COMSOL Multiphysics TOUGHREACT Numerrin Comparison in Some Modelling Tasks of Spent Nuclear Fuel Disposal

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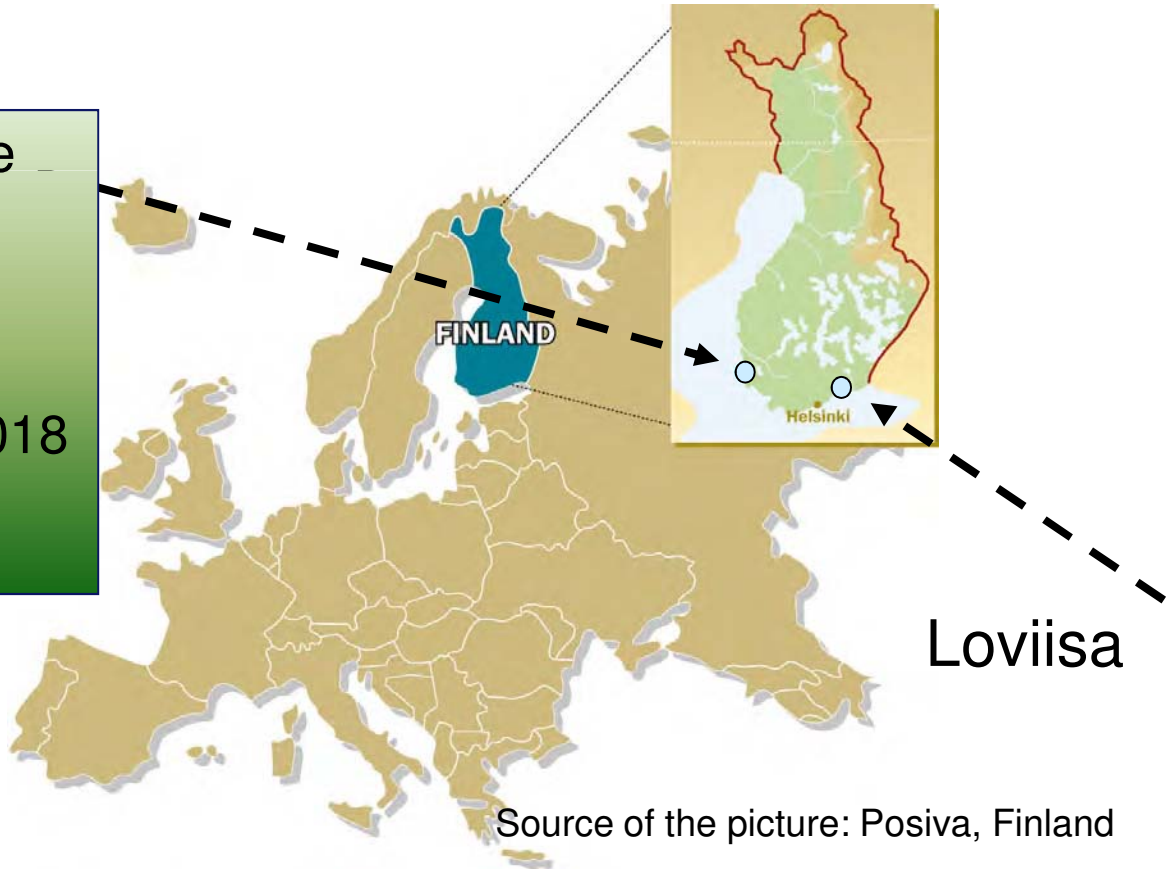
- **Nuclear power plants in Finland**
  - TVO Oy, Olkiluoto: two operating units, one under construction, one license
  - Fortum Oyj, Loviisa: two operating units
  - Fennovoima Oy, site open: one license
- Nuclear power has future in Finland

- **Olkiluoto** is the Posiva Oy's site for spent nuclear fuel disposal
- Construction license will be applied 2012
- Operation license application 2018
- Fully operational 2020 !?

