**Gesellschaft für Anlagen-
und Reaktorsicherheit
(GRS) mbH**

Heater Test in the Opalinus Clay of the Mont Terri URL

Gas Release and Water Redistribution

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH

Heater Test in the Opalinus Clay of the Mont Terri URL

Gas Release and Water Redistribution

Contribution to Heater Experiment (HE); Rock and bentonite thermo-hydro-mechanical (THM) processes in the nearfield

Norbert Jockwer
Klaus Wieczorek

with contribution of
Rüdiger Miehe
Ana Maria Fernández Diaz
(CIEMAT)

June 2006

Preface

Beside salt and granite, clay formations are investigated as potential host rocks for disposing radioactive waste. In Switzerland in the canton Jura close to the city of St. Ursanne, an underground laboratory was built in the vicinity of the reconnaissance gallery of a motorway tunnel. Since 1995, a consortium of 12 international organisations is running this laboratory for investigating the suitability of the Opalinus clay formation with regard to disposal of radioactive waste.

In 1999, the Heater Experiment B (HE-B) was started for investigating the coupled thermo-hydro-mechanical (THM) processes of the Opalinus clay in interaction with the bentonite buffer. The principal contractors of this project were the Bundesanstalt für Geowissenschaften und Rohstoffe (BGR), the Empresa Nacional de Residuos Radiactivos S. A. (ENRESA), the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, and the National Cooperative for the Disposal of Radioactive Waste (NAGRA).

GRS participated in that experiment for determining the subjects of gas generation, gas release, water content, and water redistribution in the Opalinus clay during heating. This was achieved by analysing gas and water samples from the test field before, during, and after the heating period and by performing geoelectric tomography measurements in the heated region. The in-situ measurements were supported by an additional laboratory programme.

This report deals with the work of GRS performed in this project during the years 1999 to 2005. All the results obtained in the frame of the project are presented. Additional laboratory measurements conducted by the Pore Water Laboratoy at CIEMAT in Madrid are also presented.

Table of Contents

1 Introduction

The intention pursued by the various European concepts for the disposal of radioactive waste is to dispose of radioactive waste in deep geological formations in order to isolate them from the biosphere and avoid the release of radionuclides above an acceptable limit. Besides salt and granite, clay formations are investigated.

The suitability of the Opalinus clay formation for disposing radioactive waste has been under investigation for more than ten years. In the Swiss canton Jura close to the city of Saint Ursanne, a motorway tunnel crosses the Mont Terri. Parallel to this tunnel a reconnaissance gallery with an additional underground laboratory was built. A consortium of 12 international organisations is running this laboratory, performing experiments with regard to the disposal of radioactive waste /THU 99/, /PEA 03/.

In 1999, the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) joined the consortium and participated in the Heater Experiment B (HE-B), investigating the special subjects of gas generation, gas release, water content and water redistribution in the Opalinus clay during heating.

The basic objectives of the HE-B project are /EMM 02/:

- Observation of the coupled THM processes in the near field (bentonite buffer and host rock)

- Study of the behaviour and the reliability of instrumentation and measuring techniques applicable to the type of rock formation

- Determination of the general properties of the host rock

- In-situ determination of the mechanical state of the host rock and changes induced by the experiment

- Study of the rock/bentonite interaction

- Study of the thermo-hydro-geochemical processes in the near field (bentonite buffer and host rock)

- Analysis of gases and water released from the rock by effects of heating

- Validation/refining of existing tools for modelling THM processes

The project was supported by the European Commission under the contract No. FIKW.CT-2001-00132 within the framework of the research and training programme in the field of nuclear energy (Euratom). The principal contractors were BGR (Germany), GRS (Germany), ENRESA (Spain), and NAGRA (Switzerland). Subcontractors were AITEMIN (Spain), CIMNE (Spain), ETHZ (Switzerland), COLENCO (Switzerland), and RL (Switzerland). Project coordination was under the responsibility of BGR.

This report presents the investigation programme and results of GRS, which comprise

- Gas generation and release in the test field as a result of elevated temperature

- Water content of the Opalinus clay

- Water resistribution in the Opalinus clay as a result of heating

The work was performed in the years 1999 to 2005.

2 General Information on the HE-B Test

The HE-B test was carried out in a specially excavated niche located in the shaly facies of the new gallery (Figure 2-1) /EMM 02/. The orientation of that niche is almost parallel to the striking with a dip of the bedding of approximately 40°. The niche with the dimensions of about 5 m width by 7 m length was excavated in March 1998. Drilling and installation of all the boreholes were performed in 1999. A plan view of the niche with the boreholes is shown in Figure 2-2.
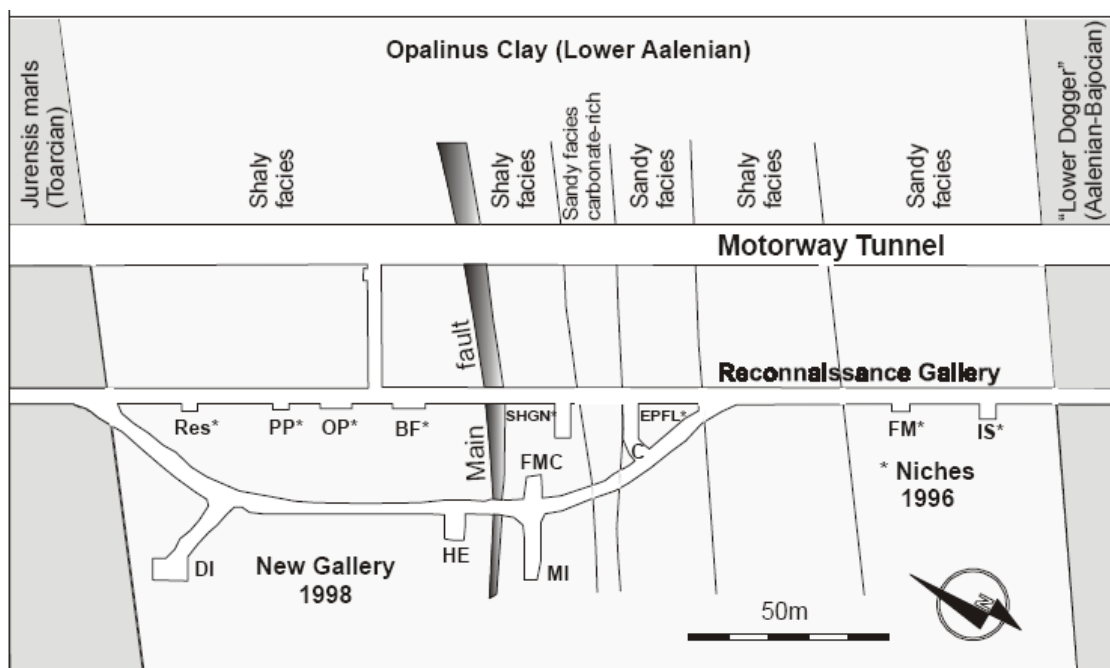

Figure 2-1   Plan view of the Mont Terri Underground Laboratory with the HE-B niche /EMM 02/
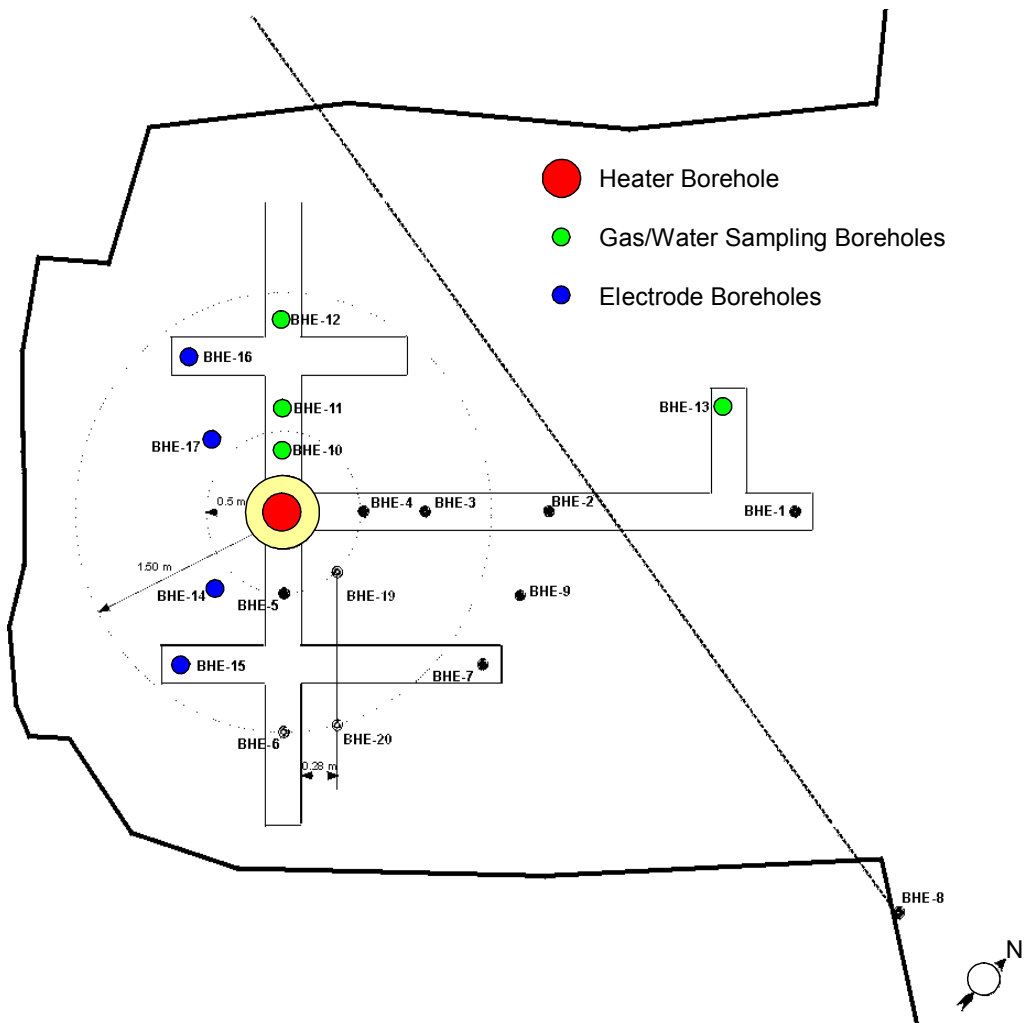
Figure 2-2 Plan view of the HE-B niche with the instrumentation boreholes

The central vertical heater borehole (BHE-0) with a diameter of 300 mm and a depth of 7.5 m was drilled into the floor of the niche. The arrangement of the different components in this borehole is shown in Figure 2-3. A heater tube was installed in the centre surrounded by a cylindrical ceramic filter and ring shaped bentonite buffer blocks (engineered barrier) with a dry density of 1800 kg/m³. For resaturating the bentonite with water, four capillaries run from the open niche into the ceramic filter. Sensors for determining temperature, total pressure, pore pressure and humidity were installed in the bentonite blocks. The gap between the bentonite blocks and the host rock as well as the upper and bottom part of the borehole were filled with sand. Inside the heater tube, the removable heater was installed with its top at 4 m below the niche floor. The borehole was sealed gastight with a plug of resin and cement and was covered with a lid anchored to the floor as shown in Figure 2-4, in order to withstand the thrust generated by the swelling pressure of the bentonite.

4

Figure 2-3   Cross section of the central borehole BHE-0 with the electrical heater and the buffer materials
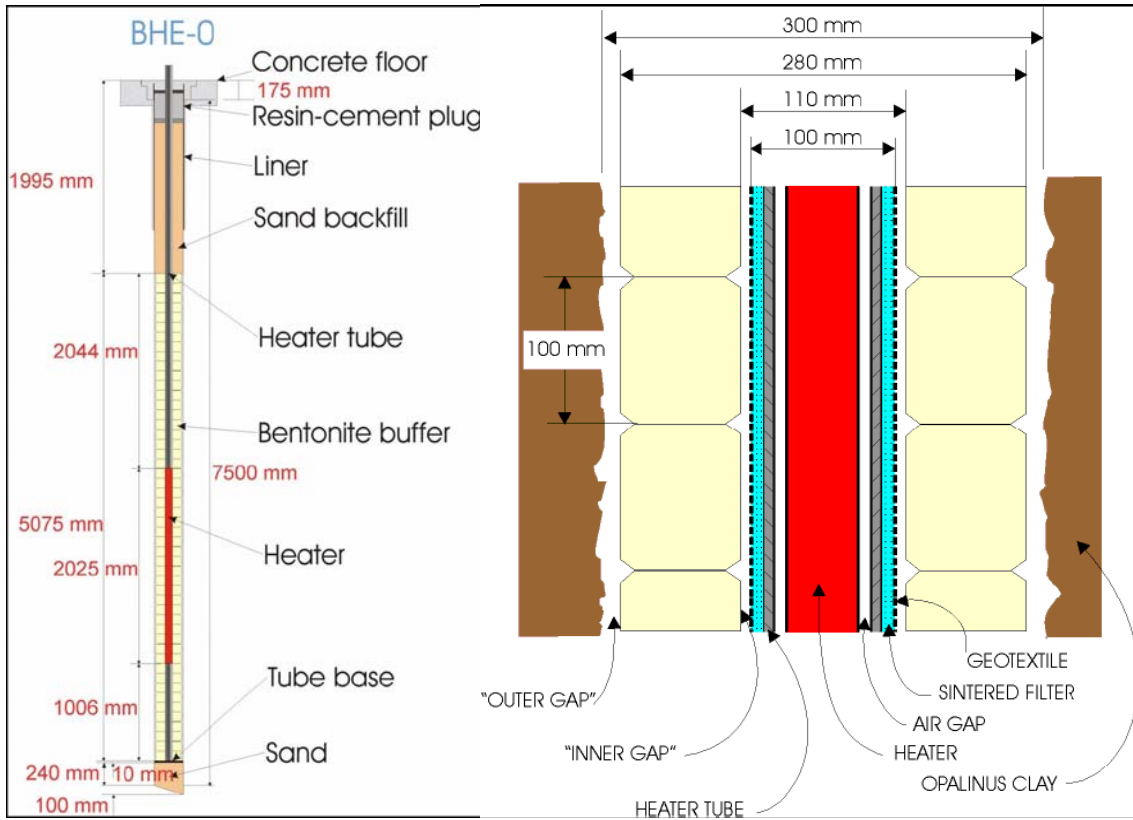

Figure 2-4   Heater borehole BHE-0 with the steel lid anchored to the rock

Additionally, a total of 19 boreholes were drilled into the niche floor for determining the relevant parameters of the host rock, such as temperature, total pressure, pore pressure, permeability to water, water content, electric resistivity, and gas release.

Table 2-1 shows the chronology of the experiment with its work phases. Due to the reduced amount of water provided by the host rock, the bentonite buffer was artificially hydrated to reach water saturation as soon as possible. For this hydration synthetic pore water (Pearson water type A1) was used, which is chemically similar to the water in the Opalinus clay formation. Hydration started in October 1999, but it caused a general problem to the whole experiment as water ran into the interior of the heating tube though an untight joint. This led to a total failure of the heater. A second tube and new designed heater had to be installed. Therefore, heating started only on February 22, 2002. At the beginning, the power was set to 140 Watts and increased to 650 watts within the first two months. Afterwards, the heating power was in the range between 550 to 650 Watts, which led to a maximum temperature of 68 °C at the bentonite blocks. Heating lasted for 18 months until September 2003. Afterwards, cores were drilled from the host rock and the whole test field was dismantled for further laboratory investigations /GOE 04/.

Table 2-1   Chronology of the HE-B test

Phase	Date
Installation of experiment	May - June 1999
Hydration of the bentonite buffer	May 1999 - March 2002
Heating	February 22, 2002 - September 01, 2003
Cooling	September 2003
Drilling of cores	October 2003
Dismantling of the test	October to December 2003

3 Field Installation for Gas Release and Water Redistribution Measurements

Four boreholes (BHE-10 – BHE-13, see Figure 2-2) were equipped with packers to enable gas and pore-water sampling. Another four boreholes (BHE-14 – BHE-17, see Figure 2-2) were used to install electrode chains for geoelectric measurements, which were applied to determine pore-water redistribution.

3.1 Gas and water sampling boreholes

Water and gases such as carbon dioxide, hydro carbons, hydrogen and hydrogen sulphide may be released out of the Opalinus clay at ambient and elevated temperature. In order to determine these components qualitatively and quantitatively, 4 boreholes were drilled into the test field with different distances between 0.5 and 3.64 m to the central borehole (see Figure 2-2). These boreholes are identified BHE-10 to BHE-13. They have a diameter of 86 mm and a depth of 7.00 m each and they were sealed with special packers for taking gas and water samples which are not influenced by the mine air.

These boreholes were drilled May 1999. Installation of the packers with three stainless steel sampling tubes was performed right after drilling. For expanding of the packers a rubber hose was connected to the inflation chamber via a quick connection and a one-way valve. After the packer was let down to the bottom of the borehole, it was raised again by 10 cm and then inflated with water to 10 bar using a manual pump. Afterwards, the rubber hose fixed to the packer by the quick connection was disconnected. The remaining borehole above the packer was then backfilled in four steps:

1. 20 cm of Sikadur 52 (special resin) were pumped in by a rotary pump. After about one hour to let the resin harden backfilling was continued.

2. 50 cm of Opalinus clay powder from drilling were filled in via a rubber hose, then an electronic humidity sensor from ANDRA with an electric wire was let down to the top of the backfill and afterwards another 50 cm of Opalinus clay powder was injected.

3. 20 cm of Sicadur (special resin) were pumped in. After about one hour to let the resin harden backfilling was continued.

4. The remaining borehole was backfilled up to the gallery floor with the special plastic concrete of cement, Opalinus clay powder and water. This concrete was designed by the Spanish engineering company AITEMIN.

The packers installed in the boreholes are shown in Figure 3-1.
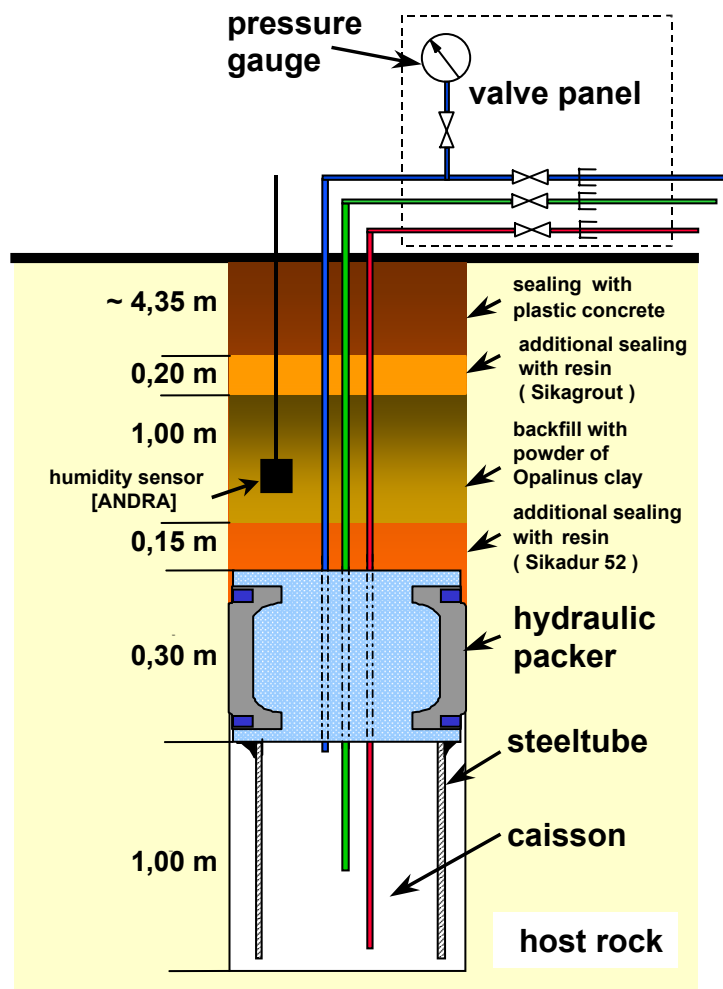
Figure 3-1   Packer with caisson for taking gas and water samples installed in the boreholes BHE-10 to BHE-13

    - top sampling tube ending 2 cm below the packer body (caisson top)

    - mid sampling tube ending in the centre of the caisson

    - bottom sampling tube ending 5 cm above the bottom of the caisson

The three stainless steel tubes from each packer run via cable channels to a valve panel fixed to the niche wall. The valve panel consists of three horizontal aluminium

ledges on which the valves, pressure gauges, and quick connections are mounted as shown in Figure 3-2.

Figure 3-2   Valve panel with Swagelok valves, pressure gauges and
                   quick connections

The sampling tubes are stainless steel type Wn.1.4571/DIN 2461/N, seamless 6/1, inner diameter 4 mm, outer diameter 6 mm.

