Radioactive Wastes and Disposal Options

Joint EPS-SIF International School on Energy 2017, Varenna, Italy

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https://sanonofresafety.files.wordpress.com/2012/02/onedaysonallthiswillbeyours.jpg?w=318&h=259

Outline

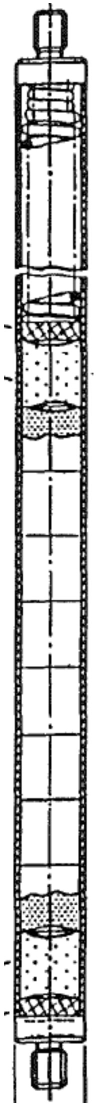
- Origin, type and amount of radioactive wastes
- Fuel Cycle: Reprocessing or
  direct disposal of used (spent) nuclear fuel
- Classification of wastes
- Disposal – International disposal projects
- Safety analysis, data and models
- Conclusions

Nuclear fuel

Fuel materials:

- Uranium oxide
  UO_2
  < 5% ^{235}U

- MOX
  ~5.5 % Pu



Rod:
Zircaloy tube
L: ~ 4.4 m
$\varnothing_{out}$: 9.5 - 11.0 mm
Active length:
~ 3.85 m

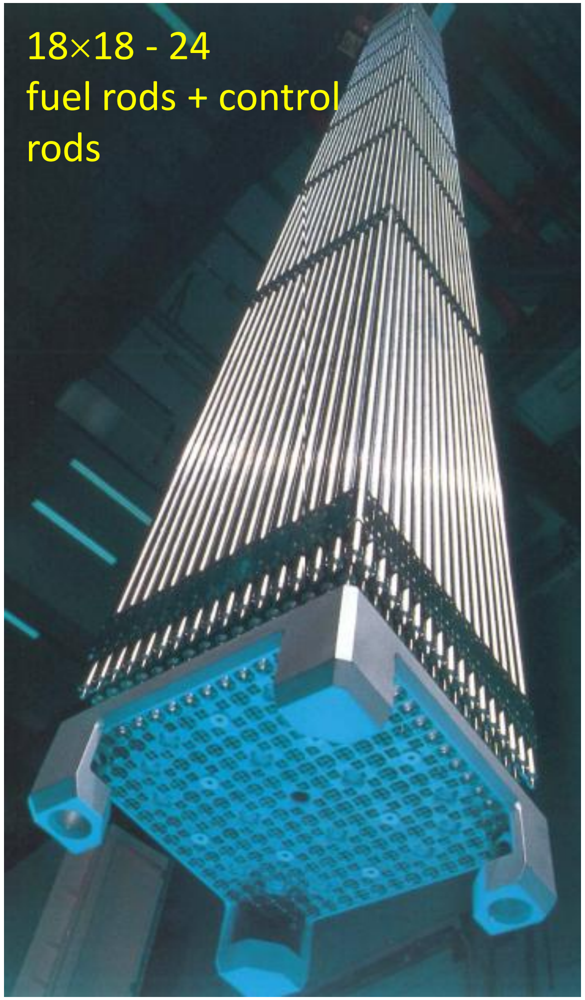
Plenum:
Steel spring
He pressure ~ 20 b

UO_2 Pellet:
H ~ 10 mm
m=10 g

> 272 pellets
per rod

BWR fuel element

PWR fuel element



Power and temperatures of NPP fuels

Pellet	KKG	GKN II	KKL
Burn-up$_{av.}$ [GWd/tHM]	64	58	58
∅ [mm]	9.3	8.05	8.5
Lin power [W/cm]	228	167	184
ΔT [K]	725	528	585
$T_{coolant}$ [deg.C]	325	305	263
T_{center} [deg.C]	**1050**	**833**	**848**

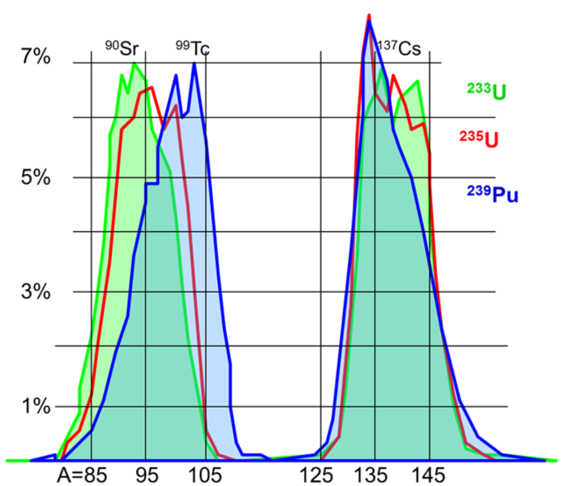
$\lambda_{irrad\ fuel} = 2.5\ W\ m^{-1}\ K^{-1}$ (Lucuta 1996)

$\Delta T = \text{lin. power}/(4\pi \cdot \lambda)$

Power ramp	
Lin. Power Change [W/cm]	100
ΔT [K]	**318**

Composition of Spent / Used Nuclear Fuel

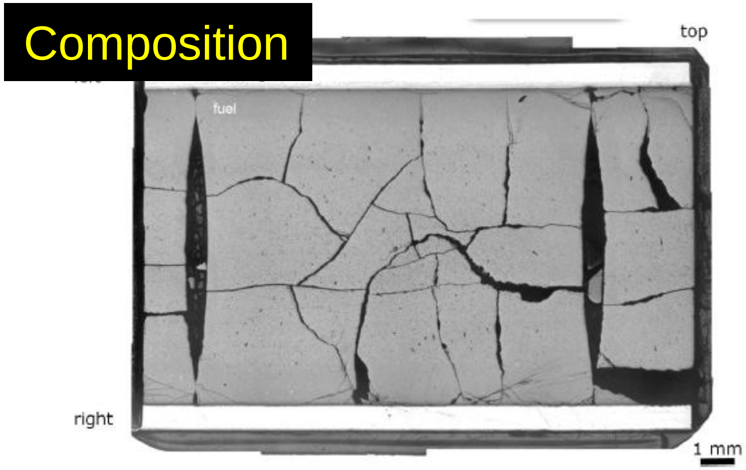
- Disturbed UO_2 matrix.
- Fission gases (Kr, Xe), not reactive.
- Rare earth elements and Y, Zr, Ba and Sr, oxides form solid solutions with UO_2, or single phase precipitate.
- Mo, Cs and Rb, oxidized or not, depending on the O/U ratio.
- Noble elements like Ru, (oxides unstable) forming metallic precipitates within the UO_2
  ε phase: Mo, Ru, Rh, Pd, and Tc

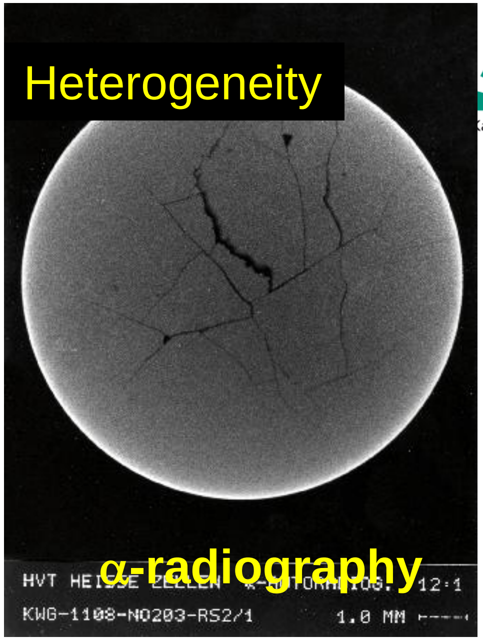


Effects of fission:

- Swelling of UO_2 lattice
  (50 GWd/tHM→5.5% FIMA)
- Distortion of lattice
- Oxygen redistribution
- Heat conductivity
- Specific heat
- Mechanical properties
  fracture patterns
- Formation of pores
  Diffusivity

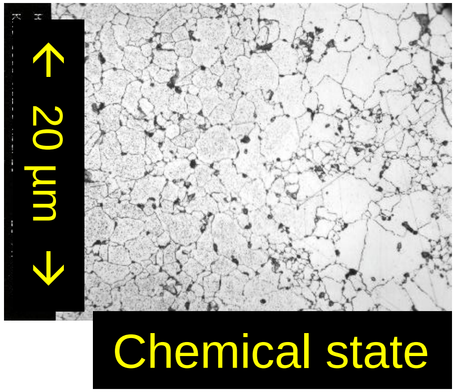
High burn-up spent nuclear fuel





Used nuclear fuel is one of the most complex materials

- Elemental composition
- Chemical state of the elements
- Heterogeneity



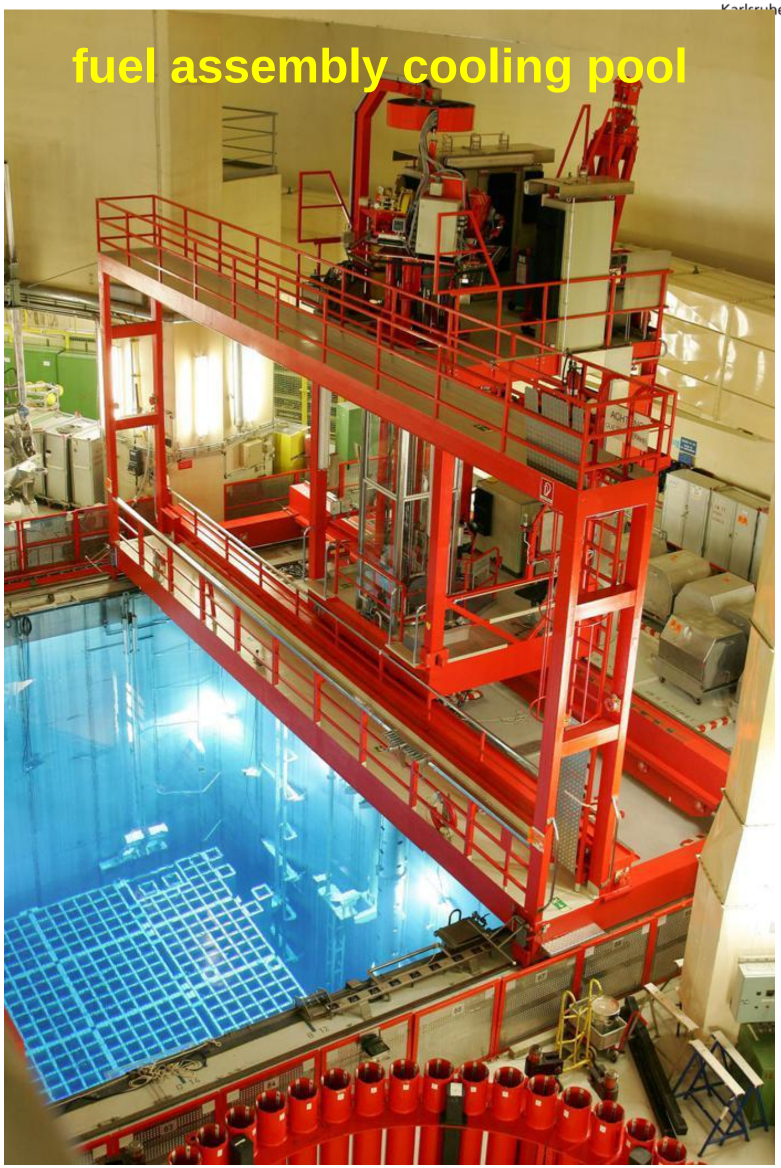
Example: Ø 10 mm, m: 10 g, 26 yrs. after discharge

β/γ activity:	2.6×10^{11} Bq,
γ dose rate:	4 Sv/hr
α activity:	9.7×10^{9} Bq

Interim storage

Heat generation of SNF:

Discharge: 200 kW/t
after 1 yr.: 13 kW/t
10 yrs: 2 kW/t
100 yrs. 0.5 kW/t



Dry interim storage in CASTOR casks

L: 6.1 m
∅: 2.5 m
0.45 m wall thickness
m: ~100 Mg
n: 19 PWR Ass./ 52 BWR Ass.

