Modelling the thermo-hydro-chemical behavior of compacted bentonite for nuclear waste disposal

Andrés Idiart

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Outline

- Background
- THC model of EBS evolution in the KBS-3V
- Main results
- Conclusions and perspectives
Background: KBS-3V concept

1. Compacted bentonite buffer surrounding canister
2. Backfill filling deposition tunnels
Many coupled processes in reactive porous media at play

- *Radioactive decay of SNF: a significant heat source*
- *Long-term heat dissipation involved a large rock volume*
- *Fractured granite gradually saturates the EBS components*
- *Groundwater and porewater in chemical disequilibrium: alteration of the EBS*
- *Temperature increase trigger a set of chemical reactions in the system*
- *Swelling of the clay components*

Geochemical processes

- Focus on geochemical processes expected to play a role during the evolution of the repository (e.g. Arcos et al., 2008):
  - Thermal and liquid saturation (mixing) effects
  - SiO$_2$(am) formation (cementation may prevent clay swelling)
  - Anhydrite redistribution during the thermal period
  - Cation exchange: Ca increase may affect swelling ability; coupling with carbonates
  - Surface complexation (protonation-deprotonation reactions) and carbonates (calcite, siderite, dolomite): pH buffers
- Not considered: montmorillonite long-term dissolution, copper-bentonite interaction, redox reactions
- Assumption: all reactions under thermodynamic equilibrium (i.e. no reaction kinetics)
## Background: THC processes in the KBS-3V

Geochemical processes: experience from in-situ experiments

### Previous modelling efforts

<table>
<thead>
<tr>
<th>Test</th>
<th>LOT A0</th>
<th>LOT A2</th>
<th>FEBEX In situ</th>
<th>ABM1</th>
<th>ABM2</th>
<th>Mock-Up-CZ</th>
<th>TBT</th>
<th>Prototype Rep.</th>
<th>CRT</th>
</tr>
</thead>
<tbody>
<tr>
<td>In-situ site</td>
<td>Asp0 HRL</td>
<td>Asp0 HRL</td>
<td>Grimsel</td>
<td>Asp0 HRL</td>
<td>Asp0 HRL</td>
<td>CEG, CTU</td>
<td>Asp0 HRL</td>
<td>Asp0 HRL</td>
<td>Asp0 HRL</td>
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<tr>
<td>Duration (years)</td>
<td>1.5</td>
<td>1.5</td>
<td>2.5</td>
<td>2.5</td>
<td>7</td>
<td>3.75</td>
<td>4.85</td>
<td>8</td>
<td>5</td>
</tr>
<tr>
<td>Concept tested</td>
<td>KBS 3V</td>
<td>KBS 3V</td>
<td>Horizontal</td>
<td>KBS 3V</td>
<td>KBS 3V</td>
<td>KBS 3V</td>
<td>KBS 3V</td>
<td>KBS 3V</td>
<td>KBS 3V</td>
</tr>
<tr>
<td>Diameter/Length (m)</td>
<td>0.3 / 4</td>
<td>0.3 / 4</td>
<td>2.28 / 17.4</td>
<td>0.3 / 3</td>
<td>0.3 / 3</td>
<td>0.8 / 2.24</td>
<td>1.75 / 8</td>
<td>1.75 / 8</td>
<td>1.76 / 7.05</td>
</tr>
<tr>
<td>Bentonite type</td>
<td>MX80</td>
<td>MX80</td>
<td>FEBEX</td>
<td>Multiple types</td>
<td>Multiple types</td>
<td>Rokli *</td>
<td>MX80</td>
<td>MX80</td>
<td>Volclay MX80</td>
</tr>
<tr>
<td>Artificial hydration</td>
<td>gw from borehole nearby</td>
<td>gw from borehole nearby</td>
<td>Natural wetting from the rock</td>
<td>gw from borehole nearby</td>
<td>gw from borehole nearby</td>
<td>synthetic</td>
<td>gw from borehole nearby</td>
<td>Natural wetting from the rock</td>
<td>natural Na-Ca-Cl</td>
</tr>
<tr>
<td>Max temperature (°C)</td>
<td>120-150</td>
<td>120-150</td>
<td>97</td>
<td>120-140</td>
<td>120-140</td>
<td>97</td>
<td>130-150</td>
<td>85</td>
<td>75-95</td>
</tr>
<tr>
<td>Initial dry density (kg/m²)</td>
<td>1570</td>
<td>1570</td>
<td>1700</td>
<td>1700-2150</td>
<td>1700-2150</td>
<td>1760-1770</td>
<td>1700</td>
<td>1570</td>
<td>1700-1780</td>
</tr>
<tr>
<td>Initial water content (w%)</td>
<td>14</td>
<td>14</td>
<td>14</td>
<td>14</td>
<td>14</td>
<td>7</td>
<td>8</td>
<td>17</td>
<td>17</td>
</tr>
</tbody>
</table>

