

## Selfsealing barriers of clay/mineral mixtures - the SB-project at the Mont Terri rock laboratory

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### Abstract

Moderately compacted clay/mineral mixtures may represent a reasonable alternative to highly compacted bentonite buffers currently studied and considered in some concepts of underground repositories for high-level radioactive wastes. In contrast to highly compacted buffers clay/sand mixtures exhibit a high permeability to gas in the unsaturated state and a comparably low gas entry/break through pressure in the saturated state while providing an adequate self-sealing potential due to swelling of the clay minerals after water uptake from the host rock. By using optimized material mixtures, the evolution of high gas pressure in the repository near-field due to corrosion of the waste containers will be avoided and possible migration of radionuclides from the waste matrix in the liquid phase through the buffer will be diffusion controlled just like in the host rock. On basis of promising laboratory results gained in GRS' geotechnical laboratory it was decided to test and demonstrate the sealing properties of clay/mineral mixtures under realistic in-situ conditions at the Mont Terri Underground Rock Laboratory (MTRL). The paper presents details about the envisaged in-situ experiments and material data obtained from laboratory investigations. First results of full-scale mock-up tests are presented as well.

In addition, information is given about further laboratory investigations and scoping calculations that have been performed to analyze whether it would be possible to achieve and demonstrate the required sealing properties within the comparably short run time of the project. It has been found that clay/sand mixtures with clay contents between 35% and 50% are suitable for the envisaged in-situ tests at the MTRL (and most likely also for adequate sealing of disposal rooms in repositories).

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*Keywords:* buffer, clay/sand mixtures, permeability, in-situ testing, modelling

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### 1. Introduction

Since about two decades, geological clay formations are investigated on their suitability to host a repository for high-level radioactive waste.

To completely seal the waste from the biosphere various disposal concepts relying on a multiple barrier system including the geological barrier and engineered barrier systems (EBS) have been developed. Some of these concepts involve highly compacted bentonite buffers between the waste containers and the wall of almost circular disposal drifts or boreholes.

Considering these concepts in more detail, there is a concern about gas pressure built-up in the near-field once formation water reaches the waste canisters after re-saturation of the buffer. Hydrogen gas will be generated by anaerobic corrosion of the canisters and/or by radiolysis of the formation water. According to Rübél et al. (2003), clay formations like the Opalinus clay provide enough water to completely corrode the vitrified HLW canisters in a disposal borehole. Up to 481 m<sup>3</sup> of hydrogen gas would be produced per canister by its complete corrosion. Because of the high swelling pressure of the bentonite buffer and its very low permeability to gas after re-saturation, gas migration is hindered and a higher gas pressure may evolve. In case the least principal stress and the tensile strength are exceeded fracturing and disintegration of the host rock may take place.

Although there are good reasons to assume that the gas pressure might be limited because of low gas generation rates and continuous gas transport by advection/diffusion and two phase flow through the EBS and the host rock, engineering measures can be used to make the system more robust.

Moderately compacted clay/sand mixtures have been found to represent a suitable alternative to the afore-described concepts. In contrast to highly compacted bentonite buffers clay/sand mixtures exhibit a high permeability to gas in the unsaturated state and a comparably low gas entry/break through pressure in the saturated state while providing an adequate self-sealing potential due to swelling of the clay minerals after water uptake from the host rock. By using optimized material mixtures, the evolution of high gas pressure in the repository near-field will be avoided and possible migration of radionuclides in

the liquid phase from the waste matrix through the buffer will be diffusion controlled just like in the host rock.

The sealing properties of clay/sand mixtures have been investigated in detail in the geotechnical laboratory of GRS within two projects, the "Two-Phase Flow" Project (Jockwer et al., 2000) and the KENTON project (Miehe et al., 2003). Seal properties such as permeability to water and gas, gas entry and break-through pressure, and swelling pressure have been determined for different mixing ratios and different degrees of compaction. Adequate sealing properties have been obtained by proper adjustment of the clay/sand ratio.

A further advantage of the clay/sand mixtures lies in the fact that they can be loosely emplaced in conventionally mined disposal drifts with non-circular, but irregular shape and rough surfaces and then reasonably compacted to get a homogeneous seal with the required density. Hence, conventional mining would be sufficient and costly mining of circular disposal drifts is not required.

