UFZ HELMHOLTZ Centre for Environmental Research

AMPHOS 21 SCIENTIFIC AND STRATEGIC ENVIRONMENTAL CONSULTING

Near field evolution of a spent fuel repository in an argillaceous rock formation and impact on radionuclide migration

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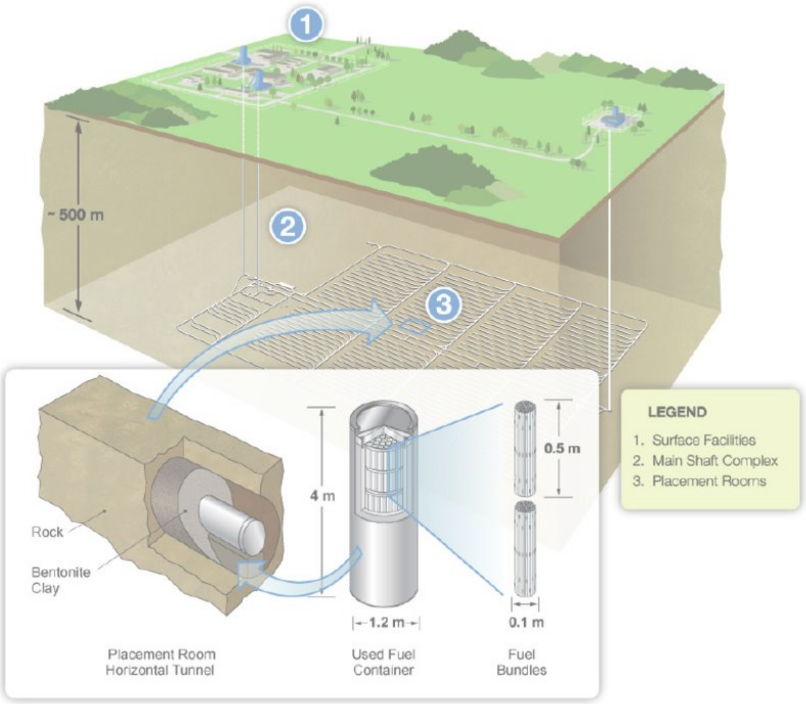
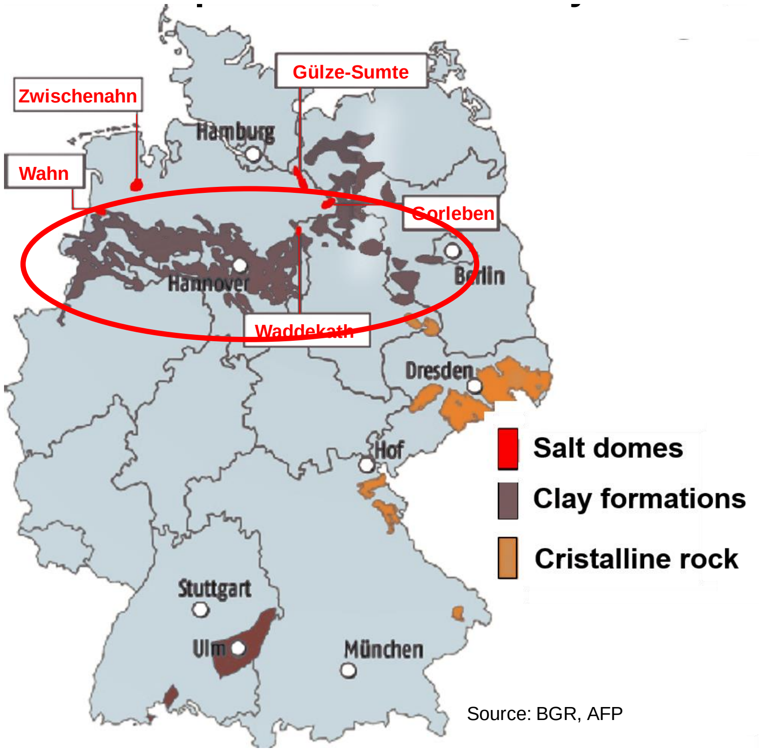