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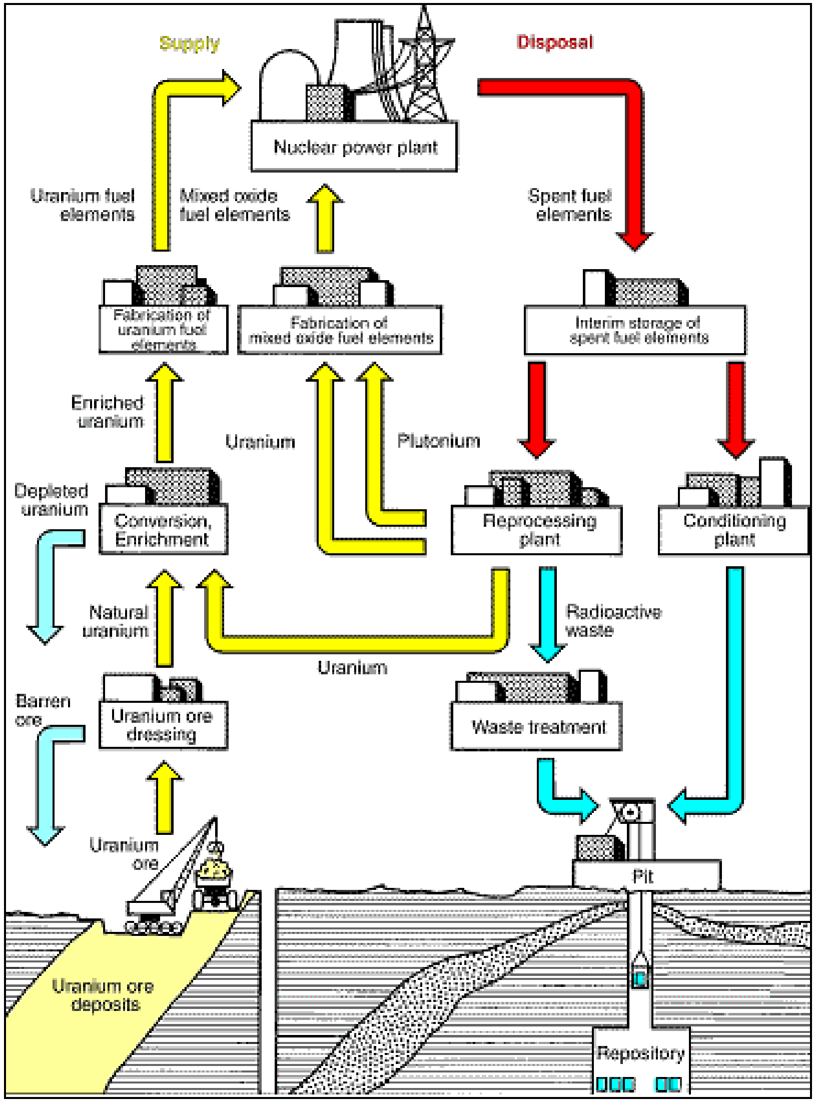
Wastes from fuel cycle and other sources



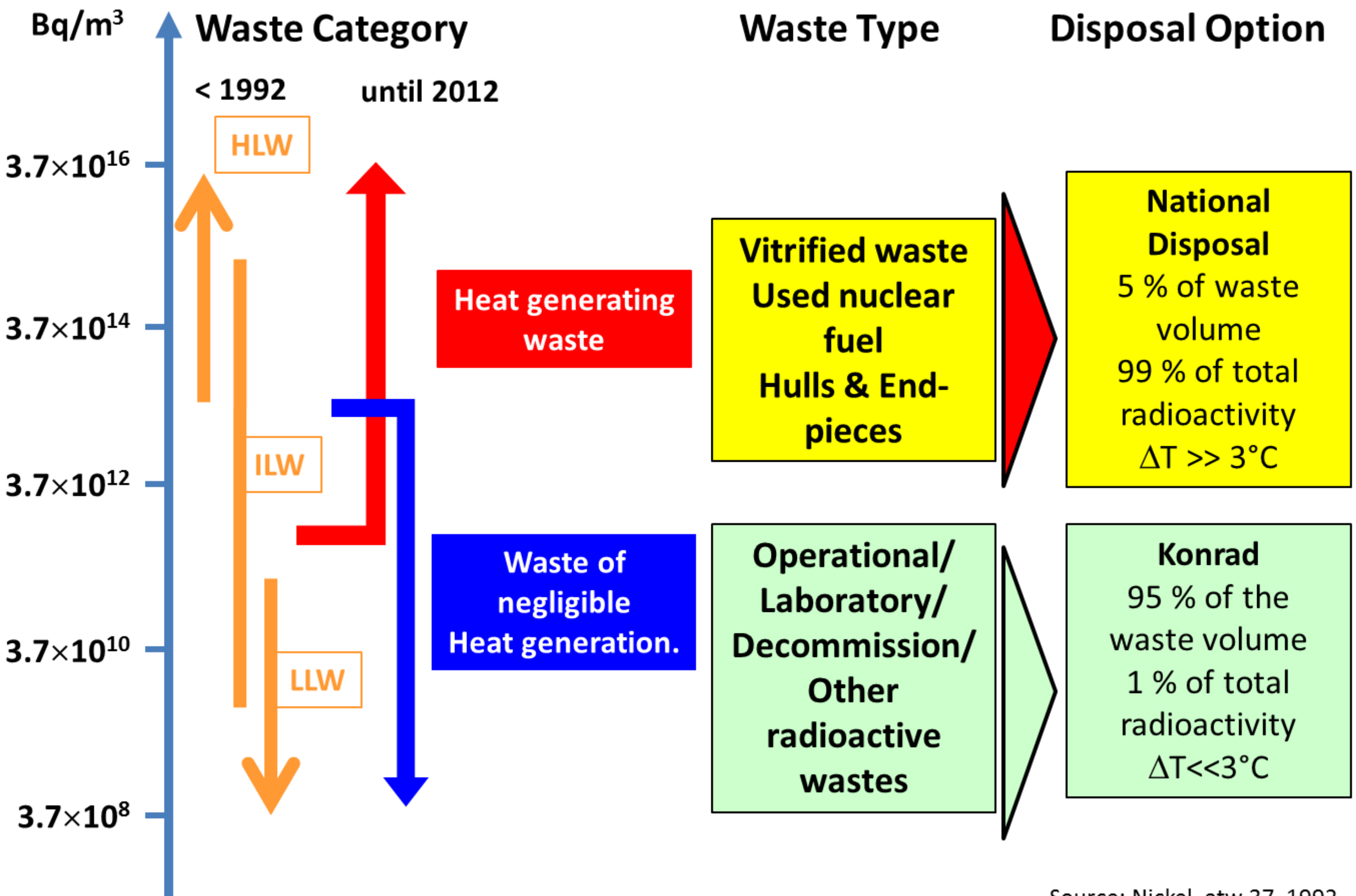
Waste streams from reprocessing:

- Hulls/cladding (solids),
- High-level waste, HNO_3 solution (liquid),
- Intermediate-level wastes acidic solutions (liquid),
- Low-level wastes, HTO-bearing liquids decontamination detergents (liquid)
- Solid wastes, e.g. filters, etc.

Other sources

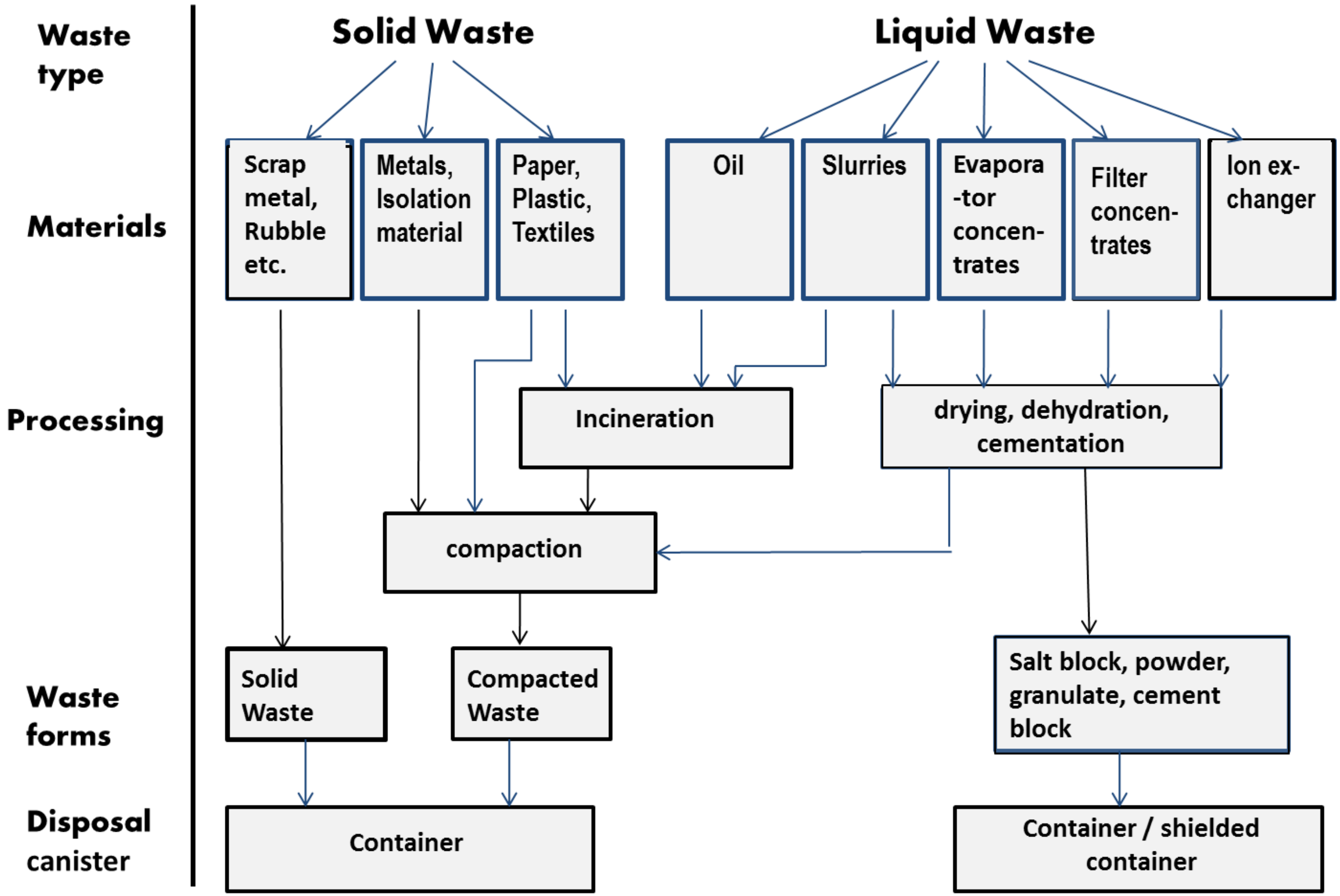
- NPP operational wastes (~300 m^3/yr.)
- Industry (sources)
- Research
- Medical diagnostic/therapy
- Decommissioning
- Scales (geothermal energy)

