UNIVERSITY OF CALIFORNIA AT BERKELEY

NUCLEAR ENGINEERING DEPARTMENT

TRANSMUTATION CAPABILITY OF MOLTEN SALT REACTORS FUELED WITH TRU FROM LWR

Massimiliano Fratoni

Ehud Greenspan

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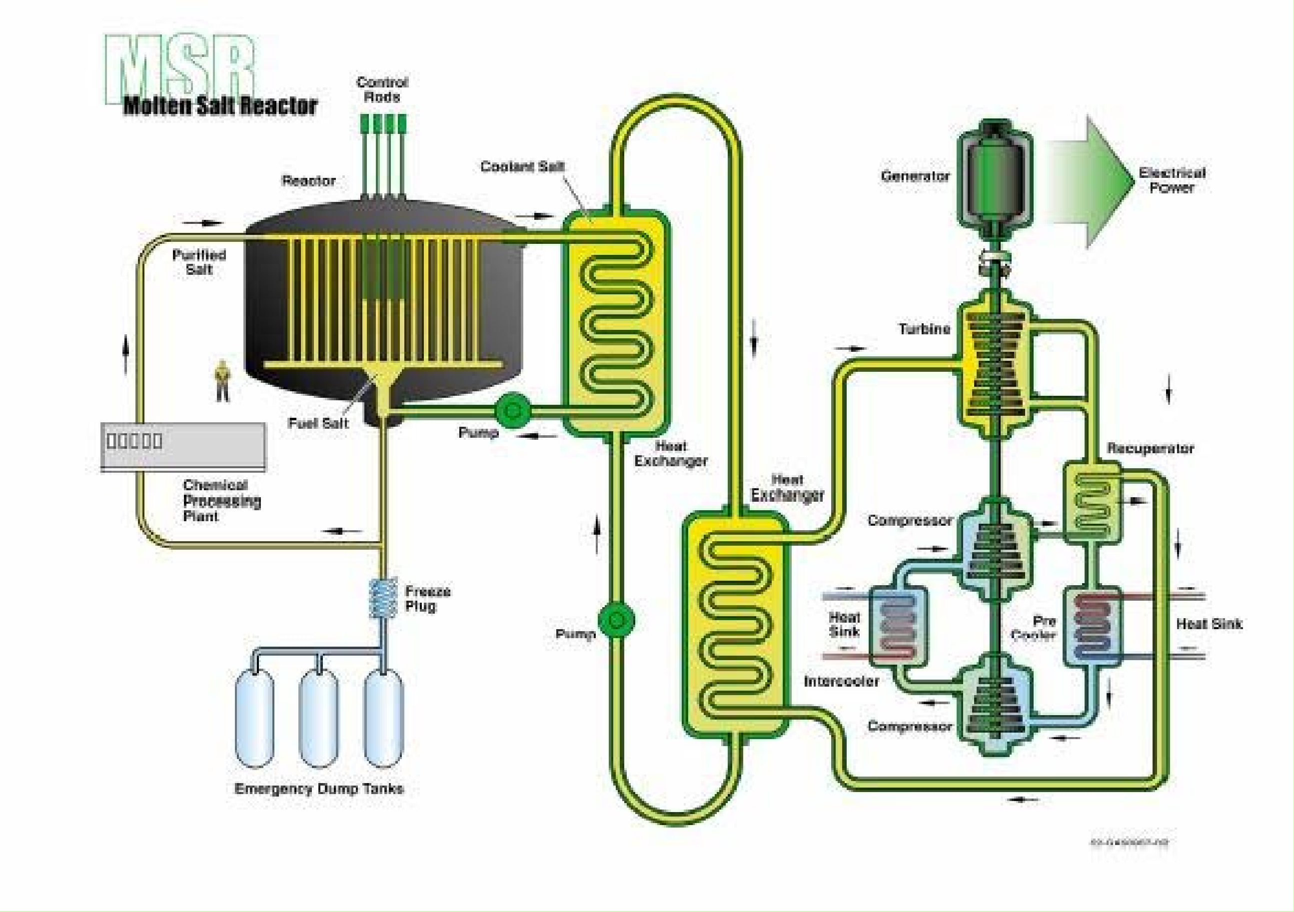
MSR Features



NaF - ZrF_4 – AcF_3

600 ÷ 700 °C

Once-through

Suggested by Charles Bowman

Fed by TRU from spent fuel from LWR after a burnup of 33 GWD/t_{HM} and 10 years of cooling and after U isotopes extraction

Objectives

1. *Assess the feasibility of attaining criticality in a finite core*

2. *Evaluate MSR performance sensitivity to design variables*

3. *Evaluate repository implications*

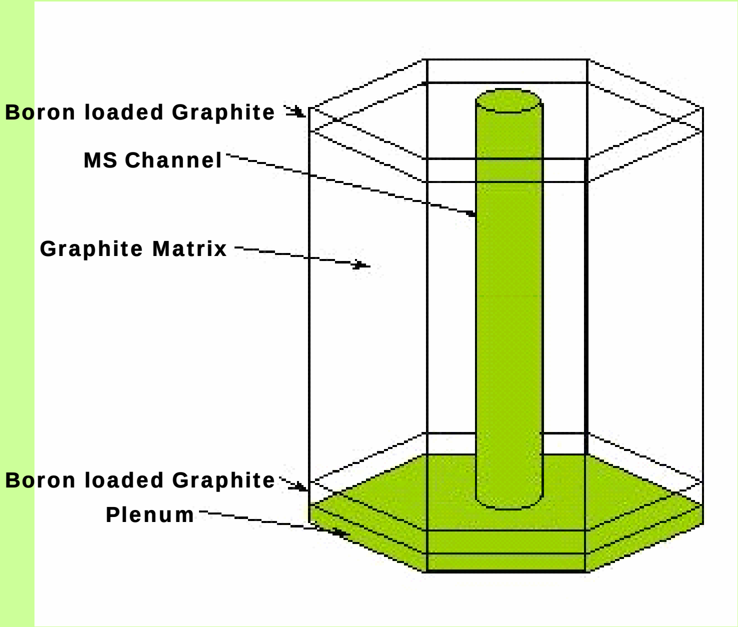
4. *Compare transmutation efficiency for different TRU burner systems*

Previous work 1. Finite Model

Graphite-to-MS volume ratio (C/MS) = 3

$D_{channel}$ =7 cm, H = 420 cm

k_{eff} = 1.032 (with axial leakage)

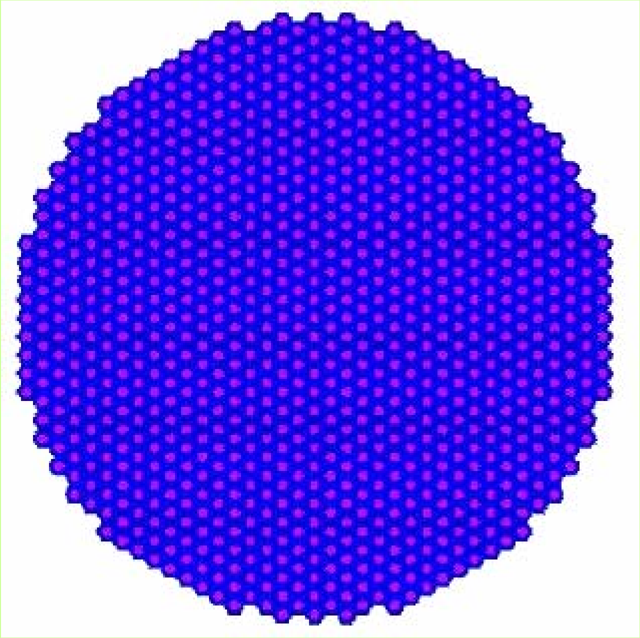


H/D_{core} = 0.924, 955 channels

5733 MW_{th} (390 W/cm^3)

k_{eff} = 1.00247 ($\pm$0.00067)

MCNP4C

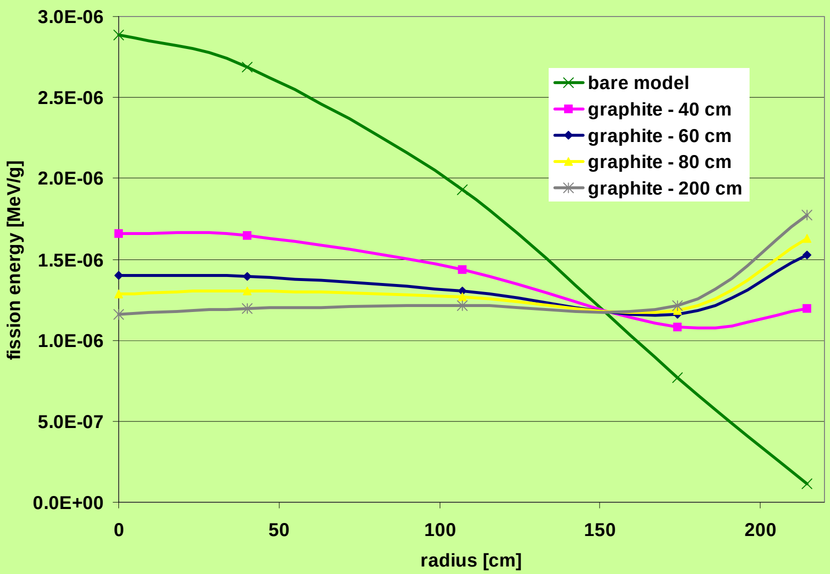


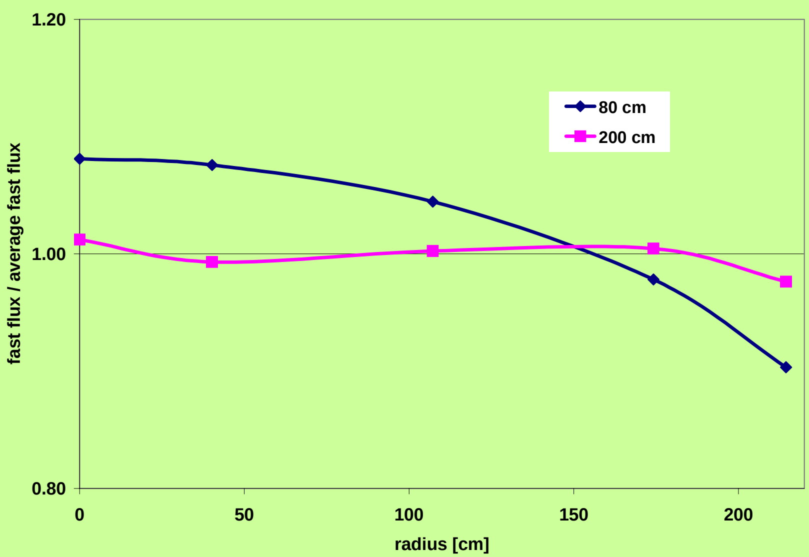
1. Radial Reflector

Graphite Thickness [cm]	k_{eff}	Radial Leakage Probability
0	1.00247 (± 0.00067)	2.56 %
40	1.01916 (± 0.00059)	2.74 %
60	1.02574 (± 0.00059)	2.85 %
80	1.02848 (± 0.00058)	2.78 %
200	1.03088 (± 0.00059)	2.79 %

(Due to statistics)

***Limit*:**
Maximum fast neutrons fluence (> 0.01 MeV) in the graphite
$3 \cdot 10^{22}$ neutrons/cm²

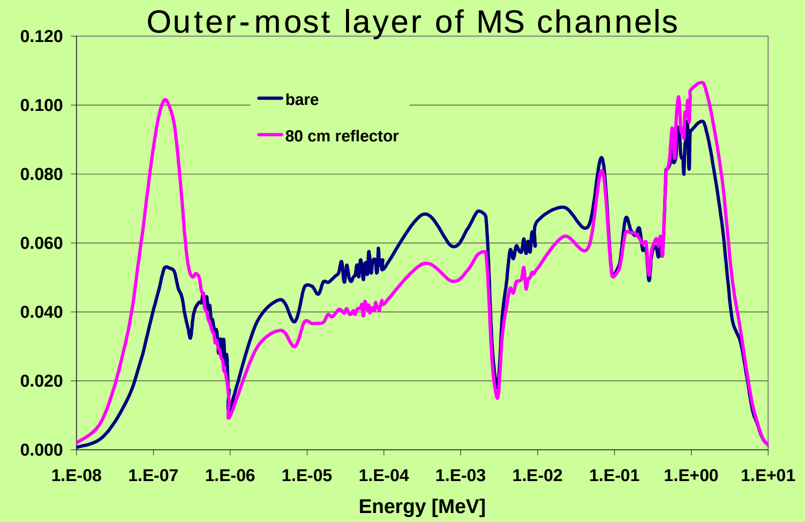


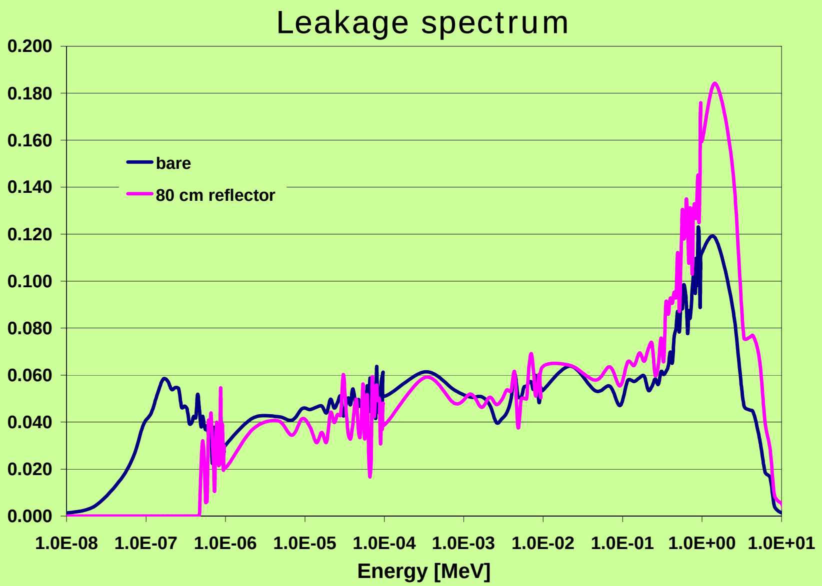


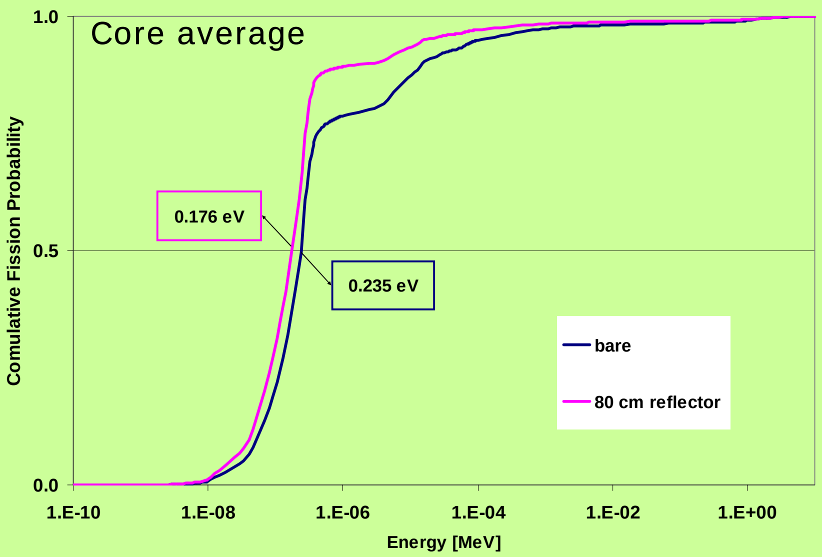
1. Leakage Probability

Graphite double effect:

- *reflector*
- *moderator*





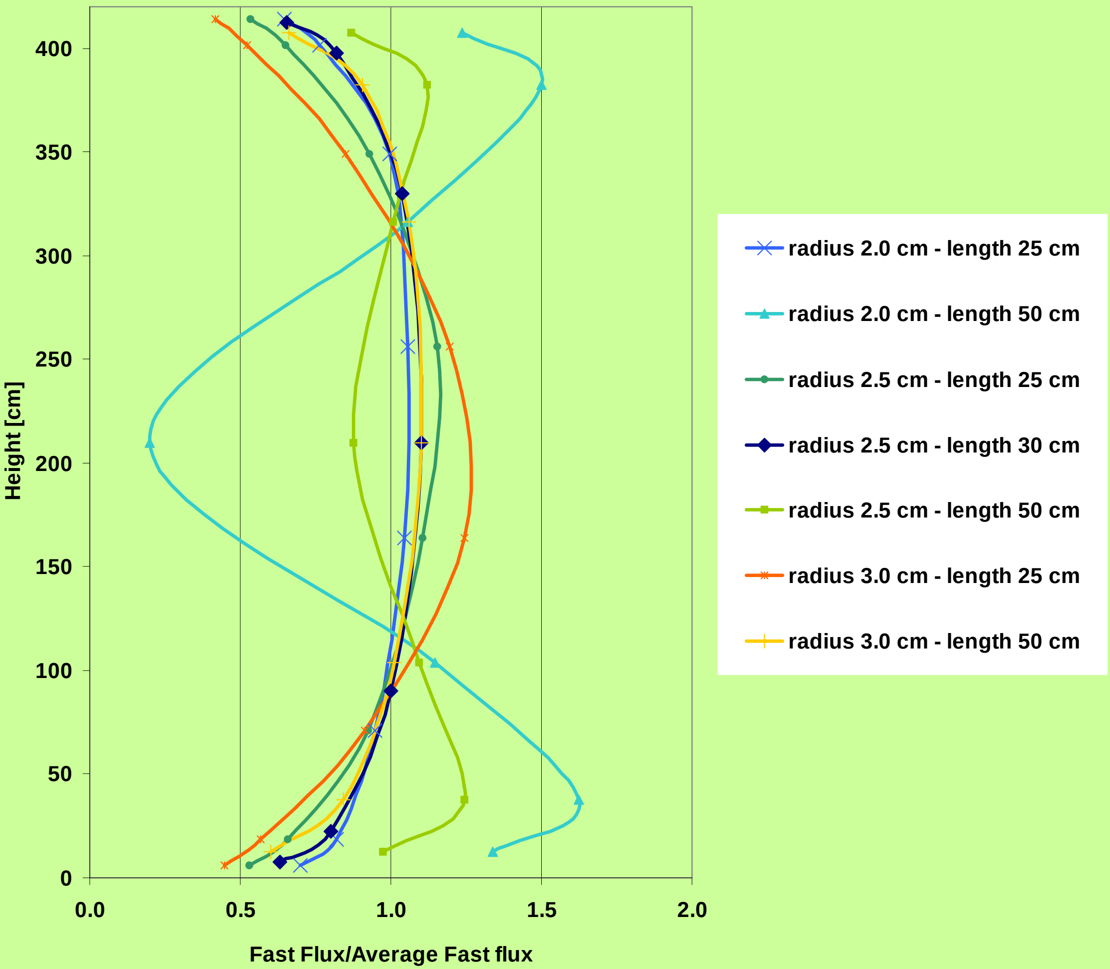


1. Axial Reflector

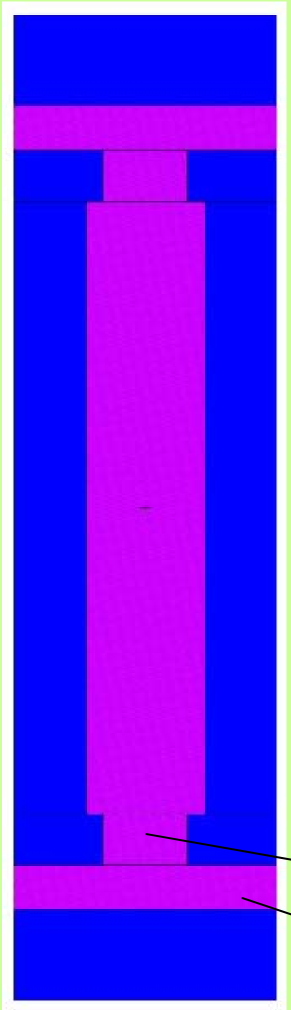
Problem:
plenum (27 cm) to collect the salt below and above the core

Graphite beyond plenum is ineffective reflector

Solution: to narrow the channels diameter at the extremities



1. Final Design



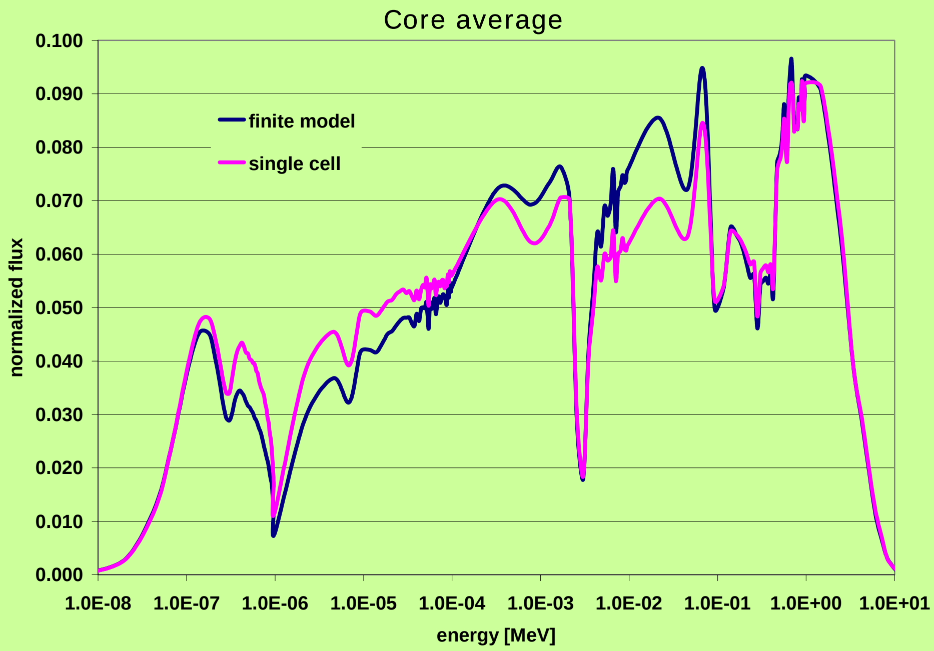
Maximum-to-average fast flux in the graphite ***1.079***

k_{eff} **= 1.04047**

The reactivity excess should compensate for:

1. *neutrons absorption by fission products*
2. *fraction of delayed neutrons released out of core*

1. Equilibrium Composition



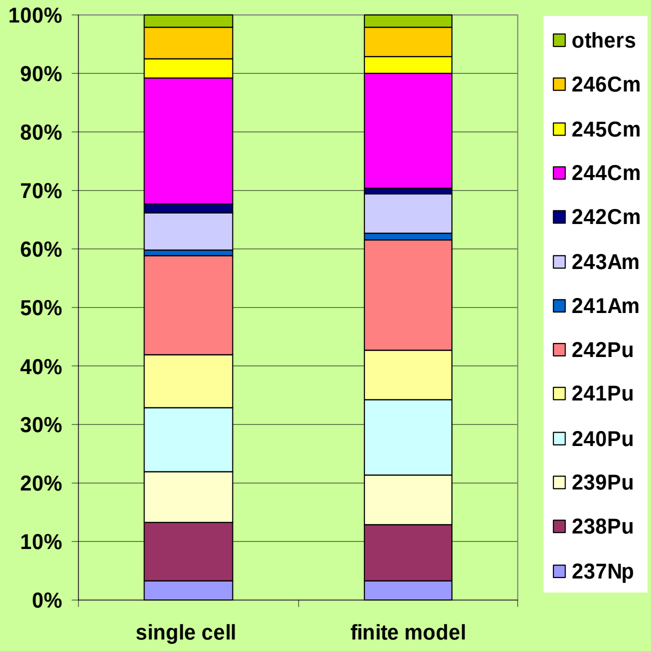
Single cell $k_{eff\ (updated)}$ = 1.02351

Finite model k_{eff} = 0.98517

- **radial leakage (2.53 %)**
- **increased absorption in non-actinides salt components**

C / MS	2.8	3.0	3.2	3.4
k_{eff}	0.98267	0.98517	0.98910	0.98799

∴ Finite core can not be critical with NaF-ZrF_4 even if fission products are extracted immediately



