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

1774/10

Definition of Environmental Impact and Mathematical Models for Fuel Cycle

&

Modeling for Assessment of Environmental Impact

& Future Direction

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Workshop on
Role of Partitioning and Transmutation in the Mitigation of
The Potential Environmental Impacts of Nuclear Fuel Cycle

(1) 14:00-14:45, Tuesday: Definition of Environmental Impact (EI) and Mathematical Models for Fuel Cycle
(2) 15:30-17:30, Wednesday: Modeling for Assessment of Environmental Impact
(3) 15:45-16:45, Thursday: Future Direction

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The Abdus Salam International Center for Theoretical Physics (ICTP)
Trieste, Italy

Definition of Environmental Impact

“Environment”*

- Natural environment
  - Physical-chemical environment (P-C)

    Soils, geology, topography, surface water, groundwater, water quality, air quality, climatology,…
  - Biological environment

    Flora, fauna, species, diversity, overall ecosystem stability, threatened and endangered species, …
- Man-made environment
  - Cultural environment

    Historical and archeological sites
  - Socioeconomic environment (SE)

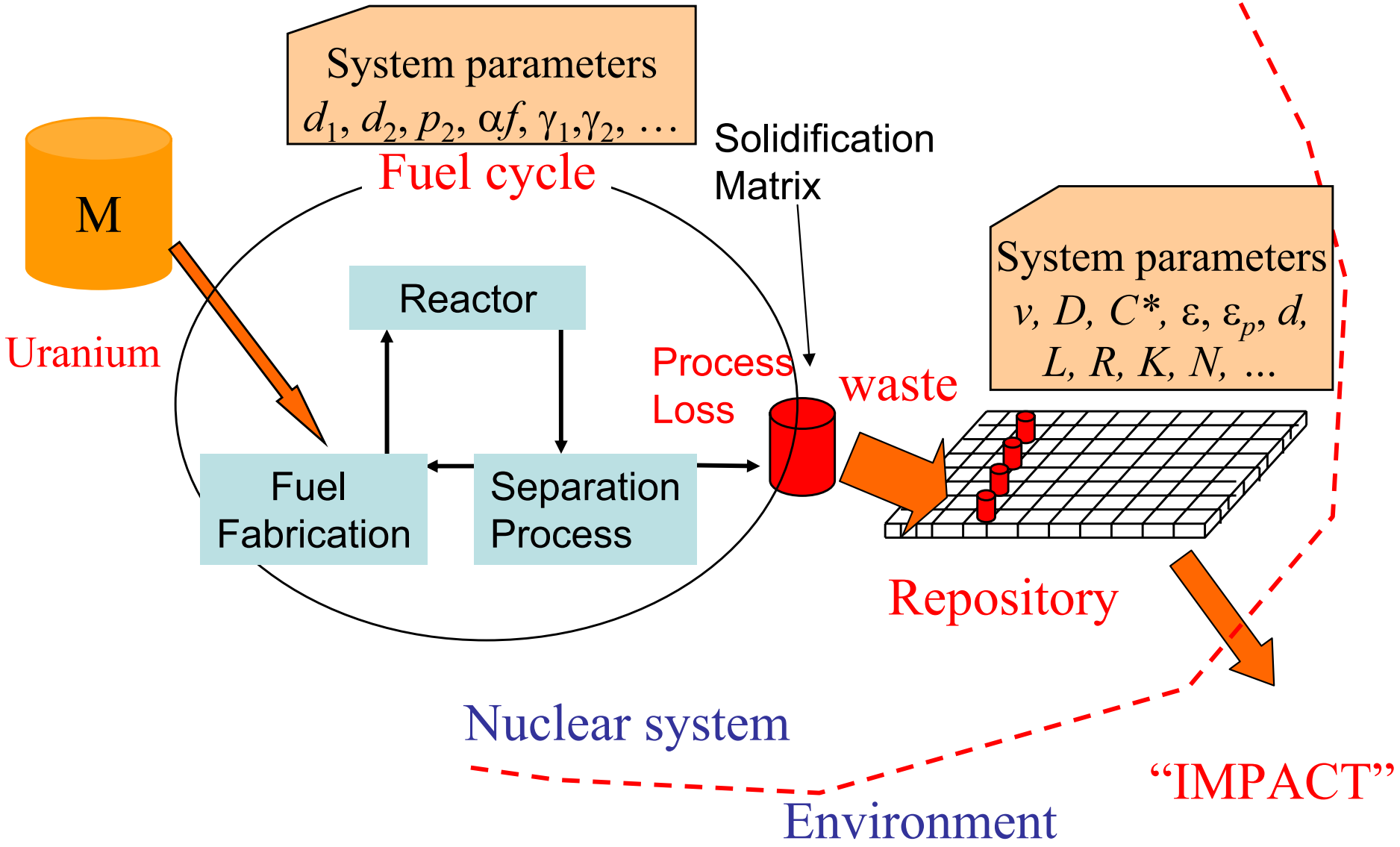
    Human health and welfare, population, economic indicators, educational systems, transportation networks, water supply, wastewater disposal, solid-waste management, public services, safety, …

* L. W. Canter, “Environmental Impact Assessment,” Second Ed., McGraw-Hill, 1996, ISBN 0-07-009767-4

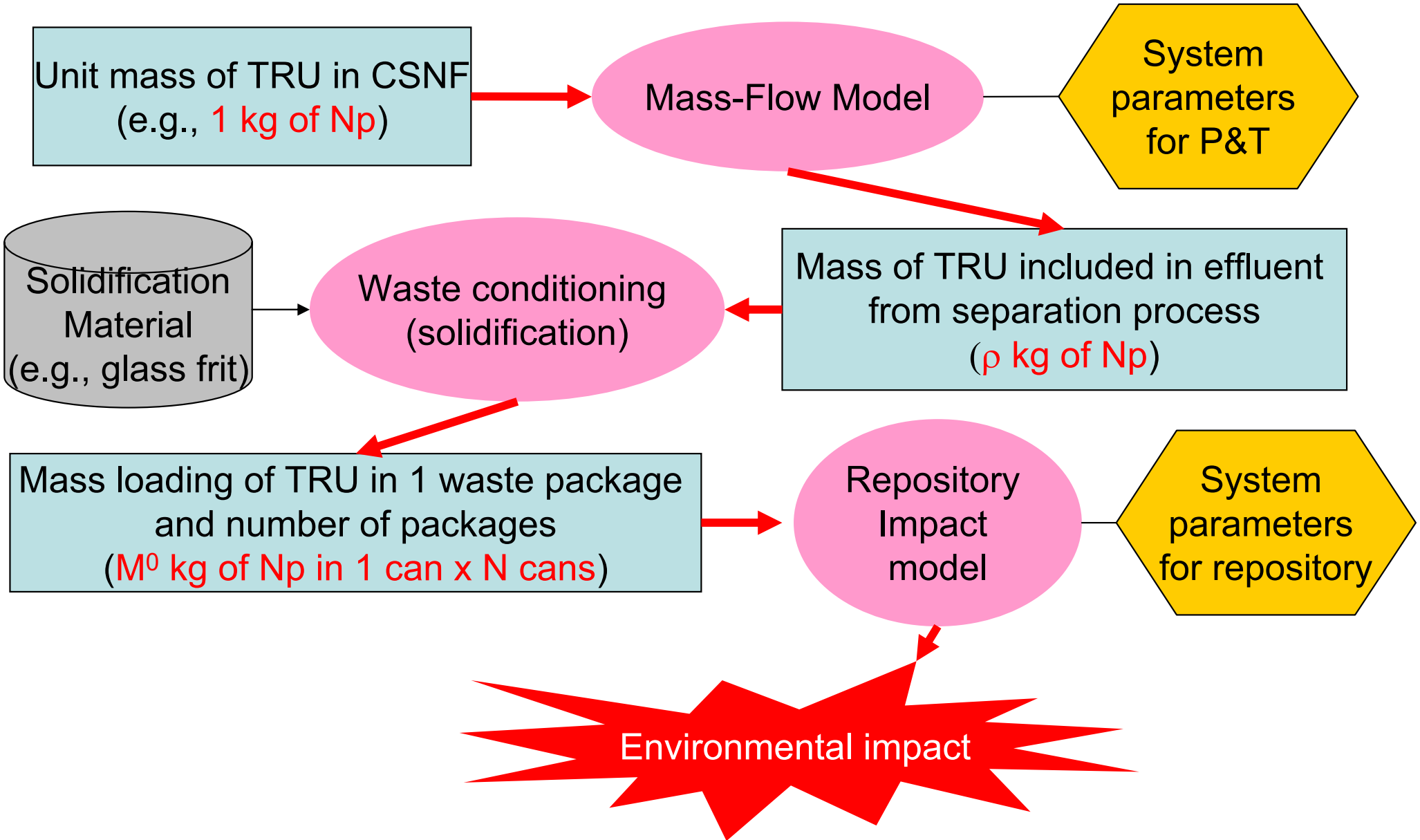
Environmental impact from P&T

- Impacts on human health and quality of life (P-C, SE)
  - Release of radionuclides from HLW generated from Partitioning processes in a geologic repository
  - Reduction of natural uranium requirement, resulting in reduction of mill tailings and depleted uranium
  - Release of radionuclides from P&T facilities operation and transportation
    - Normal operation conditions
    - Accidental conditions (e.g., criticality accident)
- Impacts on cost and institutional systems (SE)
  - Licensing for P&T facilities in addition to existing fuel cycle facilities
  - Cost for operation of P&T facilities
  - Repository capacity expansion due to reduction of waste volume and mass
- Impacts on international politics (SE)
  - Proliferation of nuclear weapons materials
  - Reduction of fissile materials in HLW

"Environmental Impact" and Separation Process Loss



Model Structure



Conventional impact measures

- Radiotoxicity of spent fuel/HLW
  - Is often used to compare risk associated with waste generation from various fuel cycles
  - Potential hazard of waste itself
  - Does not include effects of repository confinement
- Exposure dose rate to the public
  - Is used for regulations for nuclear facilities, including geologic repositories (40CFR197, 10CFR63)
  - Needs to know pathways of radionuclides to a representative group of people
  - Becomes greatly uncertain due to heterogeneity of geosphere

Toxicity Index

$$\text{Toxicity index} = \sum_i \frac{\lambda_i N_i}{C_{i,k}} \quad [\text{m}^3]$$

where

$C_{i,k}$: radioactivity concentration limit for nuclide *i* in medium *k* (*k* = water or air) [Bq/m^3],

Ni: the number of atoms of nuclide *I*

If more than one radionuclide is involved, a summation is performed over all the isotopes present in the mixture.

Toxicity index is the volume of air or water with which the mixture of radionuclides must be diluted so that breathing the air or drinking the water will result in accumulation of radiation dose at a rate no greater than 0.5 rem/year.

Maximum Permissible Concentration and Exposure Dose of Radionuclides

- MPC (Ci/m^3): the radioactivity concentration limit of a given radionuclide in air or in water an individual who obtains his or her total intake of air or water from this source will receive a radiation dose from this radionuclide at the rate of 0.5 rem/yr.
  - NRC 10CFR20 lists MPCs for all radionuclides.
- Dose (rem) = Absorbed dose (rad) • Quality factor • Distribution factor

  [rem] = defined in conjunction with rad

  [Sv] <= [Gy], 1 rem = 0.01 Sv

  Absorbed dose = energy abosrbed by the material

  [rad] = 0.01 J / kg-material

  [Gy] = 1 J/kg, 1 rad= 0.01 Gy

  Quality factor = determined based on LET [eV/m]

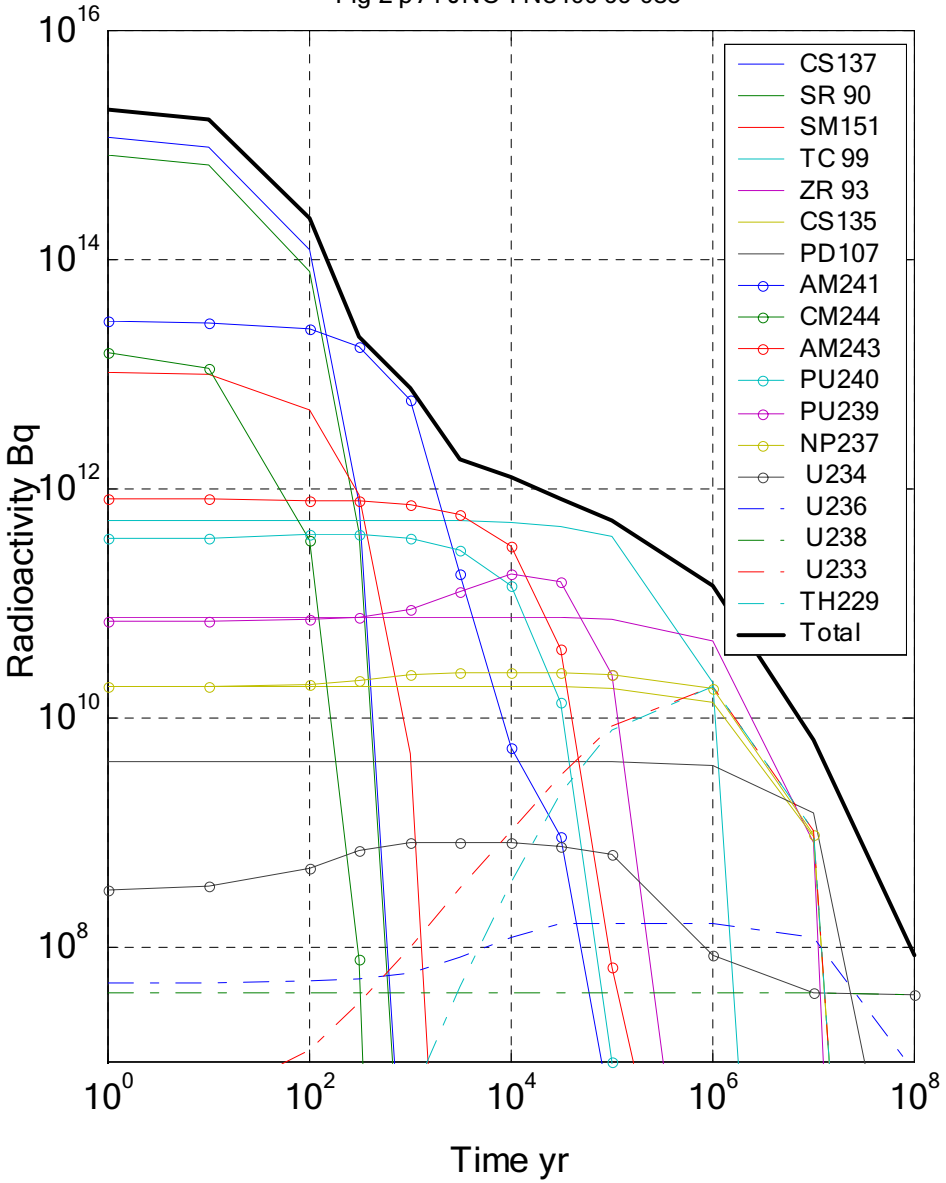
  1 MeV neutron : 10, recoil nuclei: 20

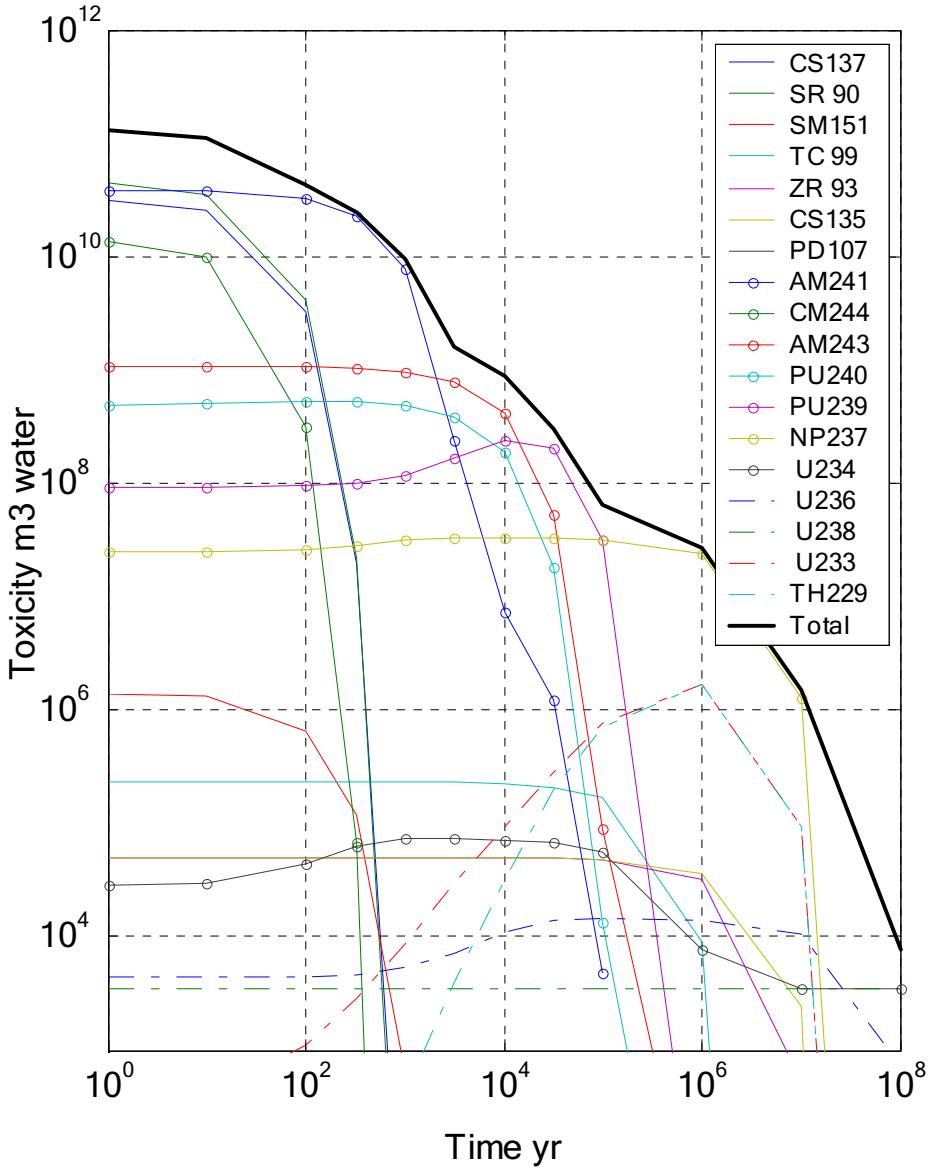
MPCs for U-238, U-235 chains

Nuclide	Half-life	MPC for ingestion (μCi/ml)	Nuclide	Half-life	MPC for ingestion (μCi/ml)
U-238	4.51E9 y	3E-7	U-235	7.1E8 y	3E-7
Th-234	24.1 days	5E-6	Th-231	25.5 h	5E-5
Pa-234	1.17 min	3E-5	Pa-231	3.25E4 y	6E-9
U-234	2.47E5 y	3E-7	Ac-227	21.6 y	5E-9
Th-230	8.0E4 y	1E-7	Th-227	18.2 days	2E-6
Ra-226	1602 y	6E-8	Fr-223	22 min	---
Rn-222	3.821 days	---	Ra-223	11.43 days	1E-7
Po-218	3.05 min	---	Rn-219	4.0 sec	---
Pb-214	26.8 min	1E-4	Po-215	1.78 ms	---
Bi-214	19.7 min	3E-4	Pb-211	36.1 min	2E-4
Po-214	164 micro sec	---	Bi-211	2.15 min	---
Pb-210	21 y	1E-8	Tl-207	4.79 min	---
Bi-210	5.01 days	1E-5	Pb-207	stable	
Po-210	138.4 days	4E-8			
Pb-206	stable				

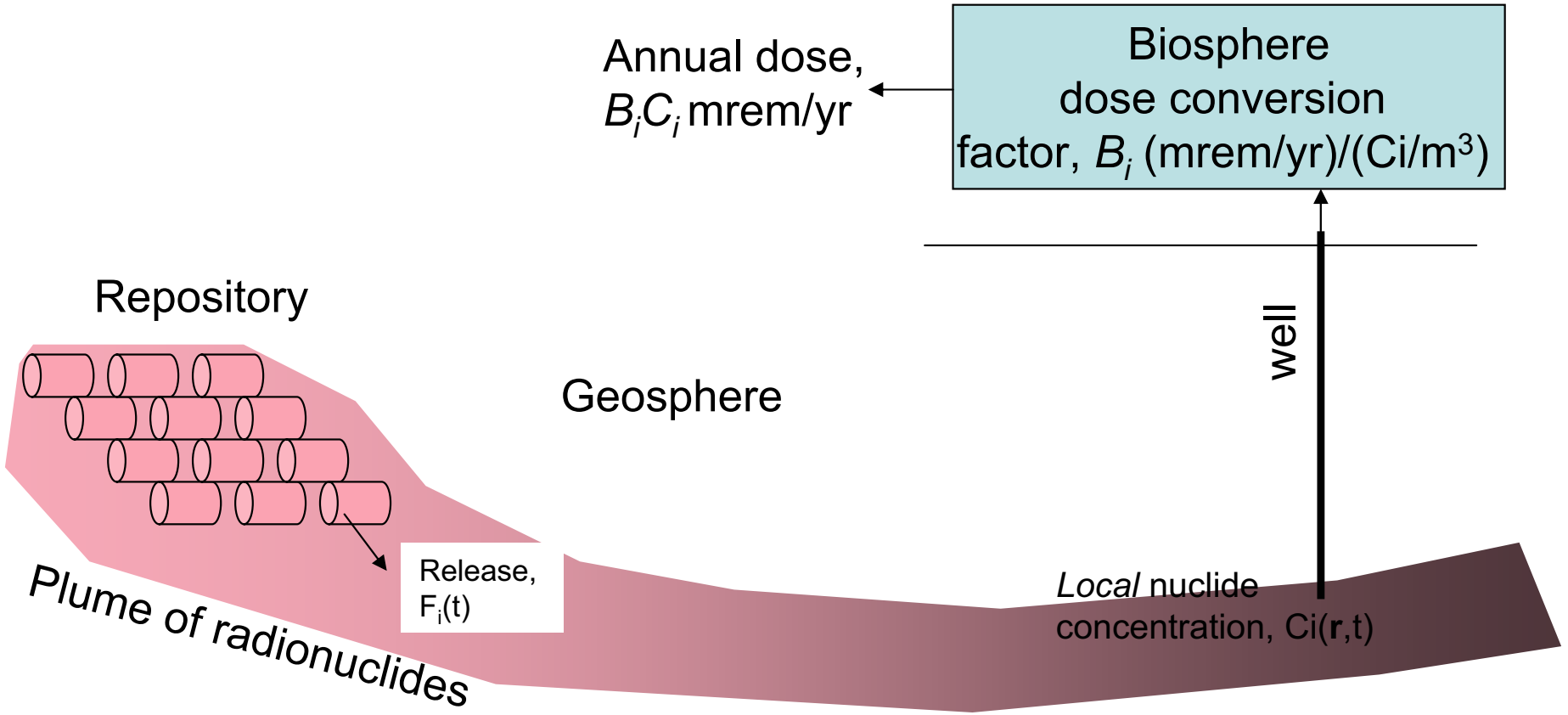
Radioactivity and Radiotoxicity of vitrified HLW from reprocessing of 1 ton of CSNF

