Reactive transport processes in compacted bentonite

Application to a prototype experiment of underground repository for nuclear waste

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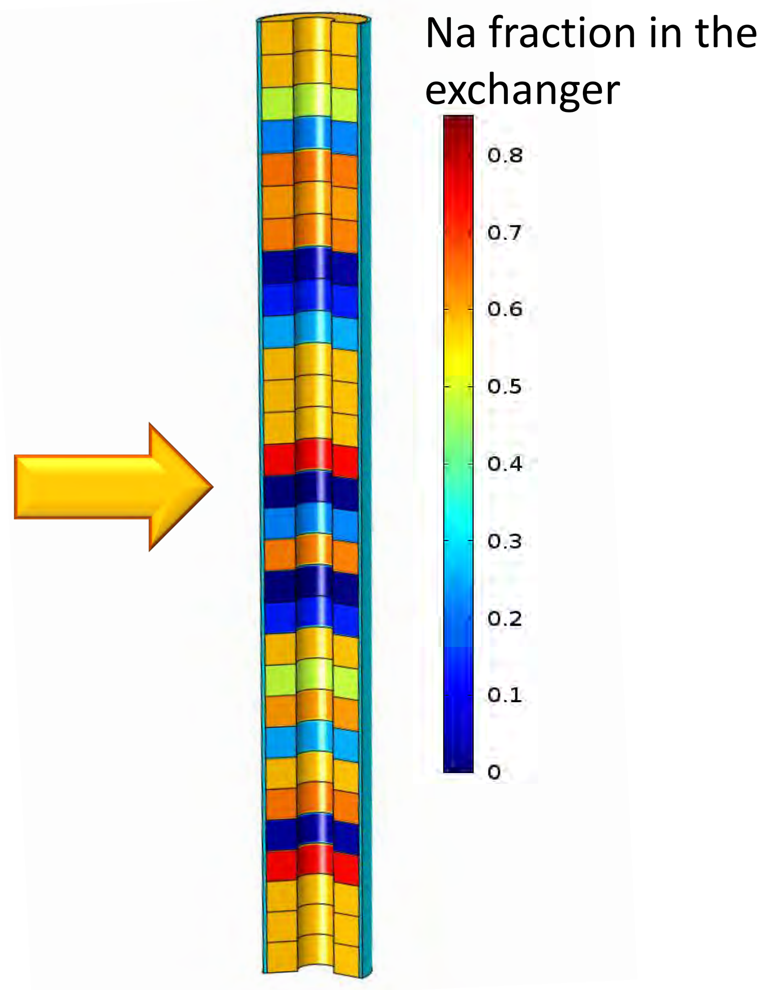
Introduction