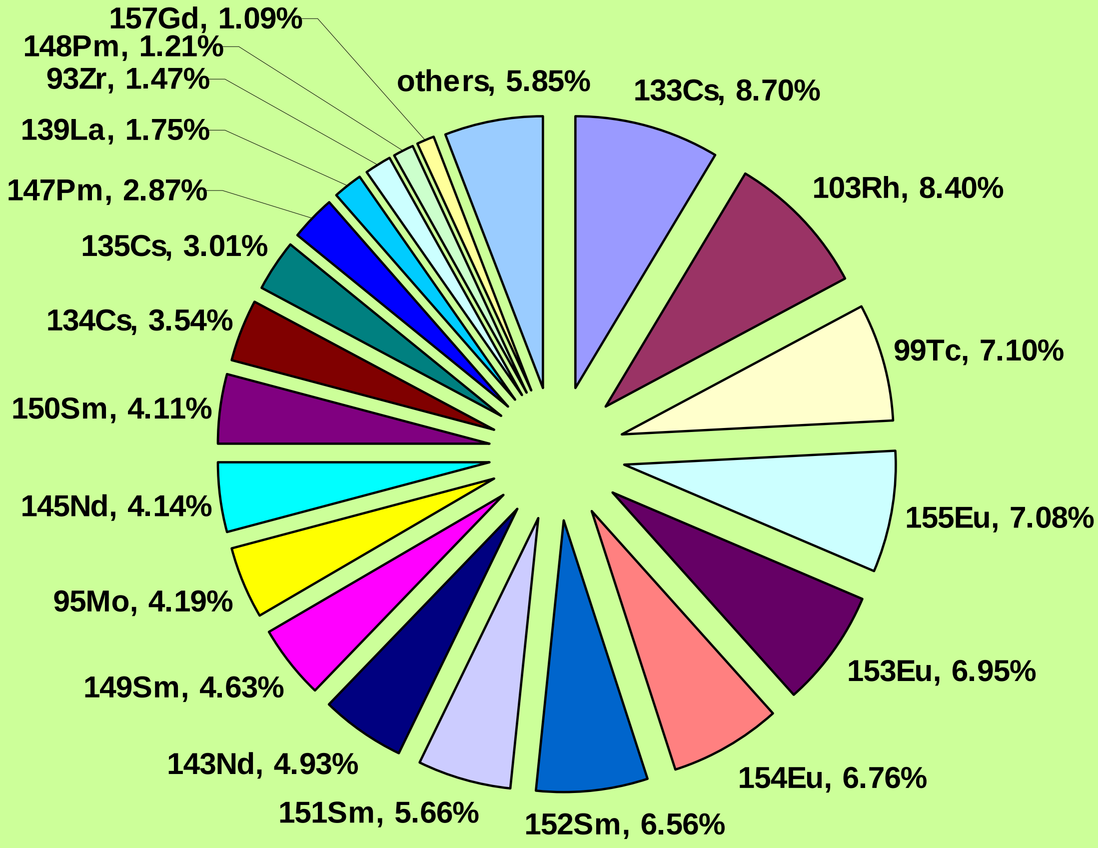
Equilibrium composition

1. Fission Products ρ Effect

No Extraction

If FP are not removed at all, Δk_{eff} = - 0.222

Not manageable



Fractional absorption by FP

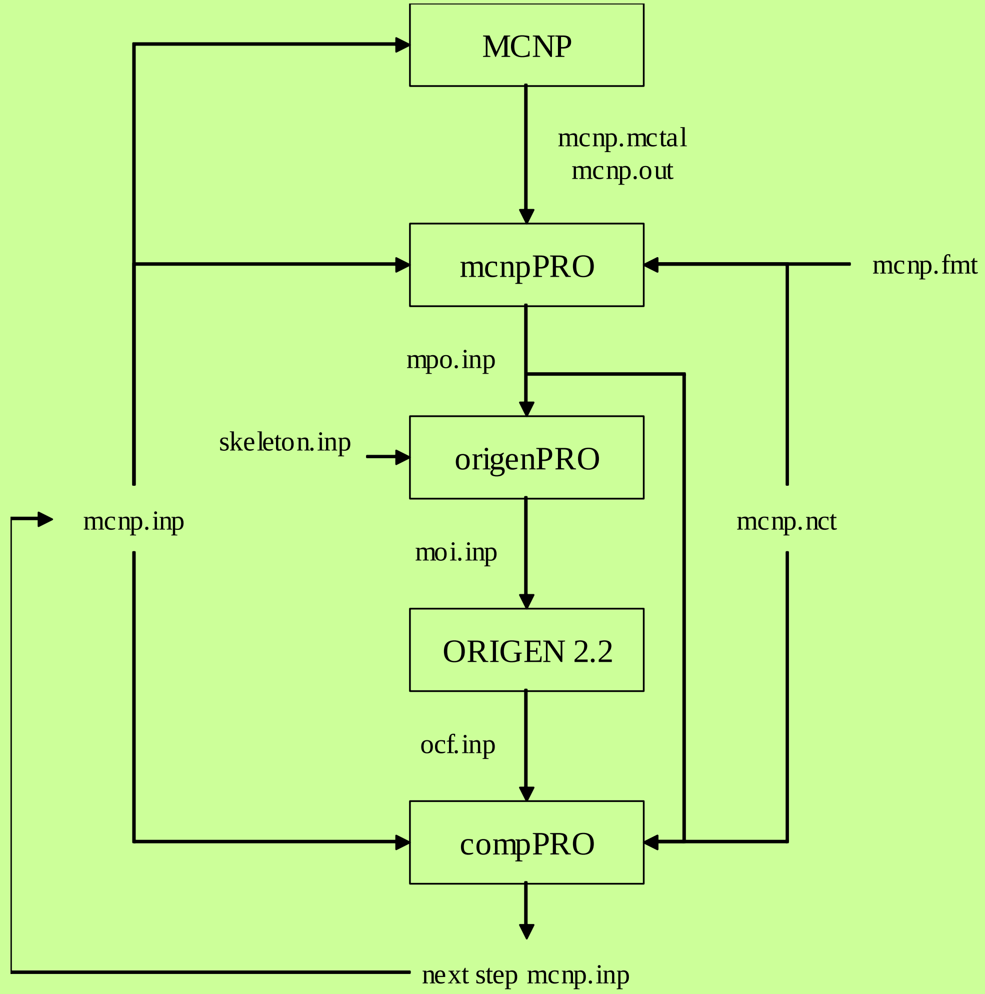
1. Fission Products ρ Effect

Fission products residence time

FP	Residence Time
Kr, Xe	50 s
Nb, Mo, Ru, Rh, Tc	2.4 h
La, Ce, Pr, Nd, Pm, Sm, Eu	< 50.4 d
Y	1 y
Sr, Ba, Cs	Salt residence time

$$\Delta k_{eff} = -0.018$$

Manageable!



2. Sensitivity Analysis

Design variables:

1. *C/MS (spectrum)*
2. *Feed Rate*
3. *Power Density*

Performance parameters considered:

1. *K_{eff} (finite unit cell)*
2. *Graphite lifetime*
3. *Transmutation Efficiency*

$$FT = \frac{\sum_i N_{i,feed} - \sum_i N_{i,equilibrium}}{\sum_i N_{i,feed}}$$

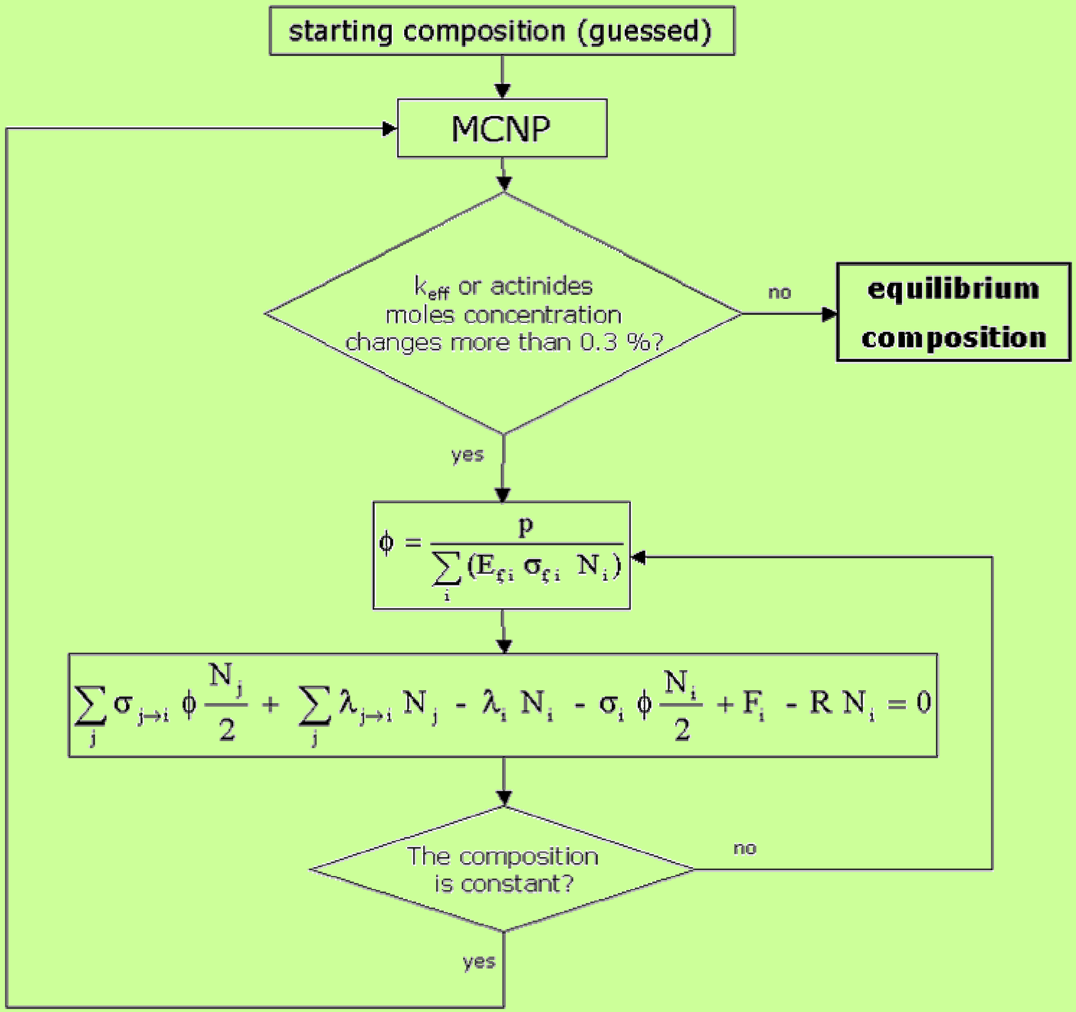
2. Eq. Composition Determination

34 actinides

reaction: fission, (n,γ), (n,2n), (n,3n)

***Constraints**:*

– *Actinide fluorides maximum solubility in the salt **1.56 mol%***
– *maximum fast neutrons fluence (> 0.01 MeV) in the graphite **$3 \cdot 10^{22}$ neutrons/cm^2***



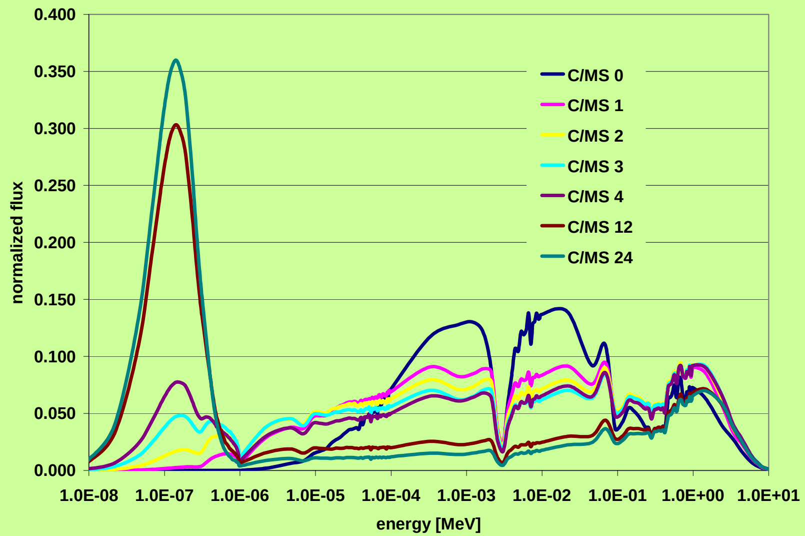
2. Feed Rate Effect

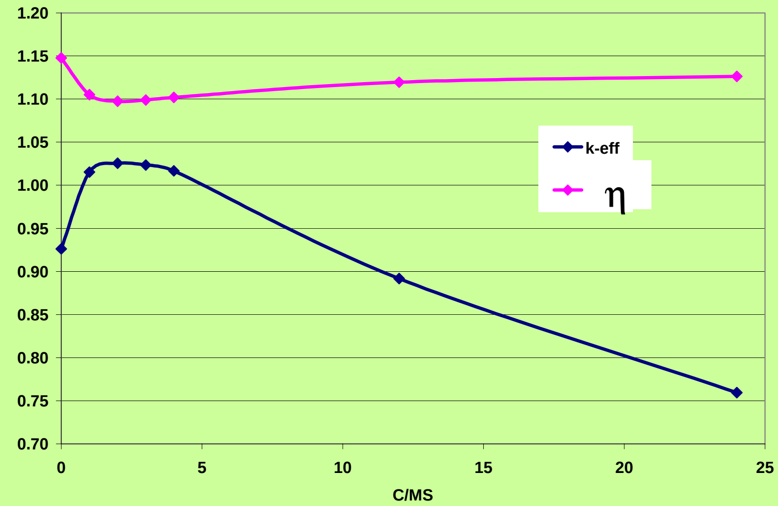
C/MS=3, power density=390 W/cm^3

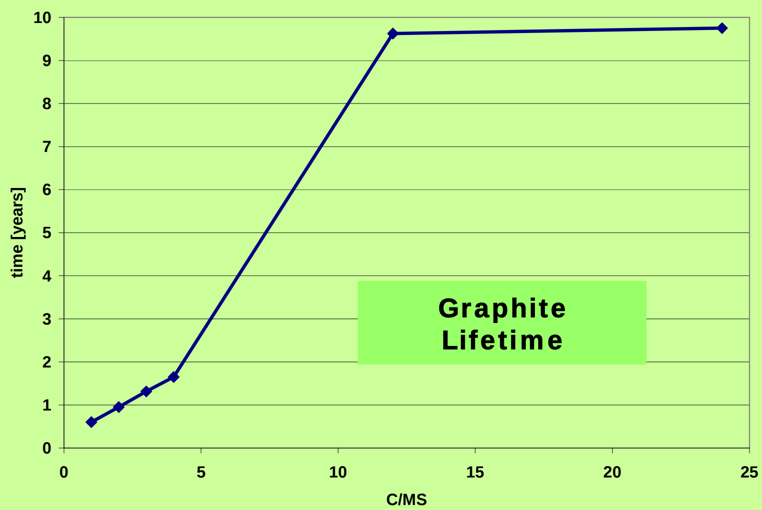
Characteristics	MS Feed Rate [l/day-GW_{th}]			
	0.133	0.267	0.533	1.067
k_{eff}	1.01869	1.01760	1.02031	1.02351
Graphite Lifetime [years]	1.371	1.371	1.374	1.379
MS Residence Time [years]	105	53	26	13
Transmutation Efficiency	98.6%	97.2%	94.5%	89.6%

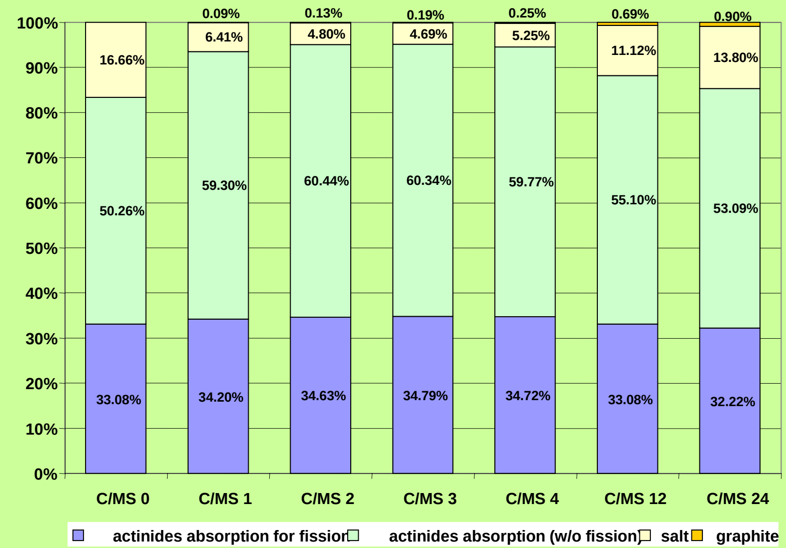
The transmutation efficiency strongly depends on MS feed/extraction rate

