The Abdus Salam
International Centre for Theoretical Physics

United Nations
Educational, Scientific
and Cultural Organization

International Atomic
Energy Agency

**Workshop on**

# ROLE OF PARTITIONING AND TRANSMUTATION IN THE MITIGATION OF THE POTENTIAL ENVIRONMENTAL IMPACTS OF NUCLEAR FUEL CYCLE

**20 - 24 November 2006**

ICTP - Trieste, Italy

**1774/2**

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# Overview of the P&T Strategies

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# Overview of the P&T Strategies

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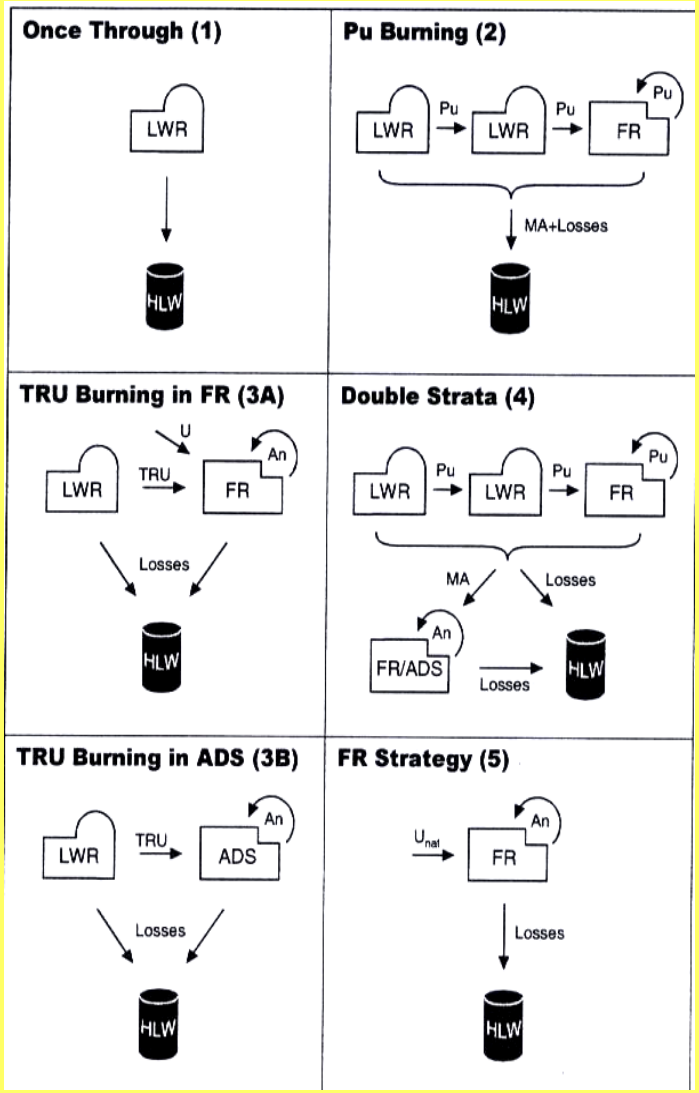
*Originally, only Pu- recovered from spent LWR fuels- was foreseen to initiate the "U-238 - Pu breeding cycle" in FBR.*

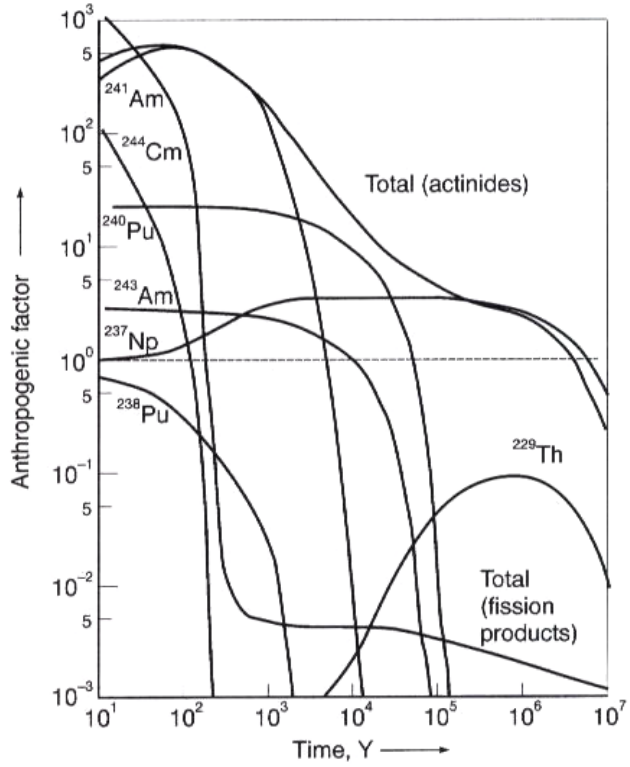
*For future fuel cycle schemes it is likely, that Pu together with the minor actinides (MA) will be partitioned from spent fuel by pyro- or advanced aqueous reprocessing technology and transmuted.*

# Nuclear Fuel Cycle Schemes

- Scheme 1 + 2 are present fuel cycles
- Scheme 3(A + B) recycles all An in FR/ADS
- When FR will have replaced LWR economy (Scheme 5), then all self generated An are recycled
- In between Pu might be burned in LWR and FR, while the An are transmuted in dedicated Burners (Scheme 4)



- *If the radiotoxicity of the needed U-ore is taken as reference, we have to reduce the An by two orders of magnitude*
- *Even if the radiotoxicity of f.p. decreases after 200a below that for U-ore, we have to consider that some f.p. may migrate out of the repository*




Time-dependence of radiotoxicity in a spent LWR fuel 33 GWd/t normalized to the radiotoxicity of the uranium ore (dashed line) mined to produce the fuel

List of long-lived radionuclides considered as candidates for nuclear transmutation

| Nuclide | $T_{1/2}$ [a] |
|---|---|
| $^{14}C$ | $5.7 \cdot 10^3$ |
| $^{36}Cl$ | $3.0 \cdot 10^5$ |
| $^{129}I$ | $1.6 \cdot 10^7$ |
| $^{135}Cs$ | $2.0 \cdot 10^7$ |
| $^{79}Se$ | $6.5 \cdot 10^4$ |
| $^{93}Zr$ | $1.5 \cdot 10^6$ |
| $^{90}Sr$ | $2.9 \cdot 10^1$ |
| $^{121}Sn$ | $5.0 \cdot 10^1$ |
| $^{126}Sn$ | $1.0 \cdot 10^5$ |
| $^{137}Cs$ | $3.0 \cdot 10^1$ |
| $^{99}Tc$ | $2.1 \cdot 10^5$ |