Introduction

Clay formations in Germany



North German Basins:

- Risk of deep glacial trench formation

  ➔ Emplacement depth ~ 600-700 m
  (Extended mine construction/lining required)

- Geological formation:
  Lower cretaceous, upper jurassic

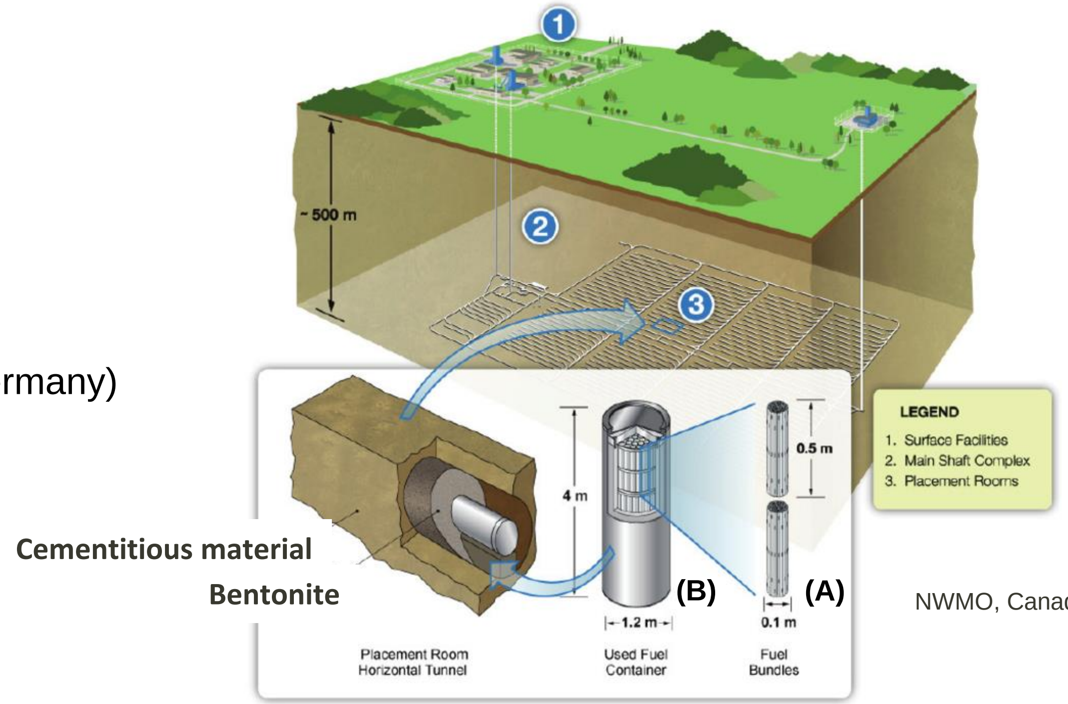
Introduction

Repository concept in Clayrock for Spent Nuclear Fuel

– **Multibarrier system**

A) SNF (Spent Nuclear Fuel)
B) the steel container (Pollux 10)
C) bentonite (MX-80)
D) cement plug (Low pH concrete)
E) cement liner (OPC)
F) surrounding clay rock (North Germany)

Radionuclide migration



NWMO, Canada

Reactive solute transport

Numerical concept

Total mass balance of water and chemical species in variable saturated porous media

$$\boldsymbol{\omega}\frac{\partial c_i}{\partial t} = -\psi\, \boldsymbol{q_l}\cdot\nabla c_i + \nabla\cdot(\psi\, \boldsymbol{D_l}\cdot\nabla c_i) - c_i\nabla\cdot(\boldsymbol{\rho_l D_l}\nabla\omega_l^w) - f_{ext}^w c_i + f_{ext}^w c_i^* + \boldsymbol{\omega} r_{eq} + \boldsymbol{\omega} r_{kin}$$