2. Spectrum Effect









2. Power Density Effect

C/MS=3, power density=39 W/cm^3

Characteristics	Feed Rate [l/day-GW$_{th}$]			
	0.133	0.267	0.533	1.067
K$_{eff}$	**0.95481**	**0.95443**	**0.95514**	**0.96159**
[vs k$_{eff}$ for 390 W/cm^3]	*1.01869*	*1.01760*	*1.02031*	*1.02351*
Graphite Lifetime [years]	**13.881**	**13.882**	**13.887**	**13.908**
Transmutation Efficiency	**98.55%**	**97.15%**	**94.37%**	**89.47%**

$$\sum_j \sigma_{j\rightarrow i}\frac{\phi}{2}N_j + \sum_j \lambda_{j\rightarrow i} N_j - \lambda_i N_i - \sigma_i \frac{\phi}{2} N_i + F_i - R\, N_i = 0$$

$$^{238}\mathrm{Np} \xrightarrow[\text{2.12days}]{} {}^{238}\mathrm{Pu} \qquad {}^{244}\mathrm{Am} \xrightarrow[\text{10hours}]{} {}^{244}\mathrm{Cm}$$

Actinide	Cross Section [barns]	
	Capture	Fission
^{238}Np	11	105
^{238}Pu	21	2
^{244}Am	36	150
^{244}Cm	18	0.6

3. Waste

Radio-toxicity

defines radiation related hazard

Decay Heat

defines the concentration limit of actinides in the repository

^{237}Np and precursors

long term radiation hazard due to leakage out of repository

Fissile Pu

Proliferation hazard

Methodology: Exponential Matrix to follow decay

$$\frac{dN_i(t)}{dt} = \sum_j \lambda_{j\rightarrow i} N_j(t) - \lambda_i N_i(t)$$

$$\frac{d\vec{N}(t)}{dt} = A\vec{N}(t)$$

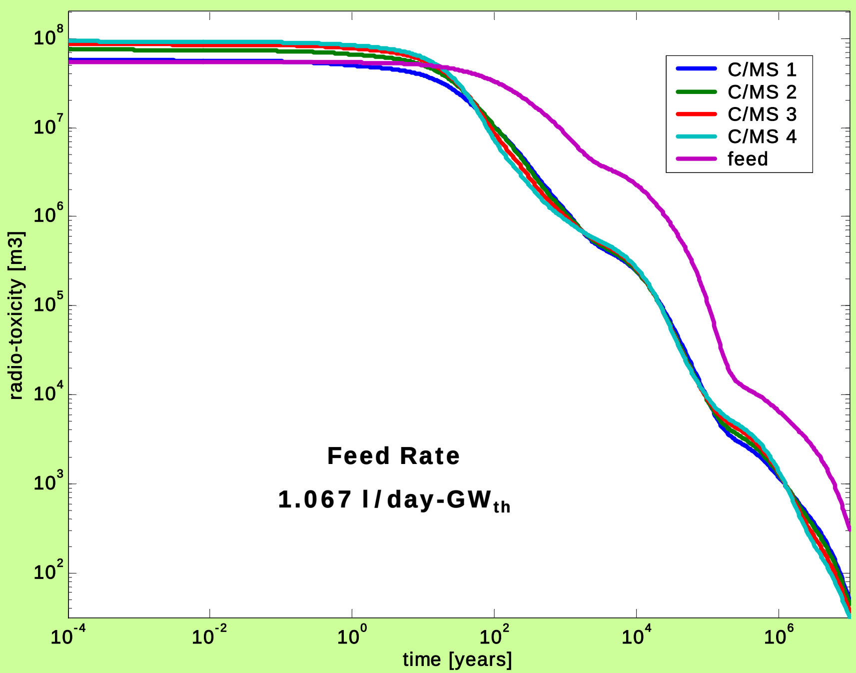
$$\vec{N}(t) = \vec{N}_{t=0} e^{At}$$

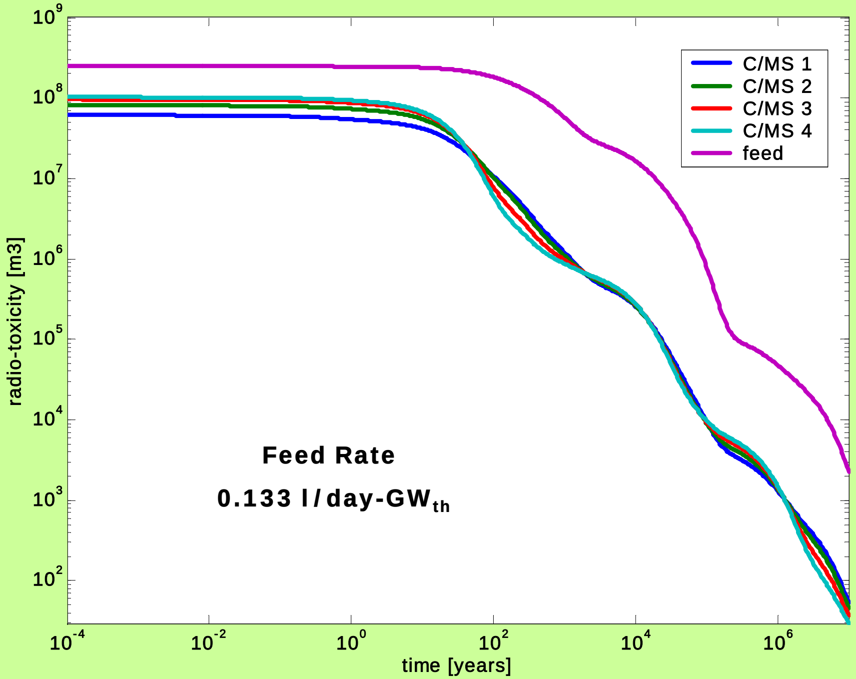
3. Radio-toxicity

Ingestion Toxicity Index

the volume of water with which the mixture of radio nuclides must be diluted so that drinking the water will result in accumulation of radiation dose at a rate no greater than 0.5 rem/year

$$TI = \sum_i \frac{\lambda_i N_i}{C_i}$$

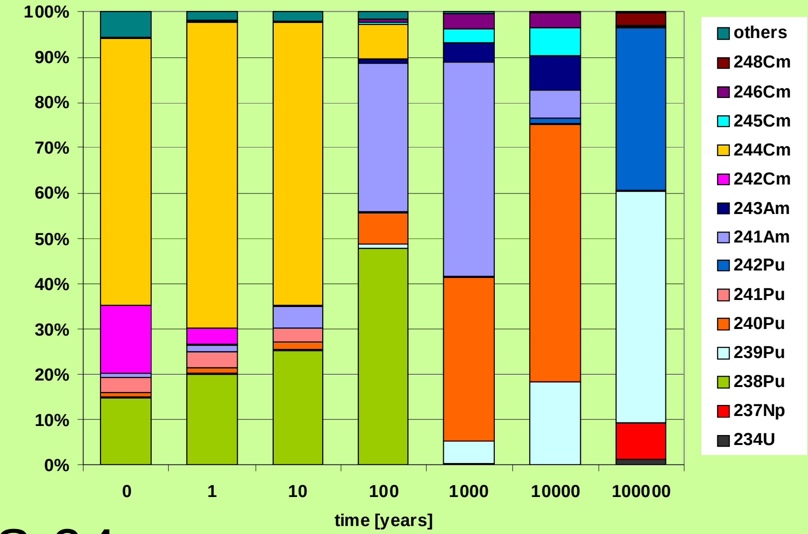




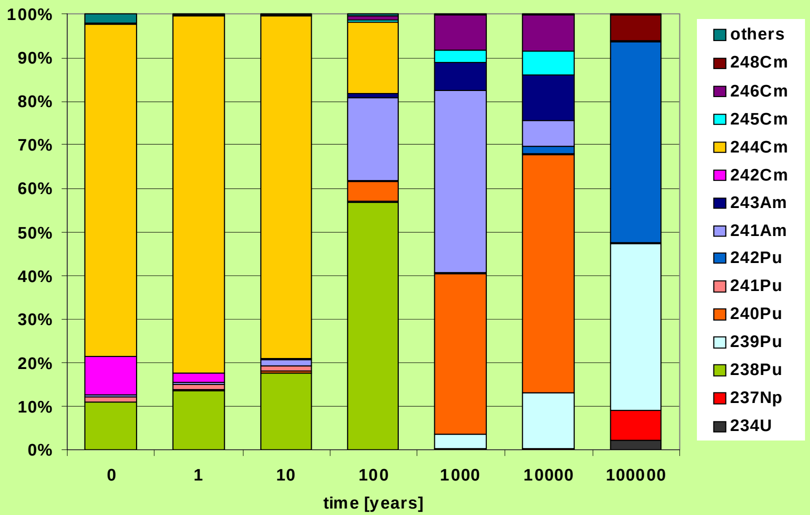
3. Radio-toxicity

Fractional Contribution to radio- toxicity

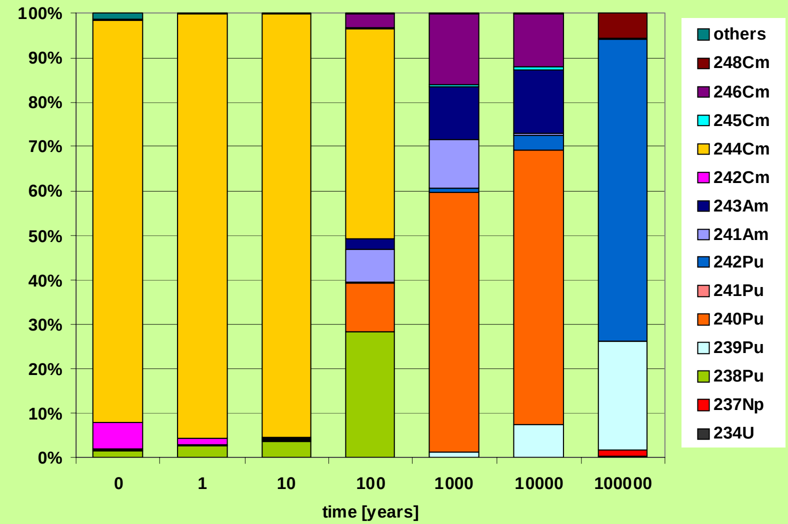
C/MS 0



C/MS 3



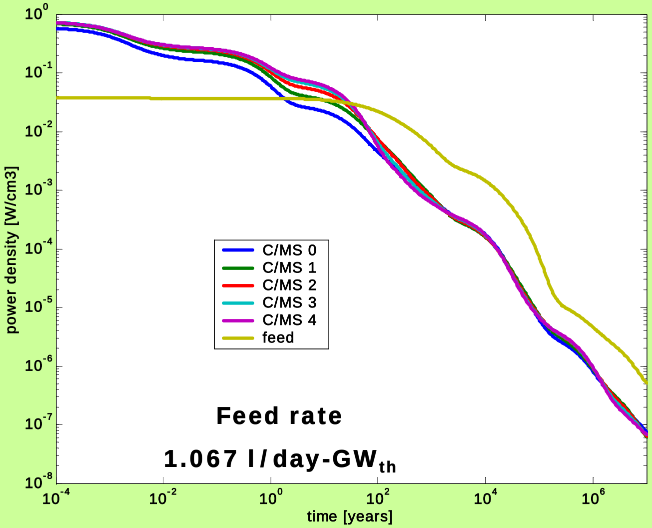
C/MS 24



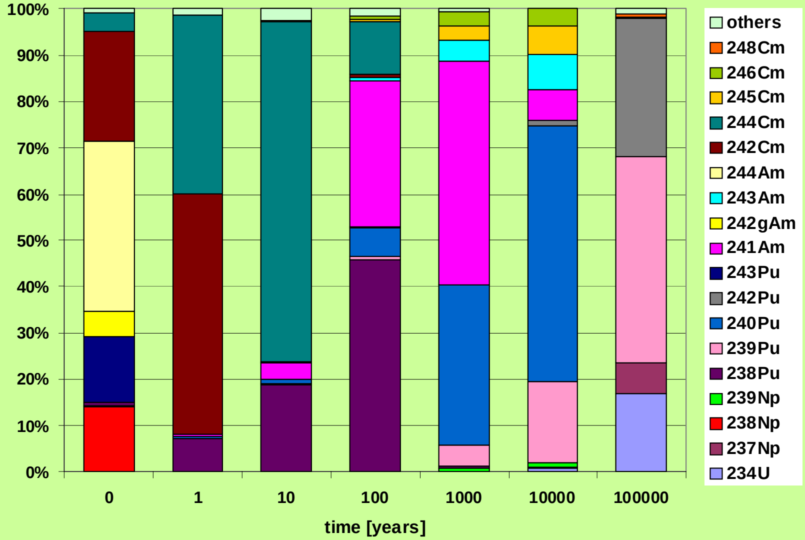
3. Decay Heat

Power Density
[W/cm³ salt]