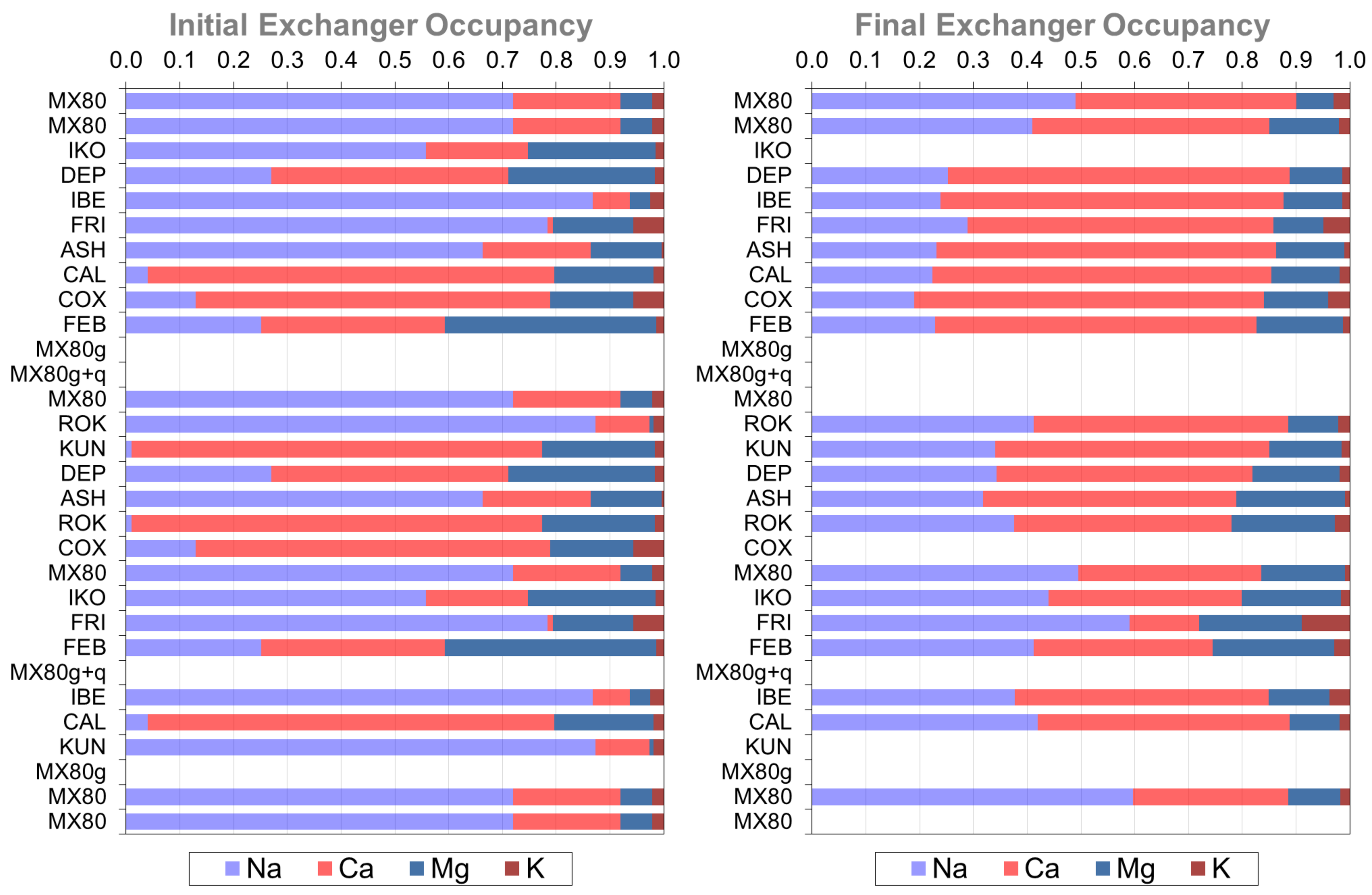
SKB, Swedish Organization for Radioactive Waste (RW) Management, is in the process of license application for the disposal of RW in a deep geological repository. Bentonite clay is planned to be used around the cylindrical RW packages as buffer material. To assess alternative clay materials, SKB started a field experiment in 2006 [1]. A package of 11 different types of compacted bentonite blocks was saturated and heated to target values, and ran for ~2.5yr.

Installation of the package



Initial block distribution

Post-mortem analysis: initially contrasting cation-exchanger compositions between bentonite blocks significantly homogenized after 880 days [1]:



Initial (before) and final (after the experiment) exchanger composition in terms of unit fraction Na, Ca, Mg and K occupancy (empty: no data available).

Objectives of the modeling study

(1) Verify whether a reactive transport model (RTM) coupling diffusion, cation-exchange & temperature evolution can explain the observed homogenisation of cation-exchanger composition

