# Gas Migration In The Opalinus Clay As A Function Of The Gas Pressure - 8169

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## ABSTRACT

Clay formations have long been proposed as potential host rocks for nuclear waste disposal. After the operational phase of a repository the openings, e.g., boreholes, galleries, and chambers, have to be backfilled in order to avoid the release of radionuclides into the biosphere. After healing and resaturation of the excavation disturbed zone (EDZ) and saturation of the backfill, the waste containers and the metallic components will corrode resulting in a generation of hydrogen. Additionally, carbon dioxide will be released as a result of oxidation and thermal decomposition of the organic components in the waste and in the clay. If the disposal boreholes and chambers are sealed gas-tight, high gas pressure may be produced leading to the potential generation of fractures in the host rock which could influence the integrity of the repository. Therefore it is essential that the gases migrate through the technical barriers (backfill) or into the surrounding host rock at lower pressure and without any irreversible damage of the repository.

In order to estimate the consequences of the gas generation the knowledge of the gas paths in the host rock and the knowledge of the parameters which influence the gas migration in the host rock are important.

During an ongoing project at the underground research laboratory Mt. Terri in Switzerland the gas migration in the undisturbed over-consolidated Opalinus Clay is investigated.

## INTRODUCTION

Clay formations are being considered worldwide as host medium for the disposal of radioactive waste. In order to prove the feasibility of the disposal concepts proposed by the countries like for instance France, Switzerland, and Belgium, a number of underground research laboratories (URL) are in operation in different clay formations. In these URLs, a large number of full-scale in-situ experiments have been performed and are currently planned to be conducted in the future. In the recent years, basic research on clay formations has also been initiated in Germany, in accordance with the R+D programme defined by the German Federal Ministry of Economics and Technology (BMWi).

After the operational phase of a repository the openings, e.g., boreholes, galleries, and chambers, have to be backfilled in order to avoid the release of radionuclides into the biosphere. After healing and resaturation of the excavation disturbed zone (EDZ) and saturation of the backfill, the waste containers and the metallic components will corrode resulting in a generation of hydrogen. Additionally, carbon dioxide will be released as a result of oxidation and thermal decomposition of the organic components in the waste and in the clay. If the disposal boreholes and chambers are sealed gas-tight, high gas pressure may be produced leading to the potential generation of fractures in the host rock which could influence the integrity of the repository. Therefore it is essential that the gases migrate through the technical barriers (backfill) or into the surrounding host rock at lower pressure and without any irreversible damage of the repository.

For long term safety aspects of a repository and especially for the design and construction of sealing elements the knowledge of the consequences of the gas generation, the knowledge of the gas paths in the host rock, and the knowledge of the parameters which influence the gas migration in the host rock are important. On one hand, the sealing elements should not be more gas tight than the surrounding host rock and on the other hand, the sealing elements and the host rock should be permeable enough that no critical gas pressure may be generated in any part of the repository.

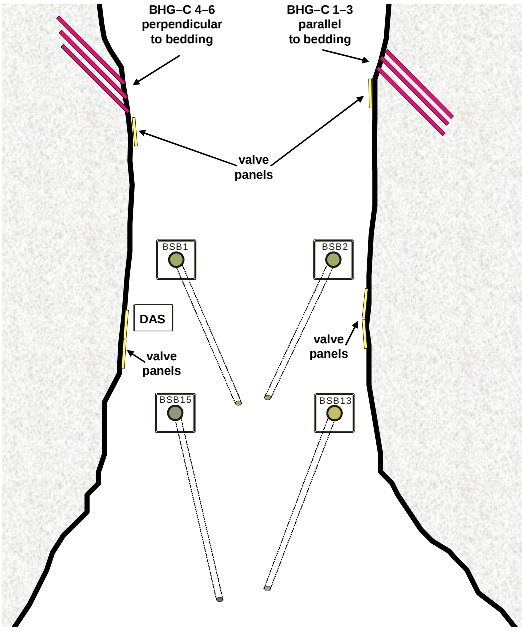
Previous tests in the Mont Terri underground laboratory [6] indicated that gas migration in the Opalinus Clay starts already in the range of gas pressure below 2.0 MPa as a result of dissolution of the gas in the interstitial water and diffusion in the liquid phase. Furthermore, advective gas flow into potentially unsaturated sand or calcite layers may occur. At gas pressures above 2.0 MPa the pore water of the clay will be displaced or gas fracs will be generated almost parallel to the bedding.