$\boldsymbol{q_l} = 0$ Advection

Diffusion

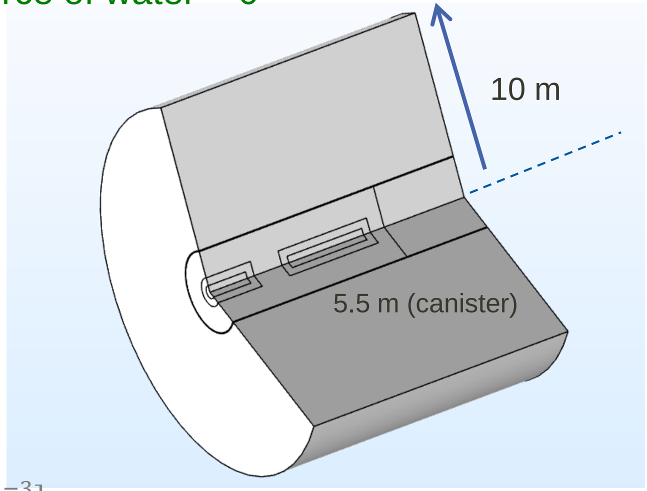
$\nabla\omega_l^w = 0$

Source of water = 0

$$\boldsymbol{D_l} = \boldsymbol{D_{disp}} + \boldsymbol{D_{dif}} = \phi\, S_l D_s \tau = \phi\, D_s\tau = \phi\; 1\; 10^{-9}\;\tau$$

$$\boldsymbol{\omega} = \phi\, S_l \boldsymbol{\rho_l}\omega_l^w = \boldsymbol{\rho_l}\phi$$

$$\psi = \boldsymbol{\rho_l}\omega_l^w$$



ϕ : *porosity* $[m^3m^{-3}]$ $\boldsymbol{S_l}$: *liquid saturation* $[m^3m^{-3}]$ $\boldsymbol{\rho_l}$: *liquid density* $[kg\ m^{-3}]$

Geochemistry

Nuclear waste (Radionuclide inventory)

55 GWd/tHM burn-up after **50 years** in PWR (UO_2 + MOX)

(Gigawatts-day /tonnes)

Initially: **$UO_{2\ (matrix)}$**: Activity: 7.74 $x10^{15}$ Bq/tHM

Max. $[\ ]_{aq}$

	Half life (a)	**Act.** (Bq/tHM)	**Mass** (g/tHM)	**Redox state**	**Key Chem. Parameters**	**Solub.** (mol/Kgw)
^{243}Am	7 370	$1.71x10^{12}$	$2.32x10^{2}$	+III	pH, HCO_3^-	$7.88x10^{-6}$
^{230}Th	75 380	$1.07x10^{9}$	1.40	+IV	HCO_3^-	$3.50x10^{-9}$
^{238}U	$4.468x10^{6}$	$1.14x10^{10}$	$9.17x10^{5}$	+IV, +VI	HCO_3^-, Eh	$3.79x10^{-9}$
^{237}Np	$2.144x10^{6}$	$6.18x10^{10}$	$2.37x10^{3}$	+IV,	HCO_3^-	$1.01x10^{-9}$
^{242}Pu	373 300	$1.49x10^{11}$	$1.02x10^{3}$	+III, +IV, +V	pH, HCO_3^-, Eh	$1.91x10^{-9}$
^{99}Tc	211 000	$8.01x10^{11}$	$1.27x10^{3}$	+IV, +VII	Eh	?