After installation of the valve panel all valves were closed. The atmosphere in the sealed borehole volume was not exchanged, that means, it was f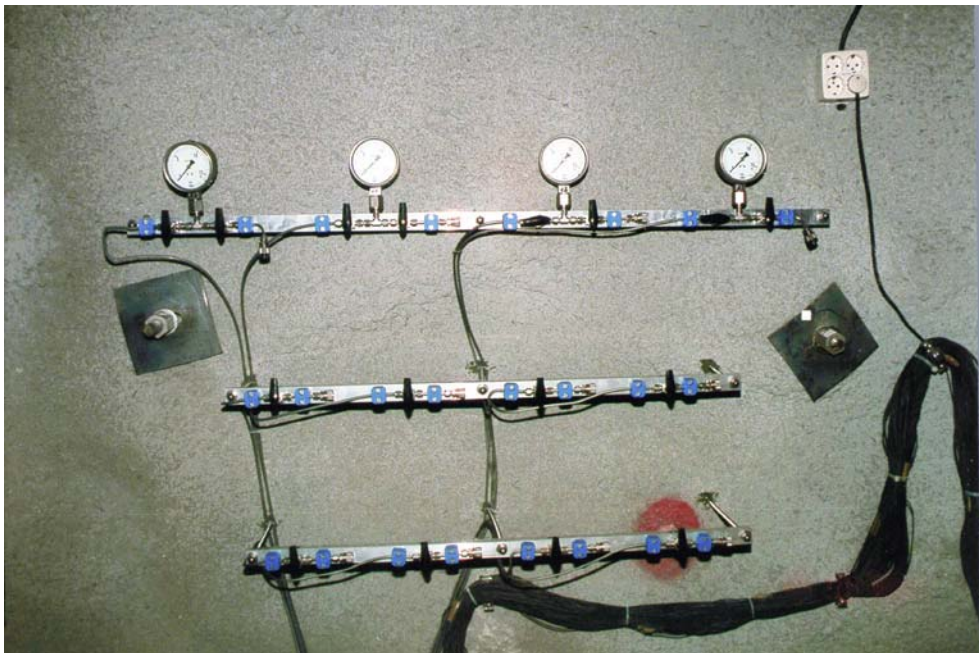illed with mine air and the gases released from the surrounding host rock. The aim of the investigation was to determine the released components at the physico-chemical conditions (temperature, pore pressure, humidity, ...) of the test field.

The installation of the packers and the valve panel is described in detail in /JOC 01-1/.

3.2   Boreholes for installation of the geoelectric array

The boreholes BHE-14 to BHE-17 with a diameter of 86 mm and a depth of 8.0 m each were drilled on May 1999. Immediately after drilling the boreholes, the electrode chains, each consisting of 33 electrodes with a spacing of 25 cm, were emplaced and then grouted with a special plastic concrete (see Figure 3-3).

The geoelectric monitoring system was installed in the test field on June, 1999. It consisted of an automatic computer-controlled measurement and data acquisition unit with multiplexers for adressing the electrodes and an external injection voltage supply. The geoelectric system was placed on a table next to the gas sampling rack. Mains, telephone line, and electrode connectors were plugged. The installation of the electrode chains and the geoelectric system is described in detail in in /JOC 01-2/.

Depth / m		Electrode No.
0.0	•	x033
0.25	•	x032
0.5	•	x031
0.75	•	x030
1.0	•	x029
1.25	•	x028
1.5	•	x027
1.75	•	x026
2.0	•	x025
2.25	•	x024
2.5	•	x023
2.75	•	x022
3.0	•	x021
3.25	•	x020
3.5	•	x019
3.75	•	x018
4.0	•	x017
4.25	•	x016
4.5	•	x015
4.75	•	x014
5.0	•	x013
5.25	•	x012
5.5	•	x011
5.75	•	x010
6.0	•	x009
6.25	•	x008
6.5	•	x007
6.75	•	x006
7.0	•	x005
7.25	•	x004
7.5	•	x003
7.75	•	x002
8.0	•	x001

Figure 3-3   Electrode locations in the boreholes BHE-14, BHE-15, BHE-16, and BHE-17 (x is 4, 5, 6, 7)

4   Methods of Investigation

The measurements performed in situ are explained in Section 4.1. They were complemented by additional laboratory experiments described in Section 4.2.

4.1   In-situ measurements

4.1.1   Gas and water sampling

Gas sampling from the sealed boreholes BHE-10 to BHE-13 was combined with the determination of the relative humidity and the temperature of the atmosphere which was extracted out of the boreholes and transferred into special gas bags /JOC 02/. For this procedure the following components were used:

- Humidity measuring instrument type TESTO 650 with sensor which is installed in a glass cell

- Manual pump as shown in Figure 4-1

- Gas tank with nitrogen

- Linde gas sampling bags of 1 liter with valve.

This equipment was arranged as shown in Figure 4-2 and connected to the tube at the valve panel out of which the gas sample was taken. After opening the valve of the sampling tube the pressure of the borehole atmosphere indicated at the manual pump was determined. Then the system was purged with one pump volume (half a litre) and this gas was discharged. Then about one litre of borehole atmosphere was pumped into the sampling bag and again the pressure of the borehole atmosphere indicated at the manual pump was determined. Afterwards nitrogen was injected into the borehole up to the pressure before sampling. The gas sampling bags with the valve closed gastight was sent to GRS Braunschweig for analysis.

Gas sampling was performed as described in "GRS Qualitätsmanagement, Prüf-anweisung Gasprobenahme und Feuchtigkeitsmessung in den Bohrlöchern BHE-10, BHE-11, BHE-12 und BHE-13 im HE-Versuchsfeld", dated September 25, 1999.
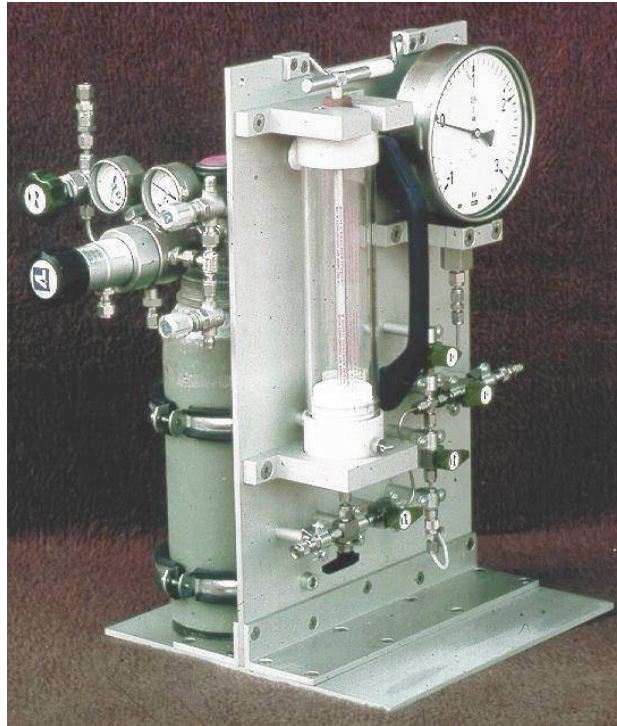
Figure 4-1 Manual pump with pressure gauge for gas sampling
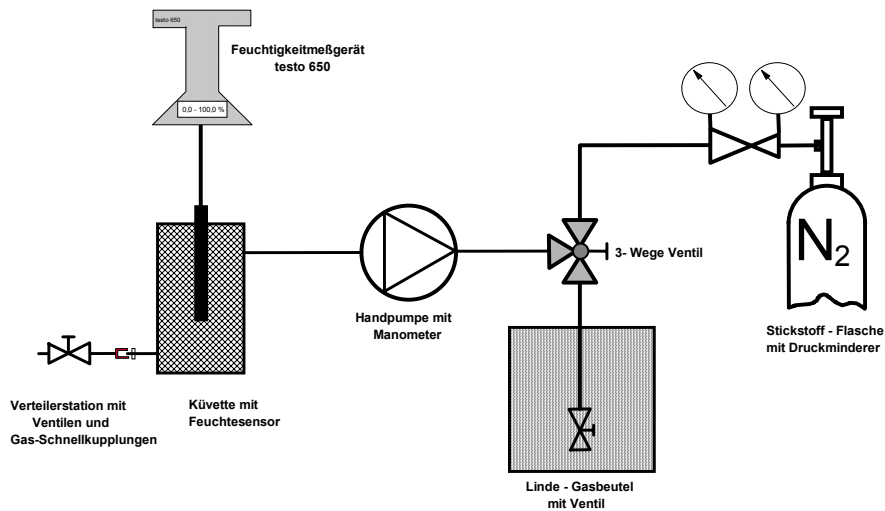
Figure 4-2 Arrangement of the equipment for gas sampling and humidity determination

In July 2003, a portable gas chromatograph was installed in the test field for on-site analyses. The advantage of that method was that analyses were performed immediately after extraction, and only a small amount of gas was needed. The influence of sampling was therefore minimized. For comparison, samples were taken for analysis in the GRS laboratory in Braunschweig.

The data of the portable gas chromatograph system are summarized in Table 4-1. Figure 4-3 shows the gas chromatograph in the HE-B test field in operation.

Table 4-1   Data of the gas chromatograph system

Type	VATIAN CP4900
Length/ width/height	65/30/40 cm
Weight	25 kg
Separation capillary columns	4
Thermal conductivity conductors	4
Carrier gas	helium and argon
Power supply	220 V AC
Control	Laptop computer

Figure 4-3   Gas chromatograph in operation in the HE-B test field

After gas sampling in August and in October 2003, the water in the boreholes was extracted by inflating the residual volume of the sealed borehole with nitrogen to 1 bar via the capillary ending at the top of the caisson. At the valve panel, a Linde sampling

bag was connected to the capillary ending at the bottom of the caisson. After opening the valve of that capillary, the water from the bottom of the borehole ran into the sampling bag without any contact to the mine atmosphere. The amounts of the water extracted from the boreholes were determined by weighing the bags. For chemical analysis the water samples were sent to CIEMAT in Madrid.

At the CIEMAT laboratory (Pore Water Laboratory), the bags were placed into an anoxic glove box for measuring pH, Eh, and electrical conductivity. Other water aliquots were acidified, depending on the type of chemical analysis required, and then the chemistry of the water was analysed outside the anoxic chamber. The total alkalinity of the samples (expressed as mg/l of HCO_3^-) was determined by potentiometric titration using a Metrohm 682 titrator. The major cations were analysed by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) in a Perkin-Elmer Elan 5000 spectrometer. Sodium and potassium were determined by flame atomic emission spectrometry, AAS-Flame, in a Perkin Elmer 2280 spectrometer, and trace elements were determined by ICP-MS (Finningan Mat SOLA). Anions were analysed by ion chromatography (Dionex DX-4500i). The silica was determined using a UV-Vis spectrophotometer by means of the silico-molybdic colourimetric method. It was estimated that the maximum analytical errors were ±10% for the major ions.

The aim of that investigation was to determine if the water extracted in the test field was formation water or the water injected into the central borehole to saturate the bentonite buffer, giving an information about the permeability to water of the Opalinus clay. Furthermore the idea was to determine whether the formation water from the Opalinus clay changed its composition as a result of heating.

4.1.2 Geoelectric measurements

Making use of the dependence of electric resistivity on the water content of rocks, potential desaturation of the rock caused by heating can be monitored by geoelectric measurements. In order to determine the resisitivity distribution in the area of interest, electrodes have to be installed. By injecting a current into the rock via two electrodes and measuring the resulting potential difference between other electrodes, an apparent resistivity value is obtained. Varying both the injection and the potential dipoles, a high number of single measurements can be performed and the resistivity distribution can be calculated from the apparent resistivity values.

In the frame of the Mont Terri Heater Experiment, geoelectric measurements were performed in order to monitor possible desaturation of the Opalinus clay due to heating of the rock. An issue of heating was that an elevated temperature of the clay and thus of the pore liquid leads to a decrease of resistivity, while desaturation leads to an increase. Therefore, calibration measurements on clay samples at different temperatures were performed in the laboratory.

The geoelectric measurements were performed as dipole-dipole measurements. Two electrodes were used for injecting a low-frequency alternating current into the formation, while the resulting potential difference between pairs of other electrodes was measured, giving an apparent resistivity for each single measurement. The injection and measurement dipoles were located in the same or in different boreholes. By varying both the injection dipole and the measurement dipole, a total of 2548 single measurements made up one dataset. One complete measurement was performed each day and took about 165 minutes.

The resulting data of each pair of boreholes were used as input for inverse finite element modelling. From the vector of apparent resistivities the resistivity distribution in the plane of the two considered boreholes was calculated as best fit between measured data and calculated response. This is why this measurement method is called geoelectric tomography.

Changes in water content due to desaturation were expected to be detectable. Resisitivity changes, however, could also be due to temperature changes, since the test field was heated. Therefore, laboratory calibrations were necessary in order to quantify the effects of temperature and of potential desaturation on the clay resistivity.

4.2 Additional laboratory programme

In the GRS laboratories in Braunschweig measurements on Opalinus clay samples on the gas generation and release, the mineralogy, and the electric resistivity were performed at defined physico-chemical conditions. Investigations were performed on samples taken prior to heating as well as after the heating phase. The results of the laboratory measurements are essential for interpretation of the behaviour of the gases in the test field, for calibration of the electric tomography, and for determination of the changes that occurred in the clay during heatin with regard to mineralogy and saturation.

4.2.1 Gas generation and release

For determination of the gas generation in the Opalinus clay and the release of the generated gases into the residual volume at different conditions, stainless steel containers and glass ampoules were used.

For determining the concentration of equilibrium of the released gases with the host rock, five almost identical Opalinus clay core samples of 3.2 kg were emplaced in five steel containers of 2.2 litres. The volume of the cores filled up almost the volume of the container which means that the relation between the amount of the sample and the residual volume into which the gases could be released is comparatively high. The five steel containers were sealed gastight and stored in an oven for 1, 3, 10, 30, and 100 days respectively at a temperature of 95 °C. After the envisaged time one container was taken out of the oven and via a septum at the lid the gas in the residual volume was extracted by a syringe and injected into the gas chromatograph for analyses. The results show the velocity of gas release and the concentration of equilibrium.

For determining the gas release from the Opalinus clay as a function of exposure time and gas in the residual volume of the reaction vessel, glass ampoules with a volume of 500 ml as shown in Figure 4-4 were used. Through the injection tube at the top of the ampoule 1 to 550 grams of ground material was filled in and additionally the atmosphere in the residual volume could be change by evacuation of the air and flooding with another gas of interest. Afterwards the ampoule was sealed gastight by welding the injection tube and etching an identification number on the outer surface. The expose periods in the over at 95 °C varied between 1 and 842 days.
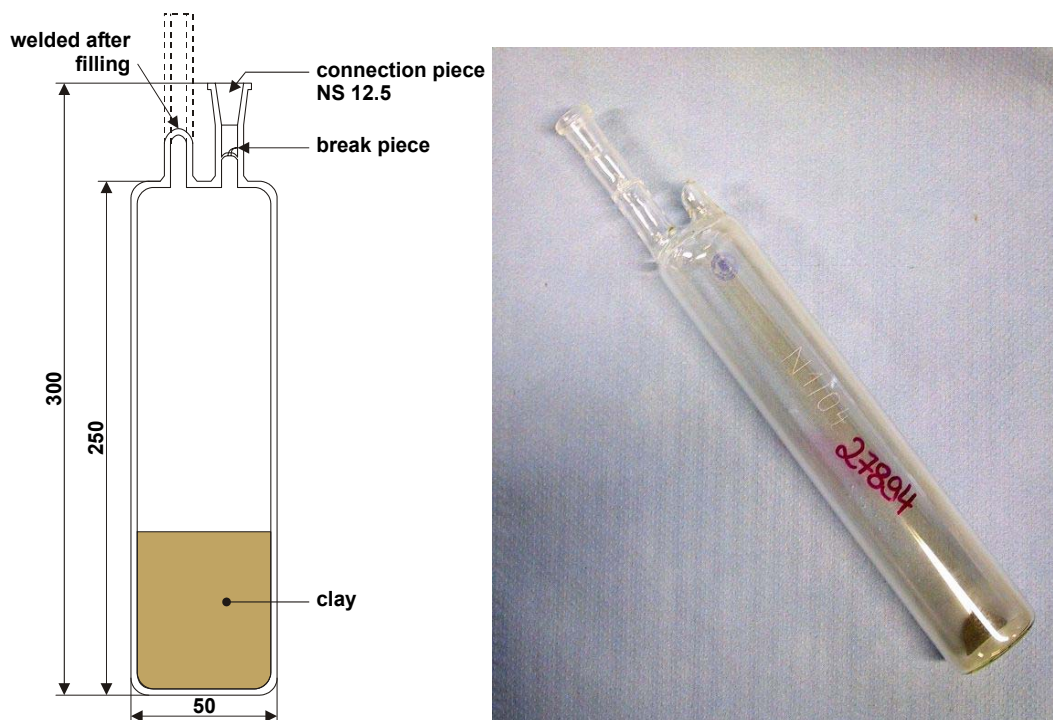
Figure 4-4   Ampoule for investigating the gas generation in the clay and the release of the gases into the residual volume

After storage at 95 °C for the envisaged time, the ampoules were withdrawn from the oven for analysis of the generated gases, which had been released into the residual volume. Each ampoule was connected to a pump stand consisting of a glass transfer tube with connectors to which two glass bulbs with a defined volume was attached, as shown in Figure 4-5. Each bulb was equipped with a valve and a septum. Gas lines were connected to the end of the transfer tube valves, one for evacuating the whole system and the other for purging it with nitrogen. The entire system was evacuated to an absolute pressure of about 100 Pa by an electric pump, then it was refilled with nitrogen to atmospheric pressure and evacuated again. This procedure was repeated three times, and at last the system was evacuated.

Afterwards, the break seal on the ampoule was opened by dropping a small Teflon®-coated magnet onto it; this allowed the gas in the residual volume of the ampoule to enter into the void volume of the transfer tube and into the two connected bulbs. Then nitrogen was added through the purge line to adjust the pressure throughout the system to 0.2 MPa absolute. The valves of the glass bulbs was closed and then disconnected from the transfer tube. The gas in the bulbs was extracted with a gas syringe through a septum and was injected into a gas chromatograph (GC) for

analysis. The pressure values after evacuation, after opening the ampoules, and after adding the nitrogen were recorded for the evaluation of the released gas amount.
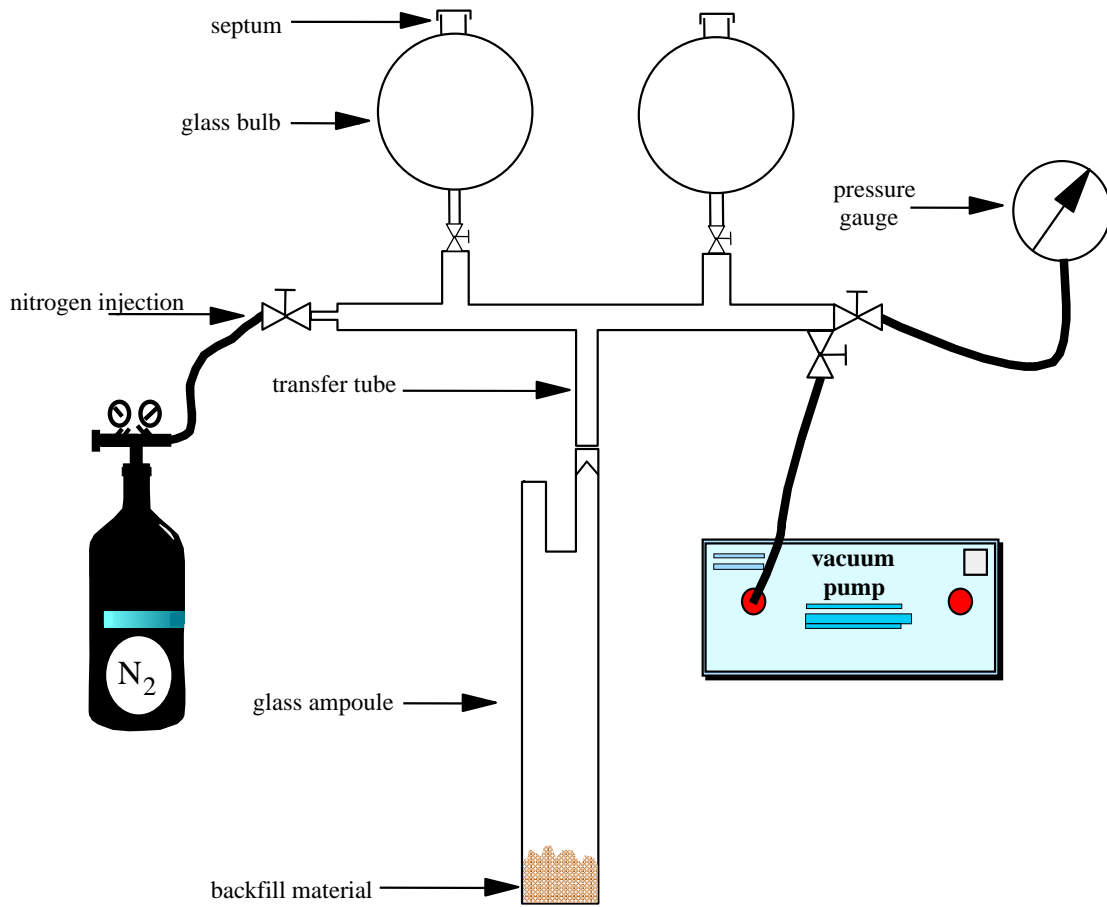

Figure 4-5   Pump stand with transfer tube and glass bulbs for extracting the generated gases from the attached ampoules

The analysis of the gas which was injected in the gas chromatograph delivers the concentration in the whole system consisting of the residual volume in the ampoule, the volume of the transfer tube and the volume of the two bulbs. The total volume of the system out of which the gas was extracted for analysis is:

$$V_{sys} = V_{resamp} + V_{trans} + 2 \cdot V_{bulb} \qquad [m^3]$$

and

$$V_{resamp} = V_{amp} - V_{ben} \qquad [m^3]$$

$$V_{ben} = m_{ben} \cdot \rho_{ben} \qquad [m^3]$$

with

V_{sys}	volume of the whole system	[m³]
V_{resamp}	residual volume of the ampoule	[m³]
V_{trans}	volume of the transfer tube	[m³]
V_{bulb}	volume of the bulb	[m³]
V_{amp}	total volume of the empty ampoule	[m³]
V_{ben}	volume of the Opalinus clay in the ampoule	[m³]
ρ_{ben}	density of the Opalinus clay	[kg/m³]
m_{ben}	mass of Opalinus clay filled into the ampoule	[kg]

The results of the analysis are the concentrations c_i of the component i. It is quoted in vpm which is 1 ml gas of the component i in 1 m³ matrix gas or 10^{-6} m³/m³. The total amount of the component i in the volume of the system is:

$$V_{isys} = c_i \cdot V_{sys} \cdot \frac{p}{p_0} \qquad [\text{m}^3]$$

with

V_{isys}	normal volume of the component i in the system	[m³]
c_i	concentration of the component i in the system	[vpm]
p_0	atmospheric pressure	[MPa]
p	pressure in the system before extracting the gas	[MPa]

As the air which was originally in the residual volume of the ampoule contains already 330 vpm carbon dioxide, the generated and released amount of that component has to be corrected by the amount:

$$V_{CO_2 corr} = V_{resamp} \cdot 330 \qquad [\text{m}^3]$$

$$V_{CO_2 rel} = V_{CO_2 sys} - V_{CO_2 corr} \qquad [\text{m}^3]$$

For the other gases such as hydrogen, methane, ethane, propane, and butane

$$V_{isys} = V_{irel}$$

because air and nitrogen do not contain these components.

