Some impacts of advanced fuel cycle options on waste management and long term disposal safety

Bernd Grambow

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June 7, 2021

Presentation to the National Academies of Sciences, Engineering, and Medicine

Study of the “Merits and Viability of Different Nuclear Fuel Cycles and

Technology Options and the Waste Aspects of Advanced Nuclear Reactors”

Potential positive effects of advanced fuel cycle options on the waste disposal

- Reduction in the radionuclide inventory in the final waste caused by recovery and re-use of fissile isotopes or even by change of burnup
- Reduction in the radiotoxicity of the waste
- Reduction in the volume of waste
- Reduction in the thermal power of the waste
- Increase in the durability of the waste form (HTR)

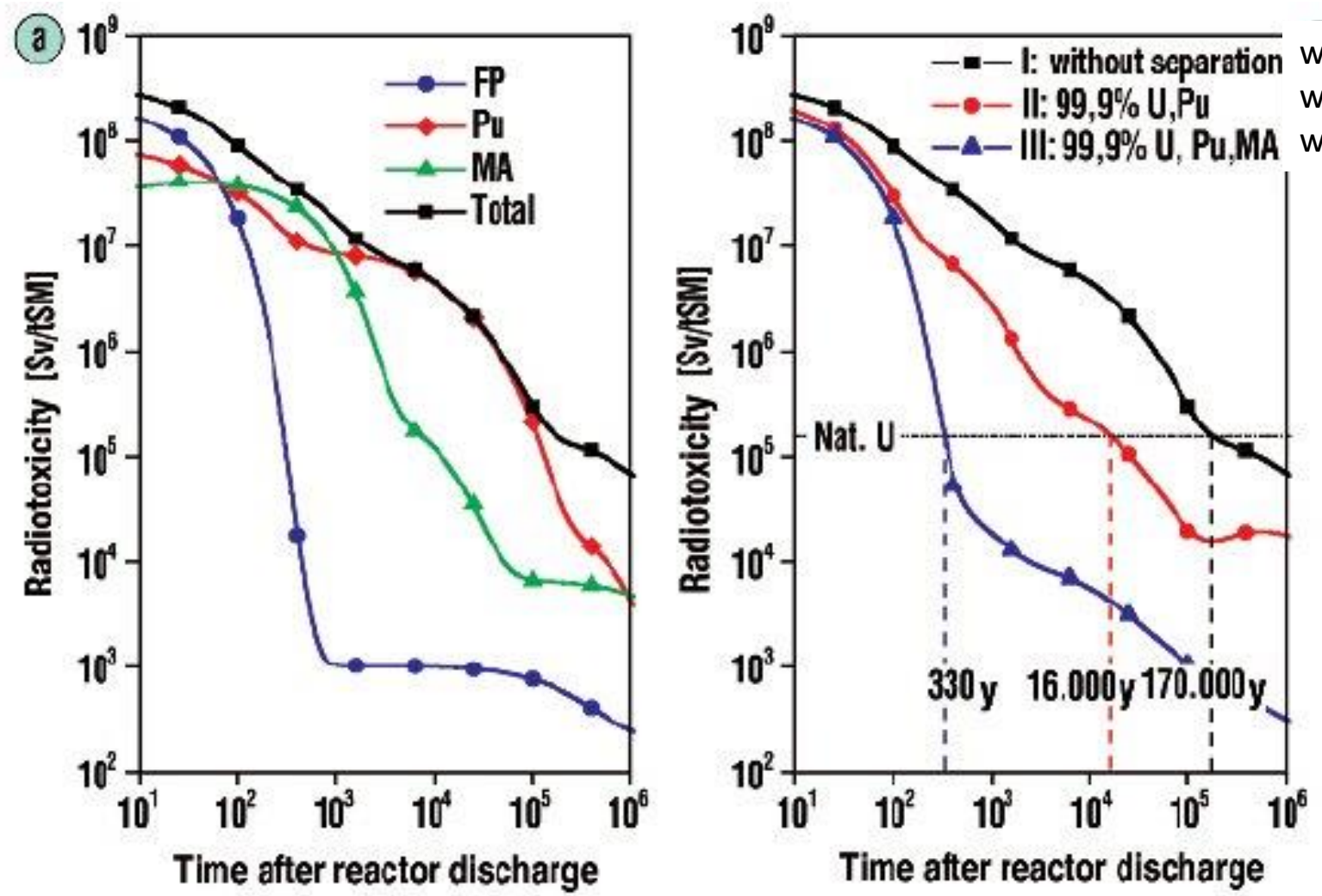
Safety indicators for radioactive waste I

- Radiotoxicity inventory
- Radiotoxicity isolation
- Mobile fraction of radionuclide inventory
- Maximum end-point dose
- Comparison of released concentrations to natural ones: U, Ra…

Safety indicators II

- **Radiotoxicity**
  - Independent on barrier function and exposure scenario
  - No quantification of risk (potential dose)
  - In consequence : valid as indicator only for overall inventory: **Sum of toxicity of waste and of inventories remaining in surface installations**, meaningless only for waste
- **Radiotoxicity isolation of many RN until decay**
  - Geochemical and hydraulic barrier functions
- **Mobile fraction of radiotoxicity**
  - Migration to the accessible environment
  - Dose by coupling to exposure scenario

Radiotoxicity evolution of 1 ton PWR fuel



without reprocessing
with reprocessing
with transmutation

Attention: this is only the toxicity of the waste. In case of transmutation a larger fraction of toxicity inventories compared to the current cycle stays with facilities at surface

Limits of ingestion in Europe: 10^{-3} Sv/yr

Hartwig Freiesleben (2013) The European Physical Journal Conferences 54:01006-DOI:0.1051/epjconf/20135401006,

Flow chart of French nuclear fuel cycle (1999-2010) including reprocessing

Average Burnup:
45 GWd/tU
(1999-2010)

exemple d'empilement de galettes de déchets compactés dans un CSD-C

Extraction
Unat
7958 tU/yr
Fluorination
UF6
7918 tU/yr
Enrichment
3,9%
Unat depleted 0.26%
UF6 enriched
978 tU/yr
Fuel (UNE)
fabrication
Unat
depleted
0.26%
UO2 enriched
978 tU/yr
Reactors
400 TWh
4360 tons
URE
Fuel assemblies
37 t U/yr
MOX
Fuel assemblies
98 tHM/yr
Spent fuel
978 tHM/yr
Stock 12000 tons
2018
Interim
storage
Fuel (URE)
fabrication
Fuel (MOX)
fabrication
Depleted U
89.5 tU/yr
Reprocessing
850t HM/yr
UF6
enriched
36,7 tU/yr
URT depleted
0.3%
243 tU/yr
PuO2
8,5 tPu/yr
Pu
8.5 t/yr
Stock 56 tons 2018
Enrichment
4.1%
U (URT)
805 t/yr
20000 tons (2018)
I-129
Cl-36
into ocean
UF6
279 tU/yr
Fluorination
284 tU/yr
Interim storage
Depleted U
6851t U/yr
318000 tons (2018)
Spent URE
37 t HM/yr
Spent MOX
98 t HM/yr
Spent UNE
128 t HM/yr
Cladding+
structures
850 CSD-C/yr (170t/yr)
Waste glass
723 CSD-V/yr (143 m³)
Cemented +
Bitumen waste
U3O8
521 tU /yr

