Effects of Repository Conditions on Environmental-Impact Reduction by Recycling

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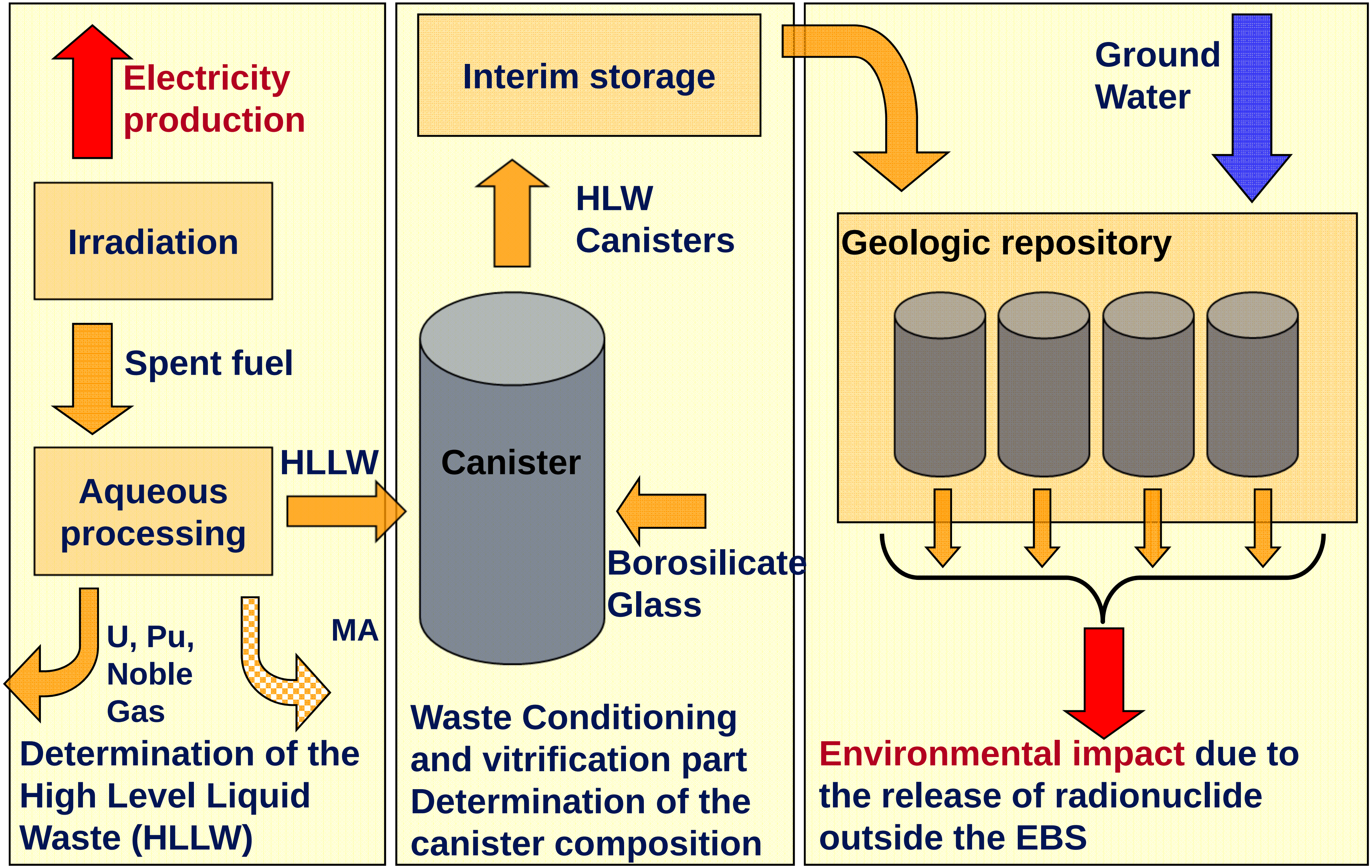
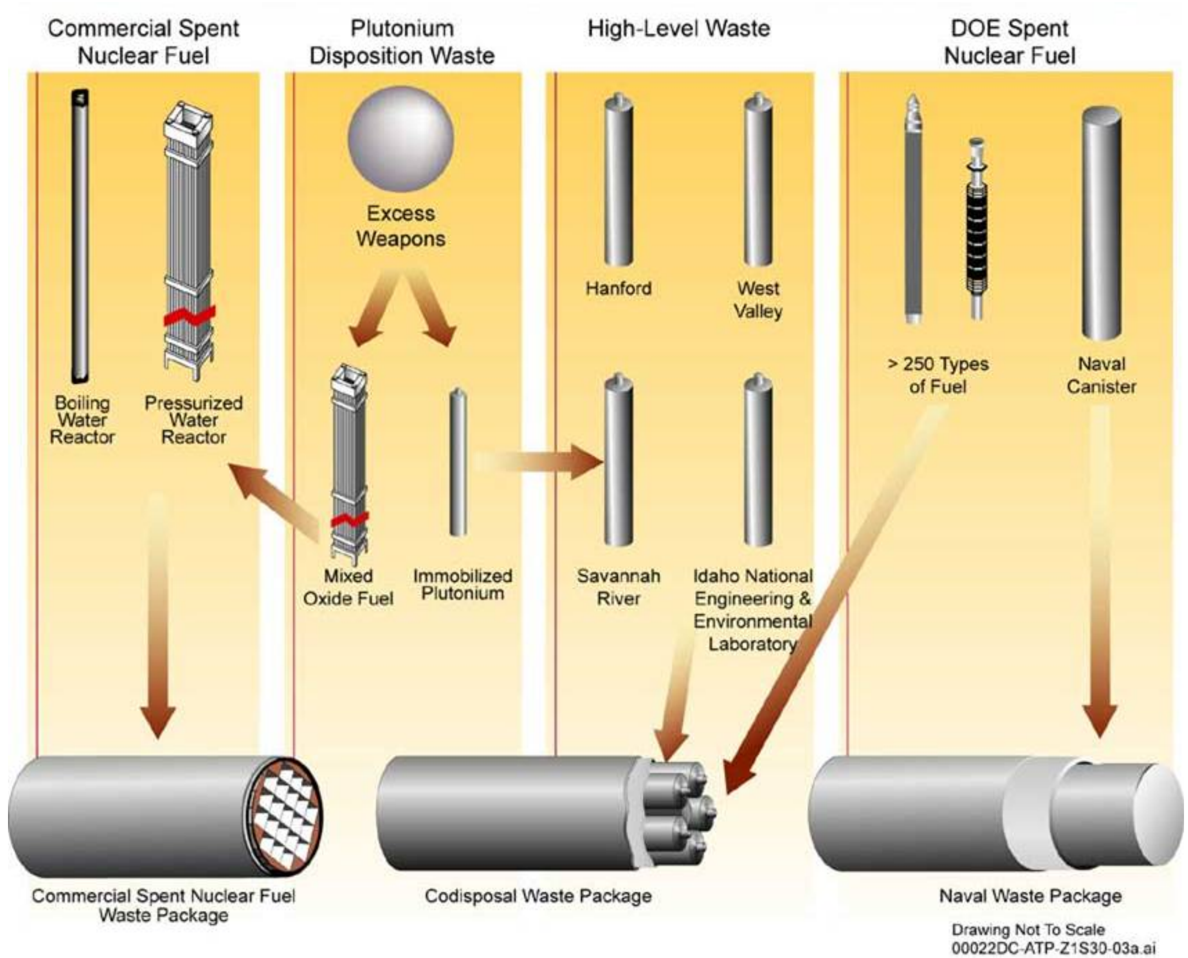
Objectives

