Session 4

Gaz and Liquid Transfer
Chair: Paul Marschall / Scott Altmann
GAS MIGRATION THROUGH BENTONITE/SAND: LESSONS LEARNED FROM THE GMT IN-SITU TEST

T. Shimura¹, S. Vomvoris², G.W. Lanyon³, P. Marshall², K. Ando⁴, S. Yamamoto⁴, H. Asano¹

1. RWMC, No. 15 Mori Building, 2-8-10 Toranomon, Minato-ku, Tokyo, Japan
2. NAGRA, Hardstrasse 73, Wettingen 5430, Switzerland
3. Fracture Systems Ltd, Tregurrian, St Ives, UK
4. Obayashi Corporation, Shinagawa Intercity Tower B, 2-15-2 Konan, Minato-ku, Tokyo, Japan

INTRODUCTION
The Gas Migration Test in Engineered Barriers (GMT), which was performed in Nagra’s Grimsel Test Site in Switzerland under the leadership of the Radioactive Waste Management Funding and Research Center in Japan (RWMC), was completed in March 2007. The project focused on issues related to waste-generated gas migration through the engineered barriers in a silo-type disposal (Fig. 1). Of particular interest was the evaluation of the bentonite/sand buffer behavior under in-situ conditions; the capability of current modelling concepts and tools to capture the relevant process; the required laboratory tests to allow extrapolation to other bentonite/sand ratios.

The components of the field experiment (see Fig.1) were [1]: a silo cavern, a concrete silo emplaced within the cavern, a gas vent in the top of the concrete silo, bentonite/sand backfill around the silo (20% bentonite and 80% sand) and, finally, sand/gravel backfill in the upper cavern above the silo cavern. The bentonite/sand was emplaced as a series of “lifts” which were compacted in situ. At the tops of selected lifts, monitoring instruments were located, defining a series of Levels and associated Layers numbered 2-12.

The saturation phase of the EBS was started in August 2001 and was ended at the start of October 2002 as the region expected to be the focus of gas flow was highly saturated (although parts of Layers 2-6 and 11 had shown little increase in saturation and their saturation continued throughout the whole duration of the experiment). The major experimental steps included:

- A series of water tests (WT1) before the gas injection (approx. 3 months) (also [1])
- A series of gas injections (approx. 9 mo.) -- nitrogen was injected into the silo at increasing mass flow rates from 0.025 to 5 mg/s. The injections were designed to target different processes: dilution, two phase flow, interface opening and pathway dilution ([1]).
- A post gas injection water test (WT2) -- a repetition of water testing, which showed no influence on the EBS properties from the gas migration (indication of increased resistance to flow in/out of the silo ([1]).

Figure 1: The GMT Concept (left) and the as-built EBS (right).
• A second series of gas injections – nitrogen and selected tracers were used (4 months) ([2])
• Excavation and characterisation of the EBS

LESSONS LEARNED
• Disposal System
  – The 20/80 bentonite/sand provided a low permeability stable buffer
  – There was no measurable increase in permeability due to the gas injection
  – The gas was able to escape from the EBS at relatively low over-pressure
  – Only small volumes of water were expelled from the concrete silo
  – Saturation behaviour requires further investigation
  – The buffer was more highly compacted than was indicated by standard measurements
• Geosphere
  – “Natural” sealing processes occurred within the geosphere during the experiment resulting in an ongoing reduction in leakage from the GMT cavern
  – While gas may have migrated into parts of the geosphere, overall the geosphere acted as a barrier to gas migration
  – Heterogeneity within the geosphere had limited effect on the EBS saturation
• Gas migration in a composite system
  – Interfaces played a significant role in gas migration and were associated with channellised flow
  – Interpretation of results requires consideration of coupled 2-phase-flow and mechanical process

The interfaces were observed both in the supporting laboratory test program and in the excavation to exercise the main control for the gas migration for the conditions of the GMT experiment. Fig. 2 shows an example from one of the embedded rhodamine pads in the interface between Level 8 and Level 9. The horizontal hydraulic paths along the interface can be seen at the left, developed along the interface; however only mm wide ‘diffusive rims’ are detectable in the vertical planes through the pads (right). The gas pathways were shown to be even more concentrated and channellised.