The results of GRS' laboratory investigations were found quite promising and it was thus concluded to qualify and quantify the sealing function of clay/sand mixtures under representative in-situ conditions. Hence, in summer 2003, GRS started the SB (Self-sealing Barriers) project, the in-situ part of which to be performed at the MTRL.

## 2. Test objectives

The overall objective of the project is to test and demonstrate that the sealing properties of clay/sand mixtures determined preliminarily in the laboratory can technically be realized and maintained under repository relevant in-situ conditions (installation density, saturation conditions, swelling pressure, gas entry as well as break-through pressure, excavation disturbed zone).

As indicated in the introduction the following material properties are to be assured:

- High permeability to gas

One way to avoid the development of a high gas pressure in the disposal rooms is to allow the generated gases to migrate through the seal. According to the preceding laboratory investigations, the permeability to gas in the unsaturated state ranges between  $1\text{E-}13$  and  $1\text{E-}15\text{ m}^2$  and remains above  $1\text{E-}17\text{ m}^2$  after gas breakthrough in the saturated state.

- Low permeability to water

After water uptake from the host rock and saturation, the water permeability of the material reduces because of the swelling of the clay minerals. An initial value of about  $1\text{E-}17$  and  $1\text{E-}18\text{ m}^2$  is considered sufficient in analogy to the permeability of  $1\text{E-}14$  and  $1\text{E-}16\text{ m}^2$  of the excavation disturbed zone (EDZ) in the host rock (Bossart et al., 2002). It is expected that the permeability to water will reduce further as a result of ongoing rock deformation towards the backfilled drift with healing of the EDZ and further compaction of the sealing material.

- Gas entry/break-through pressure lower than the gas entry pressure of the host rock

In the experimental concept, the gas entry/break-through pressure of the sealing material must be low enough in comparison to the gas entry pressure of the host rock to ensure gas migration through the seal. According to NAGRA (2002), the gas entry pressure in the undisturbed Opalinus clay at some 600 m depth below ground amounts to about 5 MPa and thus the gas entry/break-through pressure of the seal in such a situation should be lower than 5 MPa to ensure the seal acting as a gas vent.

The conditions at the MTRL differ from these conditions. According to Thury et al. (1999), the overburden pressure at Mont Terri yields a vertical stress of only 7.25 MPa with a horizontal minor stress component of about 2 MPa. Also the porewater pressure amounts to only about 2 MPa so that the gas entry/break-through pressure of the seal in the envisaged SB-experiment at MTRL is to be kept well below 2 MPa which can be considered a conservative design value if the necessary sealing

effectiveness can be demonstrated for this condition.

- Adequate swelling pressure to obtain the desired sealing effectiveness

After water uptake, the sealing material must seal itself by swelling. It fills the entire space between the waste canister and the drift wall and any gap remaining from seal construction. High swelling pressure and the capacity for large volumetric strains under free swelling conditions are considered very advantageous (Pellegrini et al., 1999). On the other hand, laboratory experiments suggest that gas penetration of an initially water-saturated clay buffer occurs only when the gas pressure slightly exceeds the sum of the swelling pressure and the groundwater pressure (Rodwell et al., 1999). Consequently, in order to cause the gas to flow preferentially through the seal and not into the host rock, the swelling pressure should not exceed the gas entry pressure of the host rock.

### 3. Work programme

The envisaged strategy for a successful execution of the project has been set up in three steps which are:

1. Continuing laboratory investigations for final selection of suited material mixtures and development of installation/emplacement techniques;
2. Large-scale mock-up tests under well controlled, but realistic conditions for the development of suited material installation techniques and testing of measuring instrumentation;
3. In-situ testing in boreholes under representative in-situ conditions in the Mont Terri Rock Laboratory.

Details on the work done so far and envisaged in the next years are given below:

### 3.1 Laboratory investigations for selection of suited clay/sand mixtures

In these preceding laboratory investigations the material mixtures exhibiting the desired material properties with regard to installation density, swelling pressure, permeability to gas and water, and gas break-through pressure have been determined first. Then, the saturation behaviour of the selected material mixtures has been determined with special respect to the time needed for achieving full saturation of the seal in the mock-up and the in-situ tests. These first investigations have been done on small samples of 5 cm diameter and 10 cm length. In addition to this, material parameters needed for model calculations to predict the full-scale mock-up tests and the field experiments will be determined. Modelling will involve design calculations, model calibrations, and simulations for supporting adequate interpretation of the in-situ experiments.