- Compare the Environmental impacts of two different repository configurations coupled with fuel cycles
  - A water-saturated repository coupled with recycle with PWR-UO_2, PWR-MOX and FR.
  - Yucca Mountain Repository coupled with UREX+ and advanced fuel cycle for minor actinide recycle
- For these objectives, we have developed models and codes for:
  - Quantitatively determining the composition vector of vitrified HLW in a canister for final disposal, and
  - Quantitatively estimating radionuclide release rates from failed waste packages for the environmental impact assessment.

High Level Waste Flow

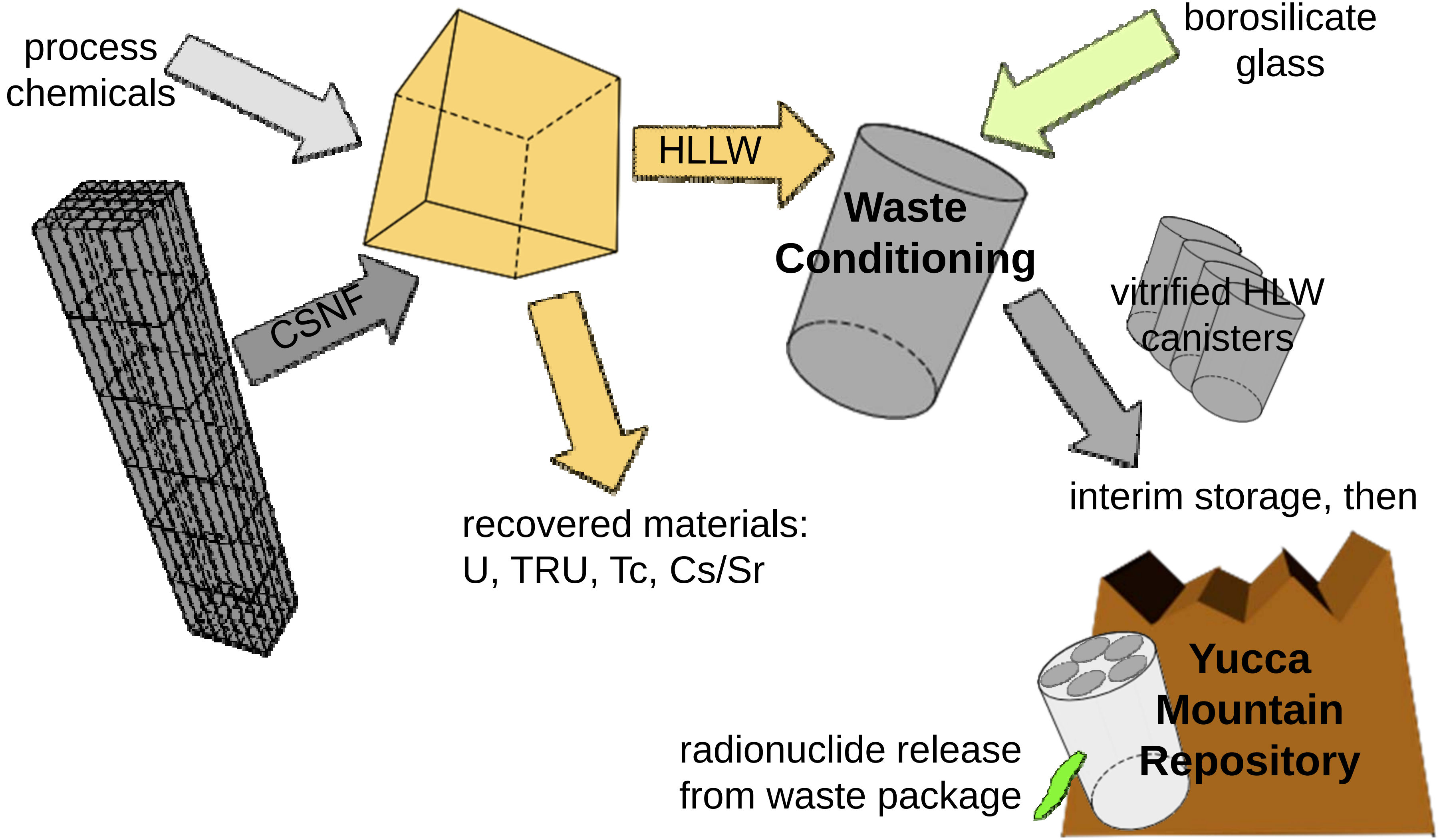


Three Types of Waste Packages



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

UREX1+ Combined with YMR



Models and Codes

Computer codes

ORIGEN Input data for PUREX cases

	PWR UO_2	PWR MOX	FR (Core/Axial)
	Cases (1)(2)	**Case (3)**	**Case (4)**
Burn-up Conditions			
Fuel composition before irradiation (g/MTHM)			
U-234	450	0	0
U-235	45000	1856	1722/833
U-236	250	0	0
U-238	954300	926144	571583/276942
Pu-238	0	1224	1637
Pu-239	0	40608	80568
Pu-240	0	16632	47798
Pu-241	0	8064	6404
Pu-242	0	4248	5812
Np-237	0	0	744
Am-241	0	1224	2981
Am-243	0	0	1488
Cm-244	0	0	1488
ORIGEN cross section library numbers			
	604/605/606	210/211/212	311/312/313 (core); 314/315/316 (blanket)
Thermal output (MW/MTHM)			
	38	37.7	35.9
Operating days (EFPD)			
	1184	1592	3200
Discharged burn-up / Core Average (GWd/MTHM)			
	45	60	115/150
Power allotment (core/axial blanket, %)			
	---	---	94.4/5.6
Capacity factor, C_{factor}			
	0.9		0.8
Conversion efficiency, C_{eff}			
	0.33		0.42

	PWR UO_2	PWR MOX	FR (Core/Axial)
	Case (2)	**Case (3)**	**Case (4)**
PUREX Conditions			
Cooling time before reprocessing, T_b (yr)			
	3	10	7
Cooling time between reprocessing and vitrification, T_a (yr)			
	1	1	1
Fractions removed from HLLW by PUREX (%)			
U	99.5	99.5	99.5
Pu	99.5	99.5	99.5
Np	0	0	99.5
Am	0	0	99.5
Cm	0	0	99.5
H	100	100	100
C	100	100	100
I	99	99	99
Cl	100	100	100
He	100	100	100
Ne	100	100	100
Ar	100	100	100
Kr	100	100	100
Xe	100	100	100
Rn	100	100	100