With respect to modelling significant advances were made within the GMT project with respect to a better understanding - and incorporation into the models - of the coupling between two-phase flow and coupled mechanical processes (dilation). This understanding is very valuable to allow evaluation of the abstraction of system models for application in modelling within the Safety Case.

References
MODELING APPROACHES OF TWO-PHASE FLOW PHENOMENA ASSOCIATED WITH CORROSION OF SF/HLW CANISTERS IN A PROPOSED REPOSITORY IN OPALINUS CLAY, SWITZERLAND

R. Senger¹, T. Xu², P. Marschall³, S. Finsterle⁴

1. INTERA Incorporated, 9111A Research Blvd., Austin, Texas 78757, rsenger@intera.com
2. Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA, Tianfu.Xu@lbl.gov
3. NAGRA, Hardstrasse 73, CH-5430 Wettingen, Switzerland, paul.marschall@nagra.ch
4. Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA, SAFinsterle@lbl.gov

INTRODUCTION

During Nagra’s Geosynthesis study (Nagra 2002a), scoping calculations were conducted to assess the gas pressure build-up in a SF/HLW repository in the low-permeability Opalinus Clay. Few studies considered both gas generation and water consumption associated with the corrosion of the metal waste canister (Lassabatiere et al., 2004). Water consumption or the supply of water is critical for maintaining the corrosion reaction and could result in reduced gas pressures. The corrosion of the steel canisters produces hydrogen gas (H₂) and a solid corrosion product magnetite (Fe₃O₄) under anaerobic conditions as the more likely chemical reaction, which is described by the following redox reaction: \( 3 \text{Fe} + 4\text{H}_2\text{O} \rightarrow \text{Fe}_3\text{O}_4 + 4 \text{H}_2 \). This chemical reaction indicates that for each mole of \( \text{H}_2 \) generated 1 mole of \( \text{H}_2\text{O} \) is consumed. Previous estimates of gas generation were based on the stoichiometry of the redox reaction and on the geometry of the cavern and canister, and potential variation in the loading of canisters in the drift. However, iron corrosion and \( \text{H}_2 \) generation rates can vary with time, depending on factors such as water chemistry, water availability, and water contact area.

As part of Nagra’s R&D project “Development of advanced modeling tools for gas transport in the EBS”, two approaches are being developed: (1) The first approach investigates the relevant two-phase flow phenomena associated with hydrogen gas migration and water consumption in the EBS and surrounding host rock. (2) The second approach investigates the coupling between the chemical reaction of steel corrosion, gas generation, and water consumption.

INVESTIGATIVE APPROACHES

In the first approach, the different two-phase flow processes associated with generation and migration of waste-generated gas were investigated using the reference design for the SF and HLW canisters for the proposed SF/HLW repository (NAGRA, 2002b). The modeling of \( \text{H}_2 \) gas generation associated with corrosion of the waste canister and the potential impact of \( \text{H}_2\text{O} \) consumption on gas migration is implemented using the two-phase flow code TOUGH2 (Pruess et al., 1999). The \( \text{H}_2 \) gas migration assuming initially fully water-saturated conditions is simulated using the Equation-of-State module EOS5 for two-phase flow of hydrogen and water. This required simplifying assumptions for the interface between the waste canister representing the source of \( \text{H}_2 \) and sink of \( \text{H}_2\text{O} \) associated with corrosion and the surrounding bentonite backfill.

The second approach involves the coupling of the chemical reaction with the two-phase flow code. For this, the chemical reaction code TOUGHREACT (Xu et al., 2006) was coupled with the EOS5 module. The reaction products in terms of \( \text{H}_2 \) and \( \text{Fe}_3\text{O}_4 \) are explicitly considered in terms of the gas
Figure 1: Sensitivity runs on pressure buildup in the SF/HLW tunnels for (a) H₂ generation only and (b) H₂ generation and H2O consumption. The hydraulic conductivity of the host rock varies between 1E-13 and 1E-15 m/s (corrosion rate is assumed the same for all simulations).

source and the change in porosity of the canister due to the volumetric difference between iron and magnetite. Moreover, the kinetic rate law implemented in TOUGHREACT allows consideration of various factors affecting gas generation, which include (1) reduction of iron volume, (2) chemical equilibrium conditions, and (3) reduction of water contact areas.

