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INVERSE MODELING OF GAS, WATER, AND HEAT FLOW IN BENTONITE/CRUSHED ROCK BACKFILL

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ABSTRACT

Swelling clays play a major role in current concepts for the underground disposal of high-level nuclear waste in deep geological formations. In one of the multi-barrier concepts for preventing the escape of radioactive substances from a high-level nuclear waste repository, the barrier consists of a copper container, compacted bentonite as buffer and backfill (the engineered barrier), and the repository host rock. Corrosion of the copper canister and radiolysis of water both produce hydrogen. When the buffer and backfill are saturated with water and the permeability of the bentonite is reduced by swelling, any hydrogen that is produced can accumulate in the space between the container and the engineered barrier. This will result in pressures exceeding the entry pressure of the bentonite and passage of gas through the engineered barrier. An experimental program was developed to investigate the thermal and hydraulic properties of the buffer and backfill under conditions expected to exist in a permanent repository for radioactive waste. Water retention curves were measured from low to high saturation using a pressure cell and a thermohygrometer. Non-isothermal drainage experiments were also designed. The measured water outflow, pressure and temperature values were used for inverse modeling using iTOUGH2. The capillary pressure curves obtained with iTOUGH2 were consistent with those obtained with the pressure cell and thermohygrometer. The influence of salinity on the capillary pressure curves was investigated.

INTRODUCTION

The Äspö Hard Rock Laboratory, located on an island in southern Sweden, is one of the underground test laboratories designed to investigate the storage of high-level radioactive waste. The high-level waste will be stored in canisters with an outer, 10-cm-thick copper wall and an inner steel lining. Storage methods are being tested at a depth of approximately 500 m in crystalline basement rock. The temperatures due to radioactive decay are expected to reach a maximum of 90 °C at the outer surface of the canisters.

The canisters will be enclosed in a buffer of bentonite; the repository drifts will be backfilled with a bentonite/crushed rock mixture. The repository host rock is the third element of the multi-barrier concept (Fig. 1).

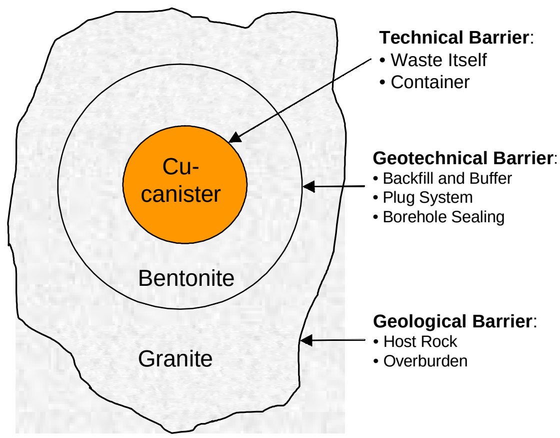

Figure 1. Multi-barrier concept for the underground disposal of HLW.

Bentonite provides the hydraulic, chemical, and mechanical properties required for a buffer and backfill material (Fig. 1). The sodium bentonite used for the buffer has a dry density of 1.62 g/cm^3 and a pore ratio of 0.71; it contains 26 % water when saturated. The 30 % sodium bentonite/70 % crushed rock (diorite from the Äspö site) mixture used for the backfill has a dry density of 1.7 g/cm^3 and contains 23 % water when saturated.

The function of the engineered barrier is being tested in the “Prototype Repository” at the Äspö site (Fig. 2) under the conditions that prevail at 500 m depth, where temperatures up to 90 °C and pressures up to 4.0 MPa prevail and the groundwater has a relatively high salinity. The heat-generating waste is simulated by six heating elements in a 100-m-long section of the “Prototype Repository”.

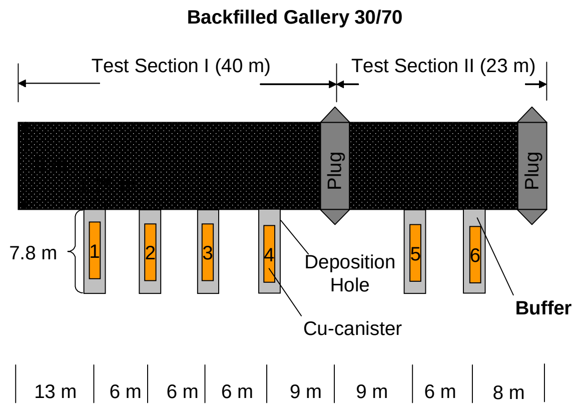

Figure 2. "Prototype repository" at the Äspö test site.