ORIGEN Input data for UREX cases

Burn-up Conditions for Cases (5) (6-1) (6-2) (6-3)	
Fuel composition before irradiation (g/MTHM)	
U-235	43000
U-238	957000
ORIGEN cross section library numbers	219/220/221
Thermal output (MW/MTHM)	40
Operating days (EFPD)	1250
Discharged burnup (GWd/MTHM)	50
Capacity factor, C_{factor}	0.9
Conversion efficiency, C_{eff}	0.33

UREX1a+ Conditions			
Cooling time before UREX1a+, T_b (yr)			15
Cooling time between UREX1a+ and vitrification, T_a (yr)			0
Fractions removed from HLLW by UREX1a+ (%)			
	Case (6-1)	**Case (6-2)**	**Case (6-3)**
U	95	99	99.5
Pu	95	99	99.5
Np	95	99	99.5
Am	95	99	99.5
Cm	95	99	99.5
Tc	95	99	99.5
Cs	95	99	99.5
Sr	95	99	99.5
H	100	100	100
C	100	100	100
I	100	100	100
Cl	100	100	100
He	100	100	100
Ne	100	100	100
Ar	100	100	100
Kr	100	100	100
Xe	100	100	100
Rn	100	100	100

Solidification of HLW

Reprocessing

HLLW

Solidification process

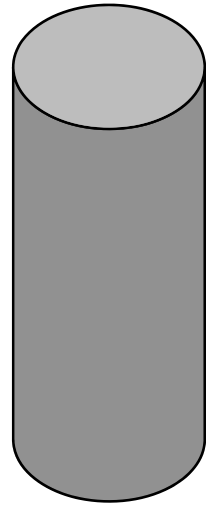
HLW in oxide forms
(Fission products,
actinides,
activation products,
corrosion products,
process chemicals, etc.)

Mass: M_W

Composition vector: $\vec{N}_W$

$$\vec{N}_W = \begin{bmatrix} x_{W,1} \\ . \\ . \\ x_{W,i} \\ . \end{bmatrix}$$

canister



Borosilicate glass

Mass: M_G

Composition vector: $\vec{N}_G$

$$\vec{N}_G = \begin{bmatrix} x_{G,1} \\ . \\ . \\ x_{G,i} \\ . \end{bmatrix}$$

Repository

Interim storage

Solidified HLW

Mass: $M_S = M_W + M_G$

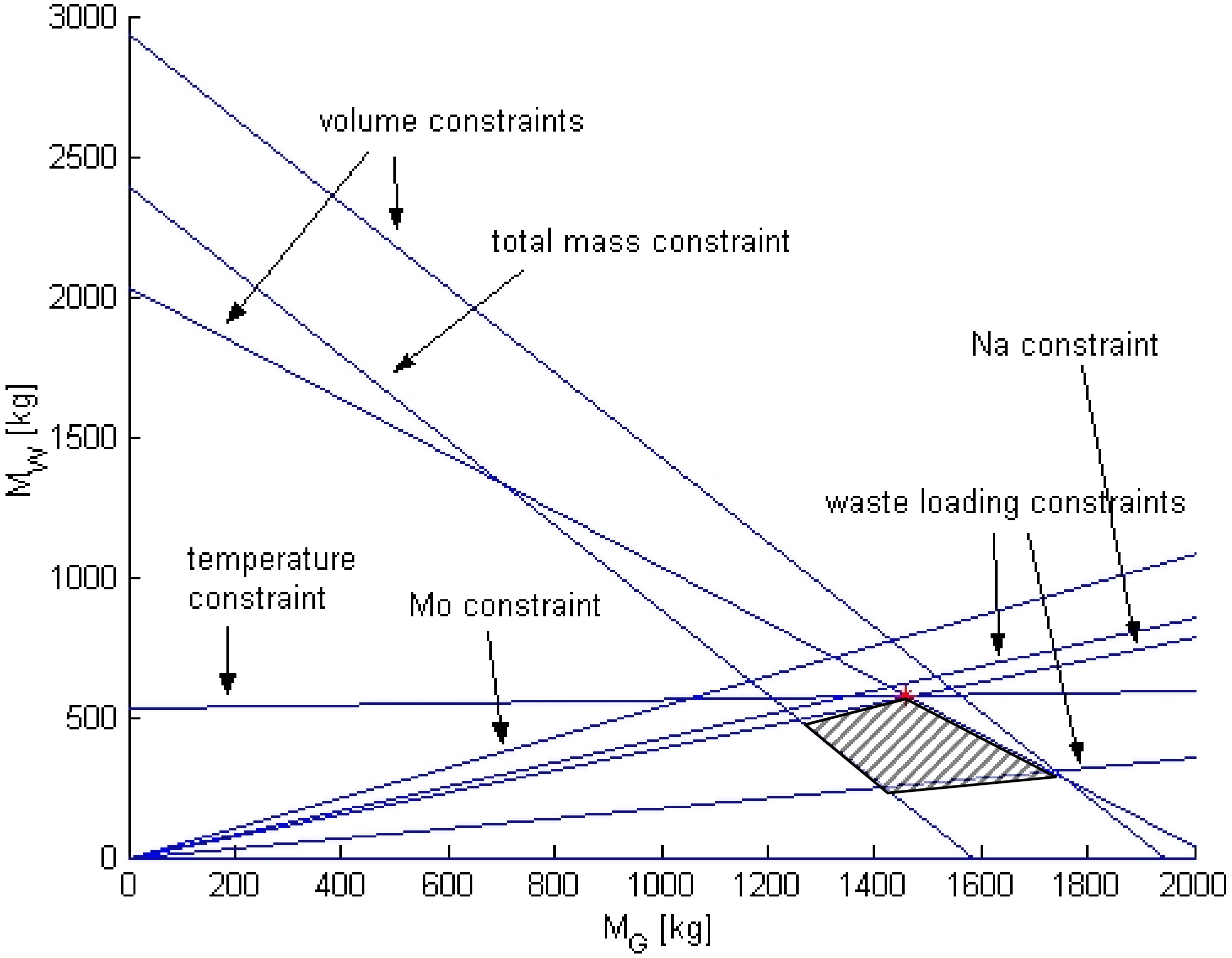
Composition vector: $\vec{N}_S = \theta \vec{N}_W + (1-\theta)\vec{N}_G,$

$$\text{where } \theta \equiv \frac{M_W}{M_W + M_G}$$

Constraints for Optimization

Specifications/Constraints	Water-saturated	YMR
Canister height (m)	1.34	3
Canister outer radius (m)	0.215	0.305
Canister thickness (m)	0.006	0.01
Canister volume, V_c (m^3)	0.15	0.82
Empty canister weight (kg)	100	467
Total mass of a package (kg)	<500	<2,500
Mass fraction of Na_2O (wt%)	<10	<10
Mass fraction of MoO_3 (wt%)	<2	<2
Concentration of Pu (kg/m^3)	<2.5	<2.5
Heat emission (kW/canister)	<2.3	----
Maximum temperature in glass (^oC)	----	<400
Volume of vitrified HLW (m^3/canister)	$< V_c$	$0.8V_c < V < V_c$