# List of radiotoxic nuclides

| Yearly uptake of elements [g] | nuclide | $T^1/_2$ [a] | ALI [Bq] | Isotopic abundance [%] |
|---|---|---|---|---|
| 1.1 E5 | $^{14}C$ | 5.7 E3 | 1.5 E6 | 1 E-8 |
| 1.9 E3 | $^{36}Cl$ | 3.0 E5 | 1.3 E4 | 5 E-7 |
| 7.3 E-2 | $^{129}I$ | 1.6 E7 | 3.3 E2 | 7 E-2 |
| 3.6 E-3 | $^{135}Cs$ | 20 E6 | 5.1 E4 | 3 E1 |
| | $^{79}Se$ | 6.5 E4 | | |
| | $^{93}Zr$ | 1.5 E6 | | |
| | $^{90}Sr$ | 28.5 | | |
| | $^{121}Sn$ | 50 | | |
| | $^{126}Sn$ | 10.5 | | |
| | $^{137}Cs$ | 30.17 | | |
| | $^{99}Tc$ | 21 E5 | | |
| | $^{237}Np$ | 2.1 E6 | | |
| | $^{238}Pu$ | 87.7 | | |
| | $^{239}Pu$ | 2.4 E4 | | |
| | $^{240}Pu$ | 6.5 E3 | | |
| | $^{241}Pu$ | 14.4 | | |
| | $^{242}Pu$ | 3.7 E5 | | |
| | $^{241}Am$ | 432 | | |
| | $^{242}Am$ | 141 | | |
| | $^{243}Am$ | 7.3 E3 | | |
| | $^{243}Cm$ | 28.5 | | |
| | $^{244}Cm$ | 18.1 | | |
| | $^{245}Cm$ | 8.5 E3 | | |
| | $^{246}Cm$ | 4.7 E3 | | |
| | $^{247}Cm$ | 1.5 E7 | | |
| | $^{248}Cm$ | 3.4 E5 | | |

Nuclides forming monovalent ions with natural isotopic diluent

Nuclides forming no monovalent ions

Nuclides with half-lives below 100a decaying into stable nuclides

Nuclides and parents forming monovalent ions with no natural diluent

$$\frac{dN}{dt} = \sum (f\sigma^{*}\phi + z\lambda^{*})N^{*} - (\sigma\phi + \lambda)N$$

*Shows the formation of N from all parent (radioactive) nuclides N* and its depletion in pile and by decay*

*The in pile depletion of a nuclide can be expressed as half-life and compared with its natural decay*

$\sigma\phi = \lambda \quad (\lambda = \ln 2/T_{1/2})$,

$\sigma$ *is the integral neutron cross section*

*In a FR the transmutation half-lives,* $T_{1/2}$ *of An are between 1 to 4 years, for f.p., however, considerably larger; in case of Tc-99, 10 to 30 years*

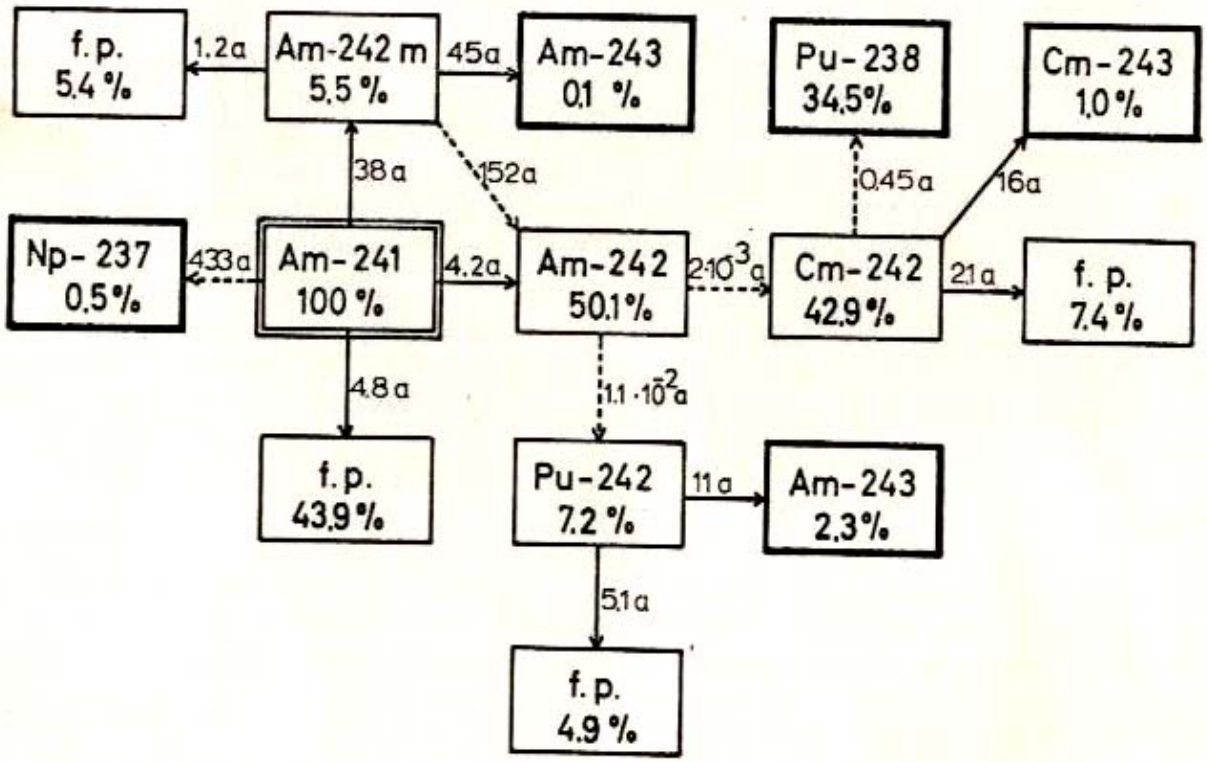



Figure 2 : Transmutation, incineration and decay of Am-241 in a fast reactor neutron flux of $7 \cdot 10^{15}$n $sec^{-1}$ $cm^{-2}$

