

Modelling coupled processes in bentonite: recent results from the UK's contribution to the Äspö EBS Task Force

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ABSTRACT

A range of potential concepts for the geological disposal of high level wastes and spent fuel are being studied and considered in the UK. These include concepts that use bentonite as a buffer material around the waste containers. The bentonite will be required to fulfil certain safety functions, the most important being (1) to protect the waste containers from detrimental thermal, hydraulic, mechanical and chemical processes; and (2) to retard the release of radionuclides from any waste container that fails. The bentonite should have a low permeability and a high sorption capacity.

These safety functions could be challenged by certain features, events and processes (FEPs) that may occur during the evolution of the disposal system. A consideration of how these FEPs may affect the safety functions can be used to identify and to prioritize the important areas for research on bentonite. We identify these important areas (which include hydration of compacted bentonite, illitization and erosion of bentonite), and describe how they are being investigated in current international research on bentonite.

The Äspö EBS Task Force is a collaborative international project designed to carry out research on bentonite. In 2011, the Nuclear Decommissioning Authority Radioactive Waste Management Directorate joined the EBS Task Force partly to benefit from its collective experience. The work of the EBS Task Force is split into two research subareas: (1) the THM subarea, which includes tasks to understand homogenization of bentonite as it resaturates, to investigate the hydraulic interaction between bentonite and fractured rock, and to model *in situ* experiments; and (2) the THC subarea, which includes tasks to investigate the issue of understanding transport through bentonite, and to model *in situ* experiments. In particular, the bentonite rock interaction experiment is a large-scale *in situ* experiment concerned with understanding groundwater exchange across bentonite rock interfaces, with the objective of establishing better understanding of bentonite wetting. In this paper, we describe our work to model the spatial and temporal resaturation of bentonite buffer in a fractured host rock.

KEYWORDS: events and processes (FEPs), bentonite, engineered barrier system (EBS), coupled processes, thermal (T), hydraulic (H), mechanical (M), chemical (C), resaturation, buffer.

Introduction

A number of the proposed engineered barrier system (EBS) concepts for the UK's geological disposal facility (GDF) involve the use of clay-based buffer or backfill in the high-level waste/

spent fuel areas. Bentonite, a clay mineral with a high proportion of sodium montmorillonite, which swells in water and endows bentonite with a low hydraulic conductivity, is most commonly considered as a buffering material in the EBS surrounding the disposal containers. Bentonite is being proposed as a buffer material in the EBS in Sweden (SKB, 2006), Finland (TILA-99, 1999), Switzerland (Nagra, 2002) and France (Andra, 2005).

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In each of these cases or concepts, the bentonite is expected to perform a similar role to that taken in the illustrative engineering designs of a UK GDF (Nuclear Decommissioning Authority, 2010a); namely as a buffer material emplaced in a deposition hole; a component of the backfill; or as a component of low permeability seals.

Safety functions

The EBS is expected to fulfil certain safety functions (Nuclear Decommissioning Authority, 2010b). These safety functions are different for disposal facilities hosted in higher strength and lower strength sedimentary rocks. Generic safety functions for the EBS have been considered previously (Nuclear Decommissioning Authority, 2010c). The safety functions for the EBS of a GDF in a higher strength host rock (Nuclear Decommissioning Authority, 2010b), (Rautioaho and Korkiala-Tanttu, 2009) are summarized here. The two main safety functions that the bentonite buffer will be expected to fulfil are described below.

The first function is that the buffer should protect the disposal containers from detrimental thermal (T), hydraulic (H), mechanical (M) and chemical (C) processes by: (i) being plastic enough to mitigate the effects of small rock movements on the container; (ii) being stiff enough to support the disposal container and maintaining its position; (iii) protecting the host rock from detrimental thermal effects; (iv) limiting the transport of dissolved corroding agents to the canister; and (v) being dense enough that microbes are barely active metabolically and so cannot give rise to unfavourable chemical conditions at the surface of the container.