(2) Check the feasibility of implementation of a RTM into COMSOL

Conceptual model

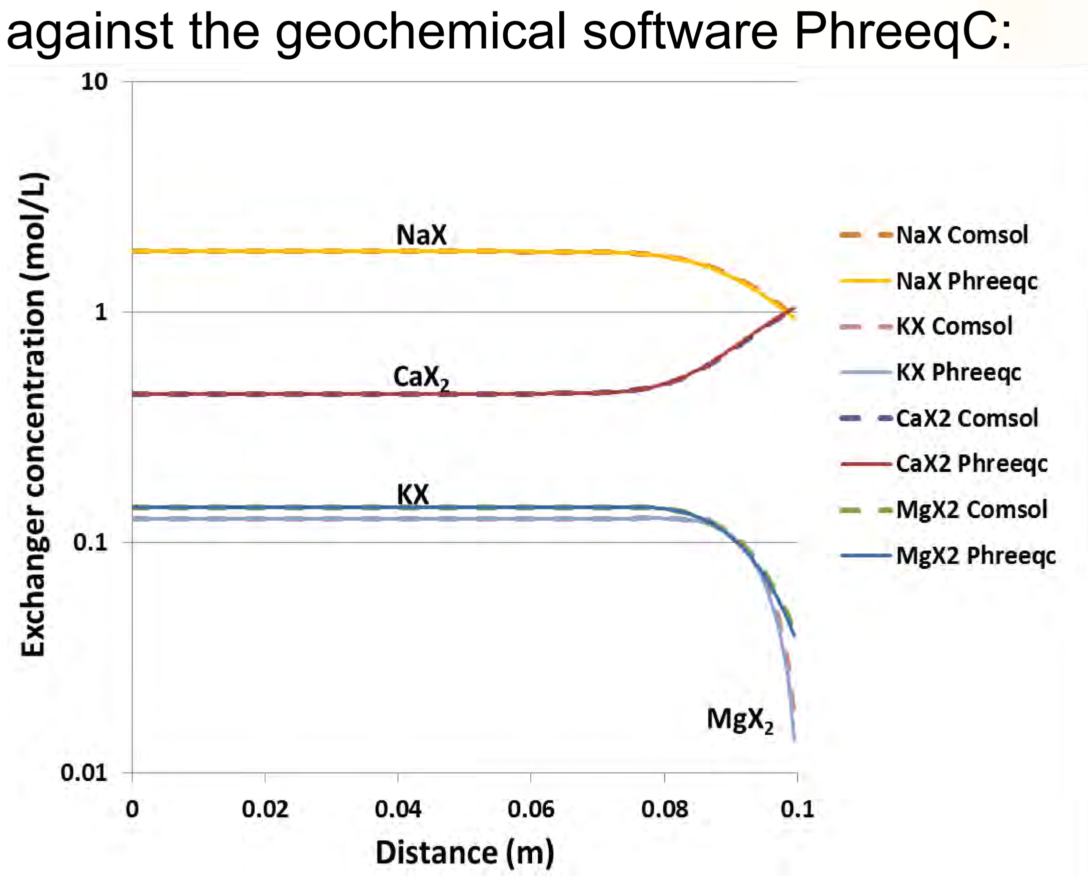
- Initial porewater composition in chemical equilibrium for each clay
- Fickian diffusion of only 5 ionic species: Na^+, Ca^{2+}, Mg^{2+}, K^+ and Cl^-
- Cation-exchange reactions (based on Gaines-Thomas convention)
- Temperature-diffusion coupling based on experimental data (temperature evolves as a function of time)
- Sensitivity scenarios considered: temperature evolution, boundary conditions (No flow, Dirichlet, combination of both)

Implementation into Subsurface Flow Module

- Diffusion using the "Solute Transport" Interface
- Cation-exchange through a system of coupled non-linear algebraic differential equations (solved using Newton-Raphson method)
- Transport-Chemistry Coupling using the approach proposed in [2] and solved using Operator Splitting (Sequential Iterative Approach)

Model verification

The RTM implemented in COMSOL was satisfactorily benchmarked against the geochemical software PhreeqC:



Comparison of exchanger composition for a 1D problem using COMSOL (dashed lines) and PHREEQC (solid lines) after 1 year . Zero and 0.1 m distance correspond to no-flux and Dirichlet boundary conditions, respectively.

Numerical results

In conjunction with a composite boundary type (see figure below) based on fracture logging measurements the simple model is capable of reproducing the experimental data with a reasonably good accuracy, especially for Ca and Na (dominant cations). A relatively poorer fit for Mg is observed in the top (although still unclear, it could be due to additional chemical reactions or altered groundwater composition).