Radioactive Waste Classification in Germany

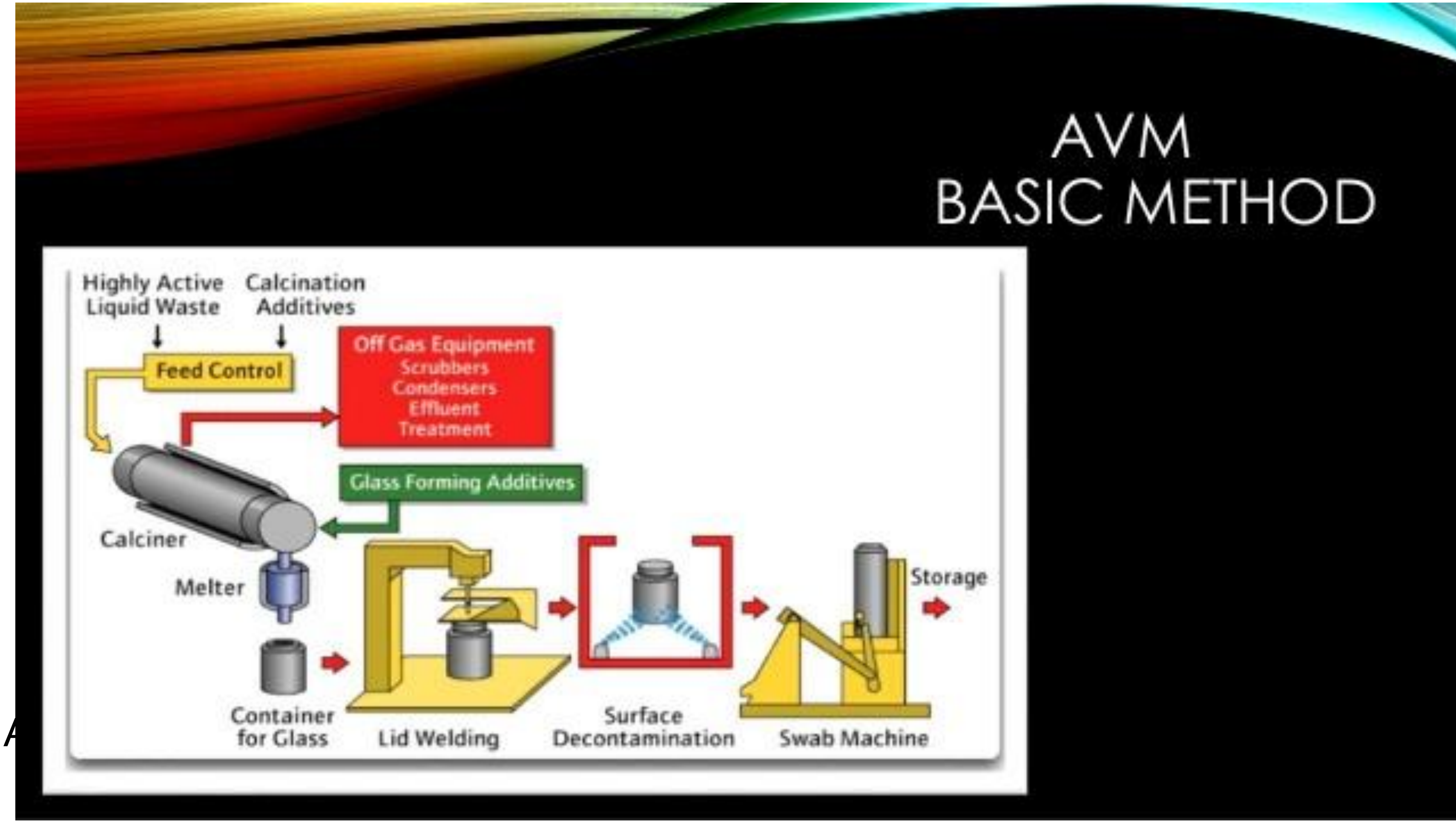


Source: Nickel, atw 37, 1992

LLW / ILW Treatment



Vitrification of high-level liquid waste



A

AREVA vitrified waste returned to Germany

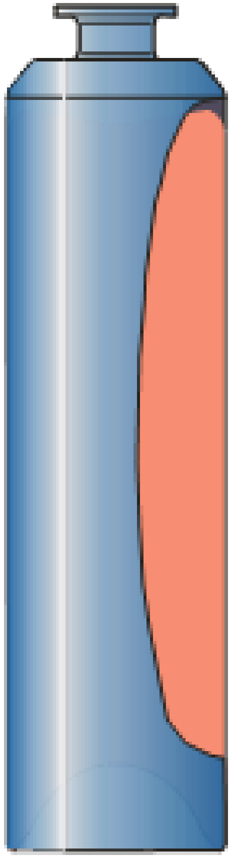
β/γ **- activity:** 65 TBq for ^{137}Cs
115 TBq for $^{90}Sr/^{90}Y$
75 TBq for ^{241}Pu

α **- activity:** 3.3 TBq for Pu
2.0 TBq for ^{244}Cm
4.2 TBq for nuclides with half-life > 50 yrs.

β/γ **- contamination:** 4.0 Bq cm^{-2}
α **- contamination:** 0.4 Bq cm^{-2}

Other parameters

Dose rate:	< 150 Gy/h
Thermal power	< 90 W
Weight	< 850 kg



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Disposal strategies

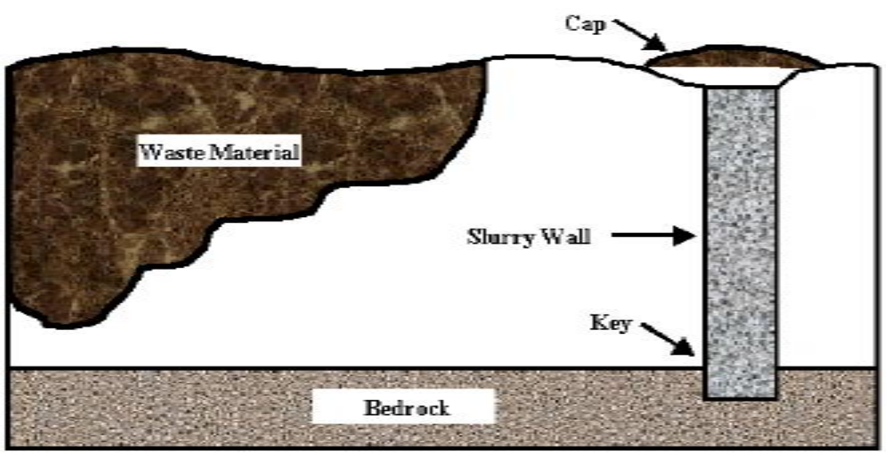
- LLW / short-lived ILW (R&D, industry, operation of NPP)
  Reminder: $A/A_o = \exp(-\ln 2/t_{1/2} \cdot t)$
  for $t = 10 \cdot t_{1/2}$: $A/A_o = 0.001$
  - Until 1970ies: sea disposal
  - International: shallow land disposal
  - Germany: geologic disposal (Asse salt mine since 1967)

- HLW, SNF (long-lived & α-bearing wastes):
  - International consensus:
    Geologic disposal in deep layers of the underground