### 3.2 Large-scale laboratory mock-up for development and testing of suitable material installation techniques and adequate instrumentation

Before going in situ, proper installation techniques assuring the realization of the required installation densities as well as determination of the time needed to reach full seal saturation in the in-situ experiments within a reasonable period of time is considered important.

Currently, respective tests are performed at GRS' surface laboratories in Braunschweig.

The detailed objectives of these mock-up tests are to:

- develop and test adequate methods for mixing clay/sand mixtures with adequate homogeneity
- develop and test methods to install the clay/sand mixture with the pre-determined dry installation density

- determine the saturation velocity of the clay/sand seal at different water injection pressures as well as the permeability to water and gas as a function of water saturation.
- test pre-selected instrumentation and a data collection system for measuring such test parameters like gas and water injection, gas and water flow, and swelling pressure
- select and test a filter material at the boundary between the porous medium and the seal which avoids penetration of fine bentonite particles into the porous medium and which ensures a homogeneous flow of water and gas through the seal

The mock-up tests are being performed in vertically arranged steel tubes with the same diameter of 0.31 m as the in-situ boreholes. The tube of 2.5 m length and the sealing material is installed in layers of about 5 to 10 cm. Different techniques (hand stamping, vibrator technique) are being tested and the achievable density is determined. In addition, the permeability to gas and water, the saturation velocity, the gas break-through pressure, and the swelling pressure will be determined in order to provide adequate experiences for the design of the in-situ experiments at Mont Terri.

### 3.3 In-situ Experiments at the Mt Terri Underground Rock Laboratory (MTRL)

The SB experiments will be conducted in a test niche of 5 m width, 4 m height and 8 m length in the MTRL (Fig. 1). In up to four boreholes of 0.31 m diameter drilled and instrumented sequentially, the sealing materials tested in the laboratory will be used and their functioning demonstrated under representative in-situ conditions. First, the installation and measuring techniques will be tested in two preceding test boreholes which will be incorporated in the

experimental programme if found representative and successful.

After seal installation in the single experiments, measurements to determine

- the gas and water permeability and
- the gas break-through pressure in the saturated stage in interaction with the surrounding rock

will be performed.

All the boreholes will be equipped with instruments for measuring different hydro-mechanical parameters. No instruments, however, will be installed in the SB seal itself to avoid any negative impact on the sealing properties. Figure 2 shows the principle design of a borehole sealing test in the niche.

The lower part of the boreholes, the injection volume, will be filled with a porous material (e.g. sand). At top of the porous medium a filter frit will be placed for ensuring a homogeneous distribution of the injected water over the entire borehole cross section. Above the filter frit, the clay/sand-seal will be installed in several layers to a height of 1 m. Above the seal a further filter frit will be installed for water and gas collection. The whole borehole will be sealed against the ambient atmosphere by a gastight packer. The most upper part of the test borehole is grouted for keeping the packer in place at higher swelling pressure of the SB seal.

For saturation or desaturation of the seal, water or gas can be injected through an injection tube running from a valve panel in the test room via an inclined borehole into the lower injection volume.

The water or gas flowing through the seal is collected in the upper collection volume by a further tube running back to the control valve panel where gas and water flow rates and pressures will be controlled and measured.

After termination of the in-situ tests, samples will be drilled from the seal and the surrounding host rock for post-test laboratory analyses of the conditions achieved in the demonstration tests with regard to saturation, homogeneity of the saturation, porosity, etc.

#### 4. Actual project results

So far, after start up of the project in 2003, the laboratory investigations on the selection of the suited material mixtures have been terminated and the large-scale mock-up tests were started in summer 2004.

##### 4.1 Laboratory investigations

In order to select optimized material mixtures, preceding laboratory experiments were performed on material mixtures with clay contents of 35 %, 50 % and 70 %. The most important material properties as criteria are the installation density and the porosity, respectively, the water permeability, the gas break-through pressure, and the gas permeability after break-through.

Table 1 summarizes ranges and mean values (in parentheses) of the determined properties for the investigated mixtures and compares them to the requirements described in section 2. It is obvious that the 35clay/65sand and 50clay/50sand mixtures meet the requirements completely. It can be expected that the gas break-through pressure may reduce further in the case of significantly lower gas generation rates which are expected in a real repository. The extrapolation of the test results suggests that the 70clay/30sand mixture may have higher swelling and gas break-through pressure than the given upper limit.