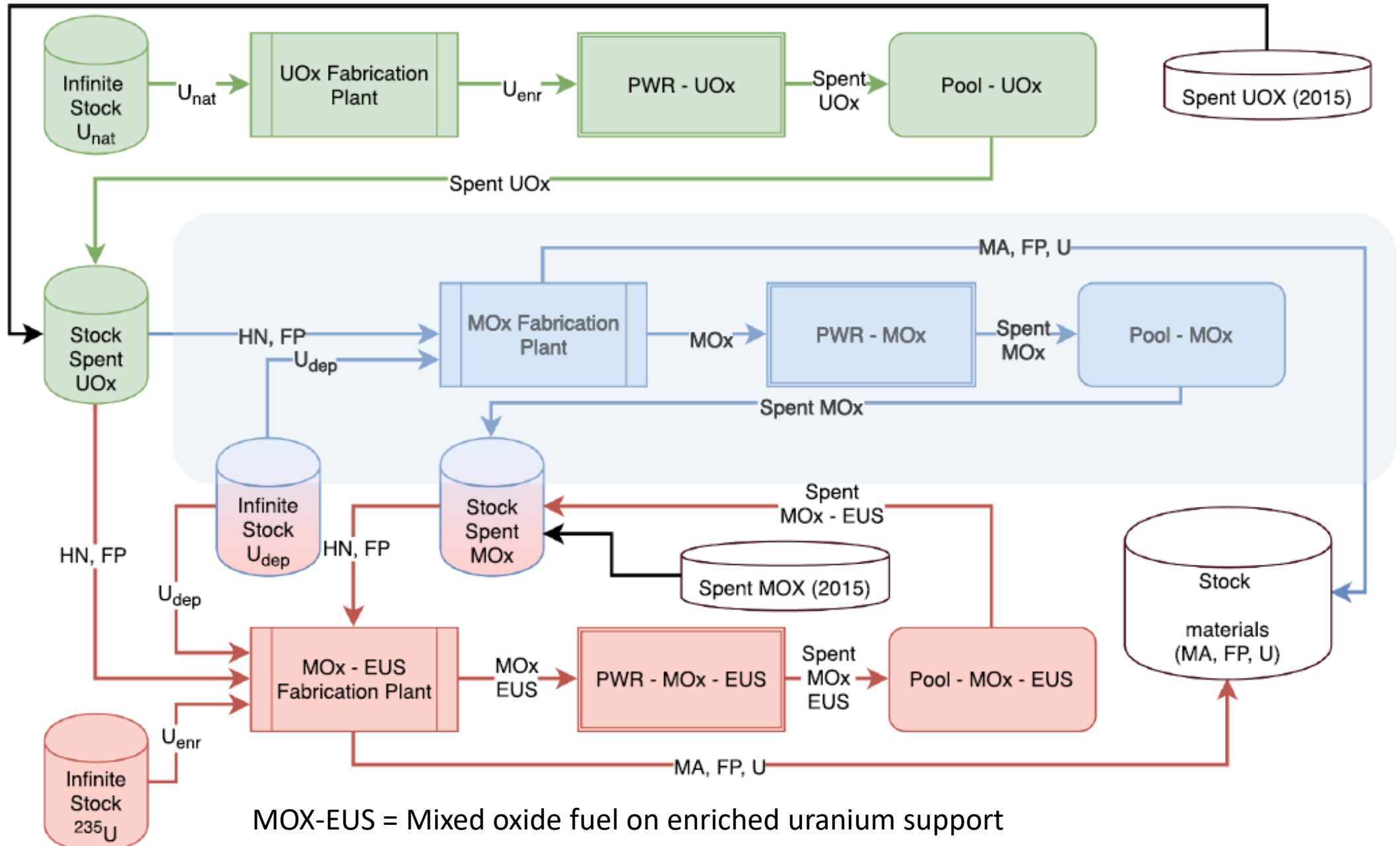
Conditioned waste waiting for disposal

Reprocessing produces various waste streams

Assessment of plutonium inventory management in the French nuclear fleet with the fuel cycle simulator CLASS

(recent article by Courtin, Thiollière et al., SUBATECH) https://doi.org/10.1016/j.nucengdes.2020.111042

- Current French policy:
  - Pu is a resource that is used to operate PWR and that may be used later to load SFR.
  - In 2015, 22 PWRs 900 are loaded with around 30% of MOX fuel
  - Ultimate waste would be Minor Actinides (MA) and Fission Products (FP)
- However, the ASTRID French SFR project to demonstrate the feasibility of the Pu multi-recycling in a fast spectrum was abandoned by CEA.
  - SFR will not become economically viable in the next decades
  - Need to avoid pile-up of MOX fuel, containing low grade Pu that cannot be transformed in new MOX
- Alternatives for Pu management: Pu multi-recycling in PWR
  - Pu can be stabilized within 30% of PWR using multi-reprocessed plutonium in MOX (8-13% Pu) on enriched uranium (1-4%) fuel, the rest being composed of PWR loaded with UOX.
  - Transuranic (Pu+MA) stabilization involves Pu incineration. Around 50% of PWR using multi-reprocessed Pu is required and the nuclear power has to decrease.



MOX-EUS = Mixed oxide fuel on enriched uranium support

U235 input would compensate for its net consumption in fissile isotopes

Stabilization of Pu and MA inventories

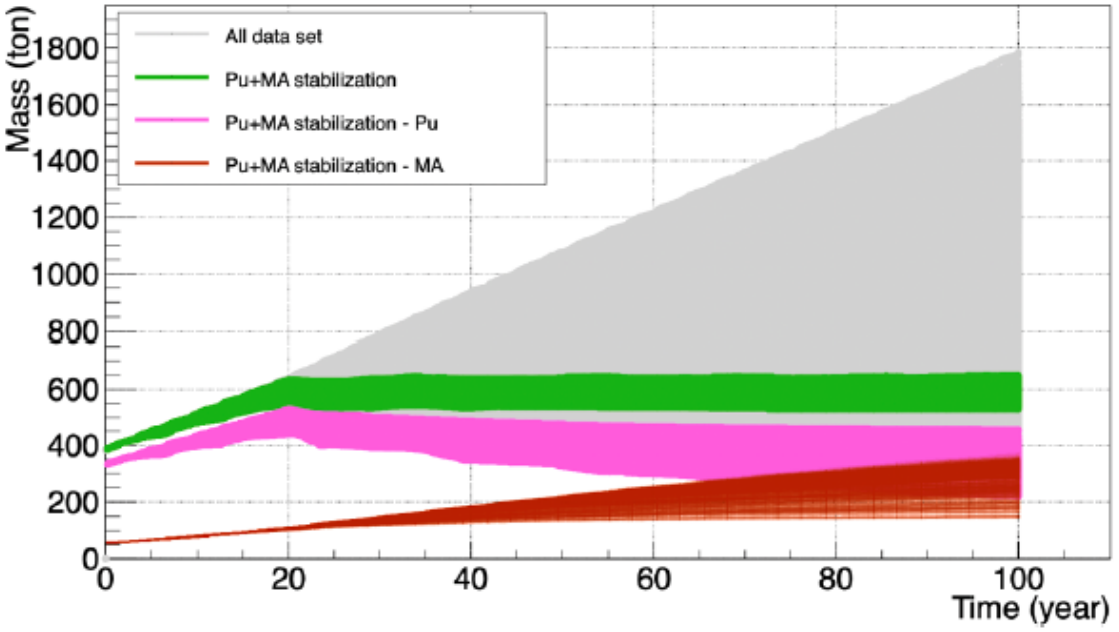


Fig. 14. Total transuranic mass evolution in the fleet (gray lines). The green lines represent the sum of plutonium and minor actinides masses for trajectories obtained by stabilization criteria. Purple lines are plutonium mass evolution and red lines are minor actinides mass evolution after the application of the same criteria.