Fig 2 p74 JNC TN8400 99-085



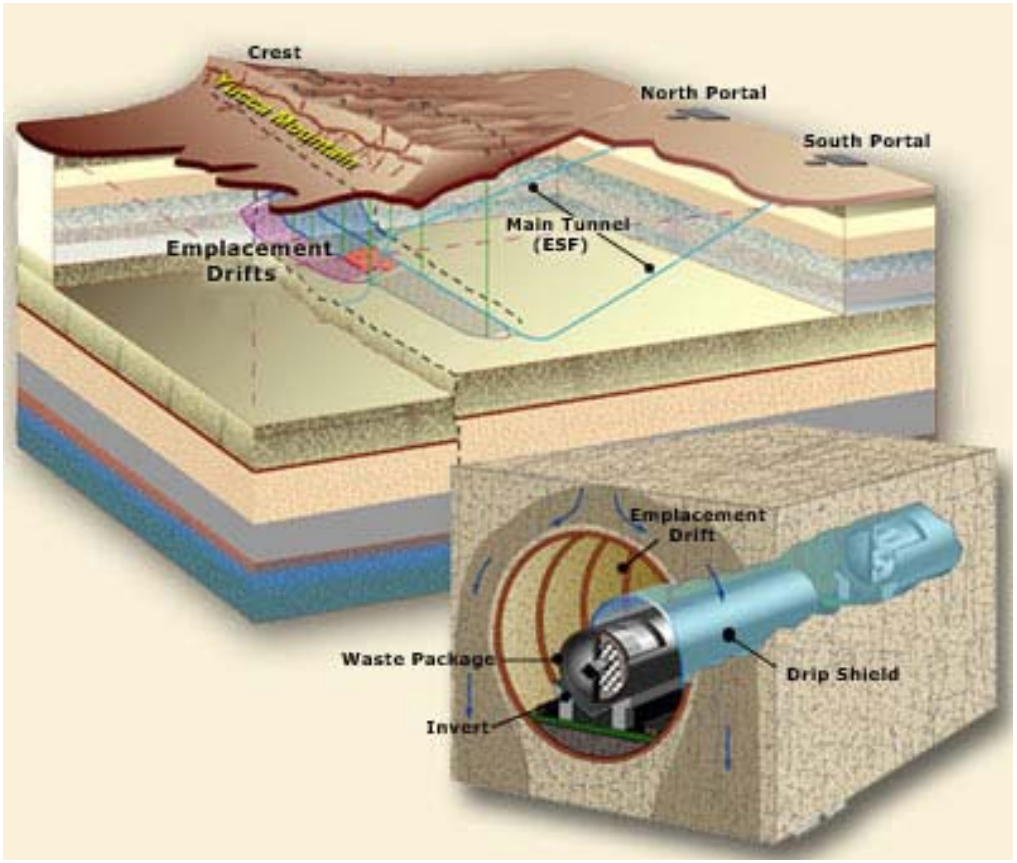


Annual Exposure Dose

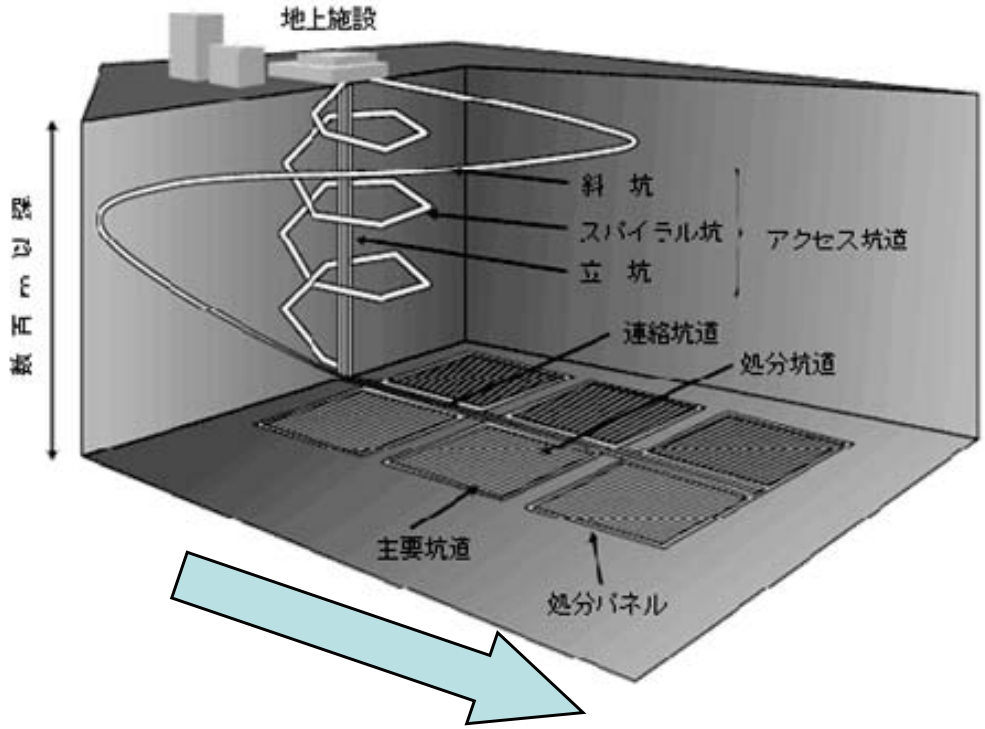


Repository Concepts

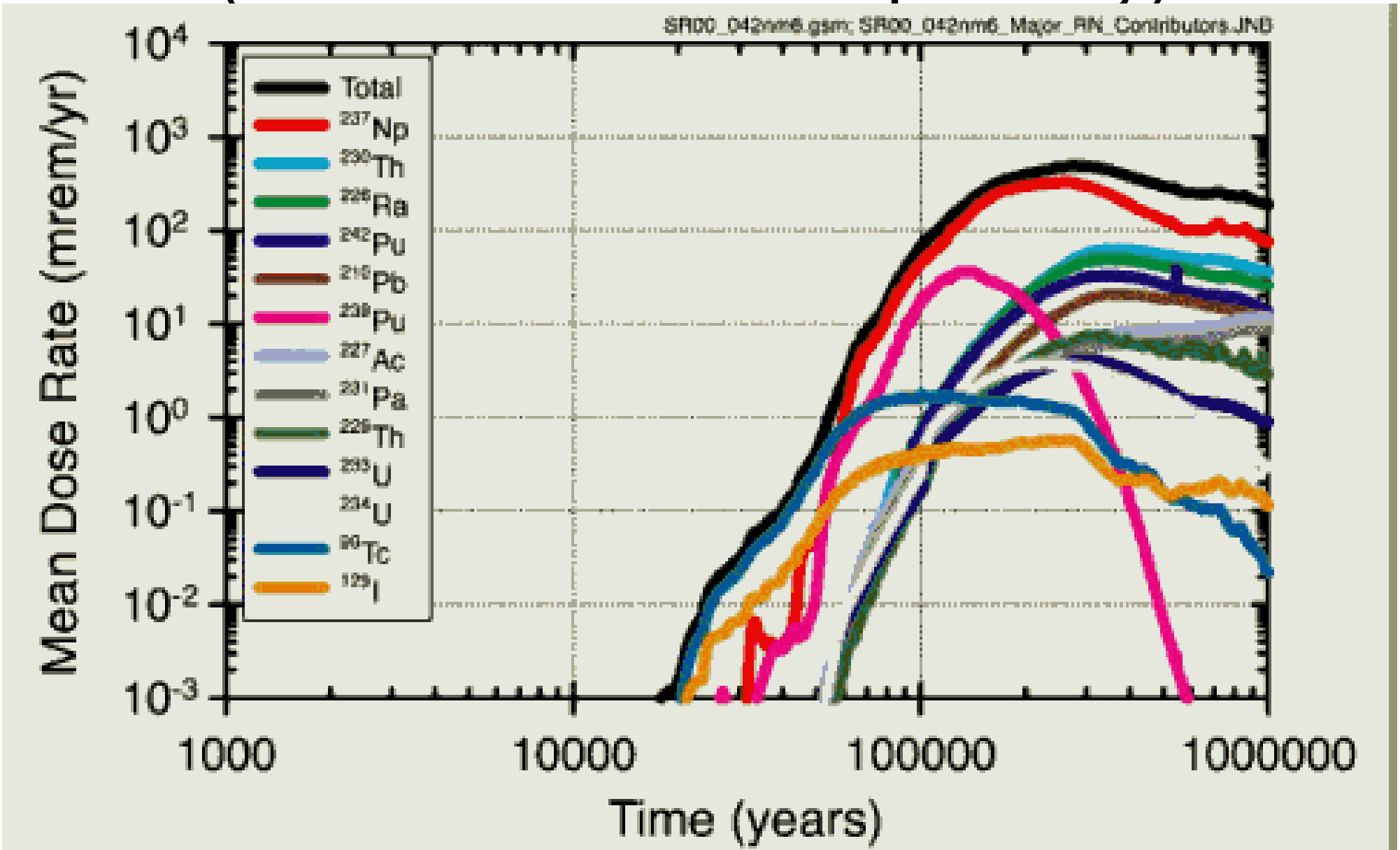
Yucca Mountain Repository



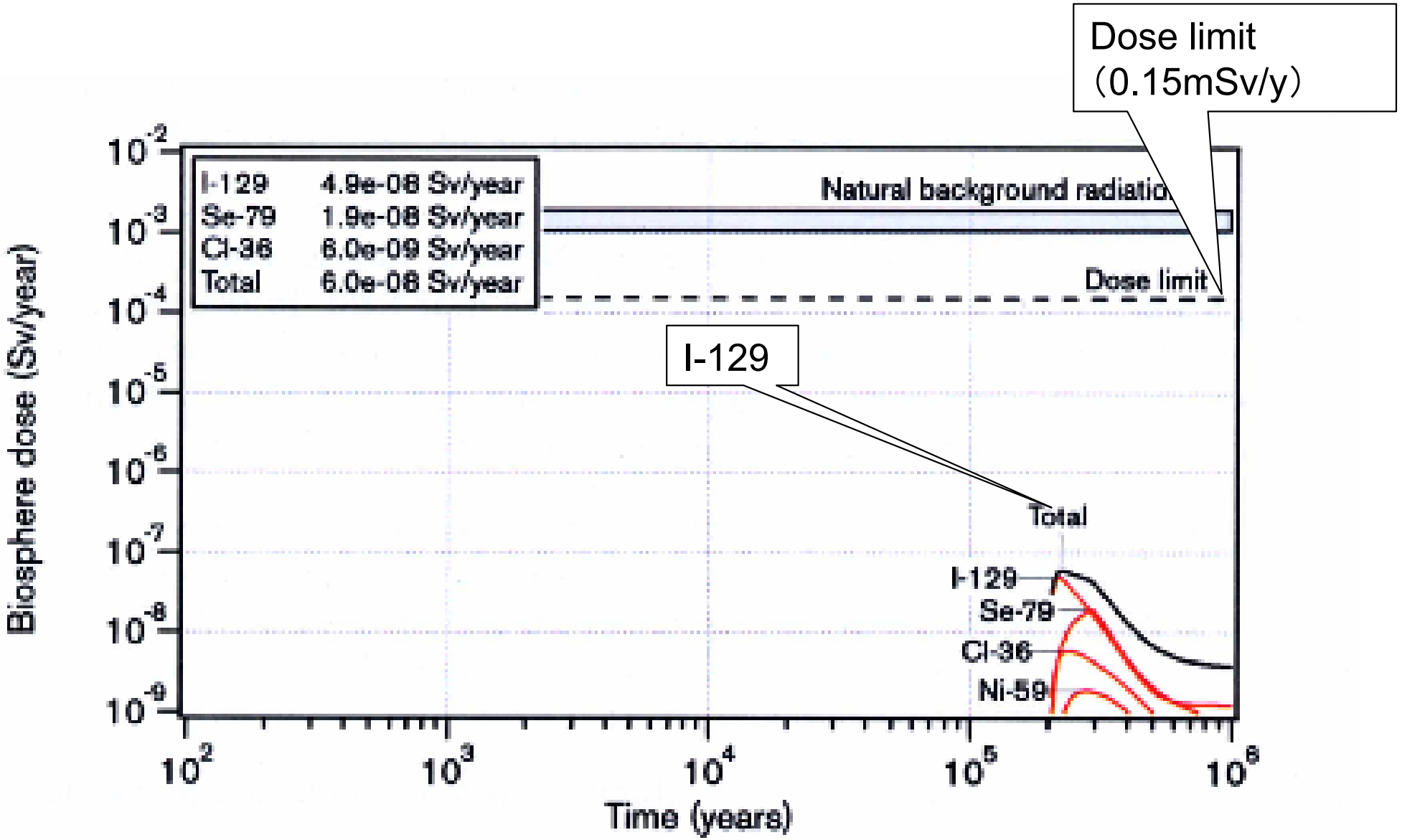
Water-saturated Repository Concept



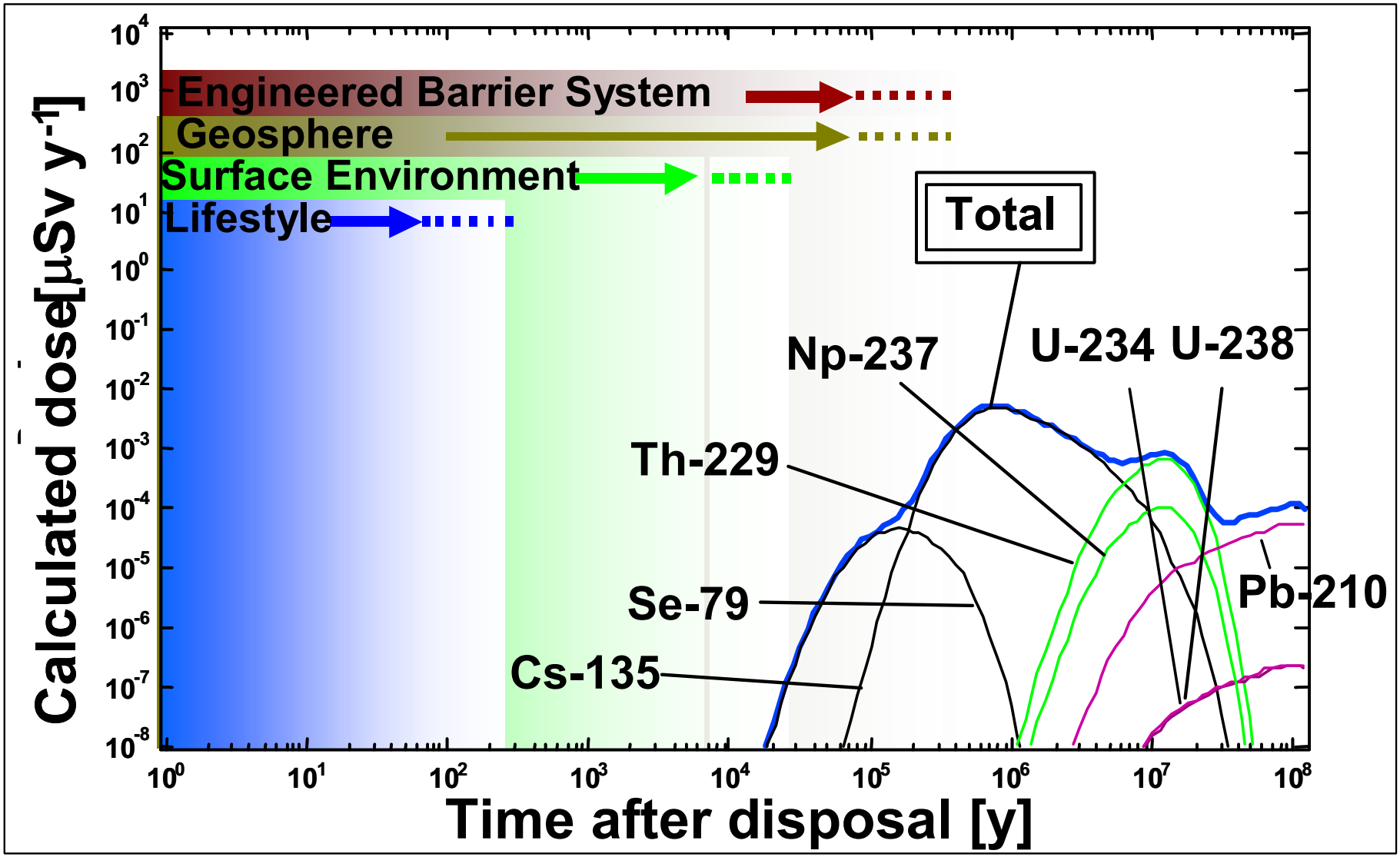
TSPA-SR Results for Nominal Performance case (Yucca Mountain Repository)



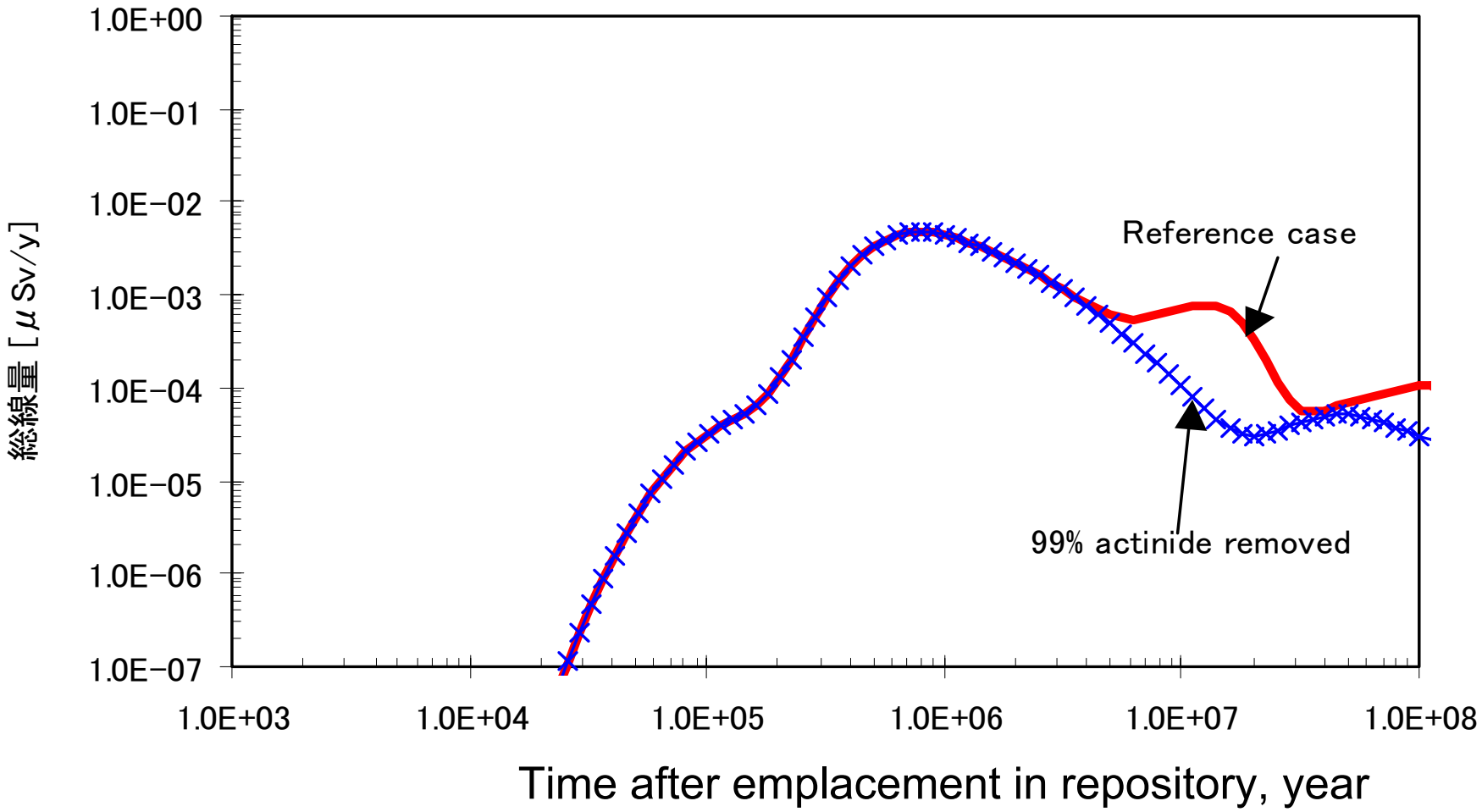
Results of Swedish repository-performance study



Water-Saturated repository (Japanese repository concept, H12)

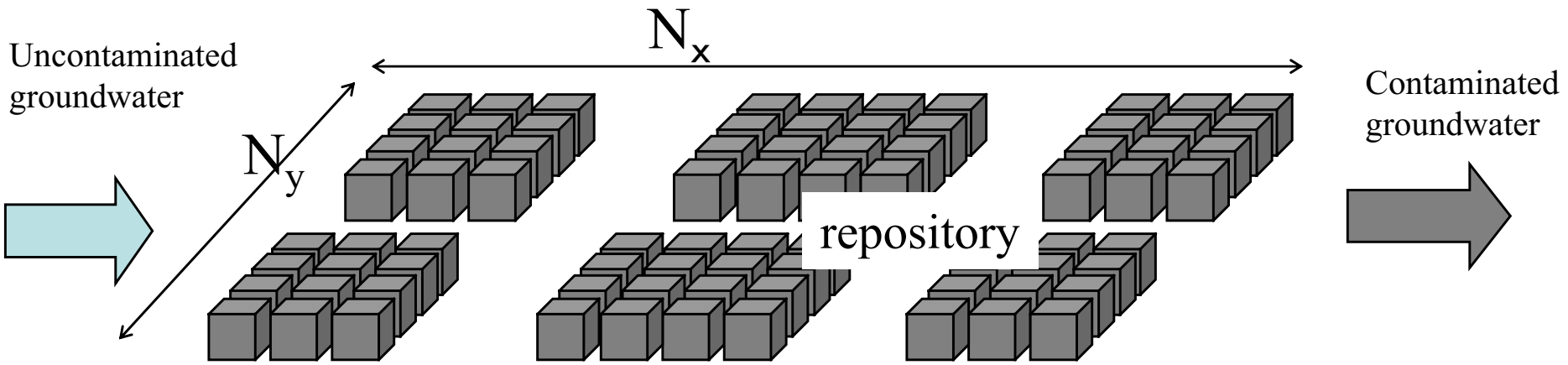


Effects of P&T in terms of exposure dose rate (H12)



Wakasugi, et al., Personal communications

Environmental Impact from nuclide i



Mass loading in a canister $\hat{M}_i^o$

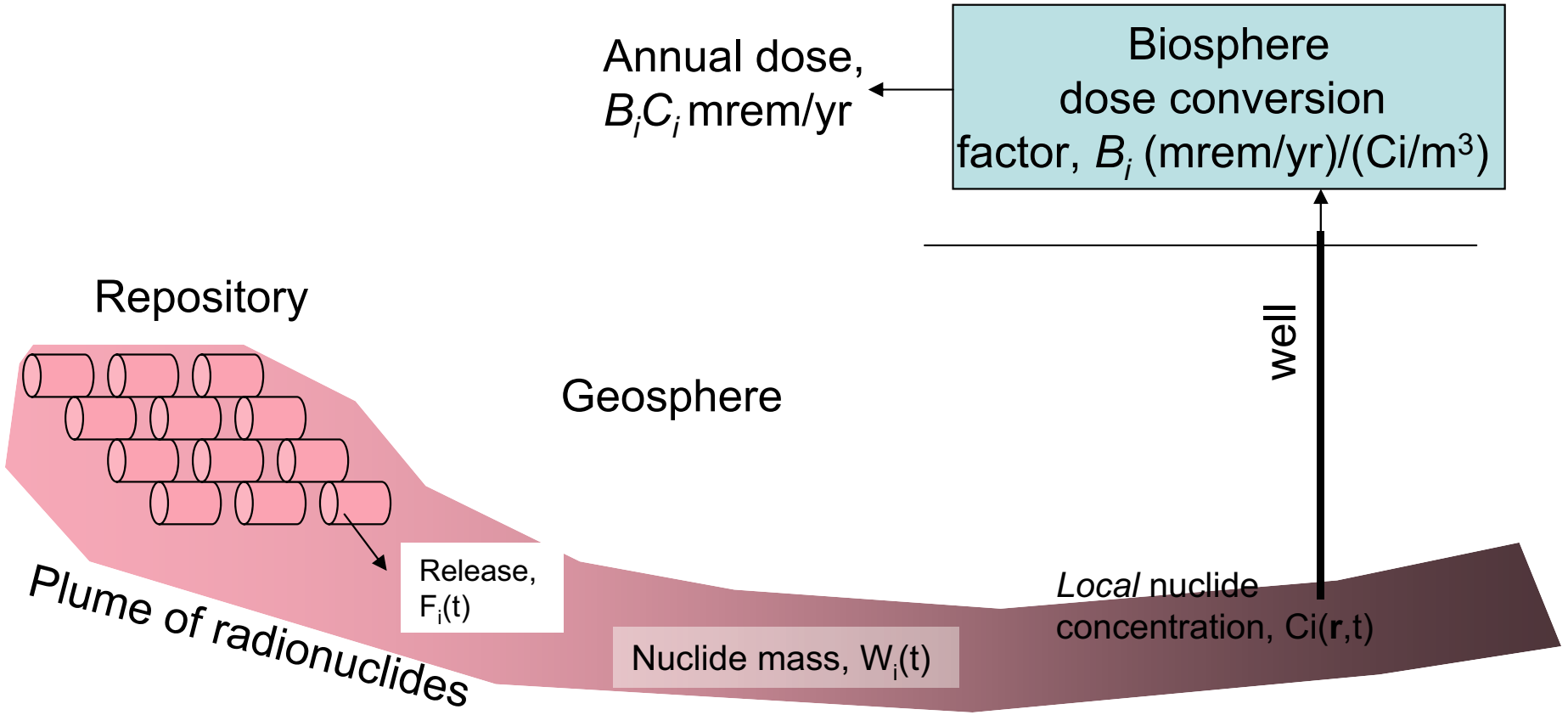
Radionuclide mass: $M_i(t)$

Environmental Impact, $$I_i = \left(N_x N_y \hat{M}_i^o \right) C_i P_i$$

$$C_i \left[\mathrm{m^3\text{-}water/kg\text{-}nuclide} \right] \equiv \frac{\lambda_i \left[/\mathrm{s} \right] \cdot 1000 \left[\mathrm{g/kg} \right] \cdot N_A \left[/\mathrm{mol} \right]}{M_i \left[\mathrm{g/mol} \right] \cdot 3.7 \times 10^{10} \left[\mathrm{Bq/Ci} \right] \cdot \left(\mathrm{MPC} \right)_i \left[\mathrm{Ci/m^3} \right]}$$

P_i is the ratio of the peak mass in the environment to the total initial loading in the repository, of radionuclide i. This function includes repository parameters, and are determined primarily by release rates of radionuclides from the repository region.

Annual Dose and “Environmental Impact”

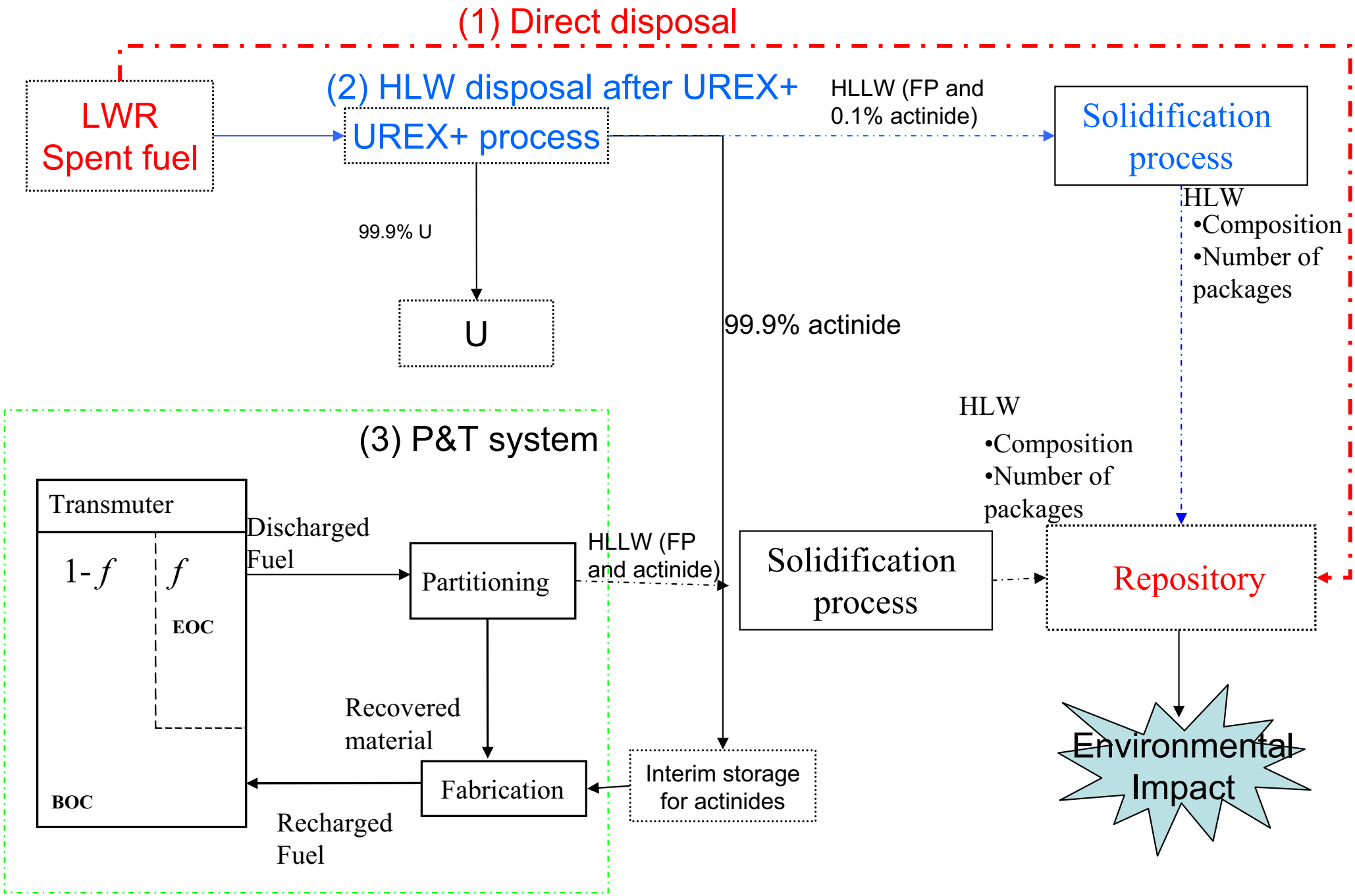


Annual Dose and “Environmental Impact”