Graphical representation for the feasible solution space
(US vitrification process)



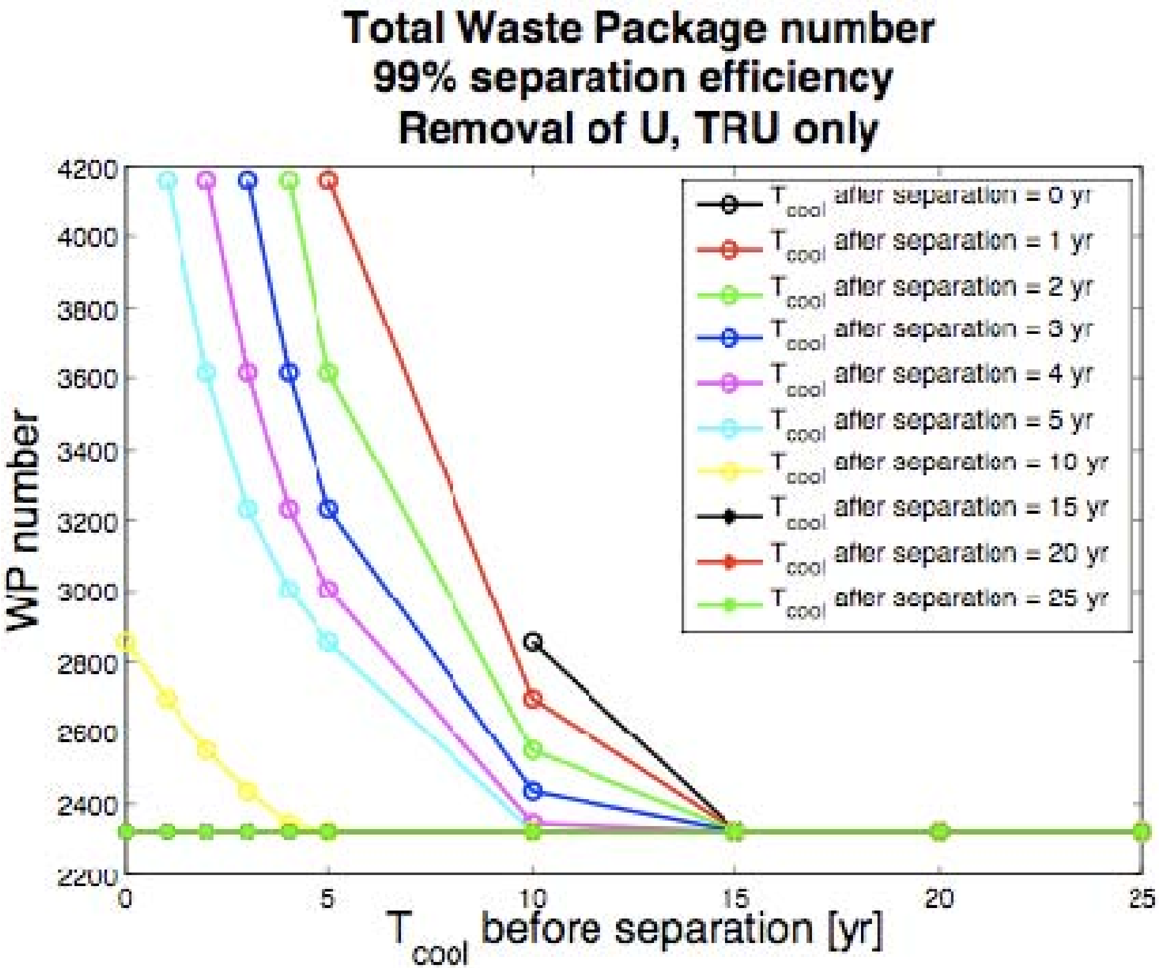
Cooling time before reprocessing and vitrification = 15 years

Results for optimized vitrification

PUREX cases			
	PWR UO_2	PWR MOX	FR (Core/Axial)
	Case (2)	**Case (3)**	**Case (4)**
Number of Canisters per MTHM of Fuel (Can/MTHM)			
	1.27	2.00	1.97

UREX cases			
Number of packages for HLW generated by UREX1a+ processing of 63,000 MTHM of Fuel			
	Case (6-1) 95%	Case (6-2) 99%	Case (6-3) 99.5%
	2994	2324	2324

Waste Package Number vs. Cooling Time (YMR)

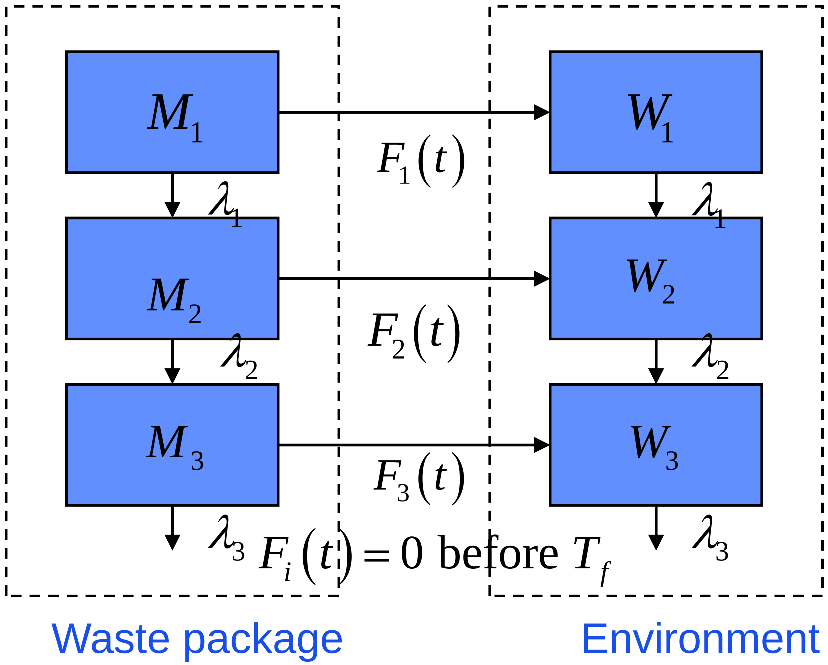


- effect of Cs/Sr not significant at longer than 15 years
- choosing 15 yr cooling time ensures minimization of waste package number
- consistent with DOE Environmental Impact Statement

Environmental Impact per GWyr

- Total toxicity index of radionuclides observed outside the Engineered Barrier Systems (EBS)
- Its peak value will be referred as the PEI (Peak Environmental Impact)

Mass Balance Equations



Environmental Impact due to Nuclide i

$$I_i\ [m^3] \equiv \frac{\lambda_{Np} W_i\ [Ci]}{MPC_i [Ci/m^3]}$$

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:

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

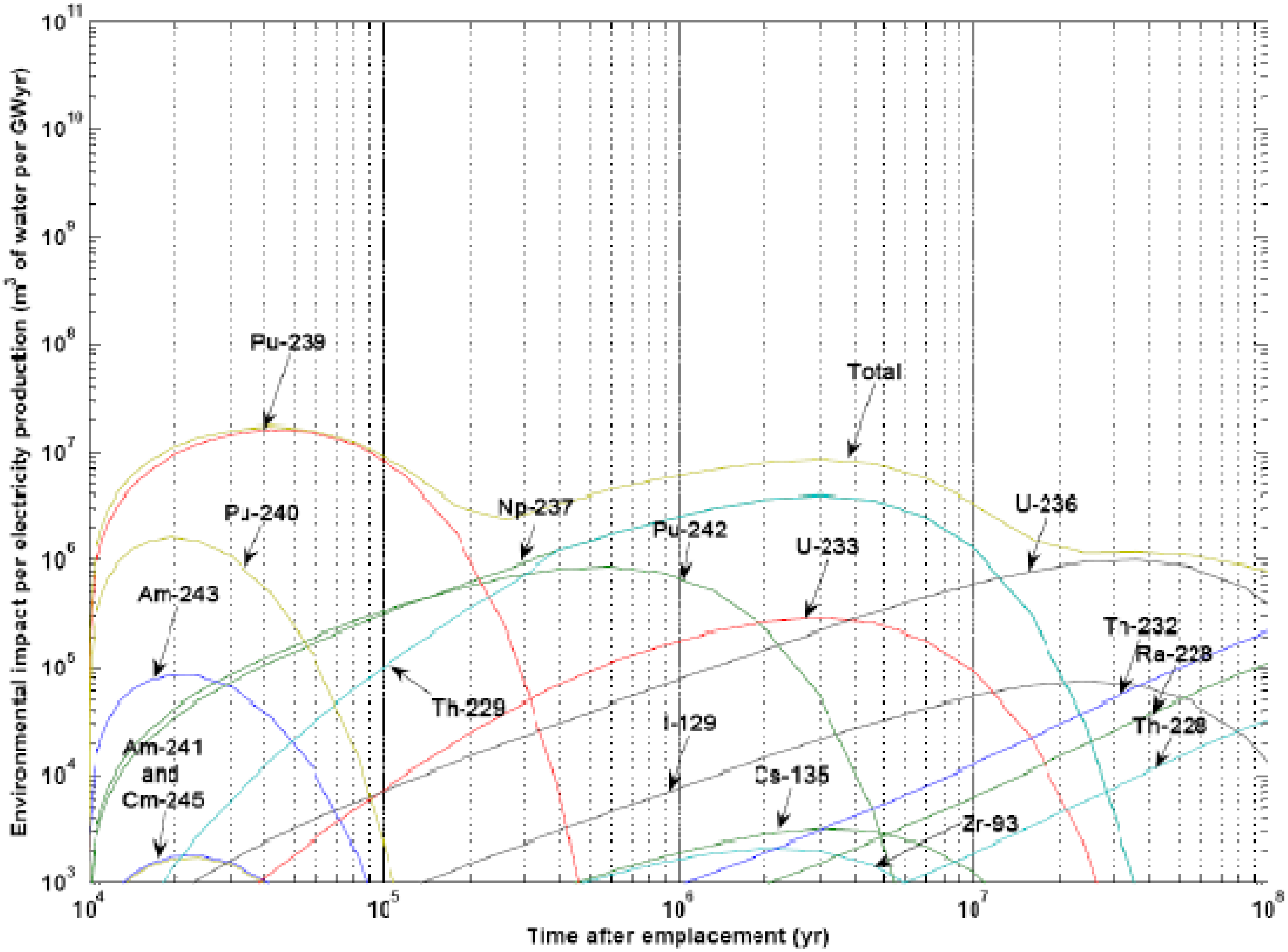
N: number of packages per GWy

Input data for cases considered

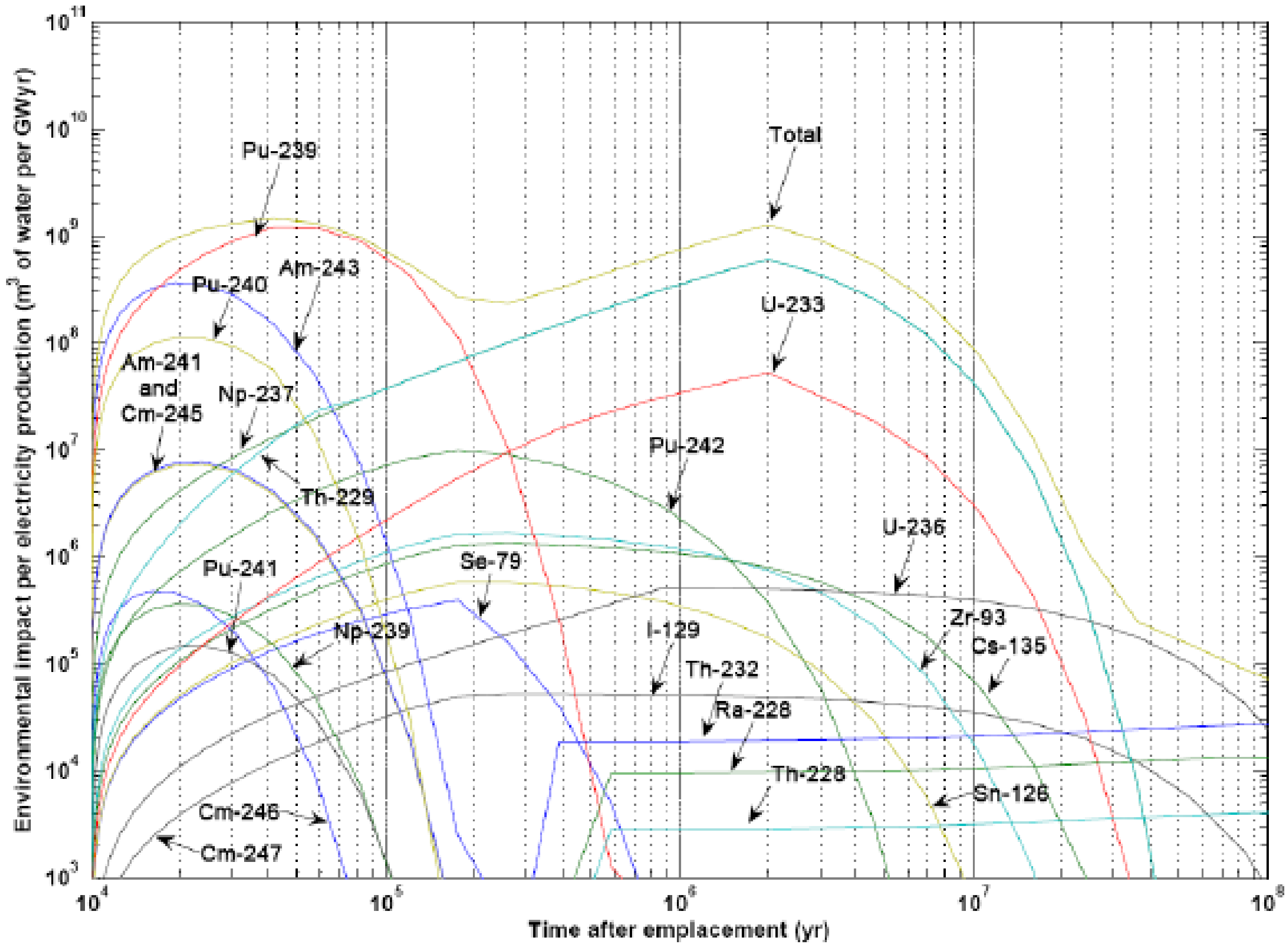
Parameters		Water-saturated	YMR
Canister/Package failure time, T_f (yr)		10,000	75,000
Radius of waste package (m)		0.21	---
Length of waste package (m)		1.34	
Pore velocity of groundwater in surrounding geologic formations (m/yr)		1	0.77
Porosity of the surrounding medium		10%	10%
Diffusion coefficient in the surrounding medium (m^2/yr)		3E-2	3E-2
Solubility in groundwater (mol/m^3)	Se	3.0E-06	1.0E+02
	Zr	1.0E-03	6.8E-07
	Nb	1.0E-01	1.0E-04
	Tc	4.0E-05	high
	Pd	1.0E-06	9.4E-01
	Sn	1.0E-03	5.0E-05
	Cs	high	high
	I	high	high
	Sm	2.0E-04	1.9E+02
	Pb	2.0E-03	1.0E-02
	Ra	1.0E-09	2.3E-03
	Ac	2.0E-04	1.9E+02
	Th	5.0E-03	1.0E-02
	Pa	2.0E-05	1.0E-02
	U	8.0E-06	4.0E-01
	Np	2.0E-05	1.6E+01
	Pu	3.0E-05	2.0E-01
	Am	2.0E-04	1.9E+02
	Cm	2.0E-04	1.9E+02
	Si	0.21	2.1