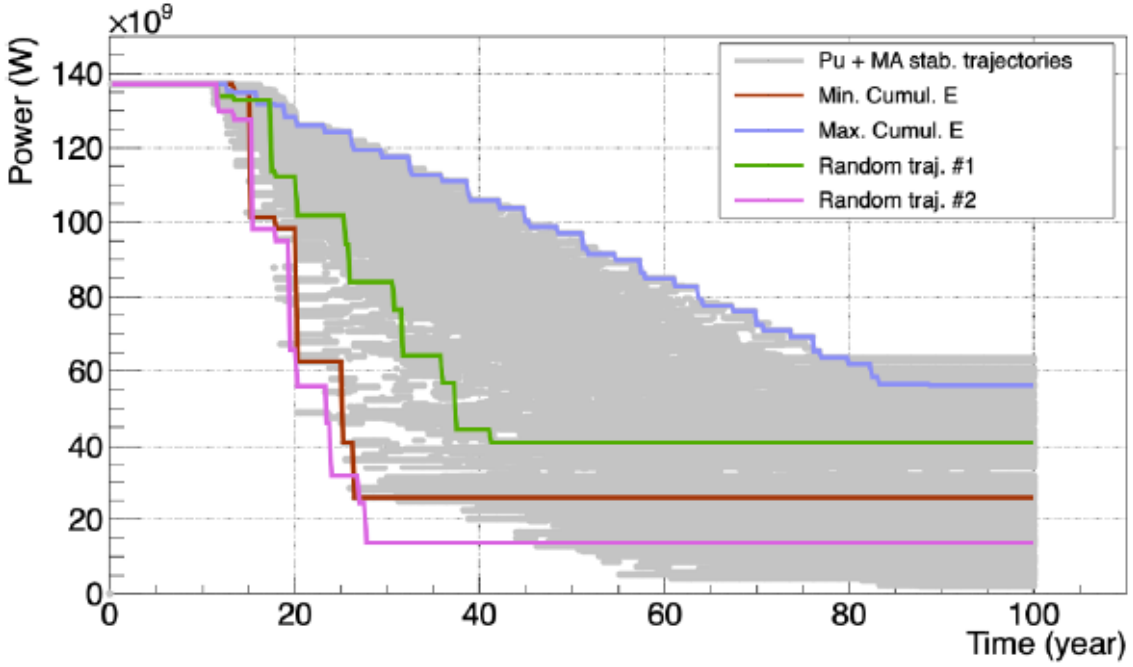


Fig. 15. Thermal power evolution for the FiFo data set obtained from the application of plutonium stabilization criteria. Gray lines represent the whole data set. The red line (resp. blue line) is the trajectory that involves the smallest (resp. highest) value of ΔE_{th}. Two random examples (green and pink lines) have been added.

P_0 = 137.13GWth (total installed thermal power 188.11GWth, load factor of 0.729)
Fuel fabrication 2 years, Pu separation 100%. Reactor $k_{threshold}$ 1.037 with 4 fuel loadings.
The MOX fraction, the BU and the MOX spent fuel cooling time are respectively 10%, 45 GWd/t and 8 years.

Challenge of fuel cycle options: toxicity reduction

Example: French reactor fleet

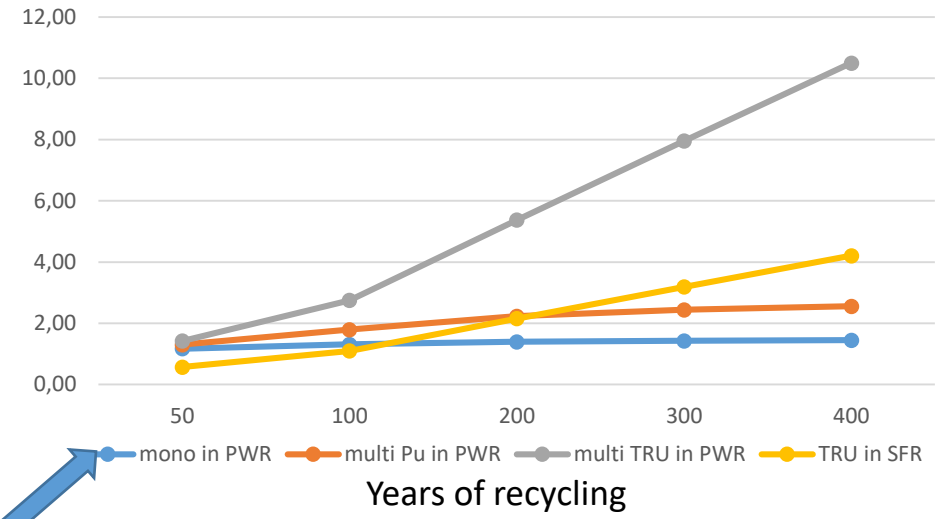
influenced by burnup

reactors	PWR (400 TWh)				SFR
fuel cycle concept	open cycle	mono reprocessing	multi reprocessing		
		MOX	only of Pu	of all TRU	of all TRU
	permanent inventory (tons) in surface installations (reactors, processing…)				
Pu	35	150	220	300	800
Np	2,4	6	5,3	13	4
Am	1	4	14	34	32
Cm	0,5	2	7,2	47	8
	mass of TRU in final waste (kg/yr)				
Pu	10500	7000	17	23	57
Np	740	760	660	1	0,3
Am	290	740	1800	2,6	2,5
Cm	150	370	900	3,6	0,6
	gain in reducing waste radiotoxicity compared to open cycle				
1000 yr	1	1,2	3	390	210
10000 yr	1	1,5	3	350	150
	total inventory (tons) after 100 yr				
Pu	1085	850	221,7	302,3	805,7
Np	76,4	82	71,3	13,1	4,03
Am	30	78	194	34,26	32,25
Cm	15,5	39	97,2	47,36	8,06
	gain reducing total radiotoxicity accumulated over 200 yr compared to open cycle				
1000 yr	1	1,05	1,79	3,07	1,54
10000 yr	1	1,31	1,79	2,75	1,10
	indiv. dose mSv/yr for 63 GWe installed (conversion UNSCAER 2016),				
surface installations	8,19E-02	1,20E-01	1,29E-01	1,29E-01	1,29E-01
deterministic	dose > 0.1 -1 Myr France (Andra Dossier 2005) / Switzerland (P. Zuidima)				
disposal (maximum dose)	1E-5	1E-9 to 1E-5	similar to mono reprocessing		

source: CLEFS CEA N°53 (2005-6)

Radiotoxicity

Factor of reduction of remaining total radiotoxicity (surface installations and cumulated waste) after 10000 yr as a function of fuel recycling option and recycling time compared to open cycle=1



Radiological risk

Likely a negative (but weak) overall impact as the number of surface installations increases and the contribution of disposal risk to overall risk remains negligible:

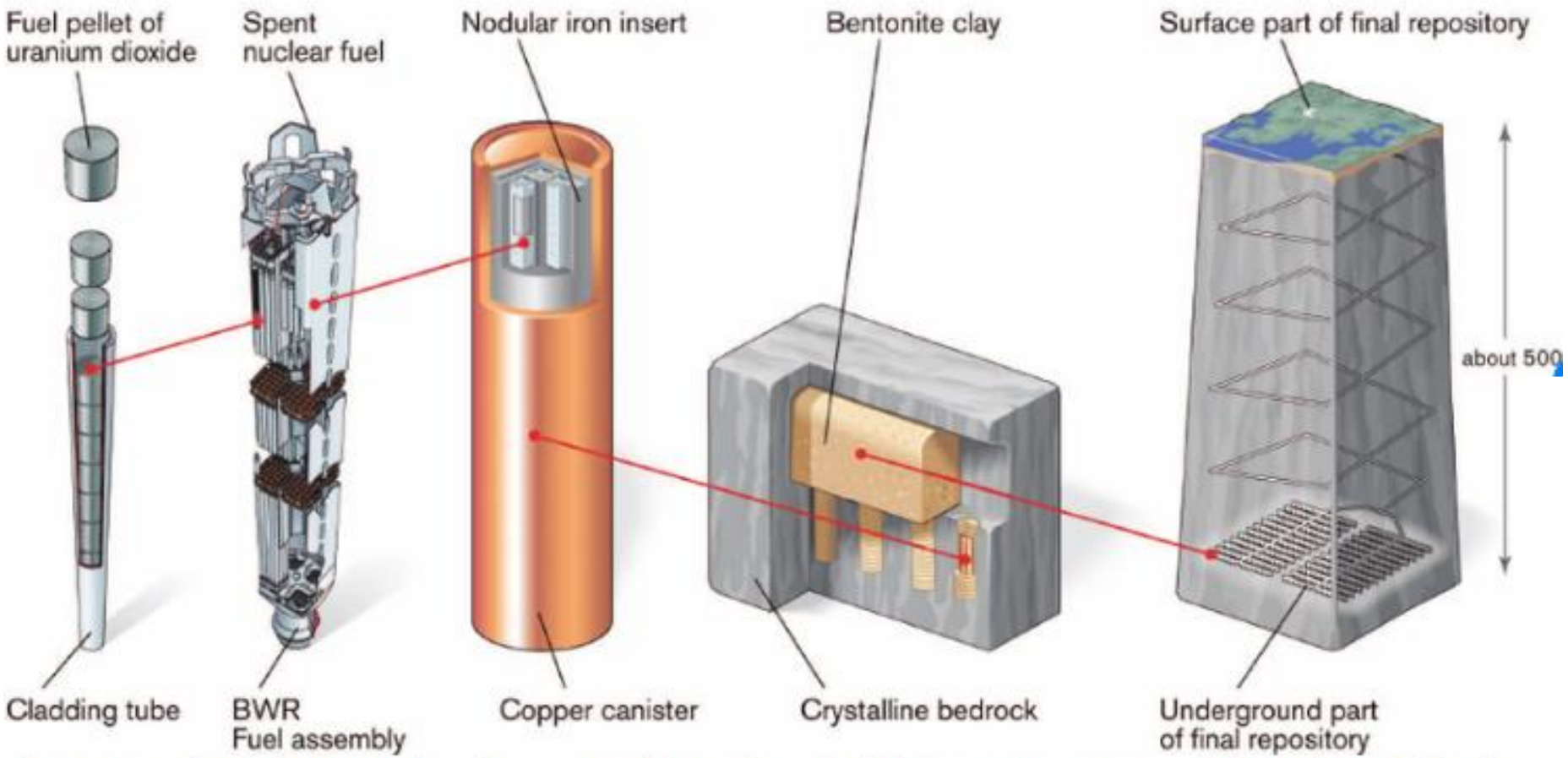
Geological disposal is a radioprotection measure: it reduces overall radiological risk