The specific amount of the released components can be calculated by:

$$V_{ispec} = \frac{V_{irel}}{m_{ben}} \qquad\qquad [m^3/kg]$$

4.2.2 Mineralogical analyses of the Opalinus clay

In addition to the gas release measurements, mineralogical analyses of the Opalinus clay were performed in order to determine the alteration as a result of heating and to gain knowledge about the origin of the thermally generated and released gases.

The mineralogical investigations were carried out with an X-ray diffractometer obtaining the qualitative and quantitative composition of the crystalline components in the whole sample.

For the X-ray analyses, a full protection equipment (Philips) with a diffractometer of the type X'Pert-MPD with sample changer and copper tube was available at GRS. The sample material was ground down and consolidated by means of a glass plate on an aluminium sample carrier to produce a smooth surface. The measurements were accomplished in the 2ϑ-angle between maximal 5° and 75° with a voltage of 40 kV and a current of 55 mA.

The total carbon content (TC), the inorganic carbon content (TIC), and the organic carbon content (TOC) were determined by means of the equipment type C-Mat 5500 of the company Ströhlein. The determination of the TOC was conducted according to EN 1484 (part H3) /EN 97/.

4.2.3 Geoelectric calibration

In order to enable adequate interpretation of the geoelectric in-situ measurements to be performed within the scope of the Heater Experiment (HE) in the Mont Terri Underground Research Laboratory, calibrations were carried out at GRS' geotechnical laboratory in Braunschweig. The calibrations included the determination of the resistivity of Opalinus clay samples from the HE-test field in dependence on their water

content at different temperatures. Both samples taken before start-up of heating and at the end of the heating phase were investigated.

The resistivity of the samples was determined using the four-point method. Here, the current I is injected at the end surfaces of the sample and the difference in the electrical potential U is measured at the voltage electrodes M and N. Figures 4-6 and 4-7 show the measurement set-up.
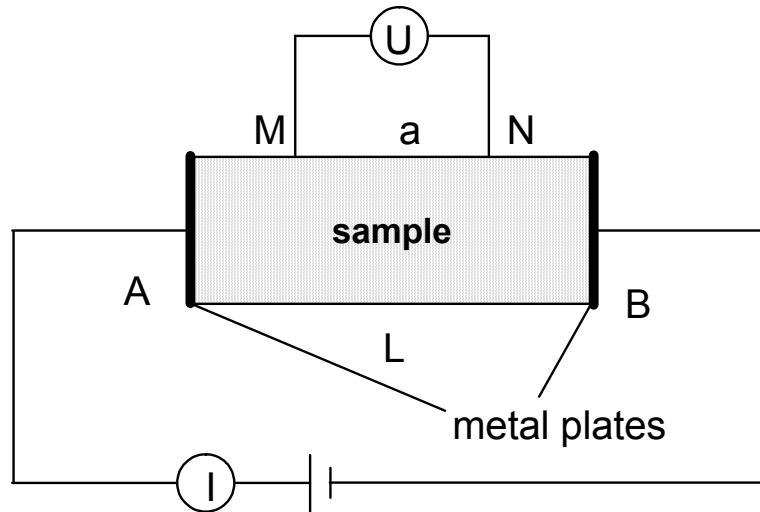

Figure 4-6 Schematic view of the 4-point arrangement for determination of sample resistivity
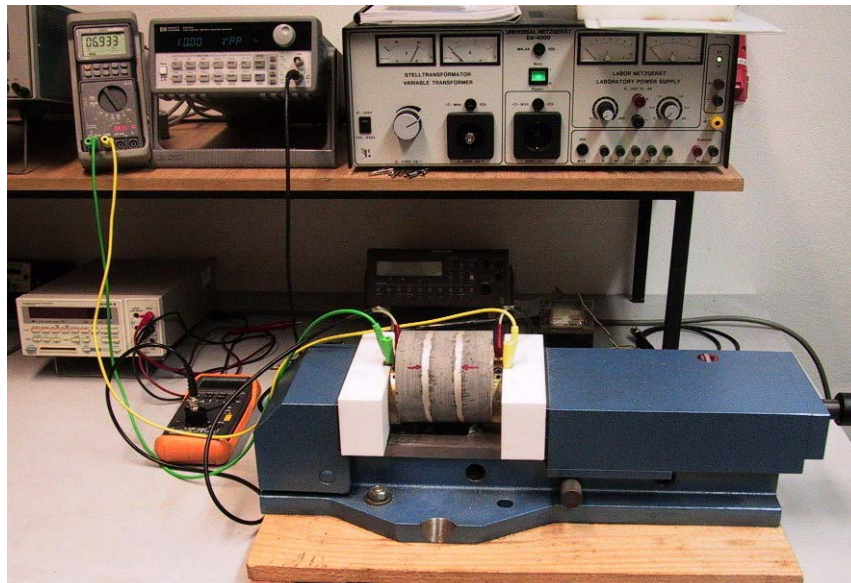

Figure 4-7 Measurement set-up for determination of clay sample resistivity

The resistivity is calculated according to Ohm's law /TEL 90/:

$$\rho = \frac{U \cdot A}{I \cdot a}$$

with

ρ	resistivity	$[\Omega m]$
U	electrical potential	[V]
I	current	[A]
a	distance of the electrodes M and N	[m]
A	cross section area	$[m^2]$

For a better coupling of the electrodes to the rock samples, the end surfaces were coated with conductive silver lacquer. For the voltage electrodes M and N, two rings of conductive silver lacquer were applied over the circumference of the samples (see Figure 4-8).

Figure 4-8   Prepared samples with conductive silver lacquer at the end surfaces and rings over the circumference

As the first step, the BHE-samples were investigated at the state of delivery without any additional saturation in order to determine the starting resistivity of each Mont Terri clay sample. Then, in order to achieve different saturations, the samples were stored in an exsiccator (see Figure 4-9) at air humidities of about 20 %, 42 %, 76 %, 90 %, and

100 %. To achieve the required air humidities, different salt solutions and water as summarized in Table were used.
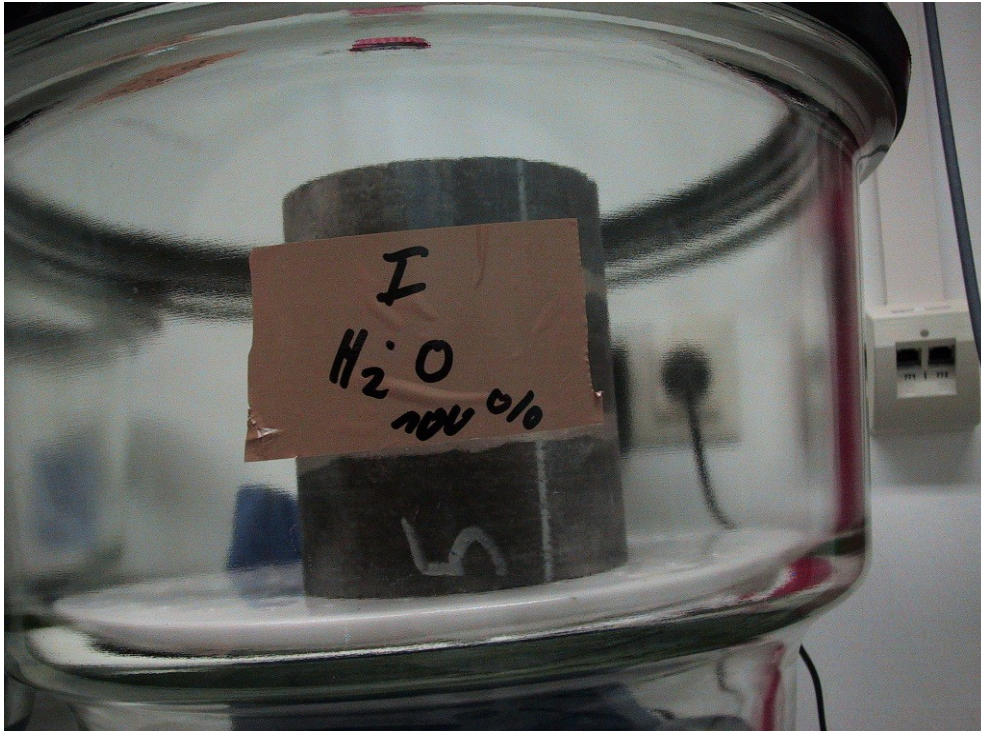

Figure 4-9   Exsiccator with a clay sample at 100 % air humidity

Table 4-2   Solutions and corresponding air humidities

air humidity [%]	solution
20	potassium acetate
42	zinc nitrate
76	sodium chloride
90	zinc sulfate
100	water

To investigate the temperature influence on the resistivity at each water content, the exsiccator with the sample was heated in an oven. Ensuring almost constant saturation conditions during the heating process, the samples were stored at the different air humidities. At each stage of saturation and temperature, the resistivity was measured and the water content of the sample was determined.

Since the water content w is given by the individual wet mass m_w in relation to the dry mass m_d, the samples were finally dried at 105 °C to constancy of mass which is assumed to represent the completely dry mass. The actual water content was then calculated on basis of the actual mass at the different saturation conditions by

$$w = \frac{m_w - m_d}{m_d} = \frac{m_w}{m_d} - 1$$

with

w	water content,	[-]
m_w	mass of the wet sample	[kg]
m_d	mass of the dry sample	[kg]

4.2.4 Density and porosity

In order to determine the stage of water saturation the knowledge of the porosity of the samples is essential. Therefore, the parameters density and porosity were determined.

The porosity of the samples was calculated by the grain density and the bulk density

$$\Phi = 1 - \frac{\rho_b}{\rho_g}$$

with

Φ	porosity	[-]
ρ_g	grain density	[kg/m^3]
ρ_b	bulk density	[kg/m^3]

The grain density was determined on the remaining clay material of the samples. The clay was dried at 105 °C and grinded. Afterwards, the grain density of the grinded clay was measured with helium using an air comparison pycnometer after Beckman (see Figure 4-10).
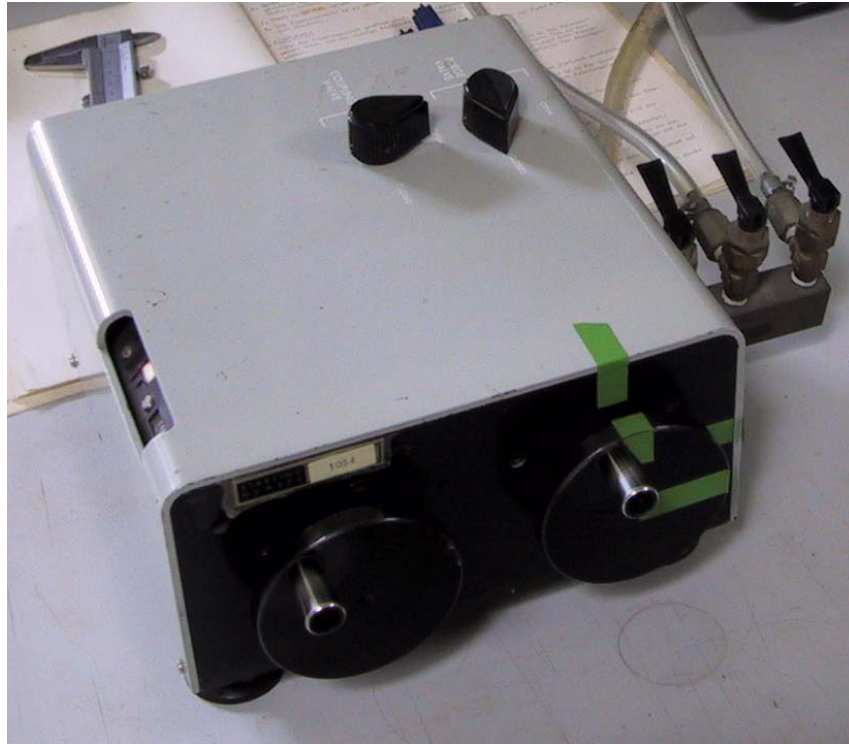
24

Figure 4-10 Air comparison pycnometer (after Beckman)

The bulk density was determined from the dry mass and the volume of the samples. The porosity of each sample was calculated using the bulk density and the grain density of the grinded material.

4.2.5 Sample locations and sampling procedure

The laboratory investigations on gas release and electric resistivity lasted from 1999 until 2004. Samples for these investigations were taken both prior to heating and after finishing the heating phase.

For determining the concentration of equilibrium, the total amount, and the time dependence of the released gases from the non heated Opalinus clay, samples from the boreholes BHE-10 to BHE-19 (see Figure 2-2) were taken during installation of the test field from levels between 2 m and 8 m below the floor (outside the EDZ).

For calibration of the geoelectric in-situ measurements, the resistivity of core samples was determined in the laboratory of GRS at Braunschweig on samples from different areas of the URL Mont Terri prior to heating. In order to avoid any drying of the

samples during storage and transportation they were wrapped up in plastic foil. In Table 4-3 the data of the samples are summarized.

Table 4-3 Data of the samples for calibration measurements of the electric resistivity of the Opalinus clay

sample	from depth [m]		sample length [cm]	sample diameter [cm]	direction of drilling°
BHE-B1/22a I	0.69	0.81	10.47	10.04	parallel to bedding
BHE-B1/22a II	0.82	0.93	9.37	10.05	parallel to bedding
BHE-B3/43a V	8.92	9.05	10.31	10.04	45° to bedding
BHE-B3/43b VI	9.47	9.60	12.25	10.04	45° to bedding
BHE-B3/43b VII	9.63	9.77	12.36	10.04	45° to bedding

At the beginning of the dismantling phase in September 2003, after 18 months of heating, two additional boreholes (BHE-26 and BHE-27, see Figure 4-11) with a distance of 0.5 and 1.50 metre to the centre of the heater were drilled by BGR. From these boreholes, cores with a diameter of 101 mm were taken from different depths. The core samples obtained were used for determining the alteration of the Opalinus clay with regard to gas generation and gas release as a result of heating and to verify the results of the geoelectric tomography measurements.

For the laboratory investigations it is essential to have specimens without significant alteration with regard to the mechanical behaviour, the mineralogical-chemical composition, and the fluid content in the pores. During sampling, storage, and transportation the specimens should remain under conditions which are similar to the conditions in the original environment. In general, specimens were wrapped up in plastic foils or bags and were stored in open metal or wooden boxes. Very often these methods have not been sufficient, as the plastic foil or the specimens were damaged mechanically. The specimens dried, oxidised by contact with air, or lost their adsorbed gases.
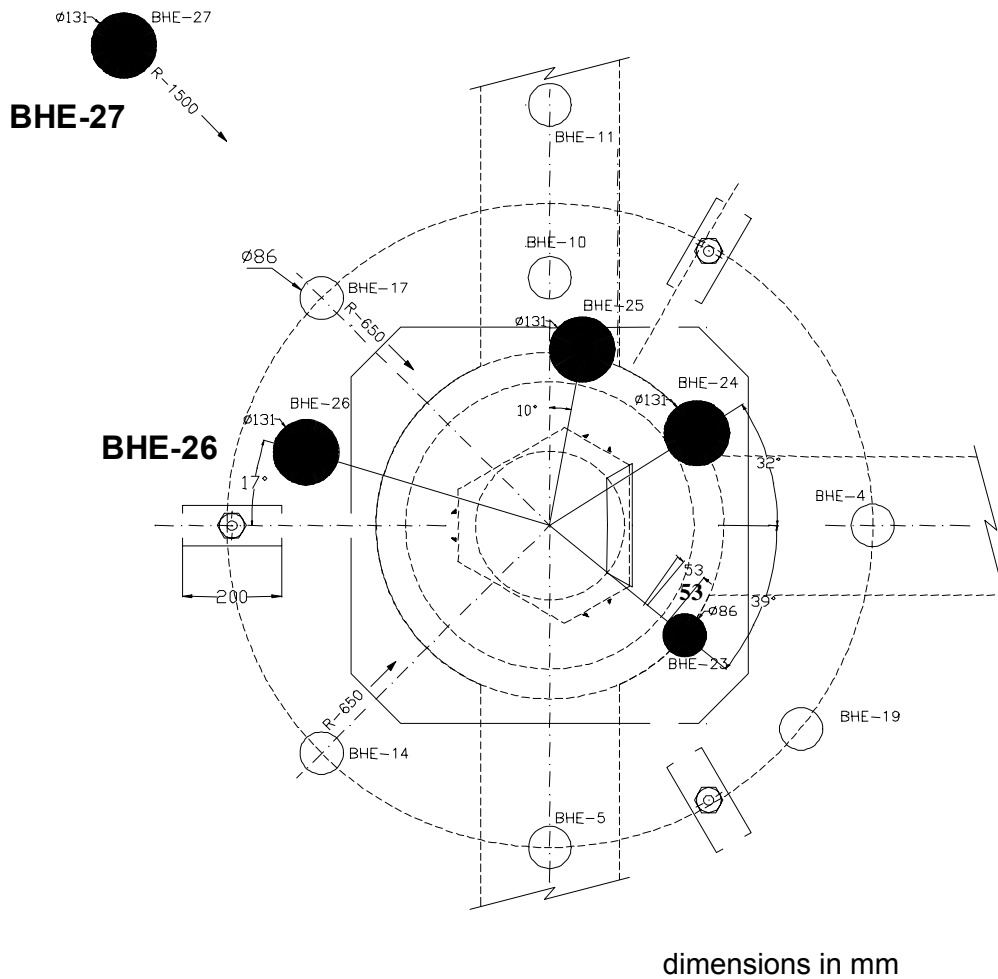
Figure 4-11   Position of the boreholes BHE-26 and BHE-27

A new method has been developed within the HE-B test for storage and transportation of specimens. Cylindrical tin boxes with 63 to 230 mm diameter and 62 to 260 mm height can be used. The size of the box can be adjusted to the diameter, length, and amount of the specimen. The residual volume in the box can be backfilled with drilling fine or special protection material. Drilling fine of the same material as the specimen guarantees an atmosphere of equilibrium regarding the relative humidity and the gas composition within a short time. The specimen is protected against drying, external humidity and air.

