
Qualification of Clay Barriers in Underground Repository Systems

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Abstract: After closure of radioactive waste repositories all residual openings like boreholes, entrances to emplacement chambers, galleries, and shafts are to be sealed to avoid a release of radionuclides by the migration of fluids into the biosphere. Among others, compacted clay and clay-sand mixtures are currently investigated as major seal materials.

When considering fluid migration, two-phase flow effects have to be taken into account because the pore volume of the seal materials is filled with air or gases generated by anoxic corrosion, and while in the long-term stage water or brine might flow into the repository. Within a R&D programme the parameters of single- and two-phase flow for clay-sand mixtures are determined at the GRS laboratory in Braunschweig/Germany.

In order to analyse the important fluid distribution in the seal materials or geotechnical barriers, respectively, GRS performs geoelectrical monitoring in the laboratory as well as in situ. In-situ measurements are currently performed or prepared at different test sites, as for instance at Tournemire (France), Mont Terri (Switzerland), and ÅSPÖ (Sweden), where saturation and desaturation effects in various barriers or host rocks are of primary interest.

This paper summarises the results of the laboratory and in-situ measurements obtained so far and describes the further envisaged in-situ investigations.

1. INTRODUCTION

The long-term safety of a repository for radioactive waste in deep geological formations is to be ensured by means of a sealing system consisting of natural and technical barriers. Typical seal components are borehole or drift seals, dam constructions, and shaft seals.

One important boundary condition is the possible gas generation from the waste or from the anoxic corrosion of waste containers. For reasons of safety, seals must be designed in such a manner that an unacceptable increase of the gas pressure and an uncontrolled release of gases and radionuclides into the repository are avoided. On the other hand, more or less impermeable seals are required to avoid water or brine inrush into repository sections. The water content of rocks and seal materials has significant influence on other petrophysical parameters, e.g., effective permeability. Hence, measurement of the water content of the rock and seal materials and its changes due to mining activity is an important task to assess the long-term petrophysical properties of the rock. Among others, geoelectrical monitoring is applied for this purpose.

In order to provide relevant experimental data on the mechanic, hydraulic, and electric behaviour of claystone and clay-seal materials GRS Braunschweig has performed respective laboratory and in-situ investigations for a couple of years.

2. LABORATORY INVESTIGATIONS OF CLAY-SAND MIXTURES AND CLAYSTONE

In areas where a sudden water or brine inrush is considered possible, for instance in the shaft of a repository, an instantaneous sealing may be required. This can be achieved by use of the swelling potential of clay-sand mixtures. The hydraulic behaviour of representative mixtures was therefore investigated in a laboratory investigation programme. Since clay formations are also considered as potential host rock formations, the hydraulic behaviour of claystone was investigated, too. [1].

In advance to geoelectrical in-situ measurements laboratory investigations were performed to assess the resolution and applicability of the tomographic method and to investigate the relation between the water content and the resistivity of the clay.

2.1 Determination of the Hydraulic Properties of Clay-Sand Mixtures and Claystone

For the description of the hydraulic properties of clay-sand mixtures and claystone the single and two-phase flow behaviour and the effect of diffusion are to be investigated.

2.1.1 Clay-sand mixtures

The starting materials for these investigations were ordinary sand with grain sizes between 0 and 2 mm and a bentonite clay (Calcigel). The components were mixed with the sand-clay ratios of 90/10, 75/25 and 50/50 and additional water with the constant clay-water ratio of 2.5. From these mixtures solid samples were pressed in a mould at pressures up to 100 MPa. Afterwards the samples were dried at room temperature with silicagel to constant weight. Thus, the excessive water added for the preparation was withdrawn. The porosity of the samples was evaluated from their bulk density and the grain densities of the sand and clay. The bulk densities of the samples were in the range between 1.97 and 2.07 g/cm³ and the porosities between 20.4 % and 22.6 %. Both parameters were not significantly dependent on the composition and preparation pressure. Obviously, the sand particles act as supporting frame that prevents any further compaction.

The porosity was also determined from the amount of water necessary for the complete saturation of the samples. The resulting values were by 2 to 5 % higher than those determined from the densities. This may be caused by water adsorption onto the intermediate layers of the mica minerals. The permeabilities of the dry samples to dry nitrogen were in the range between $6 \cdot 10^{-14}$ to $3 \cdot 10^{-13}$ m². No significant dependence on the sand-clay ratio, the preparation pressure, the confining pressure and the gas injection pressure has been found (Figure 1).

Permeability measurements with nitrogen of 55 % and 100 % relative humidity indicated that the samples adsorbed 50 % of the water from the gas flow but the permeability showed no significant decrease (Figure 2).

The single-phase water permeabilities were dependent on the sand-clay ratio. Samples with 10 % clay had permeabilities in the range between $5.3 \cdot 10^{-15}$ and $2.9 \cdot 10^{-17}$ m² and those with 25 % clay in the range between $7.7 \cdot 10^{-18}$ and $2.4 \cdot 10^{-18}$ m². Samples with 50 % clay could not be saturated with water in a reasonable time and therefore any determination of water permeability was impossible. This might be explained by the swelling of the dry clay in contact with water, and in the sample with 50 % clay all pores may have closed. Therefore water migration and water flow was hindered.

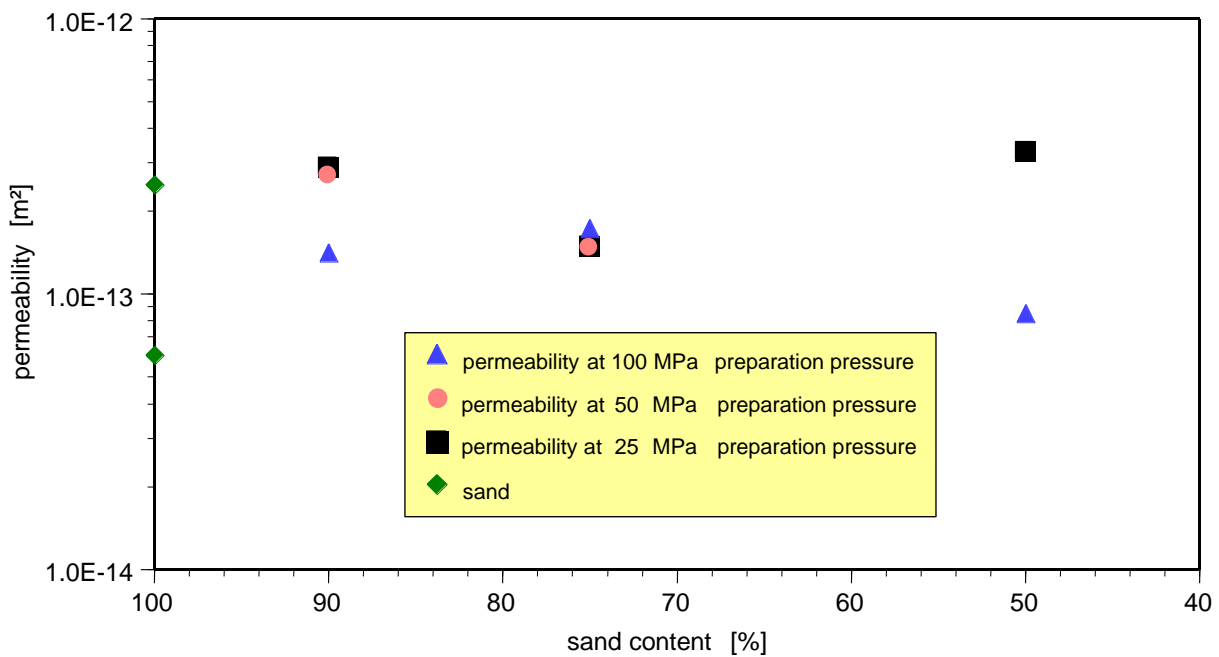


Figure 1 Gas permeability of the dry samples for various sand-clay ratios and preparation pressures