Based on these results, mixtures with clay/sand ratios of 35/65 and 50/50 are currently used in the mock-up tests.

Table 1: Comparison of the measured parameters to the requirements given in section 2

Measured parameters at installation conditions					
Clay/sand ratio of sample	Gas permeability under dry conditions	Initial water permeability at full saturation	Gas break-through pressure	Gas permeability after gas break-through	Swelling pressure
	m <sup>2</sup>	m <sup>2</sup>	MPa	m <sup>2</sup>	MPa
35/65	1.2E-13	3.3E-17 - 9E-18 (5.2E-18)	0.4 - 1.1 (0.75)	1.1E-17 - 1.6E-17 (1.4E-17)	0.2 - 0.4 (0.28)
50/50	7.5E-14	1.1E-18 - 4.3E-18 (2.2E-18)	0.4 - 2.8 (1.83)	5.5E-18 - 6.2E-18 (5.9E-18)	0.3 - 0.5 (0.35)
70/30	1.2E-15	5.5E-19		n.d.	0.4-?
Requirements					
	Gas permeability under dry conditions	Initial water permeability at full saturation	Gas break-through pressure	Gas permeability after gas break-through	Swelling pressure
	high	1E-17 - 1E-18	2	high	2

## 5 Modelling

Preceding both the mock-up and in-situ experiments are scoping calculations to enable proper design of the experiments and to validate the used computer codes and THM models by comparing modelling and test results.

GRS applies the code CODE\_BRIGHT developed by the Technical University of Catalonia (UPC) in Barcelona. The theoretical framework employed in the code is presented in (Olivella 1996) and (UPC 2002) and reviewed in (Gens 1998), (Alonso 2002), and (Zhang 2004).

The Barcelona Basic Model (BBM) implemented in CODE\_BRIGHT is an elastoplastic model able to represent many mechanical features of unsaturated soils in a consistent and unified manner. In the framework of the project it was used for the assessment of the mechanical behaviour of the sealing materials and the Opalinus clay.

Gas and water flow was modelled according to Darcy's law and the molecular diffusion of water vapour is governed by Fick's law. The mass of water vapour per unit volume of gas is determined via the psychrometric law and the solubility of air

in water is controlled by Henry's law. The hydraulic parameters for the clay/sand mixtures such as relative permeability and capillary pressure as functions of saturation were established by extrapolation of the two-phase flow data obtained in the KENTON project (Jockwer 2000) and additionally validated through special laboratory saturation tests on small samples.

The scoping calculations for designing the tests have been conducted by using material parameters preliminarily determined during the preceding laboratory tests (Rothfuchs et al., 2005) and taken from literature. The parameters for the Opalinus Clay were taken from the literature as well (Zhang 2004).

The calculations focused on prediction of testing conditions such as adequate injection pressures for water and gas, duration of water saturation, ranges of measuring parameters (gas and water flux, swelling pressure, total pressure etc.), and determination of initial and boundary conditions in the in-situ test field. In the scoping calculations, the materials installed in the mock-up and in-situ tests were assumed as homogeneous and isotropic. Processes prevailing in the materials during the tests were considered as coupled THM

processes, so that the following balance equations were to be solved: balance of energy, balance of water mass, balance of air mass, and balance of momentum (equilibrium).

### 5.1 Scoping calculations for the mock-up tests

Figure 3 shows the experimental set-up in GRS' laboratory, the FE-model, and the test procedure of the mock-up tests, which have been started summer 2004. The mock-up tests are designed as a full-scale replica of the envisaged in the in-situ experiments (Fig. 2). The only deviation from reality is the steel tube simulating the 0.31 m wide and 3 m deep test borehole at MTRL. According to the results of the scoping calculations (Fig. 4) full saturation of a 35Clay/65sand seal may be reached after about 170 days if an injection pressure of 1 MPa is applied. This was confirmed by the first mock-up test, which was started summer 2004. After about 4 months (120 days), the water outflow rates are still a bit smaller than the inflow rates (Fig. 5) indicating that the seal is still not fully saturated and is still absorbing small amounts of water. The rates, however, are more and more equalizing and full saturation and steady state will be reached soon.