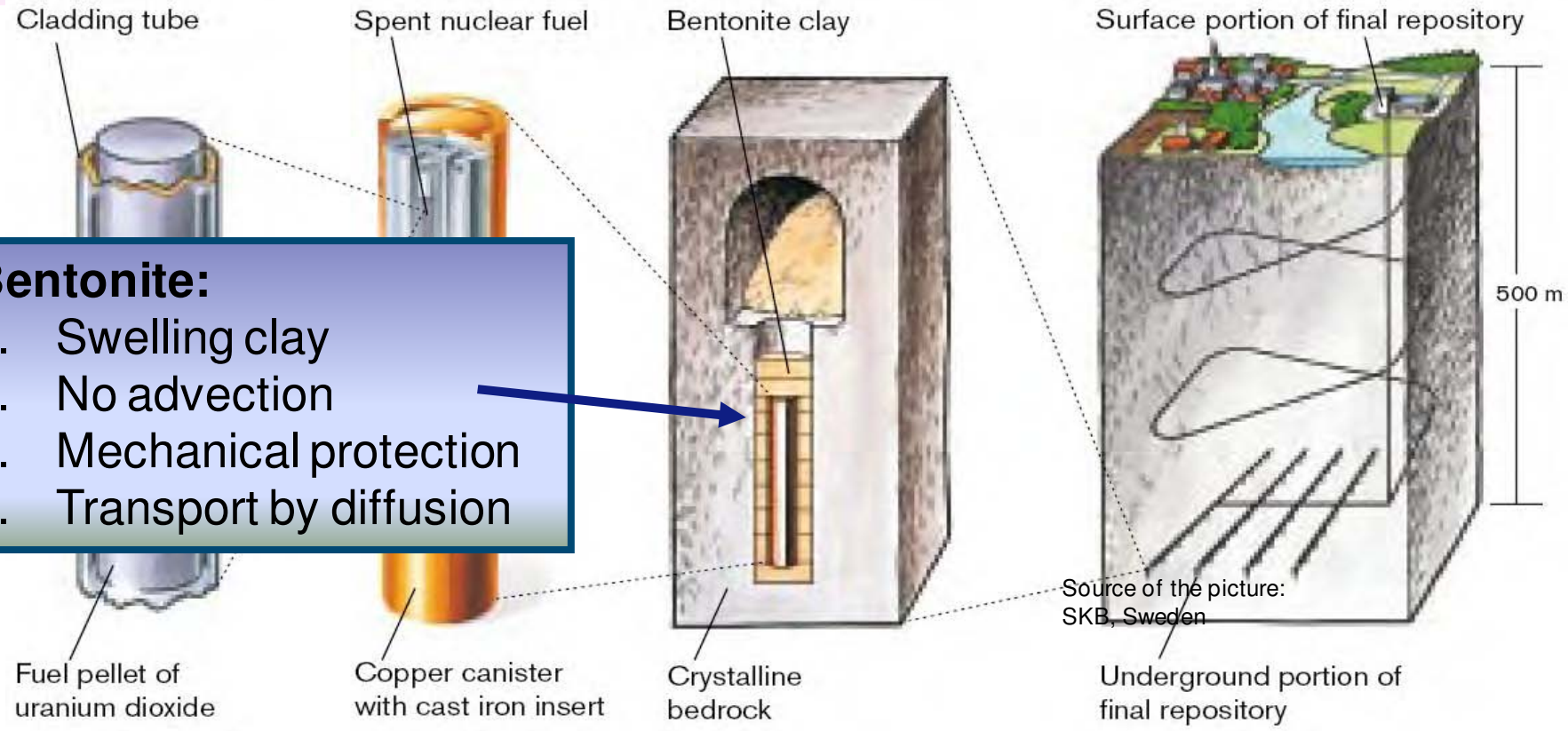


Source of the picture: Posiva, Finland

KBS-3V Concept

- KBS-3V proposed to be used in Finland like in Sweden
- An essential part of the whole system is bentonite buffer
- Bentonite may wet, swell, deform, dissolve and react chemically

Bentonite:
1. Swelling clay
2. No advection
3. Mechanical protection
4. Transport by diffusion



GOALS

- Typically, mass transport in the bentonite buffer occurs only by molecular diffusion. In some special cases, like initial saturation, convection is an important transport mechanism.
- During wetting the main problem lies in the proper and accurate calculation of the variably saturated flow field under temperature gradient.
- This work includes modelling
  - the initial saturation (wetting) of the bentonite buffer,
  - cation exchange between Na and Ca, and
  - mass transport of Na, Ca and Cl.