In the Mont Terri underground laboratory in Switzerland the objects of the diffusive and advective gas migration in the Opalinus clay as a function of the gas pressure is investigated within an ongoing project.

## LAYOUT OF THE TEST FIELD

In the year 2004, a niche was build for testing the gas and water permeability of different clay sand mixtures in the dry and wet stage. For these tests four vertical boreholes with a diameter of 30 cm and a depth of 3 m were drilled into the floor of this niche. Additionally, the niche was equipped with electricity, a data acquisition system (DAS) and a modem for data transfer.

For investigating the gas migration in the host rock as a function of gas pressure, six additional boreholes with a diameter of 76 mm and a length of 10 m were drilled at the back end of the niche. Three of these named HG-C 1 to 3 at the south-east wall with a dipping of 40 ° parallel to the bedding and three named HG-C 4 to 6 at the north-west wall also with a dipping of 40 ° but perpendicular to the bedding. The central boreholes (HG-C 1 or HG-C 4) were used for gas injection. Boreholes HG-C 2 and HG-C 5 have a horizontal distance of about 30 cm to the injection boreholes and the boreholes HG-C 3 and HG-C 6 have a vertical distance of about 30 cm to the corresponding injection borehole. Figure 1 shows a plan view and a cross section of the niche with the boreholes and the equipment.



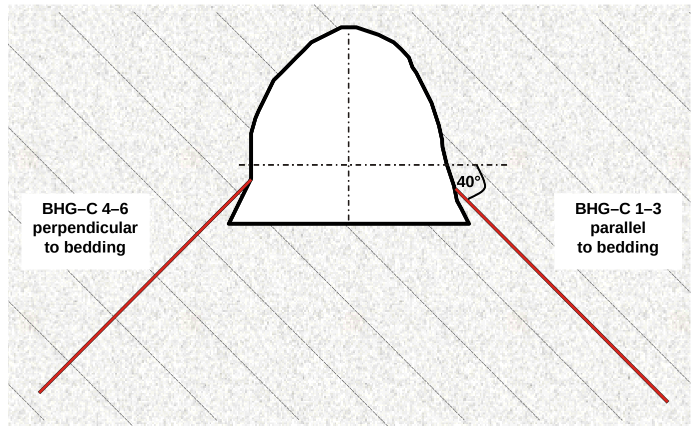




**Fig. 1. Layout of the SB test niche with the boreholes and the equipment.**
**Left: plan view Right: cross section**

Right after drilling, the quadruple packer systems with three test intervals as shown in figure 2 were installed into each borehole. The central interval with a length of 500 mm is used for gas injection whereas the other two guard intervals are for monitoring the tightness of the system. Each interval consists of a ceramic tube with a porosity of 40 % mounted on the main tube in order to minimize the gas volume and to withstand the convergence of the borehole. Into each interval two capillaries run from a valve panel in the open gallery. Via these capillaries tracer gases are injected, gas samples for qualitative and quantitative analyses are taken and the gas pressures within the intervals are determined. Additionally one capillary runs to each sealing element for inflation with water to a pressure of 4.0 MPa. The valve panel is equipped with optical pressure gauges for inflation of the sealing elements and with optical and electronic pressure gauges for determination of the gas pressure in the sample and guard intervals as shown in figure 3.