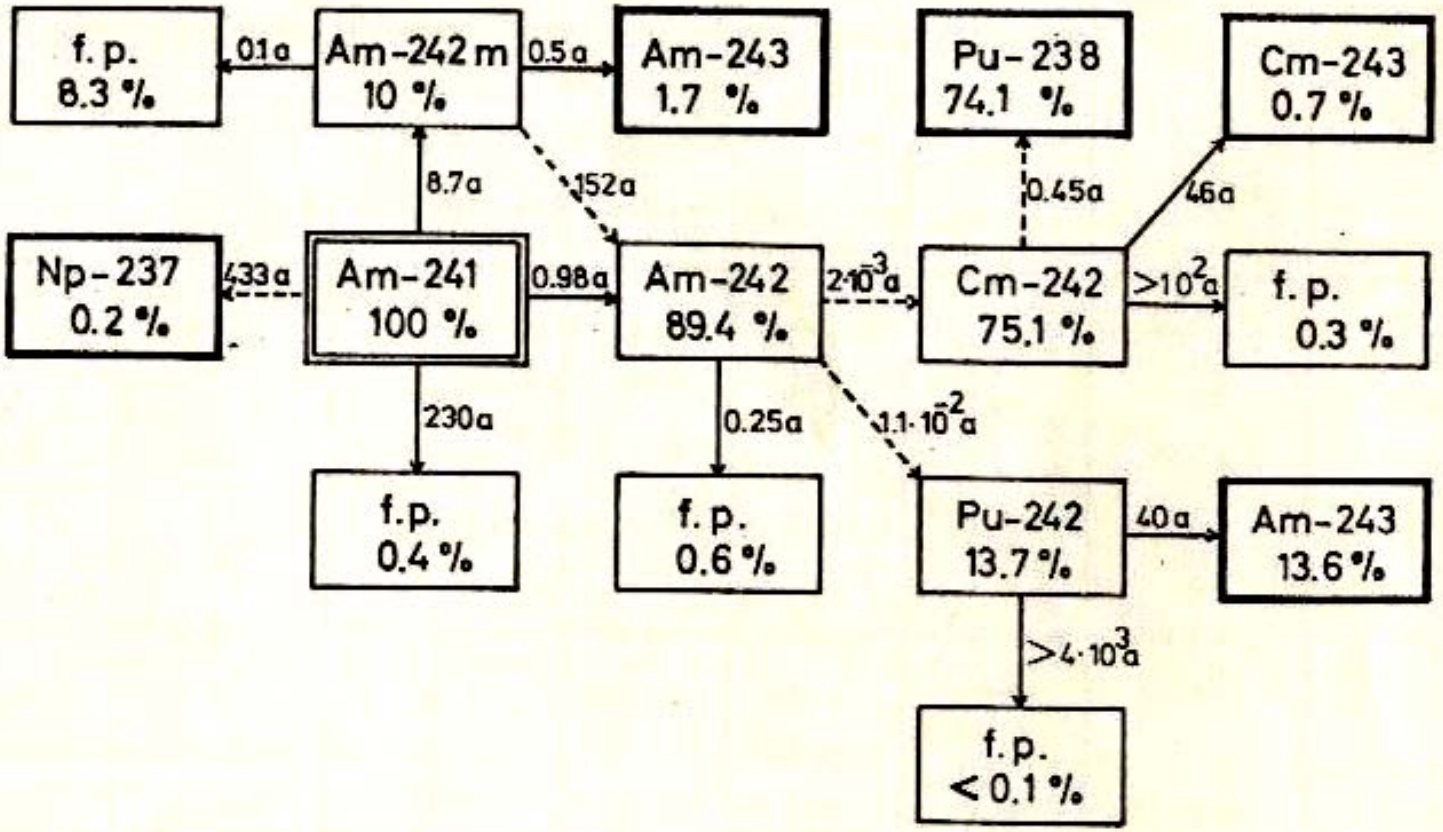



Figure 1 : Transmutation, incineration and decay of Am-241 in a thermal neutron flux of $3 \cdot 10^{13}$ n $sec^{-1}$ $cm^{-2}$

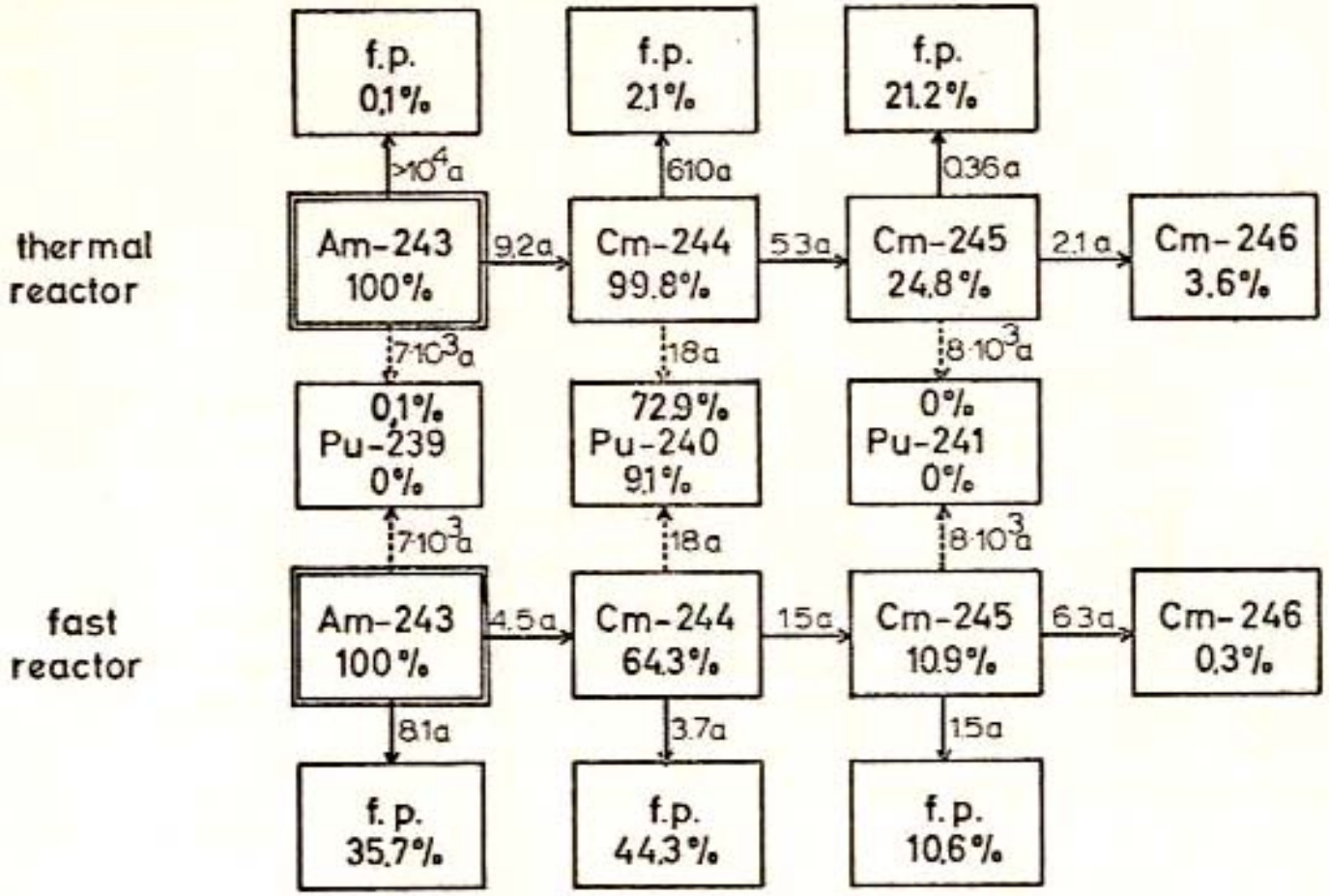
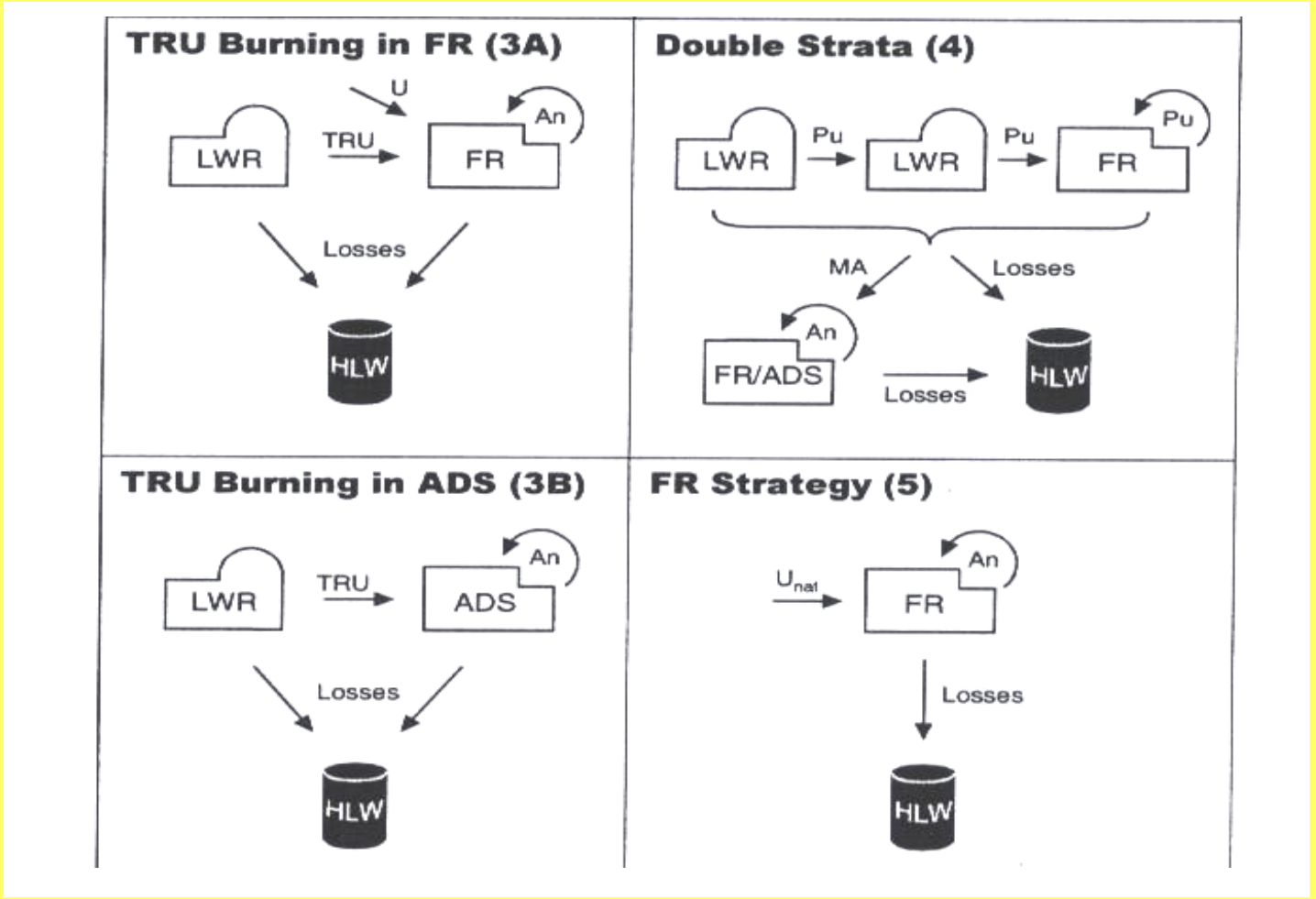