RESULTS AND INTERPRETATION
Numerical simulations of gas migration from a repository associated with corrosion of waste canisters indicate significant pressure buildup for a range of corrosion rates and for a range of permeability of the Opalinus Clay. With increasing corrosion rates and decreasing host-rock permeabilities, pressure buildup increased, as expected. The simulations taking into account water consumption show that the pressure buildup is reduced compared to the simulation considering only gas generation (Figure 1). The reduction in pressures is enhanced for lower permeability of the Opalinus Clay and for higher corrosion rates, which correspond to higher gas generations rates and higher water consumption rates. The numerical simulations of gas migration from a waste container indicate complex flow dynamics through time. At late time, water flow is toward the canister opposite the outward H₂ gas migration, with the water flow rate corresponding to the prescribed water consumption rate.

Preliminary analysis considering the coupling of chemical reaction model with the two-phase flow model indicates significant reduction of the H₂ gas generation rate and corresponding reduced pressure buildup when considering reduced water contact areas, and chemical equilibrium conditions, and iron-volume. More detailed analyses are being conducted to refine the kinetic rate law in TOUGHREACT for representing the anaerobic corrosion of the waste canister in a deep SF/HLW repository.
APPROACHES FOR MODELLING GAS TRANSPORT IN CLAY FORMATIONS BASED ON IMPROVED DARCY AND NON-DARCY FLOW CONCEPTS

H. Alkan, W. Müller

Istec, Institute for Safety Technology, 50667 Cologne, Germany (hakan.alkan@grs.de)

INTRODUCTION
One of the important issues of a waste repository in deep clay rich formations is the determination of the hydraulic properties and the sensitivity of these properties to thermal, mechanical and chemical perturbations in and around the storage locations. Modelling of transport phenomena in disturbed as well as in undisturbed portions of clay formations is more complex than that of other host rocks. In basic, the transport of gases in repository systems is modelled by diffusion and dispersion supported advection. The Darcy concept for one and/or two-phase flow provides the basic formulation for gas advancing either dissolved in liquid phase or together with liquid phase. There is no significant lack of knowledge regarding phase behaviour of gas-liquid mixtures, except data supporting the empirical parameters of equations of state such as solubility of gases in water with various ionic activities. However, the insufficiency of the Darcy formulation for modelling the transport in clay formations is obvious. The dominating effect of the capillary forces, the local and global heterogeneities, the interactions of percolating fluids with clay minerals are some of the features avoiding the direct application of Darcy formulation. Many physical and numerical attempts have been made for more representative formulations of gas transport in clay formations.

APPROACHES FOR MODELLING GAS TRANSPORT IN CLAY
Empirical correlations are used to describe the gas breakthrough pressure and flow rate as function of gas saturation, vertical stress and temperature. The attempts of defining gas entry pressure in terms of permeability can also be cited among the empirical respectively semi-empirical formulations. These approaches are mainly applied for plastic clay formations such as Boom and Pontida clays (Rodwell et al., 1999). Compressible behaviour of both the solid matrix and the pores are modelled essentially based on Terzaghi’s theory of consolidation and Biot’s effective stress concept. Case studies are published as saturated and unsaturated single phase flows by coupling the pore pressure, solid displacement (Terzaghi concept, elastic and plastic) and heat flow. Episodic or intermittent flow is the generally accepted physical concept for modelling the pulse like non-Darcy flow behaviour in Clay formations. A simple numerical approach to episodic pathway flow is given by Horseman (Horseman et al., 1996) using the cubic law for determining the flow rate through a planar fracture in which the aperture of the pathway (fracture) is a function of the applied overburden (confining) stress and pore pressure.