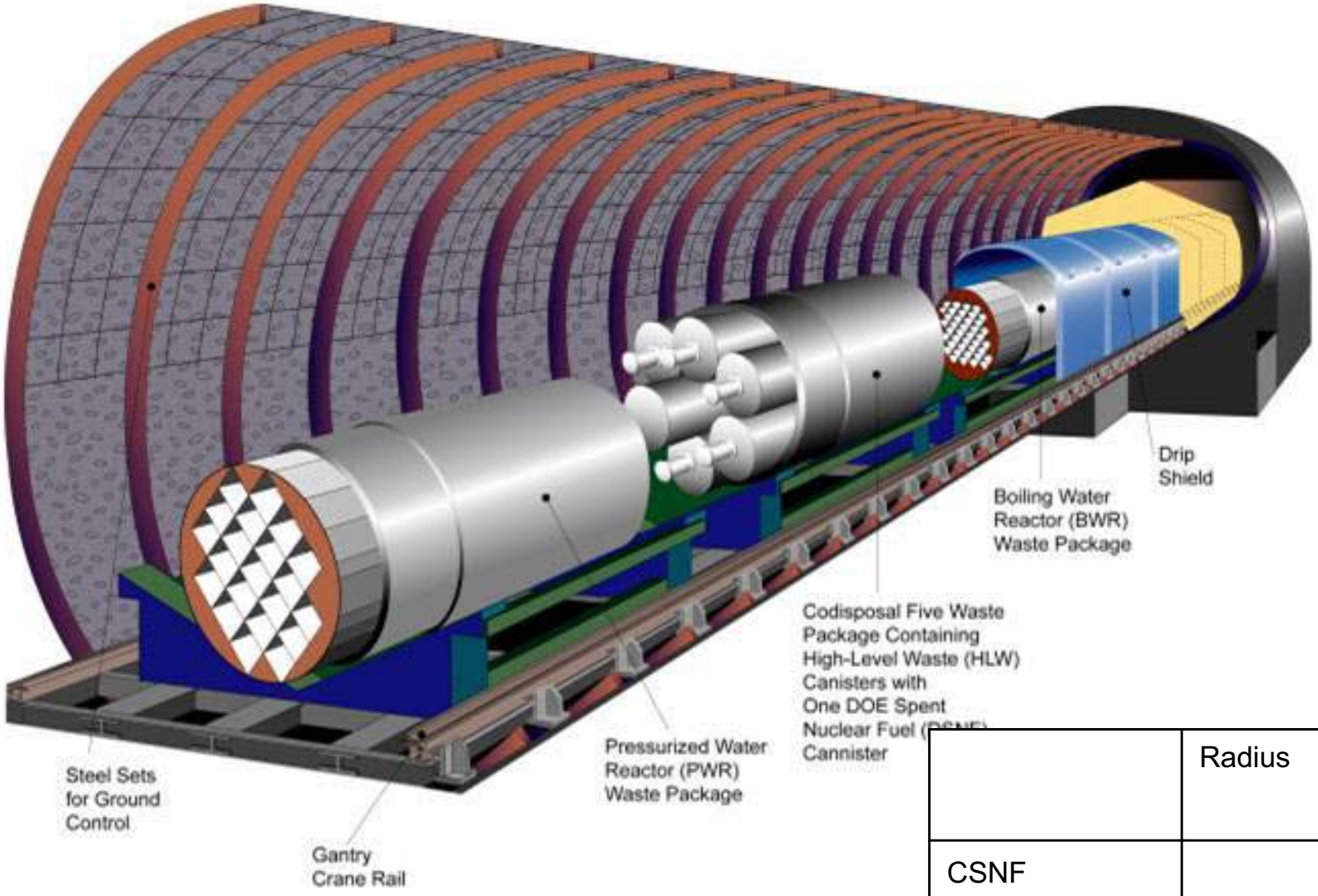
- Annual dose as a repository performance measure
  - Based on assumptions for radionuclide transport to the maximally exposed individual
    - The environment is considered as the natural barrier.
- “Environmental impact” expressed as radiotoxicity of nuclides existing in the environment
  - Based on mass of radionuclides existing in the region exterior to the repository
  - Associated with smaller uncertainties
    - Radionuclide migration in the geosphere and biosphere is not included.
  - Sensitive to reduction of initial mass loading in the waste – the major efforts to be made by advanced fuel cycles

Modeling for Assessment of Environmental Impact
for Yucca Mountain Repository

(1) Commercial Spent Nuclear Fuel (CSNF) + Defense Wastes (DW) – Base case --
(2) Vitrified HLW from reprocessing for CSNF
(3) P&T Mass Flow

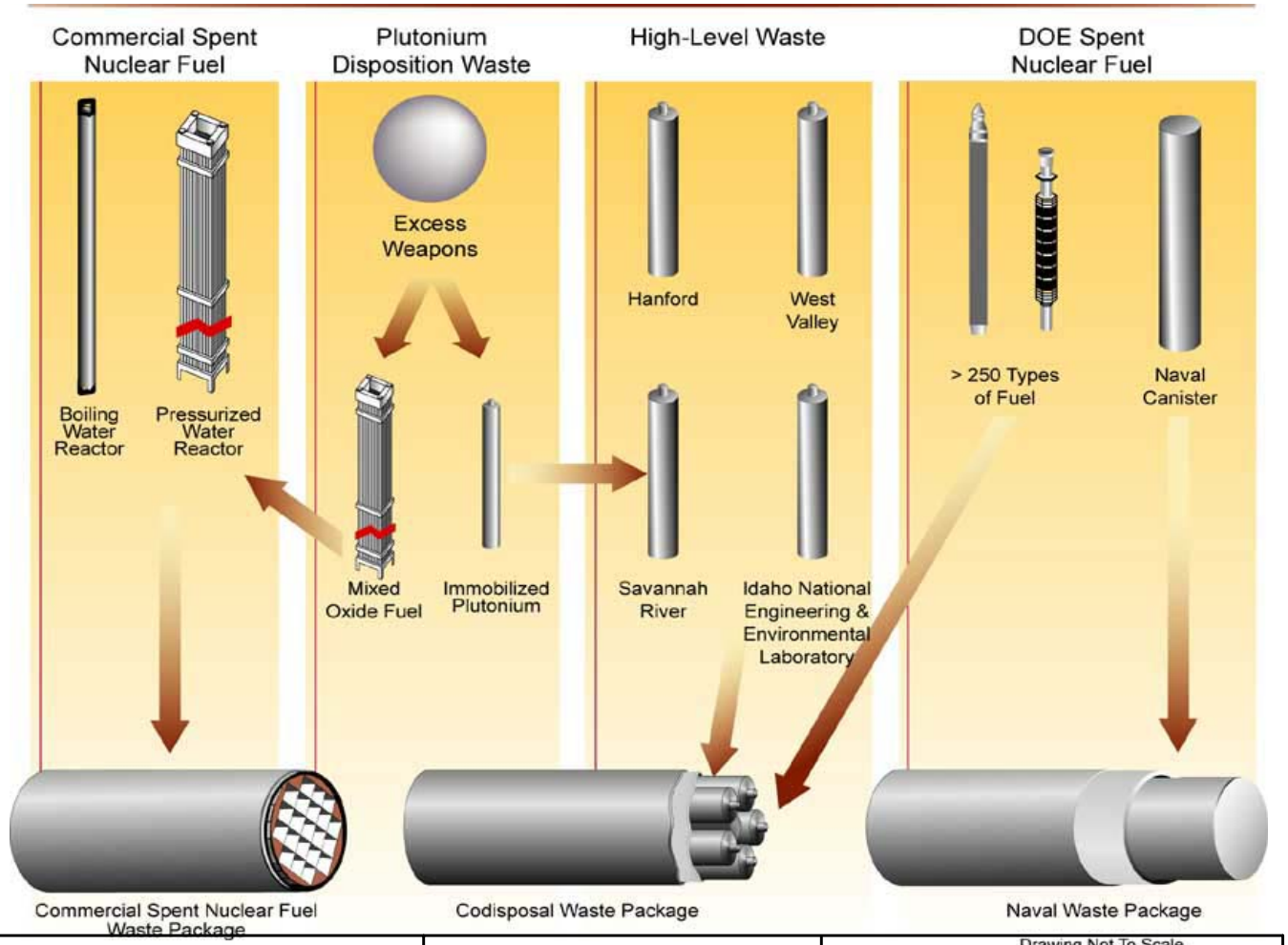

(1) Direct disposal
(2) HLW disposal after UREX+
LWR
Spent fuel
UREX+ process
HLLW (FP and
0.1% actinide)
Solidification
process
HLW
•Composition
•Number of
packages
99.9% U
U
99.9% actinide
(3) P&T system
HLW
•Composition
•Number of
packages
Transmuter
1-f
f
EOC
BOC
Discharged
Fuel
Partitioning
HLLW (FP
and actinide)
Solidification
process
Repository
Recovered
material
Fabrication
Interim storage
for actinides
Recharged
Fuel
Environmental
Impact

Yucca Mountain Repository Design



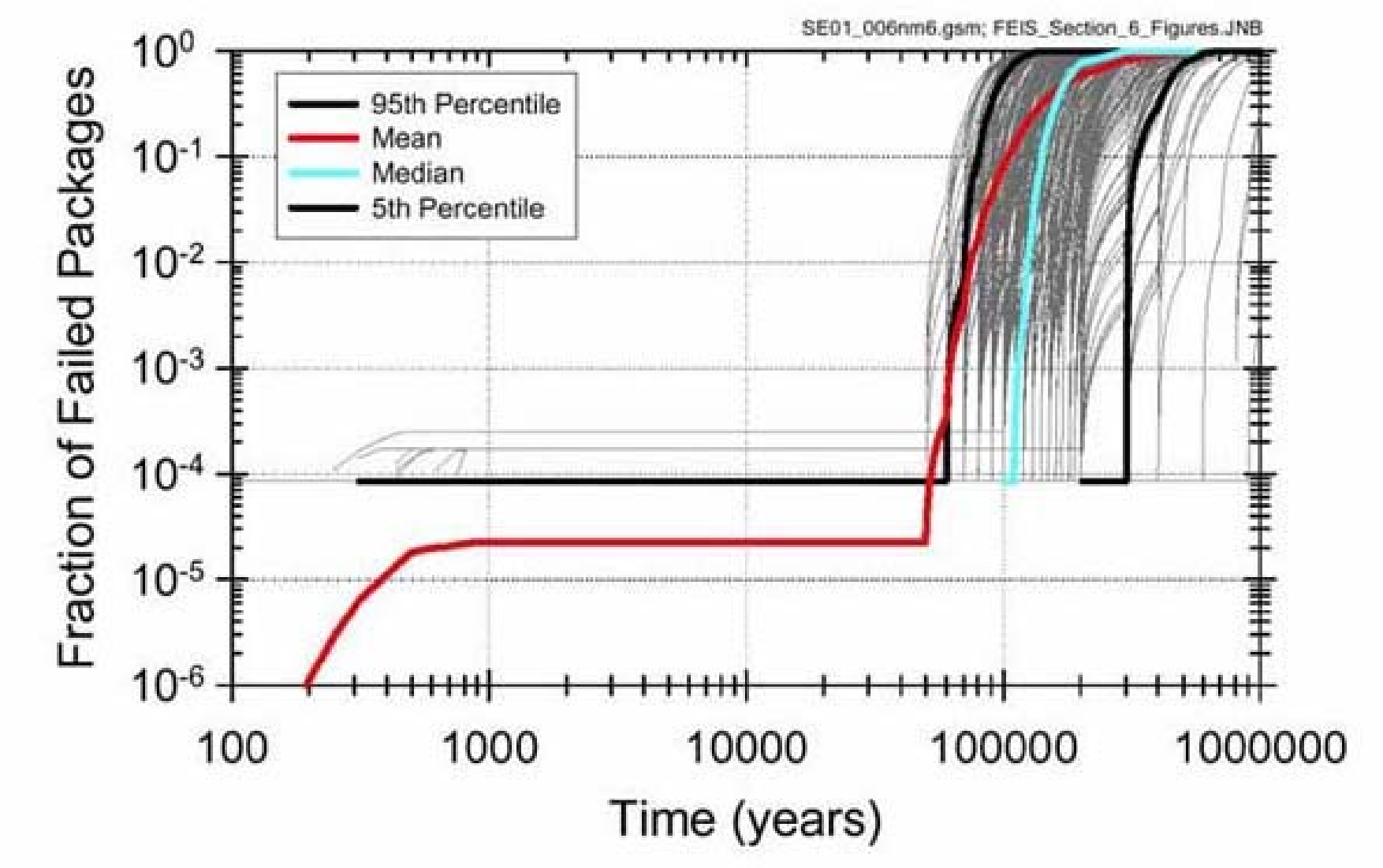
		Radius	Length
CSNF		0.79 m	5.2 m
DW	Co-disp		
	Naval	0.975 m	6.1 m

Waste Package Configurations

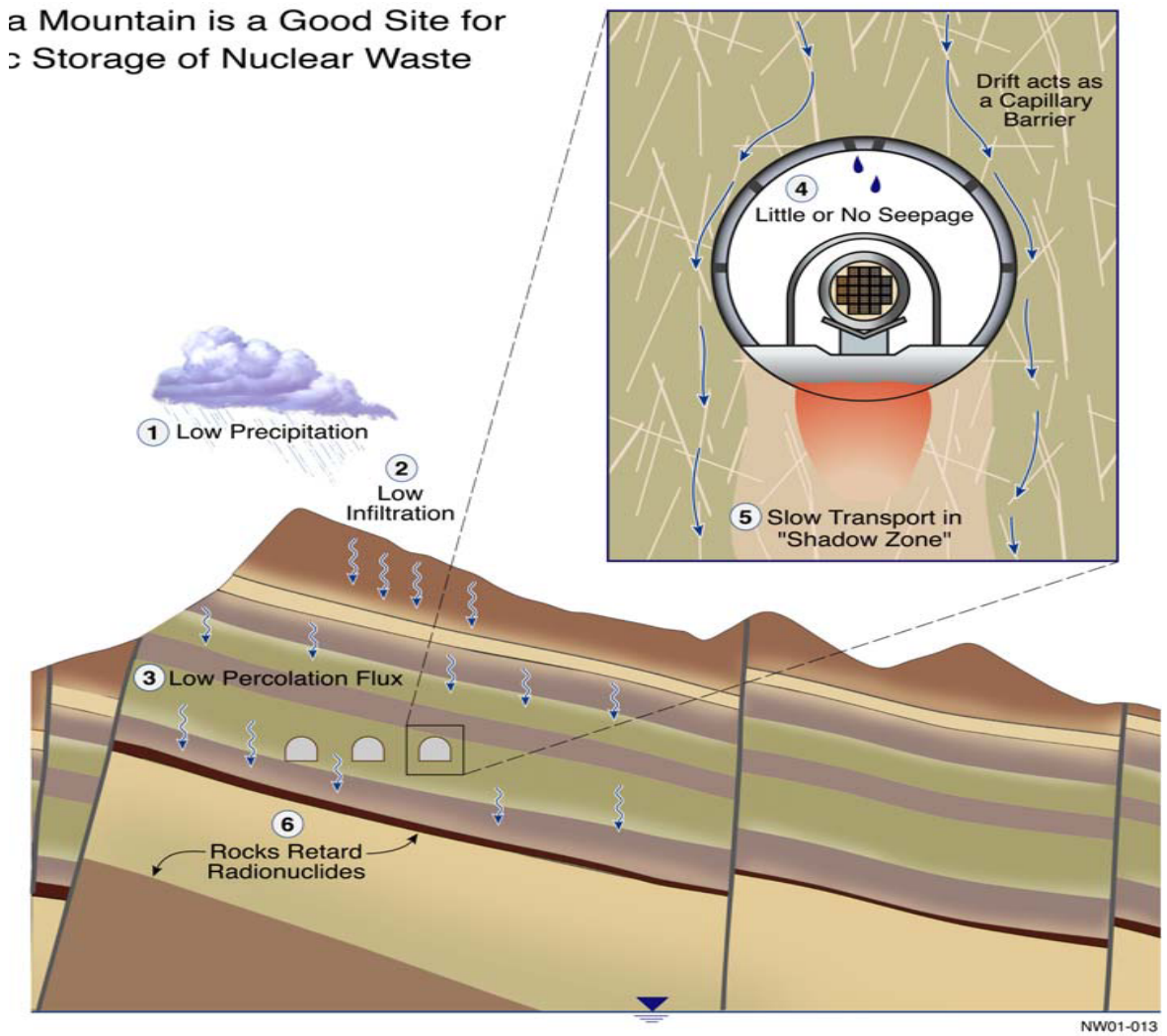


Waste Package Type	CSNF	Co-disposal	Naval SNF
Percent Distribution	67	30	3
Total # of Packages	7886	3511	353

Package Failure Time Statistics



“Drift Shadow”

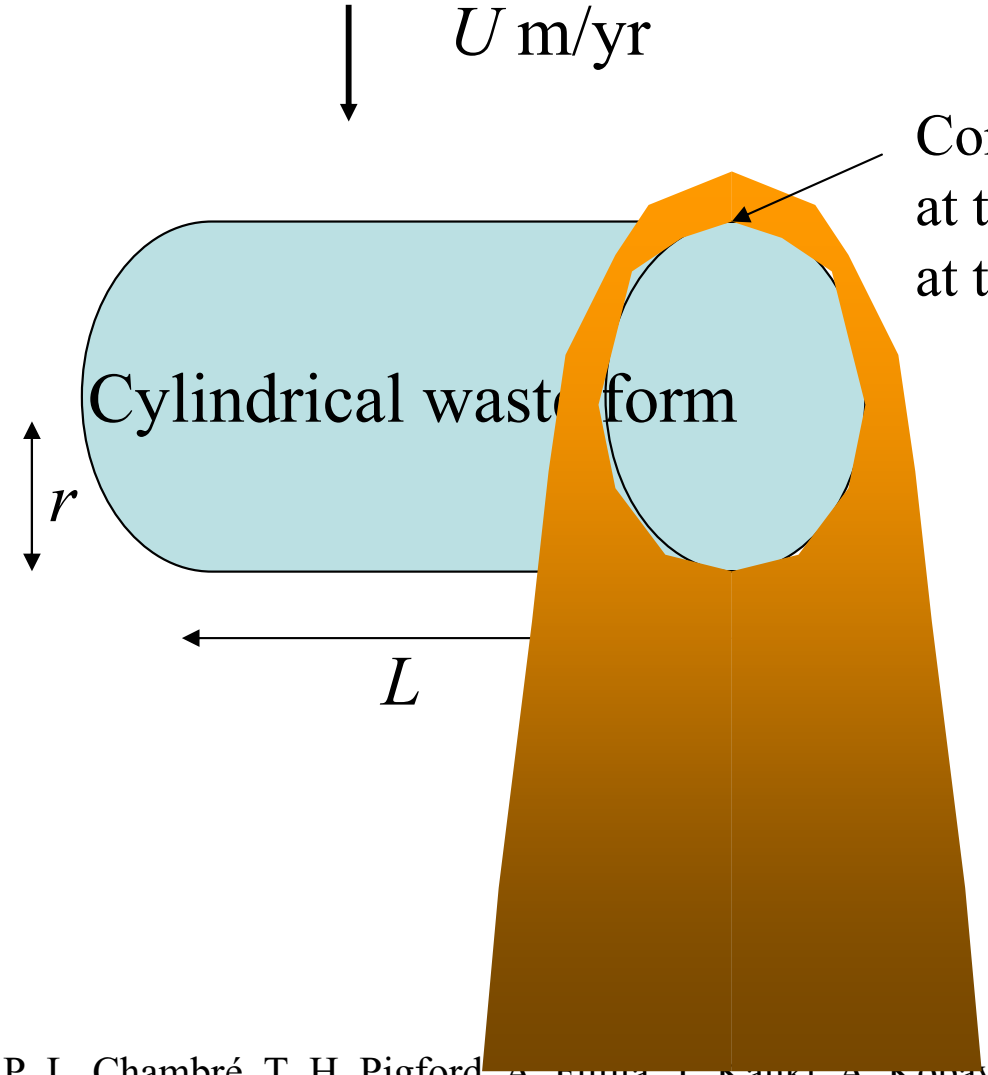


Rates of Release of Radionuclides from Failed packages

- Congruent release:
  - Radionuclides are released at the same fractional release rate with the matrix.
  - Most FP nuclides
- Solubility-limited release:
  - If solubility of radionuclide is so low, then release is limited. The fractional release rate is smaller than that of the matrix.
  - Most TRU nuclides
  - Zr, Sn

•Whichever is smaller is the release rate for the nuclide.

Solubility-Limited Release Rate



Concentration of dissolving species at the surface of the cylinder is constant at the solubility

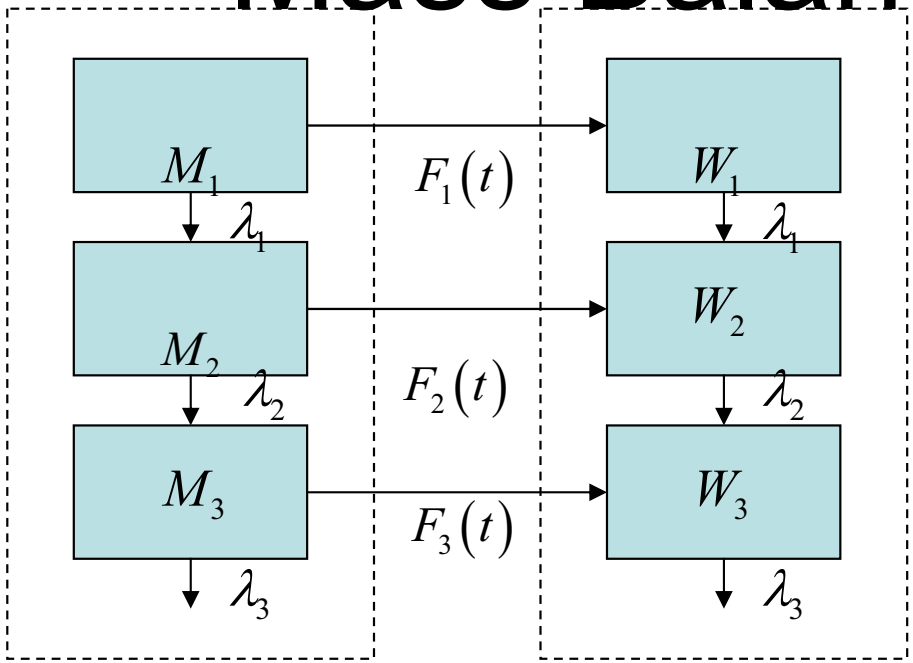
$$\dot{m}_i^* = 8\varepsilon D_e C_i^* L\sqrt{Pe/\pi},$$

for $Pe > 4$,

where $Pe = \dfrac{Ur}{D_e}$

P. L. Chambré, T. H. Pigford, A. Fujita, T. Kanki, A. Kobayashi, H. Lung, D. Ting, Y. Sato, and S. J. Zavoshy, Analytical Performance Models for Geologic Repositories, LBL-14842, Lawrence Berkeley Laboratory, October 1982.

Mass Balance Equations



$F_i(t) = 0$ before T_f

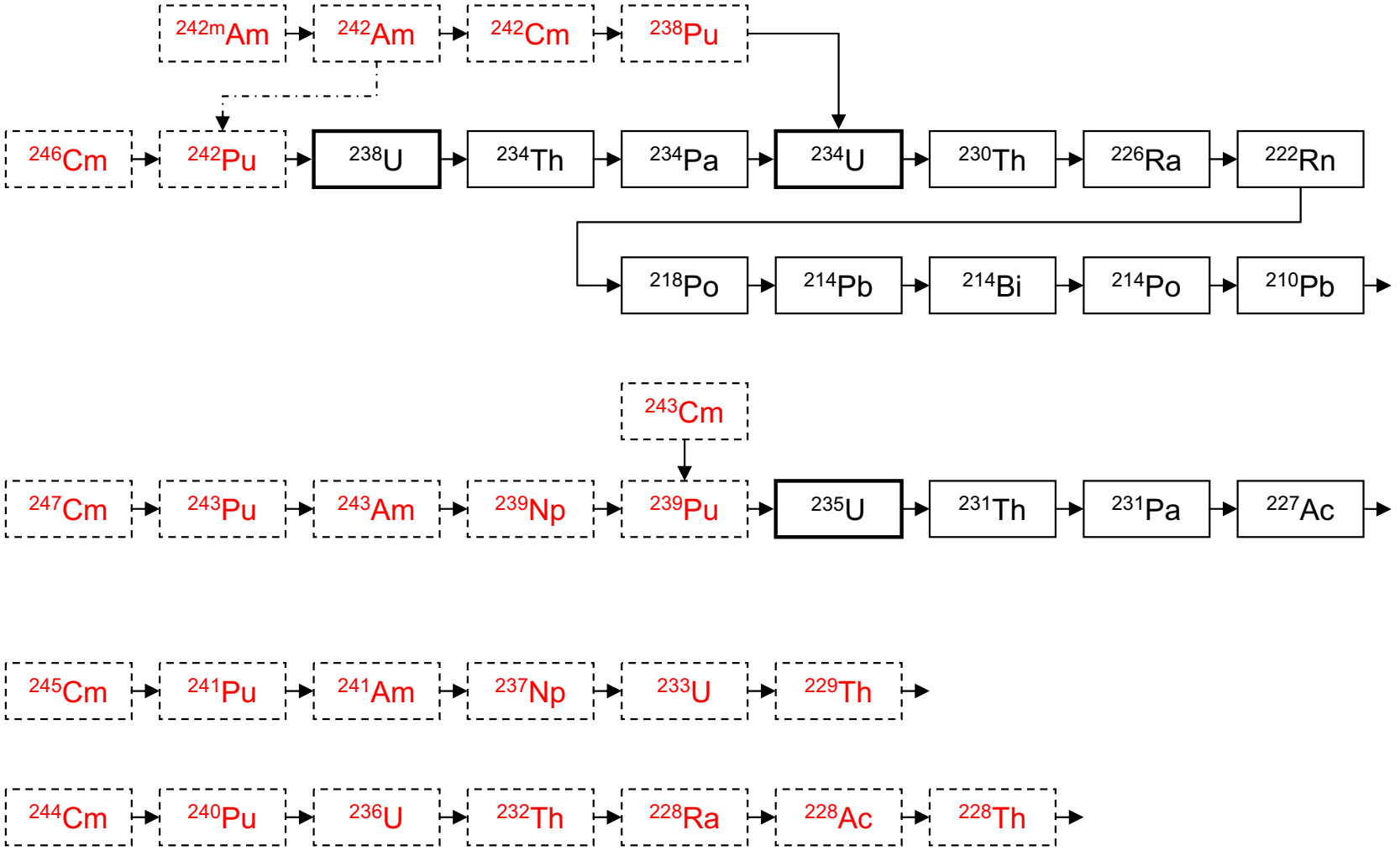
For mass of nuclide *i* in a single waste package:

$$\frac{dM_i(t)}{dt} = -\lambda_i M_i(t) + \lambda_{i-1} M_{i-1}(t) - F_i(t),\ t > 0,\ i = 1,2,\ldots,\ \lambda_0 \equiv 0,$$