## INVESTGATION METHODS

At both tests sites (perpendicular and parallel to the bedding) the central interval of one packer probe was inflated with atmospheric pressure with a tracer gas mixture consisting of 88 vol% of nitrogen and 2 vol% of hydrogen, helium, neon, krypton, iso-butane and sulphur hexafluoride each. Three days after injection the gas was extracted and analysed by a gas chromatograph with regard to the tracer gas components. Afterwards the intervals were inflated again at the tracer gas mixture with atmospheric pressure, but extraction and analyses were performed about 32 days later. Additionally, the gas of all the other intervals was extracted and analysed with regard to the tracer gas components.

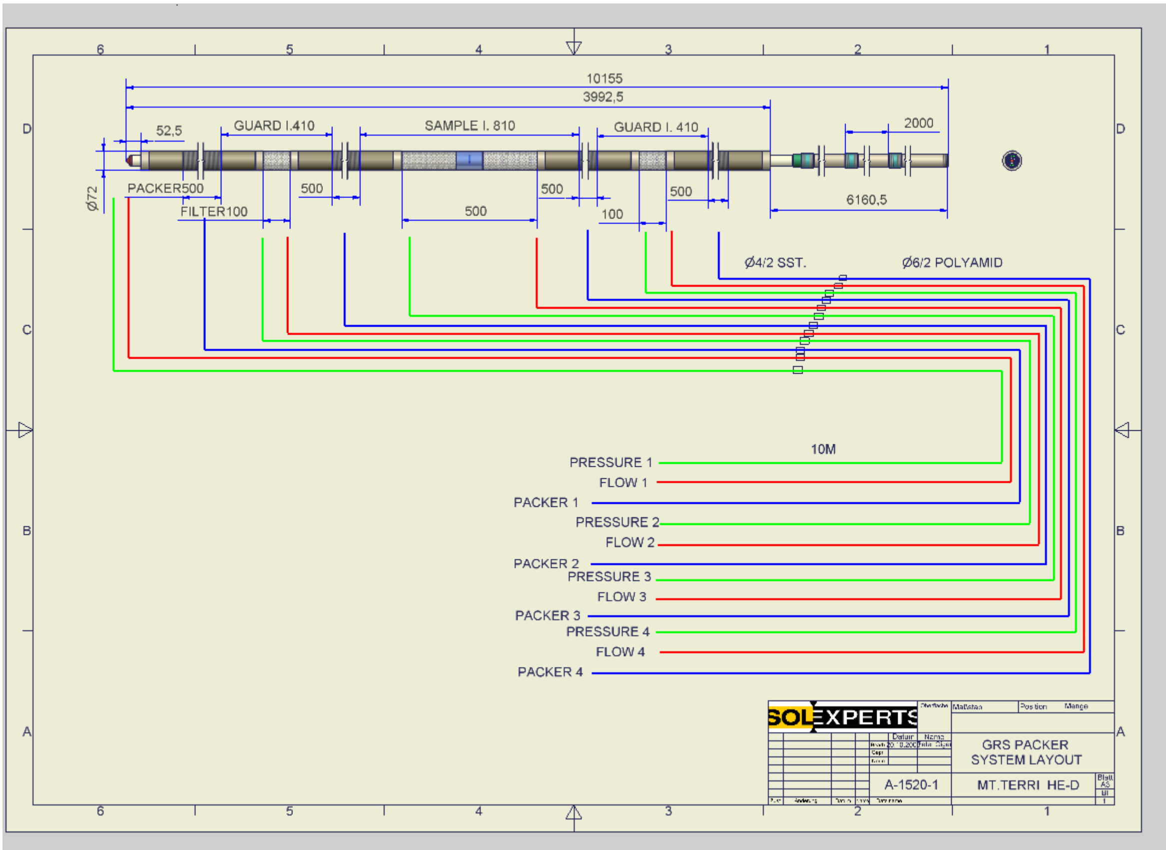



**Fig. 2. Principle drawing of the quadruple packer system with the capillaries for inflation (PACKER 1 to PACKER 4), gas injection (FLOW 1 to FLOW 4), and pressure measurements (PRESSURE 1 to PRESSURE 4).**

Aim of this investigation was to determine if the different gases with different molecular weights and different solubility to water show different migration behaviour. In table I the molecular weights, the solubility in water, and the factor $1/\sqrt{m}$ are compiled. The factor $1/\sqrt{m}$ is proportional to the diffusivity of the gas component.