From Fig. 5 it can be taken that the permeability will range between  $1\text{E-}17\text{ m}^2$  and  $1\text{E-}18\text{ m}^2$ . This result is promising and a similar seal behaviour can be expected in the in-situ experiments. The final steps in the first mock-up test after having reached full water saturation (Fig. 3) will be to determine the seal swelling pressure at reduced water injection pressure and to measure the gas break-through pressure together with the resulting effective permeability to gas.

### 5.2 Scoping calculations for the in-situ-experiments

Scoping calculations for the in-situ experiments are necessary to obtain an assessment of initial and boundary conditions and the duration of the envisaged flow experiments. Due to

excavation and ventilation of the test niche at the MTRL, the hydro-mechanical state of the surrounding rock is disturbed. Additionally, the THM conditions around the boreholes drilled down from the floor of the test niche (Fig. 2) are to be assessed. During injection of water and gas into the seal, coupled hydro-mechanical processes will occur not only in the seal but also in the surrounding rock.

The finite element mesh, the boundary conditions, and the different materials installed in the borehole are shown in detail in Fig. 6.

In the model, the materials were assumed homogeneous and isotropic. Both clay/sand mixtures with clay contents of 35% and 50% were considered in the calculations. Because of a lack of data for the injection chamber, packer and concrete, the properties and mechanical parameters of the clay rock were used for them. A high permeability of  $1\text{E-}12\text{ m}^2$  was applied to the injection chamber, while the packer was assumed to be impermeable. Such simplifications are considered acceptable for the purpose of scoping calculations focusing on the determination of hydro-mechanical processes in the seal and the surrounding rock.

In the calculations, the following in-situ conditions at the MTRL were taken into account. The temperature in the rock and in the niche is  $17\text{ }^\circ\text{C}$  for the initial state. A vertical total stress of 6 MPa applied on the top boundary and the gravity effect result in an initial vertical total stress equal to 6.48 MPa at the level of the niche floor. Assumption of a ratio  $K_0 = 0.77$  of effective horizontal stress to vertical stress leads to an initial total horizontal stress of 5.2 MPa at the floor level. A water pressure of 0.8 MPa supplied to the top boundary and its hydrostatic distribution in the model region result in an initial pore water pressure of 1.0 MPa at the floor level. An atmospheric pressure of 0.1 MPa was taken as the initial gas pressure. Flow of water and gas through the other boundaries is not allowed.

The hydraulic response of the rock mass to excavation and ventilation of the niche and borehole six months after niche excavation and

eight days after borehole drilling is shown in Fig. 7. Just after the niche excavation, the porosities in the zones over the roof and under the floor of the niche expand somewhat due to the stress relaxation. This induces a sudden reduction of the pore water pressure even to a negative value (suction) of  $-1$  MPa. In contrast to this, highly concentrated stress near the lower corner of the borehole compresses the material and hence generates a high pore water pressure of up to 6 MPa. During ventilation with an relative humidity of 85 %, the pore water pressure reduces steadily. Six months later, the zone with negative pore water pressure extends to about 1 m from the niche wall into the rock mass. The borehole drilling induces an additional dilatancy of the surrounding rock and hence a further reduction of the pore water pressure. Due to excavation and ventilation of the niche and the borehole, the surrounding rock is desaturated. Figure 8 shows the respective distribution of water saturation in the surrounding rock at the end of borehole drilling and ventilation. The de-saturated zone with a water saturation level of less than 95% is limited to 0.5 m to the niche wall. The de-saturation mainly caused by the dilatancy of the clay rock is not significant.

After installation of the seal in the borehole, the water injection phase is simulated by applying a water pressure of 0.5 or 1 MPa to the lower porous chamber. Figure 9 illustrates the evolution of water saturation at some selected points in the 35clay/65sand seal at an injection pressure of 1 MPa. The time needed for full saturation at an injection pressure of 1 MPa is about 10 months which is slightly longer than the saturation times of 6 months in the mock-up tests. Because the permeability of the seal is higher than that of the surrounding clay rock (EDZ was not simulated here), the water flow occurs mainly through the seal. This situation will be assessed during the in situ experiments by comparing the inflow and outflow rates.

Generally, the modelling data indicate that - provided the modelling parameters can be realized in situ reasonably - the required sealing function of

the considered clay/sand-mixtures can be successfully demonstrated.

## 6. Summary and conclusions

In the GRS-project “Selfsealing Barriers of Clay/Mineral-Mixtures (SB)” clay/sand mixtures are being investigated as an alternative to highly compacted bentonite rings being considered as EBS in the design of underground repositories for radioactive wastes.