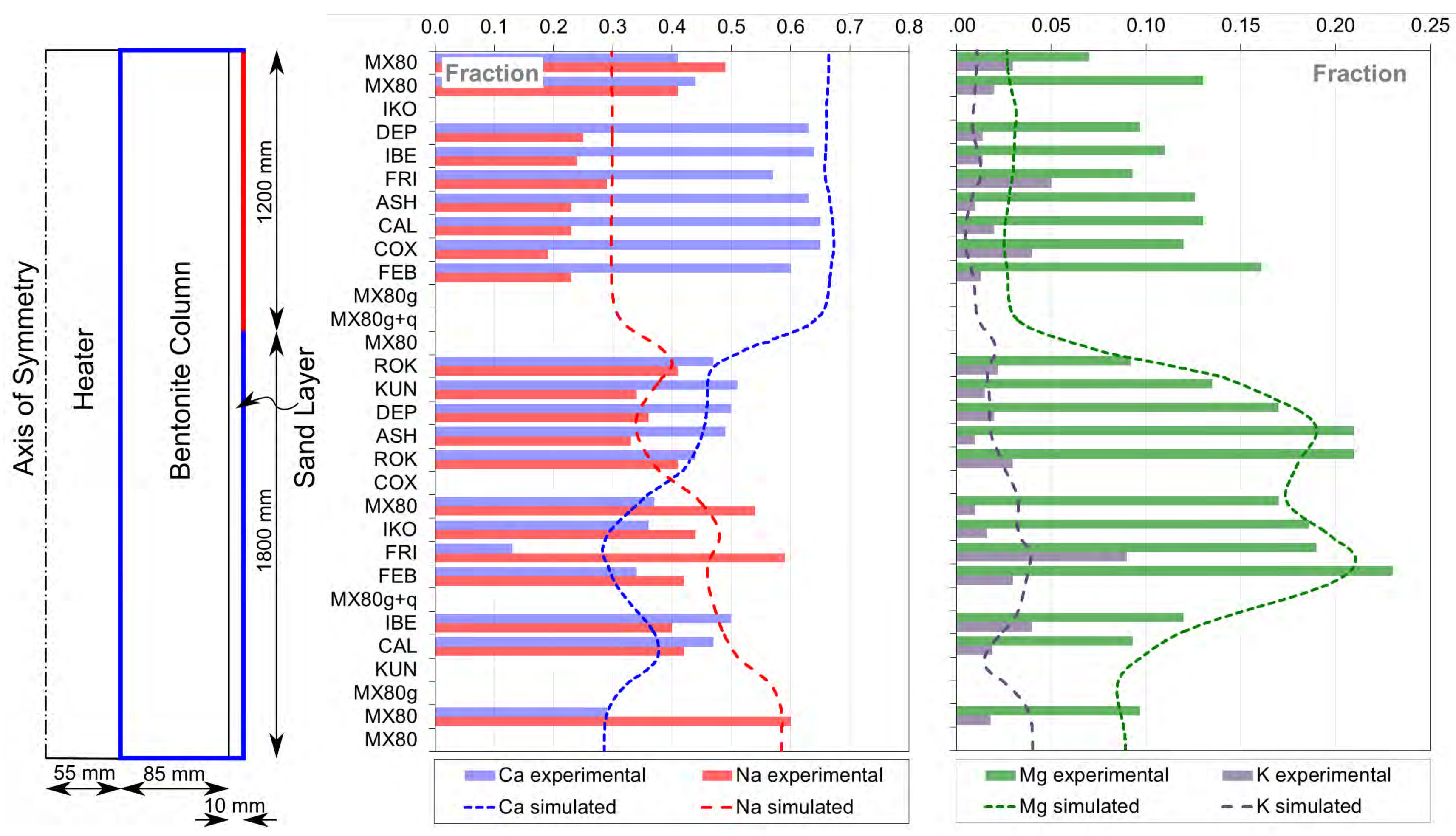
Conclusions

- A RTM has been successfully implemented into COMSOL
- The model has proved very useful to gain insight in the experiment evolution and served as a basis for discussion with experimentalists
- Comparison of measured patterns of final exchanger composition with RTM results corroborate the hypothesis of distinct regimes of reactive transport in the top and bottom sections of the bentonite column