The function of the bentonite barrier in the multi-barrier concept is:

- to prevent migration of water, which would transport corrosive substances at the waste canister,
- to guarantee a chemical environment in which corrosion rates and solubility of radionuclides are low,
- to delay or limit release of any radionuclides from the container in the case of damage,
- to guarantee the best possible uniformity of the stresses acting on the canister even when subjected to uneven loading and/or uneven distribution of water content in the bentonite.

To fulfil these functions, the bentonite must have the following material properties:

- low permeability,
- limit migration of substances to diffusion,
- good swelling properties,
- high sorption capacity,
- high plasticity,
- high long-term stability (up to 100,000 years),
- high heat conductivity,
- low shrinkage.

Three thermo-hydraulic processes occur in the backfill and buffer. There is an initial drying out phase in the backfill due to the increase in temperature at the canister surface. The rise in temperature causes the thermal conductivity of the bentonite barrier to decrease, leading to further drying (Fig. 3). The bentonite shrinks and fractures as it dry, providing pathways for migration of radionuclides (Radhakrishna et al., 1992).

The inflow of groundwater through joints in the surrounding host rock will result in swelling of the bentonite (Fig. 3). When bentonite swells, its porosity, and hydraulic conductivity are reduced, and it becomes nearly water-impermeable. This in turn delays corrosion of the canisters, which would allow radionuclides to escape. The swelling of the bentonite will also maximize the long-term mechanical and chemical stability of the buffer and backfill.

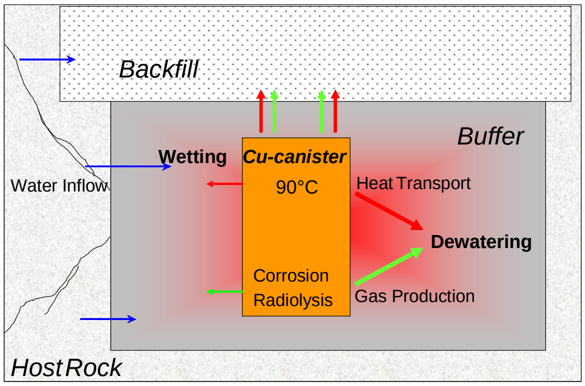

Figure 3. Thermo-hydraulic processes investigated in this study.

Gases are produced by a number of processes in a nuclear waste repository, depending on the physical and chemical conditions, the design of the repository, as well as the composition of the waste. The following gas-generating processes are possible:

- corrosion,
- radiolysis,
- microbial decomposition (in low- and intermediate-level waste),
- gas release from the buffer, backfill and host rock due to the rise in temperature, and
- radioactive decay.

The production of gas can greatly increase the pressure in the near field of the container. Experiments by Telander & Westerman (1997) indicate that this may result in hydrostatic pressures up to 30.0 MPa. New pathways for groundwater and radionuclides may be created once the entry pressure of the bentonite barrier is reached.

To assess the suitability of the bentonite as a engineered barrier for a high-level waste repository, it is necessary to know how water is taken up by bentonite, the conditions necessary for gases to migrate through the bentonite, and how the compacted bentonite reacts with the gases. This requires an understanding of the physics of two-phase flow in saturated and unsaturated media as a function of temperature, pressure, salinity, and composition of the bentonite in the barrier.

2. EXPERIMENTAL PROCEDURE

2.1 Studied Material

We performed experiments with a specially constructed permeameter to estimate the thermal parameters and the unsaturated hydraulic parameters of the backfill. The analyzed backfill material consists of 30 wt.% sodium bentonite and 70 wt.% crushed rock. The finely powdered sodium bentonite, (product name SPV Volclay) contains 75 % montmorillonite with 85.5 % exchangeable sodium (Müller-vonMoos & Kahr, 1983). The crushed rock consists of granite with a maximum grain size of 5 mm. We used water with different ionic contents: pure water and water with an ionic content similar to that of the groundwater at Äspö HRL (called Äspö water in this paper).