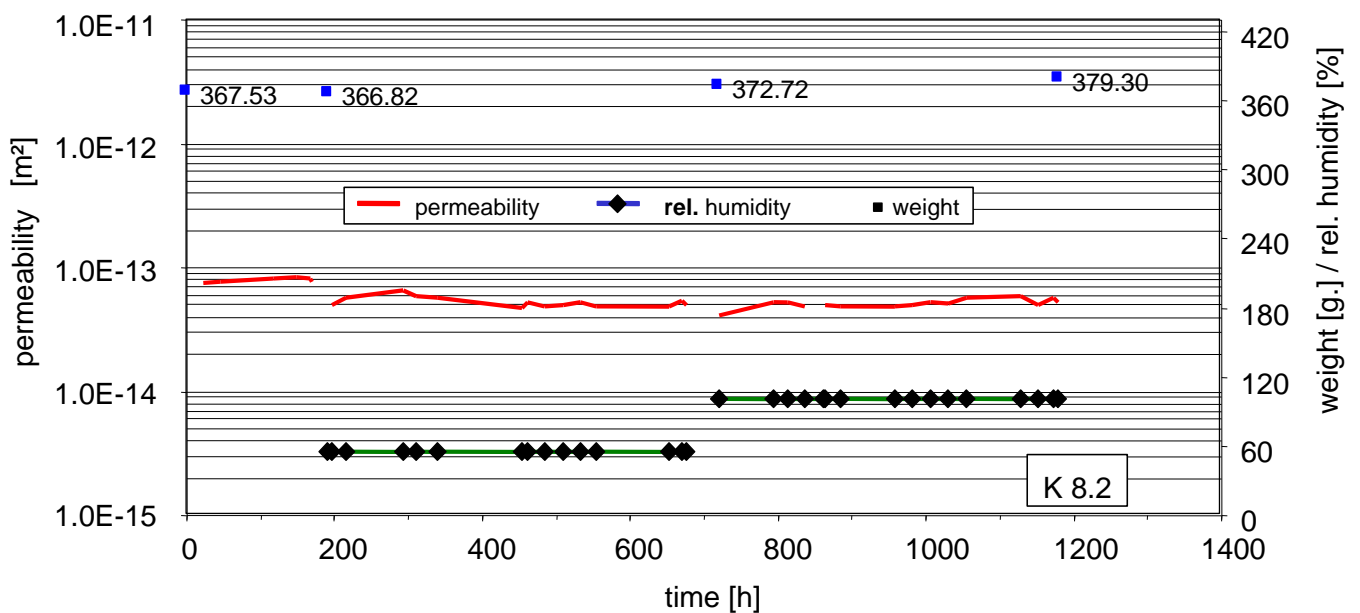


Figure 2 Gas permeability as a function of relative humidity of the nitrogen of 55 and 100 % (sand-clay ratio of the sample 75/25, preparation pressure 50 MPa, hydrostatic confining pressure 0.5 MPa)

The investigations on two-phase flow showed that the water-saturated samples with 10 % clay have gas breakthrough pressures between 0.03 and 0.05 MPa. At those pressures 80 % of the water in the pore volume was displaced. A pressure increase to 0.3 MPa only displaced another 5 to 10 % of the water. That means that the pore volumes have a residual water saturations of 10 to 20 %. These results indicate that about 80 % of the porosity is interconnected and has an almost uniform pore size distribution [2].

The samples with 25 % clay showed a non-uniform behaviour. The breakthrough pressure varied between 0.05 and 0.2 MPa and showed no plateau level in the capillary pressure-saturation relation. An increase of the gas pressure led to a further desaturation of some samples, whereas for other samples the increase of the gas injection pressure up to 0.5 MPa gave no further desaturation. Below the gas

injection pressure of 0.5 MPa the effective gas permeability was in the range of 10^{-16} m^2 . All these results indicate that water saturation leads to a swelling of the clay. Below a clay content of 25 % this swelling is too low to completely close the porosity of 20 to 22 %, and some porosity remains open for gas and water flow. Above 25 % of clay content the entire porosity is filled up by swelling and allows no advective flow. For desaturation very high pressures are necessary.

In a gas-water system the parameters of two-phase flow, such as the relative permeability and the capillary pressure, can be described by the relations of Brooks-Corey [3] or van Genuchten [4]. With a fit of these relations to the measured values of gas and water permeability, remaining gas and water saturation, as well as capillary pressure, the parameters of two-phase flow were determined. The comparison of these fits indicates that the relation of van Genuchten (Figure 3) is more adequate to describe the experimental results.

The investigations of the sand-clay mixtures indicate that the permeability of that kind of sealing material can be designed with respect to expected gas generation in a repository by choosing mixtures with defined clay content below 50 %.

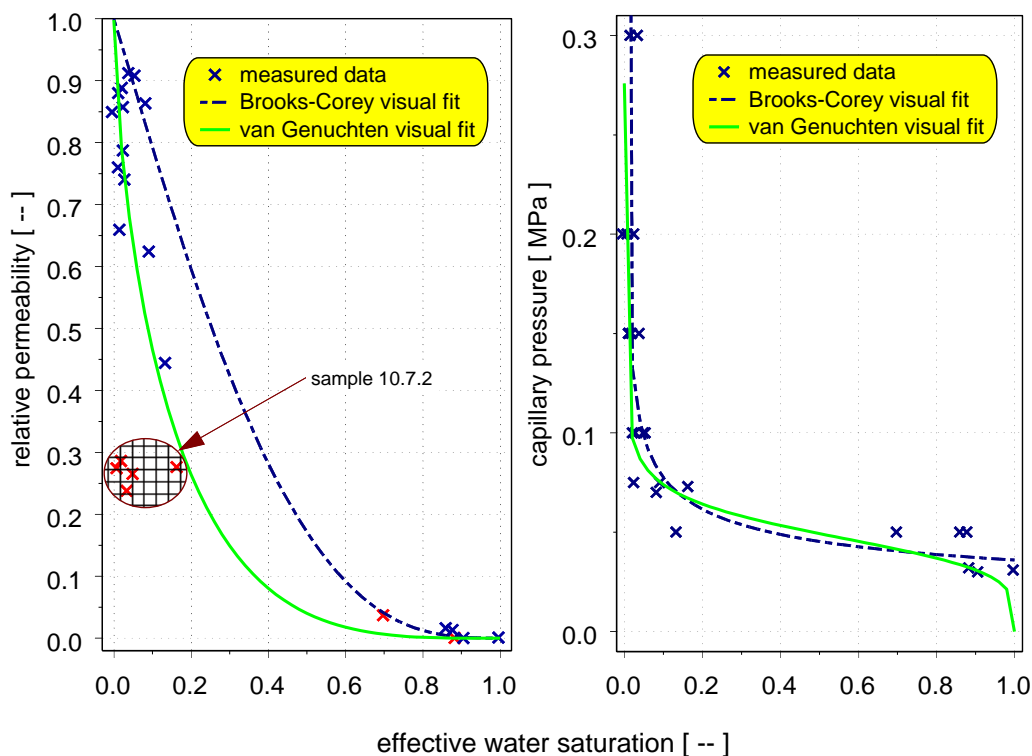


Figure 3 Relative permeability and capillary pressure in dependence on the effective water saturation for the samples with 10 % clay. Fits to Brooks-Corey and van Genuchten relation

The diffusivity of the gases hydrogen, helium, methane, and carbon dioxide were determined in the dry and water-saturated stage of the compacted samples. In the dry stage with air in the pores the diffusivity was found to be in the range between $1.4 \cdot 10^{-8}$ and $4.15 \cdot 10^{-7} \text{ m}^2/\text{s}$. Figure 4 shows the diffusivity of hydrogen for different sand contents.