The specimen can be emplaced in the box and sealed gas-tight with the folding machine as shown in Figure 4-12 without time delay at the location where it is taken. The box can be opened easily by a tin opener for extraction of the specimen and for further investigation.

27

Figure 4-12   Folding machine for tin boxes with tin boxes of 113 mm diameter and 205 mm height

The samples for investigation of gas release were placed into these special gastight steel containers or into glass vessels for storage and transportation in order to avoid degassing and drying immediately after removal from the drilling machine. The steel vessels had an internal height of 300 mm and a diameter of 102 mm, the glass vessels had an internal height of 265 mm and a diameter of 145 mm. The following samples were investigated:

- Borehole BHE-26 (0.5 m to heater)

  – From 2 to 4 m depth (above the heater): 300 mm of material in a steel vessel, 265 mm of material in a glass vessel

  – From 4 to 6 m depth (heater area): 300 mm of material in two steel vessels, 265 mm of material in two glass vessels

  – from 6 to 7 m depth (beneath the heater): 300 mm of material in two steel vessels, 265 mm of material in two glass vessels

- Borehole BHE-27 (1.5 m to heater):

  – From 2 to 4 m depth (above the heater): 300 mm of material in a steel vessel, 265 mm of material in a glass vessel

  – From 4 to 6 m depth (heater area): 300 mm of material in a steel vessel, 265 mm of material in two glass vessels

  – From 6 to 7 m depth (beneath the heater): 300 mm of material in two steel vessels, 265 mm of material in two glass vessels

In addition, one Opalinus clay sample was taken directly from the contact zone to the highly compacted bentonite blocks.

The gas in the residual volume of the containers was mine air. During transportation and storage in the GRS laboratory in Braunschweig the samples were exposed to a temperature of about 20 °C.

In order to verify the results of the geoelectric tomography measurements and determine the effect of heating during the in-situ test on the water distribution and therefore on the resistivity of the Opalinus clay, four samples were taken from each of the boreholes BHE-26 and BHE-27 and prepared directly in the Mont Terri tunnel. Then, the resistivity of the samples was determined on site directly after the preparation.

5 Measurement Results and Discussion

5.1 In-situ results

This section contains all the results regarding gas release and geoelectric measurements obtained during the pre-heating phase from May 1999 to February 2002 and for the heating period from February 2002 to August 2003.

5.1.1 Gas analyses

The results of the gas composition in the 4 investigation boreholes and in the mine air of the HE-niche are compiled in the Figures 5-1 to 5-5 and additionally in the Tables 10-1 to 10-9 in the Annex. In the beginning right after sealing the boreholes (May 1999), the residual volume of the boreholes was filled with mine air (about 80% N_2, 20 % O_2, 1% Ar and 400 to 500 vpm CO_2). During the pre-heating phase, carbon dioxide, some helium, and hydrocarbons were released at mine temperature (about 14 °C). As a result of oxidation of organic material in the clay, the concentration of oxygen decreased from 20% to a few percent. The concentration of the gas components varied significantly up to a factor of 15 in the different boreholes which are only 0.35 m apart (BHE-10 to BHE-11). This fact indicates that the Opalinus clay which seems to be homogeneous macroscopically is not homogenous regarding the gas content.

Heating of the central borehole started on February 22, 2002. Significant influence of heating is only seen in borehole BHE-10 which has a distance to the heater of about 0.5 m. The concentration of carbon dioxide increased from 814 vpm (ml/m^3) in February 2002 to 36000 vpm (3.6 vol%) in February 2003. The hydrocarbons (CH_4, C_2H_6, C_3H_8, C_4H_{10}) increased from 234 to 3303 vpm and helium from 21 to 114 vpm. Hydrogen sulphide (H_2S) which was generated after heating was found in borehole BHE-10 up to 680 vpm and in the other boreholes with less than 12 vpm. This gas component is of importance concerning the disposal of radioactive waste even at low concentrations, as it leads to corrosion of the waste and the waste container in combination with water in the liquid or gaseous phase.
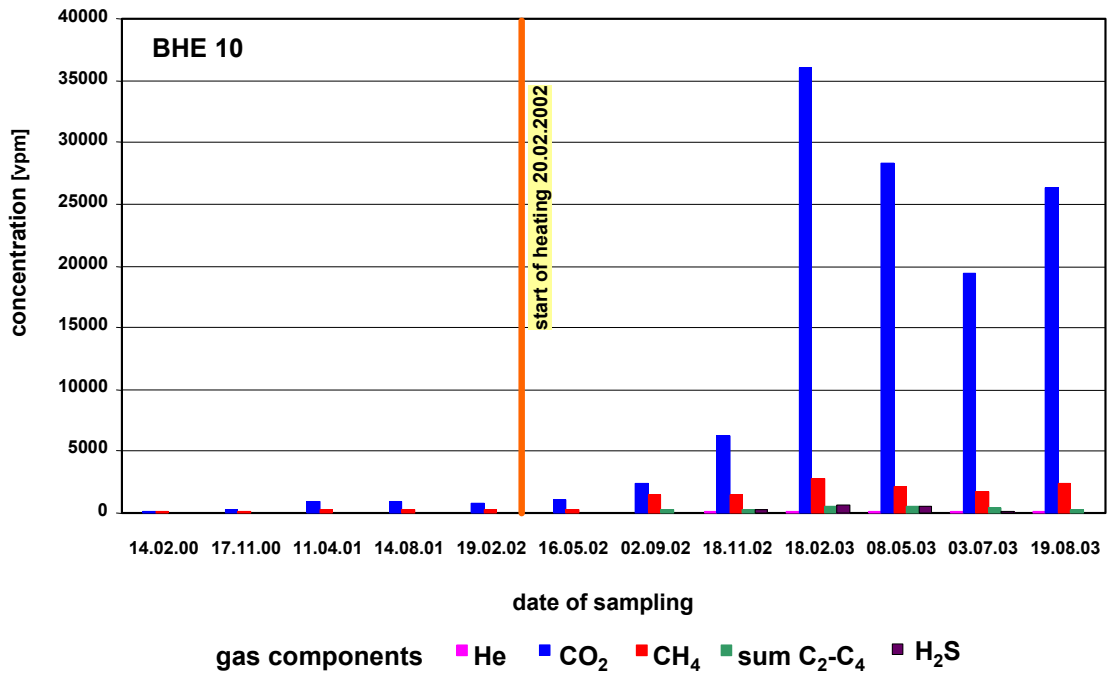
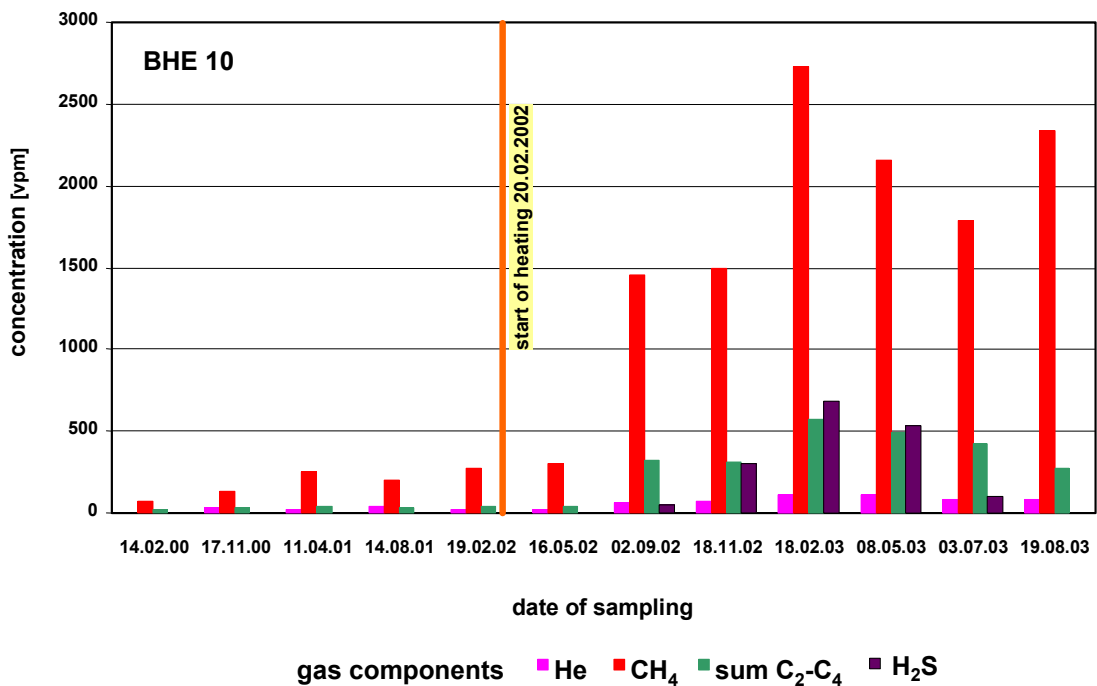**Figure 5-1**   Concentration of the gas components in borehole BHE-10

Figure 5-2   Concentration of the gas components without CO_2 in borehole BHE-10
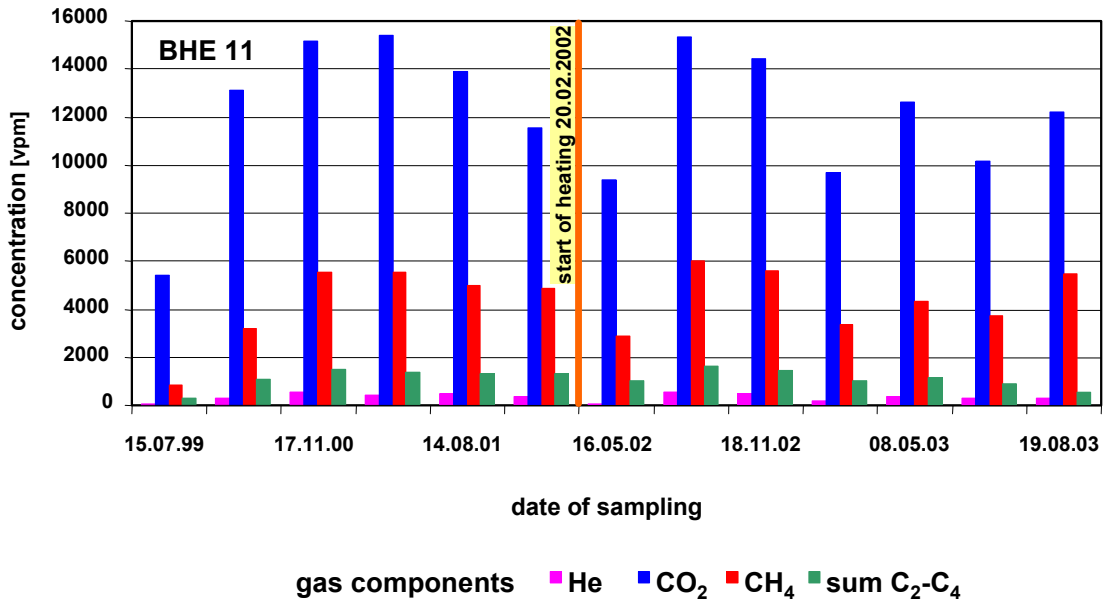
Figure 5-3   Concentration of the gas components in borehole BHE-11
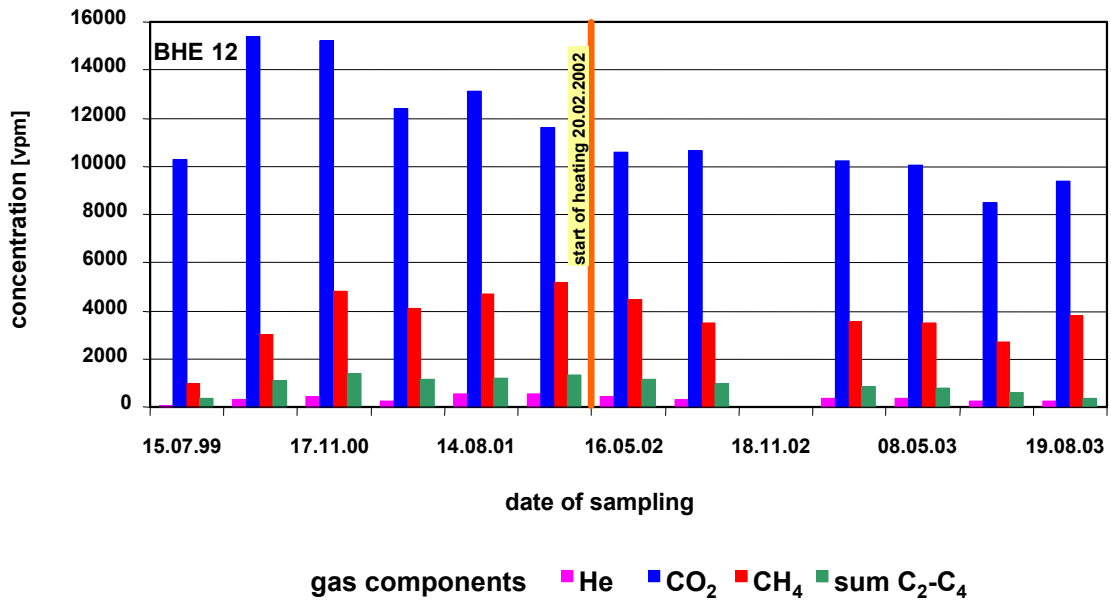

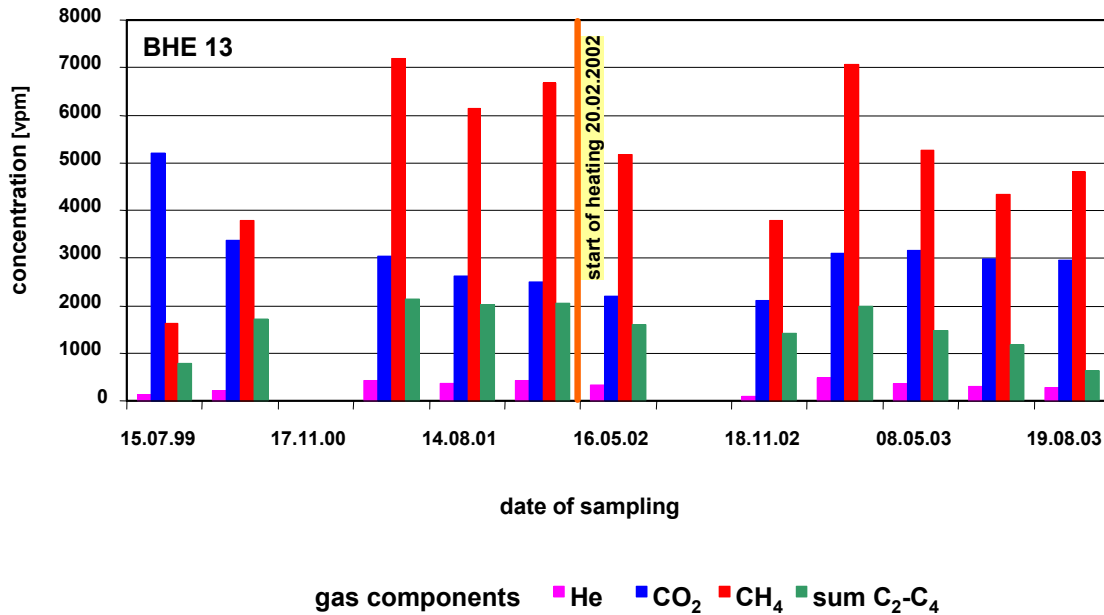**Figure 5-4**   Concentration of the gas components in borehole BHE-12

Figure 5-5   Concentration of the gas components in borehole BHE-13

Since February 2003 the concentrations of the components carbon dioxide, helium and the hydro carbons showed a slight decrease and oxygen showed a slight increase. This can especially be seen in borehole BHE-10. Furthermore, no significant pressure increases in the boreholes were observed as a result of gas release. Obviously, the host rock is not absolutely gas-tight.

The laboratory investigations indicated that gas release from the Opalinus clay is a very slow process and therefore sampling more often than three to four times a year rarefies the gas concentration in the system.

For comparison and for testing the analytical system, additional gas samples were taken from the mine air in the HE-niche. Analysis of these samples indicated that even the mine air compared to the open air has an increased concentration with regard to helium (20 vpm) and carbon dioxide (800 vpm), as a result of gas release from the open surfaces of the host rock. Large variations are caused by different intensities of ventilation. The variation of the oxygen content in the range between 20.1 to 22.0 % is caused by sampling and by the measuring error of the analytic system which is in the range between 3 and 5 %.

The measurements on site with the portable gas chromatograph system and samples transferred in gas bags to the GRS laboratory in Braunschweig for analysis did not

show significant differences. The advantage of the portable gas chromatograph and analysing on site is that the whole system is not disturbed so much, for only a small amount of gas is needed. Furthermore, falsifications by sampling and transporting the gas bags to the laboratory are excluded.

The absolute humidity in the residual volumes of the boreholes varies in the range between 7.9 and 13.5 gram water per m^3 air which is influenced mainly by the temperature in HE-niche. This temperature itself is influenced by the season and the intensity of ventilation.

5.1.2 Water analyses from the boreholes BHE-10 to BHE-13

After the last gas sampling in the boreholes BHE-10 to BHE-13 in August 2003, the water that had accumulated in the boreholes was extracted as described in Section 4.1.1. The amounts are shown in Table 5-1. For chemical analysis this water was sent to CIEMAT in Madrid.

Table 5-1 Amounts of water extracted from the gas sampling boreholes

Borehole	Amount of water extracted in August 2003	Amount of water extracted in October 2003
BHE-10	2231 ml	0,0 ml
BHE-11	658 ml	0,0 ml
BHE-12	1160 ml	213 ml
BHE-13	335 ml	139 ml

The analyses and the comparison with the Opalinus clay formation water and the synthetic formation water injected into the borehole BHE-0 for saturation of the bentonite buffer were performed by the CIEMAT Pore Water Laboratory.

The chemical composition of the waters analysed is shown in Table 5-2. The chemical composition of the Opalinus Clay pore water in the BWS-A1 borehole is shown for comparison. This borehole has been selected due to its proximity to the HE-experiment. In the BWS-A1 borehole, the pore water was obtained ex situ from squeezing at laboratory from unaltered core samples, and in situ, collected inside a borehole where the water was allowed to seep into a packed-off interval /PEA 03/.

Also, the water samples were compared with the A1-type synthetic water injected by AITEMIN in the BHE-0 borehole in order to saturate the bentonite buffer.

Table 5-2   Chemical composition of the rock waters in the HE-B test field at Mont Terri URL

Parameters	Borehole BHE10	Borehole BHE11	Borehole BHE12	Borehole BHE13	Borehole BHE12	Borehole BHE13	BWS-A1 seep water	BWS-A1 squeezed water
Date	August 2003				October 2003			
pH	7.04	7.9	7.16	7.2	7.4	6.9	8.67	7.48
Eh (mV)	-66.3	--	-76.5	-87.7	--	65.6	-88 (SHE)	--
Cond. (mS/cm)	23.1	25.6	27.3	27.6	28.2	27.3	--	--
Cl^- (mg/L)	9600	10500	11.000	11.000	12000	12000	10958	10300
SO_4^{2-} (mg/L)	790	1600	1800	1600	1900	1600	1280	2120
HCO_3^- (mg/L)	414	234	236	158	221	107	52.0	41-113
Br^- (mg/L)	21	15	26	26	27	26	33.0	33.1
F^- (mg/L)	0.45	0.68	0.48	0.73	0.65	0.63	0.59	--
I^- (mg/L)	2.2	2.3	2.7	2.9	2.7	2.6	2.16	--
NH_4^+ (mg/L)	12	14	13	13	14	13	1.1	--
SiO_2 (mg/L)	15.5	10	10.2	7.5	--	7.7	27.8	--
Ca (mg/L)	505	685	775	811	--	757	600	793
Mg (mg/L)	352	385	533	441	--	525	424	537.8
Na (mg/L)	5100	6000	6100	6000	--	6000	5575	5650
K (mg/L)	75	75	82	81	--	79	42.8	34.7
Mn (mg/L)	0.10	0.21	0.2	0.24	--	0.2	0.025	0.27
Al (mg/L)	0.1	0.08	0.11	0.1	--	0.05		0.357
Fe (mg/L)	0.262	--	2.3	1.1	--	< 0.05	--	0.74
Fe^{2+} (μg/L)	0.262	--	2.3	1.1	--	< 0.05	--	--
Sr (mg/L)	26	31	37	41	--	35	35.0	51.7

The pH of the waters is neutral and slightly lower than that expected for the original rock water. The ion concentrations in the different boreholes are shown in Figure 5-6. Significant influence of heating or the bentonite proximity is only seen in borehole BHE-10 which has a distance to the heater of about 0.5 m and is the closest to the BHE-0

borehole. According to the chloride concentration, a dilution of the rock water composition is observed in the BHE-10 borehole. This can be also observed in the concentration of sulfate and cations. However, an increase of bicarbonate is observed in this borehole (BHE-10), which seems to indicate a water movement from the bentonite, during the saturation phase, to the formation and dissolution of the carbonate phases of the Opalinus clay or the bentonite. Due to this process, some exchange reactions could be occurring, increasing the Na content in the rock water and decreasing the Ca and Mg contents. By other hand, the bentonite could have adsorbed some ions of the rock pore water during its saturation. The bentonite is not saturated at the end of the experiment, so the rock water in the BHE-10 borehole did not become recovered again.