2.2 Laboratory Equipment

The permeameter we used to study heat and gas flow in the backfill is shown in Fig. 4. The permeameter consists of a 30-cm-long steel column (3) with a diameter of 5 cm. There is a heating element (2) in a steel chamber at the bottom of the column. We maintained either a constant pressure or pressure gradient in the sample column using a water reservoir (1) that can be connected to the bottom and/or to the top of the sample column to produce the required pressure in the column. The water in the reservoir can be pressurized up to 2.0 MPa. A burette (3) was attached to the top of the sample column. The pressure in the burette can be held at atmospheric pressure or increased up to 2.0 MPa.

Äspö water was added to the column of the permeameter. Backfill material (described above) was then added and compacted with a piston to avoid air entrapment. With this procedure we obtained a compaction corresponding to a dry density of 1.6 g/cm³ and a porosity of 48 %. Before filling the column, we placed a layer of fine sand, a geotextile, and a metal filter above the heating element at the bottom of the column and then again at the top of the column when it had been filled and compacted. This was done to assure one-dimensional flow and prevent the bentonite from being washed out of the sample. The column was wrapped with insulating material to minimize heat loss.

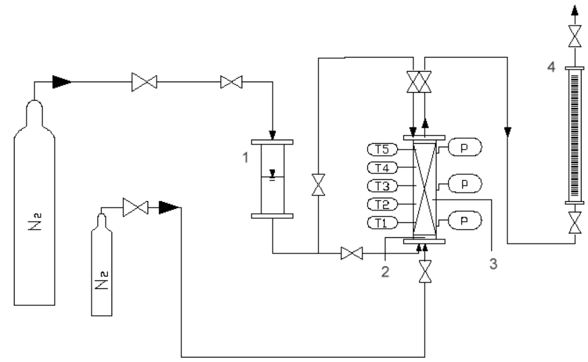

Figure 4. Experimental set-up (T = temperature sensors, P = manometers).

2.3 The Thermal Experiment

The sample was pressurized to 1.0 MPa at the top and bottom using the water reservoir (1 in Fig. 4). The valve of the burette was closed during the thermal experiment. The heating element in the steel chamber is surrounded by water. The thermostat of the heating element was set to 90 °C. Five temperature sensors recorded the initial temperature increase in the column and the time-invariant temperature distribution during the three-week experiment (Fig. 5).

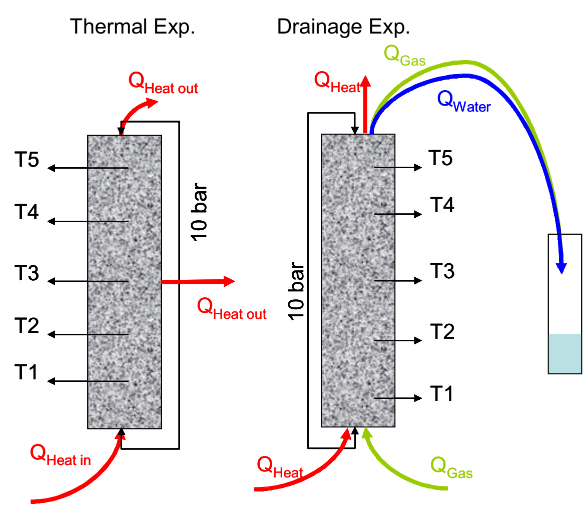

Figure 5. Schematic drawing of the experimental procedure.

2.4 The Non-Isothermal Drainage Experiment

Four days after starting the thermal experiment we replaced the water surrounding the heating element by air. Then nitrogen was injected from below into the already heated and pressurized sample. The nitrogen injection pressure was increased stepwise.

We started to inject nitrogen at 1.15 MPa in order to remain below the entry pressure of the backfill material. After 4 days we increased the injection pressure to 1.5 MPa, and after 10 days to 2.0 MPa. During the drainage experiment we opened the valve of the burette, and pressurized it initially to 1.0 MPa. The pressure in the burette increased as water, and eventually gas entered the burette. Before the burette valve was opened, 51 mL of Äspö water was placed in the burette, corresponding to 41 % of its maximum water capacity. Fig. 5 shows a schematic drawing of the experimental procedure.

