CHEMICAL EVOLUTION OF BENTONITE BUFFER IN A FINAL REPOSITORY OF SPENT NUCLEAR FUEL DURING THE THERMAL PHASE – MASTER OF SCIENCE THESIS

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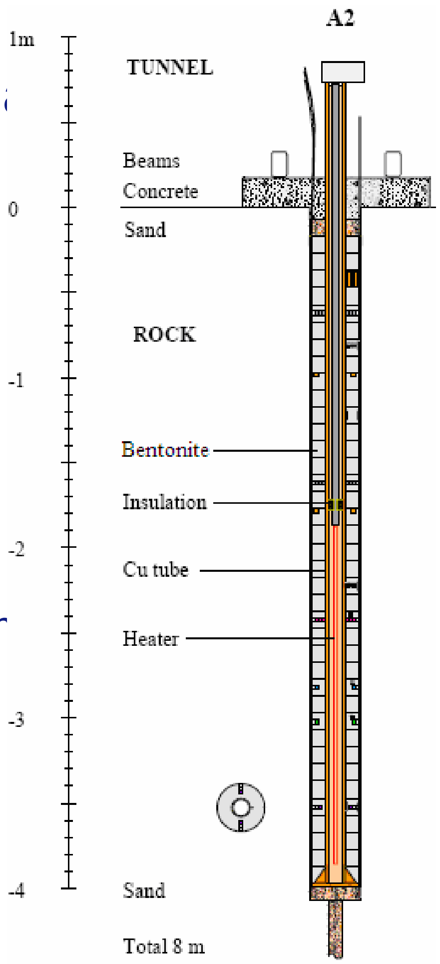
Experimental concept

- LOT A2-parcel test at Äspö HRL (6 years) used as a reference
- Adverse conditions (120-150 ^{o}C)
- Dimensions:

  hole height 8m
  depth 450 m
  water pressure 1.2 MPa
  hole diameter 300 mm
  copper tube length 4,7 m
  copper inner diameter 100 mm and wall thickness 4 mm
  bentonite cylinder diameter 280 mm.



Model and the phenomena

- PetraSim interface and TOUGHREACT EOS3 was used to model the reactive unsaturated transport processes in 1-D and the grid was pitched at uniform intervals.
- Model includes:
  - Solid, liquid (water) and gaseous phases (water/air) Chemical equilibrium of aqueous phase
  - Mineral dissolution/precipitation (kinetics)
  - Cation exchange
  - Transport processes (advection, diffusion, thermal gradient)
  - Unsaturated flow by Darcy's Law
    - Van Genuchten capillary pressure and relative permeability parameters.
  - Constant diffusion coefficient
  - Porosity changes are included but do not affect the flow of water

Excluded phenomena and chosen limitations

- Swelling and mechanical phenomena (constant material parameters) not included
- Time is limited to 10 years
- Montmorillonite is solid base and not dissolving
- Surface complexation (protonation/deprotonation) is not included
- Different kind of (pore)waters are not applied

Used Flow and Transport equations

General continuity equation: $\dfrac{\partial G^i}{\partial t} = -div\ f^i + q^i$

Water: $G_w = \phi(S_l\rho_l X_{wl} + S_g\rho_g X_{wg})$ $\quad \overrightarrow{f_w} = X_{wl}\rho_l\overrightarrow{u_l} + X_{wg}\rho_g\overrightarrow{u_g}$ $\quad q_w = q_{w_l} + q_{wg}$

$Air:\quad G_a = \phi(S_l\rho_l X_{al} + S_g\rho_g X_{ag})$ $\quad \overrightarrow{f_a} = X_{al}\rho_l\overrightarrow{u_l} + X_{ag}\rho_g\overrightarrow{u_g}$ $\quad q_a = q_{al} + q_{ag} + q_{ar}$

$Heat:\ e = (1-\phi)\rho_R C_R T + \phi\sum_{\beta=l,g} S_\beta\rho_\beta u_\beta$ $\quad \vec{q} = -k\nabla T + \sum_{\beta=l,g} C_\beta T\rho_\beta\vec{u}$ $\quad q_h$

Chemical Components in liquid phase:

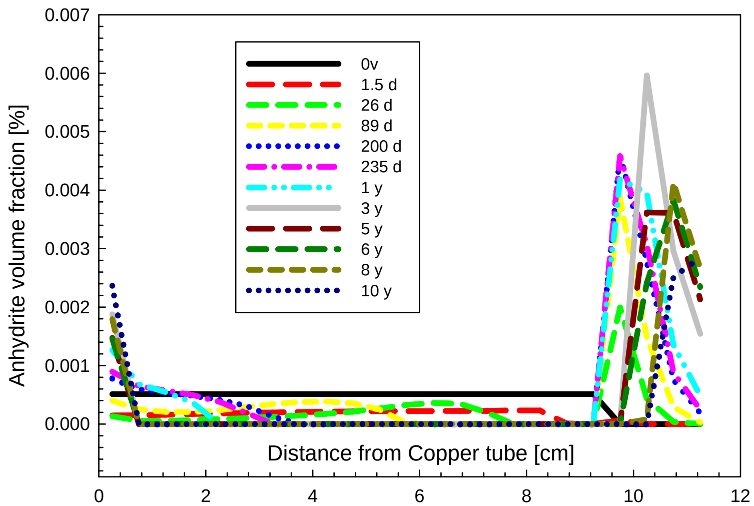
$C^i = \phi\ S_l c_{il}$ $\quad \overrightarrow{b_i} = -D_i\nabla c_{il} + \overrightarrow{u_l}c_{il}$ $\quad q_i = q_{il} + q_{is} + q_{ig}$

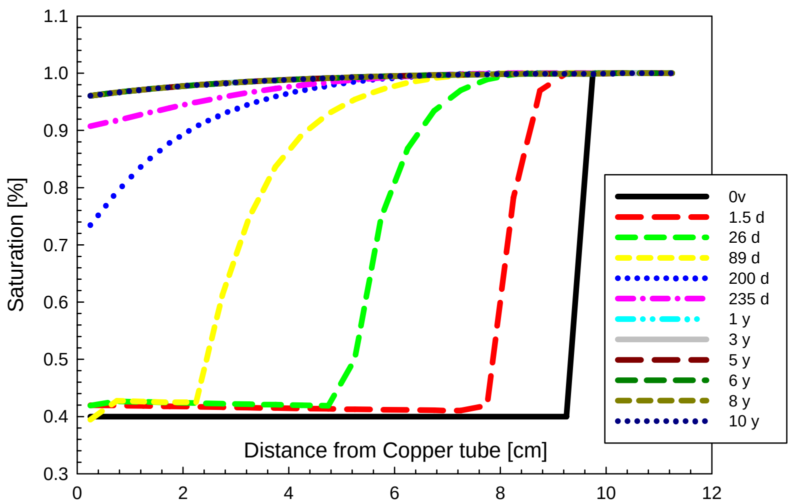
$$\vec{u} = -\mathbf{K}\nabla h = -\frac{\boldsymbol{\kappa}}{\mu}(\nabla P - \rho\vec{g}) \qquad \boldsymbol{\kappa} = \kappa_{r\beta}\boldsymbol{\kappa}_{\mathbf{a}} \qquad P = P_r + P_c$$

$\kappa_{r\beta}$ and P_c are Van genuchten functions

Results 1

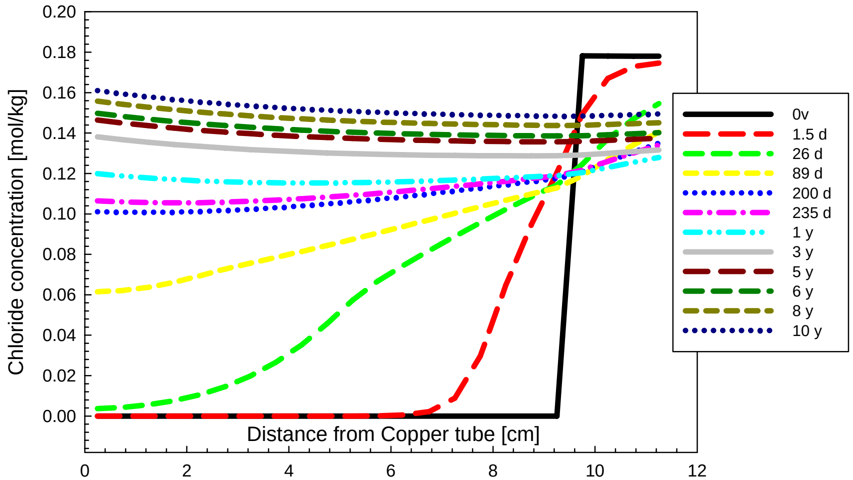
- Gypsum/Anhydrite minerals are redistributed (Gypsum dissolves and anhydrite precipitates (left figure) near the heater and the bentonite-fracture interface)
- Saturation in this model happens almost fully by liquid water (right figure)