Geochemistry

Pore water compositions

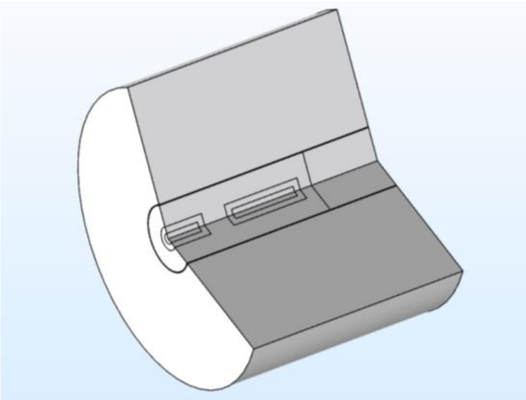
	Host rock	Bentonite	OPC Cement	Low pH concrete	Processes
Na	3.70	2.12	$5.45x10^{-10}$	$1.91x10^{-2}$	Exchange
K	$5.44x10^{-3}$	$8.91x10^{-2}$	$1.25x10^{-9}$	$3.42x10^{-2}$	Exchange
Ca	0.23	0.93	$2.76x10^{-7}$	$5.24x10^{-3}$	Exchange/precip/dissolu
Cl	4.22	4.22	$1.00x10^{-9}$	$1.00x10^{-10}$	--
SO_4	$2.59x10^{-2}$	$1.89x10^{-2}$	$5.01x10^{-10}$	$3.05x10^{-2}$	Precipitation/dissolu
Si	$1.80x10^{-4}$	$1.80x10^{-4}$	$2.02x10^{-3}$	$2.02x10^{-3}$	Precipitation/dissolu
Al	$4.99x10^{-9}$	$4.99x10^{-9}$	$1.44x10^{-4}$	$1.44x10^{-4}$	Precipitation/dissolu
Fe_{Total}	$4.98x10^{-5}$	$4.98x10^{-5}$	$5.45x10^{-8}$	$5.45x10^{-8}$	Precipitation/dissolu
Mg	0.114	0.114	$3.73x10^{-7}$	$3.73x10^{-7}$	Exchange/precip/dissolu
CO_3	$8.92x10^{-5}$	$4.34x10^{-5}$	$1.50x10^{-5}$	$1.50x10^{-5}$	Precipitation/dissolu
Eh (mV)	-111 (Fe(II)/Fe(III))	-111 (Fe(II)/Fe(III))	-27 mV	-27 mV	Precipitation/dissolu
pH	7.77	8.1	13.31	10.68	Precipitation/dissolu/surf

Modelling results

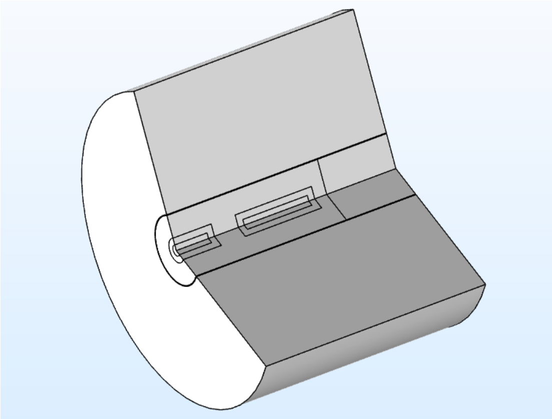
Canister Corrosion

$3\ Fe\ (s) + 4\ H_2O = \mathbf{Fe_3O_4\ (s)} + 4H_2$ → (Magnetite, but $FeCO_3$,…)