2.5 Pressure Cell

A specially designed pressure cell (3.7 cm high and 5.4 cm in diameter) with a sample volume of 84.3 cm³ was constructed to determine capillary pressures in the range between 0 and 10.0 MPa (Fig. 6). There is a semipermeable cellulose membrane at the bottom of the pressure cell that separates the gas and water phase at pressures below 10.0 MPa. SPV Volclay and crushed rock were added to Äspö water in the sample chamber and compacted with a piston, obtaining a compaction corresponding to a dry density of 1.6 g/cm³. At each step of the desaturation experiment, a given gas pressure is applied to the sample chamber and 4 mL of water is allowed to drain into a connected burette. The pressure in the water at the bottom of the chamber below the semipermeable membrane is then measured. A drainage curve is then plotted using the capillary pressure calculated for each desaturation step. The entry pressure of the membrane was reached at approximately 40 % saturation, requiring the use of a different method.

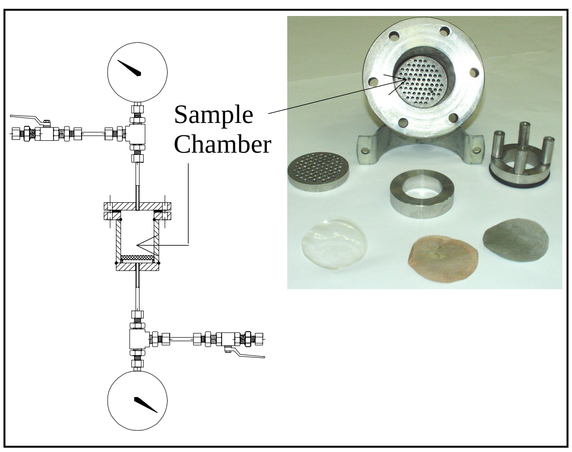

Figure 6. Schematic drawing and parts of pressure cell apparatus.

For pressures above 10.0 MPa, a thermohygrometer was used to measure capillary pressures. Äspö water was added stepwise to an air-dried sample and allowed to equilibrate with the air inside a closed container. The relative humidity *h* in the container was measured and the capillary pressure calculated using Kelvin's equation. Using the data from both methods, it was possible to plot a capillary pressure curve over a wide range of saturation (Figs. 7 and 8).

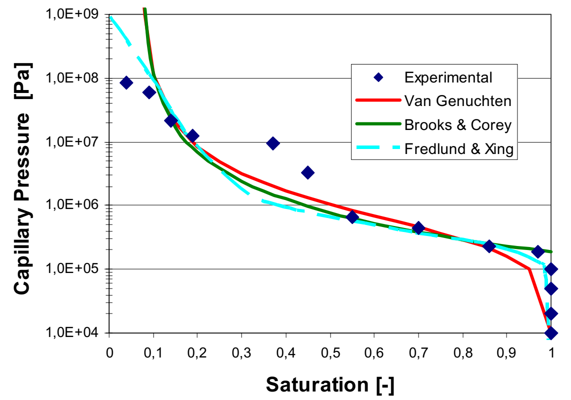

Figure 7. Measured capillary pressure curve of SPV Volclay/crushed rock mixture with pure water.

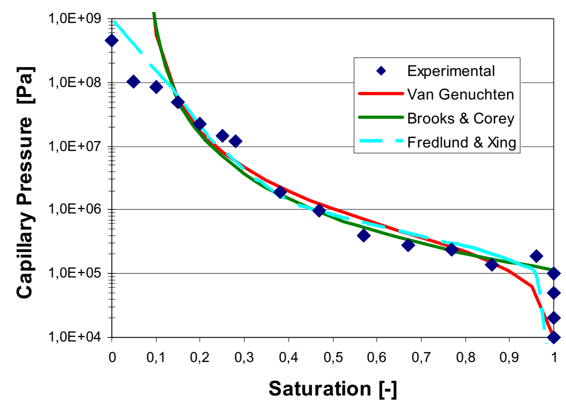

Figure 8. Measured capillary pressure curve of SPV Volclay/crushed rock mixture with Äspö water.

Three different capillary pressure-saturation functions were fitted to the measured data:

- *Brooks-Corey* (Brooks & Corey, 1964), model parameters: P_d, λ, S_{wr}, S_{nwr};
- *van Genuchten* (van Genuchten, 1980), model parameters: α , n (m), S_{wr}, S_{nwr};
- *Fredlund-Xing* (Fredlund & Xing, 1994), model parameters: a, m, (n) S_{wr}, S_{nwr}.