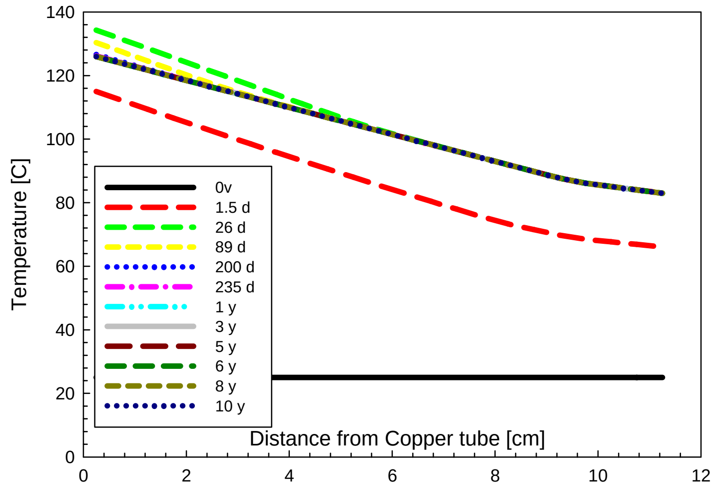




Results 2

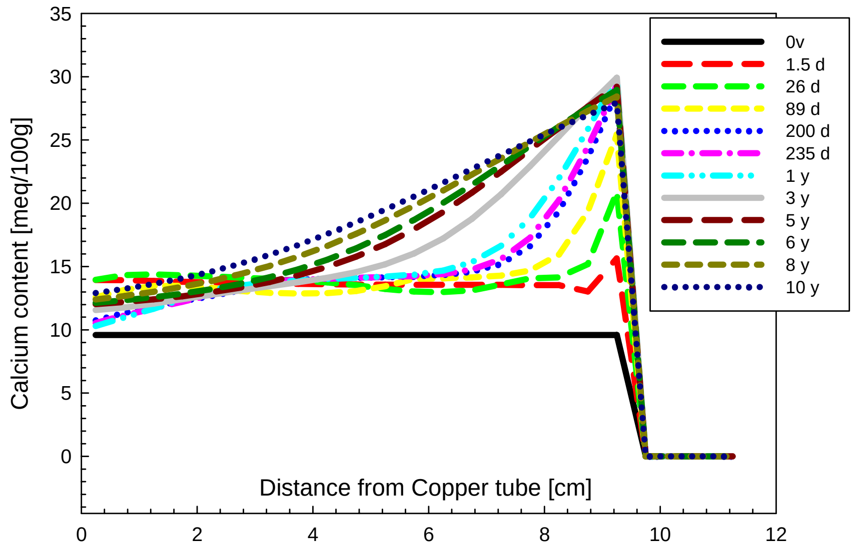
- Chloride concentrations match with this saturation
- Experimental results are not necessarily from the fracture point and for example cation exchanger composition differs from calculated
- There are still some problems with the newer version of TOUGHREACT which have to be consulted with Tianfu Xu
- Since the thermal gradient in the model exceeded that in real repository, processes are expected to be much slower in standard condition holes.

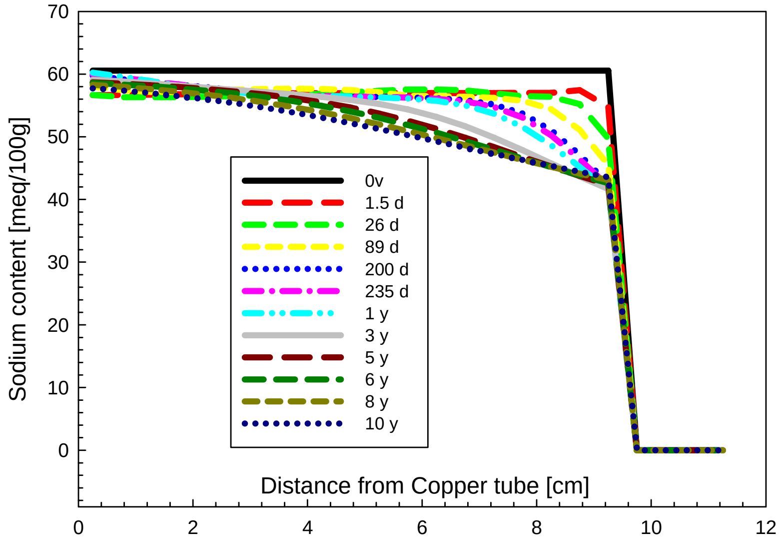




Results 3

- Calcium diffuses from groundwater to bentonite which causes the change from Na-Montmorillonite to Ca-Montmorillonite.
- All other ion concentrations in cation exchanger gets smaller except calcium.
- Calcium and sodium content in the cation exchanger is shown below.





Conclusions

- Majority of the results appear to be qualitatively correct
- Results from the model are from the fracture position, thus the changes are at maximum at this point. (Experimental results are not necessarily from fracture point)
- Cation exchanger coefficients are from laboratory tests and not thoroughly studied for compacted bentonite, thus more experimental data is needed.
- It should be studied more how exactly does the water intrude the bentonite during saturation

Future Aspects

- Further study is needed to confirm the validity of the results.
- Results should be compared to other modeling programs
- More experimental data is needed
- Including mechanical phenomena and surface complexation to models will increase the reliability of the results
- Comparing initially saturated and unsaturated states and results to each other
- Moving from 1-D to 2-D and 3-D problems.