The values do not depend on the gas component, but on the compaction pressure and the sand-clay ratio. The diffusivity of the samples with a clay content of 25 or 50 % is lower by a factor of 2 to 3 compared to that of the samples with a clay content of 10 %. By increasing the compaction pressure from 25 to 100 MPa, the diffusivity decreases by a factor of 5 to 30. These results indicate that sand-clay mixtures in the dry stage are open systems for gas diffusion.

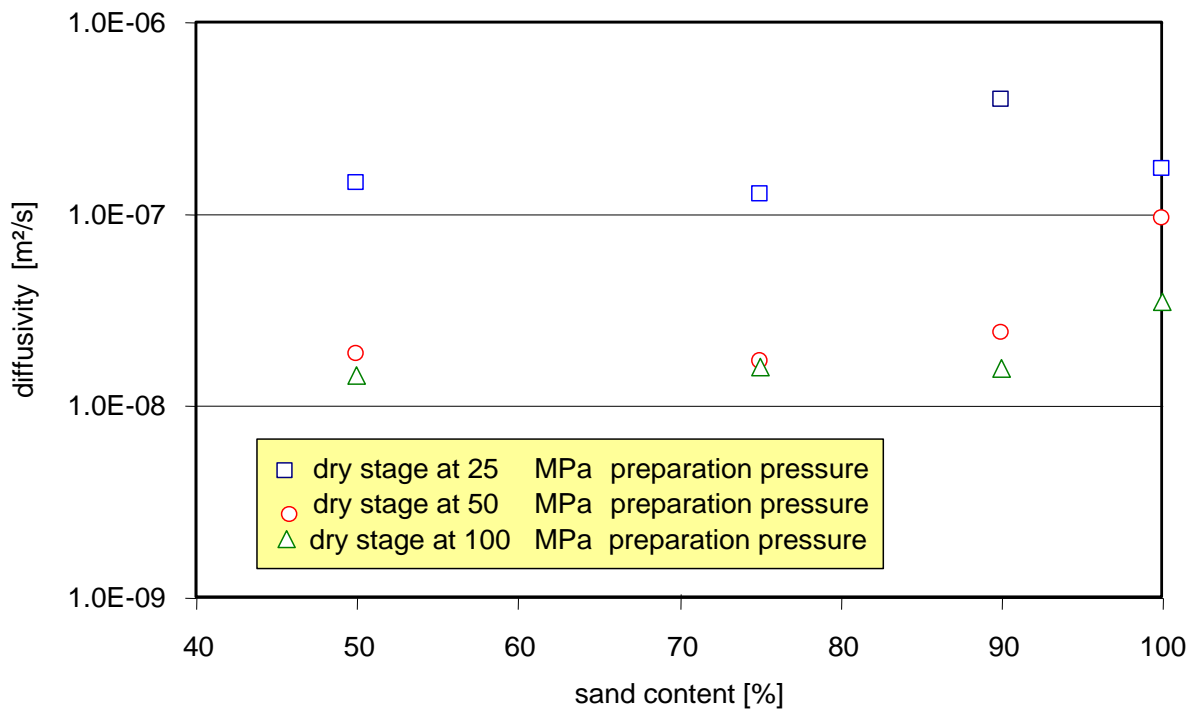
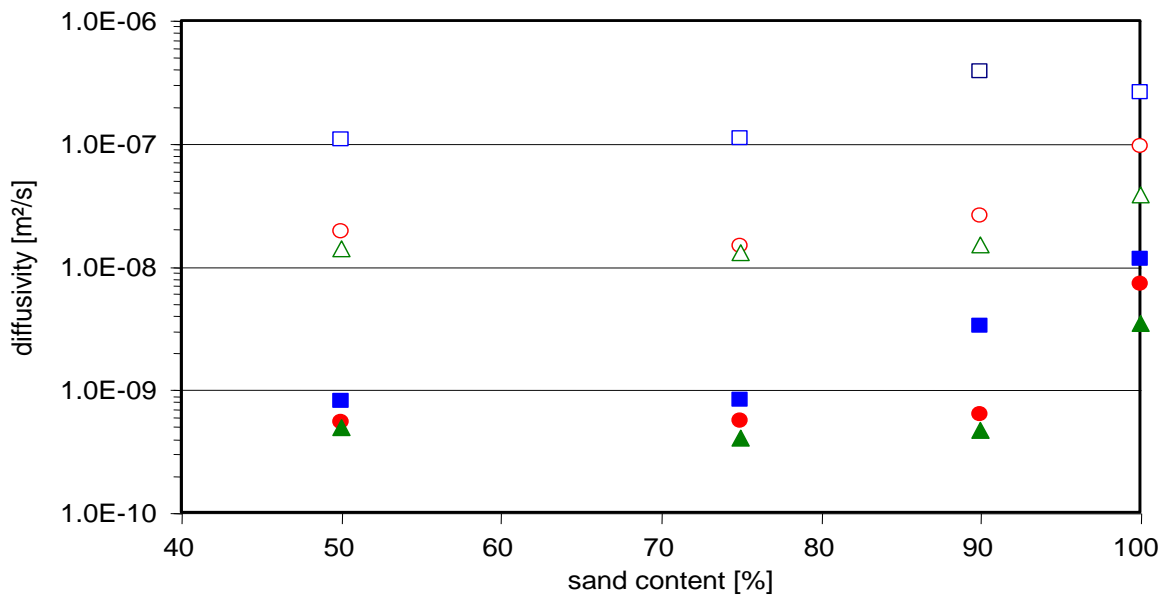


Figure 4 Diffusivity of hydrogen in sand-clay mixtures in the air dry stage as a function of sand content for different preparation pressures

In the water-saturated stage the gas diffusion is highly dependent on the sand-clay ratio and the compaction pressure. In Figure 5, this result is demonstrated for the diffusivity of carbon dioxide. Furthermore, the diffusivity of carbon dioxide is higher than that of hydrogen, helium and methane. The reason is that the solubility of carbon dioxide in water is by a factor of 25 to 105 higher than of the other gases.



□ dry stage at 25 MPa preparation pressure
 ○ dry stage at 50 MPa preparation pressure
 △ dry stage at 100 MPa preparation pressure

 ■ wet stage at 25 MPa preparation pressure
 ● wet stage at 50 MPa preparation pressure
 ▲ wet stage at 100 MPa preparation pressure

Figure 5 Diffusivity of carbon dioxide in sand-clay mixtures in the air dry stage and water-saturated stage as a function of compaction and sand content for different preparation pressures

The diffusivity of hydrogen, helium, and methane could only be determined in samples of pure sand or with a clay content of 10 % and a compaction pressure of 25 MPa. The values are in the range between $2.5 \cdot 10^{-9}$ and $2.7 \cdot 10^{-9}$ m²/s. Samples with higher clay content and higher compaction pressure have a diffusivity for the component hydrogen, helium and methane below the detection limit of 10^{-10} m²/s. The diffusivity of carbon dioxide ranges between $4 \cdot 10^{-10}$ and $1.14 \cdot 10^{-8}$ m²/s. It decreases with increasing clay content and compaction pressure.