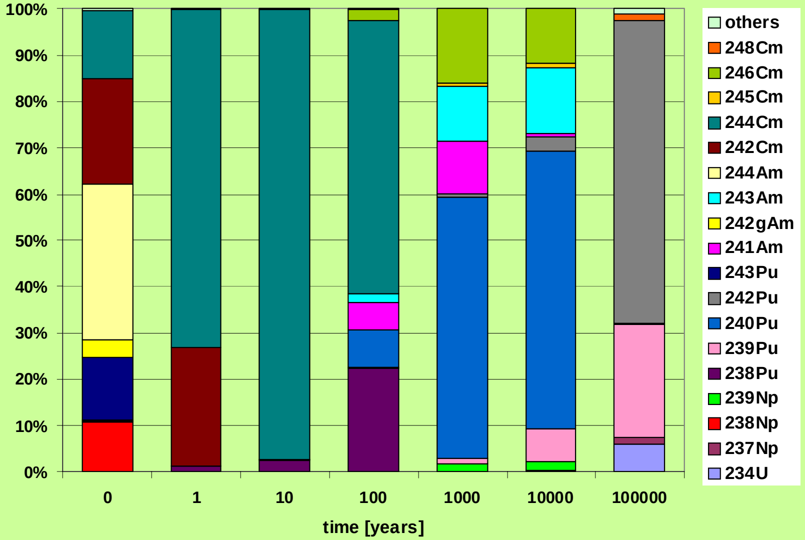
$$p = \sum_i E_i \lambda_i N_i$$



C/MS 0



C/MS 24



3. ^{237}Np & Pu

^{237}Np ($T_{1/2}$ 2.144·10^6 years) and precursors:

$$Cf^{249} \xrightarrow[351y]{} Cm^{245} \xrightarrow[8500y]{} Pu^{241} \xrightarrow[14.35y]{} Am^{241} \xrightarrow[432.2y]{} Np^{237}$$

	Feed	C/MS = 0	C/MS = 3	C/MS = 24
^{237}Np and precursors	1.0	0.105	0.090	0.024
FT ^{239}Pu	-	96.64 %	98.21 %	99.65 %
FT ^{241}Pu	-	83.49 %	87.19 %	97.46 %
(^{239}Pu + ^{241}Pu)/Pu	64.46 %	35.42 %	28.87 %	7.14 %

Softer spectrum is preferred

4. Systems Comparison

Objective: Compare the transmutation effectiveness dependence on transmuting reactor spectrum

Model: Unit cell continuously fed with TRU from LWR SF and TRU is continuously removed to establish equilibrium concentration

Constraints: - same total fractional transmutation within the same residence time
- fuel dimensions and linear heat rate (power density) as of reference reactor

Systems:

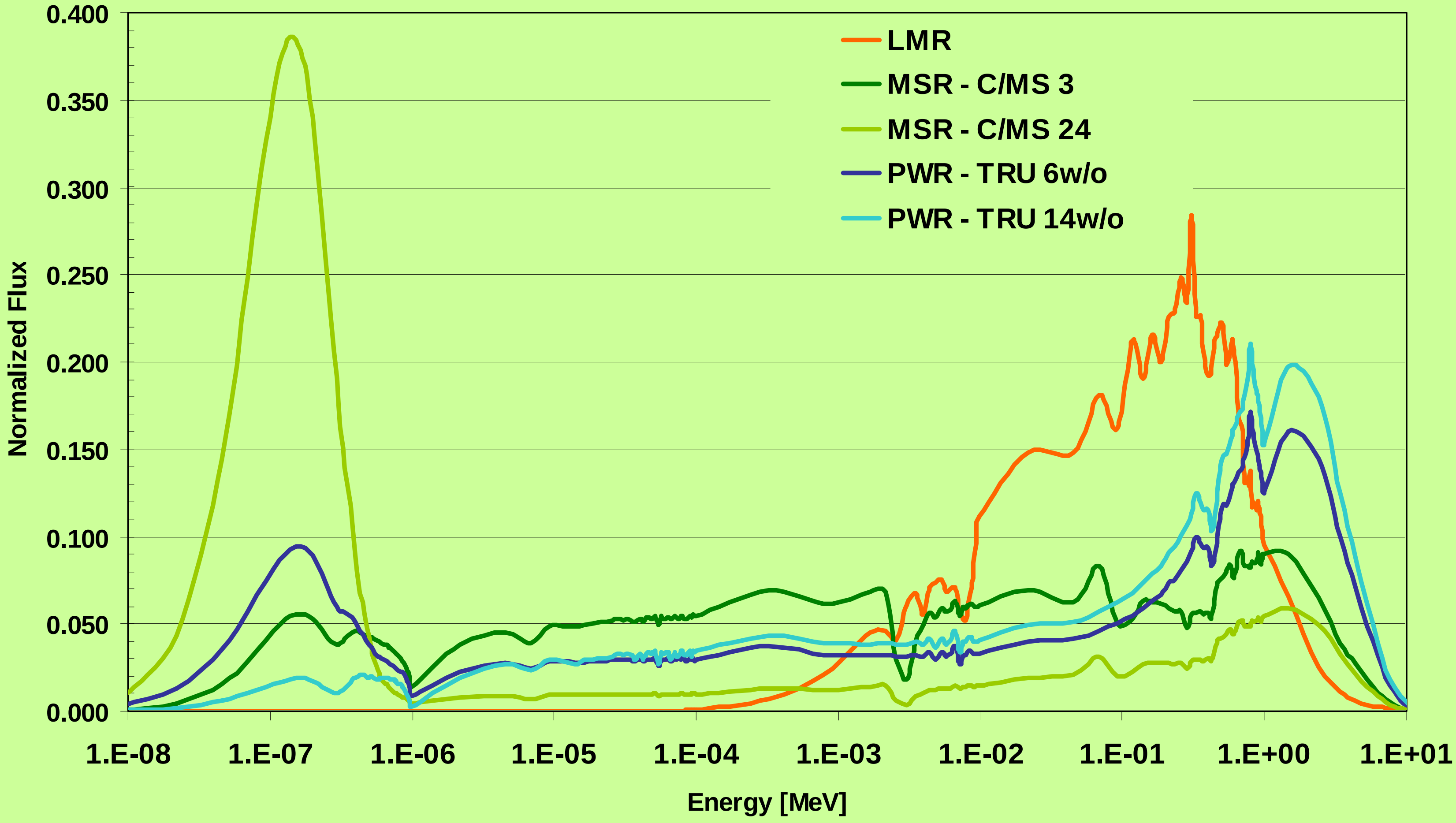
1. Liquid Metal Reactor, Pb-Bi cooled (TRU/Zr fuel)
2. MSR with epithermal spectrum (TRU fluorides)
3. MSR with thermal spectrum (TRU fluorides)
4. PWR with hard spectrum (TRU oxide – 14w/o + ZrO_2)
5. PWR with soft spectrum (TRU oxide – 6w/o + ZrO_2)

4. Reactors Characteristics

Characteristic	LMR	MSR C/MS 3	MSR C/MS 24	PWR TRU 6 w/o	PWR TRU 14 w/o
k_{eff} (equilibrium)	1.039	1.009	0.670	0.962	1.025
Fast flux [n/cm^2-s]	1.08×10^{16}	2.04×10^{15}	1.63×10^{15}	5.59×10^{14}	4.41×10^{14}
Power density [W/cc] *	181.7	97.50	15.60	110.19	110.19
Specific power [W/g]	766	3435	3403	821	329
Total FT*	99.53%	99.53%	99.53%	99.53%	99.53%
Actinides in core at equil. [atoms/cm^3]	5.94×10^{20}	2.82×10^{20}	2.84×10^{20}	1.00×10^{21}	2.52×10^{21}

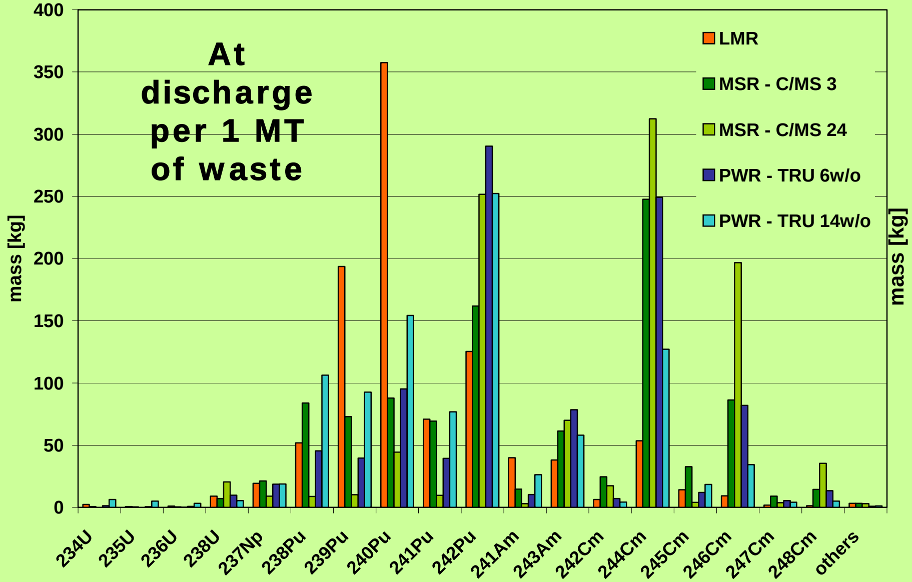
* imposed

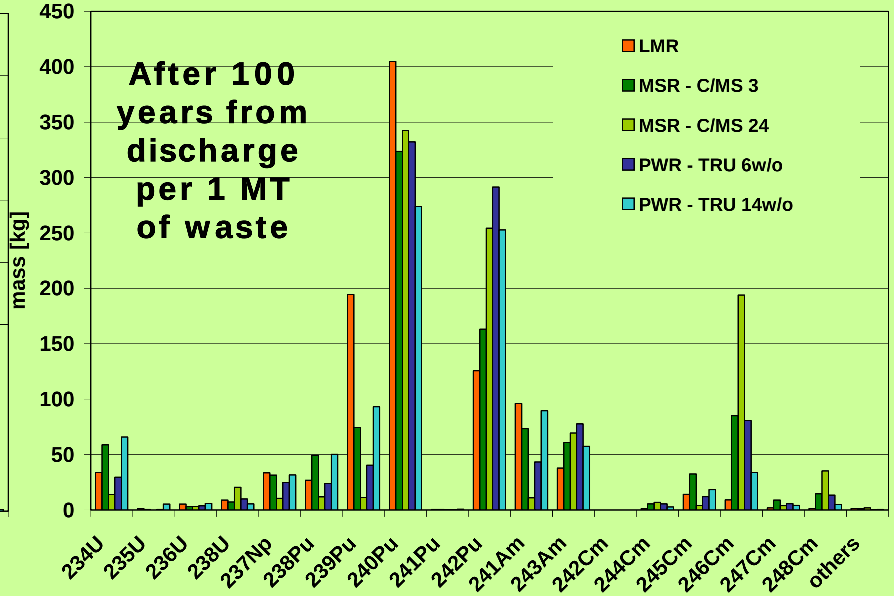
4. Spectra

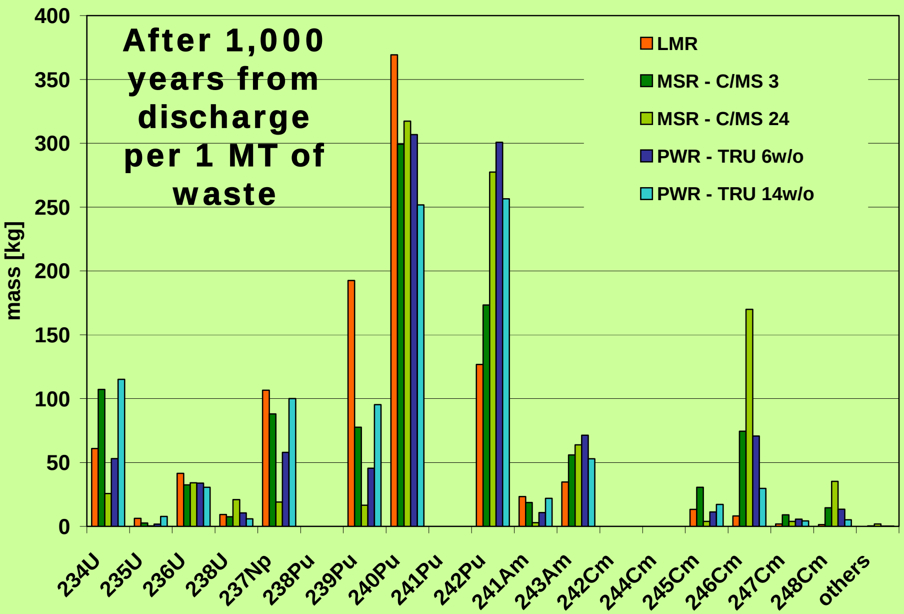


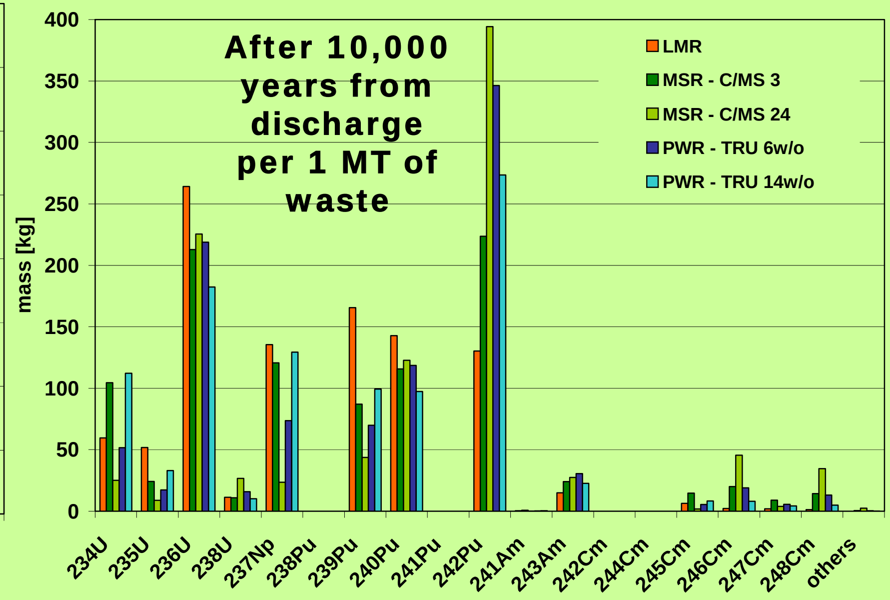
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
Normalized Flux
0.400
0.350
0.300
0.250
0.200
0.150
0.100
0.050
0.000
1.E-08
1.E-07
1.E-06
1.E-05
1.E-04
1.E-03
1.E-02
1.E-01
1.E+00
1.E+01
Energy [MeV]