With the results of the decrease of the concentration in the injection interval and the increase in the other intervals the gas diffusivity of the different gas components was calculated with the computer code ANSYS.

After the measurements for determination of the gas diffusivity all intervals were purged with nitrogen. Then the central intervals of the injection boreholes were inflated with the tracer gas composition and a pressure of 0.1 MPa (0.2 MPa absolute). All the other intervals were inflated with nitrogen at atmospheric pressure (0.1 MPa absolute). The valves to all the intervals were closed and the pressures in the intervals were recorded by the electronic pressure gauges and the DAS. After one month the gas in the intervals was extracted and analyses.

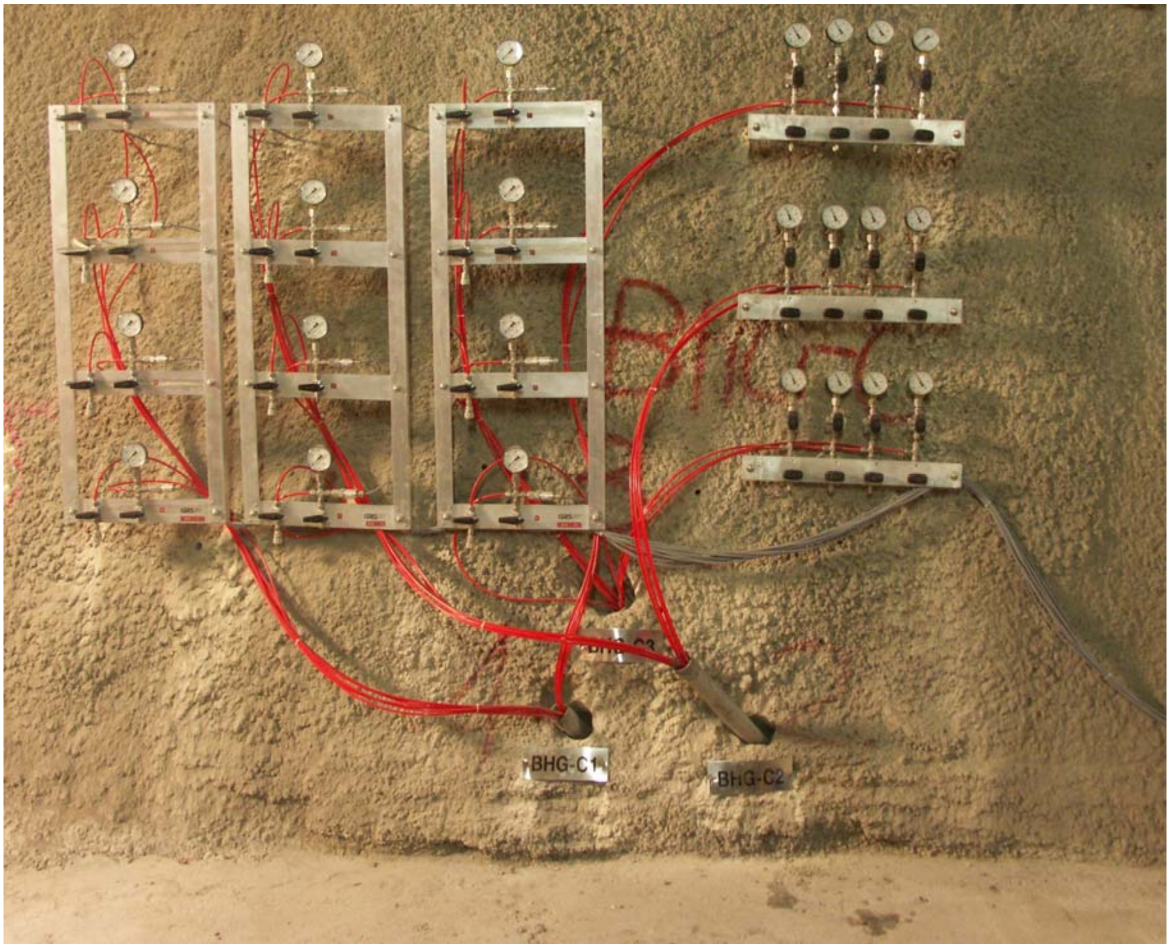



**Fig. 3. Valve panel for gas injection into or extraction from the intervals (left) and inflation of the packers (right) with the pressure gauges (optical and eletronical).**