Reflections on multi-reprocessing

- 1) The multi-reprocessing will allow stabilizing the inventories of Pu in the surface installations (reactors + reprocessing…) at a rather high level (about 10 times more of TRU in the cycle than today) and one can produce a HLW, containing no Pu and only MA+FP (in reality the separation is not 100% but 99.8% for Pu)
- 2) However, the total toxicity of the sum of the accumulated waste and the permanent inventories in the reactors and other fuel cycle installations decrease over 100 yr at maximum by a factor of 3.
- 3) This is very little, considering that the radiological risk of exposure is not proportional to this small reduction and may actually increase due to the increased number of surface installations.
- 4) In France the orientation is to integrate the fuel cycle in the increase of more renewable energies. This will reduce the flux of nuclear fuels and increase the need to operate the fuel cycle very flexible. This need of flexibility must be integrated into multi-reprocessing schemes.

Alternative for example: the Swedish concept KBS 3 for direct disposal of spent nuclear fuel without reprocessing

lFrom: P. Selline et al. clays and Clay Minerals 61(6):477-498 · March 2014



The KBS-3 method. The method involves encapsulating the spent fuel in copper canisters which are then emplaced, surrounded by a buffer of bentonite clay, in deposition holes in a tunnel system at a depth of 400 À 700 m in the bedrock (SKB, 2011, reproduced with permission). BWR: boiling water reactor.

Impact of radionuclide inventories on disposal risk

- In the absence of human intrusion that directly might expose waste to humans, the long-term risk of the disposal system is not governed by the inventory of the most toxic nuclides but a complex interplay of toxicity and mobility
  - In planned European repositories in reducing clay formations the long-term risk is dominated by the inventory of most mobile nuclides like ^{129}I, ^{79}Se and ^{36}Cl not by immobile Pu, governing toxicity.
    - Change of Pu inventory has no effect on disposal risk
    - Reprocessing and vitrification have a strong effect on risk as only 1% of the inventories of Cl36 and I129 are vitrified, the rest being released to the ocean, reducing risk by isotope dillution
  - In repositories in granite (Sweden, Finland) under reducing conditions, peak dose is governed by more mobile Ra226 rather than by low mobile Pu.
  - Dose estimates for the oxidizing repository in Yucca Mountain show dominant contributions to risk from Pu caused by higher mobility of Pu. But Pu release is governed by solubility constraints and sorption, so inventory reduction will have little or no effect on disposal risk

Geological disposal as radioprotection measure assured by multiple natural and engineered barriers

Sensitivity of analysis on the impact of the barriers in the near field

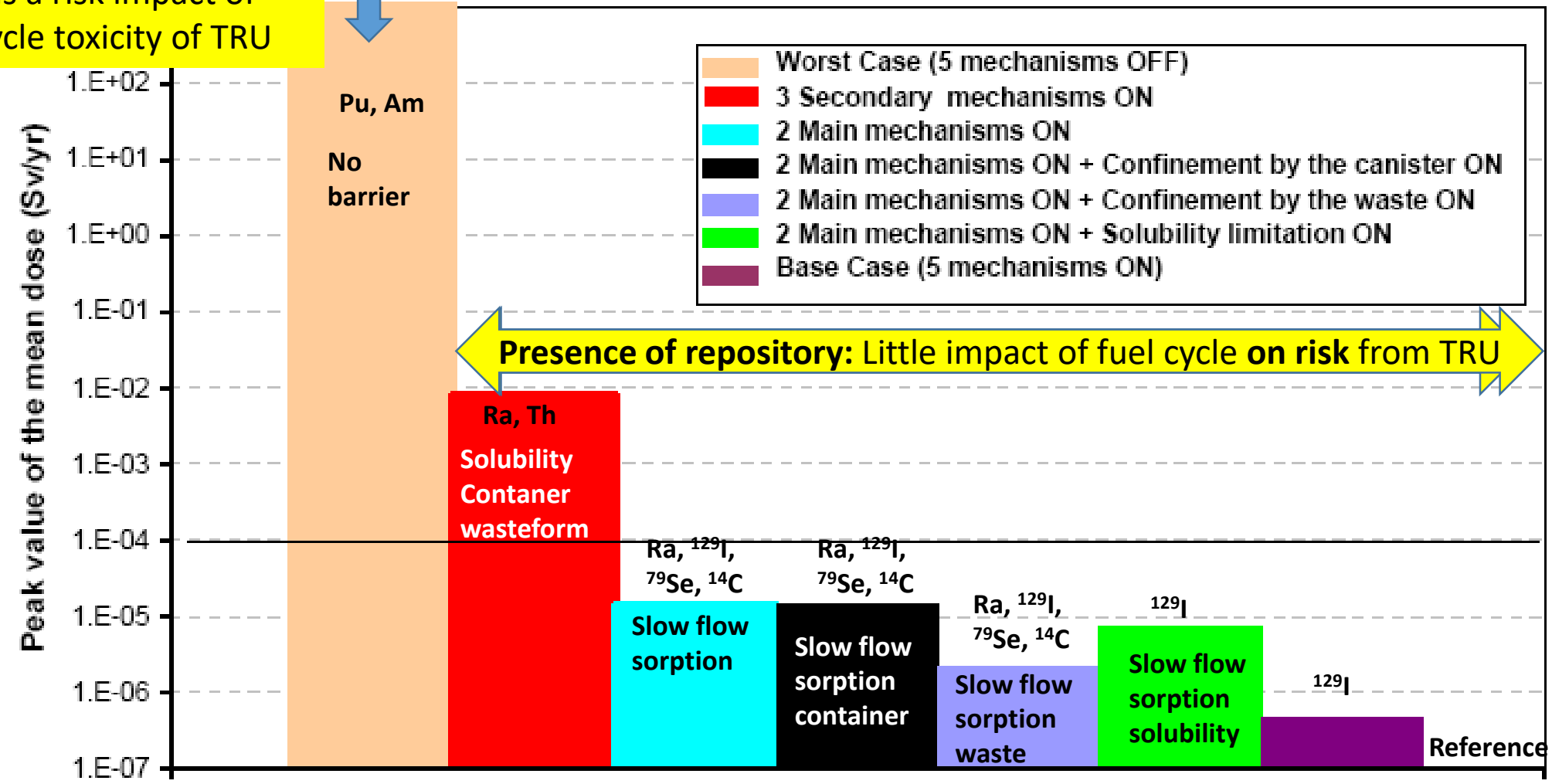
example Granite:

(Alonso et al. NFPRO ID No 5.1.5 2006)

- Assess barrier functions by eliminating them:
  - Confinement by container
  - Confinement by waste matrix
  - Solubility limitation
  - Sorption on bentonite
  - Restricted water flow in rock zones disturbed by engineering works