The second function is that the buffer should limit or retard the release of any radionuclides from the canisters in the event that the canisters fail by: (i) providing a low enough permeability that any transport is dominated by diffusion; (ii) filtering colloids derived from the wastefrom or from the disposal canister corrosion; (iii) providing favourable chemical and/or physical properties to control the rate at which dissolved radionuclides could migrate from the wastefrom into the surrounding host rock; and (iv) providing a large surface area for sorption.

In addition to protecting the disposal containers and containing any radionuclides released, the buffer should not give rise to any processes that could adversely affect the containers or the host

rock. The buffer material should also have a sufficient swelling pressure that it will completely fill the space between the disposal container and the rock and will 'self-seal' in the event that it is damaged or eroded.

In a lower strength sedimentary host rock, the function of the bentonite is somewhat different as it has similar properties to the host rock (Nagra, 2002).

Features, events and processes

The safety functions, described above, can be influenced by features, events and processes (FEPs) that could affect the bentonite performance. Previous work has reported the FEPs that are relevant to the bentonite buffer (Miller and Marcos, 2007; SKB, 2010) and some thought has also been given to the UK-relevant FEPs (Wilson *et al.*, 2010). Rather than duplicate the previous work, this paper draws from previous work to present the processes that are important to the evolution of the bentonite buffer.

Thermal processes (T)

Spent fuel and high level waste will generate heat and the heat will be transported from the surface of the disposal container to the bentonite buffer. Initially, the buffer will be unsaturated and there will be gaps, for example, between the bentonite rings that surround the disposal container and between the disposal container and the bentonite. These gaps will be sealed by the homogenization (self-sealing) of the bentonite as it resaturates. Due to the fact that air has a lower thermal capacity than water, heat will be transported less rapidly through the bentonite when it is unsaturated. The temperature the bentonite buffer reaches will be dependent on the size and contents of the disposal container, as well as separation of the deposition holes and the thermal diffusivity of the host rock.

Hydraulic processes (H)

The transport of water in a saturated bentonite will be different to that in an unsaturated bentonite. Initially the bentonite will be unsaturated and the most important factors in the uptake of water will be the permeability of the surrounding rock mass (i.e. access to water); and a negative capillary pressure in the pores of the buffer. Water uptake (i.e. resaturation) will

depend on a number of factors. These factors include the temperature, smectite content and then-current degree of saturation of the buffer.

For a GDF in a fractured host rock, the buffer resaturates as groundwater flows primarily through the fractures that are intercepted by the deposition hole. As the bentonite resaturates, it forms a gel that increases in density as the water goes deeper into the bentonite. It is possible that the rate at which water flows through the fracture and into the bentonite will be greater than the rate at which the bentonite can absorb the water. If this is the case then there will be a pressure exerted on the bentonite gel by the inflowing water. This pressure may be high enough that the water can force its way through the bentonite gel and, as it flows, the water may erode the bentonite.

Mechanical processes (M)

As the bentonite resaturates it swells. Three components of the swelling have been identified (Pusch, 2002): (1) the stored elastic energy of compressed particles; (2) the hydration potential of the interlamellar space; and (3) the osmotic pressure between electrical double layers at the surfaces of stacks of montmorillonite flakes. In the case of bentonite with high bulk densities, the first two processes dominate, whereas in the case of lower bulk densities, the last process dominates.

If the bentonite is unable to expand freely, a swelling pressure develops. The swelling pressure results in a mechanical pressure on the host rock and the disposal container. The effectiveness of the buffer is dependent on the swelling and thus adequate swelling pressure must be maintained at all times.

Chemical processes (C)

Alteration of the sodium montmorillonite can affect the performance of the buffer and, therefore, its ability to fulfil the safety functions. A summary of the main processes causing the alteration of montmorillonite have been reported previously (Karlund and Birgersson, 2006) and those that are relevant to the buffer are illitization, chloritization, silica release and iron reactions.

Illitization is the name given to the process by which montmorillonite transforms into illite. Partial replacement of silica with aluminium in the montmorillonite results in a higher layer charge. Although this higher charge could be

compensated by interlayer cations, the electrostatic forces overcome the cation hydrating forces if the layer charge reaches a critical level. The critical charge is lowest for potassium and it can become bound to the mineral surface resulting in the collapse of the interlamellar space. As it has the lowest critical charge, potassium montmorillonite (illite) is the most common transformation product. As a result of the collapse of the interlayer, the ability of illite to absorb water is significantly less than that of montmorillonite. This means that the swelling pressure of illite is not as high as that of montmorillonite and this affects the performance of the buffer.

