration of all LLFP presented above, if preliminary isotope separation of fission product in LWR discharge has been realised.

To incinerate all Tc, I, Cs without isotopic separation, one needs about 0.15 (neutron/fission in nuclear park). Isotopic separation of ^{99}Tc, ^{129}I and ^{135}Cs allows to reduce this neutron consumption to 0.08 (neutron/fission in NP) where 0.009 and 0.056 neutrons per fission are needed, respectively, for ^{129}I and ^{99}Tc.

To transmute elements such as Tc, I, Cs in a fast spectrum transmuter one needs to know the neutron surplus G available and to the fraction (f) of these transmuters in a Nuclear Power Park (NPP).

Taking into account a "standard" value of the neutron parasitic capture (CM) and the neutron leakage (L) as CM + L = 0.3 neutron/fission (which is valid for a fast reactor of an intermediate size and traditional composition), one gets for the neutron surplus (for example in a sub-critical system):

$$G = -\sum_J \varepsilon_J \times D_J - (CM + L) + \mu,$$

where ε_J is a fraction of J-nucleus in fuel, μ is a neutron spallation source ($\mu \approx 0.15$ neutron/fission if $k_{eff} = 0.95$).

It is obvious that the neutronic potential of a fuel cycle scheme for the transmutation of fission products depends on the fraction of fission product transmuters in the scheme. Table 8.5 quantifies this potential for the three principal transmutation schemes, assuming that the fission products are transmuted in the TRU or MA burners.

Table 8.5. **Performance of transmutation schemes for fission product transmutation**

	TRU burning in FR (Scheme 3a)	TRU burning in ADS (Scheme 3b)	Double Strata (Scheme 4)
Contribution of TRU or MA burner to total thermal power	0.37	0.222	0.0581
k_{eff}, averaged over reactor cycle		0.946	0.945
Neutron excess (-D from Table 2.1)	1.48	1.29	0.79
Overall neutron balance corrections:			
– for capture and leakage	-0.3	-0.3	-0.3
– for sub-criticality	0	0.15	0.15
Neutrons available for transmutation:			
– per fission in ADS	1.18	1.14	0.64
– per fission in fuel cycle scheme	0.44	0.253	0.037

These results show that the ADS-TRU burning scheme could, in principle, transmute all "transmutable" and "questionable" LLFPs whereas the ADS-MA burning scheme would be able to handle the transmutation of [129]I and, in addition, 50% of the [99]Tc. In contrast, the FR-TRU scheme would be able to transmute all "transmutable" and "questionable" LLFPs.

Finally, the maximum rate of transmutation (RT, measured in number of transmutations per fission) of any isotope in a given transmuter can be easily evaluated if the neutron surplus G available for LLFP transmutation is known:

$$RT_J = \frac{G \ (neutron/fission)}{D_J^* \ (neutron/transmutation)}$$

One can then calculate the maximum rate R_J^{max} of transmutation of a given isotope J per GWth × year of a transmuter:

$$R_J^{max} \ (kg/GWth \times year) \approx 1.6 \times A_J \times RT_J$$

where A_J is the atomic number of isotope J.

For example, D_J^* is equal to 1.01 for ^{99}Tc and, in a fast spectrum sub-critical transmuter based on TRU from LWR-UOX, G = 0.96. Then, $R_J^{max} \simeq 160$ kg/GWth × year if all the neutron surplus is devoted to ^{99}Tc transmutation.

In practice, for the sake of transmuting LLFP, one can imagine coupling the most favourable characteristics (i.e. high flux and high cross-sections) in a neutron field. This has been achieved for example in the so-called LSD (leakage with slowing down) concept, illustrated in Figure 8.1 and demonstrated in the TARC-experiment [94].

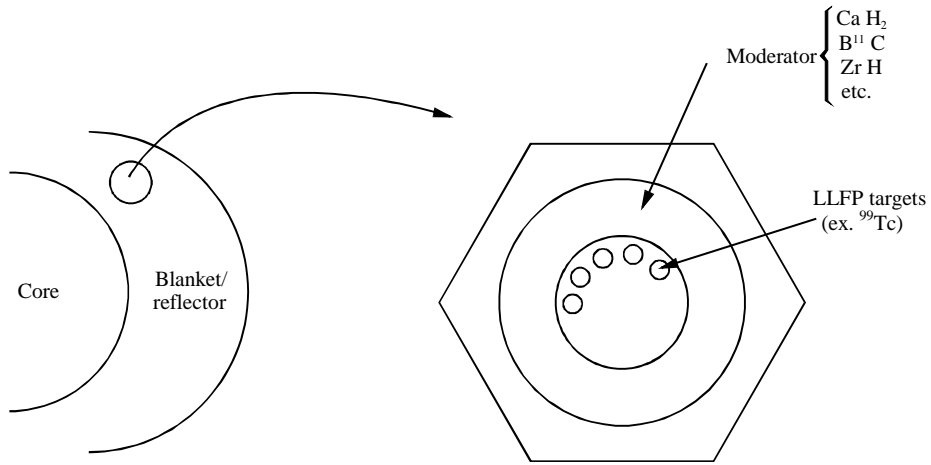Figure 8.1a. **Leakage slowing down concept for LLFP transmutation**