According to the chloride contents (see Figure 5-6 and 5-7), a salinity increase is observed in the rock (BHE-11, 12 and 13 boreholes) as a function of time and the distance to the BHE-0 borehole, i.e., there is an increase of the salinity towards the BHE13 borehole. This salinity increase is more accentuated in the second sampling campaign, whose values are higher than the concentrations obtained in the pore water samples of the BWS-A1 borehole (Figure 5-7), and similar to the highest values of chloride found in the Opalinus Clay at the contact to the Jurensis Marl formation.

There is an increase of sulfate in all the boreholes, except for the BHE10, which indicate a slightly oxidation of the rock. This is in agreement with the Eh measured in the waters (see Table 5-2, in which SHE values are not shown), whose values are oxidized. This does not correspond to the reducing conditions of the Opalinus Clay system. Thus, as was stated above, the rock is not completely gas-tight. On the other hand, the rock-water system in the boreholes was oxidized at initial conditions of the heating test. This could have perturbed the system and provoked the oxidation of the rock. However, the reducing conditions seem not to recover with time.

Regarding the CO_2 contents, a key parameter of the rock water composition, the partial pressure of CO_2 is always higher than that of air (Figure 5-8). The average amount of Log P_{CO2} is –2.09. The BHE-11 and BHE-12 boreholes follow a similar behaviour regarding to the Log P_{CO2}, with an average value of –1.83 atm. The BHE-13 borehole has an average value of –2.57 atm. However, the BHE-10 borehole, close to the bentonite buffer, has a different behaviour. It seems that there is a significant influence of heating, which is only seen in this BHE-10 borehole. The minimum value of Log P_{CO2} is –3.09 and the maximum value is –0.55, with a final average value of –1.65 atm. All

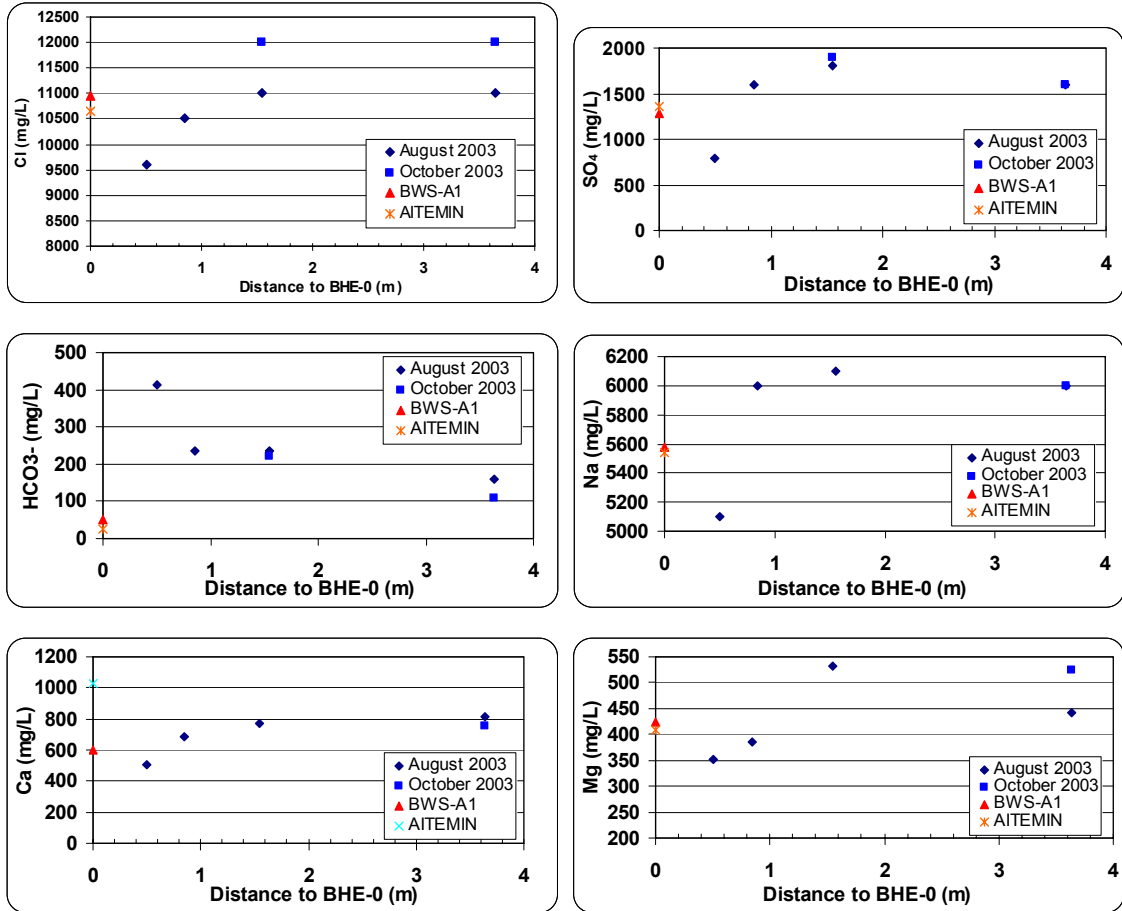the data are in the range of the measured values in the Opalinus Clay rock in other experiments /PEA 03/.

Figure 5-6 Ion Concentration in the boreholes BHE-10, 11, 12, and 13 and comparison to the A1-type synthetic water injected by AITEMIN and the water of the BWS-A1 borehole
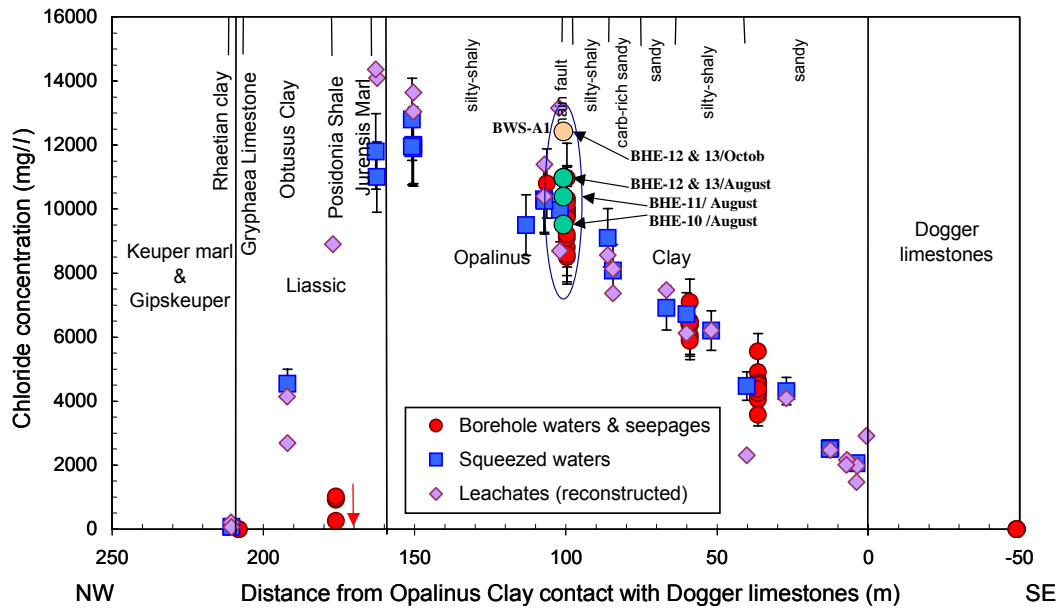
Figure 5-7   NW-SE cross-section of chloride concentration in Opalinus clay pore waters and comparison with the data obtained in the HE-Experiment /PEA 03/
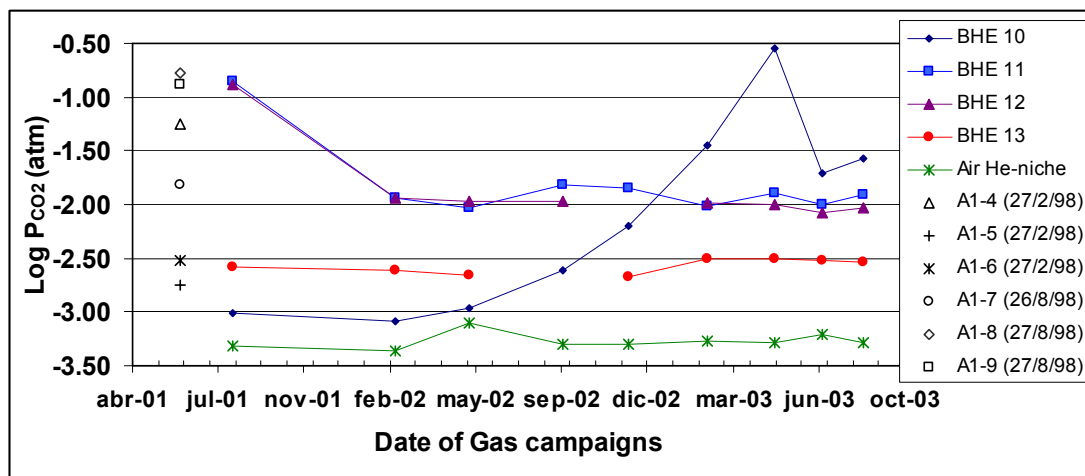

Figure 5-8   Evolution of the partial pressure of CO2 (atm) in the boreholes BHE-10 to BHE-13. Data are compared with the chemical composition of head-space gas (A1-x samples) in the packed-off interval of the borehole BWS-A1 /PEA 03/

5.1.3 Geoelectric tomography and water redistribution

Geoelectric measurements were performed since June, 1999 in the plane between the boreholes BHE17 and BHE16, and in all planes between the four boreholes since July, 1999. Measurements stopped at the end of August, 2003, when heating was terminated and post-test investigations started.

Evaluation of the measurements was first performed using the code RESITOMO /HAR 96/. From 2002 on, the modern code SensInv2D /FEC 01/ was used. The Figures 5-9 to 5-12 show inversion results for the planes BHE17-BHE16, BHE14-BHE15, and BHE14-BHE17 in three-dimensional graphs, for the time between January 2002 and August 2003, all generated with SensInv2D. The electrode chains in the boreholes are depicted as black lines in the figures.
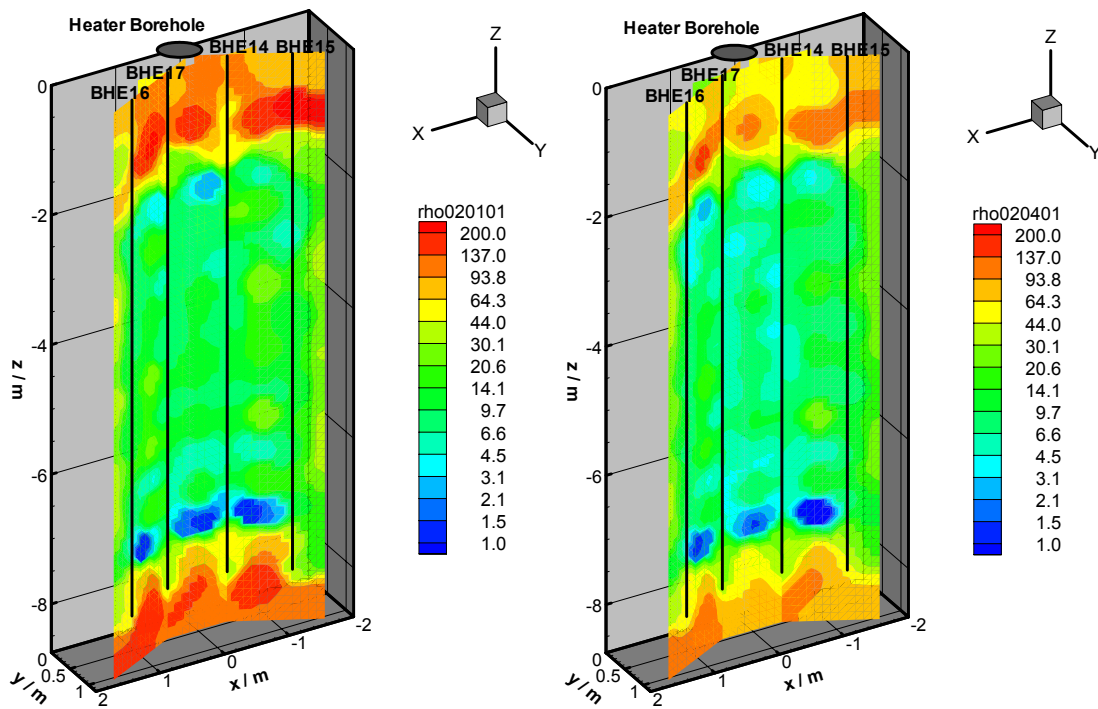
Figure 5-9   Resistivity tomograms obtained on January 1, 2002 (prior to heating – left) and on April 1, 2002 (during heating – right)

Figure 5-9 shows on the left side the result of a measurement performed in January 2002, prior to start-up of heating. Apart from the upper and lower ends of the model which are influenced by boundary effects, the resistivity distribution is rather homogeneous, with values of 7 to 20 Ωm that can be related to full saturation. A low resistivity anomaly around 7 m depth is caused by the metallic heater.

40
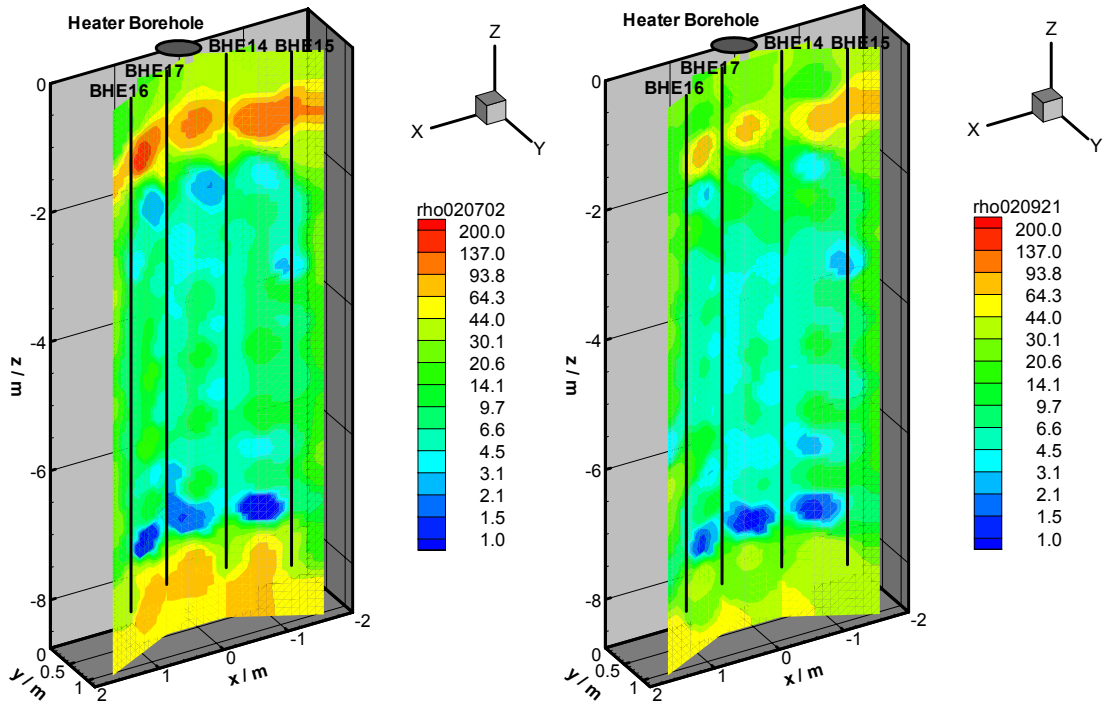
Figure 5-10 Resistivity tomograms obtained on July 2, 2002 (left) and on September 21, 2002 (right)
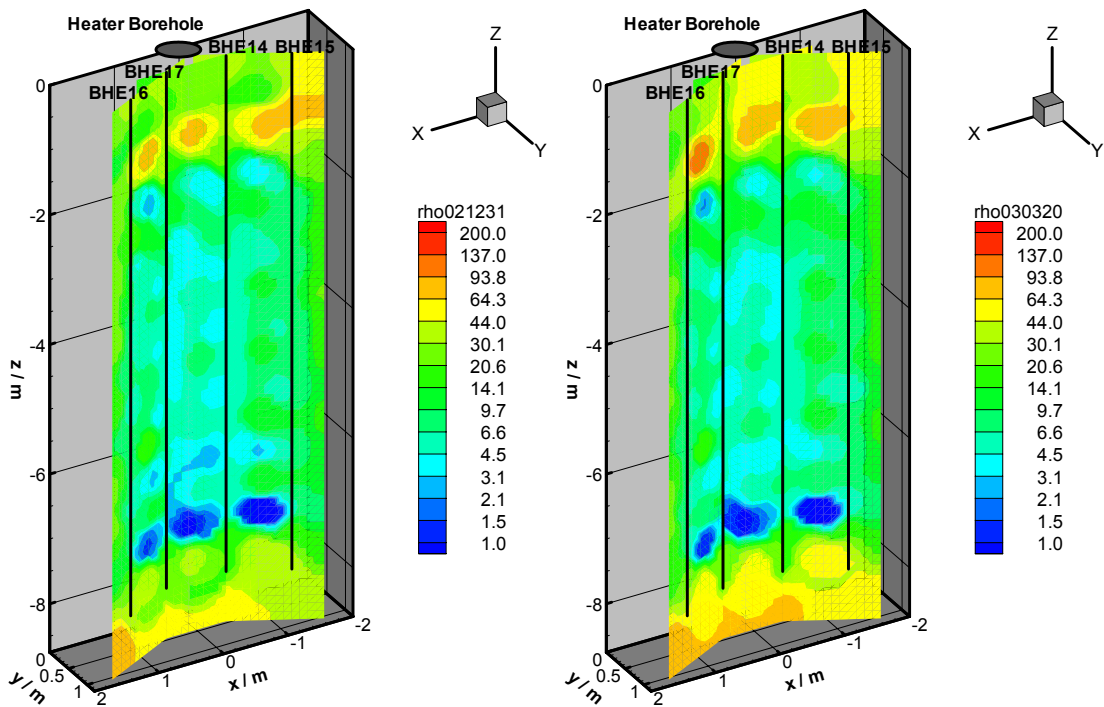

Figure 5-11 Resistivity tomograms obtained on December 31, 2002 (left) and on March 20, 2003 (right)
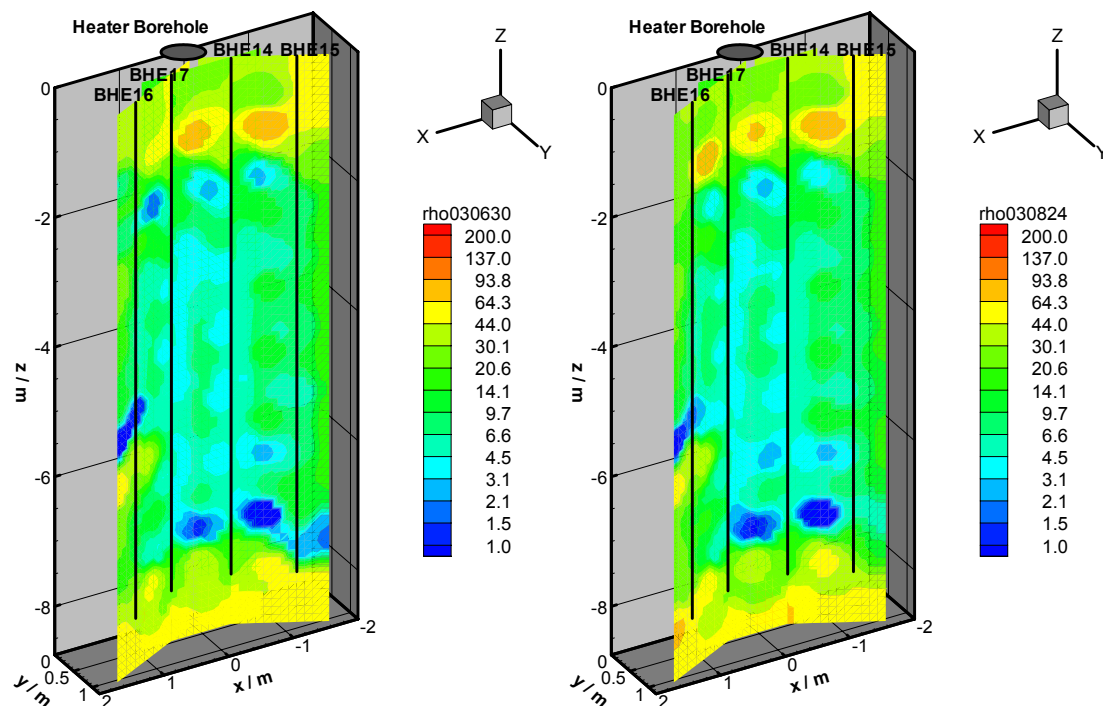
41

Figure 5-12   Resistivity tomograms obtained on June 30, 2003 (left) and on August 24, 2003 (last measurement – right)

In February 2002, heating of the lower part of borehole BHE0 was started. The tomograms from April 2002 (Figure 5.9, right hand side) to September 2002 (Figure 5.10, right hand side) show a progressive reduction of resistivity, especially in the plane BHE17/14 close to the heater borehole, which is probably due to an increase in conductivity of the pore water with increasing temperature. Laboratory testing at elevated temperature confirmed this interpretation (see Section 5.2.2.3). In September 2002 the resistivity in this plane ranged from 3 to 7 m.