Chlorite is a non-swelling sheet silicate. Chloritization requires a high temperature (>200°C) and a high pH. The chlorites that are produced by this process have the same basic structure as montmorillonite but have a higher layer charge.

At high pH the solubility of silica increases dramatically. The equilibration of the silica will therefore be dependent on the pH and the diffusive removal or precipitation of silica can affect the rate at which the tetrahedral layer charge evolves. If the charge increases sufficiently the results may be similar to those for illitization.

If a steel disposal container becomes exposed, iron leaches into the porewater as a by-product of the corrosion. The montmorillonite can be transformed into other clay minerals if the aluminium in the montmorillonite is replaced by iron. These minerals are intermediate steps in the transformation to chlorite (for details see Wilson *et al.*, 2010).

Microbial processes (B)

Microbial processes can give rise to the formation of gas and sulfides. The formation of gas will lead to an increase in the gas pressure and this can affect the swelling of the bentonite if it reaches a critical level (Skoczylas, 2010). Sulfide from sulfate-reducing bacteria can corrode copper canisters.

In order to understand which of the processes will be coupled, an understanding of the buffer variables that affect each of the processes must be gained. Posiva (Miller and Marcos, 2007) and SKB (SKB, 2010) have already developed this understanding and have highlighted buffer variables upon which the FEPs may depend. In particular, it was concluded there is a large dependence of the FEPs on the THMC variables.

International research and the UK context

International research

Several international projects have contributed to the understanding of THMC behaviour of bentonite in EBS applications. These include the following.

(1) THERESA. This project set up under the Sixth Framework Programme (FP6) of the European Commission in 2007 and ran for three years. The aim of the THERESA project was to develop a scientific methodology for evaluating the capabilities of mathematical models and computer codes used in performance assessment.

(2) DECOVALEX. The development of coupled models and their validation against experiments project was established in 1992 by SKI, the Swedish Nuclear Power Inspectorate, with participation of several international organizations. The general goal of this project is to encourage multidisciplinary interactive and cooperative research on modelling coupled processes in geological formations, in support of the performance assessment for underground storage of radioactive waste.

(3) FORGE. The fate of repository gases project focusses on understanding gas generation and migration through bentonite.

(4) LOT. In 1999, SKB (Svensk Kärnbränslehantering AB) started a series of long term buffer material tests (LOT) at the Äspö Hard Rock Laboratory (HRL). These experiments are designed to test the behaviour of the buffer under conditions that are expected in a KBS-3 repository.

(5) The EBS Task Force was set up by SKB in 2005 to study coupled THMC processes in the EBS. The current projects in the Task Force are outlined in the following text.

UK context

It is noted that there are specific issues that may arise for spent fuel. For example, higher burn-up leads to the spent fuel producing more heat. For a given cooling time and emplacement conditions, the higher burn-up fuel will be hotter and will require longer cooling or the selected buffer materials will have to be robust to such conditions. Any site that is considered in the UK will have unique features and the properties of the host rock and groundwater will need to be taken into account.

EBS Task Force

The EBS Task Force was set up to study coupled THMC processes in the EBS. The Task Force focusses on two aspects of coupled modelling, one THM processes, and the other THC processes. The modelling activities are largely underpinned by either laboratory based experimental work, or *in situ* experiments at the Äspö HRL.

Current THM research

The latest phase of research that is being conducted by the THM team has four sub-tasks described below.