2.1.2 Claystone

Concerning the permeability to gas and water the samples of the Boom clay and Opalinus clay from the underground laboratories HADES (Mol/Belgium) and Mont Terri (St. Ursanne/Switzerland) were different from the compacted clay-sand mixtures.

The Boom clay samples were water saturated and showed a plastic behaviour. Up to a confining pressure of 5.0 MPa and a gas injection pressure of 4.75 MPa no gas flow through the samples could be measured (Figure 6). That means that the permeability is below 10^{-22} m².

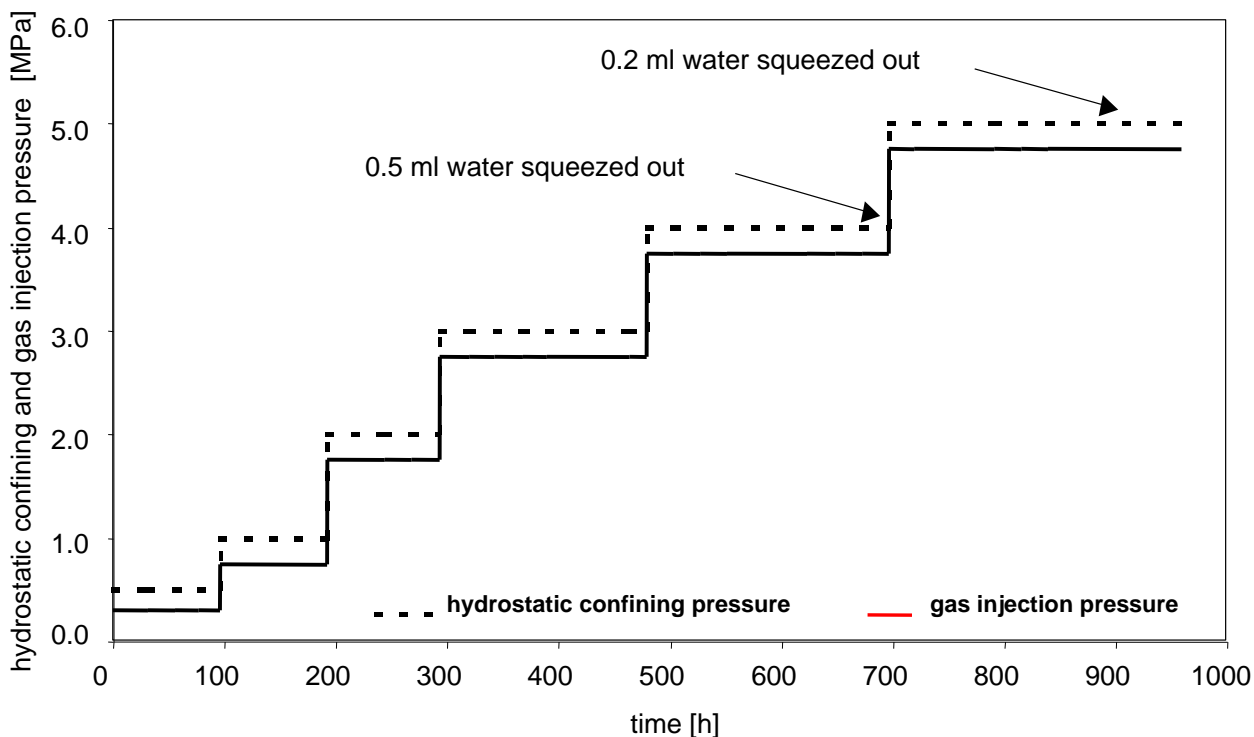


Figure 6 Investigation on permeability for different gas injection and confining pressures of a Boom clay sample from the underground laboratory Hades (Mol/Belgium)

The Opalinus clay showed a brittle behaviour and the gas permeabilities of the five investigated samples were different. At a confining pressure of 0.5 MPa two of the samples had a gas permeability of 10^{-13} m², whereas the other three had a permeability in the range between 10^{-17} to 10^{-16} m². Increasing the confining pressure up to 5.0 MPa the permeability decreased by 1 to 3 orders of magnitude. Reducing the confining pressure again to 0.5 MPa the permeability increased, but was by a factor of 2 to 10 lower than the initial permeability (Figure 7), which represents a clear elasto-plastic behaviour of the Opalinus clay.

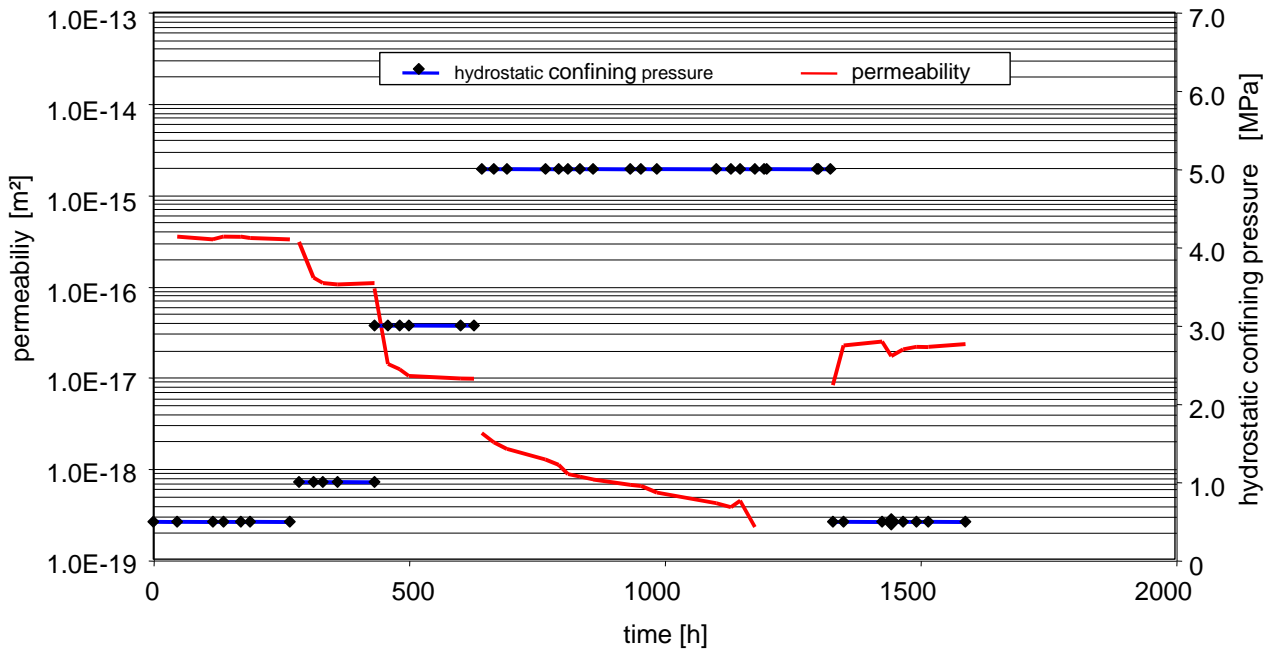


Figure 7 Permeability of Opalinus clay samples from the underground laboratory Mont Terri (St Ursanne, Switzerland) for different confining pressures between 0.5 and 5 MPa

Water and two phase permeabilities could be determined neither for the Boom clay nor for the Opalinus clay samples. If the clay comes into contact with water, swelling of the clay minerals starts and all existing pores are closed. In contrast to that, in clay-sand mixtures a two-phase flow exists at lower clay content and can be influenced by the clay-sand ratio. At higher clay content, however, a two-phase flow was also not observed.