Clay/sand mixtures exhibit a high permeability to gas in the unsaturated state and a comparably low gas entry/break-through pressure in the saturated state while the permeability to water is low ranging in the same order of magnitude like that of the host rock. Thus, the evolution of undesired high gas pressures in the repository will be avoided and possible migration of radionuclides from the waste matrix through the buffer in the liquid phase will be diffusion controlled in the sealing material as in the host rock.

Under consideration of the required sealing properties and the comparably short testing time available within the scope of the project, both preceding lab investigations and scoping calculations have shown that clay/sand-mixtures with clay contents between 35% and 50% may be suitable for both, in-situ testing at the Mt. Terri Rock Laboratory and reasonable sealing of repositories.

## 7 Acknowledgements

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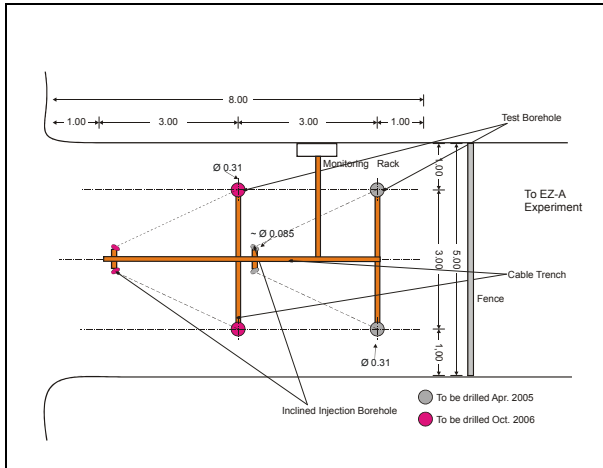