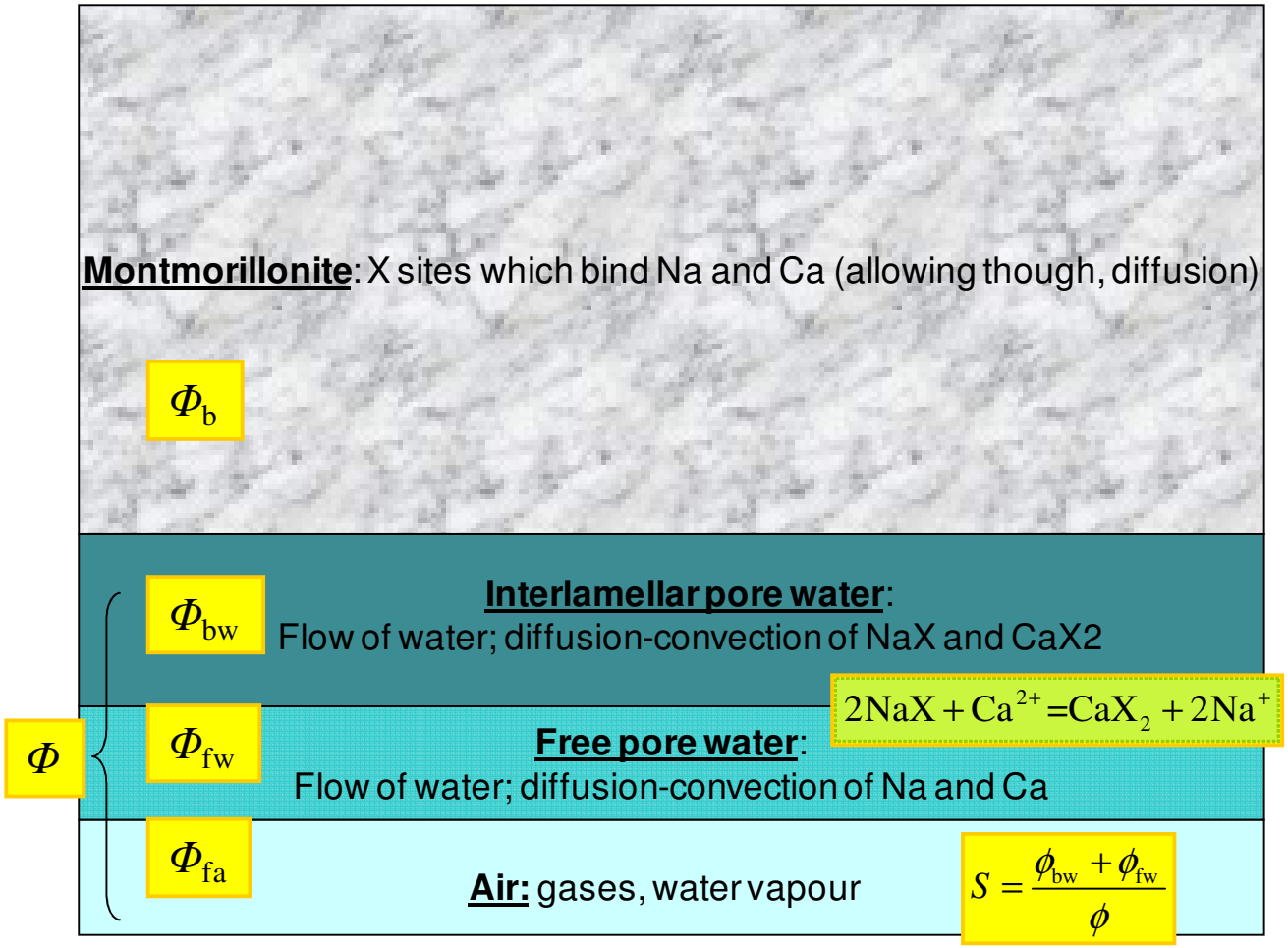
MODEL CONCEPT

- Compacted bentonite consists of montmorillonite (b), water (w), and air (a)
- Part of water is free (fw) and other part is bound on montmorillonite surfaces (bw)
- The chemical model is simple: sodium (Na), calcium (Ca), chloride (Cl) and cation exchange site (X). Na^+ and Ca^{2+} form complexes NaX and CaX_2 with X^-:

ϕ_i volume fraction of component *i*: $\sum_{i=\mathrm{b,bw,fw,fa}} \phi_i = 1 = \phi_\mathrm{b} + \phi$

ϕ volume fraction of all fluids: $\phi = \phi_\mathrm{bw} + \phi_\mathrm{fw} + \phi_\mathrm{fa}$

γ ratio of volume fractions of free and total water volume: $\gamma = \phi_\mathrm{fw} / (\phi_\mathrm{bw} + \phi_\mathrm{fw})$



GOVERNING EQUATIONS: Cation exchange

Reaction at equilibrium:

$$2\mathrm{NaX} + \mathrm{Ca}^{2+} = \mathrm{CaX}_2 + 2\mathrm{Na}^{+}$$

$$\frac{\phi_{\mathrm{bw}}}{\phi_{\mathrm{b}}}\frac{\alpha\gamma_0^2 K_{\mathrm{NaCa}}}{2\rho_{\mathrm{b}} CEC} = \frac{c_{\mathrm{CaX2}} c_{\mathrm{Na}}^2}{c_{\mathrm{NaX}}^2 c_{\mathrm{Ca}}}$$

K_{NaCa} selectivity constant (=0.63mol/L)

γ_z activity coefficient for ion of charge z

α=1 000 L/m^3

Mass balance:

$$\overline{c_{\mathrm{Na}}} = \phi_{\mathrm{fw}} c_{\mathrm{Na}} + \phi_{\mathrm{bw}} c_{\mathrm{NaX}}$$

$$\overline{c_{\mathrm{Ca}}} = \phi_{\mathrm{fw}} c_{\mathrm{Ca}} + \phi_{\mathrm{bw}} c_{\mathrm{CaX2}}$$

$$\overline{c_{\mathrm{Cl}}} = \phi_{\mathrm{fw}} c_{\mathrm{Cl}}$$

Charge balance in both phases

$$c_{\mathrm{NaX}} + 2c_{\mathrm{CaX2}} = c_{\mathrm{X}}$$

$$c_{\mathrm{Na}} + 2c_{\mathrm{Ca}} = c_{\mathrm{Cl}}$$

Calculation of activity coeffiecient

$$I = \frac{1}{2\alpha}\left(\mathrm{c}_{\mathrm{Na}^+} + 4\mathrm{c}_{\mathrm{Ca}^{2+}} + \mathrm{c}_{\mathrm{Cl}^-}\right)$$

$$\lg\gamma_1 = 0.51\cdot\left[\sqrt{I}\Big/\left(1+\sqrt{I}\right) - 0.3I\right]$$

$$\lg\gamma_z = z^2 \lg\gamma_1 \quad\rightarrow\quad \gamma_2 = \gamma_1^{z^2} = \gamma_1^4$$

Kinetics

Forward: $R_1 = k_1 \frac{\gamma_0^4}{\alpha^3}\left(\frac{\phi_{\mathrm{bw}}}{\phi_{\mathrm{fw}}}\right)^2 c_{\mathrm{NaX}}^2 c_{\mathrm{Ca}}$

Backward: $R_{-1} = k_{-1}\frac{\gamma_0^2}{\alpha^3}\frac{\phi_{\mathrm{bw}}}{\phi_{\mathrm{fw}}} c_{\mathrm{CaX2}} c_{\mathrm{Na}}^2$

At equilibrium both rates are equal, $R_1 = R_{-1}$

$$\gamma_0^2 \frac{k_1}{k_{-1}}\frac{\phi_{\mathrm{bw}}}{\phi_{\mathrm{fw}}} = \frac{c_{\mathrm{CaX2}} c_{\mathrm{Na}}^2}{c_{\mathrm{NaX}}^2 c_{\mathrm{Ca}}} \Rightarrow K^r = \frac{k_1}{k_{-1}}\frac{\phi_{\mathrm{b}}}{\phi_{\mathrm{fw}}}\frac{2\rho_{\mathrm{b}} CEC}{\alpha}$$