Since September 2002, the tomograms changed only insignificantly (see Figure 5.11 to 5.12). This may be the consequence of a steady state temperature field being reached. The mean resistivity evolution in the heated area between the boreholes BHE17 and BHE14 is shown in Figure 5.13 in order to make the resistivity evolution better visible. Here, the resistivity decrease during the first half year of heating as a temperature effect (until a steady temperature field is reached) and the succeeding steady-state resistivity afterwards is clear. In particular, no resistivity increase which could be attributed to desaturation effects was detected at any time.
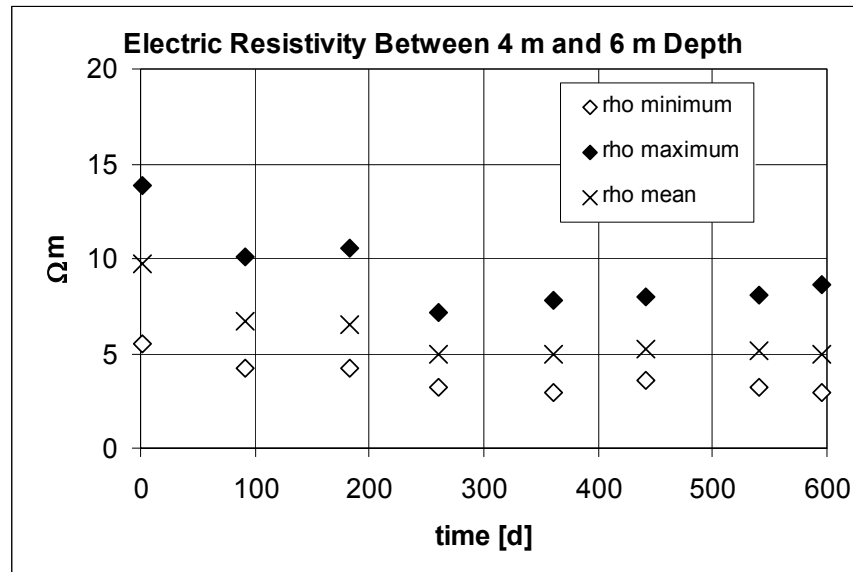
Figure 5-13   Evolution of electric resistivity between 4 m and 6 m depth in the plane
BHE14-BHE17

Overall, it can be stated that the geoelectric measurements give no indications of heat-induced desaturation of the clay rock until end of the heating period. The only effect detected is a reduction of resistivity due to temperature increase. This result is confirmed by the post-test investigations performed on samples from the test field (see Section 5.2.2.3).

5.2   Laboratory results

5.2.1   Pre-test investigations

This section contains all the results of gas release and geoelectric measurements on the samples taken from the test field during installation (prior to heating) from the boreholes BHE10 to BHE-19.

5.2.1.1   Gas release from the clay samples

The aim of the laboratory investigations was to determine the relevant parameters of gas release within defined physico-chemical conditions.

The investigations indicated that the most important gas which was generated in the Opalinus clay and released into the residual volume of the reaction vessels is carbon dioxide. This component is primarily present in the clay as it is released already at low temperature (20 °C). Furthermore it is generated by thermal decomposition of the organic components within the Opalinus clay and by oxidation of these organic components at room and elevated temperature. Additionally, small amounts of gaseous hydrocarbons, especially methane, ethane, propane, and pentane were released. In contrast to the in-situ investigations helium was not analysed in concentrations significantly above the detection limit of the gas chromatography system (about 1 vpm). The reason is that Opalinus clay contains only a small amount of helium and might also have escaped during drilling, storage, and preparation of the samples, as it is a very mobile component.

For determining the concentration of equilibrium of the released gases 3.2 kg of Opalinus clay from the borehole BHE-19 were emplaced in a stainless steel container of 2.2 litres. That means the relation between the amount of the sample and the volume into which the gases are released is comparatively high. The results of that investigation are shown in Figure 5-14. At a temperature of 95 °C the concentration of equilibrium of the released gases was reached almost after 10 days. Further heating did not lead to further gas release into the closed volume.
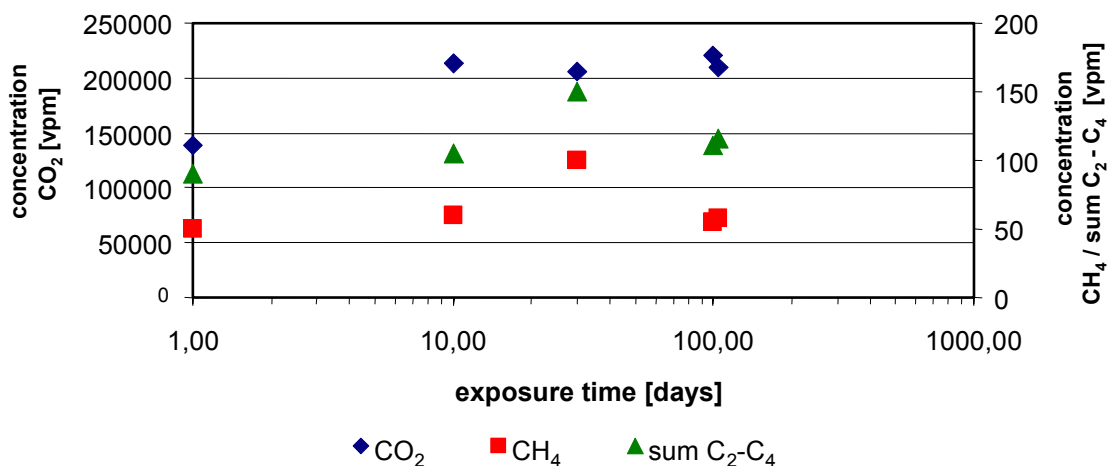

Figure 5-14 Development of the gas concentration in the stainless steel degassing container at 95 °C with a big amount of Opalinus clay sample (3.2 kg) and low residual volume (600 ml)

The concentrations of equilibrium are:

- Carbon dioxide about 22000 vpm = 2.2 vol%

- Methane about 70 vpm

- Further hydrocarbons about 120 vpm

Table 5-3   Data of the ampoules for gas release measurements on Opalinus clay from borehole BHE-19/2

ampoule No.	amount of Opalinus clay [g]	gas in the residual volume	time period of exposure [days]	date of exposure	date of retrieval and analyses
301 to 303	1	air	1	04.11.2002	05.11.2002
304 to 306	1	air	3	04.11.2002	07.11.2002
307 to 309	10	air	3	04.11.2002	07.11.2002
310 to 312	1	air	10	28.10.2002	07.11.2002
313 to 315	1	air	30	07.10.2002	06.11.2002
316 to 318	10	air	30	07.10. 002	06.11.2002
319 to 321	1	air	101	01.10.2002	10.01.2003
322 to 324	1	air	300	06.11.2002	02.09.2003
325 to 327	10	air	300	06.11.2002	02.09.2003
328 to 330	1	air	800	01.10.2002	09.12.2004
331 to 333	1	N_2	1	04.11.2002	05.11.2002
334 to 336	1	N_2	3	08.11.2002	11.11.2002
337 to 339	10	N_2	3	08.11.2002	11.11.2002
340 to 342	1	N_2	10	04.11.2002	14.11.2002
343 to 345	1	N_2	30	14.10.2002	13.11.2002
346 to 348	10	N_2	30	01.10.2002	13.11.2002
349 to 351	1	N_2	101	01.10.2002	10.01.2003
352 to 354	1	N_2	300	07.11.2002	03.09.2003
355 to 357	10	N_2	300	07.11.2002	03.09.2003
358 to 360	1	N_2	800	01.10.2002	09.12.2004
361 to 362	0	air	300	08.11.2002	04.09.2003
363 to 364	0	N_2	300	08.11.2002	04.09.2003

For determining the gas release from the Opalinus clay as a function of exposure time and gas in the residual volume of the reaction vessels glass ampoules of 500 ml volume were used. In Table 5-3 the data of these ampoules with their identification numbers are compiled. The Opalinus clay for that investigation was taken from core sample of the borehole BHE-19/2. The natural dry clay was ground and homogenised. 1 or 10 grams were transferred into the ampoule with air or nitrogen in the residual, respectively. For statistic reasons three replicates of each condition and additionally for blind tests ampoules without clay only with nitrogen or air were prepared.

After exposure the gas in the residual volume of the ampoules was analysed and evaluated as described in Section 4.2.1. The results of this investigation for the component carbon dioxide are shown in Figure 5-15.
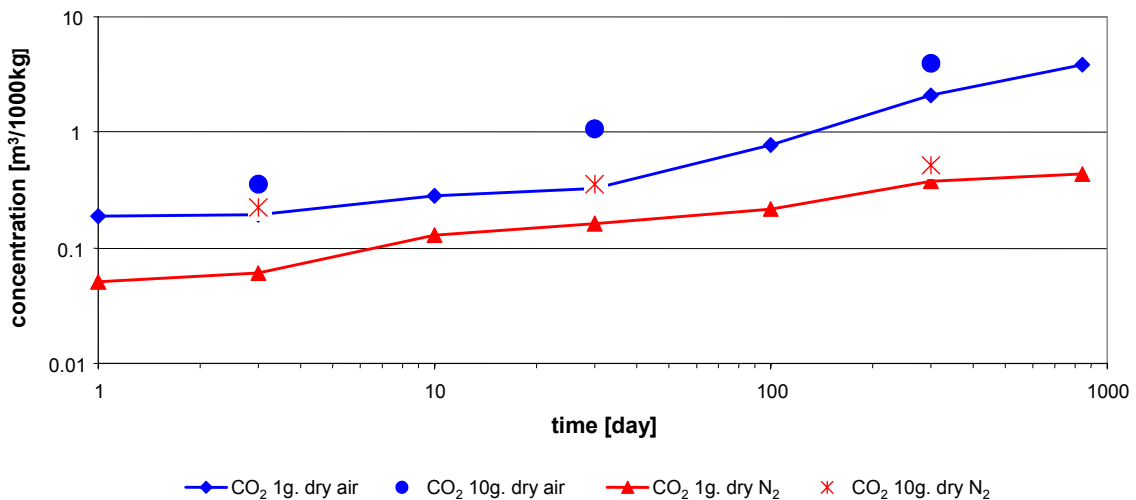
Figure 5-15   Release of carbon dioxide from the Opalinus clay of the borehole BHE-19/2 at 95 °C as a function of time

It is most likely that carbon dioxide is generated by oxidation of organic material in the clay as its amount is much higher in those ampoules with air in the residual volume than in those with nitrogen in the residual volume. Additional carbon dioxide may be generated by thermal decomposition of carbonates in the clay. Furthermore, carbon dioxide is primarily present in the clay adsorbed to the internal surfaces and will be released at elevated temperature.

After 842 days of exposure at a temperature of 95 °C, 3.8 m³ carbon dioxide per 1 000 kg of clay were released in the aerobic and up to 0.43 m³ per 1 000 kg of clay in the un-

aerobic stage. The release was not finished after 842 days as the concentration of equilibrium had not been reached yet. The amount of the clay in the ampoules did not have significant influence. For 1 gram the values were a little bit less than for 10 grams. It might be that small amounts of gas were adsorbed to the surface of the ampoule.

From the laboratory investigation on gas generation and release at elevated temperature it can be concluded that the concentration of equilibrium for carbon dioxide in the pore volume will be in the range of 2 to 3 vol%. If the host rock is gas-tight, only a small amount of organic matter will be oxidised or decomposed. If the host rock is not gas-tight and the released carbon dioxide can migrate into residual volumes of the repository, all the organic material will be oxidised if oxygen is available. Also, the thermally instable carbonates will be decomposed. It should be taken into account that up to 10 m³ carbon dioxide per 1.000 kilograms of clay may be released.

5.2.1.2 Resistivity measurements

In the state of delivery, the water content of all BHE-samples ranged between 4.6 wt% and 6.2 wt%. The corresponding resistivities of the samples taken from the cores BHE-B3/43b were about two to three times higher than the others. The resistivities varied between 15.7 Ωm to 31.9 Ωm and 6.9 Ωm to 7.5 Ωm, respectively. This observation could not be explained by different water contents, because one of the BHE-B3/43b samples had a water content of 4.6 wt% and the other 6.2 wt%. The water content of the samples were in order of 5.5 to 6.1 wt%.

Drying at ambient temperature led to water contents of about 2.1 wt% to 3.31 wt%. In correspondence to the results described above, the resistivities of the BHE-B3/43b samples with values of 114 Ωm and 118 Ωm were again higher than the BHE-B1/22a and BHE-B3/43a (29.7 Ωm to 34.4Ωm). These results are summarized in Table 5-4. The measured resistivities at each step of water content are plotted in Figure 5-16. For different water contents, the resistivity measurements of the BHE samples were performed at 25 °C, 30 °C, 50 °C, and 70 °C. The results are presented in Figure 5-17.

Table 5-4 Resistivities and water contents of Mont Terri He-samples at state of delivery and after drying at ambient conditions

sample	state of delivery		after drying at ambient conditions	
	water content [wt%]	Resistivity [Ωm]	water content [wt%]	Resistivity [Ωm]
BHE-B1/22a I	6.09	7.3	3.31	29.6
BHE-B1/22a II	5.61	6.9	2.77	34.4
BHE-B3/43a V	5.51	7.5	2.70	32.4
BHE-B3/43b VI	4.64	31.9	2.06	117.8
BHE-B3/43b VII	6.20	15.7	2.61	114.4
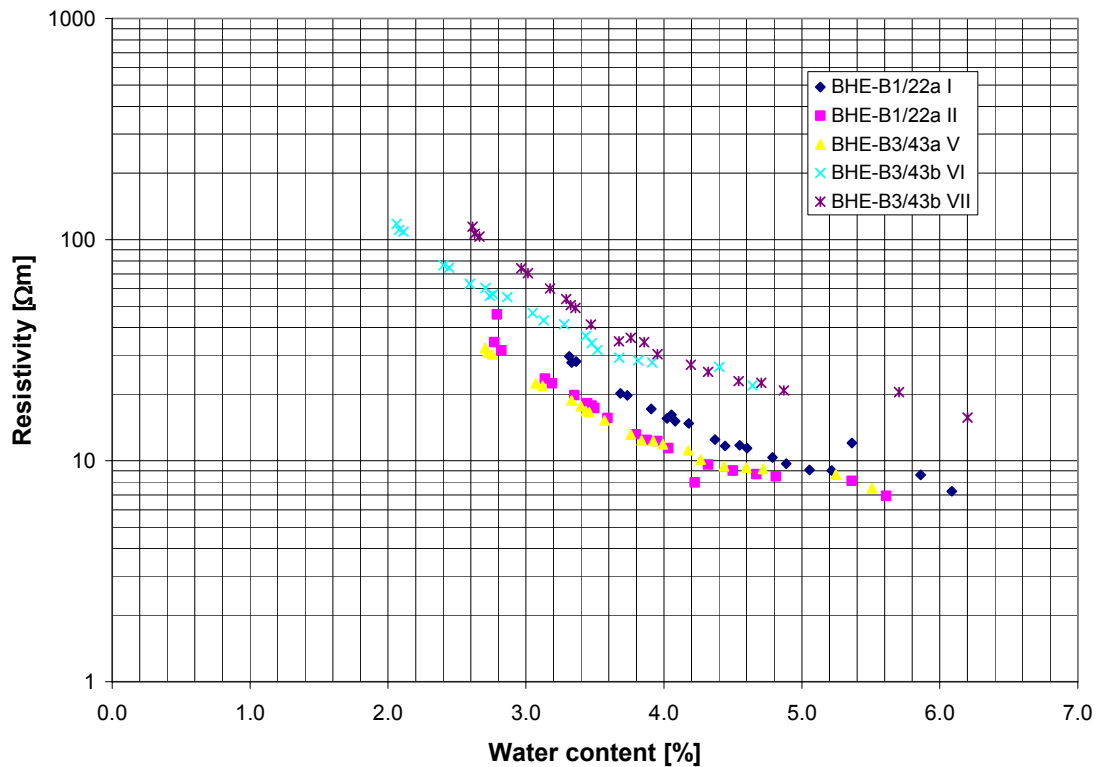
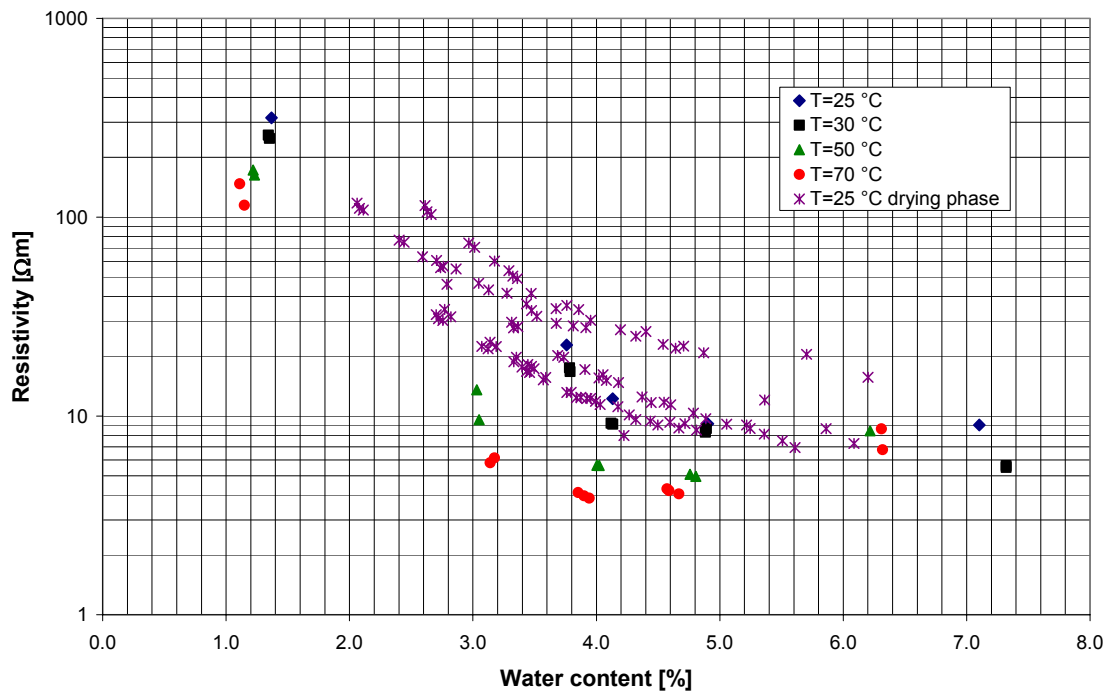**Figure 5-16** Results of the resistivity measurements at the state of delivery without additional saturation
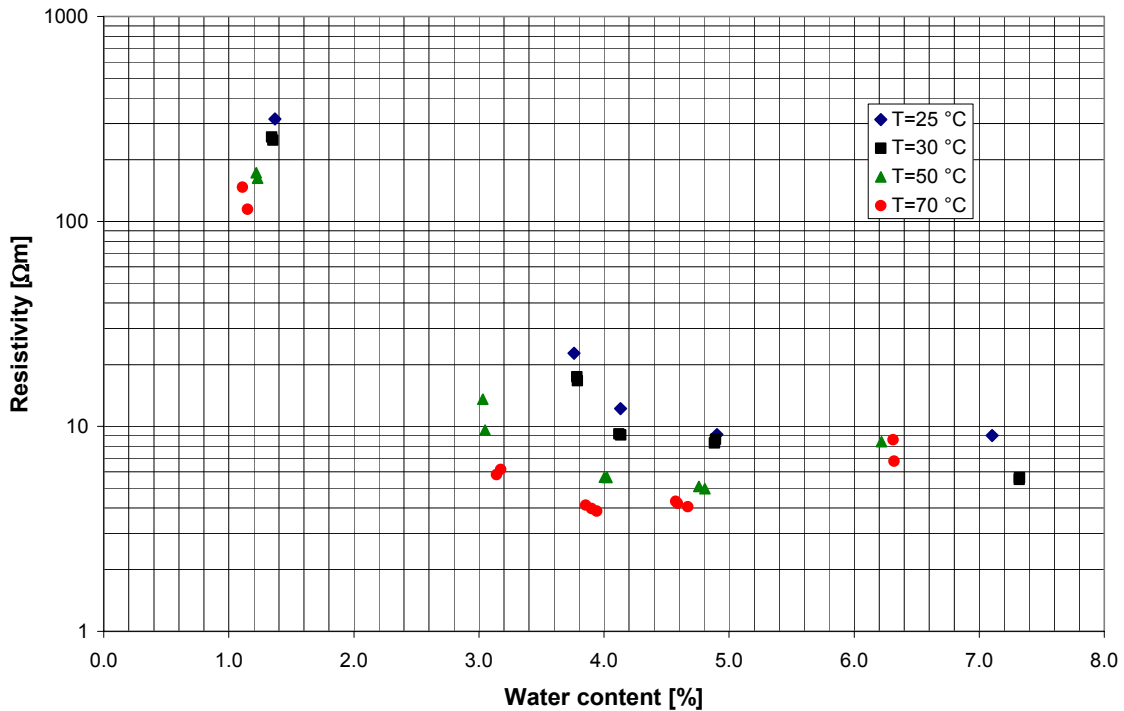
Figure 5-17   Results of the resistivity measurements at different temperatures in dependence of the water content after saturation of the samples

All four resistivity curves show a decrease in resistivity with increasing water content. The resistivities measured at 25 °C range between 315.9 Ωm and 9 Ωm at water contents of 1.37 wt% and 7.1 wt%. The resistivities measured at 30 °C are a little lower and show values between 258.3 Ωm and 5.5 Ωm at comparable water contents. At 50 °C the resistivities vary between 172.9 Ωm and 8.4 Ωm. The resistivities at 70 C are the lowest and show values between 147 Ωm and 6.7 Ωm. The water contents of about 1.1 wt% and 6.3 wt% at 50 °C and 70 °C are comparable.