Figs. 7 and 8 show the best fits of these three models to the measured capillary pressure curve for backfill mixed with pure water and with Äspö water, respectively. The models differ mostly in the low saturation range. The Fredlund-Xing model (based on thermodynamic considerations, e.g., Richards, 1965) gives the best fit to the measured capillary pressure for this low saturation. The improved fit is obtained by introducing a correction function for low saturation. The entry pressures obtained with these three models are given in Table 1. Another important parameter for describing the behavior of the backfill material is the pore-size distribution, which can be represented by an inverse function (Fredlund & Xing, 1994)

3. NUMERICAL MODEL USING ITOUGH2

The measured rate of water flow into the burette, the gas pressure in the burette, and the temperature distribution in the column were used to calibrate the model. The inverse modeling and calibration was carried out with the iTOUGH2 program (Finsterle, 1999). We determined the values for the thermal parameters and unsaturated hydraulic parameters of the backfill material by fitting the calculated to the measured system behavior.

In the iTOUGH2 simulation a two-dimensional, radial model was developed to calculate fluid and heat flow along the axis of the column, as well as radial heat loss through the top, bottom, and sides of the column. The column was discretized along the axis into 60 blocks with a thickness of 0.5 cm. The thermal and gas-injection experiments were simulated in a single model run. The column itself was initially saturated with water at a pressure of 1.0 MPa and a temperature of 19.4 °C. Appropriate thermal parameters for the insulation material to account for heat loss were specified so that the calculated temperature distribution gave the best fit with the experimental data (Fig. 9). We set the temperature of the lowest temperature sensor (T1) as a time-dependent temperature boundary condition.

When the nitrogen injection experiment was simulated, the top element was connected to another element representing the burette. This arrangement allowed us to model the observed increase in pressure due to flow of liquid or nitrogen into the burette. The initial and boundary conditions of the simulation with iTOUGH2 are shown in Fig. 9.

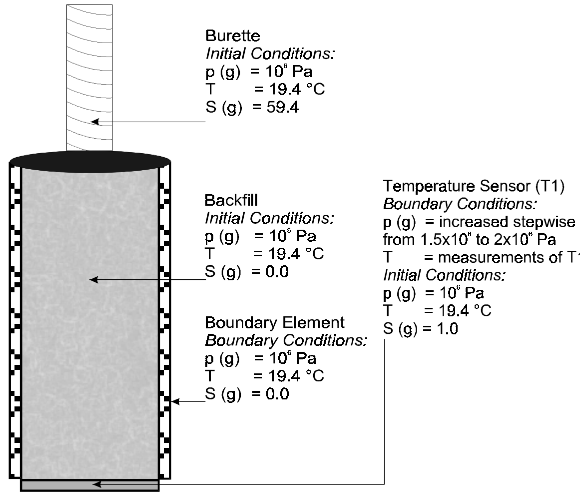

Figure 9. Initial and Boundary Conditions used for the iTOUGH2 simulation.

The Brooks-Corey (1964), van Genuchten (1980), and Fredlund-Xing (1994) models, were used to describe the two-phase relative permeability and capillary pressure functions of the backfill material during the drainage experiment. To avoid overparameterization, the pore-size-distribution indices • and n (m) and the residual saturation values S_{wr} and S_{nwr} were set to given values in the inversion. These values were taken from the fit to the measured capillary pressure-saturation curve. Only four parameters were estimated by inverse modeling: absolute permeability, gas entry pressure (entry pressure P_d [BC model] or capillary strength parameters 1/• [VG model], and *a* [FX model]), thermal conductivity, and specific heat.

4. RESULTS AND INTERPRETATION

4.1. Temperature Distribution

All three models (BC, VG, and FX) show a good fit to the measured temperature values. The fit is independent of the model and the salinity of the water. A total heat flow through the column of about 45.0 W was calculated with the calibrated forward model, and the total heat loss was about 2.1 W.

4.2. Drained Water Volume

For the experiment with Äspö water, the Fredlund-Xing model provides the best fit to the measured volume of water collected in the burette (Fig. 10). The van Genuchten and Brooks-Corey models significantly underestimate the amount of displaced water during the first 10 days of this experiment. The higher number of independent parameters in the Fredlund-Xing model is probably responsible for the more exact match of the measured volume of water entering the burette.