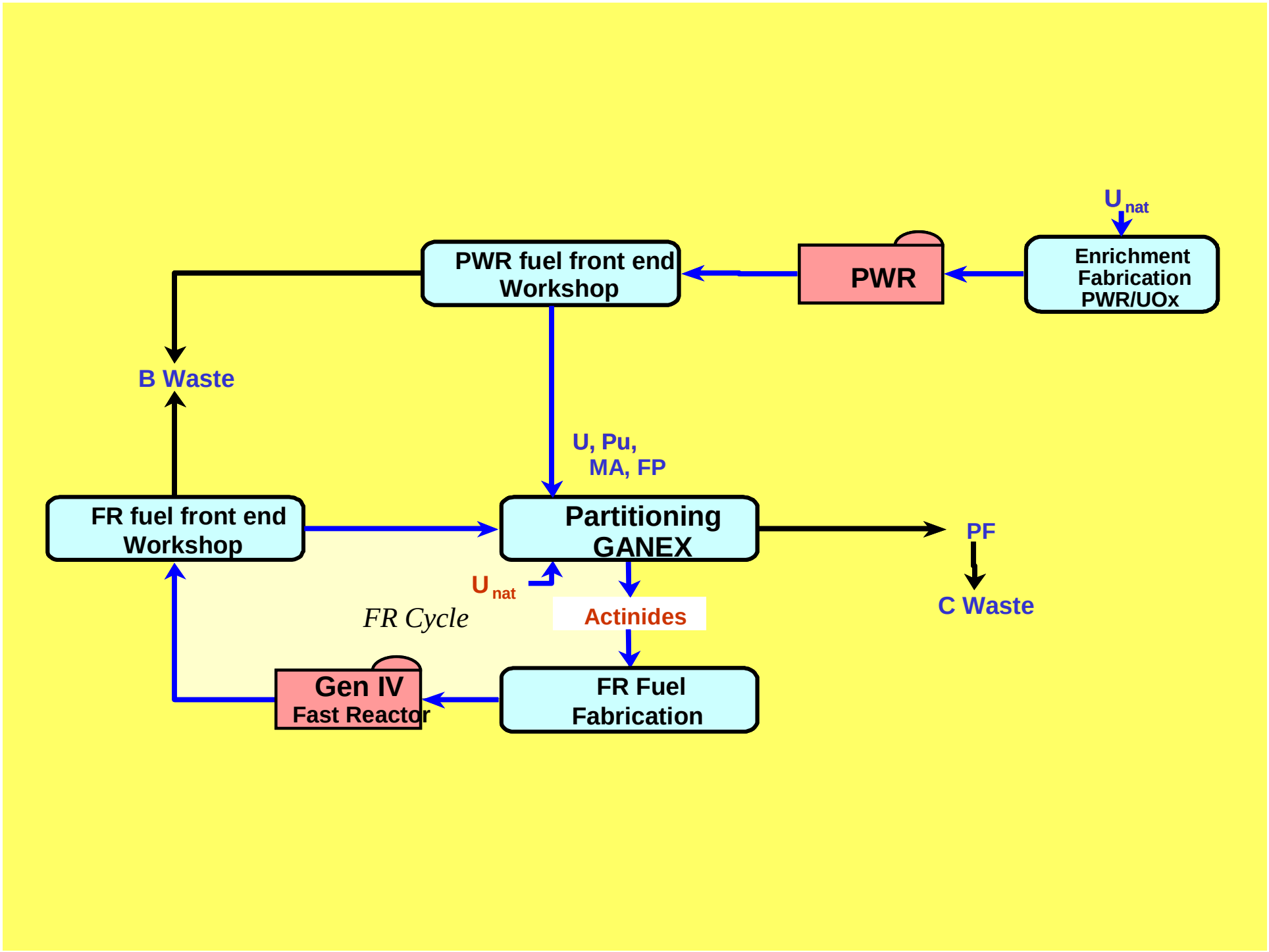

Figure 3 : Decay, transmutation and incineration of Am-243 and Cm-244 in a thermal neutron flux of $3 \cdot 10^{13}$ n $sec^{-1}$ $cm^{-2}$ and a fast reactor neutron flux of $7 \cdot 10^{15}$ n $sec^{-1}$ $cm^{-2}$