Homogenization

Homogenization refers to the ability of the bentonite blocks to swell and self-seal in such a way a produce a uniform homogeneous buffer. The swelling and homogenization of the buffer is important in order to guarantee that the buffer fulfils the safety functions identified above. It will be important to predict the final state of the bentonite after resaturation (and swelling), and to understand the homogenization that occurs. The task identifies two parts to the modelling of homogenization. The first part requires modelling of the swelling of bentonite in three simple geometries: axial swelling in a device with constant radius and limited height; radial swelling of the outer surface in a device with constant height and limited radius; and radial swelling into a cylindrical cavity in a device with constant height and radius. All of the bentonite samples are on a 50 mm scale. The second part of the task requires modelling of the self-healing of a larger cavity in a bentonite block. The aim of the experiment is to have a cavity that is large enough to have a good resolution for the sampling, but to have a bentonite block that is small enough that the time to saturation is reasonable.

Buffer–rock interaction experiment (BRIE)

The buffer–rock interaction experiment (BRIE) is being conducted at the Äspö HRL. The modelling to support this project concerns predictions of inflow into a series of boreholes in the TASO tunnel (a portion of HRL tunnel located at an elevation of -417 m). The location of the TASO tunnel and the suggested model domain is shown in Fig. 1 along with the five probing holes

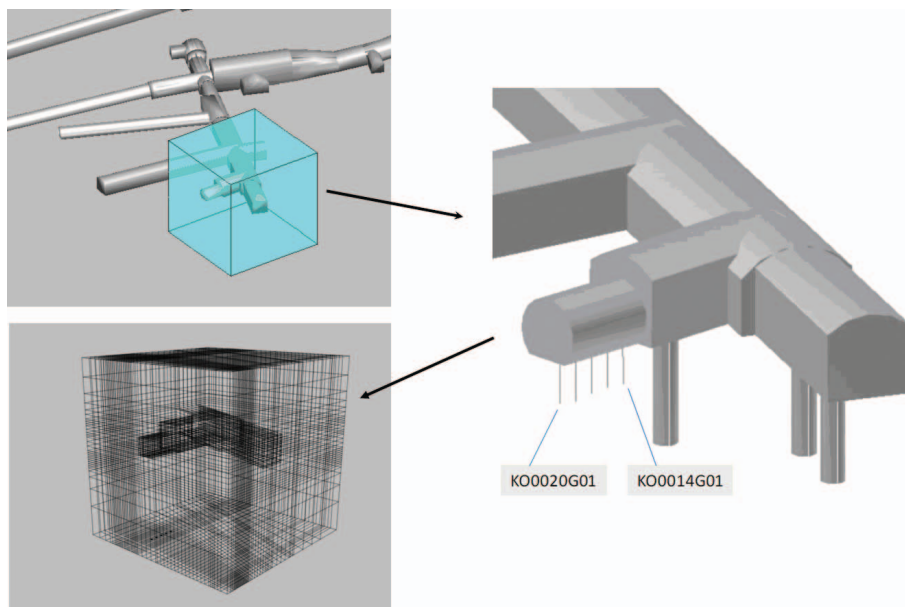


FIG. 1. The location of the BRIE experiment in the TASSO tunnel and the local-scale model domain.

that form part of the BRIE experiment. The modelling task aims to produce a scientific understanding of the exchange of water across the buffer/rock surface; better predictions for the wetting of the buffer; and better characterization methods of canister boreholes.

Sensitivity analyses

The sensitivity analyses task is a series of modelling studies intended to understand features of a THM model. The modelling carried out for this task focuses on the THM evolution of a single deposition hole. The numerical representation of the coupled THM model is intrinsically complicated. The task aims to systematically analyse the various couplings to determine the relevant key coupled processes. The sensitivity analyses studies key parameter variations in the THM properties of the rock and buffer.

Prototype repository

The prototype repository is an experiment that is being carried out at the Äspö Hard Rock Laboratory (HRL). The prototype repository has been designed to simulate a KBS-3 design repository system and aims to demonstrate the integrated function of the repository components. The experiment is highly instrumented and will

provide a full scale reference for comparison with the numerical models.

Current THC research

In 1999, SKB started a series of long term buffer material tests (LOT) at the Äspö Hard Rock Laboratory (HRL). These experiments are designed to test the behaviour of the buffer under conditions that are expected in a KBS-3 repository (an abbreviation of kärnbränslesäkerhet, a technology for disposal of high-level radioactive waste developed in Sweden by SKB). In total there are seven parcels. Each parcel consists of a heater in a central copper tube surrounded by pre-compacted bentonite blocks and instruments in a vertical borehole. These parcels are exposed to 'repository conditions' for one, five and twenty years. The temperature, total pressure, water pressure and water contents are measured during the heating period.