GOVERNING EQUATIONS: Momentum

- Can be modelled by single equation: van Genuchten in diffusion form

$$\frac{\partial S}{\partial t} = \nabla \cdot \left(D_\omega \nabla S \right)$$

$$D_\omega = -p_0 \frac{\kappa_s}{\eta \phi} \frac{1-\lambda}{\lambda} \left(\sqrt{S} \left[1 - \left(1 - S^{1/\lambda} \right)^\lambda \right]^2 S^{-1/\lambda - 1} \left[1 - S^{-1/\lambda} \right]^{-\lambda} \right)$$

GOVERNING EQUATIONS: Mass transport by diffusion

- Set of five equations, own diffusivity for bound (s) and free (p) water; traditional notation followed

$$\partial_t\left(\phi_{\mathrm{fw}} c_{\mathrm{Cl}}\right) = \nabla\cdot\left[\left(\phi_{\mathrm{fw}} D_{\mathrm{p}}\nabla - \mathbf{u}_{\mathrm{f}}\right) c_{\mathrm{Cl}}\right]$$

$$\partial_t\left(\phi_{\mathrm{fw}} c_{\mathrm{Na}}\right) = \nabla\cdot\left[\left(\phi_{\mathrm{fw}} D_{\mathrm{p}}\nabla - \mathbf{u}_{\mathrm{f}}\right) c_{\mathrm{Na}}\right] + 2R$$

$$\partial_t\left(\phi_{\mathrm{bw}} c_{\mathrm{NaX}}\right) = \nabla\cdot\left[\left(\phi_{\mathrm{bw}} D_{\mathrm{s}}\nabla - \mathbf{u}_{\mathrm{b}}\right) c_{\mathrm{NaX}}\right] - 2R$$

$$\partial_t\left(\phi_{\mathrm{fw}} c_{\mathrm{Ca}}\right) = \nabla\cdot\left[\left(\phi_{\mathrm{fw}} D_{\mathrm{p}}\nabla - \mathbf{u}_{\mathrm{f}}\right) c_{\mathrm{Ca}}\right] - R$$

$$\partial_t\left(\phi_{\mathrm{bw}} c_{\mathrm{CaX}}\right) = \nabla\cdot\left[\left(\phi_{\mathrm{bw}} D_{\mathrm{s}}\nabla - \mathbf{u}_{\mathrm{b}}\right) c_{\mathrm{CaX}}\right] + R$$

COMSOL, Numerrin and TOUGHREACT

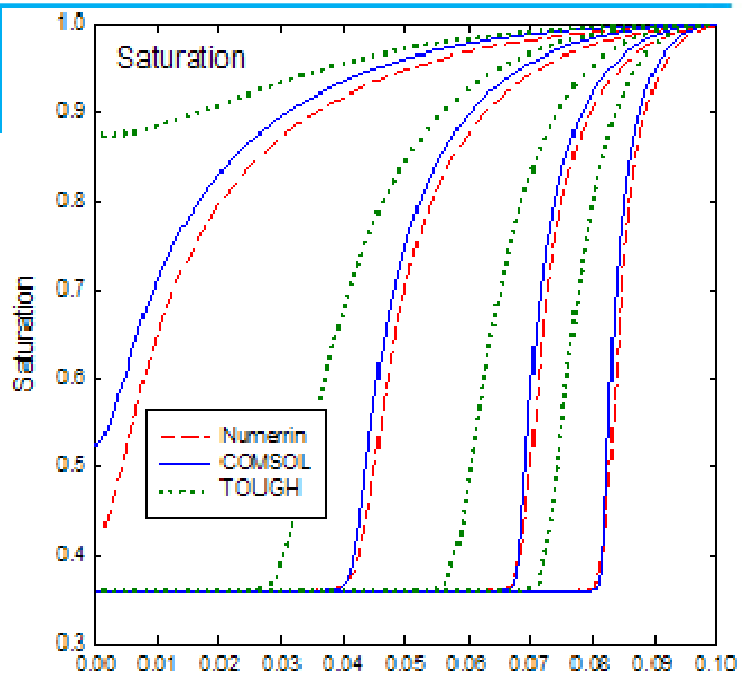
- Major advantages of COMSOL are possibility to handle complicated geometrical structures, advanced and flexible mesh generation, and options in solvers, and possibility to test how chosen numerical methods affect solutions.
- PetraSim is user interface which is interactive pre/post-processor for TOUGH2, T2VOC, TMVOC, TOUGHREACT, TOUGH-Fx/HYDRATE and TETRAD simulators. PetraSim helps users to develop models and view result of models based on nonisothermal flows of multicomponent/-phase fluids in one, two, and three-dimensional porous and fractured media. TOUGHREACT adds chemical reactions to TOUGH2. It lacks the tools to construct realistic and modern geometries.
- Numerrin is mathematical modelling software developed at Numerola Oy. In Numerrin, mathematical models are written in modelling language which gives the user full control of the models and numerical methods.