LLW / ILW disposal close to the surface

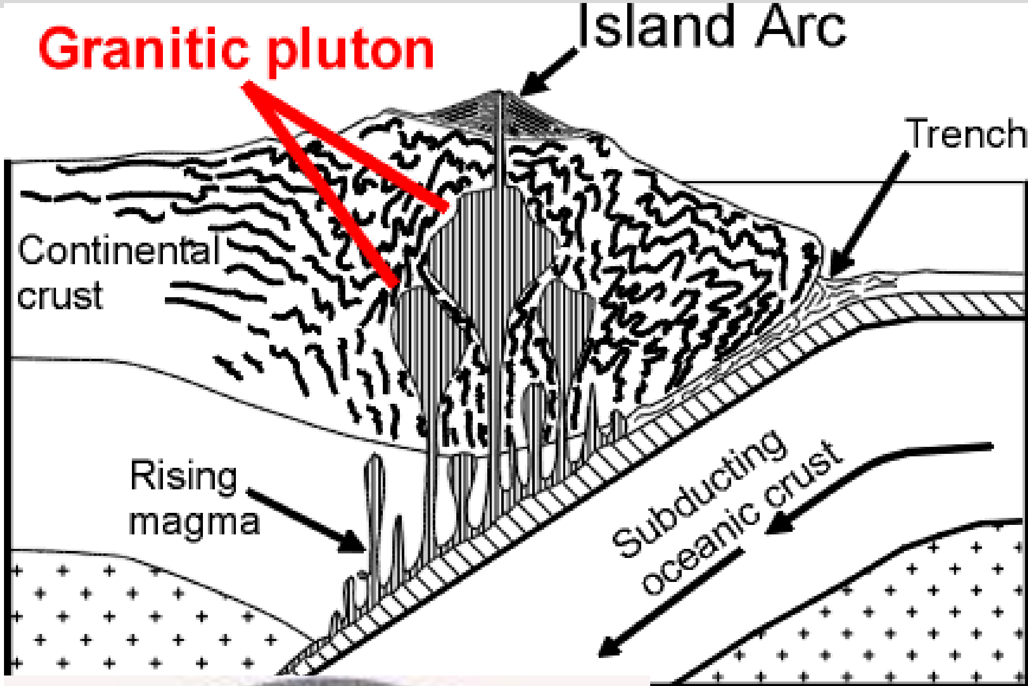
Drigg, UK

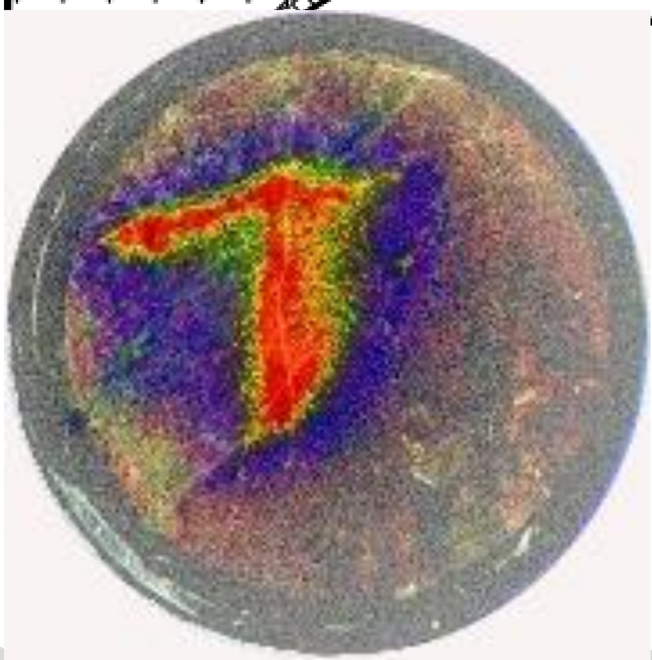
since 40 yrs. in opreation until ~ 2050



HLW / SNF: Potential Host Rocks

Rock salt	Clay rocks	Granite
+ Impermeability + Visco-plastic behaviour (convergence) + Heat conductivity + Temperature load + Age of salt domes + Experiences	+ Impermeability + Plasitc behaviour (swelling) + Insoluble + High retention capacity	+ Stable mechanics + High stress load + Moderate heat conductivity + Experience
- Solubility - Low retention capacity - Dissolution of salt - Uplift (~ 0,02 mm/a)	- Low heat conductivity - Low temperature load - Complicated mining (EDZ) - Diffusion processes	- Water bearing fractures - Low retention - Technical barriers required (bentonite) →low temperature load





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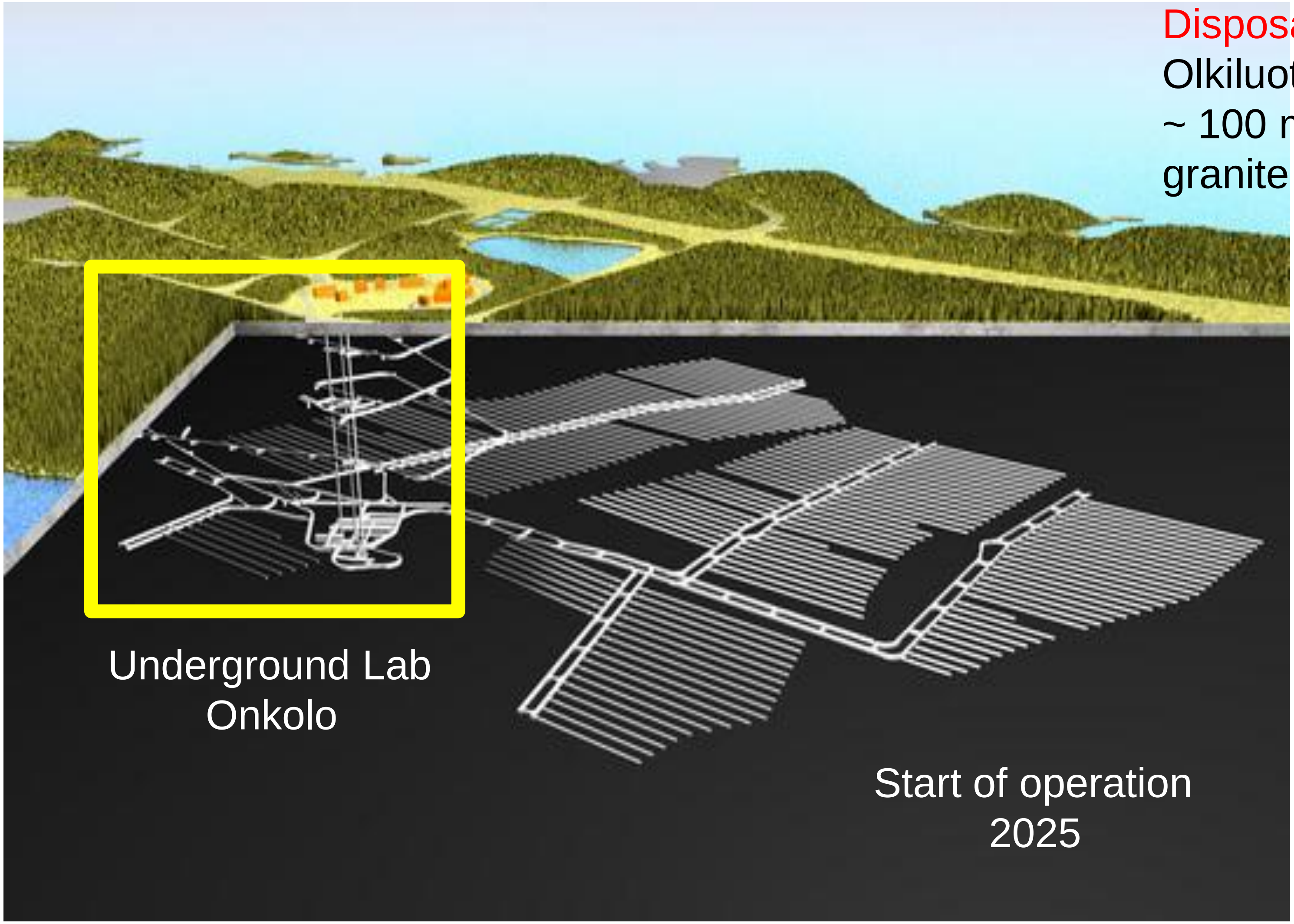
Multi-barrier system

- Waste form (glass, spent fuel, ...)
- Disposal cask
- Engineered barriers (backfill, sealing, …)
- Host rock
- Surounding / Overlaying rocks

Requirements for engineered barriers

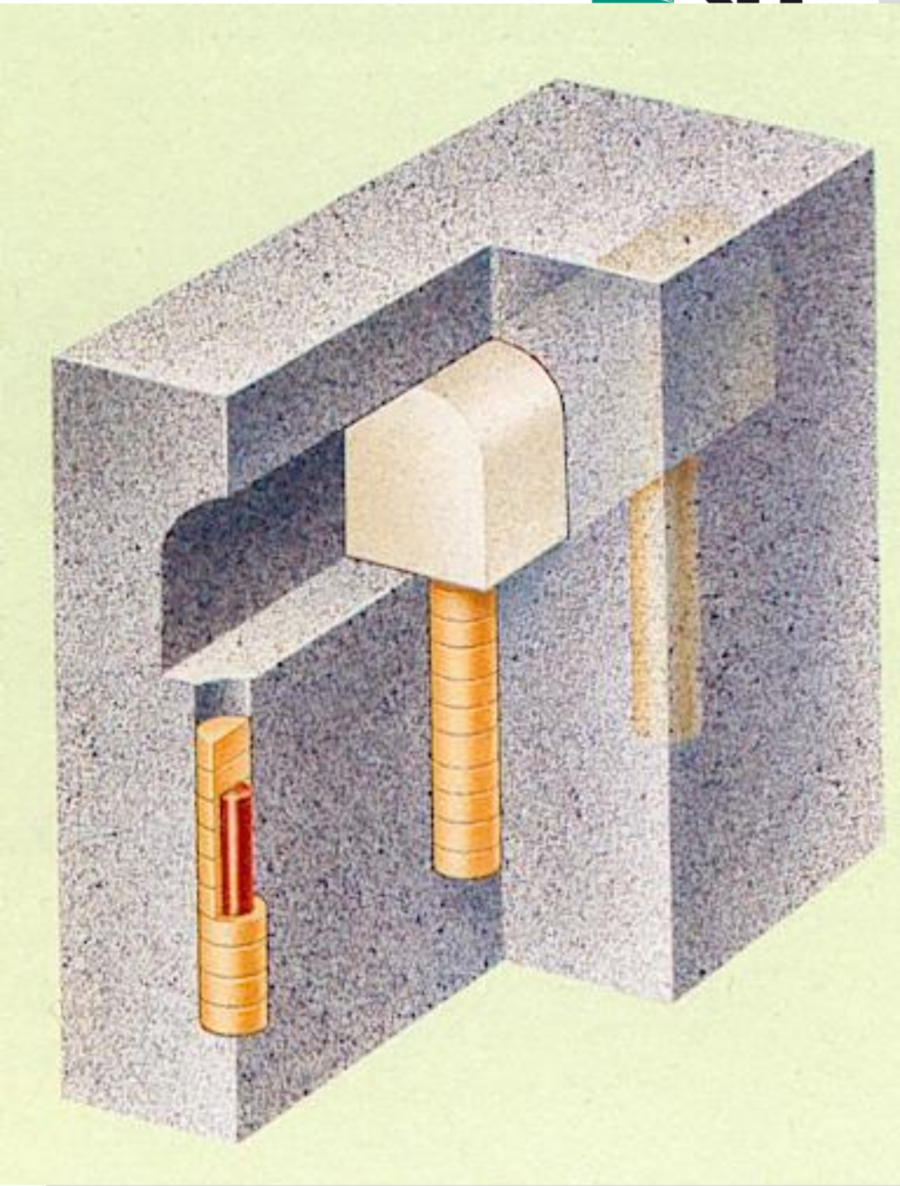
- Barrier against penetration of groundwater / solutions
- Sorption capacity for RN
- Chemical "buffering" (pH, Redox, composition of solutions…)
- Mechanical stability
- Heat conductivity (HLW, SNF)
- In granite: decoupling of cask from rock (earthquake)

Finland SNF Disposal (Posiva)