The geochemical data obtained at the end of the LOT test (and previous modelling, Arcos *et al.*, 2006) indicates that there are two main geochemical processes: (1) precipitation/dissolution of carbonate, sulfate and silica minerals; and (2) cation exchange in the montmorillonite interlayer.

The THC modelling of this parcel has been carried out (Senna *et al.*, 2010) with the numerical results predicting the dissolution–precipitation of anhydrite, calcite and silica in the heated bentonite. Current tasks include:

Benchmark 4 (HC couplings)

The bentonite used in the experiment was previously part of the LOT experiment and has a length and diameter of 50 mm. The bentonite is kept at a constant pressure of 8 MPa and an external water pressure gradient was imposed over the bentonite. The pressure difference between the inlet and outlet sides was controlled to be in the range 5–6 MPa during most of the time of the test. The water passing through the bentonite was collected and analysed in order to determine its electrical conductivity and chemical composition. The modelling work that is conducted as part of benchmark 4 requires that the properties of the collected water are correctly predicted.

Uphill diffusion (HC couplings)

A possible future study consists of modelling an experiment consisting of a layer of sodium montmorillonite sandwiched by a filter (to hold it in place) with a solution of NaClO₄ in either side. The concentrations of the NaClO₄ solutions differ, with a concentration of 0.5 M on one side and 0.1 M on the other. The sodium is then seen to migrate against the concentration gradient. Work is currently being undertaken to model this phenomenon.

The BRIE experiment and task 8

Modelling task 8 consists of modelling of the buffer–rock interaction experiment (BRIE) and requires predictions of groundwater inflow to a series of five probe boreholes in the TASO tunnel (Bockgård *et al.*, 2010), and the prediction of the resaturation of bentonite subsequently emplaced into the larger scale deposition boreholes at the same location as the probe boreholes. The emphasis of the work reported in this paper is to describe the overall modelling approach to describe the interaction of the groundwater flow, which takes place in conducting fractures in the granite, as a precursor to the prediction of the resaturation of the buffer.

Overall modelling approach

Three numerical codes are used to predict and analyse the BRIE experiment: *ConnectFlow*

(Serco, 2011), *TOUGH* (Pruess *et al.*, 1999) and *FLAC* (Itasca, 2011). *TOUGH* and *FLAC* have been coupled (referred to as *TOUGH-FLAC*). These coupled codes are able to simulate multiphase fluid flow and geo-mechanics. The coupled codes link two established software suites: *TOUGH2 v2*, which is used extensively in the radioactive waste management industry to simulate multiphase fluid and heat flows in both porous and fractured media; and *FLAC3D*, which is used in rock and soil mechanics. *TOUGH-FLAC* includes the most advanced constitutive model of bentonite behaviour [the modified *Basic Barcelona Model* (BBM)] (Alonso *et al.*, 1990) and is capable of modelling coupled thermal, hydraulic, mechanical and gas processes. The BBM has recently been implemented into the *TOUGH-FLAC* code and its capability to simulate THM processes in bentonite demonstrated (Steeffel *et al.*, 2010). Figure 2 shows the relationships between the numerical models. The remainder of this paper is concerned with the initial modelling of the flow in the fractured rock and predicted inflow to the boreholes.