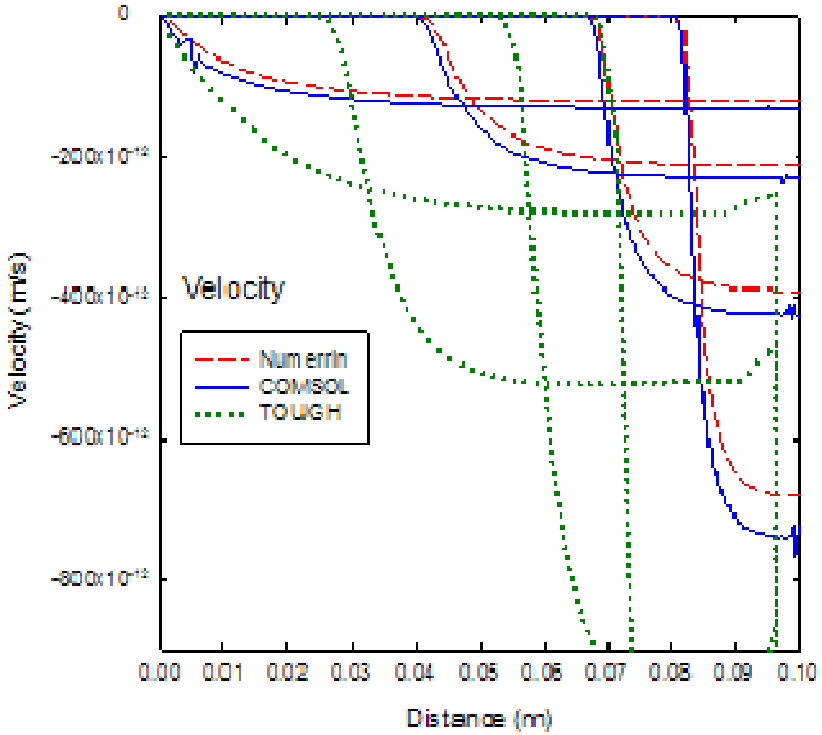
COMPARISON CASES

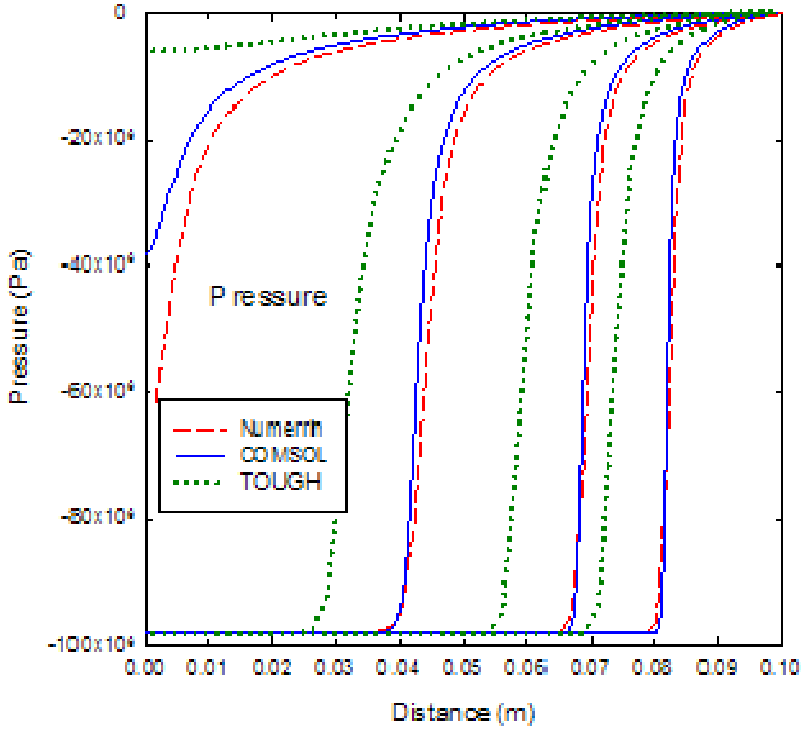
- First case was a modelling of saturation in bentonite by van Genuchten approach. The initial water saturation S in bentonite was selected to be 0.4.
- Chemical reactions and diffusion modelling case was a case where bentonite is fully saturated (i.e. S=1) but cation exchange in bentonite is happening. The system consisted only of Na-Ca exchange.Equivalent fractions for NaX and CaX2 were 0.8 and 0.2, respectively.
- The third case “combines” the two cases above, the saturation and cation exchange in bentonite. The initial saturation is 0.4 as in modelling case 1.