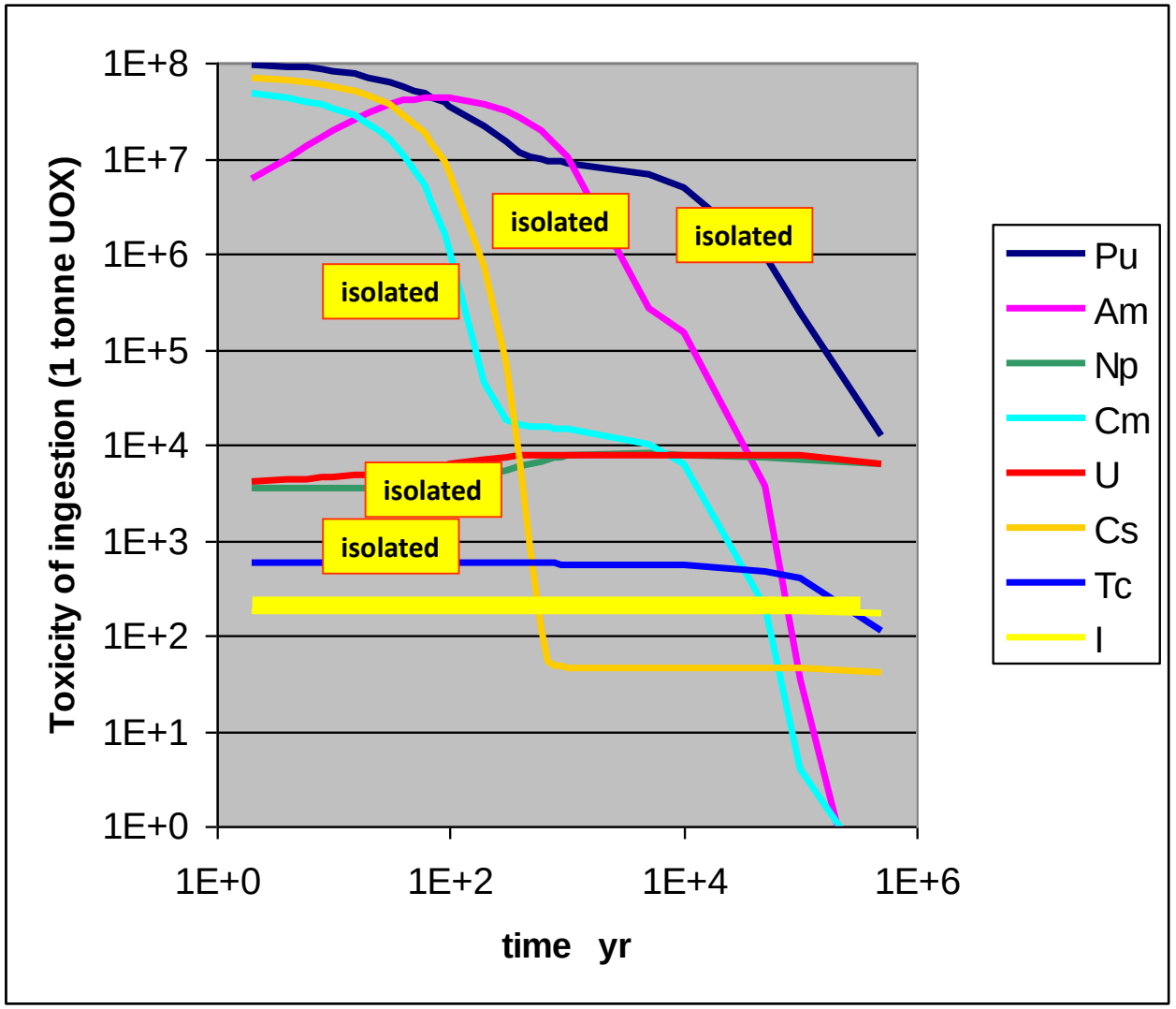
Role of combining geological and engineered barriers in reducing the exposure risk at given radiotoxicity inventory: example granite

Only in absence of barriers **(absence of repository)** there is a risk impact of fuel cycle toxicity of TRU



Source of data: NF-PRO ID N 5.1.5 Alonso et al. 2006

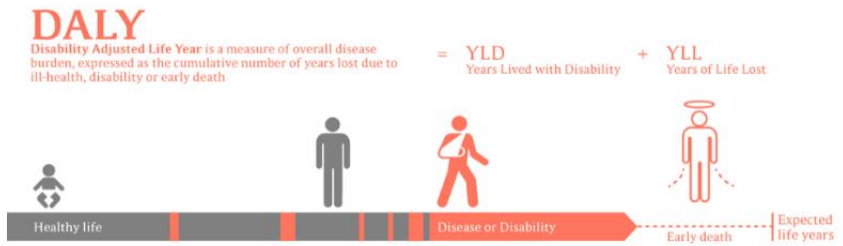
Near field isolation of radiotoxicity
A key safety indicator: reducing conditions



Isolation until decay
Isolation until decay
Isolation likely until decay
Isolation until decay
Conc. < natural
Slow release to far field
Isolation until decay
Slow release to far field

B. Grambow, Mobile fission and activation products in nuclear waste disposal, Journal of Contaminant Hydrology, (2008) 102, 180-185

Radiological risks in perspective of all risks and causes of DALY (disability adjusted loss of life years):
Japan 2017, all ages and sexes



DALY (2017) = 27366 years lost for 100000 people, without the effect of Fukushima

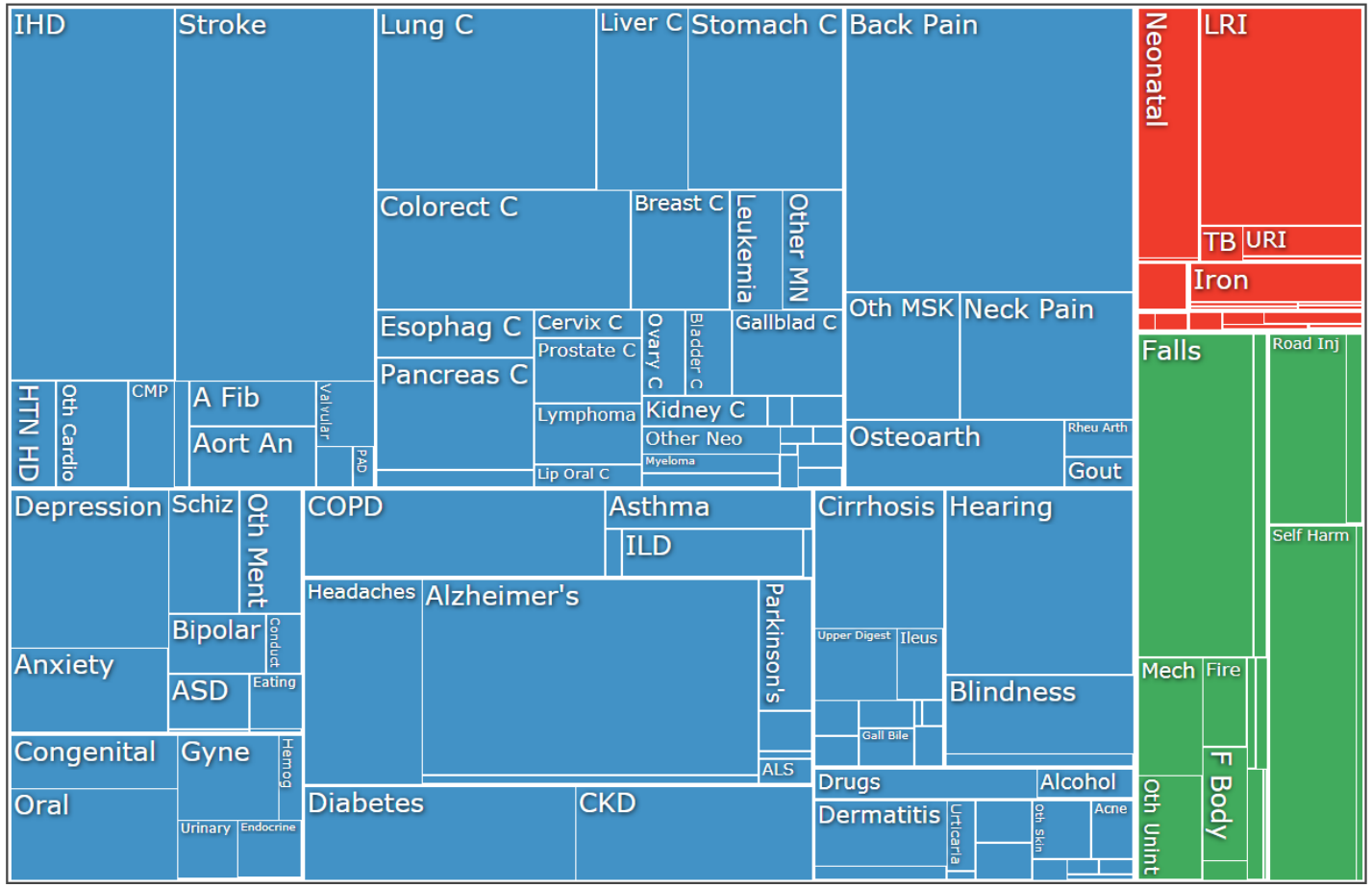
Relative risk expressed by size of the area

for 20 mSv/yr

Treshold for zoning and for allowing people to return

1mSv/yr natural radiation

Radiological risk from nuclear installations (For repository in clay 10000 x lower, Yucca mountain 100 x lower)