4. Fractional Transmutation

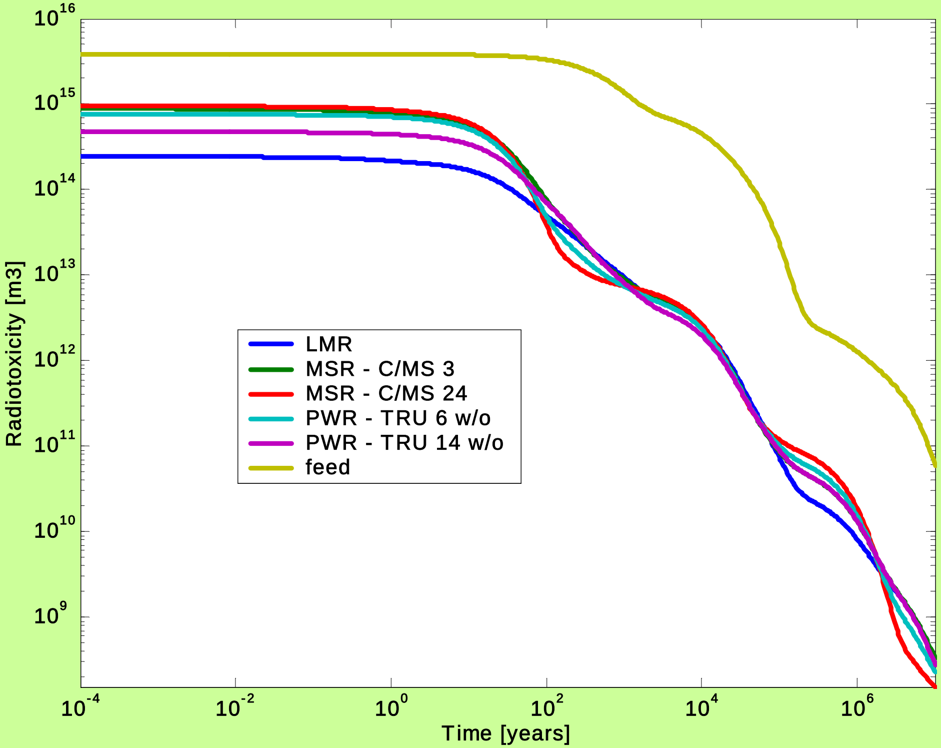




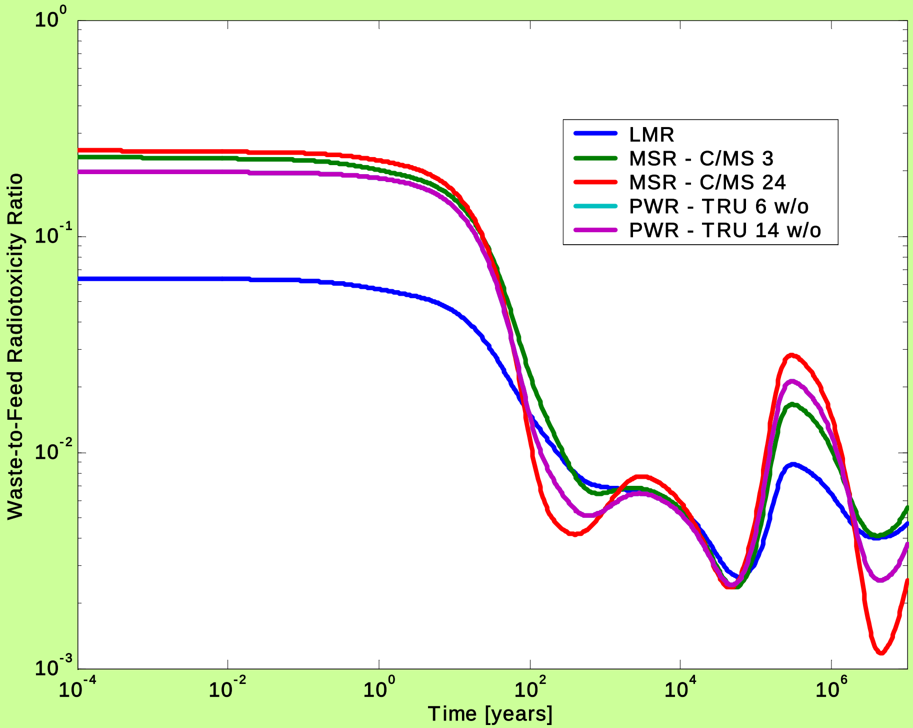




4. Toxicity

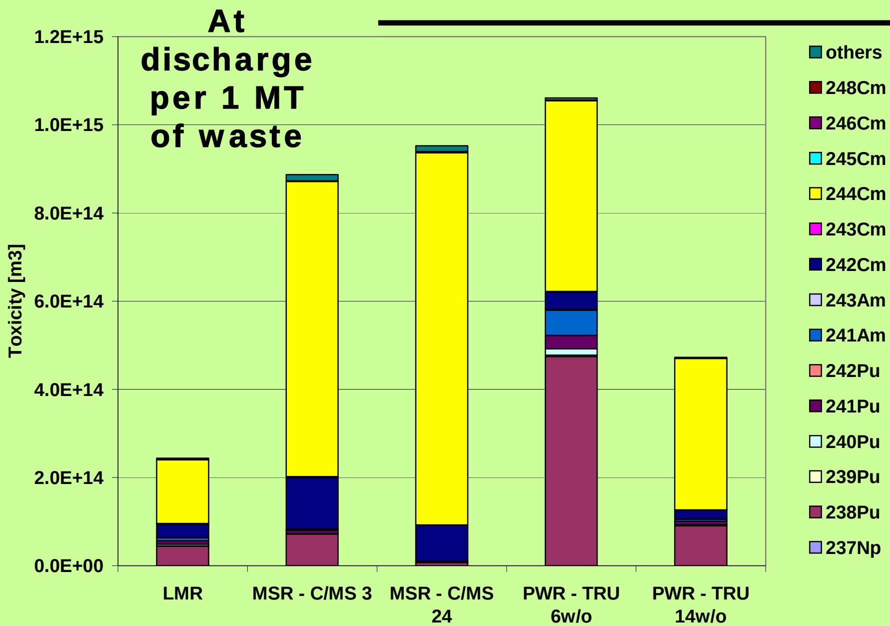


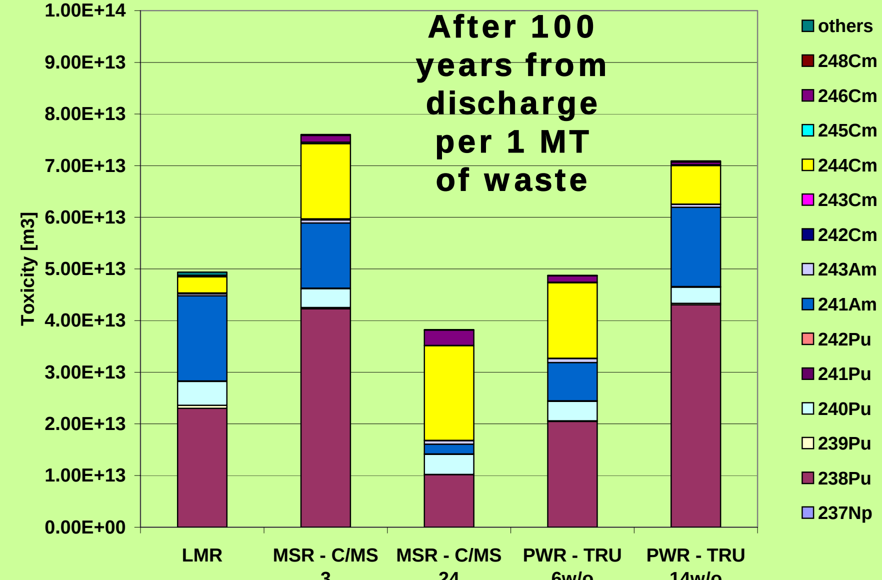
Radiotoxicity [m3]
Time [years]
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6 w/o
PWR - TRU 14 w/o
feed

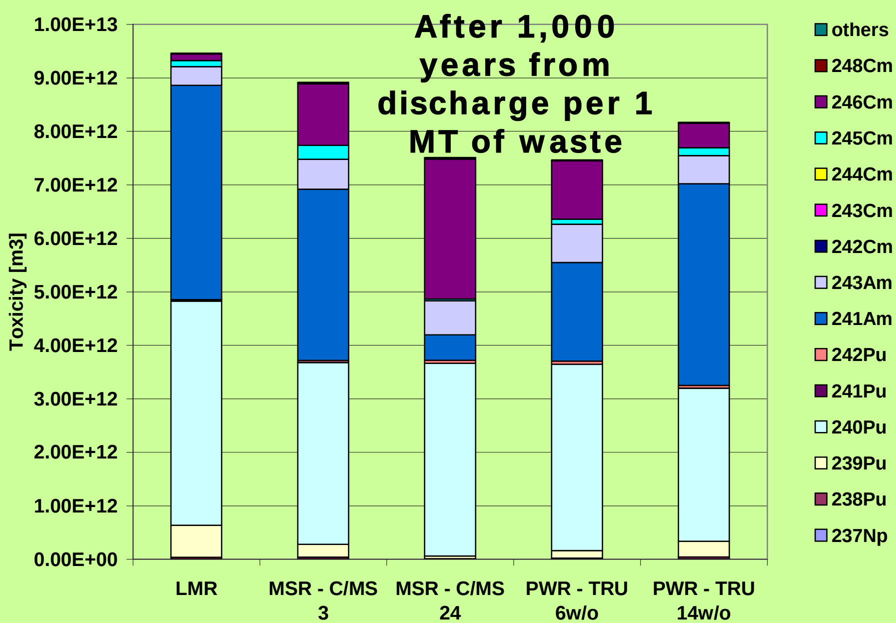


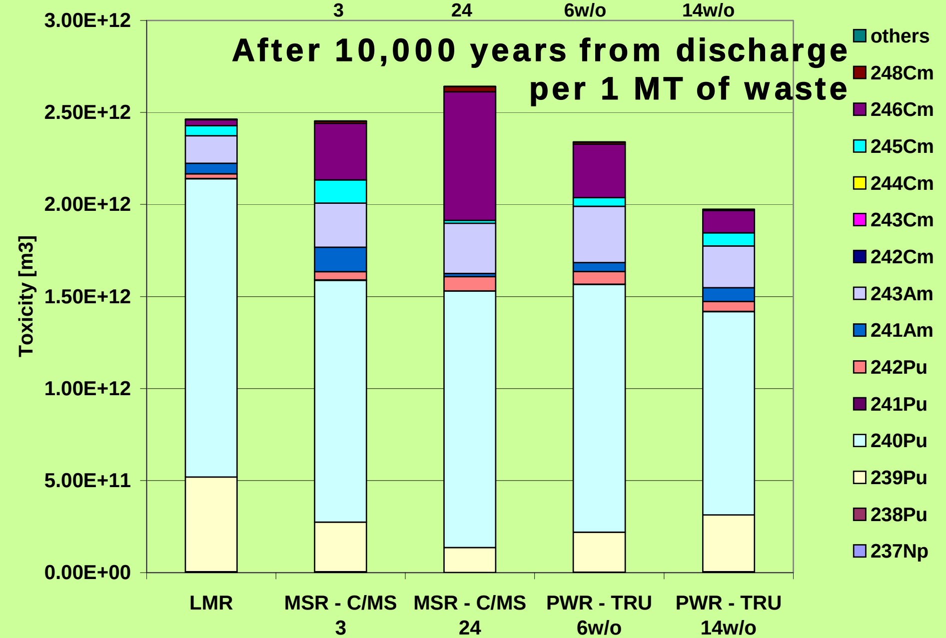
Waste-to-Feed Radiotoxicity Ratio
Time [years]
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6 w/o
PWR - TRU 14 w/o

4. Toxicity

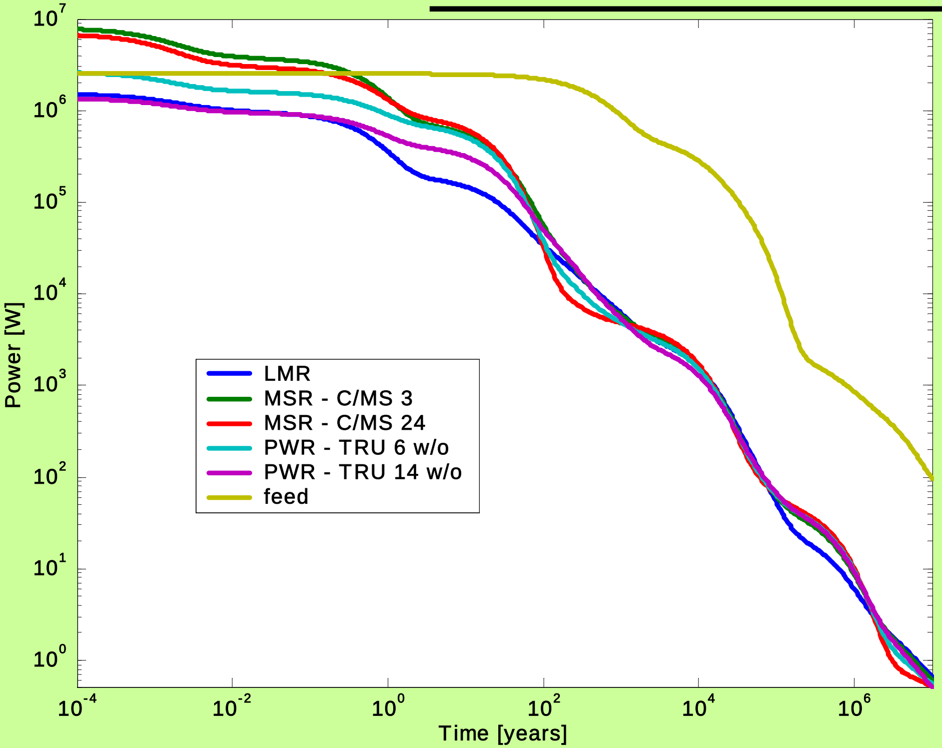

At discharge per 1 MT of waste
Toxicity [m3]
1.2E+15
1.0E+15
8.0E+14
6.0E+14
4.0E+14
2.0E+14
0.0E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
245Cm
244Cm
243Cm
242Cm
243Am
241Am
242Pu
241Pu
240Pu
239Pu
238Pu
237Np