Simplified conceptual models by using capillaries (capillary bundles) and fractures as pathway with stress dependent geometry are applied for determining gas, two-phase flow in plastic as well as in indurated clays with and without TC coupling (Ortiz et al. 1996, 1997). The number of paths is set by an input channel density. Some other approaches conceptualize the clay formations as crack-fracture networks where the opening of the cracks is governed by deformation.

A model proposed to be used especially for the EDZ in clay formations takes the linear elastic fracture mechanics as the theoretical basis. The cracks represented by ellipsoidal cavities with dimensions as suction-dependent variables are arranged in a regular pattern (Duppendecker, 1997) The model presented by Alonzo et al. (2006) assumes the clay formation as a finite element composed by a rock matrix and a series of n fractures. Liquid and gas flow is calculated using Darcy’s law. The reversible and irreversible deformation as well as the variations in capillary pressure due to the changes in fracture aperture is taken
into consideration with basic formulations. Some examples to show the capabilities of the formulation are presented in Delahaye et al. (2002).

The flow behaviour through bentonite buffers, on the other hand, is conceptualized mainly as a conventional porous medium, with gas flow governed by conventional concepts of capillary pressure and relative permeability. Microfissuring of the clay, in which small fissures are created or opened by the invading gas to provide the pathways for the gas to enter the clay, and/or macroscopic fracturing in which the scale differs from the microfissures are proposed as the mechanisms for gas invasion of water saturated compacted bentonite (Hoch et al., 2004).

RESULTS
An executive compilation of the modelling efforts of gas transport in clay formations is presented. Modelling concepts are evaluated and compared. Their implementations and validations are summarized. The application potential of these models for safety analysis in repository management is discussed. The models to describe the flow and the effect of the internal and external factors in bentonite buffers are also briefly examined (Alkan and Müller, 2006).

It is concluded that more experimental and theoretical studies should address the following issues: capillary forces and their relation to other petrophysical properties, the configuration of the natural and induced fracture networks and the effect of the physical and chemical mechanisms (swelling, osmosis,) etc on the retention and diffusion of gas. The hydraulical-mechanical coupling of the EDZ and the effect of self sealing on the long-term-behaviour can be cited as other important subjects to be investigated for safety analysis of repositories in clay formations.

References

ACKNOWLEDGEMENT
The work was conducted as a project of the BMWi (Federal Ministry of Economics and Technology, Germany). The financial support is kindly acknowledged.
DERIVING LARGE SCALE HYDRAULIC BEHAVIOUR FROM BOREHOLE PRESSURE HISTORY DATA

Cristian Enachescu, Jacques Delay, Martin Cruchaudet, Remi de la Vaissiere

Several of the boreholes drilled from the underground rock laboratory at Bure are equipped with multipacker installations and a wealth of long term pressure monitoring data is available. These long term pressure measurements are typically interrupted by hydraulic tests (usually pulse tests) conducted to achieve different objectives.

The core idea of the paper is that the pressure response measured during a hydraulic test is not only influenced by the small scale formation properties in the vicinity of the borehole but also by the large scale flow model whose response is basically “stored” into the long term pressure history data measured before the test. The paper explores how sensitive the response of a relatively short test to different assumptions concerning the large scale flow model is. In exploring this question, the flow model of the Callovo-Oxfordian clay is conceptualised using different composite model assumptions regarding the change in hydraulic properties and flow dimension at large scale.

The paper describes the theoretical base of the approach as well as interpretation results.
INTERPRETATION OF INTERSTITIAL PRESSURES IN CLAY MEDIA: APPLICATION TO THE CALLOVO-OXFORDIAN

P. Rousseau-Gueutin\textsuperscript{1,2,4}, J. Gonçalvès\textsuperscript{1}, M. Cruchaudet\textsuperscript{2}, S. Altmann\textsuperscript{3}, V. de Greet\textsuperscript{4}, S. Violette\textsuperscript{1}

1. UMR 7619 Sisyphé, Paris, France (gueutin@ccr.jussieu.fr, julio.goncalves@ccr.jussieu, sophie.violette@ccr.jussieu.fr);
2. ANDRA, Bure, France (martin.cruchaudet@andra.fr);
3. ANDRA, Châtenay-Malabry, France (scott.altmann@andra.fr);
4. UMR 7649 LMS, Palaiseau, France (greet@lms.polytechnique.fr)

The significance of interstitial pressures in clay media is a crucial point in repository studies. It is analyzed in the context within the problem of water and solute transfer in the clay media.