2.2 Determination of the Electric Properties of Clay

Resistivity measurements to determine the water content of rocks and sealing materials were applied by GRS to many different types of rock (e.g., clay, rock salt, anhydrite, granite). In hard rocks and seal materials, the electric resistivity is influenced mainly by the fluid content, the porosity, the structure of porosity, and the resistivity of the pore fluid. With decreasing fluid content, the resistivity increases. Most clays show a relatively low resistivity even if there is no free water in the pore space. This is due to the integration of water into intermediate layers of clay minerals which contributes to the electric, but not to the hydraulic conductivity.

But even if it is difficult to estimate a quantitative relation between water content and electric resistivity, it is still possible to state that in most in-situ systems higher resistivity is caused by less water in the formation. Since it is not possible at this time to determine a quantitative relation between the water content and the electric resistivity of clay, the objective of the laboratory measurements was to check qualitatively if the decrease in the water content in a clay sample can be correlated with a change in its electric resistivity.

For the determination of the resistivity as a function of the saturation the 4-point method was used. For the measurement of the average resistivity a sample with the cross area A was set between two metal plates (Figure 8). Between these two plates which serve as electrodes, a direct current I ran through the sample. A potential difference U was measured at the electrodes M and N which had been installed at the surface of the sample. From the current I , the voltage U , the cross area A , and the spacing L between M and N , the resistivity ρ of the sample can be calculated by

$$\rho = \frac{U \cdot A}{I \cdot L} \quad [1]$$

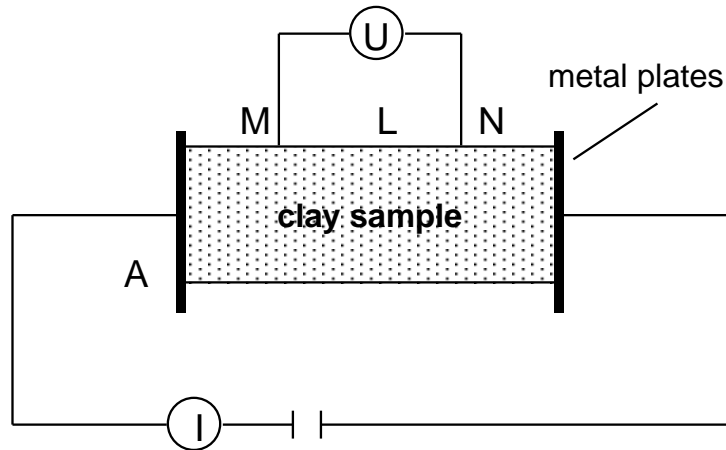


Figure 8 Schematic experiment layout for the determination of the average resistivity of a clay sample (4-point method)

First investigations were carried out on compacted MX-80 bentonite samples which were saturated with a solution being representative for ÄSPÖ granite formation water. The sample water content varied between 5 and 20 %.

Figure 9 shows the results of the measurement on the compacted MX 80 samples. The resistivity increases with decreasing solution content. The highest increase was determined at a solution content between 0 to 5 %. At higher solution contents of 10 % and 20 % the resistivity changed only in a small range. The resistivity was about 34 Ωm at the dry state and about 1.6 Ωm at a solution content of 20 %.

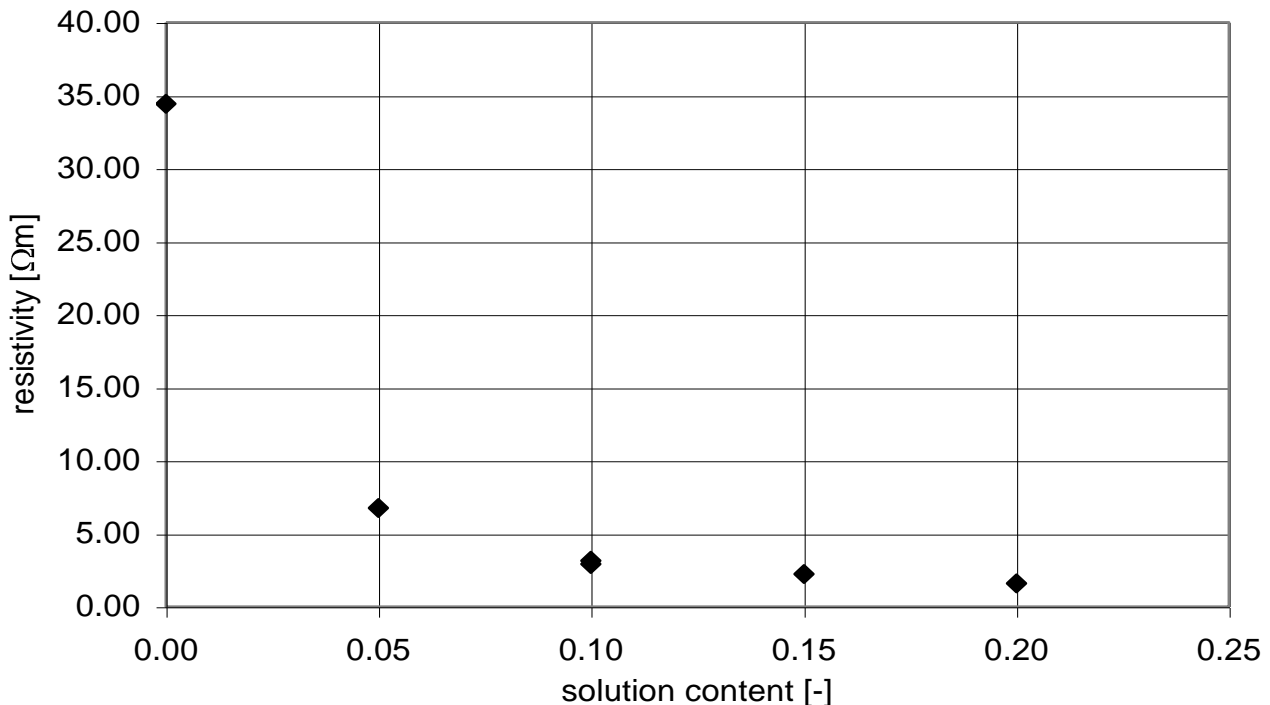


Figure 9 Resistivity-water content relation of compacted MX-80 samples saturated with a solution representative for ÄSPÖ formation water

3. GEOELECTRICAL IN-SITU INVESTIGATIONS OF CLAY

In the following sections, examples for the in-situ determination of saturation and desaturation processes in claystone and clay-bearing seal materials are presented.

3.1 Tournemire Test Site

For the monitoring of the potential desaturation process induced by ventilation, the electric resistivity around a 20 m deep continuously ventilated borehole was observed. The ventilation test is being performed by the Institute de Protection et de Sûreté Nucléaire (IPSN), the owner and operator of the Tournemire test site.

To allow the application of tomographic measurements, two boreholes, each being 0.6 m apart from the ventilated borehole, are necessary. The plane between these two boreholes includes the ventilated borehole (Figure 10). All three boreholes are 20 m deep. The electrode chains were installed in the lower 4 m of the electrode boreholes. Each chain consists of 21 single electrodes with 0.2 m spacing between them. For a single measurement 4 different electrodes are required.