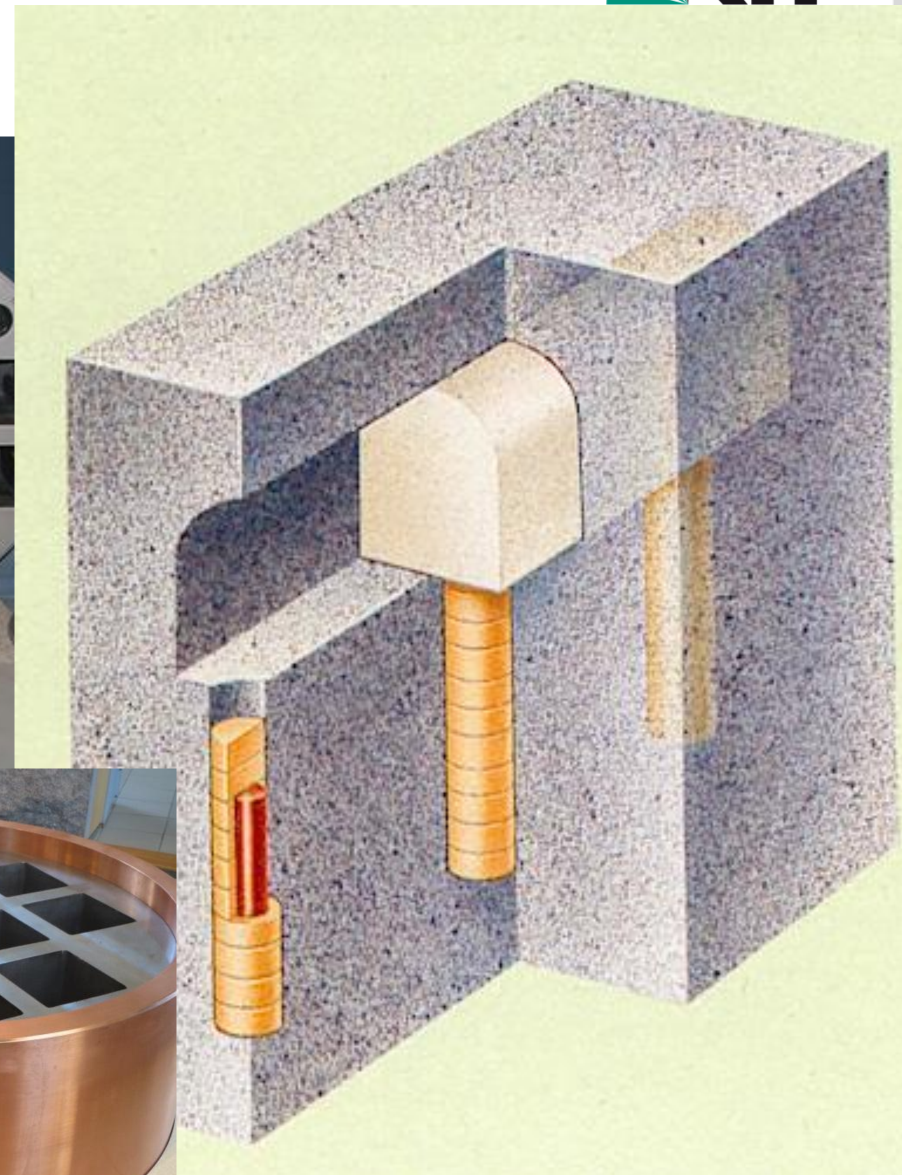
Disposal for LLW:
Olkiluoto and Loviisa
~ 100 m depth
granite

Disposal Concepts: Granite (Sweden, Finland)



KBS-3 concept

Disposal Concepts: Granite (Sweden, Finland)



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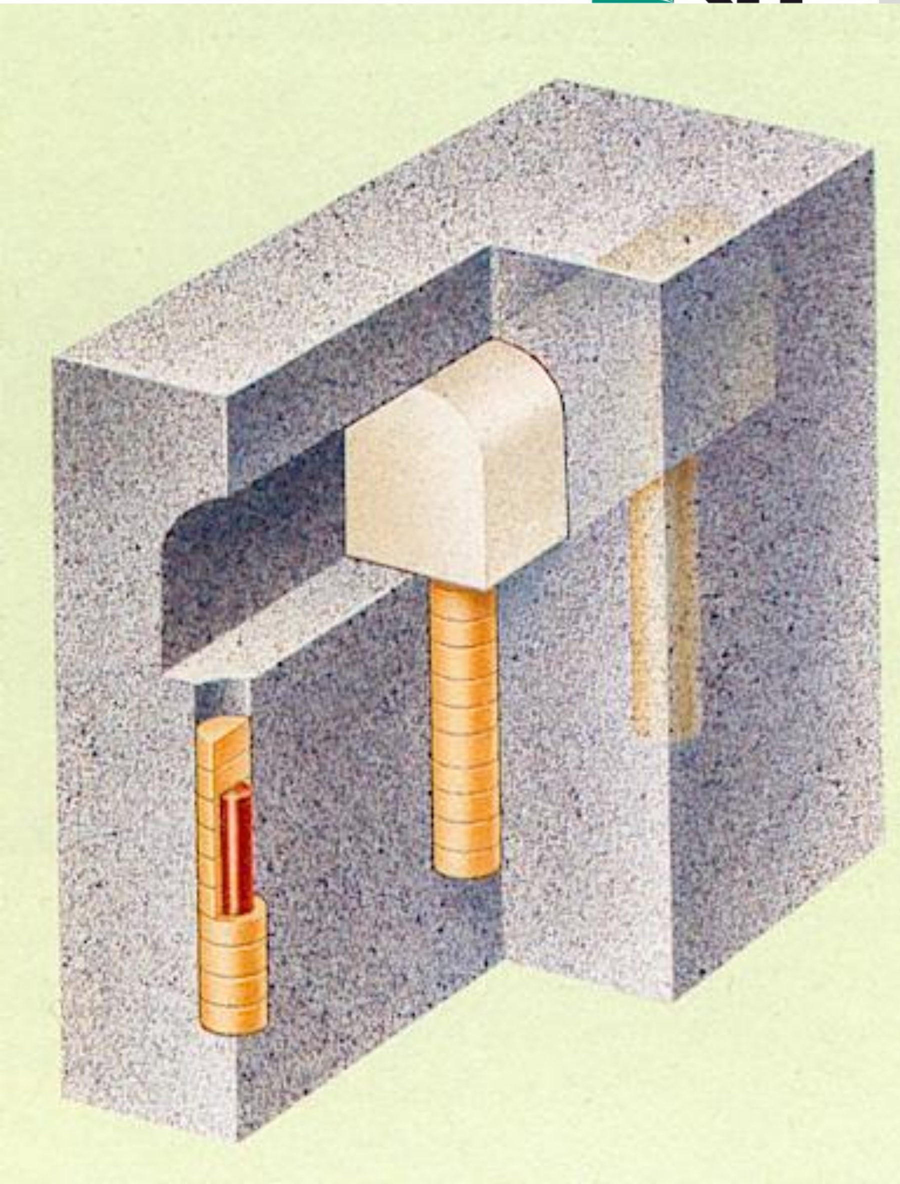
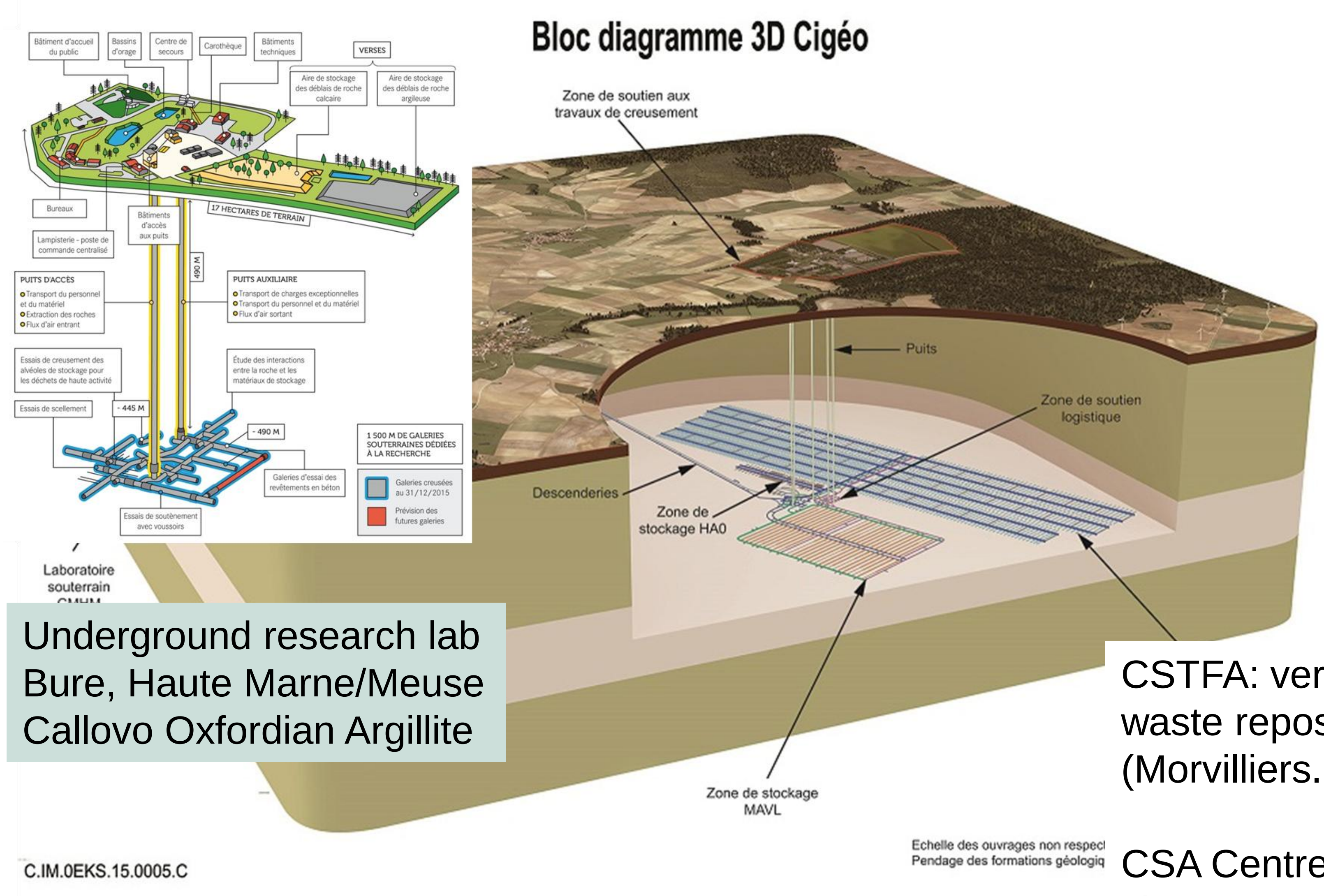