After 100 years from discharge per 1 MT of waste
Toxicity [m3]
1.00E+14
9.00E+13
8.00E+13
7.00E+13
6.00E+13
5.00E+13
4.00E+13
3.00E+13
2.00E+13
1.00E+13
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
245Cm
244Cm
243Cm
242Cm
243Am
241Am
242Pu
241Pu
240Pu
239Pu
238Pu
237Np

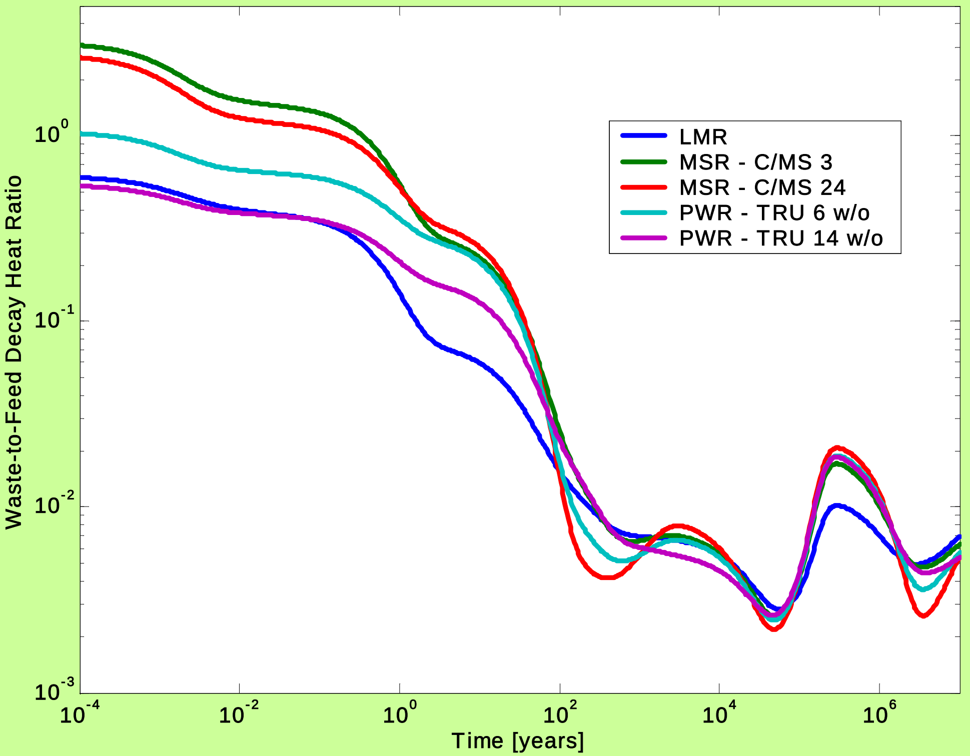

After 1,000 years from discharge per 1 MT of waste
Toxicity [m3]
1.00E+13
9.00E+12
8.00E+12
7.00E+12
6.00E+12
5.00E+12
4.00E+12
3.00E+12
2.00E+12
1.00E+12
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
245Cm
244Cm
243Cm
242Cm
243Am
241Am
242Pu
241Pu
240Pu
239Pu
238Pu
237Np


After 10,000 years from discharge per 1 MT of waste
Toxicity [m3]
3.00E+12
2.50E+12
2.00E+12
1.50E+12
1.00E+12
5.00E+11
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
245Cm
244Cm
243Cm
242Cm
243Am
241Am
242Pu
241Pu
240Pu
239Pu
238Pu
237Np

4. Decay Heat

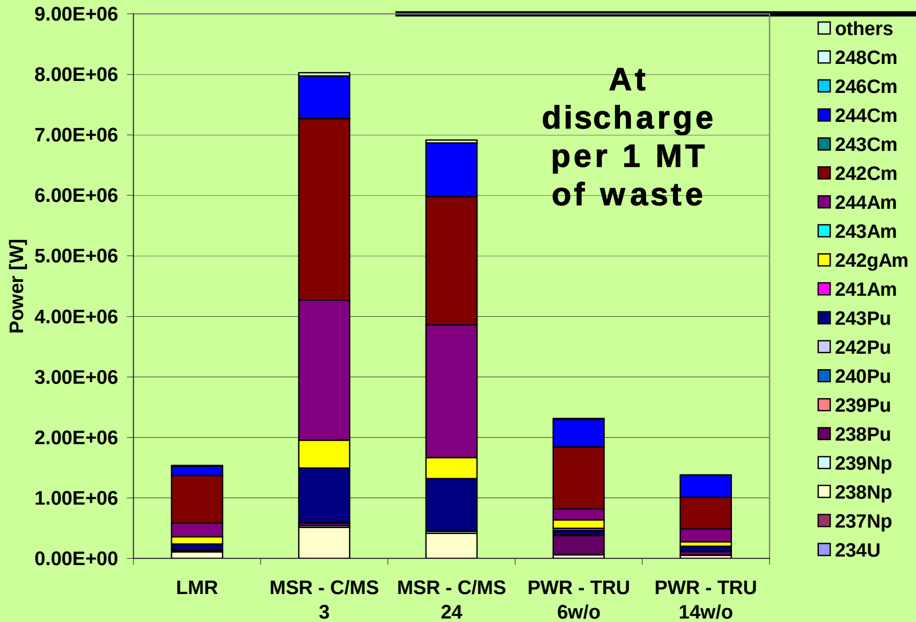


Power [W]
Time [years]
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6 w/o
PWR - TRU 14 w/o
feed

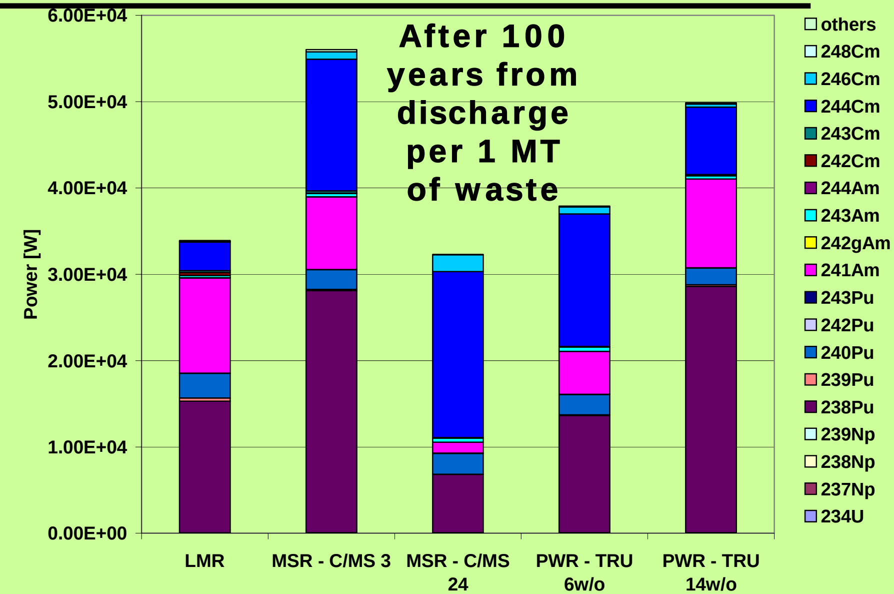


Waste-to-Feed Decay Heat Ratio
Time [years]
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6 w/o
PWR - TRU 14 w/o

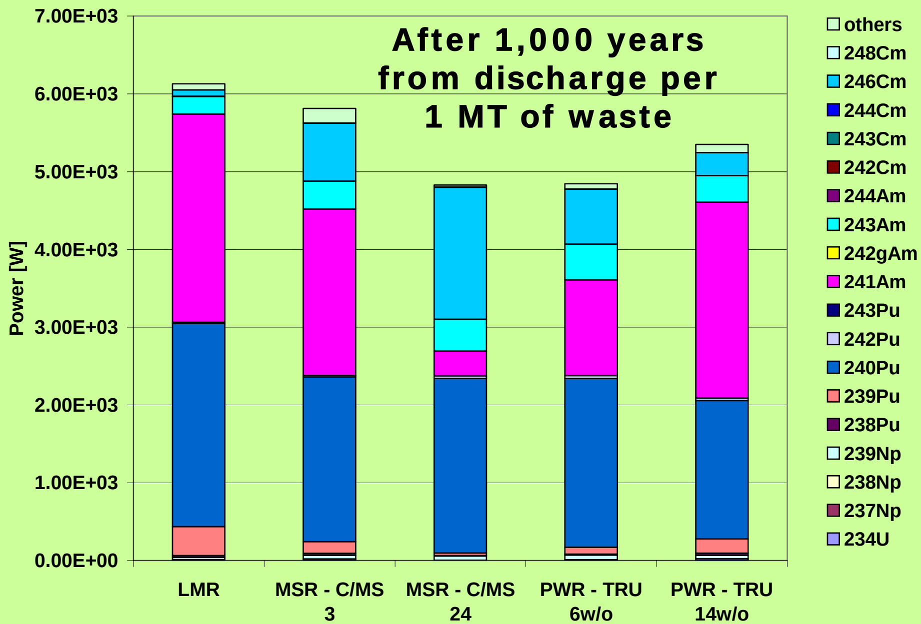
4. Decay Heat



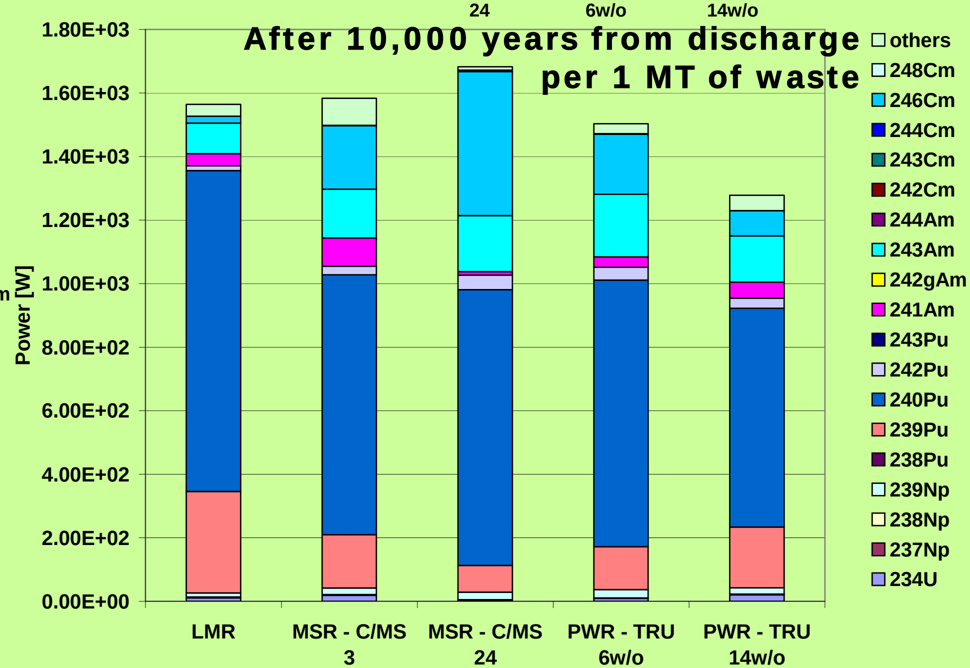
At discharge per 1 MT of waste
Power [W]
9.00E+06
8.00E+06
7.00E+06
6.00E+06
5.00E+06
4.00E+06
3.00E+06
2.00E+06
1.00E+06
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
244Cm
243Cm
242Cm
244Am
243Am
242gAm
241Am
243Pu
242Pu
240Pu
239Pu
238Pu
239Np
238Np
237Np
234U



After 100 years from discharge per 1 MT of waste
Power [W]
6.00E+04
5.00E+04
4.00E+04
3.00E+04
2.00E+04
1.00E+04
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
244Cm
243Cm
242Cm
244Am
243Am
242gAm
241Am
243Pu
242Pu
240Pu
239Pu
238Pu
239Np
238Np
237Np
234U



After 1,000 years from discharge per 1 MT of waste
Power [W]
7.00E+03
6.00E+03
5.00E+03
4.00E+03
3.00E+03
2.00E+03
1.00E+03
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
244Cm
243Cm
242Cm
244Am
243Am
242gAm
241Am
243Pu
242Pu
240Pu
239Pu
238Pu
239Np
238Np
237Np
234U