Numerical Results

EI per GWy for Direct disposal in water-saturated repository: Case (1)

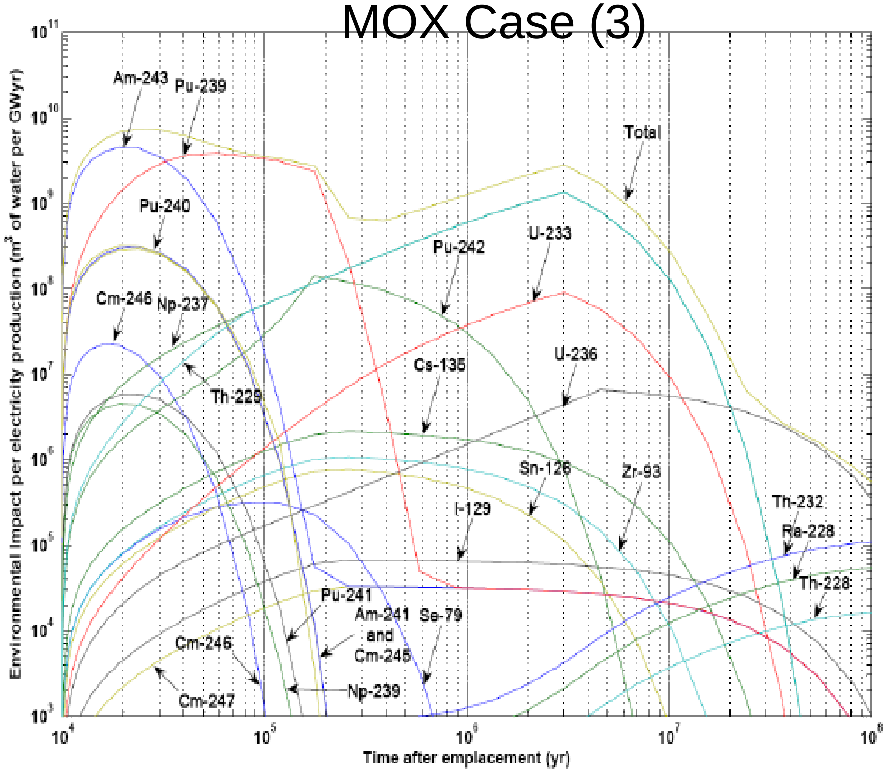


EI of HLW from PWR-UO_2 in water-saturated repository: Case (2)

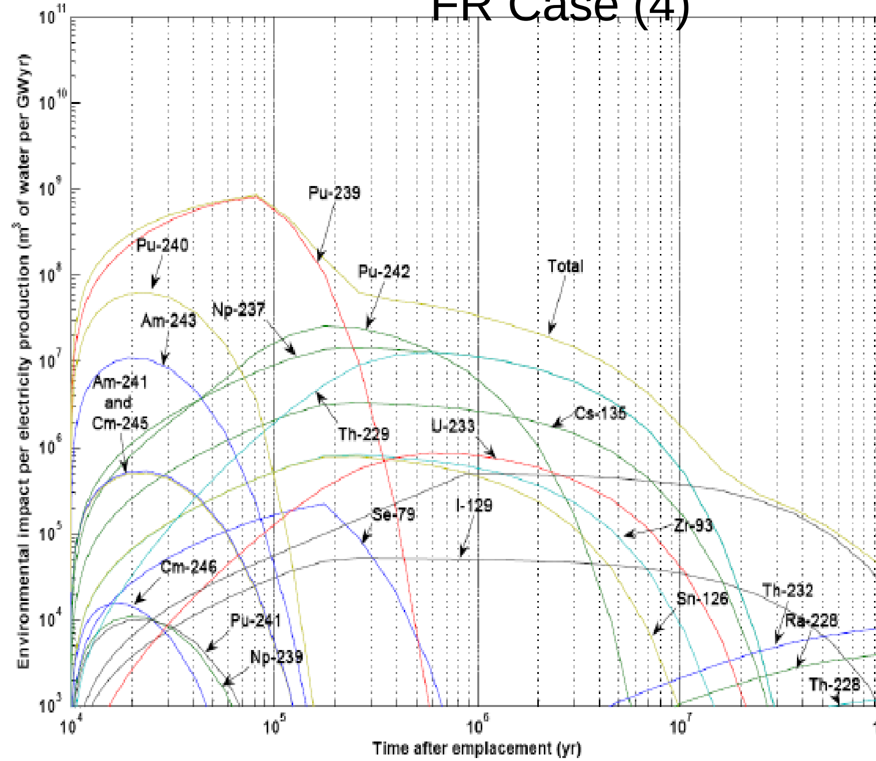


EI per Gwy in water saturated repository

MOX Case (3)