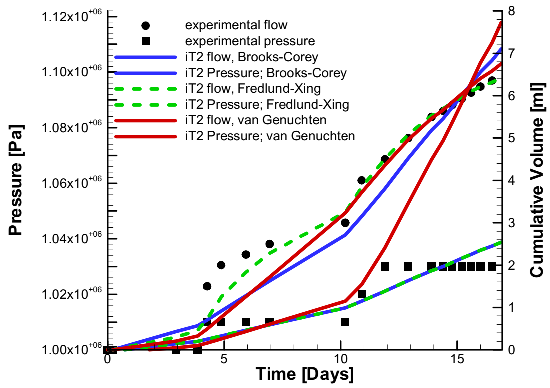

Figure 10. Measured and calculated system behavior for the non-isothermal experiments with Äspöwater.

For the experiment with pure water, the van Genuchten and Fredlund-Xing functions provide equally good fits to the measured non-isothermal drainage data (Fig. 11).

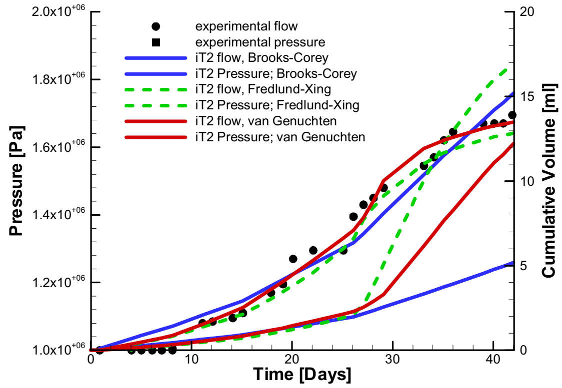

Figure 11. Measured and calculated system behavior for the non-isothermal experiments with pure water.

The development of open clusters when Äspö water is added to the backfill may be the reason for the discrepancies between the modeling results for the two experiments using water with different salinities:

- In pure water, the edges of the clay crystals have a positive electrical charge, and repulsive forces between the particles dominate. The bentonite generally shows a homogeneous, dispersed structure where micropores dominate.
- In contrast, when water containing calcium is used the thickness of the diffuse double layer is reduced. The positively charged edges are drawn towards the negatively charged surfaces of the crystals, forming weakly bonded flocculates. (Jasmund & Lagaly, 1993). The flocculated structure shows flocs with a random orientation of individual particles and open clusters (Yong & Warkentin, 1966). These open clusters may lead to secondary pore system inside the bentonite.

4.3. Pressure Increase In The Burette

While the volume of water pressed into the burette is reasonably well estimated by all three models, there are significant differences for the increase in pressure in the burette. The models show an abrupt change in the rate of pressure increase on arrival of the gas phase in the burette.

For the simulation of the experiment with pure water the van Genuchten and Fredlund-Xing models generate a dispersed gas distribution that reaches the top of the column after about 27 days with a relatively low gas saturation of 28 % or 10 % near the injection point, respectively (Figs. 11 and 12). The Brooks-Corey model predicts that the nitrogen front moves through the column with a piston-like displacement, resulting in a high saturation of gas in the injection zone (Fig. 12). The relatively sharp gas–liquid front within the column is preventing gas breakthrough within the simulation time (Fig. 12).

For the simulation of the experiment with Äspö water, the Brooks-Corey and van Genuchten models yield the same results as for pure water. Like with the Brooks-Corey model with pure water, the Fredlund-Xing model predicts no gas breakthrough within the simulation time. However, no gas breakthrough was evident in either experiment.

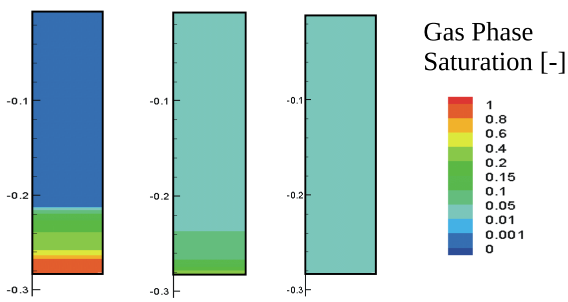

Figure 12. Simulated gas distribution after 17 days for the experiment using Äspö water.