**Geochemical alterations observed**

- **Transformation of expandable clay changes**
  - no evidence of structural changes
  - no evidence of structural changes tentatively, anhydrite formation
  - minor transformation (illite/beidellite)
  - no evidence of structural changes
- **Anhydrite/Gypsum formation**
  - distributed near the heater
  - distributed near the heater
  - redistributed near the heater
  - redistributed near the canister
- **Cristobalite**
  - no significant redistribution
  - no significant redistribution
  - dissolved near the heater
  - dissolved near the heater
- **Calcite**
  - no clear tendency
  - measured decrease
  - no clear tendency
  - no clear tendency
- **CEC changes**
  - no change
  - extensive equilibration with gw
  - extensive equilibration with gw
  - decrease in Mg in cold part
- **Cation exchange**
  - increase in Mg near the heater
  - increase near the heater
  - increase near the heater
  - low solubility Mg-phase forms near the heater
- **Non-exchangeable Mg**
  - increase near the heater
  - increase near the heater
  - increase near the heater

 gw = groundwater; * mix: 85% Czech Ca-Mg bentonite (Rokli), 10% quartz sand; 5% graphite

**Modelled using e.g. Phreeqc, PHAST, Toughreact, Crunchflow, Comsol**
Objective - Multiphysics and Geochemistry

iMaGe
interfacing geosciences
an Amphos21 initiative
http://image-modelling.net/

Providing solutions to couple multiphysics and geochemistry
Objective - Multiphysics and Geochemistry

Projects

- Miscible and immiscible multiphase flow formulations for $\text{CO}_2$, $\text{H}_2$, and $\text{CH}_4$
- Multiphase flow and reactive transport
- Development of solid mechanics capabilities for reactive porous media

(Nardi et al. 2014)
THC model of the evolution of the EBS in the KBS-3V

Geometry: 2 distinct chemically reactive domains included

1. Buffer (MX-80 bentonite) surrounding copper canister
2. Backfill (mainly Friedland clay) filling deposition tunnels
THC model of the evolution of the EBS in the KBS-3V

Thermo-hydro-chemical boundary conditions

Heat source

Groundwater source
THC model of the evolution of the EBS in the KBS-3V

Thermo-hydro-chemical boundary conditions

<table>
<thead>
<tr>
<th>Species</th>
<th>Groundwater</th>
<th>Backfill</th>
<th>Buffer</th>
</tr>
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<tbody>
<tr>
<td>pH</td>
<td>7.385</td>
<td>7.14</td>
<td>7.146</td>
</tr>
<tr>
<td>pe</td>
<td>-2.246</td>
<td>-2.904</td>
<td>1.151</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>11</td>
<td>11</td>
<td>11</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Species</th>
<th>Totals (M) Groundwater</th>
<th>Totals (M) Backfill</th>
<th>Totals (M) Buffer</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>1.49E-03</td>
<td>5.18E-03</td>
<td>5.19E-03</td>
</tr>
<tr>
<td>Ca</td>
<td>1.62E-02</td>
<td>1.83E-02</td>
<td>1.80E-02</td>
</tr>
<tr>
<td>Cl</td>
<td>0.1134</td>
<td>0.005676</td>
<td>0.005752</td>
</tr>
<tr>
<td>Fe</td>
<td>6.45E-06</td>
<td>8.49E-05</td>
<td>4.94E-11</td>
</tr>
<tr>
<td>K</td>
<td>4.86E-04</td>
<td>2.18E-03</td>
<td>1.32E-03</td>
</tr>
<tr>
<td>Mg</td>
<td>7.50E-03</td>
<td>1.53E-02</td>
<td>6.09E-03</td>
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<tr>
<td>Na</td>
<td>0.0766</td>
<td>0.1347</td>
<td>0.1579</td>
</tr>
<tr>
<td>S</td>
<td>4.79E-03</td>
<td>9.69E-02</td>
<td>9.85E-02</td>
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<tr>
<td>Si</td>
<td>1.45E-03</td>
<td>1.42E-03</td>
<td>1.42E-03</td>
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<tr>
<td>Z</td>
<td>8.46E-11</td>
<td>3.06E-10</td>
<td>9.89E-11</td>
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<tr>
<td>Ca/Na</td>
<td>0.211</td>
<td>0.136</td>
<td>0.114</td>
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</table>