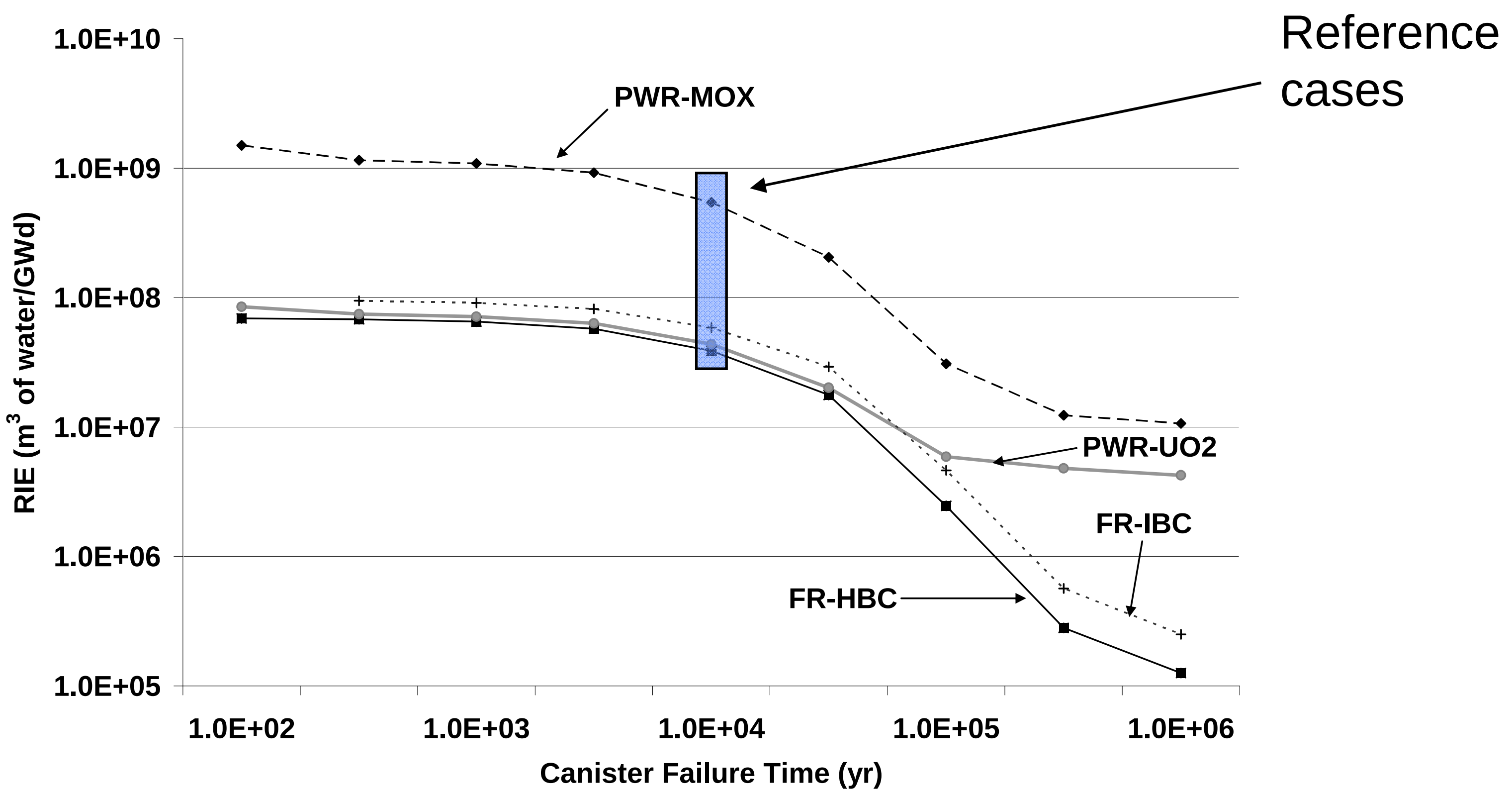
FR Case (4)



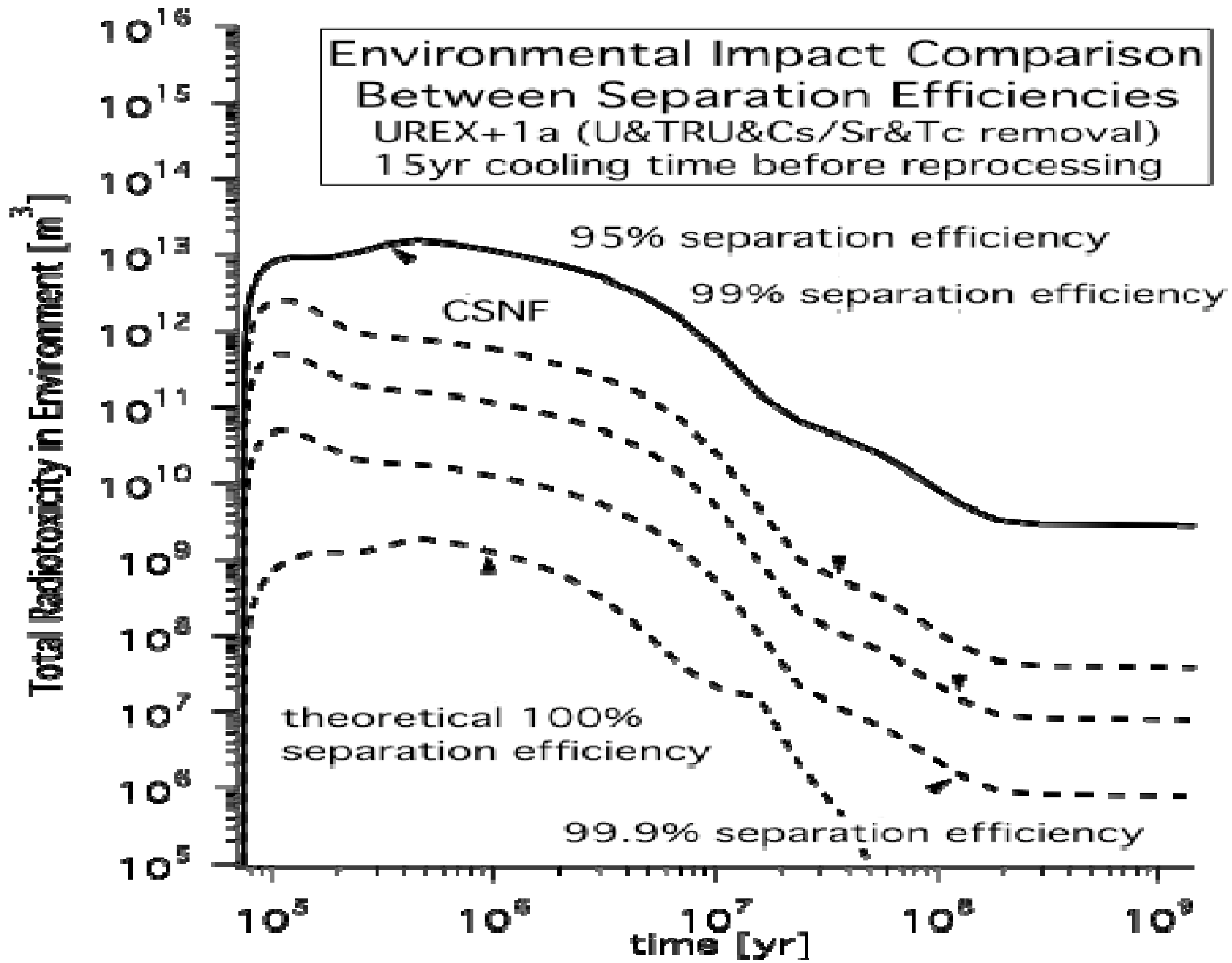
	PWR-MOX	FR
■ Am removal:	0%	99.5%
■ Cm removal:	0%	99.5%