In the clay media the interstitial pressure is currently higher than the hydrostatic pressure, predicted by the theory (e.g. Neuzil, 1995; Gonçalvès et al., 2004). In the literature several origins to explain these overpressures are invoked such as diagenesis, hydrocarbon production, or osmotic processes. In the hypothesis of osmotic processes, these overpressures could interpreted as a consequence of transient coupled fluxes or the occurrence of a “structural overpressure” at equilibrium, known as the disjoining pressure (Langmuir, 1938; Derjaguin, 1987). Theoretical and experimental estimations are possible for these two options. Transient coupled fluxes can be measured on a sample submitted to a chemical perturbation; structural overpressure can be measured on a sample at mechanical equilibrium and is equivalent to the so-called swelling pressure (Viani, 1983). The disjoining pressure ($\Pi_0$) is described as an excess pressure in the porosity of the clay layers relative to that of a reservoir at the same depth.

Clay media characterized by weak water and solute fluxes are presently studied in the context of the research program on the feasibility of a deep geological repository for nuclear waste. In such negatively charged surface media, the hydraulic conductivities are very small, less than $10^{-11}$ m.s\textsuperscript{-1} (Marsily, 1986). If the boundary conditions applied to these clay layers are changed, either in terms of hydraulic gradient, or in terms of salinity composition, the classical expression of the Darcy’s law does not describe accurately the water flow in the medium (Marsily, 1986; Revil and Pessl, 2002). Instead of this classical law, a generalized Darcy law involving coupled flows is used.

In the transient osmotic process, the electrochemistry/hydraulic coupling are considered. The pressure gradient is no longer the only driving force for fluid flow but it is associated with the chemical potential gradient. The water flow due to a chemical potential gradient through a semipermeable membrane is the so-called chemical osmosis. The clay medium can behave as a semipermeable membrane (Bolt, 1979; Fritz, 1986), as the consequence of the negatively charged surfaces of the clay minerals (Mitchell, 1993). This surface charge modifies the ionic distribution in the pore solution. In the vicinity of the negative surfaces, there is an excess in cations (counter-ions) and a deficit in anions with respect to the concentrations in the equilibrium solution (the “bulk solution”). This redistribution has an impact on the ionic flow and thus on the water flow.

The identification of the coupling parameters, in the clay medium, is fundamental in order to estimate the water flow. This characterization requires the estimation of two coupling parameters $k_l$: the intrinsic permeability $k_l$ and the osmotic coupling parameter $k_r$. These coupling parameters can be estimated using two approaches:
(i) theoretical modeling method:
- porosity/intrinsic permeability relationships, defined for a clay medium, are used to estimate the intrinsic permeability, in Kozeny type petrophysical laws.
- an electrochemical model is used to estimate the osmotic coupling coefficient. The model used is a triple-layer model (Leroy and Revil, 2004). We implement this electrical model to simulate the interactions between the charged surfaces of the clays and the solution present in their vicinity.

(ii) by measurements:
- at the sample scale.
- at the field scale.

In clay media in equilibrium with surrounding reservoirs the disjoining pressure is a result of various mechanisms e.g. Van de Wall’s forces, electrostatic forces. The disjoining pressure can be estimated by a theoretical approach which also implies the use of the electric model and by an experimental approach, based on the swelling of clay.

(i) theoretical modeling method:
- Thermodynamic equilibrium between the clay media and the surrounding reservoirs is assumed. The interstitial pressure is then related to the different structural parameters of the clays (cation exchange capacity, porosity, cimentosion factor…).
- An electrochemical triple layer model accounting for the diffuse layer overlap allows the calculation of the variables invoked in the thermodynamic equations, such as the electrical potential within the diffuse layer.