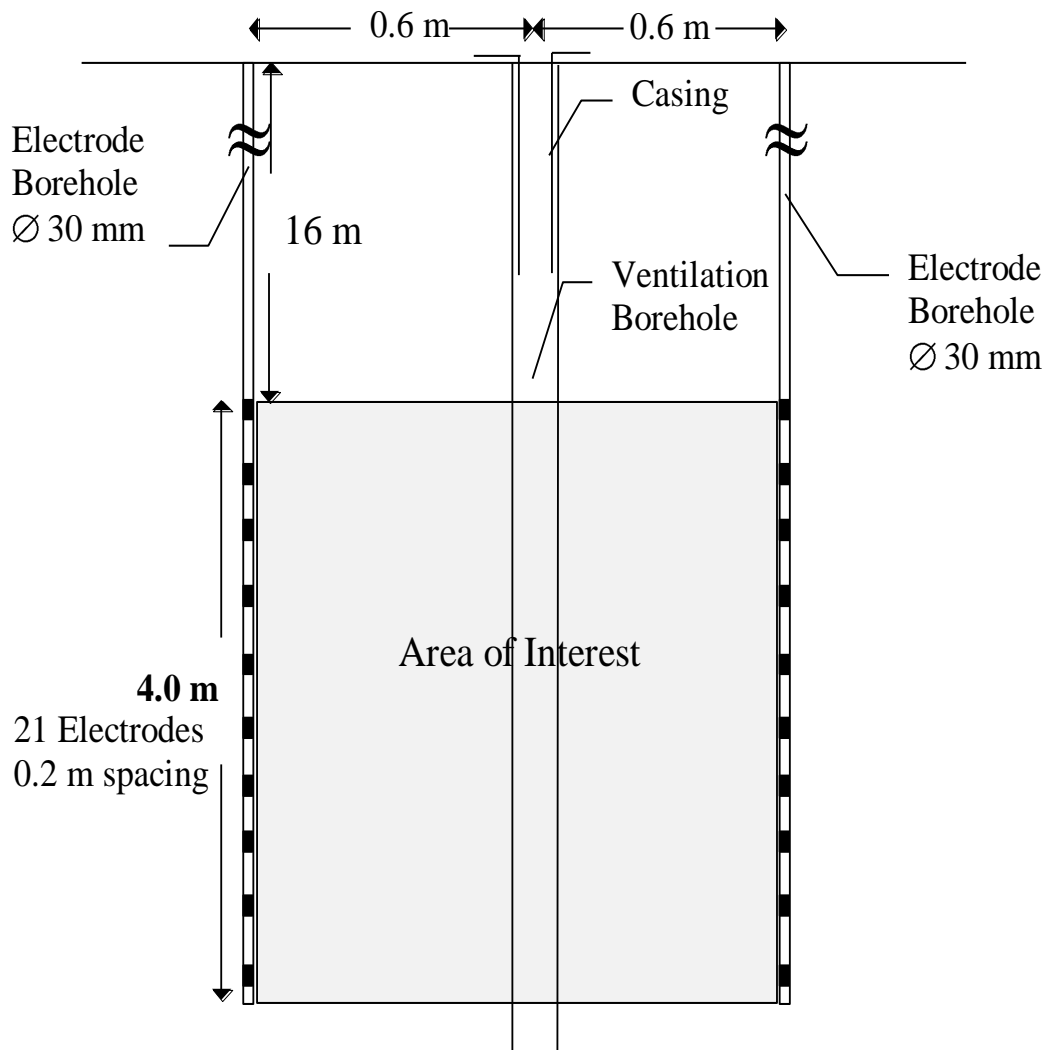


Figure 10 In-situ experiment layout at Tournemire (France)

The resistivity has been measured before and during the ventilation experiment; between August, 1998, and now. Until start-up of the ventilation on February 5, 1999, the changes in the resistivity distribution were not significant. The left hand side of figure 11 shows the result of a measurement taken in January, 1999 for an example. One can recognize a rather homogeneous resistivity around $50 \Omega\text{m}$ in the center of the model, while the resistivity along the electrodes which are grouted in the boreholes is slightly higher. The higher resistivity values at the upper and lower end of the model are boundary effects.

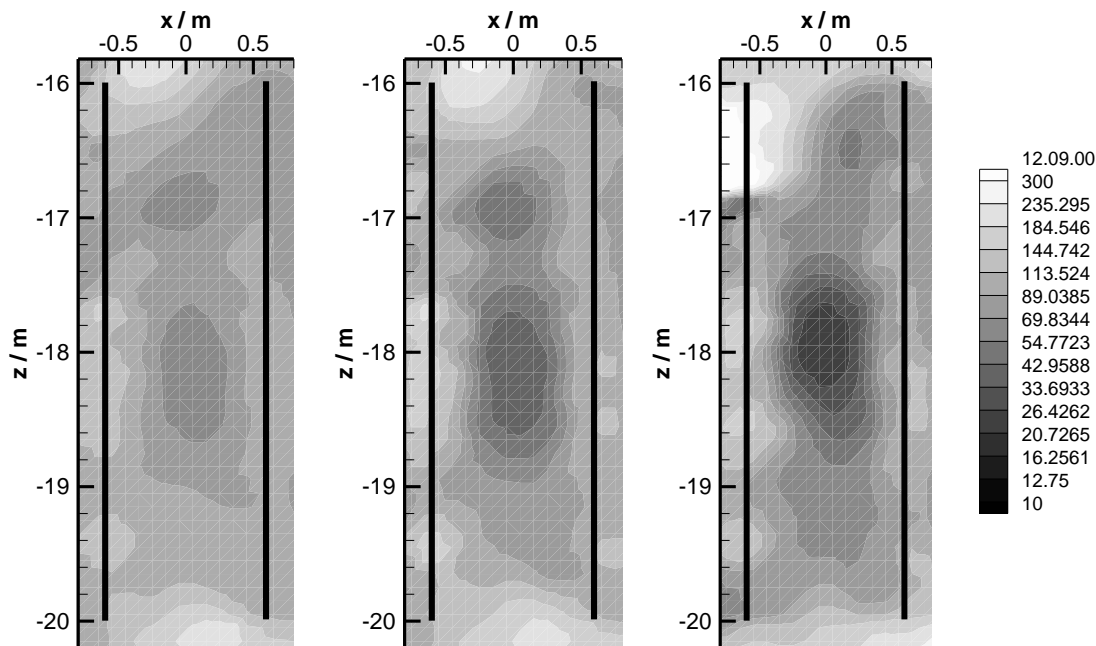


Figure 11 Resistivity distribution in the plane between the electrode boreholes at Tournemire (France), measured on January 27, 1999 (left), on April 19, 1999 (center), and on September 12, 2000 (right)

The tomograms in the center and on the right hand side of figure 11 show the resistivity distribution in April, 1999 (after 10 weeks of ventilation) and in September, 2000, respectively. The resistivity along the electrodes has partially increased, which may be due to further drying of the grout. This is particularly obvious at the upper left electrodes. A resistivity increase in the center of the model, however, which could be associated with drying of the clay, cannot be detected. This means that a significant desaturation of the clay has not taken place.