The resistivities at 25 C and 30 °C are not significantly different, because the temperature difference is too low. The decrease of the resistivities at higher temperatures is explained by the temperature dependence of the pore solution. The conductivity of most electrolyte solutions increases with increasing temperature. Still, a temperature influence of the resistivity of the clay rock material is also possible.

The resistivity curves at 50 °C and 70 °C show that the resistivities at a water content of approx. 6.2 wt% are about two times higher than at 4.6 wt%. An explanation of this result might be a mistake at the measurements caused for instance by evaporation of

water at the surface of the samples. Possibly, this effect is more pronounced at higher water contents. Furthermore, the small difference of two times is in the tolerance of the resistivity measurement. For a better interpretation, appropriate investigations would be necessary.

The combination of the measurements at state of delivery and at the various resaturation steps is shown in Figure 5-18. In spite of the different procedures, the results are quite comparable. Obviously, the saturation had no influence on the behaviour of the samples. The measurements at comparable water contents were repeatable.

Figure 5-18   Combination of the results of resistivity measurements at different temperatures in dependence of the water content after saturation of the samples, and at the state of delivery without additional saturation

5.2.1.3   Density and porosity

The grain density of the BHE clay was determined to 2.75 g/cm^3. The mean bulk densities of the BHE-samples ranged from 2.24 g/cm^3 to 2.29 g/cm^3 (average: 2.26 g/cm^3). The resulting porosities were 16.7 % to 18.5 % (average: 17.9 %). The determined bulk densities and porosities are summarized in Table 5-5.

Table 5-5 Bulk densities, porosities, and water contents of the investigated BHE samples; grain density: 2.75 g/cm^3

Sample	Bulk density (dried at 105 °C) [g/cm³]	Porosity [%]	Water content at state of delivery [%]
BHE-B1/22a I	2.24	18.5	6.09
BHE-B1/22a II	2.24	18.5	5.61
BHE-B3/43a V	2.24	18.5	5.51
BHE-B3/43b VI	2.27	17.5	4.64
BHE-B3/43b VII	2.29	16.7	6.20

The grain density reported in /HOH 98/ is about 2.64 g/cm^3 (carbonate-rich facies). The bulk densities range between 2.47 g/cm^3 to 2.55 g/cm^3 (carbonate-rich facies) and 2.37 g/cm^3 to 2.33 g/cm^3 (shaly facies), respectively. Porosity values from 12.25 % to 17.1 % (shaly facies) were observed. For the measurements, different kinds of preparation were described. The water content of the samples was determined by drying at 105 °C and ranges between 6.6 % and 7.5 % for the shaly facies.

The values in the present report are in good agreement with those described in /HOH 98/. Differences might be explained by different methods of measurements and preparation as well as by different facies.

5.2.2 Post test investigations

After switching off the electrical heaters in August 2003, the whole test field was dismantled /GAR 03/ and core samples for further laboratory investigations were drilled by BGR in order to determine the alteration of the Opalinus clay as a result of heating (see chapter 4.2.5).

5.2.2.1 Gas release measurements

Within the post test investigation programme, Opalinus clay samples were investigated with regard to gas generation and gas release from the heated area of the test field. Therefore samples were taken from the contact zone between host rock and backfill of the heater borehole which had experienced a maximal temperature of about 50 °C, from different levels of borehole BHE-26 with a maximal temperature of about 30 °C,

and from borehole BHE-27 with a maximal temperature of 20 °C. Right after drilling or mining, the samples were emplaced in gas-tight stainless steel or glass containers with a low residual volume filled with mine air (see Section 4.2.4). The containers were then transported to the GRS laboratory in Braunschweig where they were stored for about four weeks at room temperature (about 22 °C). Afterwards the gas compositions in the residual volumes were extracted in order to determine the gas release from the Opalinus clay at low temperature, but in contact with air. The results of the gas compositions (C_1-C_4 and CO_2) are compiled in Figure 5-19.

Again, carbon dioxide is the most important gas with concentrations in the residual volume of the containers between 2 000 and 20 000 vpm (0.2 to 2.0 vol%), which correspondents to a released amount of 10^{-3} and 10^{-2} m^3 per 1.000 kg clay.

The concentrations of the hydrocarbons ranged at:

- methane between 100 and 10 000 vpm; corresponds to10-3 m3 per 1.000 kg

- ethane between 35 to 4100 vpm; corresponds to10-3 m3 per 1.000 kg

- propane below 10 vpm; corresponds to10-6 m3 per 1.000 kg

- iso-butane between 500 and 1 700 vpm; corresponds to10-4 m3 per 1.000 kg

The concentrations of hydrogen and hydrogen sulphide in the containers were always below the detection limit of 30 vpm.

The concentration of helium varied between 10 and 205 vpm which correspondents to a released amount of 10^{-5} and 10^{-4} m^3 per 1.000 kg clay.

The concentration of oxygen in the residual volumes of the container decreased from 20 vol% (mine air) to a concentration below 5 vol%. This means that up to 4 vol% oxygen was consumed by oxidation of the hydrocarbons generating up to 2 vol% carbon dioxide and up to 15 vol% was adsorbed to the internal surface of the clay samples.
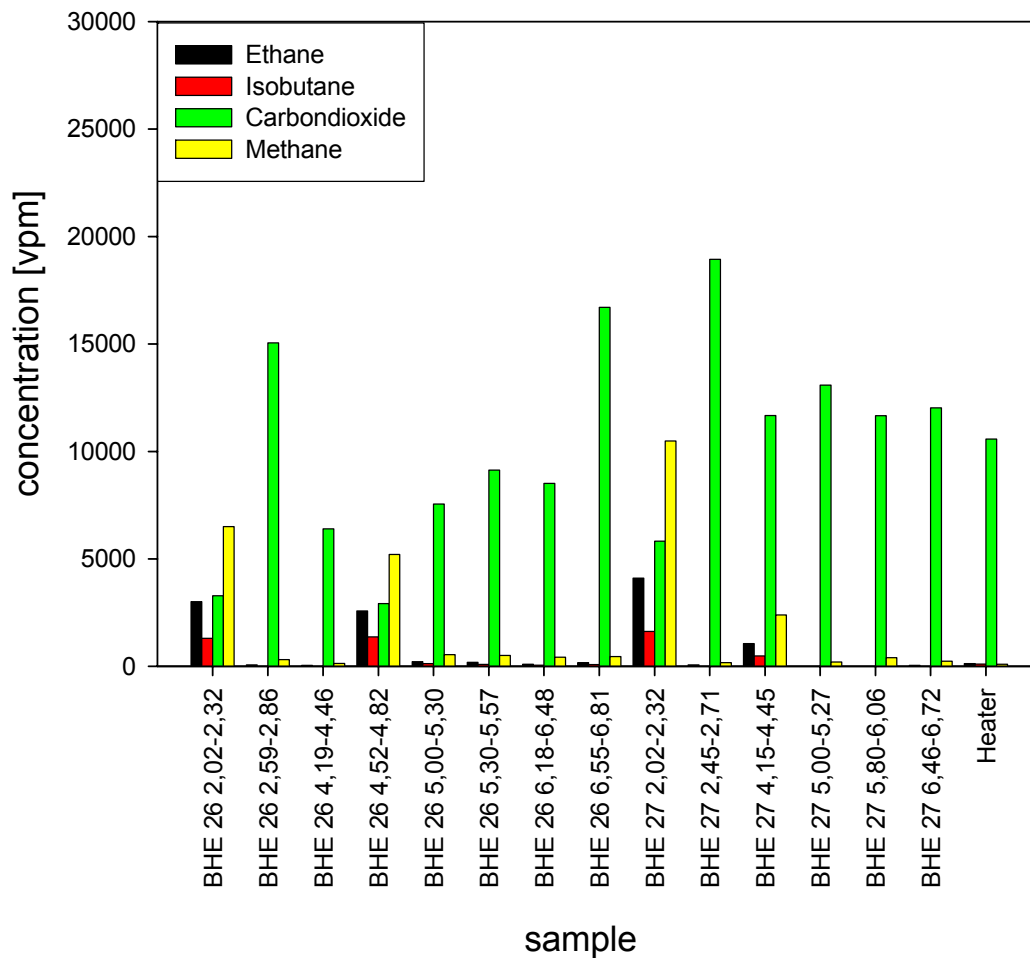
Figure 5-19   Results of gas concentrations in the containers with the samples of different levels from boreholes BHE-26 and BHE-27 and from the contact zone host rock-backfill of the heater borehole (heater)

The concentration of the most important components carbon dioxide and methane reached equilibrium in the residual volume of the containers, as the amounts of the samples were comparatively high and the volumes were comparatively low. Therefore, the specific amounts of the released gases were comparatively low. Further exchange of the gases in the residual volume of the containers would lead to further gas release.

The investigation on gas release at room temperature showed no obvious heat-induced alteration of the samples from the different areas of the test field. Furthermore, this investigation indicated the heterogeneity of the Opalinus clay with regard to the gas content.

After the investigations at room temperature the gas release at 95 °C was determined in the glass ampoules as a function of time. Additionally, the gas composition in the residual volume and the amount of the exposed material were varied.

As an example, Figure 5-20 shows the results on the release of carbon dioxide for the samples:

- Borehole BHE-27, depth 2.45-2.71 m, with a maximal temperate during the in-situ test of 20 °C

- Borehole BHE-26, depth 2.59-2.86 m, with a maximal temperate during the in-situ test of 30 °C

- Contact zone heater borehole/host rock, with a maximal temperate during the in-situ test of 50 °C
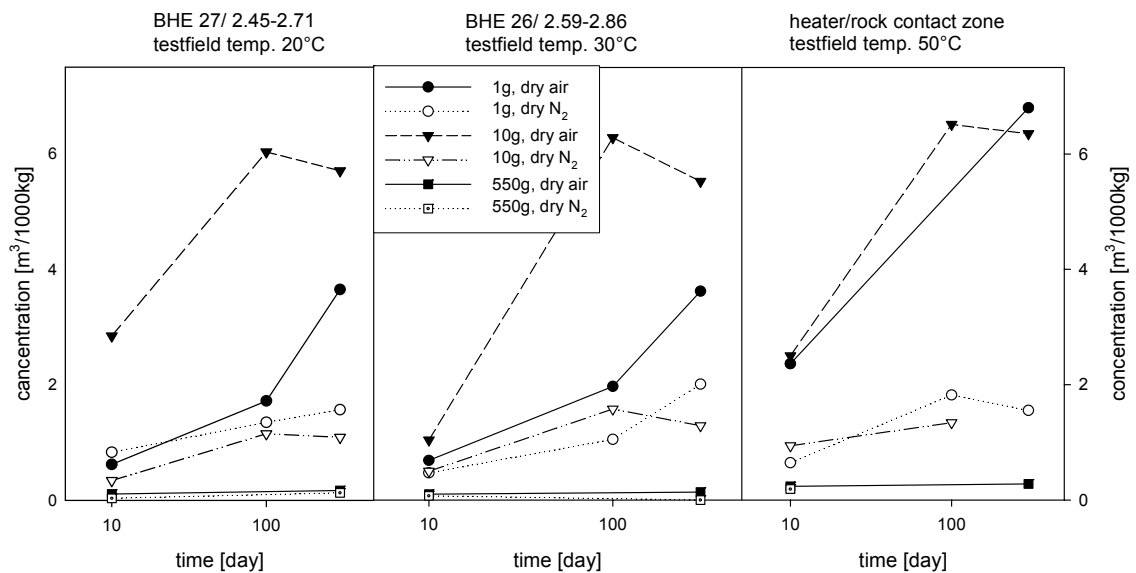
Figure 5-20   Release of carbon dioxide from the Opalinus clay from different areas of the test field at 95 °C
- BHE-26 / depth 2.59-2.86m
- BHE-27 / depth 2.45-2.71m
- contact zone heater borehole/host rock.
- exposure time 10, 100, and 300 days
- amount of clay in the ampoules 1, 10, and 550 grams
- with air or nitrogen in the residual volume of the glass ampoules.

Again, no significant alterations of the Opalinus clay as a result of heating up to 50 °C for 18 months were detected. The results agree well to those described in Section 5.2.1. The specific amount of the gases released into the residual volume of the ampoules for 1 gram is again lower than for 10 grams, for the concentrations of equilibrium in the residual volume were not reached and gases were adsorbed to the inner surface of the ampoules. The absolute amount of that adsorption is almost the same but the relative amount for the ampoules with 1 gram is higher than for those with 10 grams. The ampoules with 550 grams of clay have a low specific amount of released gases, for the residual volumes were very small and the equilibrium of the gas concentrations for carbon dioxide was reached.

5.2.2.2 Mineralogical compositon of the Opalinus clay

The phase analysis of the Opalinus clay as received from the test field indicates the presence of quartz, kaolinite as well as muscovite. Carbonates are fixed in magnesium calcite or calcium carbonate. The carbonates are considered to be important minerals in geological barrier concerning the influence of the geochemical milieu, especially the pH. In Figure 5-21 the spectrum of sample BHE-26/2.59 – 2.86 m is depicted. In Table 5-6 the determined mineral phases, formulas are given.

Table 5-6 Mineralogical components of the Opalinus clay sample BHE-26/ 6.55-6.81 as received from the test field

PDF-No.	Mineral	Formula
86-1628	Quartz low	SiO_2
89-1304	Magnesium calcite	$(Mg_{0.03} Ca_{0.97})(CO_3)$
29-1488	Kaolinite	$Al_2Si_2O_5 (OH)_4$
01-1098	Muscovite	$H_2KAl_3(SiO_4)_3$
26-0801	Pyrite	FeS_2
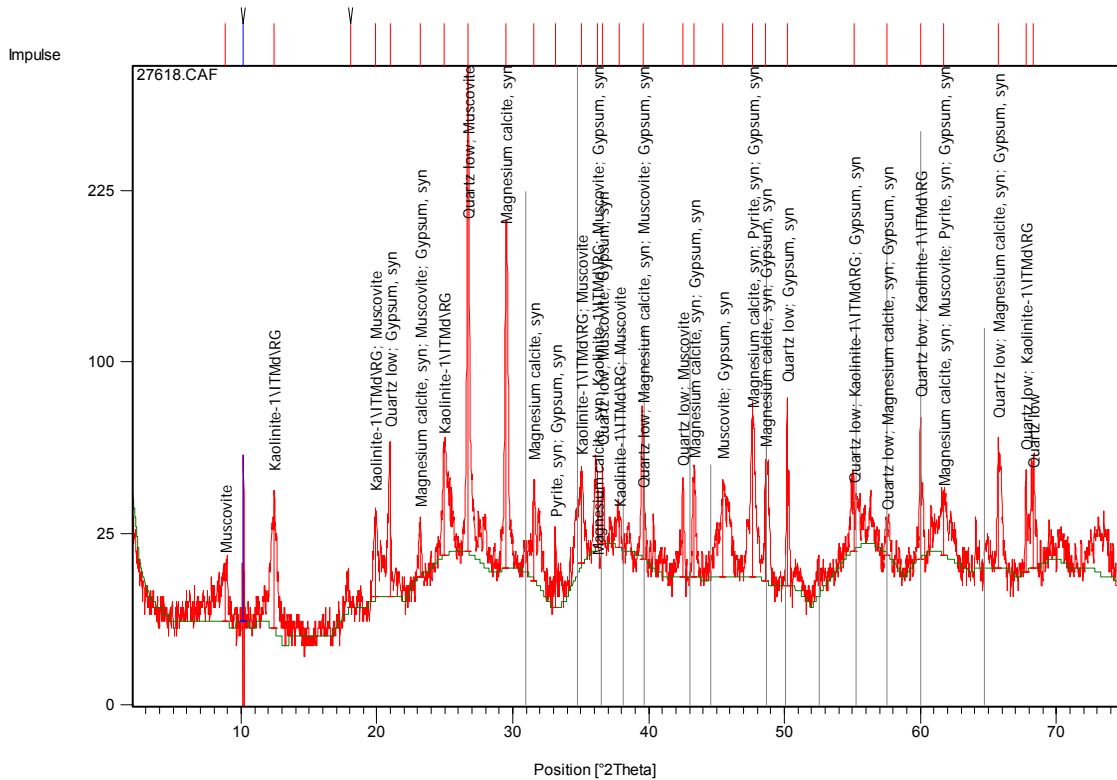
Figure 5-21 X-ray diffractometer spectrum of Opalinus clay sample BHE-26/ 2.59-2.86 as received from the test field

The results are in good agreement with the results given by BGR for boreholes B1 – B3 /KAU 03/. Some of the minerals obtained by BGR, e.g., chlorite and siderite, could not be detected in BHE-26 and BHE-27.

The Opalinus clay exposed to 95 °C for 300 days showed the same mineralogical composition qualitatively. This fact indicated that within that time period at 95 °C not detectable alteration of the clay took place.

With regard to the carbon content 8 samples of Opalinus clay from the boreholes BHE-26 and BHE-27 were investigated. The samples as delivered from the test field had an inorganic carbon content (TIC) of 1.7 wt%, an organic carbon content (TOC) of 1.1, and a total carbon content (TC) of 2.8 wt%. Similar samples exposed to 95 °C in the glass ampoules for 300 days with air in the residual volume have had significant lower carbon contents. The corresponding values are 1.5 wt% of inorganic, 1.0 of organic, and 2.5 of total carbon content. In Table 5-7 these carbon contents with the standard variation and the contents of similar unheated samples analysed by BGR /KAU 03/ are compiled. Both investigations are in comparatively good agreement.

Table 5-7 Carbon content of the Opalinus clay as delivered from the test field and after the exposure for 300 days at 95 °C within air atmosphere (average values of 8 samples)

	as delivered [wt.-%]	after exposure [wt.-%]	average values of BGR [wt.-%]
inorganic carbon (TIC)	1.7 ± 0.2	1.5 ± 0.1	1.9
organic carbon (TOC)	1.1 ± 0.1	1.0 ± 0.1	0.9
total carbon (TC)	2.8 ± 0.3	2.5 ± 0.3	2.8

As a result of heating, about 0.3 wt% of the total carbon content is transferred. This is equivalent to 3 kg per 1.000 kg clay. With the assumption that all these carbon is transferred to carbon dioxide, 5.6 m³ per 1.000 kg clay are generated. This value is in the range of the carbon dioxide generation in the glass ampoules after heating for 842 days at 95 °C with aerobic conditions described in Section 5.2.1 and in this section. The corresponding values are 3.8 and 6 to 7 m³ per 1.000 kg clay. Additionally, it has to be taken into account that the gases like carbon dioxide and hydro carbons are primarily present in the Opalinus clay adsorbed to internal surfaces.

5.2.2.3   Resistivity measurements

The results of the resistivity measurements on unheated and heated Opalinus clay samples are summarized in Table 5-8 and plotted in Figure 5.22. For comparison, the resistivities of samples taken from the boreholes BHE-B1/22a, BHE-B3/43a, and BHE-B3/43b before starting the heater experiment are shown as well.