Table I. Physical data of the tracer gas components

| Tracer gas | Molecular weight m | $\frac{1}{\sqrt{m}}$ | Diffusion in air $10^{-6}$ [$m^2 \cdot s^{-1}$] | Diffusion in water $10^{-9} \cdot$[$m^2 \cdot s^{-1}$] | Solubility in water (at 1 bar) [l Gas·$kg^{-1}$ water] |
|---|---|---|---|---|---|
| Hydrogen | 2.0 | 0.707 | 7.0 | 3.81 | 0.0176 |
| Helium | 4.0 | 0.5 | 6.98 | 5.8 | 0.0083 |
| Neon | 20.0 | 0.22 | 3.07* | 2.8 | 0.01 |
| Krypton | 83.8 | 0.11 | 1.49 | 1.276* | 0.59 |
| Iso-Butane | 56.1 | 0.13 | 1.54* | 1.508* | 0.0325 |
| Sulphur-hexafluoride | 146.0 | 0.08 | 0.75 | 0.928* | 0.0056 |
| Nitrogen | 28.0 | 0.19 | - | 2.34 | 0.0156 |

*All values from literature, except those marked with an asterisk, which are calculated [2], [4].

Afterwards the pressures in the injection intervals were increased stepwise to 0.4; 1.1, 1.5; 2.0, and 3.0 MPa by injecting the tracer gas mixture. In the inflated gas-tight sealed intervals the pressure drops were recorded over a period of at least one month.

With the results of the pressure drop in the injection interval the permeability of the surrounding host rock was calculated with the computer code WELTEST.

**RESULTS**

Gas migration into the Opalinus clay controlled either by gas concentration gradient or by gas pressure gradient between the injection intervals and the surrounding host rock was investigated. For this investigation the gas pressure in the injection intervals was varied between atmospheric pressure and 3.0 MPa.

At atmospheric pressure of the gas mixture in the injection intervals, no significant pressure increase was observed within the time period of one month. This fact indicates that neither gas nor water was released from the surrounding Opalinus clay into that interval. Otherwise a pressure increase should have been observed.

The concentration of the tracer gases hydrogen, helium, neon, krypton, iso-butane, and sulphur-hexafluoride decreased significantly with time. Right after injection all components were in the same concentration of 2 vol% in the matrix of 88 vol% nitrogen. Within three days they decreased to a level between 0.2 and 0.8 vol%. Under the assumption of an almost water-saturated host rock this fact means that the gas components became dissolved in the interstitial water of the Opalinus clay. Afterwards, the dissolved gases migrate in the dissolved stage in the water of the clay. The decrease is a result of the solubility of the gases in water (Bunsen coefficients) [4] and of the diffusion of the gases in the aqueous phase (see table 1).

With the finite element code ANSYS the diffusion coefficient of the component hydrogen, which is the most important gas in a repository, was determined in the range of $10^{-9}$ $m^2s^{-1}$. The other components have diffusion coefficients in the range between $10^{-9}$ and $10^{-10}$ $m^2s^{-1}$. A definite dependence on the molecular weight or on the solubility in water could not be determined for the influences of both corresponding parameters do overlap.

Figure 4 show the evaluation of the gas pressure in the injection interval when inflated stepwise up to 3.0 MPa. Each step lasted between 27 and 50 days. At each pressure step significant pressure decay was observed. The pressure decay is the result of the two complementary gas migration effects in the clay:

1. Diffusion in the aqueous phase as a result of solution of the gas in the interstitial water. The resulting diffusion coefficients are in the range of $10^{-9}$ $m^2s^{-1}$ at low gas pressure and increase to $10^{-8}$ $m^2s^{-1}$ at higher gas pressure.
2. Advection in the non water saturated pore volume of the clay. The resulting permeability is in the range of $10^{-19}$ $m^2$.