SATURATION RESULTS

- TOUGHREACT differed more than COMSOL or Numerrin
- Still working with this

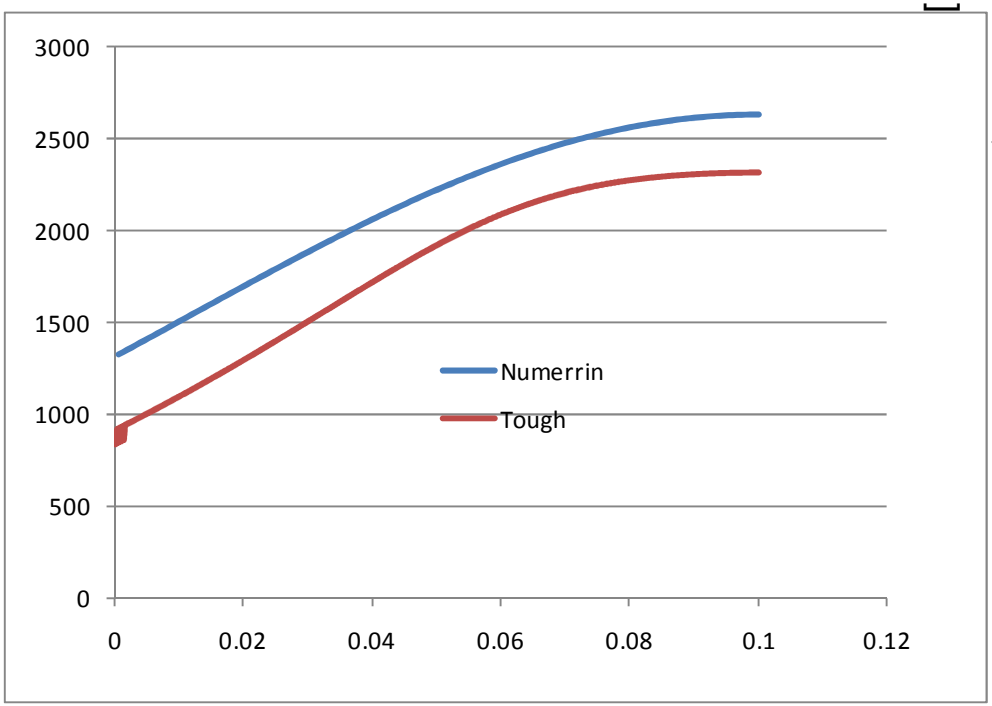


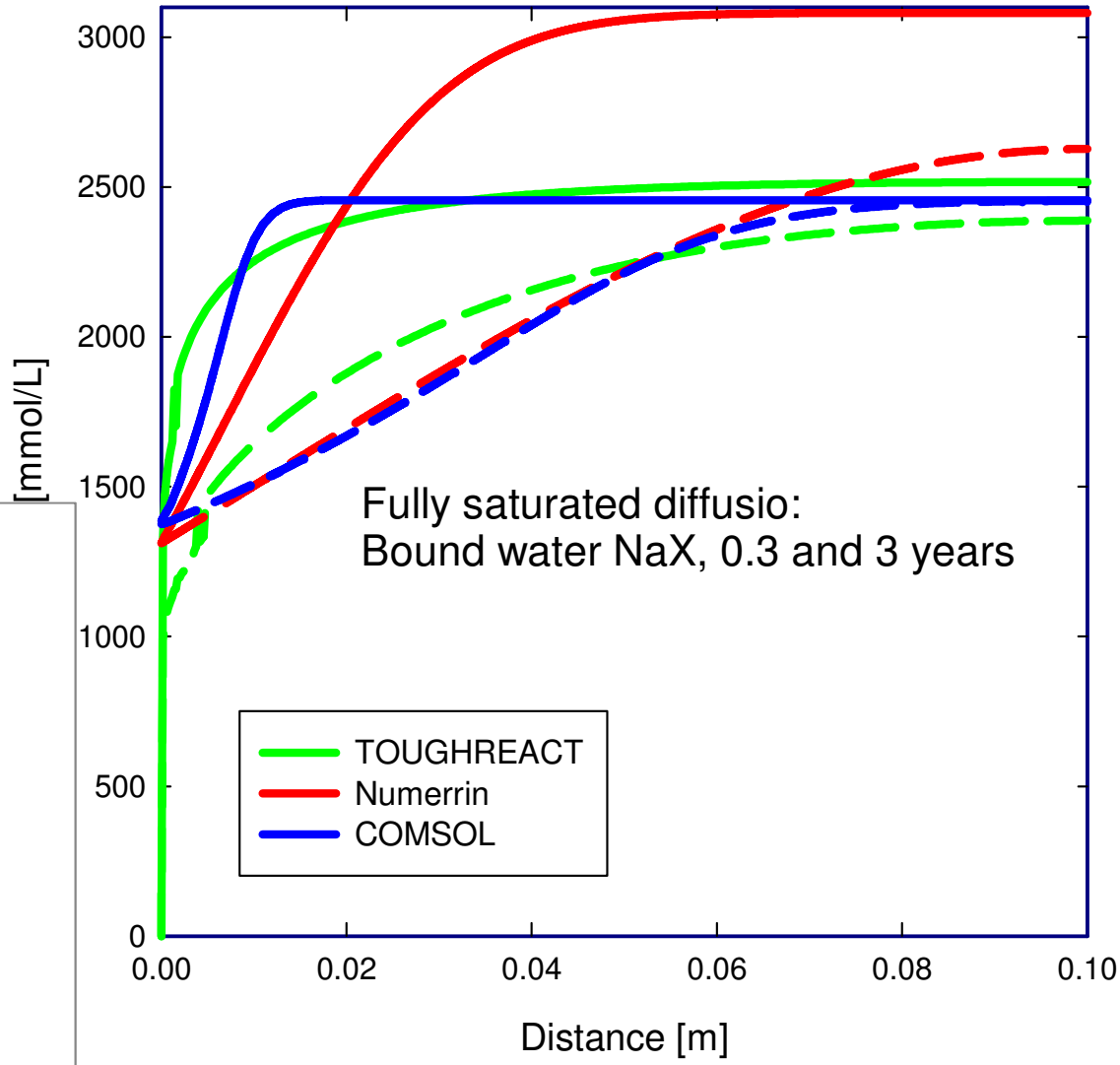




FULLY SATURATED DIFFUSION

- Much bigger differerences
- Partly due to different approaches
- Still working with this





CONCLUSIONS

- Model comparison between the applied three computing systems (COMSOL, TOUGHREACT and Numerrin) is not a straightforward task due different approaches in the model setup. Therefore, we have tried to write down all the equations and via parameterisation of those equations to create model descriptions that are at least near each other.
- Some differences were already observed for the simplest system: wetting of bentonite by van Genuchten approach. The two are cases appeared to be much more difficult and our work on them continues.

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