The results show that the resistivities before and after heating are not significantly different. The slightly lower water content of the samples obtained prior to heating might be explained by the different duration of storage before the measurements (the pre-test samples had not been tested directly after drilling, but had been taken to the GRS laboratory first). The somewhat higher resistivity of the post–test samples might be an effect of drilling or of cooling of the clay rock; these effects cannot be separated.

Table 5-8   Resitivities of Mont Terri HE samples, before heating and after heating

Sample	Water content [wt.%]	Resistivity [Ωm]
Before heating		
BHE-B1/22a I	6.09	7.3
BHE-B1/22a II	5.61	6.9
BHE-B3/43a V	5.51	7.5
BHE-B3/43b VI	4.64	31.9
BHE-B3/34b VII	6.20	15.7
After heating		
BHE-27 I	7.01	18.7
BHE-27 II	7.10	33.7
BHE-27 III	7.20	25.0
BHE-27 IV	6.29	19.5
BHE-26 V	7.61	17.2
BHE-26 VI	7.19	15.7
BHE-26 VII	7.20	18.5
BHE-26 VIII	7.53	14.8
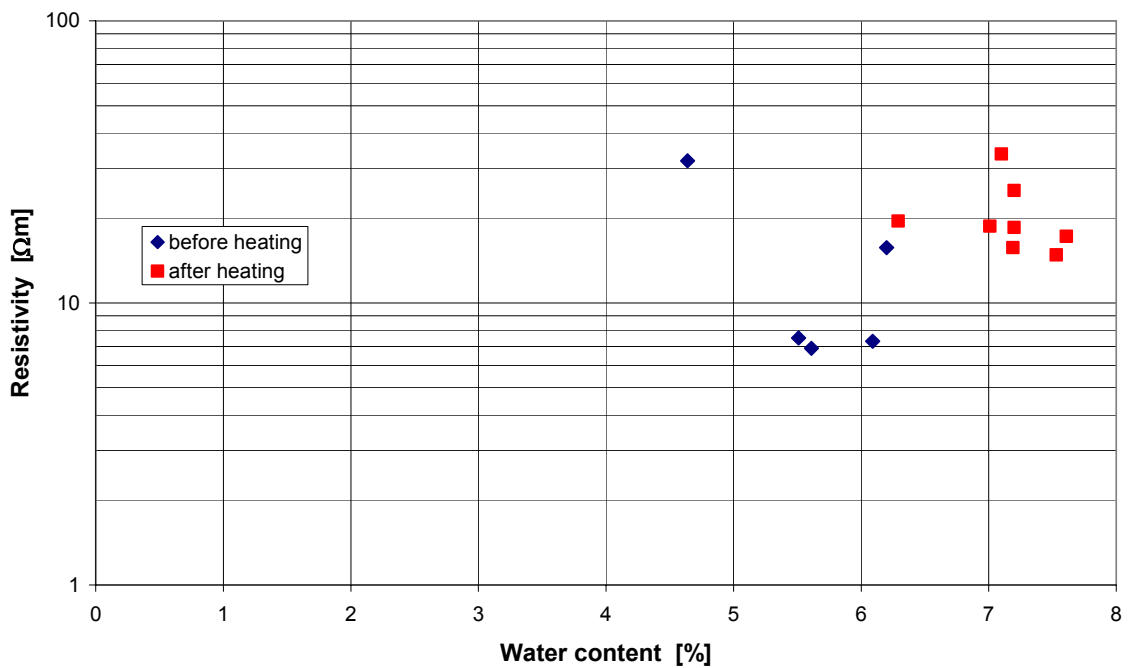
Figure 5-22   Resistivity measurement results of samples obtained before and after heating

5.2.2.4 Dismantling of packers from the boreholes BHE-10 and BHE-11

For dismantling of the HE-B test a vertical shaft with a diameter of 3.00 m and depth of 7.00 m was build in order to retrieve the heater tube, the bentonite buffer of the central borehole with the contact zone to the Opalinus clay, and the sensors installed in the vicinity to the central borehole. During these actions the packers installed in boreholes BHE-10 and BHE-11 with their stainless steel capillaries were also retrieved.

Figure 5-23 shows these packers. An optical inspection indicated that all components of the packer such as capillaries, fittings, welding seams, screws, and the main body itself did not have any corrosion. Even the rubber jacket did not show any visible alteration.

This fact indicates that measuring devices build of stainless steel (V4A) can be used for installation in test fields similar to the HE-B test.

Figure 5-23 Dismantled packers from the boreholes BHE-10 and BHE-11

6 Summary and conclusions

Beside salt and granite, clay formations are investigated as potential host rocks for disposing radioactive waste. In Switzerland in the canton Jura close to the city of St. Ursanne, an underground laboratory was built in the vicinity of the reconnaissance gallery of a motorway tunnel. Since 1995, a consortium of 12 international organisations is running this laboratory for investigating the suitability of the Opalinus clay formation with regard to disposal of radioactive waste.

From 1999 to 2005, the Heater Experiment B (HE-B) was conducted for investigating the coupled thermo-hydro-mechanical (THM) processes of the Opalinus clay in interaction with the bentonite buffer. The Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) participated in that experiment for determining the subjects of gas generation, gas release, water content, and water redistribution in the Opalinus clay during heating. This report deals with the work of GRS performed in this project during the years 1999 to 2005.

The experiment was carried out in a separate niche 5 m wide and 7 m long parallel to the striking with a dip of the bedding of 40° in the shaly facies of the Opalinus clay. The central vertical heater borehole with a diameter of 300 mm and a depth of 7.5 m was drilled into the floor of the niche. Between the heater tube and the host rock, ring shaped bentonite buffer blocks were installed. The gap between the buffer and the host rock as well as the top of the borehole were backfilled with sand. For flooding the buffer with water, capillaries ran into the sand filled gap. At the top the borehole was sealed gas-tight with resin and cement. For determining the relevant parameters of the host rock, such as temperature, total pressure, pore pressure, permeability, electric resistivity, and gas release, 19 additional boreholes were drilled into the floor of the niche. Installation of the test field was performed in 1999. For determination of the gas release from the host rock, four boreholes with a distance to the heater between 0.5 and 3.64 m were sealed with gas-tight packers. Capillaries from the open gallery ran into the residual volumes of these boreholes for extracting gas and water for analyses. The gas samples were sent to GRS laboratory or were analysed on site with a portable gas chromatograph. In another four boreholes close to the heater, electrode chains for geoelectric tomography measurements were installed. The geoelectric monitoring system consisted of an automatic computer-controlled measurement and data acquisition unit.

Hydration of the buffer started in October 1999. Heating with 650 Watts started in February 2002 and lasted 18 months until September 2003. The maximal temperature in the bentonite buffer blocks reached 68 °C. After switching off the heater, cores were drilled from the host rock and the whole test field was dismantled for post-test laboratory investigation.

Additionally to the investigations in the test field, laboratory measurements on gas generation and release and on electric resistivity were performed on Opalinus clay samples obtained during installation (prior to heating) as well as on samples with the influence of heating obtained after heater shut-down.

At the early beginning the residual volumes of the boreholes for determining the gas release were filled with mine air. The results of these analyses indicated that at mine temperature (about 14 °C) carbon dioxide, some helium, and hydrocarbons are released. As a result of oxidation of organic material in the clay the concentration of oxygen decreases from 20% to a few percent. The concentration of the gas components varies significantly up to a factor of 15 in the different boreholes which have a distance of 0.35 m. This fact indicates that the Opalinus clay which seems to be homogeneous macroscopically is not homogenous regarding the gas content. Significant influence of heating was only seen in the borehole which was close to the heater. The concentration of carbon dioxide increased from 814 vpm (ml/m^3) to 36000 vpm (3.6 vol%). The hydrocarbons increased from 234 to 3303 vpm and helium from 21 to 114 vpm. Hydrogen sulphide (H_2S) which is generated after heating was found in the borehole close to the heater up to 680 vpm and in the other boreholes less than 12 vpm. This gas component is of importance concerning the disposal of radioactive waste even at low concentrations as it leads to corrosion of the waste and the waste container in combination with water in the liquid or gaseous phase.

In the beginning right after sealing the boreholes (May 1999), the residual volume of the boreholes was filled with mine air. During the pre-heating phase, carbon dioxide, some helium, and hydrocarbons were released at mine temperature (about 14 °C). As a result of oxidation of organic material in the clay, the concentration of oxygen decreased from 20% to a few percent. The concentration of the gas components varied significantly up to a factor of 15 in the different boreholes which are only 0.35 m apart. This fact indicates that the Opalinus clay which seems to be homogeneous macroscopically is not homogenous regarding the gas content.

Significant influence of heating is only seen in the borehole which was closest to the heater. The concentration of carbon dioxide increased from 814 vpm (ml/m^3) to 36000 vpm (3.6 vol%). The hydrocarbons increased from 234 to 3303 vpm and helium from 21 to 114 vpm. Hydrogen sulphide (H_2S) which was generated after heating was found in the borehole close to the heater up to 680 vpm and in the other boreholes with less than 12 vpm. This gas component is of importance concerning the disposal of radioactive waste even at low concentrations, as it leads to corrosion of the waste and the waste container in combination with water in the liquid or gaseous phase.

After one year of heating, the concentrations of the components carbon dioxide, helium and hydrocarbons showed a slight decrease and oxygen showed a slight increase. No significant pressure increases in the boreholes were observed as a result of gas release. These facts indicate that the host rock is not absolutely gas-tight.

At the end of heating and before dismantling of the test, up to 2.2 litres of water were extracted out of the gas monitoring boreholes. The amounts of the water extracted and the analysis of this water indicated that the host rock had a relevant permeability to water. A significant dilution of the anions and kations was observed in the borehole close to the heater. This fact indicates that ions of the synthetic formation water were adsorbed by the bentonite buffer blocks surrounding the heater and that no resaturation took place during migration of that water to the borehole close to the heater.

The laboratory investigation on gas generation and release from the Opalinus clay indicated again that carbon dioxide is the most important gas component. It is most likely that carbon dioxide is generated by oxidation of organic material in the clay, as its amount is much higher if the clay had contact with air than in contact with an inert gas (nitrogen). Additionally, carbon dioxide may be generated by thermal decomposition of carbonates in the clay. Furthermore, carbon dioxide in primarily present in the clay adsorbed to the internal surfaces. It will be released if the equilibrium is disturbed by mining or by elevated temperature. It can be concluded that the concentration of equilibrium for carbon dioxide in the pore volume will be in the range of 2 to 3 vol% which agrees with the in situ measurements. If the host rock is gas-tight, only a small amount of organic matter will be oxidised or decomposed. If the host rock is not gas-tight and the released carbon dioxide could migrate into residual volumes of the repository, all the organic material will be oxidised if oxygen is available. Also the thermally instable carbonates will be decomposed. It should be taken into account that

up to 10 m³ carbon dioxide per 1.000 kilograms of clay may be released. Hydrocarbons are of minor importance as their concentrations of equilibrium are below 1 vol%, that means below an ignitable concentration.

The geoelectric tomography measurements make use of the relation between electric resistivity and solution content of the rock. For the interpretation of the in-situ measurements, calibration measurements on samples with varying saturation were performed at different temperatures. The geoelectric measurements gave no indications of heat-induced desaturation of the clay rock until end of the heating period. The only effect detected was a reduction of resistivity due to temperature increase, which is mainly caused by the higher conductivity of the pore solution at higher temperature. Post-test investigations regarding saturation and resistivity performed on samples obtained from the test field after heater shut-down confirmed this result.

The in-situ and laboratory investigation indicate that the Opalinus clay is neither water- nor gas-tight. For long-term safety aspects it has to be taken into account that generated and released gases can migrate into open areas of the repository and water can migrate within the Opalinus clay formation. Within the heating period of 18 months up to 50 °C, no significant alteration of the Opalinus clay with regard to gas and water content or mineralogical composition was observed. For much longer time periods within the temperature range up to 90 °C alteration cannot be excluded.

7 References

/EMM 02/ Emmerich, K.; Jockwer, N.; Huertas, F.; Alonso, E.; García-Siñeríz, J.L.; Kennedy, K.; Ploetze, M.; Croise, J.; Ammon, Ch.: Test Plan (D1) Heater Experiment (HE) – Rock and bentonite thermo-hydro-mechanical (THM) processes in the near field (2002)

/EN 97/ Europäische Norm EN 1484 H3: Anleitung zur Bestimmung des gesamten organischen Kohlenstoffs (TOC) und des gelösten organischen Kohlenstoffs (DOC)

/FEC 01/ Fechner, T. (2001): SensInv2D-Manual, Geotomographie, Neuwied

/GAR 03/ García-Siñeríz, J.L.; ReyMazon, M.: Dismantling Operation Plan (D16) Heater Experiment (HE) – Rock and bentonite thermo-hydro-mechanical (THM) processes in the near field (2003)

/GOE 04/ Göbel, I.; Alheid, H.J.; Alonso, E.; Ammon, Ch.; García-Siñeríz, J.L..; Jockwer, N.; Klubertanz, G.; Mayor, J.C.; Ploetze, M.; Weber, H.P.: Heater Experiment (HE) Final report to European Commission Contract No: FICK-CT-2001-00132, to be printed

/HAR 96/ HarbourDom GmbH: "RESITOMO – Programmpaket zur 2.5D Modellierung und 2D Inversion von DC-Widerstandsdaten, HarbourDom Consulting – Geophysikalische Forschungs- und Beratungsgesellschaft mbH, Köln, 1996

/HOH 98/ Hohner, M.; Bossart, P. (1998): Mont Terri Project: Geological, geochemical, geomechanical and hydraulic parameters of Opalinus Clay derived by in-situ and laboratory experiments (Phase 1, 2 and partly 3)- Technical Note 98-49, April 1998

/JOC 01-1/ Jockwer, N.: Technical Note 2001-4, July 2001, Geotechnisches Institut Bern

/JOC 01-2/ Jockwer, N.; Wieczorek, K.: Technical Note 2001-24, July 2001, Geotechnisches Institut Bern

/JOC 02/   Jockwer, N.; Wieczorek, K.: Untersuchungen zur Gasfreisetzung und Wasserumverteilung im OPALINUS-Ton des Mont Terri, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, GRS – 181, ISBN 3-931995-49-6

/KAU 03/   Kaufhold, S.; Dohrmann, R.; Bitzer, C.; Pletsch, T.; Klosa, D.: Laboratory test results (D6/2a) Heater Experiment (HE) – Rock and bentonite thermo-hydro-mechanical (THM) processes in the near field (2003)

/PEA 03/   Pearson, F.J.; Arcos, D.; Bath, A.; Boisson, J. Y.; Fernández, A.Mª; Gäbler, H-E.; Gaucher, E.; Gautschi, A.; Griffault, L.; Hernán, P.; Waber, H.N.: Geochemistry of water in the Opalinus Clay Formation at the Mont Terri Rock Laboratory. Swiss Federal Office for Water and Geology Series, Nº 5, (2003)

/TEL 90/   Telford, E. M.; Geldart, L. P.; Sheriff, R. E. (1990): Applied Geophysics. - Second Edition. Cambridge University Press.

/THU 99/   Thury, M.; Bossart, P.: Mont Terri Rock Laboratory, Result of the Hydro-geological, Geochemical and Geotechnical Experiments Performed in 1996 and 1997, Bundesamt für Wasser und Geologie, Geologische Berichte Nr. 23, Bern 1999

8   List of Figures

9   List of Tables

10 Annex: Tables

In this annex, the measured data of gas composition in the investigation boreholes BHE-10, BHE-11, BHE-12, BHE-13 and in the air of the HE-niche are given.

Table 10-1 Date of Sampling: 14.08.2001

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	$i\text{-}C_4H_{10}$ [vpm]	H_2S [vpm]		
BHE-10	6.2	37	980	200	26	8	n.d.	n.a.	10.2	14
BHE-11	1.5	480	13900	4990	625	575	140	n.a.	11.1	14
BHE-12	1.3	540	13100	4710	590	550	130	n.a.	11.5	14
BHE-13	2.6	370	2620	6150	880	900	230	n.a.	11.3	14
Air HE-niche	21.6	20	480	n.d.	n.d.	n.d.	n.d.	n.a.	10.3	14

Table 10-2 Date of Sampling: 19.02.2002

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	$i\text{-}C_4H_{10}$ [vpm]	H_2S [vpm]		
BHE-10	1.9	21	814	271	31	9	n.d.	n.a.	7.9	13
BHE-11	3.7	371	11564	4885	624	562	133	n.a.	8.2	13
BHE-12	2.1	541	11614	5171	633	575	136	n.a.	8.3	13
BHE-13	2.9	414	2484	6663	915	906	229	n.a.	8.5	13
Air HE-niche	22.0	n.b.	431	n.d.	n.d.	n.d.	n.d.	n.a.	6.2	13

Table 10-3 Date of Sampling: 16.05.2002

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	$i\text{-}C_4H_{10}$ [vpm]	H_2S [vpm]		
BHE-10	1.1	25	1071	305	34	8	n.d.	n.a.	11.3	15.0
BHE-11	10.2	68	9335	2915	458	454	113	n.a.	12,1	15.0
BHE-12	3.0	409	10605	4478	548	500	119	n.a.	12.3	15.0
BHE-13	5.0	340	2182	5158	704	709	182	n.a.	10.3	15.0
Air HE-niche	21.5	n.d.	803	7	n.d.	n.d.	n.d.	n.a.	9.0	15.0

Table 10-4 Date of Sampling: 02.09.2002

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	i-C_4H_{10} [vpm]	H_2S [vpm		
BHE-10	1.77	57	2450	1450	165	123	38.	49	11.1	14.9
BHE-11	1.17	550	15350	6000	775	710	1653	n.d.	11.9	14.9
BHE-12	3.78	280	10660	3500	440	405	97	n.d.	11.6	14.9
BHE-13	no determination								11.0	14.9
Air HE-niche	21.77	n.d.	500	n.d.	n.d.	n.d.	n.d.	n.d.	9.2	14.9

Table 10-5 Date of Sampling: 18.11.2002

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	i-C_4H_{10} [vpm]	H_2S [vpm		
BHE-10	0.52	75	6295	1490	170	105	35	297	8.2	12.9
BHE-11	0.77	510	14460	5620	700	620	140	<5	9.3	12.9
BHE-12	no determination								8.9	12.9
BHE-13	10.1	90	2100	3800	595	630	175	<5	8.5	12.9
Air HE-niche	21.7	n.d.	495	15	<5	<5	<5	n.d.	6.4	12.9

Table 10-6 Date of Sampling: 18.02.2003

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	i-C_4H_{10} [vpm]	H_2S [vpm		
BHE-10	1.0	114	36000	2730	363	148	62	680	10.5	12.5
BHE-11	7.6	177	9690	3340	474	440	102	<5	10.0	12.5
BHE-12	1.5	360	10210	3545	412	338	74	<5	9.7	12.5
BHE-13	1.3	477	3100	7055	898	870	225	<5	9.6	12.5
Air HE-niche	21.7	26.	530	15	<5	<5	<5	n.d.	6.6	12.5

Table 10-7 Date of Sampling: 08.05.2003

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	i-C_4H_{10} [vpm]	H_2S [vpm		
BHE-10	1.8	114	28300	2160	302	132	58	530	10.9	15
BHE-11	1.5	345	12640	4325	549	470	104	<5	12.2	15
BHE-12	1.3	335	10020	3500	411	328	69	n.d.	12.2	15
BHE-13	1.8	369	3150	5260	665	636	161	<5	11.7	15
Air HE-niche	21.8	<15	525	542	<5	<5	<5	n.d.	10.9	15

Table 10-8 Date of Sampling: 03.07.2003

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	i-C_4H_{10} [vpm]	H_2S [vpm		
BHE-10	2.28	80	19380	1790	250	118	50	103	9.9	17
BHE-11	1.22	308	10160	3700	450	380	80	n.d.	13.5	17
BHE-12	1.49	230	8460	2685	311	240	49	12	13.2	17
BHE-13	2.1	300	2980	4330	540	500	125	<5	12.6	17
Air HE-niche	21.7	n.d.	620	<15	<5	<5	<5	n.d.	10.9	17

Table 10-9 Date of Sampling: 19.08.2003

| Borehole | Gas Component | | | | | | | | Absolute Humidity [g/m³] | Temp. HE-niche [°C] |
	O_2 [Vol %]	He [vpm]	CO_2 [vpm]	CH_4 [vpm]	C_2H_6 [vpm]	C_3H_8 [vpm]	i-C_4H_{10} [vpm]	H_2S [vpm		
BHE-10	1.0	80	26360	2340	n.a.	235	40	<5	11.2	15
BHE-11	0.4	320	12240	5500	n.a.	480	75	n.d.	12.4	15
BHE-12	1.4	245	9360	3790	n.a.	315	37	n.d.	11.3	15
BHE-13	0.6	280	2960	4800	n.a.	510	107	n.b.	10.7	15
Air HE-niche	20.1	<10	510	85	n.a.	n.d.	n.b.	n.d.	13.4	15

n.a. = not analysed
n.d. = not detectable

75