How low is low enough? Fuel cycle changes cannot reasonably be justified by gains in repository safety

Translation of radiological risks to the DALY scale by the approach of Shimada et al. 2015

Only limited risk/impact reduction for inadvertent human intrusion IHI by fuel cycle changes

- IHI is sometimes used as selling argument for proposing fuel cycle changes, but….
- ICRP(2013)
  - While drilling into the facility, could lead to direct exposure of the intruder and nearby populations:
    - Best protection by reducing the possibility of such events, by location repository at great depth.
    - The employment of probabilities to future human behavior is unappropriated
    - ICRP relevant dose and risk constraints shall not be applied
- To the best of my knowledge, no safety organization worldwide has proposed transmutation of minor actinides to reduce the risk/consequences of IHI

New fuel cycles leading to more stable waste forms?

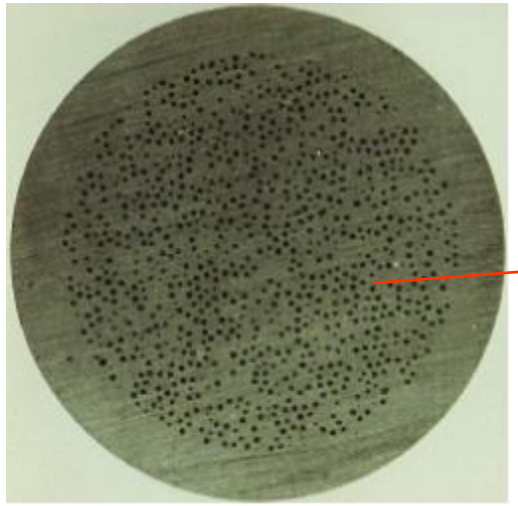
Europe: 5th FWP: HTR-N, HTR-N1 work with real spent fuel

6th FWP: Raphael

7th PCRD: Carbowaste

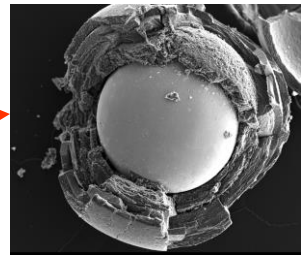
Industrie: AREVA-NC

Fuel element



6 cm

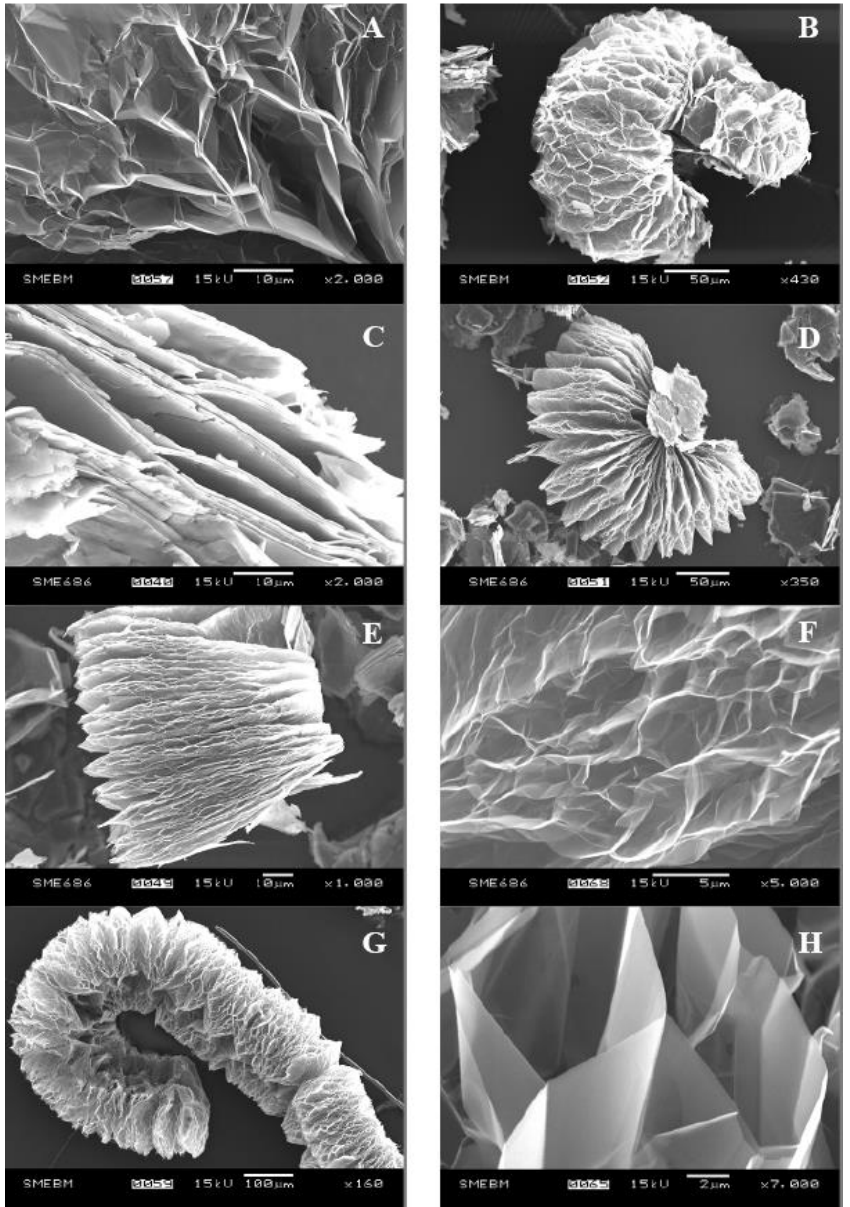
Fuel particles
TRISO



900 µm

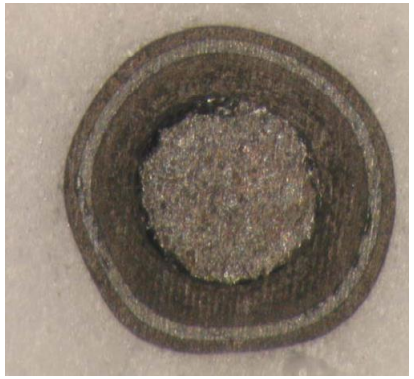
Despite stability of SiC chemical separation for reprocessing is possible

- Graphite intercalated compound (GIC) stage n, where H_2SO_4 is intercalated between each graphene using HNO_3/H_2SO_4 mixtures
- Reaction in microwave /sono chemistry works as well
- Exfoliation (via degazation at 1000°C) leads to further increase of specific surface area (final product 3 m^2/g)
- Improvement of process using H_2O_2/H_2SO_4 : volume increase by factor 600, S= 30 m^2/g
- The volume expansion leads to release of fuel particles