When increasing the gas pressure in the injection intervals a sudden increase of gas flow was observed at 2.3 MPa in the borehole parallel to the bedding and at 3.0 MPa in the borehole perpendicular to the bedding. This gas flow resulted in an increase of gas pressure in the adjacent intervals. After stopping gas injection the pressure stabilised in all the intervals. This effect is caused by the gas break-through in the almost water saturated clay by displacing the interstitial water in the pore volumes.

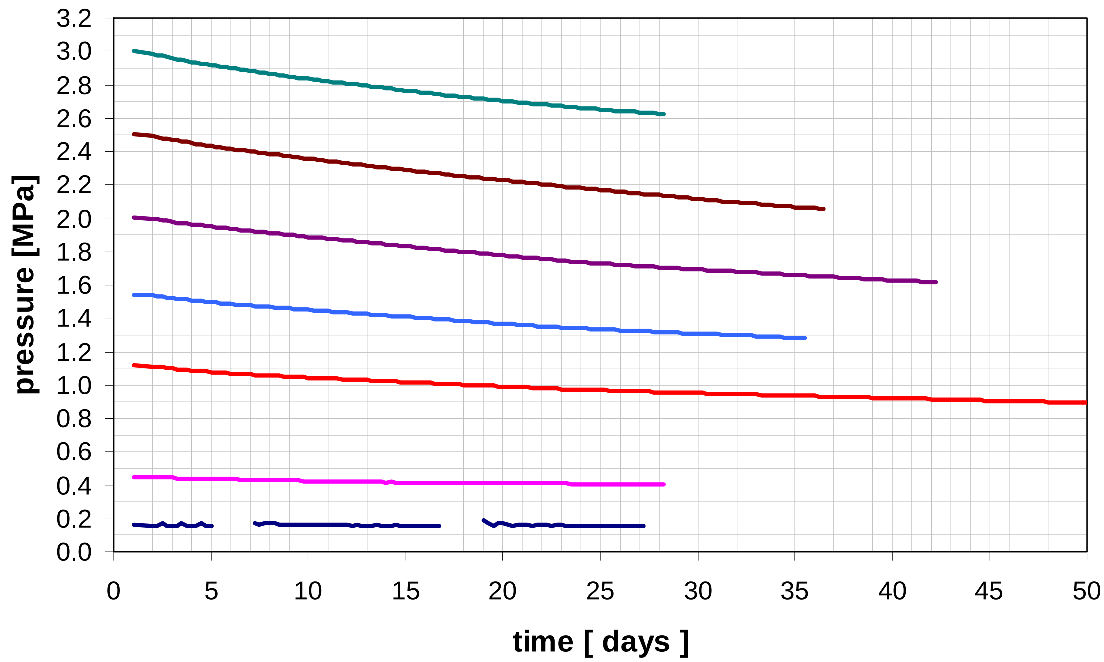



**Fig. 4. Pressure trends in the injection interval of the borehole perpendicular to bedding at different pressure steps between 0.18 and 3.0 MPa.**

## CONCLUSIONS

The gas injection tests in the undisturbed water saturated Opalinus clay outside the excavation damaged zone (EDZ) indicated that already at low gas pressure (atmospheric pressure) gas migration into the surrounding host rock takes place. With increasing injection pressure the migration rate increases. At gas pressures between 2.3 and 3.0 MPa gas break-through was observed, which means that the interstitial water of the clay was displaced by the gas.

These results indicate that gas generation in a repository as a result of corrosion of the metallic waste or by thermal and microbial degradation of the organic waste should not be a serious problem. Nevertheless further investigations on the gas generation rate in sealed areas of a repository and on the gas migration rates under the geometric and geologic conditions of the repository have to be performed in order to exclude a critical gas pressure build up.

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