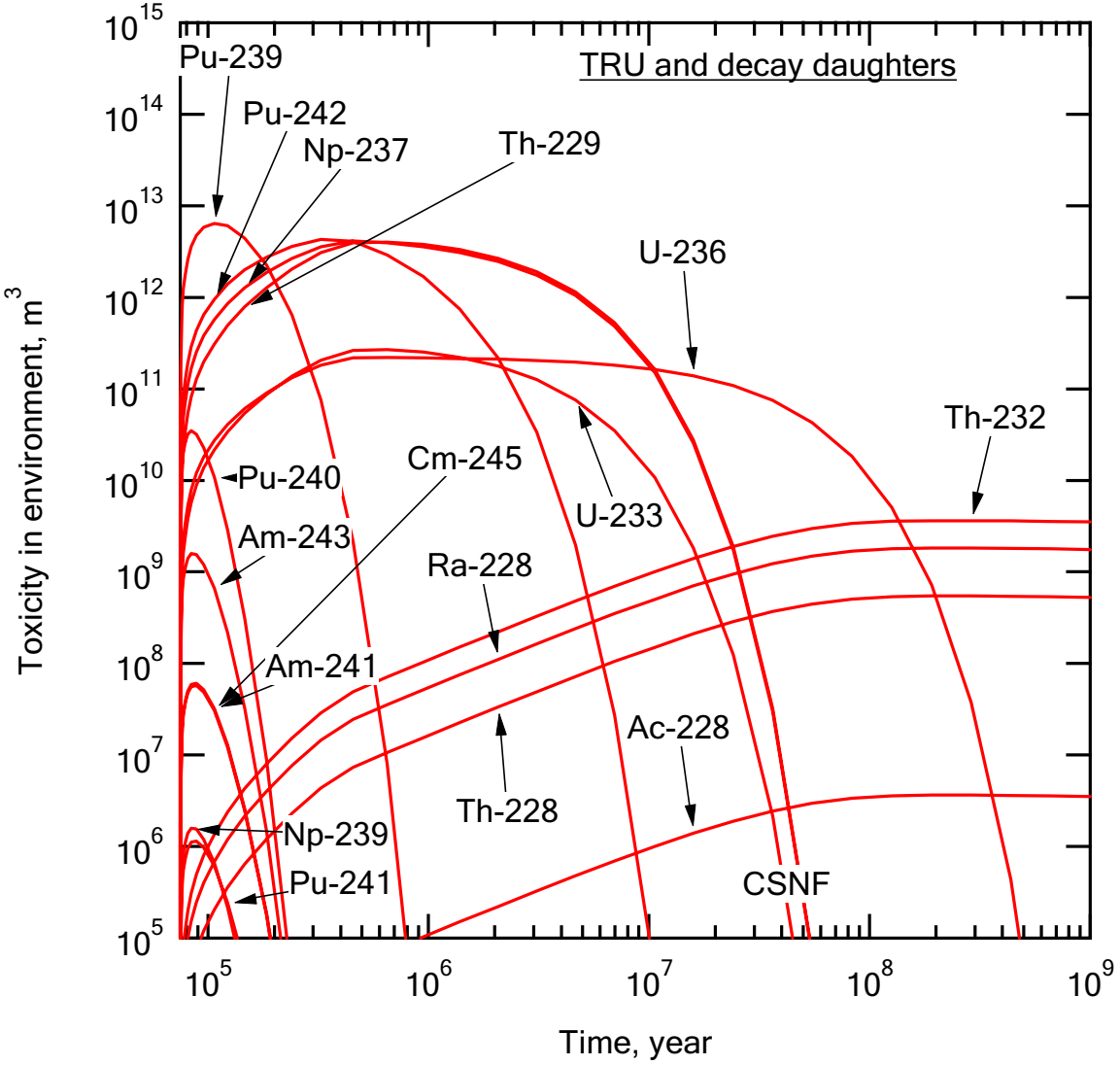
For mass of nuclide *i* in the environment:

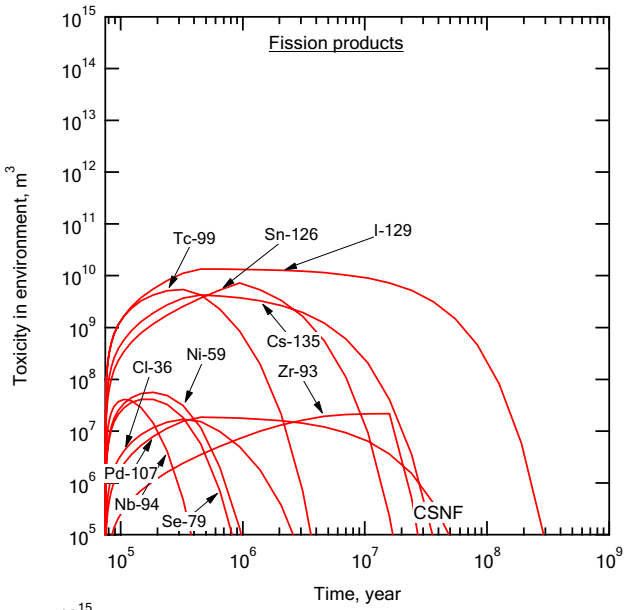
$$\frac{dW_i(t)}{dt} = -\lambda_i W_i(t) + \lambda_{i-1} W_{i-1}(t) + NF_i(t),\ t > 0,\ i = 1,2,\ldots,\ \lambda_0 \equiv 0,$$

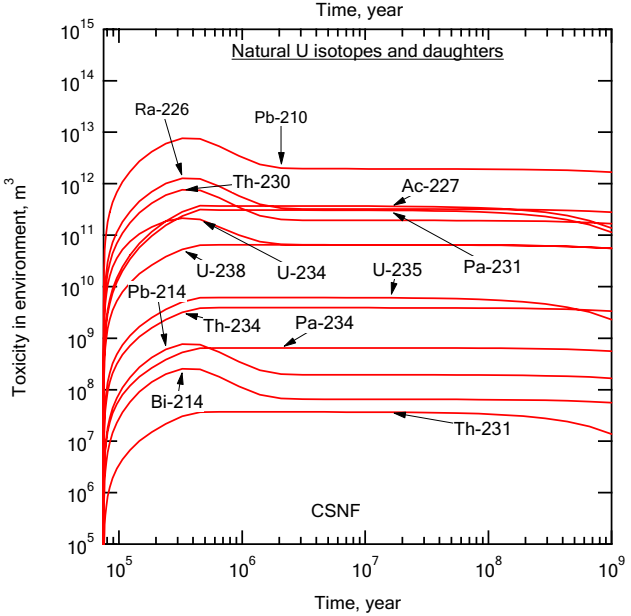
Decay Chains for Actinides



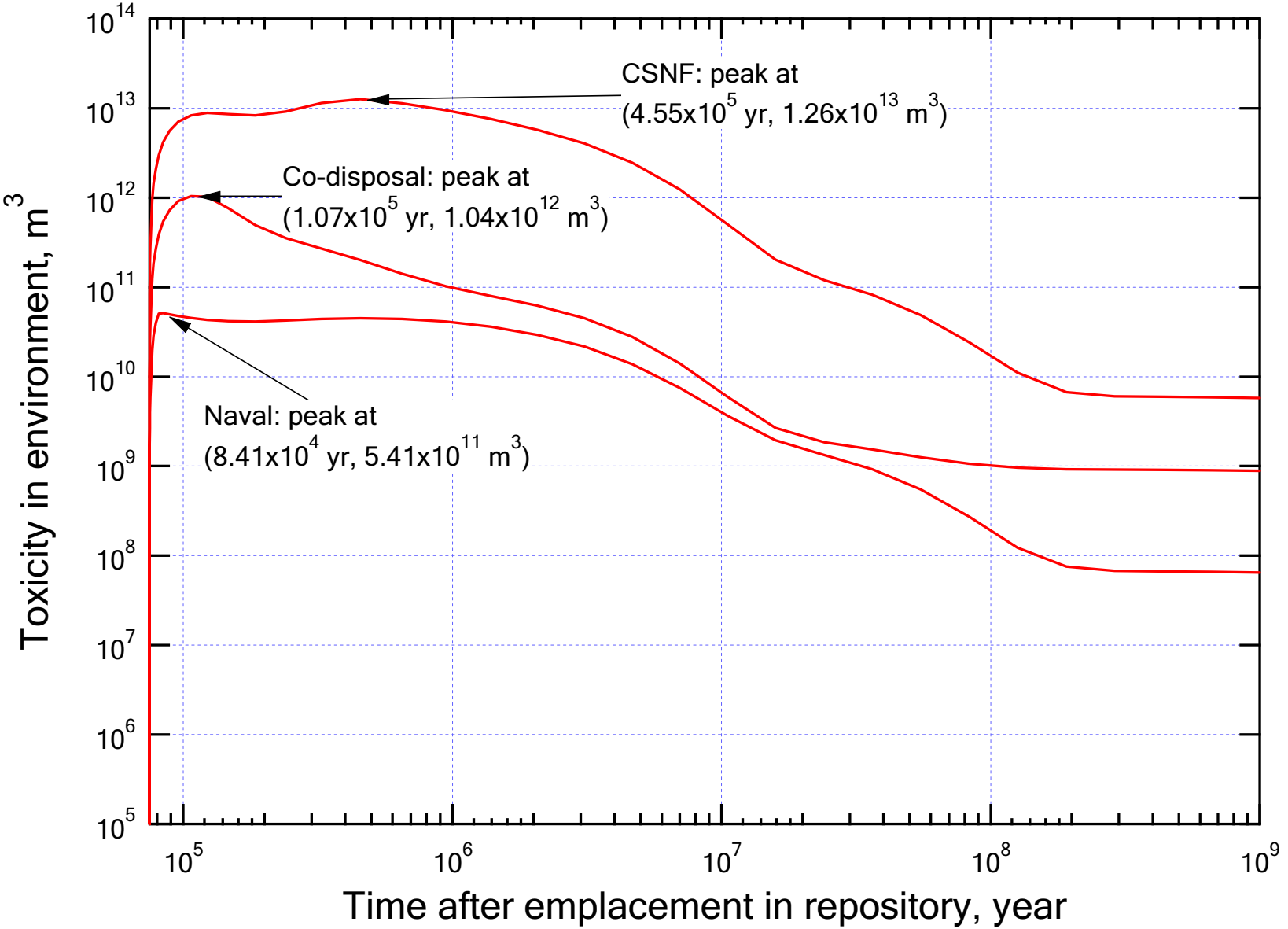
Radiotoxicity in Environment from CSNF







Comparison of EI from CSNF, Co-Disposal, and Naval SNF



Observations for YMR

- Contribution from CSNF packages is dominant for Repository's Environmental Impact.
  - The contribution of the Defense Waste (DW) packages (Co-disposal and Naval SNF packages) is about 10% of the total environmental impact.

➔Environmental impact from DW packages would become invariant base load, and can be used as a reduction target for the CSFN part.

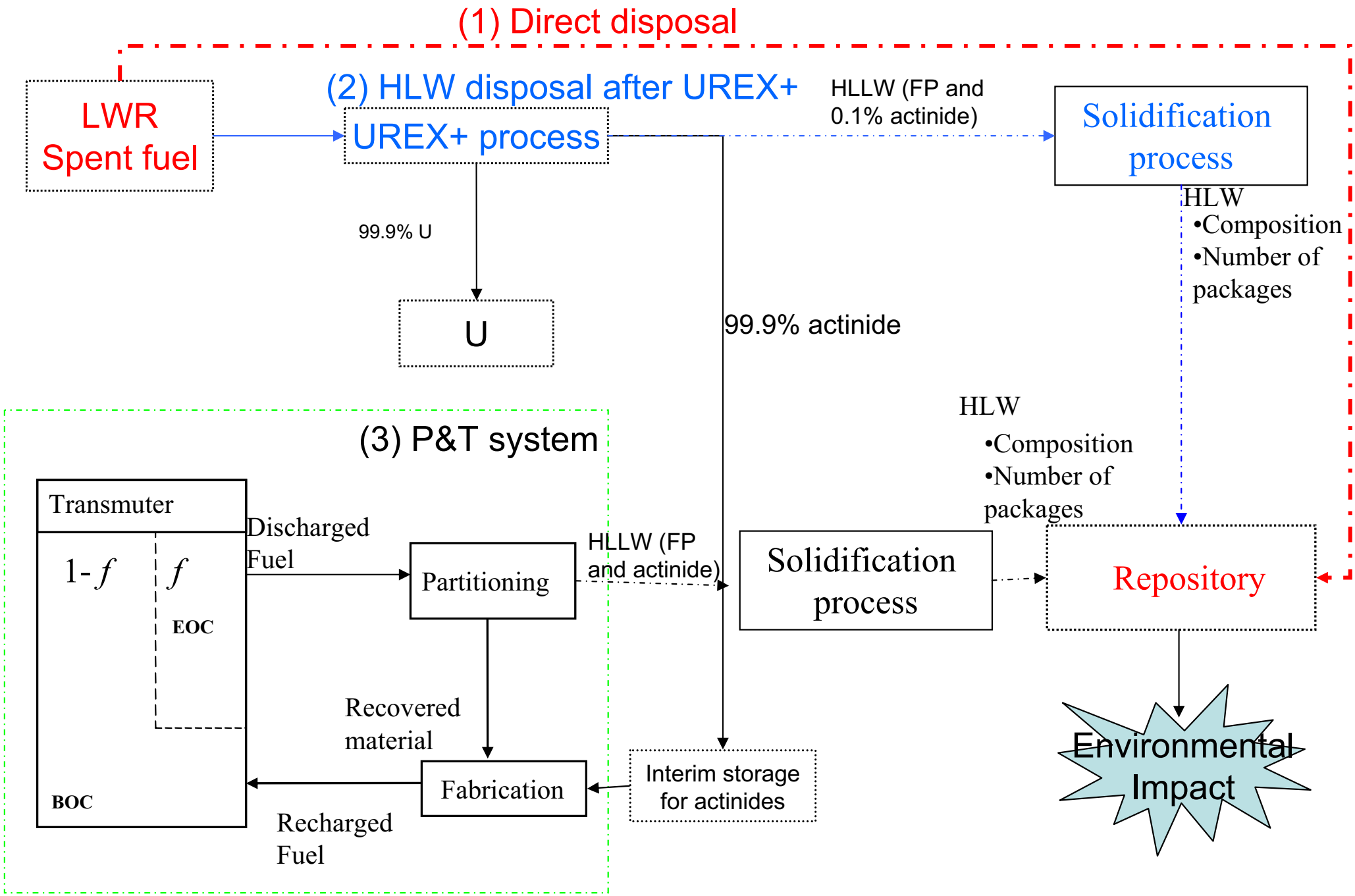
- TRU including their decay daughters give significantly greater impact than FP nuclides.

➔Primary targets for P&T are TRU nuclides.

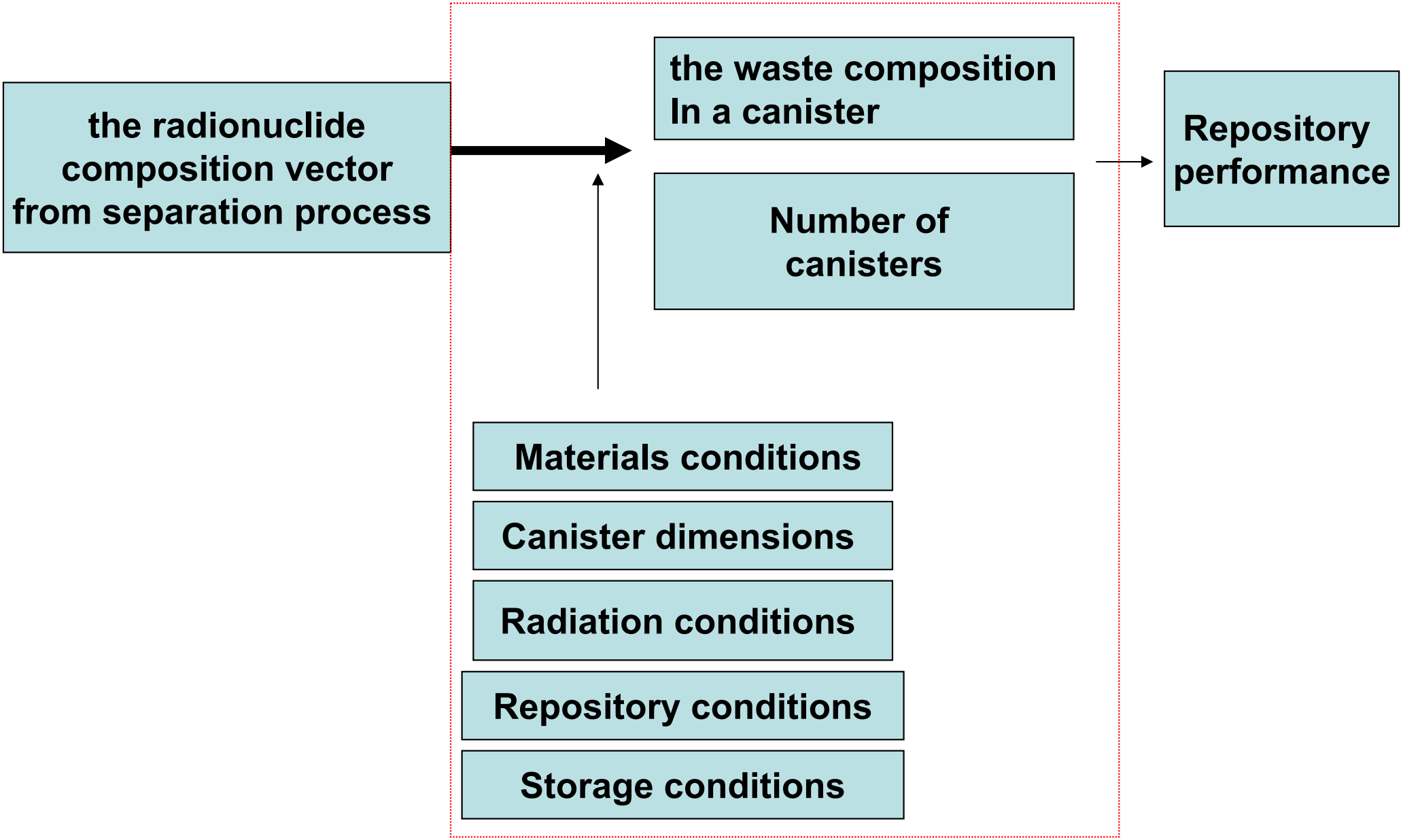
(1) Commercial Spent Nuclear Fuel (CSNF) + Defense Wastes (DW) – Base case --

(2) Vitrified HLW from reprocessing for CSNF

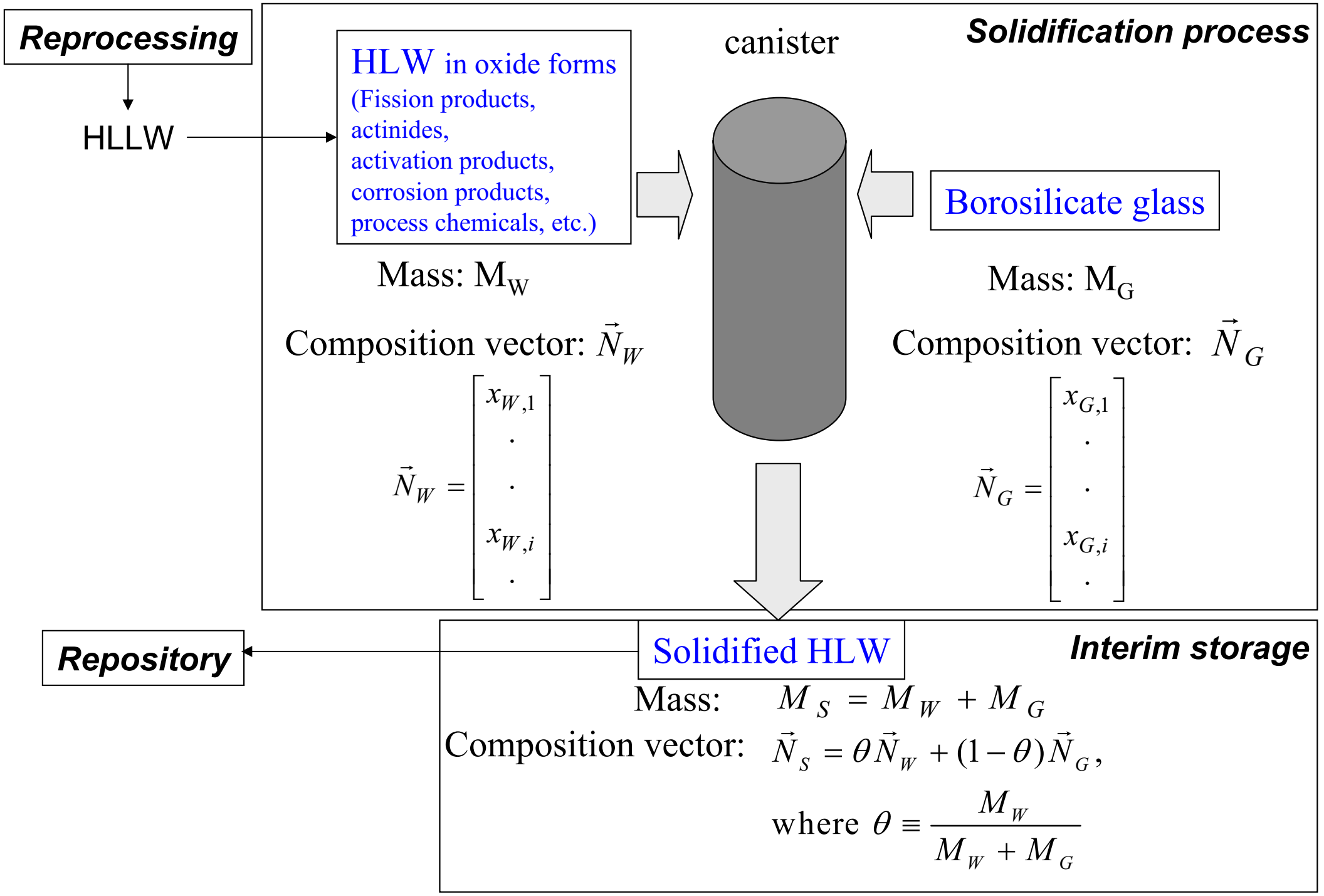
(3) P&T Mass Flow


(1) Direct disposal
(2) HLW disposal after UREX+
LWR
Spent fuel
UREX+ process
HLLW (FP and
0.1% actinide)
Solidification
process
HLW
•Composition
•Number of
packages
99.9% U
U
99.9% actinide
(3) P&T system
HLW
•Composition
•Number of
packages
Transmuter
1-f
f
EOC
BOC
Discharged
Fuel
Partitioning
HLLW (FP
and actinide)
Solidification
process
Repository
Recovered
material
Fabrication
Interim storage
for actinides
Recharged
Fuel
Environmental
Impact

Waste conditioning model to determine initial mass loading in waste package



Solidification of HLW



Borosilicate glass

Oxide	Wt%
SiO_2	62.30
B_2O_3	19.00
Al_2O_3	6.70
Li_2O	4.00
CaO	4.00
ZnO	4.00
Total	100.00

• density: ρ = ~ 2.2 g/cm3
• thermal conductivity: ~0.01 W/cmK
Cf. UO2 = 0.03, stainless steel =0.20
• softening temperature:500 ~ 600 oC
(viscosity becomes 1E11 ~ 1E12 Poise)
• de-vitrification
original glass is amorphous (i.e., non crystalline)
re-crystalization is significant in 3 day at 600oC.

• mechanically, vitrified waste should not fragment into very small pieces (< ~ 0.1 mm) on impact.
• irradiation (due to alpha particles from actinide decays)
(1) devitrification
(2) stored energy due to displaced atoms; temperature excursion.
Both shown to be negligible
• Resistance to leaching by water: leach rate *j* = 1E–5 g-glass/cm2-day

Linear Programming (LP) Model

- Standard form of LP problem

$$\textit{Maximize} \quad f = \mathbf{cx}, \qquad \text{——} \quad \textbf{Objective function}$$

$$\textit{subject to} \quad \mathbf{Ax} \le \mathbf{b} \ \textit{and} \ \ \mathbf{x} \ge \mathbf{0} \qquad \text{——} \quad \textbf{Constraints}$$

where **c** = row vector of coefficients of **objective function**,
x = column vector of **independent variables**,
A = matrix of coefficients of **constraint** inequalities,
b = column vector of RHS of constraint inequalities.

- LP model for optimizing HLW conditioning

  - For objective function: $\mathbf{c} = [1, 0]$, $\mathbf{x} = [M_W, M_G]^T$
  - For constraints: **A** and **b** are determined based on regulations/specifications imposed on solidified HLW products.