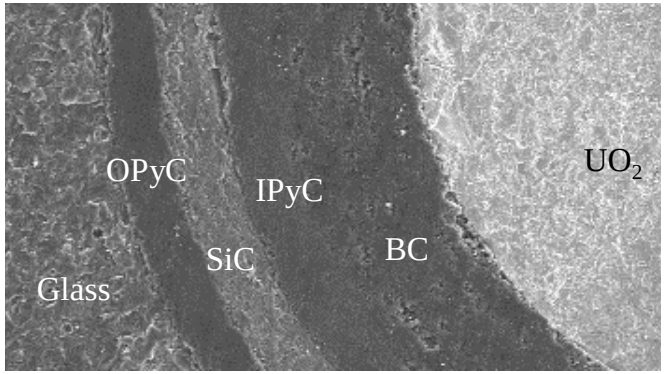


Embedding of coated particles in sintered glass

Sintered glass with OPyC



Sintered glass with pretreated particles



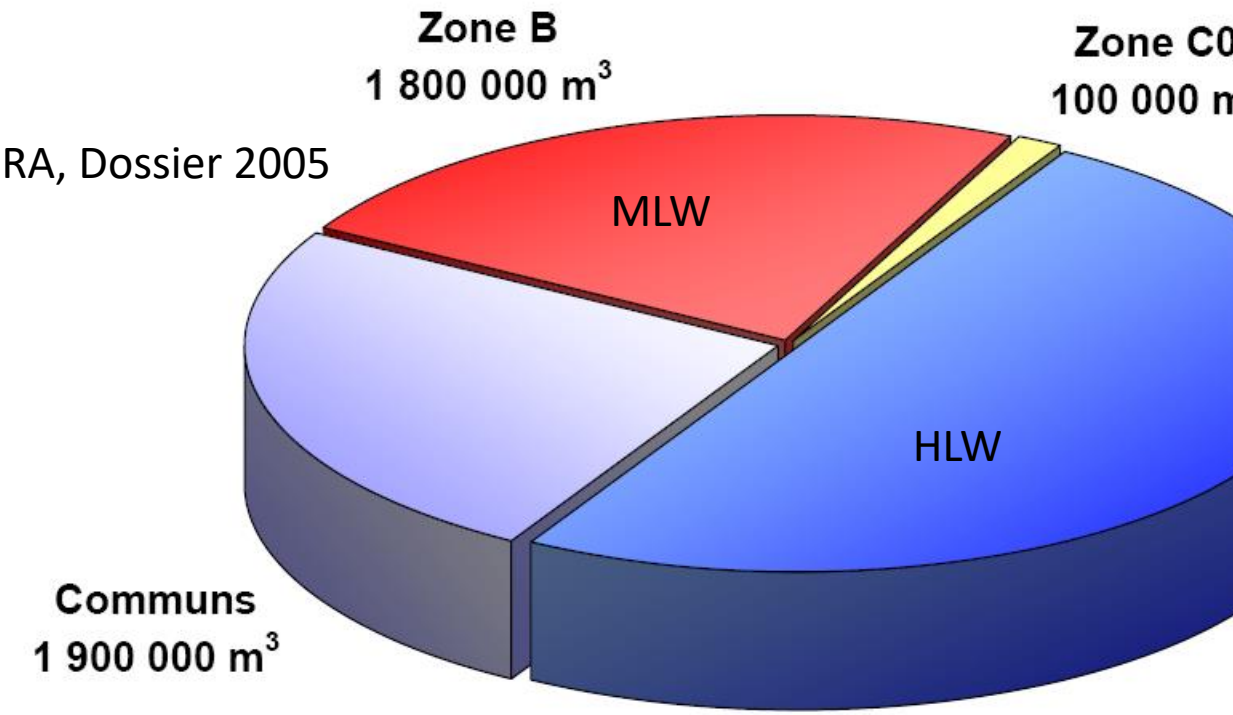
- Absence of OPyC oxidation
- Good OPyC/glass contact
- Good SiC glass contact for pretreated particles
- Glass can be replaced by molten SiC

Stability of (V)HTR waste

- Compared to direct disposal of HTR pebbles, large gain in repository volume by separating + confining coated particles
- Feasibility and effectiveness of confinement demonstrated for embedding particles in SiC and in glass
- High stability of fuel elements and confined coated particles expected for repository :
  - Early container failure:
    - scenario less relevant for conditioned particles then for “normal” spent fuel
  - The case of container failure after 10^3 to 10^4 yr
    - In graphite with SiC 6^E4 to > 1 Myr life for UO_2 particles, 10^4 yr for UCO particles
    - The Instant release fraction may be blocked from release

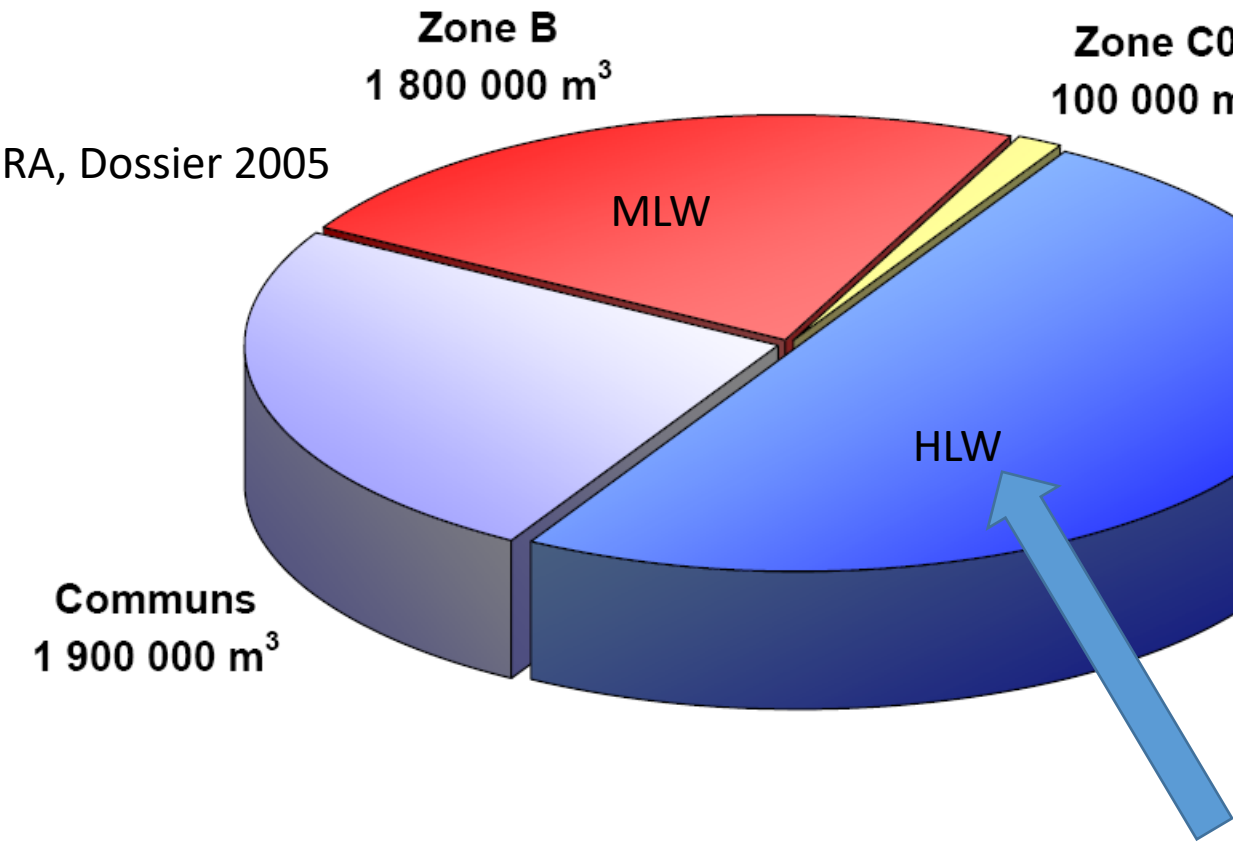
The disposal volumes foreseen in geological formations depend less on the waste volume and more on heat generation. Example France.