(ii) by measurements:
- at the sample scale.

Preliminary results of all these approaches will be presented.

References
POROSITIES ACCESSIBLE TO HTO AND IODIDE ON WATER-SATURATED COMPACTED CLAY MATERIALS AND RELATION WITH THE STATES OF WATER

G. Montavon1, E. Alhajji1, D. Lebotlan2, Z. Guo1, C. Tournassat3 and B. Grambow2

1. Subatech laboratory, 4 rue Alfred Kastler, 44340 Nantes
2. LAIEM, 2 rue de la Houssinière, BP 92208, 44322 Nantes Cedex 03
3. BRGM; 3, avenue Claude Guillemin 45060 Orléans-cedex 2
* Corresponding author: denis.lebotlan@chimbio.univ-nantes.fr

INTRODUCTION

The transport of contaminants in consolidated, water-saturated porous clay systems is controlled by sorption and diffusion processes (e.g. [1-3]). The knowledge of the porosities accessible to the different contaminants is a prerequisite for the development of transport models to be used in performance assessment. These information are generally derived from in-diffusion experiments performed with conservative radioactive [1]. The relation between the measured porosities and the states of water (bound/free, external/internal water) remains a subject of discussion. For anions, like halogens, it is generally considered that they cannot access, for electrostatic reasons, neither in the interlayers nor in the largest part of the electric double layer developed at the external surface of the clay particles. This behavior is qualified as the “anionic exclusion” effect (e.g. [1]). The water in this exclusion space is considered as bound water whereas the porosity accessible to the anions is constituted of free water [4]. The anions may therefore be a good probe to experimentally determine the pore water volume, a fundamental parameter to be considered for sorption studies in water-saturated and compacted clay systems.

The goal of the work was to assess the reliability of the above assumption by comparing accessible porosities measured for iodide and HTO on water-saturated compacted clay media (illite, montmorillonite, MX-80 bentonite) with the characterization of the states of water in these pore spaces (bound/free water, external/internal water).

RESULTS

Low resolution proton NMR was used to characterize and quantify the states of water. Its low field limits the field gradient inside heterogeneous samples, while the short 90° pulse width enables protons of both solid and liquid phases to be irradiated, giving quantitative or semi-quantitative information about them in a relative and absolute manner. Furthermore, the relaxation time is a function of the mobility of the protons and allow a characterization of water.

The NMR method allows to differentiate the three different populations (structural OH, external bound and internal bound water) on hydrated clays (no free water) considering the difference in proton mobility. The quantitative description appeared constrained by (i) the mathematical treatment of multi-exponential relaxation curves when relaxation times were to close (ratio below 2) and (ii) when the relaxation time was too short. In these cases, only an estimation could be given. A deeper description of the water states within the different populations appeared not possible when water molecules are in contact (or close in the space) because of the occurrence of rapid exchange reactions. For the same reasons, it was not possible to use the method to quantify bound and free water in fully water-saturated compacted clay media. This latter information could however be determined when measuring the samples at −25°C. At this temperature, the distinction based on the difference in mobility could be done since bound water remained in a semi-liquid state whereas free water freezes.
Accessible porosities for anions and HTO were determined by an isotopic dilution method using capillaries to confine the materials. HTO was shown to probe the whole pore volume (i.e. the space made of bound and free water). The affinity of iodide for clay materials was shown to be very weak: in the pH range (4-8) and iodide concentration (10^{-7} to 5 \times 10^{-2} M) studied, Kd values were below 0.1 L/Kg. Conditions were chosen to neglect the sorption in the data interpretation for the determination of the porosities. When the bound water volume was mainly constituted of interlayer water (case of montmorillonite and bentonite), iodide was shown to be located in the pore space made of free water. When the interlayer water was not present (case of illite), the results showed that iodide could access to a small fraction of the bound water volume localized at the external surface of the clay particles.

References