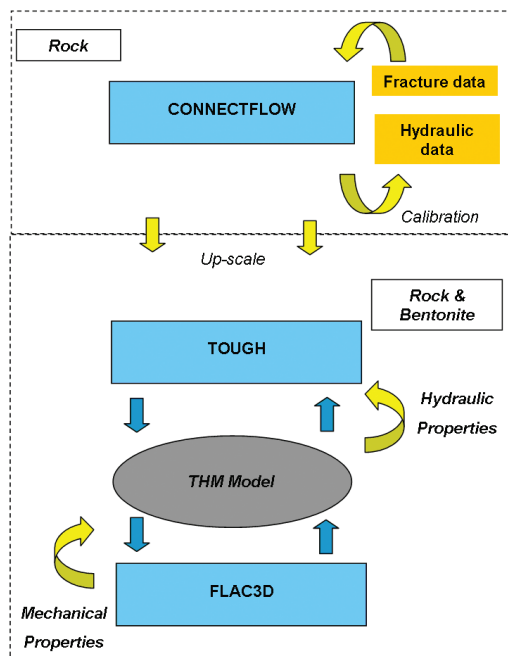


FIG. 2. Overview of the modelling process and tools used in the buffer–rock interaction experiment.

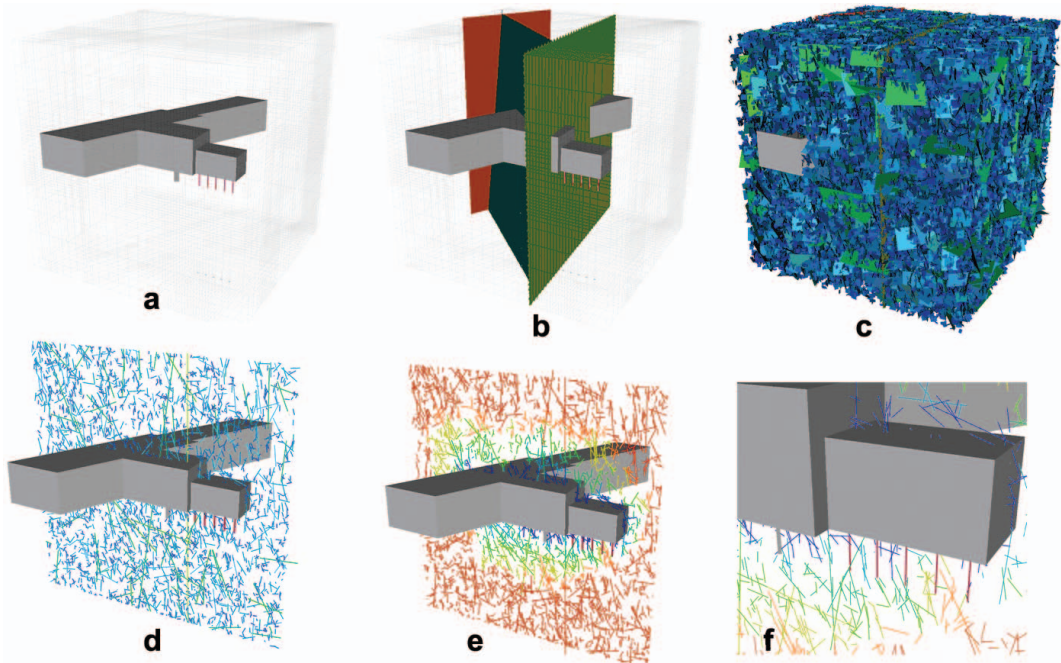


FIG. 3. Illustration of the development of the model, (a) model domain, (b) major fractures, (c) stochastic background fractures, (d) trace of fractures on a vertical slice, (e) hydraulic head distribution, and (f) hydraulic head in the vicinity of the probe boreholes.

Fracture characterization

The vicinity of the TASO tunnel at the Äspö HRL is the primary investigation area. As part of the initial characterization of the site, three deterministic structures have been identified which are contained within a local model domain, as shown in Fig. 3. Fracture statistics (orientations) have been compiled from the immediate TASO location (i.e. neighbouring tunnel locations and two deposition holes in close vicinity of the BRIE experimental

site). An initial estimate of the conductive fracture frequency, P32 (fracture area per unit volume), has been provided by the EBS Task Force (Bockgård *et al.*, 2010). This provides an estimate of the fracture area per unit volume of rock.

Model domain

The model domain is a 40 × 40 × 40 m block in the near vicinity of the TASO tunnel, as shown in Fig. 1. This shows an explicit representation of

TABLE 1. The fracture statistics for the TASO tunnel.