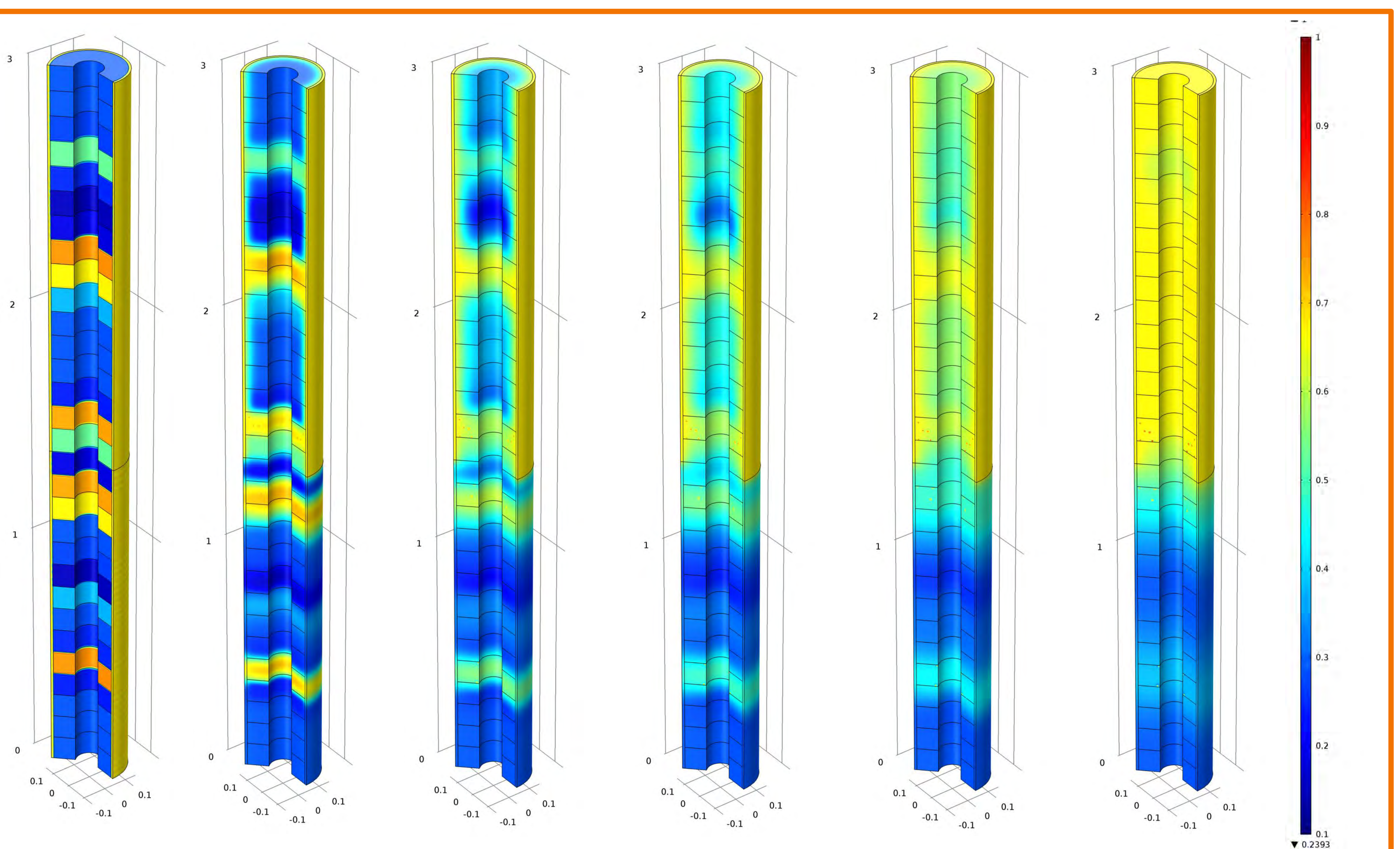
References:

[1] Svensson D. et al., 2011. Alternative Buffer Material – Status of the ongoing laboratory investigation of reference materials and test package 1. SKB TR-11-06.

[2] Saaltink M. et al., 1998. A mathematical formulation for reactive transport that eliminates mineral concentrations. Water Resources Research, 34, 1649-1656.



Left – schematic representation of the model domain and its boundaries (blue: no-flux, red: constant concentration). Right – final experimental and simulated exchanger compositions (after 880 days) across the bentonite column.



Time evolution of **Ca unit fraction** on the cation exchanger. The time increases from left (initial state) to right (final state – after 880 days).