3.2 Mont Terri Test Site

Another test site where GRS employs geoelectrical monitoring to investigate desaturation effects in claystone is located in the Mont Terri underground laboratory. Here the Spanish Empresa Nacional de Residuos Radiactivos, S. A. (ENRESA) currently prepares a heater test to simulate the disposal of high level waste [5]. It is assumed that heating of the claystone formation will lead to a desaturation of the host rock. The measurements will be performed in the surroundings of a central borehole containing the heater (Figure 12).

The system installed consists of 4 electrode chains and an automatic multi-channel data acquisition system. The electrode chains are installed in four boreholes (BHE14 – BHE17). An electrode chain consists of 30 equidistant metallic spheres of about 2 cm diameter. The electrode spacing is 25 cm. Tomography between the boreholes as well as Wenner measurements along the electrode chains are performed.

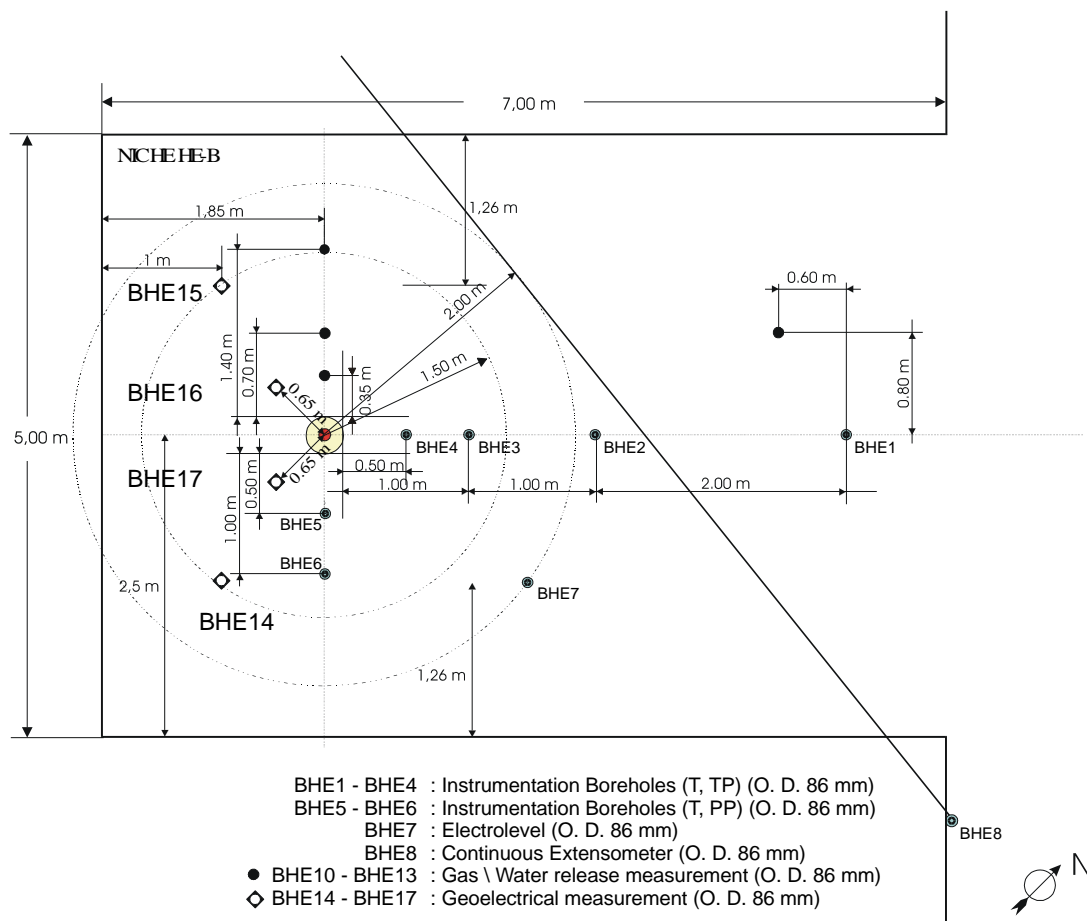


Figure 12 Plan view of the test field with the boreholes for gas release and geoelectric measurements (Mont Terri, St Ursanne/Switzerland)

The electrodes were installed in June, 1999. After a period of setting of the grout a stable resistivity distribution was obtained, as is shown for the section including the boreholes BHE15 and BHE16 in figure 13 on the left hand side. There is a high-resistivity anomaly in the upper left corner of the tomogram; this is, however, due to a bad coupling of the uppermost electrodes to the surrounding rock. The rest of the tomogram shows a rather homogeneous picture with a resistivity around 10 Ωm .

The right hand side of figure 13 shows the resistivity distribution one year later, in September, 2000. A drying effect can be seen over the whole tomogram, and the resistivity increased to values around 50 Ωm . Future measurements, which will be performed when heating is started, will show whether a more pronounced drying zone will develop near the heater.

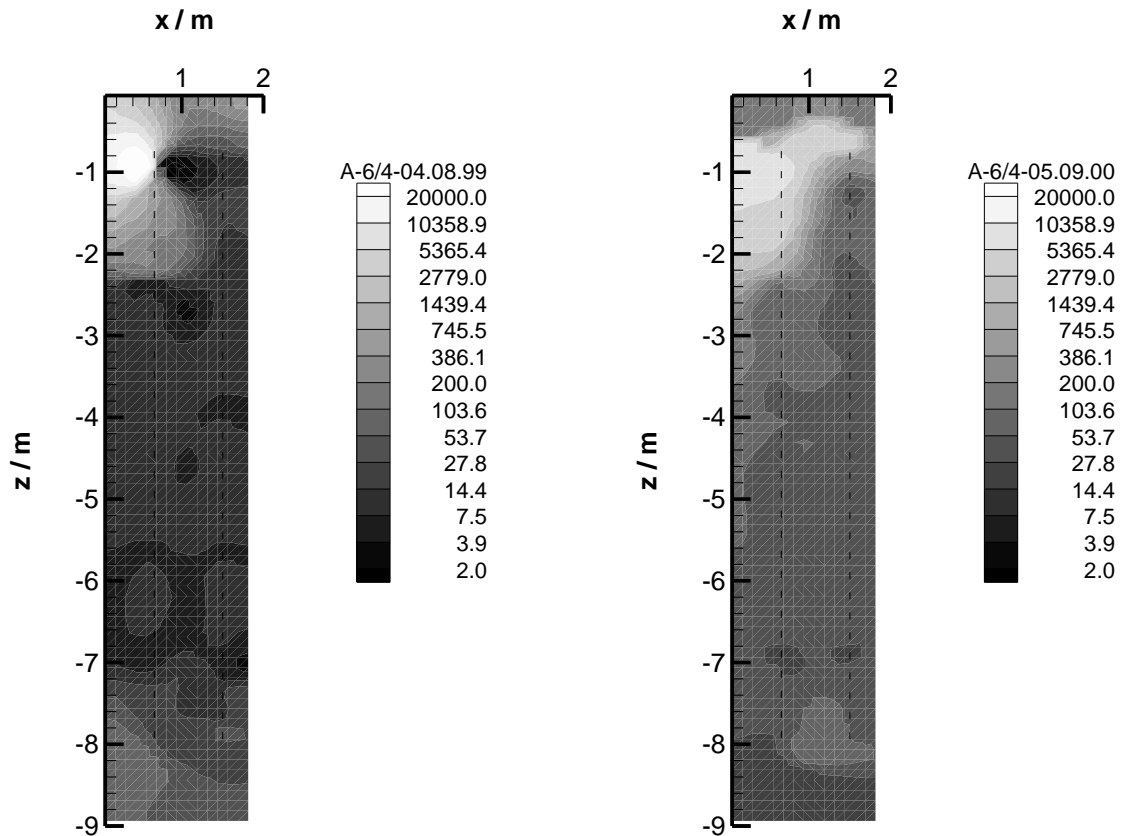


Figure 13: Resistivity distribution in the plane between the boreholes BHE17 and BHE16 of the Mont Terri Heater Test (St. Ursanne/Switzerland), measured on August 4, 1999 (left) and on September 5, 2000 (right)