Figure 1: Arrangement of boreholes in a niche at the Mt. Terri Rock Laboratory

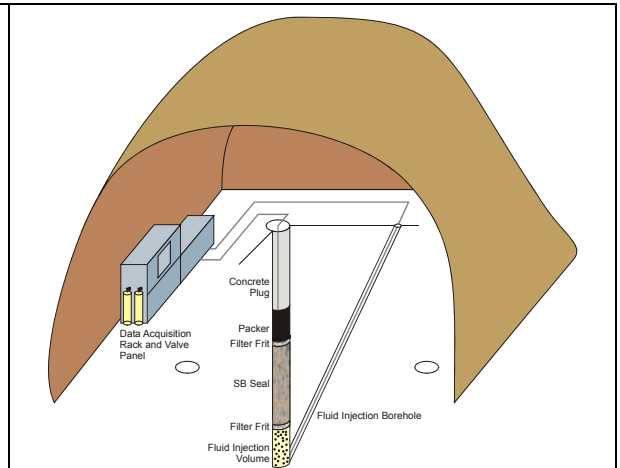


Figure 2: Principle design of a SB-borehole sealing test

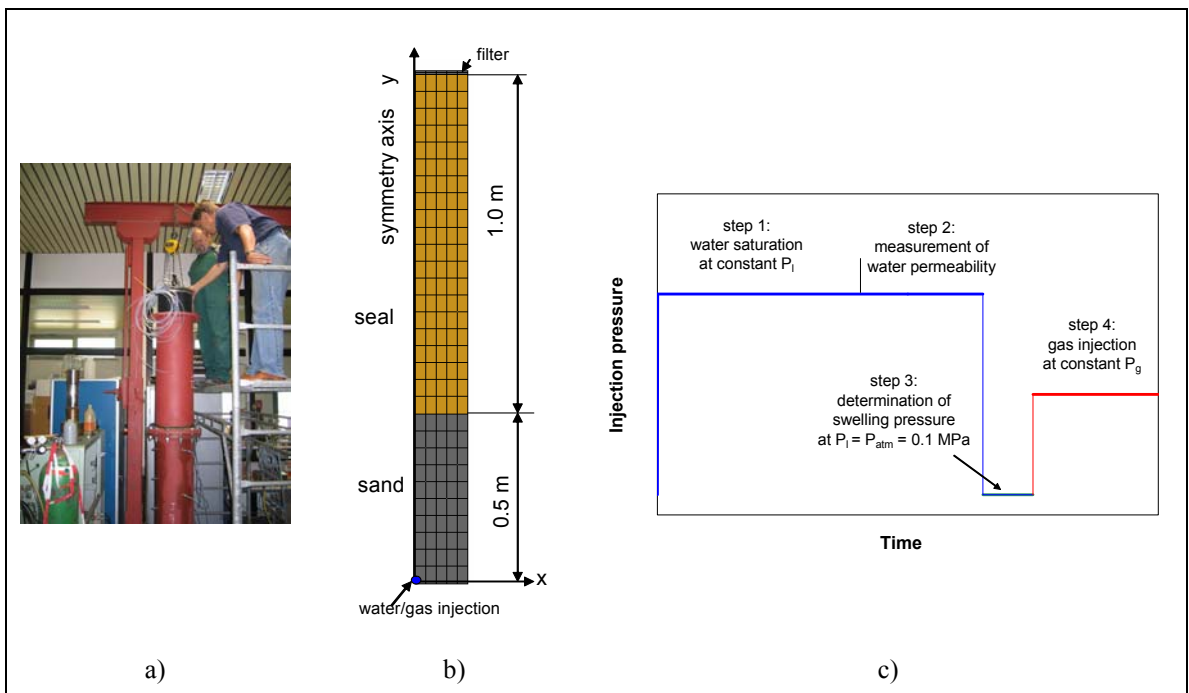


Figure 3: a) Experimental set-up, b) FE-model, and c) test procedure of the SB Mock-Up tests at GRS' geotechnical laboratory in Braunschweig/Germany

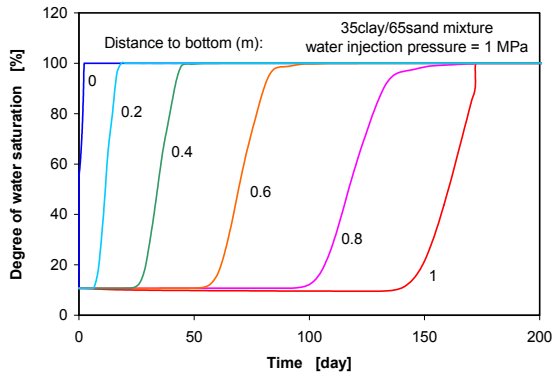


Figure 4: Evolution of water saturation in 35clay/65sand seal at an injection pressure of 1 MPa

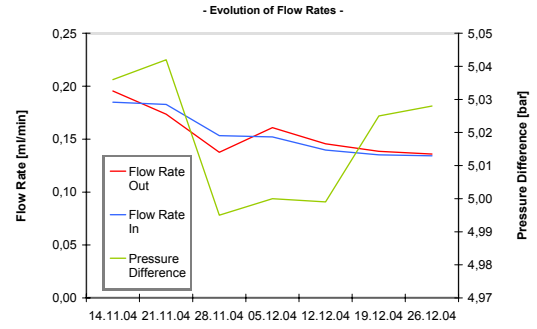


Figure 5: Water inflow and outflow rates measured in the first SB Mock-Up experiment