Set	Orientation trend	Plunge	Fischer concentration	P32	P32 0.5 m < L < 10 m
1	280	20	10	1.1	0.85
2	20	10	15	2	1.55
3	120	50	10	0.75	0.58
All Sets				3.85	2.99

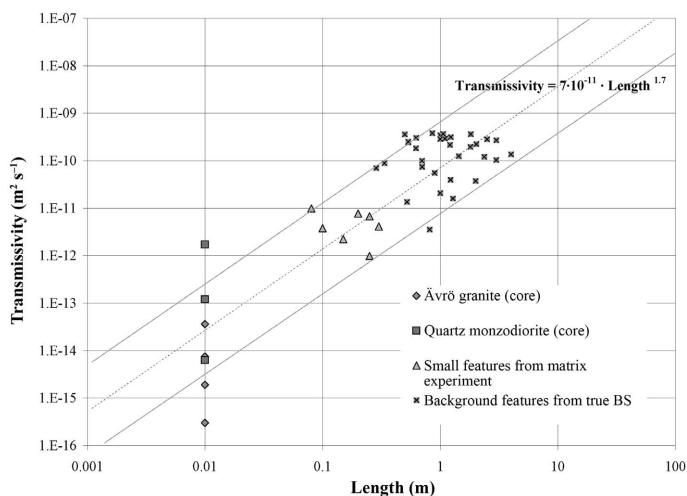


Fig. 4. Empirical power-law relationship between length and transmissivity.

the tunnel, probe boreholes and deposition holes. The boundary conditions on the outer boundary of the model have been provided by (Bockgård *et al.*, 2010) from a larger scale model of the Äspö tunnel system. The tunnel system is open to atmospheric pressure. A stochastic fracture network is simulated in the modelled domain using the fracture set descriptions given in Table 1. For the purpose of this study we have adopted a simplified power-law relationship between fracture size and transmissivity. This empirical relationship, illustrated in Fig. 4, is consistent with estimates derived from a variety of fracture scales at the Äspö site, justified using smaller-scale information. An important question is: can this statistical information (orientation, intensity and transmissivity) provide an adequate description of the fracture system to simulate the inflows into the probe holes?

Initial inflow estimates

Initial estimates of inflow (arising from opening up the holes to atmospheric pressure) into the five probe boreholes (located along the centreline of the TASO tunnel) have been modelled, shown in Fig. 3.

Ten stochastic simulations of fracture networks have been performed to simulate inflow into the probe holes. Each of these simulations generates approximately 200,000 stochastic fractures in the model domain. The results of the simulations are shown in Table 2. They show variability in inflow between boreholes, these range from below detection limit $<0.1 \text{ ml min}^{-1}$ of inflow to over 10 ml min^{-1} , as shown in Fig. 5. The range of inflows is surprisingly consistent with the range one may expect to observe statistically. In particular, one of the realizations, as shown in Table 2, is reasonably close to the observed

TABLE 2. Table of inflows to each borehole, averaged over 10 realizations, with an indication of the 'best' realization.

Borehole	Average inflow (ml min^{-1})	Realization 2	Measured inflow (ml min^{-1})
KO0020	3	0	–
KO0018	3.6	0	–
KO0017	1.5	1.3	0.5
KO0015	1.6	0.1	–
KO0014	2.6	0.9	1

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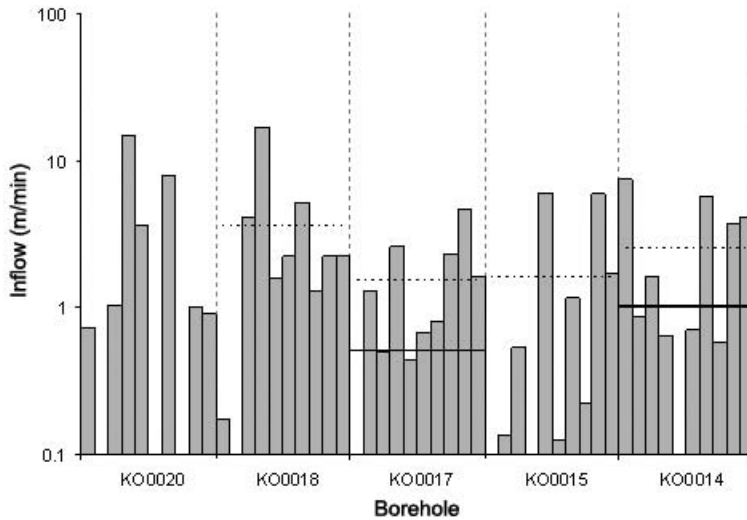