Constraints Considered for US-DOE vitrification

- Canistered waste weight $\leq$ 2500 [kg]
- 80 % of empty canister < Canistered waste volume

  <100% of an empty canister

  $V_{can} = 0.82\ m^3$
- Mass fraction of glass frit must be between 70% and 85%
- Temperature in vitrified HLW must be below 400˚C (YMR) to avoid devitrification
- Criticality safety: concentration of Pu is limited to 2.5 kg/m^3
- MoO_3 content $\leq$ 2 wt%
- Na_2O content $\leq$ 10 wt%

Summary of Constraints for Vitrification

(1) $M_W + M_G \le 2033 \quad kg$ Filled canister weight

(2) $0.8V_c \le \dfrac{M_W + M_G}{\rho_{WG}} \le 0.98V_c \ \text{m}^3$ Filled HLW glass volume

$$\rho_{WG} = 1230\left(\frac{M_W}{M_W + M_G}\right) + 2419 \ \text{kg/m}^3$$

$2393.7 \le M_W + 1.508M_G \le 2932.3$ approximate

(3) $0.7 \le \dfrac{M_G}{M_W + M_G} \le 0.85, \quad 0.1765M_G \le M_W \le 0.4286M_G$

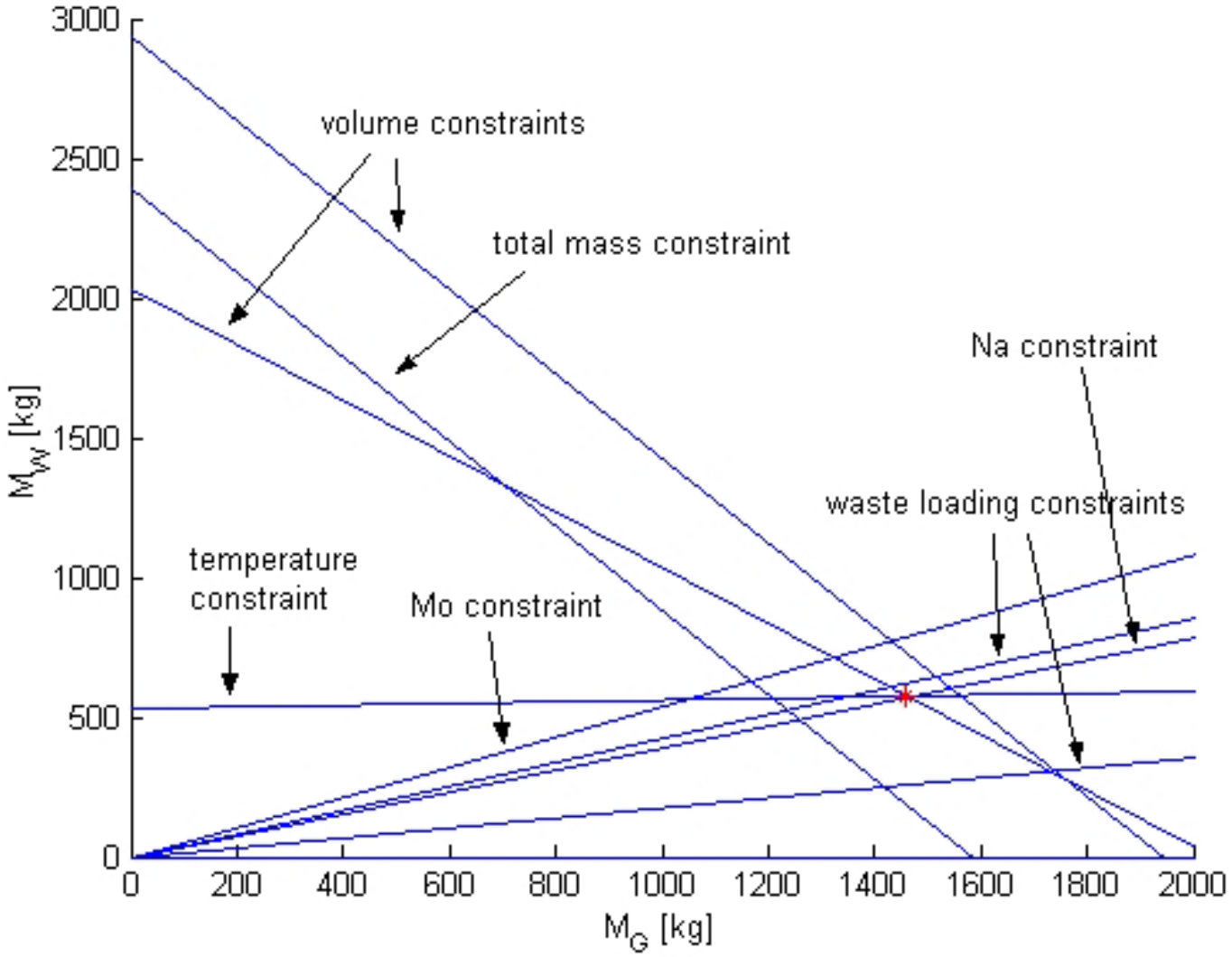
(4) $M_W \le K_1 M_G + K_2, \ 0 \le M_G \le 3000 \ (\text{kg})$ Maximum center temperature

(5) $M_W x_{W,Pu} / V_c \le 2.5 \ \text{kg/m}^3$ Pu concentration limit

(6) $M_W - 0.544M_G \le 0$ Mo-limit

(7) $M_W - 0.417M_G \le 0$ Na-limit

Graphical representation for the feasible solution space



Cooling time before reprocessing and vitrification = 15 years

HLW Glass Compositions & Number of Canisters per ton of CSNF

- Composition Vector of HLW Glass Product:

$$\vec{N}_{WG} = r\vec{N}_W + (1-r)\vec{N}_G$$

where $\vec{N}_W$ = composition vector of HLW before vitrification (known)

$\vec{N}_G$ = composition vector of glass frit before vitrification (known)

r = HLW waste loading fraction (determined by LP model)

For $r = 0.282$

$$\vec{N}_{WG} = \frac{573}{573+1460}\vec{N}_W + \frac{1460}{573+1460}\vec{N}_G$$

$$= 0.282\vec{N}_W + 0.718\vec{N}_G$$

- Canisters produced from 1 MTU of PWR-Spent Fuel

  = 0.184 [Canister/MT]

- Number of Canisters from 63,000 MTHM

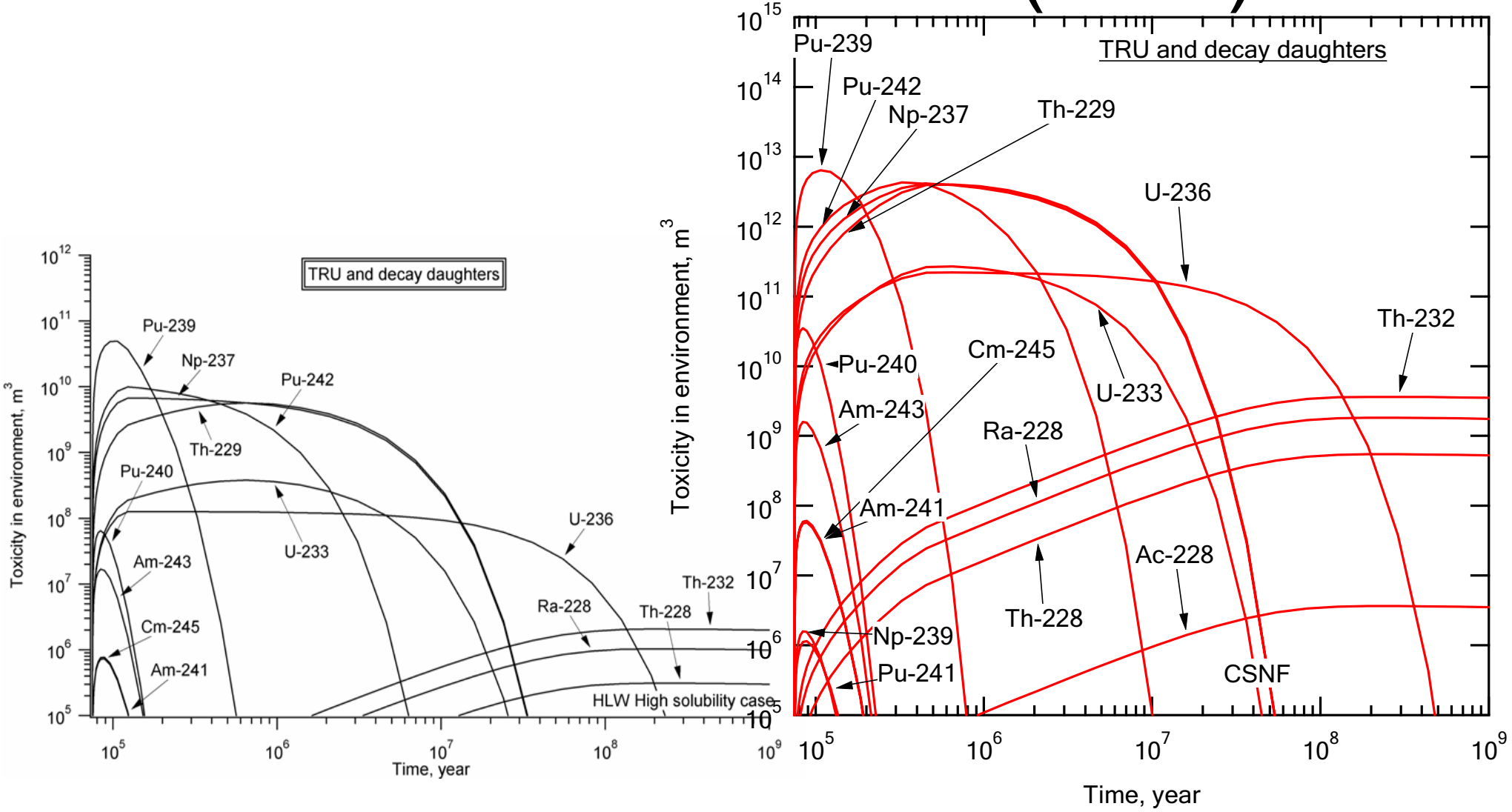
  = 63000 [MT] × 0.184 [Canister/MT]

  =11600 canisters

- Number of Packages from 63,000 MTHM

  =11600 canisters/5 =2320 packages

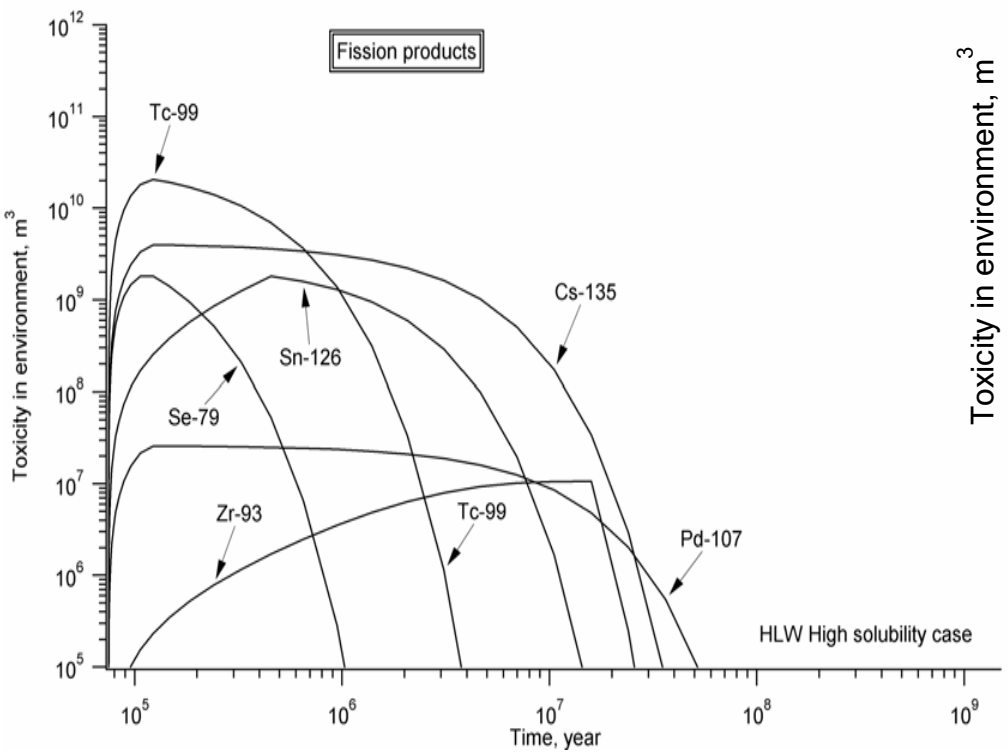
Environmental Impact from Vitrified HLW after UREX+ (TRU)



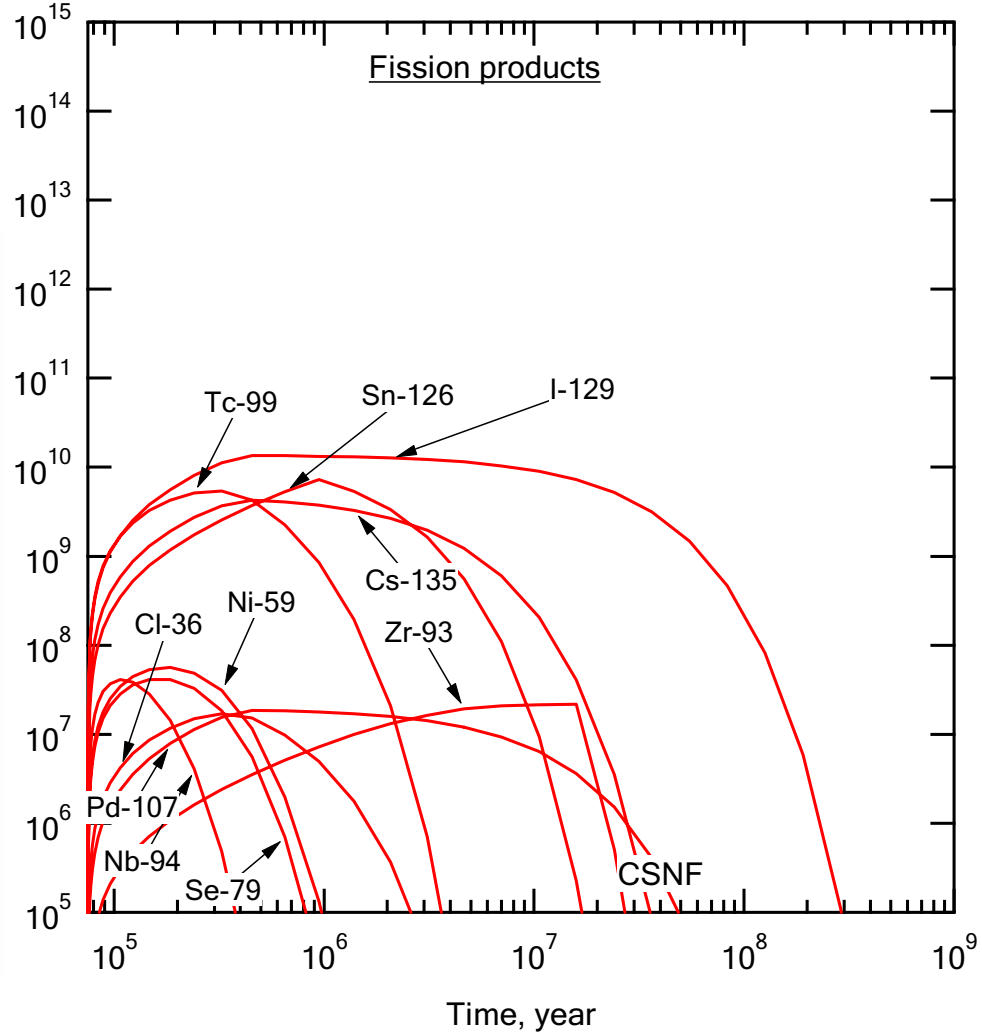
Vitrified HLW

CSNF

Environmental Impact from Vitrified HLW after UREX+ (FP)

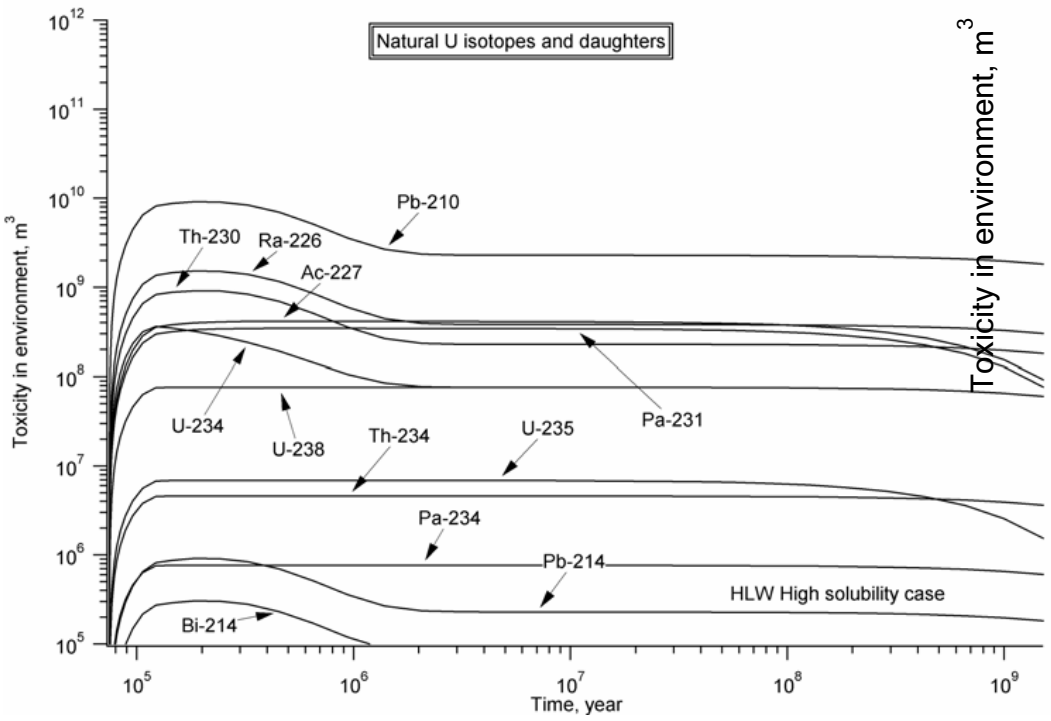


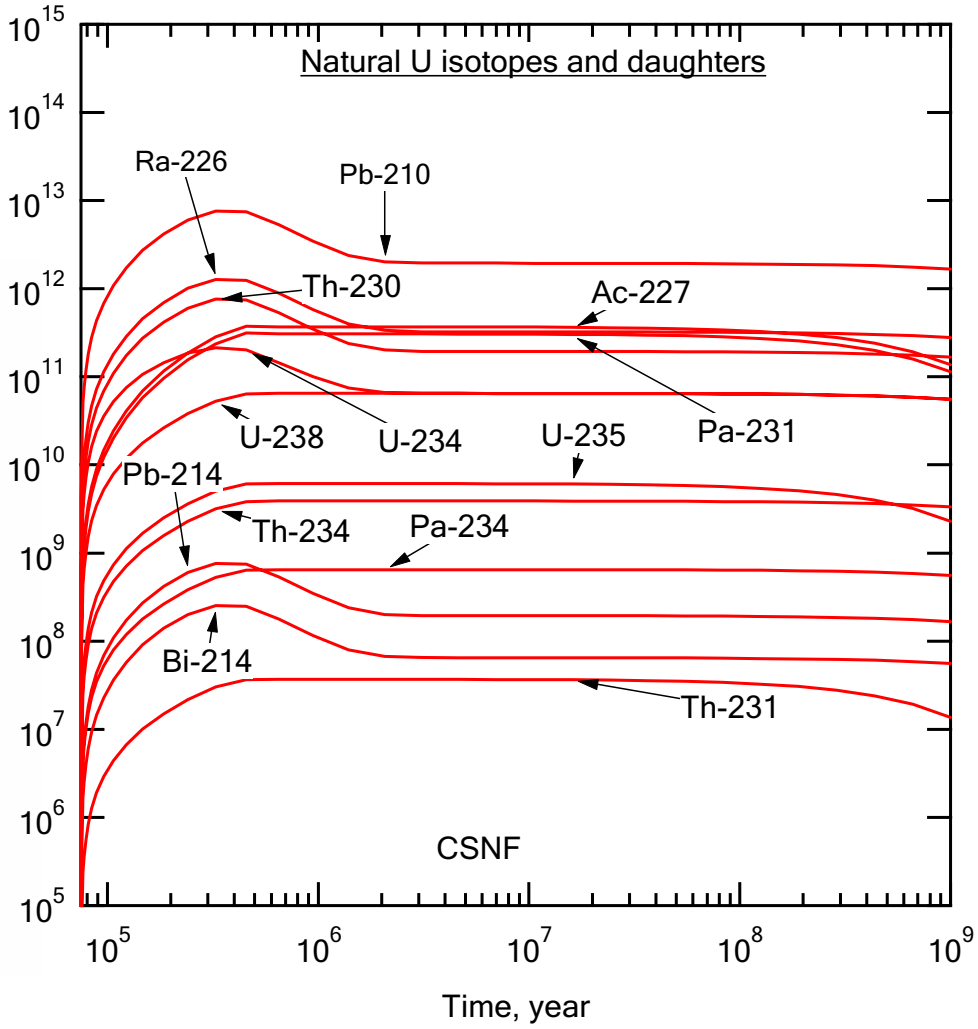
Vitrified HLW



CSNF

Environmental Impact from Vitrified HLW after UREX+ (Natural U)

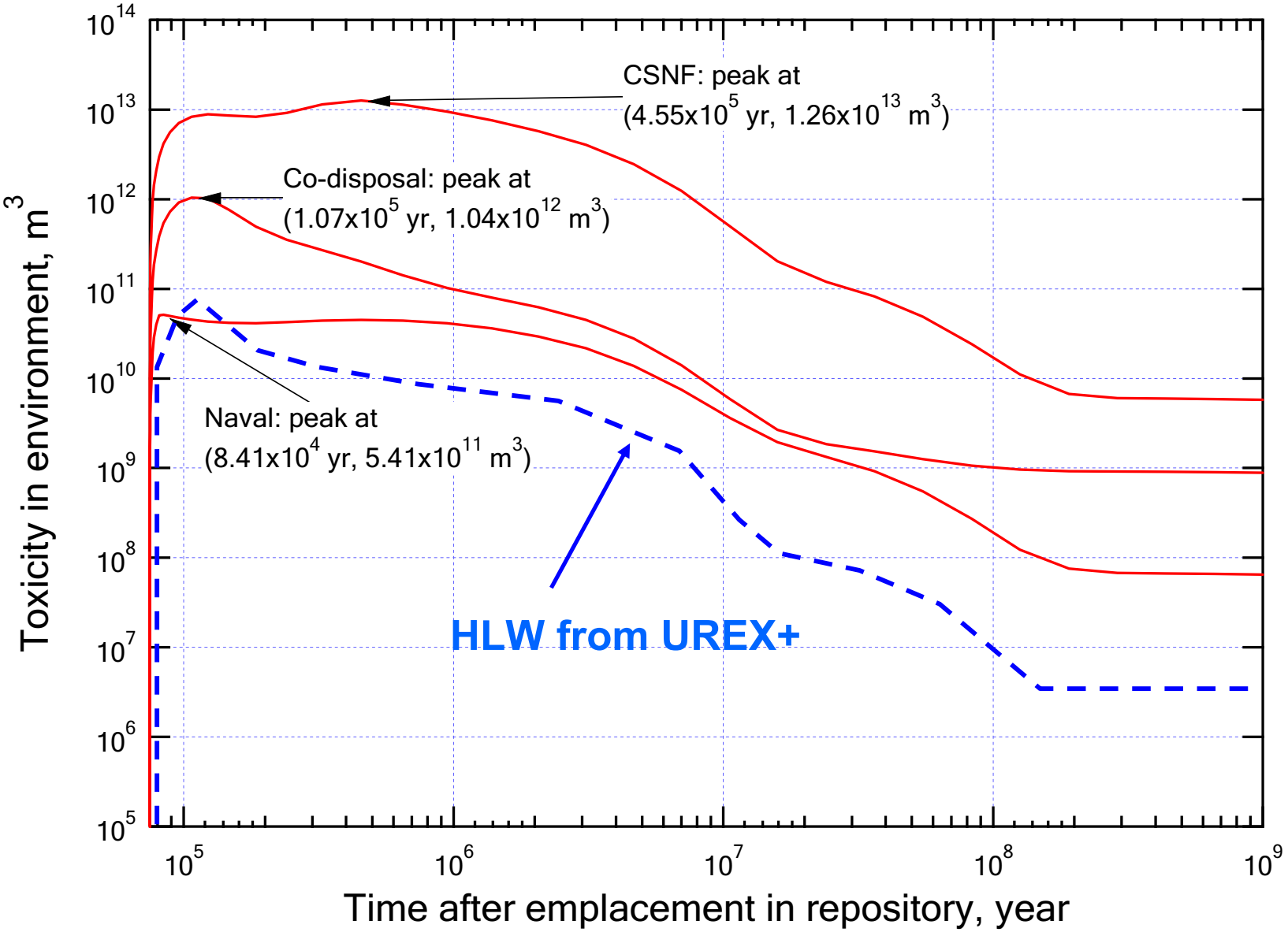




Vitrified HLW

CSNF

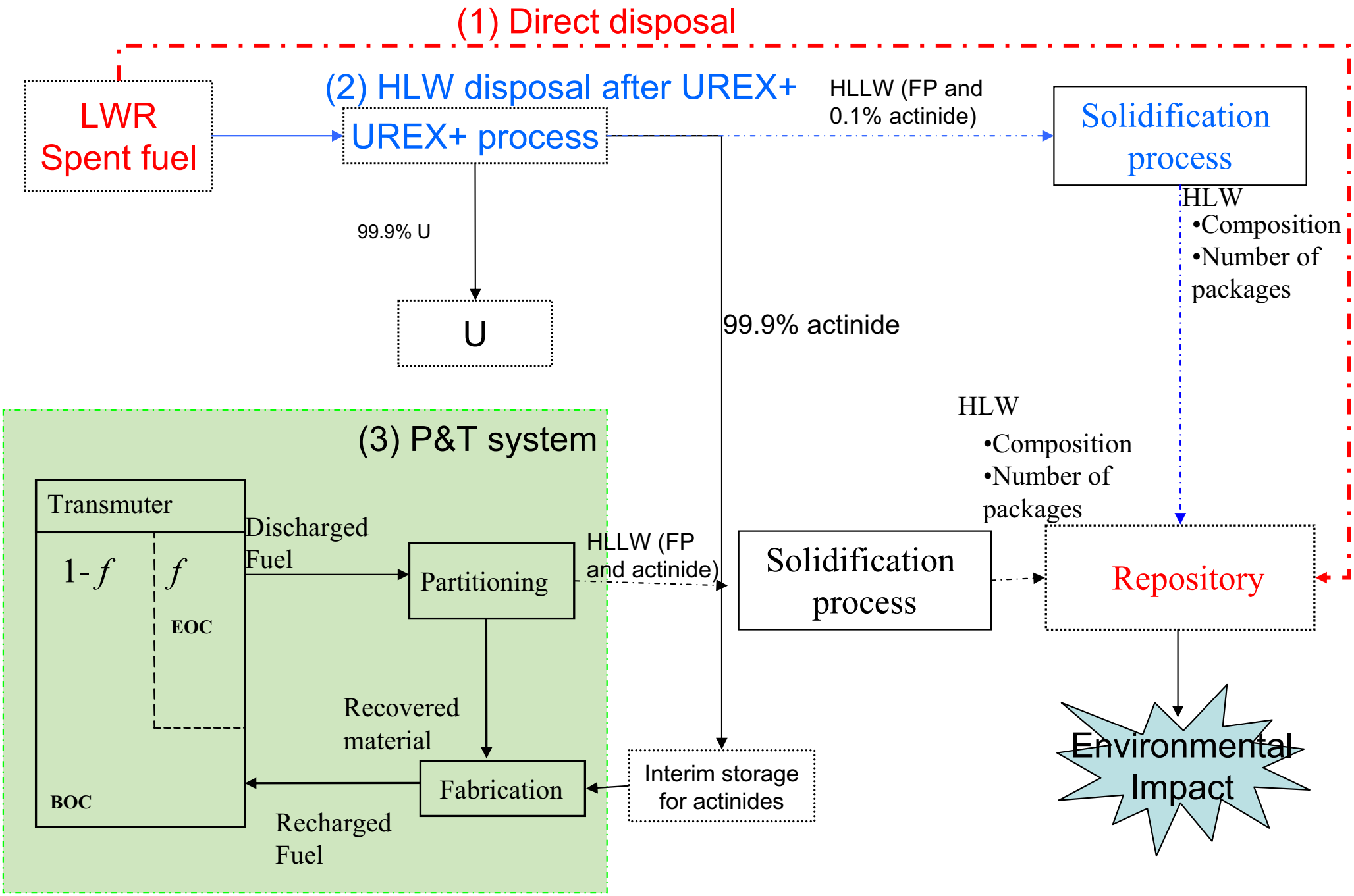
Comparison of HLW from UREX+ and CSNF



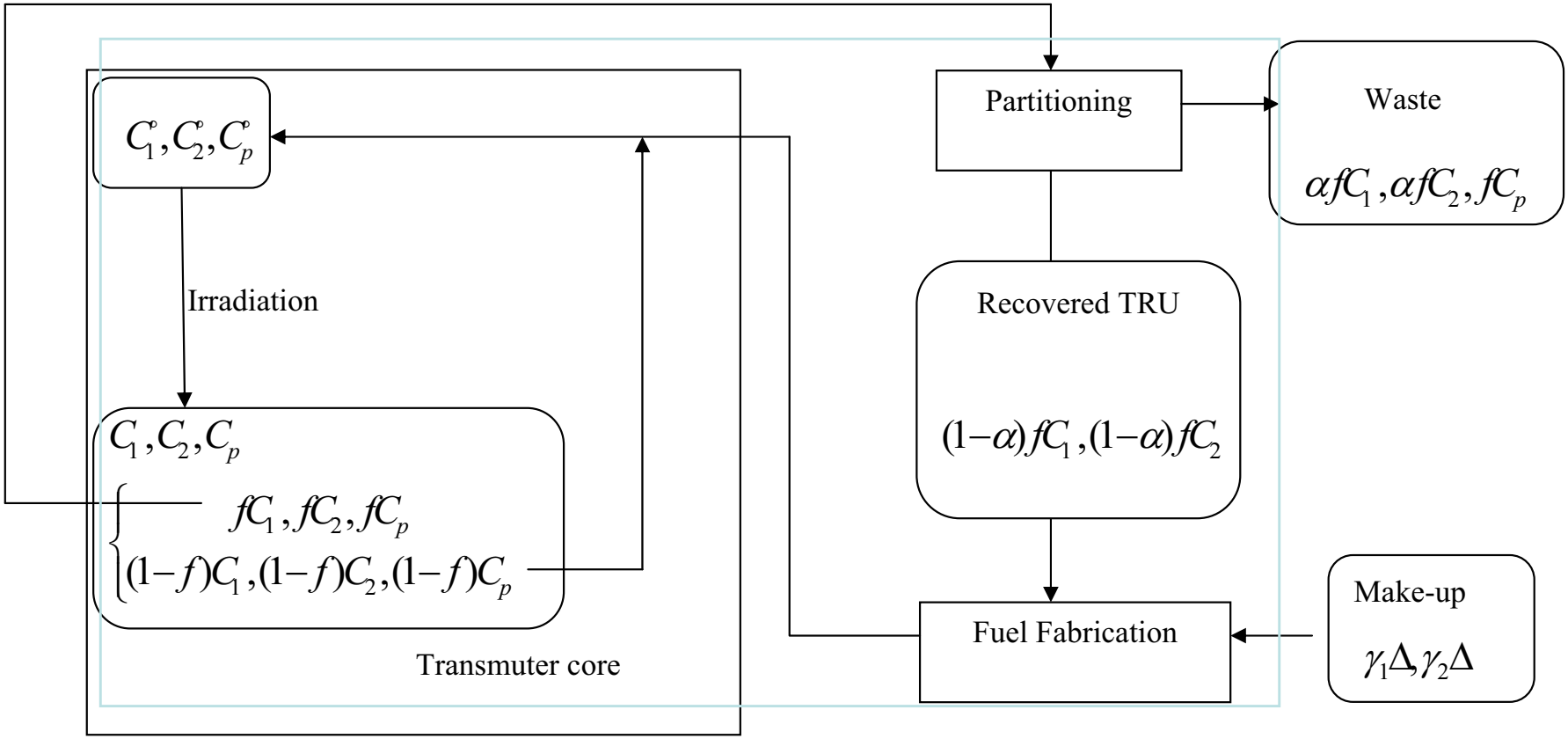
Observations for effects of UREX+ on Env. Impact Reduction

- For the number of waste packages,
  - Reduction from 7890 to 2320 (a factor of 3.4)
  - Because 63,000 MT of spent fuel is considered to generate approximately 3,000 GWy of electricity, the same repository can accommodate additionally 3,000 x 2.4 = 7,200 GWy.
  - The environmental impact from the repository fully loaded with HLW packages is significantly smaller than that from the repository with CSNF packages. (See below)
  - EI from HLW is smaller than WI from Defense Wastes.
- For impacts from TRU nuclides and their decay daughters,
  - Impacts are reduced by a factor of ~ 100 as a result of UREX+ application.
- For fission-product isotopes,
  - Impacts do not significantly decrease by UREX+.
  - I-129 is not included in the HLW; it would be included in intermediate or low-level wastes, but eventually become environmental impact.
- For natural uranium isotopes and their decay daughters,
  - Impacts are reduced by a factor of ~ 1000 as a result of UREX+ application.