<table>
<thead>
<tr>
<th>Phase</th>
<th>Formula</th>
<th>Saturation Indexes</th>
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<tbody>
<tr>
<td>Anhydrite</td>
<td>Ca(SO4)</td>
<td>-1.0 0 0</td>
</tr>
<tr>
<td>Calcite</td>
<td>CaCO3</td>
<td>0.0 0 0</td>
</tr>
<tr>
<td>Dolomite</td>
<td>CaMg(CO3)2</td>
<td>-0.3 0 0.4</td>
</tr>
<tr>
<td>Gypsum</td>
<td>CaSO4·2H2O</td>
<td>-0.7 0.3 0.3</td>
</tr>
<tr>
<td>Pyrite</td>
<td>Fe2Z</td>
<td>0.0 0 0.0</td>
</tr>
<tr>
<td>Siderite</td>
<td>Fe(CO3)</td>
<td>-1.2 0.0 -6.3</td>
</tr>
<tr>
<td>SiO2</td>
<td>SiO2</td>
<td>0.0 0 0.0</td>
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</tbody>
</table>

REFERENCE 2 MX-80 BENTONITE BLOCKS

<table>
<thead>
<tr>
<th>Mineral</th>
<th>data wt%</th>
<th>Mw (kg/mol)</th>
<th>mol/l_medium</th>
<th>Mv (cm3/mol)</th>
<th>VOL FRAC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Smectite</td>
<td>88.2</td>
<td>0.4767</td>
<td>3.1456</td>
<td>232.5</td>
<td>73.135%</td>
</tr>
<tr>
<td>Illite</td>
<td>0.1</td>
<td>0.4018</td>
<td>0.0042</td>
<td>141.34</td>
<td>0.060%</td>
</tr>
<tr>
<td>Calcite</td>
<td>0.2</td>
<td>0.1001</td>
<td>0.0340</td>
<td>36.934</td>
<td>0.125%</td>
</tr>
<tr>
<td>Biotite</td>
<td>0.233</td>
<td>0.3983</td>
<td>0.0099</td>
<td>0</td>
<td>0.000%</td>
</tr>
<tr>
<td>Dolomite</td>
<td>0.0144</td>
<td>0.1844</td>
<td>0.0000</td>
<td>64.365</td>
<td>0.000%</td>
</tr>
<tr>
<td>Quartz</td>
<td>3.467</td>
<td>0.0601</td>
<td>0.9809</td>
<td>22.69</td>
<td>2.226%</td>
</tr>
<tr>
<td>Cristobalite</td>
<td>0.1</td>
<td>0.0601</td>
<td>0.0283</td>
<td>25.74</td>
<td>0.073%</td>
</tr>
<tr>
<td>Tridymite</td>
<td>0.0</td>
<td>0.0601</td>
<td>0.0000</td>
<td>0</td>
<td>0.000%</td>
</tr>
<tr>
<td>Gypsum</td>
<td>0.367</td>
<td>0.1722</td>
<td>0.0362</td>
<td>74.69</td>
<td>0.271%</td>
</tr>
<tr>
<td>Goethite</td>
<td>0.0</td>
<td>0.0889</td>
<td>0.0000</td>
<td>20.82</td>
<td>0.000%</td>
</tr>
<tr>
<td>Hematite</td>
<td>0.033</td>
<td>0.1597</td>
<td>0.0035</td>
<td>30.3</td>
<td>0.011%</td>
</tr>
<tr>
<td>Pyrite</td>
<td>0.867</td>
<td>0.1200</td>
<td>0.1229</td>
<td>23.94</td>
<td>0.294%</td>
</tr>
<tr>
<td>Plagioclase</td>
<td>2.867</td>
<td>0.2622</td>
<td>0.1859</td>
<td>0</td>
<td>0.000%</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>2.4</td>
<td>0.2622</td>
<td>0.1556</td>
<td>0</td>
<td>0.000%</td>
</tr>
</tbody>
</table>
THC model of the evolution of the EBS in the KBS-3V

Different domains for different physics

Maintaining the compatibility of the different variables over the common domains
Main results: TH processes

Temperature after 25 years
Point A: cold buffer spot
Point B: hot buffer spot

Liquid degree of saturation
Main results: Chemical reactions

Cementation due to silica redistribution

Porosity changes due to chemical reactions
Main results: Chemical reactions (2)

Cation exchange is the most important geochemical process in the EBS

Cation exchange: Ca to Na

Point A: cold buffer spot
Point B: hot buffer spot
Conclusions

- The iCP framework has been developed as a tool that aims facilitating the interaction between engineers and (geo)chemists
- The tool has been used to revisit the thermo-hydro-chemical evolution of the EBS in a KBS-3V system
- Overall, small geochemical changes are observed even after $10^5$ years:
  - Redistribution of secondary minerals
  - Cementation is not significant
  - Increase in Ca in the montmorillonite exchanger

Perspectives

- On-going developments aim at studying other sources of coupling in this system (multi-phase flow, swelling)
- THMC tools are necessary to assess the effect on safety of e.g. hydrogen generation due to steel corrosion processes in deep geological repositories
Thank you

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