FIG. 5. Inflows to each borehole for each of the 10 flow realizations performed. The average modelled inflow is indicated as a dashed black line. The measured non-zero values are indicated by a solid line.

inflow. No direct calibration to individual boreholes has taken place in these simulations.

(Jackson *et al.*, 2000). An illustration of this, for an uncalibrated model simulation, is shown in Fig. 6.

Calibrations

The next steps in the modelling will be concerned with calibrating the models. Amongst those methods which will be explored is a new automated method using a Bayesian updating method (Cliffe *et al.*, 2011). Once the model has been suitably calibrated an up-scaling to effective properties on a suitable sub-scale will be used

Upscaling and integration with TOUGH-FLAC

The upscaled effective continuous porous media (EPM) model (effective fracture permeability on a block-scale) is then provided to *TOUGH-FLAC*, as illustrated schematically in Fig. 2. This EPM allows simulation of the rock–bentonite resaturation. This is the subject of ongoing work as part of the Äspö Task Force.

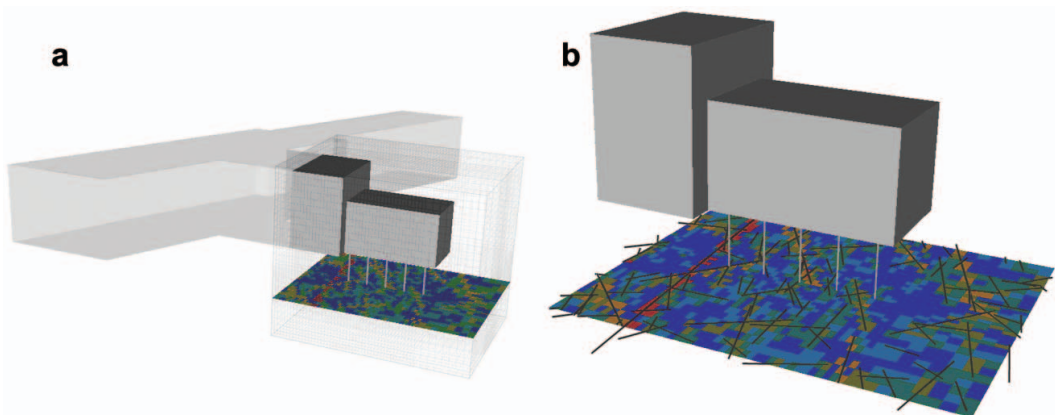


FIG. 6. (a) Horizontal slice of the upscaled permeability, computed on a sub-grid, in the vicinity of the probe holes. (b) Upscaled permeability on a ~ 0.5 m scale, with the traces of the fractures indicated.

Conclusions

In this paper research into bentonite as a buffer material has been presented in the context of its safety functions. This allows research to be articulated in terms of their barrier function (i.e. what the bentonite contributes to safety). The THMC processes that relate to the safety have been described. This leads naturally to describing the research activities in terms of understanding key couplings. This is consistent with research adopted as part of a range of international projects.

In the UK, international collaboration is important for the development of skills. The Äspö Engineered Barrier System research programme offers a good vehicle through both experimental activities and structured modelling tasks, by pooling resources, to develop the understanding of THMC coupling.

The BRIE project investigates the interaction between the rock and the bentonite. To model BRIE we have proposed a new overall modelling methodology, by integrating the description of flow in the fractured rock, calibration of the inflow measurements on a local scale, and the use of the up-scaled continuum model on a suitable sub-scale to allow the simulation of the swelling bentonite. These steps provide an integrated approach to potentially allow the use of a range of simulators to model the THM coupling, and preserve the key features of the flow in the fractured rock.

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