(1) Commercial Spent Nuclear Fuel (CSNF) + Defense Wastes (DW) – Base case --
(2) Vitrified HLW from reprocessing for CSNF
(3) P&T Mass Flow


(1) Direct disposal
(2) HLW disposal after UREX+
LWR
Spent fuel
UREX+ process
HLLW (FP and
0.1% actinide)
Solidification
process
HLW
•Composition
•Number of
packages
99.9% U
U
99.9% actinide
(3) P&T system
Transmuter
1- f
f
EOC
BOC
Discharged
Fuel
Partitioning
HLLW (FP
and actinide)
Solidification
process
HLW
•Composition
•Number of
packages
Repository
Recovered
material
Fabrication
Interim storage
for actinides
Recharged
Fuel
Environmental
Impact

Mass-Flow Model



$$C_1(t) \equiv \frac{\hat{C}_1(\hat{t})}{\hat{\bar{C}}},\ C_2(t) \equiv \frac{\hat{C}_2(\hat{t})}{\hat{\bar{C}}},\ C_p(t) \equiv \frac{\hat{C}_p(\hat{t})}{\hat{\bar{C}}},$$

$$C_1^\circ \equiv \frac{\hat{C}_1^\circ}{\hat{\bar{C}}},\ C_2^\circ \equiv \frac{\hat{C}_2^\circ}{\hat{\bar{C}}},\ C_p^\circ \equiv \frac{\hat{C}_p^\circ}{\hat{\bar{C}}},$$

$$\hat{\bar{C}} \equiv \hat{C}_1^\circ + \hat{C}_2^\circ + \hat{C}_p^\circ.$$

$$t \equiv \frac{\hat{t}}{\hat{T}},\ d_1 \equiv \hat{d}_1\hat{T},\ d_2 \equiv \hat{d}_2\hat{T},\ p_2 \equiv \hat{p}_2\hat{T}$$

Two-member decay chain

$$M_1 \xrightarrow{d_1 - p_2} [\text{Transmutation product-1}]$$

$$M_1 \xrightarrow{p_2} M_2 \xrightarrow{d_2} [\text{Transmutation product-2}]$$

$$\frac{dC_1(t)}{dt} = -d_1 C_1(t),$$

$$\frac{dC_2(t)}{dt} = -d_2 C_2(t) + p_2 C_1(t),$$

$$\frac{dC_p(t)}{dt} = \left(d_1 - p_2\right) C_1(t) + d_2 C_2(t)$$

$$C_1(0) = C_1^\circ,\ C_2(0) = C_2^\circ,\ C_p(0) = C_p^\circ$$

Concentrations after Transmutation

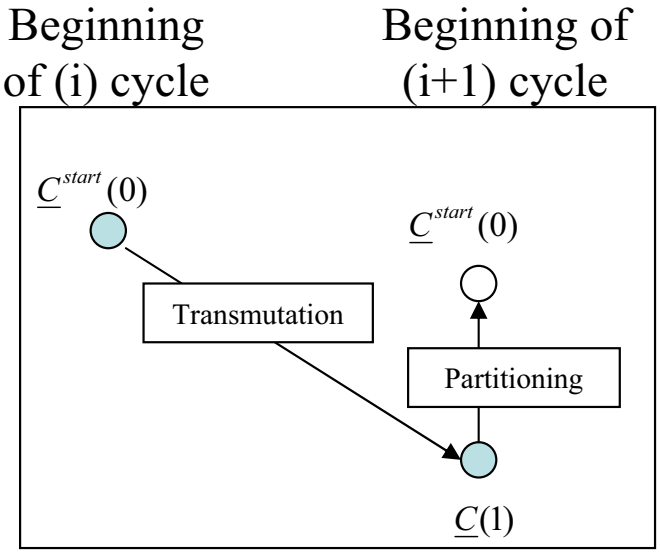
$$\underline{C}(1) = \underline{\underline{A}}\,\underline{C}^{start}(0)$$

where

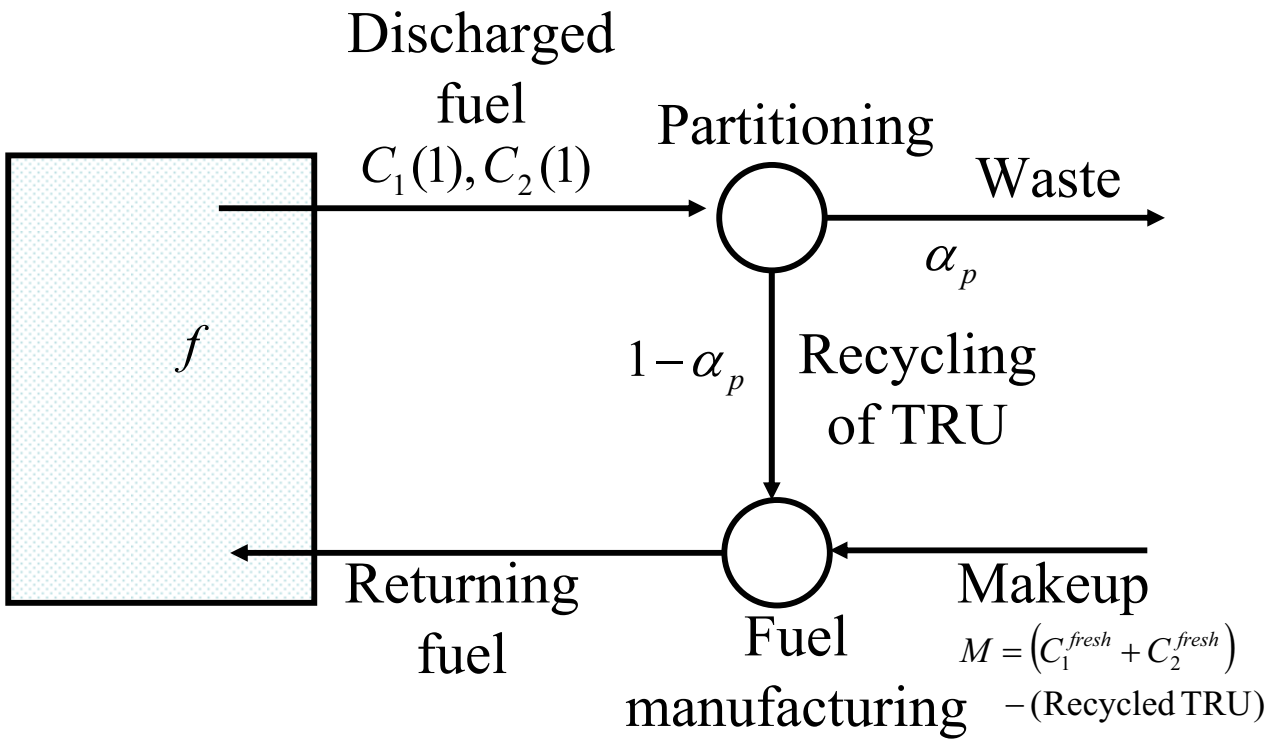
$$\underline{C}(1) \equiv \begin{bmatrix} C_1(1) \\ C_2(1) \end{bmatrix}, \quad \underline{C}^{start}(0) \equiv \begin{bmatrix} C_1^{start}(0) \\ C_2^{start}(0) \end{bmatrix}$$

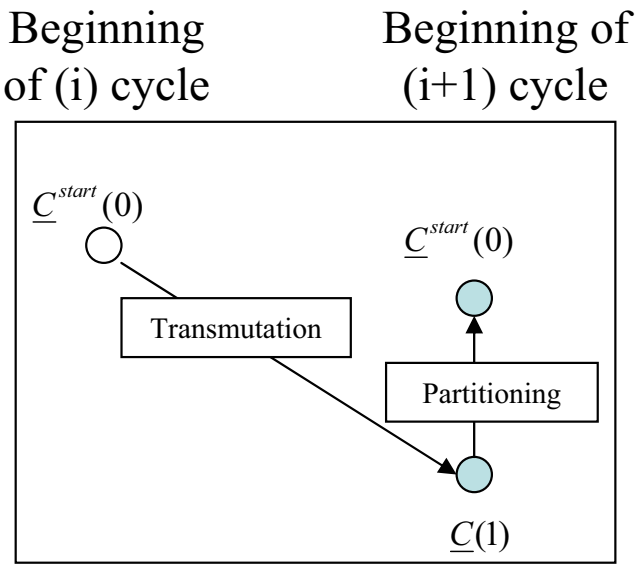
$\underline{\underline{A}} \equiv$ Transmutation Matrix

$$= \begin{bmatrix} e^{-d_1} & 0 \\ \dfrac{p_2}{d_2 - d_1}\left(e^{-d_1} - e^{-d_2}\right) & e^{-d_2} \end{bmatrix}$$



Concentrations after Partitioning





$$\underline{C}^{start}(0) = \underline{\underline{B}}\,\underline{C}(1) + f\,\underline{C}^{fresh}$$

where

$$\underline{\underline{B}} \equiv \text{Partitioning Matrix} = \begin{bmatrix} 1-\alpha_p f-(1-\alpha_p)\gamma_1 f & -(1-\alpha_p)\gamma_1 f \\ -(1-\alpha_p)\gamma_2 f & 1-\alpha_p f-(1-\alpha_p)\gamma_2 f \end{bmatrix}$$

Recursive Form of Concentrations

$$\underline{C}^{(i)}(1) = \underline{\underline{A}}\,\underline{C}^{(i-1)}(0) \qquad \text{after Transmutation}$$

$$\underline{C}^{(i)}(0) = \underline{\underline{B}}\,\underline{C}^{(i)}(1) + f\,\underline{C}^{fresh} \qquad \text{after Partitioning}$$

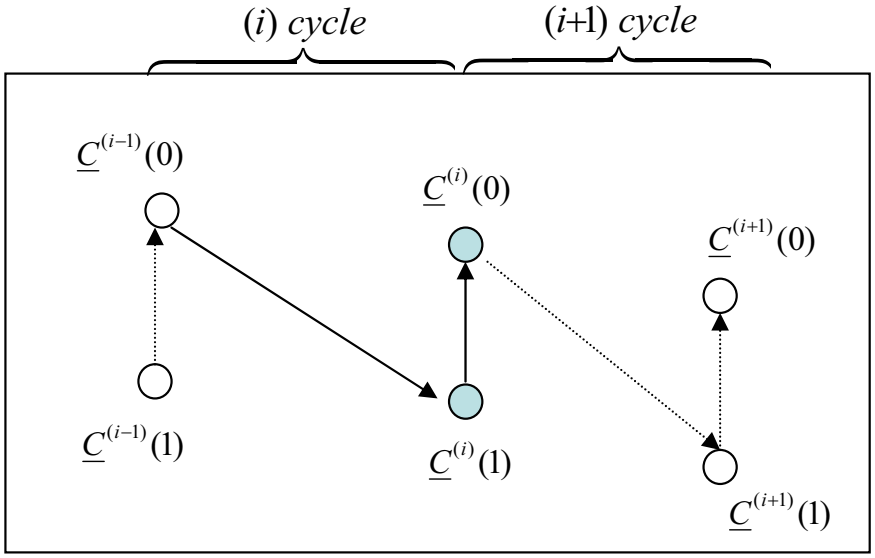
$i = 1,2,3,\ldots$ (Cycle Number)

where

$\underline{C}^{(0)}(0) = \underline{C}^{fresh}$; Conc. at TRU Inventory

$\underline{\underline{A}}$ = Transmutation Matrix

$\underline{\underline{B}}$ = Partitioning Matrix



Concentrations at the i-th Cycle

at the 1st cycle

$\underline{C}^{(1)}(T) = \underline{\underline{A}}\,\underline{C}^{fresh}$ after Transmutation

$\underline{C}^{(1)}(0) = \underline{\underline{X}}\left(\underline{\underline{D}} + f\,\underline{\underline{I}}\right)\underline{\underline{X}}^{-1}\underline{C}^{fresh}$ after Partitioning

at the 2nd cycle and onward

$\underline{C}^{(k+2)}(1) = \underline{\underline{A}}\,\underline{\underline{X}}\left(\underline{\underline{D}}^{k+1} + f\sum_{j=0}^{k}\underline{\underline{D}}^{j}\right)\underline{\underline{X}}^{-1}\underline{C}^{fresh}$ after Transmutation

$\underline{C}^{(k+2)}(0) = \underline{\underline{X}}\left(\underline{\underline{D}}^{k+2} + f\sum_{j=0}^{k+1}\underline{\underline{D}}^{j}\right)\underline{\underline{X}}^{-1}\underline{C}^{fresh}$ after Partitioning

$k = 0,1,2,...$

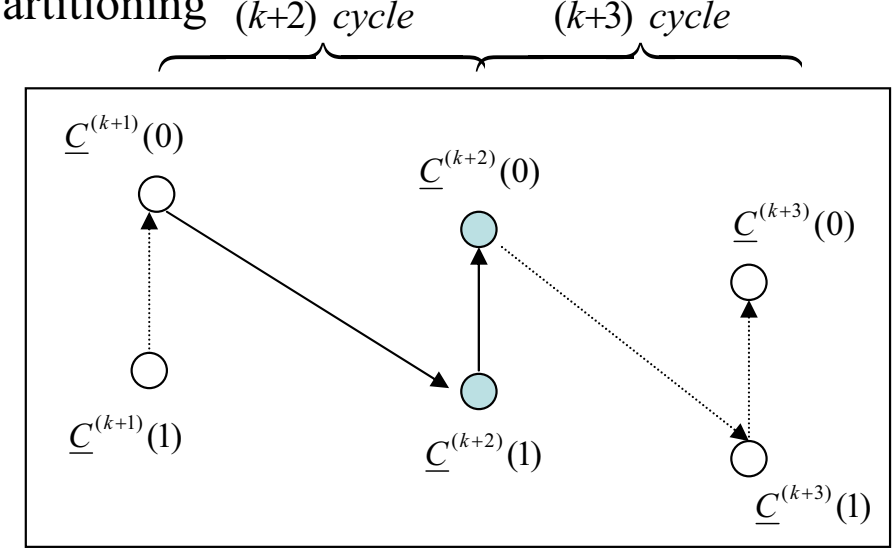
where

$\underline{\underline{D}}^{0} = \underline{\underline{I}},\ \underline{\underline{X}}\,\underline{\underline{D}}\,\underline{\underline{X}}^{-1} = \underline{\underline{R}}$

$\underline{\underline{D}} = \text{Eigenvalue Matrix of } \underline{\underline{R}}$

$\underline{\underline{X}} = \text{Eigenvector Matrix of } \underline{\underline{R}}$

$\underline{\underline{R}} = \underline{\underline{B}}\,\underline{\underline{A}}$ System Matrix



System matrix

$$\underline{\underline{R}} \equiv \underline{\underline{B}}\,\underline{\underline{A}} = \begin{bmatrix} r_{11} & r_{12} \\ r_{21} & r_{22} \end{bmatrix}$$

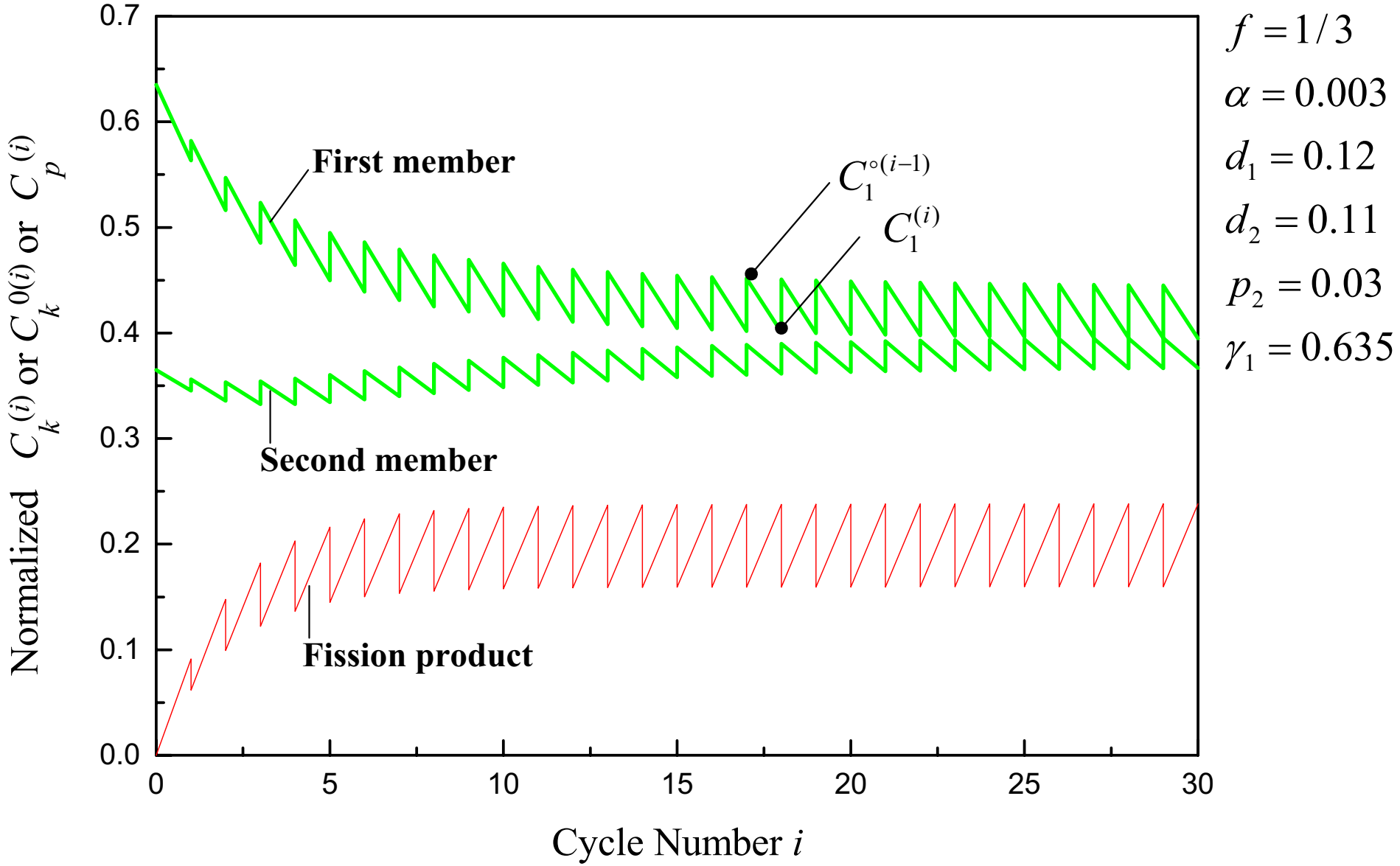
$$\underline{\underline{A}} \equiv \begin{bmatrix} a_{11} & 0 \\ a_{21} & a_{22} \end{bmatrix},$$

$$\underline{\underline{B}} \equiv \begin{bmatrix} 1-\alpha f-(1-\alpha)\gamma_1 f & -(1-\alpha)\gamma_1 f \\ -(1-\alpha)\gamma_2 f & 1-\alpha f-(1-\alpha)\gamma_2 f \end{bmatrix}$$

$$r_{11} = \left[1-\alpha f-(1-\alpha)\gamma_1 f\right]e^{-d_1} - (1-\alpha)\gamma_1 f a_{21},$$
$$r_{12} = -(1-\alpha)\gamma_1 f e^{-d_2},$$
$$r_{21} = -(1-\alpha)\gamma_2 f e^{-d_1} + \left[1-\alpha f-(1-\alpha)\gamma_2 f\right]a_{21},$$
$$r_{22} = \left[1-\alpha f-(1-\alpha)\gamma_2 f\right]e^{-d_2},$$

$$a_{11} = e^{-d_1},$$
$$a_{21} = \begin{cases} \dfrac{p_2}{d_2-d_1}\left(e^{-d_1}-e^{-d_2}\right), \text{for } d_1 \neq d_2, \\ p_2 e^{-d_2}, \text{ for } d_1 = d_2, \end{cases}$$
$$a_{22} = e^{-d_2}.$$

Concentration Profile of TRUs



TRU Reduction Ratio

- TRU Reduction Ratio

$$\delta_k^{(i)} \equiv \frac{L_k^{(i)}}{S_k^{(i)}}, \quad k = 1,2, \text{ and } i = 1,2,3,...$$

  - As *i* increases, the ratio approaches a constant value, independent of *i*.
  - Small TRU reduction ratio mean less waste.

- Cumulative TRU in waste

$$L_k^{(i)} \equiv f\alpha \sum_{m=1}^{i} C_k^{(m)} \qquad k = 1,2 \text{ and } i = 1,2,3,...$$

- Cumulative TRU feed

$$S_k^{(i)} \equiv \gamma_k \sum_{m=1}^{i} \left[f - (1-\alpha) f \left(C_1^{(m)} + C_2^{(m)} \right) \right]$$

$$k = 1,2 \text{ and } i = 1,2,3,...$$

  - Both waste and feed increase with *i*
  - For a sufficiently large value of *i*, both increase linearly with *i*.