From: ANDRA, Dossier 2005



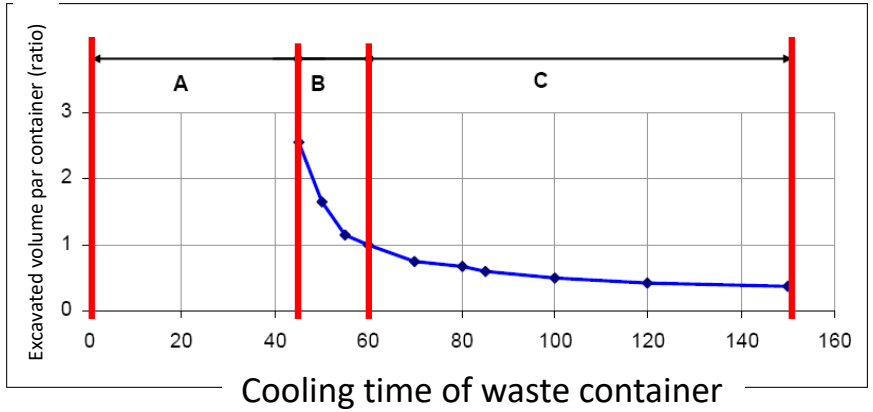
The disposal volumes foreseen in geological formations depend less on the waste volume and more on heat generation. Effect of cooling time

From: ANDRA, Dossier 2005



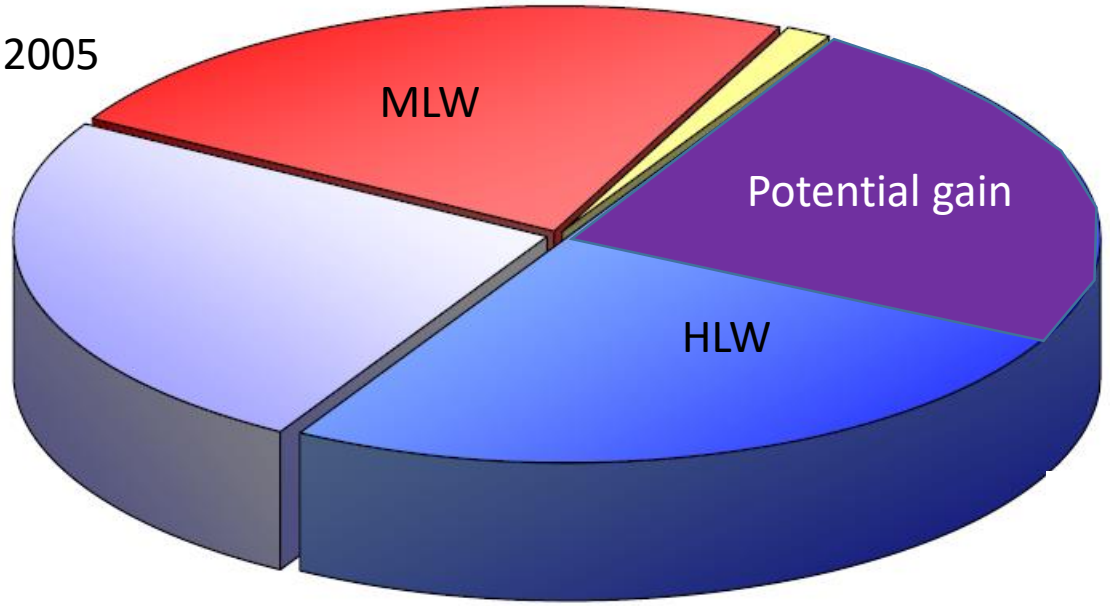
The time of interim storage of strongly exothermic wastes influences the required repository volumes in order to respect maximum temperature of 90°C

50% gain in HLW volume if cooling times increases from 50 to 100 yr



The disposal volumes foreseen in geological formations depend less on the waste volume and more on heat generation. Effect of fuel cycle.

From: ANDRA, Dossier 2005



If alternative fuel cycles involve reprocessing, the space requirement is likely to be rather similar

- As reprocessing involves large volumes of secondary wastes, the zone MLW may actually increase
- For a cooling time of 60 yr the maximum gain in repository volume dedicated to HLW is 53%, in total repository volume 25%

reactors	PWR (400 TWh)				SFR
fuel cycle concept	open cycle	mono reprocessing	multi reprocessing		
		MOX	only of Pu	of all TRU	of all TRU
	permanent inventory (tons) in surface installations (reactors, processing...)				
Pu	35	150	220	300	800
Np	2,4	6	5,3	13	4
Am	1	4	14	34	32
Cm	0,5	2	7,2	47	8
	mass of TRU in final waste (kg/yr)				
Pu	10500	7000	17	23	57
Np	740	760	660	1	0,3
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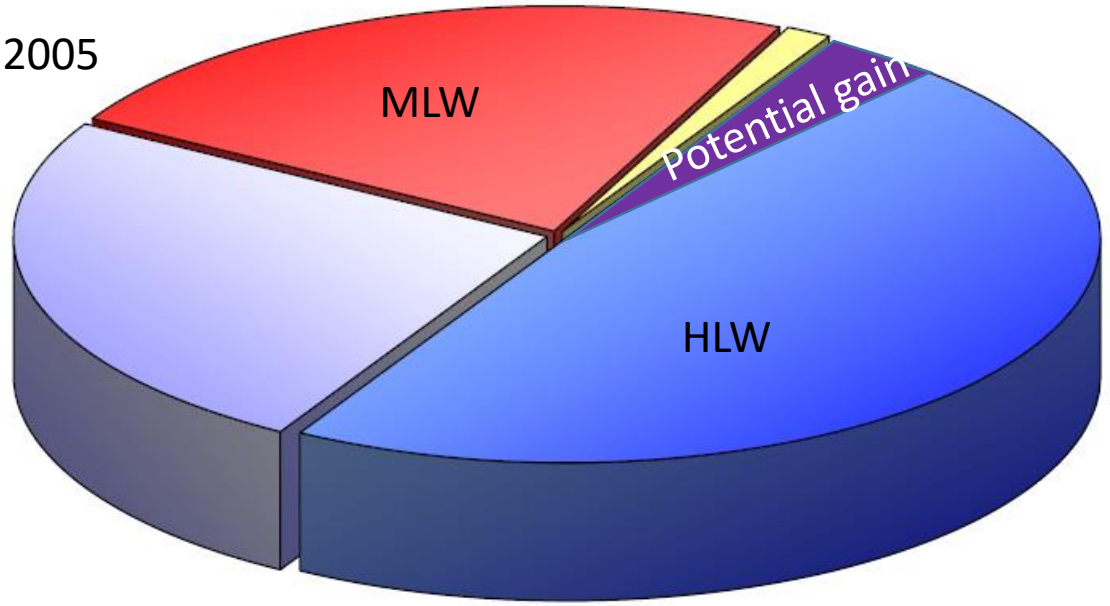
Cm	150	370	900	3,6	0,6

(calculation of heat production and associated repository volume reduction in comparison to an open cycle with an average burnup of 47,5 GWd/tHM)

The disposal volumes foreseen in geological formations depend less on the waste volume and more on heat generation. Considering as well end of life inventories remaining in reactors:

From: ANDRA, Dossier 2005



- Looking at total inventory (waste+ installations) after 100 yr:
  - For a cooling time of 200 yr the maximum gain in HLW volume is 13%, in total repository volume it is 7%

reactors	PWR (400 TWh)				SFR
fuel cycle concept	open cycle	mono reprocessing	multi reprocessing		
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Am	30	78	194	34,26	32,25
Cm	15,5	39	97,2	47,36	8,06

(calculation of heat production and associated repository volume reduction in comparison to an open cycle with an average burnup of 47,5 GWd/tHM)

Conclusions

- Actual fuel cycle options including (or not) reprocessing, reduce resource consumption or increase energy output, but they likely offer no clear benefit for waste management and long term disposal risks
- Radiological risks stemming from waste in disposal locations is governed by mobility or radionuclides and not by toxicity inventory
  - Extremely low radiological risks in disposal locations actually reduce overall risk, which is already low but is governed by surface installations
- Repository barriers are more important than fuel cycle
  - Key factors for long term safety are reducing repository conditions, container and waste form stability, groundwater mobility, solubility and sorption of radionuclides. None of these factors can be changed significantly by changing the fuel cycle
- Reduction of heat production in advanced fuel cycle lead only to limited reduction of repository volumes, longer cooling times are more efficient (see also talk of P. Swift).
  - Fuel cycles involving reprocessing need to consider the large repository volumes associated to secondary MLW wastes.
- Potential gains in waste form stability can be achieved without changing the fuel cycle