  - □ Cm-244->Pu-240
  - □ (Cm-243->Pu-239)
- ■ Am and Cm removals lead to a lower PEI for FRs.
  - □ Impact of Am-243

Environmental impact per electricity generation vs. Canister Failure Time



Comparison of Total EI for YMR (Different Separation Efficiencies)



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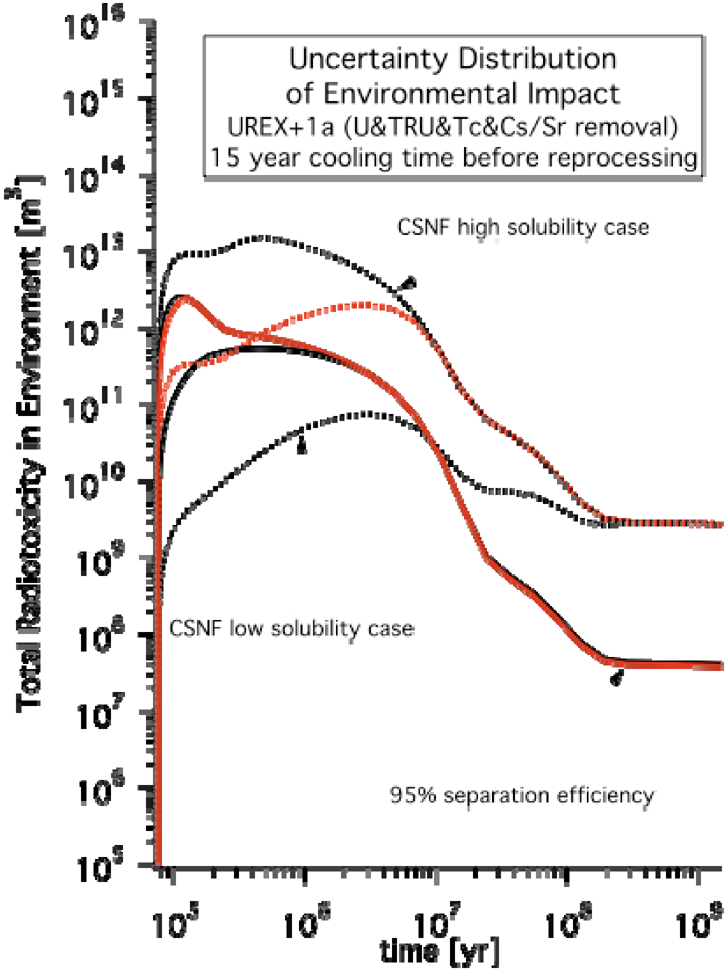
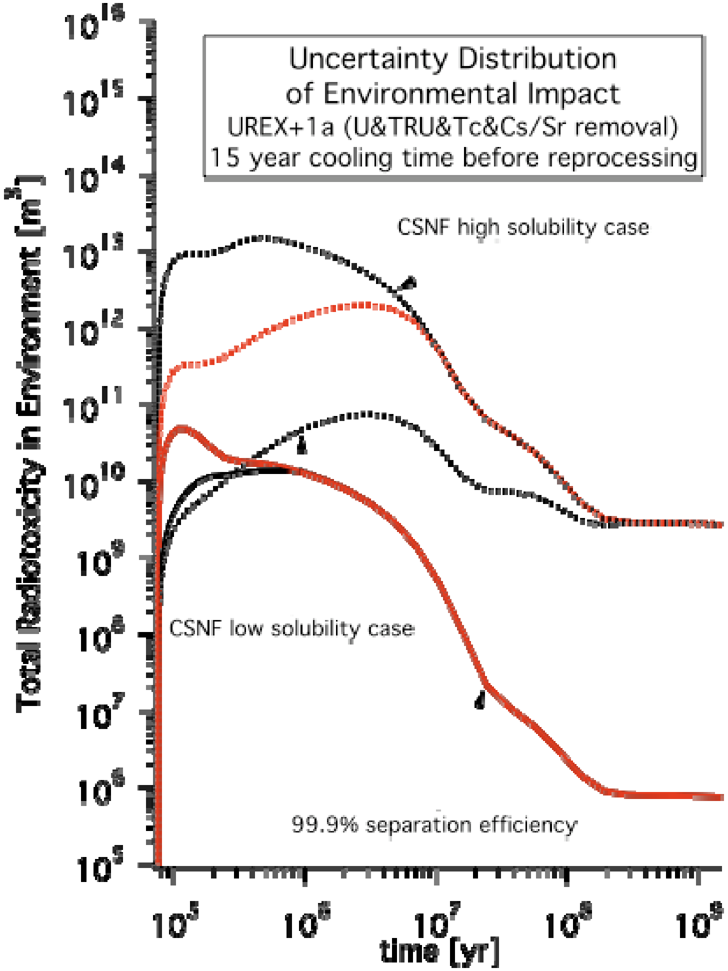
Effects of Solubility Uncertainty



- Comparison: CSNF vs. 95% separation efficiency
- At early times, mean EI of HLW greater than that of CSNF
- Low separation efficiency case indistinguishable from CSNF case?

Effects of Solubility Uncertainty



- ●Comparison: CSNF vs. 99.9% separation efficiency
- ●Although early part of the HLW curve still within the envelope of CSNF uncertainty distribution, means are distinctly different

Effects of separation of TRU

Water-saturated repository	(1) PWR UO_2 spent fuel	Vitrified HLW from PUREX		
		(2) from PWR UO_2 with 99.5% removal for U and Pu	(3) from PWR MOX with 99.5% removal for U and Pu	(4) from FR with 99.5% removal for each actinides
EIE (m^3/GWyr)	1.7E7	1.4E9	7.5E9	8.3E8
	1	82	440	49

Yucca Mountain Repository	(5) LWR UO_2 spent fuel	Vitrified HLW from LWR UO_2 by UREX+		
		(6-1) 95% removal for each actinide	(6-2) 99% removal for each actinide	(6-3) 99.5% removal for each actinide
EIE (m^3/GWyr)	4.9E9	1.2E9	2.3E8	1.2E8
	1	0.24	0.047	0.024

Summary

- Without separation of TRU, the level of the environmental impact normalized by electricity generation would be significantly dependent on repository conditions and solidification matrix.
  - UO2 spent fuel in the Yucca Mountain Repository would cause a greater potential impact than a water-saturated repository, where reducing environments are assumed.
  - In water-saturated environments, uranium oxide is considered to be thermodynamically stable and solubilities of actinides are significantly smaller than those in YMR conditions.
- The environmental impacts per GWyr from vitrified HLW after removal of TRU elements would be similar between both repository conditions.
  - This results from the assumption that borosilicate glass dissolves in a similar rate in either reducing or oxidizing environments due to thermodynamically unstable amorphous structure, and that radionuclides are released congruently with matrix dissolution.
- For the YMR, the effects of separation efficiencies appear proportionally on the environmental impact.