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

Cementitious material
Bentonite

NWMO, Canada
Reactive solute transport

Numerical concept

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

\[
\omega \frac{\partial c_i}{\partial t} = -\psi q_l \cdot \nabla c_i + \nabla \cdot (\psi D_l \cdot \nabla c_i) - c_i \nabla \cdot (\rho_l D_l \nabla \omega^w_l) - f_{ext}^w c_i + f_{ext}^w c_i^* + \omega_{eq} + \omega_{kin}
\]

\[q_l = 0\]

Advection

\[D_l = D_{disp} + D_{dif} = \phi S_l D_s \tau = \phi D_s \tau = \phi \times 10^{-9} \tau\]

Diffusion

\[\nabla \omega^w_l = 0\]

Source of water = 0

\[\phi: \text{porosity} \ [m^3 m^{-3}] \quad S_l: \text{liquid saturation} \ [m^3 m^{-3}] \quad \rho_l: \text{liquid density} \ [kg m^{-3}]\]
Geochemistry

Nuclear waste (Radionuclide inventory)

55 GWd/tHM burn-up after 50 years in PWR (UO₂ + MOX)
(Gigawatts-day /tonnes)

Initially: UO₂(matrix): Activity: 7.74 x10¹⁵ Bq/tHM

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Half life (a)</th>
<th>Act. (Bq/tHM)</th>
<th>Mass (g/tHM)</th>
<th>Redox state</th>
<th>Key Chem. Parameters</th>
<th>Solub. (mol/Kgw)</th>
</tr>
</thead>
<tbody>
<tr>
<td>²⁴³Am</td>
<td>7 370</td>
<td>1.71x10¹²</td>
<td>2.32x10²</td>
<td>+III</td>
<td>pH, HCO₃⁻</td>
<td>7.88x10⁻⁶</td>
</tr>
<tr>
<td>²³⁰Th</td>
<td>75 380</td>
<td>1.07x10⁹</td>
<td>1.40</td>
<td>+IV</td>
<td>HCO₃⁻</td>
<td>3.50x10⁻⁹</td>
</tr>
<tr>
<td>²³⁸U</td>
<td>4.468x10⁶</td>
<td>1.14x10¹⁰</td>
<td>9.17x10⁵</td>
<td>+IV, +VI</td>
<td>HCO₃⁻, Eh</td>
<td>3.79x10⁻⁹</td>
</tr>
<tr>
<td>²³⁷Np</td>
<td>2.144x10⁶</td>
<td>6.18x10¹⁰</td>
<td>2.37x10³</td>
<td>+IV,</td>
<td>HCO₃⁻</td>
<td>1.01x10⁻⁹</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>373 300</td>
<td>1.49x10¹¹</td>
<td>1.02x10³</td>
<td>+III, +IV,  +V</td>
<td>pH, HCO₃⁻, Eh</td>
<td>1.91x10⁻⁹</td>
</tr>
<tr>
<td>⁹⁹Tc</td>
<td>211 000</td>
<td>8.01x10¹¹</td>
<td>1.27x10³</td>
<td>+IV, +VII</td>
<td>Eh</td>
<td>?</td>
</tr>
</tbody>
</table>
## Geochemistry

### Pore water compositions

<table>
<thead>
<tr>
<th></th>
<th>Host rock</th>
<th>Bentonite</th>
<th>OPC Cement</th>
<th>Low pH concrete</th>
<th>Processes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>3.70</td>
<td>2.12</td>
<td>5.45x10⁻¹⁰</td>
<td>1.91x10⁻²</td>
<td>Exchange</td>
</tr>
<tr>
<td>K</td>
<td>5.44x10⁻³</td>
<td>8.91x10⁻²</td>
<td>1.25x10⁻⁹</td>
<td>3.42x10⁻²</td>
<td>Exchange</td>
</tr>
<tr>
<td>Ca</td>
<td>0.23</td>
<td>0.93</td>
<td>2.76x10⁻⁷</td>
<td>5.24x10⁻³</td>
<td>Exchange/precip/dissolu</td>
</tr>
<tr>
<td>Cl</td>
<td>4.22</td>
<td>4.22</td>
<td>1.00x10⁻⁹</td>
<td>1.00x10⁻¹⁰</td>
<td>--</td>
</tr>
<tr>
<td>SO₄</td>
<td>2.59x10⁻²</td>
<td>1.89x10⁻²</td>
<td>5.01x10⁻¹⁰</td>
<td>3.05x10⁻²</td>
<td>Precipitation/dissolu</td>
</tr>
<tr>
<td>Si</td>
<td>1.80x10⁻⁴</td>
<td>1.80x10⁻⁴</td>
<td>2.02x10⁻³</td>
<td>2.02x10⁻³</td>
<td>Precipitation/dissolu</td>
</tr>
<tr>
<td>Al</td>
<td>4.99x10⁻⁹</td>
<td>4.99x10⁻⁹</td>
<td>1.44x10⁻⁴</td>
<td>1.44x10⁻⁴</td>
<td>Precipitation/dissolu</td>
</tr>
<tr>
<td>Fe₉₅₉</td>
<td>4.98x10⁻⁵</td>
<td>4.98x10⁻⁵</td>
<td>5.45x10⁻⁸</td>
<td>5.45x10⁻⁸</td>
<td>Precipitation/dissolu</td>
</tr>
<tr>
<td>Mg</td>
<td>0.114</td>
<td>0.114</td>
<td>3.73x10⁻⁷</td>
<td>3.73x10⁻⁷</td>
<td>Exchange/precip/dissolu</td>
</tr>
<tr>
<td>CO₃</td>
<td>8.92x10⁻⁵</td>
<td>4.34x10⁻⁵</td>
<td>1.50x10⁻⁵</td>
<td>1.50x10⁻⁵</td>
<td>Precipitation/dissolu</td>
</tr>
<tr>
<td>Eh (mV)</td>
<td>-111 (Fe(II)/Fe(III))</td>
<td>-111 (Fe(II)/Fe(III))</td>
<td>-27 mV</td>
<td>-27 mV</td>
<td>Precipitation/dissolu</td>
</tr>
<tr>
<td>pH</td>
<td>7.77</td>
<td>8.1</td>
<td>13.31</td>
<td>10.68</td>
<td>Precipitation/dissolu/surf</td>
</tr>
</tbody>
</table>
Modelling results
Canister Corrosion

$$3 \text{Fe (s)} + 4 \text{H}_2\text{O} = \text{Fe}_3\text{O}_4 (s) + 4\text{H}_2$$  (Magnetite, but FeCO$_3$, …)

$$3\text{Fe}^{2+} + 6\text{OH}^- = \text{Fe}_3\text{O}_4 (s) + 2\text{H}_2\text{O} + \text{H}_2$$

$r_n = M_w m \ k_n A$

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

100 years
1 000 years
10 000 years

0.005 μm yr$^{-1}$
Modelling results

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

$$3 \text{Fe} \ (s) + 4 \text{H}_2\text{O} = \text{Fe}_3\text{O}_4 \ (s) + 4\text{H}_2$$ (Magnetite, but FeCO$_3$, …)

100 years  1 000 years  10 000 years
Modelling results
Transport of a non-reactive tracer ($^{36}\text{Cl}^-$)
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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