# Longlived Radiotoxicity confined in the Nuclear Generating System instead in a Geological Repository

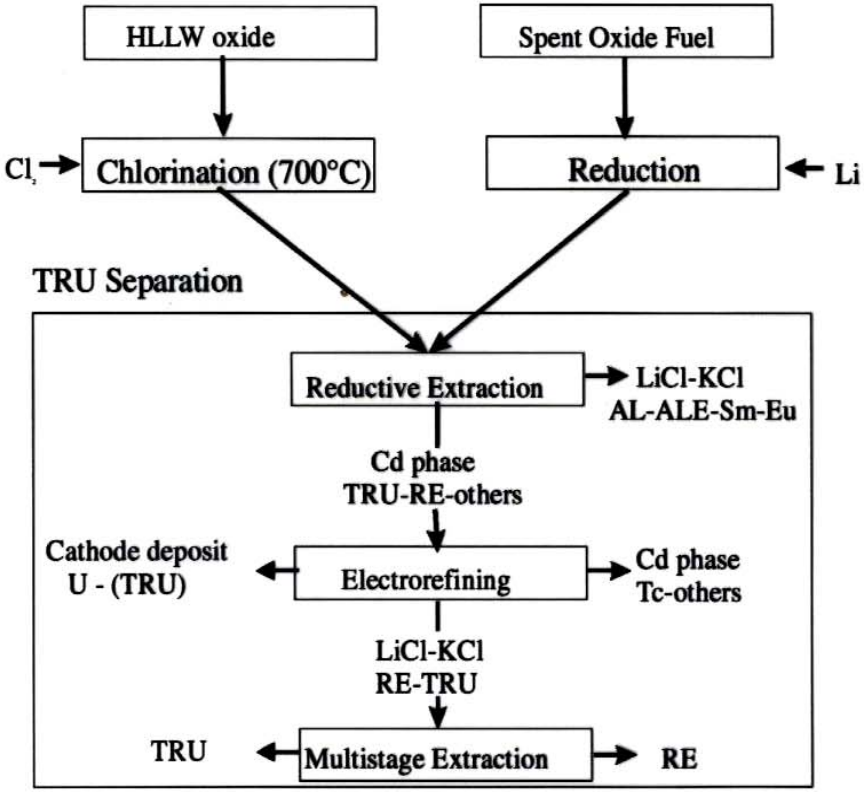
| Actinide | Natural Decay [a] | Decay in FR [a] |
|---|---|---|
| $^{237}Np$ | $2 \cdot 10^6$ | 2.5 |
| $^{239}Pu$ | $2 \cdot 10^4$ | 1.5 |
| $^{240}Pu$ | $6 \cdot 10^3$ | 3.1 |
| $^{242}Pu$ | $4 \cdot 10^5$ | 3.4 |
| $^{243}Am$ | 152 | 1.2 |
| $^{245}Cm$ | $8 \cdot 10^3$ | 1.5 |

*All advanced reactor concepts foresee a separation of long living radiotoxic waste. The group separation of all actinides must be preferred because of proliferation risk (will be explained later)*

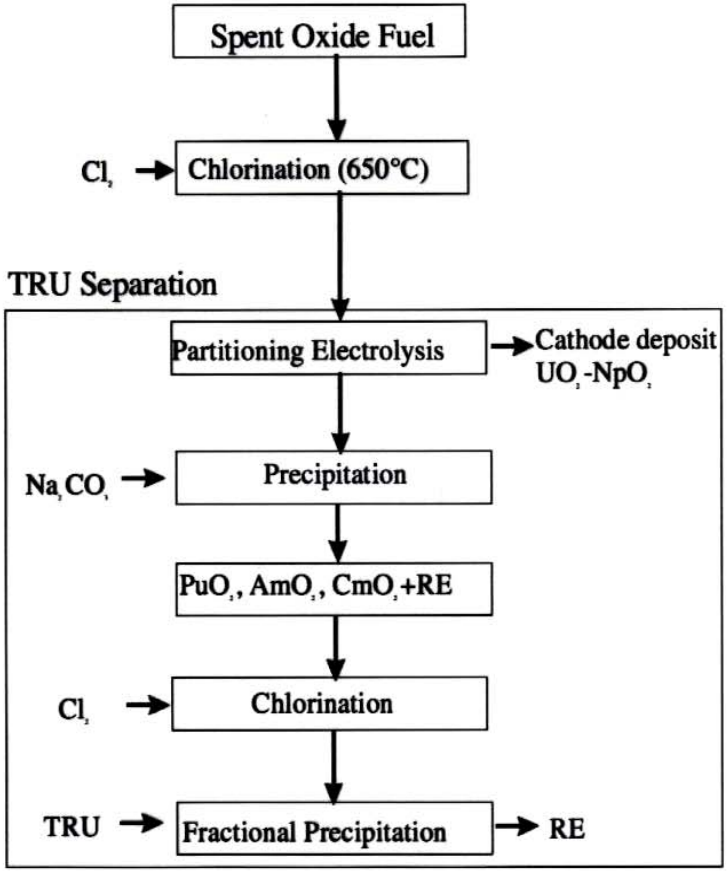


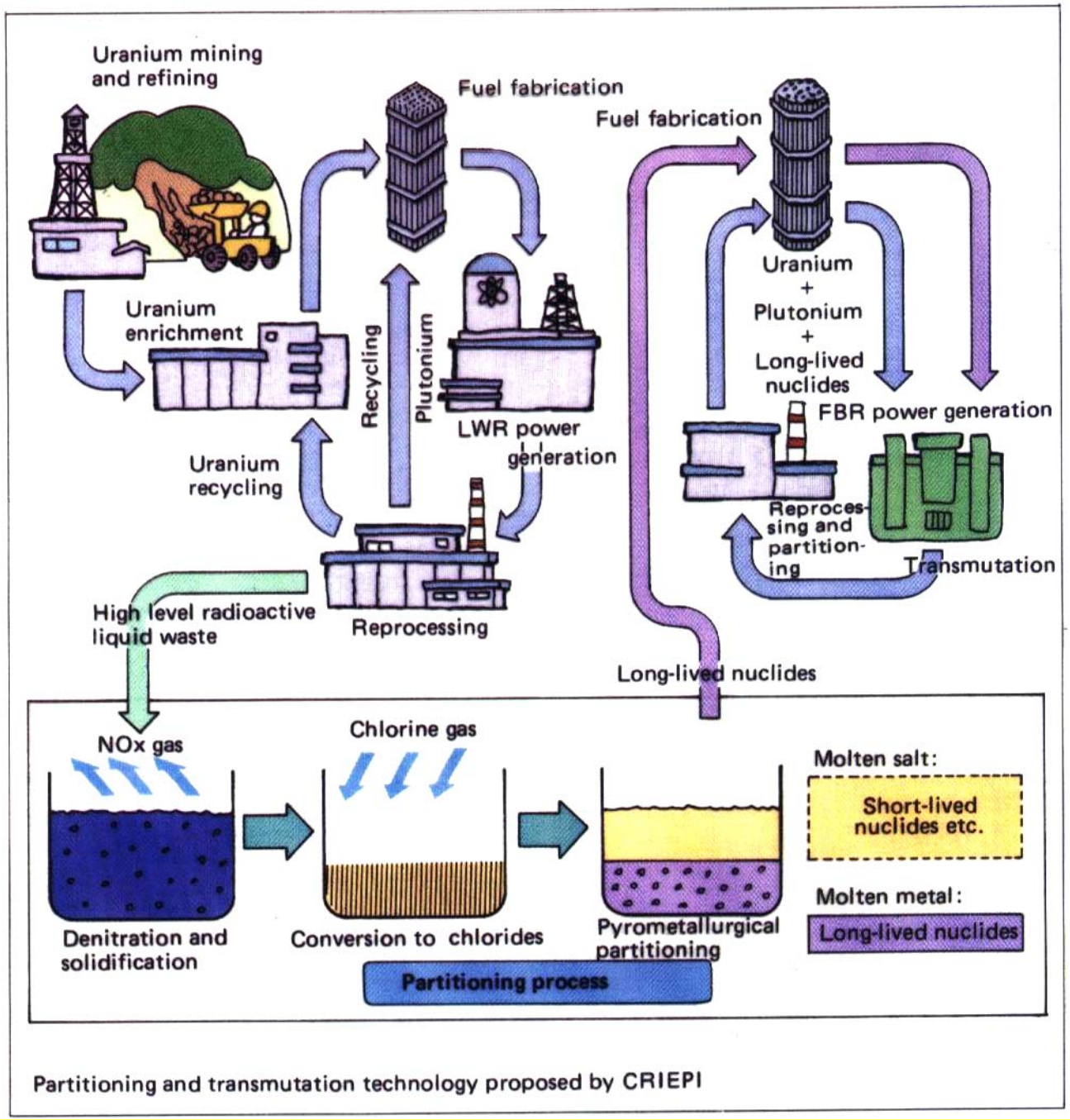

U nat
Enrichment Fabrication PWR/UOx
PWR
PWR fuel front end Workshop
B Waste
U, Pu, MA, FP
FR fuel front end Workshop
Partitioning GANEX
PF
C Waste
U nat
FR Cycle
Actinides
Gen IV Fast Reactor
FR Fuel Fabrication