TRU Reduction Ratio at Steady State

- For the first member:

$$\delta_1^{(\infty)} = \frac{\alpha f e^{-d_1}}{1-(1-\alpha f)e^{-d_1}}$$

- For the second member:

$$\delta_2^{(\infty)} = \frac{\alpha f\left\{\frac{\gamma_1}{\gamma_1} a_{21} + e^{-d_2}\left[1-(1-\alpha f)e^{-d_1}\right]\right\}}{\left[1-(1-\alpha f)e^{-d_1}\right]\left[1-(1-\alpha f)e^{-d_2}\right]}$$

where

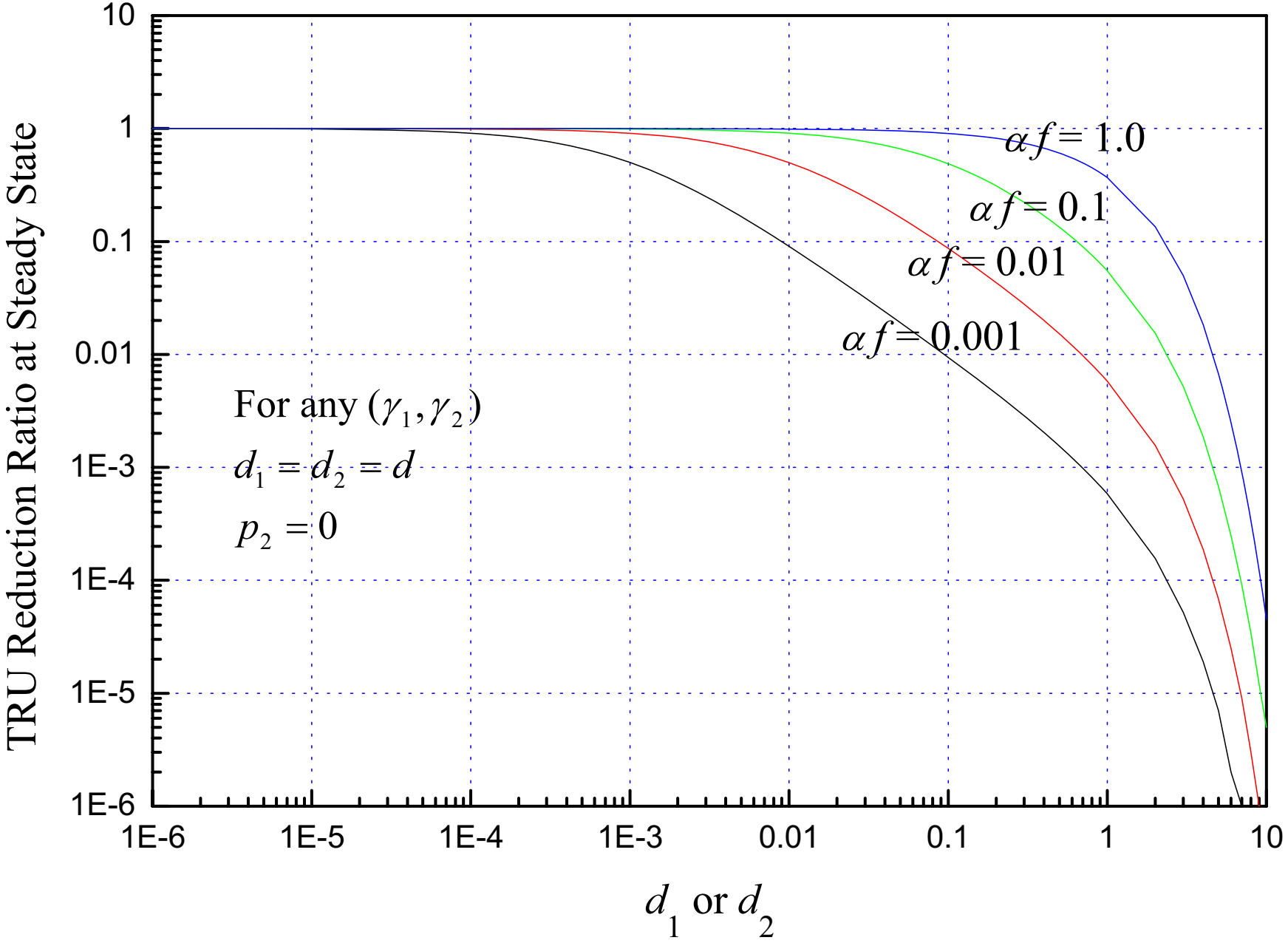
$$a_{21} = \begin{cases} \dfrac{p_2}{d_2-d_1}\left(e^{-d_1}-e^{-d_2}\right), & d_1 \neq d_2 \\ p_2 e^{-d}, & d_1 = d_2 = d \end{cases}$$

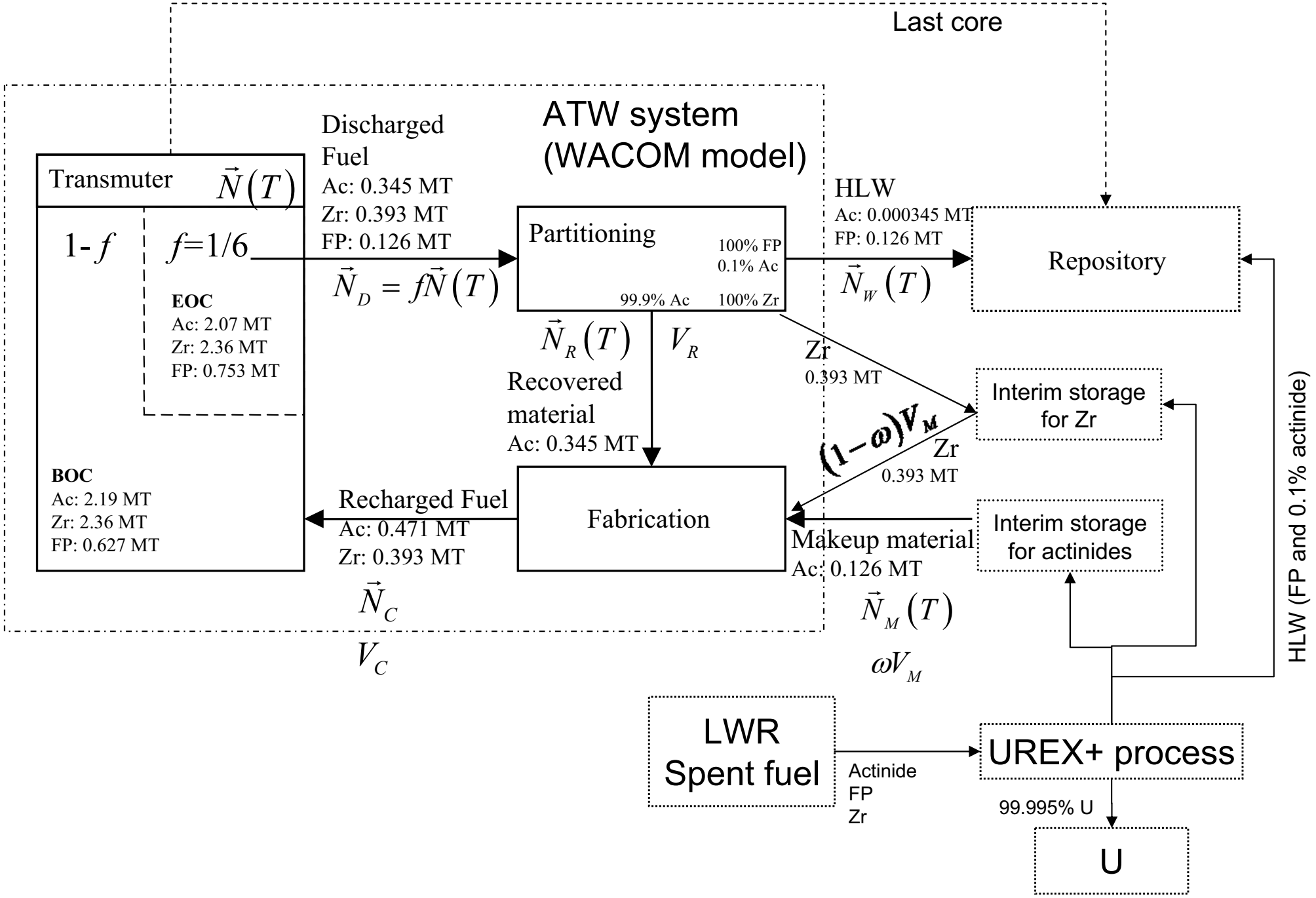
$d_1, d_2, p_2, (\alpha f)$, and γ_1 (or $\gamma_2 = 1-\gamma_1$)

  - There are five system parameters:
  - If $d_1 = d_2 = d$ and $p_2 = 0$, both expression becomes identical (only two independent parameters).

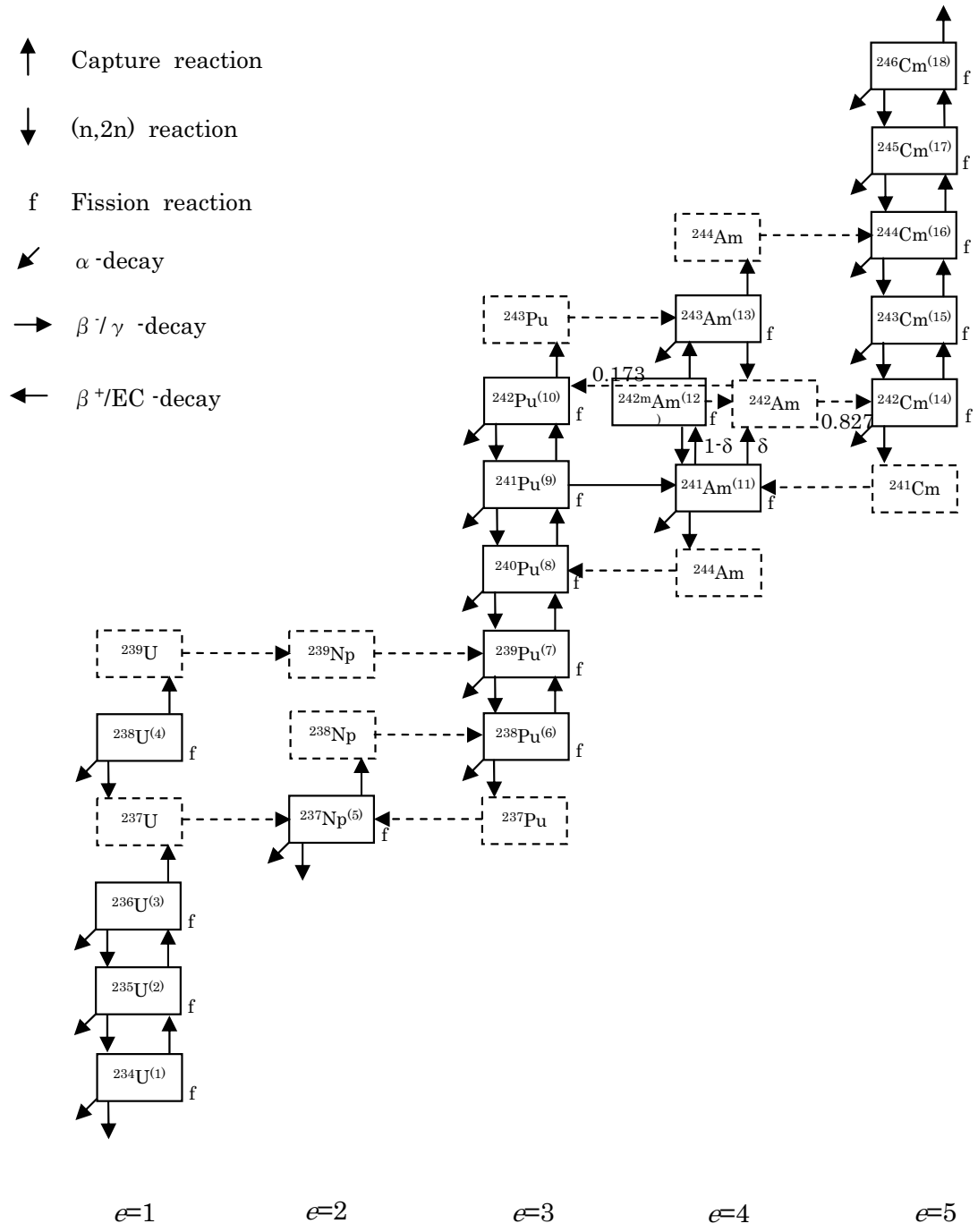
$$\rho_{1,2}^{(\infty)} = \frac{\alpha f e^{-d}}{1-(1-\alpha f)e^{-d}}$$

Effects of d and (αf)

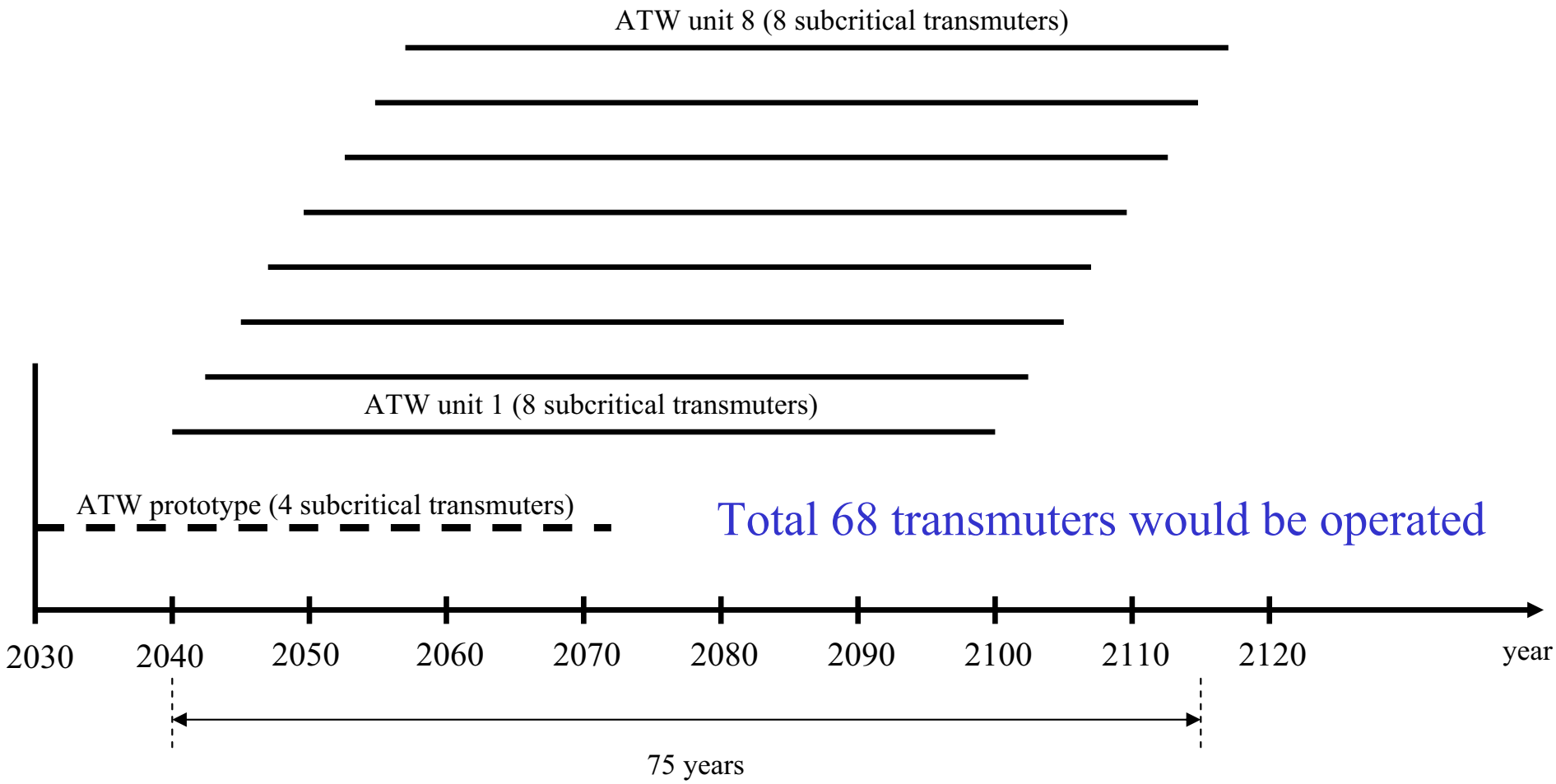



Last core
ATW system
(WACOM model)
Transmuter $\vec{N}(T)$
1- f
f=1/6
EOC
Ac: 2.07 MT
Zr: 2.36 MT
FP: 0.753 MT
BOC
Ac: 2.19 MT
Zr: 2.36 MT
FP: 0.627 MT
Discharged
Fuel
Ac: 0.345 MT
Zr: 0.393 MT
FP: 0.126 MT
$\vec{N}_D = f\vec{N}(T)$
Partitioning
100% FP
0.1% Ac
99.9% Ac
100% Zr
$\vec{N}_R(T)$
V_R
Recovered
material
Ac: 0.345 MT
HLW
Ac: 0.000345 MT
FP: 0.126 MT
$\vec{N}_W(T)$
Repository
Zr
0.393 MT
Interim storage
for Zr
$(1-\omega)V_M$
Zr
0.393 MT
Recharged Fuel
Ac: 0.471 MT
Zr: 0.393 MT
Fabrication
Interim storage
for actinides
Makeup material
Ac: 0.126 MT
$\vec{N}_C$
V_C
$\vec{N}_M(T)$
ωV_M
HLW (FP and 0.1% actinide)
LWR
Spent fuel
Actinide
FP
Zr
UREX+ process
99.995% U
U

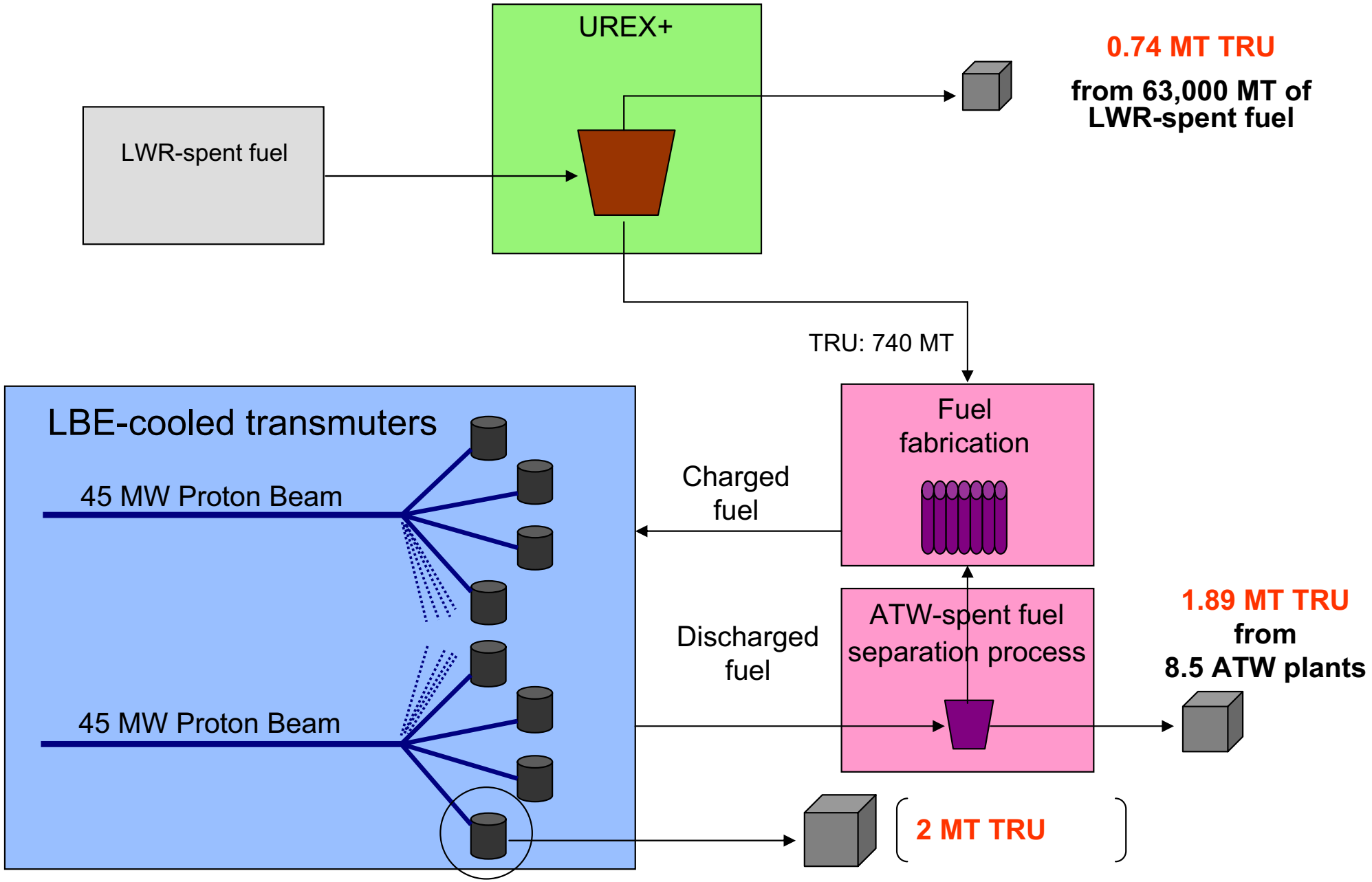
18-member decay chain considered



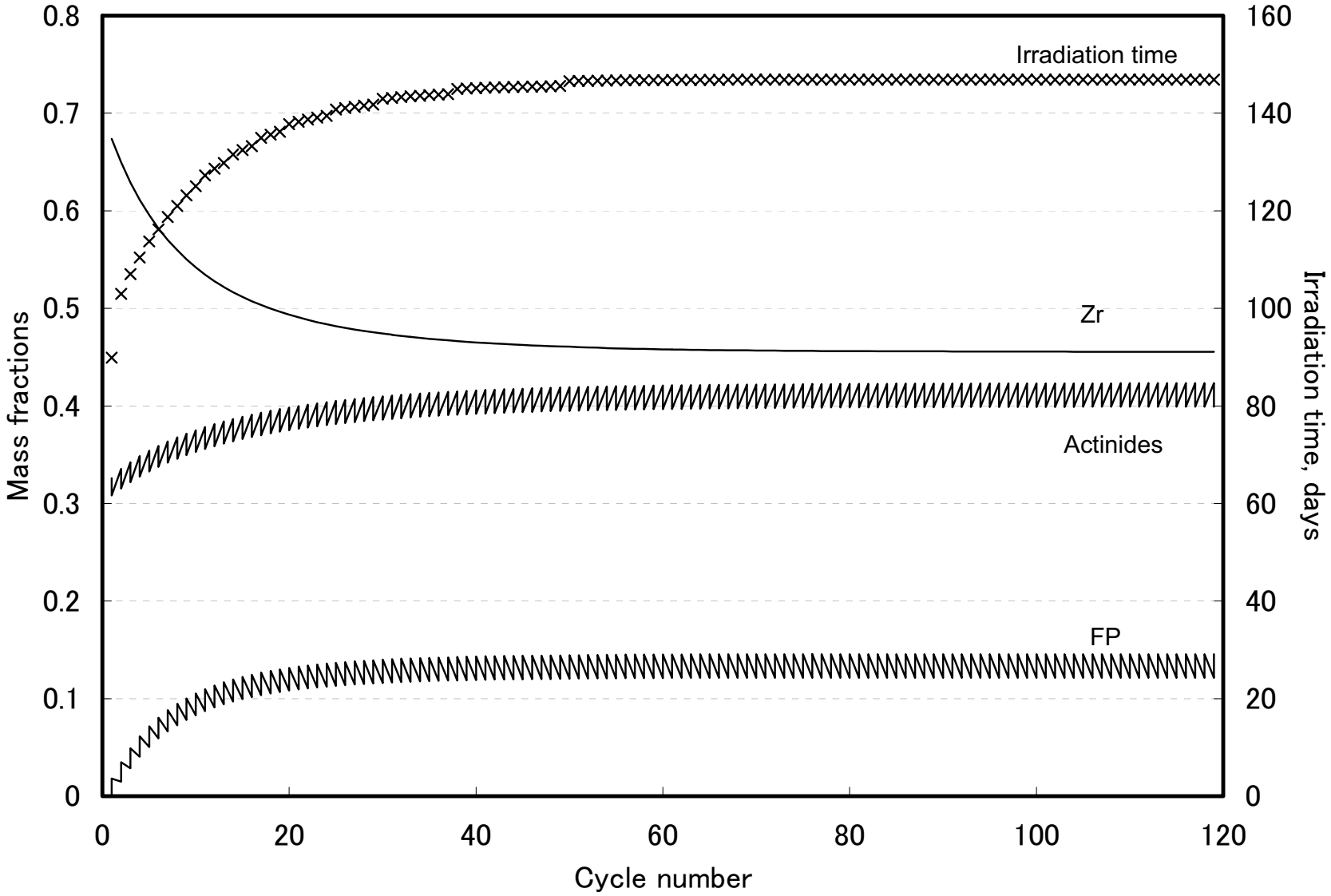
Deployment Scenario of Reference ATW Plants



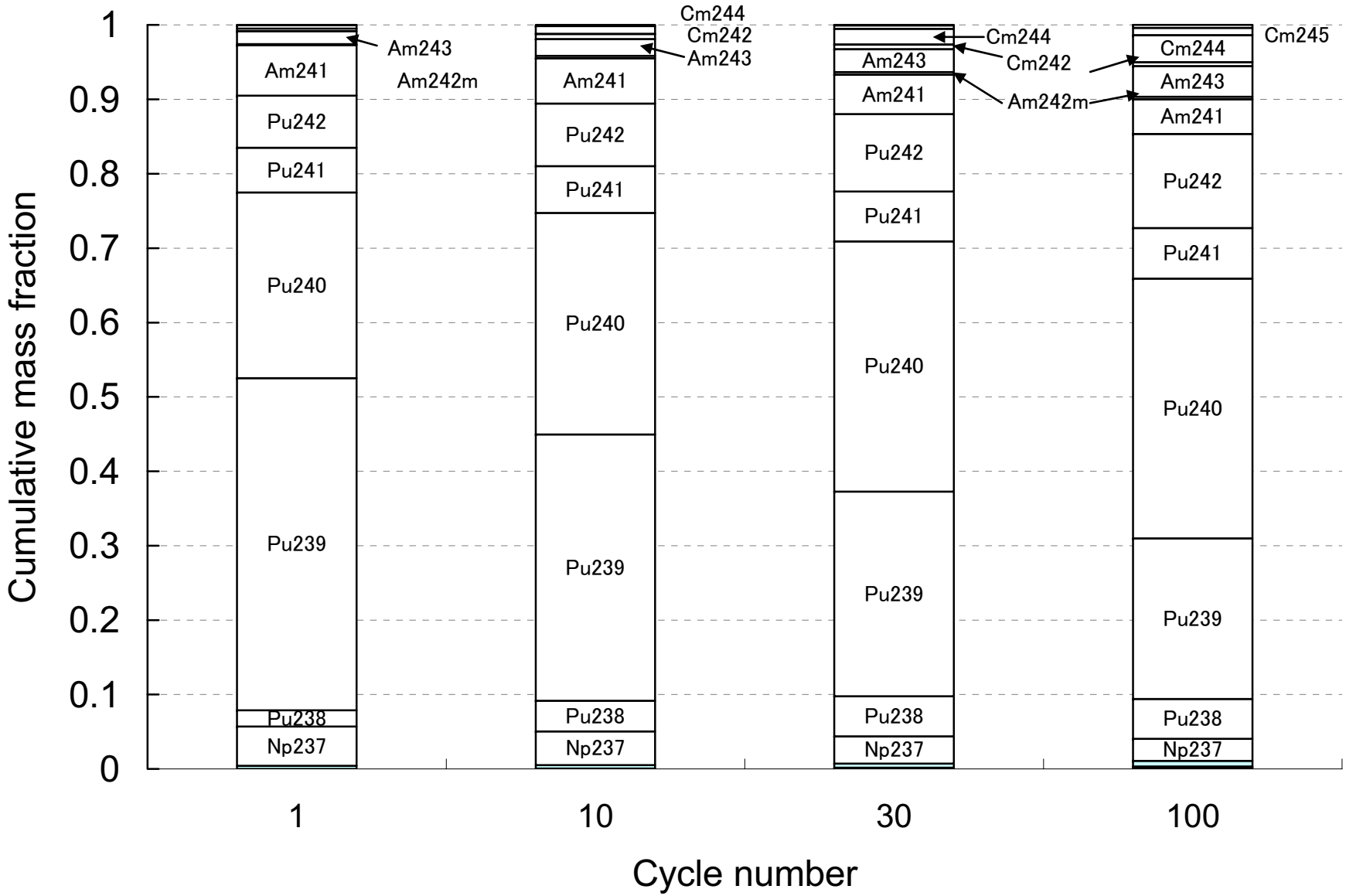
TRU Waste from the Reference ATW Fuel Cycle