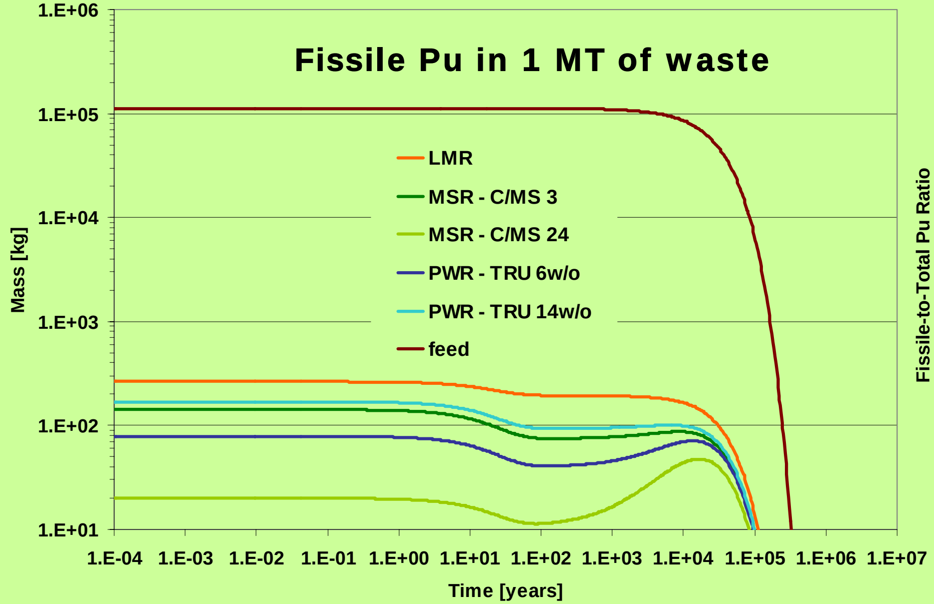


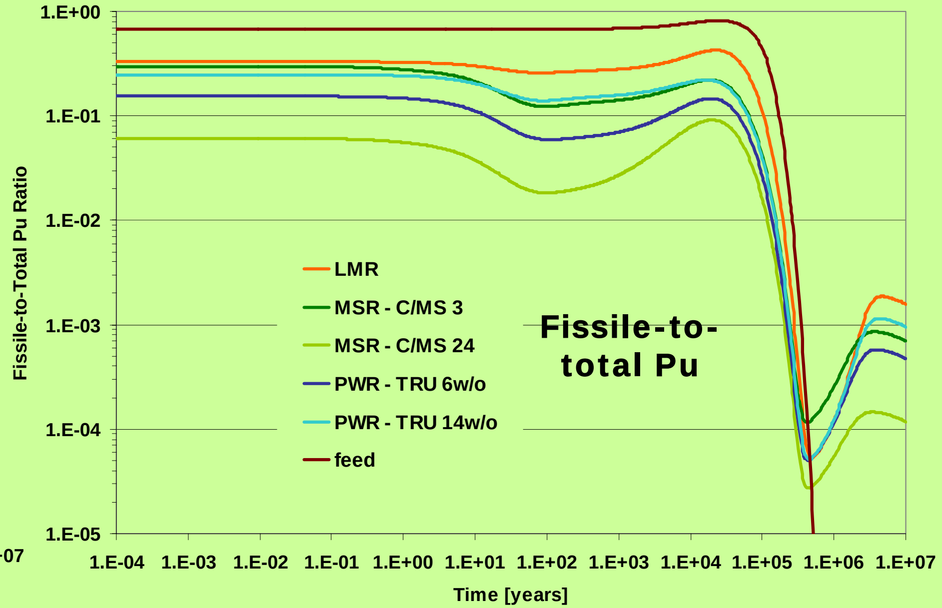
After 10,000 years from discharge per 1 MT of waste
Power [W]
1.80E+03
1.60E+03
1.40E+03
1.20E+03
1.00E+03
8.00E+02
6.00E+02
4.00E+02
2.00E+02
0.00E+00
LMR
MSR - C/MS 3
MSR - C/MS 24
PWR - TRU 6w/o
PWR - TRU 14w/o
others
248Cm
246Cm
244Cm
243Cm
242Cm
244Am
243Am
242gAm
241Am
243Pu
242Pu
240Pu
239Pu
238Pu
239Np
238Np
237Np
234U

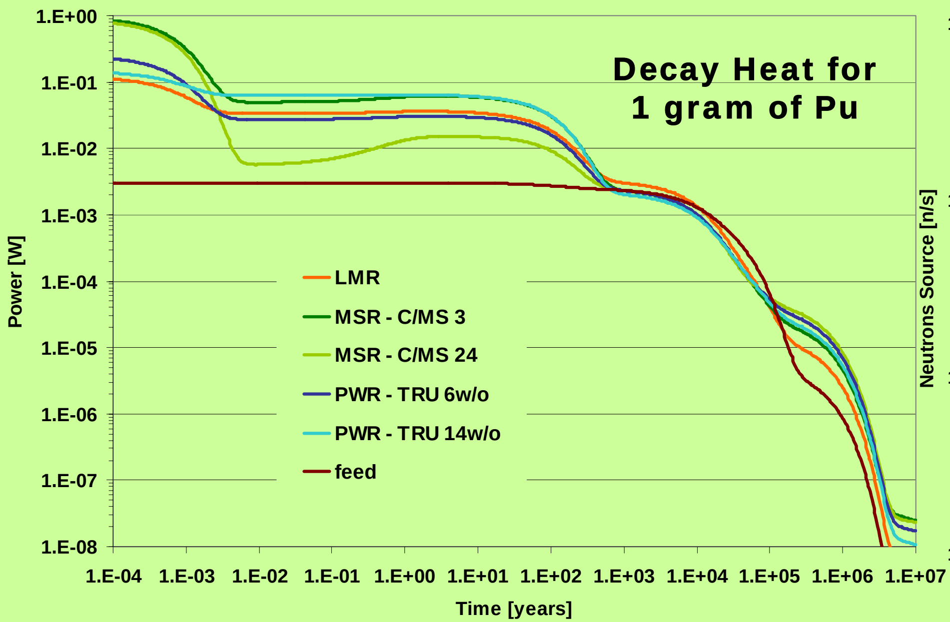
4. Np Inventory

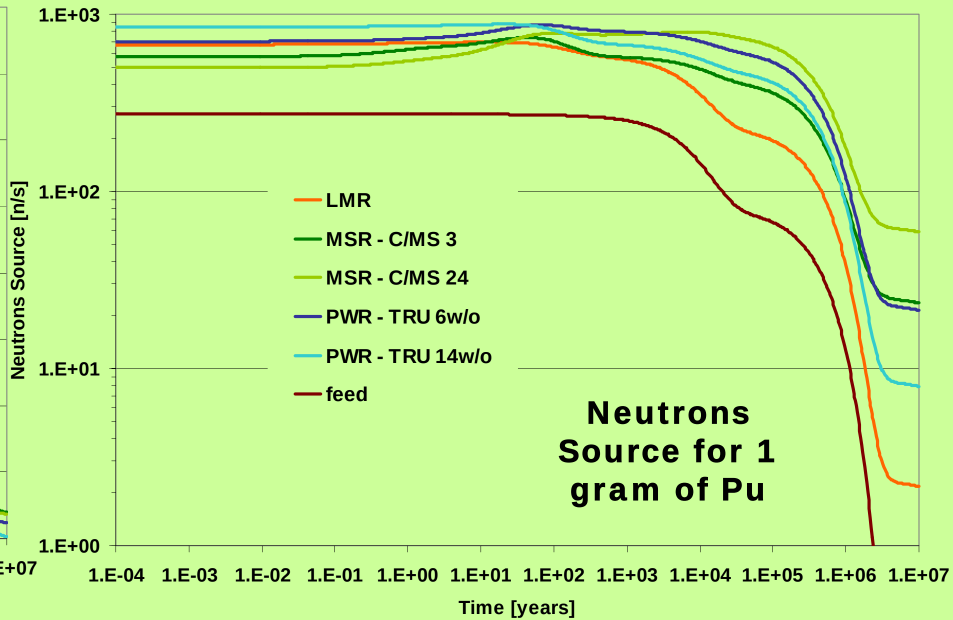
Actinide	LMR	MSR C/MS 3	MSR C/MS 24	PWR TRU 6 w/o	PWR TRU 13 w/o	Feed
^{237}Np [kg]	19.42	21.25	8.96	18.73	18.92	10674
^{241}Pu [kg]	71.00	69.30	9.65	39.34	76.79	8051
^{241}Am [kg]	39.86	14.80	3.05	10.36	26.20	19066
^{245}Cm [kg]	14.24	32.71	4.05	12.05	18.37	19
^{249}Cf [kg]	1.35	14.52	35.30	13.38	4.99	0
Total [kg]	145.87	152.58	61.02	93.86	145.27	37810

4. Pu Inventory

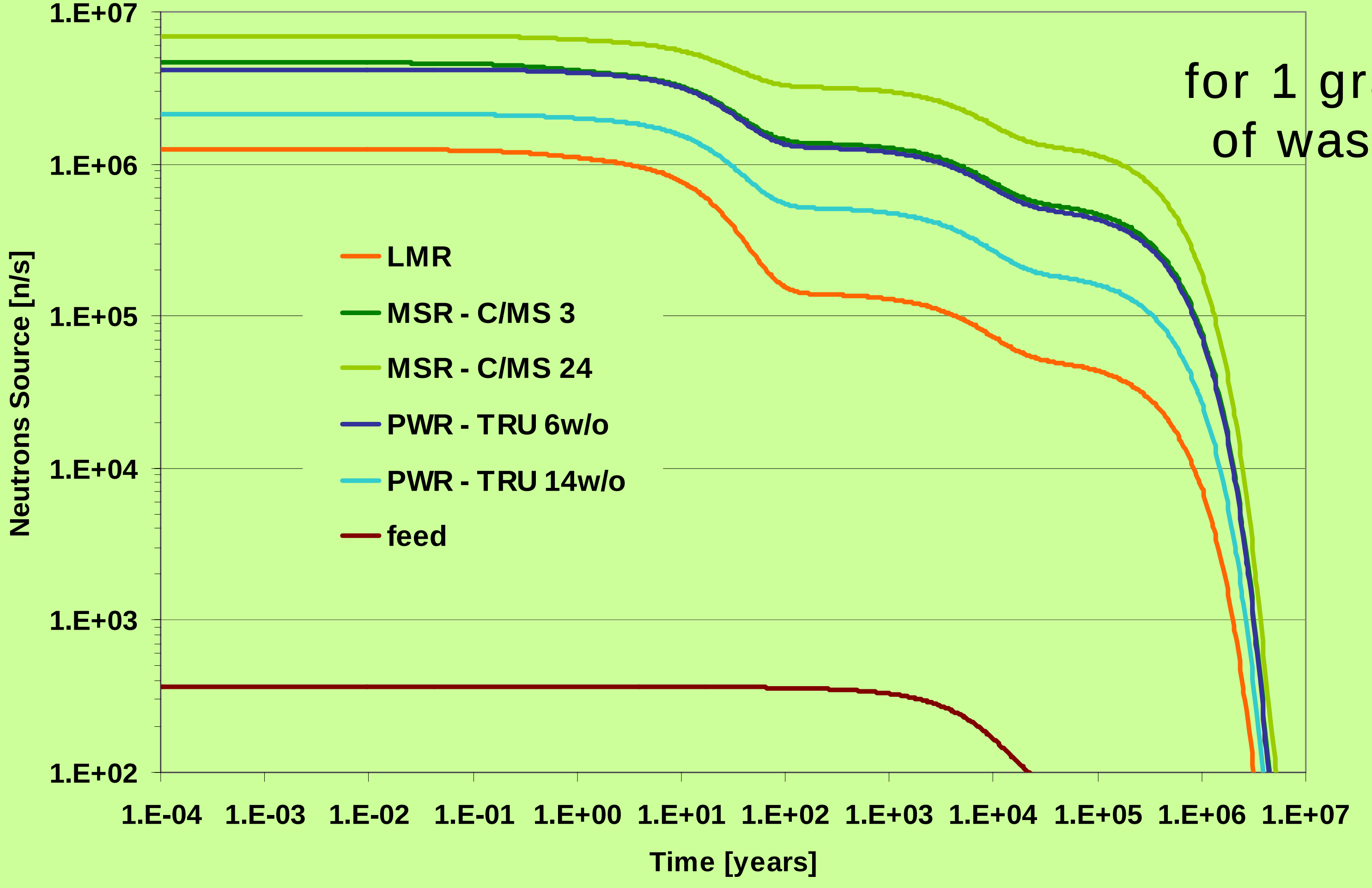








4. Neutrons Source



for 1 gram of waste

Conclusions: MSR Design

1. *It is not possible to design a NaF-ZrF_4 MSR fed by LWR SF TRU to be critical, but very close to critical – k_{eff} ~0.97, provided fission products are continuously extracted*
2. *Maximum k_{eff} is obtained when C/MS = 3.2*
3. *Using graphite reflectors it is possible to make the radiation damage to graphite very uniform*
4. *Decreasing the MS feed rate increases the transmutation efficiency without significantly affecting k_{eff}, provided fission products are continuously extracted*
5. *Power density reduction enhances the graphite lifetime but strongly decreasing k_{eff}*
6. *Softer spectrum is better for reducing long term inventory of ^{237}Np, fissile Pu and Pu(fissile)/Pu ratio*
7. *Hard spectrum is better for reducing initial radio-toxicity and decay heat but after ~100 years of cooling softer spectrum is preferable*

Conclusions: Comparison

Characteristic	Preferred System
Neutron economy	LMR, PWR(14), MSR(3)
Specific power	MSR
Radio-toxicity	LMR $<10^2$ y; MSR(24) $<10^3$ y; LWR(14) $<10^5$ y
Decay-heat	LMR $<10^2$ y; MSR(24) $<10^3$ y; LWR(14) $<10^5$ y
Neutron source (low)	LMR, PWR(14), MSR(3)
^{237}Np & precursors	MSR(24), LWR(6), rest
Fissile Pu	MSR(24), LWR(6), MSR(3)
Fissile/total Pu	MSR(24), LWR(6), MSR(3)
Specific Pu decay heat (low)	MSR(24), LWR(6), LMR
Specific Pu neutron source	MSR(24), MSR(3) <10 y; LMR, MSR(3) >10 y

Discussion

Issues to be considered:

- *Feasibility of criticality using other MS*
- *Graphite replacement strategy*
- *TRU losses during recycling of C & FP*
- *Handling of fission products*
- *Approach to equilibrium*
- *Economics*
- *Technology maturity*