(a) CRIEPI Process: LiCl-KCl, T=450°C

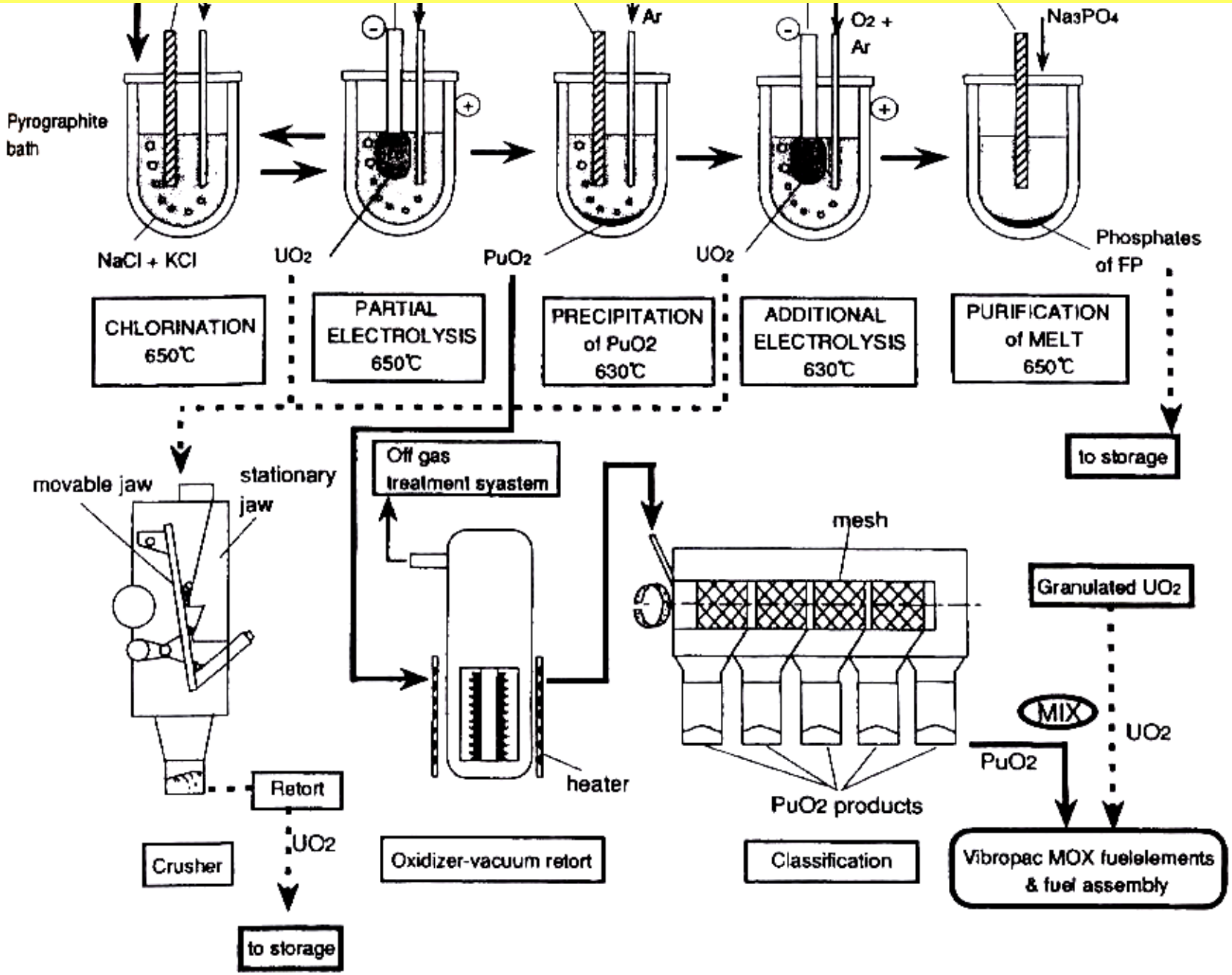



(b) Dovita Process: NaCl-KCl, T=650°C






Partitioning and transmutation technology proposed by CRIEPI


Pyrographite bath
NaCl + KCl
UO2
PuO2
Ar
O2 + Ar
Na3PO4
Phosphates of FP
CHLORINATION 650℃
PARTIAL ELECTROLYSIS 650℃
PRECIPITATION of PuO2 630℃
ADDITIONAL ELECTROLYSIS 630℃
PURIFICATION of MELT 650℃
to storage
movable jaw
stationary jaw
Retort
Crusher
UO2
to storage
Off gas treatment syastem
heater
Oxidizer-vacuum retort
mesh
PuO2 products
Classification
Granulated UO2
MIX
UO2
PuO2
Vibropac MOX fuelelements & fuel assembly

# Fuel Types

- Oxides Pellets (He bonded)

  Vibro-packed

  Coated particles — no reprocessing intended
- Zr or Mo based alloys
- Nitrides Pellets
- Inert Matrices Pellets

  Oxides of Mg, Be, Al

  Spinel type — no reprocessing intended
- Cermets

## Up to 2.5% MA can be loaded to present FR design; even for flattened cores 5% seems the maximum

- TPu decrease delayed neutron fraction
- Due to spectrum hardening void effect increases for MA containing fuels
- If U-238 is substituted by MA, Doppler effect drops