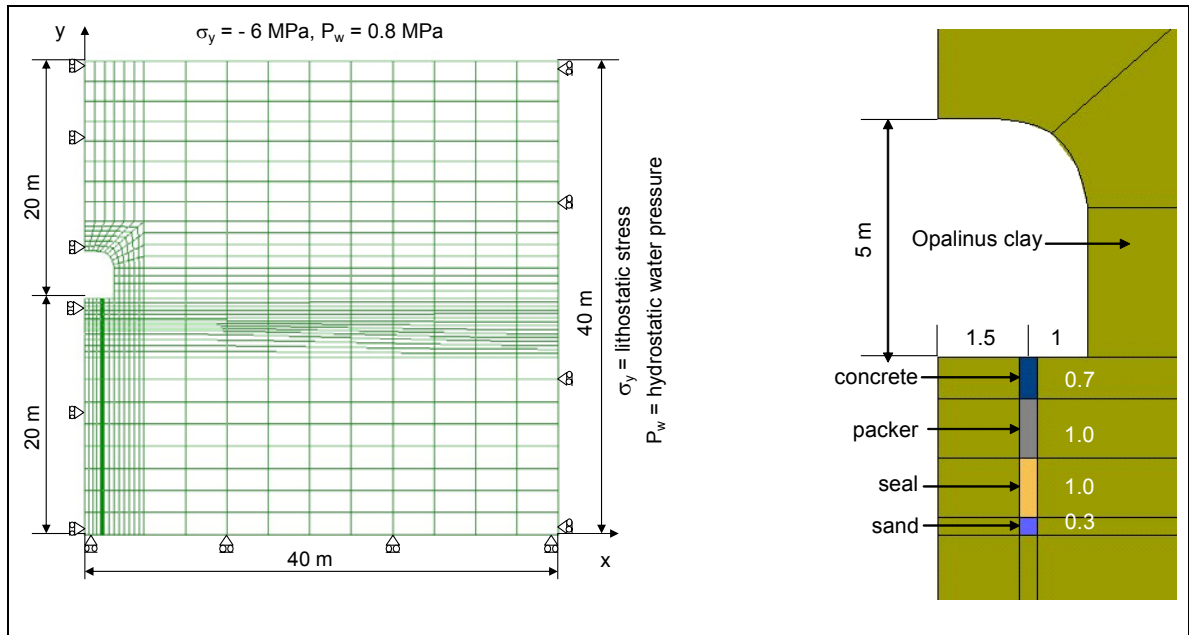


Figure 6: Numerical model and materials considered in scoping calculations

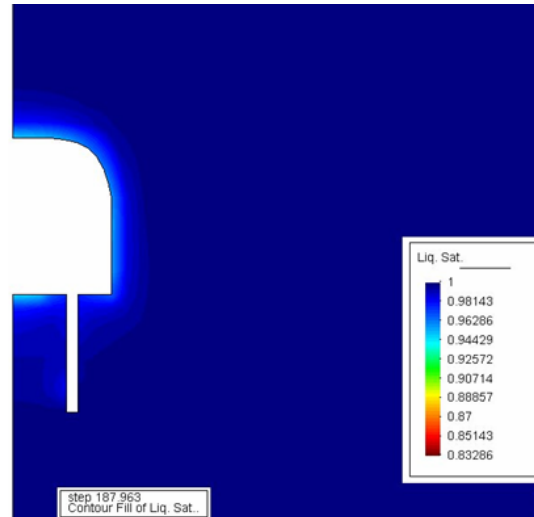
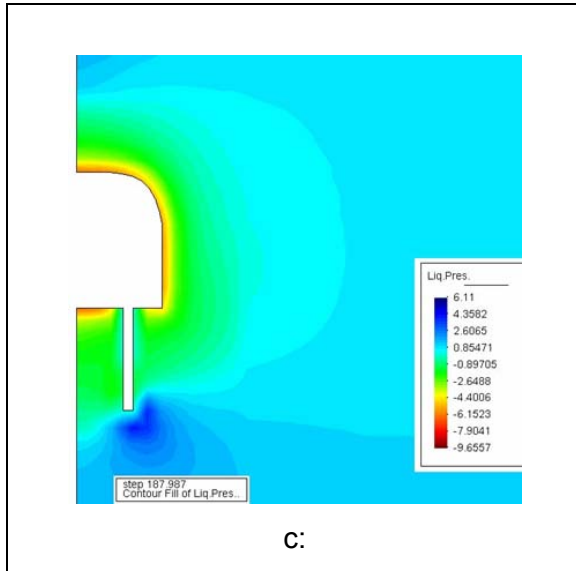


Figure 7: Redistribution of pore water pressure in the surrounding rock induced by excavation and ventilation of SB niche and borehole, 8 days after borehole drilling

Figure 8: Distribution of water saturation in the surrounding rock

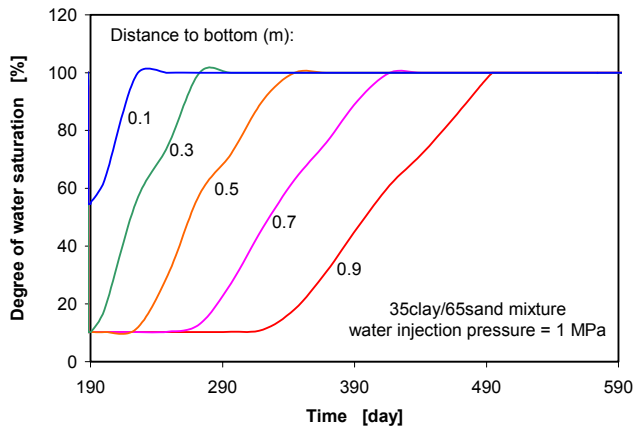


Figure 9: Evolution of water saturation in 35clay/65sand seal at an injection pressure of 1 MPa