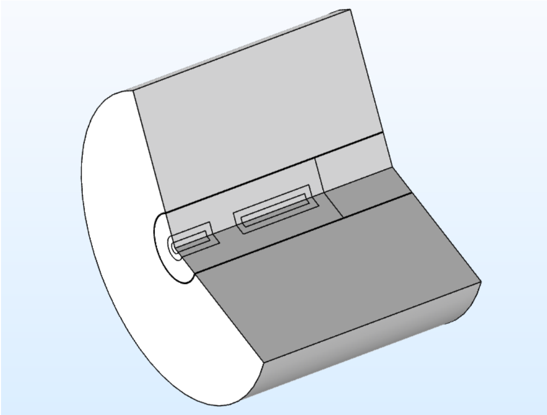
100 years



1 000 years



10 000 years



$3Fe\ (s) + 6\ H_2O = 3\ Fe^{2+} + 6OH^- + 3H_2$

$3\ Fe^{2+} + 6\ OH^- = Fe_3O_4\ (s) + 2H_2O + H_2$

$$r_n = M_w m\ \boldsymbol{k_n}\ A$$

0.005 µm yr^{-1}

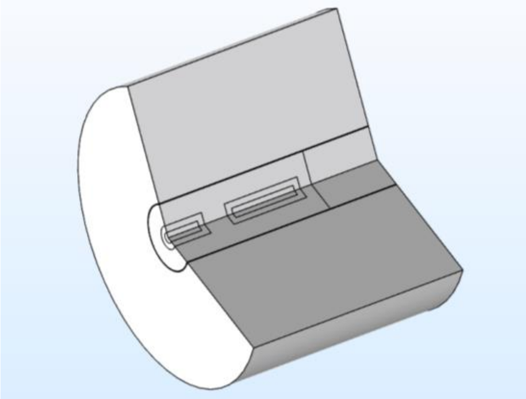
$\boldsymbol{r_n}: reaction\ rate\ [mol\ s^{-1}]$

Modelling results

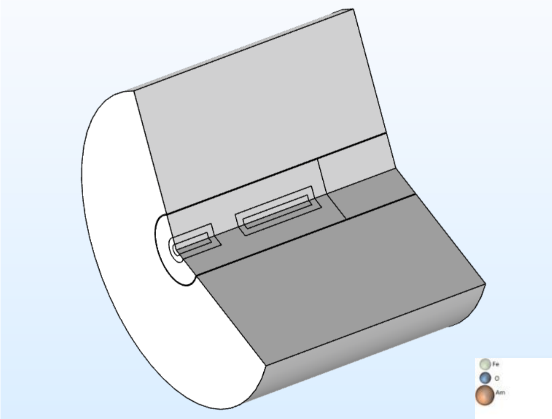
Transport of a reactive tracer (^{243}Am)

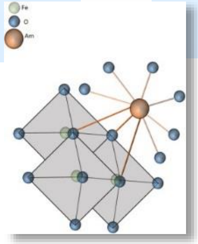
3 Fe (s) + 4 H_2O = **Fe_3O_4 (s)** + 4H_2 → ($Magnetite, but FeCO_3$,…)

100 years

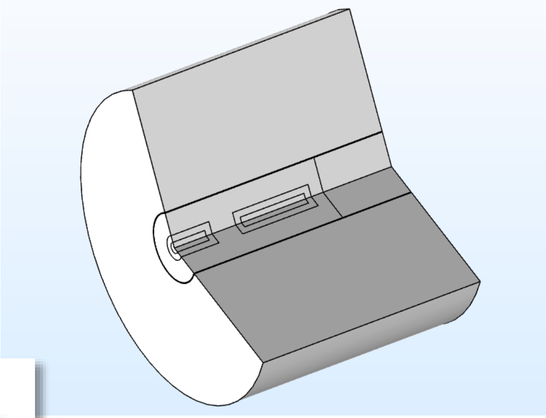


1 000 years





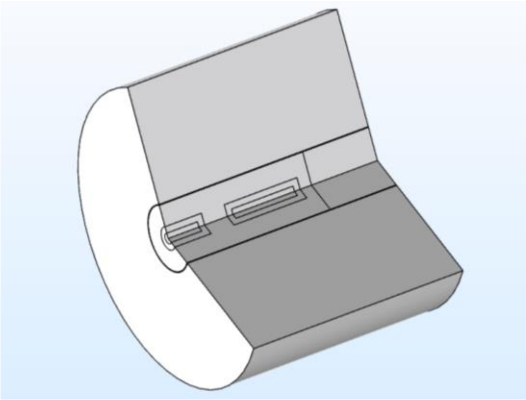
10 000 years



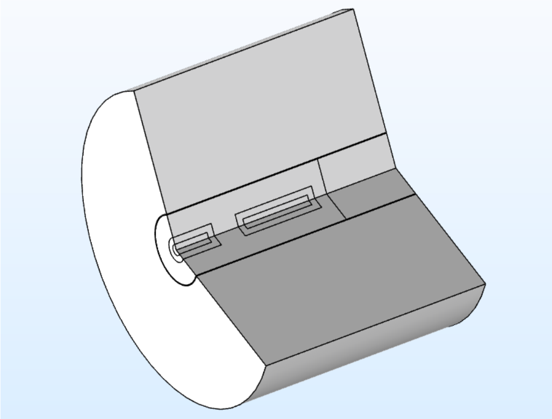
Modelling results

Transport of a non-reactive tracer ($^{36}Cl^{-}$)