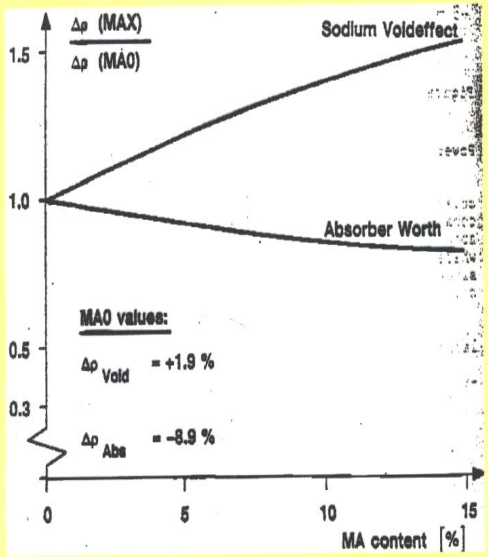
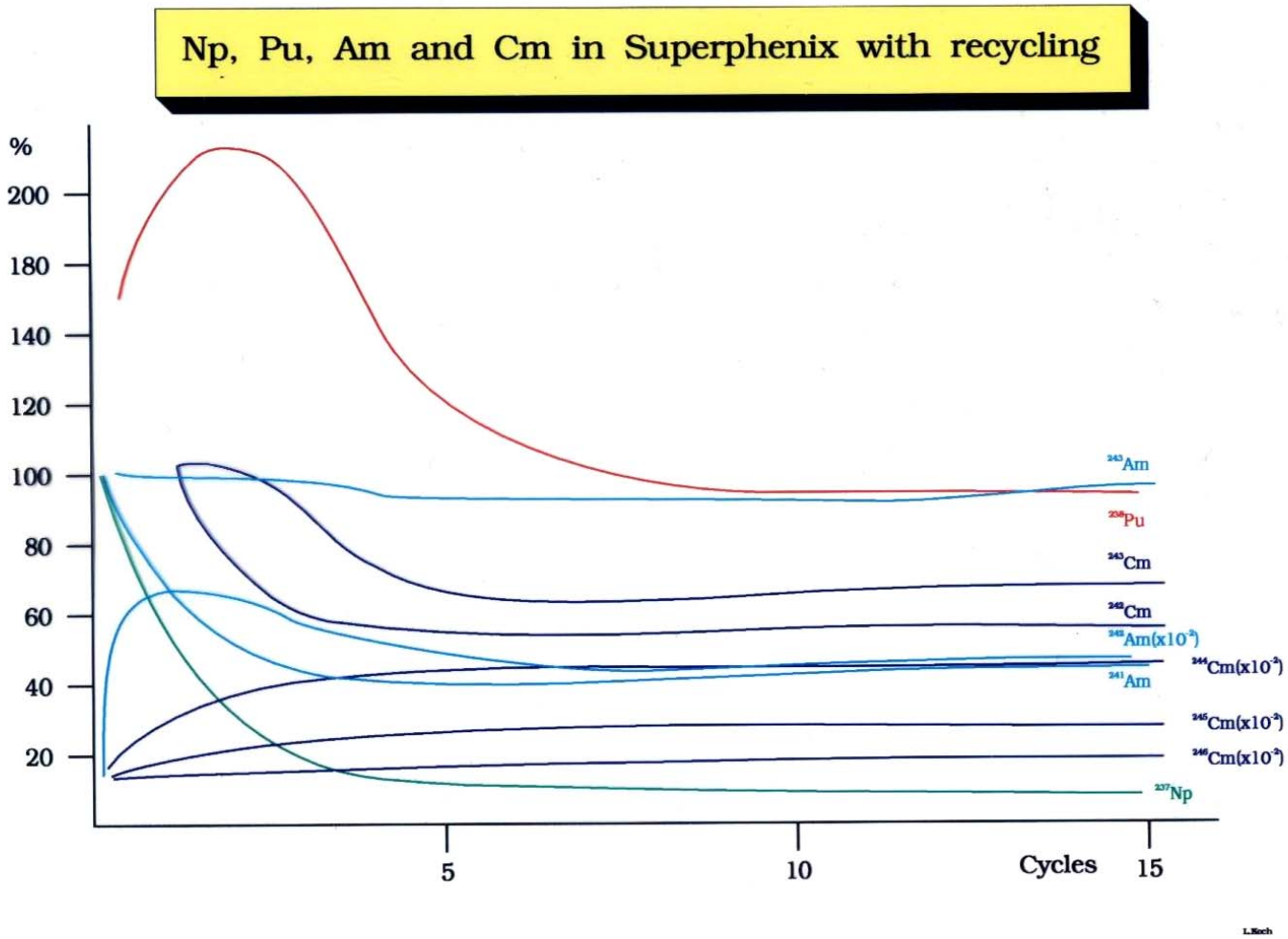
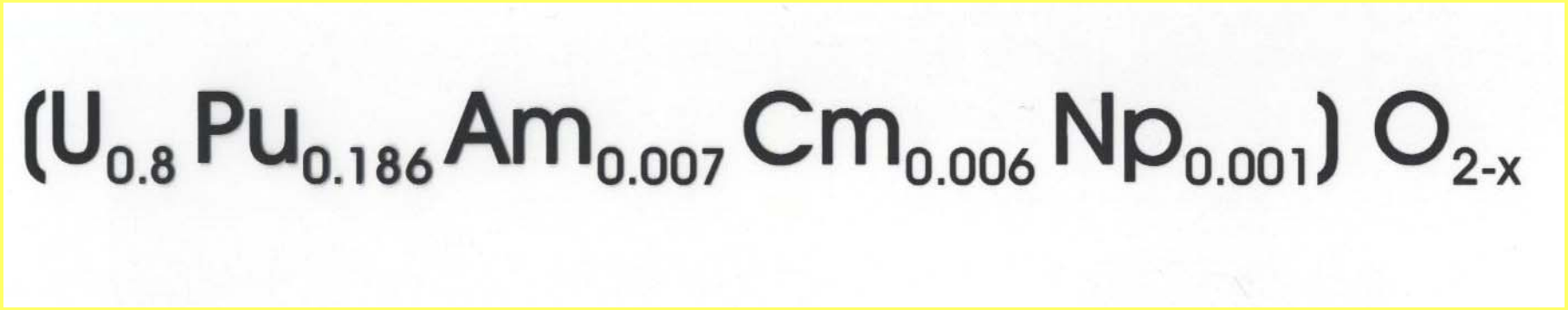



Figure 4: Influence of MA content on sodium void effect, absorber worth and Doppler constant

From: W. Balz, L. Koch, W. Löhr, U.K. Wehmann, Core Design and safety aspects of large LMFBRs with minor actinide recycling, Proceed. Int. Fast Reactor Safety Meeting (1990).