4.4. Parameter Estimation

Table 1 summarizes the best-estimate parameter sets obtained from the inversion for both experiments. The inversion of the data of the experiments using Äspö water or pure water results in an entry pressure at 90°C of 0.14 MPa or 0.21 MPa for the van Genuchten model, 0.03 MPa for the Brooks-Corey model, and 0.31 MPa or 0.43 MPa for the Fredlund-Xing model respectively.

Table 1. Calculated and measured parameter values

Parameter	VG	BC	FX
Experiment using pure water Measured data			
Entry Pressure [MPa]	0.31	0.19	0.28
Pores Size Distribution Index [-]	1.6	0.55	0.7
Res. Water Sat. [-]	0.07	0.07	0.2
Res. Gas Sat. [-]	0.0	0.0	0.0
Parameter estimation using iTOUGH2			
Entry Pressure [MPa]	0.21	0.03	0.43
Permeability [m²]	8.3×10^{-19}	7.6×10^{-19}	6.25×10^{-19}
Experiment using Äspöwater Measured data			
Entry Pressure [MPa]	0.189	0.11	0.25
Pores Size Distribution Index [-]	1.44	0.4	0.5
Res. Water Sat. [-]	0.07	0.07	0.01
Res. Gas Sat. [-]	0.0	0.0	0.0
Parameter estimation using iTOUGH2			
Entry Pressure [MPa]	0.14	0.03	0.38
Permeability [m²]	1.3×10^{-18}	8.3×10^{-19}	1.4×10^{-18}

The inversion with the van Genuchten model results in an same entry pressure calculated by iTOUGH2 as the measured one when the influence of temperature on capillary pressure is taken into consideration (Lenhard & Parker, 1987). The value of 0.03 MPa obtained with the Brooks-Corey model seems to be independent of the salinity of the pore water. The calculated parameter value is significantly lower than the measured one at 20°C (Table 1). The lower entry pressure results from the strong correlation of the hydraulic parameters with each other in the inversion employing this model. The entry pressure calculated with the Fredlund-Xing model parameters is about 50 % higher than the measured value.

The influence of salinity on the capillary pressure is obvious in Table 1: The capillary pressure increases with decreasing ionic content of the liquid phase. This observation can be explained by hydrogen bonding of water molecules in the diffuse double layer. With decreasing ionic content the diffuse double layer of the bentonite increases in thickness. The water in the diffuse double layer is bonded more strongly to the clay surface and its viscosity is increased (Yong et al., 1992). This results in a reduction of the mobility of the water molecules. Therefore, the entry pressure must be higher in the case of water with a low ionic content for the gas to be able to displace the water from the pore spaces.

All three models show an increased pore size distribution index with decreasing salinity. This is related to a more uniform pore size distribution, supporting the assumption that no open clusters exist when sodium bentonite is mixed with pure water.

5. CONCLUSIONS

The hydraulic parameters of unsaturated SPV Volclay/crushed rock mixtures were calculated in the presence of water with different ionic concentrations. The hydraulic conductivity is 36 % lower with pure water than with Äspö water. The pressure cell and thermohygrometer experiments indicate that the entry pressure is also about 39 % lower with pure water than with Äspö water. The influence of salinity on the hydraulic parameters is more significant than for a temperature increase from 20°C to 90°C, which reduces the entry pressure and hydraulic conductivity by only about 20 %.

The influence of the swelling pressure on the capillary pressure is obvious from the experimental data. The use of pure water increases the swelling pressure and decreases the porosity of SPV Volclay/crushed rock mixture relative to that when water with a higher salinity is used. The reduced pore space increases the entry pressure.

The inversion with the van Genuchten model results in parameter estimates that are consistent with the values obtained from the experimental data. For both drainage experiments, the Fredlund-Xing model provides the best fit of the calculated system to the measured system behaviour. However, both the Fredlund-Xing and the van Genuchten model predict a gas breakthrough that was not evident in the experimental data. The hydraulic parameter values are strongly correlated in the results of the Brook-Corey model, and the entry pressure is strongly reduced. The parameter correlation was shown by the sensitivity and error analysis of the iTOUGH2 program. Therefore, it is very useful when analyzing experimental data to have information about the impact of the model parameters on the system behaviour.

6. ACKNOWLEDGMENTS

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