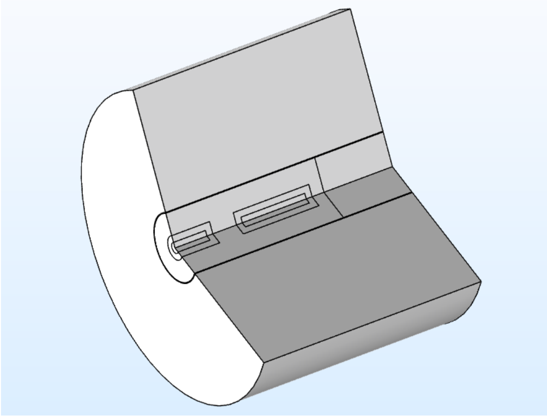
100 years



1 000 years



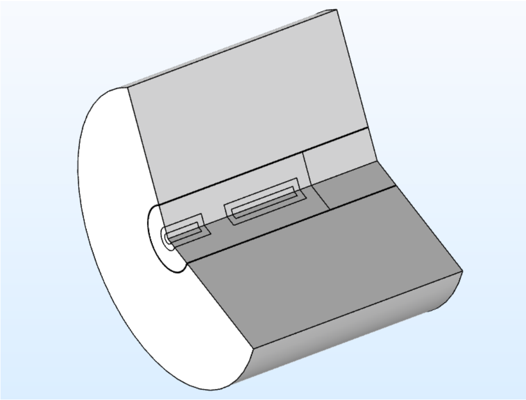
10 000 year



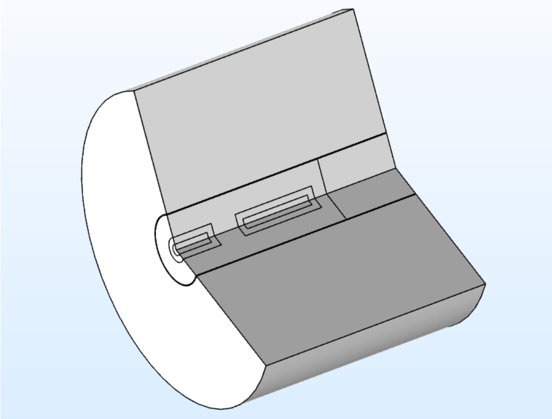
Modelling results

Processes at the bentonite barrier

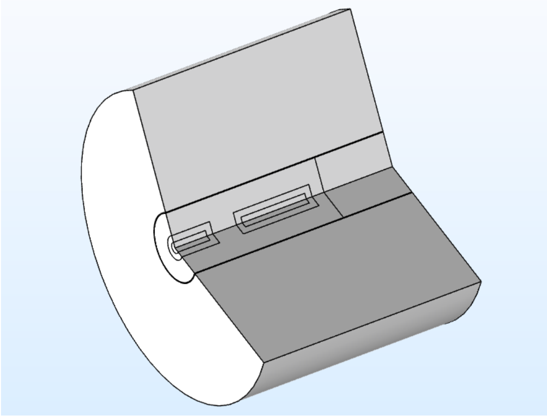
100 years



1 000 years



10 000 year



Conclusions

- **Numerical tools need to be improved** in order to handle coupled mass transport with reactions in long term simulations (10 000 years) and big domains (9000 mesh elements, 5000 nodes)

- Geochemical processes happening next to the **canister/bentonite** interface are the most relevant for most of the radionuclides present in the waste inventory

- Processes happening in the cement liner seems to be less relevant for radionuclide migration, but important for barrier integrity studies.

Thank you for your attention!

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German Federal Ministry of Education and Research (Grant 02NUK053A) and the Initiative and Networking Fund of the Helmholtz Association (Grant SO-093) within the iCross project.

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