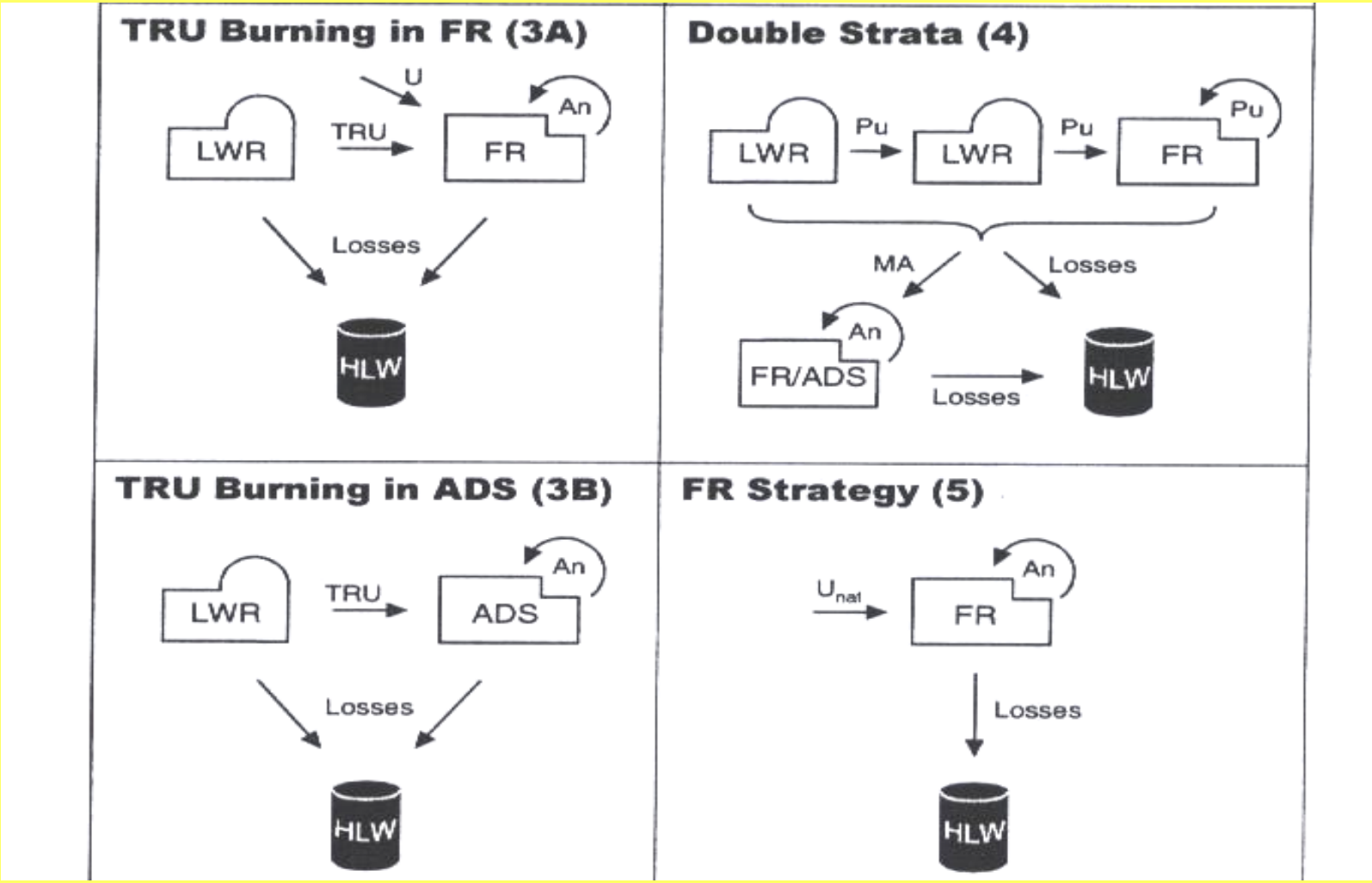

Np, Pu, Am and Cm in Superphenix with recycling
%
200
180
140
120
100
80
60
40
20
5
10
Cycles
15
243Am
238Pu
243Cm
242Cm
243Am(x10-3)
244Cm(x10-3)
241Am
245Cm(x10-3)
246Cm(x10-3)
237Np
L.Koch


(U0.8 Pu0.186 Am0.007 Cm0.006 Np0.001) O2-x

| **Fuel composition** | $(U_{0,74}\ Pu_{0,24}\ Np_{0,02})O_2$ | $(U_{0,74}\ Pu_{0,24}\ Am_{0,02})O_2$ | $(U_{0,55}\ Np_{0,45})O_2$ | $(U_{0,6}\ Np_{0,2}\ Am_{0,2})O_2$ |
|---|---|---|---|---|
| **Percent transmutation** | | | | |
| $^{237}$Np theor./exp. | 24,0/29,0 | | 25,4/27,0 | 27,0/32,0 |
| $^{241}$Am theor./exp. | | 22,6/24,8 | | 25,4/27,0 |
| **burn-up** atom % | 6,8 | 6,8 | 4,8 | 4,3 |
| **Cladding (PIE)** | | | | |
| $\Delta r/r[\%]$ | 0.58 | 0.55 | 0.84 | 0.55 |
| **Fuel (PIE)** | | | | |
| $\Delta r/r[\%]$ | 4.1 | 5.9 | 4.9 | 4.3 |
| **Fuel restructuring (PIE)** | | | | |
| Zone 1: outer ring $[r/r_0]$ | 1-0.66 | 1-0.66 | 1-0.58 | 1-0.67 |
| Zone 2: equiaxial grain growth | 0.66-0.50 | 0.55-0.26 | 0.58-0.22 | 0.67-0.48 |
| Zone 3: columnar or elongated grain growth | 0.50-0.11 | 0.26-0.00 | 0.22-0.00 | 0.48-0.11 |
| Zone 4: central void | 0.11-0.00 | | | 0.11-0.00 |

| **Fuel composition** | **$(U_{0,74}\ Pu_{0,24}\ Np_{0,02})O_2$** | **$(U_{0,74}\ Pu_{0,24}\ Am_{0,02})O_2$** | **$(U_{0,55}\ Np_{0,45})O_2$** | **$(U_{0,6}\ Np_{0,2}\ Am_{0,2})O_2$** |
|---|---|---|---|---|
| **Fabrication** | 400°C/2 h argon | 400°C/2 h argon | 400°C/1 h argon | 400°C/1 h argon |
| drying of kernels | 400°C/2 h air | 400°C/2 h air | 400°C/4 h air | 400°C/4 h air |
| calcination | 850°C/2 h air | 900°C/4 h air | 865°C/3 h air | 900°C/ h air |
| | 850°C/2 h Ar 5% $H_2$ | 1050°C/2 h Ar 5% $H_2$ | 865°C/4 h Ar 5% $H_2$ | 900°C/3 h Ar 5% $H_2$ |
| pellet pressing | 5,1 - 6,4 t/cm$^2$ | 4,4 - 5,6 t/cm$^2$ | 6,1 t/cm$^2$ | 6,1 t/cm$^2$ |
| green density | 58,6 % TD | 58,0 % TD | 55,6 % TD | 56,4 % TD |
| sintering | 1620°C/6h Ar 5% $H_2$ | 1620°C/6h Ar 5% $H_2$ | 1620°C/6h Ar 5% $H_2$ | 1620°C/6h Ar 5% $H_2$ |
| **Fuel** | | | | |
| diameter of pellet | 5,363 ± 0,052 mm | 5,417 ± 0,029 mm | 5,418 ± 0,037 mm | 5,434 ± 0,013 mm |
| height of pellet | 7,119 mm | 7,392 mm | 7,102 mm | 7,372 mm |
| density of pellet | 97,5 ± 0,8 % d$^{th}$ | 96,8 ± 0,7 % d$^{th}$ | 95,1 ± 0,6 % d$^{th}$ | 95,9 ± 10 % d$^{th}$ |
| O/M-ratio | 1,973 | 1,957 | 1,996 | 1,926 |
| crystal parameters | 5,4578 ± 5 Å | 5,4570 ± 5 Å | 5,4545 ± 5 Å | 5,4737 ± 5 Å |
| **Doserate fresh fuel** | | | | |
| γ doserate meas. | $7 \cdot 10^{-3}$ mSv/h | 0,04 mSv/h | < 0,01 mSv/h | 0,26 mSv/h |
| γ doserate pred. | $8 \cdot 10^{-3}$ mSv/h | 0,06 mSv/h | $2 \cdot 10^{-4}$ mSv/h | 0,26 mSv/h |
| n doserate pred. | $2 \cdot 10^{-4}$ mSv/h | $2 \cdot 10^{-4}$ mSv/h | $10^{-7}$ mSv/h | $3 \cdot 10^{-4}$ mSv/h |
| **Doserate spent fuel** | | | | |
| (cooling 5a) | | | | |
| γ doserate meas. | 270 mSv/h | 250 mSv/h | 150 mSv/h | 150 mSv/h |
| γ doserate pred. | 300 mSv/h | 300 mSv/h | 100 mSv/h | 100 mSv/h |
| n doserate meas. | 0.04 mSv/h | 0,04 mSv/h | 0,04 mSv/h | 0,09 mSv/h |
| n doserate pred. | 0.02 mSv/h | 0,02 mSv/h | 0,03 mSv/h | 0,06 mSv/h |
| **Pin:** length, diameter, | 1793 mm | 6.55 mm | | |
| fuel stack | | | 400 mm | |

*In some advanced reactor strategies the Accelerator Driven Systems, ADS are foreseen*



# Scheme of an accelerator driven reactor for transmutation of radiotoxic nuclear waste

- A proton- accelerated up to GeV- produces about 30 neutrons in a Pb-Bi target
- The spallation neutrons start a subcritical reactor
- No limitation on MA loading like in FR
- Some strategies foresee only TPu transmutation
- U free fuel to avoid Pu built-up