KBS-3 concept

HLW disposal in France (ANDRA)

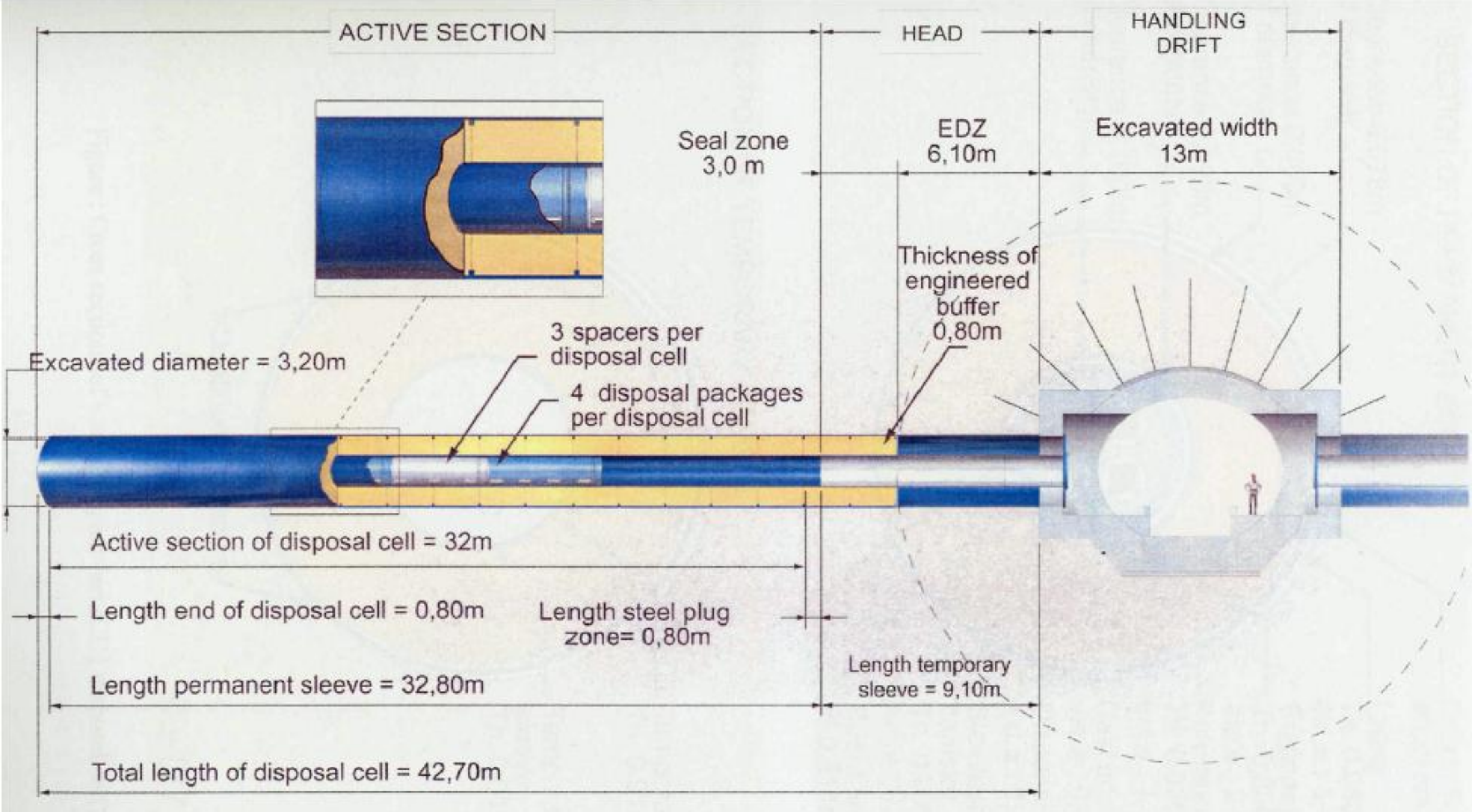


Underground research lab
Bure, Haute Marne/Meuse
Callovo Oxfordian Argillite

CSTFA: very-low-level waste repository (Morvilliers. Aube)

CSA Centre de l'Aube shallow land disposal

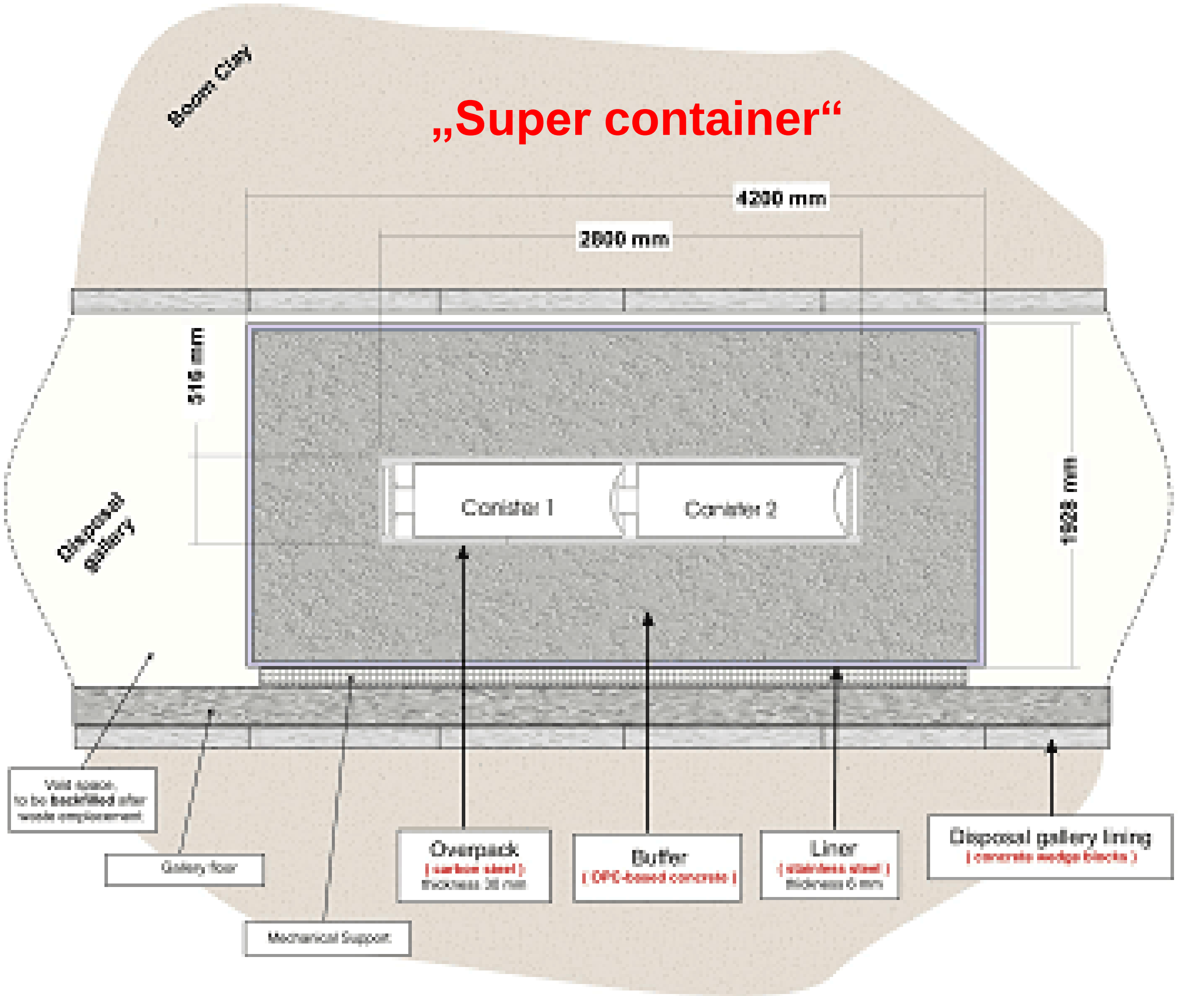
Disposal Concept in Argillite (France) - Retrievability for ~200 yrs.



Host rock Clay

Host rock Clay

„Super container“



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Why disposal in deep underground?

Technical reasons:

- Concentration of radiotoxic materials
- Isolation from biosphere
- Protection of environment and population
- Protection against intended and unintended access.
- Safeguards (nonproliferation of fissile materials)

Ethical and societal reasons :

- Passive safety
- Unattended (maintenance free)
- No burdens for future generations

What do we need to know for safety analysis?

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Assumption: Water facilitates radionuclide mobilization & transport

- Basics
  - Thermodynamics of radionuclides
  - Solids, complexation & redox reactions
- Waste form behavior
  - Radionuclide mobilization from the waste forms
  - Corrosion processes
  - Solubility
  - Formation of secondary phases
- Radionuclide migration
  - Sorption
- Performance under natural/realistic conditions
- Modelling tools

Safety Research for Nuclear Waste Disposal
- Basic Investigations -

Molecular Process Understanding

- Aquatic chemistry/thermodynamics of actinides and LLFP
- Interaction of actinides with mineral surfaces
- Secondary phase formation
- Stability of colloids/interaction with actinides
- Quantum chemical calculations

Safety Research for Nuclear Waste Disposal
- Applied Investigations -

Radionuclide retention in the multibarrier system

- Waste Form (Spent fuel, HLW-glass)
  Canister, Backfill Material,
  Host Rock/Geological Formation

- Radiation chemical effects

- Field experiments
  (Underground laboratories)