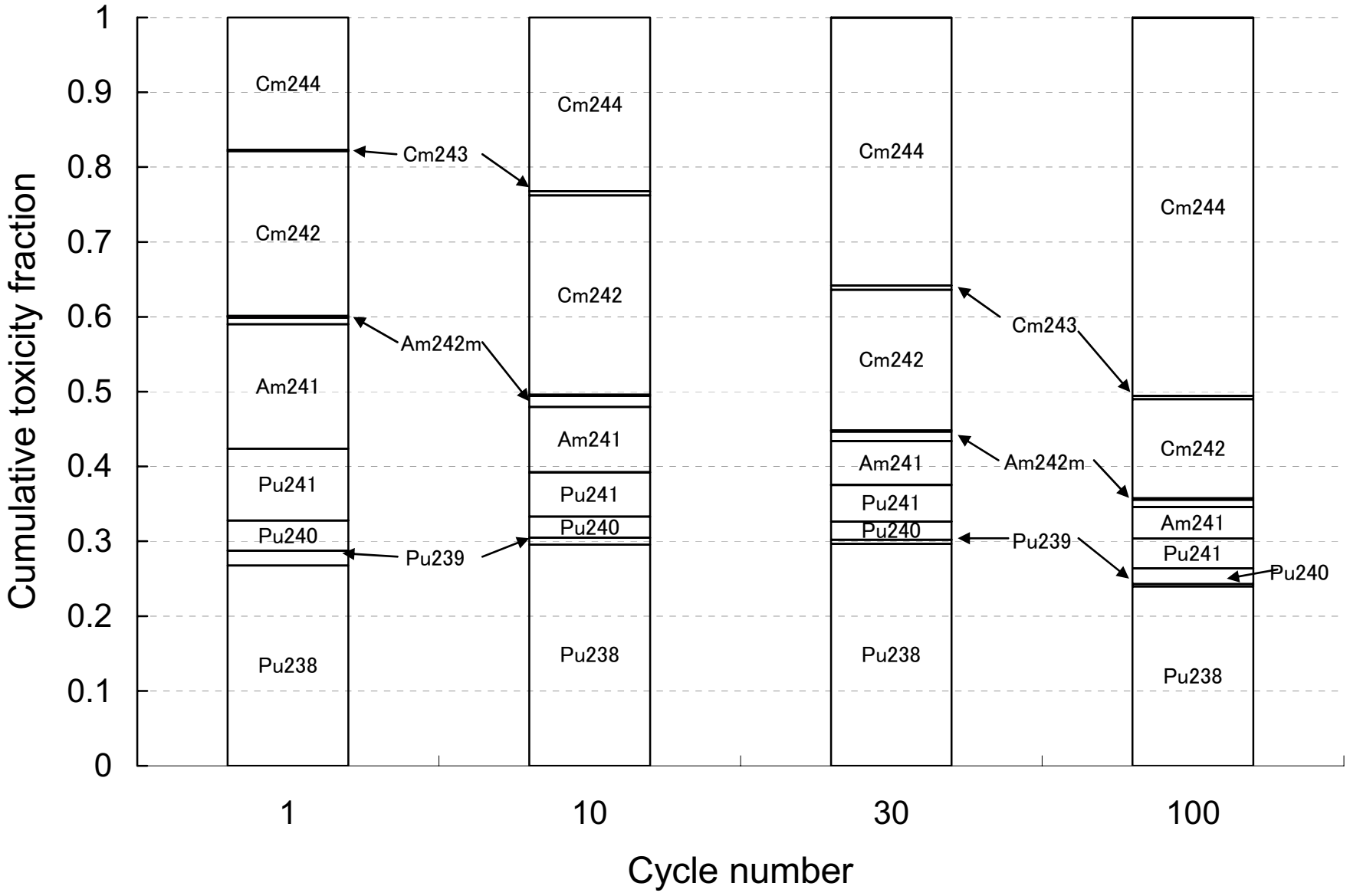
Change in mass fractions for multiple cycles



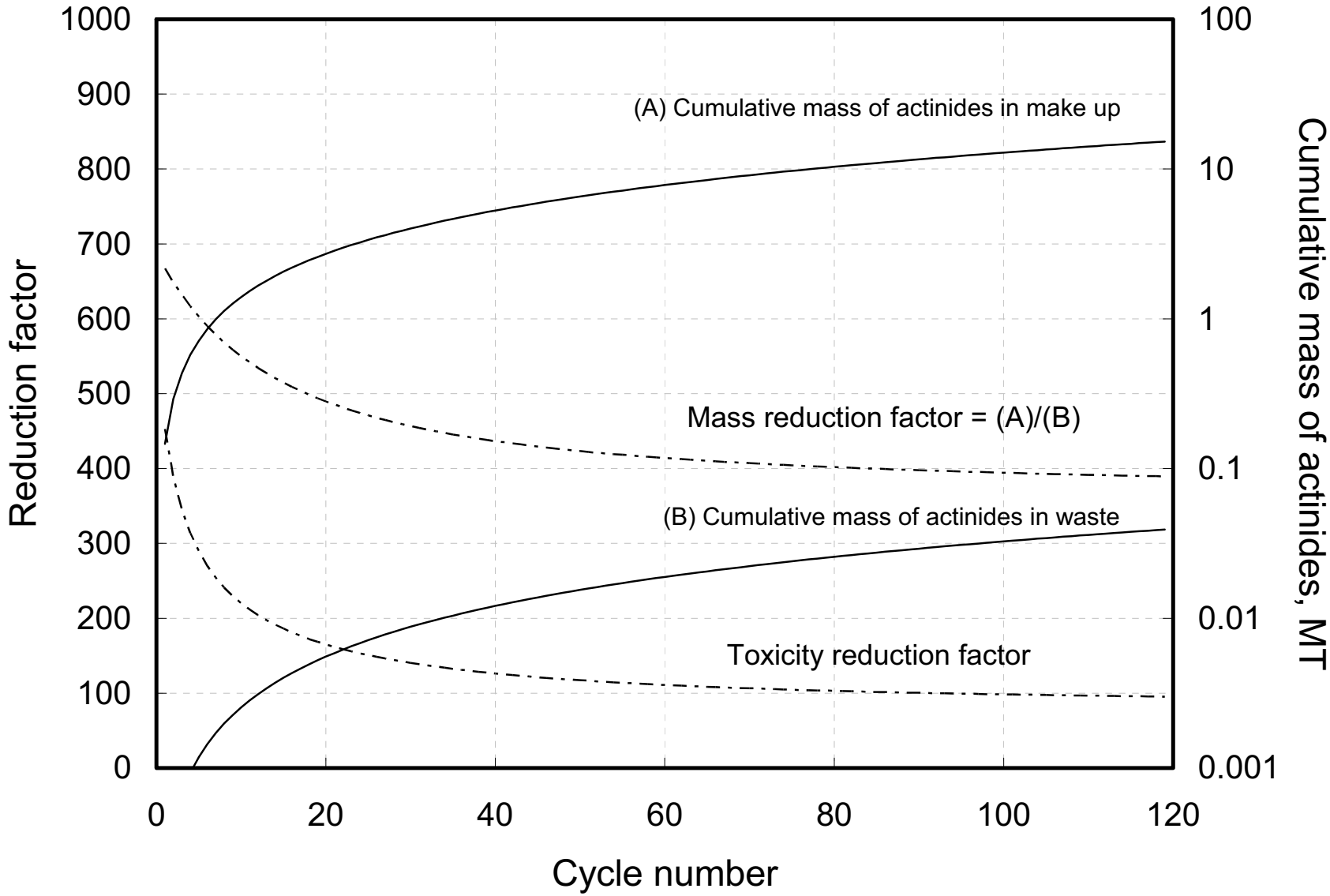
Evolution of mass fractions in HLW from P&T



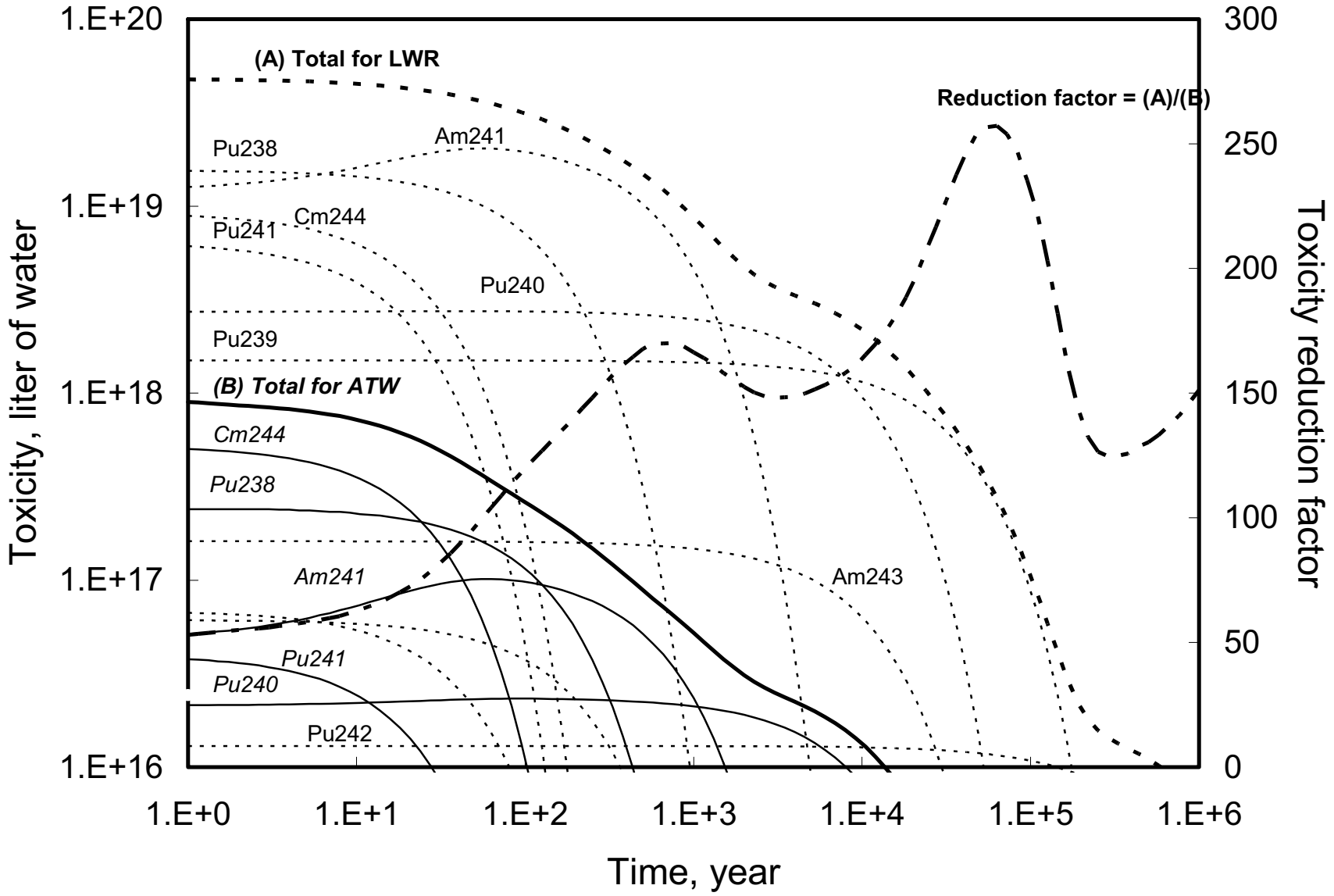
Evolution of cumulative toxicity of HLW from P&T

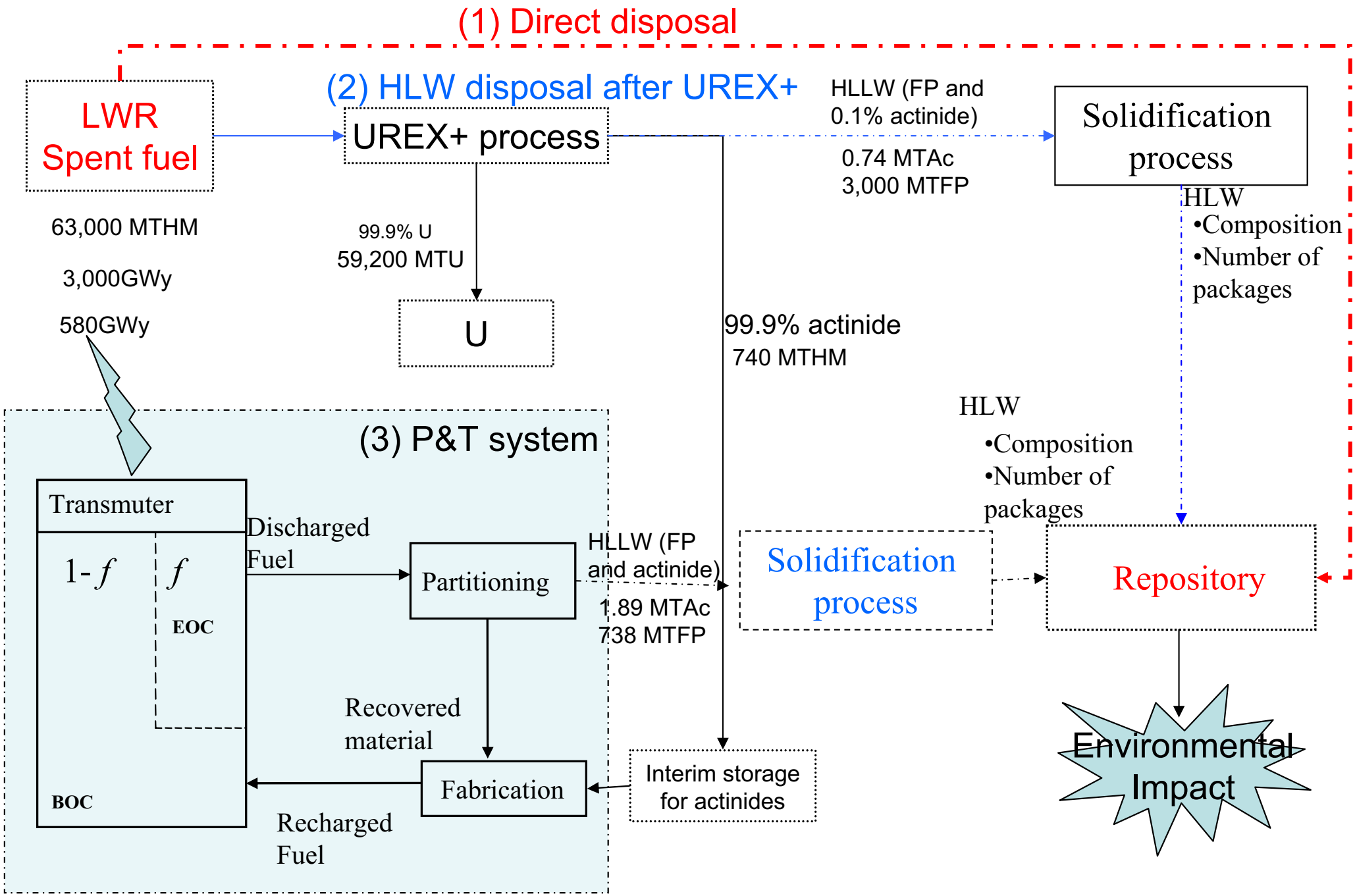


Reduction factor for TRU



Radiotoxicity of HLW from P&T




(1) Direct disposal
(2) HLW disposal after UREX+
LWR
Spent fuel
63,000 MTHM
3,000GWy
580GWy
UREX+ process
99.9% U
59,200 MTU
U
HLLW (FP and
0.1% actinide)
0.74 MTAc
3,000 MTFP
Solidification
process
HLW
•Composition
•Number of
packages
99.9% actinide
740 MTHM
(3) P&T system
Transmuter
1-f
f
EOC
BOC
Discharged
Fuel
Partitioning
HLLW (FP
and actinide)
1.89 MTAc
738 MTFP
Recovered
material
Fabrication
Recharged
Fuel
Interim storage
for actinides
HLW
•Composition
•Number of
packages
Solidification
process
Repository
Environmental
Impact

So, what is the environmental impact of HLW from P&T?

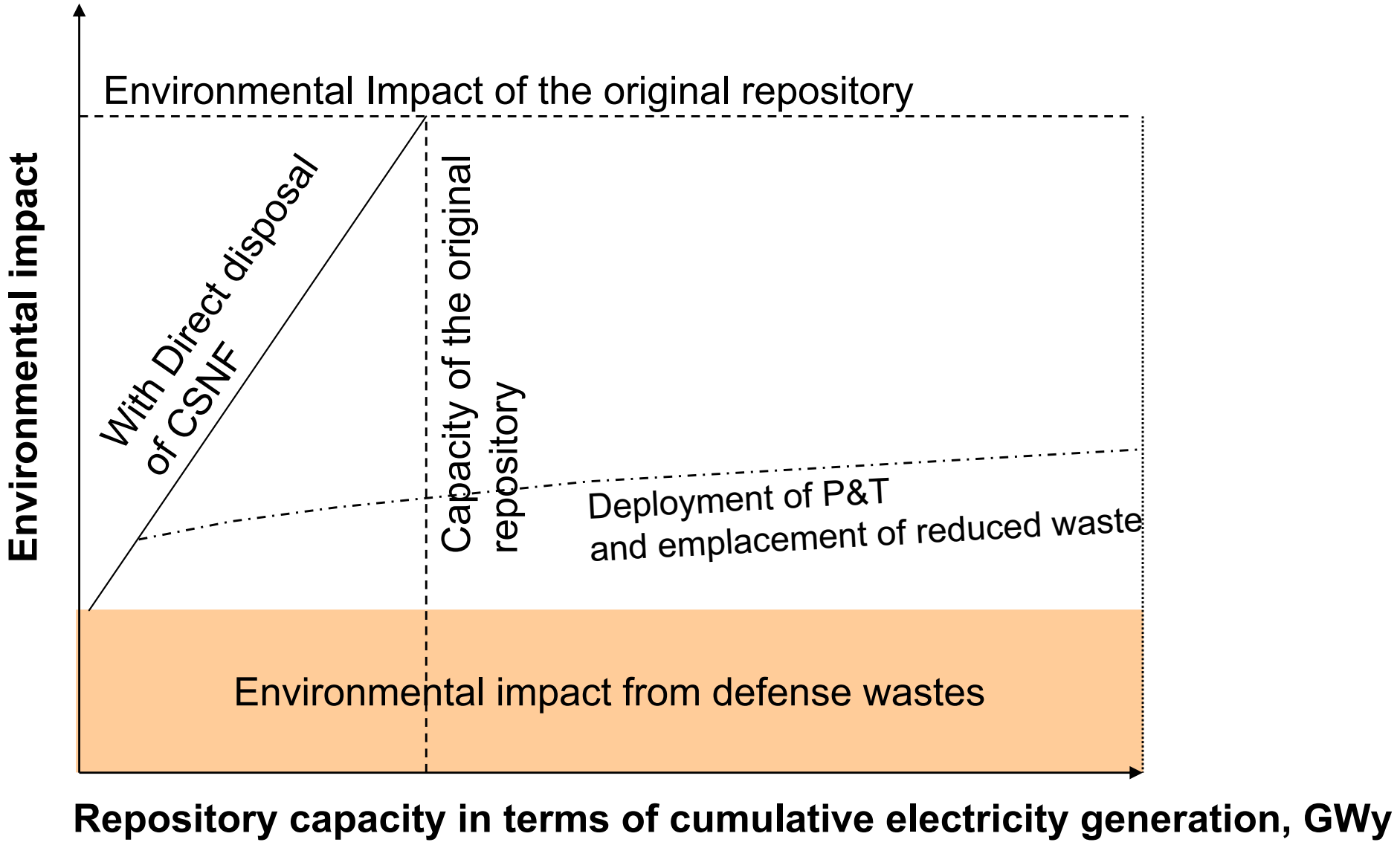
- Need to know:
  - Detailed composition of HLLW from Partitioning including FP composition
  - Solidification method (matrix material, etc)
- But, from the results obtained above, it can be said that:
  - Because short-lived Cm isotopes dominant in the HLLW decay in the waste package before it fails, environmental impact will be significantly smaller than that from HLW of UREX+.
  - Thus, the total environmental impact with UREX+ and P&T application will be approximately those for the UREX+ case.

Future Direction

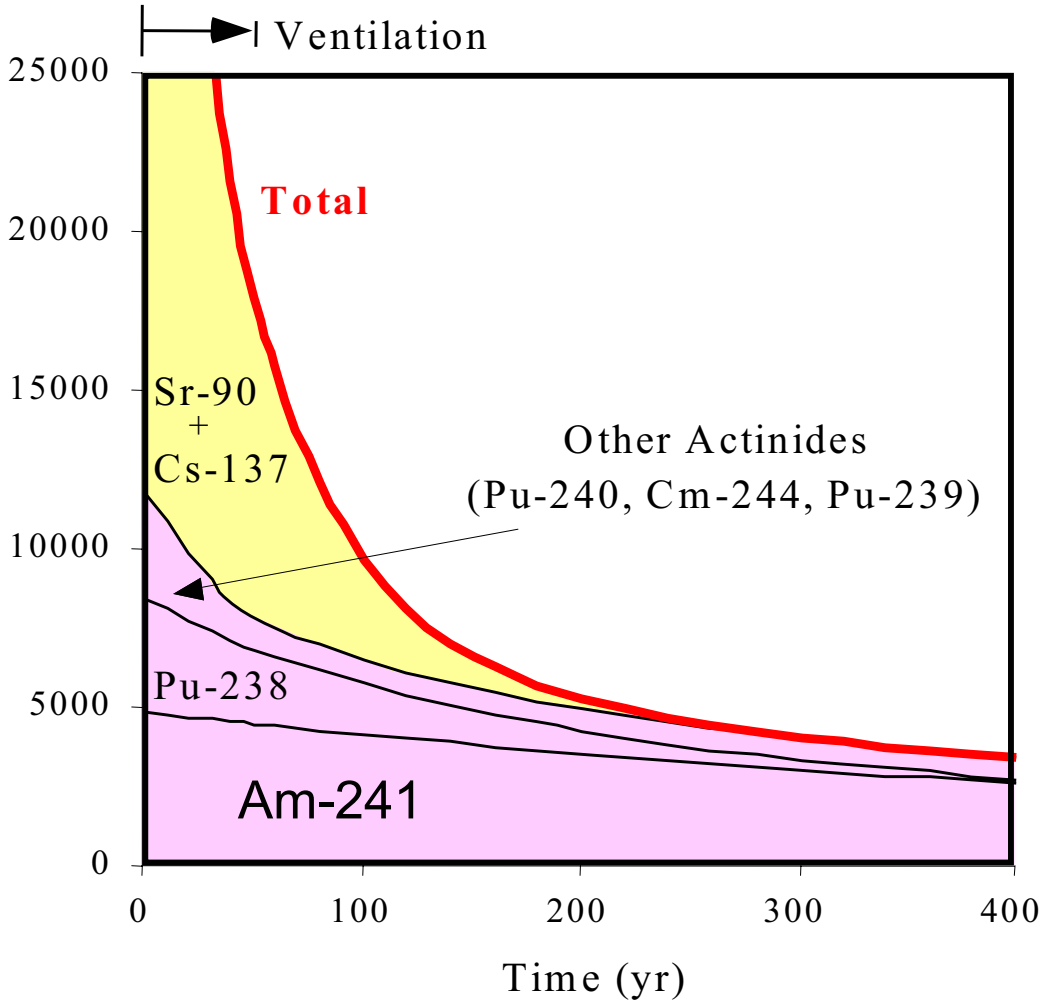
Benefit and challenges of P&T applications

- Toxic TRU isotopes are transmuted, resulting in approximately a factor of 100 smaller environmental impact from the repository.
- Mix of TRU after P&T in HLW consists of short-lived Cm isotopes, with much reduced masses of Np, Pu, and Am.
  - Lower heat emission
  - Lower radiotoxicity
  - Lower proliferation risk
- Additional electricity is gained from transmutation.
- Repository capacity can be expanded within the impact level set by the original repository. (see next slide)
- Cladding hulls and other solid wastes
- Destination of separated uranium still needs to be determined.
- Fuel manufacturing with high Cm concentration is difficult due to heat emission and radiation from Cm isotopes.
- P&T system is large and requires long-term complex operation.

Benefit of P&T on repository-capacity expansion (YMR)



Heat emission from CSNF



LWR Spent Fuel Heat Generation

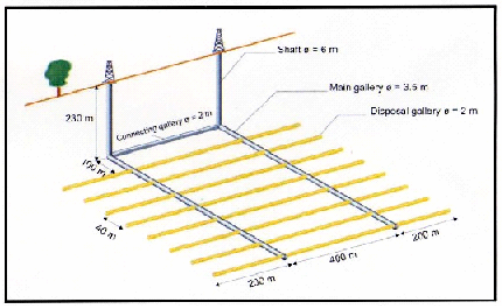
Reducing heat from waste packages results in …

- reducing ventilation in pre-closure period at YMR
- reducing repository footprint per Waste package
- improving stability of bentonite in Water-saturated repository

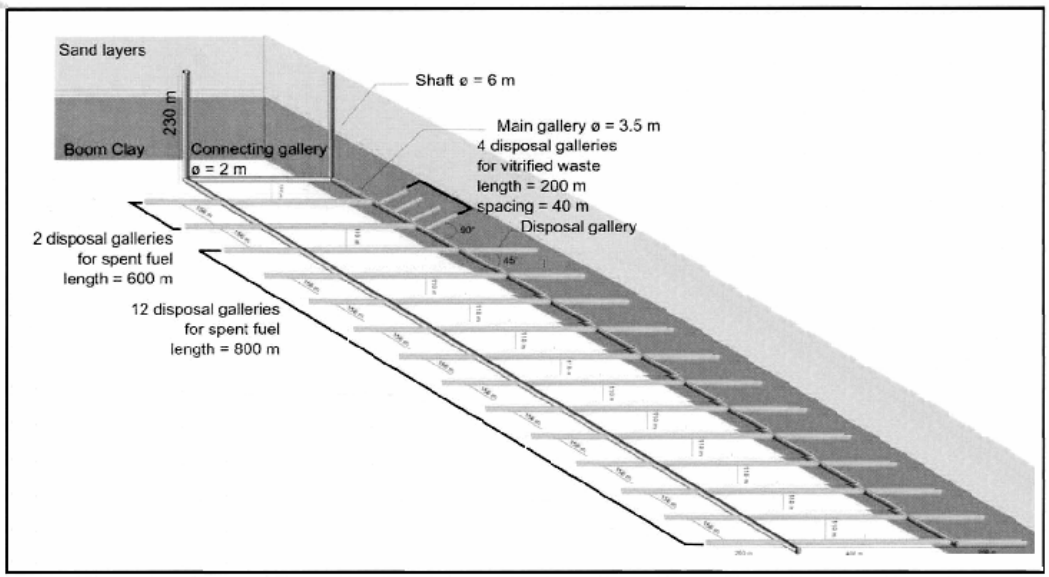
Direct Disposal vs. Vitrified HLW Disposal - Belgian Case -

- SAFIR 2 Report (December 2001)
- Direct disposal requires 6 times larger space than vitrified waste does.

Belgium stopped reprocessing and switched over to direct disposal policy in 2001



0.224 km^2 for
3,915 glass canisters
(420 + 3,495 canisters)



1.3 km^2 for 9,859 spent fuel assemblies
+ 0.024 km^2 for 420 glass canisters

Summary (1)

- Proposed definition of environmental impact could successfully be applied
  - to identify critical radionuclides for minimization of environmental impact,
  - to find separation criteria for those critical radionuclides, and
  - to understand benefits of P&T

  from the viewpoint of long-term environmental impact from HLW geologic disposal.

Summary (2)

- Benefits of P&T have been found in:
  - that repository capacity could be significantly expanded without increasing environmental impact;
    - Less socioeconomic impact than having more repositories
  - that radiotoxicity in the repository decreases faster
    - due to transmutation of long-lived Np-237 and its precursor Am-241
  - that handling of HLW could become significantly rationalized;
    - More waste packages in the same footprint,
    - Less ventilation in pre-closure period,
    - More stable repository performance,
  - that weapons materials that goes into a repository reduces significantly,
    - Creation of “Pu mine” can be avoided,
    - Proliferation resistance of waste-handling systems increases.

Summary (3)

- If a P/T system is applied to the LWR system to reduce the environmental impact from the repository, the target nuclide would be Np-237, Am-241, and their precursors.
- Iodine 129 could be contained either in HLW, Low or Intermediate Level Wastes. In either cases, due to its long half-life, it will become environmental impact.
- A scoping study shows that treatment of CSNF of YMR capacity requires operation of a P&T system including:
  - 68 transmuter reactors,
  - Partitioning and fuel fabrication facilities,
  - UREX+ facility
  - Interim storages for separated TRU, Zr, and HLWs

  for more than 100 cycles. No nuclear fuel cycles have been operated at this size and level of complexity.

  ➔ We need a realistic and optimized plan for realization of P&T!!!