Moderator
{
Ca H$_2$
B^{11} C
Zr H
etc.
}

Core

Blanket/ reflector

LLFP targets
(ex. ^{99}Tc)

Figure 8.1b. **Example of LSD for the reaction rate of ^{99}Tc**

Reaction rate $\sigma*\phi$

Fast neutron spectrum rate
Thermalized spectrum rate

Integral values:
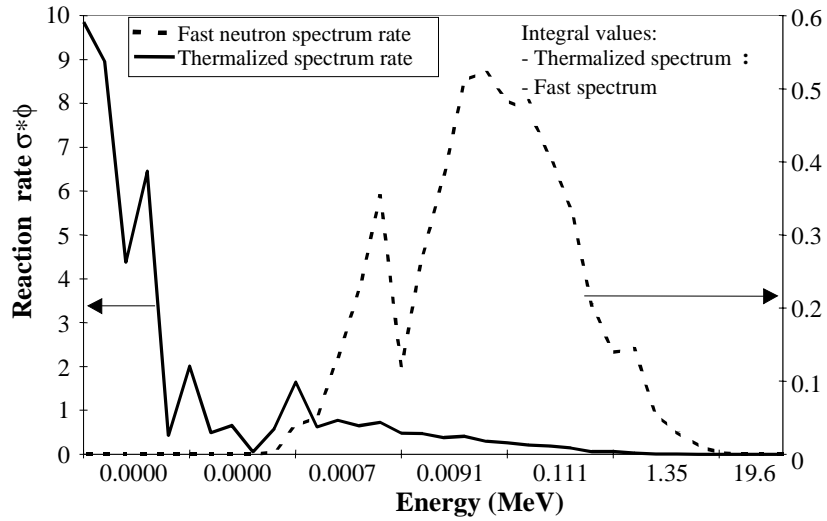- Thermalized spectrum :
- Fast spectrum

Energy (MeV)

Table 8.6 gives some quantitative values for the transmutation of ^{99}Tc and ^{129}I in both a pure fast and moderated spectra.

Table 8.6. **Transmutation of ^{99}Tc or ^{129}I**

	Masses (kg/TWhe)			σ_c	Φ	$T_{1/2}$ (transm.)
	Loaded	Transmuted	%	Barns	10^{15} n/cm^2/s	Years
In fast spectrum						
^{99}Tc	60.5	6.13	10.1	0.32	0.758	91
^{129}I	(26.0)	(3.96)	15.2	0.31	1.21	59
In moderated spectrum.						
1% ^{99}Tc	1.17	1.08	92	11.9	0.493	3.7
5% ^{99}Tc	5.86	3.62	62	5.85	0.372	10
10% ^{99}Tc	11.73	4.88	42	3.46	0.353	18
20% ^{99}Tc	23.46	6.19	26	2.19	0.317	32
To be compared to productions:		≈ 3 kg/TWhe (^{99}Tc)				
		≈ 0.7 kg/TWhe (^{129}I)				

8.3 Conclusions

- Excess neutrons produced by critical and sub-critical burners can, in principle, be utilised to transmute fission products. With the neutron fluxes available in these systems, it is theoretically possible to transmute the long-lived fission products; the transmutation of the more abundant short-lived fission products, however, is impracticable due to insufficient transmutation rates. In practice, the necessity of isotopic separations and difficulties in the preparation of targets present difficult obstacles for the fission product transmutation which currently reduce the number of candidate nuclides to only one or two, i.e. ^{99}Tc, and, possibly, ^{129}I.

- Minimising the fraction of specialised transmuters in the reactor park can result in an insufficient neutronic potential for transmuting the long-lived fission products of the entire park. The present study shows that critical and sub-critical TRU burners perform similarly in this respect. If the transmutation would be limited to ^{99}Tc and ^{129}I, all TRU burning strategies could, in principle, accomplish the task; however, a neutron shortage would not allow these fission products to be completely transmuted in the minor actinide burners of a double strata scheme.