- Reactive transport modelling

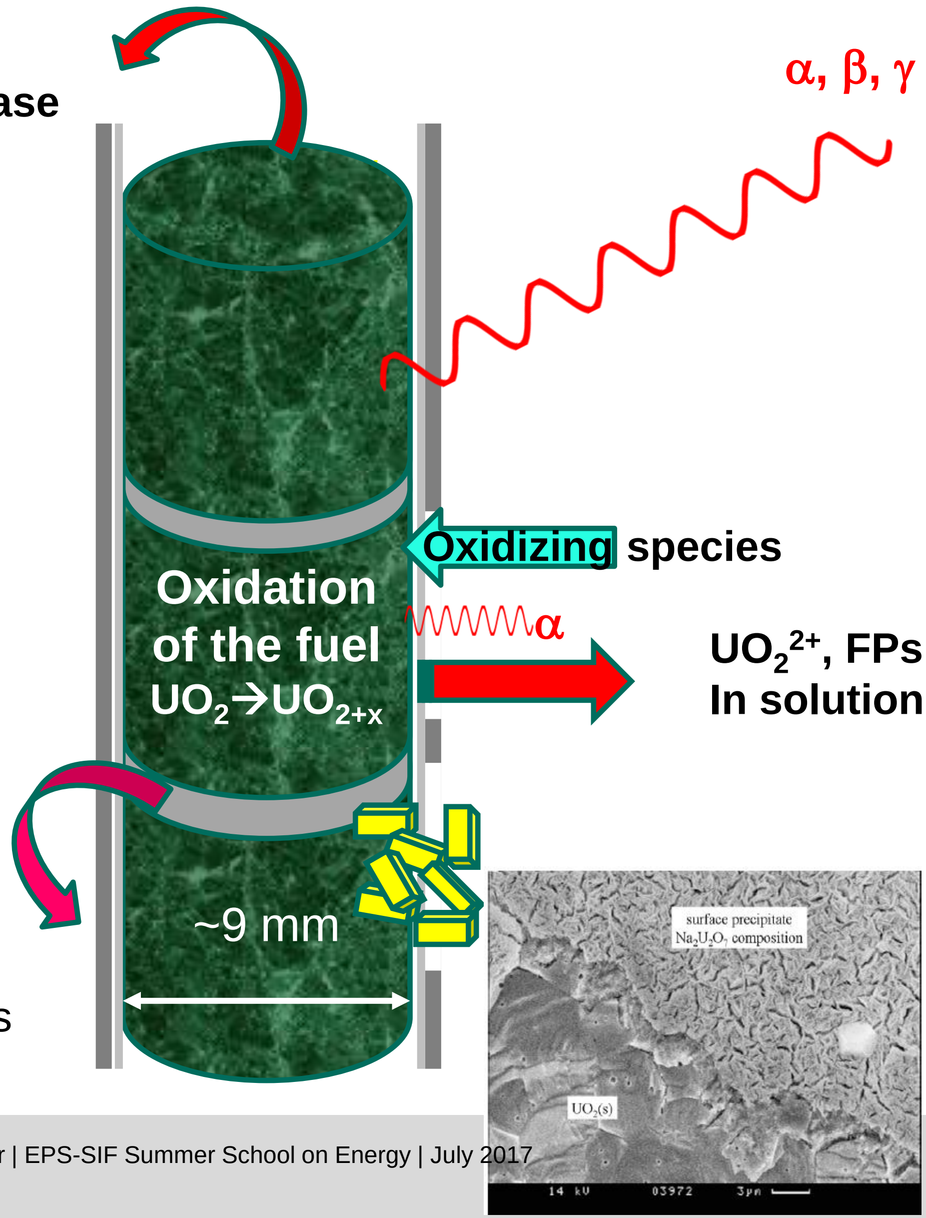
Radionuclide Mobilization from SNF

- Instant release fraction.
- Matrix dissolution.
  - Radiolysis effect.
  - Secondary phase formation.
  - H_2 effect.
  - ε phase effect
- Effect of cladding

Radionuclide mobilisation from spent nuclear fuel non-reducing conditions

Fission gas release
Xe, Kr

Gap release
FPs, fission gases



Radiolysis of water
Generation of oxidants
(H_2O_2, $OH^•$, HO_2, O_2)
and reductants
(H, H_2, e^-_{aq})
diffusion of H, H2 leaving the reaction zone

Matrix dissolution
oxidation of UO_2,to UO_{2+x}
dissolution of U(VI) + RNs

Secondary phase precipitation
U(VI) phases may incorporate other FPs and RNs

Radionuclide mobilisation from spent nuclear fuel under reducing conditions

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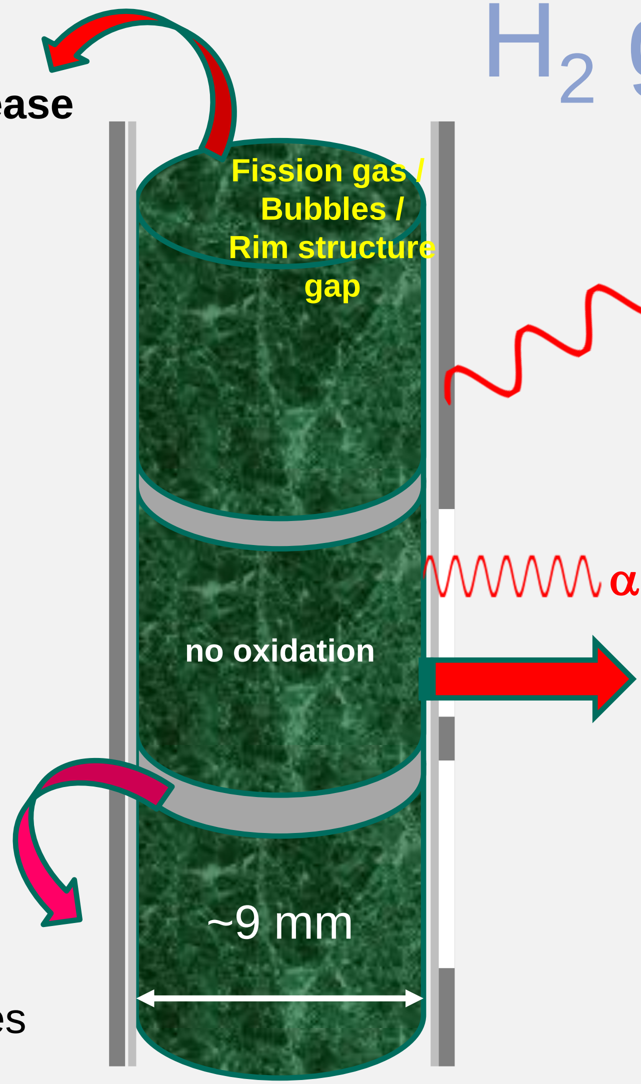
H_2 gas

Fission gas release
Xe, Kr

Closed system



U(IV), FPs dissolution

Gap release
FPs, fission gases

Radiolysis of water
Generation of oxidants (H_2O_2, $OH^\bullet$, HO_2, O_2) and reductants (H, H_2, e^-_{aq})

Recombination
oxidizing and reducing species

Matrix dissolution
UO_2, other RNs and FPs into solution

Dissolved U(IV) by thermodynamic solubility

Safety Analysis for a Nuclear Waste Disposal

Assumption: The future is unknown,
safety analyses are build-up on the concept of scenarios

A scenario is specified by a combination of some FEPs which characterize **one single potential evolution** of the disposal system.

FEP: Features, Events, Processes

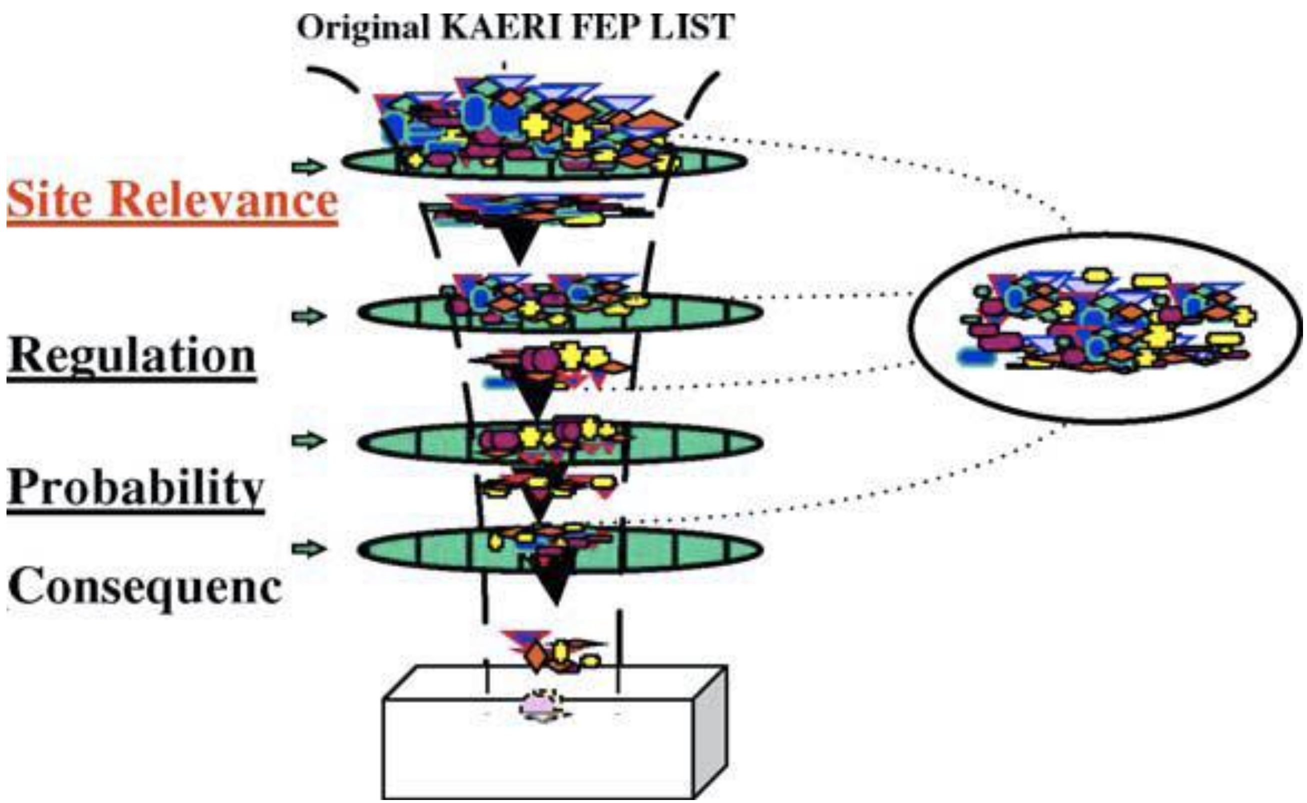
Example scenario

A scenario is specified by a combination of some FEPs which characterize one potential evolution of the disposal system.