3.3 ÄSPÖ Test Site: Prototype Repository

The Swedish Nuclear Fuel and Waste Management Co. (SKB) intends to perform a full scale spent fuel disposal test in a prototype repository at the ÄSPÖ Hard Rock Laboratory (HRL) in a granite formation in southern Sweden [6]. The underground test field in the granite formation consists of drift in which six deposition boreholes are located. The annulus in the boreholes surrounding the simulated spent fuel canisters will be backfilled with highly compacted MX-80 bentonite buffer blocks. The drift above the deposition boreholes will be backfilled with a bentonite-granite gravel mixture. In this test GRS will again perform measurements of electric resistivity to monitor water uptake in the borehole buffer and the drift backfill and desaturation effects around one of the deposition boreholes.

Three areas will be instrumented (Figure 14). Four measuring chains with eight electrodes each with a spacing of 0.2 m will be installed at the top of deposition borehole #5. The resistivity distribution in the buffer will be determined in the planes between the electrode chains. Two ring profiles with 36 single electrodes each with a spacing of 0.6 m will be installed on the surface of the tunnel sections I and II. These arrays will monitor the resistivity distribution in the backfill. The resistivity distribution in the immediate vicinity of the boreholes will be monitored with three electrode chains in vertical boreholes between deposition boreholes #5 and #6.

The installation of measuring equipment will begin in the summer of 2001. In preparation of the in-situ measurements, laboratory calibration measurements of the dependence on electrical resistivity on the water content of the buffer and the backfill are currently underway at the GRS geotechnical laboratory in Braunschweig (compare chapter 2.2).

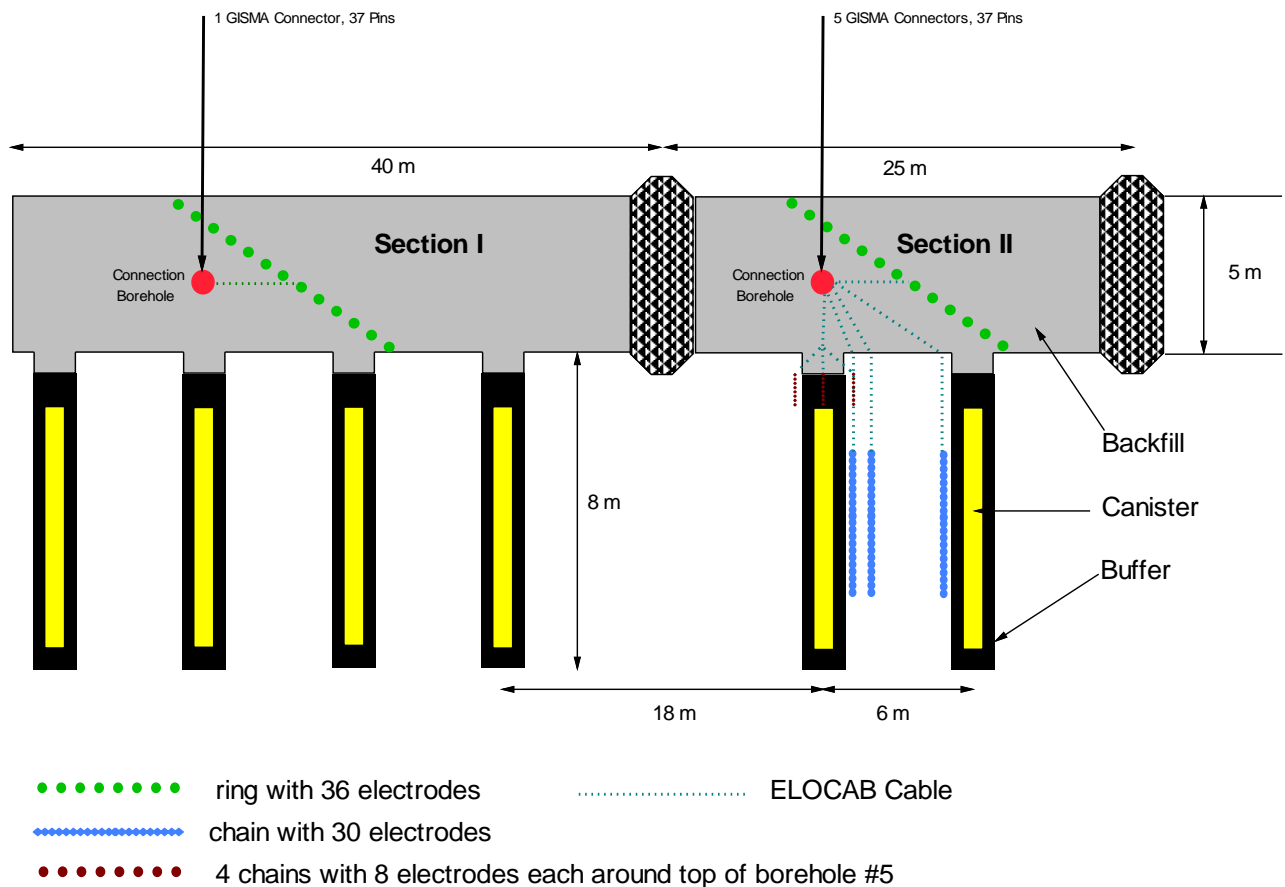


Figure 14 Planned arrangement of electrodes (ÄSPÖ/Sweden)

4. SUMMARY

Sand-clay mixtures are found suitable for backfilling and sealing of repository sections where instantaneous sealing is required in the case of a sudden water inflow.

The dry material has a permeability to gas in the range between 10^{-14} to 10^{-13} m^2 , not depending significantly on the sand-clay ratio, the preparation, or the gas pressure.

The single phase water permeability, however, is significantly dependent on the sand-clay ratio. Mixtures with a clay content of 10 % have a water permeability in the range of 10^{-15} to 10^{-16} m^2 , whereas mixtures with a clay content of 50 % are almost impermeable to water. Above 25 % of clay content, the entire porosity of 20 to 22 % is consumed by swelling, and any advective flow is impossible.

The investigation of two-phase flow indicated that sand-clay mixtures have an almost uniform pore size distribution. The two-phase flow parameters such as relative permeability and capillary pressure can be better described by the relation of van Genuchten than by the relation of Brooks-Corey. The investigations reveal that it should be possible to design optimum sand-clay seals with defined permeabilities to gas and water by choosing specific material ratios. In contrast, water and two-phase permeabilities could be determined neither for plastic Boom clay nor for Opalinus claystone samples.

For the in-situ determination of saturation and desaturation processes in claystone geoelectric measurements were performed at Mont Terri. The results obtained so far reveal that drying effects induced by evaporation can be detected with sufficient success.

Investigations of pure clay seal materials like MX-80 bentonite are planned at ÄSPÖ. Preceding measurements of the average resistivity in compacted MX-80 samples with defined saturation showed that the resistivity increases with decreasing solution content. The highest increase was observed at lower solution content.

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