FEP 1	FEP 2	FEP 3	FEP 4	FEP 5	....
Water penetra-tion	Geochem. Milieu	Corrosion canister	corrosion waste form	Radionuclide mobilization	RN Löslichkeit Sorption
$\dot{q} = f(t)$	pH, Eh, c(macro c) c(trace)	r_B=f(T,c,pH,..)	r_M=f(t,T,c,pH,..)	r_M=f(t,T,c,pH,..)	c_{RN}=f(pH,c...)
Value: (yes/no)	Numerical function	Numerical function	Numerical function	Numerical function	Numerical function

$$\dot{D}_{Scen1} = f\left(\dot{q}_{FEP1}, geochem_{FEP2}, t_{cask\,life-time}, r_M, r_{RN}, c_{RN}, pH, Eh, \ldots\right)$$

Four FEP screening criteria (example KAERI)

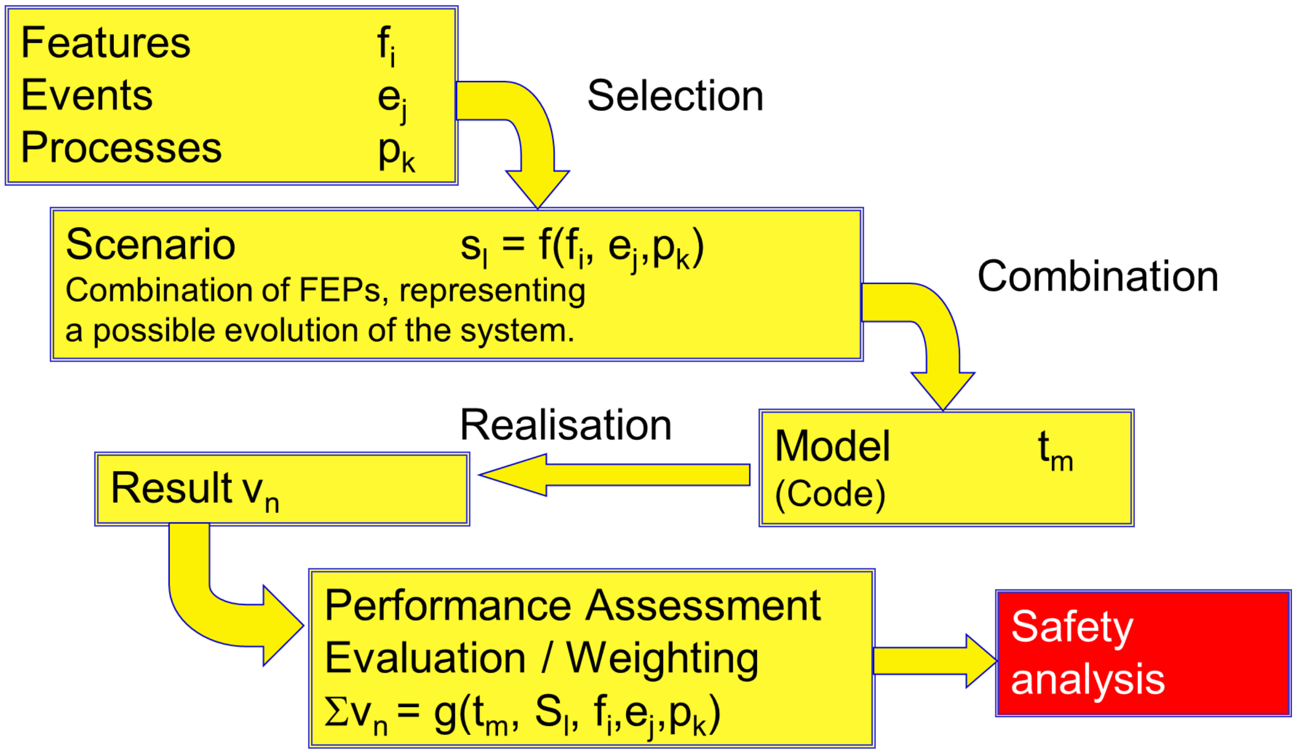


Application of modells in PA

Model:

No ab-initio modells, but
- Modells describe parts of reality,
- on basis of free parameters (determined by experiments)

PA :



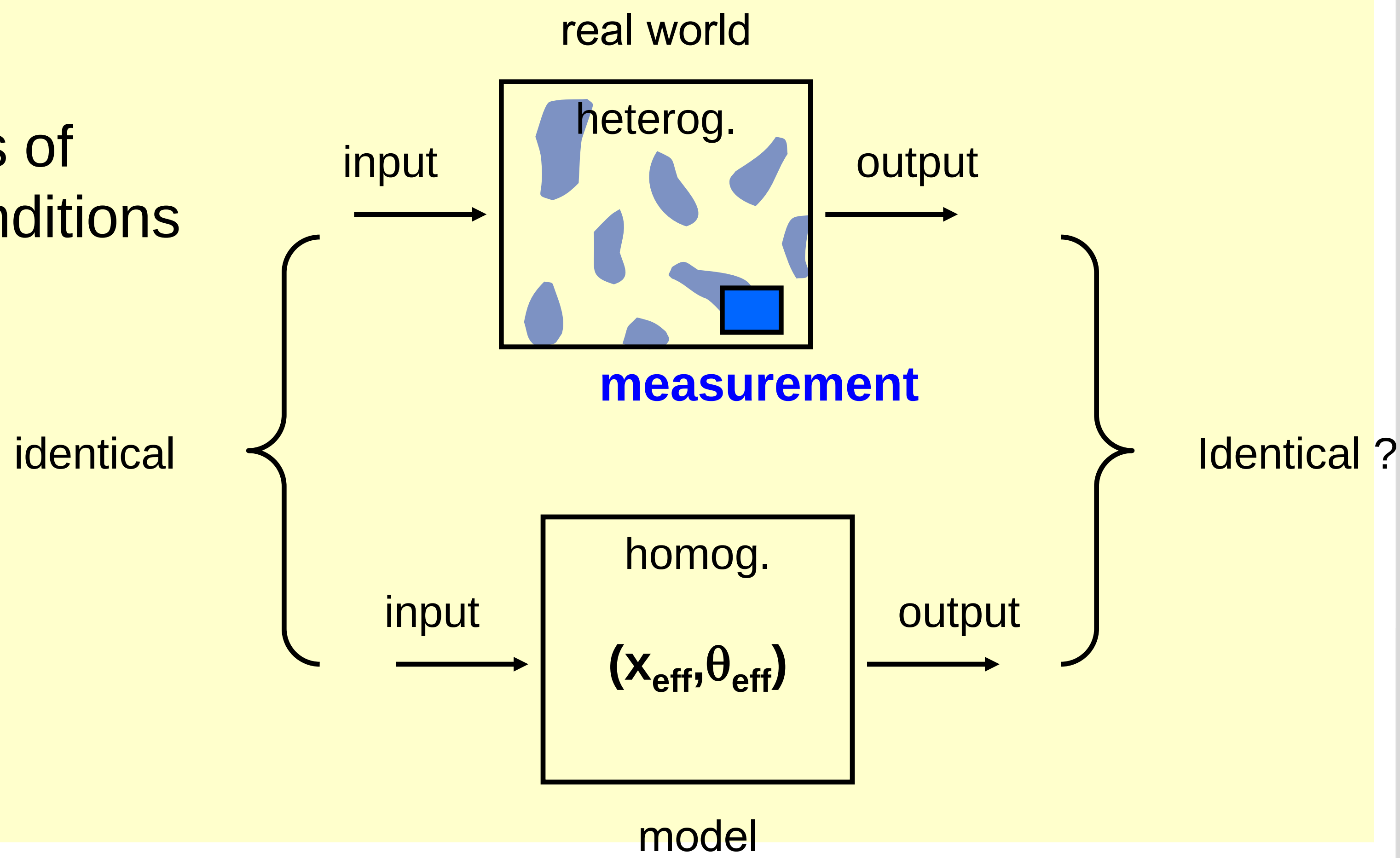
Restrictions:

No extrapolation exceeding the parameterized set of features and processes.

Sources of Error and Uncertainty

- Model Structure
- Parameters
- Data
- Forecasts of future conditions

George H. Leavesley, Hydrologist, USGS, Denver, CO



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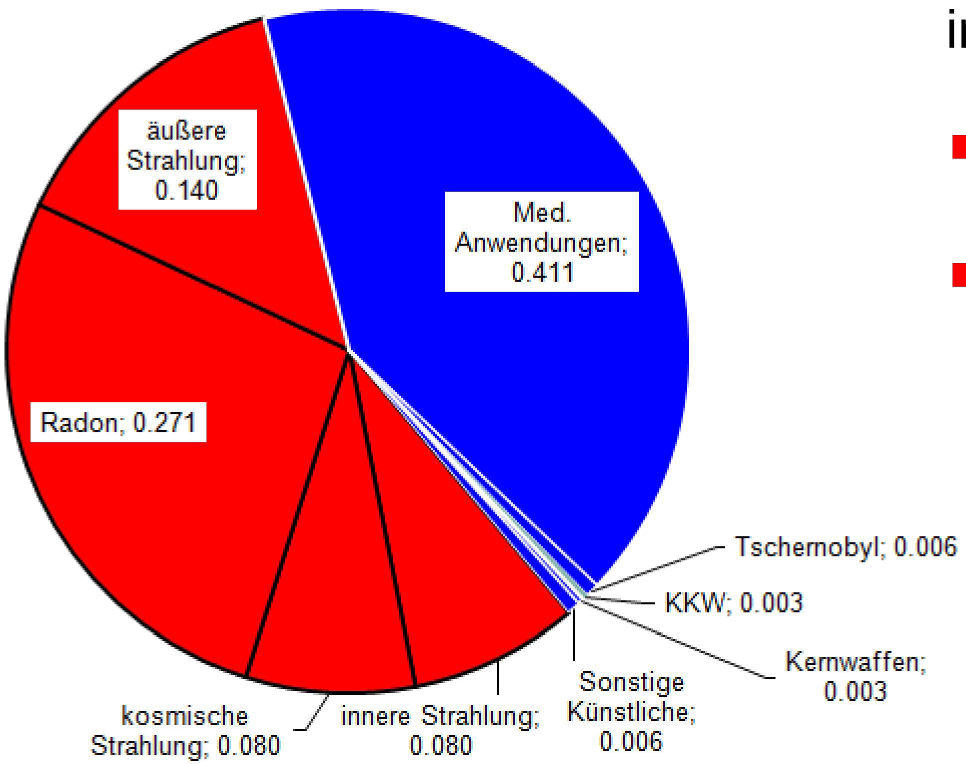
Disposal and technical demonstration of safety is an
interdisciplinary approach
including
physics, (radio-)chemistry, geology, mining, …

Open questions

- Convincing the public: NIMBY
- Safety vs. Retrievability / Reversibility
  - Reasons for retrieving the HLW:
    - Unexpected behavior of the disposal (in which time frame?)
    - Resources (Pu)
    - Improved waste management techniques (research?)
  - Monitoring
  - Preservation of knowledge, knowhow, techniques, …
- Future human actions

Risk by nuclear waste disposal

- Average natural dose rate in Germany: 2.4 mSv/a (variation 1.4 -5.7 mSv/a)



Safety requirements for a disposal in Germany:

- Exposure rate by probable scenarios: 1.8 – 7.1 ‰
- Low probable scenarios: 1.8 – 7.1 % of natural burden

Thank you for your attention